

GEOCHEMICAL MODELS OF MOBILITY OF COMPONENTS
FROM LOW-LEVEL WASTE DISPOSAL SITES

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ABSTRACT

Accurate predictions of transport of radionuclides and other contaminants away from low-level waste disposal sites must reflect an understanding of retardation processes. This paper (1) presents the geochemical concepts of sorption and precipitation; (2) relates the geochemical processes involving the major chemical composition to the retardation of trace components, (3) presents a compartmentalized approach for determining the various retardation mechanisms dominating along the flow path; and (4) discusses the retardation behavior of the Tc-99 and I-129. Tc-99 and I-129 are long-lived isotopes associated with low-level radioactive waste that are often poorly retarded because of their anionic form. Existing data on the retardation show that Tc-99 and I-129 are retarded under specific geochemical conditions. Optimum conditions for retardation should be determined for a waste site and incorporated into the design. Retardation is determined by the set of geochemical conditions created by contaminated seepage, soil mineralogy, and groundwater chemistry. The dominant retardation processes along the flow path change as the character of the geochemical parameters vary. Precipitation and dissolution reactions of the major chemical components create substrates for specific adsorption and coprecipitation of trace components in the zone of mixing seepage with groundwater. In units where chemical steady-state is maintained, ion-exchange and physical adsorption often are the primary controls of mobility. A method for incorporating retardation into solute transport modeling uses the concentrations of trace components exiting the mixing zone as source terms and retardation coefficients for the area away from the mixing zone.

INTRODUCTION

The "Licensing Requirements for Land Disposal of Radioactive Waste," 10 CFR 61, Subpart D¹, state that the disposal site shall be capable of being characterized, modeled, analyzed, and monitored. The technical position (NUREG-0902), prepared to interpret and explain the meaning and intent of this requirement, explains that the site characteristics must be such that limited site characterization can adequately define the site characteristics spatially across the disposal site and that site characteristics should vary with a sufficiently narrow range so that the input to modeling is representative of the hydrogeologic units and the assumptions underlying the modeling are valid. In addition, Subpart D of 10 CFR 61 states that a preoperational monitoring program must provide basic environmental data on the disposal site characteristics including ecology, meteorology, climate, hydrology, geology, geochemistry, and seismology of the disposal site.

The required data and modeling are to demonstrate that reasonable assurance exists and that the performance objectives will be met by the disposal site and design. The performance objective focused upon in this paper is the "protection of the general population from releases of radioactivity." Although several pathways for potential release of radioactivity are included in this objective, retardation of radionuclides along the groundwater pathway is the subject herein.

Total isolation, although a goal of the design of a disposal site, is not possible because of practical constraints and uncertainties in the geohydrological system. Therefore, release of radioactivity via the groundwater pathway is predicted by contaminant transport models. Because of the long half lives of many of the isotopes associated with low-level waste and the nearness to the accessible environment in shallow land disposal, the low-level waste industry should borrow from the high-level waste² and uranium industry^{3,4} and consider the retardation of long-lived isotopes as important parameters in models of solute transport. Over long periods of time the uncertainties in a design for hydrologic isolation increase and, therefore, the retardation of long-lived isotopes should not be left to chance. The geochemical properties controlling retardation need to be understood, and conditions promoting retardation should be incorporated in the design process.

This paper presents conceptual models of retardation processes from geochemical principles and relates the various models to the known retardation behavior of the radionuclides specified for waste classification in 10 CFR 61. In addition, a procedure for predicting the retardation expected in the volume around the disposal site as a function of the chemical character of the contaminated seepage, the groundwater, and the minerals along the groundwater flow path is presented. The volume along the flow path is compartmentalized as a function of geochemical character to decrease the variation in retardation

along the flow path. As background for the discussion, basic information on retardation processes and the application of retardation in solute transport models are reviewed. Suggestions are made as to the data needed in the monitoring program in order to apply the geochemical model of retardation. The concepts presented are applicable to the technical requirements of a new site, as well as to the understanding of the retardation processes occurring at existing sites.

RETARDATION MECHANISMS

The primary chemical mechanisms removing trace components from groundwaters include⁵:

1. Precipitation as a pure mineral, coprecipitation, and precipitation as a solid solution.
2. Physical, electrostatic (ion-exchange), and specific adsorption, collectively termed sorption.
3. Diffusion into pores and into the solid matrix.

Because of very slow rates, diffusion is generally not important in groundwater flow systems and therefore is not considered in this paper.

Precipitation

Precipitation reactions are controlled by thermodynamic solubility constants and occur in concert with dissolution reactions. Precipitation and dissolution reactions are dependent upon the pH, Eh, and chemical activities of the reacting species in solution. Generally, precipitation and dissolution reactions involve directly only the major components in solution. The concentrations of trace components are usually less than the solubility limits for precipitates of trace components, with a few exceptions. For example, the soluble concentrations of plutonium are often controlled by the solubility of PuO_2^6 .

Precipitation and dissolution reactions, however, indirectly affect trace components by the production and destruction of minerals in the solid phase. These secondary minerals are generally effective scavengers of trace components by coprecipitation and sorption. Hydroxides and oxides of iron, manganese, aluminum, and silica are especially effective substrates for sorption.

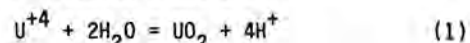
Coprecipitation and precipitation as a solid solution are the processes of incorporation of trace components into the crystal lattice of a mineral by substitution of the major elements by a trace element. These mechanisms are restricted to trace elements that are similar with respect to size and charge as the major cations in the precipitate.

Geochemical computer codes based upon thermodynamics of reactions are available to predict the precipitation and dissolution reactions that occur when two solutions of different chemical character mix. Two commonly used reaction path geochemical models are PHREEQE⁷ and EQ3/6⁸. The function of these codes are similar in that they have the option to mix two solutions and calculate the quantities of minerals transferred between the aqueous and solid phases in order to maintain a specified degree of equilibrium. From this information, the important dissolution and precipitation reactions can be determined.

Geochemical computer codes have the advantage over simplistic thermodynamic calculations because they calculate simultaneous multicomponent, multiphase equilibria; i.e., the results consider the effects of pH, Eh, ionic strength, complexing ligands, and precipitating and dissolving minerals simultaneously.

Although the numerical methods and some of the data requirements vary between PHREEQE and EQ3/6, they are both derived from speciation-solubility models that calculate the distribution of aqueous species. For example, the distribution of uranium species may include $\text{U}(\text{OH})_5^-$, U^{+4} , $\text{UO}_2(\text{CO}_3)_2^{2-}$, $\text{UO}_2(\text{OH})_2$, $\text{UO}_2(\text{SO}_4)_2^{2-}$, UO_2^{+2} , and $\text{UO}_2(\text{OH})^+$. The significance of determining the predominant species is that the sorption behavior varies for each species. For example, hydroxide complexes generally have greater sorption potentials than carbonate or sulfate complexes. Also, complexed cations such as $\text{Nb}(\text{OH})_3^{2-}$ are more susceptible to specific adsorption; whereas, free cations are typically involved in electrostatic adsorption. The advantage of specific adsorption is that it is more irreversible than electrostatic adsorption.

From the distribution of aqueous species, the degree of saturation with respect to specified minerals are calculated by PHREEQE as a saturation index. The saturation index (SI) is defined as the logarithm of the ratio of the ion activity product (IAP) of the component ions of the solid in solution to the equilibrium solubility product (K_{eq}) of the solid. For example, the SI of uraninite is calculated according to the following equations:



$$\text{IAP} = \left\{ a_{\text{U}}^{+4} \right\} \left\{ a_{\text{H}^+} \right\}^4 \quad (2)$$

$$\text{SI} = \log \frac{\text{IAP}}{K_{\text{eq}}} \quad (3)$$

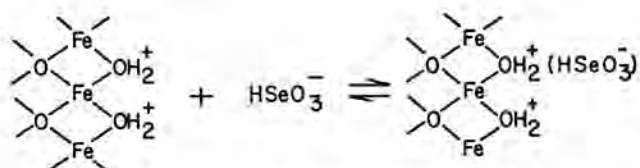
where K_{eq} is the thermodynamic solubility product of Eq. (1).

Sorption

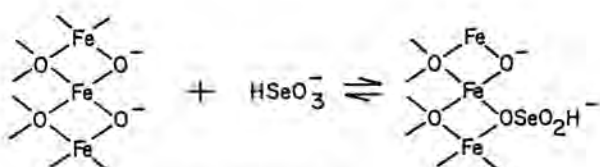
Ideally, three types of sorption can be defined, although sorption in natural systems is generally a hybrid of two or more sorption mechanisms. Sorption depends upon the charge and chemical composition of the solid phase and of the trace components in solution.

1. Physical adsorption arises from nonspecific forces of attraction between the charge distribution of the trace component and the surface of the solid substrate. Metal hydroxide complexes with neutral charge are often adsorbed by this process. This is a rapid reversible process and of little importance in retardation of trace components.
2. Electrostatic adsorption results from coulombic forces of attraction between charged solute species and the adsorbing substrate and referred to as ion-exchange. Uncomplexed cations often are adsorbed by ion-exchange. The capacity of a substrate for electrostatic adsorption of trace elements and its reversibility depend upon the competition for the sites by the major cations in solution.

3. Specific adsorption is the formation of chemical bonds between chemical groups exposed on the surface of the substrate and species in solution. This is generally an irreversible sorption process. An example of specific adsorption is the adsorption of charged metal hydroxide complexes or anionic species by ferric and manganese oxides (Fig. 1)⁹.



Electrostatic Adsorption At $\text{pH} < \text{Isoelectric Point}$



Specific Adsorption At $\text{pH} > \text{Isoelectric Point}$

Fig. 1. Sorption of anionic species by iron hydroxide at varying pH⁹.

All of the sorption mechanisms are dependent upon the chemical form of the trace components, i.e. the type of complexes, Eh and pH. The pH is a very important variable because it has strong effects on the charges of the solid surface and the aqueous species. At a near neutral pH, hydroxide complexes often are the most important; at acidic pH, other complexes such as SO_4 , F, Cl, NO_3 dominate; and at alkaline pH, carbonate complexes are often dominant. In addition, physical adsorption is highly dependent upon the ionic strength of the solution. At higher ionic strengths, major cations occupy more of the sites, leaving fewer sites for trace components.

Clay minerals are generally considered as good sorbing substrates because they have large highly charged specific surfaces. Plane surfaces of clay minerals maintain a net negative charge due to isomorphous substitution of trivalent aluminum for tetravalent silicon in the tetrahedral layer and substitution of divalent cations for aluminum in the octahedral layer. This gives rise to cation exchange sites and interlayer cations to balance the charge (cation exchange capacity). Edges of clay minerals are positively charged because of discontinuities in the XY plane. Typically, these positive charges are balanced by hydroxide ions. The net charge on the edges are a function of pH of the solution¹⁰. The isoelectric point is defined as the pH at which the edges maintain a neutral charge. Under pH conditions greater than the isoelectric point, the net charge is positive; whereas, in solutions with a pH greater than the isoelectric point, the net charge is negative. The isoelectric point of clay minerals vary with

composition and structure but generally range between 6 and 8.

Clays, therefore, can sorb trace components by exchange with interlayer cations, electrostatic sorption of cations on the plane surfaces, and specific sorption of cations and anions on the edges as a function of pH. In addition to the sorption potential, Jenne¹¹ proposed that the most significant role of clay-sized minerals in trace element retardation is as a mechanical substrate for precipitation of secondary minerals. Abundant evidence is presented relating the content of iron and aluminum hydroxides to the proportion of clay-sized particles. Manganese hydroxides are more likely to be found as discrete particles in the interstitial space. In general, the hydroxides of manganese and iron are more important than those of aluminum because the former have greater sorption capacities. The high sorption capacities of iron and manganese hydroxides result from their amorphous nature. Rapid oxidation and precipitation rates of iron promote amorphism. Recrystallization of these amorphous hydroxides will occur over time, but incorporation of trace components tends to inhibit recrystallization.

RETARDATION IN SOLUTE TRANSPORT

Solute retardation in transport models is generally modeled as sorption, expressed as a distribution coefficient defined by the linear function, $K_d = S/C$, where S (in milligrams per gram) is the total mass of the solute sorbed per unit mass of soil at equilibrium, and C is the solute concentration in the aqueous phase (in milligrams per liter). Distribution coefficients are determined by either batch or column tests¹². A trace component is added to a solution in equilibrium with a solid material. After equilibrium is reached between the trace component in the solid and solution, the ratio of the trace component in the two phases is measured.

Theoretically, distribution coefficients describe the distribution of a trace component between two immiscible phases (solid and aqueous), and are derived from thermodynamic consideration of equal chemical potentials of a trace component in both phases at equilibrium¹³. Consequently, a thermodynamic distribution coefficient is (1) independent of concentration of a trace component over the dilute range; and (2) specific to a single combination of an aqueous and solid phase at equilibrium. Distribution coefficients do not describe reactions involving precipitation because precipitation depends upon the concentration of the trace component and the solution composition, and is independent of the solid phase.

Even though distribution coefficients are conceptually sound, they rarely describe the dynamics of the geochemical reactions of groundwater/solute transport systems¹⁴. The solid media along the flow path is rarely spatially uniform and chemical disequilibrium is usually the rule because of the differences in chemical character of contaminated seepage and groundwater. As mixing occurs between seepage and groundwater, the resulting chemical disequilibrium conditions cause precipitation and dissolution reactions to occur³.

The need to incorporate precipitation, as well as adsorption, in transport models is widely recognized. Recently, there has been efforts to mathematically combine the mass action equations describing precipitation and adsorption with transport equations^{15,16,4,17}. The chemical transport model

either (1) couples existing geochemical computer codes with transport equations and iterates the mass action equations for each time step; or (2) inserts the chemical mass action equations into the transport equations and produces one set of equations. The advantages and disadvantages of these two techniques are discussed by Jennings et al¹⁵ and Miller and Benson¹⁶, with the conclusion that one set of equations describing the chemical reactions and transport is the preferred method. Narasimhan et al⁴, however, coupled the geochemical code, PHREEQ7 with transport equations to model seepage from uranium mill tailings. PHREEQ7 mixes two solutions, such as groundwater with contaminated seepage, and calculates from thermodynamic constants, the aqueous speciation and the mass transfer by precipitation-dissolution reactions. Volume elements are defined along the flow path where seepage mixes with the groundwater. The mixing proportions in each volume element are determined by the previously calculated fluid fluxes. For each volume element, chemical equilibrium is assumed and algorithms from PHREEQ7 compute the composition of the resulting solution.

Although coupling thermodynamic equations with flow equations is an important accomplishment, this approach (1) is generally restricted to situations when the aqueous concentrations of a contaminant of interest is controlled by a thermodynamic solubility constant; and (2) does not account for the interactions between sorption and precipitation.

RETARDATION AS A FUNCTION OF GEOCHEMICAL REACTIONS

Conceptual Relationship

Understanding and prediction of retardation as a function of geochemical processes is important in modeling transport of radionuclides from low-level waste disposal sites because the mechanisms that dominate retardation along the flow path away from the

disposal site vary and, therefore, retardation is not a linear function. The dominant retardation mechanism at any point along the flow path depends upon the geochemical character of the seepage, groundwater, and soil or rock. As the chemical character of these variables change along the flow path, the dominant retardation mechanisms change. Precipitation occurs in locations of chemical disequilibrium with respect to the major components. Sorption can be active in locations of disequilibrium, but often coprecipitation is the dominant mechanism controlling the concentrations of trace elements. Sorption is most important in locations where the major components in the aqueous and solid phases are in a quasi-chemical equilibrium (steady-state).

In many solute transport problems, three distinct units exist: (1) the source of contamination; (2) the zone of mixing contaminated seepage with groundwater; and (3) the local groundwater system (Fig. 2). The chemical character of seepage from the source of contamination is a function of the chemical reactions between infiltrating water and the soil cover and waste material.

The mixing zone between seepage and groundwater is typically where precipitation and dissolution reactions occur. In natural systems, a chemical equilibrium or at least a quasi-equilibrium (steady-state) exists between the water and the minerals in contact with the water^{18,19}. This implies that under conditions of chemical equilibrium, the minerals in contact with the groundwater control the composition of the groundwater. Seepage of a solution of a different chemical character into the groundwater causes a change in the chemistry of the groundwater and, consequently, disequilibrium between the groundwater and the minerals. LeChateliers principle requires chemical reactions to proceed to readjust the perturbed system toward chemical equilibrium.

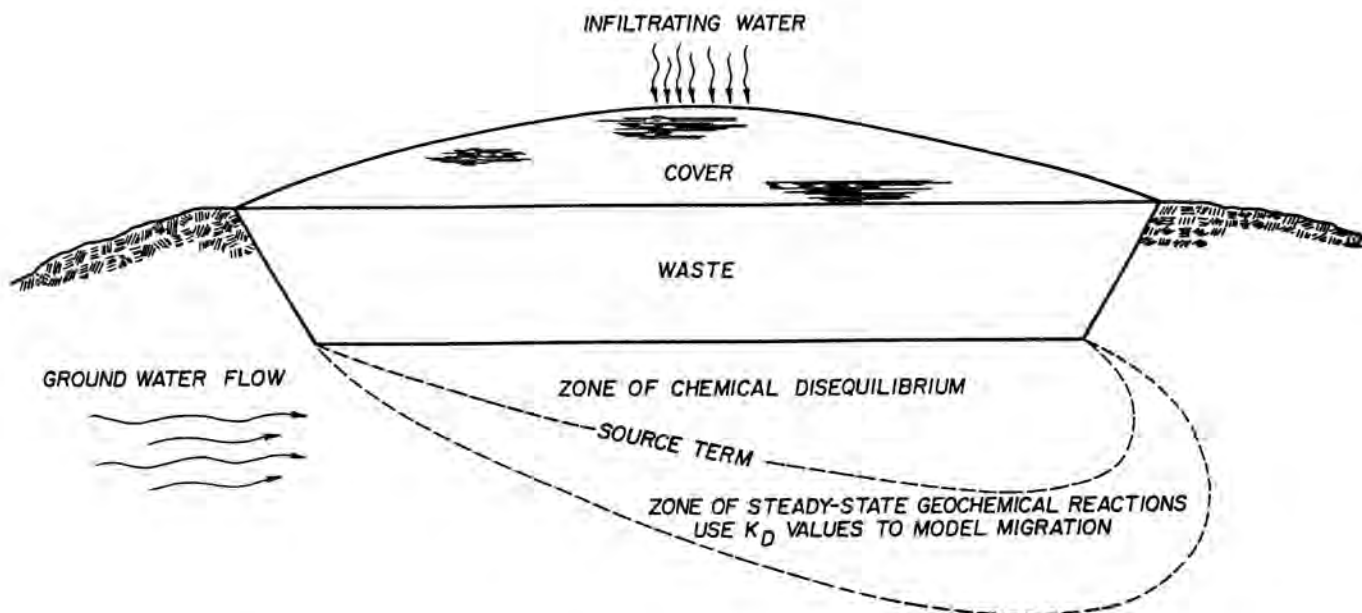


Fig. 2. Conceptual model of the interactions of the seepage from waste with the groundwater/soil system with respect to retardation processes.

The extent of reactions depend upon the difference in the chemical character of the seepage and the groundwater. Assuming rapid kinetics of the chemical reactions relative to the flow rate, the distance across the mixing zone depends upon the capacity of the soils and groundwater to react with a quantity of seepage. For example, if the pH of the seepage is less than the pH of the soil moisture, minerals such as calcite will dissolve to neutralize the acid. As the pH increases, metal hydroxides such as iron and manganese will precipitate; and if phosphates or sulfates are present, the calcium from the dissolution of calcite will be available to precipitate as gypsum or apatite minerals. These secondary minerals often provide the high sorption capacities of clay soils. The type and quantities of precipitates filling the fractures and pore space will affect the retardation of trace components, as well as the porosity of the material.

Conceptually, it is important to recognize that according to the laws of equilibrium controlling the precipitation and dissolution reactions across the interface zone, the major composition of the groundwater flowing out of the mixing zone depends primarily upon the minerals in contact with the water. This implies that the proportion of mixing is not important in controlling the concentrations of elements involved in thermodynamically controlled reactions.

The mechanisms controlling the concentrations of trace metals exiting the mixing zone vary for different elements. Trace elements that are involved in coprecipitation and solid solutions are generally dependent upon the quantity of precipitate of the major elements; whereas, trace elements retarded by sorption are dependent upon the quantity of substrate available for sorption and the chemical conditions determining the aqueous species. Only the concentration of elements that are conservative, i.e., elements that are not retarded by geochemical processes, are strictly dependent on the proportion of seepage mixing with the groundwater.

Procedure and Methodology

Prediction of the precipitates and the effects on retardation require (1) the expected chemical composition of seepage from waste material; (2) the chemical composition of groundwater; and (3) a qualitative assessment of the mineralogy along the flow path. Chemical analysis of the seepage and groundwater should include pH, Eh, major cations and anions, and radionuclides and trace metals of interest. The expected chemical composition of the seepage will undoubtedly be difficult to define because of the variability of the waste material and the solubility of the various waste forms. Laboratory leach tests of samples of waste or analysis from groundwater collection trenches or drains may provide estimates of the chemical character of the seepage. In arid regions when the site is to be located well above the saturated zone, laboratory leach tests may also be required to define the chemical character of the groundwater.

The precipitation and dissolution reactions of the major components resulting from the mixing of seepage with groundwater and soils can be predicted by geochemical codes such as PHREEQE. The supersaturation of iron, manganese, and aluminum hydroxides indicate that effective substrates for sorption will be present. Supersaturation of minerals with calcium are important in the removal of strontium from solution. Dissolution of carbonate minerals indicate that carbonate minerals are needed for pH adjustment from

the seepage to groundwater or from the acidity produced by precipitation of hydroxide minerals.

In addition, the degree of saturation (saturation index) with respect to the precipitates of trace components are calculated by geochemical computer codes. If minerals of trace metals are supersaturated, the solubility of the minerals should be considered as controlling the concentrations of trace components exiting the mixing zone, which serve as source terms for solute transport. Sorption of trace components that are undersaturated with respect to precipitates in the mixing zone must consider the substrates formed by precipitation reactions in developing source terms for the model.

Also, geochemical computer codes predict the aqueous species of the trace components and the chemical character of the groundwater in steady-state with the soils exiting the mixing zone. The aqueous species and the chemical character of the groundwater should be used to select representative distribution coefficients to model the solute transport in the groundwater system. In the volume downgradient from the mixing zone, few geochemical reactions are expected and the probability of a single coefficient accurately describing the retardation processes increases.

APPLICATION TO A MODEL OF A DISPOSAL SITE

The design of a shallow land disposal site typically includes trenches several tens of feet deep lined with low permeability material. The base of the trenches are either in the B or C horizons of the soil profile or in weathered bedrock. Waste, converted to a solid material and contained in mechanically stable packaging, is emplaced in the trenches and covered to minimize infiltration of water. Historically, problems with soil covers have produced surface subsidence and cracking, leading to water infiltrating the waste²⁰.

Typically, the composition of low-level waste includes rags, paper, metallic materials, sludges and corrosion products, detergents, resins, and organics²¹. In addition, specific types of reactor wastes are enriched in boric and nitric acids and sodium sulfates²². The chemical form of the waste is highly dependent upon treatment used for volume reduction and stabilization. Contaminated seepage from the trenches results from infiltration of meteoric water and dissolution of certain components from the waste. Although the solubility of the waste depends upon the chemical form of the packaging and waste material, the presence of sodium salts, acids, corrosion sludges, and detergents create a potential for a slightly acidic high salt seepage containing metals, phosphates, and other complexing anions.

Based upon the half-lives of the isotopes specified in 10 CFR 61 to classify low-level wastes (Table I), C-14, Ni-59, Nb-94, Tc-99, and I-129 require the greatest concern for retardation. Hydrologic isolation for the hazardous life of isotopes with shorter half lives can be modeled and designed with greater certainty. Nickel and niobium generally exist as cationic species and a nearly ubiquitous sorption of nickel and niobium have been shown¹². The distribution of C-14 depends upon the solubility of the organic compounds containing the carbon isotope. The isotopes of technetium and iodine are often poorly retarded because their anionic forms are generally not sorbed. The chemical conditions that determine the retardation of Tc-99 and I-129 are discussed below. Existing data on retardation of

TABLE I

Half-Lives of Isotopes Specified in 10 CFR 61
to Classify Low-Level Radioactive Waste

C-14	5740 Years
Ni-59	8×10^4 Years
Nb-94	2×10^4 Years
Tc-99	2.12×10^5 Years
I-129	1.7×10^7 Years
Pu-241	13.2 Years
Cm-242	163 Days
H-3	12.26 Years
Co-60	5.26 Years
Ni-63	92 Years
Sr-90	28.1 Years
Cs-137	30.23 Years

these isotopes show that these anionic isotopes can be effectively retarded under the proper geochemical conditions. Iron hydroxide and organic material are effective substrates for sorption at a pH less than the isoelectric point to create a positively charged surface and, thus, anion adsorber. An ideal case of retardation of Tc-99 and I-129 is shown by studies on the Oklo reactor site. In the two billion years since the reactor was operating, these isotopes migrated about 10 meters through partially saturated sedimentary rocks²³.

Technetium-99

Technetium exists in many valence states, the most stable being TcO_4^- . The chemical reactivity of TcO_4^- is very similar to MoO_4^{2-} . Except for very reducing sulfide environments where Tc_2S_7 may form, TcO_4^- is quite soluble. The sorption of TcO_4^- increases with increasing organic content of soils, being the greatest under slightly acidic pH conditions⁶. The sorption is lowered by bicarbonate in solution, because of the competition for positively charged sorption sites. Iron hydroxides has been shown to be an effective scavenger of TcO_4^- . The mechanism is probably specific adsorption as illustrated in Fig. 1 for selenium.

Iodine-129

Iodine exists as I^- in near neutral to acidic solutions and as IO_3^- in oxidizing alkaline solutions. Iodine is not a strong complexing agent for cations. Precipitates of iodine are quite soluble, except for salt of Pb, Sb, As, Hg, and Ba. Laboratory and field studies summarized by Ames and Rai⁶ demonstrate that the sorption of iodine is often positively correlated with organic content in soils, and inversely correlated to pH. At mildly acidic pH conditions, the edges of clays and hydroxide precipitates have greater positive charges and, thus, are stronger anion sorbers. In addition, I^- is effectively sorbed by iron hydroxides.

CONCLUSIONS

Uncertainty of hydrologic isolation increases with the time required for isolation and, therefore, retardation of long-lived isotopes associated with low-level radioactive waste must be considered in solute transport modeling. Two isotopes with long half lives, Tc-99 and I-129, are especially difficult to retard because they exist as anionic species. Retardation of anions, however, is possible under the correct set of geochemical conditions. Existing data indicate that pH, the precipitation of iron hydroxides, and organic material in the soils are important in controlling the sorption of Tc-99 and I-129. The resulting pH and the potential for iron hydroxide upon mixing seepage with groundwater can be modeled by geochemical computer codes such as PHREEQE. With this information, a qualitative assessment of the retardation potential for I-129 and Tc-99 can be made and the disposal site designed accordingly.

The geochemical conditions below a disposal facility and, thus the retardation potential, depend upon the chemistry of the soil and groundwater and on the chemical environment resulting from mixing seepage with the soil/groundwater system. Thermodynamic geochemical models are available to predict the chemical reactions and chemical conditions that occur at the interface of the waste seepage and soil. If the waste seepage is slightly acidic and the soils contain carbonate minerals to neutralize the acid, iron hydroxide generally precipitates. This serves as a very effective scavenger for anionic species of trace metals and may control the concentration of radionuclides available for further transport (i.e., source terms). The retardation of the radionuclides along the flow path away from the mixing zone is controlled by other sorption mechanisms. The probability of a chemical steady-state along the flow path increases when the mixing zone is not included. Therefore, distribution coefficients determined are more likely to describe the retardation of trace components.

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