

Sorption of Yttrium (III) From Aqueous Solution by Some Natural Clays and Ionic Exchangers

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ABSTRACT. The sorption of yttrium (III) by some natural clays, synthetic exchangers and metal oxides has been investigated. The extent of sorption was found to depend on time of contact, pH, concentration of the sorbed ion, solution temperature, shaking velocity and the solvent. Some cations were added to compete with Y^{3+} but an effect was only observed for trivalent ions at relatively high concentrations. Acid treatment of the clays was found to have no or little effect on sorption, while heat treatment decreases the extent of sorption. The results were found to follow Freundlich isotherm.

Yttrium is one of the fission products and it is estimated that its amount with lanthanum is $\sim 10^4$ g in 5 m³ of high level liquid waste produced from the reprocessing of 1 tonne of LWR fuel (Choppin and Rydberg 1980). Yttrium has many radioactive isotopes and all have short half-lives except ⁸⁸Y with a half life-of 107 days and is a γ -emitter (Friendlander *et al.* 1981).

In spite of the fact that yttrium promises to have an interesting future in the field of nuclear technology due to the low cross-section for neutron capture and high temperature stability (Shivade and Shinde 1987) there seems to be little work concerning its decontamination by natural clays or synthetic exchangers. Most work on yttrium was in the field of solvent extraction (Korkisch 1969, De *et al.* 1970 and Sekine and Hasegawa 1977). The separation of yttrium from strontium was reported by Das (Das 1987) using ascorbic acid and nitrilotriacetic acid as eluents. The adsorption of Y^{3+} on glass was also reported by others (Davydov *et al.* 1983). In

nuclear power plants, Yttrium as well as other radioisotopes were removed from the liquid stream with some ion exchangers (Clinton *et al.* 1984). The aim of this work is to study the possibility of using natural clays as sorbents for yttrium in order to remove its isotopes along with other radioactive isotopes. The clays were chosen because they are cheap and naturally abundant.

Experimental

All chemicals were of analytical grade and water was doubly distilled. The general methodology of preparing solutions, treating the clays and instrumentation has been reported elsewhere (Al-Suhybani 1988). The chemical composition of the clays is shown in Table 1.

The synthetic exchangers used in this work are Amberlite IRA-904(C1)(E₁) Amberlite IRA-400 (E₂), Dowex 50W-X8 (E₃) and Ag-1-X₂ (E₄). The sorption studies were carried out at 25°C, the shaking velocity was 125 vib. min⁻¹ and shaking time is 60 min. unless otherwise stated.

The yttrium isotope ⁸⁸Y was supplied by Amersham International as Y(NO₃)₃ and the carrier was yttrium nitrate. This isotope shows no sign of precipitation up to at least pH 10. The analysis for this isotope was based on γ spectroscopy using a well type 7.5 cm x 7.5 cm NaI(Tl) detector connected to a sample changer Amberg Model 1185 and MCA model 35 plus (Canberra).

Results and Discussion

Effect of Shaking Time

The effect of shaking time on the percent uptake of Y³⁺ by some clays, some metal oxides and some synthetic exchangers is shown in Table 2. For all systems used there is virtually no effect of contact time indicating that, the sorption process is very fast. It may be noticed, however, that % uptake by C₁ shows a slight dependence on time. It is very clear that for two clays the equilibrium uptake is ~95% and for the other two clays the uptakes were ~65 and 84 percent. All are generally better decontaminants than most of metal oxides. For synthetic exchangers only E₃ has an uptake comparable to C₄ and C₅ while the rest of exchangers have rather low uptakes.

Table 1. Chemical Composition of local Clay Samples

Sample	Colour	% Chemical composition										
		SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	TiO ₂	P ₂ O ₅	H ₂ O	L.O.I
C ₁	Light yellow	7.38	3.68	5.66	23.4	16.63	1.21	1.05	–	–	1.05	39.07
C ₂	White	1.6	0.35	0.12	55.48	0.07	0.07	0.07	≤0.05	13.85	0.76	27.08
C ₃	White	58.49	0.15	0.29	22.57	0.09	0.23	≤0.05	≤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	Variegated Shale	54.07	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 ₉	Light brown	21.46	2.12	0.34	27.50	0.91	0.20	0.07	–	17.67	–	4.77
C ₁₀	Gray	38.8	16.7	5.2	4.5	–	12.5	13.0	–	–	1.5	2.0

Table 2. Variation of % uptake of 10^{-3}M Y^{3+} with contact time by natural clays and some synthetic exchangers $\text{pH}_i = 3.25$ $\text{pH}_f = 3.65-6.00$

Sorbent	% Uptake at:				
	0.5 min.	3 min.	15 min.	30 min.	60 min.
C ₁	46.00	47.30	51.50	63.00	65.40
C ₄	89.00	91.40	91.40	92.30	96.60
C ₅	90.00	91.50	91.80	92.00	94.70
C ₉	77.30	80.20	81.00	81.50	84.00
Fe ₂ O ₃	16.00	17.00	17.00	18.00	18.00
MnO ₂	53.00	64.00	67.00	68.00	72.00
TiO ₂	60.50	66.00	67.00	71.20	73.20
Al ₂ O ₃ (N)	27.90	39.85	56.00	70.48	94.21
Al ₂ O ₃ (A)	28.90	37.36	50.10	70.00	91.20
Al ₂ O ₃ (B)	00.00	2.00	3.50	6.00	8.00
SiO ₂	5.50	10.10	10.75	12.95	16.00
E ₁	50.00	51.40	52.40	52.90	54.00
E ₂	51.80	51.80	52.00	52.00	52.30
E ₃	99.00	99.10	99.30	99.30	99.30
E ₄	53.40	54.00	54.30	55.00	55.50

Effect of pH

The sorption behaviour of different materials with initial pH was found to vary considerably as can be seen in Table 3. Thus for all clays the uptake increases with increasing pH till pH ~7 where it becomes maximum. For all metal oxides the general behaviour is similar to that of clays, though the extent of sorption is less. For synthetic exchangers only E₃ has ~100% at all pH values, while for E₁, E₂ and E₄, the uptake increases and becomes maximum at pH ~6.

The behaviour can be partly explained on the basis that these materials have different final pHs, as can be seen in Table 3. Thus for clay C₁, the final pH is very much different from the initial and for this clay the percent uptake is more dependent on pH. For those clays where the final pH is close to the initial value, the uptake is less pH dependent as with the clays C₄, C₅ and C₉. For different oxides the % uptake is what would be expected, if one takes into account the competition by H⁺. At pH > 10, Y³⁺ starts to precipitate and the removal of the ions is by sorption as well as by precipitation.

Table 3. Effect of pH uptake of 10^{-3}M Y^{3+} by some natural clays and other sorbents

Sorbent	% Uptake (different initial pH)											
	0.65*	pH _f	1.70*	pH _f	5.60*	pH _f	7.60*	pH _f	10.1*	pH _f	10.5*	pH _f
C ₁	20.10	4.2	40.20	5.9	66.00	6.7	91.70	7.8	99.60	9.9	99.50	10.6
C ₄	18.40	0.5	68.20	1.2	97.00	6.8	97.60	7.6	99.90	10	99.90	10.1
C ₅	19.30	0.5	86.30	1.6	95.00	6.5	97.30	7.6	98.00	9.9	98.80	10.2
C ₉	65.60	0.75	92.80	2.8	94.00	8	98.90	8.8	99.30	9.9	99.30	10.1
Fe ₂ O ₃	11.50	0.5	15.00	1.8	18.00	3.8	18.50	6.2	20.00	8.8	20.00	9.0
MnO ₂	4.40	0.5	56.60	3.2	72.50	6.6	73.00	7.4	73.00	9.8	75.00	10.2
TiO ₂	10.50	0.5	35.60	2.0	73.00	4.5	75.00	8.8	75.00	10.4	76.00	10.6
SiO ₂	6.00	0.5	7.10	1.9	15.00	5.5	21.00	6.7	21.00	8.8	22.00	9.6
E ₁	1.00	1.6	20.00	2.7	52.00	6.5	52.00	7.6	52.00	9.8	54.00	10.4
E ₂	1.00	0.3	25.00	0.9	35.00	4.5	46.00	6.7	52.00	8.7	52.30	9.5
E ₃	97.40	0.75	98.70	1.7	98.80	4.6	99.20	6.5	99.20	8.5	99.30	9.6
E ₄	1.00	0.45	5.50	1.6	20.00	5.5	35.00	7.8	52.50	8.8	54.80	9.0

* Initial pH.

Effect of Y³⁺ Concentration

The variation of the percent uptake with (Y³⁺) by different clays and other sorbents is shown in Table 4. For all except E₃, the percent uptake decreases with increasing concentration. Thus at 10⁻³M all have uptakes close to 100%, but at 10⁻²M Y³⁺ the uptake varies considerably and for clays it ranges from 2.5% to 70% reflecting the differences in capacities for sorption. For different sorbents the final pH varies between 3.75-6.75 and in this range of pH the uptake becomes maximum. These results were found to follow the Freundlich isotherm which is written in the form:

$$\log C_s = \log A + \frac{1}{n} \log C_b$$

where C_s is the amount of Y³⁺ sorbed, C_b is the concentration of the ion in solution at equilibrium, A and n are constants. The above relationship gave one straight line indicating the applicability of the Freundlich isotherm as can be seen in Fig. 1.

Effect of Competing Ions

Seven competing ions were examined with respect to their abilities to compete with Y³⁺ for sorption by some selected clays. These ions are Al³⁺, Cr³⁺, Fe³⁺, Mg²⁺, Ca²⁺, K⁺ and N_a⁺. The results are shown in Table 5 for two ion concentrations (10⁻³ and 10⁻²M). For the 10⁻³M all ions show almost no competing effect and at 10⁻²M of the competing ion the behaviour is largely dependent on the clay and the competing ion. Thus for most clays the trivalent ions are rather effective, though to different extents, but their effect does not follow any certain rule. A previous work on other ions under similar conditions has shown some regularity in the behaviour of these ions, *i.e.* Al³⁺ is better than Cr³⁺ and the latter is better than Fe³⁺ (Al-Suhybani 1988, 1990 and 1992). This trend was not observed here. Similar behaviour was also observed for di- and monovalent ions. Since the ionic radii of the trivalent ions are close to each other and so are the hydration energies, the behaviour of these ions may be expected due to the similarity in the charge of the displaced ion and those competing with it.

Effect of Acid Treatment of Clays

The sorption of 10⁻³M Y³⁺ by the clays treated with acids was followed at conditions similar to untreated clays and the results are shown in Table 6.

Table 4. Effect of concentration of $[Y(NO_3)_3]$ on the uptake of Y^{3+} by natural clays and other sorbents

Sorbent	% Uptake at $[Y(NO_3)_3]$ M									
	$1 \times 10^{-4}M$ $pH_i = 4.16$	pH_f	$5 \times 10^{-4}M$ $pH_i = 3.70$	pH_f	$1 \times 10^{-3}M$ $pH_i = 3.25$	pH_f	$5 \times 10^{-3}M$ $pH_i = 3.10$	pH_f	$1 \times 10^{-2}M$ $pH_i = 2.7$	pH_f
C ₁	94.70	5.9	76.50	5.5	65.40	4.9	15.10	4.8	12.40	4.5
C ₂	97.20	6.0	96.40	6.5	82.00	5.5	78.00	5.2	70.00	4.4
C ₃	94.50	5.8	80.80	5.2	68.80	4.5	37.20	4.8	18.50	4.2
C ₄	96.60	6.2	96.00	6.0	96.00	5.5	70.50	5.5	18.60	4.8
C ₆	94.90	5.0	85.60	4.9	75.00	4.9	36.10	4.5	20.00	4.0
C ₇	95.00	6.2	70.40	4.8	59.50	4.5	40.00	4.4	15.00	4.7
C ₉	95.60	6.8	86.10	6.5	74.00	5.0	33.80	4.8	20.00	5.0
C ₁₀	97.30	6.2	87.10	5.9	75.00	5.9	03.30	4.8	02.50	4.8
Fe ₂ O ₃	20.00	2.9	20.00	2.7	18.00	2.7	15.00	1.9	05.00	1.5
TiO ₂	83.00	2.5	79.00	2.5	73.20	2.3	60.00	1.5	22.00	1.2
SiO ₂	30.00	4.65	25.00	4.6	16.00	4.5	06.00	4.20	01.00	4.2
E ₁	60.00	5.8	56.00	4.8	54.00	4.8	06.00	4.8	01.00	4.3
E ₂	62.00	2.85	55.00	2.70	52.00	2.75	06.00	2.7	01.00	2.5
E ₃	99.10	5.5	99.00	5.2	99.00	5.2	98.10	5.0	96.60	4.4
E ₄	64.00	3.95	58.00	3.55	55.00	3.55	06.00	3.55	01.00	3.9

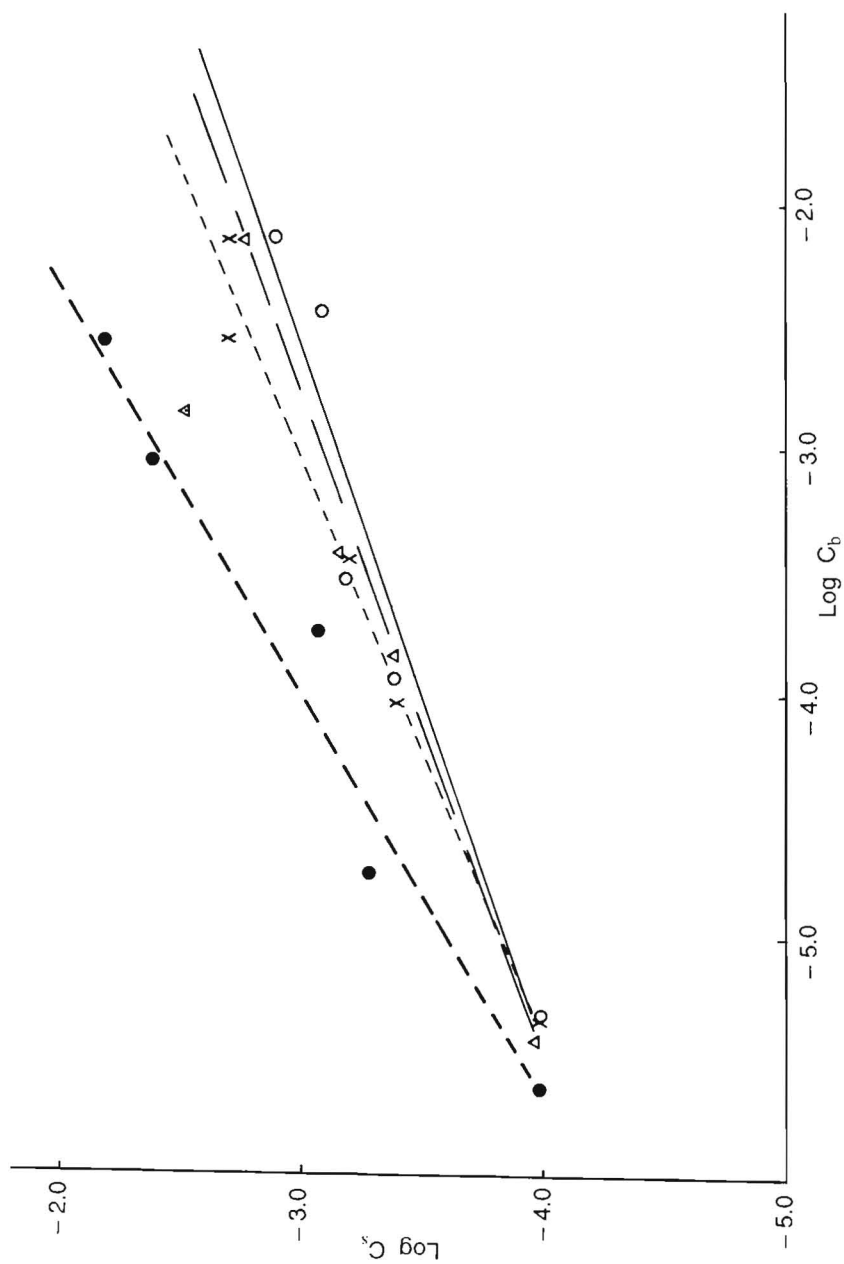


Fig. 1. Application of Freundlich equation for Yttrium Sorption / o C_1 , ● C_2 , x C_3 , Δ C_6 .

Table 5. Effect of competing ions on the sorption of 10^{-3}M Y^{3+} by natural clays from aqueous solutions

Competing ion	% Uptake by								
	[ion]M	C ₁	C ₂	C ₃	C ₄	C ₅	C ₆	C ₇	C ₁₀
Cr ³⁺	10 ⁻³	99.00	98.9	81.6	93.4	95.00	70.00	77.00	59.4
	10 ⁻²	97.30	96.00	80.4	32.00	79.70	22.00	06.00	03.50
Fe ³⁺	10 ⁻³	99.00	98.70	98.10	57.00	98.80	49.00	27.00	08.00
	10 ⁻²	86.90	80.00	72.10	03.80	02.00	07.00	07.00	07.00
Al ³⁺	10 ⁻³	98.80	98.40	63.50	85.50	80.60	80.00	70.40	46.00
	10 ⁻²	86.90	86.50	27.00	19.00	23.40	13.90	02.00	09.00
Mg ²⁺	10 ⁻³	98.10	95.80	81.00	98.30	94.80	79.40	78.50	74.70
	10 ⁻²	65.60	90.00	73.00	90.50	79.10	72.30	65.00	71.30
Ca ²⁺	10 ⁻³	96.50	98.60	84.60	97.80	87.00	91.00	85.90	88.00
	10 ⁻²	70.00	90.00	95.80	90.80	77.00	71.00	65.70	81.70
K ⁺	10 ⁻³	96.00	96.80	74.00	96.40	74.50	86.00	80.60	75.20
	10 ⁻²	65.90	89.00	96.40	95.00	97.00	74.20	80.00	96.70
Na ⁺	10 ⁻³	97.00	97.50	74.20	96.40	74.60	86.30	78.50	75.60
	10 ⁻²	70.90	91.00	97.00	86.40	91.80	84.40	88.20	83.90

Table 6. Effect of acid treatment on the uptake of Y^{3+} by natural clays. $10^{-3}M$; $pH_i = 3.25$, $pH_f = 2.00-2.90$

Clay	% Uptake for clays treated with				
	Untreated	HNO ₃	HCl	H ₂ SO ₄	H ₃ PO ₄
C ₁	65.40	58.90	59.20	99.00	97.90
C ₃	98.80	97.00	97.30	96.00	97.20
C ₄	96.00	89.40	71.30	80.00	86.20
C ₆	75.00	75.40	44.40	93.30	85.55
C ₉	74.00	00.00	00.00	00.00	00.00
C ₁₀	75.00	00.00	00.00	00.00	00.00

It is obvious that the sorption of Y^{3+} by these clays is varying considerably. Thus the uptake decreases a little for C₁, C₄ and C₆ when the clays are treated with HCl but the extent of decrease in the uptake is less if the clays are treated with HNO₃. When the clays are treated with either H₂SO₄ or H₃PO₄ there is a good improvement in the uptake for the clays C₁, C₂ and C₆ but a little decrease in case of C₄. The most striking observation is the zero uptake of C₉ and C₁₀ when they are treated with the four mineral acids. No explanation could be offered at this stage but this could be done when the chemical analysis of the treated clays is completed.

Effect of Thermal Treatment of Clays

The effect of thermal treatment on the uptake of Y^{3+} is shown in Fig. 2. The general behaviour is similar to the behaviour of previously studied ions (Al-Suhybani 1988, 1990 and 1992) *i.e.*, the uptake is almost constant in the temperature range 25-400°C and it then decreases considerably. For example, C₄, C₅ and C₆ have uptakes close to 100% in the temperature range 25-400°C and they decrease to > 45% at 800°C. The mechanism of action of treated clays is expected to be similar to the mechanism encountered with other ions (Th⁴⁺, Co²⁺, Sr²⁺, ... *etc.*) (Al-Suhybani 1988, 1990 and 1992). Grim (Grim 1968) has discussed the factors affecting such a mechanism. These factors include movement of exchangeable

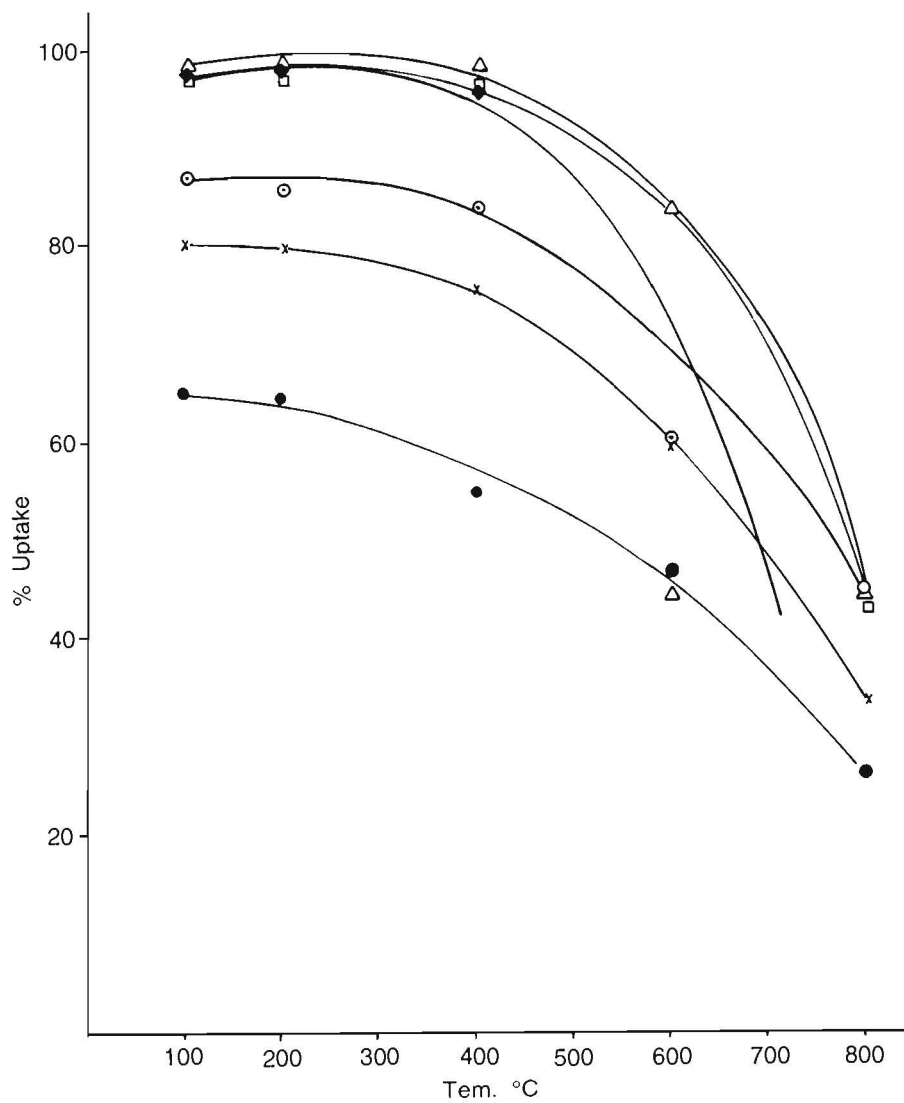


Fig. 2. Effect of Thermal treatment on Y³⁺ Uptake by natural clays. (Y³⁺) = 10⁻³M, shaking time = 60 min., velocity = 125 min.⁻¹ Initial pH = 2.48.

● C₁, x C₂, □ C₄, ■ C₅, △ C₆, ○ C₇,

cations inside the clay lattice, dehydration effects ... *etc.*

Effect of Shaking Velocity

Table 7 shows the results of the effect of shaking velocity on the uptake of Y^{3+} by some selected clays and oxides.

Table 7. Effect of velocity of shaking on the uptake of $10^{-3}M Y^{3+}$ by some natural clays and some oxides. Time of shaking = 3 min. $pH_i = 3.25$, $pH_f = 4.60-6.60$

Sorbent	Velocity, vib. min ⁻¹			
	30	75	125	225
C ₁	41.6	44.50	51.50	78.80
C ₄	88.00	91.40	93.20	97.00
C ₅	86.00	91.00	92.00	98.60
C ₉	68.40	70.70	81.00	86.30
Fe ₂ O ₃	19.00	19.80	20.30	20.30
MnO ₂	44.20	57.70	64.00	77.20
TiO ₂	63.50	65.60	67.00	78.80

It is clear that the uptake of the clays varies, to some extent, with increase of shaking velocity. This variation depends on the clays and it ranges from ~10% to 40% but for Fe₂O₃ the change is in fact within experimental error. For the first case the change is probably due to an increase in the surface area of the clay with shaking rather than a change in sorption mechanism.

Effect of Solution Temperature

Seven clays were selected to find out if there is any change in the uptake with the temperature of solutions. The runs were carried out at 5, 25 and 50°C and the results are shown in Fig. 3. The data indicate that for five clays the sorption increases with temperature and decreases again and the other two behave differently. For the first category the initial increase of sorption with temperature may in part reflect a small activation energy basis for sorption. But at higher temperatures, the decrease in sorption may be a result in change in elemental species.

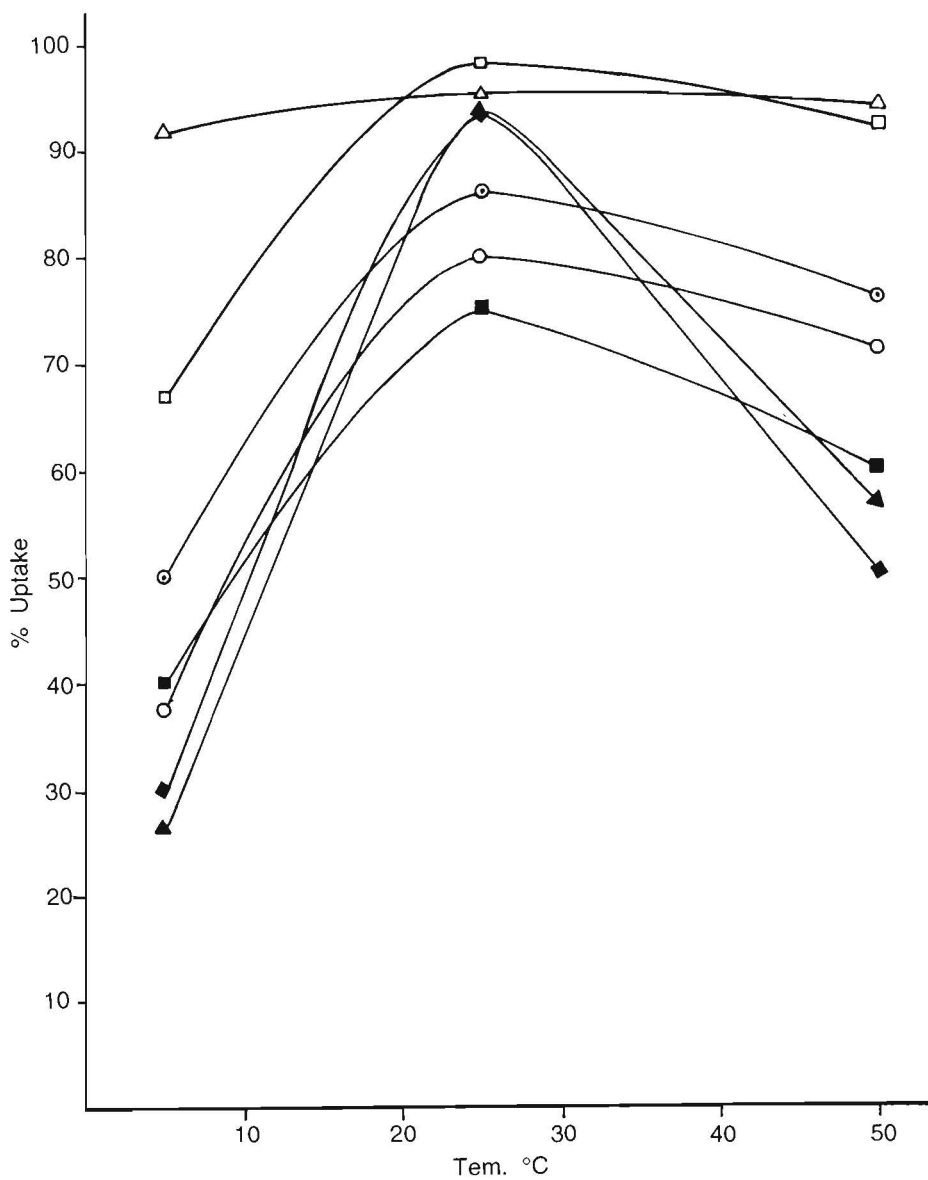


Fig. 3. Effect of solution temperature on the Y^{+3} Uptake by some natural clays. (Y^{+3}) = $10^{-3}M$, shaking time = 3 min., Initial pH = 3.25, final pH ~ (3-6).

○ C₃, □ C₄, ◆ C₅, △ C₆, ⊙ C₇, ▲ C₉, ■ C₁₀

Effect of Solvent Type

In addition to water, pure methanol, mixtures of water and methanol and tributyl phosphate (TBP) were used as solvent for sorption of Y^{3+} using six different clays and the results are shown in Table 8.

Table 8. Solvent effect on the Uptake of $10^{-3}M Y^{3+}$ by some natural clays

Clay	% Uptake for the solvent			
	Water	50% Methanol	100% Methanol	TBP
C ₁	65.40	47.32	37.90	1.30
C ₄	96.60	76.17	64.46	4.80
C ₅	94.70	48.45	44.21	6.70
C ₆	75.00	70.92	83.17	0.00
C ₇	70.00	60.96	41.51	0.00
C ₁₀	75.00	48.80	19.87	3.70

Apart from TBP, the uptake of Y^{3+} by the clays increases with increasing water content and this is probably due to the fact that in aqueous media one is dealing with hydrated ions and the hydration seems to facilitate the sorption as was observed with other cations.

Conclusions

The sorption of Y^{3+} by some sorbents has revealed the following:

The removal of Y^{3+} by clays, some metal oxides and synthetic exchangers is generally very fast. The uptake increases considerably with pH and reaches its maximum value at ~pH 7. The increase in carrier concentration leads to a decrease in the percent uptake due to the saturation of sorption sites and the results was found to obey the Freundlich isotherm. Only trivalent ions (Cr^{3+} , Fe^{3+} , Al^{3+}) were found to be effective as competing ions and this effect is clear at rather high concentrations.

For some clays, the acid treatment led to some improvement in the uptake while the thermal treatment decreases the extent of sorption. For some clays the sorption increases with rise in solution temperature and reaches a maximum and decreases again and it increases with shaking velocity due to an increase in surface area.

In general natural clays are recommended as sorpents for Y^{3+} because they are cheap and naturally abundant.

Acknowledgement

This work is supported by King Abdulaziz City for Science and Technology (KACST), under project No. RT-7-32. The author highly appreciates the technical assistance of Mr. O. Alhems.

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(Received 30/10/1994;
in revised form 11/11/1995)

إمتزاز اليتريم الثلاثي بواسطة بعض المبادلات الأيونية

عبد العزيز بن عبد الله السحبياني

قسم الكيمياء-كلية العلوم-جامعة الملك سعود

ص.ب. (٢٤٥٥)-الرياض ١١٤٥١-المملكة العربية السعودية

لقد تم في هذا البحث دراسة إمتزاز أيونات (Y^{3+}) بواسطة بعض الأطيان الطبيعية وبعض المبادلات المصنعة وأكاسيد المعادن . وقد وجد أن مدى الإمتزاز يعتمد ودرجات متفاوتة على الزمن والأس الهيدروجيني (pH) وتركيز الأيون الممتز ودرجة حرارة المحلول وسرعة الهز ونوع المذيب . وعند إضافة بعض الأيونات المنافسة للأيون الممتز وجد أن هناك تأثير ملحوظ عند التركيزات العالية للأيونات ثلاثية التكافؤ . وينتج عن معالجة الأطيان بالحموض المعدنية عدم تغير الخصائص الإمتزازية للمعادن أو حدوث تغير طفيف ، أما المعالجة الحرارية للأطيان فتؤدي عموماً إلى نقص الكمية الممتزة . ومن ناحية أخرى دلت النتائج على إنطباق أيزوثيرم لانغميور .