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Development of the Conceptual Models for Chemical Conditions and Hydrology Used in the 1996 Performance Assessment for the Waste Isolation Pilot Plant

Kurt W. Larson

WIPP Technical Integration Department 6848

Sandia National Laboratories

P.O. Box 5800

Albuquerque, NM 87185-0779

Abstract:

The Waste Isolation Pilot Plant (WIPP) is a US Department of Energy (DOE) facility for the permanent disposal of defense-related transuranic (TRU) waste. US Environmental Protection Agency (EPA) regulations specify that the DOE must demonstrate on a sound basis that the WIPP disposal system will effectively contain long-lived alpha-emitting radionuclides within its boundaries for 10,000 years following closure. In 1996, the DOE submitted the *40 CFR Part 191 Compliance Certification Application for the Waste Isolation Pilot Plant* (CCA) to the EPA. The CCA proposed that the WIPP site complies with EPA's regulatory requirements. Contained within the CCA are descriptions of the scientific research conducted to characterize the properties of the WIPP site and the probabilistic performance assessment (PA) conducted to predict the containment

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properties of the WIPP disposal system. In May 1998, the EPA certified that the TRU waste disposal at the WIPP complies with its regulations. Waste disposal operations at WIPP commenced on March 28, 1999.

The 1996 WIPP PA model of the disposal system included conceptual and mathematical representations of key hydrologic and geochemical processes. These key processes were identified over a 22-year period involving data collection, data interpretation, computer models, and sensitivity studies to evaluate the importance of uncertainty and of processes that were difficult to evaluate by other means. Key developments in the area of geochemistry were the evaluation of gas generation mechanisms in the repository; development of a model of chemical conditions in the repository and actinide concentrations in brine; selecting MgO backfill and demonstrating its effects experimentally; and determining the chemical retardation capability of the Culebra. Key developments in the area of hydrology were evaluating the potential for groundwater to dissolve the Salado Formation (the repository host formation), development of a regional model for hydrologic conditions, development of a stochastic, probabilistic representation of hydraulic properties in the Culebra Member of the Rustler Formation; characterization of physical transport in the Culebra; and the evaluation of brine and gas flow in the Salado. Additional confidence in the conceptual models used in the 1996 WIPP PA was gained through independent peer review in many stages of their development.

1.0 Introduction:

The WIPP is a DOE facility for the permanent disposal of defense-related TRU waste. The facility is located about 42 km east of the town of Carlsbad in southeastern New Mexico (Figure 1), a region with semi-arid climate. The WIPP repository is excavated 655 m underground in the Salado, a Permian bedded salt (Figure 2). WIPP TRU waste is created by defense-related industrial activities.[1] TRU waste is defined as waste containing more than 100 nanocuries of alpha-emitting transuranic isotopes, with half-lives greater than twenty years, per gram of waste, and not falling into another category. TRU waste with a surface dose rate less than or equal to 200 millirem per hour is defined as contact-handled TRU (CH-TRU). CH-TRU intended for WIPP is currently stored in 55-gallon mild steel drums or approximately 300-gallon standard waste boxes. About 850,000 containers of CH-TRU waste containing about 13,000 kg Pu will be disposed in WIPP. TRU waste with a contact dose rate greater than 200 millirem per hour is defined as remote-handled TRU (RH-TRU). RH-TRU with contact dose rates up to 1,000 rem per hour can be disposed at WIPP. No more than 6.2 million cubic feet of CH- and RH-TRU can be disposed at WIPP, and the total activity of RH-TRU is limited to 5.1 million curies. TRU waste containing chemicals regulated by the Resource Conservation and Recovery Act is referred to as mixed waste.

Political and regulatory actions have established the context for evaluating whether the disposal of radioactive waste at the WIPP is in accordance with public safety and environmental protection interests. Within this context, the concept of the WIPP disposal system has been developed. The WIPP disposal system comprises the volume of rock beneath a designated 16-square mile tract of land controlled by the DOE. The disposal system includes the repository, shafts, and other excavations associated with waste disposal operations, seals and other engineered barriers, the disturbed rock adjacent to excavations, and a large volume of intact rock. The EPA regulations specify that the DOE must demonstrate that the WIPP disposal system will effectively contain long-lived alpha-emitting radionuclides within its boundaries for 10,000 years following closure.[2,3,4] In 1996, the DOE submitted the CCA to the EPA.[5] The CCA proposed that the WIPP facility complies with EPA's regulatory requirements. Contained within the CCA are descriptions of the scientific activities conducted to characterize the WIPP site and the PA conducted to predict the containment properties of the WIPP disposal system. In May 1998, the EPA certified that the TRU waste disposal at the WIPP complies with its regulations.[6] Disposal of CH-TRU began at WIPP on March 28, 1999.

Geotechnical activities to select a site for a deep geologic repository in southeastern New Mexico began in 1972, and the present site was selected in 1975.[7,8] Since 1975, Sandia National Laboratories has been designated the Scientific Advisor for this project to the DOE and its precursor organizations, the Atomic Energy Commission and the Energy Research and Development Agency. During this period of time, environmental policies in the US were evolving, affecting not only how the decision whether to open WIPP would be made, but also the goals and activities of scientific research conducted. The evolution of national policy on nuclear waste disposal and its effects on WIPP is discussed in more detail by Rechard.[8]

From 1974 to 1996, shifts in the high-level goals and strategies of scientific research occurred, allowing three phases to be defined in general terms. Like colors in the electromagnetic spectrum, these phases represent portions of a continuous process and definitive events marking the transition points do not exist. Early investigations were mainly of a general nature to gain a basic understanding and to confirm the absence of unacceptable features in the vicinity of the WIPP site, although specific and focussed experiments were conducted in certain disciplines.[9,7] Through the 1980's to the early 1990's, perhaps spurred by the emphasis on quantitative predictions in 40 CFR Part 191,[2] scientific investigations became more focussed on characterizing the extent and nature of processes occurring at the site, especially along potential release pathways and in the disposal horizon, which was first excavated in 1982. The realism of WIPP performance predictions improved as data became more definitive and the system-level models became more comprehensive and sophisticated.[10,11] This second phase culminated in several system-level PAs providing evidence that WIPP complied with the EPA's regulations.[12,13] The third phase commenced about 1992 and focussed on augmenting the existing data base and models with specific additional information required to confirm compliance with the EPA's containment requirements, and in developing the CCA license application itself. The third phase was strongly influenced by a management assessment called the Systems Prioritization Method (SPM), which was conducted to identify important sources of uncertainty and the technical activities that could reduce those uncertainties.[14,15,16] Experimental and model development activities will continue until the WIPP is closed to support EPA regulatory oversight, site and facility monitoring, and mandatory recertification of the WIPP facility with EPA's regulations every five years.[17,18]

The National Research Council has concluded that if the WIPP remains undisturbed by human actions, isolation of actinides from the biosphere is provided by the favorable properties of the Salado, primarily its geologic stability and its isolation capability.[19] However, regulations specify that performance of the WIPP shall be predicted based on extrapolating current land-use practices in the vicinity of WIPP to 10,000 years.[18,6] Furthermore, although administrative controls will be implemented and passive markers will be built to deter incompatible activities, the EPA regulations require the assumption that controls and markers are ineffective beyond 100 years after closure.[18,] Because potash and hydrocarbon resources occur in the WIPP vicinity and are currently being developed and extracted, predictions of WIPP performance must include the potential effects of such development inside the disposal system. These considerations lead to the disturbed-performance (or human-intrusion) scenario as the dominant factor in evaluating compliance with the EPA regulations (Figure 3). In the 1996 WIPP PA, the disturbed-performance scenario incorporates: (1) the removal of waste and actinides to the land surface when human intrusion occurs by one or more boreholes drilled through the repository; (2) the long-term effects of plugged, abandoned, and degraded boreholes on fluid flow in the disposal system, including possible penetration of high-pressure brine in fractured reservoirs in the underlying Castile Formation; and (3) the effects on flow in the Rustler due to possible mining of potash in the McNutt Potash Zone of the Salado (Figure 3).[5 Section 6.2,20,21]

Chemistry and hydrology are two of the general scientific disciplines applied to evaluate the containment properties of the WIPP disposal system. Chemical conditions control the formation of dissolved and colloidal actinides and the interaction of these actinides with solids in the disposal system. Hydrology controls the movement of gas and liquid through the disposal system, including the movement (or transport) of actinides with and through those fluids. Together, the disciplines of chemistry and hydrology define many of the conditions necessary to understand the movement of actinides from the repository to the boundaries of the disposal system. During site characterization, general hydrologic and chemical information was gathered for all rock types from the land surface to about 2000 m depth (Figure 4), and some stratigraphic layers were characterized in great detail.[22] Information about hydrology and chemistry of stratigraphic units was collected *in situ* by a variety of sampling and testing techniques, as well as in laboratory experiments. *In situ* data were gathered in boreholes drilled from the surface (Figure 5), and in boreholes and observations made from the access shafts and repository excavations.

Conceptual models describe with words and diagrams the processes that occur within the disposal system (or have a reasonable likelihood of occurring). It is recognized that conceptual models can be formulated at varying levels of complexity and realism, and this has occurred on the WIPP Project. In many cases, the conceptual models developed to explain detailed observations are much more complicated than the conceptual models explicitly represented in the 1996 WIPP PA. The requirements of the system-level PA model in most cases precluded the use of conceptual models accurate enough to also be used to interpret experiments or understand the fine detail of process interactions. Thus, the detailed understanding gained from experiments and complex conceptual models is used to identify and justify the required simplifications.[23] Conceptual models form the basis for the selection of mathematical models, which in turn govern the selection and creation of numerical models and computer codes.

2.0 Conceptual Models of Chemical Conditions

Chemical reactions in the disposal system will control the concentration of actinides in brine in the repository, the sorption of actinides on rocks along flow paths, and, to a large extent, the pressure within the repository. Thus, chemical reactions are expected to play an important role in the containment properties of the disposal system. A basic evaluation program to determine and evaluate the *in situ* chemical composition of disposal system rocks and their pore fluids through sampling began with the earliest site investigations and continued for decades. Four specific programs supplemented the basic evaluation program. A research program for gas generation due to waste and container decomposition began in the 1970's, was curtailed for many years in the mid-1980's, and was renewed in the late 1980's. Research into the possible concentration of actinides and actinide complexes in disposal system brines began in the late 1980's and continues. Backfill materials were proposed and debated over the years, and in some cases tested, as it was generally expected that a backfill material would be emplaced around and over the waste for fire protection and to improve confidence in the long-term mechanical, hydrologic, or chemical properties of the waste disposal area. Formal designation of a backfill material was not made until 1996 when MgO pellets were chosen for their chemical effects. An investigation into the specific effects of the MgO pellet backfill began in 1996 and continues. Experiments were conducted to investigate sorption of actinides on geologic materials in the disposal system in the late 1970's and early 1980's, were initiated again in 1988 and significantly increased through the early 1990's. These topics are discussed in this section, except for sorption, which is discussed in Section 3.2.2.

The major lithologies present in or adjacent to the repository horizon are "clean" halite, polyhalitic halite, argillaceous halite, mixed argillaceous-polyhalitic halite, and anhydrite interbeds. Halite lithologies contain at most 5 wt % insoluble (non-NaCl) minerals. Anhydrite interbeds contain anhydrite, halite, polyhalite, magnesite, and clay. Mineral constituents in these lithologies include halite NaCl, quartz SiO_2, magnesite $MgCO_3$, anhydrite $CaSO_4$, gypsum $CaSO_4 \bullet 2H_2O$, polyhalite $K_2MgCa_2(SO_4)_4 \bullet 2H_2O$, and clays.[24]

Samples of natural brine in the Salado have been collected from boreholes of various dimensions drilled into the back, floor, and walls of the excavation.[25,26,27,28] Intragranular brine occurs as fluid inclusions, and intergranular brine occurs at halite crystal interfaces, in small fractures, and between halite crystals and trace minerals. The major ionic constituents of Salado brine are Na^+, Mg^{2+}, K^+, Cl^-, and SO_4^{2-} with smaller amounts of Ca^{2+}, carbonate, borate, and bromide. Its total dissolved solids concentration is 345,000 to 385,000 mg/l,[29] and ionic strength is about 7.66 molal (M).[30] It is saturated with respect to evaporite minerals and has little potential for dissolving Salado minerals.[31] Heterogeneity in brine composition over short distances in the Salado indicates lack of mixing or flow over long periods of time.[32]

Brine from the Castile or halite-unsaturated groundwater from units overlying the Salado could enter the waste disposal area in the disturbed-performance scenario through human-intrusion boreholes. The major ionic constituents in Castile brine are Na^{+}, Ca^{2+}, Cl^{-}, and SO_4^{2-}.[31] Castile brine is less concentrated than Salado brine with an ionic strength of approximately 6.14 M,[30] and contains significantly less Mg than Salado brine (0.02 M versus about 1 M). Groundwater in the Culebra and Magenta and the Dewey Lake Formation overlying the repository is undersaturated with respect to halite. If undersaturated groundwater entered the repository by flow through intrusion boreholes, it would dissolve halite and become compositionally similar to Castile brine.[33]

2.1 Gas Generation

Gas-generation research and experiments have focused on identifying the mechanisms and rates of gas generation in the repository, taking into account the variability in chemical composition of the waste and possible future conditions. In the late 1970s, experiments were conducted to investigate the radiolytic degradation of cellulose and similar materials (cellulosics), thermal decomposition of organic materials, microbial degradation, chemical corrosion, and helium generation from radioactive decay.[34] These experiments tested the range of conditions from TRU waste to the high-temperature environment created by high-level waste. The main conclusions were drawn from data for an essentially dry repository environment, because appreciable brine flow into the excavation rrom the Salado was not then expected and undisturbed performance was considered dominant. In a dry repository, microbial degradation was the most important and uncertain process, followed in importance by radiolysis of cellulosics and chemical corrosion. Thermal decomposition and dewatering were important at elevated temperatures but probably insignificant at 25°C, approximately the temperature expected for TRU waste-disposal areas. Gas generated by alpha decay was inconsequential for all conditions. If the repository contained brine, the dominant gas would be H_2 from anoxic corrosion.[34]

The WIPP Final Environmental Impact Statement (FEIS)[35] included an analysis of the potential effects of gas generation in the WIPP repository. The analysis included processes of gas flow in Salado salt, fracturing of Salado salt due to high gas pressure, and salt creep in response to high gas pressure. Based on data available at the time, the analysis concluded "there is little possibility of repository failure from overpressurization at gas-generation rates less than 5 moles per year per drum. Since these conclusions depend on the gas-permeability and mechanical properties of the repository medium, they will be subject to some revision when data are available from the actual underground workings."[35, p. 9-156] Based on this analysis, further gas-generation experiments were not conducted pending new data from underground workings.

Beginning in 1982, observations of brine "weeps" on the walls of the excavations[36] and brine collecting in relatively short boreholes drilled from the repository excavations[37] challenged the prevailing hypothesis that the repository would be dry, and caused a series of scientific investigations. The observation of free-flowing brine combined with other data led to the hypothesis that intercrystalline, brine-saturated porosity exists in the Salado.[38] Based on this hypothesis, estimates of Salado permeability were made.[38,39,40] These estimates indicated Salado permeability to be about 10^{-21} m^2, several orders of magnitude lower than the values used in the FEIS analyses of gas-generation consequences. Furthermore, the earlier FEIS estimates of Salado permeability were found to be unreliable.[11, p. 3-6] The prospect of brine in the waste, as well as the new permeability data indicating the potential for high pressures to develop in the waste due to gas generation without gas dissipation, led to a re-start of gas generation experiments in the late 1980's.[41]

The late 1980's and 1990's gas-generation program focused on the processes of anoxic corrosion and microbial degradation.[42,43,44] Anoxic corrosion of ferrous metals in the waste and containers was renewed as a concern because brine seepage from the Salado, as well as the human-intrusion scenario, suggested presence of brine in the repository. Microbial degradation was investigated further because of its importance in the experiments of the 1970's.[34] Radiolysis of seepage brine as a source of H_2 was investigated and found to be inconsequential.[45,46] Based on the 1970's experiments and conclusions, radiolysis of cellulosics was not considered due to low consequence. Interest in thermal decomposition as a gas-generating process waned when the Nuclear

Waste Policy Act of 1982 [47] designated the WIPP exclusively for TRU waste, which generates little heat.

A series of laboratory tests up to 24 months in duration was conducted from 1989 until 1995 to investigate metal corrosion and gas generation. These tests investigated corrosion by reaction between brine or water vapor and major metals in current or potential future waste (low-carbon steel, copper alloys, titanium alloys, and aluminum alloys). The initial composition of the gas phase was varied to simulate potential conditions in the WIPP. Data collected from these tests confirmed that anoxic corrosion of low-carbon steel waste containers could be a significant source of H_2 gas if brine is present in disposal rooms, but found gas generation rates in humid conditions to be negligible. Reduction in the rate of corrosion reactions due to formation of mineral coatings (i.e., "passivation") was found to occur in some situations, but such coatings generally did not persist as chemical conditions evolved.[48,44]

Based on analogs from natural environments,[49,50] a conceptual model of sequential usage of electron acceptors has been applied to microbial degradation in the WIPP repository since 1990.[29] In the WIPP repository, denitrification, SO_4^{2-} reduction, fermentation, and methanogenesis are potentially significant microbial processes. The gaseous products of these reactions are N_2, H_2, H_2S, CO_2, and CH_4.[29] To quantify the possible rates of microbial degradation, a series of laboratory experiments up to 3.4 years in duration was conducted from 1991 until 1995.[51,43] Various substrates, including cellulose and irradiated plastics and rubbers, were examined for brine-saturated and humid conditions.

Other experimental variables were the initial gas composition (aerobic or anaerobic), and the presence of microbial inoculum, nitrate nutrients, and bentonite, a proposed backfill material. Cellulose degradation generated CO_2, N_2O, and H_2 (in some cases). CH_4 and H_2S were not detected. Total gas production was negligible under humid conditions for all substrates. Total gas production from plastic and rubber was insignificant, and unaffected by radiation damage.

Considering all gas generation processes simultaneously, the dominant gas expected in the repository is H_2, with lesser amounts of CH_4, N_2 (or nitrous oxide intermediaries), and H_2S. CO_2 produced by microbial degradation will be removed from the atmosphere by reaction with MgO backfill (see Section 2.3). To represent the creation of these gases in the repository, the 1996 WIPP PA used the "average-stoichiometry" model.[12,52] The average-stoichiometry model uses general equations for gas generation from anoxic corrosion and microbial degradation. The equations are written so that uncertainty in the quantities of reactants and products can be evaluated through parameter sampling.[53] Furthermore, the rates of reaction are determined by sampled parameters, and the occurrence of gas generation is dependent on the presence of brine. In the case of anoxic corrosion, brine consumed by reaction is removed from the model, preserving an important feedback between gas generation and brine saturation. The parameters governing rates and stoichiometry of the anoxic corrosion and microbial degradation models are determined by analysis of experimental data such as those described in preceding paragraphs. The average-stoichiometry model does not explicitly calculate the composition of gases produced by corrosion and degradation reactions.

Although microbial degradation of cellulose has been demonstrated in laboratory experiments,[51,43] "the occurrence of significant microbial gas generation in the repository will depend on: (1) whether microbes capable of consuming the emplaced organic materials will be present and active; (2) whether sufficient electron acceptors will be present and available; [and] (3) whether enough nutrients will be present and available."[54] Uncertainty in the long-term viability of microbial degradation reactions was acknowledged in the 1996 WIPP PA by assigning a 0.5 probability to the occurrence of significant microbial activity. Similarly, even though irradiated plastic and rubber did not degrade in laboratory experiments, the long-term chemical stability and therefore biodegradability of plastics and rubbers, plus the possibility of cometabolism with cellulose, led to the assignment of a 0.5 probability of plastic and rubber degradation conditional on the occurrence of significant microbial activity.[54,53]

2.2 Actinides in Solution

Actinides in solution refers to the total concentration of actinides in liquid in the repository, including dissolved actinides, actinide-organic molecule complexes, and colloidal (suspended) actinides. Serious consideration of the concentration of actinides in the repository brines began in the late 1980's when it was recognized that credible PAs to demonstrate compliance with EPA regulations required reliable estimates of actinide concentrations in brine. However, the earliest considerations of actinides in solution occurred in the 1970's, when deterministic dose assessments were conducted for several scenarios of containment failure. The scenarios involving movement of brine from the disposal system included a very approximate model for actinide concentrations in brine, in which it was assumed that the bulk mass of the waste, including actinides, could dissolve at the same rate as halite into unsaturated groundwater circulating through the repository via boreholes.[35]

A systematic program to collect data and develop models relevant to repository chemical conditions and radionuclide chemistry began in 1989.[29] The aim of this program was to define the potential chemical conditions, dominant chemical reactions, and actinide speciation and solubility in the repository over 10,000 years. Experiments to investigate colloidal actinides were started in 1992,[29,55] and were significantly expanded when the 1994 SPM confirmed the that reducing uncertainty in this area would enhance confidence in compliance with the EPA's regulations.[14,15,16] Although there are many radioactive elements included in the WIPP waste inventory,[1] plutonium, americium, uranium, thorium, and neptunium were the focus of the actinide solubility program because these elements comprise more than 99% of the solid actinide inventory.[56,77]

Given the absence of actinide concentrations based on specific data and models, until the 1996 WIPP PA actinide solubilities were estimated based on scientific experience and extrapolation from literature data to WIPP conditions. A 1989 assessment of system performance used bounding solubility limits, based on consideration of possible chemical conditions. The solubility limits were 10^{-4} M for all actinides in Salado brine, and 10^{-6} M for all actinides in Castile brine.[11] A 1989 PA methodology demonstration used 10^{-6} M as a median value for a range of solubilities from 10^{-9} M to 10^{-3} M.[57] Preliminary PAs conducted in 1991[12] and 1992[58] included uncertainty in actinide concentrations developed by a process of expert elicitation.[59,60] Lack of actinide solubility data in the literature for high ionic strength brines contributed to the overall uncertainties, as did the wide range of plausible conditions for the waste-disposal area at the time (see Figure 6). The expert elicitation process produced wide ranges of concentrations for use in the 1991 and 1992 PAs; for example, Pu(IV) as $(Pu(OH)_5)^-$ was assigned a range from 2 x 10^{-16} M to 4 x 10^{-6} M with a median value of 6 x 10^{-10} M; Am(III) as $(AmCl_2)^+$ was assigned a range from 5 x 10^{-14} M to 1.4 M with a median value of 1 x 10^{-9} M.[59,60] Due to the absence of a comprehensive perspective on colloidal actinides, colloid effects were not included in the 1991 and 1992 preliminary PAs.[60]

At the outset of specific experimental and modeling studies to determine chemical conditions and actinide concentrations in WIPP disposal rooms, there was great uncertainty in the possible conditions under which reliable estimates of actinide concentrations would be required. As shown in Figure 6, uncertainty in pH ranged from about 3.5 to 11.5, and uncertainty in oxidation potential (Eh) encompassed the full spectrum of water stability.[61] Accordingly, the objectives of laboratory studies initiated to study radionuclide chemistry were to: "(1) quantify the speciation of Pu, Am, Th, and U in neutral, acidic, and basic solutions of high ionic strength under a wide range of redox conditions … for calculations of the solubilities of these elements in WIPP brines; (2) determine, if necessary, the solubilities of Pu-, Am-, Th-, and U-bearing solids under similar conditions … to validate the results of the speciation study…."[29] The conceptual model for actinide concentrations in brine would be developed in three steps: first, data would be collected and analyzed to define the chemical reactions that might occur in the range of possible chemical conditions in waste disposal rooms; second, a data base would be developed along with speciation-solubility models for those conditions; and, finally, the conditions in disposal rooms would be modeled using speciation-solubility models to determine actinide concentrations for specific conditions.

The conceptual model for actinide concentrations in brine was implemented in the 1996 WIPP PA through random sampling of a limited number of parameters. These parameters allow estimation of actinide concentrations by simple algebraic equations rather than by speciation-solubility calculations conducted during PA. Temporal and spatial variation in the chemical conditions influencing actinide concentrations could be incorporated only through gross generalizations. Thus, developing parameters to directly represent the concentration of actinides in brine required a sophisticated understanding of the potential liquid saturation and composition and gas pressure and composition in the waste-disposal rooms prior to development of parameter ranges. Estimates of the variability in these physical conditions were provided during the course of experimental studies by preliminary PAs.[12,13]

The conceptual model for dissolved actinide concentrations is described in Novak and others[31] and the CCA.[33] The overall chemical conditions in the waste-disposal area are strongly influenced by the composition of Salado brines that could seep into the waste disposal area and by Castile brines that could enter the repository through an intrusion borehole[20]. The pH of the waste-disposal area is expected to be moderated by MgO backfill to approximately 9.5, and the oxidation potential of the disposal system is low, due to corrosion of metallic iron in the waste containers and possible microbial activity. Other potential influences on the aqueous chemical conditions are the presence of cementitious materials in seals and some of the waste, and commercial organic complexants (e.g., EDTA) in some of the waste.

The chemical conditions in the waste-disposal area are too complex to be described exactly. Assumptions were made to make the problem tractable, using conservative assumptions where necessary. Key assumptions include (1) complexation and precipitation/dissolution (nonredox) reactions can be described with equilibrium thermodynamics, (2) the effect of sorption of actinides on immobile substrates in the waste-disposal area can be ignored, and (3) actinide conversion among oxidation states (redox reactions) is not an equilibrium process[31].

Except for actinides in the VI oxidation state, the Pitzer formalism[62] using the thermodynamic data bases of Harvie and others[63] and Felmy and Weare[64] was the starting point for estimating equilibrium actinide solubilities in the WIPP disposal rooms. This initial model was extended to the WIPP system by adding to it the standard chemical potentials of the aqueous and solid chemical species containing actinides, and the specific ion interaction parameters required to describe the interactions between those species and other constituents of disposal-room brines. Parameters for organic waste constituents were also added to the data base. Data and supporting information were compiled for the actinide (III) data base extensions used for Am(III), Pu(III), Cm(III),[65 p. 1498,66,67,68,69,70] the actinide (IV) data base extensions used for Th(IV), U(IV), Pu(IV), and Np(IV),[71,72,73] and the actinide (V) data base extensions used for Np(V).[74] At the time of the CCA, the actinide (VI) data base was insufficiently developed to support equilibrium solubility calculations. An empirical measurement of U(VI) concentration was used instead of model predictions.[33,75]

The FMT computer code implemented the Pitzer formalism and was used to calculate the solubility of Am(III), Th(IV), and Np(V), which are chemical analogs for actinides of interest in the III, IV, and V oxidation states.[76] An oxidation state analogy, which asserts that actinides in the same oxidation state exhibit similar chemical behavior, was used to extend the behavior of analogue actinides to other elements (Table 1). Solubilities were calculated for Salado and Castile brine. Implementation of the dissolved actinide model in the 1996 WIPP PA is described in Stockman and others.[77]

Colloidal particles are generally defined as particles with at least one dimension between 1 nm and 1 μm that are maintained in suspension in a liquid through Brownian motion.[78,79] Colloids can form by a variety of physical and chemical mechanisms including mineral fragmentation, intrinsic colloidal formation by actinides themselves or microbial degradation of cellulosic materials (e.g., humic acids). Additionally, microbes themselves may be considered to be colloidal particles because, though generally larger than the specified colloidal size range, their specific gravity is generally equal to the solution so they will not tend to settle out of solution by gravity. These four types of colloids – mineral fragments, actinide intrinsic, humic acids, and microbes – were each separately investigated with respect to their potential to increase actinide concentrations in brine.[55]

The colloidal actinide research results indicated that both mineral fragments and actinide intrinsic colloids have little potential to enhance mobile dissolved actinide concentrations under the expected chemical conditions at the WIPP site.[33] Microbes and humic acids will complex actinides and contribute to the actinide concentrations in brine. The concentrations of actinides complexed by microbes and humic acids were represented in the 1996 WIPP PA using two parameters.[53] The first parameter was a proportionality constant representing the microbial and humic contributions to actinide concentration in brine. These proportionality constants were multiplied by the dissolved species concentrations to obtain values for the microbial and humic contribution to the total species concentration. A second parameter was included to indicate a maximum value for complexation (or metal toxicity on microbes) via these two colloidal mechanisms. Sorption of actinides onto fixed substrates in the waste-disposal area was not considered because it would not affect actinide concentrations in brine unless most of the actinide inventory were sorbed, which is considered unlikely.

2.3 MgO Backfill Effects

The potential for backfill to improve the long-term mechanical, chemical, and hydraulic properties of the WIPP repository is well recognized, and is reflected in the research conducted in this topic and in the political and regulatory framework of WIPP.[80] Until salt was no longer considered for the disposal of high-level waste,[8] the retardation of actinide and fission-product movement by precipitation, sorption, and slow diffusion in bentonite backfills was investigated,[81,82,83,84] as well as the mineralogic stability of bentonite backfill.[85] Although the possible use of crushed salt backfill to reduce subsidence and fire potential was recognized early,[86,87,88] concern over brine seepage from the Salado in the late 1980's gave rise to sustained investigations on the potential of crushed salt and bentonite backfills to reduce liquid flow to and through the waste disposal rooms.[89,90,91] Furthermore, other aspects of backfill characteristics and options were evaluated to varying extent, including gas getters and grouts.[11,29,92,93] Thus, by 1991, the advantages of many options for backfill materials had been considered or investigated, in variable detail, for mechanical, chemical, and hydraulic properties.

As discussed in Section 2.1, if gas generation by microbial degradation of carbon-containing waste material occurs, a significant quantity of CO_2 may be introduced into the waste-disposal rooms. Carbonate ions are known to bind strongly to actinides, forming stable, relatively soluble species. In late 1995 and early 1996, concerns were forming that the relatively high solubilities might cause non-compliance with the EPA containment requirements. Based on the backfill research available at that time, and in cooperation with operations engineers at the management and operating contractor, Westinghouse Electric Corporation, a backfill of MgO (periclase) pellets was selected to control CO_2 concentrations and limit actinide solubility.[94] MgO reacts with water,

$$MgO\ (s) + H_2O\ (l) = Mg(OH)_2\ (s), \tag{1}$$

forming brucite. Brucite reacts with carbonic acid,

$$Mg(OH)_2\ (s) + H_2CO_3 = MgCO_3\ (s) + 2\ H_2O, \tag{2}$$

forming magnesite and water and virtually eliminating CO_2 from the disposal-room environment. Furthermore, the MgO hydration reactions effectively buffer the disposal room to a pH between 9 and 10, conditions of relatively low dissolved actinide solubility. The only effects of MgO incorporated in the 1996 WIPP PA were its reduction of total concentration of actinides in solution and elimination of CO_2 from the gas phase, reducing the quantity of gas in the disposal rooms. Other effects were ignored on the basis that they were beneficial, such as removal of water from the disposal room during hydration, and the potential for the precipitated phases such as brucite and magnesite to bind waste materials together, which could reduce the volume of cavings and spallings releases.[95,96]

The actual reaction mechanism for the formation of magnesite, $MgCO_3$, is more complicated than suggested in equations (1) and (2). Figure 7 illustrates the four main steps in the sequence as the formation of brucite by hydration, dissolution of brucite, carbonation, and dehydration, which is a maturation process that may include formation and destruction of several metastable phases.[97]

The reaction mechanism for the formation of magnesite from periclase needed confirmation in WIPP-relevant conditions before it would be accepted by a mandated peer-review panel.[98,99,100] The basic reaction was confirmed by immersing MgO pellets in relevant brine solutions and exposing them to CO_2 by bubbling. Hydromagnesite, $a\mathrm{MgCO_3{\cdot}Mg(OH)_2{\cdot}}b\mathrm{H_2O}$, $\{a,b\} = \{3,3\},\{4,4\},\{4,3\}$, formed in the experiments as the intermediary phase during the maturation process; magnesite was not formed in the experiments, due to suspected kinetic inhibition. The morphology of precipitated minerals was investigated to evaluate whether precipitated minerals would shield the interior of MgO pellets from further reaction; scanning electron microscope pictures show no shielding (Figure 8). Data to support the eventual formation of magnesite in the repository were obtained from laboratory studies and natural analogue studies reported in the open literature.[97]

3.0 Conceptual Models of Hydrology

The conceptual models of hydrology used in the CCA result from nearly continuous investigation of northern Delaware Basin hydrology since 1974 conducted specifically for the WIPP. During this period, the general hydrologic conditions from the surface to depths exceeding 2,000 m were established through testing and monitoring programs conducted from the land surface. For the geologic units that are plausible transport pathways, specific hydrologic conditions and transport characteristics were determined from testing and monitoring programs conducted in boreholes drilled from the surface, in underground boreholes drilled from shafts or excavations, and in laboratories. Data have been collected on hydraulic potentials (heads), major element (chemical) composition, and, in some cases, isotopic compositions.[101] Direct examination of rock samples from drill cores and in shaft exposures has aided the development of conceptual models for the flow and transport properties of the subsurface. Especially since the mid-1980's, hydrologic models have been used extensively to guide testing activities, to estimate conditions in locations not directly tested, and to evaluate the effects of slow processes, such as climate change, that cannot be directly tested. At the WIPP site, a monitoring program for hydraulic potentials and chemical composition of groundwater in the near-surface units is implemented, and activities occurring in the region with the potential to affect subsurface fluid potentials are monitored.

3.1 Regional Hydrologic Conditions

A primary objective of locating the WIPP repository in the Salado was to isolate the waste from circulating groundwater. Accordingly, an early goal of site characterization activities was to evaluate the potential for evaporite dissolution to affect waste isolation. Dissolution has long been recognized as a primary process affecting landforms in the vicinity of WIPP.[102,103] For example, Nash Draw, about eight kilometers west of WIPP (Figure 5), is a valley caused by dissolution of the upper Salado and subsidence and erosion of overlying rocks. The investigation of dissolution features in the early years of the project was extensive. However, because most dissolution was found to be of low consequence to the isolation of waste,[46,20] it is not central to the theme of this paper. Summaries of the dissolution investigations conducted during characterization of the

WIPP site are presented elsewhere.[22,20,7,104,105,106] Only dissolution as it affects the hydraulic properties of the Culebra is discussed in following sections.

Since dissolution does not threaten to breach the disposal horizon, all evidence suggests that the TRU waste will remain isolated from the biosphere for a sufficient period of time if it is not disturbed by human intrusion. However, in the human-intrusion scenario considered in the 1996 WIPP PA, processes are assumed to occur that may move radionuclides into geologic formations containing circulating groundwater. Groundwater circulation is influenced by factors such as topography, climate, and geological conditions that extend well beyond the boundaries of the WIPP site. Regional hydrologic conditions have been investigated mainly through computer modeling, but *in situ* hydraulic testing in the WIPP area, chemical and isotopic analyses of groundwater samples, and electromagnetic geophysical soundings augment the model interpretations. Characterization was directed at the more permeable units bounding the Salado (see Figure 4) – the overlying Dewey Lake Formation, Magenta, and Culebra, and the underlying Castile and Bell Canyon Formations. Early in site characterization, the unnamed lower member of the Rustler Formation and its underlying Rustler-Salado contact zone were thought to be a potential transport pathway, but interest in this potential pathway waned after several tests in the unit found relatively low transmissivity at the WIPP site.[11, p. 3-28 et seq.] Similarly, a few tests were conducted in the Tamarisk and Forty-niner Members of the Rustler Formation to confirm that they have very low potential to produce or conduct water.[107,108]

The conceptual model for regional hydraulic conditions supporting the CCA is based on well-developed concepts of regional groundwater flow in groundwater basins.[109] A groundwater basin is a three-dimensional closed hydrologic unit bounded on the bottom by a rock unit with much lower permeability than the units above (i.e., an "impermeable" unit), on the top by the ground surface, and on the sides by groundwater divides. Differences in the elevation of the water table across the groundwater basin provide the driving force for groundwater flow. All recharge to the basin is by percolation of precipitation to the water table and all discharge from the basin is by flow across the water table to the land surface. The pattern of groundwater flow depends on the lateral extent of the basin, shape of the water table, and the heterogeneity of rock properties, chiefly permeability, within the basin.[109]

The concepts of regional groundwater flow to develop models for fluid flow in the WIPP area were first used by Davies in the late 1980's.[110] His series of two-dimensional numerical analyses investigated the magnitude of gravity and density driving forces for groundwater flow, the role of vertical flux, the potential effects of river elevation changes, and the role of long-term climate change. These results supported the idea that long-term climate change could influence present-day flow systems.

In the 1990's, a three-dimensional groundwater basin conceptual model was created for the WIPP region above the top of the Salado.[109] The model was implemented with a computer code called SECO3D,[111] and had the capability to model transient motion of the water table. Hydrostratigraphic units in the model were defined based on experience with

WIPP rock types and structure-contour maps developed from geophysical logs.[109] Geologic data for analogue rock types were used to develop "intact" hydraulic properties resulting from depositional and diagenetic processes. The effects of post-depositional alteration that has occurred in the region were incorporated through modified parameter values. Uncertainty in the values of rock properties was assessed, and its impacts evaluated through deterministic simulations.

The regional groundwater basin model was used to simulate how changes in infiltration rates due to climate change[112] would influence the flux of groundwater laterally and vertically through hydrostratigraphic units at the WIPP site. Climate change over the next 10,000 years was found to increase lateral flux through the Culebra by a factor of 1.0 to 2.25.[113] This information was used to create the simple climate change model used in the 1996 WIPP PA. Regional groundwater flow modeling also predicted that all discharge from the Culebra at the WIPP site occurs by lateral flow, indicating that contaminants potentially released into the Culebra will move toward the accessible environment within the Culebra and not move upward or downward into other units. Based on this modeling result, a confined-aquifer groundwater flow model was considered reasonable for simulating flow and transport in the Culebra in the 1996 WIPP PA.[114]

3.2 Culebra Fluid Flow and Transport

Geohydrologic investigations from 1975 to the mid-1980's were intended to gather basic data on all of the rock types and stratigraphic units in the WIPP region. Beginning in the mid-1980's, however, geohydrologic investigations began to focus intensively on two stratigraphic units – the Salado hosting the repository, and the Culebra. The Culebra became the focus of investigation for several reasons. The Dewey Lake Formation was thought to be unsaturated or partially saturated with perched groundwater, which is not conducive to lateral transport.[115] (The upper Dewey Lake is now thought to be saturated and fractured in some places, but resistant to radionuclide transport because the iron-bearing minerals it contains will retard radionuclide movement by sorption).[116] Interest in the Bell Canyon and other formations below the Salado waned as it was determined that Bell Canyon rock properties and hydraulic gradients were not conducive to rapid lateral transport.[115,107,35 p.7-87] Hydraulic tests conducted in the Magenta indicated lower transmissivity than the Culebra, generally homogeneous porous medium behavior on the scale of well tests, and regional transmissivity variation.[108,117] Models of groundwater flow in the Magenta indicated slow groundwater velocities.[118] In contrast, as discussed in the next section, by the mid-1980's, *in situ* well tests, regional pumping tests, and tracer tests revealed the Culebra to: (a) be the most transmissive unit in the stratigraphic sequence; (b) transmit water pressure pulses quickly over kilometers in some areas; (c) have east-west transmissivity variation of more than six orders of magnitude in the vicinity of WIPP; and (d) have hydraulically important fractures.

The hydraulic properties of the Rustler and Culebra have been affected by dissolution. Precipitation and dissolution of gypsum in fractures accounts for some of the regional variation in Culebra transmissivity.[119] Halite dissolution in the Forty-niner, Tamarisk, and unnamed lower member may also account for some transmissivity variation in the

Rustler generally, however the timing of this dissolution is uncertain. Based primarily on isopach maps of the Rustler Formation members and the relation between halite margins and Nash Draw, the westward-thinning of halite in the Rustler has been attributed to post-depositional dissolution.[120,121,122] However, based on examination of Ruster sedimentary features, the same phenomenon has been attributed to lateral variation in depositional processes or syndepositional dissolution.[123] Disagreement persists over the genesis of the observed distribution of halite in the Rustler. However, even in the absence of consensus regarding the origin of Rustler hydrostratigraphic features and properties, it is generally accepted that the program of *in situ* testing and analysis have determined the Culebra hydraulic properties sufficiently for the 1996 WIPP PA.[98 p. 53]

3.2.1 Culebra Groundwater Flow

A network of 57 wells had been completed to the Culebra by 1988, in which numerous types of tests had been conducted, yielding information about its hydraulic properties and conditions in the vicinity of WIPP.[124,115,107,125,126,108,127,128,129,130] The testing techniques used included drillstem tests, slug tests, pumping tests, hydraulic-interference tests, and long-term monitoring programs. The drawdown and recovery of Culebra hydraulic head in response to drainage into repository shafts was monitored, which was used in inverse calibration techniques to provide information on hydraulic properties over a large region of the central portion of the WIPP site.[131,132]

Groundwater models were also important in developing the conceptual model of groundwater flow in the Culebra used in the 1996 WIPP PA. Inverse techniques were first used to infer site-scale hydrologic properties of the Culebra in the late 1980's and have been used ever since.[133,132,134,135,136] Haug and others modeled present-day groundwater flow in the Culebra using a kriged transmissivity field derived from *in situ* measurements. Through modeling steady-state conditions and transient flow caused by drainage of the Culebra during construction of the WIPP shafts, they found that observed head distributions could be matched only by increasing transmissivities in a region southeast of the waste panels.[132] Geological data and *in situ* tests had previously indicated a trend of higher transmissivity there, but model fits could not be obtained without using transmissivity values quite in excess of any observed in previous tests or inferred from geologic data. The high-transmissivity zone "discovered" in the groundwater models sparked a series of surface-based geophysical investigations[137,138] and *in situ* hydrologic testing in new boreholes H-17 and H-19 with the goal of defining the properties of the high-transmissivity zone. Despite the additional characterization efforts, the exact dimensions of the high-transmissivity zone remain unknown.

To assess the uncertainties caused by the high-transmissivity zone and Culebra heterogeneity generally, stochastic models using inverse techniques have been developed and applied.[135,136] For the 1996 WIPP PA, the inverse technique was used to create 100 realizations of Culebra transmissivity in the vicinity of WIPP.[136,114] Using the stochastic transmissivity fields as a sampled input parameter, the 1996 WIPP PA results indicate that the most-likely pathway for transport of radionuclides away from intrusion boreholes is the Culebra high-transmissivity zone.[114]

Within the WIPP site but not directly over the repository, the upper Salado contains potash. Potash mining is ongoing and is expected to continue, although mining within the WIPP site is currently prohibited. The EPA regulations for the WIPP specify a conceptual model for the effects on groundwater flow in the Culebra of subsidence over existing and hypothetical potash mines.[5] The EPA developed its model of subsidence effects by reviewing studies of analogs to WIPP mining and subsidence and through its own modeling. Data was identified in the literature for New Mexico potash mines and for longwall coal mines in West Virginia, Illinois, and Pennsylvania. Modeling analyses in the literature were also identified, and supplemented by strain-modeling for conditions relevant to the WIPP.[139 pp. 9-30 et seq.] These literature reviews and modeling analyses led the EPA to specify that mining effects could be incorporated solely by modifying the transmissivity of the Culebra.[5] The DOE incorporated the EPA model by multiplying the intact hydraulic conductivity of the Culebra above potash deposits similar to those being mined currently by a factor randomly sampled between 1 and 1000.[114] The probability of future potash mining within the currently withdrawn WIPP disposal system is described in Helton and others.[21]

3.2.2 Radionuclide Transport in the Culebra

The potential mechanisms of radionuclide transport in the Rustler generally, and in the Culebra in particular, have been of interest since the WIPP project was established, and many studies have been conducted. For example, sorption data were collected for samples from the Magenta and Culebra, with liquids of various compositions, in the late 1970's and early 1980's for many relevant actinides.[140,141,142] In 1980, soon after the transmissivity of the Culebra was recognized to be strongly influenced by fractures,[124] a dipole tracer test was conducted in the H-2 wells, and later other *in situ* conservative tracer tests were conducted to identify diffusion properties of the fractured rock.[143] However, over time it was realized that the early chemical retardation data did not differentiate between sorption mechanisms (adsorption and ion-exchange) and other retardation mechanisms such as precipitation of actinide-bearing solids.[144] Similarly, it was realized that the initial tracer tests in the Culebra were ambiguous because experimental procedures, fracture network heterogeneity, or matrix diffusion all could explain the observed results.[145,146]

Theoretical and modeling studies investigating parameter sensitivity and importance for radionuclide transport in the Culebra were conducted as well. For example, it was calculated that natural flow in the Culebra and the size of the disposal system are such that physical retardation by diffusion of solute into matrix blocks effectively balances advection in fracture networks, allowing use of equivalent single-porosity models.[147]

By 1992 there was a feeling of confidence, but an inability to demonstrate conclusively, that radionuclides entering the Culebra would diffuse from fractures into the rock mass and sorb to mineral surfaces. Insufficient data existed to eliminate alternative conceptual models in which there was little diffusion into the matrix or no chemical retardation (Figure 9). The effect of this conceptual model uncertainty was tested in a 1992 preliminary PA, showing significant differences among the three alternative models considered (Figure 10).[148] Some experiments to reduce uncertainty in actinide sorption were initiated in the early 1990's. However, a comprehensive approach was not adopted until the 1994 SPM confirmed that reducing uncertainty in the Culebra transport model would significantly enhance confidence in compliance with the EPA regulations.[14,15,16] Thus, although the basic ideas were in place more than a decade before, a rigorous basis for the CCA conceptual model for radionuclide transport in the Culebra was developed mainly by investigations conducted since 1994.

WIPP scientists charged with characterizing the radionuclide transport properties of the Culebra divided chemical retardation into a separate program of investigation from physical transport and retardation. This is reasonable from the perspective that physical transport and retardation are affected only by the physical structure (e.g., fracture surface area, matrix porosity, and tortuosity) of the medium while chemical retardation (sorption) is affected by the chemical properties of the transporting fluid and rock (e.g., mineralogy). Physical transport and retardation were investigated with a program of *in situ* conservative tracer tests and laboratory diffusion tests. Chemical retardation was investigated in a program of laboratory sorption tests. Despite the strong encouragement for and even recommendation of an *in situ* sorbing tracer test by outside review groups, the scientists involved believed such a test presented many practical problems in both procedures and analysis that could cause ambiguous results. Considering its fielding and interpretation problems, its high cost, and the presence of a reasonable alternative approach, the *in situ* sorbing tracer test could not be justified. As discussed in the following sections, coupled physical transport and retardation and chemical-retardation experiments were conducted in the laboratory with flow-through experiments using intact core from the Culebra and some of the actinides of interest.

Culebra Physical Transport and Physical Retardation

Solute transport occurs in the pore space of rocks. In the Culebra, the pore structure is heterogeneous, as stated by Holt:[149]

"Within a fractured geologic material, such as the Culebra, pore space is present in both the interconnected network of fractures and the blocks of geologic medium. In the Culebra, this relationship is further complicated because several scales of fracturing are present [[123,119]], and several types of porosity occur within the fracture-bounded blocks. In addition, fractures may not be the only advective transport path in the Culebra, as interbeds of poorly cemented, silty dolomite may actively participate in advective transport of solutes. Furthermore, diffusion and slow advection into the fracture-bounded blocks of dolomite may significantly affect solute transport in the Culebra."

In the context of this complex pore structure, several lines of investigation were pursued to characterize and quantify the mechanisms of transport in the Culebra. The basic conceptual model was developed by describing in detail the pore structures of the Culebra (Figure 11), with conceptual connections among diffusion processes and the different types of porosity, including a mathematical representation allowing comparison of observed transport behaviors at different spatial and temporal scales. This framework created a consistent basis for comparing results from the laboratory with those from the field, and for extending interpretations to the larger distance and time scales necessary in the 1996 WIPP PA.[149]

To characterize transport properties *in situ*, the H-19 seven-well cluster was installed at the north end of the high-transmissivity zone in the most likely off-site flow path (Figure 5). The H-19 well cluster provided geologic data from drill cores, downhole geophysics, and borehole imaging.[149] At the H-19 well cluster, hydraulic properties were characterized with flow tests.[130] Two types of conservative tracer tests were conducted at the H-19 and H-11 well clusters.[150] The multi-well convergent-flow tracer tests characterized the relationship between heterogeneous advective porosity and diffusive porosity by varying pumping rates and tracer diffusion constants.[151,152,153] The single-well injection-withdrawal tracer tests reduced the effects of advective porosity and provided better discrimination of diffusion mechanisms and rates.[151,154] Simulations were conducted to determine if fracture network heterogeneity alone could explain the observed results. These calculations demonstrated that variability in fracture conductivity could not reproduce experimental results.[151]

The tracer-test analyses show that *in situ* observations cannot be matched with a single-porosity, fracture-flow conceptual model, which holds that physical retardation by diffusion into slow-moving or stagnant pore fluids does not occur. Furthermore, the experiments provided conclusive evidence of multiple rates of diffusion in the pore structure of the Culebra (Figures 12 and 13). Experiments to image the diffusion process in the laboratory were also conducted to confirm that diffusion occurs at multiple rates into different types of porosity in the rock matrix.[155] The *in situ* flow and tracer tests at H-11 and H-19, post-test analyses, and laboratory diffusion studies demonstrate the mechanisms and rates of mass-transfer through different types of porosity in the Culebra in the most-likely flow path. All of these lines of evidence were used to support the choice of computer models and parameter values used in the 1996 WIPP PA.[114]

Culebra Chemical Retardation

Post-1992 sorption research focussed on the mineral dolomite [$MgCa(CO_3)_2$], because it comprises > 95% of the Culebra. The potential for additional sorption on minor mineral phases such as clay was not emphasized because the models of Culebra pore distribution and physical transport were not being developed in sufficient detail to substantiate transport to the locations of minor mineral phases.[156]

The mechanisms of sorption and the shape of the sorption isotherms were investigated in three laboratory approaches.[55] The empirical (batch) sorption study was conducted to rapidly establish minimum K_ds for a variety of WIPP-relevant conditions using crushed

Culebra dolomite and manufactured chemical analogues for natural groundwater types found at the WIPP site.[55,156] The mechanistic sorption study investigated the effects of ionic strength, CO_2 concentration, pH, and actinide oxidation state on adsorption behavior using carefully controlled solution chemistry and crushed samples of pure dolomite.[157,158,156] The intact-core column experiment used large-diameter intact cores and WIPP-relevant brines to investigate the effects of advective fluid flow on sorption in the Culebra.[159,160] Because a linear-isotherm (K_d) model was required for the 1996 WIPP PA to allow significant reduction in computational effort,[114] the laboratory data were used to choose effective K_ds, even though, in some cases, more complicated isotherms were observed.[156]

3.3 Salado Fluid Flow and Transport

The initial interest in the hydraulic properties of the Salado was to estimate the quantity of brine that could enter the waste-disposal rooms. The presence of approximately 0.1 to 1 wt % of free (not chemically bound) pore fluid in the Salado was recognized early in site characterization.[161,9,162,163,164] However, this pore fluid was thought to be contained in intracrystalline pores with little interconnection.[9 p. 6-20] *In situ* permeability tests of the Salado conducted from the surface at AEC-7 evaluated the hydraulic conductivity of the Salado,[165] but these estimates were later determined to be unreliable.[129] In the early-to-mid 1980's, observations from the underground of brine seepage, as well as other tests, led to the identification of intercrystalline porosity containing brine within the Salado. Meaningful estimates of the permeability associated with Salado intercrystalline porosity were not obtained until access to the underground was achieved in shafts and in the repository excavation. Estimates of Salado permeability were derived from *in situ* observations and tests, including flow into open boreholes,[39,40,166] tests in shafts and around seals,[167,168,169] and hydraulic tests.[129,170]

Due to brine seepage and possible high-pressure gas (see Section 2.1), it was recognized that fluid movement in and near the repository could occur as two-phase flow. Laboratory and field experiments and modeling studies were conducted to build a conceptual model of two-phase flow in the Salado. Because halite-rich stratigraphic layers were considered to be impenetrable to gas due to capillary effects,[171] efforts focused on determining the two-phase characteristics and behavior of anhydrite interbeds. The capillary pressure and relative permeability characteristic curves of Salado anhydrite were determined from tests on small rock samples in the laboratory.[172] The gas-threshold pressure of anhydrite interbeds was determined through *in situ* tests.[173] Two-phase flow computer codes were adapted to the WIPP situation to conduct PAs and sensitivity studies,[174,58,175,176] as well as to investigate the potential importance of countercurrent flow in the slightly dipping strata,[177] to evaluate the effects of fine details of stratigraphy,[178] and to investigate the errors incurred in interpreting brine-inflow experiments with single-phase flow models.[179]

The interaction of rock mechanics and fluid movement in the disturbed rock zone around the excavation was recognized as important and several characterization investigations were completed during the late 1980's and early 1990's.[180,181,182,183] To validate models based on small-scale observations, a large-scale brine-inflow experiment was conducted in Room Q, an approximately 3-m-diameter, 100-m-long horizontal borehole. Room Q was instrumented with resistivity arrays to observe conductivity changes caused by deformation and fluid flow,[184] and included sumps for collection of brine.[185] A network of boreholes was installed around it in which hydraulic properties were measured and pore pressures monitored.[186] Long-term trends in Room Q data were analyzed from the perspective of rock mechanics[184] and hydrology.[187,188] Room Q data and analysis indicate that gas-filled porosity caused by dilation in the disturbed rock zone can become saturated with brine flowing slowly from lower permeability surrounding rock. This slow resaturation may provide a source of brine to the disposal rooms.

Although parameters for undisturbed rock in the Salado were determined for the 1996 WIPP PA as accurately as possible from measured data, the properties assigned to the DRZ are conservative to account for possible future disruptive events such as earthquakes, that otherwise would have been difficult to eliminate from consideration.[46,20]

There was little interest in predicting radionuclide transport in the Salado for the first decade of site characterization. However, as discussed in Section 2.1, the higher than expected brine inflow and lower than expected Salado permeability discovered in the mid-1980's suggested that hydrogen from anoxic corrosion could attain high pressure in the repository, which could drive outward liquid flow.[189] Furthermore, the US EPA issued 40 CFR Part 191, which formally implemented the disposal system concept and the cumulative-release performance standard, in which any movement of radionuclides out of the disposal system is considered important even such movement does not threaten the biosphere.[2,3]

The low permeability of the Salado indicates that fluid flow into intact Salado rock will be slow and may not be capable of relieving pressure caused by gas generation. If the repository pressure rises to lithostatic in the repository, the rock may respond mechanically by hydrofracture. In 1992, development began of a phenomenological model for incorporating the hydraulic effects of fracturing into the BRAGFLO two-phase flow computer code.[52] Confidence in the appropriateness of the model was developed by review from an external group of rock mechanics and hydrology experts.[190] Observations taken in the Salado form the basis for values of model parameters selected for the hydrofracture model incorporated in BRAGFLO. The anhydrite interbeds in the Salado contain interconnected, partially healed, thin, subhorizontal fractures.[191] *In situ* hydraulic tests of the anhydrite interbeds demonstrate pore-pressure dependence of permeability,[173] indicating dilation of pore space, probably by the opening of pre-existing fractures, as the local effective stress changes. Liquid hydrofracture tests conducted from the repository horizon indicate pervasive alteration of the interbeds once pressure is high enough to initiate new fractures or interconnect existing fractures, accompanied by a permeability increase of about 5 orders of magnitude from initial values.[192,193]

The low permeability and high capillary pressure of halite-rich horizons indicates that lateral flow more than a few meters from the repository into the Salado can be expected to occur only in the anhydrite interbeds. A radial model for flow in the anhydrite interbeds was adopted based on a lack of regional trends in hydraulic properties suggested by the continuity of fine stratigraphic details and similarity of Salado rock fabrics and textures in the vicinity of WIPP.[5 Section 6.4,194] Since significant releases are not expected to occur via flow through the anhydrite interbeds in either their intact or fractured state, the potential for retardation in the anhydrite interbeds has not been quantified. A very simple, advection-only model for actinide transport in anhydrite interbeds is implemented in the 1996 WIPP PA.[195,196,77]

4.0 Discussion: Validating Conceptual Models Used for the CCA

Shifts in emphasis of site characterization and model-building activities occurred subtly, slowly and pervasively in the WIPP project. Early investigations focused on general characteristics and unusual features with the aim of confirming the suitability of the WIPP site from the perspective of sound scientific judgement.[e.g., 7] Early safety analyses were conducted as an accessory to and in support of sound scientific judgement as a basis for decision-making.[e.g., 35] Through the 1980's, as the national policy for regulating the disposal of nuclear waste matured, the emphasis shifted to characterization with the aim of building conceptual models for use in process-level scientific modeling and total-system PAs.

The validation of conceptual models as suitable for prediction of the long-term performance of the WIPP disposal system has been a long-standing concern. As the meaning of the term can be controversial, in this discussion "validate" is defined as "to support or corroborate on a sound or authoritative basis."[197] This definition is consistent with WIPP regulations, which state "...there will inevitably be substantial uncertainties in projecting disposal system performance. Proof of the future performance of a disposal system is not to be had in the ordinary sense of the word in situations that deal with much shorter time frames. Instead, what is required is a reasonable expectation ... that compliance will be achieved."[3, at 191.13(b)] Even though certainty is not required, building confidence in the conceptual models of hydrology and chemistry has not been simple.

Experience with WIPP has shown that confidence in conceptual models depends on documented and retrievable records of work, consistency with or improvement on standard scientific and engineering practices, and reasonable consideration of alternatives.[198] Documentation is vital because documents, or *records*, form the basis of decision-making, both in science and in regulatory proceedings. Consistency with standard scientific and engineering practice is developed and tested through technical peer review processes. Internal peer review by independent qualified peers has been a consistent part of WIPP project activities since its inception, and external peer review is acquired by publishing in refereed professional journals. Reasonable consideration of alternatives is also assessed through peer review.

An important aspect of building confidence in conceptual and mathematical models has been the use of external peer review groups. In addition to peer review as part of publishing in the scientific literature, WIPP project managers and scientists have often sought out and used external peer review groups. Two standing external review groups exist for the WIPP program. Since 1978, a National Academy of Science/National Research Council Committee on the WIPP (NAS WIPP Panel) has provided review, comment, and recommendations to the federal government and the DOE regarding development at WIPP. In its most recent of 13 reports (NAS WIPP Panel reports are summarized in 5, Chapter 9), the NAS WIPP Panel expressed confidence in the safety of the WIPP if it is not disturbed by human activities, and cautioned against excessive use of subjective scenarios of human disruptions in assessing WIPP safety.[19] The State of New Mexico has since 1978 chartered the Environmental Evaluation Group (EEG) to provide independent oversight of DOE activities pertinent to WIPP. In addition to its role in evaluating operational safety and monitoring the environmental conditions in the vicinity of WIPP, the EEG has contributed to the development of CCA conceptual models of long-term performance though its comments on project documents,[199,200] its sponsorship of workshops and symposia,[201] and independent analysis of issues of importance in evaluating long-term performance.[202,203,204] The NAS WIPP Panel and the EEG have been effective in influencing the development of WIPP conceptual models, since these organizations have been involved in the conceptual evaluation, planning, execution, and analysis of most WIPP-relevant technical activities since the late 1970's.

Other external review groups have also influenced the development of WIPP conceptual models. Perhaps the most significant are external groups that have been chartered to evaluate and guide technical activities around a particular topic or issue for a period of years. Two such groups are the 1988-1993 Performance Assessment Peer Review Panel for the system-level model of the WIPP disposal system,[205] and the 1992-1996

Geostatistics Expert Group for the use of inverse methods to infer the hydraulic properties of the Culebra.[206] In some cases, these groups participated in specific tests that were conducted to evaluate WIPP models against academic or industry approaches. For example, a test of inverse models for inferring hydrologic conditions was conducted among a subgroup of the Geostatistics Expert Group.[206]

Some external review groups were convened to evaluate the suitability of WIPP conceptual r models at specific points in their development. Two of many such groups convened for project feedback prior to the CCA are the 1993 Fracture Expert Group,[190] which evaluated the development and application of the anhydrite interbed fracture model, and the Performance Assessment Review Team, which examined the suitability of the WIPP disposal system model for predicting compliance with the EPA regulations.[207] Soon after submitting the CCA to the EPA, the DOE convened a joint IAEA/OECD panel to review the CCA from an international perspective.[208] The most important independent peer review panels of the WIPP project were convened in 1996-1997 to comply with specific EPA regulations or to support specific claims in the CCA license application,[5 Chapter 9,209] two of which are important to this discussion. The Conceptual Model Peer Review Panel examined the selection of, basis for, and mathematical implementation of all conceptual models included in the 1996 WIPP PA.[5 Chapter 9,209,98,99,100] Over a one year period, this panel examined technical documents, read previous peer review group findings and project responses to those findings, and interviewed project technical staff. To address Conceptual Model Peer Review Panel concerns, additional experiments and modeling were conducted for the MgO backfill[97] and spallings conceptual models.[210,96]

Eventually, the Conceptual Models Peer Review Panel found each conceptual model used in the 1996 WIPP PA to be acceptable for use by being:

(1) adequate;

(2) inadequate but unimportant (e.g., the explanation for spatial variability in Rustler hydraulic properties is inadequate but unimportant because the data base and transmissivity fields are appropriate to describe the existing spatial variability); or

(3) in the case of the spallings model,[95,96,210] conservative in its results.[100]

The Natural Barriers Data Qualification Peer Review Panel was convened to evaluate the adequacy of data collected prior to implementation of a quality-assurance program, satisfying the specific requirements of the EPA's 1996 40 CFR Part 194.[5 Chapter 9,209]

The WIPP project has sought feedback on its conceptual models through participation in cooperative, international technical committees. For example, the WIPP project has participated in numerous workshops and symposia on hydrology organized by the Swedish Nuclear Power Inspectorate (SKI) and/or the Nuclear Energy Agency (NEA) of the Organisation for Economic Co-operation and Development (OECD). INTRAVAL was organized by SKI to evaluate conceptual and mathematical models of groundwater flow and radionuclide transport used in PA of radioactive waste repositories. INTRAVAL focused on methods for validating conceptual and mathematical models of geophysical, geohydrological, and geochemical phenomena using test cases submitted by the participating organizations. In 1990, the WIPP project submitted the brine-flow tests from the Salado as a test case to INTRAVAL to identify mechanisms controlling brine inflow. Teams from Sandia National Laboratories, the (Dutch) National Institute of Public Health and Environmental Protection, and the (French) École Nationale Supérior de Mines de Paris participated in the WIPP test case. All teams found that flow models based on Darcy's law could replicate the experimental data from the low-permeability Salado rocks. The studies concluded that Darcy-flow models can reliably be used to predict brine flow to WIPP excavations, provided that the flow modeling is coupled with measurement and modeling of the pore-pressure field around the excavations.[211] The WIPP project also participated in the NEA/SKI GEOVAL-1990[189,212]and 1994[173] symposia and the NEA GEOTRAP project.[211,213,214,23,215] Other international working groups and symposia with WIPP participation in the areas of hydrology and chemistry include:

- the INTRAVAL Culebra Test Case, with participation from AEA Technology (UK), Universidad Politécnica de Valencia (UPV), Atomic Energy Control Board of Canada (AECB), and the Bundesanstalt für Geowissenschaften und Rohstoffe (BGR);
- the NEA workshop on Excavation Response in Geological Repositories for Radioactive Waste;[216,217]
- the NEA workshop on Heterogeneity of Groundwater Flow and Site Evaluation;[218]
- the NEA workshop on Gas Generation and Release from Radioactive Waste Repositories;[189] and
- the NEA Site Evaluation and Design of Experiments (SEDE) and Performance Assessment Advisory Group (PAAG) working groups.[219]

The most rigorous external examination of the 1996 WIPP PA conceptual and mathematical models occurred during the EPA's own review of the CCA.[6] The EPA's review not only assessed the technical credibility of data and models used to support the 1996 WIPP PA, but also tested through audits that the data and information cited by the DOE were documented and available for public review. The EPA sought out the opinions of stakeholders, in some cases soliciting comments directly from specific public stakeholder groups in private meetings. The EPA requested significant additional information and explanation of data and models used in the CCA, and conducted sensitivity studies and an independent verification test of the 1996 WIPP PA to evaluate the importance of differences of opinion between the DOE's positions in the CCA and stakeholders.[220] The record of the EPA review of the 1996 WIPP PA and CCA is available in EPA Docket A-93-02. The index to Docket A-93-02 is on the World Wide Web at www.epa.gov/radiation/wipp/.

5.0 Conclusion

The 1996 CCA used a model of the WIPP disposal system to perform a probabilistic evaluation of its performance for the next 10,000 years. The performance assessment model included conceptual and mathematical representations of key hydrologic and geochemical processes. These key processes were identified over a 22-year period involving data collection, data interpretation, computer modeling, and sensitivity studies to evaluate the importance of uncertainty and of processes that were difficult to evaluate by other means. Key developments in the area of geochemistry were the evaluation of gas-generation mechanisms in the repository; development of a model of chemical conditions in the repository and the concentration of actinides in brine; selecting MgO backfill and demonstrating its effects experimentally; and determining the chemical retardation capability of the Culebra. Key developments in the area of hydrology were evaluating the potential for dissolution of evaporite rocks in the vicinity of WIPP, development of a regional model for hydrologic conditions, development of a stochastic, probabilistic representation of hydraulic properties in the Culebra; characterization of physical transport in the Culebra; and the evaluation of brine and gas flow in the Salado. Confidence in the conceptual models used to represent these processes in PA calculations was gained through the use of various types of independent peer review in many stages of their development.

6.0 Acknowledgements

This narrative is a personal perception of events that occurred over many years. Undoubtedly, others would portray some events differently. Perhaps the referencing suffices in crediting those who contributed to the evolution of thought on these subjects. In addition to the CCA, three reports are particularly useful in understanding the evolution of events from a contemporaneous perspective and are recommended to the interested reader: the *Final Environmental Impact Statement*;[35] the *Summary Evaluation of the Waste Isolation Pilot Plant Site Suitability*;[7] and the *Systems Analysis, Long-Term*

Radionuclide Transport, and Dose Assessments, Waste Isolation Pilot Plant (WIPP), Southeastern New Mexico; March 1989.[11]

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7.0 A Note on References

Many of the references for this paper are Sandia National Laboratories internal SAND reports. Referenced SAND reports are generally available by request from the Technical Library, Sandia National Laboratories, P.O. Box 5800, Albuquerque, NM, 87185, or the National Technical Information Service. Many references for this paper are to the DOE's CCA (US DOE, 1996). The approximately 100,000 page CCA is available on the World Wide Web at www.wipp.carlsbad.nm.us through the main Library link to the Carlsbad Area Office Library. Specific chapter numbers or Appendix titles have been provided to facilitate finding the referenced information. Through the World Wide Web, Chapters 1-9 and appendices can be viewed, searched by section and with keywords, or downloaded, and other attachments can be downloaded.

Table 1. Oxidation states present in the WIPP and the application of the oxidation state analogy to determine dissolved actinide concentrations in WIPP brines.

Species in WIPP	Representative Actinide (Solubility Upper Bound)
Pu(III), Am(III)	Am(III)
Th(IV), U(IV), Np(IV), Pu(IV)	Th(IV)
Np(V)	Np(V)
U(VI)	U(VI)

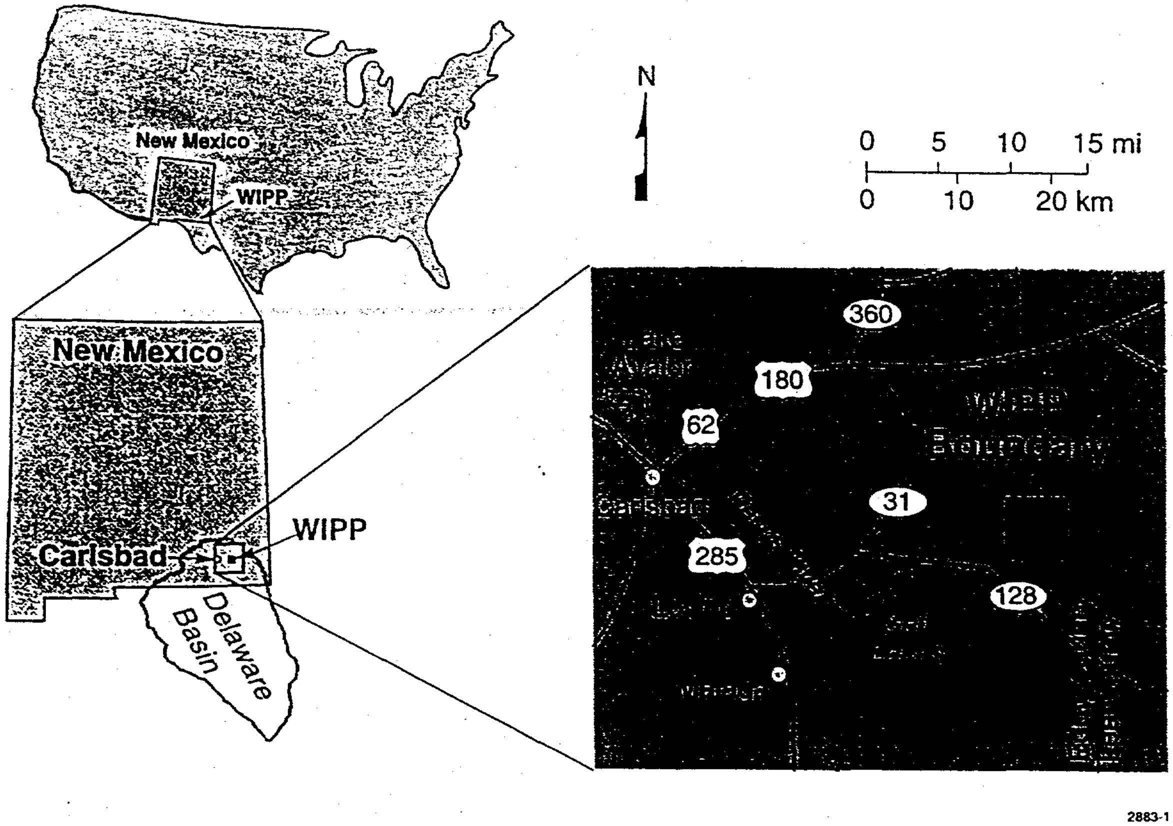


Figure 1. Location of the WIPP site in southeastern New Mexico.

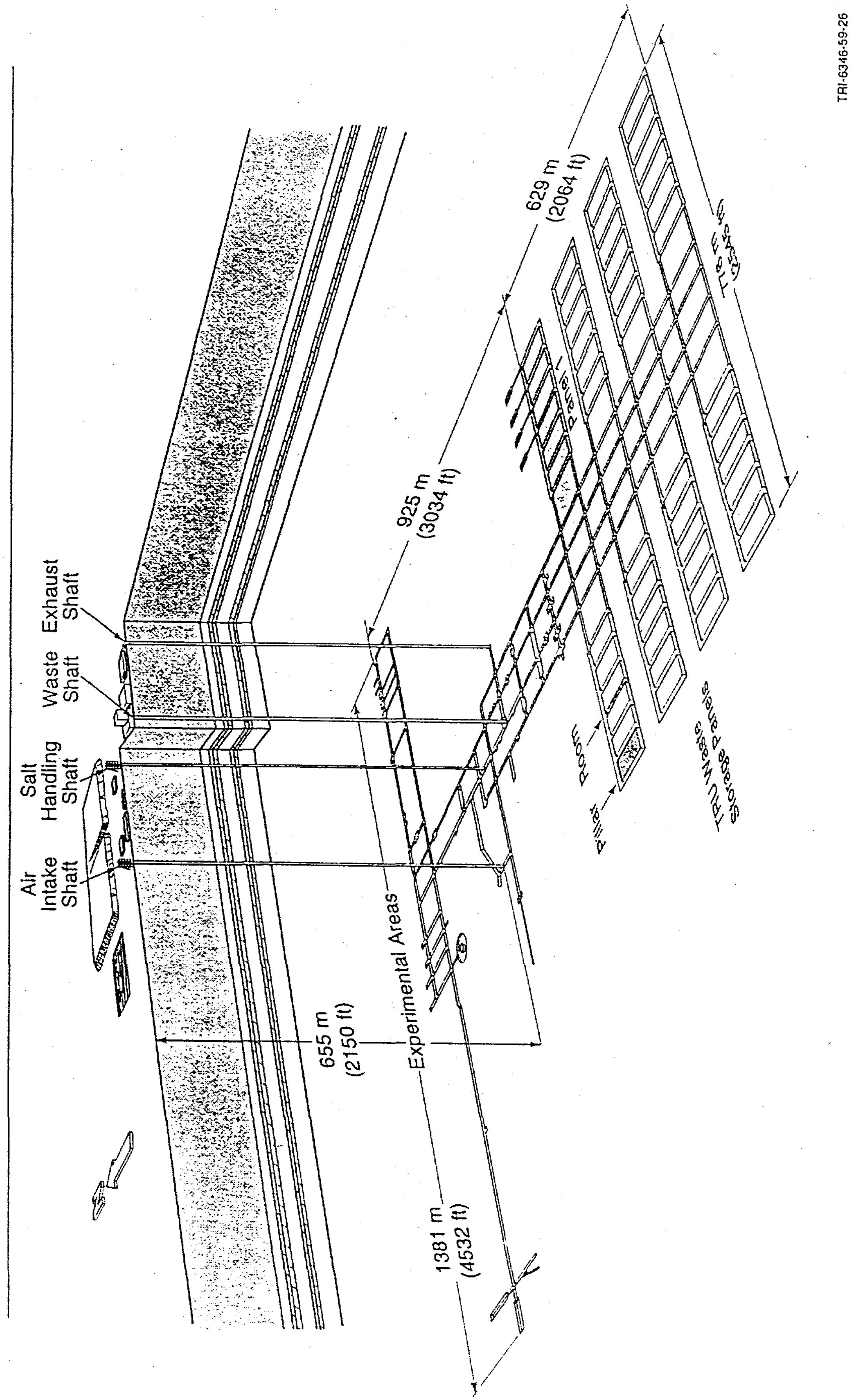


Figure 2. Cutaway diagram illustrating major features of the WIPP repository.

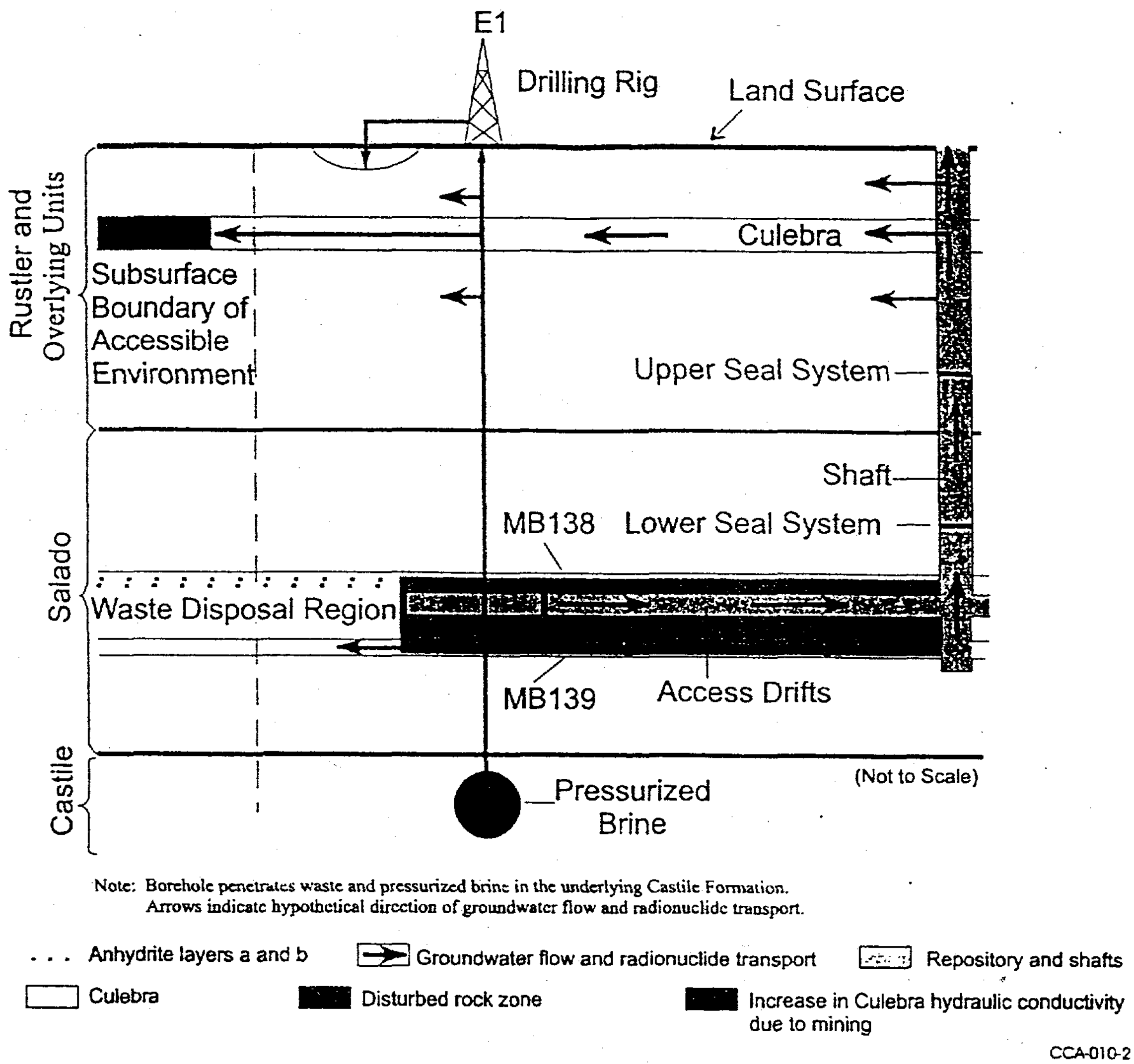


Figure 3. General features of the disturbed performance (or human intrusion) scenario for WIPP performance assessment.

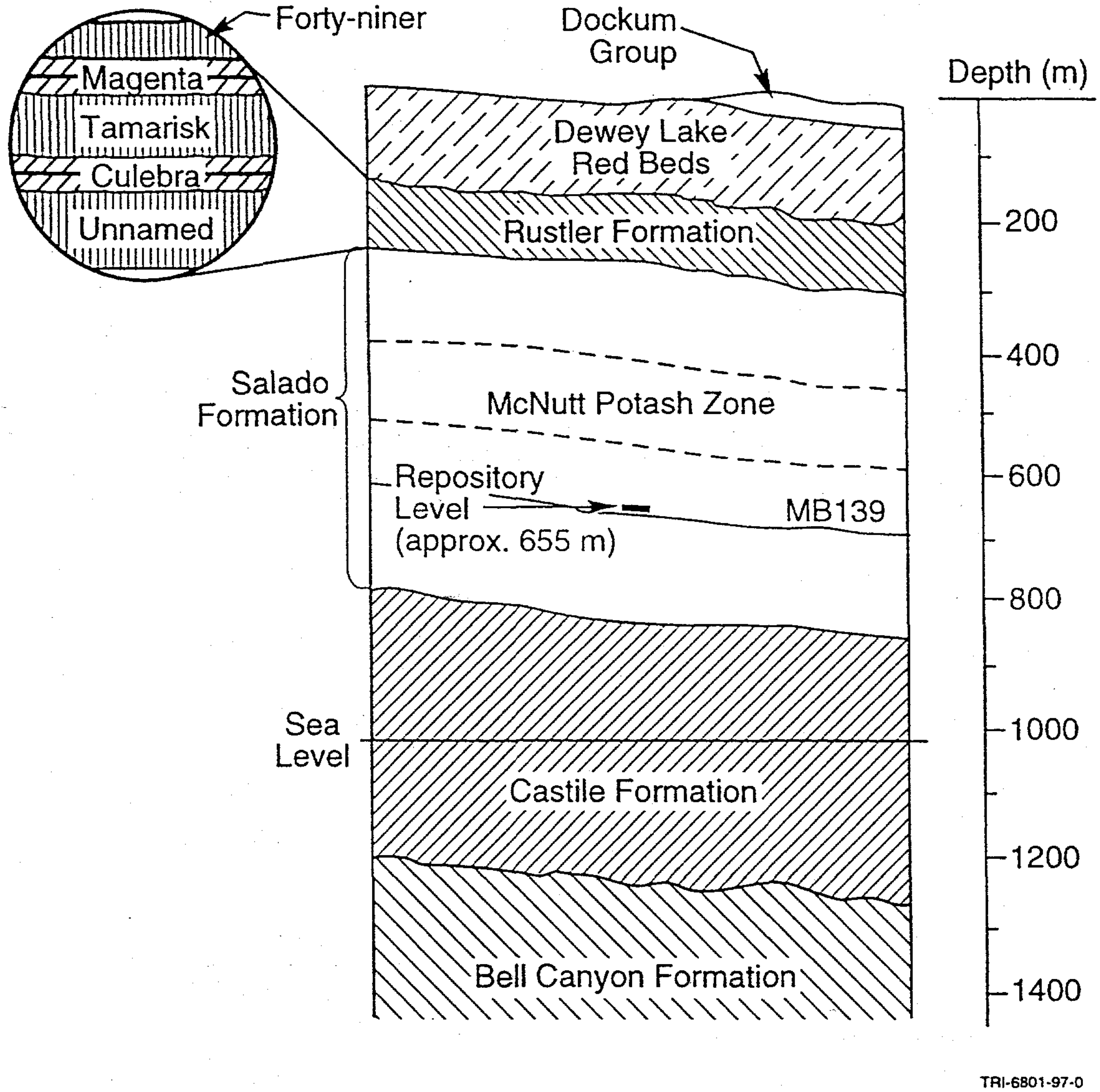


Figure 4. General stratigraphy of the WIPP site.

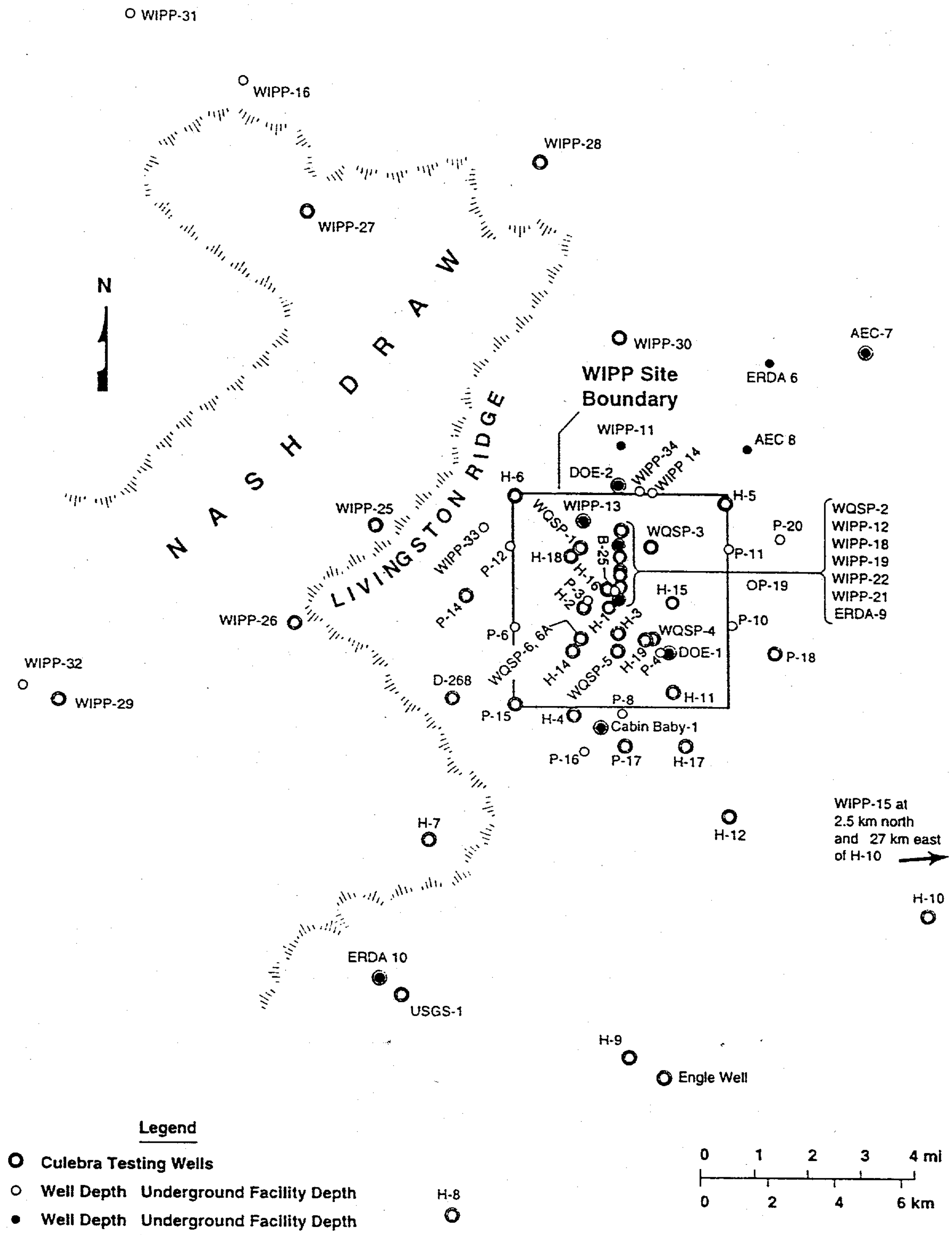


Figure 5. Location of site characterization boreholes drilled from the land surface. Filled circles indicate borehole depths less than the repository depth; open circles indicate borehole total depth greater than the repository depth.[Modified from 221]

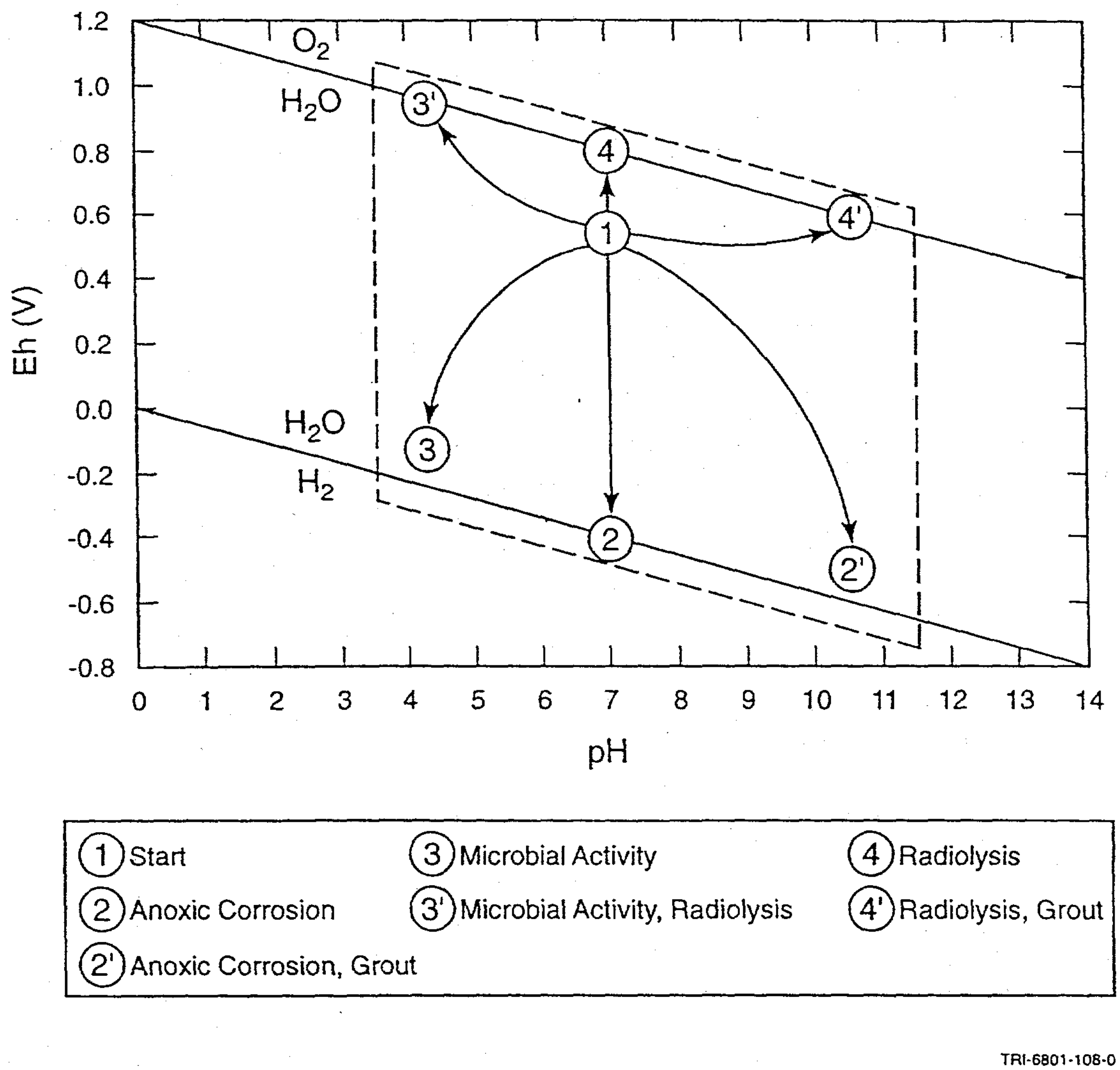


Figure 6. Uncertainty perceived from 1989 to 1995 in the range of plausible chemical conditions in WIPP repository liquids represented on an Eh-pH diagram.[61]

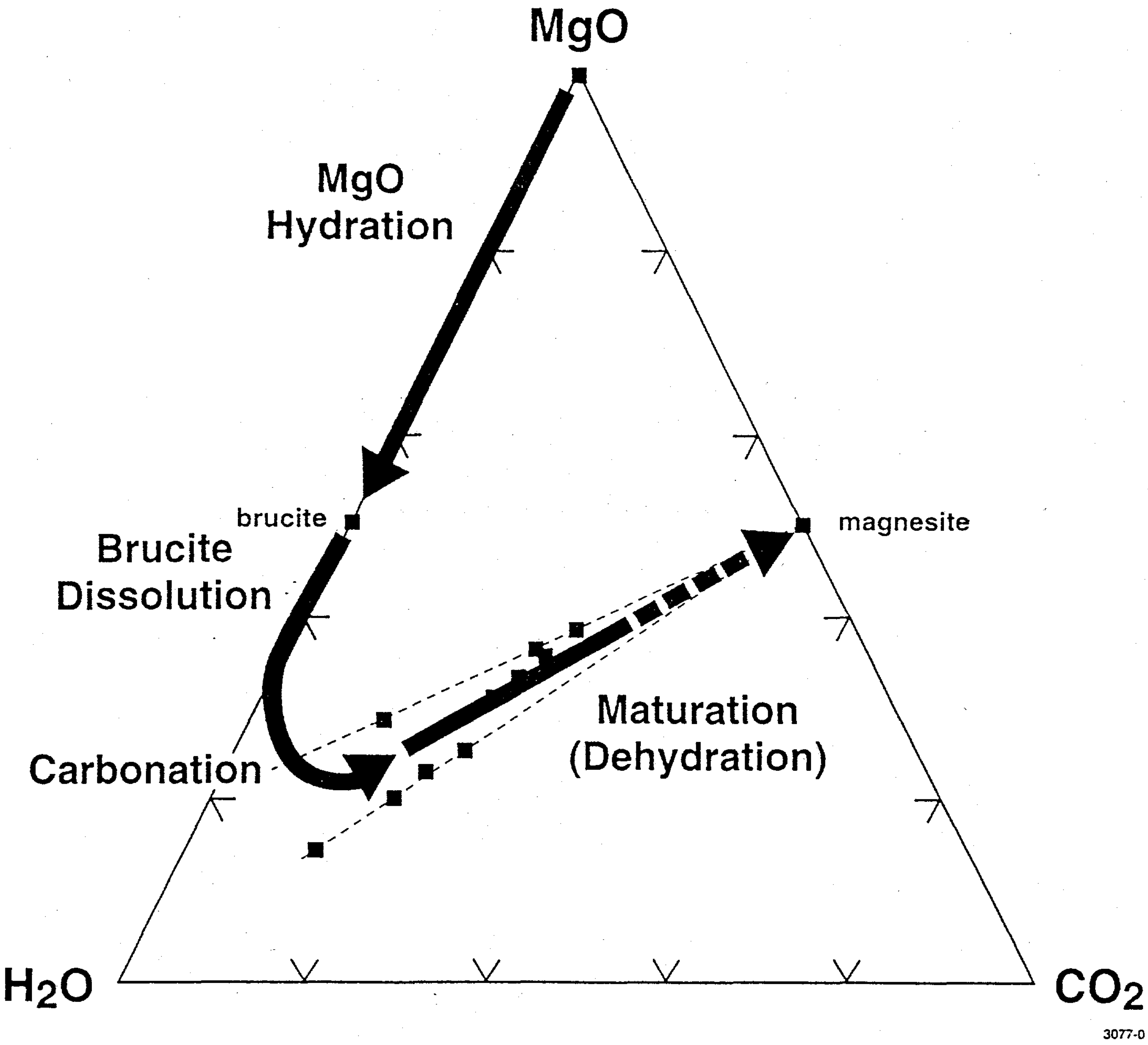


Figure 7. Ternary diagram showing steps in the reaction path for the formation of magnesite from MgO pellet backfill.[97]

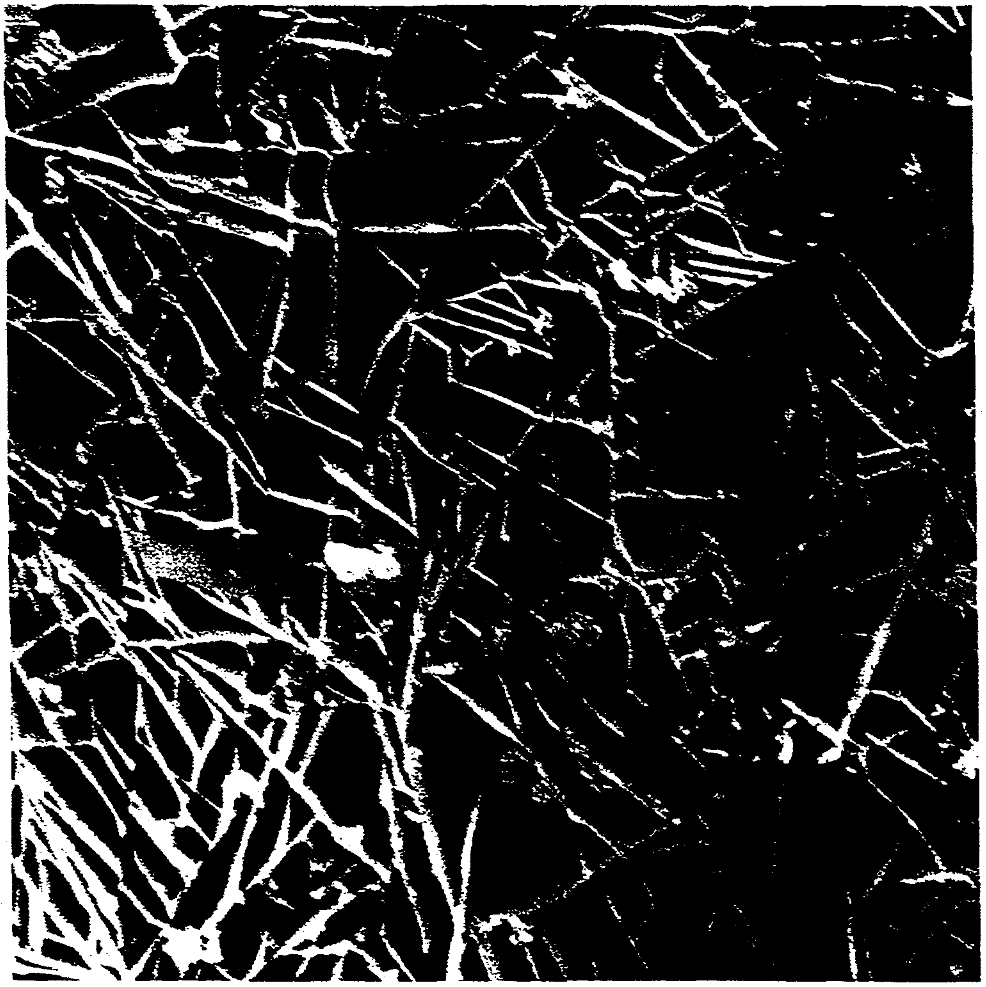


Figure 8. SEM photomicrograph of hydromagnesite precipitated from MgO pellets reacting with water and CO_2. Morphology of the crystals indicates no passivation of the interior of MgO pellets by reaction products.[97]

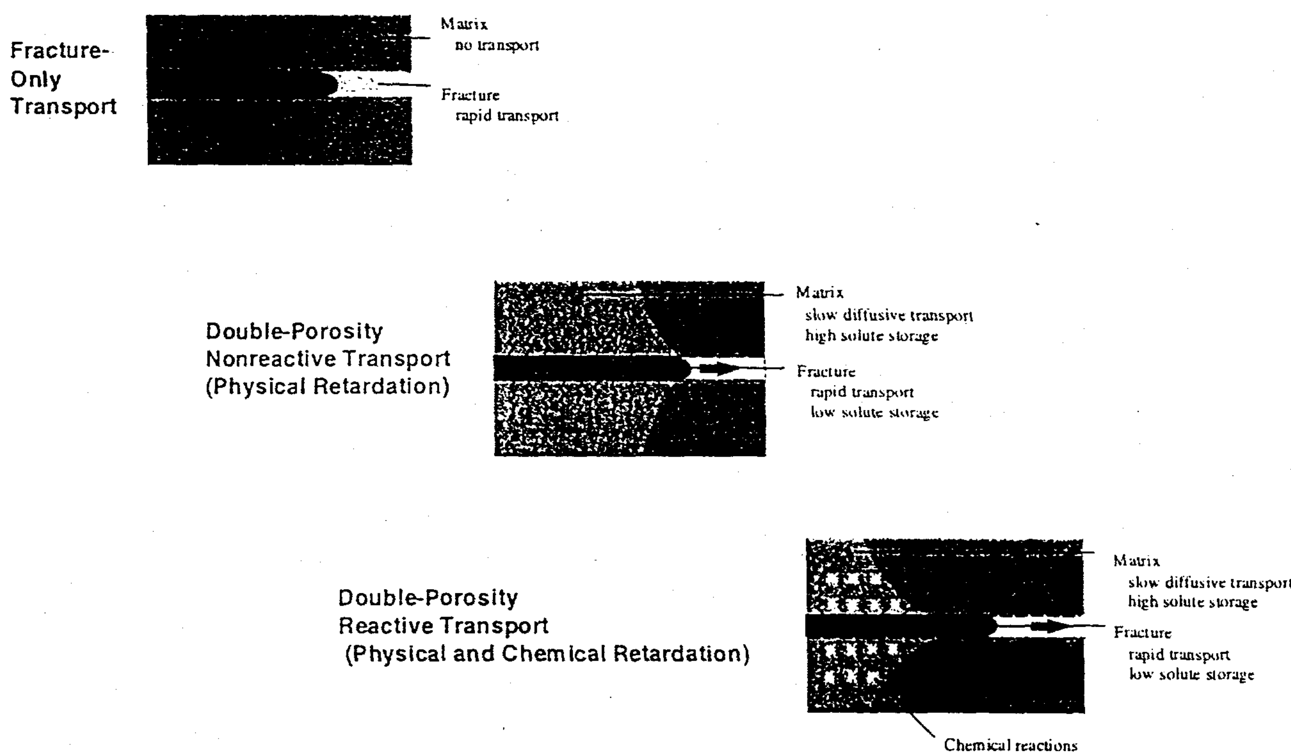


Figure 9. Perceived conceptual model uncertainty for radionuclide transport in the Culebra Dolomite during the period 1990-1994.[222]

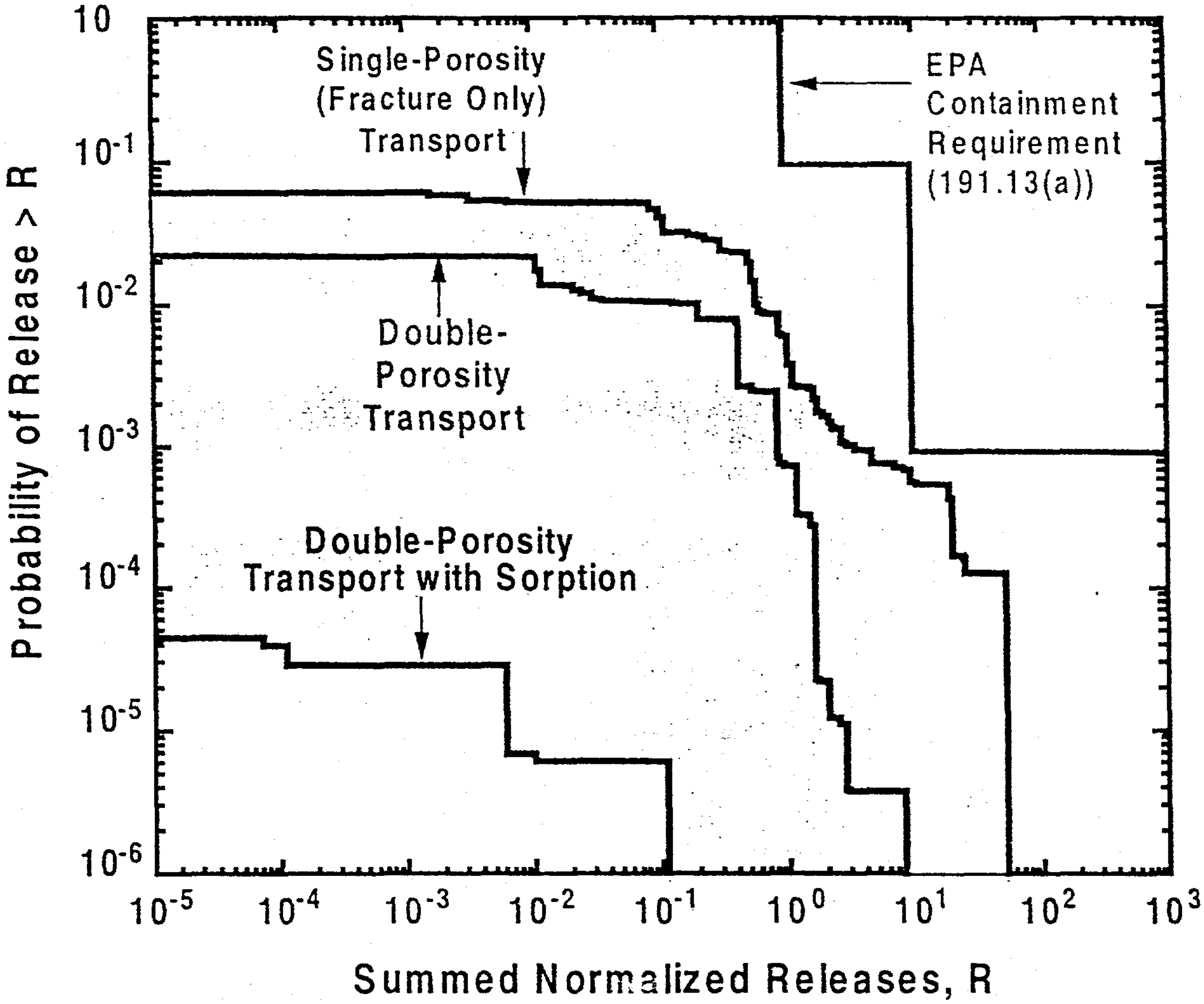


Figure 10. Sensitivity of release predictions to conceptual model uncertainty for transport in the Culebra Dolomite. Results are probabilistic and are shown in complementary cumulative distribution functions. This comparison includes only the repository-borehole-Culebra Dolomite release path. Three conceptual models are shown and compared to the EPA regulatory release limits. The effects of matrix diffusion and sorption are clearly apparent.[Modified from 148]

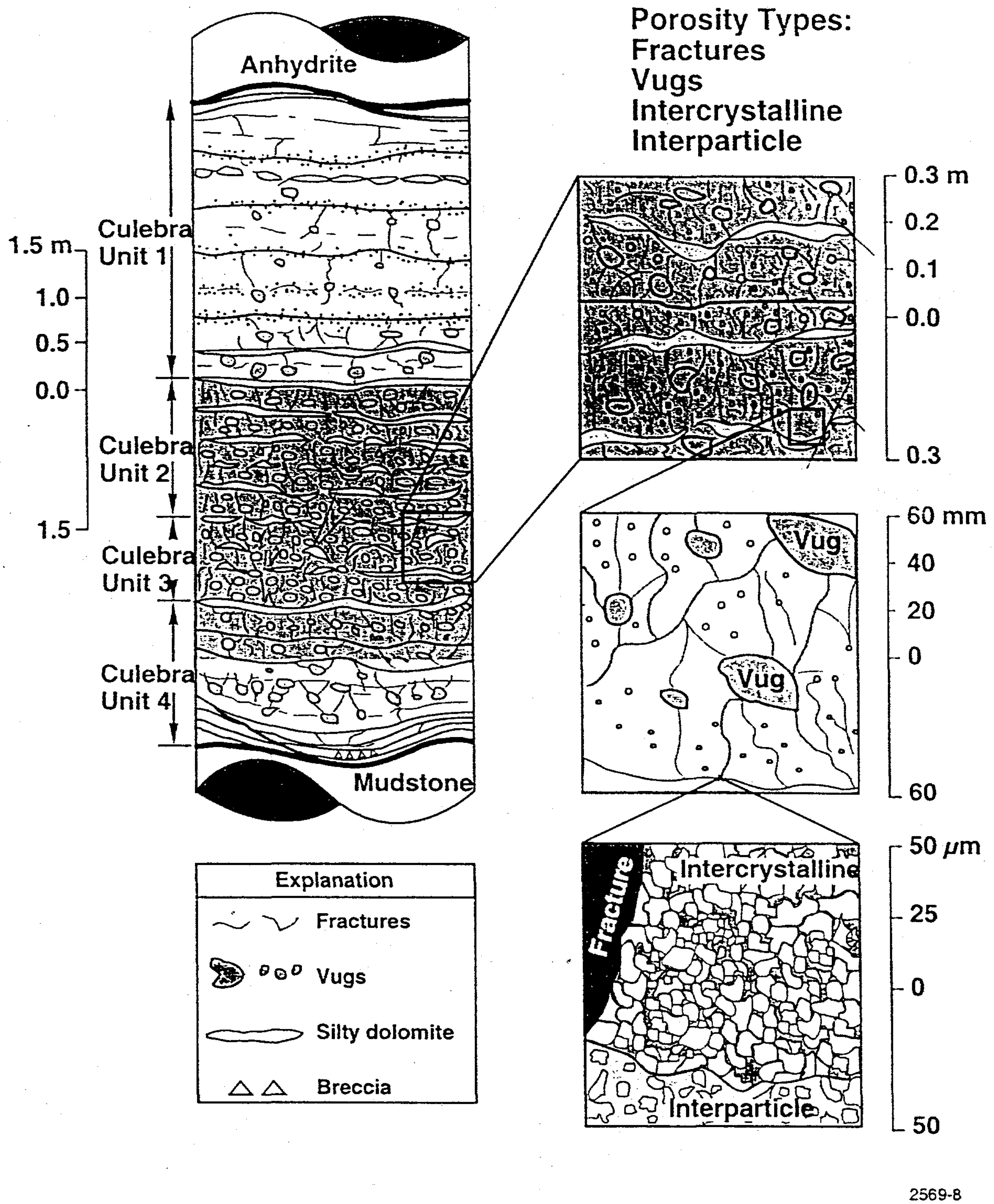


Figure 11. Conceptual representation of pore structure in the Culebra.[149]

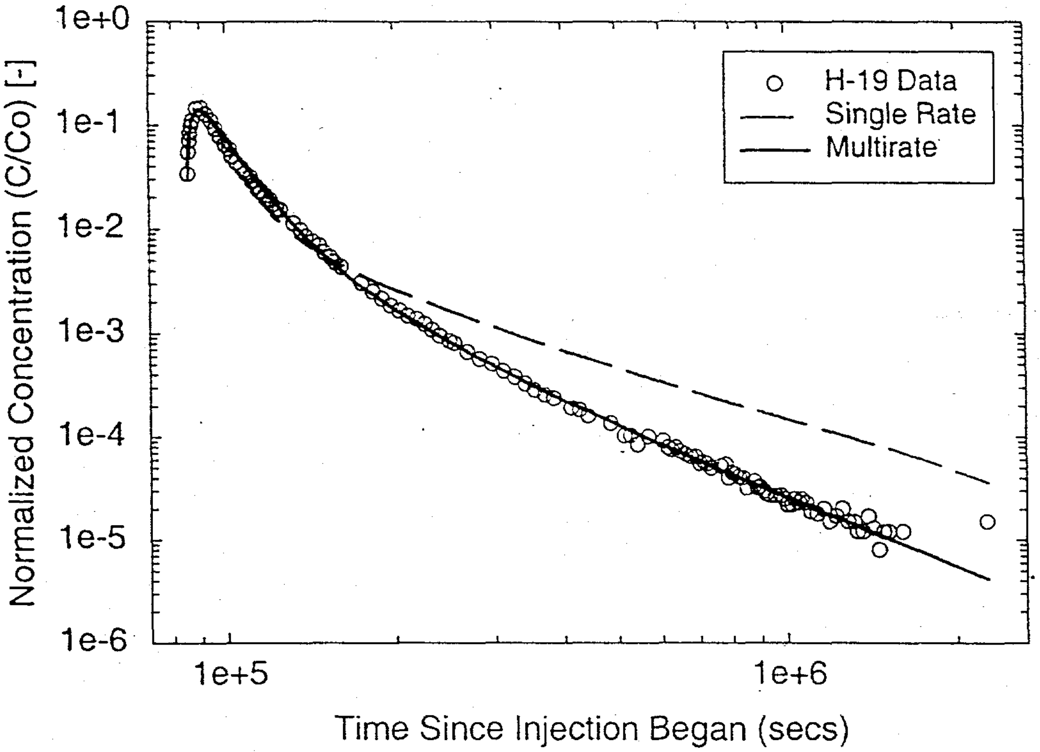
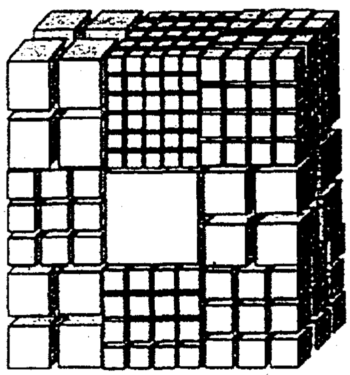
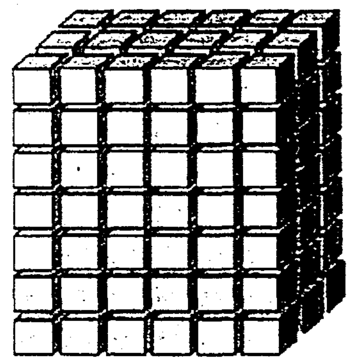


Figure 12. Analysis of the H-19 tracer test data found that double-porosity, single-rate-of-diffusion models could not match observations. The multiple-porosity, multiple-rate-of-diffusion model, which accounts for heterogeneity in matrix block size and the tortuosity of porosity, can explain the data observations.[223]



Schematic diagram of a multiple-porosity model with multiple rates of diffusion, used to interpret the H-11 and H-19 tracer tests.



Schematic diagram of a double-porosity model, used in 1996 WIPP PA calculations.

Figure 13. Double-porosity models with multiple rates of diffusion in matrix blocks are used to interpret *in situ* tests in the Culebra Dolomite. Multiple rates of diffusion are caused by heterogeneity in the size of matrix blocks and by heterogeneity in the pore structure (tortuosity) within each matrix block. Single-rate of diffusion, double-porosity models were used in the 1996 WIPP PA. Matrix block size and tortuosity were sampled randomly from a distribution derived from experimental data.[224]

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