Measures of Geologic Isolation

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ABSTRACT

Isolation in a geologic setting has been the generally favored solution to the high-level radioactive waste (HLW) problem since a scientific basis for nuclear waste management began to be formulated over half a century ago. Although general features of suitable settings have been enumerated, quantitative measures of the safety of geologic isolation of HLW are challenging to devise and to implement. Some regulatory measures of isolation for the proposed repository at Yucca Mountain, Nevada, have be devised and revised involving considerations of global releases, groundwater travel time, and time and space scales for isolation. In current Yucca Mountain specific regulations, the measure of long-term safety hinges on probabilistic estimates of radiation doses to the average member of a maximally exposed group of people living about 18 km down the groundwater flow gradient within 10,000 years after permanent closure of the repository. From another perspective, hydrogeochemical studies provide quantitative measures of system openness and the ability of geologic systems to isolate HLW. Hydrogeochemical data that bear on geologic isolation of HLW at Yucca Mountain include precipitation of radionuclides in stable mineralogical products of spent fuel alteration, ages of natural secondary mineralization in the mountain, uranium decay-series isotopic data for system openness, bomb-pulse isotope occurrences, and ambient carbon-14 distributions.

INTRODUCTION

In 2002, based on a recommendation from the U.S. Department of Energy (DOE), the President of the United States notified Congress that it considers Yucca Mountain, Nevada, qualified for submission of an application for construction of the nation's permanent geologic repository for disposal of high-level radioactive waste (HLW). The U.S. Congress ratified that recommendation over the objection of the Governor of Nevada later in 2002. The country and the world now await a license application from the DOE and its evaluation by the U.S. Nuclear Regulatory Commission (NRC) according to standards set by the Environmental Protection Agency (EPA). The necessity of a permanent solution to the HLW problem in the U.S. is made evident by continuing accumulation of wastes in numerous aging facilities, which are reaching temporary storage capacity and longevity, and by the hazardous lifespan of the wastes, which extends for a period of time longer than the present duration of human civilization. Although fanciful alternatives have been described and debated, isolation of HLW in a geologic setting has been the generally favored solution since a scientific basis for this problem began to be formulated over half a century ago.

Scientific and social concerns regarding the safety of geologic disposal of HLW (and concomitant implications for controversial nuclear technologies in general) motivate open and formalized evaluations of the measures of geologic isolation of HLW. When the Atomic

Energy Commission proclaimed in 1970 their intent to build a permanent geologic waste repository in a salt mine near Lyons, Kansas, the resulting technical and social debates and rejection of the site provided motivation to create the NRC as a federal agency with independent responsibility to assure public radiation safety. Within years after the Lyons site was abandoned, the Nuclear Waste Policy Act of 1982 set a formal and public framework for managing permanent geologic disposal of HLW in the U.S.

Geologic disposal of HLW has incomparable and compelling features of permanence and simplicity. Geologic stability, long-term isolation of wastes, and practicality are reasonable expectations of a site for permanent disposal of HLW within the context of well understood geologic systems and technical expertise. Natural analog studies reveal geologic settings and conditions that are conducive to waste isolation (and others that are adverse to isolation). Characterization of the chemically closed or open state of geologic and mineralogic systems has received intense scientific scrutiny in the context of radiometric dating. Stable geologic systems are well recognized that are vastly older than demands of radioactive waste isolation. Potential sites for disposal in the Earth are accessible by conventional mining technologies. Principles of permanent geologic disposal of HLW founded on these concepts have been expounded by eminent organizations such as the U.S. National Academy of Sciences [1] and the International Atomic Energy Agency [2].

Within this compelling context, quantitative measures of the safety of geologic isolation of HLW are challenging to devise and to implement. From another perspective, hydrogeochemical studies provide quantitative measures of system openness and the ability of geologic systems to isolate HLW. This article addresses these challenges and studies in a discussion of general measures of geologic isolation of HLW and of specific considerations for the proposed repository at Yucca Mountain.

GENERAL MEASURES OF ISOLATION

More than philosophical reasoning for geologic isolation is required to evaluate public and environmental radiation safety. General criteria and specific measures of geologic isolation must be devised. General characteristics of favorable geologic sites have been formally enumerated and suggest geological characteristics that could be used to measure geologic isolation. For example, in the French "fundamental rule of safety" two *essential* criteria are: stability of the site with respect to eventual modifications due to geologic conditions (glaciation, seismicity, neotectonism); and hydrogeology characterized by very low permeability and low hydraulic gradient. *Important* criteria according to the French law include: stable mechanical and thermal properties; geochemical properties favoring radioelement retention; deep location to avoid effects of erosion, seismicity, and human intrusion; and absence of natural resources. The International Atomic Energy Agency recently characterized suitable environments for deep disposal to have properties such as: long-term geologic stability; low groundwater content and flow; stable geochemical or hydrochemical conditions characterized by reducing conditions and geochemical equilibrium; and good engineering properties for construction and operation [2].

Favorable and potentially adverse conditions are specified among siting requirements by NRC in their regulation, 10CFR60, which formerly applied to Yucca Mountain, and currently applies to geologic disposal of HLW except at Yucca Mountain. In this approach to identify site characteristics that provide measures of the potential for geologic isolation, an appropriate combination of favorable conditions is required to assure meeting performance objectives, and an adequate dismissal, remedy, or compensation is required for potentially adverse conditions. Favorable conditions listed in 10CFR60 include: low permeability of host rocks in the saturated zone or of overlying rocks for disposal in the unsaturated zone; long groundwater travel times; and geochemical conditions that promote isolation of radionuclides. Potentially adverse conditions include: aspects of geologic instability (such as flooding, volcanism, structural deformation, and seismicity); and geochemical conditions that are not reducing or that could increase reactivity of engineered barrier systems. In 10CFR960, which applied to geologic disposal of nuclear waste at Yucca Mountain prior to the current 10CFR963, DOE identified similar favorable and potentially adverse conditions on the characteristics, processes, and events that may influence the performance of a repository after closure. In addition several disqualifying conditions for HLW sites were given in 10CFR960, including: groundwater travel time less than 1000 years; insufficient depth below the ground surface; and active dissolution or faulting that is expected to result in loss of waste isolation. General criteria for selecting a site for HLW disposal tend to be based on common sense, including requirements for multiple barriers for isolating wastes. However, applications of safety criteria to specific sites require challenging judgments and detailed understanding of the complexities of the individual settings and systems.

SOME REGULATORY MEASURES OF ISOLATION FOR YUCCA MOUNTAIN

Global Releases

The 1985 EPA standard for HLW disposal (40CFR191) specified a limit on releases of radioactive species for 10,000 years, and set individual protection requirements, which also pertained to a period of 10,000 years as the standard was amended in 1993. Attention to unique characteristics of the potential site at Yucca Mountain raised concerns about the applicability of these requirements. In particular, waste emplacement at Yucca Mountain in the water-unsaturated zone above the groundwater table could permit gaseous radionuclides, notably carbon-14 in the form of $^{14}CO_2$, to be released rapidly from a failed container and transported to the global atmosphere. The potential health risks of such releases are recognized realistically to be small. However, compliance with EPA measures of cumulative releases to the accessible environment was in doubt because they were based on an integration of doses over the world population. Explicitly recognizing this challenge, the U.S. Congress passed the Energy Policy Act of 1992 requiring EPA to adopt a dose limit for individual members of the public rather than a radiation release limit from the repository. The mandated EPA standard for Yucca Mountain (40CFR197), which was issued in 2001, accordingly prescribes a dose standard for the reasonably maximally exposed individual.

Groundwater Travel Time

A general compliance criterion for geologic isolation places a limit on the groundwater travel time (GWTT), as measured by likely migration of radioactive species from the waste emplacement zone to the accessible environment. NRC specifies this criterion in 10CFR60 for geologic repositories in general. However, with implementation of the NRC site specific regulation (10CFR63) in 2001, the GWTT rule no longer applies to the proposed repository at

Yucca Mountain. Site characterization at Yucca Mountain has led over the past twenty years to dramatic changes in scientific views of infiltration and groundwater flow, particularly in thick sequences of unsaturated, fractured rock. For example, occurrences at depth in Yucca Mountain of radionuclides associated with the twentieth century bomb pulse attest to rapid groundwater flow paths (see below).

Groundwater Contamination

EPA sets groundwater protection standards for drinking water supplies in the U.S., and sets complementary standards in 40CFR197 for groundwater contamination by radionuclides due to disposal of HLW at Yucca Mountain. The challenge of estimating concentrations in groundwater at long times in the future led EPA to specify the quantity of water that would dilute released radionuclides based on projected uses of groundwater by a community in the vicinity of Yucca Mountain.

Time, in General

Engineered systems can be designed reasonably to contain short half-life radionuclides. Geologic isolation is the solution for actinides and decay and fission products with long half-lives such as plutonium-239 (24,000 years), technecium-99 (200,000 years), neptunium-237 (2 million years), and iodine-129 (16 million years). Geologic isolation requirements in the U.S. generally specify a period of 10,000 years. EPA justified this time limit in its original HLW standard in 1985 on the basis that major geologic changes are unlikely in 10,000 years, so predictions are less compromised by geologic uncertainty. EPA also noted that levels of radioactivity will diminish to values comparable to natural geochemical occurrences in a time period of about 10,000 years. In comparison, NRC argued in support of its recent Yucca Mountain regulation (10CFR63) that 10,000 years is long enough to require consideration of changes in conditions such as seismicity, faulting, volcanism, and climate change. In its review of standards for Yucca Mountain, a National Academy of Sciences, National Research Council Committee concluded that there is no scientific basis for a 10,000 year limit on HLW standards for Yucca Mountain [3]. They recommended a regulatory time scale encompassing the time when the greatest risk to public health will occur, and argued for the feasibility of performance assessments on a time scale comparable to that of geologic stability at Yucca Mountain, about one million years. EPA originally required 100,000 year performance assessments to provide comparisons of candidate geologic sites for HLW disposal. On selection of Yucca Mountain, the current EPA standard requires performance assessments for periods longer than 10,000 years to be included in the Environmental Impact Statement. Performance assessments conducted by DOE indicate that maximum doses to exposed individuals in the vicinity of Yucca Mountain occur after hundreds of thousands of years [4]. In some assessments these estimated doses are large relative to normal safety standards.

Space, in General

Isolation refers principally to space. NRC and DOE define the geologic repository as that part of the geologic setting that provides isolation of the wastes. Controlled areas and compliance boundaries with diameters of 5 to 10 km were specified in general HLW disposal regulations. These boundaries have been replaced in current considerations of Yucca Mountain by a spatial constraint based on the location of potentially exposed individuals according to current habitation patterns.

Measures for Yucca Mountain in 2001

Prior formal measures of geologic isolation of HLW, which were codified and refined in the 1980s and 1990s by the EPA, NRC, and DOE, were superceded by new regulations specific to Yucca Mountain in 2001. In the regulations specific to Yucca Mountain, the measure of long-term safety hinges on probabilistic estimates of radiation doses to the average member of a maximally exposed group of people living about 18 km down the groundwater flow gradient within 10,000 years after permanent closure of the repository.

HYDROGEOCHEMICAL MEASURES OF ISOLATION AT YUCCA MOUNTAIN

From a hydrogeochemical perspective, two fundamental conditions conducive to geologic isolation are chemical stability and chemical closure. Isolation is afforded by thermodynamic (or secondarily kinetic) stability of waste forms, containers, and solid phases that ultimately host or encase radionuclides in the geochemical setting. Isolation is also provided by minimal hydraulic gradient and minimal permeability resulting in closed chemical conditions. Hydrologic characterization of Yucca Mountain is beyond the scope of this paper, and recent summaries are available of the evolution in understanding hydrologic features of the site [e.g., 5]. Corrosion rates of waste package materials are treated in detail in many publications in this volume and in other sources. Selected hydrogeochemical aspects of HLW isolation at Yucca Mountain are discussed in the following sections.

Stability of Secondary Minerals

Converging lines of evidence on the role of secondary minerals in the evolution of the Yucca Mountain repository come from the Nopal I natural analog site at Peña Blanca [e.g., 6], from reaction and flow experiments designed to mimic conditions at Yucca Mountain [e.g., 7,8], and from many other sources. These data indicate that oxidation of uraninite, synthetic UO_2, or spent fuel and precipitation of secondary uranyl minerals are rapid relative to migration of uranium out of the immediate vicinity of the primary reduced phase. The oxidation rate of uranium dioxide provides a generally conservative (rapid) limit to the ultimate release rate of radionuclides contained in the spent fuel matrix [6]. Secondary uranyl minerals such as schoepite, soddyite, and uranophane can be stable for millions of years under conditions comparable to Yucca Mountain [e.g., 9]. Characterization of the thermodynamic properties and stability of these mineral phases would provide a comforting and powerful theoretical basis for geochemical isolation, because their stability constrains a rate limiting process for release of uranium and potentially other radionuclides. Theoretical studies to estimate thermodynamic properties of uranium minerals [10] provide an internally coherent data set for modeling. Comprehensive evaluations of empirical data for the thermodynamic properties of uranium [11] provide limited data for uranyl minerals. Experimental studies of uranyl mineral solubilities have been conducted, but data are sometimes challenging to interpret [e.g., 12,13,14].

Challenges also attend evaluations of solubilities of phases that may provide upper limits to concentrations of radionuclides in groundwater at Yucca Mountain, and probabilistic distributions of these concentration limits for use in performance assessment models are poorly constrained [15].

Coprecipitation

Thermodynamic data for uranyl minerals may provide a basis for their stability and a limit on uranium releases, but uranium isotopes are generally small contributors to predicted doses resulting from HLW disposal at Yucca Mountain. Coprecipitation of waste radionuclides as trace solid solution components in stable secondary uranyl minerals could strongly contribute to the isolating capacity of the Yucca Mountain system [16]. Coprecipitation is well recognized in natural systems, but data that are directly applicable to the Yucca Mountain system are sketchy. Recently, experimental and analytical data were interpreted to indicate neptunium incorporation in schoepite in a ratio relative to uranium that exceeded its proportion in the spent fuel supplying both components [17]. More recently, contradictory data showed minimal incorporation of neptunium in schoepite, and the original data were reinterpreted [18]. Data for coprecipitation of trace elements in calcite, which is a relatively well studied system, show that coprecipitation partitioning depends strongly on the rate of mineral growth [19].

Natural Alteration and Ages of Secondary Minerals

Dissolution or alteration of primary minerals and precipitation of secondary minerals characterize open hydrogeochemical systems. At Yucca Mountain the predominant natural alteration is zeolitization of vitric tuffs, which mainly occurred millions of years ago [20]. Relatively recent secondary mineralization yields information on mass transfer on time scales relevant to geologic isolation. Dating microscopic quantities of secondary opal from the Exploratory Studies Facility (ESF) at Yucca Mountain showed that mineralization on fracture surfaces has occurred within the past few thousand years [21]. Reactive transport modeling suggests that reaction of glass in stratigraphic horizons well above the waste emplacement zone with infiltrating groundwater under ambient conditions has a strong effect on water chemistry throughout the unsaturated zone at Yucca Mountain [22].

Uranium decay-series isotopes

Uranium decay series isotopes can be used to identify and quantify characteristics of geochemically open systems, and in could, in principle, provide criteria for isolation characteristics of a geologic setting [23]. Systems that show secular disequilibrium (i.e., ratios of activities of decay-series isotopes deviating from unity) have been open with respect to gain or loss of the radionuclides on time scales comparable to their half-lives. Fracture and matrix tuffs at Yucca Mountain show depletion of uranium-234 relative to uranium-238, indicating that uranium-234 is released preferentially to groundwater on a time scale less than hundreds of thousands of years [24]. Groundwaters and secondary minerals at Yucca Mountain show complementary enrichment of uranium-234 relative to uranium-238 [21]. Uranium decay series data from Nopal I at Peña Blanca indicate multiple stages of uranium mobilization on a time scale of hundreds of thousands of years or less, which is consistent with late formed occurrences of uranium enriched calcite and opal from the site with radiometric dates of about 54,000 years [25].

Bomb Pulse Radioisotopes and Ambient Carbon-14

Radionuclides that are mobile in water and gas phases are particularly sensitive indicators of system openness. Yucca Mountain is currently embroiled in controversy over detection of bomb-pulse chlorine-36. Studies in the late 1990s showed bomb-pulse chlorine-36 systematically distributed in the ESF below structurally fractured zones in the overlying nonwelded Paintbrush Tuff unit, which is otherwise a barrier to fast water flow [26]. Other data from similar samples analyzed by similar methods failed to detect bomb-pulse chlorine-36 [27]. The debate over possible contamination of groundwater with bomb-pulse chlorine-36 notes possible cosmogenic values of chorine-36/total chloride up to about $1250x10^{-15}$. However, possible bomb-pulse contamination could be indicated by substantially lower values of this ratio, less than $1000x10^{-15}$ [28].

Other bomb-pulse radionuclides including carbon-14, tritium, and technetium bear on fast flow paths at Yucca Mountain. Naturally occurring carbon-14 also provides important temporal constraints on system openness. Reactions that transfer carbon between solid, gas, and water phases, and among organic and inorganic compounds make interpretation of carbon-14 complicated and informative. Values of ambient gas-phase carbon-14 in Yucca Mountain are substantially below steady-state atmospheric values, and they decrease systematically with depth in the mountain [29]. These data can be largely rationalized by gas phase diffusion of carbon-14 in CO_2 from the ground surface coupled with equilibrium exchange of carbon between gas and pore waters [30]. Carbon-14 contents of water samples are not directly indicative of water ages, particularly in the unsaturated zone at Yucca Mountain. Low carbon-14 concentrations in the gas phase indicate that large quantities of young water do not percolate through Yucca Mountain and equilibrate with the gas [30].

A MEASURED APPROACH TO GEOLOGIC ISOLATION OF HLW

Measures of HLW isolation specifically applicable to the Yucca Mountain site have been advanced invoking criteria to assure public and environmental safety based on conditions of time, space, and effect. Hydrogeochemical data from the Yucca Mountain site, from natural analog sites, and from experimental studies provide measures of physical mechanisms of waste isolation, release, and migration of radionuclides from the proposed repository. Idealized hypothetical geologic systems of isolation figured in the development of the conceptual basis for permanent geologic disposal of HLW. Functional, risk-informed applications of criteria for geologic isolation of HLW have evolved as the Yucca Mountain site was characterized and selected and as performance assessment methods were refined.

ACKNOWLEDGMENTS

The invitation from the organizers to present this paper at the Materials Research Society Symposium on the Scientific Basis for Nuclear Waste Management XXVIII, and support from the organizers and from the Department of Geological and Environmental Sciences at CSU, Chico, are gratefully acknowledged. A constructive anonymous review helped to focus the presentation.

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