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# Modeling of Sorption Characteristics of Backfill Materials

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## ABSTRACT

Sorption data analysis was carried out using the Freundlich, Langmuir, and Modified Freundlich isotherms for the uptake of sodium and potassium in an initial concentration range of 10–100 mg/L on backfill materials, viz., bentonite, vermiculite, and soil samples. The soil samples were collected from a shallow land disposal facility at Kalpakkam. The Freundlich isotherm equation is validated as a preferred general mathematical tool for representing the sorption of $K^+$ by all the selected backfill materials. The Modified Freundlich isotherm equation is validated as a preferred mathematical tool for representing the sorption of $Na^+$ by the soil samples. Since a negative sorption was observed for the uptake of $Na^+$ by commercial clay minerals (vermiculite and bentonite clay in the laboratory experiments), sorption analysis could not be carried out using the above-mentioned isotherm equations. Hill plots of the sorption data suggest that in the region of low saturation (10–40 mg/L), sorption of $K^+$ by vermiculite is impeded by interaction among sorption sites whereas sorption by soil and bentonite clay is enhanced by interaction among sorption sites. In the region of higher saturation (60–100 mg/L), sorption of $K^+$ by all three backfill materials is enhanced by interaction among sorption sites. The Hill plot of the sorption data for $Na^+$ by soil suggests that irrespective of $Na^+$ concentration, sorption of $Na^+$ at one exchange site enhances sorption at other exchange sites.

## INTRODUCTION

Backfill material as an engineered barrier for the disposal of radioactive waste is an important aspect of the multibarrier concept (1). The ultimate

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release of radionuclides from a radioactive waste repository will depend upon the natural and the man-made barriers surrounding the site. Interaction of groundwater components with the backfill material is a controlling factor for any sorption process in order to predict the rates of migration. Natural inorganic sorbents are known to act as effective barriers against radionuclide migration from radioactive waste repositories and can be used as buffers and backfill materials in nuclear waste disposal facilities.

Based on a previous study, a mixture of bentonite clay, vermiculite, and soil from a shallow land disposal facility at Centralised Waste Management Facility (CWMF), Kalpakkam, were chosen as representative materials for backfill in the reinforced concrete trenches and tile holes at the above disposal site (2).

The $Na^+$ concentration in the groundwater at the disposal site was found to vary in the 18–90 mg/L range, which appeared to be the largest range when compared to other alkali and alkaline-earth metal ions. Hence, in order to gain a thorough understanding of the sorption characteristics of the selected backfill materials, batch sorption studies were carried out for the uptake of $Na^+$ in an initial concentration range of 10 to 100 mg/L as representative of the actual levels encountered in groundwater samples. Although the $K^+$ levels encountered were very low, their sorption studies were carried out in the same concentration range in order to compare the results with $Na^+$ to understand how these ions compete in the presence of radionuclides to be adsorbed on the backfill material.

The objectives are to examine several equilibrium isotherm equations and evaluate their applicability to model sorption of the above-mentioned ions by backfill materials. The selection of the isotherm that best fits the experimental data would reflect more objective predictions when applied to transport models (3). The isotherms include two of the most commonly used isotherms (Freundlich and Langmuir) and a Modified Freundlich isotherm whose predictive power is based on theoretical considerations. The Langmuir, Freundlich, and Modified Freundlich isotherms were used in early studies to describe the sorption characteristics of bentonite (4–7), vermiculite (4, 8) and soils (9–13). In an attempt to find additional information on the interaction among sorption sites, the value of the Hill plot approach was also examined. Some relevant information on desorption characteristics of the backfill materials studied has been cited from the literature.

## SORPTION MODELS

### Langmuir Isotherm

The Langmuir equation was derived for the sorption of gases on a solid surface. Nevertheless, it has been extended to include the sorption of solutes on soils. A standard mathematical representation is

$$C/S = (1/b)C + 1/kb \tag{1}$$

where $S$ = mass of the solute sorbed by the backfill material (mg/g)
$C$ = equilibrium concentration of solute in equilibrating solution (mg/L)
$k$ = measure of the strength of the sorption bond
$b$ = maximum amount of the solute that can be sorbed by the backfill material (mg/g)

This isotherm is based on the theoretical assumption that the sorption sites are homogeneously distributed relative to energy potentials.

Throughout this paper each variable is defined when it is introduced for the first time and thereafter has the same meaning in subsequent equations.

## Freundlich Isotherm

The Freundlich isotherm has the form

$$S = KC^N \tag{2}$$

where $K$ = predicted quantity of sorption per gram of the backfill material at unit equilibrium concentration (mg/g)
$N$ = measure of nature and strength of the adsorption process and on the distribution of active sites

If $N < 1$, bond energies increase with surface density; if $N > 1$, bond energies decrease with surface density; and when $N = 1$, all surface sites are equivalent (14). The Freundlich isotherm is limited in that it only shows linearity over a relatively narrow range of concentrations.

## Modified Freundlich Isotherm

Solute exchange on soil adsorption sites is known to be heterogeneous, that is, not monoenergetic (15). Sposito, using statistical mechanics, derived rigorously the following equation with the assumptions that the exchanger surface is heterogeneous and that each class of exchange sites adsorbs individually according to the Langmuir isotherm (16).

$$S_1 = \frac{S_{\max}\alpha(a_1/a_2)^\beta}{1 + \alpha(a_1/a_2)^\beta} \tag{3}$$

where $\alpha = Aa_2^\beta/S_{\max}$
$a_1$ = activity of Solute 1 in solution
$a_2$ = activity of Solute 2 in solution

$S_{\max}$ = maximum available exchange sites or cation-exchange capacity
$S_1$ = amount of Solute 1 sorbed
$A$, $\beta$ = empirical constants

Equation (3) reduces to the Freundlich isotherm when $(a_1/a_2)^\beta \ll 1$, i.e., when the adsorbing solute (Solute 1) is in trace quantities. However, without limiting the sorbing solute to trace quantities, Eq. (3) is a modified form of the Freundlich isotherm *b*, transforming it as follows:

$$\log S_1/(S_{\max} - S_1) = \beta \log C + \log A/S_{\max} \tag{4}$$

where activities have been replaced by concentrations ($a_1$, $a_2$ for $C_1$, $C_2$, respectively).

The empirical constants $A$ and $\beta$ are determined through linear regression analysis. These empirical constants provide a means for visualizing the general sorption process relative to the distribution of sorptive energies imparted by the exchange surface.

## Distribution Coefficient

The sorption process may be expressed in terms of an empirical parameter $K_d$ (distribution coefficient) which has the unit of mL/g and is defined as the equilibrium concentration of the sorbed species divided by the equilibrium concentration of the species in solution.

The empirical parameter $A$ given by Sposito (16) can be represented as

$$A = S_{\max}K_d^\beta \tag{5}$$

The parameter $A$ is a measure of the weighted average distribution coefficient, and the parameter $\beta$ describes the spread of the distribution coefficient about the average distribution coefficient ($K_d$).

## Hill Plot

All the aforereviewed models require regressional analysis to estimate their constants. On the other hand, the Hill plot method yields information on the binding process without the need for statistical curve fitting. The Hill plot (17) may be expressed as

$$S/S_{\max} = y = K^*C^a/(1 + K^*C^a) \tag{6}$$

where $y$ = fraction of the total number of sites occupied by $C$
$K^*$, $a$ = empirical constants

These empirical constants are respectively related to the stability constants of the process and the number of binding sites.

The value of the Hill-plot approach stems from its clues on the interactions of sorption sites based on the slopes of the Hill plot (18). Three kinds of interactions can be evaluated: zero if the slope is equal to one, positive if the slope is greater than one, and negative if the slope is less than one. Zero interaction indicates that sorption at one site is independent of sorption at other sites. Positive interaction indicates that sorption at one site is enhanced by sorption at other sites, and negative interaction indicates that sorption at one site is impeded by sorption at other sites.

## MATERIALS AND METHODS

### Materials

a). Study materials include commercially available bentonite clay, vermiculite, and the soil sample from the shallow land disposal facility. The mineralogical composition of the backfill materials is given in Table 1. Since commercial bentonite clay is a very fine powder, it was used as such for batch sorption studies whereas commercial vermiculite was first ground in a ball mill and sieved to a mesh size of 610 μm. A soil sample of mesh size 610 μm was used for the above sorption studies.

b). NaCl used for the sorption studies was of analytical grade, 99.9% purity and KCl used was of analytical grade, 99% purity.

### Methods

Accurate amounts of about 1 g of backfill materials were equilibrated with $Na^+/K^+$ solutions in initial concentrations of 10, 20, 40, 60, 80, and 100 mg/L. The $Na^+$ concentration in the groundwater at the disposal site was found to vary in the 18 to 90 mg/L concentration range which appeared to be the

TABLE 1
Mineralogical Composition of Backfill Materials

| Components | Clay (%) | Vermiculite (%) | Soil (%) |
|---|---|---|---|
| $Na_2O$ | 0.65 | 0.14 | 0.43 |
| CaO | 27.4 | 15.91 | 19.58 |
| MgO | 35.12 | 68.5 | 27.06 |
| $Al_2O_3$ | 0.42 | 0.2 | 0.47 |
| $Fe_2O_3$ | 3.95 | 2.01 | 2.95 |
| $K_2O$ | 0.44 | 1.35 | 1.7 |
| $SiO_2$ | 33.13 | 12.03 | 47.7 |

TABLE 2
Groundwater Composition at Waste Disposal Site, Kalpakkam (22)

| | |
|---|---|
| pH | Neutral to mildly alkaline |
| Alkalinity | 46–287 mg/L as $CaCO_3$ |
| Hardness ($CaCO_3$) | 130–240 mg/L |
| Chlorides | 25–80 mg/L |
| Sulfates | 3–10 mg/L |
| Nitrates | 10–15 mg/L |
| Dissolved oxygen | 4–6 mg/L |
| Specific conductivity | 0.122–1.153 mμ |
| Total dissolved solids | 2000–3000 mg/L |
| Sodium ($Na^+$) | 18–90 mg/L |
| Potassium ($K^+$) | 3 mg/L |

highest range compared to other alkali and alkaline-earth ions. Hence, batch sorption studies were carried out for the uptake of $Na^+$ as well as of $K^+$ in the 10–100 mg/L initial concentration range. The groundwater composition (22) at the disposal site is given in Table 2.

After allowing equilibration of 25 days, the samples were centrifuged for 30 minutes at 6000 rpm. The supernatant solutions were passed through a 0.45-μm filter to minimize colloidal particles in the filtrate. An aliquot of the filtrate was analyzed for $Na^+$ or $K^+$ by flame photometry depending on the ion of interest. The experiments were carried out in duplicate. The amount of $Na^+/K^+$ sorbed was calculated from the initial and final concentrations in the aqueous phase. The linearized model expressions discussed in the Sorption Models Section were employed, and the sorption data are plotted accordingly.

In the present study, $K_d$ values were determined by using

$$K_d = \frac{(C_i - C_e)}{C_e} \frac{V}{M} \tag{6}$$

where $C_i$ = initial concentration (mg/L)
$C_e$ = equilibrium concentration (mg/L)
$V$ = volume of solution used for equilibration (mL)
$M$ = mass of the sorbent (g)

## RESULTS AND DISCUSSION

The sorption data obtained at equilibrium conditions for each initial concentration of $K^+$ for the three selected backfill materials were analyzed by the Langmuir, Freundlich, and Modified Freundlich equations. A similar attempt

TABLE 3
Statistical and Empirical Data Estimated from Isotherm Equations for Sorption of Sodium and Potassium[a]

| | Langmuir | | | | Freundlich | | | | Modified Freundlich | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Equation | $S = \frac{kbC}{1 + kC}$ | | | | $S = KC^N$ | | | | $\frac{S_l}{S_{max} - S_l} = \frac{A}{S_{max}} C_l^{\beta}$<br>$A = S_{max} K_d^{\beta}$ | | | | | |
| Regression | $C/S$ vs $C$ | | | | $\log S$ vs $\log C$ | | | | $\log \frac{S_l}{S_{max} - S_l}$ vs $\log C_l$ | | | | | |
| Correlation coefficient ($R^2$) | $Na^+$ | | $K^+$ | | $Na^+$ | | $K^+$ | | $Na^+$ | | | $K^+$ | | |
| Bentonite | — | | 0.071 | | — | | 0.981 | | — | | | 0.960 | | |
| Vermiculite | — | | 0.940 | | — | | 0.970 | | — | | | 0.920 | | |
| Soil | 0.028 | | 0.430 | | 0.92 | | 0.990 | | 0.997 | | | 0.920 | | |
| Empirical constants | $b$ | $k$ | $b$ | $k$ | $K$ | $N$ | $K$ | $N$ | $A$ | $\beta$ | $K_d$ | $A$ | $\beta$ | $K_d$ |
| Bentonite | — | — | 370.4 | 0.002 | — | — | 0.68 | 1.08 | — | — | — | 0.14 | 1.91 | 0.05 |
| Vermiculite | — | — | 107.7 | 0.046 | — | — | 5.75 | 0.72 | — | — | — | 3.01 | 1.49 | 0.13 |
| Soil | 434.8 | 0.002 | 222.2 | 0.014 | 0.18 | 1.35 | 0.89 | 0.92 | 0.009 | 2.66 | 0.044 | 0.23 | 1.64 | 0.04 |

[a] Units: $K_d$ in mL/g; $b$ in mg/g; $k$ in mL/mg; $K$, $N$, $A$, and $\beta$ have units corresponding to $S$ in mg/g and $C$ in mg/L.

was made in the case of $Na^+$ for soil alone. Linear regression analysis yielded the empirical parameters and correlation coefficients for the model equations, which are presented in Table 3. The Langmuir isotherms for data obtained for sorption of $Na^+$ on soil and sorption of $K^+$ on the selected backfill materials and their regression plots are presented for comparison in Fig. 1. Similar plots for the Freundlich isotherm and the Modified Freundlich isotherm are presented for comparison in Figs. 2–5.

From Fig. 1 it is observed that the Langmuir equation did not provide a linear fit except for sorption of $K^+$ on vermiculite. Also, based on the correlation coefficients ($R^2$) obtained from the regression analysis (Table 3), it is seen that the Langmuir isotherm equation did not explain the sorption phenomenon of $Na^+$ on soil, sorption of $K^+$ on bentonite, and sorption of $K^+$ on soil as compared to $R^2$ obtained for the Freundlich and the Modified Freundlich isotherm equation. In the case of $K^+$ sorption on vermiculite, $R^2$ was marginally higher compared to the Modified Freundlich isotherm equation.

From the analysis of sorption of $Na^+$ by soil (Fig. 2) it was observed that the Modified Freundlich equation appeared to fit the sorption data better. Also, by interpreting the results based on the $R^2$ data tabulated in Table 3,

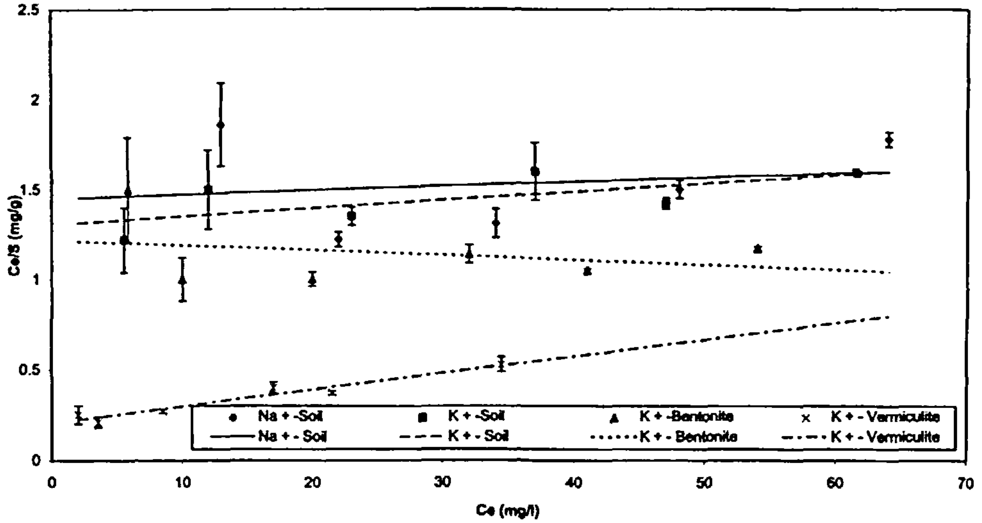

FIG. 1 Langmulr isotherm for sorption of $Na^+$ and $K^+$ on backfill materials.

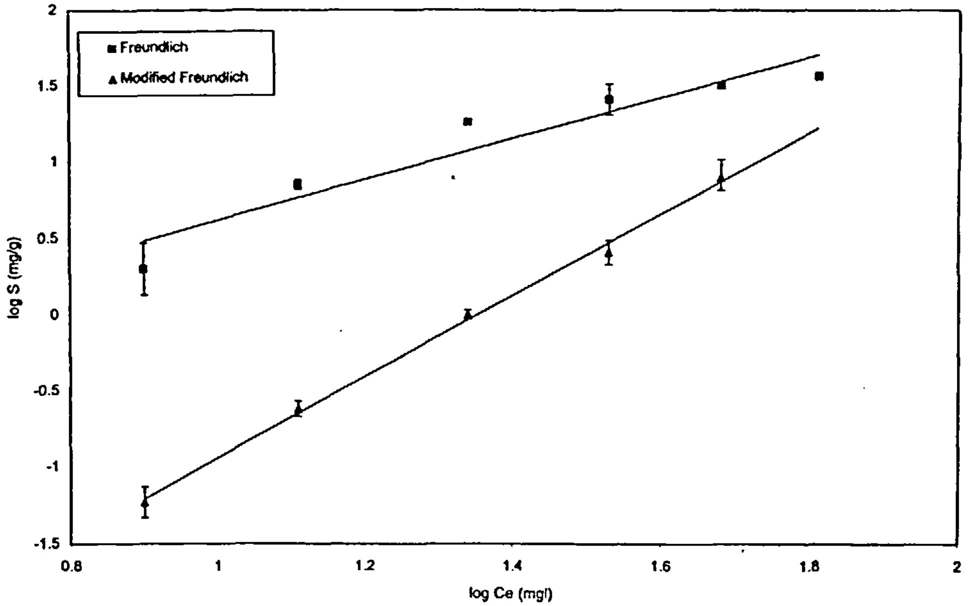

FIG. 2 Model fitting for $Na^+$ sorption on soil.

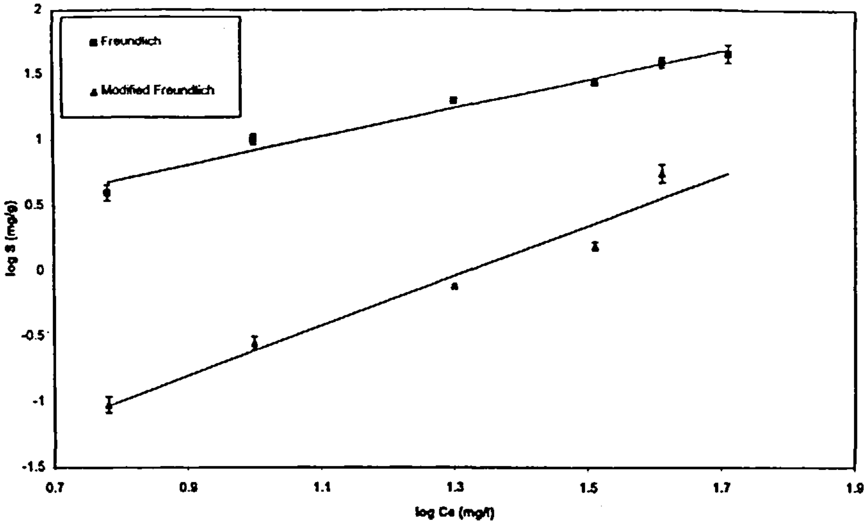

FIG. 3 Adsorption isotherms for $K^+$ sorption on bentonite clay.

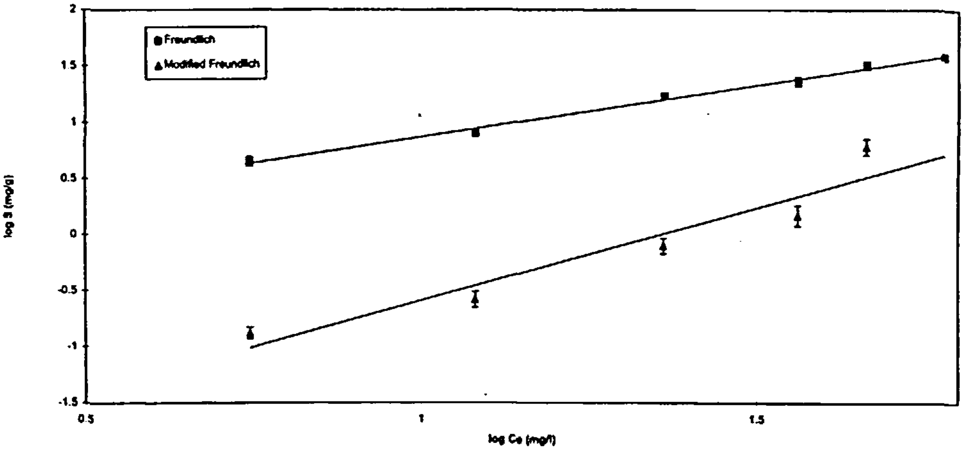

FIG. 4 Adsorption isotherms for $K^+$ sorption on soil.

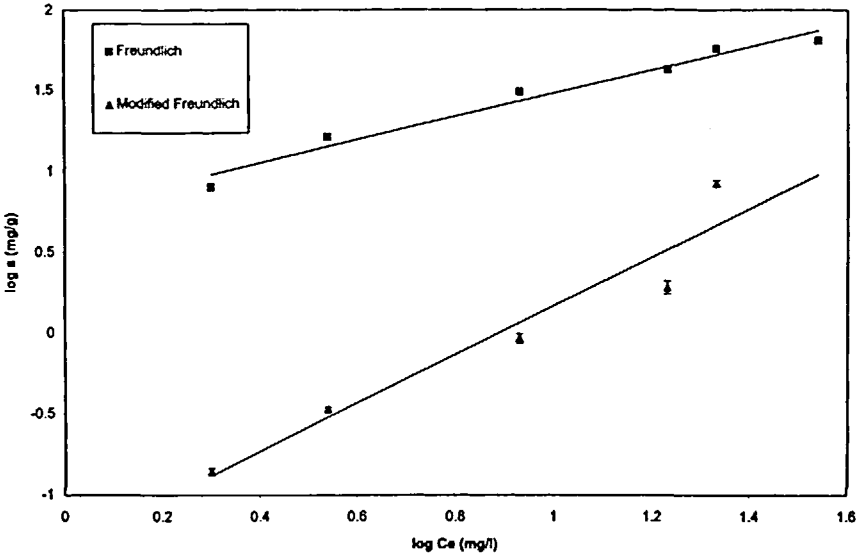

FIG. 5 Adsorption isotherms for $K^+$ sorption on vermiculite.

it was concluded that the Modified Freundlich equation explains the sorption phenomenon better than the Freundlich isotherm equation. From the sorption analysis of $Na^+$ for soil by the Modified Freundlich model, the value of its empirical constants $A$ was found to be 0.009 mL/g and $\beta$ was found to be 2.66. The empirical constant $A$ is a measure of the weighted average distribution coefficient and $\beta$ (coefficient of variation) denotes the spread of the distribution coefficient about the average $K_d$; the value of $\beta$ given above is greater than 1, indicating a higher spread. The average $K_d$ from the above equation was found to be 0.044 mL/g. A negative sorption was observed for the uptake of $Na^+$ by the commercial clay materials (vermiculite and bentonite clay) in the laboratory experiments, hence interpretation of the data is not discussed further. The $K_d$ values (Table 3) obtained using the Modified Freundlich isotherm are found to be less than 1, which indicates that these ions will not compete for sorption onto backfill materials in the presence of radionuclides (19).

A low $K_d$ of Na and K as such does not indicate that the $K_d$ for waste nuclides for vermiculite will be higher or for the other sorbents. It has been reported by Stefanova and Gradev (4) that selectivity of vermiculite for $^{137}Cs$ is high and satisfactory for $^{90}Sr$, $^{60}Co$, $^{56}Fe$, $^{144}Ce$, and $^{106}Ru$ in the presence of sodium, magnesium, and calcium, and low for these ions in the presence

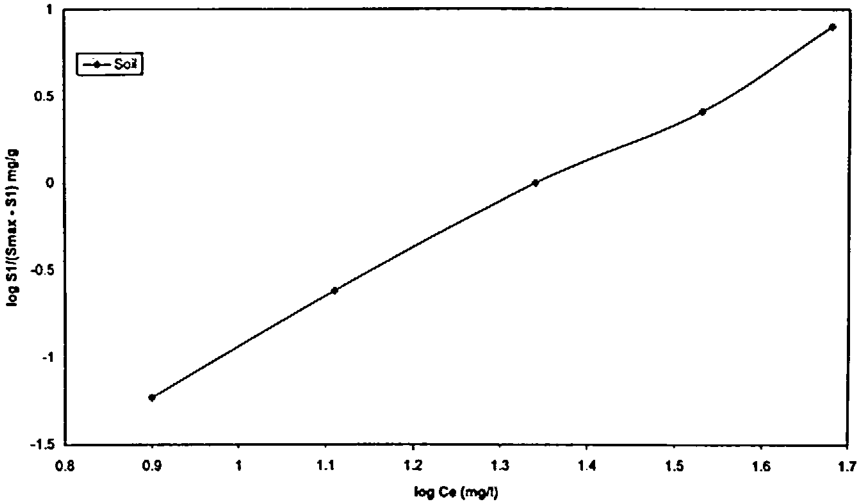

FIG. 6 Hill plot analysis for $Na^+$ sorption on soil.

of potassium. Sikalidis et al. (8) studied the selectivity of cesium and its fixation on vermiculite in the presence of competing cations. It has also been reported that vermiculite and bentonite prefer polyvalent and bivalent ions (4). The selectivity for cesium by bentonite clay decreases in the order $K^+ > Ca^{2+} > Mg^{2+} > Na^+$ (5).

It has been reported that the $K_d$ values for sorption of $^{137}Cs$, $^{90}Sr$, $^{60}Co$, $^{56}Fe$, $^{106}Ru$, and $^{144}Ce$ on vermiculite are in the $10^2$ to $10^3$ mL/g range (4). The $K_d$ values for sorption of $^{137}Cs$ and $^{90}Sr$ on bentonite were found to be 1500 and 60 mL/g, respectively (6). The $K_d$ values for sorption of $^{137}Cs$ and $^{90}Sr$ on montmorillonite and soil were found to be 3500 and 1500 mL/g and 27,000 and 400 mL/g, respectively (20).

The $K_d$ values for sorption of $K^+$ on soil, vermiculite, and bentonite were found to be 20 ± 0.5, 115.5 ± 3.1, and 24.9 ± 0.8 mL/g, respectively. The $K_d$ value for sorption of $Na^+$ on soil was found to be 15 ± 0.8 mL/g. The variation of $K_d$ values for $Na^+$ and $K^+$ on soil estimated from analysis of the Modified Freundlich isotherm was found to be rather small (Table 3), which was also the case for $K_d$ values estimated in the laboratory.

From the analysis of sorption of $Na^+$ by soil, using the Hill plot approach (Fig. 6 and Table 4), it was observed that irrespective of $Na^+$ concentration, the slope was greater than 1, suggesting that sorption of $Na^+$ at one exchange site enhances sorption at the other exchange site (18).

TABLE 4
Slopes for Hill Plots

| Backfill material | Ion: $Na^+$ Low concentration (10–40 ppm) | $Na^+$ High concentration (60–100 ppm) | $K^+$ Low concentration (10–40 ppm) | $K^+$ High concentration (60–100 ppm) |
|---|---|---|---|---|
| 1. Bentonite | — | — | 1.73 | 2.56 |
| 2. Vermiculite | — | — | 0.71 | 2.07 |
| 3. Soil | 2.79 | 2.62 | 1.24 | 2.65 |

From Table 3 it can be seen that $R^2$ data for the Freundlich isotherm are higher than for the other two isotherms studied for sorption of $K^+$ on all the three selected backfill materials. The $K$ and $N$ values obtained from the Freundlich isotherm are tabulated in Table 3. The $K$ represents the predicted quantity of $K^+$ sorption per gram of the sorbent/backfill material at unit equilibrium concentration. $K$ was found to be the highest for vermiculite, which indicates that vermiculite is the most efficient sorbent. From the values of $N$ it was observed that $K^+$ sorption on bentonite and soil is concentration-independent, the values being 1.08 and 0.92 for bentonite and soil, respectively. The value of $N$ for $K^+$ sorption on vermiculite was observed to be concentration-dependent, the value being 0.72.

From the analysis of sorption of $K^+$ using the Hill plot (Fig. 7 and Table 4), it was observed that the region of low saturation (10–40 mg/L) yields a slope of less than 1 for vermiculite, and a slope greater than 1 for soil and bentonite clay. In the case of vermiculite, sorption at one site impedes sorption at other sites (negative interaction), whereas soil and bentonite are cases of positive interaction where sorption at one site enhances sorption at other sites. Analysis of the region of higher saturation (60–100 mg/L) yields a slope greater than 1 for all the backfill materials, indicating that sorption at one site enhances sorption at other sites (positive interaction). It is concluded that these backfill materials can serve as good barriers which attenuate the pollutants effectively at higher levels (60–100 mg/L).

As discussed earlier, sorption of $K^+$ by vermiculite was found to be concentration-dependent from the Freundlich model, and this was confirmed from analysis using the Hill plot approach. The Freundlich isotherm equation itself is applicable over a limited concentration range, and the Hill plot approach is applicable over an even smaller concentration range. The negative interaction for vermiculite at low concentrations may be attributed to slow diffusion

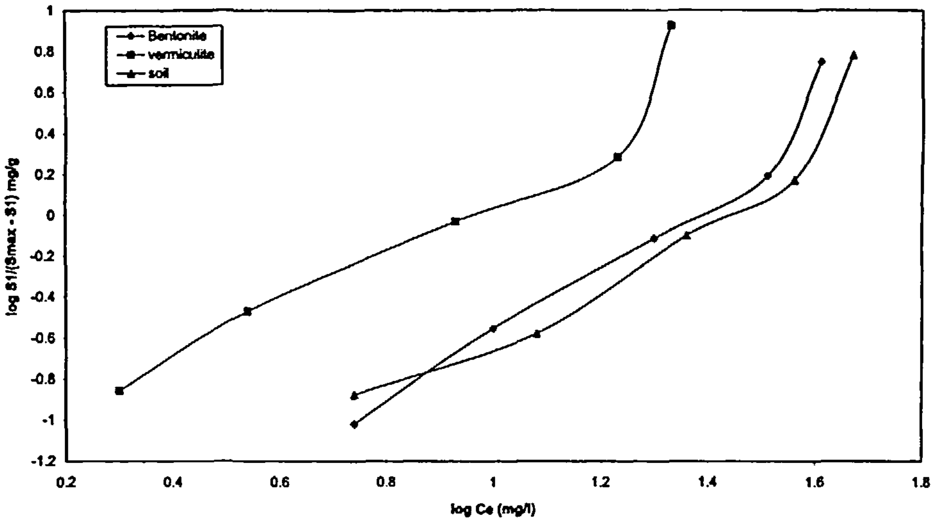

FIG. 7 Hill plot analysis for $K^+$ sorption on bentonite clay, vermiculite, and soil.

of large solvated $K^+$ ions into interlattice planes. The solvation of $K^+$ at higher concentrations becomes relatively less and promotes interaction among sorption sites. Sparks (21) reported that potassium sorption rates on montmorillonite (bentonite clay) were rapid compared to sorption rates on vermiculite. It was explained that the sorption phenomenon on vermiculite can occur by the three following mechanisms: 1) rapid adsorption on the sorption sites on external planes, 2) intermediate adsorption rates at the sorption sites on the edges, and 3) slow diffusion followed by sorption on the interlattice sorption sites.

It has been reported that desorption of radionuclides from loaded vermiculite is higher than that from bentonite, which is related to the more open structure of vermiculite (4). It has been reported that 95.5% of sorption of cesium on bentonite is irreversible due to the structure of montmorillonite which is the principal clay mineral in bentonite (5). Because of large basal spacings, the layers in montmorillonite are not bound as tightly as those in other type of clays (i.e., kaolinite), and this allows solutions in between the layers for sorption to occur.

## CONCLUSIONS

1. From evaluation and comparison of the Langmuir, Freundlich, and Modified Freundlich isotherm equations, it was concluded that the Freundlich

isotherm and the Modified Freundlich equations explain the sorption phenomenon more accurately for both $K^+$ and $Na^+$ ions compared to the Langmuir isotherm.

2. From the sorption data analysis for $K^+$, it was concluded that vermiculite is the most efficient sorbent, and that sorption of $K^+$ on bentonite and soil is independent of concentration, whereas sorption on vermiculite is concentration-dependent.
3. From Hill plot analysis it was concluded that all the mentioned backfill materials can serve as good barriers in the 10 to 100 mg/L concentration range for $Na^+$ and in the 60 to 100 mg/L concentration range for $K^+$. Hill plot analysis can be used to predict the mechanism of interaction of sorption sites with respect to concentration.

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