EVALUATION OF COUPLED THERMAL-HYDROLOGICAL-MECHANICAL- CHEMICAL PROCESSES FOR DEEP DISPOSAL IN DIFFERENT GEOLOGIC MEDIA

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The heat pulse caused by emplacement of high-level waste may alter the hydrologic, chemical, and mechanical features in the geologic media surrounding potential repositories. This is a review paper of how thermal-hydrological-mechanical-chemical (THMC) coupled processes may affect the geologic media proposed by different countries for deep geologic disposal of high-level waste. Programs from various countries have identified potential disposal sites in salt, tuff, crystalline rocks, and argillaceous formations.

Design heat loads, and thus maximum temperatures, dictate (i) the expected presence of above- or below-boiling conditions in the near-field environment and (ii) the magnitude, spatial extent, and duration of the thermal perturbation. Changes to hydrogeological and geochemical features due to coupled processes associated with the thermal perturbation may degrade of the barrier capability of host media. For example, thermal-hydrological-chemical (THC) coupled processes may result in increased solubilities and reduction in sorption characteristics in salt. Thermal-mechanical (TM) processes in salt could (i) increase thermal stresses resulting in the formation of new pathways or enhancement of existing pathways for groundwater flow; and (ii) influence the creep rate. In crystalline rocks such as granites, TM stresses can lead to deformation and changes in fracture aperture that may increase the hydraulic conductivity of the host rock. In argillaceous formations, thermal perturbation could change in hydraulic (increase in permeability), chemical (changes in sorption and diffusion properties) and mechanical (creep rate in plastic clays) properties. Similar effects are also observed in the engineered barrier system (EBS) if bentonite or a bentonite-sand mixture is used as buffer or backfill material. Because of potential deleterious effects of the high temperatures, several countries have limited the maximum temperature to some value below boiling. This temperature limit has resulted in several features, events, and processes not being evaluated in detail given their limited spatial extent and duration.

I. INTRODUCTION

This review paper summarizes international high-level waste disposal programs in different geologic media. The focus is on the evolution of THMC coupled processes associated with the heat load imposed on the geologic media by the emplaced high-level waste. The host media are expected to contribute to the safety of disposal by playing three main roles: (i) isolating wastes from the near-surface environment and the potentially disruptive processes that occur there; (ii) maintaining a geochemical, hydrogeological, and geomechanical environment favorable to the preservation and performance of the EBS; and (iii) acting as a natural barrier restricting the access of water to the wastes and the migration of mobilized radionuclides.[1] The relative importance of the three roles varies in different host media. Significant variations are possible and are dependent on the nature of the host rock and the design of the repository. Thus for a repository in hard crystalline rock, roles (i) and (ii) are likely to be the most important, while for a repository in salt deposits or argillaceous sediments, role (iii) is expected to dominate. Experience in many countries over the last 20 to 30 years has shown that acceptable conditions can be found in such diverse rock types as granitic rocks, metamorphic rocks, plastic clays, indurated clay stones, salt domes, volcanic tuffs, and sedimentary formations. It has been common practice for many years to categorize these rather loosely into crystalline rocks, argillaceous rocks, salt, and volcanic rock.[1]

This paper summarizes work performed to enhance U.S. Nuclear Regulatory Commission (NRC) knowledge and insights on high-level waste and spent fuel disposal programs outside the United States and on international organizations and international collaborative research projects dealing with high-level waste disposal issues. As this study focused on programs other than Yucca Mountain, volcanic tuff as a disposal media is not discussed further in this paper but summaries may be found elsewhere.[1,2] Table I summarizes the host media and indigenous underground research laboratories in various international programs for high-level radioactive waste disposal in geologic repositories. The host media have different properties that are both favorable (e.g., high sorption and low dissolution in argillaceous formations) and unfavorable [e.g., very low sorption for salt (Table II)] to repository performance. Coupled processes in the EBS (buffer/backfill material) as they relate to processes in the host rock are also briefly discussed.

TABLE I. Summary of International Programs for Managing High-Level Radioactive Waste and Spent Nuclear Fuel[2]

Host Media	Country	Geologic Environments Considered or Investigated for a Repository	Indigenous Underground Research Laboratories
Salt	Germany	Salt, Opalinus clay	Underground exploration of the Gorleben site was launched in 1986 but was suspended in 2000. The moratorium imposed on Gorleben by the German government in October 2000 was lifted in March 2010 (salt)
	Spain*	Granite, clay, and salt. No geological environments are currently under consideration	None
Crystalline Rocks	Canada	Granite or sedimentary rock	Pinawa Laboratory in Manitoba (granite; in the process of being decommissioned).
	China[3]	Granitic and argillaceous rocks (clay)	Beishan area is being evaluated for a potential underground research laboratory (granitic rocks).
	Finland	Granite, gneiss, granodiorite and migmatite	Construction of ONKALO underground rock characterization facility in Eurajoki began in 2004 and is continuing. Experimental work is being conducted during construction (migmatite).
	France*	Argillite and granite	Construction of the Meuse/Haute-Marne facility near the village of Bure began in 1999 (argillite).
	Japan	Granite and sedimentary rock	Tono (granite). Two laboratories are under construction: Mizunami (granite) and Honorobe (sedimentary rock).
	Korea	Granite	Korea Underground Research Tunnel at shallow depth (granite).
	Spain*	Granite, clay, and salt. No geological environments are currently under consideration.	None
	Sweden	Granite	Construction of the Aspo laboratory in Oskarshamm began in 1990 and was completed in 1995 (granite).
	Switzerland*	Clay and granite	Mont Terri near Saint Ursanne (clay) and Grimsel in the Berne Canton (granite).
Argillaceous Formations	Belgium	Clay and shale	HADES Project initiated in 1974 in Mol (clay).
	China[3]	Granitic and argillaceous rocks (clay)	Beishan area is being evaluated for a potential underground research laboratory (granitic rocks).
	France*	Argillite and granite	Construction of the Meuse/Haute-Marne facility near the village of Bure began in 1999 (argillite).
	Spain*	Granite, clay, and salt. No geological environments are currently under consideration.	None
	Switzerland*	Clay and granite	Mont Terri near Saint Ursanne (clay) and Grimsel in the Berne Canton (granite).

*Multiple host media are being considered.

TABLE II. Properties of Potential Host Media Relevant for Disposal[4]

Property	Salt	Crystalline Rocks	Argillaceous Formations
Thermal Conductivity	🟩 High	🟨 Medium	🟥 Low
Permeability	🟩 Practically impermeable	🟨 Very low (unfractured) to permeable (fractured)	🟩 Very low to low
Deformation Behavior*	🟩 Visco-plastic (Creep)	🟥 Brittle	🟨 Plastic to brittle
Strength	🟨 Medium	🟩 High	🟨 Low to medium
Dissolution Behavior*	🟥 High	🟩 Very low	🟩 Very low
Sorption Behavior*	🟥 Very low	🟨 Medium to high	🟩 Very high

*Does not include changes because of thermal perturbation.

🟩 Favorable 🟨 Average 🟥 Unfavorable

The heat load may affect (i) flow conditions in the host rock, that in turn influence water reaching the repository and transport below the repository; (ii) the stress distribution in the host rock that determines the mechanical integrity of structures, distribution of fractures, and hydrologic properties of fractures; (iii) geochemical conditions that may alter hydrologic properties; and (iv) microbial activity that in turn, influence corrosion and radionuclide transport.[1] Understanding these couplings of processes is essential for a reliable assessment of repository performance and for the evaluation of the safety case. The next sections discuss the coupled processes in salt, crystalline rocks, argillaceous formations and the EBS.

II. HOST MEDIA—SALT

Salt is being considered for a host media because of its extremely low permeability, leading to practically no groundwater flow conditions; this implies there will be less potential for radionuclide transport (Table II). However, there is potential for radionuclide transport with fluid inclusions in salt during the thermal phase. Rock salt deposits (e.g., bedded salt, salt domes) have the advantageous feature of little or no flow of water.[1] Thus, water entry into the repository is not envisioned, and transport of radionuclides away from the repository is limited to diffusion. Site selection criteria for salt domes and bedded salt deposits include the avoidance of pockets of brine that may migrate under thermally perturbed conditions or salt zones with high potential for dissolution. In salt repositories, the backfill consisting of crushed salt is expected to creep and recrystallize, becoming a part of the surrounding salt mass and completely sealing the openings.[1] Elevated temperatures may influence the rate of salt creep in the surrounding rock, alter the geochemistry of minerals and waters or brines in the system, and lead to cracking of the seals in the salt formation.

Thermal perturbation by waste emplacement may alter the flow (or lack thereof) regime.[1] High thermal conductivity of salt deposits results in relatively lower temperatures compared to other host media. Small quantities of brine occurring in disconnected fluid inclusions trapped within salt crystals may migrate along temperature gradients and could influence waste package corrosion behavior. Temperature gradients could result in thermally induced brine flow or two-phase flow. For example, temperature increases around waste located at the edges of the disposal chambers could cause evaporation of water entering from the disturbed rock zone (DRZ). This water vapor could condense on cooler waste containers in the rooms and could contribute to brine formation, corrosion, and gas generation.

Thermal perturbations can change in the hydrochemical environment in the salt deposit because (i) bound water (water of crystallization) or (ii) water present as intergranular films is released.[1] The potential effects of elevated temperatures on chemical reactions in the seals and surrounding regions include increased actinide solubilities and alteration of mineral assemblages leading to altered sorption characteristics. The effects of high temperatures on salt deposits have not been evaluated at any of the proposed sites in Table I because of restrictions on the allowable heat load.

Thermally induced stress could result in pathways for groundwater flow in the DRZ or through seals, or it could enhance existing pathways.[5] Conversely, elevated temperatures could accelerate the rate of salt creep and mitigate fracture development. Thermal expansion could also result in uplift of the rock and ground surface overlying the repository, and thermal buoyancy forces could lift the waste upward in the salt rocks. The distributions of thermal stress and strain changes depend on the induced temperature field and the differing thermal expansion of repository components based on the components' elastic properties.

III. HOST MEDIA—Crystalline Rocks

Crystalline rocks such as granite and gneiss are being considered for their hydrogeological (low permeability in unfractured rocks), mechanical (high strength), and geochemical characteristics (high sorption and low dissolution) that are beneficial to repository performance (Table II). Depending on the degree of connectivity of the fractures in the rock, the dominant radionuclide transport mechanism is advection with some potential diffusive transport. The flow regime in crystalline rocks is governed by the extent of fracture connectivity and presence of structural features such as faults. The temperature in the far field will control the rates of chemical and microbiological processes, and can influence the stress field, groundwater flow, diffusion rates and, hence, radionuclide transport. However, few geochemical and microbiological processes relevant to repository safety (other than sorption) are expected to occur in the host rock (far-field environment).

The dominant heat transfer mechanism is conduction. The thermal perturbation leads to the creation of thermo-convective cells in fractures and faults.[6] The potential of formation of thermo-convective cells is restricted by fracture network geometry, particularly continuity and connectivity. Flow continuity in a fault network is dependent on the fault dimension, orientation, and connectivity with other faults. Most repository design approaches, however, avoid major faults within the repository footprint. The temperature of the far field is largely controlled by the natural geothermal gradient, although it may be influenced by heat output from the repository and by changing climate at the surface. The heat generated in the repository will be dissipated by conduction through the rock mass and, to a much lesser extent, by advection in the moving groundwater. Consequently, the thermal impact of the repository will reduce with distance from the near field.[1,5]

The TM effects, because of emplaced waste, include differential mechanical stresses, changes to fracture apertures , and the subsequent modification of hydraulic behavior.[5] In fractured formations, mechanical deformation of the granite resulting from these stresses is absorbed by movements along the fault planes. Due to the thermal gradient arising from the repository, rock displacement could occur because of TM coupling, causing opening and closing of fractures. This could affect the groundwater flow field through changes in the permeability of the rock, causing temporary over- or underpressures. The importance of THM effects is related to the size and characteristics of the fracture pattern and the magnitude of the heat release.

IV. HOST MEDIA—Argillaceous Formations

Argillaceous clay-rich formations such as shale and mudstone have been considered as potential repository host rock because of their hydrologic (low permeability), chemical (low diffusion coefficient, high retention capacity for radionuclides), and mechanical properties (capability to self-seal fractures induced by tunnel excavation) (Table II). These formations exhibit a wide range of types: from plastic clay deposits, with transitional types, to strongly consolidated and partially fractured clay deposits. The range of types can be associated with considerable differences in deformation behavior, temperature sensitivity, and rock strength. In the absence of open fractures and stagnant pore water, diffusion is the dominant transport mechanism for radionuclides in argillaceous formations. The heat pulse causes complex mechanical, hydraulic, and chemical interactions in the host rock, and all phases (gas, liquid, solid) need to be considered.[7]

In general, temperatures above boiling, associated with high temperature gradients, increase the chemical reactivity of argillites (mineralogical transformations) and cause complex hydraulic and mechanical phenomena.[8] Thermal convection effects in clay are generally not expected because of its relatively low permeability. However, the formation of convection cells is also dependent on the repository geometry and the induced temperature gradients in the clay formation.[5,7]

The thermal perturbation leads to thermal expansion of the clay and the pore water.[7] The possible consequences are (i) pore pressure increase, (ii) change in effective stress, (iii) strength reduction of the material, (iv) lowering of threshold value for the onset of creep, and (iv) increase in creep rate. The thermally induced consolidation leads to additional flow of water and decrease in porosity and permeability of the medium. In saturated low permeability media such as clays, the displacement of water is strongly hindered and thus a pore water pressure buildup will develop as the temperature increases. The development of the pore water pressure and its dissipation depends on the ratio between the thermal and the hydraulic diffusivity. The magnitude of the increments in pore water pressure is controlled by the rate of temperature increase, the rock permeability, the rock stiffness, the boundary conditions, and construction history (excavation, ventilation, emplacement, backfill). The heating may result in a larger increase of pore water pressure than of total stress and thus in a decrease of effective stress, which modifies the strength properties of highly plastic clay deposits and leads to an increase of the plasticized zone. In stiff clay deposits, the effect of pore pressure increase will be compensated by the increase in total stress because of its thermo-mechanical properties (stiffness and coefficient of thermal expansion). The thermal perturbation can have the potential to reopen existing faults and fractures in the host clay layer. In the case where the induced stress is more considerable, it can lead to plastic deformation or even to fractures in the involved materials and in the clay formation. These phenomena are not reversible, and they might influence the performance of the repository.

The dominant transport mechanism in argillaceous formation is diffusion.[1,8] The diffusion process in response to the thermal gradients (Soret Effect) could be important if the formation is subjected to thermal load that causes significant thermal gradients.[5] Various geochemical parameters, such as density, viscosity, and diffusivity, are temperature dependent and could impact the radionuclide transport in the formation. The sorption characteristics for some species also depend on temperature and will need to be evaluated at high temperatures. The thermal perturbation can lead to (i) illitisation of the illite/smectite mixed-layer phase, (ii) acceleration of oxidation reactions, (iii) thermal maturation of organic matter, and (iv) changes in pore-water chemistry. Most chemical properties, such as solubilities, sorption coefficients, and equilibrium constants, are temperature dependent. Most of these changes are expected to be reversible for low heat loads and will not influence the behavior of the repository significantly.

Liu, et al.[9] conducted a detailed study related to coupled processes in clay formations and identified critical knowledge gaps that include topics related to

(i) representation of THM processes in evaluation of mechanical characteristics (e.g., strength and strain localization), (ii) creep in plastic clays and subcritical crack growth in indurated clays, and (iii) impact of THC processes on sorption.

V. Engineered Barrier System

EBS represents the man-made, engineered materials placed within a repository, including the waste form, waste canisters, buffer materials, backfill, and seals of access tunnels and drifts.[10] EBS plays a significant role in the containment and long-term retardation of radionuclide release. Bentonite or a bentonite-sand mixture is proposed as a buffer and backfill material in a number of countries.[1,2] The hydrological (low hydraulic conductivity), geochemical (very low diffusion, high sorption, good colloid and microbial filtration capability), and mechanical (its high swelling potential enables self-sealing of openings) properties of bentonite make it a good buffer/backfill material between the waste packages and host formation.

The heat released from radioactive waste begins to heat up the near field of the buffer-host rock system.[11] The initial pore water in the buffer will be redistributed during the early thermal phases. At the buffer-host rock interface, water is absorbed from the surrounding host rock, with potential swelling of the bentonite in this region. At the buffer-waste package interface, moisture content decreases (desiccation) with potential shrinkage. Over time, the expansion of the buffer as it is being hydrated may displace the position of the waste canister enveloped within it, as well as the position of the buffer-backfill interface. The temperature gradient and degree of saturation in the buffer determine the temperature and moisture distribution during the resaturation process. Chemically within the buffer, dissolution and precipitation of buffer minerals may occur.

The evolving temperature in the near field will also affect the groundwater chemistry because elemental solubilities are temperature sensitive. A reduction in solubility may lead to precipitation of mineral phases in pore spaces in the buffer, backfill, or near-field rock, which may in turn cause cementation and changes in hydraulic conductivities. The possibility of this process is highest during the high temperature period because the solubility of calcite decreases with increasing temperature.

If the heat load is sufficiently high to cause above-boiling temperatures, steam generation during the buffer resaturation period can induce a significant volume reduction of the clay, which can occur even at relatively short times of contact with steam at 110 °C (230 °F).[5] Hydrothermal field tests showed considerable cementation of the clay, causing brittleness and loss of expansion within a few centimeters of the steel surface of the heat source.[12] In the hottest region, anhydrite and hexa-hydrate were precipitated, probably attributable to their retrograde solubility (i.e., these phases are less soluble at higher temperatures). Slightly farther away from this hottest zone, new amorphous silica-aluminum phases formed in this slightly cooler region. The mobilization and deposition of these phases may be attributable to their solubility in a thermal gradient or to cyclic evaporation and condensation that take place in resaturating clay under a thermal gradient. Gas was also found to have filled interior voids of the backfill, probably water vapor dominantly, although some hydrogen gas may have evolved from corrosion of the steel heater in the field test. Thus, complex THMC processes that could potentially lead to degradation of the EBS support the design temperature limit to be maintained below boiling at all times.

Steefel et al.[11] conducted a detailed study and identified critical knowledge gaps related to coupled processes in the EBS that include (i) THM processes during bentonite hydration; (ii) THM behavior of bentonite pellets and pellet mixtures, and irreversible swelling or compression of bentonite in large gap fillings; (iii) impact of swelling and self-healing processes in clay buffer materials and clay host rock on reactive transport process; and (iv) effect of temperature on radionuclide sorption in bentonite.

VI. CONCLUSIONS

Thermal perturbation results in an increased influence of coupled THMC processes on repository performance, with the importance of different coupled processes dependent on the hydrogeological, chemical, and mechanical properties of the host rock. The significance and duration of coupled processes are dependent on the heat load and thermal characteristics of the EBS and the host media. The heat load due to emplaced waste is a design parameter that can be controlled by developing an appropriate thermal strategy. An important factor in the effect of coupled processes is the maximum temperature expected in the host rock. The boiling of water in the host media adds another level of complexity. Because of this added complexity, several countries have limited the maximum temperature to some value below boiling[1,6,8] [e.g., less than 90 °C (194 °F)]. Because of this limit,

- Several features, events, and coupled processes (e.g., Soret Effect) were not evaluated further in various international programs, because of low probability of occurrence or low consequence[5] given their limited spatial extent and duration

- Field heater tests also did not exceed above-boiling conditions in the host media. Several field and laboratory heater tests involved the EBS components (buffer and backfill) being subjected to above-boiling temperatures. However, the temperature in the host media was always below boiling. This is consistent for the temperature limit prescribed by the repository design. However, this has also resulted in a lack of technical information in the waste disposal scientific literature on coupled processes for temperature conditions above boiling.

The maximum temperature limit of below boiling is appropriate for countries that have relatively small amounts of waste, especially if reprocessing is to be considered. For countries that are still in the planning stage for disposal of relatively large amounts of high-level waste, temperature limits imply that longer temporary storage times must be taken into consideration and may require repository designs with larger areal extension.

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