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NUCLIDE MIGRATION IN FRACTURED OR POROUS ROCK

by

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Nuclide Migration in Fractured or Porous Rock

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Repositories built in geologic bodies are being considered for permanent disposal of nuclear wastes. In the context of deep geologic repositories, the rock formation itself may be considered as part of the mechanism retaining the wastes within the immediate neighborhood of the repository. The interactions of nuclear wastes with geologic materials are being studied in laboratory experiments to evaluate geologic media as barriers to nuclide migration. Migration caused by flowing water is considered the most credible mechanism for moving wastes from a repository in a well chosen site. The extent and rate of migration of wastes under the influence of groundwater, of course, depend on several parameters; including the type and extent of the chemical reaction of the radionuclides with the geologic media.

In previous work (1,2,3) it was found that the kinetics of sorption was an important parameter affecting the migration of nuclides in geologic media. For example, in experiments designed to measure the kinetics of reaction for radionuclides in solution with tablets of rock, it was found that periods from several minutes to several hours were required for the radionuclides to reach steady state concentrations on the rock tablets and in the solutions. Figure 1 shows the reaction curves found for the sorption of plutonium and americium from solution by a tablet of granite. The reaction rates for the sorption of plutonium and americium from solution are not the same, and both require a number of hours to reach steady state concentrations.

The observation that the sorption process progresses as a function of time implies that under conditions of solution flow through geologic media, there may not be instantaneous or local equilibrium between a nuclide bearing solution and geologic media, but rather, the nuclide would be sorbed from solution through a length or zone of the media. Following adsorption, the nuclide would desorb after an amount of water passed over the media. The nuclide would again be readsorbed downstream a distance dependent

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upon the rate of adsorption and the flow rate of the solution. Desorption and readsorption would continue in this manner and the migration of the nuclide would be defined in terms of the rate of adsorption, the rate of desorption, and the equilibrium distribution of the nuclide between solution and geologic media.

To quantify this treatment of migration as influenced by kinetics, a model has been developed in which instantaneous or local equilibrium is not assumed. The model is called the Argonne Dispersion Code (ARDISC) (4). In the model, adsorption and desorption are treated independently and the rates for adsorption and desorption are taken into account. The model treats one dimensional flow and assumes a constant velocity of solution through a uniform homogeneous media.

This paper describes an experimental study of the applicability of the ARDISC model to laboratory studies of nuclide migration in geologic media.

Basic Description of Model

The model assumes a one dimensional column divided into an arbitrary number (LEND) of units or zones. Each zone can sorb a given species such that at equilibrium the distribution of the species between the surface of the zone and the volume of solution (initially at unit concentration of the species) is

$$\alpha/\beta \quad (1)$$

where

α = amount of species sorbed on the surface

and

$$\beta = 1 - \alpha$$

If after this distribution is established, a second cycle occurs in which the solution from the initial zone is transferred to a second zone with the same characteristics as the first zone and a fresh solution containing none of the species is placed in the first zone, another equilibration, this time in both zones will occur. This process is continued by the model for additional cycles and zones.

The description of the model up to this point is analogous to a description of a chromatographic model which incorporates the concept of the theoretical plate (5).

If, however, the movement from zone to zone is fast enough that equilibrium does not occur, the fraction sorbed from solution can be calculated if the rate of sorption is known (or can be estimated). A parameter, F , is therefore defined

$$F = \frac{\text{fraction of species sorbed from soln. during one cycle time}}{\text{fraction of species sorbed from soln. at equilibrium}} \quad (2)$$

which can be related to the rate of adsorption of the species.

The parameter, F, gives directly the amount adsorbed in a zone in which the species initially is present only in the solution.

For any species adsorbed on the surface of a zone, the fraction of the sorbed species which desorbs during the time of a cycle can be measured (or estimated). Another parameter, G, is therefore defined

$$G = \frac{\text{fraction of species desorbed from surface during 1 cycle time}}{\text{fraction of species desorbed from surface at equilibrium}} \quad (3)$$

to account for desorption under non-equilibrium conditions. The parameter, G, gives directly the amount desorbed in a zone where the species is initially present only on the surface of a zone.

Thus for each zone, during a given cycle, the adsorption-desorption process is separated into two distinct events with F or G describing the kinetics of each event. Such an approach is of course valid only for first order rate reactions. In the limit of low concentration, (such as that resulting from slow leaching from a repository) the reaction sites on the rock will not approach saturation and the number of reaction sites can be considered to remain constant during adsorption. Therefore, for a single species in solution at tracer concentrations the reaction should approximate a first order reaction (i.e., where no complications such as concentration effects, step-wise dehydration, dissociation, etc., are present).

If a calculated cycle is defined as a partial equilibration between the species on the surface of a zone and the species in the solution of a zone, and is followed by an instantaneous transfer of the solution to the next zone, the following recursive formulae apply:

$$R(\ell, Z) = G\alpha R(\ell-1, Z) + (1-G)R(\ell-1, Z) + F\alpha S(\ell-1, Z) \quad (4)$$

$$S(\ell, Z+1) = F\beta S(\ell-1, Z) + (1-F)S(\ell-1, Z) + G\beta R(\ell-1, Z) \quad (5)$$

or rearranging

$$R(\ell, Z) = [G\alpha+1-G]R(\ell-1, Z) + F\alpha S(\ell-1, Z) \quad (6)$$

$$S(\ell, Z+1) = [F\beta+1-F]S(\ell-1, Z) + G\beta R(\ell-1, Z) \quad (7)$$

where $R(\ell, Z)$ = amount of species sorbed on the surface in the Zth zone after the ℓ th cycle.

$S(\ell, Z+1)$ = amount of species in solution in the (Z+1)th zone after the ℓ th cycle. (the solution in the Zth zone is transferred to the (Z+1)th zone at the end of the ℓ th cycle.)

Thus if α , F, and G are known and if ℓ and Z are specified, the distribution of the species both in solution and on the media can be calculated by the model. For equations 6 and 7, one must have

an initial amount of species present.

Experimental

If the rate of adsorption, the rate of desorption, and the equilibrium partitioning of a nuclide between a solid medium and solution are known, then the rate of migration of a nuclide through the medium can be predicted with the ARDISC model.

The rate of adsorption and the rate of desorption are assumed to be dependent on the geometric relationship of the rock and solution (i.e., dependent upon the volume of solution and on the shape and area of the solid medium in contact with the solution). Because the dependence of sorption kinetics on the geometric relationship is not known, the rates for sorption are determined by experiment for the particular geometry (surface area of rock to volume of solution) for which the prediction of nuclide migration is desired.

The applicability of the ARDISC model to laboratory migration experiments was studied in two sets of experiments. In the first set of experiments, the rate of adsorption, the rate of desorption, and the equilibrium partitioning of americium III between solution and simulated fissures were measured in static and batch experiments. The measured parameters were used in the model to predict the migration of americium III through simulated fissures. The predictions were compared with experimental results. In the second set of experiments, strontium was eluted through a column of glauconite at six solution flow rates ranging from 14.4 to 721 centimeters per hour. To obtain input data for the ARDISC model the equilibrium partitioning of strontium between solution and glauconite was determined from the resulting curves and the rate for adsorption and the rate for desorption were determined by curve fitting the results of one of the column infiltration experiments. The parameters that were determined in this way were used in the model to predict the migration of strontium through the column of glauconite for four of the solution flow rates. The predictions were compared with experimental results.

Fissures

Fissures were fabricated by cutting slabs of rock from a core of gray hornblende schist. X-ray diffraction analysis of gray hornblende schist identified chlorite and an amphibole (hornblende) as the major mineral phases present and feldspar as a minor constituent. The slabs were cut into rectangles approximately 2.54 cm wide by 5.08 cm long. Each piece was polished on one side with successively finer sized abrasive with the final abrasive size being 600 mesh. To form a fissure, two rectangular pieces were put together. Wax gaskets were placed along the long edges of the rectangular pieces to form a 0.027 cm gap between

the two pieces. The surface area to volume ratio produced was 74 cm² to 1 mL and the surface area to volume ratio was constant for all the fissures used.

Fissure Adsorption Experiments. To perform fissure sorption experiments, one end of each fissure was connected via small bore tubing to a microliter pipetter as is shown in Figure 2. Rock-equilibrated water, described below, was injected into each fissure and the fissure walls were allowed to equilibrate with the solution for a period of two days before the adsorption experiments were performed.

Rock-equilibrated water was produced by allowing distilled water to be in contact with granulated gray hornblende schist for a period of several days. The solution produced was filtered through a 0.4 μ m NUCLEOPORE filter before use. Analysis of the resulting solution showed that rock equilibrated-water contained 48 mg/L total solids (dried at 130°C). The E_h of the solution was measured to be 0.275 V. The pH of the gray hornblende schist equilibrated water was measured to be 7.6 at the time of the first adsorption experiment. Thereafter the solution pH was maintained at 7.5-7.7.

Adsorption experiments were performed by removing rock-equilibrated water from the fissures and injecting stock solution which was made by dissolving tracer amounts of americium-241 in rock equilibrated water. The stock solution was allowed to equilibrate within the fissures for different periods of time and was then removed from each fissure. The stock solution was assayed before injection and after removal from the fissures so that the change in americium concentration was determined for a different time in each fissure. Ten adsorption experiments were performed in this manner and the results are presented in table I. Figure 3 is a graphical representation of the initial part of the adsorption curve.

Equilibrium Partitioning Experiments. The equilibrium partitioning of americium-III between gray hornblende schist and rock equilibrated water was determined in batch partitioning experiments with rectangular blocks of gray hornblende schist (5). The surface area sorption coefficient, K_s , was determined to be $4.5 \pm .5$ mL/cm² where

$$K_s = \frac{\alpha}{\beta} \frac{\text{volume of solution}}{\text{surface area of sample}} \quad (8)$$

α from equation 1 is the fraction of nuclide concentration adsorbed from solution.

β from equation 1 is the fraction of nuclide concentration remaining in solution; $1 - \alpha$.

Solving equation 8 for the geometric surface area to volume ratio

found in the fissures, the fraction of the nuclide concentration predicted to be adsorbed by the fissure surfaces at equilibrium in fissure sorption experiments is 0.997.

Fissure Desorption Experiments. Desorption experiments were performed with two fissures by injecting rock equilibrated water into the fissures immediately after removing the americium-bearing solution from the fissures in adsorption experiments. After injection, the rock-equilibrated water was allowed to react within the fissures for a period of time before removal. The rock-equilibrated water was assayed after removal from the two fissures and the amount of nuclide desorbed as a function of contact time was determined. Between five and seven desorption experiments were performed for each of six time periods between ten seconds and sixty minutes for each of the two fissures. The results of the desorption experiments are presented in table 2 as the percent of β (at equilibrium) that desorbed from the fissures in a period of time.

Because most of the americium remains on the rock even after desorption is completed, the ratio of activity desorbed to that expected at equilibrium is subject to large uncertainty. The results of the desorption experiments are so varied that no quantitative conclusion can be drawn about the rate for desorption. However, the results do indicate that desorption may occur with a rate equal to or less than the rate for adsorption.

Fissure Elution Experiments. The migration characteristics of americium by water transport in fissures fabricated from gray hornblende schist were determined. Fissures not used in the previous sorption experiments were used for these elution experiments. A diagram of the experimental apparatus is shown in Figure 4. Solution reservoirs were attached above the fissures and the small bore tubes affixed to the bottom of the fissures were connected to solution metering pumps.

Before the elution experiments were performed, the "schist"-equilibrated water was pumped through the fissures for two days to allow the fissure walls to interact with the rock-equilibrated water.

Two sets of fissure elution experiments were performed. In the first set of experiments, the rock equilibrated water contained in the reservoirs above three fissures was first replaced with americium bearing stock solution. The solution metering pumps were left running in order not to interrupt the flow while the exchange was made. The flow was slow enough that air was not drawn into the fissures during the exchange. Flow was continued until an arbitrary volume, 0.67 fissure volumes, of stock solution were drawn into each fissure and then the solution metering pumps were turned off. This volume was chosen to permit the observance of a leading edge of activity, if it existed, moving with the water front. Stock solution was drawn into the fissures at linear

flow rates of 1.13, 2.29, and 4.77 cm/hr respectively. After stopping the flow of stock solution into the fissures, it was removed from the reservoirs above the fissures and the solution in each fissure was left stagnant for a period of twenty-four hours. Based on the kinetic data in table 1, it is believed that the americium in the stock solution becomes equilibrated with the surface of the fissures during the twenty-four hour period that the stock solution remained stagnant. At equilibrium 0.997 of the americium is adsorbed; it is therefore expected that the americium distribution on the fissure walls, after the twenty-four hour period, represents the migration of americium into the fissures during the 0.67 fissure volume elutions.

After the twenty-four hour period of contact, the solution in the fissures was rapidly withdrawn. The surfaces of the fissures were dried under vacuum at 25°C and then dismantled. The distributions of americium on the fissure surfaces were determined first qualitatively using autoradiographs of the fissure surfaces with POLAROID LAND black and white 3000 ASA type 47 film. The autoradiographs are shown in Figures 5, 7, and 9. The lighter areas on the autoradiographs represent areas of americium adsorption.

The distributions of americium on the fissure surfaces were then quantitatively determined by scanning the face of each fissure with a NaI scintillation crystal through a 0.3 cm slit in lead shielding. The 59 keV gamma ray emitted by ^{241}Am was monitored. Histograms of the americium distributions on the fissure surfaces were produced and are presented in Figures 5, 7, and 9.

The second set of fissure-elution experiments was performed in the same manner as described for the first set of fissure-elution experiments. The difference in the second set of experiments was that after 0.67 fissure volumes of stock solution were drawn into the fissures at their respective flow rates, the stock solution in the reservoirs was replaced with rock-equilibrated water. The solution metering pumps were not turned off during the exchange. Subsequently, a total of twenty fissure volumes of solution was drawn through each fissure before the metering pumps were turned off. After the metering pumps were stopped, the rock-equilibrated water was removed from the reservoirs and the solution in the fissures was rapidly drawn off.

The fissures were dried under vacuum at 25°C and were dismantled. The distributions of americium on the fissure surfaces in the second set of experiments were determined as they were in the first set, that is: first qualitatively by autoradiography and then quantitatively by gamma scanning the fissure faces through a 0.3 cm slit.

Autoradiographs of the fissure surfaces from the second set of experiments are presented in Figures 6, 8, and 10. Histograms representing the quantitatively determined distribution of americium on the fissure surfaces are also presented in Figures 6, 8, and 10.

Model Predictions. The rate for desorption of americium from the fissure surfaces into solution was assumed to equal the rate for the adsorption of americium from solution by the fissure surfaces. The sorption rate and the equilibrium fractionation of americium that were determined in the static experiments were used to determine input parameters to the ARDISC model. The ARDISC model predictions for the distributions of americium on the fissure surfaces in both sets of experiments are presented in Figures 5 through 10 along with the autoradiographs and the experimental histograms representing the various distributions of americium on the fissure surfaces.

Strontium Migration Experiments in Glauconite

The work that was performed in this set of experiments was an extension of work performed by Inoue and Kaufman (7). In the previous work, the migration of strontium in glauconite was modeled using conditions of local equilibrium for flows up to 6.3 kilometers per year (72 cm/hr). The differences between the predicted and experimental results in the experiments performed by Inoue and Kaufman may be due to the existence of non-equilibrium behavior.

Column Infiltration Experiments. Six infiltration experiments, each at a different flow, were performed with one column of glauconite. The apparatus used in the infiltration experiments consisted of a mineral column contained in a stainless-steel tube connected to a sample-injection valve and a solution-metering pump. Glauconite [a hydrous silicate, nominally $(K,Na)(Al,Fe^{+3},Mg)_2(Al,Si)_4O_{10}(OH)_2$] was prepared by sifting glauconite sand to obtain a fraction of uniform size (<40 and >70 mesh size). The sand was washed repeatedly in distilled water to remove dust adhering to the particles and was packed in a stainless-steel tube to form a column 15 cm long by 1.0 cm diameter with 53 percent porosity.

The column was conditioned by eluting it with a solution of 0.01 M $CaSO_4$ and 0.0001 M $SrCO_3$. The solution used in this set of experiments was produced to simulate the solution used by Inoue and Kaufman.

Strontium-85 with $SrCl_2$ carrier was added to an aliquot of the calcium-strontium solution to form a radioactive solution with 1×10^{-6} M ^{85}Sr . At the start of an experiment, a small quantity (20 μl) of the radioactive solution was injected into the solution stream above the glauconite column and the column was eluted with solution until all the radioactive strontium had moved through the column. The eluent was collected in fractions and each fraction was analyzed to determine the migration characteristics of the strontium in the column.

The radioactive strontium in solution vs. the eluate fractions for each flow rate is plotted in Figure 11. The strontium

peaks have a shape that is dependent on the flow rate of the eluent. The sharpest peak is produced with the slowest flow rate; the widest peak is produced with the fastest flow rate. The strontium peaks were delayed relative to the advancing water front (which would traverse the column in the first 6.5 mL of eluent). Moreover, each of the strontium peaks seen in Figure 11, even the one produced with the slowest flow is wider than predicted by the peak width (2 mL full width at half maximum) that was seen for peaks of tritiated water that were eluted through the column (8). Therefore, the strontium has interacted with the glauconite column and, as indicated by the different peak shapes, interacted differently at each flow. The flows given in Figure 11 were measured by collecting eluate over specific periods of time. In each experiment, all the radioactive strontium was eluted in the single peak seen in the figure.

To relate the six curves obtained experimentally with the ARDISC model, the rate for adsorption, the rate for desorption and the equilibrium distribution of the nuclide between glauconite and solution had to be determined.

The relative migration rate of the peak concentration of strontium at a solution flow rate of 0.1 mL per minute was used to calculate the equilibrium fractionation of strontium between glauconite and solution. It was determined that at equilibrium 0.958 of the strontium would be adsorbed by the glauconite and 0.042 of the strontium would be in solution (mobile phase).

The rate for adsorption and the rate for desorption were determined by curve fitting with ARDISC. It was assumed that the rate for desorption equaled the rate for adsorption. The α and β parameters were held constant at 0.958 and 0.042 respectively. The F, G, and zone length input parameters to the ARDISC model were varied to fit the strontium migration data at a flow rate of 2 mL per minute.

Assuming first order reaction kinetics, the sorption rate that was determined for adsorption and desorption was 0.187 sec^{-1} . A reaction rate of 0.187 sec^{-1} implies a half time of reaction of 3.7 seconds.

The curves predicted by the model for four of the six flow rates are presented in Figure 12.

Discussion and Conclusions

Figures 5, 7, and 9 show the americium distributions on fissure surfaces after the administration of 0.67 fissure volumes of stock solution at flow rates of 1.13, 2.29, and 4.77 cm/hr. The peak concentrations of americium were adsorbed at the top of the fissures and leading edges of americium were extended into the fissures. In general, at faster flow rates the relative quantities of americium sorbed at the top of each fissure decreased and the length and relative amount of the leading edge extending into each fissure increased.

The ARDISC model predictions for 0.67 fissure volume additions of stock solution into fissures at the three flow rates also show that the peak concentrations of americium will be sorbed at the top of each fissure. The leading edges of the nuclide are predicted to extend into the fissures. The ARDISC model also predicts that at faster flow rates, the relative amount of americium sorbed in the peak concentration at the top of each fissure should decrease. Also, the length and relative amount of the leading edge extending into each fissure is shown to increase.

Figures 6, 8, and 10 show that the distributions of americium on the fissure surfaces after the addition of 0.67 fissure volumes of stock solution followed by the elution of 20 fissure volumes of "schist"-equilibrated water through the fissures at flow rates of 1.13, 2.29, and 4.77 cm/hr. A comparison was made between the americium distributions found on the fissure surfaces after the addition of americium stock solution and that found after elution of the americium by 20 fissure volumes of "schist"-equilibrated water. It was found that after the initial loading of americium into the fissures in the first 0.67 fissure volumes of solution, the peak concentrations of americium that were sorbed at the top of the fissures decreased in their relative concentration. The leading edges of the detectable nuclide concentration extending into the fissures had increased in length and relative concentration with subsequent elution by rock equilibrated water through the fissures. The ARDISC model predicted the same relationships.

The ARDISC model qualitatively predicted the shape of the curves representing the distribution of americium on the fissure surfaces. However, the model was not able to predict accurately the distribution of americium on the fissure surfaces in each experiment. A plot of the natural logarithm of the fraction of the nuclide remaining in solution vs. the absorption time does not produce a straight line (Figure 13) for the first five data points obtained in the static fissure absorption experiments. A plot of the natural logarithm of the fraction remaining in solution vs. time should produce a straight line if the reaction were first order. The ARDISC model assumes that the sorption reactions are first order. The discrepancy between the experimental and predicted results may be due to higher order sorption kinetics. In addition, differences between the predicted and the experimental results may also be due to a number of factors such as non-homogeneous media, non uniform flow through the fissures, small pH fluctuations, precipitation reactions, and colloid formation by some of the nuclide.

In the column infiltration experiments with strontium, the model predictions closely resemble the experimental curves for the four flow rates compared. The input parameters to the ARDISC model were derived from experimental data obtained in infiltration experiments. The model predictions were based on the assumptions that the rate for adsorption and the rate for desorption were equal and that the sorption reactions were both first order.

The results of the experiments presented in this paper demonstrate that the migration of nuclides in geologic media can be studied experimentally and treated in at least a semiquantitative fashion using kinetic and partitioning data. The ARDISC model is a useful aid in analyzing nuclide migration data obtained in laboratory experiments. But the ARDISC model is limited to first-order sorption kinetics.

Phenomena such as nuclide transport by particles or nuclide transport in colloidal form will interfere with a kinetic approach to predicting nuclide migration. The results indicate that the measurement of kinetic parameters may be as important to understanding the migration of a nuclide through a geologic media as the measurement of the equilibrium-sorption value (K_d).

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Table I. ^{241}Am Adsorption by Gray Hornblende Schist:
Fissure Experiments (static)

Adsorption Time, Seconds	Fissure Number	Activity Applied in 152 λ	Percent Remaining in Solution
15	1	65 nCi	84.6 $\pm$.1
54	5	0.98 nCi	71 $\pm$ 1
150	8	0.97 nCi	63.1 $\pm$.9
300	2	1.1 nCi	50.7 $\pm$.8
600	7	1.7 nCi	36.3 $\pm$.5
3600	4	56 nCi	32.3 $\pm$.1
60120	2*	173 nCi	25.0 $\pm$.1
60120	8*	184 nCi	17.4 $\pm$.1
86400	5*	111 nCi	2.20 $\pm$.02
250200	6	0.84 nCi	0.05 $\pm$.25

*Cores #2, #5, and #8 were used twice for adsorption experiments. The activity applied during the first adsorption experiments was insignificant when compared to the activity applied in the second experiment. The reported errors are for counting statistics.

Table II. ^{241}Am Desorption from Gray Hornblende Schist:
Fissure Experiments (static)

Desorption Time, Seconds	Desorption Runs	Percent of β^* (at equilibrium) Desorbed	
		Core #2	Core #8
10	5	8 $\pm$ 5	4 $\pm$ 2
20	7	13 $\pm$ 10	13 $\pm$ 4
60	7	31 $\pm$ 47	19 $\pm$ 6
360	6	8 $\pm$ 6	13 $\pm$ 5
1200	5	13 $\pm$ 8	6 $\pm$ 2
3600	7, 6 (core #8)	14 $\pm$ 19	22 $\pm$ 16

* β is the fraction of a nuclide in solution at equilibrium. The desorption values were determined assuming that 0.003 of the nuclide would be in solution at equilibrium. The values that are presented are the arithmetic mean and standard deviations found for the number of experiments performed for each time interval.

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Fig. 1

Fig. 2

Fig. 3

Fig. 4

A. AMERICIUM DISTRIBUTION ON SURFACE OF FISSURE.

B. MODEL PREDICTION OF AMERICIUM ON SURFACE OF FISSURE.

C. AUTORADIOGRAPH OF FISSURE SURFACE SHOWING AMERICIUM DISTRIBUTION.

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B. MODEL PREDICTION OF AMERICIUM ON SURFACE OF FISSURE.

C. AUTORADIOGRAPH OF FISSURE SURFACE SHOWING AMERICIUM DISTRIBUTION.

A. AMERICIUM DISTRIBUTION ON SURFACE OF FISSURE.

B. MODEL PREDICTION OF AMERICIUM ON SURFACE OF FISSURE.

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A. AMERICIUM DISTRIBUTION ON SURFACE OF FISSURE.

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Fig. 11

Fig. 12

Fig. 13