

**LABORATORY STUDIES INTO THE USE OF THE SCANDIUM-
46-EDTA COMPLEX AS A TRACER FOR GROUNDWATER FLOW**

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Summary

Gamma ray emitting metal radionuclides, when complexed with an appropriate complexing agent, provide a wide choice of water tracers particularly for groundwater studies where a radionuclide of appropriate half-life suited to the particular study can be selected.

Scandium-46 has easily detectable gamma emission and a suitable half-life (84 days) for medium term studies. It has been widely and successfully used as a tracer in studies of sediment movement but has not yet introduced as a groundwater tracer.

In our experiments the chemical aspects of the preparation of Sc-46-EDTA were studied in some detail and its behaviour in various mineralogical environments was evaluated with reference to the standard tracer, tritiated water. The experimental results have shown that the scandium cation can be easily complexed with EDTA to form soluble Sc-EDTA. The complex is very stable in a wide range of pH; the adsorptive properties of Sc-EDTA in the batch studies and the retardation and recovery in the column tests in comparison with tritiated water are quite satisfactory. In general Sc-46-EDTA is a promising tracer for groundwater studies.

In this report the appropriate conditions, procedures and some rational and efficient methods for testing the purity of Sc-46-EDTA in the preparation of the tracer solution of Sc-46-EDTA are described. In addition, it has been found that the formation of the metal hydroxide colloids is the major reason for the great loss in groundwater aquifers of most trivalent metal nuclide tracers in the cationic form including scandium-46.

1. Introduction

Gamma emitting radionuclides are easily detectable in-situ and have been widely used as groundwater tracers both in hydrology and environmental protection studies. Although an element may be made radioactive to provide a tracer with a suitable half-life and radiation properties that give easy detection for the duration of the test, its behaviour in the flow system depends on its chemical form. Because tracers in the cationic form are largely lost in aquifers by adsorption and ion exchange, only tracers in the anionic form or negatively charged or neutral complexes can be considered for use in groundwater investigations. Since the first application of radioactive tracers in the beginning of the 1950's, several anions and a great number of metal complexes have been tested in laboratory and in the field to seek ideal tracers. But there seems to be a shortage of satisfactory γ -emitting tracers for medium term studies.

It is known that bromine-82 in the form of KBr (or NH_4Br) and iodine-131 in the form of NaI (or KI) are reliable tracers but only for short term investigations because of their short half-lives (bromine-82, 35h, iodine-131, 8.05 days). Among the metal complexes, two, chromium-51 in the form of Cr-EDTA and cobalt-58 or cobalt-60 in the form of $\text{KCo}(\text{CN})_6$ are relatively suitable for this purpose (half-life, chromium-51, 27 days; cobalt-58, 71 days; cobalt-60, 5.3 years) and have been quite successfully applied in many field experiments. But there are

some disadvantages in using chromium-51 as a tracer. Only about 8% of the disintegrations of chromium-51 give rise to gamma ray emissions which are of relatively low energy. Also due to the irradiation properties of chromium only low specific activities of chromium-51 are possible.

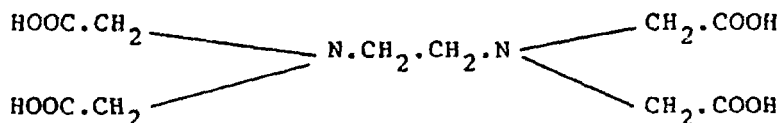
Scandium-46 is a high-energy gamma emitter of a suitable half-life (84 days) and has been the most widely employed tracer for the investigation of sediment transportation for more than thirty years. In contrast, it has not been used in ground-water studies and very little literature about it is available. The purpose of this laboratory study was to carry out a detailed investigation into the chemistry of Sc-46-EDTA and find out the appropriate conditions and procedure for the preparation of Sc-46-EDTA and evaluate its behaviour in various mineralogical environments with reference to tritiated water and to estimate its applicability.

2. Chemical basis of the preparation of scandium-EDTA complex

2.1 Formation and stability of the Sc-EDTA complex

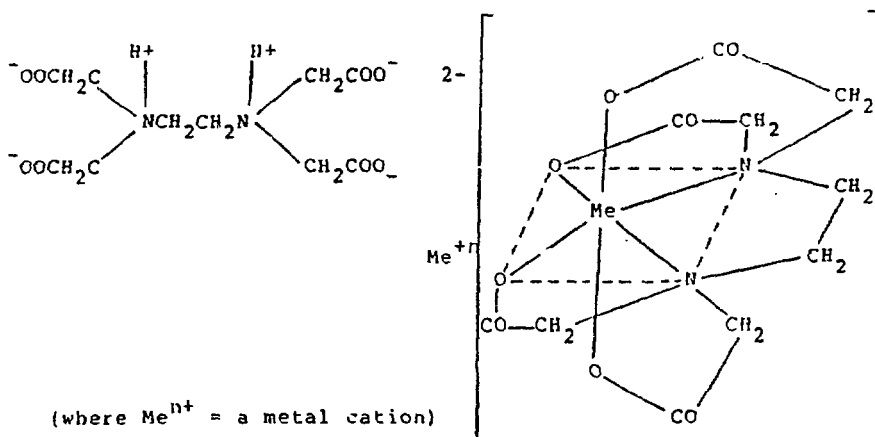
2.1.1 Complexing agent, EDTA and its metal complexes

Ethylenediaminetetra-acetic acid has the following formula:



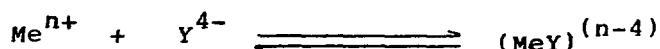
and is an outstanding metal complexing agent. Its complexing power lies in the ligand forming nitrogen atoms and the carbonyl oxygens of the acetic group. Both the acid and the salt are commonly abbreviated "EDTA".

EDTA reacts instantaneously, in nearly every case, with many polyvalent metal cations in a 1:1 ratio to form the soluble metal complexes.



This reaction results in the metal cation replacing a hydrogen atom from an acetic acid group on each of the two nitrogen atoms in the molecule. In addition co-ordinate bonds are developed with the two nitrogen atoms and carbonyl oxygens of the remaining two acetic acid groups. In this way the metal ion becomes part of the chelate rings. During past years quantitative data have been obtained to prove that the greater number of chelate rings formed, the greater the stability of complex formation is. Usually EDTA is capable of forming a co-ordination saturated complex with most metal ions.

The stability constant of the normal metal-EDTA complex formed by the general equation:



is defined by the formula

$$K = \frac{[(\text{MeY})^{(n-4)}]}{[\text{Me}^{n+}][\text{Y}^{4-}]}$$

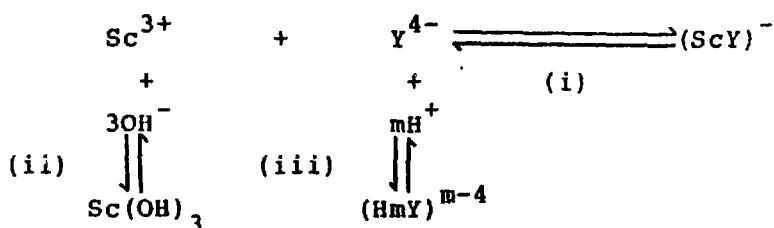
(where Y is the anion of EDTA and M is the metallic element.)

The stability contents of Sc-EDTA and some other metal-EDTA complexes are listed in Table 1.

2.1.2 Parameters affecting the formation and stability of scandium-EDTA

(1) Effect of pH

In normal experimental conditions there are three competing reactions in the formation of Sc-EDTA:



These three equilibria exist in the reaction mixture solution:

- (i) - Equilibrium of the formation of Sc-EDTA
- (ii) - Equilibrium of the precipitation (or dissociation) of Sc(OH)_3
- (iii) - Equilibrium of the formation (or dissociation) of ethylene diamino tetra-acetic acid, i.e. the EDTA anion may be regarded as a complexing agent for hydrogen ions.

It can be seen that there are two factors which will shift the equilibrium of the formation of Sc-EDTA towards the left. First, when increasing the concentration of OH^{-} (i.e. alkalinizing the solution) to such an extent that the solubility product

of $\text{Sc}(\text{OH})_3$ is reached and precipitation occurs lowering the concentration of Sc^{3+} in the solution. Secondly, there may be such an increase in the concentration of H^+ (i.e. acidifying the solution) that it complexes with Y_4^- reducing the amount of Y_4^- present in the solution. From the analysis above there must be an appropriate range of pH for the formation of Sc-46-EDTA.

In our experiment solutions of ScCl_3 at a scandium concentration of 12 ppm were complexed with EDTA at different pH values and the degree of the complex formation was determined. The results are shown in Table II.

The stability of Sc-EDTA at different pH values was also tested. The degree of the decomposition of scandium-EDTA at different pH is given in Table III.

As analysed in the schemes of the equilibria above in very strong acidic or alkaline media the equilibrium of the formation of Sc-EDTA will be considerably shifted toward the left. The data in Table III have proved the analysis. The results still show that Sc-EDTA is very stable over a quite wide range of pH.

(2) Effect of the ratio of EDTA to scandium

In the view of the chemical equilibrium and reaction dynamics, although Scandium is complexed with EDTA in the ratio of 1:1, an excess of EDTA is beneficial in the formation and stability of Sc-EDTA. The experimental results indicate that the Sc-EDTA complex formed in the ratio 2:1 is more stable over a wider range of pH than that formed in the ratio of 1:1. The comparison is made in Table IV.

(3) Effect of temperature

In general, temperature also has an effect on the formation and stability of the metal-EDTA complexes. Raising the solution temperature from 20°C to 60°C decreases the value of the stability constant of the complex by factor of 10. However, as the value of the stability constant for scandium-EDTA is very high, this effect can be neglected. On the other hand increasing the temperature is useful in increasing the speed of the complex formation. In practice at room temperature the complexing reaction occurs instantaneously and completely and the temperature has no marked effect on the formation and stability of scandium-EDTA complex.

2.2 A test to ensure the complete formation of scandium-46-EDTA in the complexing reaction

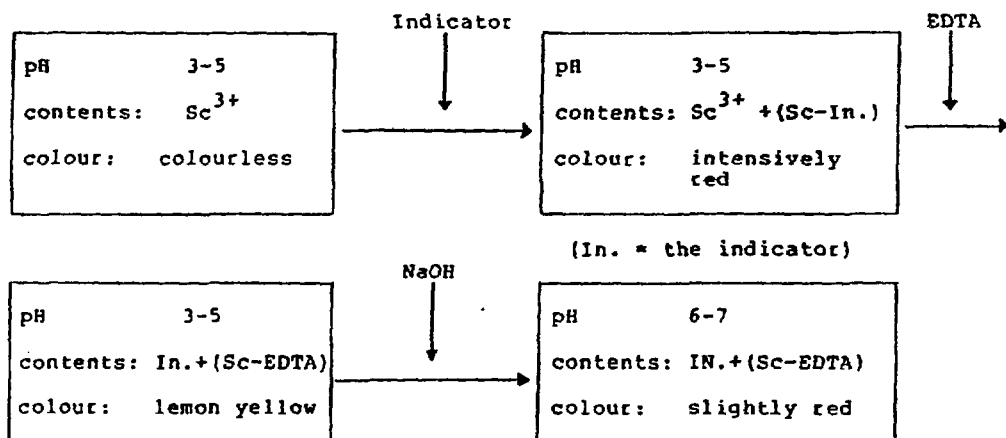
A metallochromic indicator, xylenol orange, was adopted as an indicator for measuring the completion of the complexing reaction in the preparation of scandium-46-EDTA. The metal indicator has the double function of showing the end point of the complexing reaction and ensuring the correct pH for use as a groundwater tracer.

2.2.1 Characteristics and the functions of xylenol orange in the preparation of the tracer solution of Sc-46-EDTA

Xylenol orange (3,3-bis-N), N-di (carboxymethyl-aminomethyl-o-cresolsulphonphthalein) is a conventional acid-base indicator. Xylenol orange is lemon yellow in acid media and undergoes a colour transition in the range of pH 5.4 to 7.4 to red. The indicator can also form complexes with many metals including scandium. These are intensively red in colour. During the complexing reaction with EDTA the metal ion is drawn from the metal-indicator complex and transferred to the metal-EDTA complex. Eventually the indicator changes its colour when it is released from the form of the metal-indicator complex to the "free" indicator.

Our experiment shows that the appropriate range of pH for xylenol orange as an indicator for the preparation of Sc-46-EDTA is 3 to 5. Below pH 2 the colour of the metal-indicator complex is very vague (It should be intensive red colour,) and at the end of the formation of Sc-46-EDTA the colour change is not sharp. Above pH 5.4 it has no function at all.

After completing the formation of Sc-46-EDTA, the solution has to be adjusted to neutral so that it can be used as a groundwater tracer. While the diluted NaOH solution is being added drop by drop to the solution the "free" indicator will change its bright lemon yellow to slightly red when the pH of 6 to 7 is reached. The behaviour and the functions of xylenol orange in the preparation Sc-46-EDTA is illustrated below:



The use of the indicator in the preparation of $^{46}\text{Sc-EDTA}$ is very important. Especially it is convenient for the preparation under field conditions.

2.3 Methods for testing the degree of the formation or decomposition of scandium-46-EDTA complex

In order to find out the optimum conditions for preparing scandium-46-EDTA as a groundwater tracer some rational and efficient methods to test the degree of the formation or decomposition of scandium-46-EDTA in the reaction mixture solution should be worked out. In our experiments two methods were employed, uptake by kaolinite or the filtration through glass microfibre filters under neutral conditions.

2.3.1 Chemical basis of the methods

It has been reported that upon addition of sodium hydroxide to scandium chloride solutions, precipitation of scandium hydro-

hide begins at a pH of 4-5 and is nearly complete at a pH 5.45^[25]. In our studies several verifying tests were made. One was to determine the degree of the precipitation of scandium hydroxide at different pH by adding 0.5M NaOH solution to the Sc-Cl₃ solution at a scandium concentration of 12 ppm. The other was to measure the degree of the precipitation of scandium hydroxide from scandium chloride solutions at different concentrations at a pH of about 7. The results are shown in Fig. 1 and Table V. From the data above it can be seen that in neutral media the scandium cation in solutions of scandium chloride at concentrations down to 0.1 ppm was fully precipitated from the solution to form colloidal scandium hydroxide.

The precipitation of scandium hydroxide from scandium salts solutions has been utilized as a method of separation of scandium from solution in analytical chemistry and can be adopted as a testing method to measure the degree of the formation or decomposition of scandium-46-EDTA. In the complexing reaction of scandium with EDTA if the formation is not complete or the formed complex is not stable and decomposes to some extent, when the reaction mixture is adjusted to neutral, the portion of the scandium in the solution which is not complexed with EDTA will be precipitated. By means of separation of the hydroxide the degree to which the complex is formed or decomposes can be determined. The two methods of separation of the colloidal hydroxide from the solution as mentioned above are described in the following section.

2.3.2 Uptake by kaolinite powder

Kaolinite ($Al_2O_3 \cdot 2SiO_2 \cdot 4H_2O$) is a clay mineral. The powder is a very fine clayey silt. The experimental results have shown that due to its extremely large specific surface area and cation exchange capacity (about 10 meq/100 gm) the powder can take up around 40% of the scandium in acidic medium (in the cationic form) and about 99% of the scandium in neutral medium (in the form of the colloidal hydroxide) but no scandium complex over a quite wide range of pH. The method can be adopted, therefore, as a testing method to determine the amount of scandium in the neutralised reaction mixture solution which is not complexed with EDTA. The method described below was employed in our studies as a major testing method.

2 to 5 g of air-dried kaolinite powder was added to a glass stoppered flask containing 100 ml of the reaction mixture solution to be tested. The flask and the contents were shaken for 1 hour. After settling, 5 ml of sample was taken from the supernatant and measured for its radioactivity. The results are given in Table VI.

2.3.3 Filtration through glass microfibre filter

The results have shown that by using this method the efficiency of separation of the colloidal scandium hydroxide from the reaction mixture could reach about 98-99% and no scandium-46-EDTA was removed from the solution. The data are shown in Table VII.

This method is so simple that after filtering the neutralized reaction mixture the proportion of scandium which is not combined with EDTA can be determined.

3. Preparation of the tracer solution of scandium-46-EDTA

3.1 Reagents and their preparation

(1) Analytical-grade scandium chloride hexahydrate ($\text{ScCl}_3 \cdot 6\text{H}_2\text{O}$)

Because the chloride is a more stable form than the metal and readily soluble in cold dilute acid it was chosen as the target material in the irradiation to produce scandium-46. Before irradiation the chloride hydrate was dehydrated by heating at 190°C for 24 hours.

(2) Analytical-grade disodium salt of EDTA ($\text{Na}_2 \text{H}_2\text{Y} \cdot 2\text{H}_2\text{O}$)

The use of the disodium salt dihydrate is more convenient than the acid because of its water solubility (for the salt, 11.1g per 100g of solution and for the acid, 0.02g in 100 ml of water). The prepared EDTA solution should be stored in a polyethylene bottle because a soft glass container in contact with EDTA will yield cations (especially calcium and magnesium) which may affect the function of the indicator.

(3) 0.5% Xylenol orange solution

The indicator is commercially available as the sodium salt. A 0.5% water solution of the salt should be prepared and is stable for some weeks.

(4) Analytical-grade HCl and NaOH solution

0.1 M HCl is needed for dissolving the irradiated tablet and 0.5 M NaOH is used to adjust the pH of the relevant solutions in the preparation of the tracer solution of scandium-46-EDTA.

(5) Double distilled water

In the preparation of scandium-46-EDTA it is suggested that only purest double distilled water or de-ionized water can be used because in "ordinary" distilled water sometimes more than trace amounts of calcium or magnesium are occasionally found which may influence the colour change of the indicator.

3.2 Appropriate procedures and conditions

(1) Irradiation

A tablet was made from the dehydrated scandium chloride powder by using a disc press (in our case, 13 mm die, 10 tons pressure). The tablet was put in an aluminium can filled with silicon cotten and the contents irradiated to obtain the specific and total activity needed for the investigation. The amount of scandium required and the total activity achieved can be estimated by using the following formula [13] with an error of less than 4% if the irradiation time is less than 10 days.

$$A = 6.55 \text{ Wt} \times 1^{-14}$$

where A - Total activity achieved in curies
 W - Weight of scandium in the compound irradiated in grams
 ϕ - effective neutron flux in sample in neutrons per cm² per second
 t - irradiation time in days

- (2) Dissolve the irradiated tablet in 0.1 M HCl to provide the required volume.
- (3) Adjust the pH by adding 0.5 M NaOH solution drop by drop to the solution until the pH is in the range of 3 to 5.
- (4) Add 3 to 5 drops of 0.5% xylenol orange solution. Then the solution will turn to intensively red colour.
- (5) Add EDTA solution drop by drop into the solution while it is being stirred. If Na₂H₂Y.2H₂O is used the appropriate ratio of EDTA:Sc should be about 10:1 by weight. After the colour of the solution has changed to bright lemon yellow continue to add one quarter more of the EDTA solution which has already been added (This means the ratio is about 10:1.)
- (6) Neutralize the reaction mixture solution by adding 0.5 M NaOH solution gently with stirring until the colour changes to a slightly red, i.e. the pH is about 6-7.

The prepared solution of scandium-46-EDTA is ready to use as a groundwater tracer.

4. Equilibrium studies

4.1 Experimental method and the minerals used

5g of the air-dried and sieved mineral were added to a glass stoppered flask with 100 ml of the solution of scandium-46-EDTA or of the tritiated water. The flask and contents were shaken overnight. After filtration 5 ml of sample was taken and measured for radioactivity. The concentration of the total scandium in the scandium-46-EDTA solution was 1 ppm and the specific radioactivity in the complex solution and the tritiated water were 3.5×10^{-3} uCi/ml and 3.5 uCi/ml, respectively. In these batch tests three clay minerals and nine rock minerals were used. The minerals used are briefly characterised in Table VIII

4.2 Expression of the uptake of the tracers and experimental results

In the equilibrium studies the uptake of the two tracers was expressed by a distribution coefficient, K_d, which is defined as the ratio of the adsorbed tracer per unit of dry mineral weight to the tracer per unit volume of equilibrated solution. In the case of the radioactive species the radio was expressed in terms of activity as:

$$K_d = \frac{\text{activity per gram of mineral}}{\text{activity per millilitre of solution}}$$

5. Column tests

The purpose of the column tests was to evaluate the losses and retardation of the tracer of scandium-46-EDTA in different mineral columns with reference to the tritiated water and the effect of the amount of the stable carrier.

5.1 Characteristics of the columns and the experimental method

In the column test three minerals, limestone, quartz sand and chalk were used. The minerals were ground to a certain range of particle size and packed in perspex cylinders of 8.0x85cm. Glass wool was used at the top and bottom of the column to prevent surface disturbance of the minerals or the loss of them. "Air-free" tap water, which was produced by filtration through a column packed with fine sand was used to prevent porosity changes during the test period. A constant hydraulic head was maintained so that a relatively stable filtration velocity in the columns was obtained in the range of 1-3 m/d. The characteristics of the columns in the tests are illustrated in Table XI. The operation of the column tests was as follows:

- (1) The "air-free" tap water was pumped through the column for several hours before the experiment started.
- (2) The water level in the column was adjusted to be just above the surface of the mineral.
- (3) 100 ml of the tracer solution was added gently on to the top of the column (The same volume for all tests).
- (4) The tap at the bottom of the column was turned on and the tracer solution was allowed to move down into the medium. Immediately after the first exposure of the surface the micropump was turned on at the designed rate and the water level was kept over the surface during the experimental period.
- (5) Samples were taken periodically and measured for radioactivity. The eluate samples of the tracer of scandium-46-EDTA were counted in a well-type NaI crystal detector and for the double tracer of scandium-46-EDTA and tritiated water in a liquid scintillation counter.

5.2 Simultaneous determination of scandium-46 and tritium in the double tracer solution by beta spectrometry

The tritiated water was added to the tracer solution of scandium-46-EDTA as a reference tracer. The simultaneous determination of the two nuclides in the eluate samples by assay in the liquid scintillation counter (Beckman LS9800) was carried out by the following methods:

- (1) Scandium-46 can be detected by liquid scintillation counting due to its radiation (0.357 Mev. ~100%) and the Compton absorption of part of its gamma emissions (0.889 Mev, 100%; 1.12Mev 100%). The spectra of the two nuclides in the counter are illustrated in Fig 2. As shown in Fig 2, no tritium will be detected above channel 400. With the discriminator setting of the channel from 450 to 850, scandium-46 could be measured separately at a high counting efficiency of 62-63%.
- (2) Tritium was analysed without interference by using such a high concentration of tritium with a relatively low discriminator setting that the contribution of scandium-46 to the total counts

in the range of the channels could be neglected. The relevant parameters are given in Table X.

5.3 Results

The column tests were carried out in two groups. One was running the double tracer through different mineral columns and the other was percolating the tracer of scandium-46-EDTA at different concentrations through quartz sand columns.

The results are expressed in both tabulated and schematic forms. In the tabulated form U_c is the average filtration velocity in the column obtained by the pumping rate and U_t is the filtration velocity calculated from the peak time in the elutriation curves of the tracers. V is the eluate volume and V_p is the pore volume of the column. In the schematic form the ρ data are plotted by using the dimensionless ratio of C/Co i.e. the concentration in the eluate relative to injected solution versus the volume throughput. The results of the column tests of the two groups are given in Table XI and Table XII, and the relevant curves are illustrated in Figs 3-11.

Note that experiment 7 tested the recovery of the double tracer over a long period; its experimental conditions were similar to those of experiment 3 (also using a chalk column), so only the recovery curves are presented for experiment 7.

6. Conclusions

1. From the precipitation tests it was found that in the range pH 6.9 to 7.8 (close to that of groundwater 6.5-7.5) the scandium cation in solutions at concentration down to 0.03 ppm was nearly fully precipitated. If the scandium cation was used as a groundwater tracer it would be largely absorbed in the aquifer as most trivalent metals such as Cr^{3+} , Co^{3+} , Fe^{3+} , In^{3+} , etc due to the extremely low solubility products of their hydroxides.
2. The appropriate conditions for the preparation of the Scandium-46-EDTA complex are: -
Range of pH: 3-5
Ratio of EDTA: Sc by weight: 10:1 (if $Na_2H_2Y \cdot 2H_2O$ is used)
Temperature: Room temperature
Indicator: Xylenol orange
3. It has been reported in some references that an ion-exchanger, for example, DOWEX X50WX8 was used for testing the degree of the formation of scandium-EDTA complex. But it should be pointed out that at pH above 6 if there is some unchelated scandium left in the solution it must be in the form of the hydroxide not the cation. In this case the metal in the colloidal state is only slightly adsorbed by the cation-exchanger, the uptake being about 5% of that of the normal cation [23] and this fact has been proved in our experiments. In comparison the uptakes by kaolinite powder or filtration through glass microfibre filter are more rational and the two methods are efficient to measure the degree of the formation of scandium-46-EDTA in the neutralized reaction solution.
4. Scandium-EDTA was not taken up by either kaolinite or montmorillonite, both clay minerals with high ion exchange capacities and extremely large specific surface areas, and the K_d value were less than those for tritiated water.

5. The elutriation of scandium-EDTA from the test columns was quite similar to that of tritiated water, with tracer recoveries of 98% and 100% for limestone and quartz sand, respectively. In the chalk column 90% could be eventually recovered after a relatively long period (8 days compared to 1 day for quartz sand).

6. The results obtained from the column tests with scandium-46-EDTA at different concentrations have shown that increasing 100 times the concentration of the stable carrier only resulted in doubling the radioactive concentration at the peak point. In theory and in practice it can be said that in a saturated zone of the groundwater aquifer for a basically unadsorbed tracer the effect of the carrier is not remarkable and the radioactive concentration and the volume of the injected tracer solution should be the decisive factors because in this case the hydraulic dispersion is always the most dominating parameter which affects the tracer concentration.

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Table I. Stability constants of some metal-EDTA complexes [Value at 20°C and ionic strength = 0.1 (KCl or KNO₃)] selected from references [23, 30]

Trivalent		Divalent		Monovalent	
Complex	LogK	Complex	LogK	Complex	LogK
(CrY) ⁻	23.0	(MgY) ²⁻	8.7	(KY) ³⁻	0.8
(ScY) ⁻	23.1	(CaY) ²⁻	10.7	(NaY) ³⁻	1.7 (c)
(InY) ⁻	25.0	(ZnY) ²⁻	16.5	(LiY) ³⁻	2.8
(FeY) ⁻	25.1	(PbY) ²⁻	18.5	(AgY) ³⁻	7.3
(CoY) ⁻	40.7 (a)	(CuY) ²⁻	18.8 (b)		

Notes: (a) u = indefinite
 (b) u = 0.03
 (c) u = 0.01

Table II. Effect of pH on the formation of scandium-46-EDTA

pH	1.8	2.5	3.5	5.0	7.4	9.4
Degree of complex formation (%)	99.0	99.8	100	99.7	100	99.6

Table III. Effect of pH on the stability of Sc-EDTA
 (concentration of Sc³⁺ = 12 ppm at 20°C)

pH	1.8	3.2	5.0	7.1	9.1	11
Degree of complex decomposition (%)	12	0	0	0	0	39

Table IV. Effect of the ratio of EDTA:Sc on the formation and stability of Sc-46-EDTA

(concentration of ScCl_3 : 12 ppm at 20°C)

		pH							
		1.8	3.3	5.2	7.1	9.0	11	12	13
EDTA:Sc									
Degree of decomposition (%)	1:1	4	0	0	0	0	39		
	2:1	0	0	0	0	0	0	0	1.4

Table V. Degree of precipitation at different concentrations in neutral media

Concentration of scandium in ScCl_3 solution (ppm)	pH	Degree of precipitation (%)
10	6.9	~ 100
1	7.3	~ 100
0.1	7.4	~ 100

Table VI. Uptake of scandium by kaolinite powder

Solution	Concentration (ppm)	pH	Uptake (%)
ScCl ₃	0.03-10	1.8	35-42
		6.9 - 7.8	99
Sc-EDTA	1.2	2 - 10	0
	1 to 20	7.0 - 7.4	0

Table VII. Remaining scandium in the solution after filtration through glass microfibre filter

Solution	Concentration (ppm)	pH	Remaining Scandium (%)
ScCl ₃	10	7-8	1
	1	7-8	2
Sc-EDTA	20	7-8	~ 100

Table VIII. Distribution coefficient, K_d, for various minerals in equilibrium with scandium-46-EDTA solution and tritiated water pH = 7 at 20°C

Type of Minerals	General formula	Particle size (mm)	K _d	
			⁴⁶ Sc-EDTA	HTO
Kaolinite	Al ₂ O ₃ · 2SiO ₂ · 4H ₂ O	Silty clay	0.16	0.28
Montmorillonite	(OH) ₄ Si ₈ Al ₄ O ₁₂ · nH ₂ O	Silty clay	0	0.99
Chalk	CaCO ₃	Silty clay	2.0	0.53
Limestone	CaCO ₃	0.50-0.85	2.2	0.14
Calcite	CaCO ₃	0.50-0.85	1.1	0.37
Augite	(CaNa)(Mg, Fe ²⁺ , Fe ³⁺ , Al) - (Si, Al) ₂ O ₆	0.50-0.85	0.12	0.60
Albite	Na(AlSi ₃ O ₈)	0.50-0.85	0	0.64
Gypsum	CaSO ₄	0.50-0.85	0.26	0
Microcline	K(AlSi ₃ O ₈)	0.50-0.85	0.12	0.39
Limonite	FeO(OH) · nH ₂ O	0.50-0.85	0.86	0.16
Quartz sand	SiO ₂	0.50-0.85	0.30	0.04

Table IX. Characteristics of the columns in the column tests

Type of Column		Limestone	Chalk	Quartz Sand			
Height (cm)		65.0	65.0	65.0	66.0		
Diameter (cm)		8.0	8.0	8.0	8.0		
Particle Size Distribution (%)	2.00-1.00 mm	57.3	20.8	7.2			
	1.00-0.50 mm	40.5	71.0	37.4			
	0.50-0.25 mm	1.2	5.1	30.6			
	0.25-0.063 mm	1.0	3.1	24.8			
Porosity (%)		43.2	62.5	35.5		37.0	
Exp. No		1	2	3	4	5	6
Pumping rate (ml/min)		7.9	2.3	10.6	6.2	7.6	7.4

Table X. Radioactive concentration, detection efficiency and contribution to the total counts

Nuclide	Specific activity (Bq/ml)	Channel	Efficiency (%)	Contribution to the total counts (%)
Tritium	1.23×10^5	0-100	3.3	99.8
Scandium-46	9.23×10^2		0.8	0.2

Table XI. Experimental results for the double tracer in the different columns
(concentration of Scandium was 10 ppm)

Column	Exp. No.	Filtration Velocity U_f (cm/s)	Type of Tracer	Mean Filtration Velocity U_t (cm/s)	Retardation (U_t/U_f)	Ratio 'Peak to Pulse' (C_{max}/C_0)	Position of Peak (V/Vp)	Relative Delay factor (^{46}Sc to ^{3}H)	Recovery mass %
Limestone	1	2.69×10^{-3}	H ₂ O	2.89×10^{-3}	0.93	0.33	0.93	1	100
			Sc-46-EDTA	2.43×10^{-3}	1.11	0.13	1.11	1.19	97.5
Chalk	2	7.69×10^{-3}	H ₂ O			0.10	0.28	1	87.6
			Sc-46-EDTA			0.06	0.30	1.07	70.4
Quartz Sand	3	3.60×10^{-3}	H ₂ O	3.57×10^{-3}	1.01	0.17	1.01	1	99.2
			Sc-46-EDTA	3.09×10^{-3}	1.17	0.08	1.17	1.16	94.1

Table XII. Experimental results for the tracer solutions of Sc-46-EDTA at different concentrations in quartz sand columns

Concentration of the tracer (µg)	Exp. No.	Filtration velocity U_f (cm/s)	Mean filtration velocity U_t (cm/s)	Retardation (U_t/U_f)	Ratio peak to pulse (C_{max}/C_0)	Position of peak (V/Vp)	Recovery mass %
1	4	2.10×10^{-3}	1.63×10^{-3}	1.29	0.07	1.29	89.3
10	5	2.58×10^{-3}	2.19×10^{-3}	1.18	0.10	1.18	99.6
100	6	2.52×10^{-3}	2.47×10^{-3}	1.02	0.12	1.02	94.4

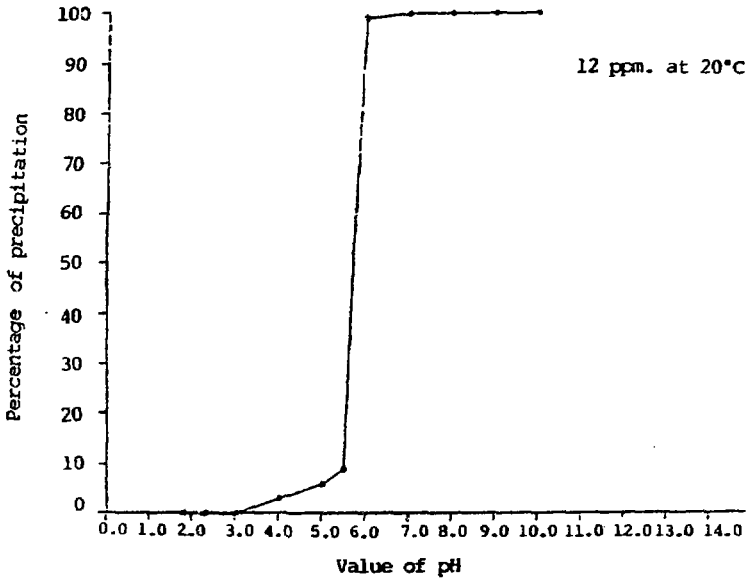


Fig. 1 Relationship between the degree of precipitation of scandium hydroxide and the value of pH

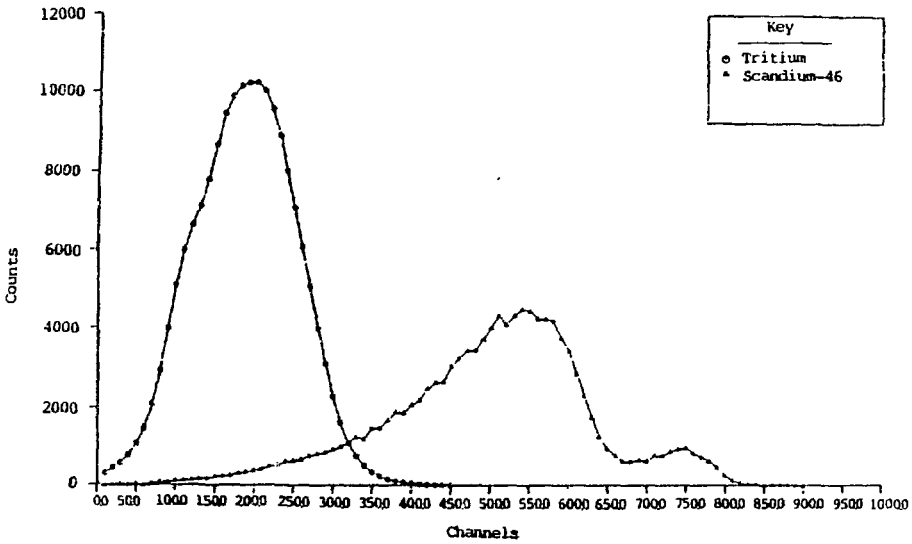


Fig. 2 Spectrum analysis of tritium and scandium-46 on beckman LS9800

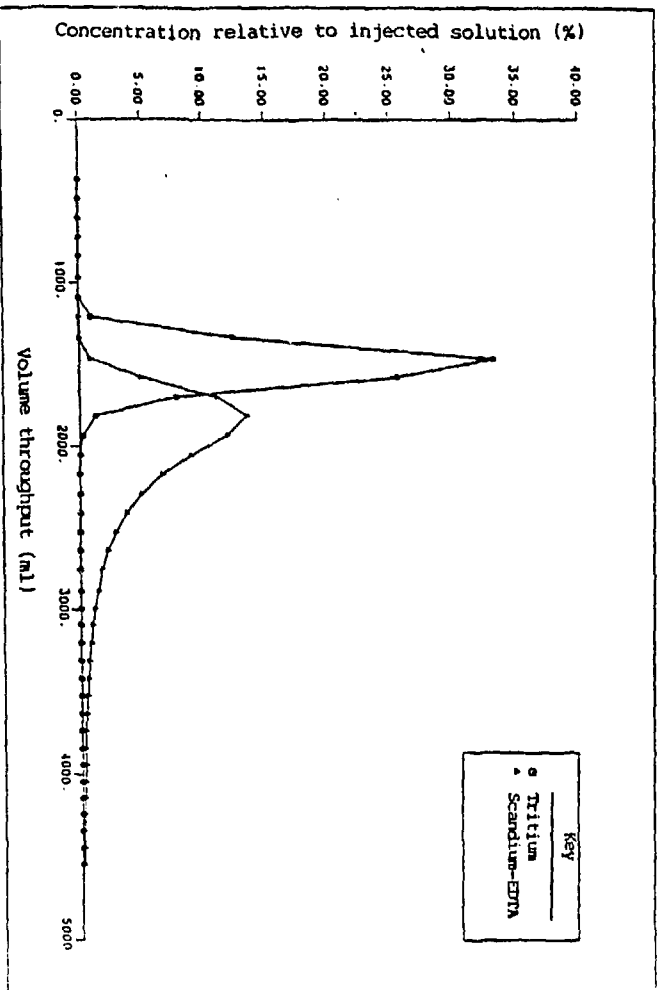


Fig. 3 Limestone column test with double tracer elutriation of tracers(Exp. No. 1)

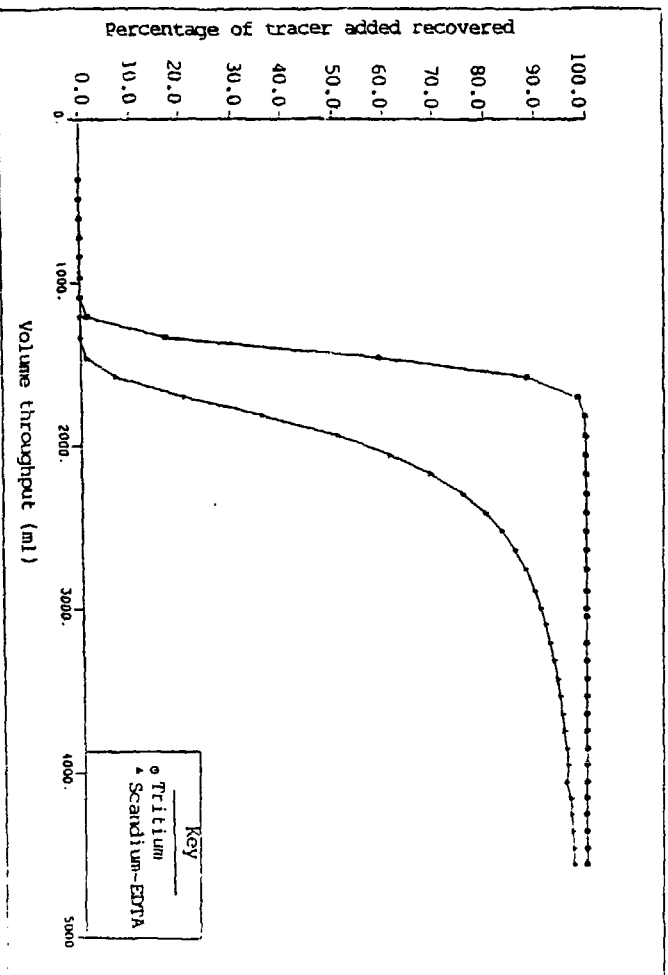


Fig. 4 Limestone column test with double tracer, tracer recovery curve(Exp. No. 1)

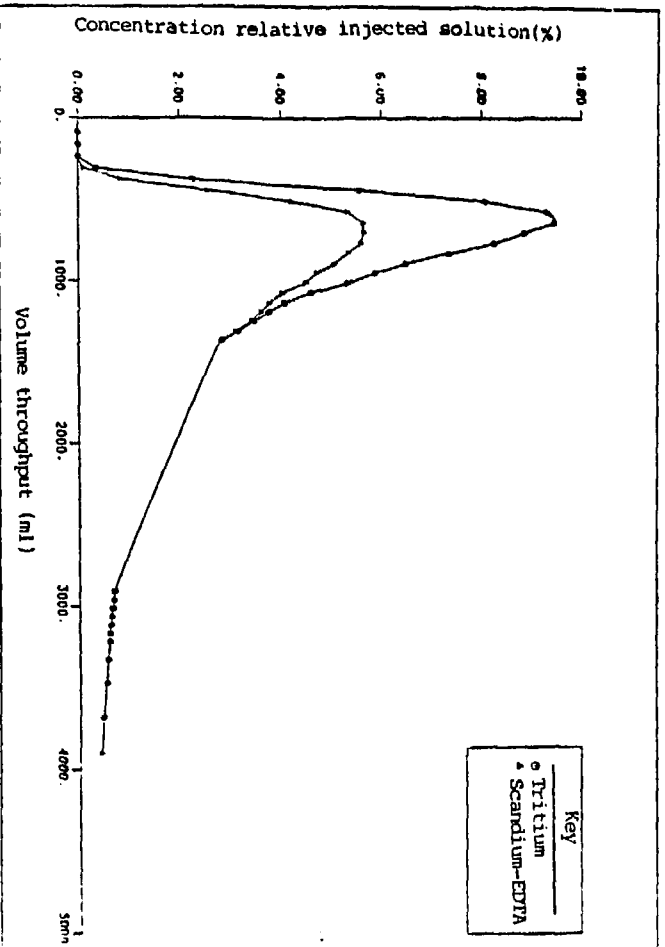


Fig. 5 Chalk column test with double tracer, elutriation of tracers (Exp. No. 2)

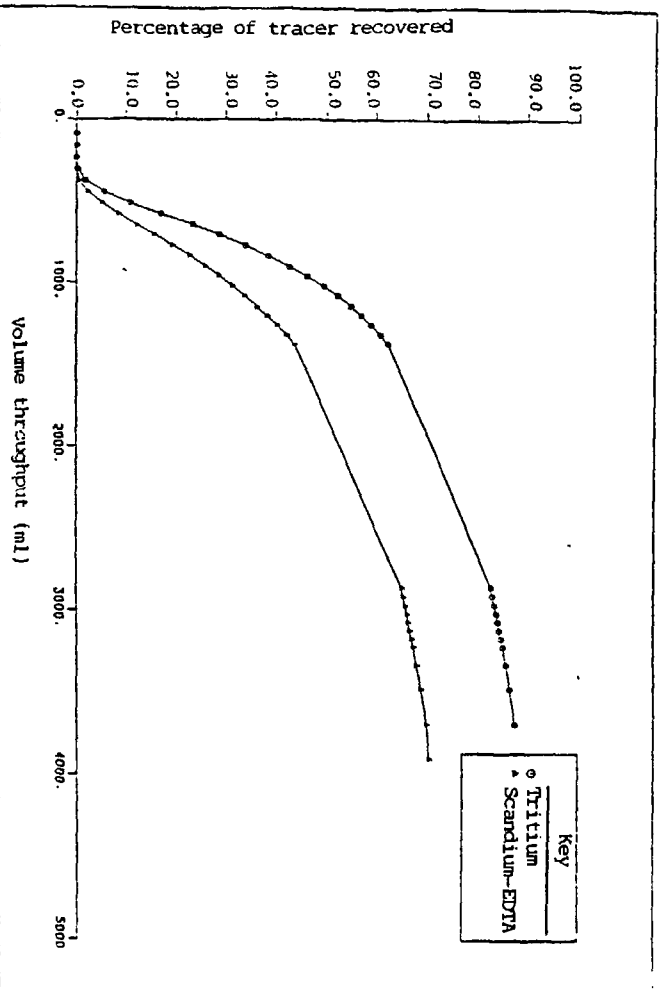


Fig. 6 Chalk column test with double tracer, tracer recovery curve (Exp. No. 2)

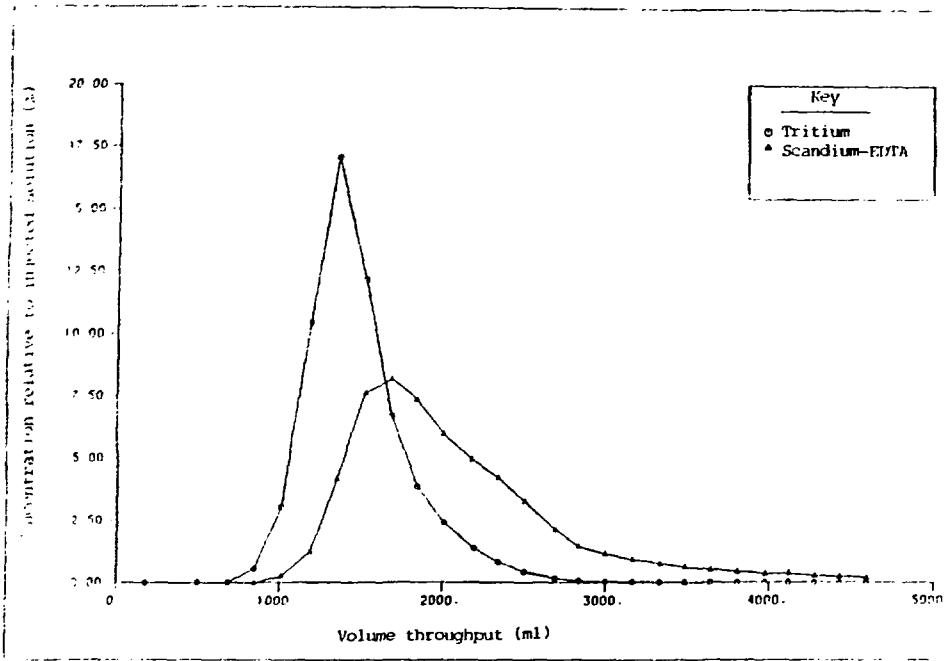


Fig. 7 Sand column test with double tracer, elutriation of tracers (Exp. No. 3)

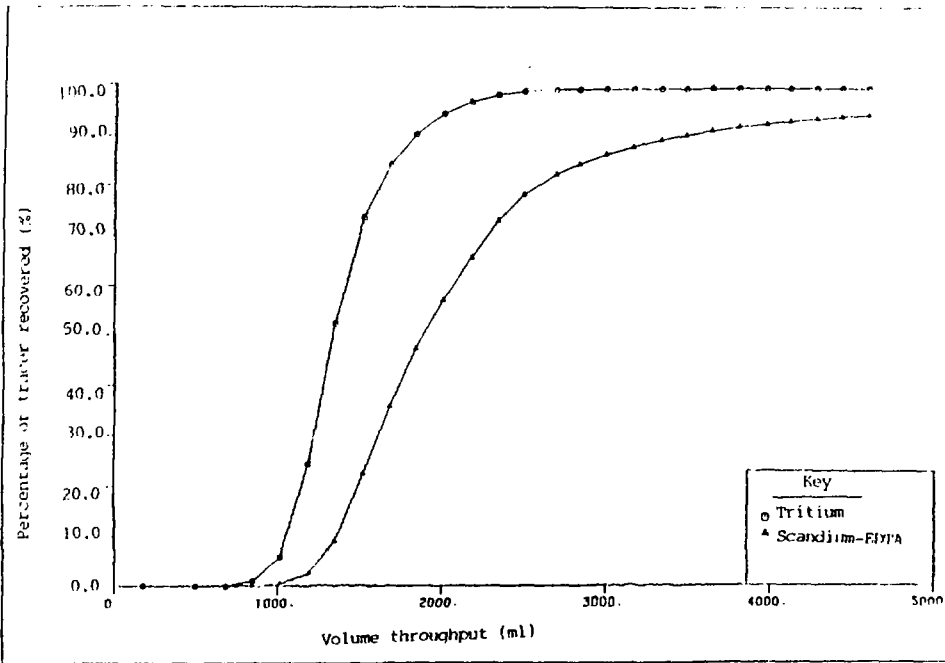


Fig. 8 Sand column test with double tracer, tracer recovery curve (Exp. No. 3)

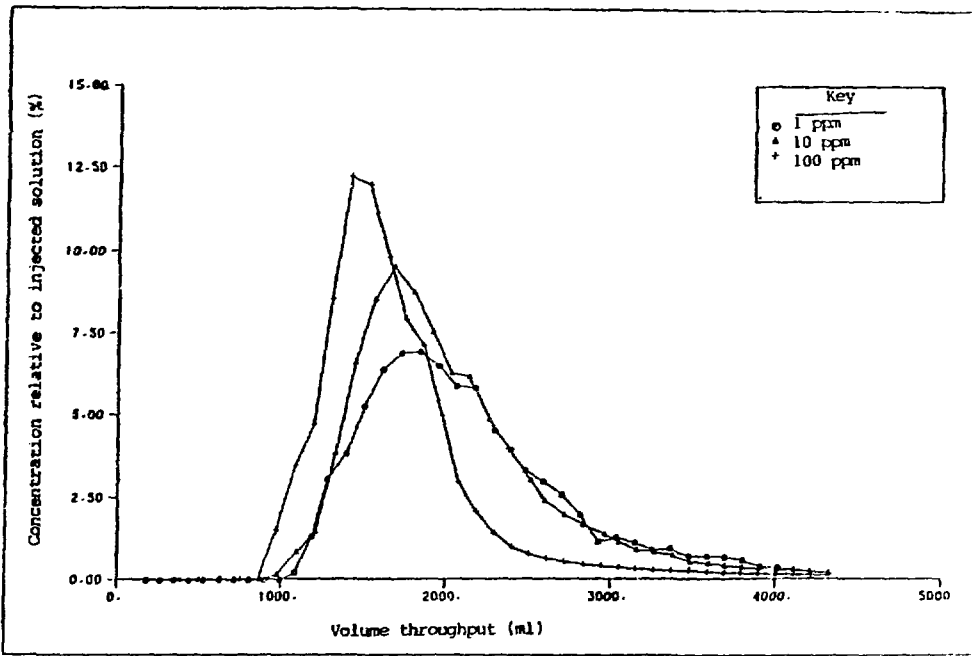


Fig. 9 Elutriation of tracer through sand columns with scandium-EDTA at different concentrations (Exp Nos 4-6)

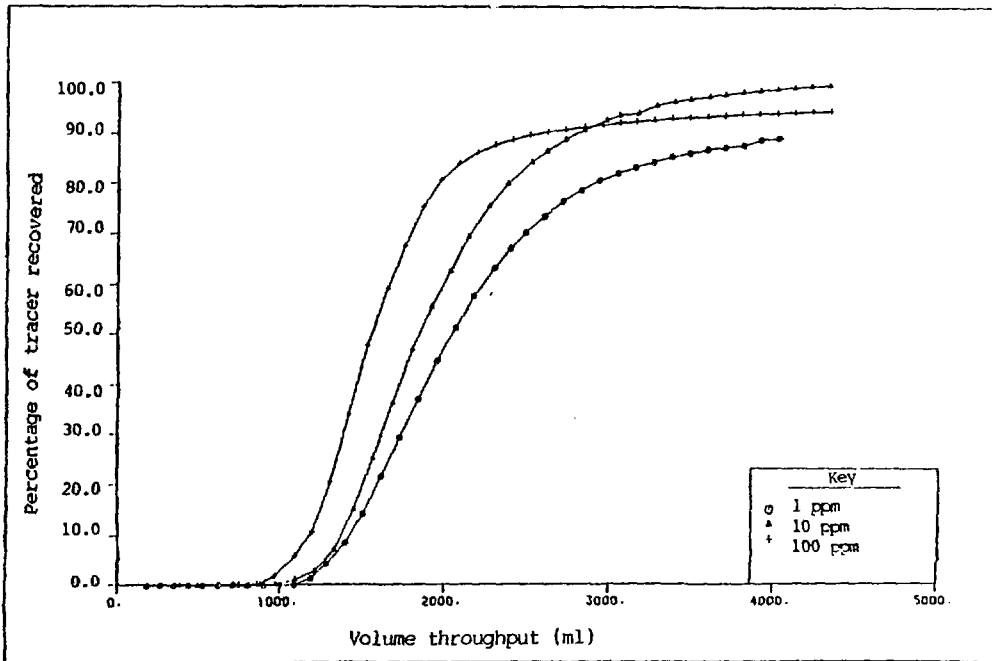


Fig. 10 Tracer recovery curves from sand column tests with scandium-EDTA at different concentrations (Exp Nos 4-6)

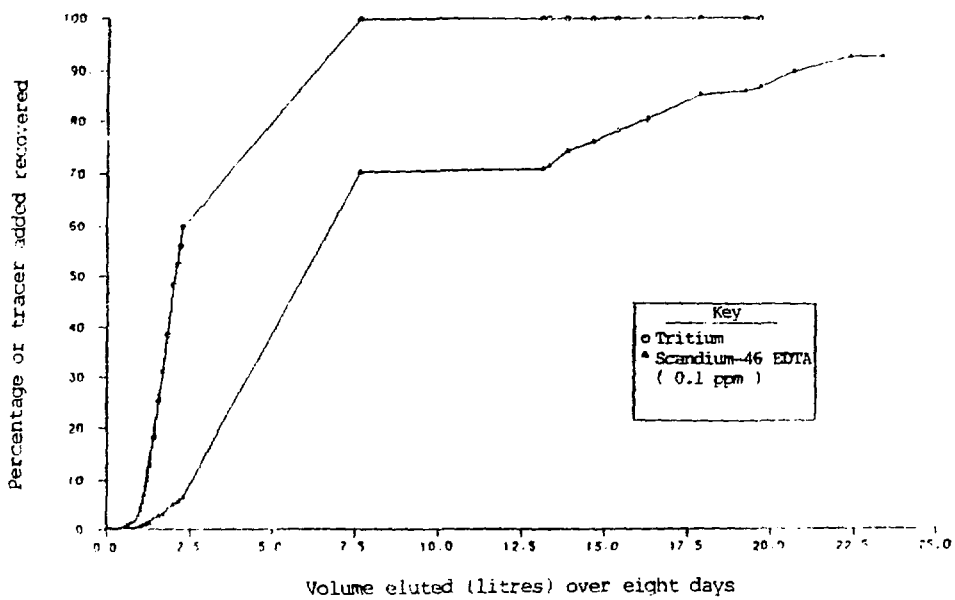


Fig. 11 Chalk column test with double tracer, tracer recovery curve (Exp. No. 7)