

# Mechanistic modelling of erosion and desiccation cracking of swelling clays

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#### Abstract

The understanding of the coupled hydraulic, chemical, and mechanical behaviour (HCM) of swelling clays has been advanced substantially over the last few decades. Less advanced, however, is the understanding, and from there the predictive modelling, of phenomena involving discontinuous processes such as erosion and desiccation cracking. The ability to describe rigorously the mechanisms leading to erosion and desiccation cracking is fundamental for the formulation of physically realistic models, which are required for safety/longevity assessment of clays as sealant or barrier materials. The research presented in this thesis aims to advance the understanding and predictive modelling of coupled hydro-chemo-mechanical processes involved in erosion and desiccation cracking at particle/micro level to macroscopic events, and the corresponding numerical implementations.

Rigorous mathematical descriptions of clay erosion and cracking must include both continuous (swelling/deformation) and discontinuous (damage/fracture, particles detachment) processes. The latter involve sequences of discrete events of finite spatial dimension, and therefore cannot be described in principle by the classical approach based on local/differential formulations of processes. A non-local approach based on Peridynamics theory (PD) is selected for the development of coupled HCM models in this work. It has been previously demonstrated that PD formulations are particularly suitable and effective for solving problems with large deformation and discontinuities. This quality of PD is further confirmed in the present work by comparing models' predictions with experimental data at all stages of model development.

A PD model which couples water flow and chemical transport in unsaturated clays is formulated first as a foundation for erosion. This is used to develop a new erosion model by coupling free swelling, detachment of clay particles and transport of detached particles by flowing water. The erosion model is used to investigate the effects of solution chemistry and flowrate on the penetration, extruded mass, and particle release rate of compacted bentonite. This model is extended by a new formulation for co-transport of accessory minerals with clay swelling, which is used to investigate the role of accessory minerals in the erosion of compacted bentonite. The erosion model is applied to study the long-term performance of clay buffer under variable hydrochemical conditions. The agreement with experimental data strongly supports the applicability of the model to account for the hydro-chemical conditions (composition and velocity) of the eroding environment. The model for erosion with co-transport of accessory minerals is shown to reproduce qualitatively experimental data, provide explanations for the effect of accessory minerals on erosion, and deliver quantitative predictions for this effect.

A PD model for desiccation cracking is formulated by coupling deformation and water transport. The validations of this model show excellent agreements between numerical and experimental results. Specifically, the desiccation cracking model shows the correlations between the shrinkage of clay, changes in displacement fields and crack growth as experimentally observed. It is shown that the cracking model captures realistically key hydraulic, mechanical and geometry effects on clay desiccation cracking models are finally integrated into a single computational framework, which is used to investigate the erosion of swelling clays assisted by piping flow in cracking channels. It is shown that the integrated model can explain the self-healing of swelling clay in piping channels and predict the mass loss of swelling clays under piping flow for backfill and plug-in material.

The agreements between model predictions and experimental data with different coupled processes and environment conditions suggest that the newly developed non-local theoretical and computational tools for analysis of clay cracking and erosion can be used under variable hydrochemical conditions. The proposed multi-physics non-local formulations provide a robust framework for incorporating a wide set of additional couplings, such as gas and heat transport, and geochemical reactions.

#### Declaration

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#### Preface

Chapter 3 has been published in *Journal of Hydrology*. (Yan, H., Sedighi, M. and Jivkov, A.P., 2020. Peridynamics modelling of coupled water flow and chemical transport in unsaturated porous media. Journal of Hydrology, 591, p.125648. https://doi.org/10.1016/j.jhydrol.2020.125648)

Chapter 4 has been published in *Géotechnique*. (Yan, H., Jivkov, A.P. and Sedighi, M., 2021. Modelling the soil desiccation cracking by peridynamics. Géotechnique, pp.1-35. https://doi.org/10.1680/jgeot.21.00032)

Chapter 5 includes two published works, one in Géotechnique and another one in Engineering Geology (Yan, H., Sedighi, M. and Jivkov, A., 2021. Modelling the effects of water chemistry and flowrate on clay erosion. Engineering Geology, 294, p.106409. https://doi.org/10.1016/j.enggeo.2021.106409; Sedighi, M., Yan, H. and Jivkov, A.P., 2021. Peridynamic modelling of clay erosion. Géotechnique, pp.1-12. DOI: https://doi.org/10.1680/jgeot.20.P.149), and a manuscript submitted for publication in a refereed journal (Yan, H., Jivkov, A. and Sedighi, M. 2021. Co-transport of accessory minerals during expansion of compacted bentonite and its impact on erosion, **under review**)

Chapter 6 is a manuscript to be submitted for publication in a refereed journal. (Title: Piping assisted erosion of swelling clays).

## CHAPTER ONE

# **1. Introduction**

#### 1.1 Background

Swelling clays are widely used or considered as sealing materials and environmental barriers, e.g., in high-level radioactive waste repositories, landfill systems, plugging of water, oil and gas wells, seismic shot holes, mining shafts and exploratory holes (James, 1996; Wheaton et al., 1994; Englehardt et al., 2001; Holl and Scheuermann, 2018; Holl, 2019; Towler et al., 2020; Yan et al., 2021; Sedighi et al., 2021). For example, compacted bentonite clays are considered as one of the engineered barriers in the geological disposal of high-level radioactive waste (HLW) (Skb, 2011; Enresa, 2000; Jnc, 2000; Nagra, 2002; Aecl, 1994; Andra, 2005; Nda, 2010). Bentonite clays have also been recommended and utilized as engineered barriers to seal abandoned oil and gas wells. Compacted clay liners can further provide low hydraulic conductivity barrier to prevent rapid migration of contaminants from landfill leachates migrating towards adjacent areas (Yan et al., 2020 & 2021). The understanding of the hydrochemical-mechanical processes in clay soils, and their effects on the clays' engineering behaviours, has progressed substantially in the last 50 years through extensive experimental investigations and theoretical developments (Sedighi, 2010). Whereas important developments have been made in coupling mechanical with hydro-chemical effects for analyses of deformation and consolidation, the progress in modelling of clay desiccation cracking and swelling/erosion has been very limited.

Clay soils are susceptible to shrink and swell due to the water loss and absorption, which may lead to geological hazards (e.g., desiccation crack and swelling/erosion damage) to buildings, infrastructure and engineering barriers and financial loss. For example, estimated damage caused by shrink–swell soils to the buildings and infrastructure exceeds £64 million during July to September in 2018 in UK (ABI, 2018). Understanding and predictive modelling of clay cracking and erosion are therefore essential for a wide range of geotechnical engineering problems. For example, cracking accelerates infiltration in road embankments and soil slopes and causes the failures of anti-seepage landfill systems (e.g., liners and covers) (Wan et al., 2018). Further, desiccation cracking can damage local ecologies as cracks increase the surface roughness and accelerate evaporation in soil, contributing to faster soil weathering and erosion leading to potential dust storms, landslides and debris flow (Zeng et al., 2020). Additionally,

desiccation cracks can create pathways for water and gas transport, which will reduce the performance of clay barriers used for waste isolation (Wan et al., 2018). In the last few decades, extensive studies have been devoted to developing experimental, theoretical and numerical methods to understand the fundamental drying cracking mechanisms and characterizing the cracking behaviour of soils, including number of cracks, crack width, depth and area, initiation and propagation of cracks etc (Tang et al., 2021). Several review papers published in recent years have synthesized the past research efforts devoted to the topics: Peron et al. (2009) reviewed the theoretical developments and global mechanisms of soil desiccation, including drying shrinkage and cracking; Kodikara and Costa (2013) presented a summary of the work undertaken on soil desiccation cracking, especially in field observations and laboratory modelling; Bordoloi et al. (2020) provided an insight into the effects of vegetation on the desiccation process considering the soil-water-plant interaction; Tang et al. (2021) reviewed the lab- and field-scale investigation approaches, cracking dynamics, and influencing factors of desiccation cracking processes. The major phenomena observed during the dynamic cracking process have been interpreted and justified via experimental and field tests; however, it remains difficult to accurately quantify or predict the initiation and propagation of cracks (Tang et a., 2021). Table 1 summarizes the effects of desiccation cracking on geotechnical related problems.

Clay erosion can also cause a series of engineering problems, from clay piping erosion in embankments (Fujisawa et al., 2009), through erosion of buffer or piping assisted erosion of backfill in geological disposal of higher activity radiative waste (HLW) (Schatz et al., 2013; Sane et al., 2013), to erosion of geosynthetic clay liners (Ashe et al., 2014) and erosion of clay plugs in boreholes (Sandén et al., 2008). The mechanisms of swelling erosion have been extensively investigated in recent years, especially their potential impacts on the long-term integrity of bentonite buffer in the geological disposal (Belbar, SKB, Posiva and Poskbar project). The recent experimental investigations of swelling clay erosion can be classified into two groups: free swelling tests and coupled expansion and erosion tests. The first group focused on investigating the swelling/expansion behaviour of clay under no flow conditions, including simulated oedometric tests (Sołowski and Gallipoli 2010), free swelling in tube tests (Dvinskikh et al., 2009; Liu et al., 2010; Li et al., 2020), extrusion behaviour in a narrow channel (Harjupatana et al., 2015) and expansion of swelling clay in artificial fracture systems (Schatz et al., 2013; ).The second group accounts for coupling swelling and erosion of expansive clay with flowing water, including pinhole test simulation (Sane et al. 2013),

dynamic erosion test (Alonso et al., 2018; Bouby et al., 2020) and erosion behaviour of swelling clay in artificial fracture system combined with the effects of various water chemistry (Schatz et al., 2013; Hdestrom et al., 2016). Table 2 summarizes the effects of erosion problems on various areas.

Table.1 Summary of the effects of desiccation cracking on various geotechnical problems

Area	Influences of desiccation cracking	References
Slope	Desiccation crack can affect vertical permeability of highly plastic clay soil and reduce unsaturated slope stability.	Khan et al. (2017)
Embankment	cracking accelerates infiltration in road embankments, leading to potential erosion pathways and piping failure.	Foster et al. (2000)
Dam	The cracks in soils can facilitate water infiltration and reduce the soil shear strength. Moreover, cracks can form part of a slip surface that has no shear strength.	Talbot and Deal (1993); Peng and Zhang (2012);Tang et al. (2021)
Foundation	Clay shrinkage causes settlement and reduces bearing capacity of the system.	Yilmaz et al. (2014)
Clay liner	Desiccation cracks can create pathways for water and gas transport. The crack opening and closure due to the wetting-drying processes can lead to the erosion damage to clay liner.	Southen et al. (2015); Wan et al. (2018); Rowe and Jabin (2021)
Nuclear waste disposal	Cracking of clay buffer may drastically increase the permeability of the clay barrier, which will reduce the performance of clay barriers used for waste isolation.	Davy et al. (2007); Gourc et al. (2010);
Mine tailing	Cracking changes the properties of mine tailing and results in an environmental impact, including creating paths for the infiltration of rainwater.	Rodríguez et al. (2007)
Highway shoulder	Once cracks develop in the paved highway shoulders due to shrinking of underlying and adjacent soils, intrusion of surface water runoff into underlying soil layers occurs and soften the base and subgrades both directly beneath the shoulders and adjacent to shoulders.	Intharasombat et al. (2007)

Area	Influences of erosion	References
Slope or dam failure	Continuous erosion of a dam can lead to a dam breach or flooding and trigger landslides.	Feng et al. (2020)
Embankment	Erosion may widen the upstream of the crest and results in overflow, which may lead to the failure process of an embankment.	Fujisawa et al., 2009
Foundation	The loss of a part of the solid phase induced by internal erosion process can lead to the modification and degradation of soil mechanical properties and risk of failure.	Yang et al. (2019 & 2020)
Geosynthetic clay liners	Wetting and drying cycles can lead to the formation of erosion features in geosynthetic clay liners. Erosion can result in an increase in hydraulic conductivity of at least one order of magnitude.	Rowe et al. (2014); Mukunoki et al. (2019)
Nuclear waste disposal	The swelling clay buffer can be eroded by the hydro-chemical interactions at the interface between the clay and the host rock, resulting in a gradual loss of the barrier and a co-transport of radionuclides into the biosphere.	Schatz et al., (2013); Sane et al. (2013); Yan et al. (2021); Sedighi et al. (2021)
Tunnel excavation	Piping erosion was reported to be especially important for the deposition tunnel backfill in the early phase of the saturation process if water flows into a deposition hole from fractures in the bedrock	Navarro et al. (2016)
Clay plugs in boreholes	Mass erosion in the annular gap between clay and rock may lead to the failure of sealing of bentonite plug in the borehole during the emplacement.	Sandén et al. (2008).

#### Table.2 Summary of the effects of erosion on various area

The developments in numerical modelling capabilities for simulating coupled hydro-chemical effects on swelling clay cracking and erosion process are the critical steps forward in the assessment and prediction of geoenvironmental solutions. Such models are of interest to a wide range of geotechnical and environmental engineering projects by coupling basic physical, chemical and mechanical processes including the impacts of water, chemistry and mechanics. Numerical simulations are widely used to assist in the study/investigation of engineering problems via developing sound mathematical models and validation with experimental observations. The validated numerical models can be applied to tackle complex engineering problems by computational simulation of various scenarios and predict future behaviour. Computer simulation is a safer and cheaper way to understand the problems we are interested in; additionally, it can speed things up or slow them down to see changes over long or short periods of time and explore unexpected problems. While these numerical models facilitate the engineering problems to a certain extent, it is critical to understand the assumptions and limitations of such computer simulations (Jing, 2003; Sedighi, 2010; Fairhurst and Long, 2018; Zendehboudi et al., 2018).

Approximate solutions to coupled hydro-chemical-mechanical processes in clays, described by the classic local theories, i.e., by partial differential equations, can be obtained by various numerical techniques, e.g., the Finite Element Method (FEM), the Finite Difference Method (FDM), and the Boundary Element Method (BEM) (Zhang, 2004; Kolditz et al., 2012; Pandey et al., 2018; Cui et al., 2018). The local formulations, however, are less efficient in analysis of problems with discontinuities (Jafarzadeh et al., 2019a; Yan et al., 2020 & 2021). For example, the analysis of clay desiccation cracking requires specification of additional conditions for crack initiation and growth, involving parameters that fundamentally depend on the specific geometry and boundary conditions. This makes the calibration of the parameters specific to the experimental setup and the predictions for the behaviour under different conditions unreliable. Even more pronounced is the inability of the local formulations to describe clay erosion, as such description must represent the strong coupling of free swelling (continuous deformation), particle detachment at clay/water interface (discontinuous particle release and moving interface) and detached particle transport.

There are strong couplings between the physical and chemical processes that govern the mechanical behaviour of clay soils, and this complexity, added to the potential emergence and evolution of material discontinuities, is the major challenge and the reason for the limited number of theoretical studies and model developments of clay cracking and erosion. Previous research on and applications of non-local theories, such as Peridynamics (PD), have demonstrated their benefits in handling large deformations and discontinuities (Silling, 2000). The PD formulation replaces the partial differential equations of the classical local theories by a set of integral-differential equations, producing a mathematically consistent description of the material behaviour even when strong discontinuities (e.g., cracks or fractures) are presents or emerge due to breaking and fragmentation of the material (Silling, 2000; Jafarzadeh et al., 2019a and 2019b;). Figure 1 shows the comparison of local approach and peridynamics. The advantages of PD compared to classical local approaches include: (i) PD based models have the capacity to capture discontinuous evolution in the computational domain (e.g. crack initiation, nucleation and propagation, dissolution and erosion at the interface); (ii) PD allows for both mesh-free and mesh-based discretization; (iii) in PD it is simple to include multiple physical fields; and (iv) Length scale can be exploited for multiscale modelling. The PD theory has been well established for prediction of fracture propagation in solids and successfully applied in different areas, including dynamic fracture evolution (Rabczuk and Ren, 2019; Ren et al., 2017), fluid driven fractures (Ouchi et al., 2015), thermo-mechanics of fracture

development for composites, rocks, ceramic and pellets (Oterkus et al., 2014; Bobaru and Duangpanya, 2010; Bobaru and Duangpanya, 2012; D'Antuono and Morandini, 2017; Wang and Zhou, 2019; Wang et al., 2018; Oterkus and Madenci 2017; Gao and Oterkus 2019), membranes and fibers (Silling and Bobaru, 2005), dynamic fracture of nanofiber (Bobaru, 2007), stress induced corrosion (Bobaru and Duangpanya, 2012; Chen and Bobaru, 2015; Jafarzadeh et al., 2019a and 2019b), hydraulic fracturing (Oterkus et al., 2014; Ouchi et al., 2015; Zhou et al., 2020) and multiphysics flow problems in unsaturated porous media (Yan et al., 2020).

Application of peridynamic theory to clay cracking and erosion is promising thanks to its ability to capture the crack initiation and propagation for failure analysis (D'Antuono and Morandini, 2017; Wang and Zhou, 2019; Oterkus and Madenci 2017; Gao and Oterkus 2019) and handle the problems with moving boundaries due to the discontinuous detachment of particle at reaction interface (Chen and Bobaru, 2015; Jafarzadeh et al., 2019a and 2019b; Sedighi et al., 2020; Yan et al., 2021). This work offers one approach to advance the understanding and modelling of the erosion and cracking of swelling clays, including hydrochemical effects on these phenomena, by developing a non-local multi-physics formulation based on the PD theory. The computational implementation of the formulation developed as part of the work is a unique tool for accurate and robust performance assessment of swelling clays.



Fig. 1. Comparison of local method (a) and peridynamics (b). In local approach, the state of a material point (x) is only influenced by its immediate neighbours (Fig.1a). However, in PD, a material point (x) is connected to other material points within a neighbourhood/horizon  $(H_x)$ .

#### 1.2 Aim and objectives

The research presented in this thesis aims to advance the understanding of coupled hydrochemo-mechanical processes involved in erosion and desiccation cracking of swelling clays and provides theoretical descriptions; linking the forces and interactions at particle/micro level and macroscopic events. The research presented in this thesis focuses on developing a solid non-local theoretical framework/tool that can be used to predict/study clay cracking and erosion damage. In particular, clay cracking model needs to capture the main features/phenomena involved in dynamic cracking process, including the initiation and propagation of cracks, crack numbers and final pattern. Erosion model needs to explain the stability of clay gel/sol particle at clay/water/rock interface. Additionally, the erosion model should have capacity to quantify the mass loss of clay due to erosion damage by coupling continuous swelling process and discontinuous particle detachment at clay/water interface. The nature of the processes involved in erosion and cracking dictates a gradual development of the framework, which is reflected in the following specific objectives:

- 1) To advance the key coupling on PD-based mathematical descriptions of water flow and chemical transport under variable saturated conditions as the base for the erosion model.
- To improve the understanding of swelling clay erosion by incorporating the process of clay particle detachment via considering the stability/strength of hydrated gel and revisiting the clay water interaction and forces involved at micro scale.
- 3) To advance the knowledge of water chemistry effects on the expansion of swelling clays
- 4) To identify the underpinning of safety/longevity assessment of clays as sealant materials via quantifying the mass loss of clay due to erosion under various conditions, including the impacts of water velocity and chemistry, in particular, the water insoluble accessory minerals on expansion/erosion process of swelling clay.
- 5) To develop an understanding of the impacts of piping flow on assisting erosion of swelling clays via applying the full modelling framework and its application for self-healing and erosion of buffer/backfill and plug-in material.

The project addresses in full the need for realistic and robust modelling of clay cracking and erosion.

#### **1.3 Scopes and limitations**

The scope of the work undertaken in this thesis and in particular the limitations that are anticipated are listed below:

- Non-local chemical transport model is applicable for single specie only. Multi-chemical transport and geochemical reaction is not considered.
- Water flow and evaporation in drying clay is described by using diffusion equation with the assumption of constant diffusion coefficient, which may limit the developed non-local clay desiccation model to the stage of shrinkage during the saturated state.
- The shrinkage of soil is described by Biot's poro-elasticity. Plastic deformation during shrinkage process is not considered in this thesis.
- Free swelling process of clay is described by a diffusion-like equation, which is developed based on the dynamic force balance. The present swelling model cannot account for the effects of external force/stress on the deformation of clay.

• The theoretical developments of erosion model are limited to the bentonite material consisting of smectite and insoluble accessory minerals, although the dissolvable minerals (e.g., feldspar, calcite, etc.) may lead to the variations of water chemistry due to the dissolution reaction during the expansion/erosion process.

#### 1.4 Overview of the thesis

A brief description of each chapter is presented below.

Chapter 2 presents a comprehensive review of the current knowledge of clay erosion and cracking, including a review of the recent experimental observations and modelling development. A selected review of the non-local approach (peridynamics, PD) used to address the erosion and desiccation problem is also presented.

Chapter 3 describes the theoretical framework for water flow and chemical transport in unsaturated soil. The main focus is to present the development of the non-local formulations for chemical transport and water flow in heterogeneous/discontinuous porous media. Modelling this coupling is essential for the development and evaluation of erosion and desiccation cracking models.

Chapter 4 presents the model development for clay desiccation cracking. The model is developed by coupling water flow and mechanical deformation in non-local framework. The model is used for investigating and explaining the mechanisms controlling the initiation and propagation of discontinuities.

Chapter 5 proposes a non-local formulation for clay erosion, which brings together clay swelling, particle detachment and detached particle transport, into a single modelling tool. The detachment criterion, an essential element of the integrated erosion model, is first tested against a series of experimental benchmarks to show the excellent agreement between calculated and experimental results. The developed model is then extended to study the effects of, mineral content, water chemistry and flowrate on erosion behaviour of clay.

Chapter 6 is the application of the models to study erosion of swelling clay in the cracks/piping channels. The swelling model is adopted to investigate the self-healing potential of MX-80 bentonite in the gaps/piping channels. The coupled swelling and erosion model is used to quantify the amount mass loss of swelling clay in cracks/piping channel by flowing water.

Chapter 7 is the conclusions of the work and suggestions for future research.

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# CHAPTER TWO

## 2. Literature review

#### **2.1 Introduction**

Clay is widely used as a sealing material in engineered barriers due to its low permeability, high sorption capacity and self-sealing properties. The understanding of the hydro-chemical processes in clay soils, and their effects on the clays' engineering behaviour, has progressed substantially in the last 50 years or so through extensive experimental investigations and theoretical developments. The accumulated experimental evidence has been used to propose constitutive relationships and to formulate predictive models for hydro-mechanical, hydro-chemo-mechanical and thermo-hydro-chemo/geo-chemo-mechanical behaviour of clays (Thomas et al., 1998; Gens et al., 1998; Guimarães et al., 2006; Gens, 2010; Sedighi et al., 2015 & 2018). While substantial progress has been made in coupling mechanical with hydro-chemical effects for analysis of, e.g., deformation and consolidation, the progress in modelling important phenomena involving discontinuous processes, such as clay erosion and desiccation cracking, has been very limited. Understanding and predictive modelling of clay erosion and cracking are important in a wide range of geotechnical engineering problems.

Clay erosion may cause a series of engineering problems, from clay piping erosion in embankments (Fujisawa et al., 2009), through erosion of buffer or piping assisted erosion of backfill in geological disposal of higher activity radiative waste (HLW) (Schatz et al., 2013; Sane et al., 2013), to erosion of geosynthetic clay liners (Ashe et al., 2014) and erosion of clay plugs in boreholes (Sandén et al., 2017). The mass loss induced by erosion may reduce the performance of clay as a sealing material and as a component in engineered barrier system (EBS). For example, during the long-term operation of disposal facilities, the swelling clay buffer can be eroded by the hydro-chemical interactions at the interface between the clay and the host rock, resulting in a gradual loss of the barrier and a co-transport of radionuclides into the biosphere (Fig.1). Piping erosion was reported to be especially important for the deposition tunnel backfill in the early phase of the saturation process if water flows into a deposition hole from fractures in the bedrock (Navarro et al., 2016). Additionally, mass erosion in the annular gap may lead to the failure of sealing of bentonite plug in the borehole during the emplacement (Sandén et al., 2017).



Fig.1 Bentonite erosion and colloid assisted transport of radionuclides.

Emergence and evolution of cracks in a complex, heterogeneous clay structure is important for many applications. For example, cracking accelerates infiltration in road embankments and soil slopes and causes anti-seepage failures of landfill barriers (Wan et al., 2018). Further, desiccation cracking can damage local ecologies as cracks increase the surface roughness and accelerate evaporation in soil, contributing to faster soil weathering and erosion leading to potential dust storms, landslides, and debris flow (Zeng et al., 2020). Additionally, desiccation cracks can create pathways for water and gas transport, which will reduce the performance of clay barriers used for waste isolation (Wan et al., 2018).

Coupled desiccation and erosion processes may cause significant damage to clays' engineering behaviour. For example, the performance of clay buffer may be degraded from desiccation behaviour under drying induced by the heat generated from HLW due to the long-term decay of nuclear waste (Tan et al., 2020). These desiccation cracks can create natural pathways to assist the erosion (mass loss) of buffer after re-saturation due to groundwater intrusion. Additionally, the desiccation cracks induced by the drying process may partially inhibit the void sealing process (Wang et al., 2021). An illustration of the erosion and desiccation involved in the long-term performance of a buffer is provided in Fig.2. GCLs as a key component for liner system may experience cyclic wetting (by hydration from the subsoil) and drying (due to evaporation). Shrinkage desiccation cracks of GCLs due to solar radiation are attributed to the formation and development of erosion channels and mass loss of GCLs (Ashe et al., 2014).



Fig. 2 Illustration of the erosion and desiccation processes involved in the long-term performance of a clay buffer in a repository for high-level nuclear waste (Chen, 2021).

Erosion and desiccation cracking of clay are fundamental geotechnical engineering problems with complex mechanisms involved (deformation, damage, and particle detachment) and challenges in their mathematical formulations (continuum deformation to discontinuous damage and erosion). The understanding and predictive modelling capability for clay erosion and desiccation is therefore critically important. The overall aim of the chapter is to present recent experimental and modelling results for the impact of clay erosion and desiccation cracking on clay behaviour. This state-of-the-art review includes five aspects. Section 2 shows the current understanding of the microstructure, force interactions, and stability of bentonite clays. Section 3 presents existing experimental observations of clay erosion and desiccation. Section 4 discusses the mechanisms and corresponding modelling efforts of clay erosion and cracking. Section 5 is dedicated to a review of the modelling approach adopted in the thesis – the non-local peridynamic theory and its applications. This section justifies the selection of peridynamics, emphasising its advantages for tackling discontinuous processes, such as clay erosion and desiccation cracking.

#### 2.2 Properties of bentonite

#### 2.2.1 Structure

The components of bentonite clay are montmorillonite and accessary minerals, e.g., kaolinite, illite, quartz, feldspars, gypsum, calcite, pyrite, and different iron oxides/hydroxides. The

chemical and physical properties of bentonite are mainly controlled by the montmorillonite content and type, e.g., sodium-montmorillonite and calcium-montmorillonite. A montmorillonite unit is built of an alumina octahedral sheet sandwiched between two silica tetrahedral sheets. The central cation of the unit octahedron is Al<sup>3+</sup> with six hydroxyls at each corner. The central cation of the unit tetrahedra is  $Si^{4+}$  with four oxygen atoms at each corner. The process of the replacement of an element with another element is referred to as isomorphous substitution without modifying its chemical structure (Uddin, 2018). For example, Si<sup>4+</sup> may be replaced by Al<sup>3+</sup> in a tetrahedron and the replacement of Al<sup>3+</sup> by Mg<sup>2+</sup>, Fe<sup>2+</sup>, Fe<sup>3+</sup> occurs in octahedron. Such substitutions create a net of negative charge for montmorillonite, which enhances cations adsorption and anions repulsion. The compensating cations determine the cation exchange capacity (CEC), whose value ranges from 0.27 to 0.87 (meq/100 g) (Karnland et al. 2006). The montmorillonite is built of repeating unit layers, e.g., tetrahedral/octahedral/tetrahedral (TOT layer). The interconnection of these montmorillonite layers determines the complexity of the geometrical and topological properties of bentonite, and therefore results in the complicated physical-chemical behaviour of bentonite (Chen et al., 2021). Figure 3 shows one unit layer and a portion of a second unit to illustrate that the space between layers contains exchangeable cation and water. The negative charge on the montmorillonite surface enables the bentonite to swell up to several times of its original volume.



Fig. 3 Diagram of the unit structure of montmorillonite (Steinmetz, 2007).

#### 2.2.2 Forces and interactions in smectite (montmorillonite)

#### Attractive forces: van der Waal's forces

The main attractive force among colloids is van der Waal's forces (vdW), which is a distance dependent interaction force between atoms or molecules. vdW forces are relatively weak with a strength of 0.4 to 4 kJ/mol, but are critically important for the structural integrity. The existing studies show that the interaction among bentonite clay particles (coin shaped colloid) is dominated by face-to-face arrangement instead of face-to-edge arrangement (Jonsson et al., 2008). The interaction energy,  $U_{VdW}$ , and the interaction force,  $F_{VdW}$ , between bentonite plate for face-to-face arrangement can be described by (Hedstrom et al., 2016)

$$U_{VdW} = -\frac{A_H S_p}{12\pi} \left[ \frac{1}{h^2} - \frac{2}{\left(h + \delta_p\right)^2} + \frac{1}{\left(h + 2\delta_p\right)^2} \right]$$
(1)

and

$$F_{VdW} = -\frac{A_H S_p}{6\pi} \left[ \frac{1}{h^3} - \frac{2}{\left(h + \delta_p\right)^3} + \frac{1}{\left(h + 2\delta_p\right)^3} \right]$$
(2)

where  $S_p$  is the surface area of the bentonite plate,  $\delta_p$  is the thickness of bentonite plate,  $A_H$  is the Hamaker constant, and *h* the separation between the plates. Typical values of  $A_H$  for montmorillonite-montmorillonite interactions across an aqueous interlayer range from 10<sup>-20</sup> J to 2×10<sup>-20</sup> J at *T*=298K (Iwata et al. 1994). It should be noted that the dissipation due to crack and chemical reactions is not considered in this study.

#### **Repulsive forces: Diffuse double layer forces**

Derjaguin-Landau-Verwey-Overbeek (DLVO) theory is extensively used to predict the repulsive forces in Na-smectite colloid system. However, it has been noted that DLVO theory may not be applicable to Ca-smectite colloid system as the effect of ion-ion correlations has to be included (Liu et al., 2009). In this section, the approximate expressions for the repulsive force interaction between Na-smectite parallel plates at constant charges is reviewed. The expression for the diffuse double layer force (DDL) for Na-smectite parallel plates within the framework of the DLVO theory is given by (Liu et al., 2009):

$$F_R = 2cRTS_p(\cosh y^m - 1) \tag{3}$$

where T is the absolute temperature, c is the ionic concentration in the pore water system, R is the universal gas constant, and  $y^m$  is the scaled electric potential in the midpoint between parallel plates. This potential is a function described by (Liu et al., 2009; Sedighi et al., 2021):

$$y^{m} = \sinh^{-1} \left[ 2\sinh(y_{\infty}^{m}) + \frac{4}{\kappa h} \sinh\left(\frac{y_{\infty}^{h}}{2}\right) \right]$$
(4)

where

$$y_{\omega}^{m} = 4 \tanh^{-1} \left[ 2 \tanh\left(\frac{y_{\omega}^{0}}{4}\right) \exp\left(-\frac{\kappa h}{2}\right) \right]$$
 (5)

$$y_{\omega}^{h} = 4 \tanh^{-1} \left[ \tanh\left(\frac{y_{\omega}^{0}}{4}\right) \exp(-\kappa h) \right]$$
(6)

The dimensionless surface potential of the isolated plate  $(y^0_{\infty})$  and the Debye length  $(\kappa)$  in the Eqns. (4)-(6) are given by:

$$y_{\omega}^{0} = 2\sinh^{-1}\left(\frac{zF\sigma^{0}}{2\varepsilon_{0}\varepsilon_{R}RT\kappa}\right)$$
(7)

$$\kappa = \sqrt{\frac{2cz^2 F^2}{\varepsilon_0 \varepsilon_R RT}} \tag{8}$$

where *F* is the Faraday's constant;  $\sigma^0$  is the surface charge density; *z* is valence of ions in the pore system and  $\varepsilon_0 \varepsilon_R$  is the dielectric constant. A good approximation and simplified expressions for the repulsive force for the case with low surface charges and not very small separation distance (e.g.,  $h\kappa > 2$ ) can be given by (Neretnieks et al., 2009)

$$\frac{\partial F_R}{\partial h} = 64\kappa cRT S_p \tanh^2(\frac{y_{\omega}^0}{4}) \exp(-\kappa h)$$
(9)

Figure 4 shows the calculated repulsive forces ( $F_R$ , Eq.3) and attractive force ( $F_{VdW}$ , Eq.2) as a function of particle separation distance for systems containing 1mM and 17 mM NaCl. The repulsive forces ( $F_R$ ) are larger than the attractive forces ( $F_{VdW}$ ) at small distances, but become weak faster as the distance between the particles increases. It is noted that  $F_R$  is very sensitive to the variation of water chemistry. Increasing ion concentration yields a substantial reduction of the repulsive forces ( $F_R$ ). The interaction between bentonite particles is critically important for the expansion behaviour, as well as for particles detachment at the clay/water interface. These points will be revisited in the following sections.



Fig.4 Variations of the repulsive force and attractive force with particle distance.

#### 2.2.3 Stability

The stability of bentonite is important when used as a buffer in geological disposal facilities for nuclear waste, as the generated colloid with high sorption capacity may assist the migration of radionuclides towards the biosphere (Missana and Adell, 2000). The bentonite stability is therefore a key factor in the assessment of the repository performance. Generally, the Critical Coagulation Concentration (CCC), defined as the minimum concentration of counterions required to induce coagulation, is used to assess the status of the colloid (Liu et al., 2009). Experiments on the stability of bentonite colloids, found in the literature, use different methods: rheological measurement (RM), adsorption method (AM), turbidity and optical density measurement (TOM), coagulation kinetic method (CKM), test tube series (TTS), and visual inspection (VI). Overall, the results show that the range of CCC for smectite in Na+ solutions is between 5 and 250 mM (Lagaly and Ziesmer, 2003; Frey and Lagaly, 1979; Chheda et al., 1992; Tombacz and Szekeres, 2004; Tombácz et al., 1989). A summary of the experimental observations of CCC for smectite in different solutions is provided in Fig.5. The results indicate that CCC is affected by the solid/liquid ratio in addition to the initial water and smectite compositions (Neretnieks et al., 2009). The measured CCC for different bentonite colloid is provided in Fig.6. The overall aggregation behaviour of bentonite colloids is similar in NaCl and CaCl<sub>2</sub> solutions. Based on experimental data, it has been proposed that CCC for different counterions is inversely proportional to the sixth power of its valence (Liu et al., 2009). The measured CCC in CaCl<sub>2</sub> (CCC<sub>Ca</sub>) and NaCl (CCC<sub>Na</sub>) ranges from 0.05 to 0.45 mM and 1 to 20 mM, respectively. This is in qualitative agreement with the theoretical ratio (CCC<sub>Ca</sub>/CCC<sub>Na</sub>=64) (Missana et al., 2018).



Fig.5 The CCC of Montmorillonite under sodium chloride and calcium chloride.Experimental data are from (a) Lagaly and Ziesmer (2003); (b) Frey and Lagaly (1979); (c)Chheda et al. (1992); (d) Tombacz and Szekeres (2004); and (e) Tombácz et al. (1989).



Fig.6 The measured CCC for different bentonite colloid in NaCl and CaCl<sub>2</sub> solution.

The prediction of CCC is typically obtained using the classical DLVO theory, which provides approximate values and is commonly employed to explain the coagulation of montmorillonite dispersions. Within DLVO theory framework, the total free energy ( $G_{TOL}$ ) between two plates is the difference between the van der Waals energy ( $G_{VdW}$ ) and electrostatic energy ( $G_{DDL}$ ) (Derjaguin, 1941). CCC is obtained when the total free energy, as well as its derivative with respect to the separation distance, vanish, i.e.,

$$G_{TOL} = 0 \tag{10}$$

And

$$\frac{\partial G_{TOL}}{\partial h} = 0 \tag{11}$$

The approximated expression given for CCC is as follow (Missana and Adell, 2000)

$$CCC = 8 \times 10^{-36} \frac{\left[ \tanh\left(\frac{2e\Psi_0}{4kT}\right) \right]^4}{A_H^2 z^6}$$
(12)

Where,  $A_H$  is the Hamaker constant, k is the Boltzmann constant, T is the absolute temperature, e is the electron charge, z is the electrolyte valence and  $\Psi_0$  is the surface potential. It should be note that the classical DLVO theory is unable to describe the pH-dependent stability of montmorillonite dispersions as it is inherently not capable of accounting for the ionizable surface groups. This issue may be addressed by using extended DLVO theory; however, the inclusion of stern-layer adsorption requires more information and increases the complexity of the system. Alternative way to assess the value of CCC can be obtained from a kinetic point of view by introducing a new key parameter referred to as the thermal deviation factor (Liu et al., 2009). The approach has delivered better predictions of CCC compared to the classical DLVO theory although the model still cannot account for the pH-dependent stability.

#### 2.3 Experimental observations

#### 2.3.1 Experimental observations of erosion

During the long-term operation of disposal facilities, the clay buffer/backfill can be eroded by the hydro-chemical interactions at the interface between the clay and the host rock, resulting in a gradual loss of the barrier and a co-transport of radionuclides into the biosphere. The erosion of swelling clays involves three main physical processes: (i) free swelling, representing the continuous transformations producing clay gel; (ii) clay particle detachment, representing the formation of clay sol; and (iii) migration of detached particles with the water flow (Sedighi et al., 2021). Long term erosion can create potential pathways for radionuclides leaving the waste form to migrate into the surrounding rock with potentially significant environmental and public health impacts. Experimental results reviewed in this section are categorized into three groups: free swelling/expansion, erosion in fracture systems, and piping erosion.

#### Free swelling/expansion

After absorbing water, the bentonite clay buffer can swell into the fractures, which leads to mass loss by erosion at the penetration front. The swelling/expansion behaviour of bentonite buffer is a critical element of the erosion assisted nuclide transport through EBS as the expansion of bentonite reduces the buffer density and degrades its performance, e.g., for colloid filtration, as a diffusion barrier, or as a load-bearing component (Tanai and Matsumoto, 2008). The swelling/expansion of bentonite buffer has therefore been extensively studied in recent years to improve the understanding of the phenomena to ensure the long-term performance of the geological disposal barrier (Tanai and Matsumoto, 2008; Dvinskikh et al., 2009; Liu, 2010; Schatz et al., 2013; Alonso et al., 2019; Li et al., 2020). Different experimental techniques have been developed for evaluating the swelling/expansion of bentonite buffer. For example, X-ray CT technique has been used to study the distribution of bentonite density during expansion process (Tanai and Matsumoto, 2008). Magnetic resonance imaging (MRI) has been used to investigate the transient expansion behaviour of MX-80 bentonite (Dvinskikh et al., 2009).

Most swelling/expansion experiments have been conducted with tube tests and artificial fracture systems (Tanai and Matsumoto, 2008; Dvinskikh et al., 2009; Liu, 2010; Schatz et al., 2013; Alonso et al., 2019; Li et al., 2020). These are summarised in Table 1. Further, the effects of water chemistry and fracture aperture on extrusion behaviour of different bentonites are shown in Fig. 7. The key findings of the existing experiment studies for bentonite buffer free swelling or extrusion into fracture systems are:

(i) Swelling behaviour of bentonite is strongly affected by the salinity of solution. This can be explained from a microscopic perspective. Decreasing concentration of solutions can enhance the DDL repulsive force, which may increase the swelling pressure and subsequently the measured swelling strain (Fig.7a). Interestingly, increasing concentration of solutions leads to increasing extruded mass, but decreasing extrusion distance (Table 1).

(ii) Generally, sodium-rich bentonite or pure sodium smectite (e.g. NaMt and Nanocor) have longer extrusion distance than natural bentonite (e.g. MX-80, Kunigel V1, Ibeco). Additionally, higher dry density of bentonite yields larger extrusion distance (Fig.7; Table 1).

(iii) The penetration of bentonite into fractures is largely affected by their aperture. Increasing fracture aperture leads to increasing extrusion distance. The experimental results indicate that the frictional forces acting at the fracture walls may play a key role in limiting the extrusion of bentonite into the fracture systems (Fig.7b).



(b)

Fig.7 Effects of (a) concentration of solutions and (b) fracture aperture on swelling behavior of different bentonite.

	Drv density	Fracture Wa	ater chemistry	Extrusion	Extruded	
Bentonite	$(g/cm^3)$	(mm) (N	aCl, mM)	distance (cm)	mass (g)	Ref.
Nanocor		0.1	1	1.43±0.12	0.3379	
Nanocor		0.2	1	2.48 + 0.12	0.7137	
Nanocor		0.4	1	2.26 + 0.17	0.9307	
MX-80		0.1	1	0.49 + 0.05	0.0205	
MX-80		0.2	1	0.82 + 0.08	0.0662	
MX-80		0.4	1	0.85 + 0.07	0.1700	
Nanocor		0.2	10	1.62		
Nanocor		0.2	100	1.05		
MCA-C clay		0.2	1	0.95		
Ibeco clay	14	0.2	1	1.05		Alonso et
Nanocor	1.1	0.1	1	1.35+0.2	0.0205	al. (2019)
Nanocor		Rough (1.0)	1	2.26+0.1	1.2509	
Nanocor		Rough (1.7)	1	1.6+0.2	0.9700	
MX-80		0.2	1	1.14 + 0.06	0.0810	
MX-80		Rough (1.0)	1	1.56 + 0.12	0.6929	
MX-80		Rough (1.7)	1	$1.48 \pm 0.12$	0.4062	
MX-80		0.2	1	1.1	0.0711	
Salt-free MX-80		0.2	1	-	0.298	
Nanocor		1.7	1	3.8	1.13	
MX-80		1.7	1	3.4	1.42	
	1.2		DI	-		
			0.1	2.43	-	
			0.5	1.27	-	
			1	1.06	-	
	1.2		2	1.00	-	
			0.1 (CaCl <sub>2</sub> )	1.69	-	
			0.5 (CaCl <sub>2</sub> )	1.26	-	
Commercial			$1 (CaCl_2)$	1.07	-	Li et al.
bentonite		Tube test	2 (CaCl <sub>2</sub> )	0.99	-	(2020)
			0.1	1.86	-	
			0.5	1.06	-	
			1	0.92	-	
	1.0		$\frac{2}{1}$	0.79	-	
			$0.1 (CaCl_2)$	1.35	-	
			$0.5 (CaCl_2)$	1.02	-	
			$1 (CaCl_2)$	0.75	-	
	1.0	0.5	$2(CaCI_2)$	0.38	-	
Kunigel V1	1.0	0.5		1.0 (at 9001) 2.2 (at 000h)	-	
	1.0	1		5.5 (at 9001) 4.5 (at 900b)	-	
	1.0	1.5		4.3 (at 9001) 0.2 (at 900h)	-	
Kunigal V1	1.0	0.5	וח	1.2 (at 900h)	-	Tanai 2008
Kuniger v I	1.0	1	DI	1.2 (at 9001) 1.7 (at 900h)	-	1 aliai 2008
	1.0	0		1.7 (at 9001) 0.1 (at 900h)	-	
30%)	1.0	0. 1		0.1 (at 900h)	-	
	1.0	1		1.0 (at 900h)	-	
	1.0	1.5	0.5 (CaCla)	1.0 (at 90011)	-	
WyNa	1.8	Tube Test	$2 (CaCl_2)$	4.5		Liu (2010)
			2 (CaCI <sub>2</sub> )	т		Reid
Nanocor	1.4	0.1	DI	3.2		(2016)
NT - N <i>T</i> -	1.6	1	DI	6 (at 700h)	1.28	<b>C</b> 1
NaMt	1.6	1	171	2.7	2.04	Schatz et
	1.6	1	17	4.6	3.8	al. (2013)

Table.1 Swelling/expansion of bentonite clay under different water chemistry, dry density, fracture openings and bentonite material
## **Erosion in fracture system (interface erosion)**

Erosion of bentonite buffer due to glacial meltwater intrusion is considered as a potential threat to the integrity of EBS by Posiva and SKB (Smith et al., 2017). For example, a small-scale lab test of bentonite erosion in fractures showed that clay extensively swelled into the fractures and clay mass was found in the effluent solutions (Baik et al. 2007, Schatz et al. 2013, Reid et al. 2015, Hedström et al. 2015, Schatz et al. 2016). Experimental studies of bentonite erosion have demonstrated that the erosion processes are highly affected by both solution chemistry and fluid velocity (Schatz et al., 2013; Sane et al., 2013; Reid et al., 2015; Navarro et al., 2016; Smith et al., 2017; Missana et al., 2018; Alonso et al., 2018; Bouby et al., 2020; Zhang et al., 2019; Bian et al., 2019, Xiang et al., 2020). The experiments conducted by Baik et al. (2007) showed that flowrate of groundwater induced a significant impact on the total eluted volume of bentonite particles in the case studies. The experimental pinhole tests conducted by Sane et al. (2013) showed that the erodibility coefficients of MX-80 bentonite at high salinity (e.g., 70 g/L) were two orders of magnitude larger than those at low salinity (e.g. 10g/L). No erosion was observed for sodium montmorillonite for solution compositions from 0.5 g/L to 10 g/L NaCl (Schatz et al., 2013). The erosion rates reported for low flowrate (0.1 ml/min) were more than an order of magnitude lower than those for high flowrate (2.84 ml/min) (Schatz et al., 2013; Reid et al., 2015). It should be noted that only a limited number of small-scale experimental tests have been conducted and the effects of up-scaling to the filed scale are still unclear (Sane et al., 2013). However, a small-scale lab test of bentonite erosion is vital for developing an understanding of the processes as it allows to as it allows to study of the individual parameters (e.g., chemistry of groundwater and flowrate etc.) associated with this complex process (Sane et al., 2013; Suzuki et al., 2013; Reid et al., 2015). The research to fully understand and characterize the erosion process in bentonite is still work-in-progress and further studies, including experimental (e.g., small-scale experiments and mock-up test) and theoretical studies on the different sub-processes (e.g., wetting, expansion etc.), are needed (Sane et al., 2013; Schatz et al., 2013;).

The results of erosion experiments are summarised in Table.2, where the data is obtained from Schatz et al. (2013), Schatz and Akhanoba (2017), ClayTechnology from (BELBaR deliverable 2.12), Schatz et al. (2016), Reid et al. (2015), Hedström et al. (2015) and Alonso et al. (2018). It should be noted that approximations (e.g., averaging estimations) are made when summarizing the experimental results as the data presented in these papers follows different

formats. Additionally, the approaches used to determine the loss of smectite are different as well, which may cause large differences in the measured rates of eroded mass. For example, eluted water samples were analysed by Photon Correlation Spectrometry (PCS) to determine the eluted mass (Schatz et al.,2013; Alonso et al., 2018), while Hedström et al. (2015) used the turbidity of the effluent, obtained by a portable turbid meter (TN-100, Eutech Instruments) to evaluate the amount of eroded montmorillonite. It should be noted that the thermal effect or change in temperature due to geochemical reactions is included in Table 2 due to lack of experimental data.

The erosion rate as a function of water velocity is shown in Fig.8. This figure covers all the erosion experiments. The erosion rate generally increases with increasing water velocity. However, the erosion rate differs significantly between different bentonites and fractures. The largest erosion rate is around  $10^{-6}$  g/s for pure sodium bentonite (NaMt) (Schatz et al.,2013), which may cause 400 kg/m<sup>2</sup>/yr loss of bentonite. Erosion behaviour of different bentonite as a function of water chemistry is shown in Fig.9. As the salinity increases, the erosion generally decreases. The trend may not be obvious as water flowrate is different for different cases. For the case with the same material and fracture thickness, increasing water chemistry can effectively reduce the erosion rate. For example, increasing water chemistry from DI to 4.3 mM NaCl for NaMt leads to the erosion rate decreasing from  $2.6 \times 10^{-6}$  to  $1.0 \times 10^{-6}$  g/s (Schatz et al., 2013). There is no erosion observed if water chemistry is larger than CCC (around 10mM for sodium bentonite). Most of bentonite have a range of erosion rate between  $10^{-8}$  and  $10^{-6}$  g/s under different water chemistry, flow velocity and fracture thickness.

Table.2 Extrusion behaviour and erosion rate of bentonite buffer under different water velocity, solution and bentonite material (Schatz et al., 2013; Schatz and Akhanoba, 2017; Reid et al., 2015; Hedström et al., 2016 and Alonso et al., 2018)

Clay So	olution NaCl, mM)	Fracture aperture (mm)	Flow velocity (m/s)	Average radial expansion (cm)	Eluted mass(g)	Eroded mass (%)	Mass loss rate (g/s)
Nanocor	1mM	0.1	1.5×10 <sup>-6</sup>	1.43±0.12	6.2×10 <sup>-4</sup>	8.5	$1.3 \times 10^{-7}$
Nanocor	1mM	0.2	6.1×10 <sup>-7</sup>	2.48±0.12	6×10 <sup>-3</sup>	18.0	$2.8 \times 10^{-7}$
Nanocor	1mM	0.4	$1.4 \times 10^{-7}$	2.26±0.17	9.9×10 <sup>-3</sup>	23.5	$3.6 \times 10^{-7}$
Nanocor	1mM	0.2	$1.35 \times 10^{-6}$	1.35±0.20	2×10 <sup>-4</sup>	2.0	$7.9 \times 10^{-9}$
Nanocor	1mM	Rough (1)	$5.6 \times 10^{-8}$	2.26±0.10	4.5×10 <sup>-4</sup>	31.6	$4.8 \times 10^{-7}$
Nanocor	1mM	Rough (1.7)	$6.3 \times 10^{-8}$	1.60±0.20	1.5×10 <sup>-3</sup>	24.5	$3.7 \times 10^{-7}$
MX-80	1mM	0.1	$1.35 \times 10^{-8}$	$0.49 \pm 0.05$	2×10 <sup>-4</sup>	0.5	$7.9  imes 10^{-8}$
MX-80	1mM	0.2	$6.7 \times 10^{-7}$	$0.82 \pm 0.08$	5×10 <sup>-4</sup>	1.7	$2.6 \times 10^{-8}$
MX-80	1mM	0.4	$3.6 \times 10^{-7}$	$0.85 \pm 0.07$	5×10-3	4.3	$6.6 \times 10^{-8}$
MX-80	1mM	0.4	$5.53 \times 10^{-7}$	1.14±0.05	1.3×10 <sup>-4</sup>	2.0	$3.1 \times 10^{-8}$
MX-80	1mM	Rough (1)	$9.8 \times 10^{-8}$	1.56±0.8	3.8×10 <sup>-4</sup>	17.4	$2.7 \times 10^{-7}$
MX-80	1mM	Rough (1.7)	$5.92 \times 10^{-8}$	1.48±0.12	1.2×10 <sup>-3</sup>	10.2	$1.57 \times 10^{-7}$
NaMt	171	1	$2.0 \times 10^{-4}$	2.75	-	<2%	-
NaMt	17	1	$1.9 \times 10^{-4}$	4.5	-	0	-
NaMt	DI	1	$2.0 \times 10^{-4}$	2.4	-	47%	$2.6 \times 10^{-6}$
NaMt	Gradient elution	1	$1.9 \times 10^{-4}$	5.3-6.2- 7.1-1.0	-	65%	$1.6 \times 10^{-6}$
NaMt	DI	1	$2.6 \times 10^{-5}$	2.3	-	34%	-
NaMt	GW	1	$6.2 \times 10^{-6}$	3.1	-	24%	$9.9 \times 10^{-7}$
50/50 Ca/NaMt	Gradient elution	1	$1.9 \times 10^{-4}$	3.9-4.5- 5.6-0.7	-	64%	$2.9 \times 10^{-7}$
50/50 Ca/NaMt	4	1	$2.0 \times 10^{-4}$	4.4	-	9%	-
50/50 Ca/NaMt	DI	1	3.3×10 <sup>-5</sup>	1.8	-	36%	$1.5 \times 10^{-6}$
50/50 Ca/NaMt	$2 \times GW$	1	$5.6 \times 10^{-6}$	4.5	-	15%	$3.7 \times 10^{-7}$
NaMt	DI	1	0	6.0	-	-	-
NaMt	2.1	1	$1.8 \times 10^{-4}$	-	-	-	$1.3 \times 10^{-6}$
NaMt	4.3	1	$1.9 \times 10^{-4}$	-	-	-	$1.0 \times 10^{-6}$
MX80	DI	Rough (0.057)	1 (ml/min)	2.15	-	24%	$1.0 \times 10^{-7}$
MX80	DI	0.1	1 (ml/min)	2.3	-	-	$6.3 \times 10^{-8}$ to $2 \times 10^{-8}$
Nanocor Nanocor	DI DI	0.1 0.1	0 (ml/min) 0.38 (ml/min	3.2 ) 2.65	-	-	-
MX80	Sea water	Rough (0.068)	1 (ml/min)	0.4	-	-	-

NaMt and CaMt represent sodium or calcium montmorillonite, respectively; Gradient elution tests: the TDS of the eluent was reduced after periods of 30 days from 0.5 to 0.25 to 0.125 g/L NaCl to DI; GW represents a Grimsel groundwater; Sea water: 299mM NaCl and 158mM CaCl<sub>2</sub>



Fig.8 Erosion rate as a function of water velocity.



Fig. 9 Erosion behavior of different bentonite as a function of water chemistry.

## **Piping erosion**

Several recent experimental investigations present evidence of potential formation of localised micro-pathways/cracks in the clay buffer/backfill, known as piping (Sanden et al., 2008; Aberg, 2009; Sane et al., 2013; Suzuki et al., 2013; Abe et al., 2017; Jo et al., 2019). These new observations are important as they indicate potential loss of a critical component of the barrier system as the result of water percolation at the rock-clay interface (through micro-fractures or wormholes formations), which can subsequently enhance the escape of highly toxic radionuclides. Piping erosion could be especially important in the early phase of the saturation process if water flows into a deposition hole from fractures in the bedrock, causing sub-vertical mass transport into the deposition tunnel backfill (Navarro et al., 2016). SKB has identified piping and colloid generation in the buffer and backfill as processes that could have significant effects on the practicality of EBS emplacement and on the long-term performance of the disposal system. For example, the piping phenomena may affect requirements for rock characterization, water inflow and water management during the installation phase, and may also affect buffer material properties and buffer installation methodology.

Recent experimental investigations mainly focus on exploring: i) the conditions of piping formation; ii) the evolution of piping channel; iii) the effects of inflow rate; and iv) the effect of piping on buffer properties and eroded mass (Suzuki et al., 2013). The experimental results (Suzuki et al., 2013; Abe et al., 2016) indicate that the preferred conditions for piping formation are flow rates above 0.001 l/min and the presence of a gap between buffer and acrylic, e.g. bentonite block or pellets. Additionally, the presence of non-reactive minerals, e.g., sand, in the bentonite block can assist sedimentation on the surface and protect the sealing function of bentonite (Suzuki et al., 2013), which may play an important role in preventing piping in the long-term assessment. The effects of water salinity on the piping formation are not well understood although experiments have shown that the swelling performance of the bentonite buffer decreases due to water salinity and Ca type conversion (Abe et al., 2016). The effects of the inflow rates, water salinity and test length on water pressure, piping and erosion rate in the buffer material are summarised in Table.3. The eroded mass of bentonite buffer induced by piping and erosion, obtained by Sanden et al. (2008), Suzuki et al. (2013) and Jo et al. (2019), is shown in Fig.10. Generally, the piping/erosion volume of the buffer material increases with increasing total drainage volume except for the case with 0.5M NaCl bentonite pellet (orange circle in Fig.10), where the self-sealing or swelling of bentonite filled the piping channel and

no erosion is observed after 30 L. The eroded mass of bentonite buffer induced by piping assisted erosion process in the field can reach several kilograms (Sanden et al., 2008), which is expected to reduce the performance of bentonite as EBS.

Test No.	Type of water liquid (g/l)	Flow rates (ml/min)	Test lengt <i>h</i> (m)	Maximum Water pressure (kPa)	Subsequent changes in the water pressure(kPa)	Piping and erosion	Erosio rate (g/h)	n Ref.
1		0		7	7	×	-	
2		0.05		13	12→15	$\checkmark$	-	
3		0.1		20	18→19	✓	-	
4		0.2		30	25→29	✓	-	
					Gradually			
5	8.5	0.3	0.1	42	decrease to 22	✓	-	Jo et al.
6		0.4		27	6	$\checkmark$	-	(2019)
7		0.5		7	6	$\checkmark$	-	
8		0.65		10	5	✓	-	
9		0.8		8	5	$\checkmark$	-	
10		1.1		12	Gradually decrease to 3	√	-	
11	29.3	0.1	0.05		60	$\checkmark$		
12		0.1-0.001	0.05		25-38	√		
13		0.1	0.05		2.5	√		
14		0.1	0.05		2.5	<b>√</b>		
15		0.1	0.05		1-5	<b>√</b>		
16		0.01	0.05		2.5	<b>√</b>		
17	DI	0.005	0.05		2.5	✓		
18		0.001	0.05	3	3000	×		Suzuki
19		0.1	0.05		7.5	<b>√</b>		et al.
20		0.1	0.05		7.5-15	✓		(2013)
21		0.1- 0.0001	1		25-300	√		
22		0.1- 0.0001	1		17.5-45	√		
23	29.3	0.1	1		12.5-20	$\checkmark$		
24	DI	0.001- 0.05	1	0.2-0.5-1-2- 2.5-3	200-3000	×		
25	Tap water	0.01	1	18	10	✓	2.5	
26		0.01	0.2	8	8	$\checkmark$	4.3	
27		0.001	0.5	650	0-650	$\checkmark$	1.1	Sande
28	10	0.01	1	60	60-10	$\checkmark$	10.2	n et al.
29	10	0.1	1	20	8	√	65	(2008)
30		1	1	80	-	√	65.1	
31		0.01	2.5	45	8	√	0.45	
32	35	0.01	1	115	8-115-8	$\checkmark$	59	

Table 3. The effects of the inflow rates, water salinity and test length on water pressure,piping and erosion rate in the buffer material



Fig. 10 Mass of eroded bentonite as a function of amount of water outflow.

The experimental results provide some insights into the piping formation process. Generally, the process depends on groundwater salinity, flow rate, pressure head, bentonite composition, gap and dry density. Theoretically, the formation of a piping channel in the swelling bentonite buffer requires a large hydrostatic head, which can initiate channels and keep them open against the swelling pressure of buffer material. Current understanding is that the pressure head needed to maintain a channel is orders of magnitude smaller than the swelling pressure of the clay (Sane et al., 2013). Fig.11 shows the evolution of flow in, flow out and water pressure with time. The corresponding hydration and piping channel formation processes are shown in Fig.12. The initial water pressure is small and produces a constant flow rate, e.g., 0.001 l/min. However, after 32h a sharp increase in water pressure with no change of the inflow rate is observed. Specifically, the water pressure starts increasing at 32h to reach a peak of 600 kPa at 56h and then decreasing to a very small value. The increase of water pressure after 32h may be induced by self-sealing or swelling of bentonite pellets during the hydration process. The swelling of bentonite pellets can fill the gaps between them and reduce the pathways for water flow. A larger water pressure is therefore required to keep the constant water flowrate. However, when water pressure is larger than the hydraulic resistance of interfaces, piping occurs and creates a preferential flow path for water transport, which leads to a sudden decrease of water pressure. If the water pressure is lower than that of hydraulic resistance, no piping is expected and the gaps are completed sealed.



Fig. 11 The inflow, water pressure and the measured outflow of the sample (Sanden et al. 2008).



Fig.12 (a) the hydration process of bentonite pellet after 3 hours during water filling; and (b) the final stage of erosion test where the channel is situated on the top of the sample (Sanden et al. 2008).

# 2.3.2 Clay desiccation

The understanding of desiccation cracking in clay soils, and its effect on soil behaviour, has substantially progressed in the last few decades through experimental investigations. Desiccation has been experimentally investigated in slurries (Li and Zhang, 2011), clay soils (Tang et al., 2011; Wei et al., 2016) and starch water mixture cakes (Kodikara et al., 2004). Desiccation cracking experiments have mainly been focused on characterising the cracks'

geometries including depth, thickness, spacing and aperture of the cracks (Najm et al., 2009; Peron et al., 2009; Wan et al., 2018; Amarisiri et al., 2014; Zhao et al.,2020; Xu et al., 2020). The results of these studies have contributed to the understanding of soil cracking processes, but their relevance is limited to the specific laboratory or field conditions. The existing experimental results from laboratory and field tests are summarised in Table 4. These results provided qualitative and quantitative description of the dynamic cracking process and the evolving crack pattern (Tang et al., 2021).

Experimental studies of soil desiccation are usually performed by using environmental chambers to control environmental variables, e.g., humidity and temperature (Amarasiri et al., 2011; Tran et al., 2019). The analysis of desiccation cracking patterns formation process requires monitoring and characterization tools, e.g., camera, laser scan, CT scan and SEM (Tang et al., 2021). The key factors affecting the desiccation behaviour of clay are the material properties, the boundary conditions, and the surrounding environment. The existing experimental tests for clay desiccation can be categorized into three major groups - restrained ring test, long bar test, and thin clay layer test – shown in Fig.13. The restrained ring test is used as a method to determine the tensile strength of desiccating clay soils by isolating the initiation of a single crack (Amarisiri et al., 2014; Shannon et al., 2015). Thin clay layer test is used to identify soil mass area, shrinkage ratio, crack ratio, average crack width, total crack length, number of crack segments, and critical stress intensity factor, i.e. fracture toughness (Konrad et al., 1997; Mal et al., 2008; Tang et al., 2008, 2011 & 2019; Trabelsi et al., 2012; Sanchez et al., 2013; Julina and Thyagaraj, 2019). The long bar tests are used to study the initiation of cracks, final crack patterns, crack spacings and numbers, apparent electrical resistivity (Peron et al., 2008, 2009 & 2013; Tang et al., 2008, 2011 & 2019; Li and Zhang, 2010; Costa et al., 2018; Lecocq et al., 2002).

Test	Geometry (mm) Radius ®, Height (H), Length (L), Width (W)	Characterization parameter	References	
D' T (	$66.5~(R_{out})\times 20~(R_{int})$	Determining fracture properties during desiccation The initiation of a single crack	Amarisiri et al. (2014)	
King Test	$53 (R_{out}) \times 21 (R_{int})$ 220 (R <sub>out</sub> ) × 150 (R <sub>int</sub> )	Measuring the stresses at failure Geometry effects on cracking patterns	Najm et al. (2009)	
	$295 \times 49 \times 12$ $295 \times 15 \times 15$	Initiation of cracks, final patterns, crack spacing, crack numbers	Peron et al. (2008, 2009, 2013)	
Long bar Test L × W ×H	$290 \times 30 \times 20$	Apparent electrical resistivity Quantitative analysis of soil properties and crack morphology	Tang et al. (2018)	
	$400 \times 7 \times 5$	Cracks pattern, crack opening, nucleation time and position	Lecocq et al. (2002)	
	$250 \times 25 \times 18$ $200 \times 9 \times 9$	Crack spacing, crack depth, cracking patterns Crack polygons density, crack opening and orientation	Costa et al. (2018) Li and Zhang (2010)	
	117 × (2.5–5)	Soil mass area, shrinkage ratio, crack ratio, average crack width, total crack length, number of crack segment	Tang et al. (2008; 2011; 2019)	
Thin clay layer test R ×H	100 × 5	Crack intensity factor, crack opening	Trabelsi et al. (2012)	
	97 × 13	volume change, water loss and evolving crack network's morphology	Sanchez et al. (2013)	
	$75.4 \times 20$	Volume change of cracks, settlement, crack patterns	Julina and Thyagaraj (2019)	
	120 × 5	Cracks forming in a radial external electric field	Mal et al. (2008)	
Field test	15m depth	Time domain reflectometry probes Large desiccation polygons	Konrad et al. (1997)	
	820 (L)× 530 (W)	crack network, crack porosity, crack aperture, and crack density	Li and Zhang (2011)	

Table 4. Experimental observation of soil cracking behaviour



Fig.13 Typical experimental results of soil desiccation for restrained ring, thin clay layer and long bar tests.

## 2.4. Mechanisms and model developments of erosion and desiccation

#### 2.4.1 Clay erosion

Despite recent developments in understanding of the underlying processes, mainly through extensive experimental studies in last 10 years, predictive modelling of erosion and piping of bentonite is very limited. Most of the existing numerical models for the erosion processes use semi-empirical or phenomenological approaches describing mostly individual processes, e.g. clay gel generation (Pusch, 1999), mechanical erosion caused by the water flow (Grindrod et al., 1999), erosion of particles and dispersion-flocculation behaviour of clay particles (Kurosawa et al., 1999), expansion of the clay solid (Alonso et al., 1990, Navarro et al., 2017), colloid generation (Degueldre et al., 2009) and swelling of bentonite (Neretnieks et.al. 2009; Liu et al., 2009). The theoretical calculations for erosion can be categorized into two groups: modelling swelling/expansion behaviour; and modelling of coupled swelling and erosion.

# Swelling/expansion

Free swelling process of clay represents the continuous transformations from solid to clay gel due to the continuous water uptake. The swelling/expansion of highly compacted clay is induced by the large repulsive forces exerting among the negative particles when wetted by the fluid, which leads to the clay buffer swelling into or filling out the fracture in the surrounding environment. In free swelling process, the main forces acting on the particles include gravitation, buoyancy, diffusion, electrical double layer repulsive forces (DDL) and Van der Waals (VdW) attractive forces (Neretnieks, 2009). The study of swelling capacity and swelling rate of bentonite clay is critical for a wide range of geotechnical engineering projects since the swelling process is thought as "self-healing" capability of bentonite. Swelling is therefore critical part for investigating erosion behaviour of clay buffer/backfill as the swelling front is the location where mass lost is induced by the erosion.

From macroscopic (continuum) perspective, the swelling of bentonite clay by water can be described as a series of phase changes: an initially solid phase is hydrated, leading to a continuous transformation into a swelling paste, which in turn is continuously transformed into a gel, and eventually discontinuously transformed into dispersed colloidal particles (sol) in the water. Figure 14 provides a schematic of the swelling process of bentonite clay and its corresponding processes. The free swelling step is characterised by continuous reduction of the clay density. The free swelling process of bentonite is largely dependent on the properties of the material, e.g., the exchangeable cations, dry density, and mineral compositions, on the fracture apertures and on the water chemistry (Birgersson et al. 2009). Additionally, the swelling capacity of a bentonite with dominating monovalent cations, e.g., Na+, K+, is larger than a bentonite with dominating multivalent cations, e.g., Ca2+, Mg2+ (Hedström et al. 2011). How these key factors affect the extrusion behaviour of bentonite is extensively studied in recent years via experimental or numerical studies (Schatz et al., 2013; Sane et al., 2013; Reid et al., 2015; Navarro et al., 2016; Smith et al., 2017; Missana et al., 2018; Alonso et al., 2018; Bouby et al., 2020; Zhang et al., 2019; Bian et al., 2019, Xiang et al., 2020).



Fig.14 Swelling behavior of bentonite into fracture system

Modelling deformation/swelling of compacted bentonite is commonly developed in the framework of thermo-hydro-chemo-mechanical (THCM) coupling, which incorporates mechanical stress evolution with water migration, chemical transport, and heat transfer (Sane et al. 2013). Recent models include those by Sánchez et al. (2014), Morena et al. (2018),

Navarro et al. (2014 & 2021), and Nasir et al. (2017). However, the existing THCM models are based on the small strain theory and may not be applicable to cases where the bentonite deformations are large, especially for the case with free swelling. Efforts have been made in recent years for modelling free swelling of bentonite by establishing a direct connection between the micro- and macro-structures (dual porosity model) in THCM framework (Sánchez et al., 2014; Navarro et al., 2014 & 2021). Formulations based on well-established elastoplastic theory using experimentally calibrated parameters have shown good agreement with several free swelling tests.

Based on the experimental observations of bentonite clay free swelling, Kanno et al. (2001) modelled the bentonite extrusion behaviour as a solid particle diffusion process, in which the diffusion coefficient was obtained by fitting the experimental data. Borrelli et al. (2008 & 2011) extended the model by incorporating tip movement process while physical driving forces for the swelling process were not explained. Many recent numerical studies of chemical erosion were developed based on DLVO electro-static forces theory, which was first presented by Liu et al. (2009) and Neretnieks et al. (2009) (called KTH model). KTH model was developed based on dynamic force balance among bentonite colloid particles, which showed the capacity to capture the free swelling process of sodium-bentonite under different water chemistry. KTH model was successfully applied to capture the expansion/extrusion behaviour of bentonite when its density was low and saturated. The free expansion/swelling formulation of the KTH model can be represented by a diffusion-like equation. Although KTH model cannot describe the entire swelling process from an initial unsaturated to well saturated bentonite, the effective diffusivity-based models are significantly simpler than THMC models. Many recent studies of swelling behaviour of bentonite were developed based on KTH model (Liu 2010; Li et al., 2021; Liu et al., 2021; Chiou et al., 2021). An alternative approach for chemical erosion presented by Birgersson et al. (2009) was developed based on a balance between the swelling pressure of the buffer and the friction force exerting on the bentonite colloid. A preliminary analysis using the model indicated that bentonite would penetrate a maximum distance of only a few mm into a fracture, and that the corresponding erosion rate would be independent of fracture aperture (Birgersson et al., 2009).

As a summary, the models for bentonite swelling proposed in the literature are expected to be valid either at the solid end of free swelling (THM models) or at the wet end (effective diffusivity models), but no model adequately covers the entire processes of swelling and

#### wetting (Sane et al., 2013).

#### **Coupled swelling and erosion model**

Theoretical studies of clay erosion are very limited due to the computational complexity of the problem, including phase changes and detachment of particles at clay/water interface. Liu and Neretnieks (2006) presented the theoretical models to quantitatively address the influence of water chemistry on colloid generation rate and erosion process. The dynamic swelling, the viscosity and water flow model was first coupled together to study the mass loss rate of bentonite due to gel expansion and particle releases at the clay/water interface (Neretnieks et al. 2009; Moreno et al. 2011; Schatz et al., 2013). Particle release and transport was modelled as advection-diffusion process. Fig.15a shows a good agreement between the numerical simulation results and experimental data for bentonite solid content distribution. However, the theoretical calculation of mass erosion rates by Moreno et al. (2011) was significantly lower by a factor of 3 to 10 (Fig. 15b), because only smectite particle diffusion processes were considered in their model and the detachment processes were not included. The erosion model presented by Moreno et al. (2011) has been later revisited by Neretnieks et al. (2017) by introducing a rim interface to account for the release mechanism of clay particles. An experimental fitting parameter of  $\phi_{Rim}$  at the water/clay interface has been introduced to account for particle detachment processes at clay/water interface. This has led to closer correlation with experimental data. However, since their model relies on fitting to experimental data, it can be used only when the data for specific conditions is available (Schatz et al., 2013).

Additionally, unsaturated soil mechanic models built on the Barcelona Expansive Model (BEM) was applied by Navarro et al. (2016) to model the coupled processes of deformation (e.g. swelling) and mechanical erosion of compacted MX-80 bentonite. The loss of mass at the clay/water interface was described by the experiment fitted erosion rate. An alternative novel method based on a particle-based method using CFD-DEM (computational fluid dynamics and discrete element models) was recently presented by Laviña et al. (2018). The model was still immature and not able to quantify the particle release process due to the excessive computational cost.



Fig. 15 Typical numerical simulation results compared to experimental data (a) bentonite solid content and (b) eroded mass.

However, critical missing element in the existing models is related to the detachment of particles at the solid/fluid interface. Fig.16 shows the erosion test results for bentonite in an artificial fracture. The sharp boundaries between inner (continuous solid phase) zones and outer (discontinuous particle detachment) zones are clearly observed (Schatz et al., 2013). Additionally, the aggregation and sedimentation of eroded bentonite particles increase with time in the outer zones. The experimental observations indicate that erosion mechanisms involve mass transfer of bentonite particle at solid/liquid interface (boundaries between inner and outer zones) due to the flowing water. The generation of mass loss is expected to be initiated if shear forces induced by flowing water are higher than the cohesive forces in the bentonite colloid when wetted/swollen bentonite clay has penetrated some distance into the fracture mouth (Sane et al., 2013). Detachment of clay particles at water/clay interface is required to link dynamically the solid swelling and particles release processes at the solid/liquid interface. A general approach is to compare the shear forces at the solid/liquid interface induced by water flow with a material parameter limiting the solid behaviour of the gel, typically called the yield stress (Laxton and Berg, 2006, Sane et al., 2013). At shear stresses lower than the yield stress the gel behaves like a solid. At shear stresses above the yield stress, the clay particles exhibit sol behaviour and can be transported away by seeping water. This concept agrees with experimental observations (Eriksson and Schatz, 2015). The interface erosion mechanisms are adopted to develop comprehensive erosion models in Chapter 5.



Fig.16 Erosion behaviour of bentonite in an artificial fracture test (a) the complete inner and outer zones and (b) the sharp boundary and particle detachment at the interface of inner and outer zones (Eriksson and Schatz, 2015)

# 2.4.2 Clay desiccation

Understanding the desiccation-induced cracking in soil has been improved over the last 20-30 years through experimental studies and theoretical models. Clay desiccation is triggered by the loss of moisture which can subsequently cause shrinkage in clays with potential cracking. Desiccation process is complicated due to the multi-physics nature of the processes involved: water evaporation; volumetric shrinkage; emergence, and propagation of cracks. Three distinct stages can be identified during soil desiccation (Peron et al., 2009; Tang et al., 2011). Stage I is characterised by a significant shrinkage, but the soil remains saturated over a certain range of moisture content. In Stage II air gradually replaces the moisture in the soil pore system, but the shrinkage is substantially smaller than in Stage I. Stage III is characterised by continued loss of moisture, but without further shrinkage.

The existing knowledge from experimental and theoretical studies showed that desiccation cracks are the results of the accumulation of internal stresses exceeding a critical strength (Tang et al., 2021). Under drying, water continuously evaporates, resulting in increasing suction and shrinkage of soil skeleton. The failure of soil is triggered when the tensile stress in certain regions becomes larger than the tensile strength of the soil (Klar and Linker, 2010; Tang et al., 2010). Cracks are reported to initiate at weak points, e.g. micro-defects or air-filled micro pores, on the clay surface (Tang et al., 2011). These weak points show different volumetric deformation compared to their neighbours leading to significant local stress concentrations (Nahlawi and Kodikara, 2006). The propagation of cracks is complex and influenced by many factors, including boundary conditions, initial water content, evaporation rate, density, salinity,

compositions of soil and temperature (Tang et al., 2021).

The challenges for modelling are the multi-physics nature of the processes involved and the mathematical description of problems that include emerging and propagating strong discontinuities in a material with complex, heterogeneous structure. Predicting the desiccation cracking under in situ/field conditions requires physically realistic models that can capture the complex and coupled processes involved. Many numerical methods and theoretical frameworks are developed in recent decades for the prediction of the clay desiccation behaviour, including initiation and propagation of cracks and their thickness and final patterns (Amarasiri and Kodikara, 2013; Sima et al., 2014; Sánchez et al., 2014; Zhang and Jivkov, 2016; Morrison et al., 2016; Vo et al., 2017; Dassios et al., 2018; Cajuhi et al., 2018; Tran et al., 2019 & 2020; Wang et al., 2020; Zhu et al., 2020). Existing numerical studies can be categorized into three major groups: mesh-based; mesh-free; and hybrid approaches (Tang et al., 2021). These numerical methods are summarised in Table 5 and typical simulation results are shown in Fig.17.

It is noted that numerical methods for solving the continuum physical or mechanical problems, such as finite element method (FEM), cannot represent the crack initiation without the introduction of special elements. These are typically cohesive elements inserted at the interfaces between the standard continuum elements, which require specific remeshing techniques at the locations of possible crack initiation (Wang et al., 2020). The propagation of existing cracks within an FEM framework is possible by different enhancements, such as the extended FEM (Vo et al., 2017) and mesh fragmentation technique (Sánchez et al., 2014). Modelling crack propagation also requires specific calibration of a crack extension criterion, and as for initiation the calibration is expensive and typically not universally valid. For example, calibration with experimental data for one specific stress state may not correctly predict the behaviour for other stress states (Zhang and Jivkov, 2016). Some of the difficulties of FEMbased methods can be avoided by discrete methods, such as lattice methods (Zhang and Jivkov, 2016; Morrison et al., 2016; Dassios et al., 2018), lattice spring methods (Zhu et al., 2020) and discrete element methods (Amarasiri and Kodikara, 2013; Sima et al., 2014; Tran et al., 2020). Discrete particle-based approaches show the capacity to capture the desiccation pattern formation, e.g. hierarchical structure. It however remains challenging for the approach to capture the multi-physical processes involved in desiccation process induced by moisture evaporation and heat and mass exchange. Alternative approaches, including phase-field

methods (Cajuhi et al., 2018) and smoothed-particle hydrodynamics (Tran et al., 2019), have also been adopted for soil desiccation problems. However, these non-FEM-based methods inherit a significant requirement from FEM – introduction and calibration of failure initiation and propagation criteria – which is dictated by the local (differential) nature of the underlying governing equations (Wang et al., 2019).



Fig. 17 Numerical method used for desiccation cracking modelling and its typical results.

	Numerical method	Model results/crack criterion	References
Mesh- based approach	FDM	Cracking was analysed by both stress relief and fracture energy release approach.	Costa et al. (2018)
	FEM/ XFEM	Analysis of deformation of the material under drying with pre-crack inside Tensile failure	Peron et al. (2007) Lee et al. (1988)
	MFT	Purely mechanical model Cracking was analysed by both fracture energy and tensile strength.	Sanchez et al. (2014)
	CEM	Hydro-mechanical model Damage-elastic cohesive fracture law	Vo et al. (2017)
	PFM	Poro-mechanical coupled with a phase-field model. Energy-minimization-based failure law	Cajuhi et al. (2018);
Mesh-free approach	DEM	Crack initiates when the contact traction or shear force exceeds the normal or shear contact bond strength	Sima et al. (2014)
	DiEM	The joint model used at the soil-base interface was Mohr- Coulomb failure criteria	Amarasiri et al. (2011)
	DLSM	Two-phase bond model (solid and liquid bond) Shear and tension failure but no compression failure	Gui and Zhao (2015)
	MD	Elastic-brittle material with a yield function	El Youssoufi et al. (2005)
	SNM	Solid particles connected by springs The inter-particle springs fail in tension Hydro-mechanical coupled with voronoi-based discretization technique	Asahina et al. (2014)
	SPH	Size-dependent constitutive model with an embedded cohesive fracture	Tran et al. (2019)
Hybrid approach	FEM- DEM	Mix-mode cohesive fracture coupled with tension, compression and shear material behaviour Inter-block failure criterion	Gui et al. (2016)
	PDS-FEM	Fracture defined by the loss of interaction between Voronoi blocks Pattern formation in desiccation cracking	Hirobe et al. (2016)

Table.5 Summary of numerical approach for simulating desiccation behaviour of soil

FDM: finite difference method; FEM: finite element method; XFEM: extended finite element method; MFT: fragmentation technique; CEM: cohesive zone model; DEM: discrete element method; DiEM: Distinct element method; DLSM: distinct lattice spring model; MD: molecular dynamics; SNM: spring network method; FEM-DEM: finite-discrete-element method; PDS-FEM: particle discretization scheme finite element method; PFM: phase field model; SPH: smoothed particle hydrodynamics.

#### 2.5. Peridynamics (PD) theory and its developments

Studies of erosion processes requires a comprehensive understanding the multi-physical processes involved, including free swelling of compacted clay (the continuum large deformation), phase changes (e.g., solid, gel, sol, fluid), particle detachment at clay/water interface (e.g., discontinuous particle release) and detached particle transport with flowing water. The computational complexity and challenges of the coupled problem resulted in a high demanding of the numerical approach in addressing multiple physical fields and discontinuous processes. Additionally, the challenges for modelling desiccation cracking processes are the

multi-physics nature of the phenomenon involved and the mathematical description of problems that includes emerging and propagating discontinuities in a material with complex, heterogeneous structure. The ambition of this work is to create a model for clay erosion and desiccation by integrating the hydro-chemical processes with mechanics of the problem. The existing studies and developments of PD have shown the capacity of PD in handling large deformation and discontinuities. This encouraged the author to adopt a peridynamic approach, i.e., a non-local formulations, for clay cracking and erosion. The advantages of PD compared to classical local approaches include: (i) PD is capable of handling discontinuities in the system by removing the spatial derivatives; (ii) PD allows for both mesh-free and mesh-based discretization; and (iii) PD based models have the capacity to capture discontinuous evolution in the computational domain (e.g. crack initiation and propagation, dissolution and erosion at the interface) (Zhou and Wang, 2021). PD is therefore adopted and where necessary further developed in this thesis to tackle the large deformation and discontinuous behaviour of clay under desiccation and erosion.

#### **Fundamentals of PD**

The non-local peridynamics theory first presented by Silling (2000) is very advantageous in solving discontinuity-related problems such as crack propagations. PD has been attractive to researchers as it is a non-local formulation in an integral form, unlike the local differential form of classical continuum mechanics, e.g.,

$$\rho(\mathbf{x})\ddot{\boldsymbol{u}}(\mathbf{x},t) = \boldsymbol{\sigma}_{ij,j} + \mathbf{b}(\mathbf{x},t) \quad i \& j = 1,2,3 \tag{13}$$

The new form of equation of motion in PD framework is given by

$$\rho(\mathbf{x})\ddot{\boldsymbol{u}}(\mathbf{x},t) = \int_{H_{x}} \boldsymbol{f}(\mathbf{x}' - \mathbf{x}, \boldsymbol{u}(\mathbf{x}',t) - \boldsymbol{u}(\mathbf{x},t), t) \, \mathrm{d}H_{x} + \mathbf{b}(\mathbf{x},t)$$
(14)

where  $\rho(\mathbf{x})$  and  $\mathbf{x}$  are the density of material and position vector of material points in the reference configuration ( $\mathbf{\Omega}$ ), *t* is time,  $\sigma_{ij,j}$  is the divergence of the stress tensor in CCM,  $\mathbf{b}(\mathbf{x},t)$  is body force vector field. *f* is the force state. The concept of a horizon, i.e.,  $H_x = \{\mathbf{x}' \in \mathbf{\Omega}, |\mathbf{x}' - \mathbf{x}| < \delta, \text{ is introduced which signifies, as its radius } (\delta), the finite distance over which two particles can interact and the long-range forces are non-negligible (Fallah et al., 2020). The PD theoretical framework replaces the partial differential equations (Eq.13) of the classical local theory by a set of integral-differential equations (Eq.14) (Silling, 2000; Silling and Bobaru, 2005). This results in a mathematically consistent formulation, even where strong$ 

discontinuities appear due to breaking and fragmentation of the material. An increasing interest in PD as a novel non-local approach is shown in Fig.18.



Fig.18 Number of citations of Silling (2000). The scatter points are obtained from SCOPUS and red solid line is an exponential fitted function.

A constitutive model is a relation that determines the force state f (Eq.14) in terms of the deformation state in the neighbourhood of **x** and possibly other variables. Peridynamic models can be fundamentally categorized into two groups: bond-based peridynamic (BB-PD) models and state-based peridynamic (SB-PD) models. BB-PD was originally proposed by Silling (2000), where interactions between two material points within the horizon are pairwise. The pairwise force interactions cause a restriction of Poission's ratio (e.g., 1/3 in two-dimensional and 1/4 in three-dimensional problem). The SB-PD was developed by Silling (2007) to overcome the restriction on the Poission's ratio via introducing peridynamics states. Based on the characteristic of peridynamic force states, there are two types of SB-PD: an ordinary state-based peridynamic (OSB-PD) model and a non-ordinary state-based peridynamic (NOSB-PD) model (Silling et al., 2007; Silling 2010). The direction of force vector is parallel with the bond in NOSB-PD (Yang et al., 2019). Fig.19 shows the illustration of bond-based, ordinary state-based, and non-ordinary state-based PD.



Figure 19. Illustration of the bond-based (BB) (top left), ordinary state-based (OSB) (bottom right), and non-ordinary state-based (NOSB) PD formulations (top right). The red points represent two material points (x and x), each having its own horizon ( $H_x$ ), represented by dark blue and light blue circle. The difference among BB-PD, OSB-PD and NOSB-PD lies in the magnitude and direction of the interaction forces (black arrows) between the materials points.

This thesis is focused on the development of bond-based PD (BB-PD) formulations for clay erosion and desiccation cracking as it is possible to link the bond-based PD model with the classical linear elasticity theory or transport equations to obtain the model parameters from well-established material parameters (Diehl, 2017). Additionally, the implementation of SB-PD is relatively complicated and requires extensive computational costs compared to BB-PD. Previous applications of PD to a number of fields relevant to the thesis are presented below.

# PD Theory for multi-physical problems

The nonlocal PD theory enables the coupling of different fields in the presence of discontinuities. Recent developments included heat transfer, fluid transport and corrosion. The PD theory has been successfully applied in different areas, including dynamic fracture (Rabczuk and Ren, 2019; Ren et al.,2017), fluid driven fractures (Ouchi et al., 2015), thermomechanics of fracture development for composites, rocks, ceramic and pellets (Oterkus et al., 2014; Bobaru and Duangpanya, 2010; Bobaru and Duangpanya, 2012; D'Antuono and Morandini, 2017; Wang and Zhou, 2019; Wang et al., 2018; Oterkus and Madenci 2017; Gao and Oterkus 2019), membranes and fibers (Silling and Bobaru, 2005), dynamic fracture of nanofiber (Bobaru, 2007), stress induced corrosion (Bobaru and Duangpanya, 2012; Chen and Bobaru, 2015; Jafarzadeh et al., 2019a and 2019b), hydraulic fracturing (Oterkus et al., 2014; Ouchi et al., 2015; Zhou et al., 2020) and multiphysics flow problems in unsaturated porous media (Yan et al., 2020).

*Heat transfer.* The classical local heat equation is not applicable to domains with emerging discontinuities because it contains spatial differentials of temperature. Bobaru and Duangpanya (Bobaru and Duangpanya, 2010; Bobaru and Duangpanya, 2012) first presented a PD model for thermal diffusion which adopted the bond-based version of PD and demonstrated its advantages in handling discontinuities with several applications. A state-based PD formulations for heat conduction was then proposed by Oterkus et al. (2014). Effects of the kernel choice on PD solution for heat transfer was investigated by Chen et al. (2015). The results indicated that different kernels represented the non-local properties of the material and converged to the local solution in the limit of vanishing horizon. The analysis of the effects of temperature on the deformation behaviour of materials is of interest for many applications. Coupled thermo-mechanical PD models have been therefore extensively studied in recent years. Oterkus et al. (2014) presented a fully coupled thermo-mechanics PD model based on thermodynamic theory. A weakly coupled method was then introduced in PD to solve thermomechanical problems (Wang et al., 2019; D'Antuono and Morandini, 2017). Additionally, coupled thermo-mechanical PD analysis was conducted for composites, rocks, ceramic, and pellets (D'Antuono and Morandini, 2017; Wang and Zhou, 2019; Wang et al., 2018; Oterkus and Madenci 2017; Gao and Oterkus 2019).

*Fluid flow in porous media.* The governing equation of flow in porous media is typically described by using Darcy's law employing local differential equations to establish the relationship between pressure and fluid flux. Katiyar et al. (2014) proposed a SB-PD

formulation for steady state flow of water in saturated porous media. A PD model for transient moisture flow in unsaturated porous media was also proposed by Jabakhanji and Mohtar (2015); which provided a solution to the classic Richard's equation of water flow in unsaturated porous media. Zhao et al. (2018) proposed PD model for transient advection and diffusion problems. Yan et al. (2020) presented PD formulations for coupled water flow and chemical transport in unsaturated porous media, addressing discontinuities and heterogeneities in the modelling of coupled flow. PD models for coupled flow and deformation have also been presented to study fracture initiation and propagation by a cumulative damage concept, which tracks broken mechanical bonds for material points (Oterkus et al., 2014; Ouchi et al., 2015; Zhou et al., 2020).

*Corrosion*. Corrosion and damage evolution is a major problem affecting material strength and safety of structures. A PD model for simulating corrosion by a non-local diffusion equation was presented by Chen and Bobaru (2015) to address corrosion damage issues in metals. De Meo and Oterkus (2017) designed a PD-FEM framework for the failure analysis of corrosion-resistant materials. The peridynamics formulations were extended by Jafarzadeh et al. (2019a, 2019b and 2019c) to address diffusion-type corrosion problems for evolving discontinuities or phase changes, e.g., pitting corrosion damage and mechano-chemical damage). Peridynamics modelling approach allows for autonomous evolution of the moving interface without any extra modelling efforts, e.g., interface conditions.

*Coupling schemes.* The coupling methods used in multiphysics problems include the strong coupling approach (direct coupling), and the weak coupling approach (iterative coupling) (Zhang and Cen, 2015). A weakly coupled method has been introduced in PD to solve coupled problems such as thermo-mechanical (D'Antuono and Morandini, 2017; Wang and Zhou, 2019; Wang et al., 2018). In a weakly coupled method, different time stepping strategies are generally employed for different physics/processes. PD formulations of coupled multiphysics problems have been developed which are based on sequential numerical approaches to solve the coupled problem (Agwai, 2011; Madenci and Oterkus, 2014; Oterkus et al., 2014; Oterkus et al., 2017; Ouchi et al., 2015). In these models the processes are sequentially addressed within a small-time step to satisfy convergence in the solutions. On the contrary, the strong coupling method solves the coupled equations directly and implicitly where the variable vectors of the coupled system are updated simultaneously by appropriate interface coupling methods, e.g., direct matrix assembly, direct interface coupling, multipoint constraint, and penalty method (Zhang

and Cen, 2015). The coupling methods for different coupling problems are dependent on the level of nonlinearity of the problems. For example, strong coupling approach is generally applied for systems with high nonlinearity, such as fluid–structure interaction with structural buckling and discontinuous shock wave in computational fluid dynamic field. If the nonlinearity of the coupled systems is not critical and the dependency between different physics processes is weak, such as thermal-stress and fluid-structure coupling problems, it can be solved by a weak coupling scheme (Zhang and Cen, 2015).

*PD developments and outlook in geotechnical engineering.* Non-local PD theory has been successfully applied to a wide range of geotechnical engineering problems. For example, Song and Menon (2019) presented a non-local SB-PD chemo-mechanical model for unsaturated soil using an implicit return mapping algorithm. Nadimi et al. (2016) and Zhou et al. (2020) employed the non-local PD model to study the hydraulic fracturing processes. Ha et al. (2015), Lee et al. (2017), Wang et al. (2016, 2017 and 2019) developed PD models to explain the initiation, propagation, branching and fragmentation of rock subjected to static and dynamic compression and tension. Panchadhara et al. (2017) employed peridynamics to study propellant-based well stimulation. Fan and Li (2017) developed a PD-SPH modelling scheme for blast fragmentation of soil under buried explosive loads. Recent studies of PD applications in geotechnical engineering are summarised in Table 6.

Applications	Model results	References
Geomaterial fragmentation	The PD formulation was capable to capture the dynamic responses and complex fracture processes of drained and saturated geomaterials.	Lai et al. (2015)
Sand particle crushing	The proposed PD framework produced reasonable results with respect to particle size distribution, fractal dimension, normal compression, as well as particle morphology.	Zhu and Zhao (2019a & 2019b)
Water flow	The proposed non-local model showed the capability to capture discontinuities and heterogeneities, including stationary cracks, propagating cracks, and randomly distributed permeable and impermeable inclusions.	Katiyar et al. (2014); Jabakhanji et al. (2015); Yan et al. (2020)
Clay desiccation	The results show that the model can capture realistically key hydraulic, mechanical and geometry effects on clay desiccation cracking, including initial, propagation and ultimate crack patterns.	Meng and Song (2019); Yan et al. (2021)
Clay erosion	The proposed multi-physics non-local formulation to clay erosion provides a robust framework for incorporating a wide set of additional couplings, including coupled hydro-chemical effects in the eroding environment.	Sedighi et al. (2021); Yan et al. (2021).
Strain localization analysis	The numerical results show that the proposed nonlocal model can be used to simulate the inception and propagation of shear banding as well as to capture the thickness of shear bands in geomaterials at a constant suction.	Song and Khalili (2018)
Hydraulic fracture	The peridynamics model allowed for simulating the propagation of multiple, nonplanar, interacting fractures and provided a novel approach to simulate the interaction between hydraulic fractures and natural fractures.	Nadimi et al. (2020); Zhou et al. (2020); Ouchi et al. (2015 & 2017); Oterkus et al. (2017)
Thermal damage in rock	The proposed PD model can accurately predict the crack initiation time, locations and failure patterns of rocks subjected to heating from boreholes.	Bazazzadeh et al. (2020); Wang et al. (2016, 2018 & 2019);
Stability analysis of rock and soil slopes	The developed PD model was used to characterize failure mechanisms of jointed rock masses and investigate the strength reduction of jointed rock.	Gao et al. (2021)
Rock explosion	The state-based PD formulation is successful applied to study large scale simulation of explosive fragmentation of soil up to five million particles.	Fan and Li (2017)

Table 6 PD application in geotechnical engineering

### 2.6 Conclusion and summary

This chapter presented a comprehensive review on cracking and erosion behaviour of swelling clay, including the structure and properties of bentonite clay, and recent developments in the experimentation and modelling.

The review of clay cracking showed that understanding of desiccation cracking behaviour depends on reliable experimental techniques, strong theoretical developments, and effective numerical approaches. The existing experimental tests can be categorized into three major

groups, including the restrained ring test, long bar test, and thin clay layer test, which provide qualitative and quantitative description of the dynamic cracking process and the evolving crack pattern. Theoretical models and numerical approaches applied for the analysis of clay desiccation cracking were summarized. The advantages and shortcomings of the existing numerical analysis were discussed. The main challenges for modelling clay cracking are the multi-physics nature of the phenomenon involved and the mathematical description of problems that includes emerging and propagating discontinuities in a material with complex, heterogeneous structure.

The review of clay erosion indicated that understanding the hydro-chemical effects on the clay erosion is especially important for the faithful assessment of EBS, the deposition tunnel backfill in the early phase of the saturation process and sealing of bentonite plug in the borehole during the emplacement. Understanding of erosion processes was improved in last 10 years mainly through extensive experimental studies, which demonstrated that the erosion processes were strongly affected by both solution chemistry and fluid velocity. Theoretical studies of bentonite erosion however are very limited due to the computational complexity of the problem, including the continuum large deformation (e.g., free swelling), phase changes (e.g., solid, gel, sol) and discontinuous particle release (e.g., detachment of sol at the clay/water interface).

A review of recent developments and applications of a non-local approach (peridynamics) for multi-physical problems was provided. Theoretical studies of clay erosion and cracking processes are very limited due to the computational complexity of the problem, including the continuum large deformation and discontinuous damage. The existing studies and developments of PD showed the capacity of PD in handling the large deformation and cracking and is therefore adopted as a modelling tool in this thesis.

The aim of the work is achieved by developing a solid non-local theoretical framework/tool that can be used to predict/study clay cracking and erosion damage.

- An integrating bond-based PD formulations for coupling chemical transport and water flow in unsaturated clay is presented in Chapter 3, which are fundamental elements or key factors for evaluating and developing erosion and desiccation models.
- The non-local formulations are then extended to couple deformation and water transport, which are applied to address the clay desiccation issues in Chapter 4. The proposed non-local models are validated by experimental data and used to study the mechanisms

controlling the initiation and propagation of discontinuities in clay cracking processes.

- The formulations developed in Chapters 3 and 4 are combined to incorporate the process
  of clay particle detachment by considering the stability/strength of hydrated gel and
  revisiting the clay water interaction theory, producing a single non-local modelling
  framework in Chapter 5 Section3. The proposed non-local formulations are validated by a
  set of experimental data and used to investigate the hydraulic and chemical effects on the
  penetration, extruded mass and particle release rate of swelling clays in Chapter 5 Section
  4. Chapter 5 Section 5 creates a new model for erosion with co-transport of accessory
  minerals building upon the formulation developed in Chapter 5 Section 1 and use it to
  investigate the role of minerals in the protection of erosion/expansion of swelling clay.
- Chapter 6 investigate the self-healing of swelling clay in piping channels using the full modelling framework and to quantify the amount of mass loss of swelling clays under piping flow for buffer/backfill and plug-in material.
- The PD formulations in the present work are implemented in Pyramid (multi-physics PD framework), which is a built-in MATLAB code and recently developed by the author in Yan et al. (2020).

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# CHAPTER Three

# 3. Peridynamics modelling of coupled water flow and chemical transport in unsaturated porous media

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#### 3.1 Paper introduction:

This paper presents a non-local approach by developing a bond-based formulation for coupled water flow and chemical transport in partially saturated porous media. An implicit numerical scheme was adopted to solve the coupled problems. The formulations were implemented within a computational framework (Pyramid) and tested against a set of benchmarks. The verifications demonstrated excellent agreements between the proposed approach and the classical/local formulations for homogeneous media. Extended capability of the model was demonstrated by analysis of coupled chemical transport and water flow problem in 2D domains with stationary and propagating cracks and various randomly distributed inclusions. The model was applicable to multi-physics problems in porous systems where coupled hydro-chemical degradations can create discontinuity or heterogeneity. The developments of non-local water flow and chemical transport models are key element for the study of cracking and erosion behaviour of clay.

#### 3.2 Contribution to the joint authorship

The author has contributed the following:

- Development of the main concept for this paper (80%).
- Theoretical developments for coupling water flow and chemical transport (90%).
- Model implementations and code writing (100%).
- Verifications and applications of the proposed model (90%).
- Preparation of tables and data visualisation (100%).
- Paper writing (80%).

#### Addendum

Figures 4, 5, 6, 8 and 9 in the published paper: The relative difference ( $\varepsilon$ ) is used to show the

validity of the present numerical model given as follow:

$$\varepsilon = \sum_{i=1}^{N} \left( \frac{\theta_{classical} - \theta_{peri}}{\theta_{classical}} \right)^2 \tag{1}$$

where, N is the number of all nodes used in every numerical test;  $\theta_{peri}$  is results obtained by this study;  $\theta_{classical}$  is results obtained by FEM method in this chapter.

Figure	Case	E (%)	Figure	Case	E (%)
Fig.4	<i>t</i> =1 s	12.92	Fig.8a	<i>t</i> =5 d	0.03
	t = 2 s	8.41		<i>t</i> =10 d	0.08
	<i>t</i> =3 s	0.95		<i>t</i> =15 d	0.21
Fig.5	t = 1 s	5.31	Fig.8b	<i>t</i> =5 d	10.00
	t = 2 s	0.13		<i>t</i> =10 d	15.86
	<i>t</i> =3 s	0.21		<i>t</i> =15 d	8.48
Fig.6a	<i>t</i> =5 d	0.08	Fig.9a	<i>t</i> =5 d	0
	<i>t</i> =10 d	0.05		<i>t</i> =10 d	0.05
	<i>t</i> =15 d	0.05		<i>t</i> =15 d	0.02
Fig.6b	<i>t</i> =5 d	73.5	Fig.9b	<i>t</i> =5 d	0.26
	<i>t</i> =10 d	5.25		<i>t</i> =10 d	0.17
	<i>t</i> =15 d	9.39		<i>t</i> =15 d	0.14
Fig.6c	<i>t</i> =5 d	86.32			
	<i>t</i> =10 d	20.11			
	<i>t</i> =15 d	5.88			

Table 1 The relative difference between the results from PD and FEM

### This Chapter is an exact copy of the accepted format manuscript referred to above.

## Peridynamics modelling of coupled water flow and chemical

### transport in unsaturated porous media

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#### Abstract

A wide range of phenomena in natural or engineered systems emerge from strongly coupled hydraulic, chemical and mechanical processes involving a mix of clearly discrete and quasicontinuous mechanisms. Non-local formulations of these processes, e.g. based on Peridynamics, offer significant advantages compared to classical local formulations. Existing Peridynamics models for water flow and chemical transport are applicable only to saturated systems and use loosely coupling schemes, such as explicit time stepping approaches. This work advances the non-local approach by developing a bond-based formulation for coupled water flow and chemical transport in partially saturated porous media. An implicit solution is proposed for coupling the PD formulation of chemical transport with water flow formulation. Firstly, the proposed formulation is verified against results from finite element/finite difference transient solutions for 1-D and 2-D coupled problems. The agreement between results demonstrates the accuracy of the proposed methodology. Secondly, a series of case studies are presented to illustrate the model's capability to capture discontinuities and heterogeneities, including stationary cracks, propagating cracks, and randomly distributed permeable and impermeable inclusions. The results show that the multi-physics Peridynamics-based formulations and computational model (Pyramid) provide clear advantages to classical local formulations for analyses of reactive transport in partially saturated porous media with physically realistic microstructures.

#### Keywords

Peridynamics; multiphysics, chemical transport; water flow; unsaturated, porous media

#### **1. Introduction**

Modelling approaches based on classic formulations of transport phenomena in porous material utilise the basic physics of fluid flow and chemical transport where the fluid and chemical fluxes are proportional to the gradient of a potential which are applied alongside the constitutive equations, describing the material's properties. In such formulations, the conservations of mass, momentum and energy are locally imposed via partial differential equations. Examples include Fick's law, Darcy's law and Fourier's law. For problems where processes are strongly coupled, additional complexity arises in the modelling approaches from the non-linearity and cross-coupling effects. Approximate solutions to coupled transport processes, described by the local theories, can be obtained by various numerical techniques (e.g. Finite Element Method (FEM), Finite Difference Method (FDM), and Boundary Element Method (BEM)).

The local formulations, however, are less efficient for analysing heterogeneous media or media with discontinuities (Jafarzadeh et al., 2019b). Examples include porous systems, whose fundamentally discrete structures span several length scales with associated discretely changing transport properties (e.g., permeability and diffusivity), and systems with discontinuous transport across phase boundaries (Carciopolo et al., 2020). Such underlying structures result in fast and slow transport processes existing in the same computational domain. It becomes extremely challenging to solve problems with heterogeneities and discontinuities without extra requirements such as multi-scale algorithms and adaptive multilevel schemes (Helmig et al., 2010). Moreover, in a natural heterogeneous porous medium, the moisture flux at a given location depends on the pressure distribution in a finite (not infinitesimal) size neighbourhood, which requires 'connections' in discretised domain (Delgoshaie et al., 2015; Jenny and Meyer, 2017). In the presence of discontinuities, such as cracks or shocks, additional requirements such as dependence of density on concentration, need to be imposed to represent the flow realistically (Koldoba and Koldoba, 2003). Without imposing these requirements, the solutions of the partial differential equations may not converge. Furthermore, the classical local formulations of chemical transport in porous media have limitations to capture important phenomena, such as anomalous transport observed in heterogeneous or discontinuous environments, where nonlocal effects are important (Liu et al., 2019; Liu et al., 2017; Zhao et al., 2018).

Nonlocal formulations, e.g. weakly nonlocal theories, nonlocal elasticity, nonlocal strainsoftening models and Peridynamics, have been developed in recent decades to address problems with discontinuous systems, large deformations and heterogeneities (Bažant and Chang, 1984; Eringen and Edelen, 1972; Rogula, 1982; Silling, 2000). Among those, the Peridynamics (PD) theory is receiving increasing interest among the continuum mechanics community. The theory of peridynamics (Silling, 2000) presents a set of equations of continuum mechanics to overcome the limitations of the classical local theory in solving problems with discontinuities. A major development in this theoretical framework is that integral-differential equations replace the partial differential equations of the classical local theory. This results in obtaining consistent formulation from mathematical point of view where discontinuities are developed in the porous media (e.g. fracture, fragmentation). The PD theory has successfully been applied in different areas, including fluid driven fractures (Oterkus et al., 2017; Ouchi et al., 2015), thermal fracture (Oterkus et al., 2014; Wang and Zhou, 2019), membranes and fibers (Silling and Bobaru, 2005), dynamic fracture of nanofiber(Bobaru, 2007), pitting corrosion damage (Jafarzadeh et al., 2019a) and electro-migration (Gerstle et al., 2008).

The PD theory bypasses the calculations of spatial derivatives, which can be mathematically undefined in the presence of sharp discontinuity in the field variables (such as a crack or a shock) (Madenci and Oterkus, 2014). This feature allows for developing PD formulations for physical phenomena beyond the original applications to solids behaviour proposed by Silling (2000) (Silling, 2000). For example, Bobaru and Duangpanya (Bobaru and Duangpanya, 2010; Bobaru and Duangpanya, 2012) presented a PD model for thermal diffusion which adopted the bond-based version of PD. The generic PD formulations have also been used for studying mass diffusion (Du et al., 2012; Tian et al., 2017; Tian and Du, 2013). Katiyar et al. (Katiyar et al., 2014) proposed a PD formulation for steady state flow of water in saturated porous media; addressing discontinuities and heterogeneities in the modelling of fluid flow. PD models for coupled flow and deformation have also been presented to study fracture initiation and propagation by a cumulative damage concept, which tracks broken mechanical bonds for material points (Oterkus et al., 2014; Ouchi et al., 2015). A PD model for transient moisture flow in unsaturated porous media was also proposed by Jabakhanji and Mohtar (Jabakhanji and Mohtar, 2015); which provides a solution to the classic Richard's equation of water flow in unsaturated porous media. The PD formulations presented for water flow and chemical transport problems are limited to those solving either the water flow (Jabakhanji and Mohtar,

2015; Katiyar et al., 2014) or the chemical transport (Gerstle et al., 2008) separately, or offering a weak coupling scheme to deal with the two processes only in saturated systems (Zhao et al., 2018).

The coupling methods used in multiphysics problems include the strong coupling approach (i.e., direct coupling), and the weak coupling approach (i.e., iterative coupling) (Zhang and Cen, 2015). A weakly coupled method has been introduced in PD to solve coupled problems such as thermo-mechanical (D'Antuono and Morandini, 2017; Wang and Zhou, 2019; Wang et al., 2018). In a weakly coupled method, different time stepping strategies are generally employed for different physics/processes. PD formulations of coupled multiphysics problems have been developed which are based on sequential numerical approaches to solve the coupled problem (Agwai, 2011; Madenci and Oterkus, 2014; Oterkus et al., 2014; Oterkus et al., 2017; Ouchi et al., 2015). In these models the processes are sequentially addressed within a time step which can fail to satisfy convergence in the solutions. This is often tacked at the expense of adopting a small-time step (Oterkus et al., 2014).

To the authors' knowledge, there is no PD formulation which can describe an implicit coupled water flow and chemical transport in partially saturated porous media. Such coupled hydrochemical formulation is an essential step to develop PD-based fully coupled hydro-chemicalmechanical models for studying the chemo-mechanical problems such as erosion assisted degradation of materials or hydro-chemo-mechanical behaviour of fractured systems where coupling effects are inherently combined with modelling challenges associated with discontinuity and heterogeneity of the system. By using the non-local theory of peridynamics, transport in systems with spatially varying geometrical and physical properties can be solved without resort to special multi-scale or adaptive schemes.

This paper presents the development of a PD formulation for coupled water flow and chemical transport in discontinuous and/or heterogeneous unsaturated porous media. Bond-based Peridynamics formulations for water flow and chemical transport, considering advection, dispersion and diffusion processes in partially saturated porous media, are derived and presented. The proposed coupling of water flow and chemical transport processes is implemented based on an implicit numerical scheme. The new coupled hydraulic and chemical Peridynamics model (Pyramid) is tested against several verification bench marks to ensure accurate derivation and implementation of the formulations. A series of case studies are presented of transient water flow and chemical transport in fractured and heterogeneous

unsaturated porous media, by which the effects of fractures and randomly distributed permeable and impermeable regions are investigated by the new capabilities of the proposed model.

#### 2. Bond-based Peridynamics formulations for water flow and chemical transport

This section describes a bond-based PD formulation for the transport equations. The classical continuum equations for water flow and chemical transport are provided in Appendix A. The PD considers a body occupying a region ( $\mathbf{R}$ ) as a collection of interacting material points  $\mathbf{x}$ . The points are identified with their coordinates: (x) in 1-D, (x, y) in 2-D, and (x, y, z) in 3-D problems. Specifically for transport problems, each point is assigned physical variables: volume and concentration, from where its mass is calculated; or volume and mass, from where its concentration is calculated. In the following description, we will assume that the mass is the primary variable for transport problems, instead of the alternative concentration. A material point x interacts with (is connected to) all materials points x within a certain finite region  $H_x$ , called the horizon of x (see Fig. 1). The radius of the horizon is denoted by  $\delta$ . The neighbourhood of x is divided into a region, where the points have larger mass than x, denoted by  $H_{x+}$ , and a region, where the points have smaller mass than x, denoted by  $H_{x-}$ . S denotes the equivalent mass at the surface of the horizon of x. The conductor of mass between two interacting material points x and x` is called 'mass bond' or 'm-bond' (Bobaru and Duangpanya, 2010; Bobaru and Duangpanya, 2012). The Peridynamics mass flux per unit volume along PD bond is assumed to be dependent on the distance between the material points x and x (Bobaru and Duangpanya, 2012). The pairwise mass exchange between material points x and x is denoted by  $f_h$ .



# Fig. 1. Illustration of concentration bonds (left) and Peridynamics flux between connected points (right)

The reactive chemical transport and water flow PD formulations will be presented in Section 2.1 and Section 2.2. There are four steps for the construction of PD transport formulations. Step I: we construct the peridynamic flux along the bond between material points x and x` (Fig.1). Step II: the conservation of mass law is used to develop the peridynamic transport formulations along a single m-bond between points x and x`. Step III: We integrate peridynamic flux over the horizon of material point x to obtain the peridynamic transport formulations. It is noted that x'-x or ||x'-x|| is treated as a finite distance, which is different from the way that the mass conservation equation is obtained in the classical transport model where a limit of  $x' \rightarrow x$  is taken (Bobaru and Duangpanya, 2010). Step IV: We connect peridynamics' parameters to the measurable material properties to obtain the PD micro-diffusivity and micro-hydraulic conductivity by using different influence functions, which will be presented in Section 2.3. To clarify further these points, we provide detailed derivations of 1-D pure chemical diffusion PD formulations in Appendix B.

#### 2.1. PD formulations for chemical transport

The vector of Peridynamics' chemical mass flux along the 'm bond' connecting points x and x 'is given by (Zhao et al., 2018):

$$J_{n}(\boldsymbol{x},t) = D(\boldsymbol{x},\boldsymbol{x}',t) \frac{C(\boldsymbol{x}',t) - C(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} - \bar{q}(\boldsymbol{x},\boldsymbol{x}',t) \left[C(\boldsymbol{x}',t) - C(\boldsymbol{x},t)\right] \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(1)

where, D(x, x', t) is the diffusivity of the 'm-bond' between the material points x and x', C(x', t) and C(x, t) are chemical concentrations at material points x' and x, respectively;  $\xi$ represents the distance vector between the two material points x and x', i.e.  $\xi = x' - x$  (Silling, 2000),  $\frac{\xi}{\|\xi\|}$  is the unit vector along the mass bond; and  $\overline{q}(x, x', t)$  is the average water flux

between the two material points x and x'.

In previous studies where saturated systems were of interest (e.g. (Zhao et al., 2018)) the chemical advective flux induced by the water flux are assumed to be constant. However, considering a transient advection flux (Eq. A-2 in Appendix A) and variable unsaturated hydraulic conductivity (Eq. A-4 in Appendix A) is necessary for modelling the chemical

advection in partially saturated system which creates further nonlinearity. In order to couple the transient advection induced by the water flow, an average water flux  $\overline{q}(x, x', t)$  is defined (Oterkus et al., 2017):

$$\overline{q}(\boldsymbol{x}, \boldsymbol{x}', t) = \frac{q(\boldsymbol{x}, t) + q(\boldsymbol{x}', t)}{2}$$
(2)

 $q(\mathbf{x}, t)$  and  $q(\mathbf{x}', t)$  are the liquid velocity vectors of the material points  $\mathbf{x}$  and  $\mathbf{x}'$ . We note that for 2-D problems,  $q(\mathbf{x}, t)$  has components in x and y directions:  $q_x(\mathbf{x}, t)$  and  $q_y(\mathbf{x}, t)$ . The velocity/advection vector can be obtained by solving the water transport equations, which will be given in Section 2.2.

The conservation of mass law led to the peridynamic chemical transport in a single m-bond (x, x') in the direction  $\frac{\xi}{\|\xi\|}$  as follow (Bobaru and Duangpanya, 2010):

$$\theta_{l,a}(\mathbf{x},\mathbf{x}',t)\frac{\partial C_a(\mathbf{x},\mathbf{x}',t)}{\partial t} = \frac{J_n(\mathbf{x},t)}{\|\mathbf{x}-\mathbf{x}'\|}$$
(3)

Combining Eqns. (1) and (3), the mass conservation equation for the bond along the two material points x and x' can be presented as:

$$\theta_{l,a}(\mathbf{x},\mathbf{x}',t)\frac{\partial C_a(\mathbf{x},\mathbf{x}',t)}{\partial t} = D(\mathbf{x},\mathbf{x}',t)\frac{C(\mathbf{x}',t) - C(\mathbf{x},t)}{\left\|\boldsymbol{\xi}\right\|^2} \cdot \frac{\boldsymbol{\xi}}{\left\|\boldsymbol{\xi}\right\|} - q(\mathbf{x},\mathbf{x}',t)\frac{C(\mathbf{x}',t) - C(\mathbf{x},t)}{\left\|\boldsymbol{\xi}\right\|} \cdot \frac{\boldsymbol{\xi}}{\left\|\boldsymbol{\xi}\right\|}$$
(4)

where,  $C_a(\mathbf{x}, \mathbf{x'}, t)$  is the average concentration along the m-bond between  $\mathbf{x}$  and  $\mathbf{x'}$  given as  $C_a(\mathbf{x}, \mathbf{x'}, t) = \frac{C(\mathbf{x}, t) + C(\mathbf{x'}, t)}{2}$ , and  $\theta_{l,a}(\mathbf{x}, \mathbf{x'}, t)$  is the average moisture content along m-bond  $(\mathbf{x}, \mathbf{x'})$ .

In order to obtain the mass conservation at the material point x due to a chemical flux from all the bonds attached to material point x within its horizon  $(H_x)$ , we integrate Eq. (4) over the horizon  $H_x$  of material point x:

$$\int_{H_{x}} \theta_{l,a}(\mathbf{x}, \mathbf{x}', t) \frac{\partial C_{a}(\mathbf{x}, \mathbf{x}', t)}{\partial t} d\mathbf{V}_{\mathbf{x}'} = \int_{H_{x}} D(\mathbf{x}, \mathbf{x}', t) \frac{C(\mathbf{x}', t) - C(\mathbf{x}, t)}{\|\mathbf{x} - \mathbf{x}'\|^{2}} - \overline{q}(\mathbf{x}, \mathbf{x}', t) \frac{C(\mathbf{x}', t) - C(\mathbf{x}, t)}{\|\mathbf{x} - \mathbf{x}'\|} d\mathbf{V}_{x}$$
(5)

where,  $V_{x'}$  is the volume (area in 2D, length in 1D) associated with x'. The uniform grid (spacing  $\Delta x$ ) is used for desretization in this study. The volume of x' is therefore represented by  $\Delta x^3$  ( $\Delta x^2$  for 2D,  $\Delta x$  for 1D). It is worth noting that we have implemented successfully nonuniform grids, based on Voronoi tesselations within peridynamics models. However, for the purposes of the present work, it is sufficient to use uniform grids, and we have used only those.

The relationship between the concentration at point x and time t and the average concentration in all the m-bonds connected to material point x is given by (Bobaru and Duangpanya, 2010):

$$\int_{H_{x}} \theta_{l,a}(\boldsymbol{x}, \boldsymbol{x}', t) \frac{\partial C_{a}(\boldsymbol{x}, \boldsymbol{x}', t)}{\partial t} d\boldsymbol{V}_{\boldsymbol{x}'} = \theta_{l}(\boldsymbol{x}, t) \frac{\partial C(\boldsymbol{x}, t)}{\partial t} \boldsymbol{V}_{H_{x}}$$
(6)

where,  $V_{H_x}$  is the horizon volume for the node centred at x.

Combining Eqns. (5) and (6) leads to the following equation:

$$\theta_{l}(\boldsymbol{x},t)\frac{\partial C(\boldsymbol{x},t)}{\partial t} = \int_{H_{x}} \frac{D(\boldsymbol{x},\boldsymbol{x}',t)}{\boldsymbol{V}_{H_{x}}} \frac{C(\boldsymbol{x}',t) - C(\boldsymbol{x},t)}{\|\boldsymbol{x} - \boldsymbol{x}'\|^{2}} - \frac{\overline{q}(\boldsymbol{x},\boldsymbol{x}',t)}{\boldsymbol{V}_{H_{x}}} \frac{C(\boldsymbol{x}',t) - C(\boldsymbol{x},t)}{\|\boldsymbol{x} - \boldsymbol{x}'\|} d\boldsymbol{V}_{x'}$$
(7)

The Peridynamics microscopic diffusivity and the Peridynamics microscopic liquid flux are defined respectively as:

$$d(\boldsymbol{x}, \boldsymbol{x}', t) = \frac{D(\boldsymbol{x}, \boldsymbol{x}', t)}{V_{H_x}}$$
(8)

$$\overline{v}(\boldsymbol{x}, \boldsymbol{x}', t) = \frac{\overline{q}(\boldsymbol{x}, \boldsymbol{x}', t)}{V_{H_x}}$$
(9)

The pairwise mass exchange  $f_h$  is also defined by:

$$f_{h}(C'-C, \mathbf{x}'-\mathbf{x}, t) = d(\mathbf{x}, \mathbf{x}', t) \frac{C(\mathbf{x}', t) - C(\mathbf{x}, t)}{\|\mathbf{x} - \mathbf{x}'\|^{2}} - \overline{v}(\mathbf{x}, \mathbf{x}', t) \frac{C(\mathbf{x}', t) - C(\mathbf{x}, t)}{\|\mathbf{x} - \mathbf{x}'\|}$$
(10)

By substituting Eqns.(8) and (9) into Eq. (7) and combining with Eq.(10), the bond-based Peridynamics formulation for the coupled water flow and chemical transport in partially porous media at the material point at x can be expressed in the following form:

$$\theta_{l}(\boldsymbol{x},t)\frac{\partial C(\boldsymbol{x},t)}{\partial t} = \int_{H_{x}} f_{h}(C(\boldsymbol{x}',t) - C(\boldsymbol{x},t), \boldsymbol{x}' - \boldsymbol{x},t) d\boldsymbol{V}_{\boldsymbol{x}'}$$

$$= \int_{H_{x}} d(\boldsymbol{x},\boldsymbol{x}',t) \frac{C(\boldsymbol{x}',t) - C(\boldsymbol{x},t)}{\|\boldsymbol{x} - \boldsymbol{x}'\|^{2}} - \overline{v}(\boldsymbol{x},\boldsymbol{x}',t) \frac{C(\boldsymbol{x}',t) - C(\boldsymbol{x},t)}{\|\boldsymbol{x} - \boldsymbol{x}'\|} d\boldsymbol{V}_{\boldsymbol{x}'}$$
(11)

The micro-advection term,  $\overline{v}(\cdot, \cdot, \cdot)$  in Eq. (11), is induced by the water flow due to the differences in water contents of interacting points lead the water flux into their connecting points. In order to calculate the micro-advection, a Peridynamics formulation for water flow is required to enable a coupled chemical transport and water flow is expanded. The following section describes the PD formulation for the water flow.

#### 2.2. PD formulations for water flow

Similarly to Eq. (1), the Peridynamics water flux between the two interacting material points x and x' per unit volume of x' is described as:

$$q_{l}(\boldsymbol{x}',\boldsymbol{x},t) = K(\boldsymbol{x}',\boldsymbol{x},t) \frac{H(\boldsymbol{x}',t) - H(\boldsymbol{x},t)}{\|\boldsymbol{x} - \boldsymbol{x}'\|}$$
(12)

Where,  $q_l(x', x, t)$  is the volumetric flux of liquid water; H(x, t) and H(x', t) are the hydraulic potentials at material point x and x', respectively; K(x', x, t) is the hydraulic conductivity between the material points x and x'.

Mass conservation equation, Eq. (A-1), for the bond between material points x and x' can be written within the PD framework as (Katiyar et al., 2014):

$$\frac{\partial \theta_l(\mathbf{x},t)}{\partial t} = K(\mathbf{x}',\mathbf{x},t) \frac{H(\mathbf{x}',t) - H(\mathbf{x},t)}{\|\mathbf{x} - \mathbf{x}'\|^2}$$
(13)

Eq. (13) is integrated over the horizon of x to obtain the mass conservation for point x due to flow by all the bonds attached to the material point x within its horizon $H_x$ .

$$\int_{H_{x}} \frac{\partial \theta_{l}(\boldsymbol{x},t)}{\partial t} d\boldsymbol{V}_{\boldsymbol{x}'} = \int_{H_{x}} K(\boldsymbol{x}',\boldsymbol{x},t) \frac{H(\boldsymbol{x}',t) - H(\boldsymbol{x},t)}{\|\boldsymbol{x} - \boldsymbol{x}'\|^{2}} d\boldsymbol{V}_{\boldsymbol{x}'}$$
(14)

Similar to Eq. (6), we assume that the relationship between the water content at point x and time t and the average water content in all the bonds connected to x is given by:

$$\int_{H_x} \frac{\partial \theta_l(\mathbf{x}, t)}{\partial t} \mathrm{d} \mathbf{V}_{\mathbf{x}'} = \frac{\partial \theta_l(\mathbf{x}, t)}{\partial t} \mathbf{V}_{H_x}$$
(15)

From Eqns. (13) -(15), we obtain the mass conservation equation for liquid water flow for the material point x as:

$$\frac{\partial \theta_{l}(\boldsymbol{x},t)}{\partial t} = \int_{H_{x}} \kappa(\boldsymbol{x}',\boldsymbol{x},t) \frac{H(\boldsymbol{x}',t) - H(\boldsymbol{x},t)}{\|\boldsymbol{x} - \boldsymbol{x}'\|^{2}} d\boldsymbol{V}_{\boldsymbol{x}}.$$
(16)

where,  $\kappa(\mathbf{x}', \mathbf{x}, t) = \frac{K(\mathbf{x}', \mathbf{x}, t)}{V_{H_{\mathbf{x}}}}$  is the Peridynamics' micro-hydraulic conductivity function.

The newly derived Peridynamics based formulations for the coupled water flow and chemical transport, Eqns. (11) and (16), do not involve any partial derivatives. It is also indicated that the proposed PD formulations can be applied everywhere in the domain including in cases of discontinuities of the parameters (i.e.  $d(\cdot, \cdot, \cdot)$  and  $\overline{v}(\cdot, \cdot, \cdot)$ ) and discontinuities of the domain.

#### 2.3. Peridynamics' parameters

The Peridynamics' parameters, including micro-hydraulic conductivity  $\kappa(\mathbf{x}', \mathbf{x}, t)$  and microdiffusivity  $d(\mathbf{x}', \mathbf{x}, t)$ , are non-measurable parameters. However, the classical (local) material properties including diffusivity D and hydraulic conductivity K are experimentally measurable parameters. We present  $\kappa(\mathbf{x}', \mathbf{x}, t)$  and  $d(\mathbf{x}', \mathbf{x}, t)$  as functions of the hydraulic conductivity and diffusivity, respectively to develop the connection between the Peridynamics and the classical descriptions of the material constitutive relationships.

Inside a nonlocal Peridynamics region, the micro-hydraulic  $\kappa(x', x, t)$  conductivity and microdiffusivity d(x', x, t) depend on the distance between the interating points (Chen and Bobaru, 2015). In order to assess the level (significance) of contributions of all the bonds participating in calculating the volume-dependent properties, the influence function is introduced (Seleson and Parks, 2011; Silling et al., 2007). The most common of influence functions are the shape of "uniform" and "triangular" (see Fig.2). The shape of the different influence function is important in the numerical approximations. However, investigating the specific effects that the shape of the influence functions on the overall behaviour of a system are yet to be fully investigated (Jabakhanji and Mohtar, 2015). Different influence functions have been tested by Jabakhanji and Mohtar (Jabakhanji and Mohtar, 2015) which indicate that the solutions obtained by utilising "uniform" and "triangular" influence functions correlate closely with the solutions obtained by FEM. A uniform influence function is considered in this study for both micro-hydraulic conductivity and micro-diffusivity. By using a uniform influence function  $(d(\mathbf{x}', \mathbf{x}, t) = d$  and as shown in Fig.2a and Fig.2b) the Peridynamics hydraulic conductivity and Peridynamics diffusivity will be independent to the distance between the points in the horizon which provides a simplified form of the solution.



Fig. 2. The micro-diffusivity influence functions (a) one-dimension uniform influence functions; (b) two-dimension uniform influence functions; (c) one-dimension triangular influence functions and (d) two-dimension triangular influence functions.

The Peridynamics pure diffusive flux and the classic pure diffusive flux under steady-state conditions are considered to be equal (Bobaru and Duangpanya, 2010) which enables determining the Peridynamics micro-diffusivity. Based on Fick's law of diffusion, the steady-state concentration and flux for pure diffusion at position x can be given by:

$$C(\mathbf{x}) = m\mathbf{x} + n \tag{17}$$

$$J_{diff}\left(\boldsymbol{x}\right) = -D\frac{\partial C}{\partial x} = -mD \tag{18}$$

where, *m* and *n* are constants,  $J_{diff}(x)$  is the classic chemical flux for pure diffusion.

The peridynamic flux for pure diffusion along the 'c bond' connecting points x and x' can be obtained by reducing the advection term in Eq.(1):

$$J_n(x,t) = d(\mathbf{x}, \mathbf{x}', t) \frac{C(\mathbf{x}', t) - C(\mathbf{x}, t)}{\|\mathbf{x} - \mathbf{x}'\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(19)

In order to derive the Peridynamics chemical flux in all the bonds attached to x in its horizon  $H_x$ , Eq. (19) is integrated over the horizon of material point x. For 1D and 2D diffusion cases, the Peridynamics' chemical flux can be expressed as:

$$J_n^{peri}(\mathbf{x},t) = -\int_0^\delta d(\mathbf{x},\mathbf{x}',t) \frac{C(\mathbf{x}',t) - C(\mathbf{x},t)}{\|\mathbf{x} - \mathbf{x}'\|} \,\mathrm{d}\,\mathbf{x}$$
(20)

$$J_n^{peri}(\mathbf{x},t) = -\int_{-\pi/2}^{\pi/2} \int_0^{\delta} d(\mathbf{x},\mathbf{x}',t) \frac{C(\mathbf{x}',t) - C(\mathbf{x},t)}{\|\mathbf{x} - \mathbf{x}'\|} \cos^2(\varphi) r \,\mathrm{d}r \,\mathrm{d}\varphi$$
(21)

By substituting the uniform influence function (see Fig.2) and the steady-state concentration equation, Eq. (17), into Eqns. (20) and (21), we obtain:

$$J_n^{peri}(x,t) = \begin{cases} -md\delta & \text{one-dimension} \\ -dm\pi\delta^2/4 & \text{two-dimension} \end{cases}$$
(22)

If the classic chemical flux, Eq. (18), and Peridynamics chemical flux, Eq. (22), at material point x are equal, the micro-diffusivity for 1D and 2D transport cases are obtained to be:

$$d(\mathbf{x}', \mathbf{x}, t) = \begin{cases} \frac{D}{\delta} & \text{one-dimension} \\ \frac{4D}{\pi\delta^2} & \text{two-dimension} \end{cases}$$
(23)

Similar process is also applied to obtain the micro-hydraulic conductivity. Based on Darcy's law, the steady-state water flux due to a linear hydraulic potential field (H(x) = ax + b) at position x is:

$$q_c(\mathbf{x}) = -Ka \tag{24}$$

where, *a* is constant and  $q_c(x)$  is the classic Darcy's flux.

The Peridynamics' water flux (Eq.12) in all the bonds connected to material point x within its horizon  $H_x$ , for 1D and 2D transport cases can be expressed as:

$$q_l^{peri}(\mathbf{x},t) = -\int_0^\delta \kappa(\mathbf{x},\mathbf{x}',t) \frac{H(\mathbf{x}',t) - H(\mathbf{x},t)}{\|\mathbf{x} - \mathbf{x}'\|} \,\mathrm{d}\,\mathbf{x}$$
(25)

$$q_l^{peri}(\boldsymbol{x},t) = -\int_{-\pi/2}^{\pi/2} \int_{0}^{\delta} \kappa(\boldsymbol{x},\boldsymbol{x}',t) \frac{H(\boldsymbol{x}',t) - H(\boldsymbol{x},t)}{\|\boldsymbol{x} - \boldsymbol{x}'\|} \cos^2(\varphi) r \,\mathrm{d}\, r \,\mathrm{d}\,\varphi$$
(26)

Equivalence between the classic Darcy's flux, Eq. (24), and the Peridynamics flux, Eqns. (25) and (26), at material point x, provide the micro-conductivity relations:

$$\kappa(\mathbf{x}', \mathbf{x}, t) = \begin{cases} \frac{K}{\delta} & \text{one-dimension} \\ \frac{4K}{\pi\delta^2} & \text{two-dimension} \end{cases}$$
(27)

#### 3. Numerical solution

The numerical solution for the PD equations of water flow (Eq. 16) and chemical transport (Eq. 11) are developed for a finite number of PD particles. The domain is discretized using a uniform grid with spacing  $\Delta x$ . Each node has a "volume" (length in 1D and area in 2D). Figure 3 shows the 1D discretization around a node x. In this illustration, the points located in yellow region are the points that their chemical concentrations and water contents are larger than that of the material point x. The points located in red region points are those points that their chemical concentrations and water contents are those points that their chemical concentrations are smaller than that of the material point x. N is the total number of particles in the whole domain. For a given PD particle x, the set of particles x within  $H_x$  is called  $P_x$  and defined as:

$$p_{\mathbf{x}} = \left\{ \mathbf{x}' \neq \mathbf{x} \, \big\| \, \mathbf{x} - \mathbf{x}' \big\| \le \delta \right\}$$
(28)



Fig. 3. Numerical discretization around a node  $x_i$ . The circular region is the horizon region  $(H_x)$  of  $x_i$ . The set of particles within  $H_x$  is called  $P_x$ .

The spatial discretization of Peridynamics moisture flow, Eq. (16), and Peridynamics chemical transport in partially saturated soil, Eq. (11), for the given PD particle can be approximated as:

$$\frac{\partial \theta_{l}(\boldsymbol{x}_{i},t)}{\partial t} = \sum_{p} \kappa \left(\boldsymbol{x}_{i}, \boldsymbol{x}_{p}, t\right) \frac{H\left(\boldsymbol{x}_{p}, t\right) - H\left(\boldsymbol{x}_{i}, t\right)}{\left|\boldsymbol{x}_{p} - \boldsymbol{x}_{i}\right|^{2}} V_{ip}$$
(29)

$$\theta_{l}(\boldsymbol{x}_{i},t)\frac{\partial C(\boldsymbol{x}_{i},t)}{\partial t} = \sum_{p} d\left(\boldsymbol{x}_{i},\boldsymbol{x}_{p},t\right)\frac{C\left(\boldsymbol{x}_{p},t\right) - C\left(\boldsymbol{x}_{i},t\right)}{\left|\boldsymbol{x}_{p}-\boldsymbol{x}_{i}\right|^{2}}V_{ip} - \sum_{p} \bar{v}\left(\boldsymbol{x}_{i},\boldsymbol{x}_{p},t\right)\frac{C\left(\boldsymbol{x}_{p},t\right) - C\left(\boldsymbol{x}_{i},t\right)}{\left|\boldsymbol{x}_{p}-\boldsymbol{x}_{i}\right|}V_{ip}$$
(30)

The implicitly coupled method is used for solving the coupled problems. In this method, equations for water flow and chemical transport are solved simultaneously. The spatially discretized equations for water flow and chemical transport can be combined in a matrix form. The matrix equation can conveniently be expressed as:

$$\mathbf{A}\phi + \mathbf{B}\frac{\partial\phi}{\partial t} + \mathbf{C} = \mathbf{0} \tag{31}$$

where A, B and C are the matrices of coefficients and  $\phi$  is the vector of variables.

We use the forward Euler method for the time integration of Eq. (31):

$$\mathbf{A}^{n+1}\boldsymbol{\phi}^{n+1} + \mathbf{B}^{n+1} \left[ \frac{\boldsymbol{\phi}^{n+1} - \boldsymbol{\phi}^n}{\Delta t} \right] + \mathbf{C}^{n+1} = \mathbf{0}$$
(32)

Rearranging Eq.(32) gives:

$$\phi^{n+1} = \left[ \mathbf{A}^{n+1} + \frac{\mathbf{B}^{n+1}}{\Delta t} \right]^{-1} \left[ \frac{\mathbf{B}^{n+1} \phi^n}{\Delta t} - \mathbf{C}^{n+1} \right]$$
(33)

For the numerical treatment of the implicitly coupled water flow and chemical transport Peridynamics system of equations, the same time stepping is adopted. The equation of water flow is solved for the fluid field and the chemical transport equation is solved for the concentration field. In order to approximate the solutions to both equations, we adopt an explicit time stepping scheme.

In order to illustrate the numerical implementation, the solution applied to the 1D Peridynamics coupled equations are discussed. Similar procedure has been applied to the 2D problem. In this study, the horizon size of  $\delta=3\Delta$  has been selected which is based on the efficient numerical convergence study confirmed by various researchers (Agwai, 2011).  $\Delta$  is the spacing between the material points. As shown in Figure 3, the material point of interest x is denoted by  $x_i$  which interacts with the three points to its left and right. Hence, the set point x` from left to right

within the horizon of  $x_i$  are  $x_{i-3}$ ,  $x_{i-2}$ ,  $x_{i-1}$ ,  $x_{i+1}$ ,  $x_{i+2}$ ,  $x_{i+3}$ , as shown in Fig.3. The Peridynamics' water flow and chemical transport formulations, Eqns. (29) and (30), at the n<sup>th</sup> time step (i.e., the current time step), can be presented in a matrix form as:

$$\begin{bmatrix} 1/\Delta t & 0\\ 0 & \theta_l(x_i, t^{n+1})/\Delta t \end{bmatrix} \begin{bmatrix} \theta_l(x_i, t^{n+1}) - \theta_l(x_i, t^n)\\ C(x_i, t^{n+1}) - C(x_i, t^n) \end{bmatrix} = \begin{bmatrix} \mathbf{K} \end{bmatrix} \begin{bmatrix} \mathbf{M} \end{bmatrix}$$
(34)

The details of matrix  $[\mathbf{K}]$  and  $[\mathbf{M}]$  are provided in Appendix C.

The approach proposed by Bobaru and Duangpanya (Bobaru and Duangpanya, 2012) is adopted for prescribing the boundary conditions. The nonlocal Dirichlet boundary condition is applied directly at the boundary nodes. The nonlocal Dirichlet boundary condition converges to the local one as the size of the horizon becomes smaller and approaches zero. The Neumann boundary condition is applied by equating the known volumetric fluxes with their peridynamic equivalents.

The peridynamics bonds caused by either concentration or hydraulic gradient that cross a preexisting crack are assigned different micro properties, specifically zero micro diffusivity and zero micro hydraulic conductivity. Consider an insulating crack defined by the segment *AB* and peridynamics bonds between material points *x* and *x*` denoted by  $f_{xx}$ . If the line segment *AB* intersects the line segment xx`, the peridynamics bonds  $f_{xx}$ ` is set to zero; mathematically if  $(xA \times xx`) \cdot (xB \times xx`) < 0$  and  $(Ax \times AB) \cdot (Ax` \times AB) < 0$  then  $f_{xx`} = 0$ .

For modelling heterogeneous inclusions in the system (e.g. oil and rock segments), a subdomain is generated to represent the heterogeneity. The particles located inside the sub-domain are assigned with different properties (e.g. micro diffusivity and hydraulic conductivity). Therefore, different peridynamics bonds can be defined to describe the interaction between points of different phases. For example, the peridynamics bonds between the material points xand x are defined as

- $f_{rp}$ , the material points x and x` are located in rock and porous media, respectively.
- $f_{rr}$ , the material points x and x` are both located in rock domain.
- $f_{pr}$  the material points x and x` are located in porous media and rock, respectively
- and  $f_{pp}$ , the material points x and x`` are both located in porous media domain.

#### 4. Verifications

In this section, there are two verification examples including i) chemical transport in a saturated porous medium and ii) coupled water flow and chemical transport in unsaturated porous system. Chemical transport in a saturated porous medium will be first presented. The proposed model will be verified by an existing analytical solution for 1D problem. The problem is then extended to a coupled water flow and chemical transport in unsaturated porous media. The proposed PD solutions will be compared with alternative finite element analysis of the same problem.

#### 4.1. Chemical transport in a saturated porous medium

The verification tests presented in this section focused on testing the PD model for pure chemical transport in a fully saturated system. Two problems for which analytical solution exists are considered: Case 1, where neither diffusion nor advection is dominant; and Case 2, where the transport is advection dominated.

Diffusion and advection mechanisms for chemical flow are considered. A constant velocity of liquid is considered in the simulations. The length of domain is 10 cm. The initial concentration of chemical in the domain is considered to be zero. The boundary condition at x=10 cm is assumed to be zero flux boundary conditions (i.e. Neumann boundary condition). The boundary condition at the injection side (x=0) is maintained at  $C=C_0$ . In peridynamics approach,  $\Delta$  is set to be 0.01 cm and the horizon sizes of 0.03 cm is considered. The classical solution for this problem can be found in (Cleary and Ungs, 1978; Ogata and Banks, 1961)as:

$$C(x,t) = \frac{C_0}{2} \left[ \operatorname{erfc}\left(\frac{x - Vt}{2\sqrt{Dt}}\right) + \exp\left(\frac{Vx}{D}\right) \operatorname{erfc}\left(\frac{x + Vt}{2\sqrt{Dt}}\right) \right]$$
(35)

For case 1, the diffusion coefficient and advection are assumed to be  $D=1.0 \text{ cm}^2/\text{s}$  and V=1.0 cm/s, respectively. Figure 4 shows the chemical concentration profiles at 1 s, 2 s and 3 s, obtained by the proposed formulation (solid lines) and the results by the analytical solution (symbols). The simulation results with the PD formulation are clearly in excellent agreement with the analytical solution.



Fig. 4. Comparison between analytical solution and PD analysis (this study) for simulation case 1.

For case 2, the diffusion coefficient and advection are assumed to be  $D=0.01 \text{ cm}^2/\text{s}$  and V=1.0 cm/s, respectively. The Peclet number,  $Pe = \frac{VL}{D} = 1000 \gg 1$ . This smaller dispersivity or advection-dominated transport case is in principle difficult to solve as numerical oscillations develop when advection dominates. Nevertheless, the PD formulation is able to capture the transport successfully. The chemical concentration distributions at 1s, 2s and 3s obtained by the PD model and those obtained by the analytical solution are shown in Fig.5. It can be seen that the results are again in very good agreement with the analytical solution.



# Fig. 5. The comparison between analytical solution and PD analysis (this study) for advection-dominated transport

The verification examples demonstrate that a robust implementation of the theoretical formulation of chemical transport in the computational model has been achieved.

#### 4.2. 1D water flow and chemical transport in unsaturated porous media

The problem considered is the water flow and chemical transport in a one-dimensional domain of partially saturated medium with a length of 10 cm (L=10cm). The parameters used in the simulation of water flow and chemical transport are listed in Table 1.

	Parameter/coefficients	Value
$\theta_s(-)$	Volumetric water content at saturation	0.363
$\theta_r(-)$	Residual volumetric water content	0.186
$\alpha(m^{-1})$	van Genuchten's parameters	1
n (-)	van Gendenten s parameters	1.53
$K_s(m/s)$	Saturated hydraulic conductivity	10-9
D (m <sup>2</sup> /s)	Diffusion coefficient	9×10 <sup>-11</sup>

Table 1. Parameters for the simulations

The initial water content is assumed to  $\theta_r$  and the initial chemical concentration in the domain is considered to be zero. The boundary conditioned applied at *x*=10cm for chemical transport and water flow analysis are *C*=0 and  $\theta_l = \theta_s$ , respectively. The boundary condition at injection side (*x*=0) is maintained at *C*=*C*<sub>0</sub> and  $\theta_l = \theta_s$ .  $\Delta$  is 0.05 cm and a horizon size of 0.15 cm was used for the PD simulations. The PD simulation results (Fig.6) are compared with analysis of the same problem solved by Finite Element method which is implemented in Comsol Multiphysics with the element size of 0.05 cm. It can be seen from Fig.6 that there is a good agreement between PD solutions and FE solution indicating that the proposed PD formulations can be used to simulate the water flow in unsaturated porous medium.







(b)



(c)

Fig. 6. Comparison of Peridynamics solution against the numerical solution obtained using FE smulations (a) water saturation; (b) water velocity and (c) chemical concentration

#### 4.3. 2D water flow and chemical transport in unsaturated porous system

For the 2D simulation case, the domain is 10 cm long and 10 cm wide. The parameters of simulation used are listed in Table 1. The initial chemical concentration and water content in the domain are considered to be zero and  $\theta_r$ , respectively. The boundary condition at *x*=10cm is assumed to be zero flux (impermeable boundary). The boundary condition at the injection side (*x*=0) is divided into two areas. Area One is 4 cm wide at *x*=0cm (left boundary). The area is between coordinates (0, 3) and (0, 7). Zone Two covers the excluding area one at the left boundary of domain (Fig.7). At the injection boundary in area one the chemical concentration and moisture content are maintained at *C*=*C*<sub>0</sub> and  $\theta_l = \theta_s$ , respectively. The chemical concentration and moisture content in zone two are assumed to be *C*=0 and  $\theta_l = \theta_s$ , respectively (Fig.7).  $\Delta$  is 0.1 cm and a horizon size of 0.3 cm is assumed for the PD simulations. Similar to previous verification case, the PD results are compared with Finite Element analysis of a same problem. The same time step of  $\Delta t = 10^{-4}$  d is used for PD and FE.



Fig.7 Boundary conditions and initial conditions for water flow and chemical transport in a two-dimension porous media

The results of the saturation profile and chemical concentration profile along the centre line (x=5 cm) from the PD simulation are compared with those from FE solution at t=5, 10 and 15 days in Fig. 8. Fig.9 shows the comparison of peridynamics solution against the numerical solution obtained by FE analysis along x=0.02m. It can be seen that the results obtained by the proposed PD are in excellent agreement with the FE solution for 2D cases (Figs.8 and 9). A difference of saturation and concentration profile can be found in Fig.6 and Fig.8. The results in Fig.6 show that chemical migration is faster than water transport; however, the results in

Fig.8 show a contradicting trend. These phenomena may be explained by dilution effects and transversal dispersion. Moreover, the results in Fig.8 indicated a faster migration of chemical and water compared to the results observed in Fig.6. This is because the source of chemical and water for 1D problems is a point injection; however, the injection position for 2D problems ranges from (0, 3) and (0, 7).

The computational times required for the PD and FE analysis for this simulation case study using a I7-6200 CPUs is 16 min 38 sec and 8 min 9 sec, respectively. The computational time for the PD analysis compared to the FE is approximately twice (for this case) and this is due to the fact that the PD calculations involves analysis of interactions between a large number of collocation points (Fig.3). However, it is noted that the PD can provide solutions for discontinuous or strongly heterogenous problems without any extra requirements. This point will be further clarified in Section 6. Moreover, the PD solutions can readily be implemented in a parallel programming environment by which a significant reduction in run times can be expected.



(a)



Fig. 8. Comparison of Peridynamics solution against the numerical solution obtained by FE analysis along the centre line (a) saturation (b) chemical concentration



(a)



Fig. 9. Comparison of Peridynamics solution against the numerical solution obtained by FE analysis along x=2cm (a) saturation (b) chemical concentration

#### 5. Model application

In this section, we present three examples to show the capacity of PD models to capture the transport features in discontinuous and heterogeneous unsaturated porous media. A simulation case for coupled water flow and chemical transport in unsaturated porous media with multiple insulating cracks will be presented first. The case is then extended to consider propagating cracks. Finally, two case studies of analysis of the coupled water flow and chemical transport model in heterogeneous unsaturated porous media will be shown.

The domain is 10 cm long and 10 cm wide. The initial chemical concentration and moisture content in the domain are considered to be zero and  $\theta_r$ , respectively. The boundary at *x*=5 cm is assumed to be impermeable, i.e. natural (Neumann) boundary condition of zero fluxes. The boundary at the injection side (*x*=-5cm) is maintained at  $C=C_0$  and  $\theta_l = \theta_s$ , i.e. essential (Dirichlet) boundary condition with constant concentration and moisture for chemical transport and water flow, respectively. The properties of the porous medium and the parameters used for the simulations are listed in Table 1. The problem is analysed with horizon sizes  $\delta=0.15$  cm and  $\Delta = 0.05$  cm.

#### 5.1. Chemical transport in unsaturated porous media with insulating cracks

Solving the mathematical equation of transport processes with cracks and fractures using the classical local formulations is computationally challenging due to the difficulties in solving derivatives over discontinuities. The PD eliminates such difficulties by its non-local and integrating approach. In this section, water flow and chemical transport in a system with pre-existing cracks (discontinuity) are studied. The case study was designed to demonstrate the capability of proposed PD model in solving the transport processes in a discontinuous system with insulated cracks. A 2D domain with two single insulated vertical cracks of length 2cm is considered, as illustrated in Fig. 10. The implementation of peridynamics bonds that cross the crack is described in Section 4. The concentration distribution and saturation profile are shown in Fig. 10.



Fig. 10. PD results of saturation distribution (left) and concentration distribution (right) at 20 days.

It can be seen that PD models capture successfully the shielding effects of the cracks: both the water flow and the chemical concentration profile are strongly affected by the presence of cracks. Sharp jumps of concentration and saturation near the cracks can be observed. For example, the saturation at the points (0.01-,0.02) and (0.01+,0.02) is 0.94 and 0.62, respectively. The jumps are natural consequence of the zero peridynamics bonds caused by concentration gradient or hydraulic across the cracks. This saturation behaviour in the presence of cracks/fractures is challenging for the local methods including FEM and FDM, where the cracks must be explicitly modelled as narrow slits and meshed. This is because in the local formulations the constitutive parameters, such as diffusivity and conductivity, are volume-based and hence cannot be assigned to interfaces. For example, the concentration distribution and saturation profile shown in Fig. 11 is obtained by FEM. It can be found that geometry of the domain is divided into two sections. One is the porous medium and another one is the

insulating cracks which are separated and marked as an extra boundary. The insulating cracks are considered as a 0.3cm×2.5cm slits in FEM. A slight difference can be found between Fig.10 and Fig.11. This may be due to the different approaches adopted to represent the insulating cracks by PD and FEM. The crack is represented naturally with the broken bonds in PD; however, extra boundaries are introduced to form the cracks in FEM.



Fig. 11. FE results of saturation distribution (left) and concentration distribution (right) at 20 days.

#### 5.2. Chemical transport in unsaturated porous media with propagating cracks

In this section, the problem described in section 5.1. is extended to study a dynamic discontinuous system for which the length of discontinuities in the domain is enlarged with time. We did not aim to describe the mechanics of crack propagation in this work, but to explore the capability of the present PD model to solve such problems. The domain with two growing cracks, initiated at points (-0.02, -0.035) and (0.01, 0.035), is considered. The two cracks grow with a constant velocity (3 mm/day) as shown in Figure 12.

The concentration distribution and saturation profiles at 5d, 10d, 15d and 20d are shown in Figure 12. It can be seen that the effect of cracks increases from being practically insignificant at 5 days to very significant at 15 days, where strong discontinuous profiles of saturation and concentration near the crack at x=0.1cm are observed. The distribution profiles of saturation and concentration are orthogonal to the crack surfaces. The shielding effect induced by the growing cracks is correctly captured by the peridynamic solution. A nonnegligible difference can be noticed between Fig.10 and Fig.12 at 20 days. A faster transport phenomenon is observed in Fig.10. A sharper jump of concentration and saturation near the second crack is observed in Fig.12 compared to the results in Fig.10. For example, the saturation difference

along the cracks can reach 0.32 in Fig.10; however, this value increases to 0.68 in Fig.12. This is due to the fact that a larger crack with a length of 6cm is formed in Fig.12 compared to the length of 2.5cm in Fig.10. The larger cracks show stronger shielding effects. Notably, water flow and chemical transport through a system of propagating cracks is even more challenging for local methods, where adaptive mesh refinement may be required to track the crack extension.

The examples provided in sections 5.1 and 5.2 demonstrate the ability of the proposed PD formulation to handle complex static and dynamic discontinuous systems. The PD models deal with the problem naturally. PD-based extension to include the mechanical and geochemical reaction processes for a fully coupled hydro-chemical- mechanical model is currently under development.







#### 5.3. Coupled water flow and chemical transport in heterogeneous system

The effects of heterogeneous system on water flow and distribution of chemicals are studied in this section. The case studies were designed to demonstrate the capability of proposed PD model in solving the transport processes in heterogeneous media which has permeable or impermeable inclusions. For this purpose, a 2D domain with randomly distributed circular inclusions is considered, as illustrated in Fig. 13.


Fig.13. Boundary conditions and initial conditions for water flow and chemical transport in the heterogeneous medium

In order to assign impermeable inclusions, the diffusion coefficient and hydraulic conductivity are assumed to be zero for the inclusions. Figure 14 shows distributions of degree of saturations and concentration of chemicals in the domain at t=40 days. The grey circles indicate the location of inclusions. The chemical transport and water flow are restricted only to the gaps between the inclusions. The PD model has successfully analysed the discontinuity in the domain. The chemical concentration and saturation of the impermeable circles maintain C=0 and  $S_r = 0$ , which are equal to the initial values. This shows that the implementations of PD bonds for heterogeneous system are able to solve the transport phenomena with strong impermeable inclusions. It is shown that migration of chemical and water is impeded by the existence of impermeable inclusions. For example, there is no chemical transport distance of water and chemical is observed at y=0.05m. It can be found that y=0.05m is the furthest away from the impermeable inclusions so the shielding effects is the weakest at this line and a faster transport is observed. The results indicated that the existence of impermeable inclusions slows down the transport of chemical and water.



Fig. 14. Saturation distribution (left) and concentration distribution (right) for impermeable circular inclusions at 40d

A similar domain is analysed but permeable inclusions are prescribed which have smaller diffusion and hydraulic conductivity coefficients than the rest of the domain (matrix). To prescribe the diffusion coefficient and hydraulic conductivity we use a random number (from a uniform distribution) ranging between (0, 1) for each inclusion. The black dashed circles indicate the location of permeable inclusions. Figure 15 shows the concentration distribution and degree of saturation in the domain at t=40 days. The results show that the randomly assigned diffusion coefficients and hydraulic conductivity lead to different levels of penetrations in different inclusions. For example, the back diffusion is found in the circle on the left-hand side. The diffusivity of chemical in porous media is larger than that in the inclusion, which leads to a faster transport of chemical in porous media. However, the concentration difference between porous media and permeable inclusions results in the back-diffusion phenomena observed in Figure 15.



Fig. 15. Saturation distribution (left) and concentration distribution (right) for permeable circular inclusions (with smaller diffusion coefficients and hydraulic coefficients) at 40d

It is noteworthy that the water saturation is almost 1 in one circle and two ellipses on the lefthand side of the model, while concentration is almost zero in these structures. This observation can be explained by the following two points. One is that water transport is faster compared to chemical migration. It is governed by the input diffusion coefficients and hydraulic conductivity. The same trends are observed in Figs.8, 10,11,12 and 14. Therefore, water will be saturated in porous media in a shorter time than that of chemical and permeate into the inclusions. Another point is that the randomly assigned hydraulic conductivity in the inclusions is relatively large. It leads to a faster penetration in the inclusions. The penetrations distance of water and chemical is larger in Fig.15 than that in Fig.14. For example, the penetration distance of chemical is around 6.5cm and 7cm at y=5cm in Fig.14 and Fig.15m, respectively. This shows that the impacts of inclusions are governed by the hydraulic conductivity and diffusivity of the inclusions. A smaller hydraulic conductivity and diffusivity of the inclusions will show a more significant blocking effect. The results from Figs.14 and 15 demonstrate how the inclusion in overall control of transport processes.

## **5.4.** Discussion

Heterogeneities such as permeable/impermeable inclusions and cracks in a porous medium result in discontinuities in the concentration and flow field (see Figs.10-15). The classical local model faces difficulties simulating such problems since spatial derivatives fail at the discontinuities. In classical approach such as FEM and FVM, special interface/boundary conditions must be introduced to satisfy conservation principles (Fig.11). However, in non-local peridynamics model, transport equations are valid everywhere, regardless of the presence of material discontinuities/heterogeneities. This makes nonlocal models naturally applicable to sharp interface problems without any additional interface conditions.

The heterogeneities/discontinuities of porous media are defined by using different bonds across them without any additional modification to the transport formulations or any extra computational expense. The results from Fig.10 show a jump of saturation and concentration around the discontinuous cracks region, which demonstrate that the discontinuous features in the domain are captured by the peridynamic model. For a dynamic crack growth porous media (Fig.12), the PD shows a strong ability to capture the features induced by the propagating cracks without any refinements and modifications. It further demonstrates the advantages of PD for solving heterogeneities/discontinuities in porous media.

Important insights toward transport in heterogeneities porous media are obtained in section 5.3. The simulations in Fig.15 shows the back-flow and back-diffusion phenomena which are caused by the existence of permeable inclusions, which promote the appearance of fast paths as well as regions of hold up. The extra modifications, such as stochastic approaches and continuum-scale dual porosity, are required to capture these phenomena by using the local method; in contrast, the non-local PD models are naturally applicable to such problems and capable to capture these abnormal phenomena. Development of multi-chemical reactive transport in discontinuous porous media, which is induced by stress or geochemical reaction, is of interest for future work.

## 6. Conclusions

Bond-based non-local formulations of water flow and chemical transport in porous media are proposed. Water flow is partially coupled with chemical transport by an implicit numerical scheme to allow for solving coupled problems of practical importance.

The formulations are implemented within a computational framework (Pyramid) and verified against a set of benchmarks. The verifications demonstrate excellent agreements between the proposed approach and the classical/local formulations for homogeneous media.

Extended capability of the model is demonstrated by analysis of coupled chemical transport and water flow problem in 2D domains with stationary and propagating cracks and various randomly distributed inclusions. The simulations illustrate how the model can capture naturally the effects of static and dynamics discontinuities and heterogeneities, which is problematic or computationally demanding for numerical schemes based on local formulations.

Overall, it is demonstrated that the proposed nonlocal formulation-implementation framework is efficient and effective in analysis of coupled water flow and chemical transport problems. The model is applicable to multi-physics problems in porous systems where coupled hydrochemical degradations can create discontinuity or heterogeneity. The addition of mechanical deformation, fracture nucleation and erosion is a subject of ongoing work.

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## Appendix A

# Classical (local) formulations of water flow and chemical transport in unsaturated porous media

The formulation of coupled water flow and chemical transport presented is based on mass conservation. The chemical transport is governed by diffusion and advection processes, where advection term is induced by coupled water transport. Water transport is described by Richard's equation, which is not affected by concentration field. The transition to the PD formulation requires a brief description of the local formulations in the classical theories. These are presented in the following two subsections.

## A1.1 Flow of liquid (water) phase

Water flow is considered as liquid water flow (no vapour flow), and described by Richard's equation. The water flow is considered at ambient temperature and vapour transfer is neglected due to its limited effect on the overall flow. Richard's equation provides a sound theoretical description of moisture flow at ambient isothermal flow (Simunek et al., 1999). The mass conservation for liquid water in porous medium, using classical theory, is (Richards, 1931):

$$\frac{\partial \rho_l \theta_l}{\partial t} + \nabla \cdot [\rho_l q(\theta_l)] = 0 \tag{A-1}$$

where,  $\theta_l$  is the volumetric liquid content,  $q(\theta_l)$  is the liquid flux,  $\rho_l$  is the density of liquid.

The liquid flux is considered via Darcy's law as:

$$q(\theta_l) = -K(\theta_l)\nabla h(\theta_l) \tag{A-2}$$

where,  $K(\theta_l)$  is the hydraulic conductivity which is a function of volumetric liquid content and  $h(\theta_l)$  is the hydraulic or total water potential. We adopt the van Genuchten model (Van Genuchten, 1980) to describe the relationship between the total hydraulic or water potential and the water content and also for calculating the unsaturated hydraulic conductivity. The model is given as follow:

$$\theta_l(h_m) = \theta_r + \frac{\theta_s - \theta_r}{\left[1 + (\alpha |h_m|^n)\right]^{\frac{n-1}{n}}}$$
(A-3)

$$K(\theta_l) = K_s \left(\frac{\theta_l - \theta_r}{\theta_s - \theta_r}\right)^{0.5} \left\{ 1 - \left[1 - \left(\frac{\theta_l - \theta_r}{\theta_s - \theta_r}\right)^{\frac{n}{n-1}}\right]^{\frac{n-1}{n}} \right\}^2$$
(A-4)

$$h_m(\theta_l) = -\frac{1}{\alpha} \left[ \left( \frac{\theta_s - \theta_r}{\theta_l - \theta_r} \right)^{\frac{n}{n-1}} - 1 \right]^{\frac{1}{n}}$$
(A-5)

where,  $\theta_s$  and  $\theta_r$  are the saturation and residual volumetric water content respectively. Also,  $K_s$  is the soil hydraulic conductivity at saturation,  $\alpha$  is related to the soil pore size distribution.

According to the van Genuchten model of water retention behaviour, the effective saturation is defined as follow:

$$S_r = \frac{\theta_l - \theta_r}{\theta_s - \theta_r} \tag{A-6}$$

## A1.2 Chemical transport

The transport formulation considers advective, diffusive, and dispersive mechanisms of chemical transport. The formulation of the chemical transport in unsaturated porous media can be presented as (Sedighi et al., 2015; Thomas et al., 2012):

$$\frac{\partial \theta_l C}{\partial t} + \nabla \cdot (q(\theta_l)C - D_e \nabla C - D_m \nabla C) = 0$$
(A-7)

where, *C* is the concentration of chemical in the liquid phase;  $D_e$  is the effective molecular diffusion coefficient;  $D_m$  is the matrix of the effective dispersion coefficients. For the sake of simplification in presenting the formulation, the effective molecular diffusion and effective dispersion coefficients are combined (i.e.  $D = D_e + D_m$ ) to represent combined effects of physical processes of molecular diffusion and dispersion migration of chemicals. The liquid flux in the second term of Eq. (A-7) can be replaced by Eq. (A-2) while the first term is expanded to its derivatives:

$$\theta_l \frac{\partial C}{\partial t} + C \frac{\partial \theta_l}{\partial t} = D\nabla \cdot [\nabla C] - C\nabla q(\theta_l) - q(\theta_l)\nabla C$$
(A-8)

If the liquid phase is considered to be incompressible (i.e.  $\rho_l$  is constant), Eq. (A-8) can be simplified by using Eq. (A-1), given as:

$$\theta_l \frac{\partial c}{\partial t} = D\nabla \cdot [\nabla C] - q(\theta_l)\nabla C \tag{A-9}$$

## **Appendix B**

## Steps for development of a peridynamic model for 1-D chemical diffusion in porous media

Considering a 1-D domain (peri-pipe element) with a length of l, the two sides of element are assigned with different concentrations; C and C. If we define  $C_a$  as the average concentration in the domain, then the rate of mass change in the pipe is

$$l\frac{\partial C_a}{\partial t} \tag{B-1}$$

The chemical flux in the domain can be expressed as

$$\frac{D(C-C^{*})}{l} \tag{B-2}$$

where, *D* is the diffusivity.

The mass conservation yields:

$$\frac{\partial c_a}{\partial t} = \frac{D(C - C)}{l^2} \tag{B-3}$$

In peridynamic approach, the material points are connected through the mass bonds. For example, the material point x interacts with (and is connected to) all materials points x` within the horizon of  $H_x$  (Figs.1 and 3). In order to construct the peridynamic formulations of diffusion, Eq. (B-3) can be written as follow:

$$\frac{\partial C_a(x,t)}{\partial t} = D(x,x',t) \frac{C(x',t) - C(x,t)}{\|x - x'\|^2}$$
(B-4)

where, D(x, x', t) is the diffusivity of the m-bond.

Integrating over the horizon of points x on both sides of Eq.(B-4) gives

$$\int_{H_x} \frac{\partial C_a(x, x', t)}{\partial t} dx' = \int_{H_x} D(x, x', t) \frac{C(x', t) - C(x, t)}{\|x - x'\|^2} dx'$$
(B-5)

where, D(x, x', t) is the diffusivity of the m-bond.

By substituting Eqns. (6) and (8) into Eq. (B-5), the bond-based Peridynamics formulation for chemical diffusion x can be expressed in the following form:

$$\frac{\partial C(x,t)}{\partial t} = \int_{H_x} d(x, x', t) \frac{C(x',t) - C(x,t)}{\|x - x'\|^2} dx'$$
(B-6)

The determination of peridynamic micro-diffusivity, d(x, x', t), is provided in Section 2.3.

Appendix C

$$\begin{bmatrix} H(x_{i+3},t^{n})-H(x_{i},t^{n}) \\ H(x_{i+2},t^{n})-H(x_{i},t^{n}) \\ H(x_{i+1},t^{n})-H(x_{i},t^{n}) \\ H(x_{i-1},t^{n})-H(x_{i},t^{n}) \\ H(x_{i-2},t^{n})-H(x_{i},t^{n}) \\ H(x_{i-3},t^{n})-H(x_{i},t^{n}) \\ C(x_{i+3},t^{n})-C(x_{i},t^{n}) \\ C(x_{i+1},t^{n})-C(x_{i},t^{n}) \\ C(x_{i-2},t^{n})-C(x_{i},t^{n}) \\ C(x_{i-2},t^{n})-C(x_{i},t^{n}) \\ C(x_{i-3},t^{n})-C(x_{i},t^{n}) \end{bmatrix}$$
(C-1)

$$\begin{bmatrix} \mathbf{K} \end{bmatrix} = \begin{bmatrix} k_{l+3} & k_{l+2} & k_{l+1} & k_{l-1} & k_{l-2} & k_{l-3} & 0 & 0 & 0 & 0 & 0 \\ k_{\nu+3} & k_{\nu+2} & k_{\nu+1} & k_{\nu-1} & k_{\nu-3} & k_{\nu-3} & k_{d+3} & k_{d+2} & k_{d+1} & k_{d-1} & k_{d-2} & k_{d-3} \end{bmatrix}$$
(C-2)

$$k_{l+3} = \frac{\kappa_{(i)(i+3)}V_{i+3}}{|\boldsymbol{x}_{i+3} - \boldsymbol{x}_i|^2}$$
(C-3)

$$k_{l+2} = \frac{\kappa_{(i)(i+2)}V_{i+2}}{\left|\boldsymbol{x}_{i+2} - \boldsymbol{x}_{i}\right|^{2}}$$
(C-4)

$$k_{l+1} = \frac{\kappa_{(i)(i+1)} V_{i+1}}{\left| \boldsymbol{x}_{i+1} - \boldsymbol{x}_{i} \right|^{2}}$$
(C-5)

$$k_{l-1} = \frac{\kappa_{(i)(i-1)}V_{i-1}}{|\boldsymbol{x}_{i-1} - \boldsymbol{x}_i|^2}$$
(C-6)

$$k_{l-2} = \frac{\kappa_{(i)(i-2)} V_{i-2}}{\left| \boldsymbol{x}_{i-2} - \boldsymbol{x}_{i} \right|^{2}}$$
(C-7)

$$k_{l-3} = \frac{\kappa_{(i)(i-3)}V_{l-3}}{|\boldsymbol{x}_{l-3} - \boldsymbol{x}_{l}|^{2}}$$
(C-8)

$$k_{\nu+3} = k_{l+3} \Big[ C(x_{l+3}, t^n) - C(x_l, t^n) \Big]$$
(C-9)

$$k_{v+2} = k_{l+2} \Big[ C(x_{i+2}, t^n) - C(x_i, t^n) \Big]$$
(C-10)

$$k_{\nu+1} = k_{l+1} \Big[ C(x_{i+1}, t^n) - C(x_i, t^n) \Big]$$
(C-11)

$$k_{\nu-1} = k_{l-1} \Big[ C(x_{l-1}, t^n) - C(x_l, t^n) \Big]$$
(C-12)

$$k_{v-2} = k_{l-2} \Big[ C(x_{i-2}, t^n) - C(x_i, t^n) \Big]$$
(C-13)

$$k_{v-3} = k_{l-3} \Big[ C(x_{i-3}, t^n) - C(x_i, t^n) \Big]$$
(C-14)

$$k_{d+3} = \frac{d_{(i)(i+3)}V_{i+3}}{|\mathbf{x}_{i+3} - \mathbf{x}_i|^2}$$
(C-15)

$$k_{d+2} = \frac{d_{(i)(i+2)}V_{i+2}}{\left|\boldsymbol{x}_{i+2} - \boldsymbol{x}_{i}\right|^{2}}$$
(C-16)

$$k_{d+1} = \frac{d_{(i)(i+1)}V_{i+1}}{\left|\boldsymbol{x}_{i+1} - \boldsymbol{x}_{i}\right|^{2}}$$
(C-17)

$$k_{d-1} = \frac{d_{(i)(i-1)}V_{i-1}}{\left|\boldsymbol{x}_{i-1} - \boldsymbol{x}_{i}\right|^{2}}$$
(C-18)

$$k_{d-2} = \frac{d_{(i)(i-2)}V_{i-2}}{\left|\boldsymbol{x}_{i-2} - \boldsymbol{x}_{i}\right|^{2}}$$
(C-19)

$$k_{d-3} = \frac{d_{(i)(i-3)}V_{i-3}}{\left|\boldsymbol{x}_{i-3} - \boldsymbol{x}_{i}\right|^{2}}$$
(C-20)

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# CHAPTER Four

# 4. Modelling the soil desiccation cracking by peridynamics

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## 4.1 Paper introduction:

In this paper, a study on clay desiccation cracking by developing a new model based on nonlocal theory of Peridynamics was presented. The paper presented, for the first time, a non-local PD formulation for crack initiation and development in clays induced by desiccation. The developed model was used to address the challenge of modelling the clay desiccation cracking which involves a strong degree of coupling between hydraulic and mechanics and more importantly the strong mathematical discontinuity that exists as the result of fracture development in the system. The approach presented in this manuscript was a fundamental development in hydro-mechanical modelling of geotechnical engineering problems that involve strong, random and evolving discontinuity.

The developed model was applied and tested against two sets of experimental data to explain the typical features of drying-induced cracking of clays. We demonstrated the correlations between the shrinkage of soil clay, changes in displacement fields and crack growth. Crack initiation, propagation and ultimate crack patterns simulated by the model were found to be in very good agreement with experimental observations. The results showed that the model can capture realistically key hydraulic, mechanical and geometry effects on clay desiccation cracking.

## 4.2 Contribution to the joint authorship

The author has contributed the following:

- Development of the main concept for this paper (80%).
- Theoretical developments for clay cracking (90%).
- Model implementations and code writing (100%).
- Validations and applications of the proposed model (90%).
- Preparation of tables and data visualisation (100%).

• Paper writing (80%).

## 4.3 Supplemental information

- S1: Derivation of micro-modulus function
- S2: Derivation of microscopic diffusivity function
- S3. Weakly coupled approach with multi-rate integration

## Addendum

Page 136 and 137, Figures 3 and 4 in the published paper: The relative difference ( $\varepsilon$ ) is used to show the validity of the present numerical model. The relative difference between the hoop stress in Fig.3 and Fig.4 from experimental data and PD is 4.39 and 0.005 respectively.

## This Chapter is an exact copy of the accepted format manuscript referred to above.

# Modelling the soil desiccation cracking by peridynamics

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#### Abstract

Understanding the desiccation-induced cracking in soil has been improved over the last 20-30 years through experimental studies but the progress in predictive modelling of desiccation cracking has been limited. The heterogeneous structure of soils and the multi-physics nature of the phenomenon, involving emergence and propagation of discontinuities, make the mathematical description and analysis a challenging task. We present a non-local hydromechanical model for soil desiccation cracking capable of predicting crack initiation and growth. The model is based on the peridynamics (PD) theory. Attempts to model the soil desiccation cracking by PD are limited to a purely mechanical description of the process that involves calibration of the parameters. Differently, the model presented in this paper describes soil desiccation cracking as a hydro-mechanical problem, where moisture flow and deformation are coupled. This allows for investigating and explaining the mechanisms controlling the initiation and propagation of discontinuities. The model is applied and tested against two sets of experimental data to explain the typical features of drying-induced cracking of clays. The validations use experimental parameters (Young's modulus, water retention characteristics) and avoid calibrations to test the accuracy of the model. We demonstrate the correlations between the shrinkage of soil clay, changes in displacement fields and crack growth. Crack initiation, propagation and ultimate crack patterns simulated by the model are found to be in very good agreement with experimental observations. The results show that the model can capture realistically key hydraulic, mechanical and geometry effects on clay desiccation cracking.

Keywords: Soil desiccation, peridynamics, coupled problems, shrinkage cracking

## **1. Introduction**

Soil desiccation is characterised by loss of moisture which can lead to shrinkage and potentially to cracking. Describing and analysing this phenomenon is challenging because it involves strongly coupled processes of moisture flow, deformation, emergence and evolution of cracks in a complex, heterogeneous soil structure, but is important for many applications. For example, desiccation accelerates infiltration in road embankments and soil slopes (Jamalinia et al.,2020) and causes failures of anti-seepage landfill system (Wan et al., 2018). Further, desiccation cracking can damage local ecologies as cracks increase the surface roughness and accelerate evaporation in soil, contributing to faster soil weathering and erosion leading to potential dust storms, landslides and debris flow (Zeng et al., 2020). Additionally, desiccation cracks can create pathways for water and gas transport, which will reduce the performance of clay barriers used for waste isolation (Wan et al., 2018; DeCarlo and Shokri, 2014).

The understanding of desiccation cracking in clay soils has progressed in the last few decades through experimental investigations and theoretical developments. Desiccation has been investigated in slurries (Li and Zhang, 2011), clay soils (Tang et al., 2011; Wei et al., 2016) and starch water mixture cakes (Kodikara et al., 2004). Desiccation cracking experiments have been focused on characterising the cracks' geometries including depth, thickness, spacing and aperture of the cracks (Najm et al., 2009; Péron et al., 2007 & 2009a; Wan et al., 2018; Amarisiri et al., 2014; Zhao et al., 2020; Xu et al., 2020). The results of these studies have contributed to understanding of the processes, but their relevance is limited to the specific laboratory conditions (Tran et al., 2019). Predicting desiccation cracking under field conditions requires physically realistic models that capture the complex and coupled processes involved. The aim of this paper is to address the need for realistic modelling of soil desiccation.

Numerical methods for solving continuum physical or mechanical problems, such as the finite element method (FEM), cannot capture crack initiation without the introduction of special elements. These are typically cohesive elements inserted at the interfaces between continuum elements. They require specific calibration of their behaviour and predetermine crack initiation locations and paths (Sánchez et al., 2014; Vo et al., 2017; Wang et al., 2020). Modelling crack propagation also requires specific calibration of a crack extension criterion, which is expensive and typically not universally valid. For example, calibration with experimental data for one specific stress state may not correctly predict the behaviour for other stress states. Some of the difficulties of FEM-based methods can be avoided by discrete methods, such as lattice methods

(Zhang and Jivkov, 2016; Morrison et al., 2016; Dassios et al., 2018), lattice spring methods (Zhu et al., 2020) and discrete element methods (Amarasiri and Kodikara, 2013; Sima et al., 2014; Tran et al., 2020). Alternative approaches, including phase-field methods (Cajuhi et al., 2018) and smoothed-particle hydrodynamics (Tran et al., 2019), have also been adopted for soil desiccation problems. However, these non-FEM-based methods inherit a requirement from FEM – introduction and calibration of failure initiation and propagation criteria – dictated by the local (differential) nature of the underlying governing equations (Wang et al., 2017 & 2019).

Nonlocal formulations have been developed to address problems with discontinuities, large deformations and heterogeneities (Eringen and Edelen, 1972; Bažant and Chang, 1984). A recent non-local theory (Silling, 2000), referred to as the Peridynamics (PD), is receiving increasing interest among the mechanics community, as it overcomes the limitations of the classical theory particularly for problems with discontinuities. PD avoids calculations of spatial derivatives, which are mathematically undefined at sharp discontinuities such as cracks or shocks (Kilic and Madenci, 2009a; Oterkus et al, 2014). This allows for developing PD formulations for all physical phenomena beyond the original applications to solids mechanics proposed by Silling (2000). Such formulations remain mathematically consistent irrespective of the emergence and evolution of discontinuities. The PD theory has been applied in different areas, including fluid driven fractures (Ouchi et al., 2015; Zhou et al., 2020), thermal fracture (Oterkus et al., 2014; Wang et al., 2018b; Kilic and Madenci, 2009b), clay erosion (Sedighi et al., 2021) and pitting corrosion damage (Jafarzadeh et al., 2019).

This paper presents the development of PD formulations for coupled hydro-mechanical processes to predict the desiccation cracking of soils. In Section 2 we present our PD formulations for water flow and mechanical deformation, including crack initiation and growth. In Section 3 we present validations by comparison of our predictions with three different soil desiccation tests and discuss the results. Conclusions are summarised in Section 4.

## 2. Hydro-mechanical formulations of soil desiccation cracking

### 2.1. Classical (local/differential) formulation of soil deformation induced by drying

Soil desiccation results from the loss of moisture. As the water content in clay decreases, the suction increases (total soil water potential decreases), leading to soil shrinkage. This is appropriately described by Biot's poroelasticity (Biot, 1941), which relates the stresses, strains

and fluid mass or pore pressure change (Péron et al., 2013). The total strain increment is

$$\mathrm{d}\varepsilon_{ij} = \mathrm{d}\varepsilon_{ij}^m + \mathrm{d}\varepsilon_{ij}^h \tag{1}$$

where  $\varepsilon_{ij}$  are the components of the total strain tensor,  $\varepsilon_{ij}^m$  are the components of the elastic strain tensor work-conjugate to the mechanical stresses, and  $\varepsilon_{ij}^h$  are the components of a strain tensor arising from water content changes (hydration/dehydration) and referred to as the hydric strain tensor. This is volumetric, i.e.,  $\varepsilon_{ij}^h = 0$ ,  $i \neq j$ , irrespective of coordinate system selection.

The mechanical stress increment is

$$d\sigma_{ij} = D_{ijkl} d\varepsilon_{kl}^m = D_{ijkl} (d\varepsilon_{kl} - d\varepsilon_{kl}^h)$$
<sup>(2)</sup>

where  $D_{ijkl}$  are the components of the elasticity tensor of the solid material.

Assuming the response of soil to changes in water content is isotropic, the hydric strain tensor increment can be expressed by (Péron et al., 2009a)

$$\mathrm{d}\varepsilon_{kl}^{h} = \delta_{kl}\alpha\mathrm{d}\theta \tag{3}$$

where  $\delta_{kl}$  is the Kroneker delta,  $d\theta$  represents the water content change, and  $\alpha$  is a shrinkage coefficient, which can be represented as a function of void ratio *e* and specific gravity  $G_s$  of the soil (Péron et al.,2009a)

$$\alpha = \frac{G_s}{1+e} \tag{4}$$

We assume that drying by moisture loss is a diffusion-like phenomenon. We neglect the effects of mechanical deformation on the water transport due to the small mechanical strains involved (Péron et al., 2009b). The mass conservation of water is described by Yan et al. (2020)

$$\frac{\partial\theta}{\partial t} = \nabla \cdot [D\nabla\theta] \tag{5}$$

where, t is time and D is the diffusion coefficient of water in soil.

Three distinct stages can be identified during soil desiccation (Péron et al., 2009b; Tang et al., 2011). Stage I is characterised by shrinkage but the soil remains saturated. In Stage II air gradually replaces moisture in the soil pore system but the shrinkage is smaller than in Stage I. Stage III is characterised by continued loss of moisture, but without further shrinkage. We will limit our desiccation model to Stage I (Maarry et al., 2012) where significant shrinkage is expected. The implication of this decision is that the diffusion coefficient of water will be

considered as constant.

### 2.2. Peridynamic formulations of coupled deformation and moisture flow

In peridynamics, a body occupying a region  $\mathbf{R}$  is considered to be a collection of peridynamic particles, which can be placed in a regular grid, as illustrated in Fig. 1(a), or in any other required arrangement. One of the two classical PD approaches – the bond-based PD, which we utilise here – represents the interactions between the peridynamic particles by 'bonds'. A particle at position  $\mathbf{x}$  interacts with (is connected to) all particles at positions  $\mathbf{x}'$  within a certain finite region  $\mathbf{H}_{\mathbf{x}}$ , called the horizon of the particle at  $\mathbf{x}$ . The horizon radius is denoted by  $\delta$ . Further,  $\rho(\mathbf{x})$  and  $\theta(\mathbf{x})$  denote the soil density and water content at position  $\mathbf{x}$ , respectively. The bond vector between particles at  $\mathbf{x}$  and  $\mathbf{x}'$  is  $\boldsymbol{\xi} = (\mathbf{x}' - \mathbf{x})$ , the change of water content along this bond is  $\zeta = \theta(\mathbf{x}') - \theta(\mathbf{x})$ , and the average bond water content is  $\overline{\theta} = [\theta(\mathbf{x}') + \theta(\mathbf{x})]/2$ .



Figure 1. Illustration of interaction between peridynamic particles during deformation: (a) Reference configuration; and (b) Deformed configuration.

As the body deforms, a particle at x is displaced by u(x) and the relative displacement between particles at x and x' is  $\eta = u(x') - u(x)$ . The deformed configuration is illustrated in Fig. 1(b). Note, that  $\eta + \xi$  represents the current distance vector between particles. With these notations the quantities  $\epsilon$  and n represent a micro-strain (relative change of bond length) and a unit vector along the bond between particles at x and x' in the deformed configuration, respectively:

$$\epsilon = \frac{\|\boldsymbol{\eta} + \boldsymbol{\xi}\| - \|\boldsymbol{\xi}\|}{\|\boldsymbol{\xi}\|} \tag{6}$$

$$\boldsymbol{n} = \frac{\boldsymbol{\eta} + \boldsymbol{\xi}}{\|\boldsymbol{\eta} + \boldsymbol{\xi}\|} \tag{7}$$

The micro-strain includes mechanical and hydric contributions, so that  $\epsilon = \epsilon_m + \epsilon_h$ , where  $\epsilon_h = \alpha \bar{\theta}$ .

The peridynamic version of the conservation of linear momentum is the equilibrium of forces acting on a particle at position x:

$$\rho(\boldsymbol{x})\ddot{\boldsymbol{u}}(\boldsymbol{x}) = \int_{H_{\boldsymbol{x}}} \boldsymbol{f}(\boldsymbol{\xi},\boldsymbol{\eta}) \, \mathrm{d}V_{\boldsymbol{x}} + \boldsymbol{b}(\boldsymbol{x}) \tag{8}$$

where  $\ddot{u}(x)$  is the acceleration of the particle at x,  $f(\xi, \eta)$  is a pairwise force density vector representing the interaction of the particle at x with the particles in its horizon,  $V_x$  is the horizon volume (area in 2D, length in 1D) of the particle at x, and b(x) is a body force on the particle at x. A micro-elastic material (Silling 2000) is defined by postulating a micro-elastic potential,  $\omega(\xi, \eta)$ , from which the pairwise force density is derived by

$$f(\boldsymbol{\xi},\boldsymbol{\eta}) = \frac{\partial \omega(\boldsymbol{\xi},\boldsymbol{\eta})}{\partial \boldsymbol{\eta}}$$
(9)

We consider a linear micro-elastic material, for which the micro-elastic potential is a quadratic function of the mechanical micro-strain, similarly to continuum elasticity

$$\omega(\boldsymbol{\xi}, \boldsymbol{\eta}) = \frac{1}{2} C_p(\boldsymbol{x}, \boldsymbol{x}') \epsilon_m^2$$
(10)

where  $C_p(\mathbf{x}, \mathbf{x}')$  is a peridynamic micro-modulus. Considering that  $\epsilon_m = \epsilon - \alpha \bar{\theta}$ , this gives the following expression for the force density vector

$$\boldsymbol{f}(\boldsymbol{\xi},\boldsymbol{\eta}) = C_p(\boldsymbol{x},\boldsymbol{x}') \left(\boldsymbol{\epsilon} - \boldsymbol{\alpha} \ \bar{\boldsymbol{\theta}}\right) \boldsymbol{n}$$
(11)

The relationships between  $C_p(x, x')$  and the macroscopic material properties have been proposed by Oterkus et al. (2014); for 2D problems considered here this reads:

$$C_p(\mathbf{x}, \mathbf{x}') = \frac{9E}{\pi\delta^3} \tag{12}$$

where E is the Young's modulus of the soil; details of derivation of Eq. (12) are provided in supplementary material (S1).

The peridynamic version of the conservation law for water content at position x is given by (Yan et al., 2020)

$$\frac{\partial \theta(\mathbf{x})}{\partial t} = \int_{H_{\mathbf{x}}} g(\boldsymbol{\xi}, \boldsymbol{\zeta}) \, \mathrm{d} V_{\mathbf{x}}$$
(13)

where,  $g(\boldsymbol{\xi}, \boldsymbol{\zeta})$  is the pairwise mass exchange function. This is given by

$$g(\boldsymbol{\xi},\boldsymbol{\zeta}) = D_p(\boldsymbol{x},\boldsymbol{x}') \frac{\boldsymbol{\zeta}}{\|\boldsymbol{\xi}\|^2}$$
(14)

where  $D_p(\mathbf{x}, \mathbf{x}')$  is a peridynamic micro-diffusivity (micro-transport coefficient). The relation between  $D_p(\mathbf{x}, \mathbf{x}')$  and the macroscopic diffusivity D for 2D moisture flow is given by Yan et al. (2020) and Sedighi et al. (2021)

$$D_p(\boldsymbol{x}, \boldsymbol{x}') = \frac{4D}{\pi\delta^2} \tag{15}$$

Details of derivations of Eq. (15) are provided in supplementary material (S2).

## 2.3. Failure initiation and damage parameter

The bond between particles at  $\mathbf{x}$  and  $\mathbf{x}'$  can be equipped with an indicator function  $\mu(\mathbf{x}, \mathbf{x}')$  that encodes the state of the bond as either being intact or broken (Wang et al., 2018a; Menon and Song, 2019). Mathematically, the indicator ( $\mu(\mathbf{x}, \mathbf{x}')$ ) is a Heaviside step function of the difference between a critical value of the micro-strain,  $\epsilon_f$ , and the mechanical micro-strain in the bond  $\epsilon_m$ , as follow:

$$\mu(\mathbf{x}, \mathbf{x}') = \begin{cases} 1 & \epsilon_m < \epsilon_f \\ 0 & \epsilon_m \ge \epsilon_f \end{cases}$$
(16)

This means that a bond is broken when the micro-strain exceeds the critical value. The critical micro-strain  $\epsilon_f$  can be related to the fracture energy release rate of the material  $G_f$ , measured in N/m, which is the energy required for creating a unit of new surface area (Silling and Askari, 2005; Lehoucq and Silling, 2008). For example, the critical micro-strain for plane stress problem is related to the energy release rate by

$$\epsilon_f = \sqrt{\frac{4 \ G_f}{9E\delta}} \tag{17}$$

Failure of bonds associated with particle at x redistributes the forces on this particle to the remaining intact bonds within its horizon (Gu et al., 2017). The change experienced by the particle can be described by a damage parameter  $\varphi(x)$ , defined as the ratio of the number of broken bonds and the number of all initially present bonds in its horizon (Kilic and Madenci, 2009a and 2009b)

$$\varphi(\mathbf{x}) = \frac{\int_{H_x} \mu(\mathbf{x}, \mathbf{x}') \, \mathrm{d}V_x}{\int_{H_x} \mathrm{d}V_x}$$
(18)

The damage ranges from zero, when all bonds associated with particle at x are intact, to one, when all bonds are broken.

## 2.4. Numerical implementation

The PD formulation for coupled water flow and mechanical deformation with cracking is summarised by

$$\frac{\partial \theta(\mathbf{x})}{\partial t} = \int_{H_{\mathbf{x}}} \mu(\mathbf{x}, \mathbf{x}') D_p(\mathbf{x}, \mathbf{x}') \frac{\theta(\mathbf{x}') - \theta(\mathbf{x})}{\|\boldsymbol{\xi}\|^2} dV_{\mathbf{x}}$$
(19)

$$\rho(\boldsymbol{x})\ddot{\boldsymbol{u}}(\boldsymbol{x}) = \int_{H_{\boldsymbol{x}}} \mu(\boldsymbol{x}, \boldsymbol{x}') \, C_p(\boldsymbol{x}, \boldsymbol{x}') \, (\boldsymbol{\epsilon} - \alpha \, \bar{\theta}) \, \boldsymbol{n} \, \mathrm{d}V_{\boldsymbol{x}} + \boldsymbol{b}(\boldsymbol{x}) \tag{20}$$

The spatial discretization of Eqns. (19) and (20), using the mid-point rule is (Yan et al., 2020)

$$\frac{\partial \theta(\boldsymbol{x}_i)}{\partial t} = \sum_p \mu(\boldsymbol{x}_i, \boldsymbol{x}_p) D_p(\boldsymbol{x}_i, \boldsymbol{x}_p) \frac{\theta(\boldsymbol{x}_p) - \theta(\boldsymbol{x}_i)}{\|\boldsymbol{x}_p - \boldsymbol{x}_i\|^2} V_{ip}$$
(21)

$$\rho(\boldsymbol{x})\ddot{\boldsymbol{u}}(\boldsymbol{x}) = \sum_{p} \mu(\boldsymbol{x}_{i}, \boldsymbol{x}_{p}) C_{p}(\boldsymbol{x}_{i}, \boldsymbol{x}_{p}) \left[\epsilon(\boldsymbol{x}_{i}, \boldsymbol{x}_{p}) - \alpha \overline{\theta(\boldsymbol{x}_{i}, \boldsymbol{x}_{p})}\right] V_{ip} + \boldsymbol{b}(\boldsymbol{x})$$
(22)

Iterative coupling approaches have been used in multiphysics problems (e.g. Sedighi et al. 2018). Time integration of the system is performed by a weakly coupled method which deals with large displacements and evolving discontinuities. This method has been introduced in PD to solve thermo-mechanical problems (Wang et al., 2019; Antuono and Morandini, 2017). It

uses different time stepping strategies for different physical or mechanical processes. Details of our implementation are provided in Supplementary material (S3).

#### 3. Results and discussion

We have selected two sets of experimental data from literature (Najm et al., 2009; Péron et al., 2009a and 2013) to validate the model. The first applications, presented in sub-section 3.1, deal with the dessication cracking of clay rings of different geometries. The second application, presented in sub-section 3.2, deals with the dessication cracking of a long clay bar. The data reproduced and replotted from the literature is obtained by using the function of Digitizer in Origin Pro 2016 although there may have been slight deviations as the results of digitalisation.

## 3.1. Desiccation cracking of clay rings

Restrained ring tests are convenient for studying soil desiccation cracking as they allow for obtaining clearly isolated cracks (Najm et al., 2009). In these tests, a soil sample with inner and outer radii  $R_{is}$  and  $R_{os}$ , respectively, is placed around a steel ring with inner and outer radii  $R_{ir}$  and  $R_{or}$ , respectively (i.e., the sample thickness is  $R_{or} - R_{ir}$ . The sample is not subjected to external mechanical loads, but experiences dimensional changes (shrinkage) induced by drying. The shrinkage is constrained by the steel ring, leading to the development of stresses and strains in the ring and in the sample. If the strain at the inner surface of the steel ring is recorded by strain gauges, the hoop,  $\sigma_{\varphi}(r)$ , and radial,  $\sigma_r(r)$ , stresses in the soil can be calculated analytically as functions of radial distance, r, (Najm et al., 2009):

$$\sigma_{\varphi}(r) = \varepsilon_r E_r \left(\frac{R_{or}^2 - R_{ir}^2}{2R_{or}^2}\right) \left(\frac{R_{is}^2}{R_{os}^2 - R_{or}^2}\right) \left(1 + \frac{R_{os}^2}{r^2}\right)$$
(23)

$$\sigma_r(r) = \varepsilon_r E_r \left(\frac{R_{or}^2 - R_{ir}^2}{2R_{or}^2}\right) \left(\frac{R_{is}^2}{R_{os}^2 - R_{or}^2}\right) \left(1 - \frac{R_{os}^2}{r^2}\right)$$
(24)

where  $\varepsilon_r$  is the strain at the inner wall of the steel ring and  $E_r$  is Young's modulus of the steel. Notably, the hoop stress is tensile, while the radial stress is compressive as the last term in Eq. (24) is negative at all locations in the sample. The analytical solution provided in Eqns. (23) and (24) is applicable to homogeneous isotropic linear elastic materials, hence approximate for the real experimental setup. We consider two sets of experimental data obtained with constrained ring tests for model validation. These tests are illustrated in Fig. 2 (Najm et al., 2009) and their geometrical characteristics and the steel ring elastic modulus are given in Table 1. The figure shows the effect of test geometry on the cracking behaviour. The ring with the smaller inner radius,  $R_{is}$ = 2.11 cm, and smaller thickness, 3.19 cm, referred to as Test 1, developed a single crack. The ring with the larger inner radius,  $R_{is}$ = 15 cm, and larger thickness, 7 cm, referred to as Test 2, developed multiple cracks. The initial water content in Test 1 was 44.95%. In this validation round, we aim to assess the accuracy of the model to reproduce such effects.

In the experiments water was free to evaporate, evaporation rate was recorded, and evaporation was assumed to be uniform across the sample. Cracking was observed when the soil moisture content was reduced to around 35.9%. The tensile stress in the sample, calculated at the moment of crack initiation, was 65.9 kPa. This was taken to be the tensile strength,  $\sigma_t$ , of the soil. The parameters measured during Test 1, including moisture content, evaporation rate, and strain at inner steel surface, are not available for Test 2. Therefore, we assume that they are the same as in Test 1 for our validations.



Figure.2 Desiccation cracking results of clay in restrained ring tests: (a)  $R_{is}$ =2.11 cm; and (b)  $R_{is}$ =15 cm. It is noted that Fig 2(b) is a recreation of the image presented by Najm et al., 2009 (Fig 6 in the original reference) which was digitised and plotted by OriginPro 2016 with enhanced quality and for quantitative comparison purposes with numerical results.

Test No.	Test 1	Test 2
The inner radius of soil ring, $R_{is}$ (cm) <sup>a</sup>	2.11	15
The outer radius of soil ring, $R_{os}$ (cm) <sup>a</sup>	5.30	22
Restraining ring inner radius, $R_{ir}$ (cm) <sup>a</sup>	1.95	14.84
Restraining ring outer radius, $R_{or}$ (cm) <sup>a</sup>	2.11	15
Restraining ring Young's modulus, $E_r$ (GPa) <sup>a</sup>	2.9	2.9
The Young modulus of the soil, $E$ (MPa) <sup>b</sup>	1.2	1.2
Shrinkage coefficient, $\alpha$ (-) <sup>a</sup>	0.065	0.065
Fracture energy release rate, $G_0 (N/m)^b$	5.7	5.7

Table 1. Geometry of the experimental tests and material properties

<sup>a</sup> (Najm et al., 2009); <sup>b</sup> (Jabakhanji, 2013);

For the simulation of Test 1, the soil sample was represented by a ring of inner and outer radii of 2.1 cm and 5.3 cm, respectively, populated by PD particles. The spacing between particles was  $\Delta x = 0.5$  mm. An important parameter for the numerical implementation of PD is the nonlocal ratio  $m = \delta/\Delta x$ . As described by Dipasquale et al. (2014) the dependence of the crack path on the particle spacing is reduced by selecting an appropriate m in the range 5~8. We used m = 5 and  $\delta = 2.5$  mm. Young's modulus of the soil was assumed to remain constant  $E_s = 1.2$ MPa (Jabakhanji, 2013). We note that the experiments indicated changes of Young's modulus with deceasing moisture content of clay, but consider our assumption as a reasonable approximation for the purposes of this study. Based on the experiments (Najm et al., 2009), a constant shrinkage coefficient  $\alpha = 0.065$  was adopted for this sub-section. The fracture energy release rate was taken from Jabakhanji (2013):  $G_0 = 5.7$  N/m. We note that this parameter has been reported in the literature in the range 1-10 N/m. The clay heterogeneity was accounted for by considering that the fracture energy release rate followed a Weibull distribution with shape parameter 20 (Tang et al., 2016; Oterkus and Madenci, 2017). The inner boundary of the soil was constrained, i.e., Dirichlet boundary condition was equal to zero which was achieved by adding a layer of fixed PD particles with a thickness equal to the horizon,  $\delta$  (Oterkus et al., 2014). This neglects the deformability of the steel ring, but is a reasonable approximation considering

the large difference in the elastic moduli. The outer boundary of the soil was left free, i.e., Neumann boundary condition equal to zero. The moisture content in the soil was reduced by assuming a continuous uniform water loss,  $1.43 \times 10^{-3}$  wt (%) /s, across the model at each time step. The total number of time steps was  $10^5$  with a time step of 1s.

Figure 3 shows a comparison between the hoop stress  $\sigma_{\varphi}(r)$  at r = 2.1cm (inner soil surface), calculated from our PD simulations and the hoop stress calculated by Eq. (23) using the gauge strain recorded during the experiments. There is a good agreement between the experimental and modelling values. Observable differences can be attributed to two factors: (i) Young's modulus of soil is changing with moisture content; and (ii) the clay is not a homogeneous isotropic linear elastic material for which Eq. (23) is strictly applicable. We can influence only the first factor, and this is a subject of ongoing work.



Figure 3. Hoop stress evolution with moisture content at the constrained soil surface of Test 1.

Figure 4 shows a comparison between the hoop stress  $\sigma_{\varphi}(r)$  along the radius, calculated by the PD simulations ( $\theta = 36\%$ , 39% and 42%) and those reported from the experimental data ( $\theta = 36\%$ ). The curve at  $\theta = 36\%$  corresponds to the moisture content at which the crack initiation was experimentally observed. The close agreement of the calculated and experimental values provides confidence that the peridynamic simulations predict accurately the stress distribution during desiccation under constrained shrinkage.



Figure 4. Hoop stress distribution in radial direction at different moisture content for





Figure 5. Contours of hoop (left) and radial (middle) displacements, and damage parameter (right) from Test 1 PD simulations at moisture content: (a)  $\theta = 35\%$ ; (b)  $\theta = 33.6\%$ ; and (c)  $\theta = 30.7\%$ .

The results in Fig 5 indicate a correlation between soil shrinkage and crack growth. Variations of strains and stresses as crack propagates are clearly visible. The hoop stress gradient causes the crack to emerge at the inner surface and to extends towards the outer surface. The crack path deviates from a straight line due to the heterogeneity introduced in the model by the Weibull distribution of the fracture energy release rate.

Figure 6 shows a comparison between the predicted final crack and the experimental outcome, including the damage contour from the PD simulations and a geometric representation of the ring that facilitates comparison with the experimental figure. There is a close agreement between the predicted and observed configurations, including the non-straight crack path due to soil heterogeneity.



Figure 6. Desiccation crack in Test 1: (a) damage contour from PD simulations; (b) reconstructed cracked configuration; and (c) experimental results. It is noted that Fig 6(c) is a recreation of the image presented by Najm et al., 2009 (Fig 6a in the original reference) which was digitised and plotted by OriginPro 2016 with enhanced quality and for quantitative comparison purposes against numerical results.

The simulation was repeated for Test 2 with the corresponding change of ring geometry. Figure 7 shows the final cracked configuration (numerical prediction and experimental result) for Test 2. It can be seen that the multiple cracks observed experimentally in this thicker specimen with larger inner radius are correctly predicted by the model. The emergence of multiple cracks can be explained by the larger inner radius in Test 2. One crack in Test 1 appears to be sufficient for relaxing the hoop stress along the entire inner soil surface to levels that do not allow for damage to initiate elsewhere. In contrast, the emergence of one crack in Test 2 does not relax the hoop stress along the inner entire soil surface, which allows for initiation of multiple cracks, approximately equally spaced. This is an interesting outcome, which is affected not only by the size of the inner surface but also by the thickness of the soil ring and deserves further investigation. Presently, we note the excellent agreement between the predicted and experimentally observed cracks in terms of their number and approximate spacing.



Figure 7. Desiccation cracks in Test 2: (a) damage contour from PD simulations; (b) reconstructed cracked configuration; and (c) experimental results. It is noted that Fig 7(c) is a recreation of the image presented by Najm et al., 2009 (Fig 6b in the original reference) which was digitised and plotted by OriginPro 2016 with enhanced quality and for quantitative comparison against numerical results.

## 3.2. Desiccation cracking of clay bars

A series of clay (Bioley silt) desiccation tests on long bars were conducted by Péron et al. (2009a) to study the mechanism of desiccation cracking induced by swelling/shrinkage processes. In these experiments, a clay slurry at a gravimetric water content of 1.5 times the liquid limit has been subject to drying. The clay slurry was poured into a plate mold with dimensions 295mm(length)×49mm(width)×12mm(height). The sample was not subjected to mechanical loads. The bottom of the sample was constrained by a notched metallic base. The initial water content of sample was 50% (Péron et al., 2013). Experimental results showed that the water content decreased linearly in the initial drying stage (lasting approximately 2000 min), which corresponded to a constant water evaporation rate of approximately 3 g/h, and accompanied by a decrease of void ratio. This was followed by a drying stage with non-linear water content reduction due to reaching a structural lower limit of the void ratio of approximately 0.4. Our simulations aimed to reproduce the initial drying stage.

To facilitate comparison with our PD results, the experimentally observed desiccation cracking at t = 16.5h, 17.2h and 18h, and the final crack pattern are shown in Fig. 8. These cover moments from the first crack observation at t = 16.5h, through the middle of the linear regime at t = 18h, to the final pattern realised prior to the end of the linear regime.



Figure 8. Experimentally observed evolution of cracks with time (a) and final crack pattern (b) during desiccation cracking of long clay bar. In (b), the top figure is a top view, while the bottom figure is a side view. It is noted that these are recreations of the Fig.17 in Péron et al., 2009a and Fig.2 in Péron et al., 2013; digitised and plotted by OriginPro 2016 for enhanced quality and comparison purpose against numerical reults.

The PD model was created using a 2D domain of 295mm(length)×12mm(height), populated with PD particles. The spacing and horizon values were identical to the ones in Section 3.1. The material properties are listed in Table 2. Young's modulus of clay E = 32 MPa was taken from Péron et al. (2007), where it was determined by triaxial compression tests. The fracture energy release rate  $G_0 = 1.8N/m$  was taken from Ayad et al. (1997), and as in Section 3.1 this material property was assumed to follow Weibull distribution with shape parameter 20 in order to represent the heterogeneity of clay. The drying shrinkage coefficient  $\alpha = 1.56 \times 10^{-2}$  was taken from the shrinkage experiments by Péron et al. (2013). The diffusivity of moisture in clay  $D = 2.8 \times 10^{-10}$  m<sup>2</sup>/s was taken from Hirobe and Kenji (2017).

Table 2. Soil properties of Bioley silt

Property	Value
Young's modulus of soil, <i>E</i> (MPa) <sup>a</sup>	32
Shrinkage coefficient, $\alpha$ (-) <sup>b</sup>	$1.56 \times 10^{-2}$
The fracture energy release rate, $G_0$ (N/m) <sup>c</sup>	1.8
The diffusivity of moisture, $D (m^2/s)^d$	$2.8 \times 10^{-10}$

a (Péron et al., 2007); b (Ayad et al., 1997); c (Péron et al., 2013); d (Hirobe and Kenji, 2017).

The displacements at the bottom side of the model were fixed, i.e. Dirichlet boundary condition equal to zero, to represent the effect of the notched metallic base, while the other three sides were left free, i.e. Neumann boundary condition equal to zero. Moisture flow was prohibited through the bottom, right and left sides of the sample, i.e. Neumann boundary condition equal to zero, while the top of the sample was subjected to the experimentally measured evaporation rate of 3 g/h (Péron et al., 2013). Figures 9-12 present the results for water content, horizontal displacement, vertical displacement and damage parameter at four instances: t=17h, 18.3h, 20.8h and 25h. Based on experimental observations, soil desiccation has been described by three main phases: (i) evaporation of moisture from the sample surface due to water potential difference between soil and atmosphere ; (ii) water flow from the interior to the surface due to pore-water pressure gradient created in phase (i); and (iii) shrinkage of the sample with decreasing water pressure leading to increasing internal compression of the matrix/soil

aggregate (Coussy et al., 1998; Kowalski 2003). The results presented show that the proposed PD model captures these main phases with acceptable accuracy. The surface water content decreases as the evaporation increases with time (see Fig. 9). This leads to a hydraulic gradient between the interior and the surface and moisture flow from the bottom to the top of the sample, with a subsequent reduction of the overall water content in the sample with time (Fig. 9). The contours of water content are not uniform due to the emergence and growth of cracks, which disrupt fluid flow locally (Yan et al., 2020). The shrinkage of the sample with increasing evaporation is captured by the displacement plots in Figs. 10 and 11. The horizontal shrinkage is represented by the opposite horizontal displacements at the two ends of the sample in Fig. 10, while the maximal vertical shrinkage is observed between the growing cracks in Fig. 11.

Figure 8 shows that the desiccation cracking generally starts from the top of the soil sample. The PD results Fig. 12 are consistent with this observation. For example, the first substantial crack in the PD model is observed at t=17h and starts from the top, which is very close to the initiation of cracking recorded in the experiment at 16.5h. The second substantial crack is observed at t=18.3h, followed by a number of further cracks developing as the desiccation process proceeds. The final crack pattern is also close to the experimental observation. For example, the total number of primary cracks developed in experiments was between 7 and 9 (Péron et al., 2009a) and our simulations lead to 8 primary cracks (Fig. 12). The spacings between the developed cracks differ between model and experiment, which is to be expected considering the soil heterogeneity. The number of cracks and the average spacing, however, should depend on the length and thickness of the specimen, similarly to the case considered in Section 3.1, and this dependence deserves further investigation.



Figure 9. Water content contours predicted by PD simulations at *t*=17h, 18.3h, 20.8h and 25h (from top to bottom)



Figure 10. Horizontal displacement contours predicted by PD simulations at t=17h, 18.3h, 20.8h and 25h (from top to bottom)



Figure 11. Vertical displacement contours predicted by PD simulations at t=17h, 18.3h, 20.8h and 25h (from top to bottom)



Figure 12. Damage contours, illustrating cracks development, predicted by PD simulations at t=17h, 18.3h, 20.8h and 25h (from top to bottom)

## 4. Conclusions

A model for analysis of soil desiccation cracking based on peridynamics was presented. It integrated bond-based formulations of moisture flow and mechanical deformation/failure. The coupled formulation was implemented within a computational framework Pyramid (Yan et al.
2020; Sedighi et al., 2021). The model was tested against two sets of experimental data. It was shown that the simulations reproduced the main phases of soil desiccation successfully. The results captured key features, such as stress distribution, sample geometry effects, crack initiation and propagation, including the number and the non-uniform spacing of cracks due to clay heterogeneity. Specifically, crack initiation was studied by investigating the correlation between the shrinkage of soil clay, changes in displacement fields and crack growth in ring specimens with different dimensions. In the specimen with small inner radius, a single crack emerged at the inner surface of the ring, due to the larger hoop stresses there, and extended towards the outer surface as experimentally observed. For this geometry, it appears that one crack is sufficient to relax the hoop stress along the inner surface, so that other cracks cannot be realised. In the specimen with large inner radius, multiple cracks emerged at the inner radius and propagated towards the outer surface to cause fragmentation as experimentally observed. For this geometry, it appears that the initiation of one crack is not sufficient to relax the hoop stress along the entire inner surface, which allows for initiation of multiple cracks. The simulations of desiccation cracking of long clay bars showed cracks emerging from the free surface of the specimen where evaporation was allowed in accordance with experiments. The cracks propagation and final crack patterns were found to be in close agreement with the experimental observations. The results provide confidence that the proposed modelling approach is physically realistic and can be used to investigate physical, mechanical, and geometric effects on clay desiccation cracking.

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### **Supplementary materials**

#### S1: Derivation of micro-modulus function

For the two-dimensional (2D) plane stress case, the peridynamic strain energy density at node x under isotropic expansion can be calculated by (Silling and Askari, 2005):

$$W^{PD}(\mathbf{x}) = \frac{1}{2} \int_{H_{\mathbf{x}}} \omega(\boldsymbol{\xi}, \boldsymbol{\eta}) \, \mathrm{d}\mathbf{x} = \frac{1}{2} \int_{0}^{\delta} \frac{C_{p} \, \epsilon_{m}^{2} r}{2} 2\pi h r \mathrm{d}r = \frac{\pi C_{p} \epsilon_{m}^{2} \delta^{3}}{6}$$
(S1-1)

For the 2D plane stress case, the strain energy density in classical continuum mechanics (CCM) under the same stress state is given by (Chen et al., 2019)

$$W^{CCM}(\mathbf{x}) = \frac{E\epsilon_m^2}{(1-\nu)}$$
(S1-2)

where,  $\nu$  is Poisson's ratio.

Equating Eqns. (S1-1) and (S1-2) with  $\nu = 1/3$ , gives the micro-modulus in the 2D plane stress case

$$C_p = \frac{9E}{\pi\delta^3} \tag{S1-3}$$

#### S2: Derivation of microscopic diffusivity function

In order to determine the microscopic diffusivity from measurable (apparent) diffusivity, we follow the approach proposed by Bobaru and Duangpanya (2010). We consider pure diffusion with a constant diffusivity in a homogeneous and isotropic porous medium. The peridynamics pure diffusive flux,  $J_s^{PD}(x, t)$ , and the local pure diffusive flux,  $J_s(x)$ , are equal under steady-state conditions. This enables the calculation of the peridynamics micro-diffusivity. The local steady-state flow at position x can be expressed by:

$$\theta(x) = mx + n \tag{S2-1}$$

$$J_s(x) = -D\frac{\partial\theta}{\partial x} = -mD \tag{S2-2}$$

where, *m* and *n* are constants, and  $J_s(x)$  is the classic water flux.

The peridynamics flux,  $J_s(x, x', t)$ , between two material points x and x' is given in the manuscript as follow:

$$J_s(\boldsymbol{x}, \boldsymbol{x}', t) = -D_p(\boldsymbol{x}, \boldsymbol{x}') \frac{\theta(\boldsymbol{x}', t) - \theta(\boldsymbol{x}, t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(S2-3)

In order to derive the peridynamics flux in all the bonds attached to x in its horizon  $H_x$ , Eq. (S2-3) is integrated over the horizon of material point x. For the 2D case, the peridynamics flux can be expressed as (Yan et al., 2020):

$$J_{s}^{PD}(x,t) = \int_{-\pi/2}^{\pi/2} \int_{0}^{\delta} D_{p}(x,x') \frac{\theta(x',t) - \theta(x,t)}{\|x' - x\|} \cos^{2}(\phi_{s}) r dr d\varphi$$
(S2-4)

By substituting the uniform influence function  $(D_p(\mathbf{x}, \mathbf{x}') = D_p)$  and the steady-state flow equation, Eq. (S2-1), into Eq. (S2-4), we obtain:

$$J_s^{PD}(x,t) = -m\pi D\delta^2/4$$
(S2-5)

If the local flux, Eq. (S2-2), and the peridynamics flux, Eq. (S2-5), at material point x are equal, the micro-diffusivity for 2D transport cases is obtained as:

$$D_p = \frac{4D}{\pi\delta^2} \tag{S2-6}$$

#### S3. Weakly coupled approach with multi-rate integration

The coupling methods used in multiphysics problems include the strong coupling approach (i.e., direct coupling), and the weak coupling approach (i.e., iterative coupling) (Yan et al., 2020; Sedighi et al. 2018). A weakly coupled method has been introduced in PD to solve coupled problems such as thermo-mechanical (Wang et al., 2019; D'Antuono and Morandini, 2017). In a weakly coupled method, different time stepping strategies are generally employed for different physics/processes. The weakly coupled approaches with explicit multi-rate integrations are suitable choices for the PD multiphysics problem as they can deal with large displacements and evolving discontinuities. In this study, a weakly coupled approach is adopted to solve the coupled problems of water flow and mechanical deformation.

A forward difference time marching is adopted for the moisture transport, Eq.(20); therefore, given the field variable at time  $t_n^{MT}$ , the solver can advance to the next time step as follow:

$$\frac{\theta(\boldsymbol{x}_{i}, t_{n+1}^{\theta}) - \theta_{l}(\boldsymbol{x}_{i}, t_{n}^{\theta})}{\Delta t_{n}^{MT}} = \sum_{p} \mu(\boldsymbol{x}_{i}, \boldsymbol{x}_{p}, t) d_{l}(\boldsymbol{x}_{i}, \boldsymbol{x}_{p}, t) \frac{\theta(\boldsymbol{x}_{p}, t) - \theta(\boldsymbol{x}_{i}, t)}{\left\|\boldsymbol{x}_{p} - \boldsymbol{x}_{i}\right\|^{2}} V_{ip}$$
(S3-1)

where,  $\Delta t_n^{MT}$  is the incremental time step corresponding to the moisture transport.  $\Delta t_n^{MT}$  is selected with a proper range to achieve a stable solution. The upper bound of the range of

stability condition calculated based on von Neumann stability analysis can be expressed as (Kilic and Madenci, 2010)

$$\Delta t_n^{MT} < \frac{1}{\sum_{p=1}^N \frac{d_l(\boldsymbol{x}_i, \boldsymbol{x}_p)}{\|\boldsymbol{x}_p - \boldsymbol{x}_i\|^2} \Delta \boldsymbol{A}_{\boldsymbol{x}_p}}$$
(S3-2)

At time step  $t_n^{MT} \sim t_{n+1}^{MT}$ , the quasi-static solutions of mechanical deformation and cracking are obtained using the Adaptive Dynamic Relaxation (ADR) method with the mechanical time step  $\Delta t_n^{ME} = 1$  s proposed by (Kilic and Madenci, 2010; Huang et al., 2015). Assembling the equation of each node, Eq. (21), into a global system of equations and introducing an artificial damping to the system, it can be transformed to

$$\Lambda \ddot{\mathbf{U}}(\mathbf{X}, t) + c\Lambda \dot{\mathbf{U}}(\mathbf{X}, t) = \mathbf{F}(\mathbf{U}, \mathbf{U}, \mathbf{X}, \mathbf{X})$$
(S3-3)

where  $\Lambda$  is the fictitious diagonal density matrix and *c* represents the adaptive varying damping coefficient introduced by the lowest frequency of the system using Rayleigh's quotient (Kilic and Madenci, 2010); The positions vector, **X** and displacements vector **U** in Eq. (S-3) are expressed by

$$\mathbf{X} = \langle \mathbf{x}_1, \mathbf{x}_2, \cdots, \mathbf{x}_M \rangle \tag{S3-4}$$

$$\mathbf{U} = \langle \mathbf{u}_1, \mathbf{u}_2, \cdots, \mathbf{u}_M \rangle \tag{S3-5}$$

M is the total number of nodes and  $\mathbf{F}$  represents the summation of the external and internal forces, whose *i*-th component is given by

$$\mathbf{F}_{i} = \sum_{i=1}^{M} \sum_{j=1}^{N} \mathbf{f} (x_{j} - x_{i}, u(x_{j}, t) - u(x_{i}, t), t) V_{j} + b(x_{i}, t)$$
(S3-6)

In the present study, time integration is performed by the central difference explicit integration. Given displacement and acceleration of each node  $x_i$  at  $t_n$ , velocities and displacements at  $t_{n+1} = t_n + \Delta t_n^{ME}$  can be obtained by

$$\dot{\mathbf{U}}^{n+1/2} = \left[ (2 - c\Delta t_n^{ME}) \dot{\mathbf{U}}^{n-\frac{1}{2}} + \mathbf{2}\Delta t_n^{ME} \mathbf{\Lambda}^{-1} \mathbf{F}^n \right] / (2 + c\Delta t_n^{ME})$$
(S3-7)

$$\mathbf{U}^{n+1} = \mathbf{U}^n + \Delta t_n^{ME} \dot{\mathbf{U}}^{n+1/2}$$
(S3-8)

where the integration starts with initial conditions  $\dot{\mathbf{U}}^0 = 0$  and  $\mathbf{U}^0 \neq 0$  as follow

$$\dot{\mathbf{U}}^{1/2} = \Delta t_n^{ME} \mathbf{\Lambda}^{-1} \mathbf{F}^0 / 2 \tag{S3-9}$$

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# CHAPTER Five

# 5. Erosion of Swelling Clay

This chapter contains three research articles, including (i) one published in *Géotechnique*, 2020 (DOI: https://doi.org/10.1680/jgeot.20.P.149) (section 5.3), (ii) one published in *Engineering Geology*, 2021 (https://doi.org/10.1016/j.enggeo.2021.106409) (section 5.4), and (iii) one submitted manuscript (section 5.5).

# 5.1 Paper introduction:

In this chapter, a study on theoretical and model developments for investigating hydrochemical effects on swelling clay erosion in section 5.3 is first presented. A non-local PD formulation for clay erosion is developed, which brings together clay swelling, particle detachment, and detached particle transport processes, into a single modelling framework. In addition, a new particle detachment model, which can capture hydro-chemical effects on erosion by looking into the physico-chemical interactions between colloid particles was proposed. The proposed detachment model was validated by experimental data for yield stress dependence on clay volume fraction, and for critical clay volume fraction at the solid/liquid interface.

Section 5.4 presents the applications of the proposed erosion model for assessing the effects of solution chemistry and flowrate on the penetration, extruded mass and particle release rate of clay buffer. A series of experimental data were used to validate the swelling and erosion model developed in this study. The developed and validated model appears to be a viable alternative to the classical analysis approaches, based on the strong differential formulations of the processes involved, as it captures naturally the effects of main parameters controlling erosion – clay dry density, water chemistry and water flowrate.

Section 5.5 presents a model for co-transport of smectite particles and suspended accessory mineral particles. It was developed to explain the role of accessory minerals in the erosion protection of bentonite. The accessory minerals are transported with the smectite as it swells and deposited at the location where the repulsive forces between smectite particles become week, i.e., in the interface region between the bentonite and the water. The model was validated by two sets of experimental data. It was demonstrated that the model reproduced the main experimental observations of build-up of mineral film and of its growth with time. It is

highlighted that assessments of erosion of mineral-rich bentonite based on experimental data obtained with pure smectite will be overly conservative.

# 5.2 Contribution to the joint authorship

The author has contributed the following to the paper in Section 5.3 and Section 5.4:

- Development of the main concept for this paper (80%).
- Theoretical developments for swelling clay erosion (90%).
- Model implementations and code writing (100%).
- Validations and applications of the proposed model (90%).
- Preparation of tables and data visualisation (100%).
- Paper writing (80%).

The author has contributed the following to the paper in Section 5.5:

- Development of the main concept for this paper (90%).
- Theoretical developments for co-transport model (90%).
- Model implementations and code writing (100%).
- Validations and applications of the proposed model (90%).
- Preparation of tables and data visualisation (90%).
- Paper writing (90%).

#### Addendum

1. Page 178 lines 7-9 in the published paper (Section 5.3): It can be seen that the calculated values are in good match with the experiments.

2. Page 212 lines 5-7 in the published paper (Section 5.4): Decreasing water chemistry can enhance the DDL repulsive force, which may increase the swelling pressure and subsequently the measured swelling strain.

3. Page 222 lines 5-6 in the published paper (Section 5.4): Figure 10 shows the penetration distance of clay buffer as a function of water flowrate ranging from  $10^{-7}$  m/s to  $2 \times 10^{-4}$  m/s (the maximum flowrate in the experiment).

4. The relative difference ( $\varepsilon$ ) is used to show the validity of the present numerical model given

in Table 1.

	Figure	Case	ε
Section 5.3	on 5.3 Fig.6 $v=2\times 10^{-10}$		2.33
		$v=2 \times 10^{-4} \text{ m/s}, c=8.6 \text{ mM}$	0.343
		$v=2 \times 10^{-4} \text{ m/s}, c=17 \text{ mM}$	2.50
		$v=2 \times 10^{-4} \text{ m/s}, c=171 \text{ mM}$	0.22
		$v=2.6 \times 10^{-5} \text{ m/s}, c=0 \text{ mM}$	0.1
		$v=6.2 \times 10^{-6} \text{ m/s}, c=0.83 \text{ mM}$	0.09
	Fig.11	Test 1	11.71
		Test 2	1.41
		Test 3	0.13
Section 5.4	Fig.4	$ ho_d=1.2 \text{ Mg/m}^3$	0.0035
		$\rho_d = 1 \text{ Mg/m}^3$	0.0025
	Fig.5a	<i>t</i> =4 d	10.74
		<i>t</i> =8 d	1.65
		<i>t</i> =16 d	6.18
		<i>t</i> =32 d	0.17
	Fig.5b	<i>t</i> =4 d	85.8
		<i>t</i> =8 d	0.21
		<i>t</i> =16 d	0.27
		<i>t</i> =32 d	0.19
	Fig.7	<i>c</i> =171 mM	1.58
		<i>c</i> =17 mM	0.13
		Stagnant water	10.77
	Fig.9	-	0.03

Table 1 The relative difference between the results from PD and experiments

# This Chapter is exact copies of the accepted format manuscripts (section 5.3 and 5.3) and a submitted draft (section 5.5) referred to above.

#### 5.3 Peridynamics modelling of clay erosion

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# Abstract

Experimental observations of the erosion of clays indicate a high degree of coupling between the hydro-chemical processes in the clay-water system and the mechanics of erosion. In contrast with the volume of experimental work and the significance of swelling clay erosion in many geotechnical engineering problems, limited advances have been made with the predictive modelling of this phenomenon. Critical erosion step, missing in existing models, is the particles detachment from the moving boundary of the expanding clay – a discontinuous process. This paper presents, for the first time, a non-local formulation for clay erosion, which brings together clay swelling, particle detachment, and detached particle transport, into a single modelling tool.

The detachment criterion, an essential element of the integrated erosion model, is firstly tested against a series of experimental benchmarks to show excellent agreement between calculated and experimental results. The integrated erosion model is subsequently validated by comparison with experimental data for the behaviour of compacted bentonite under several eroding environments. The results: (i) show clearly the capacity of the model to capture concurrently the free swelling of clay, the detachment of clay particles, and the transport of detached particles in the eroding environment; and (ii) support strongly the applicability of the model to account for the hydro-chemical conditions (composition and velocity) of the eroding environment. The proposed multi-physics non-local formulation, successfully validated for hydro-chemical effects on clay erosion, provides a robust framework for incorporating a wide set of additional couplings.

**Keywords**: Erosion, clay, peridynamics, numerical modelling, coupled problems, discontinuity, chemistry

# **1. Introduction**

Robust prediction of clay erosion is important for a range of geotechnical engineering problems; from clay erosion in embankments (Fujisawa et al., 2009), through erosion of clay in geological disposal of nuclear waste (Schatz et al., 2013), to erosion of geosynthetic clay liners (Ashe et al., 2014). Of particular importance and recent interest is the erosion of clay buffer and backfill in the context of geological disposal of high-level nuclear waste (HLW). The clay barriers are key components of the engineered barrier system (EBS) in the geological disposal concepts and provide critical functions in the safe isolation of HLW. During the long-term operation of disposal facilities, the clay buffer/backfill can be subject to erosion induced by the hydrochemical interactions at the interface between the clay and the fractured host rock. Localised interactions of the clay with peculating water at fracture/fissures/pre-crack in host rock varying in size from micro-fractures (below 1mm) to relatively large fractures (several mm and more), can result in gradual loss of the barrier and a co-transport of radionuclides into the biosphere. The aim of this paper is to address the need for a realistic and robust modelling of clay erosion. This is accomplished by presenting a set of new formulations, based on the non-local continuum mechanics of peridynamics (PD), which is utilised, for the first time, to couple the hydro-chemical processes with erosion of swelling clays.

Experimental studies of erosion of smectite-rich clays (highly swelling clays) have shown that the overall behaviour is strongly dependent on the fluid geochemical composition and exchangeable ions, and on the fluid velocity (Schatz et al., 2013, Smith et al., 2017). The erosion of swelling clays involves three physical processes: (i) free swelling, representing a continuous transformation of solid-type phase to gel-type material; (ii) clay particle detachment; and (iii) migration of detached particles with the water flow in the fracture. The semi-empirical or phenomenological models proposed to date do not fully address all phase changes accompanying the underlying processes. The existing models address mostly the individual processes, e.g. clay gels generation (Pusch, 1999), mechanical erosion caused by the water flow (Grindrod et al., 1999), erosion of particles and dispersion-flocculation behaviour of clay particles (Kurosawa et al., 1999) and expansion/swelling of the clay (Navarro et al., 2017; Asensio et al., 2018). Pioneering works by Neretnieks et.al. (2009) and Liu et al. (2009a; 2009b) introduced the dynamic force balance model in simulating clay expansion and extrusion. A version extending the model by flow and colloid diffusion was presented by Moreno et al.

(2011), whilst a recent development (Neretnieks et al., 2017) addressed the strong hydrochemical effects on erosion of smectite-rich clays observed in experiments (e.g. Schatz et al., 2013).

Critical missing element in the existing models is related to the detachment of particles from the gel phase at the solid/fluid interface and its strong coupling with hydro-chemical effects. Our modelling approach will introduce a new criterion for detachment based on the particle interaction forces, i.e. an intrinsic criterion, rather than prescribed phenomenologically. The particles detachment is inherently a discontinuous process taking place at the moving-boundary of continuously swelling phase (gel). The non-local formulations proposed in this work, based on peridynamics theory, are suitable to tackle the complexity of this problem. The PD theoretical framework replaces the partial differential equations of the classical local theory by a set of integral-differential equations. This results in a mathematically consistent formulation, even where strong discontinuities appear due to breaking and fragmentation of the material. The PD theory has successfully been applied in different areas, such as hydraulic fracturing (Ouchi et al., 2015), thermo-mechanics of fracture development (Oterkus et al., 2014), membranes and fibers (Silling and Bobaru, 2005) and dynamic fracture of nanofiber (Bobaru, 2007).

The aim of the work is achieved by developing and integrating bond-based PD formulations for free swelling of clay as a diffusion process and for transport of detached particles, with the important incorporation of the new detachment model. The peridynamics formulations have been shown to provide the computational capability required to address diffusion-type problems for evolving discontinuities or phase changes (e.g. heat transfer in evolving cracking system, pitting corrosion damage and mechano-chemical damage) (Bobaru and Duangpanya, 2012; Chen and bobaru, 2015; Jafarzadeh et al., 2019a and 2019b ). Furthermore, peridynamics modelling approach allows for autonomous evolution of the moving interface without any extra modelling efforts (e.g. interface conditions), which is impotant for modelling the moving-boundary of continuously swelling phase. The PD formulation can offer a computationally robust solution for describing the free swelling where the parameters of the diffusion process vary several orders of magnitude during the soil expansion. Approximation of such level of non-linearity by classical numerical methods, such as FEM and FVM, may yield gross computational errors (Jafarzadeh et al., 2019c). The PD erosion model is validated against the results of a set of experimental benchmarks, firstly to establish the realism of the detachment

model, and then to predict the behaviour of compacted smectite-rich clay under eroding environments.

#### 2. Local formulations of erosion processes

From macroscopic (continuum) perspective, the erosion of clay by water can be described as a series of phase changes: an initially solid phase is hydrated, leading to a continuous transformation into a swelling paste, which in turn is continuously transformed into a gel, and eventually discontinuously transformed into dispersed colloidal particles (sol) in the water. The material is initially partially saturated and the free swelling model approach describes the evolution of the solid volume fraction with time from the starting initial saturation stage until fully hydrated system with an assumption that process follows a diffusion-type behaviour. In this model we consider the evolution of swelling in a relatively thick fracture (> 1 mm) where the friction between the material and boundary does not induce significant effects on the overall process (Neretnieks et al., 2009).

Figure 1 provides a schematic of erosion of swelling clay and its corresponding processes. The free swelling step is characterised by continuous reduction of the clay density. The distinction between the gel and sol phases can be made on the base of previous studies on smectite rich clays.

Forces acting between the particles are shown in Figure 1 which include the diffusion forces  $(F_T)$ , attractive Van der Walls forces  $(F_A)$  and repulsive electrical double layer forces  $(F_R)$ . These forces can be described by (Liu et al., 2009a):

$$F_T = \frac{-k_B T}{\phi_s} \frac{\partial \phi_s}{\partial x} \tag{1}$$

$$F_{A} = \frac{A_{H}S_{p}}{6\pi} \left[ \frac{1}{h^{3}} - \frac{2}{\left(h + \delta_{p}\right)^{3}} + \frac{1}{\left(h + 2\delta_{p}\right)^{3}} \right]$$
(2)

$$F_R = 2cRTS_p(\cosh y^m - 1) \tag{3}$$

where  $\phi_s$  is the clay (solid) volume fraction,  $k_B$  is Boltzmann constant, *T* is absolute temperature,  $A_H$  is Hamaker constant,  $S_p$  is the surface area of the particles, *h* is the separation distance between the flat particles,  $\delta_p$  is the particle thickness, *c* is the ionic concentration in

the pore water system and *R* is universal gas constant. The function  $y^m$  represents the scaled potential in the midpoint between parallel plates of for example a fracture, described by (Liu et al., 2009a):

$$y^{m} = \sinh^{-1} \left[ 2\sinh(y_{\infty}^{m}) + \frac{4}{\kappa h} \sinh\left(\frac{y_{\infty}^{h}}{2}\right) \right]$$
(4)

where

$$y_{\infty}^{m} = 4 \tanh^{-1} \left[ 2 \tanh\left(\frac{y_{\infty}^{0}}{4}\right) \exp\left(-\frac{\kappa h}{2}\right) \right]$$
(5)

$$y_{\infty}^{h} = 4 \tanh^{-1} \left[ \tanh\left(\frac{y_{\infty}^{0}}{4}\right) \exp(-\kappa h) \right]$$
(6)

The dimensionless surface potential of the isolated plate  $(y_{\infty}^0)$  and the Debye length  $(\kappa)$  in the Eqns. (4)-(6) are given by:

$$y_{\infty}^{0} = 2\sinh^{-1}\left(\frac{zF\sigma^{0}}{2\varepsilon_{0}\varepsilon_{R}RT\kappa}\right)$$
(7)

$$\kappa = \sqrt{\frac{2cz^2F^2}{\varepsilon_0\varepsilon_R RT}} \tag{8}$$

where *F* is the Faraday's constant;  $\sigma^0$  is the surface charge density; *z* is valence of ions in the pore system and  $\varepsilon_0 \varepsilon_R$  is the dielectric constant. It is noted that the calculation of *F*<sub>R</sub> is based on the "compression" approach, which is applicable to a wide range of parameter conditions in comparison with other available approximate expression (Liu et al., 2009a; Laxton and Berg, 2006).



Figure 1. Conceptual description of clay erosion by swelling, particle detachment, and transport of detached particles

# 2.1. Free swelling of clay

The free swelling is treated as a solid diffusion process following the approach proposed by Liu et al., 2009a. It is noted that the free swelling model proposed by Liu et al (2009a) is developed for the colloidal system of clay. However, applying a diffusion-type description of free swelling for compacted swelling clay is not far from experimental observations (Moreno et al., 2011; Neretnieks et al., 2017). In the absence of gravitational forces and buoyant effects, the mass balance equation for clay solid diffusion can be simplified to:

$$\frac{\partial \phi_s}{\partial t} = \nabla \cdot \left(\frac{\chi}{f_r} \nabla \phi_s\right) \tag{9}$$

where t is time,  $\chi$  is the sum of the particles' energies, and  $f_r$  is the friction coefficient between particles and water.

The friction coefficient is given by (Moreno et al., 2011)

$$f_r = 6\pi \eta_w r_{eq} + V_p k_0 \tau^2 a_p^2 \eta_w \frac{\phi_s}{(1 - \phi_s)^2}$$
(10)

where  $r_{eq}$  is the equivalent radius of the non-spherical particles,  $V_p$  is the volume of the particles,  $k_0$  is the pore shape factor,  $\tau$  is the tortuosity of the flow channel in the clay gel,  $a_p$  is the specific surface area per unit volume of particles, and  $\eta_w$  is the dynamic viscosity of water.

The particles' energy ( $\chi$ ) due to the interaction forces  $F_T$ ,  $F_A$  and  $F_R$ , is given by (Liu et al., 2009a):

$$\chi = k_B T + \left(h + \delta_p\right)^2 \left(\frac{\partial F_A}{\partial h} - \frac{\partial F_R}{\partial h}\right) \tag{11}$$

The separation distance between the flat particles (h) is described by (Laxton and Berg, 2006)

$$h = \delta_p \left( \frac{\phi_s^{max}}{\phi_s} - 1 \right) \tag{12}$$

where  $\phi_s^{max}$  is the maximum clay solid volume fraction.

# 2.2. Particle detachment

The process of particles detachment from the solid/fluid interface is central to clay erosion. The detachment is caused by shear stresses on the particle-particle bonds of the swelled clay (gel) induced by the fluid flow along the interface. This shear stress can be related to the flow velocity by Stoke's law (Kurosawa and Ueta, 2001):

$$\tau_f = \left(\frac{6\pi\eta_w D_p}{S_p}\right) \nu_l \tag{13}$$

where,  $\tau_f$  is the shear stress,  $D_p$  is the particle diameter, and  $v_l$  is the fluid velocity.

A detachment model is required to link dynamically the solid swelling and particles release processes at the solid/liquid interface. A general approach is to compare the shear stress at the solid/liquid interface induced by water flow with a material parameter limiting the solid behaviour of the gel, typically called the yield stress (Laxton and Berg, 2006, Sane et al., 2013). At shear stresses lower than the yield stress the gel behaves like a solid. At shear stresses above the yield stress, the clay particles exhibit sol behaviour and can be transported away by seeping water. This concept is in agreement with experimental observations (Eriksson and Schatz, 2015). However, its application in continuum modelling framework requires the knowledge of both the yield stress and the solid/liquid interface, which remains a challenge. Based on experimental observations, Neretnieks et.al. (2017) developed a rim-region model which imposes an external boundary as a detachment interface with a value of  $\phi_R$  (solid volume fraction at rim). While their model was able to capture a number of experimental observations, it did not provide physical explanations of the mechanisms involved.

The detachment model proposed in this work is significantly different in two ways: 1) the yield stress is not prescribed but emerges from the balance between the shear stress on a particle and a cohesive stress arising from its interactions with the particles (in its horizon in the PD formulation) and 2) the solid/liquid interface is evolving naturally with the erosion process, rather than being assumed. With this formulation, the yield stress is a variable dependent on the local conditions at the solid/liquid interface, which equals the balancing shear and cohesive stresses.

The diffusion forces  $F_T$ , given by Eq. (1), result from changes in the chemical potential and cause a corresponding stress expressed by (Goodwin and Hughes, 1992)

$$\tau_T = \frac{k_B T}{6\pi h^3} \tag{14}$$

Using the forces  $F_A$  and  $F_R$  given by Eqns. (2) and (3), the cohesive stress is approximated by

$$\tau_c = \frac{F_A + F_R}{S_p} + \tau_T = \frac{F_A + F_R}{S_p} + \frac{k_B T}{6\pi h^3}$$
(15)

The detachment of clay (gel) particles is then postulated to take place if the shear stress induced by the water flow on a particular boundary particle exceeds the cohesive stress.

# 2.3. Migration of sol particles

The detached particles (clay sol) are transported and dispersed in the fluid by mechanisms of advection and dispersion:

$$\frac{\partial \phi_s}{\partial t} = -v_l \nabla \phi_s + \nabla \cdot \left[ (D_d + D_m) \nabla \phi_s \right] \tag{16}$$

where  $D_d$  and  $D_m$  are the diffusion and the mechanical dispersion coefficients of detached particles in the liquid, respectively.

The fluid velocity can be expressed by Darcy's law:

$$v_l = \frac{T_w \eta_w}{\eta_s} \nabla p \tag{17}$$

where p is the fluid pore pressure,  $T_w$  is the fracture transmissivity for water, and  $\eta_s$  is the viscosity of clay sol/gel. The latter is given by (Moreno et al., 2011):

$$\eta_s = \eta_w [1 + 1.022\phi_s^{cov} + 1.358 (\phi_s^{cov})^3]$$
(18)

where the co-volume fraction ( $\phi_s^{cov}$ ) is defined by (Neretnieks et al., 2009):

$$\phi_s^{cov} = \frac{2}{3} \left( D_p + 2z \sqrt{\frac{\varepsilon_0 \varepsilon_R RT}{2cF^2}} \right)^3 \frac{1}{D_p^2 \delta_p} \phi_s \tag{19}$$

The diffusivity of detached particles is calculated based on Stokes-Einstein equation, which is valid for large spherical particles (or molecules). The dispersion coefficient is considered to be a function of the flow velocity. The sum of the diffusion and dispersion coefficients is given by (Bird, 2002):

$$D_l = D_d + D_m = \frac{k_B T}{3\pi \eta_w D_p} + \alpha_L v_l \tag{20}$$

where  $\alpha_L$  is the dispersivity parameter.

The clay erosion is a discontinuous phase change process with highly non-linear coefficients. A non-local formulation development will offer significant benefit by introducing a detachment functions, which enables the discontinuous detachment autonomously evolving without the need of tracking surfaces.

#### 3. Bond-based peridynamics formulation of erosion processes

In peridynamics, a body occupying a region (**R**) is considered to be a collection of infinite particles, as illustrated in Figure 2a. In the bond-based PD, the term 'bond' refers to the interactions between the material points at spatial positions **x** and **x**'. The bond between two interacting material points is called a '*mass bond*' ( $f_h$ ), see Fig. 2. The peridynamic mass flux per unit volume along an 'f-bond' depends on the distance between the points **x** and **x**'. The material point **x** interacts with (and is connected to) all materials points **x**' within a certain finite region  $H_x$ , called the horizon of material point **x**. The radius of the horizon is denoted by  $\delta$ , and  $\xi$  represents the distance vector between the two material points **x** and **x**', i.e.  $\xi = (x' - x)$ .

Corresponding to the three distinctive processes involved in erosion (i.e. free swelling, particle detachment and detached particle transport), three different PD bonds are defined: solid-solid bonds, interfacial bonds and liquid-liquid bonds (illustrated in Fig 2b). The solid-solid bonds control the free swelling process and liquid-liquid bonds govern detached particles transport. The interfacial bonds represent the solid-liquid interaction at the interface and control the detachment process (Jafarzadeh et al.,2019a and 2019b). Once the shear stress exceeds the cohesive stress, the solid particles detaches and become liquid particles. The erosion process will switch from free swelling to detached particle transport as the phase change proceeds. Accordingly, the interface evolution (detachment) is computationally processed in the PD model of erosion. It is noted that the free swelling and detached particle transport follow two different governing equations. Therefore, the particles detachment at the moving interface is considered as a discontinuous process.



(a)



Figure 2. (a) Illustration of peridynamic horizon and force-bonds and (b) particles detachment in peridynamic models (modified from Eriksson and Schatz,2015; Jafarzadeh et al., 2019b)

# 3.1. PD formulation for clay expansion and detachment

Mass conservation equation for the bond between two material points x and x', describes the temporal variation of solid content by (Zhao et al., 2018)

$$\frac{\partial \bar{\phi}_s(\boldsymbol{x}, \boldsymbol{x}', t)}{\partial t} = -\frac{J_s(\boldsymbol{x}, \boldsymbol{x}', t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(21)

where  $\bar{\phi}_s(\mathbf{x}, \mathbf{x}', t)$  is average clay volume fraction along the bond between points  $\mathbf{x}$  and  $\mathbf{x}'$ ,  $J_s(\mathbf{x}, \mathbf{x}', t)$  is the solid mass diffusion flux, and  $\frac{\xi}{\|\xi\|}$  is the unit vector along the bond.

The gradient of clay solid content (clay concentration gradient) between two particles induces clay expansion along the bond connecting these particles. The rate of clay expansion along the bond, i.e. the solid flux  $J_s(\mathbf{x}, \mathbf{x}', t)$ , is (Bobaru and Duangpanya, 2010):

$$J_{s}(\boldsymbol{x},\boldsymbol{x}',t) = -\overline{D}_{s}(\boldsymbol{x},\boldsymbol{x}',t) \frac{\phi_{s}(\boldsymbol{x}',t) - \phi_{s}(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(22)

where  $\phi_s(\mathbf{x}, t)$  and  $\phi_s(\mathbf{x}', t)$  are the clay volume fractions at points  $\mathbf{x}$  and  $\mathbf{x}'$ , respectively, and  $\overline{D}_s(\mathbf{x}, \mathbf{x}', t)$  is the average solid diffusivity between points  $\mathbf{x}$  and  $\mathbf{x}'$ , i.e.  $\overline{D}_s(\mathbf{x}, \mathbf{x}', t) = 0.5[D_s(\mathbf{x}, t) + D_s(\mathbf{x}', t)].$ 

The conservation of mass at point x invloves the fluxes in all the bonds adjacent to x (bonds to particles within the horizon  $H_x$ ) and is obtained by integrating Eq. (21) over the horizon  $H_x$ :

$$\int_{H_x} \frac{\partial \bar{\phi}_s(\boldsymbol{x}, \boldsymbol{x}', t)}{\partial t} \, \mathrm{d} \boldsymbol{V}_{\boldsymbol{x}'} = -\int_{H_x} \frac{J_s(\boldsymbol{x}, \boldsymbol{x}', t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} \, \mathrm{d} \boldsymbol{V}_{\boldsymbol{x}'} \tag{23}$$

where  $V_{x'}$  is the horizon volume (area in 2D, length in 1D) of particle x'.

The relationship between the concentration at point x and time t and the average concentration in all the bonds adjacent to x is given by (Bobaru and Duangpanya, 2010):

$$\int_{H_{\chi}} \frac{\partial \bar{\phi}_{s}(\boldsymbol{x}, \boldsymbol{x}', t)}{\partial t} \, \mathrm{d}\boldsymbol{V}_{\boldsymbol{x}'} = \frac{\partial \phi_{s}(\boldsymbol{x}, t)}{\partial t} \, \boldsymbol{V}_{\boldsymbol{H}_{\chi}} \tag{24}$$

where  $V_{H_x}$  is the horizon volume of particle x.

Combining Eqns. (22)-(24) leads to the following equation for the evolution of clay solid content at particle x

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \int_{H_x} \frac{\overline{D}_s(\boldsymbol{x},\boldsymbol{x}',t)}{\boldsymbol{V}_{H_x}} \cdot \frac{\phi_s(\boldsymbol{x}',t) - \phi_s(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|^2} \, \mathrm{d}\boldsymbol{V}_{\boldsymbol{x}'}$$
(25)

Clay particles detachment at the solid-liquid interface is related in this work to force bond damage, as previously modelled in peridynamics (Silling and Bobaru, 2005, Bobaru and Duangpanya, 2012). A similar concept of using concentration-dependent damage to simulate a corrosion process via a peridynamic nonlinear diffusion equation is presented by Jafarzadeh et al. (2019a and 2019b). A phase change process autonomously evolves when concentration in the solid drops below the concentration in the electrolyte; and phase change evolving is governed by solid-liquid bonds. Similarly, solid-liquid bonds used in this study control the detachment processes. However, different definitions for solid-solid and liquid-liquid bonds are used in this study. Solid-solid bonds and liquid-liquid bonds govern swelling process and detached particles transport, respectively. The detachment of clay particles is postulated to occur when the shear stress ( $\tau_f$ ) induced by the flow at gel-fluid boundary exceeds the cohesive stress ( $\tau_c$ ) given by Eq. (15). To establish the relation, we introduce a detachment function:

$$\mu(\mathbf{x}, \mathbf{x}', t) = \begin{cases} 1 & \tau_c \ge \tau_f \\ 0 & \tau_c < \tau_f \end{cases}$$
(26)

The detachment function implies that when the cohesive stress calculated from the mass bonds of solid-solid points is smaller than the shear stress induced by the seeping water, the solid material points change to liquid points and follow the governing equations of transport processes. Incorporating the detachment function  $\mu(\mathbf{x}, \mathbf{x}', t)$  in Eq. (25) leads to the peridynamic equation for coupled clay expansion and particles detachment:

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \int_{H_x} \mu(\boldsymbol{x},\boldsymbol{x}',t) \frac{\overline{D}_s(\boldsymbol{x},\boldsymbol{x}',t)}{\boldsymbol{V}_{H_x}} \cdot \frac{\phi_s(\boldsymbol{x}',t) - \phi_s(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|^2} d\boldsymbol{V}_{\boldsymbol{x}'}$$
(27)

The peridynamics microscopic diffusivity is defined as:

$$d_s(\mathbf{x}, \mathbf{x}', t) = \frac{\overline{D}_s(\mathbf{x}, \mathbf{x}', t)}{V_{H_x}}$$
(28)

which transforms Eq. (27) into:

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \int_{H_x} \mu(\boldsymbol{x}, \boldsymbol{x}', t) d_s(\boldsymbol{x}, \boldsymbol{x}', t) \frac{\phi_s(\boldsymbol{x}', t) - \phi_s(\boldsymbol{x}, t)}{\|\boldsymbol{\xi}\|^2} \,\mathrm{d}\boldsymbol{V}_{\boldsymbol{x}'}$$
(29)

PD microscopic diffusivity is determined from measurable material diffusivity. Using the approach proposed by Bobaru and Duangpanya (2010) we derive the relationships between PD microscopic diffusivity and macroscopic diffusivity for 1D and 2D deformation as:

$$d_s(\mathbf{x}, \mathbf{x}', t) = \frac{D_s}{\delta}$$
(30)

$$d_s(\mathbf{x}, \mathbf{x}', t) = \frac{4D_s}{\pi\delta^2} \tag{31}$$

Details of the derivations of Eqns (30) and (31) are provided in supplementary material (S1).

## 3.2. PD formulation for transport of detached particles in water

The mass conservation equation, Eq. 16, for the bond along the two material points x and x' can be presented within the PD formulation as (Zhao et al., 2018)

$$\frac{\partial \bar{\phi}_s(\boldsymbol{x}, \boldsymbol{x}', t)}{\partial t} = -\frac{\left[J_l^d(\boldsymbol{x}, \boldsymbol{x}', t) + J_l^a(\boldsymbol{x}, \boldsymbol{x}', t)\right]}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(32)

where  $J_l^d(\mathbf{x}, \mathbf{x}', t)$  is the mass flux caused by dispersion and  $J_l^a(\mathbf{x}, \mathbf{x}', t)$  is the mass flux caused by advection. Similarly, to Eq. (22), the PD dispersion flux  $J_l^d(\mathbf{x}, \mathbf{x}', t)$  is written as follow:

$$J_{l}^{d}(\boldsymbol{x}, \boldsymbol{x}', t) = -D_{l}(\boldsymbol{x}, \boldsymbol{x}', t) \frac{\phi_{s}(\boldsymbol{x}', t) - \phi_{s}(\boldsymbol{x}, t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(33)

where  $D_l(x, x', t)$  is the diffusivity of the 'm-bond' between the material points of x and x'. The PD advection flux  $J_l^a(x, x', t)$  is given by the equations (Zhao et al., 2018):

$$J_{l}^{a}(\mathbf{x}, \mathbf{x}', t) = V_{l}(\mathbf{x}, \mathbf{x}', t) [\phi_{s}(\mathbf{x}', t) - \phi_{s}(\mathbf{x}, t)] \cdot \frac{\xi}{\|\xi\|}$$
(34)

where  $V_l(x, x', t)$  is the advection of the 'm-bond' between the material points of x and x'. Integrating over the horizon region of point x,  $H_x$ , on both sides of Eq. (32) gives

$$\int_{H_{x}} \frac{\partial \bar{\phi}_{s}(\boldsymbol{x}, \boldsymbol{x}', t)}{\partial t} \, \mathrm{d}\boldsymbol{V}_{\boldsymbol{x}'} = -\int_{H_{x}} \frac{\left[J_{l}^{d}(\boldsymbol{x}, \boldsymbol{x}', t) + J_{l}^{a}(\boldsymbol{x}, \boldsymbol{x}', t)\right]}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} \, \mathrm{d}\boldsymbol{V}_{\boldsymbol{x}'} \tag{35}$$

Substituting Eqns. (24), (33) and (34) into Eq. (35), gives the peridynamics description of transport of detached clay particles:

$$\frac{\partial \phi_{s}(\boldsymbol{x},t)}{\partial t} = \int_{H_{\boldsymbol{x}}} \frac{D_{l}(\boldsymbol{x},\boldsymbol{x}',t)}{\boldsymbol{V}_{H_{\boldsymbol{x}}}} \frac{\phi_{s}(\boldsymbol{x}',t) - \phi_{s}(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|^{2}} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{\boldsymbol{x}'} - \int_{H_{\boldsymbol{x}}} \frac{V_{l}(\boldsymbol{x},\boldsymbol{x}',t)}{\boldsymbol{V}_{H_{\boldsymbol{x}}}} \frac{\phi_{s}(\boldsymbol{x}',t) - \phi_{s}(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{\boldsymbol{x}'}$$
(36)

The peridynamics microscopic diffusivity and microscopic advection of detached particles are defined as:

$$d_{l}(x, x', t) = \frac{D_{l}(x, x', t)}{V_{H_{x}}}$$
(37)

$$v_l(\boldsymbol{x}, \boldsymbol{x}', t) = \frac{V_l(\boldsymbol{x}, \boldsymbol{x}', t)}{V_{H_x}}$$
(38)

which transforms Eq. (35) into:

$$\frac{\partial \phi_{s}(\mathbf{x},t)}{\partial t} = \int_{H_{x}} d_{l}(\mathbf{x},\mathbf{x}',t) \frac{\phi_{s}(\mathbf{x}',t) - \phi_{s}(\mathbf{x},t)}{\|\boldsymbol{\xi}\|^{2}} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{\mathbf{x}'} - \int_{H_{x}} v_{l}(\mathbf{x},\mathbf{x}',t) \frac{\phi_{s}(\mathbf{x}',t) - \phi_{s}(\mathbf{x},t)}{\|\boldsymbol{\xi}\|} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{\mathbf{x}'}$$
(39)

PD microscopic diffusivity for 1D and 2D cases is given in Eqns. (30) and (31). By using the same approach for the proposed of microscopic diffusivity we derive the relationships between PD microscopic advection and macroscopic advection for 1D and 2D case as:

$$v_l(\boldsymbol{x}, \boldsymbol{x}', t) = \frac{V_l}{\delta}$$
(40)

$$v_l(\mathbf{x}, \mathbf{x}', t) = \frac{4V_s}{\pi\delta^2} \tag{41}$$

Details of the derivations for Eqns. (40) and (41) are provided in supplementary materials (S2). Notably, the newly derived peridynamics formulations for clay solid swelling and detached clay particles transport, Eqns. (29) and (39), do not involve any spatial derivatives. This indicates that the proposed PD formulations can be applied everywhere in the domain including in cases of discontinuities and heterogeneities of the parameters (i.e.  $d_s$ ,  $d_l$  an  $v_l$ ) and discontinuities and heterogeneities of the domain.

#### 3.3. Numerical solution of the PD formulations

Numerical solutions of the equations describing clay expansion, Eq. (25), and transport of detached clay particles with the fluid, Eq. (35), are developed for infinite number of particles. The domain is discretized using a uniform grid with spacing  $\Delta$ , and particles are placed within the grid cells. A finite number of nodes is obtained as the result of the spatial discretization. Each particle/cell has an associated volume  $V_{ip}$  (length in 1D and area in 2D). In this study, the horizon size of  $\delta=3\Delta$  has been selected. Figure 3 illustrates a discretization in the vicinity of a node  $x_i$  in a 2D domain.  $\xi_{ip}$  represents the distance vector between the two material points  $x_i$  and  $x_p$ .



Figure 3. Illustration of discretization around node *x* in a 2D domain.

The spatial discretization of Eqns. (25) and (35), using the mid-point rule, are:

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \sum_p \frac{d_s(\boldsymbol{x}_i,t) + d_s(\boldsymbol{x}_p,t)}{2} \frac{\phi_s(\boldsymbol{x}_p,t) - \phi_s(\boldsymbol{x}_i,t)}{\|\boldsymbol{x}_p - \boldsymbol{x}_i\|^2} V_{ip}$$
(42)

and

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \sum_p \left[ \left( \frac{d_l(\boldsymbol{x}_i,t) + d_l(\boldsymbol{x}_p,t)}{2} \right) \frac{\phi_s(\boldsymbol{x}_p,t) - \phi_s(\boldsymbol{x}_i,t)}{\|\boldsymbol{x}_p - \boldsymbol{x}_i\|^2} \right] V_{ip} - \sum_p \left[ \left( \frac{v_l(\boldsymbol{x}_i,t) + v_l(\boldsymbol{x}_p,t)}{2} \right) \frac{\phi_s(\boldsymbol{x}_p,t) - \phi_s(\boldsymbol{x}_i,t)}{\|\boldsymbol{x}_p - \boldsymbol{x}_i\|} \right] V_{ip}$$

$$(43)$$

The forward Euler method is used for the time integration:

$$\phi_s(x_i, t^{n+1}) = \phi_s(x_i, t^n) + \dot{\phi}_s(x_i, t^n) \Delta t$$
(44)

Supplementary S3 provides the details of the numerical solution applied to the 1D Peridynamics. Supplementary S4 also provides the flowchart of erosion calculation.

#### 4. Results and discussion

# 4.1. Validation of the cohesive stress proposal

We first present a validation of the proposed cohesive stress formulation by comparison with experimental studies reported by Eriksson and Schatz (2015). The cohesive stress induced by the different particle interaction forces is calculated by Eqns. (2), (3), (14) and (15). The overall cohesive stress emerges as a combination of the forces. The experimental study reported by Eriksson and Schatz (2015) involved samples of clay with solid contents ranging from 0.1% to 15% in aqueous solutions containing 1 mM and 17 mM NaCl. The materials properties and parameters used in this section are taken from several literature sources and shown in Table 1.

Figure 4 shows the calculated cohesive stress as a function of particle distance for the case of the system containing 1mM and 17 mM NaCl (as an example to demonstrate the component of cohesive stress model). The repulsive forces ( $F_R$ ) are larger than attractive and diffusive forces ( $F_A$ ,  $F_T$ ) at small distances, and become very weak as the distance between the particles increases. The overall cohesive stress ( $\tau_c$ ) emerges as a combination of the forces. It is noted that  $\tau_c$  increases with the increase of the ion concentration as the repulsive forces ( $F_R$ ) is governed by the ion concentration.

It has been reported that that the thickness of a single particle  $(\delta_p)$  for a sodium smectite (WyNa) ranges from 0.6 nm to 2.4 nm (Cadene et al., 2005). Therefore, the simulations were performed for a rage of particle thickness including 0.6, 0.8,1, 1.2 and 2 nm. Figure 5 shows the cohesive stress calculated by the proposed formulation, Eq. (15), with solid lines, and the yield stress observed by experimental data with symbols. The experimental observations indicated that for solid contents below 4% (grey area in Figure 5), the clay exhibits the properties of sol, which is most susceptible to erosion by water flow (Neretnieks et al., 2009, Abend and Lagaly, 2000, Michot et al., 2004). The modelling results are in close agreement with the data, which provides confidence in the realism of the cohesive stress formulation – a key component of the detachment model.

		unit	value
Particle surface area <sup>a</sup>	$S_p$	$m^2$	9×10 <sup>-14</sup>
Particle diameter <sup>b</sup>	$D_p$	nm	200
Particle thickness <sup>a</sup>	$\delta_p$	nm	1.0
Maximum solid volume fraction <sup>a</sup>	$\phi_s^{max}$	-	1.0
Surface charge of particles <sup>a</sup>	$\sigma^0$	$Cm^{-2}$	-0.131
Ionic valence <sup>c</sup>	Ζ	-	1
Viscosity of water	$\eta_w$	$Nsm^{-2}$	1.002×10 <sup>-3</sup>
Relative permittivity of water	$\mathcal{E}_r$		78.54
Permittivity of vacuum	$\varepsilon_0$	$Fm^{-1}$	8.85×10 <sup>-12</sup>
Gas constant	R	$JKmol^{-1}$	8.134
Temperature	Т	K	298
Faraday's constant	F	$Cmol^{-1}$	96485
Boltzmann's constant	$k_B$	$JK^{-1}$	1.38×10 <sup>-23</sup>
Hamaker constant	$A_H$	J	$2.5k_BT$
Kozenys' constant <sup>a,c</sup>	$k_0 \tau^2$	-	5-13

Table 1. Material Properties and parameter

<sup>a</sup> Liu et al. (2009a); <sup>b</sup> Kelessidis and Maglione (2008); <sup>c</sup> Schatz el al. (2013)



Figure 4. Variations of cohesive stress and its components with particle distance for (a) c=1mM and (b) c=17mM



Figure 5. Yield stress dependence on the solid fraction calculated for (a) c=1mM and (b) c=17mM by the proposed model (line) and Experimental data (scatter) by Eriksson and Schatz (2015)

# 4.2. Critical solid volume fraction at the solid/liquid interface

The critical solid volume fraction is defined as the volume fraction at which the shear stress equals the cohesive stress. This clearly depends on the hydro-chemical conditions, so it is not a material-only dependent constant. Equating Eqns. (13) and (15) provides a critical distance between the particles,  $h_{cr}$ , from the equation:

$$\frac{F_A + F_R}{S_p} + \frac{k_B T}{6\pi h_{cr}^3} = \left(\frac{6\pi \eta_w D_p}{S_p}\right) v_l \tag{45}$$

The critical solid volume fraction,  $\phi_s^{cr}$ , is obtained from critical distance and Eq. (12):

$$\phi_s^{cr} = \phi_s^{max} \left( \frac{h_{cr}}{\delta_p} + 1 \right) \tag{46}$$

The critical soil volume fraction at the solid/liquid interface is calculated for sodium rich smectite clay with parameters and properties given in Table 1 and for a number of hydrochemical conditions - water velocity and ionic concentration. The results are presented in Fig. 6 and compared with those calculated by Neretnieks et.al. (2017) for the same conditions and material properties. It can be seen that the calculated values are in very close agreement with the experiments. Small differences can be observed only for intermediate ionic concentrations, but these are in the order of previously reported ranges of critical fractions. In can be concluded that the proposed detachment model captures successfully the effects of ionic concentration and fluid velocity on the critical solid volume fraction.



Figure 6. Critical solid volume fraction at the solid/liquid interface calculated using the proposed detachment model compared to experimental data by Neretnieks et al. (2017)

#### 4.3. Behaviour of compacted bentonite under eroding environment

A series of experimental work on erosion of compacted bentonite by Schatz el al. (2013) are used to test the accuracy of the prediction by the PD model. Erosion of 2cm diameter discs of compacted sodium rich bentonite is analysed which is exposed to either a stagnant or flowing water through a 1.0 mm thick artificial fracture(as shown in Fig. 7). Three experimental tests reported by were selected for the simulations:

- *Test 1* includes the expansion of clay in contact with deionised water under stagnant condition (no flow). A very low value for ionic concentration (0.05 mM) is considered for this case (Neretnieks et al., 2017).
- *Test 2* is the expansion and subsequent erosion of the clay disc by the flowing deionised water (average flow rate of 0.38 ml/min).
- *Test 3* represents the expansion and erosion of clay due to exposure to groundwater solution (0.04 g/L NaCl and average flow rate of 0.09 ml/min).

The domain is 24 cm × 24 cm square. The cell size,  $\Delta$ , and the horizon,  $\delta$ , are 0.12 cm and 0.36 cm, respectively. The material parameters used are provided in Table 1. The initial bentonite fraction in the domain outside of the sample is set to zero. The solid fraction and height of the initial experimental sample is 0.6 and 2cm, respectively. The solid fraction of the initial sample is assumed to be constant as the height of sample is 20 times larger than that of fracture thickness.

The left boundary (x=-12 cm) was kept at zero solid fraction and the particles would exit through the right boundary (x=12 cm). Impervious boundary conditions were prescribed at the domain boundaries y=-12 cm and y=12cm. Water inlet and outlet boundary conditions include five input and output points in the domain. Constant heads are imposed at the left and right borders; creating a steady water flow from left to the right in the domain.



Figure 7. Schematic of the system used for erosion of compacted bentonite: (left) side view showing the clay sample and fracture; (right) plan view with boundary conditions.

Figure 8 presents the solid volume fraction profile along the centre line at x=0 for the three tests analysed by the PD model. The results are compared with experimental results reported by (Schatz el al., 2013) for *Test* 1 and *Test 3* for which experimental data reported by Schatz el al., 2013). The results of the model correlate with the experiments with regards to the extrusion distance. There is no experimental data for *Test* 2, but the calculations shown in Fig. 8b show similar trend to those in Fig. 8c. As shown in Fig. 8, (i) the solid volume fraction decreases with distance from the solid/liquid interface (as expected) and (ii)the decrease is slower for stagnant water (*Test 1*) compared to flowing water (*Tes2 and Test 3*) (which agrees with experiments).


(c)

Figure 8. Solid volume fraction along the centre line at x=0 (a)*Test 1* (b)*Test 2* and (c)*Test 3* 

Figures 9 and 10 illustrate the evolution of swelling and erosion observed in the experiment and predicted by the model. It is noted that the images of the experimental tests used for comparison do not provide the evolution in the full domain. Although the simulations included the entire domain (i.e. outside detachment zone), the visual comparison is only possible for the areas where images were available.

The profiles of solid volume fraction over time obtained by the experiments and those obtained by PD are in close agreement. Figure 9 describes the evolution of the swelling front under hydration process whilst Figure 10, illustrates the combined effects when the eroding solid has been co-transferred with the water. These observations are also in agreement with the results shown in Fig. 8c. The discontinuous detachment interface is marked by dash line. It is shown that the modelling results are much smoother compared to sharp interfaces which are evidently observed by the experiment (Figs.8b-8c and Figs. 10c-10d). The difference may be caused by the sedimentation of aggregate, the formation of flocs and experimental artifacts (e.g., transport of initially loosely bound material) (Neretnieks and Moreno, 2018), which are not considered in the current model. It is noted that in *Test 2* and *Test 3* there is a difference between the outflow boundary condition of the experiment (outlet boundary is effectively a narrow point) and the model (the entire outer boundary is open to flow). This difference is believed to have limited impact due to the length of the outlet from the solid/water interface.



Figure 9. Clay swelling in *Test 1*: comparisons between simulations (bottom) and experiment (top) at (a) 24h, (b) 168h, (c) 384h and (d) 720h.



Figure 10. Clay erosion in *Test 3*: comparisons between simulations (bottom) and experiment (top) at (a) 24h, (b)168h, (c) 288h and (d) 504h



Figure 11. Time evolution of extrusion distance *Test 1*, *Test 2* and *Test 3*: simulations with solid lines and experiments with symbols

Comparisons between the average extrusion distances into the fracture obtained by PD and experiment for *Test 1*, *Test 2* and *Test 3* are shown in Fig. 11. The results of the experiment and simulations correlate well for all cases. The comparison for *Test 1* indicates the accuracy of the proposed model to simulate clay free swelling in stagnant water for the case study. The comparisons for *Test 2* and *Test 3* support further the model's accuracy to account for the hydro-chemical effects on bentonite clay erosion. Theoretically, *Test 2* with deionised water

should show larger expansion than *Test 3* with groundwater, because of the lower ionic concentration, which affects the repulsive double layer forces in Eq. (3) and the cohesive stress in Eq. (15) and Fig. 1. However, the flow rate in *Test 2* is 4.2 times larger than that in *Test 3*. This leads to a larger shear stress, which reduces the penetration distance, Eq. (13). This demonstrates how the proposed model captures the complex interplay between hydrodynamics and water chemistry to provide evolution in close agreement with the experimental data.

## 5. Conclusions

A bond-based Peridynamics formulation for clay erosion was presented. It includes descriptions of i) the free swelling of clay as a diffusion process and ii) the transport of detached particles as an advection-dispersion process, with an important addition of a new detachment model for separation of particles from the gel.

The physical realism of the new detachment model was tested against a set of experimental data. It was shown, that the model captured the impacts of ionic strength and flow rate on the detachment of a smectite-rich clay with good accuracy. The results highlighted that increasing the ionic concentrations and velocities of the eroding fluid leads to increasing critical clay volume fraction at the solid/liquid interface;

For validation, the full erosion model was applied to cases with available experimental data for erosion of compacted bentonite under three hydro-chemical conditions. Firstly, it was shown that the diffusion model was appropriate for representing the free swelling as the simulation results were in close agreements with the experimental data if *Test 1*. Secondly, by comparisons with *Test 2* and *Test 3* it was shown that the full erosion model accounted realistically for the hydro-chemical conditions of the eroding fluid.

In summary, it has been demonstrated that the proposed non-local Peridynamics-based framework is efficient and effective for analysis of hydro-chemical effect on clay erosion. Specifically, the model is able to capture the complexity of the problem arising from highly non-linear physics (input parameters vary over several orders of magnitude) and geometry (moving gel-fluid interface), as well as the discontinuous process of particle detachment. Noting the PD applications to a range of problems with discontinuity, large deformation, non-linearity and heterogeneity in solid mechanics, the existing applications to problems in soil mechanics are very limited and mainly related to granular materials. The model presented here is the first PD model related to clay behaviour, for which the effects of coupled phenomena on the overall behaviour are inherently stronger than on the behaviour of coarse-grained soils.

## Nomenclature

$A_H$	Hamaker constant
$a_p$	Specific surface area
С	Ionic concentration
$D_d$	Diffusion coefficients
$D_m$	Mechanical dispersion coefficients
$D_p$	particle diameter
F	Faraday's constant
$F_A$	Attractive Van der Walls forces
$F_T$	The diffusion forces
$F_R$	Repulsive electrical double layer forces
$f_r$	The friction coefficient
h	Separation distance between the flat particles
$k_B$	Boltzmann constant
р	The fluid pore pressure
R	Universal gas constant
r <sub>eq</sub>	Equivalent radius of the non-spherical particles
$S_p$	The surface area of the particles
Т	Absolute temperature
$T_w$	Fracture transmissivity for water
$v_l$	Fluid velocity.
$V_p$	Volume of the particles
$y^m$	The scaled potential
$y^0_\infty$	Dimensionless surface potential
Ζ	Valence of ions
κ	Debye length
$\alpha_L$	the dispersivity parameter
$ au_f$	The shear stress
$ au_c$	The cohesive stress
τ	The tortuosity
$\varepsilon_0 \varepsilon_R$	Dielectric constant
χ	The sum of the particles' energies
$\eta_w$	The dynamic viscosity of water

$\eta_s$	The viscosity of clay sol/gel
$\sigma^0$	Surface charge density
$\phi_s$	The clay (solid) volume fraction
$\phi_{\scriptscriptstyle R}$	solid volume fraction at rim
$\phi_s^{cov}$	Co-volume fraction
$\phi_s^{max}$	The maximum clay solid volume fraction
$\delta_p$	The particle thickness
δ	The radius of the horizon
$f_h$	Mass bond
$H_x$	The horizon of material point $\boldsymbol{x}$
ξ	The distance vector between the two material points
$\phi_s(\boldsymbol{x},t)$	The clay volume fractions at points $\boldsymbol{x}$
$\phi_s(\pmb{x}',t)$	The clay volume fractions at points $\boldsymbol{x}$ `
$\bar{\phi}_s(\pmb{x},\pmb{x}',t)$	Average clay volume fraction
$V_{x'}$	Horizon volume of particle $x'$
$J_l^a(\boldsymbol{x}, \boldsymbol{x}', t)$	Mass flux caused by advection
$J_l^d(\boldsymbol{x}, \boldsymbol{x}', t)$	Mass flux caused by dispersion
$J_s(\boldsymbol{x}, \boldsymbol{x}', t)$	The solid mass diffusion flux
$d_l(\boldsymbol{x}, \boldsymbol{x}', t)$	Microscopic diffusivity of detached particles
$d_s(\mathbf{x}, \mathbf{x}', t)$	Microscopic diffusivity of solid
$v_l(\boldsymbol{x}, \boldsymbol{x}', t)$	Microscopic advection of detached particles
$\mu(\pmb{x}, \pmb{x}', t)$	The detachment function

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### Supplementary materials (Section 5.3)

## S1: Derivation of microscopic diffusivity

The microscopic diffusivity function introduced in the manuscript is given as follow:

$$d_s(\mathbf{x}, \mathbf{x}', t) = \frac{\overline{D}_s(\mathbf{x}, \mathbf{x}', t)}{V_{H_x}}$$
(S1-1)

In order to determine the microscopic diffusivity, Eq. (S1-1), from measurable (apparent) diffusivity, we follow the approach proposed by Bobaru and Duangpanya (2010). We consider the pure diffusion case with a constant diffusivity in a homogeneous and isotropic porous medium. The peridynamics pure diffusive flux  $(J_s^{PD}(x,t))$  and the local pure diffusive flux  $(J_s(x))$  are equal under steady-state conditions. This enables the calculation of the peridynamics micro-diffusivity. Based on Fick's law of diffusion, the steady-state concentration and the flux of pure diffusion at position x can be given by:

$$\phi_s(x) = mx + n \tag{S1-2}$$

$$J_s(x) = -D_s \frac{\partial \phi_s}{\partial x} = -mD_s \tag{S1-3}$$

where, m and n are constants.  $J_s(x)$  is the classic solid phase diffusive flux.

The peridynamics mass flux  $(J_s(x, x', t))$  between two material points x and x' is given in the manuscript as follow:

$$J_{s}(\boldsymbol{x}, \boldsymbol{x}', t) = -\overline{D}_{s}(\boldsymbol{x}, \boldsymbol{x}', t) \frac{\phi_{s}(\boldsymbol{x}', t) - \phi_{s}(\boldsymbol{x}, t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(S1-4)

In order to derive the peridynamics mass flux in all the bonds attached to x in its horizon  $H_x$ , Eq. (S1-4) is integrated over the horizon of material point x. For the 1D and 2D diffusion cases, the peridynamics' mass flux can be expressed as:

$$J_{s}^{PD}(x,t) = \int_{0}^{\delta} d_{s}(x,x',t) \frac{\phi_{s}(x',t) - \phi_{s}(x,t)}{\|x' - x\|} dx$$
(S1-5)

$$J_{s}^{PD}(x,t) = \int_{-\pi/2}^{\pi/2} \int_{0}^{\delta} d_{s}(x,x',t) \frac{\phi_{s}(x',t) - \phi_{s}(x,t)}{\|x' - x\|} \cos^{2}(\phi_{s}) r dr d\varphi$$
(S1-6)

By substituting the uniform influence function  $(d_s(\mathbf{x}, \mathbf{x}', t) = d_s)$  (Silling et al., 2007) and the steady-state concentration equation, Eq. (S1-2), into Eqns. (S1-5) and (S1-6), we obtain:

$$J_s^{PD}(x,t) = -md_s\delta \tag{S1-7}$$

$$J_s^{PD}(x,t) = -m\pi d_s \delta^2/4 \tag{S1-8}$$

If the local mass flux, Eq. (S1-3), and peridynamics mass flux, Eqns. (S1-7) and (S1-8), at material point x are equal, the micro-diffusivity for 1D and 2D transport cases are obtained as:

$$d_s(\mathbf{x}, \mathbf{x}', t) = \frac{D_s}{\delta}$$
(S1-9)

$$d_s(\boldsymbol{x}, \boldsymbol{x}', t) = \frac{4D_s}{\pi\delta^2} \tag{S1-10}$$

#### S2: Derivation of microscopic advection

The microscopic advection function introduced in the manuscript is given as follow:

$$v_l(x, x', t) = \frac{V_l(x, x', t)}{V_{H_x}}$$
 (S2-1)

In order to determine the microscopic advection function introduced in Eq. (S2-1) from measurable material advection, we consider the mass transfer with a constant flow in a homogeneous and isotropic porous medium. Using a linear approximation, the initial solid volume fraction can be described as:

$$\phi_s(x,0) = mx + n \tag{S2-2}$$

The distribution of  $\phi_s$  with respect to time t for a constant flow  $(V_l)$  can be given as:

$$\phi_s(x,t) = m(x - V_l t) + n$$
 (S2-3)

From mass conservation, we obtain:

$$\frac{\partial \phi_s}{\partial t} = -mV_l \tag{S2-4}$$

The peridynamics mass transport induced by pure advection can be expressed as

$$\frac{\partial \phi_s(\mathbf{x},t)}{\partial t} = -\int_{H_x} v_l(\mathbf{x},\mathbf{x}',t) \frac{\phi_s(\mathbf{x}',t) - \phi_s(\mathbf{x},t)}{\|\boldsymbol{\xi}\|} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{\mathbf{x}'}$$
(S2-5)

By substituting the uniform influence function (i.e.  $v_l(x, x', t) = v_l$ ), the peridynamic mass transport equations for 1D and 2D, S2-5, can be expressed by

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = -\int_{H_x} v_l \frac{\phi_s(\boldsymbol{x}',t) - \phi_s(\boldsymbol{x},t)}{(\boldsymbol{x}'-\boldsymbol{x})} d\boldsymbol{x}'$$
(S2-6)

$$\frac{\partial \phi_s(\boldsymbol{x}, t)}{\partial t} = -\int_{H_x} \boldsymbol{v}_l \frac{\phi_s(\boldsymbol{x}', t) - \phi_s(\boldsymbol{x}, t)}{\|\boldsymbol{x}' - \boldsymbol{x}\|} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{A}_{\boldsymbol{x}'}$$
(S2-7)

For the 1D advection cases, substituting Eqns. (S2-3) into Eq. (S2-6), we obtain:

$$\frac{\partial \phi_s(x,t)}{\partial t} = -\int_0^\delta v_l \frac{m(x'-x)}{(x'-x)} dx'$$
(S2-8)

By enforcing a match between Eq.(S2-4) and (S2-8), the peridynamic 1-D micro-advection can be expressed by

$$v_l = \frac{V_l}{\delta} \tag{S2-9}$$

For the 2D advection cases, we consider the advection vector with components in x and y directions,  $v_x$  and  $v_y$ , respectively. Eq.(S2-7) becomes

$$\frac{\partial \phi_s(\mathbf{x},t)}{\partial t} = -\int_{H_x} (v_x \cos \alpha + v_y \sin \alpha) \frac{\phi_s(\mathbf{x}',t) - \phi_s(\mathbf{x},t)}{\|\mathbf{x}' - \mathbf{x}\|} d\mathbf{A}_{\mathbf{x}'}$$
(S2-10)

where  $\alpha$  is the angle between  $v_l$  and  $v_x$ .

By using the uniform influence function, the peridynamic mass transport equation, S2-10, can be expressed as:

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = -\int_{H_x} (\frac{v_x \cos^2 \alpha}{x^2 - x} + \frac{v_y \sin^2 \alpha}{y^2 - y}) [\phi_s(\boldsymbol{x}',t) - \phi_s(\boldsymbol{x},t)] \mathrm{d}\boldsymbol{A}_{\boldsymbol{x}'}$$
(S2-11)

Considering the uniform flow in x direction, we have  $v_y = 0$ , thus

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = -\int_{H_x} (\frac{v_x \cos^2 \alpha}{x - x}) [\phi_s(\boldsymbol{x}',t) - \phi_s(\boldsymbol{x},t)] d\boldsymbol{A}_{\boldsymbol{x}'}$$
(S2-12)

Substituting Eqns.(S2-3) and (S2-4) into Eq.(S2-12), we obtaine:

$$mV_x = \int_{H_x} mv_x \cos^2 \alpha \, \mathrm{d}\mathbf{A}_{x'} \tag{S2-13}$$

The micro-advection for 2D transport cases are then calculated to be:

$$v_x = \frac{V_x}{\int_{H_x} \cos^2 \alpha \, \mathrm{d}\mathbf{A}_{x'}} = \frac{4V_x}{\pi\delta^2}$$
(S2-14)

Similarly, if we consider the same uniform flow in y direction, we have

$$v_y = \frac{V_y}{\int_{H_x} \sin^2 \alpha \, \mathrm{d} A_{x'}} = \frac{4V_y}{\pi \delta^2}$$
(S2-15)

#### S3: Details of the numerical implementation

In order to illustrate the numerical implementation, the solution applied to the 1D Peridynamics is presented. In this study, the horizon size of  $\delta=3\Delta$  is used.  $\Delta$  is the spacing between the material points. The material point of interest  $\mathbf{x}$  is denoted by  $x_i$  which interacts with the three points to its left and right. The spatial discretization of PD free swelling and detached particles transport, using the mid-point rule, are given as follow:

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \sum_p \frac{d_s(\boldsymbol{x}_i,t) + d_s(\boldsymbol{x}_p,t)}{2} \frac{\phi_s(\boldsymbol{x}_p,t) - \phi_s(\boldsymbol{x}_i,t)}{\|\boldsymbol{x}_p - \boldsymbol{x}_i\|^2} V_{ip}$$
(S3-1)

and

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \sum_p \left[ \left( \frac{d_l(\boldsymbol{x}_i,t) + d_l(\boldsymbol{x}_p,t)}{2} \right) \frac{\phi_s(\boldsymbol{x}_p,t) - \phi_s(\boldsymbol{x}_i,t)}{\|\boldsymbol{x}_p - \boldsymbol{x}_i\|^2} \right] V_{ip} - \sum_p \left[ \left( \frac{v_l(\boldsymbol{x}_i,t) + v_l(\boldsymbol{x}_p,t)}{2} \right) \frac{\phi_s(\boldsymbol{x}_p,t) - \phi_s(\boldsymbol{x}_i,t)}{\|\boldsymbol{x}_p - \boldsymbol{x}_i\|} \right] V_{ip}$$
(S3-2)

Hence, the set point  $\mathbf{x}$ ` from left to right within the horizon of  $x_i$  are  $x_{i-3}$ ,  $x_{i-2}$ ,  $x_{i-1}$ ,  $x_{i+1}$ ,  $x_{i+2}$ ,  $x_{i+3}$ . Eqns. (S3-1) and (S3-2), at the n<sup>th</sup> time step (i.e., the current time step), can be presented in a discretised form as:

$$\left[\frac{\phi_{s}(x_{i},t^{n+1})-\phi_{s}(x_{i},t^{n})}{\Delta t}\right] = \left[\mathbf{K}\right] \begin{bmatrix} \phi_{s}(x_{i+3},t^{n})-\phi_{s}(x_{i},t^{n})\\ \phi_{s}(x_{i+2},t^{n})-\phi_{s}(x_{i},t^{n})\\ \phi_{s}(x_{i+1},t^{n})-\phi_{s}(x_{i},t^{n})\\ \phi_{s}(x_{i-1},t^{n})-\phi_{s}(x_{i},t^{n})\\ \phi_{s}(x_{i-2},t^{n})-\phi_{s}(x_{i},t^{n})\\ \phi_{s}(x_{i-3},t^{n})-\phi_{s}(x_{i},t^{n}) \end{bmatrix}$$
(S3-4)

and

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$$[\mathbf{K}] = \begin{bmatrix} k_{s,3} & k_{s,2} & k_{s,1} & k_{s,-1} & k_{s,-2} & k_{s,-3} \\ k_{l,3} & k_{l,2} & k_{l,1} & k_{l,-1} & k_{l,-2} & k_{l,-3} \end{bmatrix}$$

where

$$k_{s,3} = \frac{d_s(x_{i+3}, t^n) + d_s(x_i, t^n)}{2} \frac{V_{i+3}}{\|x_{i+3} - x_i\|^2}$$
(S3-5)

$$k_{s,2} = \frac{d_s(x_{i+2}, t^n) + d_s(x_i, t^n)}{2} \frac{V_{i+2}}{\|x_{i+3} - x_i\|^2}$$
(S3-6)

$$k_{s,1} = \frac{d_s(x_{i+1}, t^n) + d_s(x_i, t^n)}{2} \frac{V_{i+1}}{\|x_{i+3} - x_i\|^2}$$
(S3-7)

$$k_{s,-1} = \frac{d_s(x_{i-1}, t^n) + d_s(x_i, t^n)}{2} \frac{V_{i-1}}{\|x_{i-1} - x_i\|^2}$$
(S3-8)

$$k_{s,-2} = \frac{d_s(x_{i-2}, t^n) + d_s(x_i, t^n)}{2} \frac{V_{i-2}}{\|x_{i-2} - x_i\|^2}$$
(S3-9)

$$k_{s,-3} = \frac{d_s(x_{i-3}, t^n) + d_s(x_i, t^n)}{2} \frac{V_{i-3}}{\|x_{i-3} - x_i\|^2}$$
(S3-10)

$$k_{l,3} = \frac{d_l(x_{i+3}, t^n) + d_l(x_i, t^n)}{2} \frac{V_{i+3}}{\|x_{i+3} - x_i\|^2} + \frac{v_l(x_{i+3}, t^n) + v_l(x_i, t^n)}{2} \frac{V_{i+3}}{\|x_{i+3} - x_i\|}$$
(S3-11)

$$k_{l,2} = \frac{d_l(x_{i+2}, t^n) + d_l(x_i, t^n)}{2} \frac{V_{i+2}}{\|x_{i+3} - x_i\|^2} + \frac{v_l(x_{i+2}, t^n) + v_l(x_i, t^n)}{2} \frac{V_{i+2}}{\|x_{i+3} - x_i\|^2}$$
(S3-12)

$$k_{l,1} = \frac{d_l(x_{i+1}, t^n) + d_l(x_i, t^n)}{2} \frac{V_{i+1}}{\|x_{i+3} - x_i\|^2} + \frac{v_l(x_{i+1}, t^n) + v_l(x_i, t^n)}{2} \frac{V_{i+1}}{\|x_{i+3} - x_i\|}$$
(S3-13)

$$k_{l,-1} = \frac{d_l(x_{i-1}, t^n) + d_l(x_i, t^n)}{2} \frac{V_{i-1}}{\|x_{i-1} - x_i\|^2} + \frac{v_l(x_{i-1}, t^n) + v_l(x_i, t^n)}{2} \frac{V_{i-1}}{\|x_{i-1} - x_i\|}$$
(S3-14)

$$k_{l,-2} = \frac{d_l(x_{i-2}, t^n) + d_l(x_i, t^n)}{2} \frac{V_{i-2}}{\|x_{i-2} - x_i\|^2} + \frac{v_l(x_{i-2}, t^n) + v_l(x_i, t^n)}{2} \frac{V_{i-2}}{\|x_{i-2} - x_i\|}$$

$$k_{l,-3} = \frac{d_l(x_{i-3}, t^n) + d_l(x_i, t^n)}{2} \frac{V_{i-3}}{\|x_{i-3} - x_i\|^2} + \frac{v_l(x_{i-3}, t^n) + v_l(x_i, t^n)}{2} \frac{V_{i-3}}{\|x_{i-3} - x_i\|}$$
(S3-15)
(S3-16)

# S4: Flowchart of erosion computation



#### 5.4 Modelling the effects of water chemistry and flowrate on clay erosion

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#### Abstract

Clay buffer is a key component of the engineered barrier system (EBS) for geological disposal of higher activity radioactive waste. Experimental observations indicate the possibility of buffer erosion at the interface with host rock due to interactions with groundwater. Existing models for clay erosion are very limited in terms of addressing the hydro-chemical effects, while the assessment of the long-term performance of clay buffer requires robust predictive models covering expected environment conditions and multiphysics phenomena involved in the erosion process. The work presented here is a step towards such a predictive capability, which considers clay expansion, detachment of clay particles and transport of detached particles by groundwater within a single modelling framework. The effects of solution chemistry and flowrate on the penetration, extruded mass and particle release rate of clay buffer are investigated in this paper. A series of experimental data are used to validate the swelling and erosion model developed in this study. The results show that the extrusion distance, which is controlled by both clay swelling and detachment processes, is nearly linearly dependent on the water flowrate irrespective of the water chemistry. Similar linear dependence of the erosion rate on water flowrate is observed for flowrates less than  $10^{-5}$  m/s. Higher water flowrates are shown to induce nonlinearly increasing erosion rates in accordance with experiments. A flowrate threshold is found above which the erosion behaviour of compacted bentonite can be significantly affected. A concept of tolerance time for clay buffer is introduced as a failure criterion. The results indicate that the coupled effect of water chemistry and velocity requires further investigation for ionic concentrations below 1mM.

Keywords: Bentonite, expansion, erosion, water chemistry, water velocity, peridynamics

## Introduction

Presently, mined geological disposal concepts are the preferred options for the long-term management and safe isolation of a high-level radioactive waste (HLW). Conceptual designs for the disposal in fractured rocks are based on constructing engineered barrier systems (EBS). A key component of EBS is the clay buffer which can be subject to erosion as the result of clay interaction with groundwater at the interface between the clay and host rock during the long-term performance of the barrier. Long term erosion can create potential pathways for radionuclides leaving the waste form to migrate into the surrounding rock with potentially significant environmental and public health impacts. Understanding the hydro-chemical effects on clay erosion and encoding them in a robust predictive model is therefore critical for the faithful assessment of EBS performance over the long-term repository function (tens of thousands of years).

Fitness-for-service assessment of the clay buffer requires a comprehensive understanding of bentonite extrusion, erosion and radionuclide release. These result from the clay interaction with groundwater, whose physical and chemical conditions may vary over time. Experimental studies of bentonite erosion have demonstrated that the erosion processes are affected strongly by both solution chemistry and fluid velocity (Schatz et al., 2013; Sane et al., 2013; Reid et al., 2015; Navarro et al., 2016; Smith et al., 2017; Missana et al., 2018; Alonso et al., 2018; Zhang et al., 2019; Bian et al., 2019; Bouby et al., 2020; Xiang et al., 2020). The experiments conducted by Baik et al. (2007) show that the flowrate of groundwater induced a significant impact on the total eluted volume of bentonite particles in the case studies. The experimental pinhole tests conducted by Sane et al. (2013) and Navarro et al. (2016) showed that the erodibility coefficients of MX-80 bentonite at high salinity (e.g. 70 g/L) were two orders magnitude larger than those at low salinity (e.g. 10g/L). No erosion was observed for sodium montmorillonite for solution compositions from 0.5 g/L to 10 g/L NaCl (Schatz et al., 2013). The erosion rates reported for low velocity (0.1 ml/min) were more than an order of magnitude lower than those for high velocity (2.84 ml/min) (Schatz et al., 2013; Reid et al., 2015).

These experimental observations, which highlight the importance of both physically and chemically induced erosion, show the need for predictive modelling to estimate the long-term impacts of water chemistry and water/clay interactions on erosion. The process clay buffer erosion by groundwater includes three main physical processes: (i) the initial hydration of solid clay from solid to gel and eventually discontinuously transformed into diluted sol particles; (ii)

the diluted clay particles (sol) detached at the interface of clay/fluid; (iii) detached particles taken away be seeping water (Sedighi et al., 2021; Yan et al.,2021). This mechanistic understanding, however, is not fully reflected in the existing predictive models for buffer erosion (e.g., Pusch, 1999, Neretnieks et.al. 2009 & 2017; Moreno et al., 2011; Navarro et al., 2017; Asensio et al., 2018). They remain largely empirical, based on experimental data for specific hydro-chemical conditions, which limits the confidence with which they could be applied to systems subjected to variable geological conditions. The limited progress in erosion modelling is in sharp contrast with the important progress made in coupling mechanical behaviour with hydro-chemical effects on clays for analyses of deformation and consolidation. The key elements missed in the existing models for erosion are the mathematical description of particle's detachment at the clay/water interface and its strong coupling with hydro-chemical processes. A model with particles detachment was firstly presented by Sedighi et al. (2021) and successfully applied to bentonite erosion.

The aim of this work is to underpin future fitness-for-purpose assessments of clay buffer by providing new insights into the effects of groundwater interaction with clay on erosion. For this, we present new developments of our mechanistic modelling approach (Sedighi et al., 2021), based on peridynamics (PD). PD is a non-local approach, particularly suitable for resolving discontinuities emerging and phase evolution (Silling, 2000; Silling and Bobaru, 2005), e.g. transport phenomena in discontinuous or highly heterogeneous systems, and stress induced corrosion (Bobaru and Duangpanya, 2012; Chen and Bobaru, 2015; Jafarzadeh et al., 2019a and 2019b). Furthermore, PD has been used successfully in different areas, such as hydraulic fracturing (Ouchi et al., 2015), thermally-induced cracking (Oterkus et al., 2014; Wang et al., 2018 & 2019), dynamic fracture (Rabczuk and Ren, 2017; Ren et al., 2017; Wang et al., 2016 & 2017), and chemical transport in unsaturated fracture systems (Yan et al., 2020). The present model aims to incorporate the fundamental physical and chemical interactions controlling the erosion, with PD formulations of expansion, detachment of diluted particles and detached particles transport processes. It has been demonstrated that PD has the capacity to resolve the moving interface without any additional interface constraints, which is significant for simulating the moving-boundary of expansion of diluted clay particles (Sedighi et al., 2021). This leads to an improved understanding of the combined impacts of the chemistry and flowrate.

The paper is arranged as follows. Section 2 presents the continuum formulation of processes and the development of Peridynamics (PD) frameworks for clay buffer erosion, including PD

formulations of swelling, detachment and particle transport. Section 3 present results used for model validation and analysis of additional conditions of interest. Conclusions from the work are given in Section 4.

#### 2. Clay-water interaction and erosion

The continuum and peridynamics formulations of the erosion in clays have been presented in the earlier work by the authorship team (i.e. Sedighi et al. 2021). This section provides a brief summary of the erosion model by Sedighi et al. (2021), which intends to support the new and further developments presented here. Sedighi et al. (2021) presented the main mechanisms and theoretical aspects involved in clay erosion and demonstrated how the erosion model can capture the key processes through a number of case studies. Here, in this paper, we have extended the model to be used to quantify the effects of the key parameters controlling erosion, specifically the effects of water chemistry and velocity. Therefore the focus of the present work is rather on validations with extended set of experimental data and on furthering the potential model applications. From modelling perspective, the erosion of bentonite buffer involves three distinct processes: (i) free clay swelling, which can be seen as a continuous process producing gel; (ii) clay particle detachment, which is a discontinuous process producing sol; and (iii) detached particles taken away by fluid, which can be seen as a continuous convection-dispersion process.

## 2.1. Continuum formulation of processes

Based on the dynamic force balance model proposed by Liu et al. (2009a), the free swelling/expansion of clay is represented as a solid diffusion process by the equation

$$\frac{\partial \phi_s}{\partial t} = \nabla \cdot \left(\frac{\chi}{f_r} \nabla \phi_s\right) \tag{1}$$

where,  $\phi_s$  is the clay (solid) volume fraction,  $\chi$  is the sum of the particles' energies, and  $f_r$  is a friction coefficient between particles and water. The friction coefficient is given by Moreno et al. (2011)

$$f_r = 6\pi \eta_w r_{eq} + V_p k_0 \tau^2 a_p^2 \eta_w \frac{\phi_s}{(1 - \phi_s)^2}$$
(2)

where  $r_{eq}$  is an equivalent radius of non-spherical particles,  $V_p$  is the volume of particles,  $k_0$  is the pore shape factor,  $\tau$  is the tortuosity of the flow channel in the clay gel,  $a_p$  is the specific surface area per unit volume of particles, and  $\eta_w$  is the dynamic viscosity of water.

The particles' energy,  $\chi$ , due to the interaction forces is given by:

$$\chi = k_B T + \left(h + \delta_p\right)^2 \left(\frac{\partial F_A}{\partial h} - \frac{\partial F_R}{\partial h}\right)$$
(3)

where  $k_B$  is the Boltzmann constant, *T* is the absolute temperature, *h* is the separation distance between the particles,  $\delta_p$  is the particle thickness,  $F_A$  represents attractive van der Walls forces, and  $F_R$  represents repulsive electrical double layer forces. The expressions and the calculation of  $F_A$  and  $F_R$  are discussed in detail by Liu et al. (2009a) and Sedighi et al. (2021), which are given in Appendix A.

The expression of the high non-linear diffusivity function ( $D_S$ ) can be obtained by combining Eqns.(2) and (3) as follow

$$D_{S} = \frac{\chi}{f_{r}} = \frac{k_{B}T + \left(h + \delta_{p}\right)^{2} \left(\frac{\partial F_{A}}{\partial h} - \frac{\partial F_{R}}{\partial h}\right)}{6\pi\eta_{w}r_{eq} + V_{p}k_{0}\tau^{2}a_{p}^{2}\eta_{w}\frac{\phi_{s}}{(1 - \phi_{s})^{2}}}$$
(4)

It can be observed that  $F_R$  (Eq. A-3) is dependent on the ion concentration and type of ions in the solution. Therefore, the salinity affects the repulsive electrical double layer forces ( $F_R$ ) in the model. An increase in the salinity can increase  $F_R$  at very short distances (Sedighi et al., 2021). Further discussions about the the effects of salinity on the erosion behaviour of bentonite are provided in section 3.

The central process to clay buffer erosion is particle detachment at the clay/fluid interface. The detachment of diluted sol is induced by the shear stress applied on the bonds among the particles. The shear stress induced by the fluid flow can be described by Stoke's law (Kurosawa and Ueta, 2001):

$$\tau_f = \left(\frac{6\pi\eta_w D_p}{S_p}\right) v_l \tag{5}$$

where  $D_p$  are  $S_p$  are the particles diameter and surface area, respectively, and  $v_l$  is the fluid velocity. It is noted that the detachment model developed in this study is used to link the

discontinuous behaviours from solid expansion to particles releases at the interface of clay/fluid. A general approach is to compare the shear stress at the solid/liquid interface induced by water flow with a material parameter limiting the solid behaviour of the gel, typically called the yield stress (Schatz et al., 2013; Laxton and Berg, 2006; Sedighi et al., 2021). The clay gel behaves like a solid when the shear stress is below the yield stress. In contrast, at shear stress values larger than the yield stress, the clay particles exhibit sol behaviour and can be taken away by flowing water. Hence, the complex clay erosion modelling is still highly challenging since the model is required to handle the phase change processes and resolve the discontinuous detachment at the solid/liquid interface induced by the flowing water. In this study, the yield stress is not prescribed but emerges from the balance between the shear stress on a particle and a cohesive stress arising from its interactions with other particles (Sedighi et al., 2021). The cohesive stress can be given by

$$\tau_c = \frac{F_A + F_R}{S_p} + \frac{k_B T}{6\pi h^3} \tag{6}$$

With this framework, the yield stress considered by previous authors can be determined by the local conditions at the solid/liquid interface, i.e. it is the value of the balanced shear and cohesive stresses.

The detached clay particles in water can be modelled by advection and dispersion equation:

$$\frac{\partial \phi_s}{\partial t} = -v_l \nabla \phi_s + \nabla \cdot [D_l \nabla \phi_s] \tag{7}$$

where  $D_l$  is diffusion/dispersion coefficient, which is the sum of diffusion and mechanical dispersion coefficients of detached particles (Sedighi et al.,2021).

The flow velocity can be expressed by Darcy's law:

$$v_l = -\frac{T_w \eta_w}{\eta_s} \nabla p \tag{8}$$

where p is the fluid pore pressure,  $T_w$  is the fracture transmissivity for water, and  $\eta_s$  is the sol viscosity.

The diffusion/dispersion coefficient is given by (Bird, 2002):

$$D_l = D_d + D_m = \frac{k_B T}{3\pi\eta_w D_p} + \alpha_L v_l \tag{9}$$

where  $\alpha_L$  is the dispersivity parameter.

Notably, the clay erosion involves a discontinuous process and a process with highly non-linear coefficient,  $\chi/f_r$ , making in very challenging for solutions based on the local (differential) formulations above. A non-local (integral) framework provides significant advantages by allowing discontinuous detachment at the solid/liquid interface evolving naturally with the erosion process without the need of tracking surfaces.

#### 2.2. Peridynamic formulation of clay buffer erosion processes

Peridynamics considers material domains as collections of PD points. These are not simply geometric points, but possess certain physical properties depending on the problem at hand. A point x, interacts with all points, x, in a finite spatial region called horizon and denoted by  $H_x$ . The horizon radius is denoted by  $\delta$ . The distance vector between x and x' is  $\xi = (x' - x)$ , and the interaction between them is described by a PD bond. Horizons and bonds are shown in Fig. 1.

Three different PD bonds, including solid-solid (S-S), interfacial (S-L) and liquid-liquid (L-L) bonds, are defined to facilitate the modelling of clay erosion, which are illustrated in Fig. 1. The free swelling/expansion of solid clay is described by the S-S bonds. Within the PD framework the phase changes from hydrated solid to swelling paste and clay gel evolve naturally through the changing strengths of S-S bonds. The detachment of diluted clay particles at the interface of clay/fluid is controlled by the interfacial bonds (Jafarzadeh et al., 2019a). Once the cohesive stress among the solid particles is lower than the shear stress caused by the fluid flow, the diluted clay particles detach and become liquid particles, whose transport is controlled by the L-L bonds. The interface evolution is also natural in the proposed PD framework. The particles detachment at the moving interface is considered as a discontinuous evolution process since the free swelling/expansion and detached particle transport in water follow different governing equations. A brief description of the peridynamic formulation for the three processes is given in the following subsections.



Figure 1. Illustration of the erosion processes by swelling, particle detachment, and transport of detached particles in the peridynamic framework.

## 2.3. PD formulation for bentonite free swelling and detachment

The mass conservation equation for the bond between x and x' is given by (Zhao et al., 2018; Sedighi et al., 2021)

$$\frac{\partial \bar{\phi}_s(\mathbf{x}, \mathbf{x}', t)}{\partial t} = -\frac{J_s(\mathbf{x}, \mathbf{x}', t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(10)

where  $\bar{\phi}_s(\mathbf{x}, \mathbf{x}', t)$  is the average clay volume fraction along the bond between  $\mathbf{x}$  and  $\mathbf{x}'$ ,  $J_s(\mathbf{x}, \mathbf{x}', t)$  is the solid mass diffusion flux, and  $\frac{\xi}{\|\xi\|}$  is the unit vector along the bond.

The solid flux along the bond can be given by (Sedighi et al.,2021)

$$J_{s}\left(\boldsymbol{x},\boldsymbol{x}',t\right) = -\overline{D}_{s}\left(\boldsymbol{x},\boldsymbol{x}',t\right) \frac{\phi_{s}\left(\boldsymbol{x}',t\right) - \phi_{s}(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(11)

where  $\phi_s(\mathbf{x}, t)$  and  $\phi_s(\mathbf{x}', t)$  are the clay volume fractions at points  $\mathbf{x}$  and  $\mathbf{x}'$ , respectively, and  $\overline{D}_s(\mathbf{x}, \mathbf{x}', t)$  is the average solid diffusivity between points  $\mathbf{x}$  and  $\mathbf{x}'$ , i.e.  $\overline{D}_s(\mathbf{x}, \mathbf{x}', t) = 0.5 \left[ D_s(\mathbf{x}, t) + D_s(\mathbf{x}', t) \right]$ .

Integrating Eq. (10) over the horizon leads to the conservation of mass at material point x:

$$\int_{H_{\mathbf{x}}} \frac{\partial \bar{\phi}_{s}\left(\mathbf{x}, \mathbf{x}', t\right)}{\partial t} d\mathbf{V}_{\mathbf{x}'} = -\int_{H_{\mathbf{x}}} \frac{J_{s}\left(\mathbf{x}, \mathbf{x}', t\right)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\mathbf{V}_{\mathbf{x}'}$$
(12)

where  $V_{x'}$  is the horizon volume (area in 2D, length in 1D) of particle x'.

The relationship between the concentration at point x and time t and the average concentration in all bonds adjacent to x is given by (Bobaru and Duangpanya, 2010; Sedighi et al.,2021):

$$\int_{H_{x}} \frac{\partial \bar{\phi}_{s}\left(\boldsymbol{x}, \boldsymbol{x}', t\right)}{\partial t} \mathrm{d}\boldsymbol{V}_{x'} = \frac{\partial \phi_{s}(\boldsymbol{x}, t)}{\partial t} \boldsymbol{V}_{H_{x}}$$
(13)

where  $V_{H_x}$  is the horizon volume of particle x.

Combining Eqns. (10)-(13) leads to the following evolution of clay solid content at x

$$\frac{\partial \phi_s(\boldsymbol{x}, t)}{\partial t} = \int_{H_{\boldsymbol{x}}} d_s\left(\boldsymbol{x}, \boldsymbol{x}', t\right) \frac{\phi_s\left(\boldsymbol{x}', t\right) - \phi_s(\boldsymbol{x}, t)}{\|\boldsymbol{\xi}\|^2} \,\mathrm{d}\boldsymbol{V}_{\boldsymbol{x}'}$$
(14)

where,  $d_s(\mathbf{x}, \mathbf{x}', t)$  is PD microscopic diffusivity. The relationships between PD microscopic diffusivity and measurable macroscopic diffusivity,  $D_s$ , for 1D and 2D problem are as follow:

$$d_{s}\left(\boldsymbol{x},\boldsymbol{x}',t\right) = \frac{D_{s}}{\delta}$$
(15)

$$d_s\left(\boldsymbol{x}, \boldsymbol{x}', t\right) = \frac{4D_s}{\pi\delta^2} \tag{16}$$

Derivations of Eqns. (15) and (16) are given by Yan et al. (2020; 2021) and Sedighi et al. (2021).

Diluted clay particles detach from solid/fluid interface when the shear stress,  $\tau_f$  given by Eq. (5), induced by the flowing water is above the cohesive stress,  $\tau_c$  given by Eq. (6). To model this, a detachment function is introduced in this study as follow:

$$\mu(\mathbf{x}, \mathbf{x}', t) = \begin{cases} 1 & \tau_c \ge \tau_f \\ 0 & \tau_c < \tau_f \end{cases}$$
(17)

which ensures that when the cohesive stress calculated from the mass bonds of S-S particles is below the shear stress, the solid particles transform to liquid particles and follow the governing equations of transport processes.

Incorporating the detachment function (Eq.17) into Eq. (14) leads to PD formulation for coupled clay free swelling and particles detachment:

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \int_{H_x} \mu\left(\boldsymbol{x}, \boldsymbol{x}', t\right) d_s\left(\boldsymbol{x}, \boldsymbol{x}', t\right) \frac{\phi_s\left(\boldsymbol{x}', t\right) - \phi_s(\boldsymbol{x}, t)}{\|\boldsymbol{\xi}\|^2} d\boldsymbol{V}_{\boldsymbol{x}'}$$
(18)

#### 2.4. PD formulation for detached particles transport

The mass conservation equation, Eq. (7), for the bond between x and x' is used also for particle transport, but with a modification to the flux (Zhao et al., 2018)

$$\frac{\partial \bar{\phi}_{s}\left(\boldsymbol{x}, \boldsymbol{x}', t\right)}{\partial t} = -\frac{\left[J_{l}^{d}\left(\boldsymbol{x}, \boldsymbol{x}', t\right) + J_{l}^{a}\left(\boldsymbol{x}, \boldsymbol{x}', t\right)\right]}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(19)

where  $J_l^d(\mathbf{x}, \mathbf{x}', t)$  is the diffusive/dispersive mass flux and  $J_l^a(\mathbf{x}, \mathbf{x}', t)$  is the advective mass flux. Similar to Eq. (10), the PD diffusion/dispersion flux  $J_l^d(\mathbf{x}, \mathbf{x}', t)$  is given by:

$$J_l^d\left(\boldsymbol{x}, \boldsymbol{x}', t\right) = -D_l\left(\boldsymbol{x}, \boldsymbol{x}', t\right) \frac{\phi_s\left(\boldsymbol{x}', t\right) - \phi_s(\boldsymbol{x}, t)}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(20)

where  $D_l(\mathbf{x}, \mathbf{x}', t)$  is the diffusion/dispersion coefficient of the bond between  $\mathbf{x}$  and  $\mathbf{x'}$ . The PD advection flux  $J_l^a(\mathbf{x}, \mathbf{x'}, t)$  is given by the equations (Zhao et al., 2018):

$$J_l^a\left(\mathbf{x}, \mathbf{x}', t\right) = V_l\left(\mathbf{x}, \mathbf{x}', t\right) \left[\phi_s\left(\mathbf{x}', t\right) - \phi_s(\mathbf{x}, t)\right] \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|}$$
(21)

where  $V_l(\mathbf{x}, \mathbf{x}', t)$  is the advection of the bond between  $\mathbf{x}$  and  $\mathbf{x'}$ .

Integrating Eq. (19) over the horizon point x gives

$$\int_{H_{x}} \frac{\partial \bar{\phi}_{s}\left(\boldsymbol{x}, \boldsymbol{x}', t\right)}{\partial t} d\boldsymbol{V}_{x'} = -\int_{H_{x}} \frac{\left[J_{l}^{d}\left(\boldsymbol{x}, \boldsymbol{x}', t\right) + J_{l}^{a}\left(\boldsymbol{x}, \boldsymbol{x}', t\right)\right]}{\|\boldsymbol{\xi}\|} \cdot \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{x'}$$
(22)

Substituting Eqns. (13), (20) and (21) into Eq. (22), leads to the peridynamic description of transport of detached clay particles:

$$\frac{\partial \phi_{s}(\boldsymbol{x},t)}{\partial t} = \int_{H_{x}} d_{l}\left(\boldsymbol{x},\boldsymbol{x}',t\right) \frac{\phi_{s}\left(\boldsymbol{x}',t\right) - \phi_{s}(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|^{2}} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{\boldsymbol{x}'} - \int_{H_{x}} v_{l}\left(\boldsymbol{x},\boldsymbol{x}',t\right) \frac{\phi_{s}\left(\boldsymbol{x}',t\right) - \phi_{s}(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{\boldsymbol{x}'}$$
(23)

where,  $d_l(\mathbf{x}, \mathbf{x}', t)$  and  $v_l(\mathbf{x}, \mathbf{x}', t)$  are the PD microscopic diffusivity and microscopic advection of detached particles, respectively. PD microscopic diffusivities for 1D and 2D cases are given by Eqns. (15) and (16). The PD microscopic advection is related to macroscopic advection for 1D and 2D cases by:

$$v_l\left(\boldsymbol{x}, \boldsymbol{x}', t\right) = \frac{V_l}{\delta} \tag{24}$$

$$v_l\left(\mathbf{x}, \mathbf{x}', t\right) = \frac{4V_l}{\pi\delta^2} \tag{25}$$

Derivations of Eqns. (24) and (25) are given by Yan et al. (2020) and Sedighi et al. (2021). It is noted that the newly developed peridynamics formulations for the erosion problem (Eqns. 18 and 23) do not include any spatial derivative, which ensures that the present model is able to be applied to domains of arbitrary geometry and constitution, e.g. containing discontinuities and heterogeneities in parameters  $d_s$ ,  $d_l$  and  $v_l$ .

## 2.5. Numerical implementation

The numerical treatment of coupled free swelling and detachment, Eq. (18), and transport of detached clay particles with the fluid, Eq. (23), involves spatial discretization and time integration. The domain of interest can be discretized into subdomains by using uniform linear and square sub-grids (length in 1D and area in 2D), respectively. With this discretization, Eqns.(18) and (23) can be expressed as:

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \sum_p \mu(\boldsymbol{x},t) \frac{d_s(\boldsymbol{x}_i,t) + d_s(\boldsymbol{x}_p,t)}{2} \frac{\phi_s(\boldsymbol{x}_p,t) - \phi_s(\boldsymbol{x}_i,t)}{\|\boldsymbol{x}_p - \boldsymbol{x}_i\|^2} V_{ip}$$
(26)

and

$$\frac{\partial \phi_{s}(\boldsymbol{x},t)}{\partial t} = \sum_{p} \left[ \left( \frac{d_{l}(\boldsymbol{x}_{i},t) + d_{l}(\boldsymbol{x}_{p},t)}{2} \right) \frac{\phi_{s}(\boldsymbol{x}_{p},t) - \phi_{s}(\boldsymbol{x}_{i},t)}{\left\|\boldsymbol{x}_{p} - \boldsymbol{x}_{i}\right\|^{2}} \right] V_{ip} - \sum_{p} \left[ \left( \frac{v_{l}(\boldsymbol{x}_{i},t) + v_{l}(\boldsymbol{x}_{p},t)}{2} \right) \frac{\phi_{s}(\boldsymbol{x}_{p},t) - \phi_{s}(\boldsymbol{x}_{i},t)}{\left\|\boldsymbol{x}_{p} - \boldsymbol{x}_{i}\right\|} \right] V_{ip}$$

$$(27)$$

Time integration is performed by the forward Euler method:

$$\phi_s(x_i, t^{n+1}) = \phi_s(x_i, t^n) + \dot{\phi}_s(x_i, t^n) \Delta t$$
(28)

A sequential approach is adopted to solve the coupled free swelling, detachment and detached particles transport processes. In brief, the clay swelling equation is solved at each time step and clay solid volume fraction is updated herein. This is used for calculation of cohesive stress between particles. A detachment interface is found and naturally updated by equating cohesive stress and shear stress induced by flowing water. The transport of detached particles is then calculated. The eroded mass is obtained by summation of detached particles after each time step.

#### **3.** Validation and applications

The validation of the PD model has been performed by comparisons of model predictions with experimental data for two cases reported in the literature. The data reproduced from the literature is obtained by using the function of Digitizer in Origin Pro 2016. Section 3.1 deals with bentonite expansion in a stagnant water/solution system, including bentonite extrusion and free swelling in strong sodium chloride solution and calcium chloride solution. Section 3.2 deals with bentonite erosion in a fracture system, involving the entire model proposed. This also reports an investigation of the effects of coupled water chemistry and flow velocity on erosion rate and the performance of clay barrier.

#### **3.1 Bentonite expansion behaviour in stagnant solutions**

## Bentonite expansion in de-ionised water

We have used experimental data reported by Dvinskikh et al. (2009) who applied magnetic resonance imaging (MRI) to record the spatial distribution of clay particles during swelling of bentonite in water. Their experimental programme included swelling of bentonite pellet samples (Na-exchanged MX-80, 8 mm diameter and 3 mm height) which were compacted to 1.8 Mg/m<sup>3</sup> and fitted tightly at the bottom of an 8 mm(diameter)×80mm(length) glass tube. The swelling experiment was performed initially by adding deionized water into the tube over the pellet. We have therefore considered a very low chemistry (0.05 mM) for this case (Neretnieks et al., 2017). The material properties and parameters used for the simulations have been adopted from several literature sources and are given in Table 1.  $\Delta$ =0.4mm and  $\delta$ =1.6mm are used for the cell and horizon sizes, respectively. The Kozeny constant reported in the literature ranges from 5 to 13 for bentonite clay. A value of 10 has been selected for our simulations.

		unit	value
Particle surface area <sup>a</sup>	$S_p$	$m^2$	9×10 <sup>-14</sup>
Particle diameter <sup>b</sup>	$D_p$	nm	200
Particle thickness <sup>a</sup>	$\delta_p$	nm	1.0
Surface charge of particles <sup>a</sup>	$\sigma^0$	$Cm^{-2}$	-0.131
Viscosity of water	$\eta_w$	$Nsm^{-2}$	1.002×10 <sup>-3</sup>
Relative permittivity of water	E <sub>r</sub>	-	78.54
Permittivity of vacuum	$\varepsilon_0$	$Fm^{-1}$	8.85×10 <sup>-12</sup>
Gas constant	R	$JKmol^{-1}$	8.134
Temperature	Т	Κ	298
Faraday's constant	F	$Cmol^{-1}$	96485
Boltzmann's constant	$k_B$	$JK^{-1}$	1.38×10 <sup>-23</sup>
Hamaker constant	$A_H$	J	$2.5k_BT$
Kozeny's constant <sup>a,c</sup>	$k_0 \tau^2$	-	10

Table 1. Material properties and parameters

<sup>a</sup> (Liu et al., 2009a); <sup>b</sup> (Schatz et al., 2013)

Figure 2 shows the bentonite extrusion profiles at 2 h, 4 h, 8 h, 16 h, 32 h, 70 h,140 h, 310 h and 620 h, obtained by the model (solid lines) and the results from experimental data (symbols).

The PD simulation results are in excellent agreements with the experimental results by Dvinskikh et al. (2009), lending strong support for the first part of our model.



Figure 2. Evolution of bentonite volume fraction for upward expansion of Na-exchanged MX-80. Experimental data (Dvinskikh et al., 2009) shown with symbols, PD simulation results shown with solid line.

Two observations are made from the results: sharp reduction of clay volume fraction at the expanding front; and decrease of expansion rate with time (deceleration). By Eq. (1), expansion is controlled by the diffusivity,  $D_s = \chi/f_r$ . Figure 3 shows the calculated dependence of diffusivity on clay volume fraction for several ionic strengths, one of which is the case of deionised water (DI). In cases in Fig. 3 the diffusivity,  $D_s$ , experiences a jump at a concentration-dependent value of  $\phi_s$  – an indication for criticality. For example, for deionised water,  $D_s(\phi_s = 0.1)$  is 560 times larger than  $D_s(\phi_s = 0.001)$ . For  $\phi_s < 0.001$ , i.e., very diluted bentonite sol, the Brownian motion of individual particles is the dominant process, and the diffusivity varies from  $10^{-13}$  to  $10^{-12}$  m<sup>2</sup>/s. This condition is valid up to  $\phi = 0.02$  for ionic concentration c=10mM. At high clay volume fractions, the diffuse double layer (DDL) forces

become dominant and  $D_s$  increases rapidly. Therefore, the clay particles diffuse faster at the initial stage due to dominant DDL forces, while the front evolves more slowly with time due to the rapid decrease of  $D_s$ . These simulation results explain the experimentally observed behaviour in Fig. 2: the rapid decrease of clay volume fraction near the front, and the decelerated clay expansion.



Figure 3. Variation of diffusivity as a function of bentonite volume fraction,  $\phi_s$ 

#### Bentonite expansion in strong sodium chloride solution

A free swelling test of bentonite clay in strong sodium chloride solutions has been reported by Li et al. (2019). The mineral compositions of the bentonite used in the test include 88% montmorillonite and 12% cristobalite. In the experimental tests used for validation (Li et al., 2019), bentonite specimens were compacted quasi-statically at a velocity of about 0.2 mm/min to the target dry density (e.g.  $\rho_d = 1.0 \text{ Mg/m}^3$  and  $\rho_d = 1.2 \text{ Mg/m}^3$ ) and a thickness of 10 mm. The compacted bentonite was then confined in tubes and exposed to NaCl solutions with a range of concentrations (e.g. 0.1 to 2 mM). The parameters used for these experiments are listed in Table 1. The comparison with the experimental data uses the notion of swelling strain, defined as the ratio of the differential height of the sample at any stage of the swelling and the

initial height.

Figure 4 shows the results for bentonite swelling strain at different NaCl concentrations for the two densities studied. It is clear that the model captures effectively both effects. The main observation is that increasing ion concentration leads to decreasing bentonite swelling strain – a negative effect. This can be explained from a microscopic perspective. Decreasing water chemistry can enhance the DDL repulsive force, which may reduce the swelling pressure and subsequently the measured swelling strain. From a macroscopic perspective, the expansion is governed by the diffusivity function. The results in Fig. 3 show that the diffusivity reaches a plateau of approximately  $5 \times 10^{-10}$  m<sup>2</sup>/s for all ionic concentrations. However, this plateau is narrower for higher ion concentrations because the increase of ion concentration reduces the DDL force. This results in the dominance of thermal diffusion over a larger range of solid fraction under lower ion concentrations.



Figure 4. Dependence of bentonite swelling strain on ionic concentration and initial dry density. Experimental data shown with symbols (Li et al., 2019), the predictions by solid and dashed lines.

#### Bentonite expansion in calcium chloride solution

A series of experimental investigations of free swelling of purified and fully Na-exchanged MX-80 was reported by Liu (2010). The compacted bentonite pellets (8 mm diameter and 3 mm height) with density 1.8 Mg/m<sup>3</sup> was used in the test. The samples were confined in 8 mm(diameter)×180mm (length) glass tubes. The swelling experiments involved 0.5 mM and 2.0 mM CaCl<sub>2</sub> solutions added to the tubes. We have simulated these experiments with materials properties and parameters shown in Table 1.

The bentonite sample is expected to equilibrate rapidly due to the ion exchange when it contacted with the solution (CaCl<sub>2</sub>) (Birgersson et al.,2009) and the exchange reaction  $Ca^{2+}/Na^{+}$  is given by

$$Ca^{2+}(solution) + 2Na^{+}(Clay) \leftrightarrow Ca^{2+}(clay) + 2Na^{+}(solution)$$
(29)

According to the Gaines-Thomas convention (Gaines and Thomas,1953), the ion exchange reaction of  $Ca^{2+}/Na^{+}$  can be expressed by (Birgersson et al.,2009)

$$K = \frac{X_{Ca}}{(1 - X_{Ca})^2} \frac{[Na^+]^2}{[Ca^{2+}]}$$
(30)

where, *K* is the selectivity constant,  $X_{Ca}$  is the equivalent charge fraction of Ca<sup>2+</sup> in the ionexchange positions, [Na<sup>+</sup>] and [Ca<sup>2+</sup>] are the concentrations of sodium and calcium in the solution, respectively.

The bulk concentration of counter ions in the solution can be determined for the system under consideration by setting up the mass balances for  $Ca^{2+}$  and  $Na^{+}$ , respectively (Liu, 2010):

$$[Ca^{2+}] = [Ca_0^{2+}] - \frac{u(X_{Ca} - X_0)}{2}$$
(31)

$$[Na^+] = [Na_0^+] + u(X_{Ca} - X_0)$$
(32)

where index 0 denotes the initial conditions and

$$u = \frac{m_{clay}}{V} \text{CEC}$$
(33)

where, CEC is the cation exchange capacity of the bentonite clay,  $m_{clay}$  is mass of clay and V is the volume of the bentonite clay.  $X_0$  has been taken as zero, because the material used in the experiment was purified Na-exchanged bentonite clay.

Combining Eqns. (30)-(32) gives:

$$K = \frac{X_{Ca}}{(1 - X_{Ca})^2} \frac{\left[ [Na_0^+] + u(X_{Ca} - X_0) \right]^2}{\left[ [Ca_0^{2+}] + \frac{u(X_{Ca} - X_0)}{2} \right]}$$
(34)

Solving Eq. (30) for  $X_{Ca}$  reads

$$X_{Ca} = 1 + \frac{R - \sqrt{R(4+R)}}{2}$$
(35)

where

$$R = \frac{[Na^+]^2}{K \cdot [Ca^{2+}]}$$
(36)

Combination of Eqns. (34)-(35) gives:

$$[Ca^{2+}] = [Ca_0^{2+}] - \frac{u}{2} \left( 1 + \frac{R - \sqrt{R(4+R)}}{2} - X_0 \right)$$
(37)

$$[Na^+] = [Na_0^+] + u\left(1 + \frac{R - \sqrt{R(4+R)}}{2} - X_0\right)$$
(38)

The value of parameters used for ion exchange equilibrium calculation are shown in Table 2. It is noted that the effects of ion exchange reaction  $Ca^{2+}/Na^{+}$  on the properties of the clay (e.g. particle surface area, diameter, thickness, surface charge, CEC) are not considered in the theoretical framework.

Table 2. Parameters used for ion exchange equilibrium

		unit	value
Density of clay	$ ho_s$	Mg/m <sup>3</sup>	2.77
The cation exchange capacity	CEC	eq/kg	1
The selectivity constant	Κ	Μ	2.6

The initial equivalent charge fraction	$X_0$	-	0
The initial concentration of Na <sup>+</sup>	[Na <sub>0</sub> <sup>+</sup> ]	Μ	0

Data adopted from (Birgersson et al., 2009) and (Liu, 2010)

Figures 5(a) and 5(b) show the bentonite extrusion profiles at 4 d, 8 d, 16 d and 32 d in 0.5 mM and 2.0 mM CaCl<sub>2</sub> solution, respectively. The results obtained by the proposed formulation (solid lines) and the results from experimental data (symbols) are in very close agreement. It can be seen that the ion concentration has a dual effect on bentonite clay swelling: i) the swelling deformation of the sample decreases with increasing ion concentration; ii) the clay solid volume fraction at the expanding front is sharper for higher ion concentrations. These can also be observed in Fig. 4. The DDL repulsive force would dominate the interaction force in a wider range of  $\phi_s$  for the case with lower ion concentration. The diffusivity function shown in Fig. 3 is nearly independent of the volume fraction with a relatively large value of  $5 \times 10^{-10}$  m<sup>2</sup>/s, which leads to a faster and smoother expansion behaviour. A simulation case without considering the exchange reaction (Ca<sup>2+</sup>/Na<sup>+</sup>) at 32 d is provided in Figs.5 (red dash line) to illustrate the effects of reaction processes on bentonite extrusion behaviour. It is shown that the exchange reaction can accelerate the expansion velocity and increase the extrusion distance of bentonite. The model of coupling reaction processes shows the better capability of quantifying the main features involved in bentonite erosion.





(b)

Figure 5. Evolution of bentonite volume fraction in the experimental domain subjected to: (a)
0.5 mM CaCl<sub>2</sub> solution; and (b)2.0 mM CaCl<sub>2</sub> solution. Experimental data (Liu, 2010) shown by symbols; the predictions shown by solid lines.

The validations presented in Section 3.1 provide confidence that the proposed model can capture the effects of ion concentration, ion types and dry density on bentonite swelling behaviour (first stage of erosion). In addition, it has been demonstrated that the model accounts for both the early extrusion behaviour with a sharp expanding front, and the late extrusion behaviour caused by the domination of thermal diffusion. These observations encouraged the extension of the model to erosion behaviour of clay buffer in seeping water by introducing a detachment framework. The following sub-section is designed to understand the erosion behaviour and quantify the release of colloid particles from the eroded clay buffer, which may lead to the failure of bentonite buffer.
### 3.2 Coupled water chemistry and flowrate effects on clay erosion

The experimental data reported by Schatz et al. (2013) from a series of erosion tests is used for validations, where compacted bentonite was exposed to water under different hydraulic flow and water chemistry conditions. Specifically, the sodium montmorillonite samples of  $d_s=2$  cm diameter were placed in the centre of an artificial fracture of thickness 1mm. The samples were put in contact with stagnant water or with water flowing through the fracture.

The experimental results indicated that only for clay solid content below a certain value (e.g. 2 to 4 % of solid content) and ionic strengths below 10 mM, the sodium montmorillonite dispersions could be characterized as sols (most susceptible to erosion by flowing groundwater through removal of clay particles by shear forces), which was in agreement with previous studies (Abend and Lagaly, 2000; Michot et al., 2004; Neretnieks et al., 2009). A parameter referred to as the Critical Coagulation Concentration (CCC) defines the separation between conditions allowing for and prohibiting sol formation. When the water ion concentration is larger than CCC, the smectite is considered to form a coherent gel which does not release particles. Therefore, the results for ion concentrations larger than CCC and smaller than CCC are separately presented in this section. The value of CCC can also be obtained by using the present model. A discussion of the determination of CCC by using the present model is provided in Supplemental Material.

The calculation domain is a square ( $x \times y = 24 \text{ cm} \times 24 \text{ cm}$ ) containing the bentonite sample in the centre. The cell size,  $\Delta$ , and the horizon,  $\delta$ , are set to 0.12 cm and 0.36 cm, respectively. The material parameters are given in Table 1, with the following exceptions. The thickness of clay plate is assumed to be  $\delta_p = 1.2$ nm, which is in the range from 0.6 nm to 2.4 nm reported by (Cadene et al., 2005; Liu et al., 2009b). This choice is also supported by the previous study (Sedighi et al., 2021) where  $\delta_p = 1.2$ nm provided a better performance in simulating detachment. Furthermore, the Kozeny constant is selected to be 5, similar to the choice for granular particles (OLSEN, 1962). The initial solid fraction of bentonite at  $\sqrt{x^2 + y^2} = d_s/2$  is set to 0.6, which is imposed as a boundary condition (Fig.6). The top and bottom the domain are fixed with no-flux boundaries. The left boundary was kept at zero solid fraction and the particles can exit through the right boundary. Water inlet and outlet boundary conditions were fulfilled through five flow input and output holes in the experiment. For simplicity, the constant heads were imposed at the left and right boundary to create a water flow in the model (as shown

in Fig.6).



Fig. 6. Schematic diagram of the system used for erosion of compacted bentonite: (left) artificial fracture test system (Schatz et al., 2013); (right) plan view with boundary conditions

### Clay extrusion at high ion concentrations

Water ion concentration larger than CCC leads to the formation of a coherent and non-eroded gel. In such cases clay particle release is not observed, but the bentonite buffer undergoes expansion and extrusion. The expansion/extrusion of clay buffer is likely to create localised failure points as the clay density decreases.

Figure 7 shows the clay buffer extrusion distance in the fracture with time for flowing water with two high ion concentrations and, for comparison, for stagnant deionised water. The experimental data (symbols) and the simulations results (lines) are in good agreement for the cases with DI water and c=171 mM. The simulations with c=17mM underestimate somewhat the experimental data for reasons that will be discussed shortly. Considering the significant differences between the three cases - DI in stagnant water, c=17mM and c=171 mM in seeping water with  $v_l$ =2×10<sup>-4</sup> m/s, the good agreement is a strong demonstration of the model ability to explain the solution chemistry and fluid velocity effects on bentonite clay buffer extrusion. The observation in Fig. 6 that higher ion concentration results in a shorter penetration distance is in accord with the results presented in Figs. 4 and 5.

Figure 8 shows the extruded mass of clay buffer as a function of water ion concentration. The extruded mass of clay buffer is calculated as follow:

$$m_{ext} = \int_0^{\delta_f} \int_0^{\pi} \int_{r_i}^{\infty} 2\rho_s \phi_s \delta_f dr d\varphi dz$$
(39)

where  $\delta_f$  is the fracture thickness and  $r_i$  is the initial radius of the bentonite sample (Fig.6).

The three experimental data points in Fig. 8 correspond to the three experiments shown in Fig. 7. Notable difference between the simulation result and the experimental data is observed at c=17 mM. This is because the experimentally determined extruded mass for this case included extruded mass and retained mass (dispersed and sedimented in the fracture), which led to a larger collected extruded mass. The under-predicted extrusion distance for this case, seen in Fig. 7, can therefore be explained by the lower amount of extruded mass predicted by the model in Fig. 8.

The simulations show that the extruded mass decreases slowly for water ion concentrations smaller than CCC (10mM), while significant reduction is observed for water ion concentration larger than CCC. As an example, water with c=10 mM can induce a 3.1g mass extrusion of clay buffer (see curve in Fig. 8). This corresponds to 31% decrease of density (extruded mass/original mass), which may reduce the performance of clay buffer significantly. It can be concluded that the water chemistry has a strong effect both on the extrusion distance and extruded mass of the clay buffer. This is especially highlighted for high ion concentrations, which induce shorter penetrations of the clay buffer in the fracture, lower extruded mass and effectively delay the release of clay particles.



Figure 7. Evolution of extrusion distance of clay buffer with time for high ion concentrations and for deionised stagnant water.



Figure 8. Effect of water chemistry on extruded mass of clay buffer for high ion concentrations and for deionised stagnant water.

### Clay erosion at low ion concentrations

Water ion concentrations smaller than CCC lead to colloidal particle release from clay buffer. Experiments under such conditions allow us to validate the entire model that couples clay swelling, particle detachment and transport with water flow. The smectite loss rate  $N_{erosion}$  from the detachment at the clay-water interface is (Moreno et al., 2011; Neretnieks et al., 2017)

$$N_{erosion} = 4\rho_s \phi_{s,R} \delta_f \sqrt{D_R r_R v_l} \tag{40}$$

where,  $\phi_{s,R}$  is the critical clay solid volume at detachment interface;  $r_R$  is the extrusion distance of clay buffer and  $D_R$  is the diffusion coefficient for smectite particle at the interface. A value of  $D_R = 3 \times 10^{-10} \text{ m}^2/\text{s}$  is used for low ion concentration (e.g., <1mM) case as the diffusivity is essentially constant in such cases (Neretnieks et al., 2009 and 2017). The critical clay solid volume ( $\phi_{s,R}$ ) can be obtained by equating the shear stress (Eq.5) and the cohesive stress (Eq.6). Additionally, the extrusion distance ( $r_R$ ) is dependent on the swelling process provided in Eq. (1).

Figure 9 shows a comparison between the eroded mass obtained by experiments (red triangles), previous simulation results by Moreno et al. (2011)(blue squares), and the present simulation results by the proposed model (black circles). It can be seen that the prediction by Moreno et al. (2011) underestimates the eroded mass by a factor of 3 to 10, because only smectite particle diffusion processes were considered in their model and the detachment processes were not included. The erosion model presented by Moreno et al. (2011) has been later revisited by Neretnieks et al. (2017) by introducing a rim interface to account for the release mechanism of clay particles. An experimental fitting parameter of  $\phi_{Rim}$  at the water/clay interface has been introduced to account for particle detachment processes at clay/water interface. This has led to closer correlation with experimental data. However, since their model relies on fitting to experimental data, it can be used only when the data for specific conditions is available (Schatz et al., 2013). In contrast, the present model makes its predictions based on arbitrary values of water chemistry and flowrate.



Figure 9. Eroded mass as a function of ion concentration: comparison of experimental data, previous modelling predictions and the predictions with proposed model.

Figure 10 shows the penetration distance of clay buffer as a function of water flowrate ranging from  $10^{-5}$  m/s to  $2 \times 10^{-4}$  m/s (the maximum flowrate in the experiment). All available experimental data is plotted by symbols, while model results are given by solid lines for three ion concentrations (e.g. c= 0.05, 0.86 and 4.3 mM). Two of these equal the experimental values and one is taken to be higher to facilitate comparison with results presented later. It can be observed that the calculated values are in close agreement with experiments. The extrusion distance is a nearly linearly dependent on water flowrate and decreases with increasing ion concentration. Considering Eq. (18), it can be concluded that the extrusion distance is governed by both clay swelling and detachment processes. An increase of ion concentration can effectively decrease DDL forces, which leads to a shorter penetration distance in the fracture. The same trends are observed in Figs. 4 and 5. On the other hand, an increase of water flowrate can induce a larger shear stress at the clay/water interface, which leads to larger number of particles released and shorter penetration distance.



Figure 10. Variation of extrusion distance with water flowrate. Experimental data (Schatz et al., 2013) shown by symbols; Simulation results shown by solid lines.

Figure 11 presents the effects of water flowrate and water chemistry on the erosion rate of clay buffer. The results suggest two regimes for the erosion rate – nearly linear dependence on the flowrate for rates lower than  $10^{-5}$  m/s, and accelerated dependence for higher rates. With increasing ion concentration, the flowrate effect on the erosion rate in the linear regime diminishes, which is seen in the result for *c*=4.3 mM. For all cases, however, the non-linear dependence of the erosion rate on the flowrates higher than  $10^{-5}$  m/s is present. The results show the coupling between water flowrate and chemistry induces an important impact on the erosion behaviour.

Notably, the erosion rate for c=0.05 mM is lower than for c=0.83 mM, and both are higher than for c=4.3 mM. From Eq. (40) the erosion rate is governed by water flowrate, diffusivity of particles, critical solid volume fraction and extrusion distance. Increasing ion concentration reduces extrusion distance, which leads to a lower erosion rate (e.g. c=4.3 mM). On the other hand, a higher concentration induces a larger critical solid volume fraction at the detachment interface. Therefore, the critical solid volume fraction may be the dominant factor in the case c=0.83 mM and to lead to the largest erosion rate among all cases. These results demonstrate that the developed erosion model's capability to explain the complex behaviour combined effects of hydrodynamics and water chemistry and predicts clay evolution process in close agreement with experimental results.



Figure 11. Erosion rate of bentonite buffer as a function of water flowrate. Experimental data (Schatz et al., 2013) shown by symbols; Simulation results shown by solid lines.

# 3.3 Application to assess mass loss during erosion

The laboratory studies undertaken to evaluate the effects of complex water chemistry and flow conditions on bentonite erosion are limited to several special scenarios for ion concentration and flow velocity. Furthermore, the small temporal and spatial scales of such experiments may not be relevant to the time scale of geological disposal and the associated variability of water chemistry and hydrodynamics. The present model is a step towards a faithful quantitative assessment of erosion, necessary for the design of a stable, effective and well-operational clay buffer. Being validated by experimental data at specific experimental condition, it offers explanations for observed trends and predictions for conditions beyond the experimentally feasible.

To demonstrate a relatively simple application of the model, the concept of tolerance (breakthrough) time is used to investigate the performance of clay buffer, which is the time required to reach 5% mass loss. This value is selected as the loss of 1200 kg clay from originally

22,000 kg in one emplacement hole (5% mass loss) has been considered by SKB as a buffer failure criterion (Reid et al., 2015). The buffer material used for investigation is NaMt (sodium bentonite), which was tested by Schatz et al. (2013) through an experimental program. The properties of the material are the same as those reported in Sec. 3.2.

Our calculation of the tolerance time of a clay buffer as a function of water flowrate for different ion concentrations is given in Fig. 12. It can be seen that the tolerance time decreases with increasing of water flowrate. For example, for c=4.3 mM, the tolerance time at rate  $u_l=10^{-7}$  m/s can be 190 times larger than at rate  $u_l=10^{-4}$ . Furthermore, substantial increase of ion concentrations may effectively improve the service time of the clay buffer. For example, at flowrate  $u_l=10^{-7}$  m/s, the tolerance time increases from 100 days for c=0.83 mM to 4.8 years for c=4.3 mM. Notably, the coupled effect of water chemistry and flowrate should be investigated further for very low ion concentrations, e.g., below 1 mM, where the effect of chemistry is reversed (compare c=0.83 and c=0.05 in Figs. 11 and 12). The demonstration provided with Fig.12 shows one potential application of the proposed model to reduce the uncertainties related to erosion in the long-term performance of the clay buffer. Clearly, the model can be applied to variable in time chemistry and hydrodynamic conditions.

It should be noted that the investigation of bentonite buffer erosion in this section is applicable to lab scale and limited to homo-ionised sodium bentonite. Field experiments with real geometry are necessary to investigate the bentonite buffer behaviour under different water chemistry and velocity and provide further validation data. As more complex geochemical reaction and erosion processes are expected to operate in a natural bentonite (Reid et al., 2015), the model might require extensions to incorporate additional phenomena, such as mineral dissolution, precipitation and non-uniform erosion.



Figure 12. Variation of tolerance time with water flowrate under different water chemistry.

# 4. Conclusions

A non-local model of clay erosion was presented by developing and integrating bond-based peridynamic formulations for clay swelling, particle detachment, and detached particle transport. The model was implemented within a computational framework (Pyramid; Yan et al. 2020). The model was validated by comparing its predictions to a series of experimental test, firstly under conditions prohibiting particle detachments (bentonite expansion and extrusion) and then under conditions facilitating particle detachments (bentonite erosion).

Validations for free swelling demonstrated that the present model captured successfully the effects of ion concentration, ion types and dry density on the bentonite swelling behaviour. The simulation results indicated a dual effect of the ion concentration: i) the swelling strain decreased with increasing ion concentration; and ii) the solid volume fraction at the water-clay interface increased with increasing ion concentration. These outcomes were explained by the dominance of the DDL forces in a wider range of solid concentrations for lower ion concentrations. It was also shown that larger clay dry density increased clay expansion in accordance with experiments.

The model predictions for the coupled effect of water chemistry and flowrate on penetration,

extruded mass, and particle release rate, were validated by the available in the literature data from experimental tests. For these limited data, the model allowed for explaining the experimentally observed trends. The simulation results showed that the extrusion distance, which is controlled by both clay swelling and detachment processes, was a nearly linearly dependent on the water flowrate irrespective of the water chemistry. Similar linear dependence of the erosion rate on water flowrate was observed for rates less than  $10^{-5}$  m/s. Higher water flowrates were shown to induce nonlinearly increasing erosion rates in accordance with experiments.

In addition, the concept of tolerance time of a clay buffer was used to demonstrate one possible application of the present model for the assessment of the buffer fitness-for-service. The results showed the tolerance time is significantly affected by water flowrate and ion concentration for homo-ionised sodium bentonite with lab scale geometry. The tolerance time decreases with increasing water flowrate and decreasing ion concentration under such conditions. Clearly, further extensions of the model are necessary to account for all the geochemical processes involved and the behaviour of natural raw bentonite behaviour at the field length scale.

The developed and validated model appears to be a viable alternative to the classical analysis approaches, based on the strong differential formulations of the processes involved, as it captures naturally the effects of main parameters controlling erosion – clay dry density, water chemistry and water flowrate.

## Appendix A

Forces acting between the particles include the diffusion forces ( $F_T$ ), attractive Van der Walls forces ( $F_A$ ) and repulsive electrical double layer forces ( $F_R$ ). These forces can be described by (Liu et al., 2009a; Sedighi et al., 2021):

$$F_T = \frac{-k_B T}{\phi_s} \frac{\partial \phi_s}{\partial x} \tag{A-1}$$

$$F_{A} = \frac{A_{H}S_{p}}{6\pi} \left[ \frac{1}{h^{3}} - \frac{2}{\left(h + \delta_{p}\right)^{3}} + \frac{1}{\left(h + 2\delta_{p}\right)^{3}} \right]$$
(A-2)

$$F_R = 2cRTS_p(\cosh y^m - 1) \tag{A-3}$$

where,  $A_H$  is Hamaker constant, *c* is the ionic concentration in the pore water system and *R* is universal gas constant. The function  $y^m$  represents the non-dimensional potential in the midpoint between parallel plates of for example a fracture, described by:

$$y^{m} = \sinh^{-1} \left[ 2\sinh(y_{\infty}^{m}) + \frac{4}{\kappa h} \sinh\left(\frac{y_{\infty}^{h}}{2}\right) \right]$$
(A-4)

where

$$y_{\infty}^{m} = 4 \tanh^{-1} \left[ 2 \tanh\left(0.5 \sinh^{-1}\left(\frac{zF\sigma^{0}}{2\varepsilon_{0}\varepsilon_{R}RT\kappa}\right)\right) \exp\left(-\frac{\kappa h}{2}\right) \right]$$
(A-5)

$$y_{\infty}^{h} = 4 \tanh^{-1} \left[ \tanh \left( 0.5 \sinh^{-1} \left( \frac{zF\sigma^{0}}{2\varepsilon_{0}\varepsilon_{R}RT\kappa} \right) \right) \exp(-\kappa h) \right]$$
(A-6)

The Debye length ( $\kappa$ ) is given by:

$$\kappa = \sqrt{\frac{2cz^2 F^2}{\varepsilon_0 \varepsilon_R RT}} \tag{A-7}$$

where *F* is the Faraday's constant; *z* is valence of ions in the mis-plane and  $\varepsilon_0 \varepsilon_R$  is the dielectric constant.

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## **Supplementary Material (Section 5.4)**

In this supplementary section, an approximate solution to the Critical Coagulation Concentration (CCC) implemented in the present model is presented. The CCC can be explained as the electrolyte concentration below which expansion of colloids would always lead to the full access to the domain and above which a sharp boundary is established between a colloidal gel and pure water (Liu et al., 2009). At the latter stage, the expansion of colloid can be stopped end and the colloidal suspension can be contracted. The expression of diffusivity function (Eq.4) can be reformulated and normalized as:

$$\varsigma = \frac{D_S}{(k_B T/f_r)} = 1 + \frac{\left(h + \delta_p\right)^2 \left(\frac{\partial F_A}{\partial h} - \frac{\partial F_R}{\partial h}\right)}{k_B T}$$
(S-1)

- -

- -

where,  $\varsigma$  is the thermal deviation factor, which can be interpreted as the relative expansion rate of the colloid compared to pure diffusion.

The criterion of determination of CCC by using the thermal deviation factor ( $\varsigma$ ) was provided by Liu et al. (2009) as follow

$$\varsigma_c = 0 \tag{S-2}$$

and

$$\frac{\partial \varsigma_c}{\partial h} = 0 \tag{S-3}$$

Eqns. (S-2) and (S-3) indicate that when the salinity of solution reaches CCC,  $\varsigma$  and its derivative with respect to *h* should be zero. Thus, the CCC can be obtained by substituting Eq. (S-1) into Eqns. (S-2) and (S-3). An analytical expression of CCC has been provided by Liu et al. (2019) as follow

$$CCC = 8\varepsilon_0 \varepsilon_R RT \left[ \frac{W_{-1}(X)}{h_b Fz} \right]^2$$
(S-4)

and

$$h_{b} = -\delta_{p} + \alpha (\frac{10A_{H}S_{p}\delta_{p}^{2}}{\pi k_{B}T})^{0.25}$$
(S-5)

$$X = -\frac{h_b}{8} \left[ \frac{1}{\varepsilon_0 \varepsilon_R} \left( \frac{Fz}{RT} \right)^2 \left( \frac{30A_H \delta_p^2}{\pi (h_b + \delta_p)^7} - \frac{k_B T}{S_p (h_b + \delta_p)^3} \right) \right]^{0.25}$$
(S-6)

where,  $\alpha$  is a correction factor, which is set as  $\alpha = 1$  for simplicity.  $W_{-1}(X)$  is Lambert W function.

Fig.S1 shows the Variation of CCC as a function of the thickness of clay plate ( $\delta_p$ ). It can be observed that CCC decreases with the increase of  $\delta_p$  and surface area ( $S_p$ ). The value predicted is in the range of experimental observation reported by Lagaly and Ziesmer (2003) (e.g. 5-30 mM), Frey and Lagaly (1979) (e.g. 10-15 mM) and Tombacz and Szekeres (2006) (e.g. 250 mM).



Fig.S1 The ccc value as a function of the thickness of clay plate

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# 5.5 Co-transport of accessory minerals during expansion of compacted bentonite and its impact on erosion

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## Abstract

Compacted bentonite is considered as an engineered barrier in geological disposal of nuclear waste in fractured rocks. One concern with this concept is that the expansion of bentonite into fractures as the system saturates may lead to its erosion by flowing water. The loss of bentonite through erosion over long times may compromise the barrier. Commercial bentonite contains insoluble accessory minerals and experimental observations have shown that these are co-transported with clay minerals during expansion and form a mineral film at the eroding boundary. Quantifying the effects of the mineral film on the erosion rate is important for the assessment of the long-term sealing capacity of bentonite barriers. The paper presents a model for the extrusion of bentonite into fractures that considers key processes governing the co-transport of insoluble accessory minerals and explains why and where the mineral film forms and how it affects the erosion rate.

The model is tested against two sets of published experimental data. It is demonstrated that the model predicts the phenomena observed in experiments: the build-up of a mineral film near the extrusion boundary; and the progressive growth of the mineral film thickness. Importantly, the model allows for investigating the effects of key environmental and material parameters – water chemistry, filtration coefficient, and mineral fraction – on the expansion and erosion. The results of such investigations are presented. It is shown that higher water ion concentration, larger filtration coefficient, and larger fraction of accessory minerals retard the swelling and extrusion of bentonite, thus reducing the overall erodibility. It is further shown that the presence of accessory minerals may reduce the loss of bentonite by a factor of 2 compared to a pure bentonite.

**Keywords:** Erosion, expansion, compacted bentonite, coupled problems, accessory minerals, co-transport

## 1. Introduction

Clay buffer is a critical component of engineered barrier systems (EBS) for long-term management of high-level radioactive waste, specifically in repositories constructed in crystalline and fractured rock. The clay buffer acts as a barrier between the metallic containers containing high-level (heat generating) radioactive waste and the host rock. The clay will swell after absorbing water, fill out the gaps/fractures in the rock, and create a stable surrounding environment for containers. However, the clay buffer may be susceptible to erosion by the flowing groundwater with low salinity in the fractures due to the formation of unstable colloidal sol at the clay/water interface (Schatz et al., 2013; Sane et al., 2013; Reid et al., 2015; Missana et al., 2018; Alonso et al., 2018; Bouby et al., 2020) presented evidence that the clay buffer can be eroded at the rock-clay interface by water percolating through micro-fractures or wormholes. This could facilitate the escape of radionuclides to host rock earlier than expected (Schatz et al., 2013).

Commercially available bentonite clays widely studied in the literature, such as MX-80, FEBEX, and GMZ, contain a larger fraction of smectite mineral (usually montmorillonite and as major mineral present in the composition) and a smaller fraction (10 - 25 %) of accessory minerals, such as quartz, plagioclase, illite, kaolinite, feldspar, calcite and gypsum (Schatz et al., 2013; Saba et al., 2014; Brachman et al., 2021; Cui et al., 2012 &2019; Reid et al., 2015; Alonso et al., 2019). Accessory minerals are reported to affect the erosion of bentonite (Birgersson et al. 2009; Richard and Neretnieks, 2010; Reid et al., 2015; Schatz, 2016). Understanding their role in the expansion and erosion of bentonite is therefore important and should be incorporated in the assessment of clay buffer erosion. This paper presents a model for analysing the behaviour of insoluble accessory minerals during re-saturation and expansion of bentonite, which can underpin advanced fitness-for-purpose assessments of clay buffers.

Experimental results for expansion/swelling and erosion of natural bentonite in fractures showed that a mineral film (detritus film) was formed during the swelling that could eventually clog fractures (Birgersson et al. 2009; Richard and Neretnieks, 2010; Reid et al., 2015; Schatz, 2016). Jansson (2009) observed a dark rim of accessory minerals (mainly feldspar) evident at the gel/sol interface in artificial fracture tests. Experimental observations suggest that accessory minerals could be beneficial in reducing buffer erosion at the bentonite-rock interface. For example, experiments with disk-shaped specimens showed that an accessory mineral ring of 2

mm thickness was formed at the expansion front of bentonite, and the ring was effective in mitigating against further erosion of clay buffer (Reid, 2016). Buffer mass erosion rate from the fracture was reduced from 6.6 grams/year to 2 grams/year when this protective mineral ring was formed.

Current understanding of the processes involved in bentonite expansion and erosion has been achieved mainly through extensive experimental studies in the last decade, while the developments in predictive modelling have been rather limited (Liu et al., 2009; Neretnieks et al., 2009 &2018; Moreno et al, 2011; Navarro et al., 2017; Asensio et al., 2019; Laviña et al., 2018; Sedighi et al., 2021). Deformation/swelling of compacted bentonite is commonly modelled by thermo-hydro-chemo-mechanical (THCM) coupling (Sane et al., 2013; Navarro et al., 2014 & 2021; Nasir et al., 2017; Sedighi et al. 2015a & 2018). However, the existing THCM models are based on the small strain theory and may not be applicable to cases where the bentonite deformations are large, especially for the case of free swelling. A different approach, based on DLVO electro-static forces theory (Liu et al., 2009; Neretnieks et al., 2009), has been proposed to capture the expansion/extrusion behaviour of bentonite, specifically saturated with low density. The original DLVO models could not describe the entire swelling process from an initial unsaturated to fully saturated bentonite, but they are simpler than THCM models, as the swelling is represented as an effective diffusion process. This has been used to develop extensions by Moreno et al. (2011), Neretnieks et al. (2017), Sedighi et al. (2021) and Yan et al. (2021) that capture the combined effects of swelling and erosion of pure smectite clay. These improved models, however, do not allow for incorporating accessory minerals and their effects on the erosion process.

The present work addresses the deficiency in the existing models by developing a new formulation for interactions between smectite and water insoluble accessory minerals during swelling to explain further the behaviour observed in experimental studies (Reid et al., 2015; Reid, 2016; Schatz, 2016; Alonso et al., 2018). The theoretical developments are limited to bentonite consisting of smectite and insoluble accessory minerals. It is noted that the dissolution of soluble minerals, such as feldspar and calcite, may lead to variations of water chemistry (Sedighi et al., 2015b & 2018). The model developed in the work allows for investigating the effect of water chemistry but does not explicitly account for the presence and dissolution of soluble minerals. The focus is on understanding the formation of mineral film (detritus film) due to deposition of insoluble accessory mineral and its impact on

swelling/erosion of bentonite buffer. The paper is organised as follows. Section 2 presents the model development for the expansion of bentonite and co-transport of accessory minerals. Section 3 presents validations of the model with two sets of experimental data. Section 4 presents results for the effects of several key factors - water chemistry, filtration coefficient and mineral content - on bentonite expansion and erosion. Further, it describes an application of the model to the assessment of the long-term performance of the buffer. Conclusions from the work are drawn in Section 5.

## 2. Conceptual model development

The accessory mineral particles are mechanically and chemically stable and have less electrical charge than the smectite particles. The experimental observations have shown that the fine mineral particles are co-transported with smectite gel, when the gel swells due to the strong repulsive forces between the smectite particles (Neretnieks et al., 2009). Figure 1 presents the swelling process of bentonite (containing accessory minerals) and its conceptual representation. Figure 1a shows the transport of a mineral (gypsum) within the smectite gel with high-resolution secondary electron (SE) images (Daniels et al., 2021).





(b)



(c)

Figure 1. (a) SE image of the smectite gel. A lenticular aggregate of microcrystalline gypsum (arrowed) can be seen within the smectite gel (Daniels et al., 2021); (b) Illustration of microscopic swelling mechanism of bentonite and the development of the mathematical model; (c) Illustration of bentonite containing accessory minerals swelling into a fracture: (i) Initial state of the bentonite sample in the fracture system, (ii) co-swelling of smectite and mineral particles into the fracture, and (iii) accessory mineral detachment and film formation.
Smectite aggregates are represented by larger bronze symbols and accessory mineral particles by smaller green symbols.

The conceptual representation of the microscopic swelling of mineral rich bentonite is shown in Fig.1b. To simplify the mathematical formulation, the accessory minerals are assumed to be very fine (approximately equal to the smectite particles), suspended non-reactive particles attached to smectite plates. The mineral particles can be deposited at locations where the repulsive forces between the smectite particles are week and cannot support the accessory mineral particles, as illustrated by Fig.1c. Such locations are generally at the interface between the swelling bentonite and flowing water. The detached accessory mineral particles have been reported to gradually build up a mineral film at this interface (Reid et al., 2015). The accessory mineral film contains pores of sizes less than 300 nm and is expected to slow down the expansion and erosion of bentonite (Neretnieks and Moreno, 2018).

## 2.1 Equation of motion

Particles have irregular shapes and different sizes in raw or natural bentonite. This is simplified in the proposed model by assuming that the stacks of smectite (plate-like) and mineral (spherelike) particles have uniform dimensions and properties as shown in Fig.2a. Each unit of smectite and accessory mineral particles (see Fig.2b) is subject to several different forces, including gravity ( $F_g$ ), buoyant force ( $F_b$ ), diffusional force ( $F_\mu$ ), viscous drag force ( $F_\eta$ ), attractive van der Waals forces ( $F_A$ ) and repulsive double layer forces ( $F_R$ ). These forces are shown in Fig. 2b. These forces determine the expansion or contraction of bentonite (Liu et al., 2009). The accessory mineral particles are assumed to be attached to the smectite plates (see Fig.1 and Fig.2) during the co-transport process.

The force on the particle due to the combined influence of gravity and buoyancy can be expressed by:

$$F_s = -V_p(\rho_p - \rho_w)g\tag{1}$$

where,  $F_s$  is the sum of  $F_g$  and  $F_b$ ;  $V_p$  and  $\rho_p$  is the volume and density of the unit formed by smectite and accessory mineral particles (see Fig.2b);  $\rho_w$  is the density of water and g is acceleration of gravity.

Attractive van der Waals forces  $(F_A)$  acting between particles can be presented by

$$F_A = \frac{A_H S_p}{6\pi} \left[ \frac{1}{h^3} - \frac{2}{(h+\delta_c)^3} + \frac{1}{(h+2\delta_c)^3} \right]$$
(2)

where,  $A_H$  is the Hamaker constant,  $S_p$  is the surface area of the smectite plates, h is the separation distance between the smectite plates;  $\delta_c$  denotes the thickness of smectite particles. The separation distance between the smectite plates (h) can be linked to the solid volume fraction of smectite ( $\phi_c$ ) described by (Laxton and Berg, 2006)

$$h = \delta_c \left(\frac{\phi_c^{max}}{\phi_c} - 1\right) \tag{3}$$

where  $\phi_c^{max}$  is the maximum smectite solid volume fraction.



Fig.2 Illustration of (a) conceptual model and (b) forces acting on an arbitrary unit of bentonite plate and accessory mineral particles.

Repulsive double layer forces ( $F_R$ ) due to the presence of electrical charges on the smectite particles is given by (Liu et al., 2009; Yan et al., 2021)

$$F_R = 2cRTS_p(\cosh y^m - 1) \tag{4}$$

where, *c* is the ionic concentration in the pore water, *R* is the universal gas constant and *T* is absolute temperature; The function  $y^m$  represents the scaled electric potential at the midpoint between parallel plates. The expression of  $y^m$  is given in (Liu et al., 2009; Sedighi et al., 2021). The diffusional driving force  $F_{\mu}$  results from the gradient of the chemical potential, which may be written as (Liu et al., 2009):

$$F_{\mu} = -\frac{k_B T}{\phi_c} \frac{\partial \phi_c}{\partial x} \tag{5}$$

where  $k_B$  is the Boltzmann constant.

The frictional drag force  $(F_{\eta})$  depends on the velocity v of the unit, and it can be written as

$$F_{\eta} = f v \tag{6}$$

where f is the friction coefficient, which depends on the fluid viscosity and properties of bentonite (e.g., size and volume fraction); given by (Moreno et al., 2011; Huber et al., 2021):

$$f = \left(6\pi\eta_w (r_{eqc} + r_{eqm}) + V_p k_0 \tau^2 a_p^2 \eta_w \frac{\phi_c}{(\varepsilon - \phi_c)^3}\right)$$
(7)

where  $k_0$  is the pore shape factor,  $\tau$  is the tortuosity of the flow channel in the clay gel,  $a_p$  is

the specific surface area per unit volume of smectite,  $\varepsilon$  is the porosity of the fracture as it fills with accessory minerals (Neretnieks et al., 2018), and  $\eta_w$  is the dynamic viscosity of water,  $r_{eqc}$  and  $r_{eqm}$  are the equivalent radii of the smectite and accessory mineral particles, respectively.

For a spherical like particle, the equivalent radii equals to its radius. For a con-like particle, such as smectite plate, it can be given by (Neretnieks et al., 2009)

$$r_{eqc} = \frac{\delta_c \sqrt{\frac{4S_p}{\pi \delta_c^2} - 1}}{2\tan^{-1} \left( \sqrt{\frac{4S_p}{\pi \delta_c^2} - 1} \right)}$$
(8)

The equation of motion for a single unit can be obtained by applying Newton's second law

$$F_{net} = m_p a \tag{9}$$

where, a and  $m_p$  are the acceleration and mass of the unit, and  $F_{net}$  is the net force applied on the unit, which is the sum of all forces acting on the unit, including not only the internal but also the external forces.

We assume that only the nearest particles interact with each other and consider only motion in the vertical direction aligned with all forces on the Figure 2 (Liu et al., 2009). The expression for  $F_{net}$  is given by

$$F_{net} = -F_s - F_\eta + F_\mu + \sum_i F_A^i + \sum_i F_R^i$$
(10)

where, the summation over "i" includes all the nearest neighbors.

Under steady-state conditions, i.e., for negligible acceleration (a  $\approx$  0),  $F_{net} = 0$  and Eq.10 gives

$$F_{s} + F_{\eta} + F_{\mu} + \sum_{i} F_{A}^{i} + \sum_{i} F_{R}^{i} = 0$$
(11)

# 2.2 Equation of continuity

The force equilibrium of the unit at steady state is described by:

$$-F_{\eta} - F_s + F_{\mu} + (F_A^n - F_A^{n-1}) - (F_R^n - F_R^{n-1}) = 0$$
(12)

where, superscript "n" is the result of interaction force between the n and the n + 1 layers, and similar meaning remains for the superscript "n-1"

The difference between the van der Waals attractive forces  $(F_A)$  and diffuse double layer forces  $(F_R)$  on both sides of the unit can be approximated by

$$(F_A^n - F_A^{n-1}) = (x_n - x_{n-1}) \frac{(F_A^n - F_A^{n-1})}{(x_n - x_{n-1})} \approx (h + \delta_c) \frac{\partial F_A}{\partial \phi_c} \frac{\partial \phi_c}{\partial x}$$
(13)

$$(F_R^n - F_R^{n-1}) = (x_n - x_{n-1}) \frac{(F_R^n - F_R^{n-1})}{(x_n - x_{n-1})} \approx (h + \delta_c) \frac{\partial F_R}{\partial \phi_c} \frac{\partial \phi_c}{\partial x}$$
(14)

Substituting Eqns.(13) and (14) into Eq.(12) gives

$$F_{\eta} = F_{\mu} + (h + \delta_c) \left(\frac{\partial F_A}{\partial \phi_c} - \frac{\partial F_R}{\partial \phi_c}\right) \frac{\partial \phi_c}{\partial x} - F_s$$
(15)

Substitution of Eqns. (7) and (15) into Eq. (6) gives the expansion rate as

$$v = \frac{1}{f} \left[ -\frac{k_B T}{\phi_c} \frac{\partial \phi_c}{\partial x} + (h + \delta_c) \left( \frac{\partial F_A}{\partial \phi_c} - \frac{\partial F_R}{\partial \phi_c} \right) \frac{\partial \phi_c}{\partial x} - F_s \right]$$
(16)

The flux of smectite  $(J_c)$  and accessory mineral particle  $(J_m)$  can be written as

$$J_c = \varepsilon v_c \phi_c \tag{17}$$

$$J_m = \varepsilon v_m \phi_m \tag{18}$$

where,  $v_c$  is the velocity of smectite, which equals to the expansion rate given in Eq. (16);  $v_m$  is the velocity of smectite and accessory mineral particle and  $\phi_m$  is the volume fraction of accessory mineral particle.

The equation of continuity of smectite particles

$$\frac{\partial \varepsilon \phi_c}{\partial t} + \frac{\partial s_c}{\partial t} = -\frac{\partial J_c}{\partial x}$$
(19)

where;

$$\frac{\partial s_c}{\partial t} = k_c J_c \phi_c \tag{20}$$

where,  $s_c$  and  $k_c$  is the retention and filtration coefficient of bentonite particles. The equation of continuity of suspended accessory mineral particles is given by:

$$\frac{\partial \varepsilon \phi_m}{\partial t} + \frac{\partial s_m}{\partial t} = -\frac{\partial J_m}{\partial x}$$
(21)

where;

$$\frac{\partial s_m}{\partial t} = k_m J_m \tag{22}$$

where,  $s_m$  and  $k_m$  is the retention and filtration coefficient of suspended mineral particles. At high accumulation of solids, the change of porosity by settling of the particles can be accounted for by (Neretnieks and Moreno,2018)

$$\frac{\partial s_m}{\partial t} = -\frac{\partial \varepsilon}{\partial t} \tag{23}$$

Substituting Eqns. (16-17) and (20-23) into Eq.19, the clay expansion can be written as

$$\varepsilon \frac{\partial \phi_c}{\partial t} = \frac{\partial}{\partial x} (-\varepsilon v_c \phi_c) - \varepsilon k_m v_m \phi_m \phi_c + \varepsilon k_c v_c {\phi_c}^2$$
(24)

Similarly, the co-transport of suspended mineral particle can be written as

$$\varepsilon \frac{\partial \phi_m}{\partial t} = \frac{\partial}{\partial x} (-\varepsilon v_m \phi_m) - \varepsilon k_m v_m \phi_m + \varepsilon k_m v_m \phi_m^2$$
(25)

The velocity of suspended mineral particles is defined as

$$v_m = H v_c \tag{26}$$

where H is the Heaviside function given as follow

$$H = \begin{cases} 0 & F_R - F_A < F_{g,m} \\ 1 & F_R - F_A \ge F_{g,m} \end{cases}$$
(27)

Equation (27) takes into account the two experimental observations mentioned in the introduction (Reid, 2016; Schatz, 2016; Neretnieks et al., 2018): when the repulsive forces between smectite plates are strong, the Heaviside function takes value of 1, and the suspended mineral particles are co-transported with smectite plates; when the repulsive forces between smectite plates are weak and the plates move randomly and independently (Brownian motion), then the Heaviside function becomes zero, and the suspended mineral particles are deposited due to the gravity.

Governing equations (24)-(27) are solved in non-local framework and implemented in Pyramid (Yan et al., 2020), which is a peridynamic (PD) computational framework. PD is capable for

addressing such high non-linear transport process with the wide range of coefficient variations, which may yield gross computational errors by using meshed based approach (e.g., finite difference method). Eqns. (24) - (27) may also be solved by using finite element approach although the selection of mesh and time step should be carefully due to the nonlinearities and couplings of the model (Schatz et al, 2013).

## 3. Model validations

## 3.1 Expansion of bentonite containing accessory minerals in a fracture system

To assess the role of accessory minerals in bentonite expansion and erosion behaviour, Schatz (2016) performed a series of experiments in 1 mm-aperture artificial fracture system using MX-80 bentonite (85% smectite and 15% accessory mineral). The MX-80 bentonite was compacted into dense disk-shape samples with effective smectite dry densities of 1.6 g/cm<sup>3</sup>. The sample was then exposed to Grimsel groundwater, which mainly contains 0.04 g/L NaCl and 0.016 g/L CaCl<sub>2</sub>. The result of the X-ray powder diffraction (XRD) analyses of the bentonite mineralogical composition (Schatz, 2016) is shown in Fig.3.



Figure 3. Mineralogical composition results from MX80 bentonite after expansion/erosion test (This is a rebuild version of Fig.2 in Schatz, 2006, created by OriginPro 2016 to enhance its quality to current standards)

Experimental result showed that: (i) accessory mineral ring builds near the extrusion/erosion interface; and (ii) the thickness of the mineral ring grows progressively with time. The model described in the previous section was used to simulate this experiment. The parameters used for the simulation are listed in Table 1.

	unit		value
Particle surface area <sup>a</sup>	$S_p$	$m^2$	9×10 <sup>-14</sup>
Particle diameter <sup>b</sup>	$D_p$	nm	200
Particle thickness <sup>a</sup>	$\delta_c$	nm	1.0
Maximum solid volume fraction <sup>a</sup>	$\phi_s^{max}$	-	1.0
Surface charge of particles <sup>a</sup>	$\sigma^0$	$Cm^{-2}$	-0.131
Ionic concentration <sup>c</sup>	С	mM	0.5
Ionic valence <sup>c</sup>	Ζ	-	1
Viscosity of water	$\eta_w$	$Nsm^{-2}$	1.002×10 <sup>-3</sup>
Relative permittivity of water	E <sub>r</sub>		78.54
Permittivity of vacuum	$\varepsilon_0$	$Fm^{-1}$	8.85×10 <sup>-12</sup>
Gas constant	R	$JKmol^{-1}$	8.134
Temperature	Т	Κ	298
Faraday's constant	F	$Cmol^{-1}$	96485
Boltzmann's constant	$k_B$	$JK^{-1}$	1.38×10 <sup>-23</sup>
Hamaker constant	$A_H$	J	$2.5k_BT$
Kozenys' constant <sup>a,c</sup>	$k_0 \tau^2$	-	5
Filtration coefficient	$k_m$	1/m	40

Table 1. Material Properties and parameters

<sup>a</sup> Liu et al. (2009); <sup>b</sup> Kelessidis and Maglione (2008); <sup>c</sup> Schatz el al. (2013)

Figure 4 shows the results of the simulation for the distribution of bentonite solid volume fraction ( $\phi_c$ , Fig.4a), mineral solid volume fraction ( $\phi_s$ , Fig.4b), porosity evolution in the fracture (Fig.4c) and the ratio of bentonite/mineral along the fracture (Fig.4d). The extrusion distance of bentonite is larger than that of accessory mineral. For example, the extrusion distance of bentonite and accessory mineral is 73 cm and 69 cm. In addition,  $\phi_m$  at the expansion front increases with increasing expansion (Fig. 4b). Figure 4(d) shows a comparison between the experimental data and numerical results obtained at different filtration coefficient (10 m<sup>-1</sup>, 40 m<sup>-1</sup>, 100 m<sup>-1</sup>). The filtration coefficient providing the best prediction is 40 m<sup>-1</sup>. The results highlight the ability of the present model to capture the main experimental observations - the build-up of accessory mineral ring and its growth with time (Fig.4b). The following

sections aim at understanding the expansion behaviour of bentonite containing accessory minerals and quantifying the effects of the accessory mineral on the overall long-term behaviour of bentonite buffer.



Fig.4 Model results for the distribution along the *x* direction of (a) solid volume fraction of bentonite, (b) solid volume fraction of accessory mineral, (c) bed porosity and (d) the ratio of bentonite and accessory mineral

# 3.2. Swelling of bentonite containing sand

The experimental data reported by Cui et al. (2019) for studying the one-dimensional free swelling of GMZ bentonite-sand mixtures is used to validate the present model. The GMZ bentonite samples is a light grey sodium-rich bentonite with 63.8% smectite, 18.7% sand and

17.5% others (e.g., feldspar, cristobalite). The sample was compacted to 61.8 mm(diameter) × 20 mm(height) and placed at the bottom of the 61.8 mm(diameter) × 60 mm(height) cylindrical mould. Two cases were considered to study the effects of sand ratio on swelling performance of GMZ bentonite. Case one is the free swelling of GMZ bentonite without any extra additional sand. Case two is the free swelling of GMZ bentonite with 30% additional sand. The sand content ratio is defined as *Rs* (%), the ratio of the sand's dry mass to the bentonite's dry mass (Cui et al., 2019). The dry density of samples for case 1 and case 2 is  $1.59g/cm^3$  and  $1.81g/cm^3$ , respectively. The extrusion strain of a sample is defined as  $\epsilon = \Delta H/H_0$ , where  $\Delta H$  and  $H_0$  are the vertical deformation and the initial height of the sample, respectively. The material properties and parameters used for the simulations have been adopted from several literature sources and are given in Table 2, with the following exceptions. The thickness of GMZ bentonite plate, the Kozeny constant, surface charge of particles and filtration coefficient are assumed to be  $\delta_s=1.5nm$ ,  $k_0\tau^2=13$ ,  $\sigma_0 =-0.08$  Cm<sup>-2</sup> and  $k_s=100$  m<sup>-1</sup>, respectively (Liu et al., 2009; Xiang et al., 2019).

Table 2. GMZ bentonite properties and parameters

		unit	value
Particle surface area <sup>a</sup>	$S_p$	$m^2$	9×10 <sup>-14</sup>
Particle diameter <sup>b</sup>	$D_p$	nm	200
Particle thickness <sup>a,d</sup>	$\delta_c$	nm	1.5
Maximum solid volume fraction <sup>a</sup>	$\phi_s^{max}$	-	1.0
Surface charge of particles <sup>d</sup>	$\sigma^0$	$Cm^{-2}$	-0.08
Ionic concentration <sup>e</sup>	С	mM	40-80
Kozenys' constant <sup>a,c</sup>	$k_0 \tau^2$	-	13
Filtration coefficient	$k_m$	1/m	100

<sup>a</sup> Liu et al. (2009); <sup>b</sup> Kelessidis and Maglione (2008); <sup>c</sup> Schatz el al. (2013); <sup>d</sup>Xiang et al., 2019;<sup>e</sup> Cui et al. (2019)

Figure 5 shows the results for bentonite-sand mixtures at different NaCl concentrations for the two sand content ratios (e.g. Rs = 0% and 30%). The model captures the effects of both sand content and ion concentration.



(a)



(b)

Fig. 5 (a) Geometry of experimental setup and (b)Transient extrusion behavior of sand and bentonite mixture by the proposed model (line) and experimental data (scatter) by Cui et al.

(2019)

The experimental results indicated that the swelling behaviour of GMZ bentonite-sand mixtures were varied with the sand ratio and water chemistry. The test data demonstrated that the increase of sand ratio in the bentonite mixture may significantly decrease the bentonite swelling strain or restrain the swelling properties of bentonite. This is because that uncharged larger sand content contributes to a larger friction coefficient among bentonite particles and smaller expansion rate. This effectively limits the expansion behaviour of the bentonite-sand mixtures. The model developed in this study theoretically explains why sand does not contribute to the swelling of bentonite mixtures corresponding to the experimental observations (Cui et al., 2012) and how it inhibits the expansion of the bentonite mixtures. The results above indicate that the addition of quartz sand into bentonite may inhibit better the expansion/erosion of buffer material exposed to saline groundwater (Zhang et al., 2012). Increasing ion concentration leads to decreasing bentonite swelling strain. The effect of salinity can be explained from a microscopic perspective. Decreasing water chemistry enhances the DDL repulsive force (Eq.4), which reduces the swelling pressure and subsequently the measured swelling strain. The validations presented above provide confidence that the proposed model may be used to account for the effects of the salinity and additional mineral (e.g. sands) on bentonite swelling behaviour.

### 4. Effects of insoluble accessory minerals on the extrusion of compacted bentonite

This section presents a numerical investigation of the effects of some key variables including water chemistry, filtration coefficient and mineral ratio on the expansion and extrusion of MX-80 bentonite that contains 10% addition insoluble accessory minerals (e.g., quartz). The length of domain is 80 mm. The solid volume fraction of smectite and accessory mineral are fixed at 0.6 and 0.105 at the left boundary (x=0 mm). No flux boundary is fixed at the right boundary (x=20 cm). The initial content of smectite and accessory mineral in the domain is zero. The fracture aperture ( $\delta_f$ ) is 1mm. Fig. 6 shows the illustration of geometry and boundary conditions for the problem. The parameters used for the simulations are listed in Table 1.


Fig. 6 Illustration of geometry and boundary conditions for the problem.

#### 4.1. Effects of water chemistry

Fig.7 shows the effects of water chemistry, including c= deionised water (DI), 0.5 mM, 10 mM and 100mM, on the distribution of solid volume fraction of smectite (Fig.7a) and accessory mineral (Fig.7b) along the *x* direction.



Fig. 7 The effects of water chemistry on volume fraction of (a) smectite and (b) mineral

The results show that the water chemistry has a dual effect on bentonite and accessory mineral expansion: (i) the extrusion distance decreased with increasing ion concentration; and (ii) larger solid volume fraction of smectite is observed at sharper expansion front for the higher water chemistry. The higher volume fraction of smectite at the front may protect the buffer from erosion as the interaction force among the particles at clay/water interface is larger, which

requires a bigger shear force induced by flowing water to initiate the erosion. From a microscopic perspective, the swelling behavior is governed by the expansion velocity (Eq.15), which is largely controlled by the DDL repulsive force (Eq.10). Higher ion water chemistry induces a lower DDL force, which reduces the expansion velocity.

#### 4.2. Effects of filtration coefficient

The value of filtration coefficient, which is mainly controlled by the size of the particles and the pore size distribution, is generally obtained by the filtration experiments. The value of filtration coefficient was reported to have a range between 1 m<sup>-1</sup> and 100 m<sup>-1</sup> for the fracture systems (Tsang and Neretnieks 1998). Fig.8 shows the effect of filtration coefficient (e.g.,  $k_m$ =1 m<sup>-1</sup>, 10 m<sup>-1</sup> and 50 m<sup>-1</sup>) on the distribution of volume fraction of smectite (Fig.8a) and accessory mineral (Fig.8b) along the *x* direction. The results show that increasing filtration coefficient may delay the expansion of smectite and accessory mineral. Additionally, the solid volume fraction of accessory mineral is largely increased with the increase of filtration coefficient (Fig.8b).



Fig. 8 The effects of filtration coefficient on volume fraction of (a) smectite and (b) accessory mineral

Increasing filtration coefficient enhances the deposition of accessory mineral particles, which causes the increase of the volume fraction of accessory mineral. Additionally, the increase of accessory mineral solid fraction profile induces a reduction of bed porosity, which results in the decrease of expansion distance of smectite and the volume fraction distribution profile.

#### 4.3. Effects of accessory mineral content

Fig.9 shows the effect of accessory mineral content (e.g.,  $\phi_m$ =0, 0.06, 0.18, 0.3) on the distribution of solid volume fraction of smectite (Fig.9a) and accessory mineral (Fig.9b) along the *x* direction. The results show that, similarly to the ion concentration (Fig.7b)., the accessory mineral content has a dual effect on bentonite expansion: (i) the extrusion distance decreases with increasing accessory mineral content; and (ii) the sharper expansion fronts are observed for the larger accessory mineral content. Increasing accessory mineral content can increase the accessory mineral profile and decrease the bed porosity, which enhances the friction force (Eq.7) and the expansion rate (Eq.16). The increase of accessory mineral content directly reduces the swelling rate of smectite (Fig.9a), reflected in reduced expansion distance and smectite volume fraction distribution. For example, the swelling distance of smectite decreases from 68mm to 46 mm when the accessory mineral content is increased from 0 to 0.3. The results indicate that the presence of additional accessory minerals in bentonite can effectively limit the expansion/erosion behavior of bentonite. Clearly, neglecting the effect of accessory minerals leads to an overestimation of expansion/erosion rate.



Fig. 9 The effects of accessory mineral content on (a) solid volume fraction of bentonite and (b) solid volume fraction of accessory mineral

# 4.4. Combined effects of water chemistry and accessory mineral content on mass extrusion

The extruded mass into the fracture is directly related to the long-term behavior of bentonite due to the decrease of density of bentonite buffer. The question investigated here is the role of accessory minerals in the expansion/erosion protection of bentonite by deposition and formation of accessory mineral film. Fig.10a shows the transient extrusion behavior of bentonite buffer under different accessory mineral content (e.g.,  $\phi_m=0$ , 0.06, 0.18) and water chemistry (e.g., Groundwater, GW and Seawater, SW). Fig.10b shows the mass extrusion rate as a function of accessory mineral content and water chemistry. This is calculated by:

$$E_r = \frac{\partial m_{ext}}{\partial t} = \int_0^{\delta_f} \int_0^{\pi} \int_0^{\infty} 2\rho_s \frac{\partial \phi_c}{\partial t} r dr d\varphi dz$$
(28)

where,  $m_{ext}$  is the extruded mass of bentonite buffer.

The extrusion distance of the bentonite without accessory minerals may reach 13.5 cm after 30 days (Fig.10a). Increasing  $\phi_m$  to 0.18 leads to the reduction of extrusion distance to 12 cm at 30 days for groundwater intrusion. Additionally, increasing  $\phi_m$  from 0 to 0.18 results in a two-fold decrease of the mass extrusion rate at 30 days. More specifically, after 30 days 4.9 g and 2.65 g bentonite is lost from the original buffer for  $\phi_m = 0$  and  $\phi_m = 0.18$  due to the groundwater intrusion. The reduction of extruded mass is caused by the deposition of accessory mineral detritus in the calculated domain, which can effectively clog the pores and lead to smaller mass loss.



Fig. 10 The swelling behavior of bentonite (a) extrusion distance and (b) mass extrusion rate It is noted that the extruded mass calculated from the present model for the case without accessory minerals (e.g., 4.9 g) agrees well with the experimental observation from Schatz et

al. (2013) (e.g., Test 11, 4.83g). The extrusion distance for seawater (SW, c=175 mM) is much smaller than for groundwater (GW, c=0.5 mM). The result indicates that increasing water chemistry may be an effective way to inhibit the extrusion of buffer as higher ion concentration induces lower internal repulsive forces between particles (e.g. DDL). Interestingly, the rate of mass loss for GW and SW shows a less difference at 30 days although they differ greatly in extrusion behaviour. The result in Fig.7a showed that increasing ion concentration enhances the distribution of solid fraction of bentonite although the extrusion distance is effectively decreased. The increase of bentonite solid fraction profile can amend the decrease of mass loss induced by the reduction of extrusion distance.

#### Conclusions

A model for co-transport of smectite particles and insoluble accessory mineral particles during expansion of bentonite has been presented. The model provides explanations for the role of accessory minerals in the erosion protection of bentonite observed in experiments. The accessory minerals are transported with the smectite as it swells and deposited at the location where the repulsive forces between smectite particles become week, i.e., at the interface region between the bentonite and the water. The accuracy of the model was tested against data from two sets of experimental data form literature. It was demonstrated that the model reproduced the main experimental observations of build-up of mineral film and of its growth with time.

The model was used to study parametrically the effect of water chemistry, filtration coefficient and accessory mineral ratio on the swelling and erosion of bentonite containing accessory minerals. It was shown that higher ion concentration, larger filtration coefficient and larger mineral content reduced the swelling of bentonite. Larger filtration coefficients and larger accessory mineral content enhanced mineral deposition and increased bed porosity, which effectively reduced the swelling and erosion rates. This observation was used to determine quantitatively the mass extrusion behavior of bentonite containing accessory minerals in fracture systems. The results showed that the presence of minerals could reduce the loss of bentonite by a factor of 2 compared to pure smectite bentonite. This suggests that assessments of erosion of bentonite containing accessory minerals based on experimental data obtained with pure smectite may be overly conservative.

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## CHAPTER Six

# 6. Piping assisted erosion of swelling clays

#### This chapter contains a manuscript to be submitted for publication in a refereed journal

#### 6.1 Paper introduction:

The assessment of the performance of clay as buffer/backfill or sealing material requires robust predictive models covering expected environmental conditions and multiphysics phenomena involved in the erosion process. This study is a step towards understanding the role of piping on erosion behaviour of swelling clays. All components presented in Chapter 3 to 5 are integrated into a single framework, which is used to investigate the erosion of swelling clays assisted by piping flow in cracking channels. The model was validated by comparing its predictions to a series of experimental test, including free swelling tests and erosion assisted by piping flow. The coupled swelling and erosion model showed capacity to quantify the amount of mass loss of swelling clay under piping flow. The trend observed in the experiments, including pinhole test and erosion of plug-in material in borehole, was correctly reproduced by the present model.

#### 6.2 Contribution to the joint authorship

The author has contributed the following:

- Development of the main concept for this paper (70%).
- Theoretical developments (90%).
- Model implementations and code writing (100%).
- Validations and applications of the proposed model (80%).
- Preparation of tables and data visualisation (100%).
- Paper writing (80%).

Other contributors to this part: Majid Sedighi, Andrey Jivkov and Abdelmalek Bouazza

### Piping assisted erosion of swelling clays

#### Abstract

Recent experimental studies at laboratory and underground research laboratories highlight potential erosion and piping of the clay as buffer/backfill in geological disposal and plug in material in boreholes. Existing models for swelling clay erosion assisted by piping flow are very limited as compacted swelling clay has strong swelling potential, which is not considered in traditionally piping materials (e.g. coarse soil and sand). The assessment of the performance of clay as buffer/backfill or sealing material requires robust predictive models covering expected environmental conditions and multiphysics phenomena involved in the erosion process. The present model is validated by a series of experimental data, including the free swelling test, pinhole test and erosion of plug-in material in boreholes. The swelling model is extended to investigate the self-healing potential of MX-80 bentonite in the gaps/piping channels. The gaps may be effectively filled by MX-80 with the initial dry density of 1.6 Mg/m<sup>3</sup> and 1.8 Mg/m<sup>3</sup>. The coupled swelling and erosion model is used to quantify the amount mass loss of swelling clay under piping flow. The trend observed in the experiments, including pinhole test and erosion of plug-in material in borehole, was correctly reproduced by the present model.

Keywords: swelling clay, erosion, piping, self-healing, peridynamics model

#### **1. Introduction**

The mass loss due to erosion may reduce significantly the sealing capacity of clays and their performance as engineered barriers to waste transport. For example, during the long-term operation of nuclear waste disposal facilities, the swelling clay buffer can be eroded by hydrochemical interactions at the interface between the clay and the host rock, resulting in a gradual loss of sealing capacity and a co-transport of radionuclides into the biosphere. Piping erosion is reported to occur in the early phase of the saturation process when water flows into a deposition hole from fractures in the bedrock, causing sub-vertical mass transport into the deposition tunnel backfill (Navarro et al., 2016). Further, mass erosion in the annular gap between bentonite blocks and rock may lead to failure of sealing of bentonite plug in the borehole during the emplacement (Sandén et al., 2017). Erosion of swelling clay is a fundamental geotechnical engineering problem with complex mechanisms involved (deformation, damage, and particle detachment) and challenges in their theoretical/mathematical descriptions, where continuum deformation needs to be considered together with discontinuous damage and erosion processes. This study aims to address the need for realistic modelling of the performance of swelling clay erosion assisted by the piping flow.

Most research on erosion has been focused on non-swelling materials, and direct application of the existing methods may not be suitable for analysis of swelling clay erosion, e.g., bentonite (Sane et al., 2013). For example, mechanical erosion studies have been concentrated on the piping failure of earth dams or similar structures where the pressure difference and flow rate are very large, e.g., larger than 1m<sup>3</sup>/s (Borrelli et al., 2011). In contrast, the groundwater flow rate in rock fractures is much smaller, e.g., 10<sup>-4</sup> to 1 ml/min, with very low hydraulic conductivity in the range  $10^{-5}$  to  $10^{-8}$  m/s (Suzuki et al., 2013). The behaviour of swelling clays under such low flow rates is complex, involving large deformations, phase changes, e.g., solid to gel or sol, and channel widening or narrowing due to the balance between erosion and swelling (Sane et al., 2013; Yan et al., 2021). Moreover, the mechanism of mechanical erosion of non-swelling material is significantly different from the one of piping erosion of compacted swelling clays. The latter have strong swelling potentials, which is not considered in traditional piping materials, such as coarse soil and sand. When compacted bentonite is wetted, it swells and fills the gaps or voids, leading to permeability reduction. However, water pressure acting on the bentonite may increase if the inflow is localized in fractures leading to the continuous wetting of the bentonite buffer (Borgesson and Sande, 2006; Sandén et al., 2008). The high hydraulic pressure causes a series of hydraulic phenomena, such as erosion and piping (Suzuki et al. 2013). Piping damage and erosion were observed when the water pressure was larger than the hydraulic resistance of the bentonite buffer (Chen et al., 2016).

Experimental studies in in-house and underground research laboratories are still limited in number, but provide evidence for potential erosion and piping of the clay buffer during the resaturation process. The experimental observations from the SKB and Posiva's BACLO projects (Sweden and Finland), the LOT tests at Äspö URL (Sweden), EPSRC's SAFE Barriers (UK), BELBaR project (EU), and the in-situ tests at Horonobe URL (Japan) describe complex phenomena involved in governing the erosion and piping of clay barriers. Experimental investigations have been focussed on understanding: i) the conditions for piping formation; ii) the evolution of piping channel; iii) the effects of inflow rate; and iv) the effect of piping on buffer properties and eroded mass (Sanden et al., 2008; Suzuki et al., 2013; Abe et al., 2016; Jo et al., 2019). In situ tests indicated the eroded mass of the bentonite buffer induced by piping assisted erosion could reach several kilograms (Sanden et al., 2008), which could reduce the performance of bentonite as a barrier to radionuclide transport. Additionally, water flowing along the bentonite plug was reported to cause a large amount of mass loss in a very short time (43.5% mass loss in 1 hour) and to compromise the sealing performance of bentonite plug boreholes (Sandén et al., 2017).

A limited number of theoretical studies of piping assisted erosion of swelling clay have been undertaken to date. Navarro et al. (2016) presented a coupled swelling and mechanical erosion model for compacted MX-80 bentonites. The mass loss calculation was achieved by a simplified experimentally calibrated erosion model. The model was extended by Asensio et al. (2018) to account for the effects of water salinity. The aim of the present work is to develop further the theoretical basis for performance analysis of swelling clays as sealing materials by providing new insights into piping assisted erosion. This is achieved by substantial new developments of the mechanistic modelling approach (Sedighi et al., 2021; Yan et al., 2021) based on peridynamics (PD). The earlier work presented by the authorship team (Sedighi et al., 2021; Yan et al., 2021) mainly focused on quantifying erosion of clay buffer in artificial fracture systems where the clay buffer can penetrate several meters into the fracture (e.g., maximum 10 m for 1 m/yr water velocity) (Moreno et al., 2011; Huber et al., 2021). Here, in this paper, we have extended the model to investigate the erosion of swelling clays assisted by piping flow, which could be especially important in HLW backfill and plug in the borehole. The expansion

of swelling clays is limited to these piping channels, which generally have a small size on the order of a few millimetres/centimetres (Sanden et al., 2008; Sane et al., 2013; Suzuki et al., 2013; Navarro et al., 2016). Additionally, the piping channels/gaps may be widened if the erosion potential induced by piping flow is beyond the self-healing capacity of swelling clays; contrarily, the piping channels/gaps can be closed/filled due to its strong self-healing characteristics. The work presented here is a step towards understanding the role of piping on erosion behaviour of swelling clays, although the initiation and propagation of piping channels is not coupled into the current models. The model incorporating the fundamental physical interactions controlling the erosion, with PD formulations of swelling, detachment of diluted particles and detached particles transport processes is presented in Section 2. Section 3 present results used for model validation and analysis of erosion behaviour in piping channels. Conclusions from the work are given in Section 4.

#### 2. Peridynamic formulations for erosion of swelling clay in piping flow

The continuum and peridynamics formulations of the swelling clay erosion in fractured systems have been presented in earlier works by Sedighi et al. (2021) and Yan et al. (2021). From a modelling perspective, the main mechanisms of swelling clay erosion assisted by piping flow involve: (i) swelling of clay; (ii) clay particle detachment at clay/water interface and (iii) detached particles transport by piping flow (Figure 1). The development of free swelling model (Fig.1a top) is summarised in Section 2.1; the swelling is represented by spatially varying reduction of clay density. The erosion model is coupled to the free swelling model (Fig.1a bottom), and includes representations of particle detachment and detached particle transport summarised in Section 2.3, respectively. The differences between the erosion of swelling clay in a fractured system (Sedighi et al., 2021) and the piping assisted erosion model developed here are: (i) the water flow rate in a piping channel is much larger, 10<sup>-3</sup> m/s to 1 m/s, than in a fractured system, 10<sup>-8</sup> m/s to 10<sup>-5</sup>m/s; (ii) erosion of swelling clay in fractured systems, while piping assisted erosion may have wider implications including piping erosion in backfill and mass loss of plugs in boreholes.

Peridynamics considers material domains as collections of PD points (Fig. 1b). These are not simply geometric points but possess given physical properties depending on the problem at hand. A point x, interacts with all points, x', in a finite spatial region called horizon and is denoted by  $H_x$  (Fig. 1b). The horizon radius is denoted by  $\delta$ . The distance vector between x

and  $\mathbf{x}'$  is  $\boldsymbol{\xi} = (\mathbf{x}' - \mathbf{x})$ , and the interaction between them is described by a PD bond. Horizons and bonds are shown in Fig. 1b. We defined three PD bonds to describe the three distinct interactions involved in erosion assisted by the piping flow. Solid-solid bonds (S-S) control the swelling process; Solid-liquid interfacial bonds (S-L) govern the detachment of particles at the clay/water interface; Liquid-liquid bonds (L-L) describe the transport of detached particles with the piping flow.



Figure 1. Illustration of (a) swelling and piping assisted erosion and (b) peridynamic representation of interactions and processes (Fig.1b)

#### 2.1. PD formulation of free swelling

Free swelling can be treated as a solid diffusion process, theoretically developed by a dynamic force balance method (Liu et al., 2009). However, the solid diffusivity may change by 5 orders of magnitude as the clay solid content (density) changes. This makes the problem highly non-linear. PD is particularly suitable to handle such strong non-linearities, while numerical schemes based on a continuum formulation, such as finite elements or finite differences, may produce unreliable results. The solid diffusion process in the PD framework is given by Yan et al. (2020; 2021) and Sedighi et al. (2021) as follow

$$\frac{\partial \phi_s(\boldsymbol{x}, t)}{\partial t} = \int_{H_{\boldsymbol{x}}} d_s\left(\boldsymbol{x}, \boldsymbol{x}', t\right) \frac{\phi_s\left(\boldsymbol{x}', t\right) - \phi_s(\boldsymbol{x}, t)}{\|\boldsymbol{\xi}\|^2} \, \mathrm{d}\boldsymbol{V}_{\boldsymbol{x}'} \tag{1}$$

where  $\phi_s(\mathbf{x}, t)$  is the clay solid content, t is the time,  $\mathbf{V}_{\mathbf{x}'}$  is the horizon volume of particle  $\mathbf{x}'$ , and  $d_s(\mathbf{x}, \mathbf{x}', t)$  is the PD microscopic diffusivity. The relationships between PD microscopic diffusivity and measurable macroscopic diffusivity,  $D_s$ , for 1D and 2D problem are as follow:

$$d_s\left(\boldsymbol{x}, \boldsymbol{x}', t\right) = \frac{D_s}{\delta} \tag{2}$$

$$d_s\left(\mathbf{x}, \mathbf{x}', t\right) = \frac{4D_s}{\pi\delta^2} \tag{3}$$

where,  $D_s$  is the macroscopic solid diffusivity, which can be calculated by

$$D_S = \frac{\chi}{f_r} \tag{4}$$

where,  $\chi$  is the particles' energy and  $f_r$  is the friction coefficient. Equation (4) is obtained by developing a dynamic force balance, including the diffusion forces ( $F_T$ ), attractive Van der Waals forces ( $F_A$ ) and repulsive electrical double layer forces ( $F_R$ ) between clay particles (Sedighi et al., 2021).

The friction coefficient and particle's energy are given by (Liu et al., 2009; Neretnieks et al., 2009)

$$f_r = 6\pi \eta_w r_{eq} + V_p k_0 \tau^2 a_p^2 \eta_w \frac{\phi_s}{(1 - \phi_s)^2}$$
(5)

and

$$\chi = k_B T + \left(h + \delta_p\right)^2 \left(\frac{\partial F_A}{\partial h} - \frac{\partial F_R}{\partial h}\right) \tag{6}$$

where  $r_{eq}$  is the equivalent radius of the non-spherical particles,  $V_p$  is the volume of the particles,  $k_0$  is the pore shape factor,  $\tau$  is the tortuosity of the flow channel in the clay gel,  $a_p$  is the specific surface area per unit volume of particles,  $\eta_w$  is the dynamic viscosity of water,  $k_B$  is Boltzmann constant, T is absolute temperature,  $\delta_p$  is the particle thickness, and h is the separation distance between the flat particles. The calculation of attractive Van der Waals forces  $(F_A)$ , repulsive electrical double layer forces  $(F_R)$  and separation distance of particles (h) is provided in Appendix A.

#### 2.2. Particle detachment at clay/water interface

Erosion is characterised by detachment of particles at the clay/water interface. The challenge with modelling swelling clay erosion is in the coupling of a continuous phase change process (swelling) with a discontinuous particle detachment process. The external force causing detachment is a shear force ( $\tau_f$ ) induced by the water flowing in the pipe channel. Detachment occurs when this shear force becomes larger than the interparticle forces that keep interface solid particles attached to their surrounding solid particles (Sane et al., 2013). The interactions of an interface solid particle with its surrounding solid particles provide a cohesive strength to the particle. This is related to the interaction forces with other particles by Sedighi et al. (2021):

$$\tau_c = \frac{F_A + F_R}{S_p} + \frac{k_B T}{6\pi h^3} \tag{7}$$

With these settings, a detachment function is given by

$$\mu\left(\boldsymbol{x}, \boldsymbol{x}', t\right) = \begin{cases} 1 & \tau_c \ge \tau_f \\ 0 & \tau_c < \tau_f \end{cases}$$
(8)

It ensures that when the cohesive strength calculated from S-S bonds is less than the shear force, the solid particles transform to liquid particles and follow the governing equations for transport processes (Yan et al., 2021). Incorporating the detachment function Eq. (8) into Eq. (1) leads to PD formulation for coupled free swelling and particles detachment:

$$\frac{\partial \phi_s(\boldsymbol{x},t)}{\partial t} = \int_{H_x} \mu\left(\boldsymbol{x}, \boldsymbol{x}', t\right) d_s\left(\boldsymbol{x}, \boldsymbol{x}', t\right) \frac{\phi_s\left(\boldsymbol{x}', t\right) - \phi_s(\boldsymbol{x}, t)}{\|\boldsymbol{\xi}\|^2} \, \mathrm{d}\boldsymbol{V}_{\boldsymbol{x}'} \tag{9}$$

#### **2.3. PD formulation for detached particles transport**

The detached particles transport in piping flow is represented by the advection and dispersion equation (ADE). PD formulation of transport of detached clay particles is given by Yan et al. (2020; 2021) and Sedighi et al. (2021):

$$\frac{\partial \phi_{s}(\boldsymbol{x},t)}{\partial t} = \int_{H_{x}} d_{l}\left(\boldsymbol{x},\boldsymbol{x}',t\right) \frac{\phi_{s}\left(\boldsymbol{x}',t\right) - \phi_{s}(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|^{2}} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{x'} 
- \int_{H_{x}} v_{l}\left(\boldsymbol{x},\boldsymbol{x}',t\right) \frac{\phi_{s}\left(\boldsymbol{x}',t\right) - \phi_{s}(\boldsymbol{x},t)}{\|\boldsymbol{\xi}\|} \frac{\boldsymbol{\xi}}{\|\boldsymbol{\xi}\|} d\boldsymbol{V}_{x'}$$
(10)

where,  $d_l(\mathbf{x}, \mathbf{x}', t)$  and  $v_l(\mathbf{x}, \mathbf{x}', t)$  are the PD microscopic diffusivity and microscopic

advection of detached particles, respectively. Similar to Eqns. (2) and (3), PD microscopic diffusivities and advection for 1D and 2D cases are given by:

$$d_l\left(\boldsymbol{x}, \boldsymbol{x}', t\right) = \frac{D_l}{\delta} \tag{11}$$

$$d_l\left(\mathbf{x}, \mathbf{x}', t\right) = \frac{4D_l}{\pi\delta^2} \tag{12}$$

and

$$v_l\left(\boldsymbol{x}, \boldsymbol{x}', t\right) = \frac{V_l}{\delta} \tag{13}$$

$$v_l\left(\mathbf{x}, \mathbf{x}', t\right) = \frac{4V_l}{\pi\delta^2} \tag{14}$$

where,  $D_l$  is diffusion/dispersion coefficient, which is the sum of diffusion ( $D_d$ ) and mechanical dispersion ( $D_m$ ) coefficients of detached particles as follow

$$D_l = D_d + D_m = \frac{k_B T}{3\pi \eta_w D_p} + \alpha_L v_l \tag{15}$$

where,  $\alpha_L$  is the dispersivity parameter.

The flow velocity  $(V_l)$  can be expressed by Darcy's law:

$$V_l = -\frac{T_w \eta_w}{\eta_s} \nabla p \tag{16}$$

where, p is the fluid pore pressure,  $T_w$  is the fracture transmissivity for water, and  $\eta_s$  is the soil viscosity.

#### 2.4. Numerical implementation

A sequential approach is adopted to solve the coupled free swelling and particles detachment, Eq. (9), and detached particles transport processes, Eq. (10). The domain of interest is discretized into subdomains using uniform linear and square sub-grids (length in 1D and area in 2D), respectively. The Euler method is adopted for time integration. The discretization of domain and integration of time are presented in Sedighi et al. (2021) and Yan et al. (2021). Briefly, the swelling equation is solved first at each time step where the clay solid volume fraction profile is obtained to compute the cohesive strength of particles. The detachment interface is, therefore, naturally updated by equating cohesive strengths and shear forces. The transport of detached particles is then calculated. The eroded mass is consequently obtained by summation of detached particles after each time step.

#### 3. Results and discussion

The swelling model is first validated using free swelling experiments performed by Liu et al. (2011). Further discussion of self-healing properties of MX-80 in existing piping channels is provided based on the results. Two applications of the model, including pinhole test simulation and erosion performance of bentonite plugs in boreholes, are presented. The model is used to quantify the total mass lost due to piping assisted erosion.

#### 3.1 MX-80 bentonite swelling behaviour

Free upward expansion of compacted bentonite (e.g., MX-80) in a vertical test tube was performed by Liu et al. (2011) to study the free swelling behaviour of compacted bentonite. Their experimental programme included swelling of bentonite pellet samples (MX-80, 8 mm diameter and 3 mm height), which were compacted to 1.8 Mg/m<sup>3</sup> and fitted tightly at the bottom of an 8 mm (diameter) × 80mm (length) glass tube. The water chemistry in the tube was assumed to be 4 mM NaCl, considering that the gypsum mineral was fully dissolved during the expansion process (Liu et al., 2011). The material properties and parameters used for the simulations have been adopted from current literature and are given in Table 1.  $\Delta$ =0.1 mm and  $\delta$ =0.3 mm are used for the cell and horizon sizes, respectively.

		unit	value
Particle surface area <sup>a</sup>	$S_p$	$m^2$	9×10 <sup>-14</sup>
Particle diameter <sup>b</sup>	$D_p$	nm	200
Particle thickness <sup>a</sup>	$\delta_p$	nm	1.0
Surface charge of particles <sup>a</sup>	$\sigma^{0}$	$Cm^{-2}$	-0.10
Viscosity of water	$\eta_w$	$Nsm^{-2}$	1.002×10 <sup>-3</sup>
Relative permittivity of water	E <sub>r</sub>	-	78.54
Permittivity of vacuum	$\varepsilon_0$	$Fm^{-1}$	8.85×10 <sup>-12</sup>
Gas constant	R	$JKmol^{-1}$	8.134
Temperature	Т	K	298
Faraday's constant	F	$Cmol^{-1}$	96485
Boltzmann's constant	$k_B$	$JK^{-1}$	1.38×10 <sup>-23</sup>
Hamaker constant	$A_H$	J	$2.5k_BT$
Kozeny's constant <sup>a,c</sup>	$k_0 \tau^2$	-	10

Table 1. Material properties and parameters

<sup>a</sup> (Liu et al., 2009a); <sup>b</sup> (Schatz et al., 2013)

Figure 2 shows the MX-80 swelling profiles at 0.5 h, 1 h, 2 h, 6 h (Fig. 2a) and 24 h, 48 h, 164 h, 384 h (Fig. 2b), obtained by the swelling model (Eq.1) (solid lines) and the results from experimental data (symbols). The general swelling behaviour of MX-80 is captured well, although the predicted expansion front deviates somewhat from the measured one. For example, the predicted swelling distance for  $t \le 6$  h (Fig.2a) is shorter compared to the experimental observation. MX-80 bentonite contains 20% accessory minerals (e.g., quartz, feldspar, gypsum, calcite, etc.), which can lead to the variations of water chemistry due to the transient geochemical reaction. However, to avoid the use of an additional reaction model in the current study, a constant water chemistry (e.g., 4mM NaCl) is obtained by assuming that the gypsum was fully dissolved and ion exchanged for sodium (Karnland et al., 2006; Liu et al., 2011). This may contribute to the deviations between the model and experimental data. However, the effort of the present study is primarily focused on validating the swelling model, which is developed based on dynamic force balance theory, for transient expansion process. It can be seen that the expansion front is relatively smooth at the start of swelling (e.g., < 6 h, Fig. 2a) and becomes sharper after 24 h (Fig. 2b). At high bentonite volume fractions, the repulsive forces among

particles are dominant, which results in a rapid and smooth expansion of bentonite. When the bentonite volume fraction is below a specific value (e.g.  $\phi_s = 1\%$ , very diluted bentonite colloid) due to the expansion, a rapid change of the expansion force and viscous drag force by five orders of magnitude can be observed (Sedighi et al., 2021). At this stage, the Brownian motion of individual particles is the dominant process. The rapid decreases of expansion force and viscous drag force cause a sharp expansion front.



(a)



(b)

Fig.2 Comparison of the MX-80 swelling profiles between the proposed model and experimental data at (a) 0.5h, 1h, 2 h, 6 h and (b) 24h, 48h, 164h, 384h

The most important property of bentonite as buffer/backfill/plug-in material is its strong swelling behaviour expected to fill interstitial spaces (e.g., gaps/voids) between clay and disposal pits' walls waste containers and host rock. This swelling characteristic is referred to as self-sealing capability. Therefore, the analysis of self-sealing behaviour is important for investigating the performance of bentonite clay as buffer/backfill for HLW and sealing material in boreholes. The case considered herein is for a cylindrical bentonite sample with a size of 100 mm in diameter and 100 mm in length. A hole with the initial radius of  $r_h$  =3 mm was made in the centre of bentonite and filled with stagnant water (see Fig.1a top, a 2D axisymmetric illustration). This geometry was used for the pinhole test by Sane et al. (2013).

Figure 3 shows the swelling of MX-80 into the existing hole/piping channel in stagnant water at different time intervals. It can be seen that MX-80 fills the gaps due to the strong self-sealing potential induced by the swelling behaviour. There is a close over occurring at the interface of bentonite and gap when  $t \le 5$  h (see Fig.3a). The strong swelling capacity of bentonite results in a fast expansion of bentonite in the gap. However, the continuous extrusion of bentonite into the gap decreased the density of buffer, which leads to slowing down the expansion of bentonite (see Fig.3b). Additionally, it is noted that MX-80 can fill the gaps (e.g.,  $r_h$  =3 mm piping channel) quickly. For example, the swelling front can reach the right boundary of the gaps at around 2.5 h. However, the solid fraction of bentonite in the gaps is very low, which may not protect its surrounding environments due to very high permeability. The key point to consider to quantify the self-sealing capability of MX-80 is the swelling pressure of bentonite after filling the gaps/channels. For example, the self-sealing capacity was assumed to be effective when the swelling pressure of bentonite after filling the gaps was larger than 1000 kPa (Komine, 2019), which indicated an effective low permeability. The swelling pressure of bentonite can generally be linked to its dry density. The swelling pressure ( $P_s$ ) of MX-80 bentonite can be estimated as follows (Zeng et al., 2019):

$$P_s = 1.652 \times 10^{-4} e^{6.781\rho_d} \tag{17}$$

Where,  $\rho_d$  is the dry density of MX-80.

Fig.4 shows an example of transient self-healing behavior of MX-80 bentonite under different initial dry densities. It can be seen that the gaps may be effectively filled by MX-80 with the initial dry densities of 1.6 Mg/m<sup>3</sup> and 1.8 Mg/m<sup>3</sup>. However, the self-healing capability for lower dry density (e.g., <1.4 Mg/m<sup>3</sup>) may not be sufficient to fill the gaps and establish an effective barrier. It is noted that the higher dry density can heal the gaps sooner due to the stronger swelling capacity. For example, the time required to achieve effective self-sealing capacity for  $\rho_d$ =1.8 Mg/m<sup>3</sup> and  $\rho_d$ =1.6 Mg/m<sup>3</sup> is 107 h and 240 h, respectively. This analysis shows the capability of the present swelling model to quantify the healing performance of MX-80 in stagnant water. Further discussion of swelling and erosion behavior of MX-80 with constant piping flow injection is provided in the following section.



(a)



(b)

Fig.3 Swelling of MX-80 into the existing hole/piping channel in stagnant water at at (a) 1 h, 2.5 h, 3.75 h and 5 h and (b) 10 h, 25 h, 50 h



Fig.4 Modelling results of self-sealing capacity of MX-80 bentonite under different initial dry density

#### **3.2 Model application**

#### **Pinhole test simulation**

The experimental data reported by Sane et al. (2013) from a series of pinhole tests (Fig.5) was used to validate the coupled swelling and erosion model when MX-80 bentonite is exposed to tap water. The flow was driven through a pinhole at the centre of MX-80 bentonite with a constant flow rate of 0.1 L/min; the maximum allowed inflow in a deposition hole in Finland (Juvankoski et al., 2012). Cylindrical samples had a size of 100 mm in diameter and 100 mm in length. The initial dry density of MX-80 bentonite was 1700 kg/m<sup>3</sup>. A smooth-surface steel rod with an initial diameter  $d_h$  (either 6 or 12mm) was inserted in the cell (Navarro et al., 2016). An automated effluent collector collected effluent samples. The initial and boundary conditions shown in Fig.5 were applied to model the pinhole tests. The shear stress induced by flowing water on bentonite particles is obtained by assuming that a quasi-steady laminar flow has fully developed in the central hole (Navarro et al., 2016). The hydraulic shear stress, therefore, can be calculated based on the laws of fluid dynamics as follow:

$$\tau_f = \frac{4\eta_w Q}{\pi r_h^3} \tag{18}$$

Where,  $\eta_w$  is the dynamic viscosity of the water, Q is the water flow rate (0.1 L/min), and  $r_h$ 

is the characteristic mean radius of the central hole. Generally,  $r_h$  is a function of time and it is distributed non-uniformly along the cylinder due to the heterogeneities of bentonite. However, for the sake of simplicity, we assumed that the central hole has an effective cylinder of radius  $r_h$  (see Fig. 5a; Navarro et al., 2016). The experimental observations for the initial and final hole diameters at the top and bottom of the sample, which were both 3 mm, justified this assumption. The shear stress calculated from Eq.(18) for  $r_h$ =3mm and  $r_h$ =6mm is 0.078 Pa and 0.00975 Pa, respectively. Fig.7 shows the measured experimental data for  $r_h$ =3mm (e.g. s1a, s1b, s1c) and  $r_h$ =6mm (e.g. s3a) through pinhole tests.



(a)

(b)

Fig. 5. (a) boundary conditions used in the 2D axisymmetric simulations for pinhole test and bentonite plugs in boreholes and (b) experimental setup of the pinhole test (Navarro et al., 2016)

Fig.6 shows the solid volume fraction distribution temporal profile for  $r_h=3$  mm (Fig. 6a) and  $r_h=6$  mm (Fig. 6b) piping channel. It can be seen that there is almost no bentonite expanding into the piping channel as the swelling velocity is much lower compared to the erosion rate. The flow velocity ( $V_l$ ) for  $r_h=3$  mm and  $r_h=6$  mm is around 0.06 m/s and 0.015 m/s. The maximum expansion diffusivity of bentonite colloid ( $D_R$ ) is around 10<sup>-9</sup> m/s (Nerenieks et al., 2009; Yan et al., 2021). We introduce, herein, the dimensionless Peclet number ( $Pe=V_lL/D_R$ ) to clarify the difference between expansion rate and flow rate. The Peclet numbers for  $r_h=3$  mm and  $r_h=6$  mm are  $6\times10^6$  and  $1.5\times10^6$ , respectively, demonstrating that the erosion rate induced

by flowing water may be significantly larger than the expansion rate. It is noted that the flowing water continually erodes the bentonite mass in the piping channel. The solid bentonite fraction profile decreases as time increases (Fig.6). For example, the solid fraction of bentonite decreases from 59% to 32 % at r=40 mm if time increases from 10 h to 50 h (Fig. 6a). The decrease of volume fraction may result in a significant reduction of the self-healing potential of MX-80 bentonite.



(a)



(b)

Fig.6 Variation of solid volume fraction of bentonite with time (a)  $r_h = 3$  mm piping channel and (b)  $r_h = 6$  mm piping channel

Navarro et al. (2016) used a simplified erosion model to describe the loss of mass at the water/clay interface. Mass loss was obtained by using an experimental fitted erosion rate ( $k_e$ ). The best fitted erosion rate for  $r_h=3$  mm and  $r_h=6$  mm is 0.004 s/m. Fig.7 shows the comparison of cumulative mass loss per unit length among experimental data and the modelling results from Navarro et al. (2016) and the current investigation.

In this study, the bentonite mass loss rate ( $N_{erosion}$ ) from the detachment at the clay-water interface is obtained by (Moreno et al., 2011; Neretnieks et al., 2017):

$$N_{erosion} = 4\rho_s \phi_{s,R} L \sqrt{D_R r_R V_l} \tag{19}$$

where,  $\phi_{s,R}$  is the critical clay solid volume at detachment interface, which can be obtained by equating the cohesive stress (Eq.7) and the shear stress (Eq.18); *L* is the length of piping channel;  $r_R$  is the radius of the piping hole and  $D_R$  is the diffusion coefficient for bentonite particles at the interface. A value of  $D_R = 2 \times 10^{-10} \text{ m}^2/\text{s}$  was used for low ion concentration (e.g., <1mM) case.

Figures 7a and 7b show a comparison between the eroded mass obtained by experiments (scatters), previous simulation results by Navarro et al. (2016) (dashed black line), and the current simulation results by the proposed model (solid black line). Experiments s1a, s1b and s1c are in principle identical, but the recorded data appears to vary substantially due to limitations of the erosion test equipment and the heterogeneity of clay samples. It can be seen that the present model correctly reproduced the trend observed in two of the experiments with similar records (s1a and s1b). The average relative difference ( $\varepsilon$ ) between experiments and simulations is 1.9 and 10.6, respectively. The predictions obtained by Navarro et al. (2016) and the present study show a minor difference. The main difference is that the results by Navarro et al. are obtained after calibrating the model with the experimental data for erosion rate, while with the current model the cumulative mass loss is calculated without any calibration – it is a consequence of the swelling and detachment formulations. Specifically, the erosion process is controlled by the balance between cohesive strength (which depends on the swelling) and shear force (which depends on water velocity and chemistry). The present model shows the capacity to capture the main trend observed in the test for the case with  $r_h = 6$  mm, although the predicted mass erosion rate is large compared to the experimental results after 10 hours and before 65 hours. The over-predicted mass loss of bentonite may be attributed to the assumption of constant radius of the bentonite cylinder  $(r_h)$ .





(a)

(b)

Fig.7 Comparison of calculated mass loss per unit height and experimental results (a)  $r_h = 3$ mm piping channel and (b)  $r_h = 6$  mm piping channel

#### Erosion performance of bentonite plugs in boreholes

MX-80 bentonite is often used as a plug material for sealing deep boreholes (SKB, 2010). One potential problem that might be encountered in this type of application is that the standing water in the borehole can pass the annular gap between the rock and the bentonite by a relatively high flow velocity (e.g, 1 L/s) during the emplacements of plug, which may cause erosion. Sandén et al., 2017 performed the test of bentonite plugs erosion in boreholes usingMX-80 bentonite block with a diameter of 80 mm. The length of the bentonite block was 0.25m. The gap between the wall and the block was  $d_g=3$ mm. After installation, the bentonite block was flushed with tap water at 1 L/s for 1 hour. The shear stress at the water/clay interface induced by flowing water was around 0.5 Pa ( $\tau_f \approx \eta V_l/d_g$ ; Eriksson and Schatz, 2015). The test parameters are provided in Table.2. Fig.8 shows the experimental test installation (Fig. 8a) and eroded profile after the test (Fig. 8b). A clear visible erosion profile can be observed in Fig.8b.



Fig.8 Photos of (a) blocks ready for installation and (b) blocks profile after erosion (Sandén et al., 2017)

Test Parameters	Value
Borehole diameter (m)	0.08
Installation time (min)	60
Flow rate (l/s)	1.0
Diameter of bentonite block (m)	0.074
Length of bentonite block (m)	0.25
Area of annular gap (cm <sup>2</sup> )	7.3
Velocity in annular gap (m/s)	1.378
Mass loss (Test results)	10.3%

Table.2 Test parameters and results (Sandén et al., 2017)

Figure 9 compares the eroded mass amount obtained by experiments (scatters) and the simulation results by the proposed model (solid black line). The present model correctly reproduced the trend observed in the experiments, although the current model underestimated the eroded mass amount at the end of the test (e.g., > 50 min). The average relative difference ( $\varepsilon$ ) between experiments and simulations is 0.11 in Fig.9. It can be seen from Fig.8b that the erosion profile of the bentonite is non-uniformly distributed, especially at the bottom of the sample. The radius of the lower bentonite block was much smaller after erosion than that of the

upper block as the water was injected from the bottom of the sample. This is different from the pinhole test (Sane et al., 2013) where the radius of the bentonite at the top and bottom of the sample did not change after the test. The non-uniform erosion along the length of the bentonite block was not addressed in the current model, which may account for underestimating the mass of eroded bentonite at the end of the test. The amount of bentonite mass loss at the end of the test was 10.3% of the original mass at installation (see. Table 2). The large amount of bentonite mass loss induced by erosion can be detrimental to the sealing capacity of bentonite plugs in boreholes. The above analysis indicates that the possible loss of bentonite material should be carefully investigated at the planning and design stages.



Fig.9 Comparison of calculated mass loss and experimental results

#### 4. Conclusion

The assessment of the performance of clay as buffer/backfill or sealing material requires robust predictive models covering expected environment conditions and multiphysics phenomena involved in the erosion process. A non-local model development for coupling swelling and erosion of clay in piping flow is presented. The model was validated by comparing its predictions to a series of experimental test, including free swelling tests and erosion assisted by piping flow.

Validations for free swelling demonstrated that the present model captured successfully transient swelling behaviour of MX-80 bentonite. The swelling model is extended to study the self-healing potential of MX-80 bentonite in the gaps/piping channels. The self-sealing capability is investigated by assessing the swelling pressure of bentonite after filling the gaps. The results showed the capability of the present swelling model to quantify the healing performance of MX-80 in stagnant water. For example, the higher dry density (e.g., 1.6 Mg/m<sup>3</sup>) and 1.8 Mg/m<sup>3</sup>) can heal the gaps effectively sooner due to the stronger swelling capacity

The coupled swelling and erosion model shows the capacity to quantify the amount mass loss of swelling clay under piping flow. The trend observed in the experiments, including pinhole test and erosion of plug-in material in borehole, was correctly reproduced by the present model. Compared to the model presented by Navarro et al. (2016) using experimental fitted erosion rate, the predicted cumulated mass loss obtained by this study is directly achieved by establishing the relationship between the internal force among the bentonite particles and shear force induced by flowing water. The amount of mass loss of plug-in material in the end of test was 10.3% of the installed mass. The large amount mass loss induced by erosion was highly suspected to cause the damage of sealing behaviour of bentonite plugs in boreholes.

#### Appendix A

The separation distance between the flat particles (h) is directly controlled by the solid fraction of clay and can be described by (Laxton and Berg, 2006)

$$h = \delta_p \left( \frac{\phi_s^{max}}{\phi_s} - 1 \right) \tag{A-1}$$

where  $\phi_s^{max}$  is the maximum clay solid volume fraction.

The expression of attractive Van der Waals forces  $(F_A)$  and repulsive electrical double layer forces  $(F_R)$  is given by

$$F_{A} = \frac{A_{H}S_{p}}{6\pi} \left[ \frac{1}{h^{3}} - \frac{2}{\left(h + \delta_{p}\right)^{3}} + \frac{1}{\left(h + 2\delta_{p}\right)^{3}} \right]$$
(A-2)

$$F_R = 2cRTS_p(\cosh y^m - 1) \tag{A-3}$$

where  $A_H$  is the Hamaker constant,  $S_p$  is the surface area of the particles, the function  $y^m$  represents the scaled potential in the midpoint between parallel plates. The function  $y^m$  represents the non-dimensional potential in the midpoint between parallel plates of, for

example, a fracture, described by Liu et al. (2009) and Sedighi et al. (2021):

$$y^{m} = \sinh^{-1} \left[ 2\sinh\left(y^{m}_{\infty}\right) + \frac{4}{\kappa h} \sinh\left(\frac{y^{h}_{\infty}}{2}\right) \right]$$
(A-4)

where

$$y_{\infty}^{m} = 4 \tanh^{-1} \left[ 2 \tanh \left( 0.5 \sinh^{-1} \left( \frac{zF\sigma^{0}}{2\varepsilon_{0}\varepsilon_{R}RT\kappa} \right) \right) \exp \left( -\frac{\kappa h}{2} \right) \right]$$
(A-5)

$$y_{\infty}^{h} = 4 \tanh^{-1} \left[ \tanh \left( 0.5 \sinh^{-1} \left( \frac{zF\sigma^{0}}{2\varepsilon_{0}\varepsilon_{R}RT\kappa} \right) \right) \exp(-\kappa h) \right]$$
(A-6)

The Debye length ( $\kappa$ ) is given by:

$$\kappa = \sqrt{\frac{2cz^2 F^2}{\varepsilon_0 \varepsilon_R RT}} \tag{A-7}$$

where *F* is the Faraday's constant; *z* is valence of ions in the mid-plane and  $\varepsilon_0 \varepsilon_R$  is the dielectric constant.

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# **CHAPTER** Seven

# 7. Conclusions and Future works

The aim of the research presented has been achieved by developing a non-local theoretical framework/tool that can be used to predict/study the clay cracking and erosion-assisted damage. This chapter includes a synthesis of these developments and their relations to the objectives set at the start of the work. Key contributions and advancements will be highlighted and opportunities for further work will be identified.

### 7.1 Summary

In order to study the coupled hydro-chemical processes in clay cracking and erosion, theoretical and model developments based on a non-local approach are presented in this thesis. The proposed models are implemented within a new computational framework (Pyramid); built-in MATLAB and presented by the author in Yan et al., 2020). The presented numerical analysis tools provide valuable insights for investigating coupled hydro-chemical effects on erosion and cracking of swelling clays. The confidence of the proposed models is achieved by the validation of the framework with experimental data and its application to a large class of engineering problems. The summary of the work presented in this thesis is described as follows:

1. The comprehensive review of experimental investigations and theoretical developments on cracking and erosion of swelling clays presented in Chapter 2; provided a detailed and comprehensive ground for the establishment of the work, including:

- qualitative and quantitative descriptions of the dynamic cracking process and the evolving erosion processes;
- understanding the advantages and shortcomings of the existing mathematical formulations and numerical analysis approaches for clay desiccation cracking and erosion;
- understanding the limitations of local formulations for describing clay erosion and cracking processes due to the complexity introduced by discontinuous damage and detachment processes;
- familiarisation with the developments and applications of a non-local approach (peridynamics) for multi-physical problems;
- selection of the peridynamic theory as the basis for this project developments due to its ability to handle large deformations and discontinuities.

2. The non-local framework for coupling water flow and chemical transport in unsaturated porous media presented in Chapter 3 was successfully validated by a set of benchmarks and its predictive capability was demonstrated by analysis of coupled chemical transport and water flow problem in 2D domains with stationary and propagating cracks and various randomly distributed inclusions.

- Notably, the model is applicable to multi-physics problems in porous systems where coupled hydro-chemical degradations can create discontinuity or heterogeneity.
- The developments of non-local water flow and chemical transport models was adopted to investigate the hydro-chemical processes in cracking (Chapter 4) and erosion (Chapter 5) of clay.

3. The integration of mechanical deformation and failure presented in Chapter 4 was successfully validated with two set of experimental data, including ring test and long clay bar test.

- The results demonstrated that the model captured realistically key hydraulic, mechanical and geometry effects on clay desiccation cracking.
- Crack initiation was studied by investigating the correlation between the shrinkage of clay, changes in displacement fields and crack growth in ring specimens with different dimensions.

4. The new erosion model presented in Chapter 5, which brought together clay swelling, particle detachment, and detached particle transport, into a single modelling tool, was validated in several steps:

- The validation of the detachment part of the model with a series of experimental benchmarks showed excellent agreement between calculated and experimental results.
- The validation of the free swelling part of the model with experimental data demonstrated that the swelling part of the model captured successfully the effects of ion concentration, ion types and dry density on the bentonite swelling behaviour. The simulation results indicated a dual effect of the ion concentration: i) the swelling strain decreased with increasing ion concentration; and ii) the solid volume fraction at the water-clay interface increased with increasing ion concentration.
- The full erosion model was validated successfully with a series of experimental results for the behaviour of compacted bentonite under different eroding environments, firstly under

conditions prohibiting particle detachments (bentonite expansion and extrusion) and then under conditions facilitating particle detachments (bentonite erosion).

• The set of validations of the erosion model demonstrated that the model predictions for the coupled effect of water chemistry and flowrate on penetration, extruded mass, and particle release rate, were in good match with the available experimental data and allowed for explaining the experimentally observed trends. The simulation results showed that the extrusion distance, which is controlled by both clay swelling and detachment processes, was a nearly linearly dependent on the water flowrate irrespective of the water chemistry. The higher water flowrates were shown to induce nonlinearly increasing erosion rates in accordance with experiments.

5. The model for co-transport of smectite particles and suspended accessory mineral particles presented in Chapter 5.3, was developed to explain the role of accessory minerals in the erosion protection of bentonite clay and was validated by two sets of experimental data.

- It was demonstrated that the model reproduced the main experimental observations of build-up of mineral film and of its growth with time.
- The model was used to study parametrically the effect of water chemistry, filtration rate and mineral ratio on the swelling and erosion of mineral-rich clays, and it was shown that higher ion concentration, filtration rate and mineral content reduce the swelling of bentonite.
- The model was used to determine quantitatively the long-term behaviour of mineral-rich bentonite in repository conditions. The results showed that the presence of minerals could reduce the loss of bentonite by a factor of 2 compared to pure smectite bentonite. It was highlighted that assessments of erosion of mineral-rich bentonite erosion based on experimental data obtained with pure smectite would be overly conservative.

6. The model extension to erosion of swelling clay in cracks/piping channels presented in Chapter 6 was applied to investigate the self-healing potential of MX-80 bentonite in the gaps/piping channels, and the results indicated that channels could be effectively filled by MX-80 with high initial dry density (e.g. 1.6 Mg/m<sup>3</sup> and 1.8 Mg/m<sup>3</sup>).

• The coupled swelling and erosion model showed capacity to quantify the amount of mass loss of swelling clay under piping flow. The trend observed in the experiments, including pinhole test and erosion of plug-in material in borehole, was correctly reproduced by the present model.

• Compared to a model using experimental fitted erosion rate, the cumulative mass loss obtained by the new model was directly calculated by establishing the relationship between the internal force among the bentonite particles and shear force induced by flowing water. The amount of mass loss of plug-in material in the end of test was 10.3% of the installed mass. The large mass loss by erosion was suggested to cause damage to bentonite plugs in boreholes.

## 7.2 Conclusions

The following conclusions can be drawn from the work:

#### 7.2.1 Theoretical and formulation developments

Based on the state of the art review presented in chapter 2, the understanding of coupled hydrochemical effects on clays' engineering behaviour has progressed substantially through extensive experimental tests and theoretical developments. While substantial progress has been made in coupling mechanical with hydro-chemical effects (HCM models) for analysis of, e.g., shrinkage and swelling of expansive clay, the progress in modelling important phenomena involving discontinuous processes, such as clay erosion and desiccation cracking, has been very limited. Modelling clay erosion and cracking requires to understand the complex mechanisms involved (e.g., large deformation, damage, phase change and particle detachment at reaction interface) and tackle the discontinuities evolving during the wetting or drying period. It was realised that the traditional local approaches (e.g., finite element, finite difference and finite volume method) are less efficient in solving cracking and erosion problems, which required the assistance of the introduction of special elements in local approaches. The computational complexity and challenges of the coupled problems resulted in a high demanding of the numerical approach in addressing multiple physical fields and discontinuous processes. This leads to the ambition of this work by creating a non-local framework, which has been an evidently efficient tool in handling large deformation and discontinuities, for clay erosion and desiccation by integrating the hydro-chemical processes with mechanics of the problem.

An important development was initiated by developing a non-local framework for coupling water flow and chemical transport in variable saturated clay in Chapter 2. An implicit numerical scheme is adopted to solve the coupled problems. The developed models are verified by a set of benchmarks. The ability of non-local models for solving discontinuous or heterogeneous material are demonstrated by analysis of coupled chemical transport and water flow problem

in 2D domains with stationary and propagating cracks and various randomly distributed inclusions. The simulations illustrate how the model can capture naturally the effects of static and dynamics discontinuities and heterogeneities, which is problematic or computationally demanding for numerical schemes based on local formulations. Important back-flow and back-diffusion phenomena which are caused by the existence of permeable inclusions, which promote the appearance of fast paths as well as regions of hold up, are captured and explained by the present model.

Clay desiccation model was developed via coupling hydro-chemical and mechanical effects. The developed non-local model aims to address the challenge of modelling the clay desiccation cracking which involves a strong degree of coupling between hydraulic and mechanics and more importantly the strong mathematical discontinuity that exists as the result of fracture development in the system. The above models are relevant to the objective 1.

A major contribution of theoretical developments for erosion problem of swelling clay is that we introduced a new criterion for detachment based on the particle interaction forces, i.e. an intrinsic criterion, rather than prescribed phenomenologically, which address a critical missing element in the existing models related to the detachment of particles from the gel phase at the solid/fluid interface and its strong coupling with hydro-chemical effects. This is pertinent to the objective 2 in which the proposed detachment model linked dynamically the solid swelling and particles release processes at the solid/liquid interface via comparing the shear stress at the solid/liquid interface induced by water flow with a material parameter limiting the solid behaviour of the gel.

Revisiting the clay water interaction theory produced a non-local modelling framework that brings together clay swelling, particle detachment, and detached particle transport, into a single modelling tool. Three different peridynamics bonds, including solid-solid (S-S), interfacial (S-L) and liquid-liquid (L-L) bonds, are defined to facilitate the modelling of clay erosion. Within the PD framework the phase changes from hydrated solid to swelling paste and clay gel are evolving naturally through the changing strengths of S-S bonds. The detachment of diluted clay particles at the interface of clay/fluid is controlled by the interfacial bonds (S-L). Additionally, the detached particle transport within water is controlled by the L-L bonds. The particles detachment at the moving interface is considered as a discontinuous evolution process since the free swelling/expansion and detached particle transport in water follow different governing equations.

In order to explain the role of accessory minerals in the erosion protection of bentonite observed in experiments, a model for co-transport of smectite particles and insoluble accessory mineral particles during expansion of bentonite was developed based on a dynamic force balance, considering gravity, buoyant force, diffusional force, viscous drag force, attractive van der Waals forces and repulsive double layer forces. The mineral particles are deposited at locations where the repulsive forces between the smectite particles are week and cannot support the accessory mineral particles, which results in a mineral film protecting the erosion of bentonite.

#### 7.2.2 Desiccation cracking behaviour of clay

The non-local desiccation model presented in this thesis showed that predictions were demonstrated to be in very good qualitative and quantitative agreement with experiments, lending strong support for the model predictive capability. Firstly, the numerical model reproduced experimentally observed characteristics of desiccation cracking in two ring tests, such as stress distribution, sample geometry effects, cracking initiation and propagation, including the number and the non-uniform spacing of cracks due to clay heterogeneity. Secondly, the model captured the main phases observed in desiccation cracking of long clay bars, as well as the instance of first crack emergence and the number and the non-uniform spacing of cracks.

These outcomes suggest that the proposed model is sufficiently physically realistic to investigate outstanding questions on geometry effects identified in the thesis. Specifically, a single crack emerged at the inner surface of the ring with small inner radius (e.g., r=2.11 cm) and extended towards the outer surface. However, multiple cracks (e.g., 6 cracks) emerged at the inner radius with large inner radius (e.g., 15 cm) and propagated towards the outer surface to cause fragmentation. For the large geometry, it appears that the initiation of one crack is not sufficient to relax the hoop stress along the entire inner surface, which allows for initiation of multiple cracks. The results provide confidence that the proposed modelling approach is physically realistic and can be used to investigate physical, mechanical, and geometric effects on clay desiccation cracking.

### 7.2.3 Erosion under coupled hydro-chemical conditions

The integrated erosion model showed the capacity to capture three main physical processes involved in erosion of compacted bentonite under eroding environments, including free swelling of clay from the initial hydration solid to gel and eventually transformed into diluted sol particles by identifying the solid content of clay at different stages, diluted sol detached at clay/water/rock interface, and released particle transport within the flowing water. The simulation results supported strongly the applicability of the model to account for the hydrochemical conditions (water composition and velocity) of the eroding environment. The model can be used to quantify the effects of the key parameters controlling erosion, specifically the effects of water chemistry and velocity (objective 3).

The chemistry in the solution determines whether diluted clay particles can be released or not. This value referred to as the Critical Coagulation Concentration (CCC) can be approximated by introducing a thermal deviation factor based on the free swelling model, which can explain how the thickness of clay plate, surface area and charge of particle affected the behaviour of diluted particle at the clay/water interface under different water chemistry. The variations of these properties determined the stability of diluted clay particles. The effects of water chemistry on swelling and erosion of clay can be summarized as follow:

- The swelling deformation of the sample decreases with increasing ion concentration;
- The clay solid volume fraction at the expanding front is sharper for higher ion concentrations.
- The exchange reaction of Ca<sup>2+</sup> in clay and Na<sup>+</sup> in solution can accelerate the expansion velocity and increase the extrusion distance of bentonite. For example, the extrusion distance of bentonite considering the exchange reaction can be 1.5 times larger than that without the exchange reaction.
- A higher concentration induces a larger critical solid volume fraction at the detachment interface.

The effects of water velocity on swelling and erosion of clay can be summarized as follow:

- Increasing the velocities of the eroding fluid lead to increasing critical clay volume fraction at clay/water interface.
- The extrusion distance is a nearly linearly dependent on water flowrate.
- The results suggest two regimes for the erosion rate nearly linear dependence on the flowrate for rates lower than  $10^{-5}$  m/s, and accelerated dependence for higher rates.

Additional processes, that have been taken into account, through the modelling efforts described in this thesis, are co-transport of accessory mineral particles in raw bentonite materials (objective 4). The accessory mineral particles can lead to building up a mineral film, which may slow down or even possibly stop the loss of clay mass. The modelling results further showed that increasing additional minerals in the bentonite may significantly decrease the

bentonite swelling strain or restrain the swelling properties of bentonite. For example, the presence of accessory minerals may reduce the loss of bentonite by a factor of 2 compared to a pure smectite bentonite. This suggests that assessments of erosion of bentonite containing accessory minerals based on experimental data obtained with pure smectite may be overly conservative.

The applications of erosion model are further extended to study the erosion assisted by piping flow, specifically erosion of MX-80 bentonite in pinhole and plug-in material in boreholes (objective 5). The swelling model is extended to investigate the self-healing potential of MX-80 bentonite in the gaps/piping channels. The self-sealing capability, which is the most important characterization of bentonite as buffer/backfill/plug-in material, is investigated by using the present swelling model. The results showed the capability of the present swelling model to quantify the healing performance of MX-80 in stagnant water. For example, the higher dry density (e.g., 1.6 Mg/m<sup>3</sup> and 1.8 Mg/m<sup>3</sup>) can heal the gaps effectively sooner due to the stronger swelling capacity. Compared to the model using experimental fitted erosion rate, the predicted cumulated mass loss obtained by this study is directly achieved by establishing the relationship between the internal force among the bentonite particles and shear force induced by flowing water. The amount of mass loss of plug-in material in the end of test was 10.3% of the installed mass, which was highly suspected to cause the damage of sealing behaviour of bentonite plugs in boreholes.

#### 7.3 Suggestions for further research

(1) The developed model was applied to simulate the cracking and erosion of swelling clay under different hydro-chemical conditions, but the computational implementation was limited to 2D geometries. Experimental observations show that cracking of clay is three-dimensional, significantly affected by the geometry of the clay sample and boundary conditions, which may lead to different initiation and propagation of cracks and final crack patterns. It is therefore, recommended to extend the implementation of the theoretical framework to three-dimensional problems. A major challenge of 3D modelling of clay cracking is the difficulty of defining and measuring model parameters (e.g., heterogeneities of clay and boundary effects) as well as high computational cost.

(2) Wetting-drying circles of swelling clay may lead to the formation of desiccation cracking and erosion features in one single system. For example, geosynthetic clay liners (GCLs) can experience loss of bentonite if exposed to cyclic wetting (by hydration from the subsoil) and drying (due to evaporation). The initial wet/dry cycle caused desiccation cracks to form in the bentonite. These cracks may be filled during wetting period due to the swelling performance of bentonite and assist the erosion of bentonite particles. The cracking and erosion model developed in this thesis may be applied to study the coupled processes of erosion and cracking during the wetting/drying cyclic.

(3) The investigation of erosion behaviour of swelling clays in Chapter 5 is limited to a single straight fracture of constant width. Natural fractures have generally non-constant apertures and are spatially distributed due to the complex heterogeneous nature of the rocks. The geometries of these natural fractures can affect the groundwater flow field, which may result in different erosion behaviour of bentonite buffer. The effects of fracture geometries and fracture systems on bentonite erosion behaviour require further investigations, which can be based on the model proposed in this work with significantly more computational power.

(4) The co-transport model presented in Chapter 5 (Sec. 5.5) focuses on addressing the role of non-reactive minerals on clay erosion/expansion behaviour. However, the importance of precipitation and dissolution of minerals during erosion and gel formation is identified in the existing experimental observations. The present model can be extended to couple geochemical reactions to describe the kinetics of the problem for an accurate description of swelling and gel formations. Additionally, the influences of fracture aperture variability and surface roughness on the deposition of mineral particles and erosion behaviour of bentonite buffer should be further investigated.

(5) The erosion assisted by flow in existing piping channels was investigated in Chapter 6. The mechanisms of the formations of piping channels in swelling clays are not well understood. The piping erosion reviewed in Chapter 2 indicated that piping might occur and create a preferential flow path for water transport when water pressure is larger than the hydraulic resistance of interfaces. Further developments of the piping model can improve the understanding of erosion assisted by piping processes.

(6) The developed non-local models for cracking and erosion of swelling clays are robust and flexible computational tool which incorporated advanced theoretical and experimental knowledge into the coupled physico-chemical and mechanical modelling. The present models in general can be extended for the applications to the cracking predictions of other brittle material, dam failure, and erosion and cracking behaviour of engineering barriers.