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**STUDIES ON SORPTION BEHAVIOUR
OF TECHNETIUM IN SOILS**

March 1995

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Studies on Sorption Behaviour of Technetium in Soils

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The technetium sorption behaviour in different soils has been studied by batch experiments under aerobic conditions. The soil samples characteristics and soil pre-equilibrium water properties of pH-Eh have been also investigated. In addition, the activated carbon and reduced iron powder have been selected as the additives to the JAERI sand according to the research work, and the technetium sorption behaviour in the artificial soils has been studied in the similar conditions. The experiment results show that all soils except for gley soil have little distribution coefficient for Tc, while the artificial soils have very high distribution coefficient for Tc. The distribution coefficient K_d values have an increase trend with the additive increase in artificial soils and contact time. Physico-chemical processes of fixation and possible sorption modes are discussed as well.

Keywords: Soils, Artificial Soils, Aerobic Condition, Sorption Behaviour, Batch Experiment, Distribution Coefficient, Technetium

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土壤中のテクネチウムの吸着挙動に関する研究

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土壌の性質と土壌平衡水の性質との関係から、種々の土壌に対するテクネチウムの吸着挙動を好気条件下でバッチ法により調べた。また、原研砂に活性炭あるいは鉄粉を添加した人工的な土壌試料に対するテクネチウムの吸着挙動について同様に調べた。実験結果からグライ土を除いた全ての土壌においてテクネチウムの分配係数は非常に小さく、人工土壌試料の分配係数は非常に大きいことが分かった。この分配係数は人工土壌の混合物が多いほど、反応時間が長いほど大きい傾向が見られた。核種の固定に関する物理化学的な過程や吸着様式について検討した。

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1. Introduction

Technetium-99, produced by the fission of ^{235}U and ^{239}Pu with a thermal neutron fission yield of 6.13%, has a radiological half-life of 2.15×10^5 years. It has been dispersed in the environment via nuclear weapon testing, releases from nuclear establishments using it for medical or industrial purposes. And, it could also reach the environment via releases from nuclear fuel processing facilities and radioactive waste disposal vault. Thus, technetium could enter into environment via manifold paths, and has been discharged into water, air, and mostly in the low level radioactive waste. Only the radionuclides with the long lived could reach the biosphere, and only the most mobile nuclides could escape from the waste disposal site and surroundings. With respect to ^{99}Tc , it has both of these properties, so the migration behaviour of Tc in the geosphere is of great interest.

Technetium usually exists as anion TcO_4^- in environment. And sorption in rocks and soils was low[1,2,3]. The sorption was influenced by the atomsphere, pH, Eh, etc., because of its redox-sensitive property[4,5,6,7]. Some research works focused on minerals and artificial materials[8,9,10,11], which may be used for additives to the backfill to retard the migration of Tc. It has been found that the activated carbon, reduced iron powder, and such minerals as antimony-containing, have high sorption capacity for Tc. After the burial of radioactive wastes, Tc may return to surface environment, mostly entering highly organic deposit in lowland areas. So the sorption behaviour of Tc on highly organic soil (e.g., sediment, peat, etc.) has also been done in laboratory and field tests[1,2,3,4,5].

Activated carbon and iron powder were selected to be candidate additives to soils as artificial barriers in waste disposal site, because the activated carbon and reduced iron powder have high sorption capacity for technetium. They also are general easy-getting commerials. In the future radiowaste disposal site, it may be considered to use some additives for natural soils to improve the retardation for some special radiowaste. So the activated carbon and reduced iron powder are the best choices as the additives for the radiowaste containing technetium. The artificial soils are called for convenience.

Despite the research done to date, the sorption behaviour of Tc on Japanese soils (including natural barriers and artificial materials) have not been studied. To determine the behaviour of Tc under aerobic condition means conditions similar to those expected in a nuclear waste disposal vault; the aerobic condition exists in waste disposal vault prior to removal of atmospheric oxygen by geologic and bacteriological process, especially for low-level nuclear waste disposal by shallow soil burying and Tc transferring from disposal vault to surface land soils. Our present studies were undertaken (1) to examine the variability of Tc sorption among several different typical Japanese shallow land soil samples, some of which were from disposal sites and candidate disposal sites, and one of which was from organic soil site. (2) to further examine the effect of solution parameters (Eh, pH, etc.), and (3) to investigate the sorption behaviour of Tc on the artificial soils.

2. Materials and Methods

2.1 Selection and preparation of the samples

(1) Natural soils

The natural soils were selected from across the north Japan to represent a broad range of chemical and textural characteristics (Table 1). They include samples of different horizons of Simokita, Aomori-ken; a Kuroboku soil; a gley soil and a JAERI sand. They belong to loam, sand, rock, tuff and gley soil (containing high humic matter), respectively. The soil characteristics were showed in Table 1. Mineralogically, the soils consist mainly of quartz, meanwhile the other minerals are only present in small amounts.

The natural soils were air-dried, sieved to a 2000 μ m under . Moisture contents relative to oven-drying at 110°C for 24 hours were measured. The water content data were showed in the Table 1. Subsamples of the soils were taken out, in order to convert all weight data to an oven-dried basis. The gley soil was kept in original moisture state. The chemical characteristics of experimental soils were showed in the Table 2. Some data should be given more attention. The gley soil cation exchange capacity (CEC) value was very high. In addition, it had very high values of anion exchange capacity for SO_4^{2-} and dissolved organic matter (humic acid). All these data had significant difference from that of the other natural soils. The KUROBOKU soil had slightly high CEC value. Both CEC value and AEC value were slightly high.

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SIMOKITA loam showed its slightly high CEC value and slightly high value of anion exchange capacity (AEC) for SO_4^{2-} , while JAERI sand had slightly high exchange cation capacity value for Ca^{2+} .

(2) Artificial soils

Iron powder was product of Wako Pure Chemical Industries L.T.D., 100 mesh under, and kept in sealed box. Activated carbon was produced by Tsurumicoal Co., L.T.D., 1.64mm under, HC-42, made of coconut-husk. The two kinds of materials were weighed to add in JAERI sand in different percentage (0, 1, 5, 10, 20, 100wt%), and mixed up completely, respectively.

2.2 Experiment methods and calculation

The batch experiments used ^{99}Tc , as TcO_4^- , the long-lived beta-emitting isotope of environment concern. Initial TcO_4^- concentration was 42Bq/ml, 4.0×10^{-7} mol/l. four grams of the soils (dried-weigh) were weighed to mix up with 40ml of ^{99}Tc -solution in 50ml centrifugation tubes, then the tubes were incubated in water bath at 25°C, exposed to ambient air, shaking at 60 times/min.

After some contact time, the tubes were put into centrifuge to separate supernatant solution and solid at 3,000rpm for 20 min. 1ml supernatant solution was taken to mix up with 3ml distilled water and 12ml liquid scintillation solution in measurement tube, shaken, and then the concentration of supernatant was determined by liquid scintillation counting technique (PACKARD-2500TR Type LSC, detection limit: about 0.37 Bq/ml). All samples were repeated in triplicate counting. The concentration of ^{99}Tc in the blank was used as the reference for calculating the amount of Tc sorbed to soil in the samples, so the background and initial solution countings were also measured in the same time.

Eh and pH values were measured with pH-Eh meter in soils or mixtures weigh(g)/deioned and distilled water volume(ml)=1:10. Both values changing with contact time were also determined. The pH values after sorption experiment were checked out afterward.

K_d was calculated by the following equation from the β -radioactivities in the solution before and after equilibration:

$$K_d(\text{ml/g}) = \frac{V}{M} \cdot \frac{A-B}{B}$$

- V: Volume of aqueous phase (ml)
M: Weight of oven-dried soil (g)
A: Radioactivity concentration of initial solution (Bq/ml)
B: Radioactivity concentration of supernatant solution (Bq/ml)

Kd values showed in results were the average values. Negative Kd values occurred in some samples where sorption was minimum, the Kd values appeared in results as zero in that case because it is impossible to be negative Kd values in theory. And if the supernatant counting was very near the initial solution counting, the given Kd value was possibly inaccurate because of measurement limitation and error range. In our experiment, when the Kd value was below 0.03ml/g (Namely the sorption percent was smaller than 0.3%), the Kd was expressed as zero. There were also other cases in which the sorption was so high to give meaningful Kd values, because the supernatant counting was below the measurement limitation; in our experiments, when the Kd value was more than 3000 ml/g, i.e., the percent sorption bigger than 99.7%, we think the Kd value was inaccurate, so the Kd value appeared in results as >3,000ml/g.

3. Results and Discussion

3.1 Sorption behaviour of Tc on soils

The Kd values of the soils were shown in Table 3. No sorption was observed on the batch experiment for five soils: JAERI sand, SIMOKITA sand, SIMOKITA tuff-sand, SIMOKITA tuff-rock and TOHOKU sand-rock, even though the contact time was as long as 48 days. In contrast, the gley soil has the growing Kd values with increase of contact time, meanwhile a very small amount of Tc sorbed on SIMOKITA loam and KUROBOKU soil. This may be explained by pH-Eh diagram and soil compositions.

Because the Tc sorption is more sensitive to pH and Eh values, the measured pH and Eh values were given out in Fig. 1, as showed, the pH and Eh values have no significant changes with contact time. For SIMOKITA loam and KUROBOKU soil, the pH values of experimental soil solution range between 5--7, for gley soil between 2.8--2.9, for other five soils between 6--8. Except for gley soil, Eh values as high as 430mv, others between 200-360mv (see Fig.2). The chemical form of ^{99}Tc on these pH-Eh values existed as anionic form (TcO_4^-) under aerobic condition.

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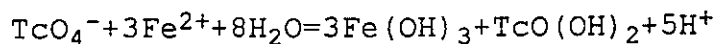
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For SIMOKITA loam and KUROBOKU soil, the little amount of sorption may be contributed by their pH values, and relatively high Fe(II) and Fe(III) mineral composition in those two kinds of soils. On one hand, as seen from Fig.1, the pH values of SIMOKITA loam and KUROBOKU soils are relatively low, Tc likely exists towards to Tc(IV) as pH values down. On the other hand, the two kinds of soils contain relatively high percentages of FeO and Fe₂O₃, so TcO₄⁻ may be reduced by Fe(II) according to the reaction



Hydrolytic adsorption of TcO(OH)₂ on Fe(III) hydroxide or coprecipitation with Fe(III) by formation of hydroxocomplexes containing Fe-O-Tc bonds can be expected^[2,6]. So the K_d values of SIMOKITA loam and KUROBOKU soil of high containing of reducing matter were little high compared with sandy soils. But in case of JAERI sand, even though the Fe(II) and Fe(III) mineral composition contents are relatively high, its K_d values show no sorption had happened. The contradictory phenomena are needed to investigate deeply.

For gley soil, the K_d values increase with contact time. This indicates the sorption equilibrium needs a long time to reach, the kinetics of sorption is relatively slow. Organic material in gley soil (see Table 2) plays a major role in decreasing Tc mobility^[6], so the humic substance in gley soil may strongly affect the migration of Tc in a swamp or bog. The highly polar positively charged organics easily hold and reduce TcO₄⁻ to insoluble tetravalent compound such as TcO₂ thereby retarding its movement through soil. After the gley soils were sampled from the site, they had been kept in laboratory for such some time that sulfur(S) and thiosulfate(S₂O₃²⁻) in the natural soil were transformed to sulfate(SO₄²⁻) in the biochemical reaction caused by thiobacillus^[12] (see Table 2). Generally, this transformation resulted in decrease of pH and increase of Eh; but the organic matter in gley soils played the more important role than Eh effect and the effect of slightly high content of Fe for technetium adsorption. In addition, the sorption behaviour of Tc on gley soils could be also useful to predict the Tc migration in bogs and marshes.

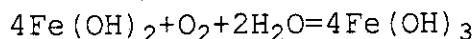
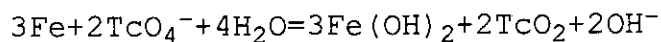
3.2 Sorption behaviour of Tc on artificial soils

The sorption behaviour of Tc on artificial soils containing JAERI sand and activated carbon or reduced

iron powder in different percentage (0, 1, 5, 10, 20, 100wt%) has been studied under the similar condition as on the natural soils. The K_d values are listed in Table 4. The Eh and pH values of artificial soil and distilled water system have been measured before adsorption and after adsorption, the results are showed in Fig. 3.

For the Eh and pH values for artificial soils containing JAERI sand and activated carbon, the Eh values range between 170-270mv and pH between 7-8.1 (see Fig.2). The significant change of pH and Eh values with contact time and adsorption process have not been observed. Even though the pH and Eh values are unfavourable for Tc sorption as seen from Fig. 2, the K_d values appeared very high, and have the increase trend with contact time and additive percentage according to the inaccurate sample counting. The activated carbon perhaps sorbed Tc on its large specific surface area by physical or chemical adsorption mechanism. And even if the percentage of activated carbon is small and contact time is short, the high K_d values can be observed. This indicates that the sorption rate is fast and sorption equilibrium is relative easy to reach, the activated carbon has high sorption capacity for technetium with relative low weight compared to other materials. The results also indicate that the high sorption capacity of activated carbon has not been influenced by its production of raw materials, because in reference[8] the activated carbon was made of apricot-husk, meanwhile the activated carbon used in this experiment was made of coconut-husk.

As for the Eh and pH values for artificial soils containing JAERI sand and reduced iron powder, Eh values became smaller as the increase of contact time and the additive percentage, while the pH values became larger with the additive percentage, kept constant values with the increase of contact time, and became a little bigger after adsorption. Its K_d values are very high, and like activated carbon, have the increase trend with the contact time and additive percentage. The results also show the sorption rate for this kind of soil was slow compared to that of artificial soil containing activated carbon in the same condition. The reduced iron powder has very low Eh value to reduce TcO_4^- to form Tc(IV) insoluble compound co-precipitated with $Fe(OH)_3$ or Tc-Fe complexes. The following reactions are expected to occur in the experimental conditions



So the artificial soils containing reduced iron powder has high adsorption capacity for Tc.

It can be generally summarized that activated carbon and reduced iron powder appear to be favourable backfill materials for retarding redox-sensitive technetium migration.

4. Conclusion

The sorption behaviour studies of Tc by batch experiments clearly showed that Tc could not be retarded by the natural soils except for gley soil, but the artificial soils containing JAERI sand and activated carbon or reduced iron powder have high sorption capacity for Tc, and the capacity increases with contact time and additive percentage. Soil properties and Eh, pH values of soil solution could influence the sorption behaviour of Tc. Column experiments about Tc migration in soils and artificial soils are on progress. Further investigation emphasis will be placed on sorption mechanisms, effect of soil properties and anaerobic condition etc., with some deep research for gley soil.

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Table 1 Selected Soil Properties

	JAERI		SIMOKITA		SIMOKITA		SIMOKITA		SIMOKITA		TOHOKU		KUROBOKU		gley	
	sand		sand	loam	tuff-sand	tuff-rock	sand-rock	soil	soil						soil	
Density, g/cm ³	2.69		2.68	2.77	2.67	2.43	2.74	2.72							2.69	
Gravel, >2000um, wt%	0.50		0.00	0.00	0.00	0.00	4.00	0.00							0.00	
sand, 74-2000um, wt%	96.50		88.00	8.50	87.50	26.00	76.00	16.00							4.00	
silt-clay, <74um, wt%	3.00		12.00	91.50	12.50	74.00	20.00	84.00							96.00	
Mineral composition																
wt%																
SiO ₂	63.00		77.48	50.56	52.29	68.62	72.89	51.59							50.51	
Al ₂ O ₃	8.50		10.68	21.16	23.77	13.68	11.35	21.28							15.11	
TiO ₂	0.87		0.19	0.91	0.57	0.23	0.29	1.23							0.71	
Fe ₂ O ₃	3.53		1.83	6.81	4.62	1.06	2.48	3.66							2.70	
FeO	3.30		1.04	2.07	0.51	1.11	1.32	6.54							3.90	
CaO	4.13		2.22	0.47	2.60	2.25	3.33	1.15							1.15	
MgO	2.45		0.91	1.30	1.73	0.67	1.46	1.71							0.59	
Na ₂ O	6.21		1.61	0.82	1.61	2.78	1.93	0.59							0.56	
K ₂ O	5.41		0.50	1.44	0.53	2.69	1.09	0.59							0.73	
P ₂ O ₅			0.10	0.20	0.06	0.05	0.08	0.32							0.07	
MnO	0.13		0.03	0.08	0.08	0.06	0.09	0.15							0.04	
H ₂ O(+)	2.04		2.68	13.46	11.19	4.51	2.25	9.48							12.42	
H ₂ O(-)			0.08	0.06	0.06	1.07	1.06	1.76							6.19	
Water content, wt%	0.3		1.9	25.2	1.7	3.2	4.2	13.6							63.1	

Table 2 Chemical Characteristics of Experimental Soils

	JAERI Sand	SIMOKITA Sand	SIMOKITA loam	SIMOKITA tuff-sand	SIMOKITA tuff-rock	TOHOKU sand-rock	KUROBOKU soil	Gley soil
Cation exchange capacity(CEC)	2.7	4.6	16.9	5.0	12.8	29.1	19.3	33.4
Exchange cation	Na ⁺	0.5	0.8	1.0	2.0	2.2	0.6	0.9
(meq/100 g dry soil)	1.1	0.4	1.5	0.5	1.7	1.1	0.5	0.3
	K ⁺	0.4	0.9	0.5	4.0	5.0	5.4	12.5
	Ca ²⁺	0.6	1.6	0.7	5.8	13.3	0.5	8.7
	Mg ²⁺	0.3	1.4	0.5	0.3	1.4	0.5	0.9
Anion exchange capacity(AEC)	0.2	1.8	2.3	0.3	0.2	0.6	1.3	68.6
Exchange anion	SO ₄ ²⁻	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
(meq/100 g dry soil)	HCO ₃ ⁻	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
	PO ₄ ³⁻	1.4	1.1	1.6	7.2	1.1	<1.0	7.0x10 ³
Dissolved organic matter (humic acid)	mgC/L	<1.0	<1.0	<1.0	<1.0	<0.5	<1.0	0.5
	mgN/L	1.4	<1.0	<1.0	<1.0	<0.5	<1.0	0.5

Table 3 Distribution Coefficient Kd of Soils, ml/g

contact time day	JAERI	SIMOKITA		SIMOKITA		SIMOKITA		SIMOKITA		TOHOKU		KUROBOKU		gley
	sand	sand	loam	tuff-sand	tuff-rock	sand-rock	soil	sand-rock	soil	sand-rock	soil	sand-rock	soil	soil
1		0	0.84	0	0	0	0.16	0	0	0	0.16	0	0	3.61
3		0	0.84	0	0	0	0.09	0	0	0	0.09	0	0	18.78
11		0	0.92	0	0	0	0.07	0	0	0	0.07	0	0	626.5
30		0	0.97	0	0	0	0.13	0	0	0	0.13	0	0	>3000
48		0	0.85	0	0	0	0.08	0	0	0	0.08	0	0	>3000

Table 4 Distribution Coefficient Kd of Artificial Soils, ml/g

contact time day	Activated carbon percent in mixture						Reduced iron powder percent in mixture					
	1%	5%	10%	20%	100%		1%	5%	10%	20%	100%	
1	237	1230	1140	815	1050		43	135	175	269	162	
3	238	1120	1210	795	1110		44	138	185	283	170	
11	553	>3000	>3000	>3000	>3000		620	930	2480	>3000	>3000	
27	443	>3000	>3000	>3000	>3000		>3000	>3000	>3000	>3000	>3000	
45	348	>3000	>3000	>3000	>3000		2390	>3000	>3000	>3000	>3000	

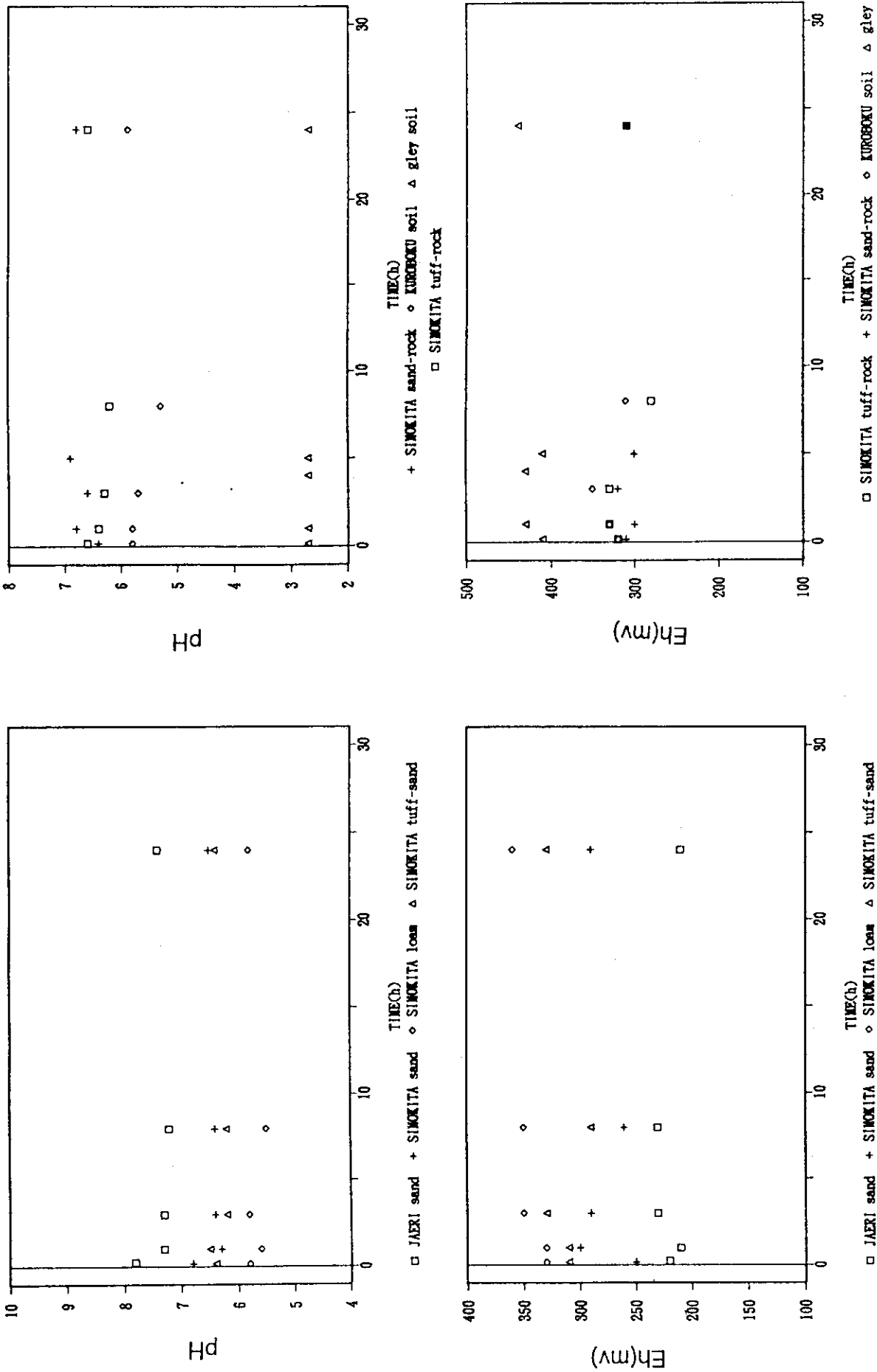


Fig. 1 Change of pH and Eh of experimental soil solution with contact time

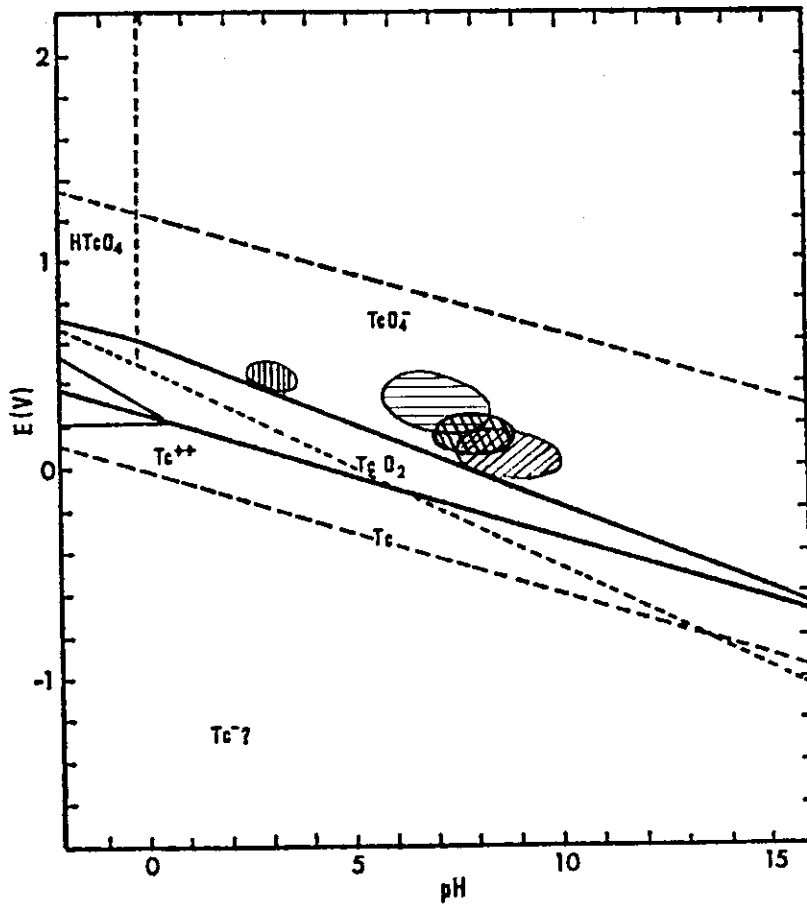






Fig.2 Potential-pH equilibrium diagram for the system technetium-water, at 25°C¹⁷⁾

-  Natural soils except for gley soil,  Gley soil
-  Artificial soil containing activated carbon
-  Artificial soil containing reduced iron powder

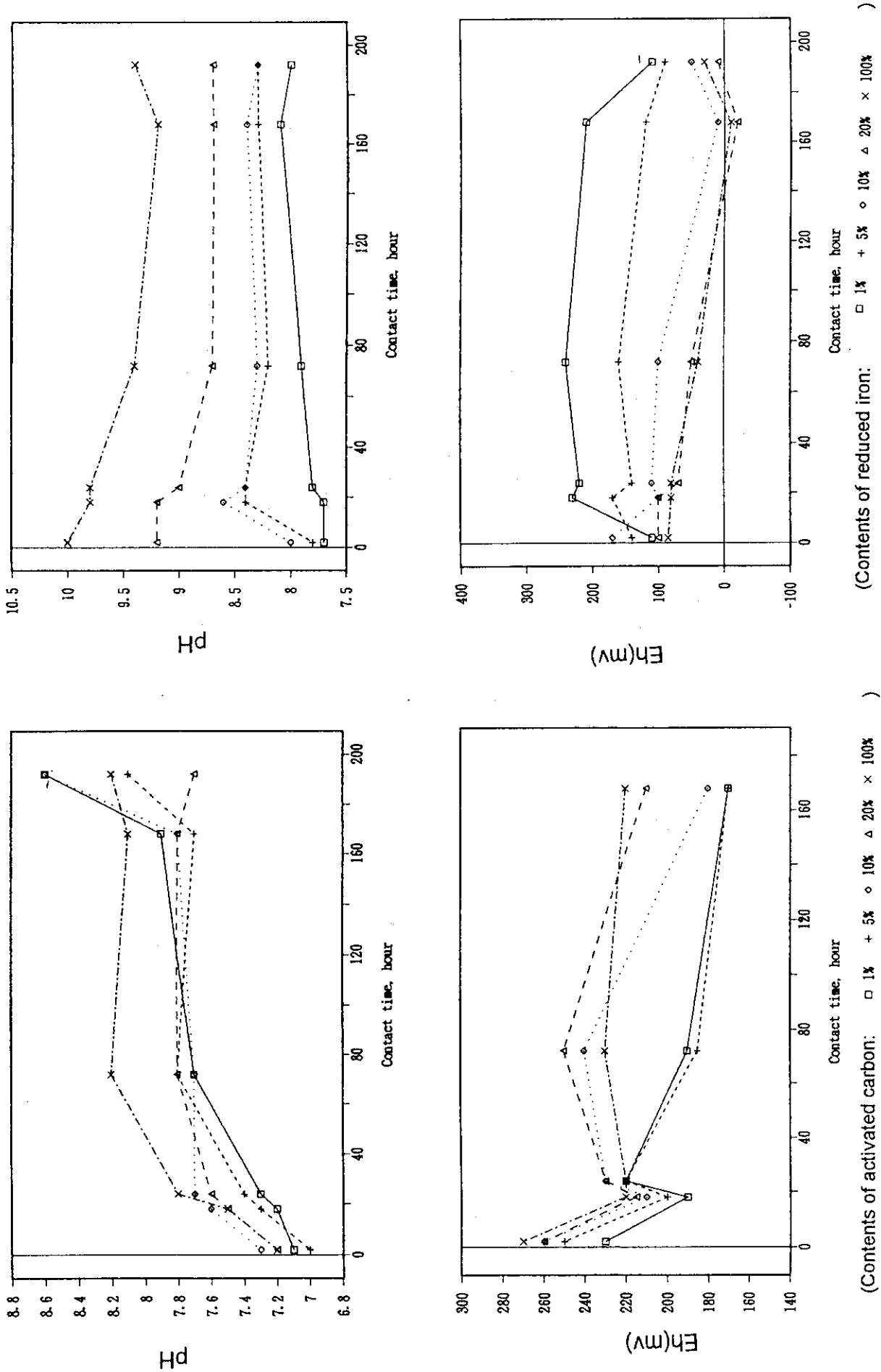


Fig. 3. Change of pH and Eh of artificial soil solution with contact time