



Editorial

Editorial for Special Issue “Clay Mineral Transformations after Bentonite/Clayrocks and Heater/Water Interactions from Lab and Large-Scale Tests”

Ana María Fernández ^{1,*} , Stephan Kaufhold ² , Markus Olin ³, Lian-Ge Zheng ⁴, Paul Wersin ⁵ and James Wilson ⁶

¹ Centro de Investigaciones Energéticas Medioambientales y Tecnológicas, 28040 Madrid, Spain

² Bundesanstalt für Geowissenschaften und Rohstoffe, 30655 Hannover, Germany; stephan.kaufhold@bgr.de

³ VTT Technical Research Centre of Finland Ltd., 02044 Espoo, Finland; markus.olin@vtt.fi

⁴ Energy Geoscience Division, Lawrence Berkeley National Laboratory (LBNL), Berkeley, CA 94720, USA; lzhang@lbl.gov

⁵ Institute of Geological Sciences, University of Bern, CH-3012 Bern, Switzerland; paul.wersin@geo.unibe.ch

⁶ Wilson Scientific Ltd., Warrington WA3 6TR, UK; jim@wilsonscientific.co.uk

* Correspondence: anamaria.fernandez@ciemat.es

This Special Issue “Clay Mineral Transformations after Bentonite/Clayrocks and Heater/Water Interactions from Lab and Large-Scale Tests” covers a broad range of relevant and interesting topics related to deep geological disposal of nuclear fuels and radioactive waste.

Most countries that generate nuclear power have developed radioactive waste management programmes during the last 50 years to emplace long-lived and/or high-level radioactive wastes in a deep underground repository in a suitably chosen host rock formation. The aim is to remove these wastes from the human environment, considering the ethical undertaking that this geological disposal should be pursued now and not left to future generations [1,2]. Because radioactive waste retains potentially harmful levels of radioactivity for hundreds of thousands of years, safely disposing of high-level nuclear waste is not a temporary problem, but it involves a million-year solution.

The earliest discussions of solutions for the disposal of nuclear waste date from the mid-1950s and the first US National Academy of Sciences (NAS) Committee on Waste Disposal report [3], where it was proposed that the waste could be disposed of hundreds of meters underground in specially constructed mined cavities. If a site were properly chosen, a repository system comprising both natural and engineered barriers would provide a high level of protection from the toxic effects of the waste. Since then, all countries having a long-term management strategy have accepted this approach [4].

The search for technically suitable and socially acceptable repository sites for high-level radioactive waste (HLRW) and spent fuel (SF) began in the mid-1960s. Nuclear nations in Western Europe and elsewhere began to plan for geological disposal during the late 1970s and 1980s. Since the mid-1980s, France and Belgium initiated projects for analysing the disposal of HLRW in clayrocks, whereas Finland, Sweden, Canada, Switzerland and Spain did the same in crystalline granitic rocks. In 1998, the European Community recommended to its members to “continue activities on siting, construction, operation and closure of HLRW repositories”. For this purpose, most countries have interacted through the International Atomic Energy Authority, which has established safety requirements for the disposal of radioactive waste [5] and guidance on how geological disposal facilities should be developed [6].

Nowadays, the site-selection process for HLRW has been reached in only three countries: Finland, Sweden and France. The first construction license for a geological disposal facility was presented by Posiva at Olkiluoto in Finland in 2012, which was accepted in 2015.



Citation: Fernández, A.M.; Kaufhold, S.; Olin, M.; Zheng, L.-G.; Wersin, P.; Wilson, J. Editorial for Special Issue “Clay Mineral Transformations after Bentonite/Clayrocks and Heater/Water Interactions from Lab and Large-Scale Tests”. *Minerals* **2022**, *12*, 569. <https://doi.org/10.3390/min12050569>

Received: 20 April 2022

Accepted: 26 April 2022

Published: 30 April 2022

Publisher’s Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

Final disposal activities are expected to start in 2025. On 16 March 2011, SKB applied for a license to build a repository for spent nuclear fuel at the Forsmark Site (Sweden, north of Stockholm), which was finally approved in January 2022. In France, the National Radioactive Waste Management Agency (ANDRA) is currently preparing its license application for the Industrial Centre for Geological Disposal (Cigeo project, Meuse/Haute-Marne Site). In Canada and Switzerland, national waste management agencies (NWMO and NAGRA, respectively) are still investigating appropriate sites through site characterisation programmes. In other countries, such as the United States, Germany and Spain, the process appears to have stalled [2,7]. In the case of Russia, the research into the searching of geological formations and sites for building an underground facility for HLRW disposal started in 1993, later than in the other countries. In 2015, it approved the programme “Maintaining Nuclear and Radiation Safety, 2016–2030”, for the construction of a high-level and intermediate-level radioactive waste isolation facility in the Nizhnekansk Granitic Gneiss Crystalline Massif (Krasnoyarsk Territory, Central Siberia) [8].

Significant research efforts world-wide have increased our knowledge and understanding of how underground disposal systems will function over very long periods of time. Characterisation works have been performed, not only at laboratory scale but also by large scale in situ tests in underground research laboratories (URLs). These URLs, conducted for experimental and demonstration programmes, have played an important role in the assessment of the geological and the engineered barrier systems, the development of research methodologies and techniques, the development of construction techniques, the demonstration of repository operations, confidence building and international co-operation [1,7].

The first URLs were already developed in the ninety-sixties and ninety-seventies. Since then, the main URLs constructed were [9]: Asse Mine (salt formation, Germany, 1965–1997), Nevada Test Site (tuff, USA, 1979–1990), Sellafield (tuff, UK, stopped in 1997), HADES-URF (clay, Belgium, since 1980), Konrad (limestone, Fe-mine, Germany, since 1980), URC Josef (volcanic and sedimentary rock, Czech Republic, since 1981), Grimsel Test Site (granite, Switzerland, since 1983), Lac du Bonnet (granite, Canada, since 1984), Gorleben (salt dome, Germany, 1985–2020), Tono (sandstone, Japan, since 1986), Äspö (granite, Sweden, since 1990), Tournemire (clay, France, since 1990), Olkiluoto (granite, Finland, since 1993), Mont Terri (clay, Switzerland, since 1995), Yucca Mountain (tuff, 1996–2010), Meuse/Haute-Marne (clay, Bure, France, since 2000), ONKALO (granite, Okiluoto, Finland, since 2003), Mizunami URL (granite, Japan, since 2004), Horonobe (sedimentary rock, Japan, since 2005), KURT (granite, Korea, since 2006) and Beishan URL (igneous rock, Gobi Desert, China, since 2021). An underground research laboratory will be built in the Nizhnekansk Granitic Massif in 2025–2030, as the first phase in constructing a HLRW final isolation facility in Russia [8].

The 17 papers published in this Special Issue show that bentonites and clayrocks are an essential component of the multi-barrier system ensuring the long-term safety of the final disposal of nuclear waste. The efficiency of such engineered and natural clay barriers relies on their physical and chemical confinement properties, which should be preserved in the long-term. From a geochemical point of view, the clayey mineral’s function to isolate the canisters from water and retard the migration of radionuclides also means maintaining a suitable chemical and mineralogical environment for canister integrity, radionuclide retention and mechanical stability over time, buffering possible alteration/deterioration processes of the nanoporous clay materials [10,11].

The selected papers can be grouped in four main investigating issues: (a) characterisation of the barrier materials and their properties [12,13], (b) impact of transient variables and external perturbations on clay minerals’ properties: cement, temperature, organics and microbial activity [14–18], (c) long-term behaviour of bentonite and clayrock barriers from large-scale tests inside URLs [19–25] and (d) long-term predictions of clay behaviour by means of reactive-transport modelling after different perturbations linked to interactions between the clay materials and the allochthonous engineered solid materials (ground-/pore-water, heat, cement/concrete, iron corrosion, organics, etc.) [26–28]. Most

of these studies are currently being performed in the framework of various international co-operation programmes, such as the Mont Terri Project and the European Commission BEACON and EURAD (tasks ACED, HITEC, CONCORD) Projects, which are a base for improving technical and scientific knowledge in the context of nuclear waste disposal.

The Guest Editors would like to acknowledge the authors and reviewers for their foremost contributions and the *Minerals* journal for the opportunity to share this valuable knowledge.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. OECD. *Geological Disposal of Radioactive Waste. Review of Developments in the Last Decade*; OECD Nuclear Energy Agency (NEA): Paris, France, 2000; 106p.
2. Ewing, R.C.; Whittleston, R.A.; Yardley, B.W.D. Geological Disposal of Nuclear Waste: A Primer. *Elements* **2016**, *12*, 233–237. [[CrossRef](#)]
3. NAS–NRC. *Disposal of Radioactive Waste on Land*; National Academy of Sciences–National Research Council Report; National Academies Press: Washington, DC, USA, 1957; 146p.
4. Metlay, D.S. Selecting a Site for a Radioactive Waste Repository: A Historical Analysis. *Elements* **2016**, *12*, 269–274. [[CrossRef](#)]
5. IAEA (International Atomic Energy Agency). *Disposal of Radioactive Waste. Specific Safety Requirements*; International Atomic Energy Agency Safety Standards Series No. SSR-5; IAEA: Vienna, Austria, 2011; 62p.
6. IAEA (International Atomic Energy Agency). *Geological Disposal Facilities for Radioactive Waste. Specific Safety Guide*; International Atomic Energy Agency Safety Standards Series No. SSG-14; IAEA: Vienna, Austria, 2011; 104p.
7. Delay, J.; Bossart, P.; Ling, L.X.; Blechschmidt, I.; Ohlsson, M.; Vinsot, A.; Nussbaum, C.; Maes, M. Three decades of underground research laboratories: What have we learned? *Geol. Soc. Lond. Spec. Publ.* **2014**, *400*, 7–32. [[CrossRef](#)]
8. Nikitin, A. *The Underground Research Laboratory in the Deep Geological Repository in the Nizhnekansk Massif, Krasnoyarsk Territory*; A Bellona Working Paper; Bellona: Oslo, Norway, 2018; 23p.
9. OECD. *Underground Research Laboratories (URL)*; OECD Nuclear Energy Agency (NEA): Paris, France, 2013; 52p.
10. Altman, S. Geochemical research: A key building block for nuclear waste disposal safety cases. *J. Contam. Hydrol.* **2008**, *112*, 174–179. [[CrossRef](#)] [[PubMed](#)]
11. Apted, M.J.; Ahn, J. *Geological Repository Systems for Safe Disposal of Spent Nuclear Fuels and Radioactive Waste*, 2nd ed.; Woodhead Publishing Series in Energy; Elsevier: Duxford, UK, 2017.
12. Daniels, K.A.; Harrington, J.F.; Milodowski, A.E.; Kemp, S.J.; Mounteney, I.; Sellin, P. Gel Formation at the Front of Expanding Calcium Bentonites. *Minerals* **2021**, *11*, 215. [[CrossRef](#)]
13. Meleshyn, A.Y.; Zakusin, S.V.; Krupskaya, V.V. Swelling Pressure and Permeability of Compacted Bentonite from 10th Khutor Deposit (Russia). *Minerals* **2021**, *11*, 742. [[CrossRef](#)]
14. Manzel, T.; Podlech, C.; Grathoff, G.; Kaufhold, S.; Warr, L.N. In Situ Measurements of the Hydration Behavior of Compacted Milos (SD80) Bentonite by Wet-Cell X-ray Diffraction in an Opalinus Clay Pore Water and a Diluted Cap Rock Brine. *Minerals* **2021**, *11*, 1082. [[CrossRef](#)]
15. Bateman, K.; Amano, Y.; Kubota, M.; Ohuchi, Y.; Tachi, Y. Reaction and Alteration of Mudstone with Ordinary Portland Cement and Low Alkali Cement Pore Fluids. *Minerals* **2021**, *11*, 588. [[CrossRef](#)]
16. Kašpar, V.; Šachlová, Š.; Hofmanová, E.; Komárková, B.; Havlová, V.; Aparicio, C.; Černá, K.; Bartak, D.; Hlaváčková, V. Geochemical, Geotechnical, and Microbiological Changes in Mg/Ca Bentonite after Thermal Loading at 150 °C. *Minerals* **2021**, *11*, 965. [[CrossRef](#)]
17. Laufek, F.; Hanusová, I.; Svoboda, J.; Vašíček, R.; Najser, J.; Koubová, M.; Čurda, M.; Ptíčen, F.; Vaculíková, L.; Sun, H.; et al. Mineralogical, Geochemical and Geotechnical Study of BCV 2017 Bentonite—The Initial State and the State following Thermal Treatment at 200 °C. *Minerals* **2021**, *11*, 871. [[CrossRef](#)]
18. Podlech, C.; Matschiavelli, N.; Peltz, M.; Kluge, S.; Arnold, T.; Cherkouk, A.; Meleshyn, A.; Grathoff, G.; Warr, L.N. Bentonite Alteration in Batch Reactor Experiments with and without Organic Supplements: Implications for the Disposal of Radioactive Waste. *Minerals* **2021**, *11*, 932. [[CrossRef](#)]
19. Bleyen, N.; Small, J.S.; Mijndonckx, K.; Hendrix, K.; Albrecht, A.; De Cannière, P.; Surkova, M.; Wittebroodt, C.; Valcke, E. Ex and In Situ Reactivity and Sorption of Selenium in Opalinus Clay in the Presence of a Selenium Reducing Microbial Community. *Minerals* **2021**, *11*, 757. [[CrossRef](#)]
20. Emmerich, K.; Bakker, E.; Königer, F.; Rölke, C.; Popp, T.; Häußler, S.; Diedel, R.; Schuhmann, R. A MiniSandwich Experiment with Blended Ca-Bentonite and Pearson Water—Hydration, Swelling, Solute Transport and Cation Exchange. *Minerals* **2021**, *11*, 1061. [[CrossRef](#)]
21. Wersin, P.; Hadi, J.; Jenni, A.; Svensson, D.; Grenèche, J.-M.; Sellin, P.; Leupin, O.X. Interaction of Corroding Iron with Eight Bentonites in the Alternative Buffer Materials Field Experiment (ABM2). *Minerals* **2021**, *11*, 907. [[CrossRef](#)]
22. Sudheer Kumar, R.; Podlech, C.; Grathoff, G.; Warr, L.N.; Svensson, D. Thermally Induced Bentonite Alterations in the SKB ABM5 Hot Bentonite Experiment. *Minerals* **2021**, *11*, 1017. [[CrossRef](#)]
23. Kaufhold, S.; Dohrmann, R.; Ufer, K.; Svensson, D.; Sellin, P. Mineralogical Analysis of Bentonite from the ABM5 Heater Experiment at Äspö Hard Rock Laboratory, Sweden. *Minerals* **2021**, *11*, 669. [[CrossRef](#)]

24. Fernández, A.M.; Marco, J.F.; Nieto, N.; León, F.J.; Robredo, L.M.; Clavero, M.A.; Fernández, S.; Svensson, D.; Sellin, P. Characterization of Bentonites from the in situ ABM5 Heater Experiment at Äspö Hard Rock Laboratory, Sweden. *Minerals* **2022**, *11*, 471. [[CrossRef](#)]
25. Yokoyama, S.; Shimbashi, M.; Minato, D.; Watanabe, Y.; Jenni, A.; Mäder, U. Alteration of Bentonite Reacted with Cementitious Materials for 5 and 10 years in the Mont Terri Rock Laboratory (CI Experiment). *Minerals* **2021**, *11*, 251. [[CrossRef](#)]
26. Bateman, K.; Murayama, S.; Hanamachi, Y.; Wilson, J.; Seta, T.; Amano, Y.; Kubota, M.; Ohuchi, Y.; Tachi, Y. Evolution of the Reaction and Alteration of Mudstone with Ordinary Portland Cement Leachates: Sequential Flow Experiments and Reactive-Transport Modelling. *Minerals* **2021**, *11*, 1026. [[CrossRef](#)]
27. Jenni, A.; Mäder, U. Reactive Transport Simulation of Low-pH Cement Interacting with Opalinus Clay Using a Dual Porosity Electrostatic Model. *Minerals* **2021**, *11*, 664. [[CrossRef](#)]
28. Chaparro, M.C.; Finck, N.; Metz, V.; Geckeis, H. Reactive Transport Modelling of the Long-Term Interaction between Carbon Steel and MX-80 Bentonite at 25 °C. *Minerals* **2021**, *11*, 1272. [[CrossRef](#)]