Advances in Geo-Energy Research

Vol. 8, No. 3, p. 141-145, 2023

Editorial

Opportunities and challenges in CO_2 geologic utilization and storage

Liwei Zhang[1,2]*, Wolfgang Nowak[3], Sergey Oladyshkin[3], Yan Wang[1], Jiancai Cai[4]

[1]*State Key Laboratory of Geomechanics and Geotechnical Engineering, Institute of Rock and Soil Mechanics, Chinese Academy of Sciences, Wuhan 430071, P. R. China*

[2]*University of Chinese Academy of Sciences, Beijing 100049, P. R. China*

[3]*Institute for Modelling Hydraulic and Environmental Systems, Stuttgart Center for Simulation Science, University of Stuttgart, Stuttgart 70569, Germany*

[4]*State Key Laboratory of Petroleum Resources and Prospecting, China University of Petroleum, Beijing 102249, P. R. China*

Keywords:
Carbon dioxide
flow and transport model
CT scanning
gas storage
3D imaging
cement degradation

Cited as:
Zhang, L., Nowak, W., Oladyshkin, S., Wang, Y., Cai, J. Opportunities and challenges in CO_2 geologic utilization and storage. Advances in Geo-Energy Research, 2022, 8(3): 141-145.
https://doi.org/10.46690/ager.2023.06.01

Abstract:
CO_2 geological utilization and storage is considered as an effective approach to deeply cut anthropogenic CO_2 emissions. It is vital to enhance the amount of CO_2 stored in the subsurface, at the same time to ensure safe and long-term subsurface storage of CO_2 without any CO_2 leakage. Science and engineering research in modeling concepts, experimental approaches, safety assurance and emerging CO_2 geological utilization and storage technologies have driven the advancement of CO_2 geological utilization and storage in recent years. In order to encourage communication and collaboration in CO_2 geological utilization and storage research worldwide, a Sino-German joint symposium titled "Opportunities and Challenges in CO_2 Geologic Utilization and Storage" was organized in Wuhan and Stuttgart from February 22 to 24, 2023, bringing together experts from China, Germany, and other countries. The symposium was jointly organized by Institute of Rock and Soil Mechanics, Chinese Academy of Sciences and Institute for Modelling Hydraulic and Environmental Systems, University of Stuttgart with financial support from the Sino-German Center for Research Promotion. A two-site hybrid meeting was held (participants in China met in Wuhan, participants in Germany met in Stuttgart, and other participants joined the meeting online), attracting more than 100 participants from around the world. The latest studies in the field of CO_2 geological utilization and storage were presented at the symposium.

1. Introduction

In recent years, global warming caused by greenhouse gas emissions such as carbon dioxide (CO_2) has become increasingly significant, resulting in frequent extreme weather, sea level rise and other serious consequences (Soeder, 2021). As a result, reducing greenhouse gas emissions has become a top priority for the international community. CO_2 capture, utilization and storage (CCUS) has been recognized by the international community as an effective carbon emission reduction technology for global economies to achieve "low-carbon" development. China currently has the world's largest CO_2 emissions, about 80% of which come from concentrated sources such as power plants and cement plants (MST, 2019). CCUS technology is specifically designed to tackle concentrated sources of CO_2 emissions, which meets China's needs. China's theoretical CO_2 underground storage capacity is up to 8 trillion tons, enough to meet China's goal of CO_2 emission reduction. Therefore, China is likely to become a "hot spot" for CCUS applications from 2020 to 2060 (IEA, 2022), and CCUS is expected to bring great environmental and economic benefits to China. On September 22, 2020, President Xi clearly stated at the United Nations General Assembly that China's carbon emissions should peak by 2030

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*Corresponding author.
E-mail address: lwzhang@whrsm.ac.cn (L. Zhang); wolfgang.nowak@iws.uni-stuttgart.de (W. Nowak); sergey.oladyshkin@iws.uni-stuttgart.de (S. Oladyshkin); ywang@whrsm.ac.cn (Y. Wang); caijc@cup.edu.cn (J. Cai).

Received April 9, 2023; revised April 25, 2023; accepted May 1, 2023; available online May 5, 2023.

and achieve the goal of carbon emission neutralization by 2060. As a result, CCUS-related research will play a key role in achieving China's carbon reduction targets. In Germany, CCUS will be an indispensable CO_2 reduction strategy in order to meet the goal of reducing Germany's CO_2 emissions to zero by 2050. Researchers have pointed out that CCUS technology must be considered to reduce CO_2 emissions from Germany's large number of irreplaceable sources of CO_2, such as those emitted from agricultural production and cement manufacturing. In 2019, the German government developed the Climate Action Plan 2030, which explicitly proposed a CCUS development plan to increase support for CCUS-related research and development, particularly offshore CO_2 storage (PIOFG, 2023). As a result, CCUS has rapidly become a hot research topic in the field of climate change in Germany in recent years.

In a whole-chain CCUS system, all CO_2 captured from emission sources needs to be handled either by CO_2 conversion and utilization, or by underground storage of CO_2. At the current stage, the amount of CO_2 that can be converted by the state-of-the-art CO_2 conversion and utilization technologies (e.g., hydrogenation of CO_2 to produce syngas, photocatalytic reduction of CO_2 to fuels, and electrochemical CO_2 reduction to fuels (Wang et al., 2021)) is limited. To achieve a significant reduction in CO_2 emissions, large amounts of CO_2 need to be captured and stored in deep subsurface formations including saline aquifers, gas and oil reservoirs, coal beds, and salt caverns, which is defined as CO_2 geological utilization and storage (CGUS). CGUS serves as a final "CO_2 sink" in the whole CCUS chain, after CO_2 capture and transportation. For point sources with large amounts of CO_2 emissions (power plants, cement plants, oil refinery plants, etc.) and with ideal geological sinks nearby, underground storage of CO_2 can be regarded as the most effective approach for deep cut of CO_2 emissions. A schematic showing the typical CGUS pathways is demonstrated in Fig. 1.

In this context, Institute of Rock and Soil Mechanics, Chinese Academy of Sciences (IRSM-CAS) in China and the University of Stuttgart in Germany jointly hosted the Sino-German symposium. The joint research symposium aimed to facilitate idea sharing and academic exchange between scholars from both countries in four key areas: modeling approaches for CGUS process analysis, experimental tools to study subsurface CO_2 flow, safety assurance after CO_2 injection, and novel CGUS technologies. Important talks delivered at the symposium are divided into the four themes and are presented in the following sections.

2. Modeling approaches for CGUS process analysis

In a talk titled "Finite Volume Neural Network: Physics-confined machine learning of flow & transport models", Prof. Wolfgang Nowak from the University of Stuttgart reported new opportunities from a series of works (Horuz et al., 2022; Karlbauer et al., 2022; Praditia et al., 2022) that enables a machine-learning assisted scientific inquiry of poorly-understood processes. In the context of CGUS process analysis, these could become a game changer in searching for appropriate constitutive relations, hysteresis models based on partial knowledge and experimental data. The underlying method is called the Finite-Volume Neural Network. In this method, the experimental domain is represented as a collection of finite volumes. For these volumes one defines state variables of interest and that they would change with incoming/outgoing fluxes. Both the required equations of state and the required flux laws are then expressed by machine-learnable functions, e.g., neural networks. These functions can then be equipped with constraints and prior knowledge if available, and finally inferred from data.

Prof. Chaozhong Qin from Chongqing University delivered a talk titled "Modelling of flow and transport in multiscale digital rocks", which discussed a pore network extraction approach from a three dimensional (3D) computed tomography (CT) scanning image. The pore network extracted can be used for CO_2-brine two-phase flow simulation in CGUS scenarios. Based on multiple convolution calculations, the proposed method increases the calculation efficiency of the traditional micro-pore region voxel grid coarsening method, ensures the accuracy of the simulation result, and decreases the number of calculation grids needed for the micropore area.

A presentation with the title "Storage of green gas in the subsurface-challenges for models and simulation" was given by Prof. Rainer Helmig from the University of Stuttgart. He spoke about the primary challenge to accurately model a subsurface hydrogen storage system-a lack of accurate transport and physical properties of hydrogen in a high pressure and high temperature subsurface system (Eller et al., 2022). To tackle this challenge, he uses an entropy scaling method together with the Perturbed-Chain Statistical Associating Fluid Theory equation of state to calculate the transport characteristics of water, hydrogen, and their mixing.

In a talk titled "The importance of hydro-mechanical coupling phenomena for (fractured) reservoir modeling", Prof. Holger Steeb from the University of Stuttgart highlighted the need to characterize subsurface fluid flow accounting for hydro-mechanical coupling between fluid-pressure variations and rock deformation. In-depth modeling considerations and numerical simulations of a compressible fluid traveling over a single compliant hydraulic conduit, such as a joint or crack, were covered in this talk.

Prof. Ke Xu from Peking University emphasized the importance of capillary pressure to immobilize CO_2 bubbles from bauyancy-driven rising in his presentation with the title "Thermodynamic stability of capillary trapping after CO_2 subsurface sequestration". Prof. Xu found that the porous structure regulates bubble evolution by modifying P_c-bubble volume correlation and decoupling mass transfer length from bubble size. Also, capillary trapping of ganglia can be stable thermodynamically, as pore-geometry reshapes ganglia morphology. When dissolution follows minimum-energy rout, inside-out dissolution may emerge that enhances trapping stability.

In a speech titled "Geochemical driven fracture alteration in GCS systems", Prof. Hang Deng from Peking University discussed the impacts of the altered porous layer on the fracture surface resulting from preferential dissolution of the

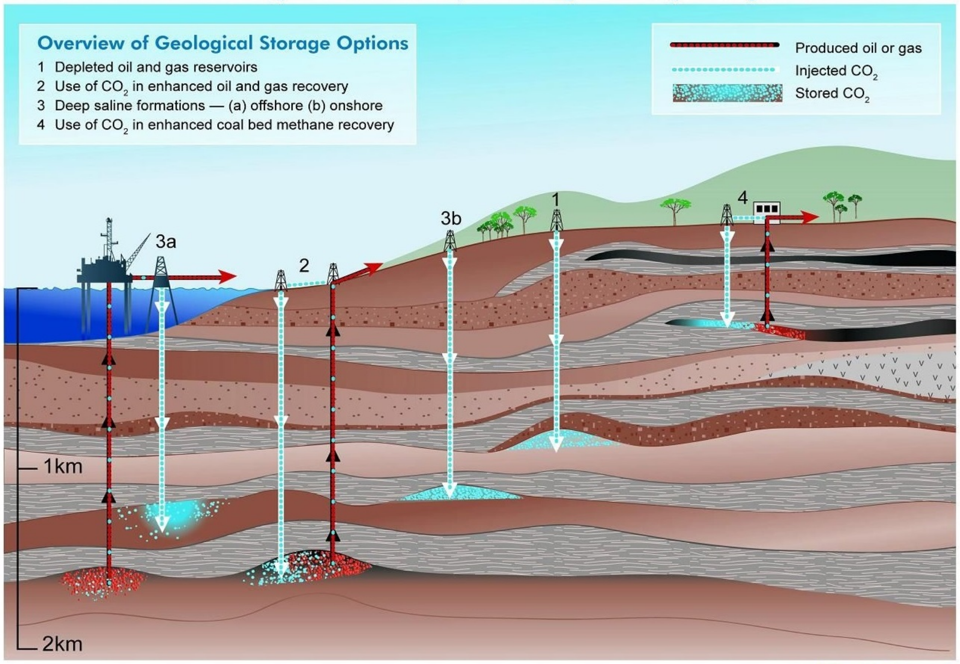

Fig. 1. Typical CGUS pathways (Metz et al., 2005).

fast-reacting minerals on subsequent chemical and physical alteration of the fractures. The modeling framework combines CrunchFlow's geochemical modeling capabilities with the Darcy-Brinkman-Stokes solver in COMSOL Multiphysics. Her modeling results support prior findings that diffusion across the altered porous layer plays an important role in the fast-reacting mineral's (calcite) dissolution (Zhang et al., 2022).

3. Experimental tools to study subsurface CO_2 flow

A talk with the title "CCUS: insights from high-pressure optical observation and in-situ Raman spectroscopic study" was given by Prof. Huirong Guo from the China University of Geosciences (Wuhan). She proposed a novel approach to use in-situ quantitative Raman spectroscopy combining with high pressure optical capillary cell to measure water solubility in CO_2 at pressures from 10 to 50 MPa and temperatures from 313.15 to 473.15 K. The unsaturated homogenized solution approach that had been previously used for measuring the CO_2 concentration in aqueous solution was modified to get the quantitative link between composition (the mole fraction of H_2O in the homogeneous phase, H_2O), and Raman peak area ratio. Most of the 45 solubility data points collected were in agreement with earlier experimental research.

Prof. Branko Bijeljic from Imperial College London used X-ray microtomography to observe CO_2 droplets trapped in the pore space as droplets surrounded by water through capillary trapping in a conversation titled "Pore-scale imaging and modelling of multiphase flow and reactive transport in CO_2 storage". Three carbonates and two sandstones were employed to image, using X-ray microtomography, the pore-scale organization of CO_2 droplets at a resolution of 6.6 μm. The size distribution of residual ganglia greater than 105 $voxel^3$ follows power law distributions, with exponents broadly consistent with percolation theory over two orders of magnitude, only with the exception of one strongly connected sample. (Andrew et al., 2014).

In a talk titled "Numerical investigation of microscopic flow mechanisms during imbibition in reservoir rock", Prof. Jianchao Cai from the China University of Petroleum-Beijing carried out numerical simulations of forced imbibition in three natural rocks at four different injection rates based on the color lattice Boltzmann model. He analyzed the interfacial evolution and in situ fluid distribution from several perspectives. His work systematically studied for imbibition of the local and global flow physics and dynamics, the mechanical nature and the resulting effects of interfacial instability under unfavorable conditions, which help to deepen our understanding of the microscopic mechanism of forced imbibition.

Prof. Zhibing Yang from Wuhan University presented his recent research in a presentation about visualization of the colloid transport and retention during immiscible two-phase flow based on confocal microscopy. The presentation has the title "Experimental and numerical investigations of multiphase flow dynamics in subsurface rocks from nanoscale to field scale". Prof. Yang discovers that the pore/grain size and flow rate have a significant impact on the colloid transport and retention behaviors. The colloids can congregate at the liquid filaments in small-grain packings at low saturation levels (high flow rate), while the wetting liquid is mostly present as pendular rings, and are uniformly dispersed in large-grain packings (Wu et al., 2023).

Prof. Liwei Zhang from IRSM-CAS presented the progress of an interaction experiment between aqueous CO_2 and cement along with X-ray computed micro-tomography characterization of pre- and post-exposure cement samples. His talk has the title “CT characterization of wellbore cement degradation under geologic CO_2 storage conditions”. To map mineral dissolution and precipitation as well as characterizing the shape of carbonate shells, an image processing framework based on CT number was proposed. By creating supplemental images of the carbonate region’s skeleton, 3D local thickness, spatial changes in the carbonate shell’s area, penetration depth, and pore/calcite/portlandite contents are quantified (Miao et al., 2020).

In a talk titled “In-situ imaging of CO_2 trapping and oil recovery in three-phase systems: dependence on pore geometry and wettability”, Ms. Yingwen Li from the China University of Petroleum (East China) studied gas-water flow patterns in various limestones via in-situ X-ray microtomography combined with numerical flow simulations. She found that the direction of fluid migration was considerably impacted by pore structural heterogeneity. While gas moves slowly through microfractures, it moves quickly through big pores and vugs. At low capillary number circumstances, however, water does not enter big pores. Instead, water flows through microfractures and microscopic pores (Li et al., 2022).

4. Safety assurance after CO_2 injection

A presentation named “The role of subsurface engineering in the net-zero energy transition” was given by Prof. Michael Celia from Princeton University. In the presentation, Prof. Celia discussed the challenges for large-scale CCUS deployment in the USA, in China and around the world, with a special focus on leakage risk assessment. In his talk, Prof. Celia presented an overview of the governing equations of CO_2 and brine migration in wellbores and faults. Then, he concentrated on some models that had been established to examine the risks of leakage connected with CO_2 injection and the potential for both CO_2 and displaced brine leaking along historic wells. These models make a number of simplifying assumptions that are suitable for the physics underlying many injection scenarios (Celia, 2019).

Prof. Qi Li from IRSM-CAS talked about microseismic events induced by fluid injection into subsurface in his talk titled “Security regulation of induced seismicity under multi-well fluid injection”. He highlighted that the microseismic events induced by fluid injection must be well managed to avoid fault re-activation. The following conclusions are reached: (1) The location of the well relative to the known fault is the main factor affecting the hydo-mechanical coupling behaviour of the fault in both single-well and multi-well injection schemes. (2) Compared with the single-well scheme, the multi-well scheme has the advantage of being more flexible. It can reduce the seismic magnitude with minimal or no extension of the construction period through a reasonable combination of different wells.

Prof. Xiaying Li from IRSM-CAS delivered a speech titled “Fracturing around dry/wet boundary in tight sandstones monitored by acoustic emission”, which discussed the micro-crack and microseismic patterns generated as a result of fluid injection. She found that the micro-crack usually initiated in the saturated region and migrated to the partially saturated and dry regions. More than 80% acoustic emissions clustered in the partially saturated and in the front of waterfront at dry regions. The heterogeneity distribution of fluid promotes the formation of complex network cracks. The dry and fully saturated rocks form a simple failure, while the partially saturated rocks form a complex microcrack network in the flooding region.

5. Novel CGUS technologies

In a talk titled “CO_2 methanation and renewable natural gas storage in depleted petroleum reservoirs-a new CCUS technology”, Prof. Zhengmeng Hou from the Clausthal University of Technology discussed the potential for renewable natural gas production by microbial-mediated CO_2 methanation, which has been identified as a key for future systems relying on renewable and zero-carbon power. Specifically focusing on the biogeochemical mechanism and possible uses for renewable energy conversion and storage, an overview of hydrogenotrophic methanation in conjunction with subsurface CO_2 utilization and renewable natural gas storage was presented. Prof. Hou found that the local methanogenic archaea can stimulate bio-methane synthesis by providing necessary functional groups and electron carriers, using injected CO_2 and H_2 as substrates and subsurface petroleum reservoir as a bioreactor.

Prof. Zhonghe Pang from Institute of Geology and Geophysics, CAS, discussed the concept and the feasibility of CO_2-enhanced aquifer thermal energy recovery (CO_2EATER) in his conversation with the name “CO_2EATER”. In a CO_2EATER operation, CO_2 is injected into the sandstone reservoir by using CO_2 as a chemical initiator, and the porosity and permeability of the reservoir are improved by partially dissolving the carbonate minerals of the reservoir, so as to improve the geothermal exploitation efficiency of the reservoir. This technology is of great significance for the sustainable development and utilization of heat storage resources in China and around the world.

Dr. Xuerui Wang from Gesellschaft für Anlagen- und Reaktorsicherheit discussed the potential for CO_2 leakage reduction through biofilm-induced calcium carbonate precipitation in her presentation titled “CO_2 leakage remediation via biomineralization-numerical approach to the coupled processes”. Based on the literature review, sporosarcina pasteurii biofilms have the desired capacity to produce $CaCO_3$ that can seal large fractures. In order to maximize the $CaCO_3$ precipitation brought on by microbial urea hydrolysis, optimized injection protocols are reviewed and discussed.

Acknowledgements

We would like to express appreciation to the financial support from the Sino-German Center for Research Promotion (No. GZ1682). We also would like to thank all the volunteer postdocs and students for their contributions to this conference: Manguang Gan, Quan Xue, Hanwen Wang, Xiaojuan Fu, Sinan Liu, Hesam Bazargan Harandi, Theogene Hakuzweyezu.

Conflict of interest

The authors declare no competing interest.

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