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OCEAN FLOOR SEDIMENT AS A REPOSITORY BARRIER: COMPARATIVE
DIFFUSION DATA FOR SELECTED RADIONUCLIDES IN SEDIMENTS
FROM THE ATLANTIC AND PACIFIC OCEANS*

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ABSTRACT

Effective diffusion coefficients for selected radionuclides have been measured in ocean floor sediments to provide data for the assessment of barrier effectiveness in subseabed repositories for nuclear waste. The sediments tested include illite-rich and smectite-rich red clays from the mid-plate gyre region of the Pacific Ocean, reducing sediment from the continental shelf of the northwest coast of North America, and Atlantic Ocean sediments from the Southern Nares Abyssal Plain and the Great Meteor East region. Results show extremely small effective diffusion coefficients with values less than $10^{-14} \text{ m}^2 \text{ s}^{-1}$ for plutonium, americium, curium, thorium, and tin. Radionuclides with high diffusion coefficients of approximately $10^{-10} \text{ m}^2 \text{ s}^{-1}$ include the anionic species pertechnetate, TeO_4^- , iodide, I^- , and selenite, SeO_3^{2-} . Uranyl(VI) and neptunyl(V) ions, which are stable in solution, have diffusion coefficients around $10^{-12} \text{ m}^2 \text{ s}^{-1}$. The diffusion behavior of most radionuclides is similar in the oxygenated Pacific sediments and in the anoxic sediments from the Atlantic. An exception is neptunium, which is immobilized by Great Meteor East sediment, but has high mobility in Southern Nares Abyssal Plain sediment. Under stagnant conditions a 30 m thick sediment layer forms an effective geologic barrier isolating radionuclides in a subseabed repository from the biosphere.

INTRODUCTION

The possibility of isolating high-level nuclear waste in the sediment overlying the base rock of the deep oceans has been the subject of research for more than a decade.¹ An obvious advantage of subseabed disposal of nuclear waste is the fact that the 60% of the earth's surface covered by water need no longer be excluded in the search for a potential repository site. Furthermore, geologically stable sediment formations can be found in the central regions of major oceans that are poor in resources and are long distances away from populated land areas. Establishment of a repository at great depth would not interfere with the sole normal commercial use of these regions of the sea, which is navigation.

The ongoing effort of U. S. Subseabed Disposal Program is aimed at the demonstration of the appropriateness and practicality of the subseabed disposal concept.² In a subseabed repository the waste, contained in

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disposal concept.² In a subseabed repository the waste, contained in suitable canisters, will be embedded in the sediment at a depth no less than 30 m below the surface. This feature sets the subseabed disposal concept off against the practice of "dumping", which involves placement of the waste material on top of the sediment at the bottom of the water column. Such dumping contravenes international convention.³

The waste in a subseabed repository will be contained in canisters that meet the requirement of maintaining their integrity for 1000 years, which has been set for land-based repositories.⁴ However, in the water-saturated environment of the ocean floor sediment corrosion will take place and will ultimately expose the waste form to leaching. When this happens, the sediment overlying the repository must be relied upon to isolate the radioactive material from the biosphere. It is therefore important to demonstrate the effectiveness of the sediment as an adequate geologic barrier.

In a number of previous reports the results of diffusion tests with selected important radionuclides in several types of sediments from the mid-plate gyre region of the Pacific Ocean have been described.⁵ In the present paper, new data obtained in two types of sediment from the Atlantic Ocean are added and compared with the earlier results. As will be shown in the following sections, all existing data point to the sediment column as being a thoroughly effective barrier for the most hazardous transuranium elements plutonium and americium, and an adequate medium for the reduction of radionuclide release even in cases of more mobile species, such as technetium and neptunium.

EXPERIMENTAL

The laboratory diffusion tests described here were carried out with five different types of sediments, two from the Atlantic Ocean and three from the Pacific. The Atlantic Ocean sediments will be referred to as SNAP and GME to indicate their respective origins, the Southern Nares Abyssal Plain, and the Great Meteor East location*. The SNAP sediment was obtained from a core taken at a point 22°54.1' N and 63°29.4' W. It consisted chiefly of clay minerals (70-90%) and sand, with minor components of calcium carbonate (1-5%) and organic carbon (0.3%). The GME sediment came from a point 30°40.9' N and 24°58.9' W. Its composition was 45-50% calcium carbonate, 10-30% clay, 10-30% sand, and 1-1.5% organic carbon.

The three sediments from the Pacific Ocean included two types of red clays, and a reducing sediment. The red clays were collected in the Pacific MPG I study area, which is centered about a point 31°30' N and 158°W.⁶ These sediments are not reducing in nature. They are made up of a mixture of clay minerals, hydrous oxides of iron and magnesium, and

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sea water. For the diffusion experiments a sediment containing the clay mineral illite, and one containing smectite were selected. These materials will be referred to as "Pacific smectite" and "Pacific illite", respectively. A detailed mineralogical description of the red clays from MPG I can be found in reference 6.

The reducing sediment from the Pacific stemmed from the shelf off the northwest coast of the United States. The reducing quality is attributed to biological activity, leading to the presence of organic carbon, sulfides and ferrous iron.

In order to preserve a reducing environment during sample handling and storage, the Atlantic Ocean sediments and the Pacific reducing sediment were handled in an atmosphere of nitrogen from which oxygen was excluded to the extent that the oxygen content never exceeded 50 parts per million (0.00005 atm).

The experimental technique applied to the determination of effective diffusion coefficients for tracer radionuclides in the sediment has been described in a previous publication.⁷ The movement by diffusion of substances dissolved in the interstitial sea water of the sediment follows Fick's law. Since the diffusing species are present at very low concentrations, the diffusion coefficient has a constant value. The numbers obtained from concentration curves observed in the bulk sediment, however, are not directly comparable to self-diffusion coefficients measured in homogeneous solution. In the two-phase system of the sediment the actual distance traveled by dissolved species in the interstitial channels is larger than the outside measurement of distance indicates. Both, the effects of tortuosity of diffusion path, and the extreme narrowness of the channels, will in general slow the rate of diffusion. Furthermore, sorptive interaction of dissolved ions with the solid mineral phases, generally expressed by distribution coefficients, K_d , has a profound influence on the diffusion process.⁸ To indicate that the coefficients reported here include the rate-reducing effects of tortuosity, channel size, and sorption, they are referred to as "effective" diffusion coefficients. These numbers, however, are descriptive of the overall diffusive spreading of the radionuclides, and can therefore be used for the assessment of the barrier effectiveness of the sediment.

The effective diffusion coefficients are evaluated from concentration-distance curves determined experimentally. A schematic representation of the technique is shown in Fig. 1. The diffusion samples are in the shape of small right cylinders with a volume of 2 cm^3 each. They are prepared by joining two equal-sized sections of the sediment intimately at a plane cross-section (No. 1 in Fig 1). The right-hand half section of the sample contains the appropriate tracer nuclide admixed in uniform concentration, c_0 . Samples of this type are conveniently made up in plastic disposable syringes of 10 cm^3 volume, the tips of which have been cut off. The plungers serve as a practical means of controlling sediment volume during preparation.

Fig. 1. Schematic representation of the technique used to determine concentration - distance curves and effective diffusion coefficients. (For an explanation of the numbers refer to the description in the experimental section. "I" and "II" in part C indicate concentration - distance curves before and after diffusion, respectively.)

The samples are stored for appropriate time periods to permit diffusion of the tracer nuclides. The concentration-distance function developed during storage time is determined by analyzing the activity of the radionuclide as a function of distance measured along the axis of the sample cylinder. This is achieved by smearing the sediment in crayon-like fashion on long strips of filter paper impregnated with a binder solution (No. 2 of Fig. 1). A section of thickness Δx results in a sediment streak of length Δl , in which the distance along the paper strip is correlated to the distance in the sample (No. 3 in Fig. 1). The strips are divided into small sections for assay of activity, each section yielding a data point on the concentration-distance curve (No. 4 and part C, Fig. 1). Specific activities on a relative scale are established by obtaining the amount of sediment on a paper section conductometrically.

Desirable isotopes for these tracer diffusion experiments are those whose decay is associated with the emission of gamma or X-radiation, and whose half-lives are in the 2 month to 2 year range. This choice eliminates the need for chemical isolation of the radionuclide from the sediment before assay, since the electromagnetic radiation is not significantly attenuated by the sample material and can be measured with scintillation counting equipment.

RESULTS

A summary listing of effective diffusion coefficients for eleven different radionuclides in sediments from the Atlantic as well as from the Pacific Ocean is given in Table I. The numbers in the last column of the table were obtained from the observed variation of the radionuclide concentration with distance after allowing diffusion to occur. Specific activities of the tracer nuclides in the individual samples were evaluated as described in the preceding section, and plotted versus distance. Typical plots of this type are shown in Figs. 2, 3, and 4.

The variation of the concentration with distance in cylindrical samples of the geometry chosen for the diffusion experiments can be calculated from the solution of the diffusion equation for the case of linear bounded diffusion at a given value of the product of diffusion coefficient and time, $D \cdot t$.⁹ This solution is in the form of a series given by:

$$\frac{C}{C_0} = 0.5 + \frac{2}{\pi} \sum_{n=1}^{\infty} \left\{ \frac{1}{2n-1} \exp \left[-D \cdot t \left(\frac{(2n-1)\pi}{h} \right)^2 \right] \right. \\ \left. \left[\cos (2n-1)\pi \cdot \left(1 - \frac{x}{h} \right) \right] \sin \left[\frac{(2n-1)}{2} \pi \right] \right\}. \quad (1)$$

The best value of $D \cdot t$ in a particular case was determined by numerical computation of the function Eq. 1, and by varying the product $D \cdot t$ incrementally until the minimum value for the variance of the experimental data points was obtained.

Figure 2, which shows the experimental data for the diffusion of pertechnetate in Atlantic SNAP sediment after a diffusion time of 2.05 hours,

TABLE I. Effective Diffusion Coefficients of Selected Radionuclides in Sediments from the Atlantic and Pacific Oceans.

Nuclide	Sediment	No. of Samples	Time (d. = days) (h = hours)	Effective Diffusion Coefficient ^a D_{eff}/m^2s^{-1}
^{237}Pu	Atlantic SNAP	3	60.82 d	$< 1.8 \cdot 10^{-14}$
(PuCl_4 ,	Atlantic GME	3	60.85 d	$< 1.6 \cdot 10^{-14}$
ox.st. +4)	Pacific smectite	4	53.7 d	$< 1.0 \cdot 10^{-14}$
	Pacific illite	6	119.9 d	$< 1.2 \cdot 10^{-14}$
^{237}Pu	Pacific smectite	1	60.0 d	$< 1.9 \cdot 10^{-15}$
(PuO_2Cl_2 ,				
ox.st. +6)				
^{241}Am	Atlantic SNAP	3	71.95 d	$< 8.0 \cdot 10^{-14}$
(AmCl_3 ,	Atlantic GME	3	71.98 d	$< 1.2 \cdot 10^{-14}$
ox.st. +3)	Pacific smectite	1	35.9 d	$< 1.0 \cdot 10^{-14}$
	Pacific illite	2	240.1 d	$< 1.0 \cdot 10^{-14}$
^{235}Np ,	Atlantic SNAP	4	53.02 d	$(3.3 \pm 1.7) \cdot 10^{-12b}$
^{237}Np	Atlantic GME	4	53.05 d	$< 1.9 \cdot 10^{-14}$
(NpO_2Cl ,	Pacific smectite	2	46.2 d	$(2.0 \pm 1.2) \cdot 10^{-12}$
ox.st. +5)	Pacific reducing	2	556.2 d	$(1.4 \pm 1.2) \cdot 10^{-12}$
^{243}Cm	Atlantic SNAP	3	108.0 d	$< 1.0 \cdot 10^{-14}$
(CmCl_3 ,	Atlantic GME	3	108.0 d	$< 1.0 \cdot 10^{-14}$
ox.st. +3)	Pacific smectite	2	47.9 d	$< 2.2 \cdot 10^{-15}$
^{95m}Tc	Atlantic SNAP	4	9.3 d	$(3.2 \pm 1.5) \cdot 10^{-11}$
(NH_4TcO_4 ,	Atlantic GME	4	9.32 d	$(2.3 \pm 1.5) \cdot 10^{-11}$
ox.st. +7)	Pacific smectite	2	17.4 d	$(3.0 \pm 1.5) \cdot 10^{-10}$
	Pacific reducing	2	65.1 d	$(3.0 \pm 1.5) \cdot 10^{-10}$
^{99}Tc	Pacific reducing	2	231.2 d	$(3.0 \pm 1.5) \cdot 10^{-10c}$

TABLE I. Continued.

Nuclide	Sediment	No. of Samples	Time (d = days) (h = hours)	Effective Diffusion Coefficient ^a $D_{\text{eff}}/\text{m}^2\text{s}^{-1}$
²²⁵ Ra	Atlantic SNAP	4	52.2 d	$(5.6 \pm 2.0) \cdot 10^{-12}$
(RaCl ₂ ,	Atlantic GME	4	52.2 d	$(4.8 \pm 2.0) \cdot 10^{-12}$
ox.st. +2)	Pacific smectite	2	27.9 d	$< 3.8 \cdot 10^{-15}$
²²⁸ Th	Atlantic SNAP	3	99.0 d	$< 1.0 \cdot 10^{-14}$
(ThCl ₄ ,	Atlantic GME	3	99.1 d	$< 1.0 \cdot 10^{-14}$
ox.st. +4)	Pacific smectite	2	55.8 d	$< 1.2 \cdot 10^{-14}$
²³³ U	Pacific smectite	2	47.05 d	$(7.4 \pm 3.5) \cdot 10^{-13}$
(UO ₂ Cl ₂ ,				
ox.st. +6)				
¹¹³ Sn	Pacific smectite	2	15.98 d	$< 1.5 \cdot 10^{-14}$
(SnCl ₂ ,				
ox.st.+2)				
⁷⁹ Se	Pacific smectite	2	24.18 h	$(1.7 \pm 1.0) \cdot 10^{-10}$
(Na ₂ SeO ₃ ,				
ox.st. +4)				
¹²⁵ I	Pacific smectite	2	8.0 h	$(2.5 \pm 1.5) \cdot 10^{-9}$
(NaI,				
ox.st. -1)				

^a Values preceded by the "<" sign are upper limits for the effective diffusion coefficient.

^b There is some evidence that the neptunium is immobilized after an initial period of diffusion. The coefficient listed characterizes the mobility during the initial phase of diffusion.

^c The long term technetium sample indicated complete equilibration of the tracer throughout the sample. The coefficient listed was derived from short-term experiments.

Fig. 2. Concentration curve of ^{99}Tc after 2.05 days of diffusion in Atlantic SNAP sediment.

Fig. 3. Concentration curve of ^{237}Pu after 0.97 days of diffusion in Atlantic SNAP sediment.

leads to a best value of $1.0 \cdot 10^{-5} \text{ m}^2$ for D.t. Accordingly, the effective diffusion coefficient of technetium is $(5.7 \pm 2.8) \cdot 10^{-11} \text{ m}^2 \text{ s}^{-1}$.

Figures 3 and 4 are plots of experimental activity data for plutonium in Atlantic SNAP sediment. They can stand as representative for plutonium and americium, as well as other non-mobile species. The data of Fig. 3 were recorded after 23.3 hours, those of Fig. 4 after 60.8 days. The two concentration-distance curves are equal within experimental error. Diffusion has not progressed as would be expected if Fig. 3 represented a true sigmoid diffusion profile. The gradual increase of activity measured over approximately a millimeter distance in the center region of the sample must be attributed to experimental uncertainty. In this case, the value for the effective coefficient of diffusion derived by fitting a curve can only be interpreted as an upper limiting value. Coefficients that fall into this category are indicated in Table I by preceding them with a "<" sign. (Conf. footnote a of Table I).

Fig. 4. Concentration curve of ^{237}Pu after 60.8 days of diffusion in Atlantic SNAP sediment.

The data listed in Table I show the important transuranium elements plutonium and americium to be characterized by a very low mobility in all sediments tested. For these two elements no evidence for diffusive spreading in the substrate has been obtained. Their effective diffusion coefficients must therefore be smaller than $1 \cdot 10^{-14}$ to $8 \cdot 10^{-14}$.

At the other end of the scale of mobility the anionic species pertechnetate, iodide, and selenite are found. Their diffusion coefficients are between 10^{-9} and $10^{-10} \text{ m}^2 \text{ s}^{-1}$. This magnitude is consistent with uninhibited diffusion in the interstitial sea water, with an apparent reduction of the mobility because of tortuosity and channel size.

The cationic species NpO_2^+ and UO_2^{+2} show clear evidence for diffusion, but at a slower rate than anions. In this case, sorptive interaction with the solid matrix of the clay and hydrous oxides reduces the diffusion rates.

An unexpected result of the diffusion measurements in Atlantic sediments is observed in the case of neptunium. This element diffuses in the Atlantic SNAP sediment, but is effectively immobilized in the Atlantic GME sediment. It is possible that the relatively high content of organic carbon in the GME sediment provides a strong reducing environment in which the pentavalent neptunyl ion is converted to the tetravalent Np^{+4} species, which is hydrolysed to form an insoluble hydroxide. The example of neptunium points to the importance of site specific studies once a repository site has been selected.

CONCLUSION

The numbers listed in Table I provide useful guidance in the assessment of the effectiveness of the sediment barrier. Current plans call for an emplacement depth of 30 m for waste canisters in a subseabed repository. It will be assumed that no upward flow of the interstitial water exists in the sediment overlying the repository, and that the only mode of movement for ionic species dissolved in the water is molecular diffusion. The waste canisters will be fabricated to meet the specification set by the U.S. Nuclear Regulatory Commission of retaining their integrity for at least 1000 years. However, in the seawater-saturated environment of the sediment corrosion cannot be avoided, and ultimately leaching of radionuclides will occur. At that point the sediment layer above the repository becomes the principal barrier isolating the radionuclides from the biosphere.

A simple model can serve to visualize the diffusive spreading of radionuclides through the sediment at rates governed by the effective diffusion coefficients listed in Table I. This model assumes the radionuclides initially to be present at a uniform concentration in a sediment layer with a thickness of the expected height of a waste canister, i.e. 3 m, and that this layer is embedded in radionuclide-free sediment at a depth of 30 m. Diffusion outward from the layer is the well-known case of diffusion from a plane sheet of given thickness. Boundary effects arising from the fact that the repository is not infinite in extent will be ignored here. Since the repository is very much larger in lateral extension than in thickness, being a square array of perhaps 10000 canisters spaced 10 m apart from each other, the approximation by a layer of infinite extent involves only a small error. Besides, the error is on the conservative side, since lateral diffusion diminishes the amount of substance diffusing upward.

Figure 5 is a schematic of the model described here. The repository level is 30 m below the sediment surface at the bottom of the ocean. All diffusing radioactive material is initially contained in a 3 m thick layer at the repository level represented by the rectangular bar. As diffusion progresses, a concentration-distance curve will develop as indicated in Fig. 5. Repository performance for land-based repositories is specified in terms of maximum permissible cumulative releases for the individual radionuclides of concern over a 10000 year period.¹⁰ With this in mind, a critical release level can be defined, which determines the distance on the 10000 year concentration-distance curve beyond which no more than the maximum permissible amount of a given radionuclide has diffused. In other words, the integral of the curve up to the critical level, indicated by the cross-hatched area, must represent all the originally embedded material minus the maximum permissible amount released.

For land-based repositories, limits for maximum permissible releases of individual radionuclides have been set by the U.S. Environmental Protection Agency.¹⁰ For instance, the cumulative activity permitted to be released over a 10000 year period is 100 curies per 1000 tons of waste for ^{239}Pu and 10000 curies per 1000 tons of waste for ^{99}Tc . These two elements are at opposite ends of the mobility spectrum with effective diffusion coefficients of $2 \cdot 10^{-14} \text{ m}^2 \text{ s}^{-1}$ for plutonium and $3 \cdot 10^{-10} \text{ m}^2 \text{ s}^{-1}$ for technetium.

Fuel removed from reactors typically contains 5.9 kg of ^{239}Pu and 0.792 kg of ^{99}Tc per ton of waste.¹⁰ A waste canister ready for emplacement in a repository could contain the waste from as much as 3 tons of spent fuel, or a lesser amount if the fuel is not reprocessed¹¹. With a spacing of 10 m between canisters in a square array repository, 1000 tons of fuel require an area of 33300 m^2 . The activities of ^{239}Pu and ^{99}Tc are 366800 and 13400 curies, respectively.

After a 10000 year period of diffusion all but 100 curies of plutonium will be contained below a critical release level that is only 0.3 m above the level of original emplacement horizon, well within the 30 m thick sediment layer covering the repository. For the more mobile element technetium, the critical level is 3 m above the repository, still comfortably below the sediment surface. In this case, the comparatively high fraction of the nuclide that is considered acceptable to be released, alleviates the problem. Other radionuclides are bracketed by these two extreme cases. If diffusion is the only mode by which radioactivity from the waste can spread, the sea floor sediment provides an adequate geologic barrier for the isolation of nuclear waste.

The simple model calculation does not take into account refinements, such as the inclusion of boundary effects of a finite repository, the canister life, and the radioactive decay during diffusion. However, all of these effects reduce the amount of activity diffusing upward from the repository. The Subseabed Disposal Program has developed the computer codes IONMIG and MARINRAD for the comprehensive assessment of radionuclide releases.^{12,13} Conclusions derived from the elaborate calculations with these codes show that in the long term there are only two nuclides of potential concern in a subseabed repository: technetium and neptunium.²

Fig. 5. "Plane sheet" diffusion of radionuclides from a subseabed repository.

Questions that remain to be addressed in the assessment of radionuclide migration in ocean floor sediment concern the influence of temperature and pressure on the rate of diffusion. The temperature in the sediment is near 2°C and the pressure is 600 atmospheres. The lower than ambient temperature is likely to reduce the values of effective diffusion coefficients that are measured at ambient temperature. The effect of the high pressure, on the other hand, is less easily assessed, and its influence on diffusion rates must be measured either in future laboratory tests, or in situ in the ocean floor.

Finally, it must be remembered that the effective diffusion coefficients are representative for the migration of radionuclides under stagnant conditions. Hydrodynamic flow through the sediment in an upward direction would easily overwhelm the slow rates of molecular diffusion and become the most important factor for the transport of radionuclides. Its absence at the repository site must be assured. Flow rates of 1 cm per year are sufficient to cause concern, and the absence of such low flow rates is quite difficult to verify. Techniques to deal with the problem of interstitial flow in the sediment are under development. Present evidence suggests that there are large regions of sediment-covered ocean floor in which no upward component of flow exists.

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