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Removal of ^{137}Cs by Some Saudi Natural Clays

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The sorption of cesium, from aqueous solution by natural clays is a very fast reaction and seems to be under film diffusion mechanism. Cesium exchanges weakly with clays of high Ca and Mg contents but strongly with silico-aluminium clays of low Ca and Mg contents. Sorption of cesium by these clays has been studied under different experimental conditions, pH, shaking time, etc. Heat and acid treatments failed to improve the uptake quality of the clays.

INTRODUCTION

Cesium is one of the alkali-metals which are the most reactive group of metals. It has many radioisotopes, the most important of which are ^{137}Cs and ^{134}Cs. Both are fission products, the half-life of the first isotope is *ca.* 30 years and the energy of its γ line is *ca.* 662 kev. The second isotope has a half-life of *ca.* 2.1 years and various photon energies of 569, 604 and 796 keV. The radiotoxicity of ^{137}Cs is classified as medium and this isotope has many important applications, such as radioimmunological tests, therapeutic treatment, industry research . . . etc.[1]

There has been a good deal of work concerning the sorption of cesium by natural and synthetic exchangers. Thus, Brat[2] has used vermiculite and mica for the treatment of radioactive waste containing ^{137}Cs and ^{90}Sr. Thomas[3] has reviewed the research and developments for the treatment of low and intermediate levels of radioactive wastes using vermiculite, bentonite, certain earths and clinoptilolite. The results indicated that these clays are useful in removing Cs, Sr and other fission products. In addition, Mitry and others[4] used natural clays, mainly montomorillonite with traces of chlorite and illite to study the uptake of long-lived fission products taking into account different factors such as pH, and contact time. Moreover, the sorption of radionuclides on natural materials used for cementation of liquid radioactive wastes has been carried out[5].

The use of synthetic exchangers to remove cesium from contaminated solutions has also attracted the attention of many workers. For example, Ampelogova[6] and Crispino[7] have studied the sorption of ^{137}Cs by metal oxides, such as magnetite, hematite and the oxides of Cr, Zr and Ti.

Regarding the local natural clays, it has been shown that the smectite type clay shows the highest uptake (*ca.* 60%) in the pH range 4–9 within 5 min. reaction time. The heat treatment of the clay to 450°C has improved the uptake by *ca.* 13%.[8]

EXPERIMENTAL

Cesium nitrate, analytical grade, from BDH Chemical Ltd. and radioactive cesium chloride in 1M HCl from Amersham International have been used in this work.

The clay samples were taken from different places from Saudi Arabia Kingdom. The chemical composition of these clays is shown in Table 1. The clays were crushed, sieved and only the size fraction (0.5–0.8 mm) was used.

The dried clays were heated for 2 hrs at various temperatures (200–800°C) in a muffle furance, cooled again at room temperature and used as such in our experiments.

Technique

2 gms of clay sample was contacted with 20 ml of 10^{-3}M radioactive cesium nitrate solution with continuous shaking, the uptake by clay was followed by measuring the activity of 1 ml of clear solution at different time intervals. Measuring the activity was carried out using CANBERRA Series 35 plus MCA connected to NaI (TL) detector Model 727. The percent uptake was calculated as

$$\%\ \text{uptake} = \frac{A_0 - A_t}{A_0} \times 100$$

where A_0 is the original activity, A_t is the activity at time t.

Doubly distilled water was used in all experiments which were carried out in duplicate and when neccessary in triplicate and average values of percentage removal were obtained.

RESULTS AND DISCUSSION

Table 2 shows the variation of % uptake as a function of shaking time. The least possible time of shaking is 1 min. It is clear that the uptake is very fast. Three clay samples show a very good uptake ($\geqslant$ 95%) while two show a low uptake. In all cases the results obtained show that the uptake by used clays is between 0–7 and 1–5 mmol/100 g of clay. C_1, mainly Ca and Mg carbonate clay (nearly 40%), as well as C_2 (55%) resisted exchange with Cs^+ as most of their exchangable sites are saturated with alkaline earth elements, C_4, C_5 and C_6 which are alumino-silicate with only small contents of CaO and MgO were best suited for cesium exchange with an uptake value greater than 95% which may be considered as a high capacity. Although C_7 has a high content of SiO_2 and Al_2O_3, its uptake is lower than expected compared with C_4, for example. It is possible that there is some change in the structure of the clay or some impurities are causing such behaviour.

Figure 1 shows the uptake of cesium ions by some clays at different

TABLE 1

CHEMICAL COMPOSITION OF LOCAL CLAY SAMPLES

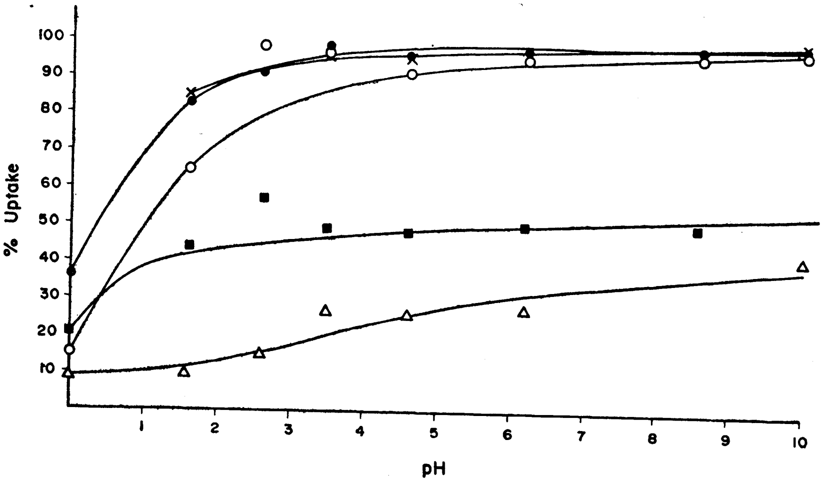
Sample	Colour	% Chemical composition										
		SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	H_2O	L.O.I.
C_1	Light yellow	7.38	3.68	5.66	23.4	16.63	1.21	1.05	0.05	—	1.05	39.07
C_2	White	1.6	0.35	0.12	55.48	0.07	0.07	0.07	$\leqslant 0.05$	13.85	0.76	27.08
C_3	White	58.49	0.15	0.29	22.57	0.09	0.23	$\leqslant 0.05$	$\leqslant 0.05$	12.96	1.57	3.36
C_4	Red Shale	49.35	25.13	10.17	0.64	1.17	0.44	2.62	1.40	—	—	9.08
C_5	White	58.42	24.43	4.28	0.5	0.18	0.66	0.28	1.69	—	—	9.26
C_6	Variegnated Shale	58.07	27.82	10.67	0.65	0.47	0.57	0.55	1.20	—	—	9.00
C_7	White (Kaolinite)	44.61	34.84	1.51	1.35	0.09	1.21	0.10	2.16	—	—	14.13
C_8		46.49	32.98	3.01	1.01	0.06	0.70	0.06	2.78	—	—	12.91
C_{10}	Gray	38.8	16.7	5.2	4.5	—	12.5	13.0	—	—	1.5	2.0
C_{11}	Brown	47.04	12.28	17.97	3.40	1.40	0.03	3.0	2.50	0.06	1.88	8.72

TABLE 2

EFFECT OF CONTACT TIME ON THE UPTAKE OF Cs^+ BY NATURAL CLAYS [$CsNO_3$] 10^{-3} M
Velocity of shaking = 125 vibrations min^{-1}

Natural clay	Time of shaking				
	1 min	5 min	30 min	60 min	120 min
C_1	55.1	54	56.2	55.3	
C_2	25.8	26.9	24.8	25.4	25.5
C_4	94.5	96.4	95.6	96.4	96.4
C_5	95.4	96.4	96.4	96.4	97.2
C_6	94.5	93.7	97.6	96.0	94.2
C_7	74.3	78.6	79.4	78.6	78.8

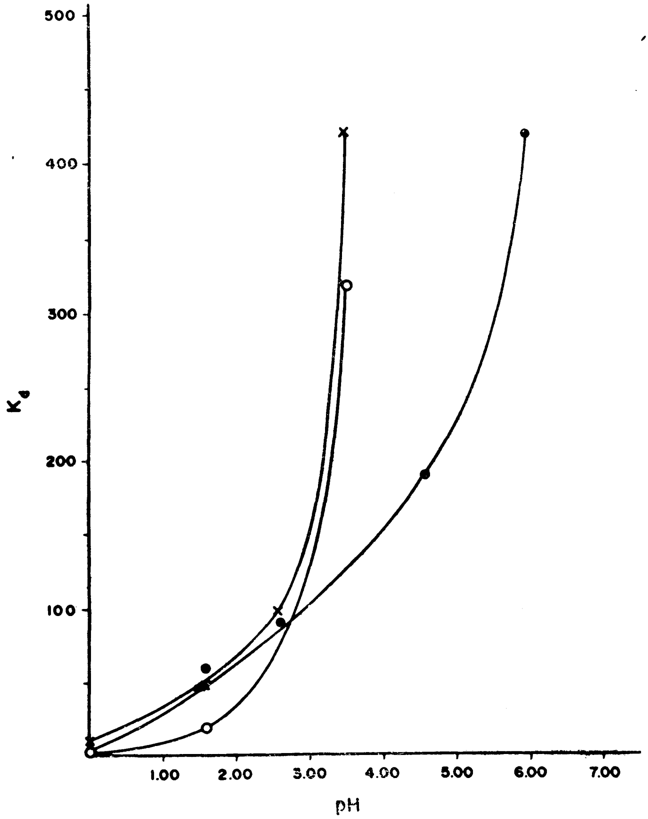
initial pH values. From the figure we can see that the uptake seems to increase with the increase of pH while it is always constant after pH 3.5. The low uptake at low pHs is attributed to the competition by H^+ and it is clear from the figure that the % uptake becomes appreciable at pH $\geqslant$ 3 i.e. at [H^+] *ca.* 10^{-3}M which means that Cs^+ starts to compete effectively with H^+ when their concentrations are about equal and this behaviour is unusual, since the diffusion coefficient of C^+ is less than that of H^+. It is also clear that the equilibrium uptake is attained at different pH values and some clays show some kind of upward curvature. From



■ C_1, △ C_3, ○ C_4, × C_5 and ● C_6

Fig. 1. Variation of % uptake of 10^{-3}M Cs^+ with pH. Contact time = 30 min., shaking velocity = 125 min^{-1}

the % uptake-pH curve the distribution coefficient K_d can be obtained and the plot of K_d-pH for some clays is shown in Fig. 2. The rapid increase of K_d with pH may be related with a rapid increase of the ionization of the hydrogens of the exchangers.



○ C_4, ×C_5 and ● C_6

Fig. 2. Plot of distribution coefficient, K_d, with pH. $[Cs^+] = 10^{-3}M$. Contact time = 30 min, shaking velocity = 125 min^{-1}

The effect of $[Cs^+]$ on its uptake was investigated in the concentration range 10^{-4}–10^{-2} M and the results are shown in Table 3. The data of Table 3 was subjected to Freundlich isotherm which can be written in the form:

$$\log C_{ad} = \log A + \frac{1}{n} \log C_{Bulk}$$

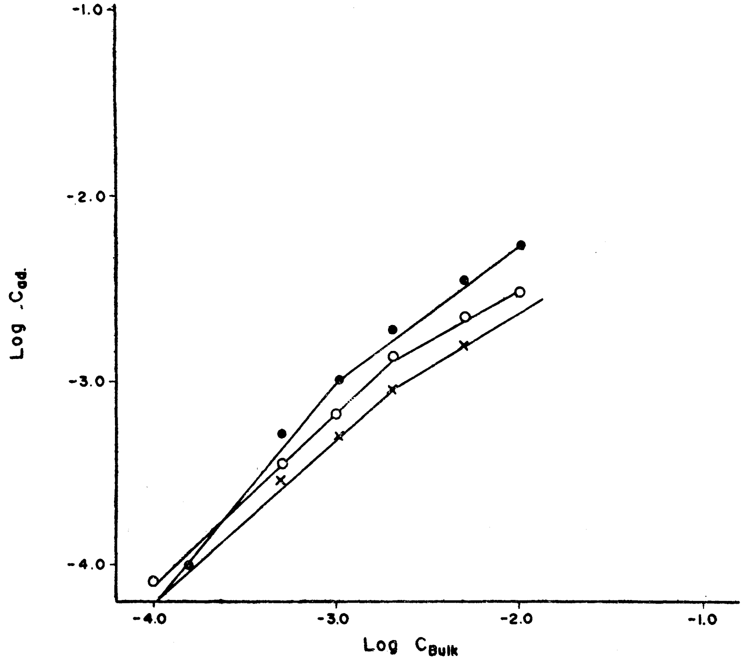
where C_{ad} is the amount of Cs^+ adsorbed on clay, B_{Bulk} is the total concentration of the ion in solution before adsorption and A and n are constants. The plot of the above relationship gave two straight lines indicating the operating of two mechanisms as can be seen in Fig. 3.

TABLE 3

EFFECT OF CONCENTRATION OF [$CsNO_3$] ON THE UPTAKE OF Cs^+ BY NATURAL CLAY

Initial pH = 2.95, Temperature (25 ± 1)°C

Clay	% Uptake at [$CsNO_3$]M					
	10^{-4}	5×10^{-4}	10^{-3}	2×10^{-3}	5×10^{-3}	10^{-2}
C_1	58.8	57.3	48.6	44.6	30.7	29.8
C_3	21.4	12.0	12.3	16.4	15.3	14.3
C_4	97.9	96.4	96.4	90.0	73.4	57.0
C_5	96.4	96.4	96.4	93.9	82.8	66.6
C_6	96.6	94.8	94.8	94.2	87.5	78.8
C_7	84.8	81.1	77.2	69.2	51.0	25.2
C_{10}	81.4	62.9	55.1	41.7	21.7	20.6



○ C_4, × C_5 and ● C_6

Fig. 3. Freundlich adsorption isotherm of Cs^+ by some clays

Table 4 shows the effect of thermal treatment of the clays on their uptake. It is clear that, for some clays, the uptake is decreasing a few % between room temperature and 200°C. At higher temperatures, the decrease with raising temperature is not the same. Thus for C_4 and C_6

TABLE 4

EFFECT OF THERMAL TREATMENT ON THE UPTAKE OF 10^{-3} M CS+

Time of shaking = 60 min., shaking velocity = 125 min^{-1}

Clay	Uptake %					
	25°C	100°C	200°C	400°C	600°C	800°C
C_2	30.0	28.0	28.0	25.0	26.0	20.0
C_3	25.5	24.0	24.5	25.0	23.0	23.0
C_4	96.4	94.0	92.9	93.6	79.7	52.3
C_5	96.4	97.8	93.9	69.7	72.3	29.0
C_6	97.7	95.8	95.3	94.8	83.3	36.1
C_7	78.6	74.0	70.0	63.9	46.8	35.2

the uptake is very high (94%) at 400°C and decreases to 80% at 600°C. The appreciable decrease is observed at 800°C. This behaviour indicates that these clays are stable against degradation at least upto 600°C. For C_5 and C_7 the appreciable decrease was observed at *ca.* 600°C which means that the stability of the last two clays is not as good as for C_4 and C_6. The rest of clays have low uptake at all temperatures.

Two clays were examined to see the effect of added ions on the uptake of Cs^+ by these clays. It is clear from Table 5 that the general behaviour is that all added ions reduce the sorption of Cs^+ though the

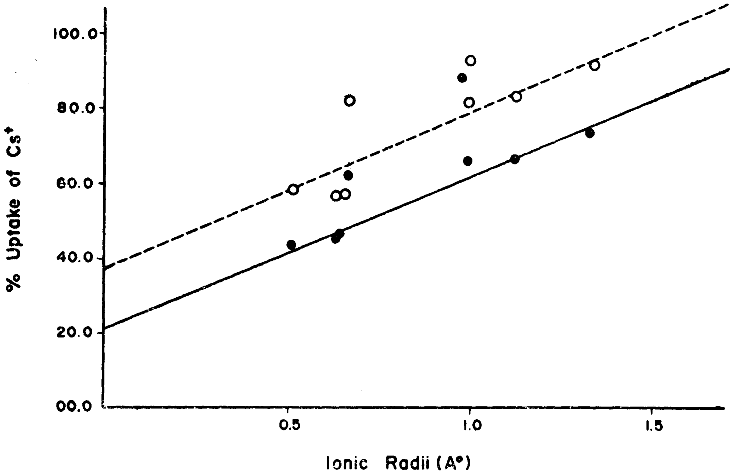
TABLE 5

EFFECT OF COMPETING IONS ON SORPTION OF Cs^+ BY SOME NATURAL CLAYS

$[CsNO_3] = 10^{-3}M$, Initial pH = 2.58

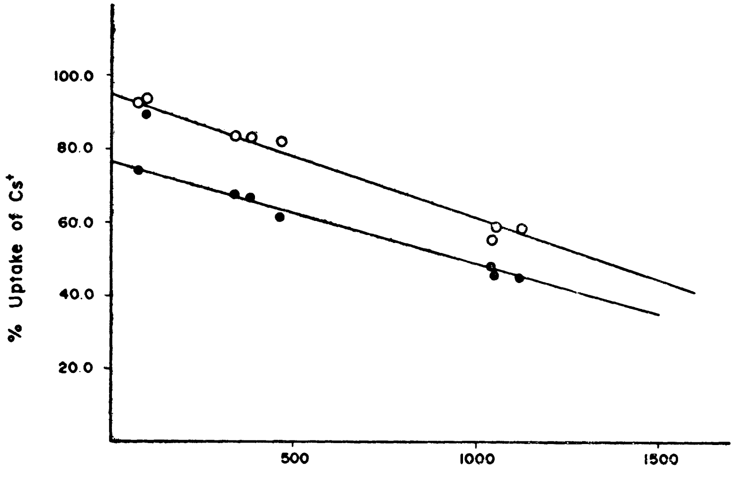
Competing ion	Natural clay	Normal	Concentration of competing ion		
			10^{-3} M	5×10^{-3} M	10^{-2} M
Na^+	C_4	96.4	86.0	88.0	89.0
	C_6	97.7	93.5	94.6	93.6
K^+	C_4	96.4	87.8	82.1	74.8
	C_6	97.7	95.4	93.6	92.4
Mg^{+2}	C_4	96.4	81.6	70.7	62.8
	C_6	97.7	93.8	86.1	82.1
Ca^{+2}	C_4	96.4	79.6	73.2	66.5
	C_6	97.7	93.0	88.5	83.1
Sr^{+2}	C_4	96.4	82.9	69.8	67.7
	C_6	97.7	87.3	91.0	83.6
Al^{+3}	C_4	96.4	63.0	45.3	44.8
	C_6	97.7	89.1	71.4	59.6
Cr^{+3}	C_4	96.4	55.3	53.1	45.5
	C_6	97.7	76.4	70.1	58.3
Fe^{+3}	C_4	96.4	74.9	46.7	46.8
	C_6	97.7	80.4	65.6	56.3

competitive sorption of these ions is not identical. Thus we can easily see that the effect of these ions can be classified in the following order: trivalent ⩾ divalent ⩾ monovalent. Within each group, the change in sorption is very small to be used for the conclusion of a well defined relationship; it is, however, interesting to mention that a good linear relationship exists between the uptake of Cs+ and ionic radii and an inverse relationship between the uptake of Cs+ and the change in hydration enthalpy of these ions. The results are shown in Figs. 4 and 5. In



○ C₄ and ● C₆

Fig. 4. The relationship between % uptake of Cs+ and ionic radii of added ions. [Cs+] = 10^{-3} M; [ion] = 10^{-2} M.



○ C₄ and ● C₆

Fig. 5. The relationship between % uptake of Cs+ and hydration enthalpy of added ions

the two clays examined, Na^+ and Mg^{2+} show some deviation from the established relationships but this is not significant in such complicated systems.

Table 6 shows the variation of Cs^+ uptake with the type of acid used in treating the clays. The most interesting point is that the clays containing phosphate are less effective in removing Cs^+ after their treatment with all acids. For other clays the Cs^+ uptake depends on the acid used for the treatment. It can be generally concluded that the acid treatment by HCl and HNO_3 gave a good uptake which is comparable to the untreated clays and these two acids have almost the same effect. For H_2SO_4 and H_3PO_4 the uptake is also comparable but it is less than for the untreated clays. In conclusion, the acid treatment is not recommended since it results in no improvement in the uptake of Cs^+.

TABLE 6

EFFECT OF ACID TREATMENT OF NATURAL CLAYS ON THE UPTAKE OF 10^{-3} M Cs^+

Shaking velocity = 125 min^{-1}
Time of shaking = 30 min

Clay	% uptake for clay treated with			
	HCl	HNO_3	H_2SO_4	H_3PO_4
C_2-	16.0	28.0	1.0	26.0
C_3	7.0	11.0	2.0	10.0
C_4	84.8	83.5	80.9	68.3
C_5	87.4	95.9	69.5	58.1
C_6	93.1	84.6	58.8	75.6
C_9	90.0	86.8	37.9	35.1

Table 7 presents some results obtained when 10^{-3} M Cs^+ was shaken with 1 g of these exchangers at different pHs. It can be seen that, in general, the uptake is not affected, for most exchangers, by pH change from 2.5 to 10.0. It is also obvious that only three have an uptake of 100% and one has an uptake of *ca.* 70% (basic Al_2O_3). The rest of exchangers have much lower uptake and the dependence on pH is not uniform. It is worth mentioning that the acid and neutral form of Al_2O_3 have very low uptakes. Silica gel, iron oxides and titanium oxide have low uptakes, *i.e.* in the range of *ca.* 30%. Since these oxides are the main constituents of the natural clays and because of the low uptakes of these oxides, this leads to the conclusion that the complexation of these oxides in the network of the clays plays an important role in improving their sorption capacities.

TABLE 7

SORPTION OF 10^{-3} M Cs^+ BY SOME SYNTHETIC EXCHANGERS AND OXIDES

Shaking velocity = 125 min^{-1}
Shaking time = 30 min

Exchanger	% uptake at pH		
	2.5	5.5	10.00
Dowex-50	99.7	99.6	99.6
Al_2O_3 acidic	10.9	4.3	2.7
Al_2O_3 Neutral	2.1	16.4	12.3
Al_2O_3 Basic	64.9	68.9	69.2
Fe_2O_3	17.7	13.5	22.2
Fe_2O_3 calcined	27.5	33.4	—
Fe_3O_4	20.0	21.4	30.5
TiO_2	30.7	34.8	30.4
Amberlite IRA 400	22.9	66	—
Amberlite 129	96.8	98.5	—
Silica gel	14.4	32.5	—
Molecular Sieve	98.9	99.2	99.4

It is possible, therefore, to conclude that for Cs^+ the natural clays are better decontaminants than many synthetic exchangers. To this is added the advantage of being cheap and needs simple or no treatment.

Effect of Organic Medium

The effect of medium on the uptake of Cs^+ by natural clays has been examined, using 50 and 100% ethanol and comparing the results with water as solvent. The results are shown in Table 8. It is clear that there is no detectable change for mostly all clays. It is not possible, at this stage, to explain the present observations concerning the similarity in the uptake of Cs^+ from aqueous and organic solvents. This similarity could be due to different effects counterbalancing each other. For example, in aqueous solutions the hydration of ions may favour sorption but the size of hydrated ion will be bigger than unhydrated ion which may render the sorption process. On the other hand, the smaller size of unhydrated ion will make its accommodation by the clay lattice easier but the loss of hydration may result in lesser affinity for sorption.

TABLE 8

SOLVENT EFFECT ON THE UPTAKE OF Cs^+ BY NATURAL CLAYS $[Cs^+] = 10^{-3}M$

Velocity of shaking = 125 min^{-1}
time of shaking = 60 mins

Natural clay	% uptake for the solvent		
	Water	50% Ethanol	100% Ethanol
C_1	54.8	50.0	48.5
C_2	30.0	28.5	25.6
C_4	97.0	83.9	98.4
C_5	96.0	91.2	98.1
C_6	97.7	92.4	98.6
C_7	79.0	89.4	96.8

Kinetics of Cs^+ Sorption

The uptake of Cs^+ seems to be dominantly under film diffusion control. This conclusion is based on the fact that no time effect was observed even if the time of contact was as short as 1 min, which is the least measured time for the present technique. In addition, there was no effect of shaking velocity on the uptake of Cs^+ when the velocity was changed considerably at different times of contact. The result of velocity is shown in Table 9.

TABLE 9

Initial pH = 5.95, final pH = 7.48,
$[CsNO_3] = 10^{-3}M$, temperature = 25 ± 1°C

Clay	Static 60 min	30 min^{-1}		75 min^{-1}	125 min^{-1}		225 min^{-1}
		3 min	60 min	3 min	3 min	60 min	3 min
C_1	44.0	46.0	48.4	47.0	54.0	48.6	55.0
C_2	17.0	13.0	22.7	11.0	16.0	25.3	14.0
C_4	84.0	90.0	93.6	84.0	93.0	96.4	93.0
C_5	94.0	96.0	86.9	96.0	96.0	97.7	97.0
C_6	82.0	76.0	86.9	80.0	95.0	97.7	98.0
C_7	70.0	65.0	72.4	63.0	74.0	77.2	80.0

Moreover, the effect of solution temperature on the uptake of Cs^+ by some natural clays was examined and very little change was observed upon changing temperature from 5 to 50°C as can be seen in Table 10. This may support the conclusion that film diffusion mechanism is dominant.

The fact that there is no effect of temperature may indicate that the rate determining step needs small activation energy.

TABLE 10

EFFECT OF SOLUTION TEMPERATURE ON THE UPTAKE OF Cs^+ BY NATURAL CLAYS SHAKING BY HAND

$[CsNO_3] = 10^{-3}$ M

Clay	Solution temperature ± 1°C		
	5.0	25.0	50.0
C_1	38.5	49.0	42.8
C_4	90.7	97.7	95.4
C_6	86.2	96.0	92.9

The sorption of Cs^+ by natural clays appears to be simpler than other ions, mainly due to the relative simplicity of the chemistry of this ion as indicated by the absence of pH effect. However, this simplicity is not as it appears from the first sight. This can be shown by using simple competition kinetics. Thus if the available site for sorption is given the symbol S_t, then the following reactions may be written:

$$S_t + Cs^+ \xrightarrow{k_1} Cs^+_{ad}$$

$$S_t + X \xrightarrow{k_2} P$$

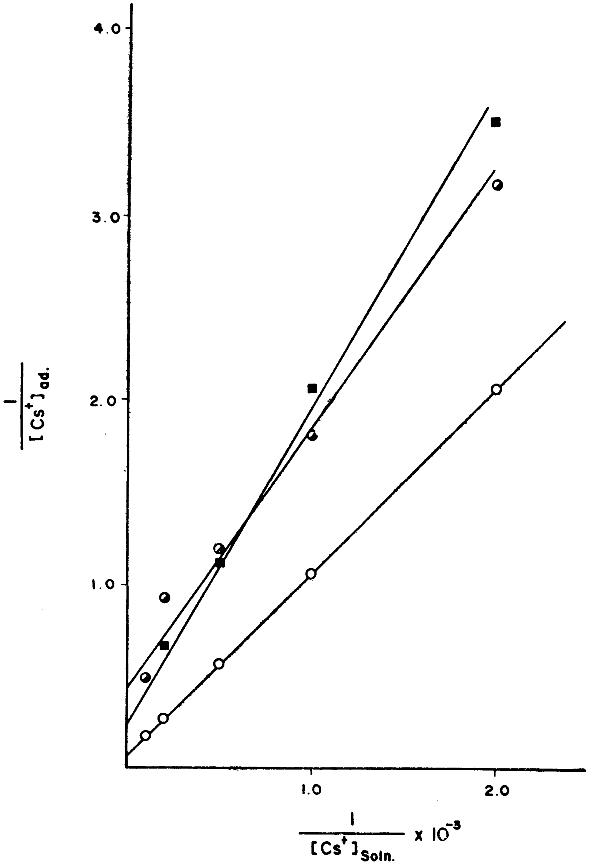
where Cs^+_{ad} will be the sites occupied by Cs^+ (simply the amount of Cs^+ sorbed) and X is another species competing with Cs^+ to give the products P (simply the total sites minus those occupied by Cs^+). The rate constants of the respective reactions are k_1 and k_2. Now, we can write:

$$[Cs^+]_{ad} = S_t \frac{k_1[Cs^+]_{soln}}{k_1[Cs^+]_{soln} + k_2[X]_{soln}}$$

This equation can be arranged to:

$$\frac{1}{[Cs^+]_{ad}} = \frac{1}{S_t}\left[1 + \frac{k_2[X]_{soln}}{k_1[Cs^+]_{soln}}\right]$$

where $[Cs^+]_{ad}$, $[Cs^+]_{soln}$ and $[X]_{soln}$ are the concentrations of Cs^+ sorbed, Cs^+ added originally and the competing species present initially. If these assumptions are correct, then a plot of $\frac{1}{[Cs^+]_{ad}}$ versus $\frac{1}{[Cs^+]_{soln}}$ should give a straight line. Fig. 6 shows that the this is in fact the case. The fact that the pHs of solutions under investigation are all about 6 indicate that the competing species is not H^+, since its concentration is at least two to three orders of magnitude less than $[Cs^+]$. In addition, the ratio k_2/k_1 can be evaluated from the graph and the obtained value was much higher than could be accounted for by the ratio of the diffusion coefficient of H^+ and Cs^+.



■ Cl, ○ C_4 and ◑ C_{10}.

Fig 6. Plot of $\frac{1}{[Cs^+]_{ad}}$ against $\frac{1}{[Cs^+]_{soln}}$ for the absorption of Cs^+ by some clays

For this reason one is forced to conclude that another species is competing effectively for sorption by these clays. If the clays are behaving as anionic exchangers, then NO_3^- may be the candidate for this process. If, however, some complexes are being formed, they may be sorbed simultaneously with Cs^+.

It is known that adsorption is a surface phenomenon and it is affected by many factors, such as the structure of the solid surface, nature of electrolyte and the structure of double layer.

A rough estimate of the free energy change of sorption can be found by the application of the suitable isotherms such as those proposed by Bockris and Swinkels[9] and Flory[10]. These were formulated originally for electrodes in contact with solutions containing some additives and were found to explain some experimental observations. It is assumed that for dilute solutions, the sorption takes place as a result of the reaction:

$$nH_2O_{ad} + solute_{soln} = solute_{ad} + nH_2O_{soln}$$

where n represents the number of water molecules sorbed by the surface which has to be removed in order to accommodate one molecule of solute. The sorption isotherms are given by:

$$\frac{\theta}{1-\theta}\,\frac{[\theta + n(1-\theta)]^n}{n^n} = \frac{C_{solute}}{55.5}\exp\left(\frac{-\varDelta G^*_{ad}}{RT}\right) \text{ (B.S. Isotherm)}$$

$$\frac{\theta}{e^{n-1}(1-\theta)^n} = \frac{C_{solute}}{55.5}\exp\left(\frac{-\varDelta G^*}{RT}\right) \text{ (F.H Isotherm)}$$

where θ is the surface coverage by the suitable solute and is defined in our system by:

$$\theta = \frac{(\%\text{ uptake})_{Eq} - (\%\text{ uptake})_c}{(\%\text{ uptake})_{Eq}}$$

Here $(\%\text{ uptake})_{Eq}$ is the maximum uptake which can be obtained from the highest concentration of solute (Cs^+) and $(\%\text{ uptake})_c$ is the uptake of solute at c concentration. The uptake here is expressed as mol l^{-1}.

Since n can take many values (1, 2, ...), then its proper value can be found by plotting the LHS of the above isotherms [$f(\theta)$] against C. The plot in which a straight line passing through the origin would give the proper value of n and hence $\varDelta G^*_{ad}$ can be calculated. Figure 7 shows the plot of the above isotherms for the sorption of Cs^+ by some clays. It is obvious that the free energy change is negative indicating strong interaction between the clays and Cs^+. It is important to point out that the above analysis is very approximate for many reasons, some of which are:

1. The solid surfaces are heterogeneous and the application of the above isotherms will be very limited.
2. The exchangers are complex systems and this will be practically true for natural clays. In addition, the impurities associated with clays will play an important role in influencing the reaction mechanism.
3. It is known that metal oxides in aqueous solutions carry a surface charge which depends on pH of solution. The potential assumed by the surface will thus affect the adsorption process. This potential is unknown for such systems.

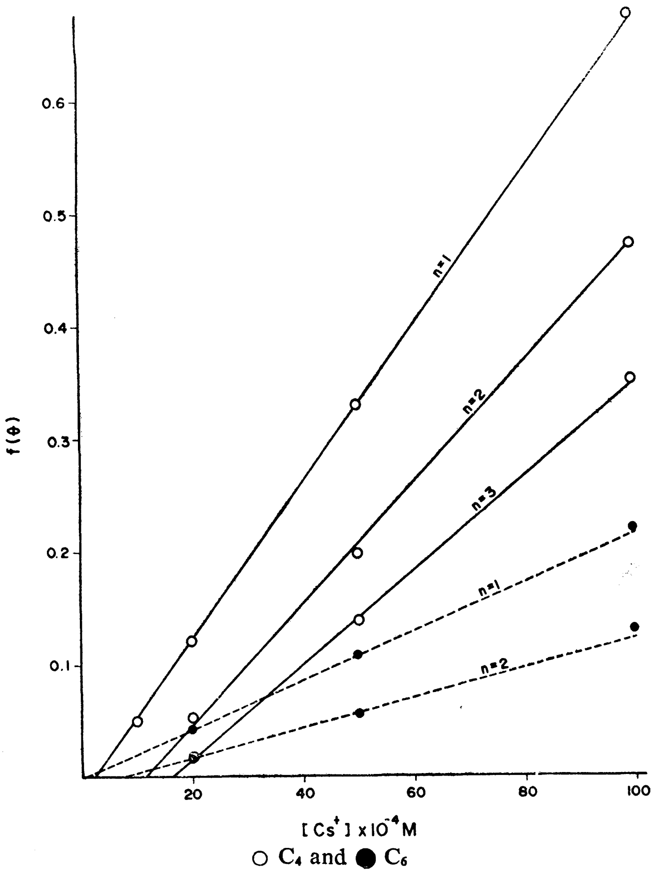


Fig. 7. Plot of F(Θ) versus $[Cs^+]_{soln}$ for Bockris-Swinkel's isotherm for various values of n

No matter what the exact situation may be, the above conclusions may stimulate further work in this field in an attempt to find out the proper kinetic and thermodynamic parameters which control the decontamination processes.

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