Analytical solution for clay plug swelling experiments

Simon A. Mathias[a,*], H. Chris Greenwell[a], Charlotte Withers[a], Ali R. Erdogan[a], Jim N. McElwaine[a], Chris MacMinn[b]

[a]*Department of Earth Sciences, Durham University, Durham, UK*
[b]*Department of Engineering Science, Oxford University, Oxford, UK*

Abstract

Clay swelling experiments frequently involve monitoring the one-dimensional displacement with time of an initially dry clay plug as it imbibes water from a supply at its base. This article presents a new analytical solution for interpreting such experiments based on Richards' equation for flow in a partially saturated porous medium combined with a linear empirical function relating moisture ratio with void ratio. The analytical solution is described by just two parameter groups. The first parameter group describes the swelling potential of the clay. The second parameter group describes the rate at which the swelling plug reaches equilibrium, which is controlled by permeability and capillary pressure. Application of the analytical solution is demonstrated by calibration to one-dimensional displacement data from clay swelling experiments for an illite and bentonite clay.

Keywords: Swelling, Unsaturated, Clay, Diffusion, Absorption, Richards' equation

1. Introduction

Compacted Wyoming bentonite, a clay rock composed predominantly of sodium montmorillonite, is widely used as a barrier material for nuclear radioactive waste repositories, as an adsor-

*Corresponding author. Tel.: +44 (0)1913343491, Fax: +44 (0)1913342301, E-mail address: s.a.mathias@durham.ac.uk

bent, and as an analogue for swelling clay mineral rich sub-sea shale formations (Komine & Ogata, 1994). The two-dimensional layered structure of aluminosilicate clay minerals results in a high surface area and, combined with the presence of exchangeable cations, gives rise to the adsorbent properties of bentonite for radionuclides (for example, Cs^+), and other contaminants (Eriksen et al., 1999).

These properties of bentonite and related clay minerals also result in a propensity to spontaneously hydrate upon contact with water. To accommodate this hydration, the bulk volume of the clay mineral must increase if the material is unconfined; if confined, a swelling pressure arises instead. Upon swelling, the transport properties of the hydrated clay mineral material also change due to the changes in pore size distribution.

One manifestation of swelling that presents significant challenges to oil and gas operations is shale instability. Oil and gas reservoirs are usually topped by an impervious cap rock, which holds the less dense hydrocarbons within the subterranean reservoir. In most cases, the caprock will be a clay mineral rich shale. Thus shales are invariably encountered when drilling the well-bore to access the oil reservoir. To clear the rock fragments (cuttings) from the well-bore during drilling, and to maintain hydrostatic pressure in the cleared hole and thus formation integrity, a technical drilling fluid is used. This fluid is colloquially known as “mud”. In the early days of oil exploration, oil-based muds (OBM) were generally used, based on crude oil. However, owing to the environmental impacts of OBMs, drilling engineers have focussed on developing less environmentally damaging water-based muds (WBM).

The clay minerals present in shale may either disperse (for example kaolinite) or swell (for example montmorillonite) upon contact with WBMs, causing instability in the well bore region,

such as swelling/sloughing, in the medium-long term, or increased plasticity and the aggregation of shale cuttings to the drill string ("bit balling") in the short term (Anderson et al., 2010). The swelling of clay is strongly controlled by the composition of pore-water (Chai et al., 2014; Chen, 2016; Chavali et al., 2017; Oren and Akar, 2017). To address these problems, the industry has tried to increase the environmental performance of WBMs through use of more sophisticated water-based technologies including small organic molecules to inhibit clay mineral/shale swelling. Compacted bentonite is frequently used as a model in which to probe the efficacy of the swelling inhibitors.

The swelling properties of clay minerals have long been studied, in aqueous brines and in organic solvents (Anderson et al., 2010), though often at the single crystal level. Clay crystal swelling has been partitioned into both crystalline and osmotic swelling regimes (Anderson et al., 2010). Studies have primarily focussed on the equilibrium swelling of the clay minerals, with the expansion of the crystal at given conditions of temperature and salinity tested against classical colloid theories, e.g. DLVO theory (Smalley, 2006). Though the crystal chemistry and swelling potential of clay minerals are relatively well understood (Wilson & Wilson, 2014; Anderson et al., 2010), the link between clay mineral crystal swelling and shale stability, where many components influence the bulk swelling (Van Oort, 2003), is less clear, and it has even been argued that clay mineral swelling properties are not relevant in the oil field and that shale swelling may not occur when artefacts of experiments are carefully accounted for (Santarelli & Carminati, 1995).

It is inherently challenging to create compacted shale-like materials that replicate the properties and behaviour of saturated shale under wellbore conditions. As such, compacted bentonite and other natural clays will remain in use as a proxy for shales to allow testing of drilling fluid

formations and additives and it is imperative to account for the artefacts introduced during production of compacted clay mineral shale analogues to be able to discern performance improvements in new drilling fluid technologies (Santarelli & Carminati, 1995). In particular, and especially to attempt to understand bit balling, the short term swelling rates of clay minerals need to be better understood.

To address this, a non-contact one-dimensional displacement meter has been developed and used to measure the short term swelling response of compacted bentonite tablets (Sellick et al., 2017). Similar experiments in this context are also reported by Chen (2016). Modelling of the swelling process can help reveal the relative importance of processes contributing to the compacted bentonite swelling. Whereas very accurate computational chemistry methods, such as atomistic molecular dynamics, have enabled swelling energetics to be probed at the clay mineral crystal scale (Suter et al., 2011), and more complex multi-scale methods have been developed (Suter et al., 2015), these are computationally very expensive. The objective of this study is to improve understanding about the kinetics of hydration processes through an analytical model, which allows rapid calculation of swelling curves to be tested against experimental data.

Our approach builds on modeling techniques previously developed to understand swelling and shrinkage of clay rich soils (Philip, 1969; Smiles and Raats, 2005; Su, 2010). Such models employ Richards' equation to describe water movement through partially saturated porous media combined with empirical relationships between moisture ratio (volume of water / total volume) and void ratio (volume of voids / total volume) (Peng and Horn, 2007). The swelling of clay gives rise to moving boundary conditions. However, these can be dealt with by using a material coordinate system, associated with a theoretical dry clay mass, as opposed to a spatial coordinate system

(Philip, 1969; Su, 2010).

In the context of the swelling experiments described above, the resulting non-linear partial differential equation (PDE) have traditionally been solved numerically using finite differences (Kim, 1999). Although a range of analytical and quasi-analytical solutions for this class of problem have been sought in the past (Raats, 2002), these have mostly been in the context of infiltration. In this article, a linear relationship between moisture ratio and void ratio is assumed such that the resulting PDE is linear and can be solved analytically. An exact analytical solution for the one-dimensional displacement of a compacted clay tablet as a function of time is then derived. Finally, the efficacy of the analytical solution is demonstrated by calibration to experimental data sets from two different clays.

2. Mathematical model

The approach used here was originally developed by Philip (1969) in the context of swelling soils, and has been frequently applied in the context of soil-deformation modeling (Kim, 1999; Smiles and Raats, 2005; Su, 2010). An explanation of how this approach can be used to derive an analytical solution for clay plug swelling experiments is described as follows.

The bulk volume of an unsaturated clay mass, V_b [L^3], can be defined by

$$V_b = V_w + V_c + V_a \tag{1}$$

where V_w [L^3], V_c [L^3] and V_a [L^3] are the volumes of water (including free and adsorbed water), clay mineral (excluding all water) and air present within the bulk clay mass of concern, respec-

tively.

The swelling of clay due to the presence of water manifests itself through a strong correlation between void ratio, e [-], and moisture ratio, ϑ (e.g. Peng and Horn, 2007; Chertkov, 2012), defined by:

$$e = \frac{V_w + V_a}{V_c} \tag{2}$$

$$\vartheta = \frac{V_w}{V_c} \tag{3}$$

A simple way to represent this correlation is to assume that e and ϑ are related by the linear function

$$e = e_I + (\vartheta_s - e_I)\left(\frac{\vartheta - \vartheta_I}{\vartheta_s - \vartheta_I}\right) \tag{4}$$

where e_I [-], ϑ_I [-] and ϑ_s [-] represent the initial void ratio, initial moisture ratio and maximum moisture ratio of a clay sample, respectively.

Consider water movement through the bulk clay associated with a mineral clay volume of δV_c. Let δQ [$\mathrm{L^3T^{-1}}$] be the net rate of out-flowing water. Assuming the water and mineral clay to be incompressible, the change in moisture ratio, $\delta\vartheta$, over a a time-period, δt [T], is found from

$$\delta\vartheta\delta V_c = -\delta Q\delta t \tag{5}$$

such that for an infinitesimal time-period and mineral clay volume

$$\frac{\partial\vartheta}{\partial t} = -\frac{\partial Q}{\partial V_c} \tag{6}$$

2.1. One-dimensional vertical flow

Now consider a vertically oriented cylindrical clay plug encased within a rigid and impermeable sleeve. The clay is initially dried to a uniform moisture ratio, ϑ_I, and uniform void ratio, e_I. The base of the clay plug is then soaked in water at a fixed pressure whilst the top of the plug is exposed to the atmosphere. In this way, the volumetric flow rate of water, Q, can be assumed to be described by the following form of Darcy's law (Philip, 1969)

$$Q = -\frac{Akk_r}{\mu_w}\frac{\partial(P_w + \rho_w gz + P_o)}{\partial z} \tag{7}$$

where A [L^2] is the cross-sectional area of the clay plug (which is constant due to the rigid sleeve), k [L^2] is the permeability, k_r [-] is relative permeability (which is a function of ϑ), P_w [$ML^{-1}T^{-2}$] is water pressure, μ_w [$ML^{-1}T^{-1}$] is the dynamic viscosity of water, ρ_w [ML^{-3}] is the density of water, g [LT^{-2}] is gravitational acceleration, z [L] is elevation and P_o [$ML^{-1}T^{-2}$] is the so-called overburden pressure associated with the work done in swelling the clay. See Raats (2002) for further discussion with regards to the overburden pressure term.

Capillary pressure, P_c [$ML^{-1}T^{-2}$], is defined by $P_c = P_a - P_w$ where P_a [$ML^{-1}T^{-2}$] is the air pressure. Importantly, capillary pressure is a function of ϑ. Analogous to Richards' equation, assuming the air pressure to be constant throughout and capillary pressure variations to be much larger than those associated with gravity head and overburden pressure (which is very likely in clay media), Eq. (7) reduces to

$$Q = \frac{Akk_r}{\mu_w}\frac{\partial P_c}{\partial \vartheta}\frac{\partial \vartheta}{\partial z} \tag{8}$$

The relevant initial and boundary conditions are:

$$\begin{aligned} &\vartheta = \vartheta_I, \quad 0 \le z \le H_I, \quad t = 0 \\ &\vartheta = \vartheta_s, \quad z = 0, \qquad\quad t > 0 \\ &Q = 0, \quad z = H(t), \qquad t > 0 \end{aligned} \tag{9}$$

where H_I [L] is the initial height of the clay plug and $H(t)$ [L] is the height of the clay plug at time, t. Note that the height of the clay plug increases with time as the clay plug swells due to water adsorption within the clay layers.

2.2. Transformation to a static boundary condition

Considering Eqs. (1) and (2), the relationship between V_c and z is defined by

$$\frac{\partial V_c}{\partial z} = \frac{A}{e+1} \tag{10}$$

Let us now consider an alternative coordinate, ζ [L], defined by

$$\frac{\partial V_c}{\partial \zeta} = \frac{A}{e_I+1} \tag{11}$$

It can be understood that

$$\frac{\partial \zeta}{\partial z} = \frac{e_I+1}{e+1} \tag{12}$$

and furthermore that

$$H(t) = \int_0^{H_I} \frac{e+1}{e_I+1} d\zeta \tag{13}$$

Eqs. (6), (8) and (9) map to the ζ coordinate system to form a non-linear diffusion problem as follows:

$$\frac{\partial \vartheta}{\partial t} = \frac{\partial}{\partial \zeta}\left(D_A(\vartheta)\frac{\partial \vartheta}{\partial \zeta}\right) \tag{14}$$

$$\begin{aligned} &\vartheta = \vartheta_I, \quad 0 \le \zeta \le H_I, \quad t = 0 \\ &\vartheta = \vartheta_s, \quad \zeta = 0, \qquad\quad t > 0 \\ &Q = 0, \quad \zeta = H_I, \qquad\quad t > 0 \end{aligned} \tag{15}$$

where D_A [$\mathrm{L}^2\mathrm{T}^{-1}$] is the apparent diffusion coefficient, defined by

$$D_A = -\frac{(e_I + 1)^2 k k_r}{(e + 1)\mu_w}\frac{\partial P_c}{\partial \vartheta} \tag{16}$$

The above set of equations can also be derived using the large deformation mathematical framework of Song & Borja (2014).

2.3. Analytical solution for linear diffusion coefficient

For the special case when D_A can be treated as a positive constant, the above problem has an analytical solution of the form (similar to Crank, 1975, p. 47)

$$\frac{\vartheta - \vartheta_I}{\vartheta_s - \vartheta_I} = 1 - 2\sum_{n=0}^{\infty}\frac{1}{\lambda_n}\exp\left(-\frac{\lambda_n^2 D_A t}{H_I^2}\right)\sin\left(\frac{\lambda_n \zeta}{H_I}\right) \tag{17}$$

where $\lambda_n = (n + 1/2)\pi$.

Substituting Eq. (17) into Eq. (4) and then into Eq. (13) leads to

$$h(t) = \alpha \left[1 - 2 \sum_{n=0}^{\infty} \frac{1}{\lambda_n^2} \exp\left(-\beta \lambda_n^2 t \right) \right] \tag{18}$$

where

$$h(t) = \frac{H - H_I}{H_I}, \quad \alpha = \frac{\vartheta_s - e_I}{e_I + 1}, \quad \beta = -\left(\frac{e_I + 1}{H_I} \right)^2 \frac{k k_r}{(e+1)\mu_w} \frac{\partial P_c}{\partial \vartheta} \tag{19}$$

and $h(t)$ [-] is hereafter referred to as the swelling ratio. Note that this is the first and only time that the linear relationship between e and ϑ (Eq. (4)) is invoked.

For a zero e_I, $\alpha = \vartheta_s$. The α parameter says something about the adsorption capacity of the clay. Higher values of α imply greater adsorption potential and hence greater swelling potential. The β parameter represents the rate at which a clay swelling experiment equilibrates. Large values of β correspond to high permeability and/or high capillary pressures. Large β values lead to the clay reaching equilibrium faster.

3. Calibration to experimental data

Artificial cylindrical plugs of clay were processed with a thickness of 3.8 mm and a diameter of 20 mm. The clay plugs were placed into circular sample holders with the base exposed to a water bath. Clay plugs within the sample holders sat on top of circular perforated disks to enable hydraulic contact between the water bath and the base of the plugs. The sample holder was also designed to constrain the plug laterally such that swelling only occurs in the vertical direction. Vertical displacement of the upper surface of the clay plug was measured using an induction sensor non-contact one-dimensional displacement meter. The displacement of the upper

surface in the course of time was monitored. Two clay plugs were examined comprised of illite and bentonite. Using illite and bentonite (predominantly sodium montmorillonite) provide near end members in terms of clay swelling response in terms of crystalline swelling. Assuming similar particle size/void space, the additional swelling of any other minerals should fall between or close to these materials.

Values of α [-] and β [T^{-1}] were obtained by calibrating Eq. (18) to the observed data. Calibration was achieved by minimizing the mean absolute error (MAE) between observed and simulated h using MATLAB's local optimization routine, FMINSEACH. A comparison of observed and calibrated model results are presented in Fig. 1. The associated calibrated parameters and MAE are presented in Table 1. It is clear that the two parameter analytical solution is capable of catching almost all aspects of the data presented

Consider again Eq. (19). The fact that α is larger for bentonite than for illite suggests that bentonite has a larger ϑ_s and hence a greater potential for water adsorption. The fact that β is larger for illite as compared to bentonite suggests that illite has a higher permeability and/or a higher capillary pressure, leading to faster capillary suction of water.

4. Conclusions

The objective of this study was to develop an analytical solution to aid interpretation of one-dimensional displacement observations from clay swelling experiments. An appropriate mathematical model was developed by combining Richards' equation for flow in partially saturated porous media with a linear empirical function relating void ratio with moisture ratio. By ignoring gravity and overburden pressure and assuming a constant relative permeability and moisture ratio

Table 1: Model calibration results.

Clay type	α (%)	β (hour^{-1})	MAE (%)
Illite	44.4	10.1	0.747
Bentonite	70.1	1.06	0.794

- capillary pressure ratio, the associated non-linear partial differential equation reduced to a linear diffusion equation for the water ratio. Making use of Crank (1975), an analytical Fourier series solution for the water ratio in the swelling clay plug was found. This solution was substituted into the linear void ratio–moisture ratio function to obtain an expression for the void ratio, which in turn was used to calculate the height of the sample in the course of time.

The analytical solution reveals that one-dimensional displacement is controlled by two important parameter groups, α and β (recall Eq. (19)). The α parameter is linked to the maximum moisture ratio for the clay. Large α values imply higher potential for water adsorption and swelling. The β parameter is linked to permeability and capillary pressure and represents the rate at which a clay swelling experiment reaches equilibrium.

The analytical solution was successfully calibrated to experimental data for two very different clays: illite and bentonite. The illite was found to have a lower swelling potential (low α) but was found to equilibrate relatively quickly (high β). In contrast, the bentonite was found to have a much greater swelling potential (high α). However, swelling in the bentonite was found to develop more slowly (low β).

Anticipated future work will look at empirical relationships between α and β and various drilling fluid compositional characteristics, with a view that these can be used to simulate drilling fluid improvements on oil-reservoir scale models of clay swelling phenomena.

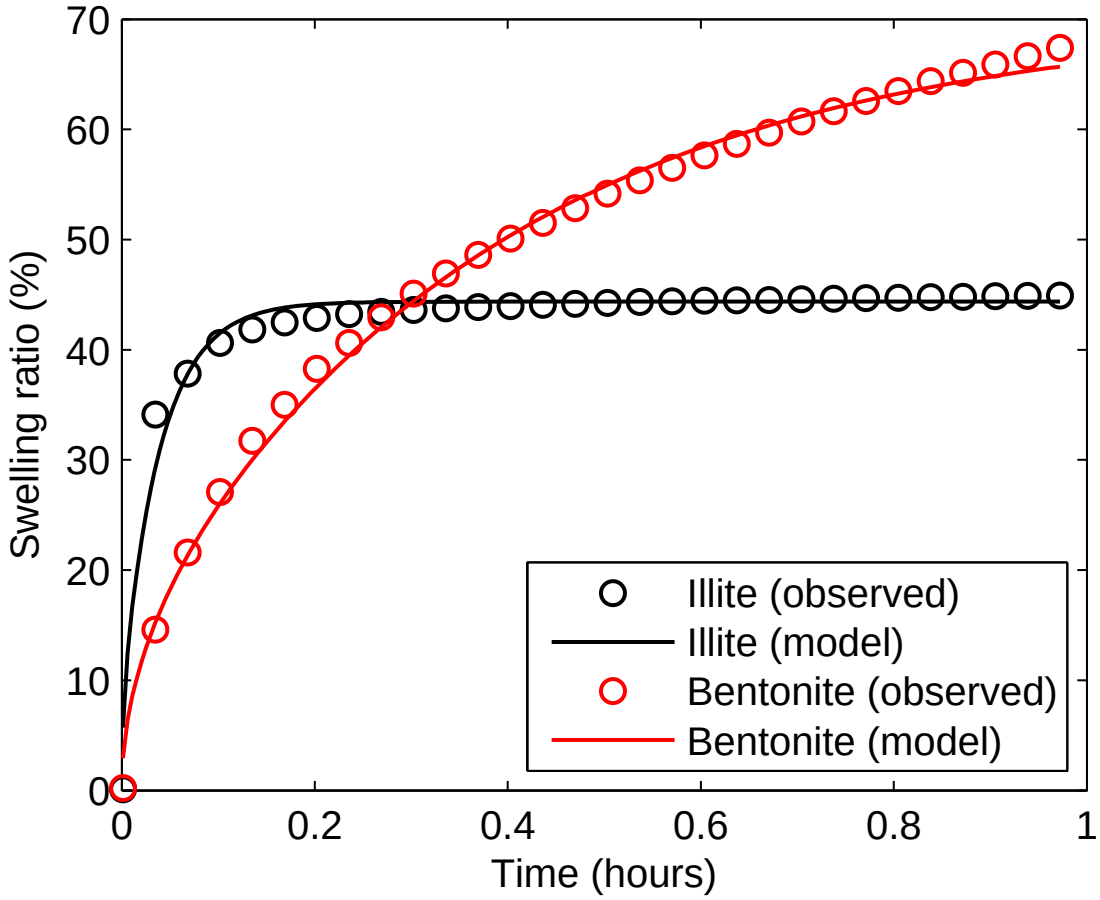

Figure 1: Plots of swelling ratio against time as observed during the experiments and as simulated from the calibrated models, according to Eq. (18).

5. Acknowledgements

We are very grateful for the useful comments provided by Peter Raats.

6. References

Anderson, R. L., Ratcliffe, I., Greenwell, H. C., Williams, P. A., Cliffe, S., Coveney, P. V. (2010). Clay swelling – A challenge in the oilfield. Earth-Science Reviews, 98, 201–216.

Chai, Z. Y., Kang, T. H., Feng, G. R. (2014). Effect of aqueous solution chemistry on the swelling of clayey rock. Applied Clay Science, 93, 12-16.

Chavali, R. V. P., Vindula, S. K., Babu, A., Pillai, R. J. (2017). Swelling behavior of kaolinitic clays contaminated with alkali solutions: A micro-level study. Applied Clay Science, 135, 575-582.

Chen, Y. G., Zhu, C. M., Ye, W. M., Cui, Y. J., Chen, B. (2016). Effects of solution concentration and vertical stress on the swelling behavior of compacted GMZ01 bentonite. Applied Clay Science, 124, 11–20.

Chertkov, V. Y. (2012). Physical modeling of the soil swelling curve vs. the shrinkage curve. Adv. Water Resour., 44, 66–84.

Crank, J. (1975). The Mathematics of Diffusion: 2nd Ed. Clarendon Press.

Eriksen, T. E., Jansson, M., Molera, M. (1999). Sorption effects on cation diffusion in compacted bentonite. Engineering Geology, 54, 231–236.

Kim, D. J., Jaramillo, R. A., Vauclin, M., Feyen, J., Choi, S. I. (1999). Modeling of soil deformation and water flow in a swelling soil. Geoderma, 92, 217–238.

Komine, H., Ogata, N. (1994). Experimental study on swelling characteristics of compacted bentonite. Canadian Geotechnical Journal, 31, 478–490.

Oren, A. H., Akar, R. C. (2017). Swelling and hydraulic conductivity of bentonites permeated with landfill leachates. Applied Clay Science, 142, 81–89.

Peng, X., Horn, R. (2007). Anisotropic shrinkage and swelling of some organic and inorganic soils. Eur. J. Soil Sci., 58, 98–107.

Philip, J. R. (1969). Hydrostatics and hydrodynamics in swelling soils. Water Resour. Res., 5, 1070–1077.

Raats, P. A. C. (2002). Flow of water in rigid and non-rigid, saturated and unsaturated soils. In: The Modeling and Mechanics of Granular and Porous Materials edited by G. Capriz, V. N. Ghionna and P. Giovine, in the series Modeling and Simulation in Science, Engineering and Technology Series edited by N. Bellomo, pp.181–210.

Santarelli, F. J., Carminati, S. (1995). Do shales swell? A critical review of available evidence. In SPE/IADC Drilling Conference. Society of Petroleum Engineers.

Sellick, C., Underwood, T. R., Erdogan, A. R., Patel, R., Skipper, N., Greenwell, H. C. (2017). Assessing compacted bentonite swelling in organic solvents using a non-contact linear swellometer. Applied Clay Science, In preparation.

Smalley, M. V. (2006). Clay swelling and colloid stability. CRC Press.

Smiles, D.E., Raats, P.A.C., (2005). Hydrology of swelling clay soils. In: Anderson, M.G. (Ed.), Encyclopedia of Hydrological Sciences. Wiley, 1011–1026 (Chapter 67).

Song, X., and Borja, R. I. (2014). Mathematical framework for unsaturated flow in the finite deformation range. Int. J. Num. Meth. Eng., 97, 658–682.

Su, N. (2010). Theory of infiltration: Infiltration into swelling soils in a material coordinate. J. Hydrol., 395, 103–108.

Suter, J. L., Coveney, P. V., Anderson, R. L., Greenwell, H. C., Cliffe, S. (2011). Rule based design of clay-swelling inhibitors. Energy & Environmental Science, 4, 4572–4586.

Suter, J. L., Groen, D., Coveney, P. V. (2015). Chemically Specific Multiscale Modeling of ClayPolymer Nanocomposites Reveals Intercalation Dynamics, Tactoid Self-Assembly and Emergent Materials Properties. Advanced Materials, 27, 966–984.

Van Oort, E. (2003). On the physical and chemical stability of shales. J. Pet. Sci. Eng., 38, 213–235.

Wilson, M. J., Wilson, L. (2014). Clay mineralogy and shale instability: an alternative conceptual analysis. Clay Minerals, 49, 127–145.