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Can Shale Safely Host U.S. Nuclear Waste?

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Even as cleanup efforts after Japan's Fukushima disaster offer a stark reminder of the spent nuclear fuel (SNF) stored at nuclear plants worldwide, the decision in 2009 to scrap Yucca Mountain as a permanent disposal site has dimmed hope for a repository for SNF and other high-level nuclear waste (HLW) in the United States anytime soon. About 70,000 metric tons of SNF are now in pool or dry cask storage at 75 sites across the United States [*Government Accountability Office,* 2012], and uncertainty about its fate is hobbling future development of nuclear power, increasing costs for utilities, and creating a liability for American taxpayers [*Blue Ribbon Commission on America's Nuclear Future,* 2012].

However, abandoning Yucca Mountain could also result in broadening geologic options for hosting America's nuclear waste. Shales and other argillaceous formations (mudrocks, clays, and similar clay-rich media) have been absent from the U.S. repository program. In contrast, France, Switzerland, and Belgium are now planning repositories in argillaceous formations after extensive research in underground laboratories on the safety and feasibility of such an approach [*Blue Ribbon Commission on America's Nuclear Future,* 2012; *Nationale Genossenschaft für die Lagerung radioaktiver Abfälle* (*NAGRA*), 2010; *Organisme national des déchets radioactifs et des matières fissiles enrichies,* 2011]. Other nations, notably Japan, Canada, and the United Kingdom, are studying argillaceous formations or may consider them in their siting programs [*Japan Atomic Energy Agency,* 2012; *Nuclear Waste Management Organization* (*NWMO*), (2011a); *Powell et al.,* 2010].

Why argillaceous formations? First and foremost, they have low permeability. Flowing groundwater transports dissolved material and is thought to be the most likely means by which nuclear waste in an underground repository could escape and contaminate the biosphere, posing a risk for future generations. Clay-rich geologic media are millions to tens of billions of times less permeable than productive aquifers and hydrocarbon reservoirs [e.g., *Ingebritsen et al.,* 2006].

Their exceptionally low permeability suggests that argillaceous formations could be effective geologic barriers to the migration of SNF and HLW constituents from a repository. What's more, argillaceous media are the most common and voluminous of sedimentary materials and often are found in relatively old and stable geologic settings. Thus, tectonically quiet sedimentary terrains with thick clay-rich sequences may have a number of desirable qualities for hosting America's substantial and growing inventory of nuclear waste.

A Different Approach to Permeability in Argillaceous Formations

While other geologic materials, including crystalline rocks like granite, often display extremely small permeabilities in tests, they are almost invariably crisscrossed by water-conducting fractures and faults [*Committee on Fracture Characterization and Fluid Flow, National Research Council,* 1996]. Moreover, determining the presence or absence of these features in repository-sized volumes of rock is a surprisingly difficult problem. They are most reliably detected with boreholes, but this is a catch-22 when the rock must not be compromised. An interesting alternate approach now appears possible in argillaceous formations and is providing evidence that many are remarkably free of water-conducting features.

Investigations of easily accessible (less than ~1 kilometer deep) argillaceous formations have yielded the surprising finding that about half have patterns of groundwater pressure that are "anomalous," referring to energy gradients driving net flow in or out of the formation without apparent cause (see Figure 1). Nowhere is this seen more dramatically than at Wellenberg, Switzerland, where pressures in an argillaceous marl are as much as 8 megapascals lower than expected [*NAGRA,* 1997]. A similar pattern has been discovered in Ontario, Canada ("Bruce" in Figure 1), where hydraulic heads 200 meters below sea level have been reliably measured in shale [*NWMO,* 2011b]. Anomalously high pressures have also been found, including those in the east Paris Basin ("Bure" in Figure 1), where heads in an argillite are higher than in the surroundings [*Delay et al.,* 2007].

Such pressures seem most readily explained as responses to forcing from geologic processes, usually manifested as strain, with the magnitude of the anomaly reflecting the balance between how strong or how recent the forcing is and the rate at which pressures normalize by flow. This can be described using a dimensionless ratio involving the rate of forcing, formation thickness, and permeability [*Ingebritsen et al.,* 2006]. Bounds can be placed on forcing rates, such as tectonic deformation, glacial ice load changes, or erosional removal of overlying material [e.g., *Neuzil,* 1995], and thereby also on permeability.

Seen in this light, pressure anomalies—both negative and positive—are essentially glimpses of large-scale permeability experiments that have run for tens of millennia. Their presence signals formation-scale permeabilities small enough to rule out water-conducting fractures. At Wellenberg, low pressures seem to have resulted when the weight of a continental ice sheet squeezed a small amount of groundwater from the rock; after ice retreat, the rock elastically dilated faster than groundwater could flow back in [*Vinard et al.,* 2001].

Argillaceous formations with anomalously low pressures are particularly interesting repository candidates because groundwater is apparently being drawn into the formation. This would enhance the isolation of waste emplaced in the formation because the inward flow, which will persist long into the future, would oppose escape of the waste to surroundings. Whether inward or outward, however, groundwater fluxes associated with the pressure anomalies are extremely small, typically resulting in only centimeters of movement per thousand years even under the largest driving forces. As a result, almost no solute transport results from groundwater flow; the little that occurs is dominantly by molecular diffusion, a process driven only by concentration differences. Clay-rich formations are advantageous even in this respect because they are ultrafilters [*Ingebritsen et al.,*

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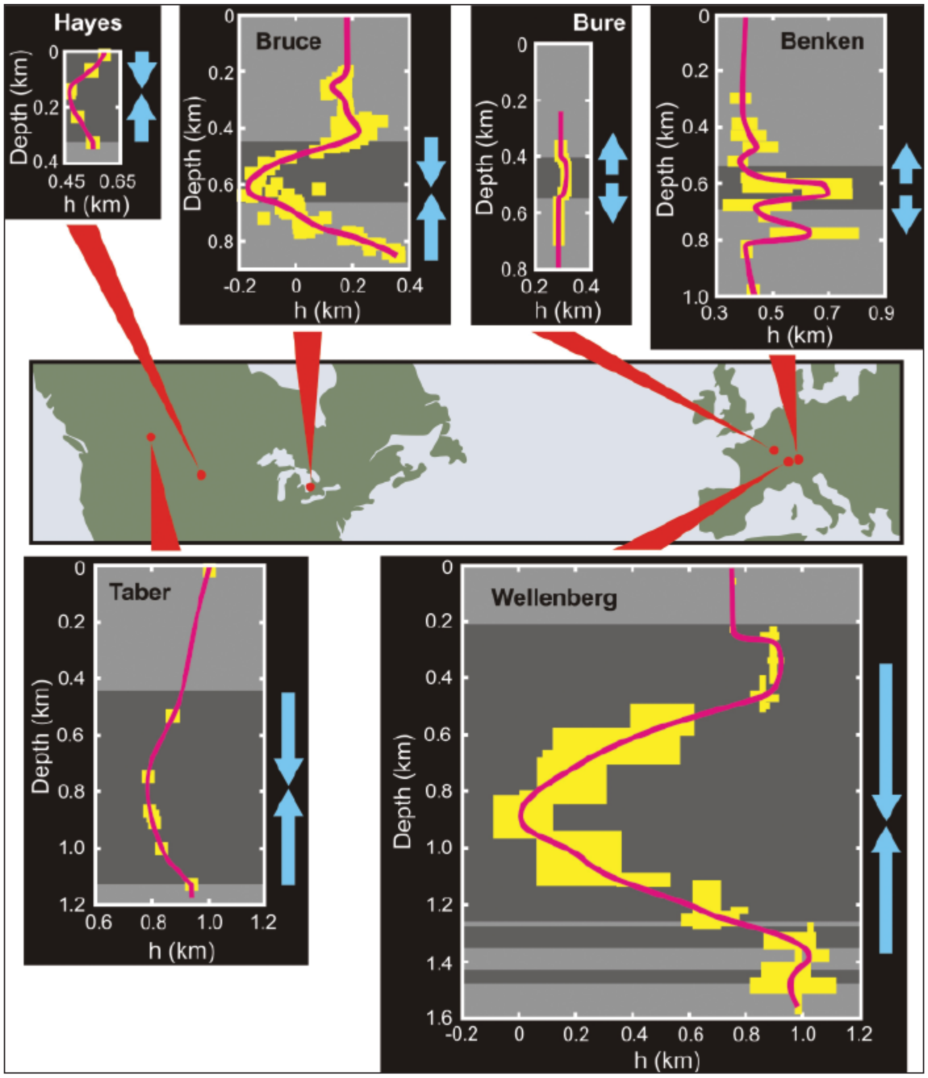

*Fig. 1. Anomalously pressured argillaceous formations exhibit great variety in scale and setting. Plots show hydraulic head (h) in kilometers relative to sea level versus depth in kilometers, with argillaceous units in dark gray. The vertical extent of each head measurement is indicated by yellow rectangle height, and reported uncertainty in the head measured at that level or the range in multiple measurements is indicated by the width. Subjective trend lines through the data are red, and blue arrows show groundwater flow. Sites are Pierre Shale in the Rocky Mountain foreland near Hayes, S. D. [*Neuzil, *1993]; Queenston, Georgian Bay, and Blue Mountain Formations on the east flank of the Michigan Basin at the Bruce Nuclear Complex in Ontario, Canada [*NWMO, *2011b]; Callovo-Oxfordian Argillite in the east Paris Basin near Bure, France [*Delay et al., *2007]; Opalinus Clay in the Molasse Basin near Benken, Switzerland [*NAGRA, *2002]; Colorado Group and Upper Mannville shales in the Taber area of the Alberta Foreland Basin in Alberta, Canada [*Corbet and Bethke, *1992]; and Palfris Formation and Vitznau Marl in the Swiss Alps at Wellenberg, Switzerland [*NAGRA, *1997].*

2006], with diffusion coefficients as much as 3 orders of magnitude smaller than in other geologic media [*Mazurek et al.*, 2003]. Waste migration out of clay-rich formations would be further slowed by the high sorption capacity of clays [*Hansen et al.*, 2010].

Like pressure anomalies, spatial distributions of groundwater constituents in argillaceous formations frequently can be viewed as experiments, in this case tracer tests, on a grand scale. In Europe, researchers found analyzable patterns in the concentrations of chloride ions, deuterium, and oxygen-18 in eight of nine argillaceous formations studied. By simulating inferred initial concentrations subjected to long periods of diffusive transport, they were able to replicate these concentration profiles, but only without noticeable transport by flowing groundwater [*Mazurek et al.*, 2009]. As such, the tracer profiles place low upper limits on groundwater flow rates and permeability in the formations.

Laboratory and field experiments show that patterns of groundwater constituents and pressures evolve at different rates in argillaceous media. Because of this, pressure anomalies and natural tracer profiles can be used as complementary indicators of formation permeability and flow history. Formation-specific data indicate that most tracer profiles have taken on the order of a million to tens of millions of years to evolve [*Mazurek et al.*, 2009], whereas groundwater flow simulations based on formation properties show that pressure anomalies are responding to influences in the last several thousand to a million or so years [*Neuzil*, 1995]. Together, pressure and geochemical patterns are a powerful tool for understanding transport and detecting permeability changes in argillaceous formations over thousands to millions of years, a time span encompassing regulatory mandates for confinement of SNF.

Practical Considerations for Shale as a Repository Host

Potentially usable argillaceous formations in the United States—those without extractable energy resources or other prohibitive circumstances—are distributed widely and occur in varied geologic and hydrologic settings [*Gonzales and Johnson*, 1985]. Indeed, the United States is in an enviable position with respect to the scale and sheer diversity in age, history, composition, and thickness of argillaceous formations within its borders. Geologically and geographically, potential choices for a repository are many.

Ironically, qualities that may make argillaceous formations desirable repository hosts also make characterizing them difficult. Procedures that are usually routine, such as permeability tests and pore fluid sampling, are technically challenging and time-consuming in these media. For example, researchers are learning how to make borehole test installations “tighter” than the formation so that the formation's permeability is measured rather than any leaks in the equipment. This is a significant feat with permeabilities as small as 10^{-22} meters squared [e.g., *NWMO*, 2011b], which is hundreds to tens of thousands of times smaller than the permeability of cement grouts. To characterize pore water geochemistry, analysts are refining methodologies that include leaching, vacuum distillation, core squeezing, and diffusive exchange, in addition to long-term borehole fluid collection.

Beyond such technological issues, research in the last 2 to 3 decades has raised interesting new questions and highlighted some old ones. Water-clay systems are complex, their physics and chemistry are incompletely understood [e.g., *Leroy and Revil*, 2004], and descriptions of water and solute movement in them are often extrapolated from experience in much more permeable materials. Applicability of Darcy's law, the constitutive relation for porous medium flow, is unsupported by direct observations in very low permeability rocks because flow rates under natural conditions have proven too small to measure. Moreover, Darcy's law describes but one of several couplings between driving forces and fluxes in clay media [e.g., *Ingebritsen et al.*, 2006], most of which have been little studied.

Research on these questions will help scientists better understand pore fluid

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transport in argillaceous formations and its role in subsurface hydrologic and geologic processes. Regardless, a strong case can be made for the ability of argillaceous formations to provide long-term confinement. Possible non-Darcian behaviors, for example, would make fluxes smaller still, and alternative interpretations of some pressure anomalies, including the presence of a gas phase [*Normani and Sykes,* 2012] and osmosis [*Delay et al.,* 2007], all imply extremely slow transport.

Perhaps the most pressing questions surrounding argillaceous formations as repository hosts are related to effects of repository excavation, sealing access openings, and thermal loading by the waste. Stress changes can increase permeability near excavations by opening small fractures in the formation, but there is evidence that both mechanical and chemical "self-sealing" often reverses such increases in clay-rich media [e.g., *Bock et al.,* 2010], a phenomenon that may also help efforts to seal access openings. For thermal loading concerns, preliminary modeling suggests that temperatures can be limited to acceptable levels by reasonable management of the form of the waste and the method of emplacement [*Hansen et al.,* 2010].

Shale and the Future of SNF in the United States

Ensuring geologic isolation of nuclear waste over tens of millennia or longer poses questions that Earth scientists continue to study. Since the decision to focus U.S. repository research on Yucca Mountain more than 25 years ago, parallel research efforts have dramatically increased understanding of the isolation afforded by shales and similar media. A result of that research is that argillaceous formations may be used to host much of the world's SNF and other HLW.

With the apparent end of plans to use Yucca Mountain, U.S. nuclear waste repository planning is at a crossroads. While interim storage of SNF at one or more secure locations is an option, permanent underground disposal will likely be necessary. Although U.S. repository research has been directed at crystalline rock, bedded salt, and the tuffs at Yucca Mountain [*Blue Ribbon Commission on America's Nuclear Future,* 2012], current research offers argillaceous formations as an emerging option—their abundance and geographic diversity suggest that they could lend significant flexibility to siting efforts.

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