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DETERMINATION OF DISTRIBUTION RATIO FOR ^{60}Co, ^{85}Sr AND ^{134}Cs IN LOESS MEDIUM

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Tadao TANAKA, Hiromichi OGAWA
Zhentang LI*, Hui WANG*
and Hideo KAMIYAMA**

日本原子力研究所
Japan Atomic Energy Research Institute

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Determination of Distribution Ratio for ^{60}Co, ^{85}Sr and ^{134}Cs in Loess Medium

Tadao TANAKA, Hiromichi OGAWA[+], Zhentang LI[*]
Hui WANG[*] and Hideo KAMIYAMA[**]

Department of Environmental Safety Research
Tokai Research Establishment
Japan Atomic Energy Research Institute
Tokai-mura, Naka-gun, Ibaraki-ken

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Distribution ratio R for ^{60}Co, ^{85}Sr and ^{134}Cs between loess and ground water was measured by using 16 loess samples taken from the field test site of China Institute for Radiation Protection, in which the field test of radionuclide migration had been performed.

The R of each radionuclide was taken nearly constant value among 16 samples. This revealed that the site can be regarded as uniform loess medium from the view point of sorption ability. The sorption ability between the three radionuclides was the order of $^{134}Cs > {}^{60}Co >> {}^{85}Sr$. The ^{85}Sr was an available nuclide for examining the migration phenomena, because ^{85}Sr might easily migrate in the loess medium due to its low sorption ability. The results of parametric check suggested that it is necessary to select the valid temperature and pH for determining the R value. The sorption of Sr on the loess was dominated by reversible ion-exchange reaction, while that of Co and Cs was dominated by irreversible one.

The study was carried out under a cooperative research between Japan Atomic Energy Research Institute and China Institute for Radiation Protection.

+ Office of Planning
* China Institute for Radiation Protection
** Socio-Energy System Co., Ltd.

Keywords: Loess, Ground Water, Distribution Ratio, ^{60}Co, ^{85}Sr, ^{134}Cs, Sorption, Desorption Sorption Mechanism, Temperature, pH, Chemical Extraction.

黄土中における ^{60}Co, ^{85}Sr 及び ^{134}Cs の分配比の測定

日本原子力研究所東海研究所環境安全研究部
田中　忠夫・小川　弘道[+]・李 禎 堂[*]
王　　輝[*]・神山　秀雄[**]

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放射性核種野外移行試験を実施してきた中国輻射防護研究院の野外試験場内16地点から採取した黄土を対象として、黄土と地下水との間における ^{60}Co, ^{85}Sr 及び ^{134}Cs の分配比を測定した。

16試料に対する各放射性核種の分配比はほぼ一定の値を示し、野外試験場は放射性核種の収着性の観点から均一な黄土層であるとみなすことができた。収着親和性の大きさは $^{134}Cs > ^{60}Co \gg ^{85}Sr$ の順であった。収着親和性の小さな ^{85}Sr は黄土層中を移行し易いので、移行現象を調べるための有効な核種であることが分かった。分配比に及ぼす種々のパラメータの影響を調べた結果、分配比の測定においては適切な温度及び pH を選定する必要があることが示唆された。また、黄土への Sr の収着は可逆的なイオン交換反応に支配されているが、Co 及び Cs の収着は非可逆的な反応に支配されていることが分かった。

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東海研究所：〒319-11　茨城県那珂郡東海村白方字白根2－4
＋　企画室
＊　中国山西省太原市中国輻射防護研究院
＊＊　㈱ソシオ・エナージー・システム

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1. Introduction

In the assessment of environmental impact on shallow land burial of low level radioactive waste, the migration of radionuclide released from the wastes into local ground water flow system is one of the important pathways. Understanding the processes of the radionuclide migration in hydrogeologic environments is therefore an important factor in the design and assessment of subsurface radioactive waste repository[(1)(2)]. We have been performed *in situ* field tracing tests examining radionuclide migration in the natural loess medium at field test site of China Institute for Radiation Protection, by using radioactive tracers: ^{60}Co, ^{85}Sr and ^{134}Cs, to evaluate the radionuclide migration in unsaturated hydrogeologic environments.

It is well recognized that many radionuclides such as ^{60}Co, ^{85}Sr and ^{134}Cs migrate at velocities less than the ground water in environment, as a result of geochemical interactions of the radionuclides with geological materials. Distribution coefficient has usually used as a convenient parameter of quantifying the geochemical interactions[(3)]. The distribution coefficient is defined as an equilibrium distribution of the radionuclides between the solid and solution phase[(4)]. The distribution coefficient for evaluating the radionuclide migration can be roughly predicted from distribution ratio, which is the ratio of radionuclide concentration sorbed on solid phase to that dissolved in liquid phase, measured under static condition by batch method. It is also known that the value of distribution ratio is affected by physico-chemical properties of radionuclide concentration, temperature, pH, *etc.*[(5)~(9)].

Laboratory experiments were made to study the variation of the distribution ratio as well as the influence of contact time of soil and solution, radionuclide concentration, pH and temperature on the distribution ratio of ^{60}Co, ^{85}Sr and ^{134}Cs in the loess medium at the test site. Furthermore, desorption properties of the radionuclides from the loess was investigated, to determine the distribution ratio on desorption process, and to elucidate sorption mechanisms of the radionuclides on the loess.

2. Experimental

2.1 Soil and Ground Water

Soil and ground water were taken from the field test site of China Institute for

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2. Experimental

2.1 Soil and Ground Water

Soil and ground water were taken from the field test site of China Institute for

Radiation Protection (**Fig.1**). Sixteen loess samples were taken from depths of 0.5, 1.0, 1.5 and 2.0 m at 4 sampling points in the test site, and they were sieved into 12 mesh under. Ground water was sampled from a well of 170 m in depth. Physico-chemical properties of the loess and the ground water are listed in **Table 1** and **2**, respectively.

2.2 Sorption Experiment

Distribution ratios were measured by the following method proposed by Relyea *et al.*[10]. Ten milliliters of ground water were added to each sample tube (made of polycarbonate and had 25 ml in volume capacity) containing 1 g of loess sample. The wash procedure was repeated by 3 times. After removal of the third wash solution, each tube must be reweighed and the weight must be recorded to determine the volume of excess solution left in each sample. Then, 10 ml of spiked ground water with ^{60}Co, ^{85}Sr or ^{134}Cs were placed in each tube. After replacing tube caps, three tubes of each parallel sample were placed in a small plastic bag, then several small plastic bags (3 to 5 small bags) were placed in a larger plastic bag to prevent any contamination caused by leaky tubes. Next, the bags were placed into a constant temperature box (25 ℃). The bags were shaken each every day with one's hand. During shake, solid and liquid were mixed until there were not any sticking in tube bottom. In addition, blank tests were conducted to detect sorption of tracer by centrifuge tube walls. After contact between soil and ground water within 168 h, the sample and blank tubes were centrifuged for 20 minutes at 6,000 rpm (5,400 G) to separate the phases. A supernatant of 5 ml was sampled into a counting tube and counting rate of it was measured with well type NaI(Tl) detector.

Distribution ratio R (ml/g) was calculated by the equation:

$$R = \frac{V}{W}\left(\frac{B}{E_s} - 1\right) - \frac{X}{W} \qquad (1)$$

where, V: volume of spiked ground water (ml), W: weight of loess sample (g), B: radioactivity of blank solution (cpm), E_s: radioactivity of supernatant in sample tube (cpm), X: volume of solution left in loess after washing (ml).

In addition, influence of various parameters on R values was examined within following ranges;

a) Effect of contact time : 1 ~ 168 h

b) Effect of concentration : 10^{-8} ~ 10^{-4} mol/l

c) Effect of pH : 5 ~ 10

d) Effect of temperature : 25 ~ 45 ℃.

2.3 Desorption Experiment

Without washing with ground water, 2.5 g of the loess was contacted with 50 ml of deionized water spiked ^{60}Co, ^{85}Sr and ^{137}Cs for 3 days. Liquid phase was separated from the loess by centrifugation and was removed. Residual loess sorbed the radionuclides was used for desorption experiments, as the tracing loess. Liquid phase prepared without the radionuclides was used for determination of the distribution ratio on desorption process, as the equilibrium water.

a) Determination of distribution ratio on desorption process

The tracing loess was contacted with 50 ml of the equilibrium water for 3 days. The mixture were centrifuged to separate the phases. Radioactivity of supernatant was measured by gamma-ray detector.

Distribution ratio R' (ml/g) on desorption process was calculated by the equation:

$$R' = \frac{V}{W} \frac{B - E_s - E_d}{E_d} \qquad (2)$$

where, E_d: radioactivity of supernatant on desorption precess (cpm).

b) Sequential chemical extraction procedure

Extracting reagents and procedures for selective extraction of the radionuclides from the tracing loess were described elsewhere in detail[11][12]. The the tracing loess were sequentially contacted with 50 ml of following 5 kinds of solution;

1st step: 3 times with 0.5 mol/l $CaCl_2$,
to remove a fraction exchanged by Ca^{2+}.

2nd step: 2 times with 0.5 mol/l KCl,
to remove a fraction exchanged by K^+.

3rd step: 2 times with 0.1 mol/l $NH_2OH \cdot HCl$,
to remove a fraction sorbed on manganese oxide.

4th step: 2 times with 0.2 mol/l K-oxalate,
to remove a fraction sorbed on iron oxide.

5th step: 1 time with 30 $_w$% H_2O_2,
to remove a fraction interacted with organic substance and calcite.

The solution extracted the radionuclides was separated from the residue by centrifugation. Radioactivities of the solution and final residue were measured.

3. Results and Discussion

3.1 Distribution of R in Loess Medium

The R values determined for 16 samples are listed in **Table 3**. The R values were not affected by the sampling points as well as the sampling depths, for all three radionuclides. The mean R value X of ^{60}Co, ^{85}Sr and ^{134}Cs were 5,400, 82 and 7,500, respectively. The standard deviation σ_n for each radionuclide was calculated from a population containing the 16 samples. The σ_n was 270 for ^{60}Co, 6.3 for ^{85}Sr and 1,200 for ^{134}Cs, as seen in Table 3. **Table 4** shows a correlation between confidence and error relating to the R values of 16 samples. Even if the confidence of $3\sigma_n$ is required, that is 99.7 %, error of three radionuclides is smaller than 48 % (at ^{134}Cs).

The R of each radionuclide was taken nearly constant value between 16 samples. This reveals that the test site can be regarded as uniform loess medium from the view point of sorption ability and is very suitable for examining the migration behavior of radionuclide.

The R of ^{85}Sr for the loess was markedly smaller than ^{60}Co and ^{134}Cs, as seen in Table 3. It has been well known that sorption of Sr is significantly affected by the coexistence of Ca and Mg ion and it is decreased with increasing concentration of the coexistent cations in solution phase[13][14]. Calcium and strontium belong to the same alkaline earth metal. So the ^{85}Sr ion adsorbed once on the loess will be easily replaced with the Ca ion in ground water by ion exchange reaction[15]. Since the loess mainly consists of Ca compounds, the ground water passed through the loess soil layer contains a great deal of Ca ion, as seen from Table 1 and Table 2. Therefore the low R value of ^{85}Sr is possibly caused by the presence of Ca ion in the ground water.

Strontium 85 having low R value will easily migrate in the loess geological formation. This suggests that ^{85}Sr is the most available radionuclide for examining the migration behavior of radionuclide in the test site.

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3.2 Influence of Various Parameters on R

a) Influence of contact time

The R profiles of ^{60}Co, ^{85}Sr and ^{134}Cs studied as a function of time are shown in **Fig. 2 (a), (b)** and **(c)**, respectively, in which R is normalized by the value measured at the longest contact time. It was observed that there are two sorption processes in the time course of the R. In the first process, the R values of the three radionuclides were immediately increased with a lapse of time. In a second process, the R values of all three radionuclides were kept in each constant, after 70 h for ^{60}Co, 140 h for ^{85}Sr and 110 h for ^{134}Cs. The results show that the main interaction of three radionuclides between the loess and the ground water was equilibrated within 140 h. Possibly the sorption mechanism of all three radionuclides might be mainly contributed to sorption on the external accessible surface of the loess, because the interaction was equilibrated rather immediately[9].

b) Influence of concentration

The result of concentration effect on R is shown in **Fig. 3**, in which R is normalized by the value measured at 10^{-8} mol/l. The R values of ^{134}Cs were constant up to a certain concentration, but in a concentration region higher than 10^{-6} mol/l the R values were decreased with increasing its concentration. On the other hand, the R values of ^{60}Co were not affected by its concentration. Such tendency is possibly caused from that the background concentration of ^{60}Co is rather high, as seen in Table 1. As to ^{85}Sr, the effect of concentration was not examined, because the background concentration of stable strontium is significantly high in both the loess and the ground water, as seen in Table 1 and 2.

c) Influence of pH

Figure 4(a), (b) and **(c)** show R values of ^{60}Co, ^{85}Sr and ^{134}Cs over a pH range between 5 and 10, respectively, in which R is normalized by the value measured at pH 8.1. The R of ^{85}Sr was significantly increased with pH. The R values of ^{134}Cs were also gradually increased with pH, excepting the R of YS-3D. The R values of YS-3D were decreased with increasing pH in a pH range higher than 8.1. It is well known that the negative charge density on the surface of many soils is increased with pH in the solution[6][9]. This suggests that the ^{85}Sr dissolved in the solution of higher pH is adsorbed on the surface of the loess by surface charge attraction[9][16], while the ^{134}Cs may be sorbed

by a specific interaction mechanism besides the surface adsorption.

The R values of ^{60}Co were little affected by pH and its sorption mechanism may be not reflected by the surface adsorption based on surface charge attraction.

d) Influence of temperature

The temperature dependency of R is given in **Fig. 5**, in which R is normalized by the value measured at 25 ℃. The R values of ^{60}Co were most affected by temperature. The R values of ^{60}Co were decreased with increasing temperature but those of ^{134}Cs were opposite, for both the loess samples YS-1D and YS-3D. As to ^{85}Sr, the R values at low temperature were larger than those at high temperature for YS-1D while it was opposite for YS-3D.

The increase of temperature generally cause to increase chemical reaction rates and to decrease physical reaction rate[17]. As to the interactions between minerals and radionuclides, some become more stable at higher temperature and others become opposite[10]. Whatever the direction of a change in the stability of reaction, the stability may be altered as changes of R with temperature.

3.3 Desorption Properties of Radionuclide

Distribution ratio R' determined on the desorption process is compared with R determined on the sorption process in **Table 5**. The R' value of ^{60}Co and ^{137}Cs was significantly larger than the R. The R' value of ^{85}Sr agreed well with the R. These results suggest that the interaction of ^{85}Sr with the loess is based on a reversible equilibrium reaction, while the interaction of ^{60}Co and ^{137}Cs is not so.

Figure 6 shows the concentration of radionuclides desorbed by each extraction step. The results of chemical extraction is summarized in **Table 6** as a fraction percent of radionuclides desorbed from the tracing loess by each reagent.

All of ^{85}Sr sorbed on the loess was desorbed by $CaCl_2$ and KCl. The sorption of ^{85}Sr on the loess was dominated by reversible ion-exchange reaction. The fact that the sorption ratios of ^{85}Sr on both the sorption and desorption process were closed and the ^{85}Sr was sorbed on the loess by reversible ion-exchange reaction, is expected that the migration of ^{85}Sr through the loess medium must be well expressed by the equilibrium sorption model using the distribution coefficient.

The ^{60}Co sorbed on the loess was extracted with $NH_2OH \cdot HCl$ and K-oxalate by 20.7 and 27.6 %, respectively. The result revealed that most dominant sorption mechanism of

^{60}Co is an association with ferro-manganese oxide. Since ferro-manganese oxide do not easily dissolve into ground water in the natural environmental condition, the association with forro-manganese oxide can be regarded as irreversible sorption. In addition, about 20 % of the sorbed ^{60}Co was not extracted by all the extracting procedure. More than 80 % of ^{137}Cs sorbed on the loess could not be extracted and 11.4 % of that was extracted with KCl. This shows that the most dominant sorption of ^{137}Cs can be attributed to the interaction with layer silicates. Most of ^{137}Cs interacted with layer silicates is irreversibly fixed, though K^+ can partially replace Cs^+. The reason that the R' of ^{60}Co and ^{137}Cs did not agree with the R is attributable to irreversible sorption of ^{60}Co and ^{137}Cs onto the loess. So, the migration of ^{60}Co and ^{137}Cs in the loess medium seems to be difficult to evaluate by the equilibrium sorption model using the distribution coefficient.

4. Conclusion

Distribution ratio R for ^{60}Co, ^{85}Sr and ^{134}Cs between loess and ground water was measured by using 16 loess samples taken from the field test site of China Institute for Radiation Protection.

The distribution data of R in the test site indicated that the test site can be regarded as uniform loess medium from the view point of sorption ability.

The R between the three radionuclides was the order of $^{134}Cs > {}^{60}Co >> {}^{85}Sr$. The ^{85}Sr is an available nuclide for examining the migration behavior, because ^{85}Sr will be easily migrated in the loess medium due to its low sorption ability.

The main interaction between the loess and the ground water was equilibrated within 140 h. The R value was not affected by its concentration in extremely low concentration region. The R was affected by temperature and pH. When the adequate distribution coefficient for evaluating the radionuclide migration is determined from R values measured by batch method, the effects of temperature and pH on R have to be taken into consideration.

The sorption of Sr on the loess was dominated by reversible ion-exchange reaction, while that of Co and Cs was dominated by irreversible one.

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REFERENCES

1 IAEA; *IAEA Safety Series No.53*, (1981).

2 J.F.McCarthy and J.M.Zachara; *Environ.Sci.Technol.*, **23**[5], 496 (1989).

3 Y.Inoue and W.J.Kaufman; *Health Phys.*, **9**, 705 (1963).

4 H.M.Selim, J.M.Davidson and P.S.C.Rao; *Soil Sci.Soc.Am.J.*, **41**, 3 (1977).

5 A.J.Weiss and P.Colombo; *NUREG/CR-1289, BNL-NUREG-51143*, (1979).

6 T.Tanaka, T.Yamamoto, S.Koto and H.Kazama; *J.At.Energy Soc.Japan*, **33**[4], 373 (1991)(in Japanese).

7 T.Tanaka and T.Yamamoto; *ibid*, **30**[10], 933 (1988)(in Japanese).

8 R.A.Sallach; *SC-RR-67-861*, (1967).

9 B.Torstenfelt, K.Andersson and B.Allard; *Chemical Geology*, **36**, 123 (1982).

10 J.F.Relyea, R.J.Serne and D.Rai; *PNL-3349*, (1980).

11 T.Tanaka, K.Sriyotha and H.Kamiyama; "*Proceedings of the third international conference on Nuclear fuel reprocessing and waste management (RECOD'91)*", 1011 (1991).

12 T.Tanaka, K.Sriyotha and H.Kamiyama; *JAERI-M 93-109*, (1993).

13 S.Uchida and H.Kamata; *Hoken Butsuri*, **22**, 179 (1987)(in Japanese).

14 T.Tsutsui and K.Nishimaki; *ibid*, **10**, 79 (1975)(in Japanese).

15 D.B.Hawkins and H.L.Short; *IDO-12046*, (1965).

16 M.Saiki; "*Environmental Radioactivity*", 170 (1984), Soft Science, Inc., (in Japanese).

17 T.Keii; "*Kyu-chaku*", 3 (1983), Kyoritsu, (in Japanese).

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REFERENCES

1 IAEA; *IAEA Safety Series No.53*, (1981).

2 J.F.McCarthy and J.M.Zachara; *Environ.Sci.Technol.*, **23**[5], 496 (1989).

3 Y.Inoue and W.J.Kaufman; *Health Phys.*, **9**, 705 (1963).

4 H.M.Selim, J.M.Davidson and P.S.C.Rao; *Soil Sci.Soc.Am.J.*, **41**, 3 (1977).

5 A.J.Weiss and P.Colombo; *NUREG/CR-1289, BNL-NUREG-51143*, (1979).

6 T.Tanaka, T.Yamamoto, S.Koto and H.Kazama; *J.At.Energy Soc.Japan*, **33**[4], 373 (1991)(in Japanese).

7 T.Tanaka and T.Yamamoto; *ibid*, **30**[10], 933 (1988)(in Japanese).

8 R.A.Sallach; *SC-RR-67-861*, (1967).

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10 J.F.Relyea, R.J.Serne and D.Rai; *PNL-3349*, (1980).

11 T.Tanaka, K.Sriyotha and H.Kamiyama; "*Proceedings of the third international conference on Nuclear fuel reprocessing and waste management (RECOD'91)*", 1011 (1991).

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13 S.Uchida and H.Kamata; *Hoken Butsuri*, **22**, 179 (1987)(in Japanese).

14 T.Tsutsui and K.Nishimaki; *ibid*, **10**, 79 (1975)(in Japanese).

15 D.B.Hawkins and H.L.Short; *IDO-12046*, (1965).

16 M.Saiki; "*Environmental Radioactivity*", 170 (1984), Soft Science, Inc., (in Japanese).

17 T.Keii; "*Kyu-chaku*", 3 (1983), Kyoritsu, (in Japanese).

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REFERENCES

1 IAEA; *IAEA Safety Series No.53*, (1981).

2 J.F.McCarthy and J.M.Zachara; *Environ.Sci.Technol.*, **23**[5], 496 (1989).

3 Y.Inoue and W.J.Kaufman; *Health Phys.*, **9**, 705 (1963).

4 H.M.Selim, J.M.Davidson and P.S.C.Rao; *Soil Sci.Soc.Am.J.*, **41**, 3 (1977).

5 A.J.Weiss and P.Colombo; *NUREG/CR-1289, BNL-NUREG-51143*, (1979).

6 T.Tanaka, T.Yamamoto, S.Koto and H.Kazama; *J.At.Energy Soc.Japan*, **33**[4], 373 (1991)(in Japanese).

7 T.Tanaka and T.Yamamoto; *ibid*, **30**[10], 933 (1988)(in Japanese).

8 R.A.Sallach; *SC-RR-67-861*, (1967).

9 B.Torstenfelt, K.Andersson and B.Allard; *Chemical Geology*, **36**, 123 (1982).

10 J.F.Relyea, R.J.Serne and D.Rai; *PNL-3349*, (1980).

11 T.Tanaka, K.Sriyotha and H.Kamiyama; "*Proceedings of the third international conference on Nuclear fuel reprocessing and waste management (RECOD'91)*", 1011 (1991).

12 T.Tanaka, K.Sriyotha and H.Kamiyama; *JAERI-M 93-109*, (1993).

13 S.Uchida and H.Kamata; *Hoken Butsuri*, **22**, 179 (1987)(in Japanese).

14 T.Tsutsui and K.Nishimaki; *ibid*, **10**, 79 (1975)(in Japanese).

15 D.B.Hawkins and H.L.Short; *IDO-12046*, (1965).

16 M.Saiki; "*Environmental Radioactivity*", 170 (1984), Soft Science, Inc., (in Japanese).

17 T.Keii; "*Kyu-chaku*", 3 (1983), Kyoritsu, (in Japanese).

Table 1 Physico-chemical properties of loess soil

Term	Unit	Measured value
Density	(g/cm^3)	2.64～2.74
Porosity	(%)	45.3～53.0
Permeability	($\times 10^{-4}$ cm/s)	0.64～6.93
CEC	(meq/100g)	11.7～21.3
Chemical composition	(%)	
SiO_2		58.2～66.0
Al_2O_3		10.9～11.5
Fe_2O_3		3.0～3.6
TiO_2		0.63～0.70
CaO		6.3～9.6
MgO		1.6～2.2
K_2O		2.0～2.3
Na_2O		1.7～2.0
MnO		
Element	(%)	
Co		0.002
Sr		0.03

Table 2 Physico-chemical properties of ground water

Term	Unit	Measured value
pH		7.07～8.00
Eh	(mV)	336～372
K^+	(mg/l)	0.40～5.02
Na^+	(mg/l)	64.6～110.9
Ca^{2+}	(mg/l)	23.0～563.5
Mg^{2+}	(mg/l)	18.2～166.0
CO_3^{2-}	(mg $CaCO_3$/l)	0.00～37.6
HCO_3^-	(mg $CaCO_3$/l)	118.4～294.7
Cl^-	(mg/l)	23.8～51.9
Co	($\times 10^{-3}$ mg/l)	<5
Sr	(mg/l)	0.58～6.62
Cs	($\times 10^{-5}$ mg/l)	<5

Table 3 Distribution of R in field test site

Sample No.	Sampling position	Sampling Depth (m)	R (ml/g)		
			^{60}Co	^{85}Sr	^{134}Cs
YS-1A	YS-1	0.5	5.0×10^3	7.7×10	7.6×10^3
YS-1B	YS-1	1.0	5.3×10^3	8.5×10	7.9×10^3
YS-1C	YS-1	1.5	5.7×10^3	8.1×10	1.0×10^4
YS-1D	YS-1	2.0	5.7×10^3	7.7×10	9.7×10^3
YS-2A	YS-2	0.5	4.9×10^3	7.5×10	7.2×10^3
YS-2B	YS-2	1.0	5.5×10^3	9.8×10	9.2×10^3
YS-2C	YS-2	1.5	5.4×10^3	8.0×10	6.5×10^3
YS-2D	YS-2	2.0	5.7×10^3	8.0×10	7.3×10^3
YS-3A	YS-3	0.5	5.2×10^3	8.8×10	6.9×10^3
YS-3B	YS-3	1.0	5.4×10^3	7.5×10	5.8×10^3
YS-3C	YS-3	1.5	5.7×10^3	8.2×10	6.6×10^3
YS-3D	YS-3	2.0	5.5×10^3	9.1×10	8.6×10^3
YS-4A	YS-4	0.5	5.2×10^3	7.4×10	6.8×10^3
YS-4B	YS-4	1.0	5.1×10^3	8.1×10	5.6×10^3
YS-4C	YS-4	1.5	5.6×10^3	7.8×10	7.5×10^3
YS-4D	YS-4	2.0	5.8×10^3	8.6×10	7.5×10^3
Average, X			5.4×10^3	8.2×10	7.5×10^3
σ_n			2.7×10^2	6.3	1.2×10^3

Table 4 Correlation between confidence and error on R of 16 samples

Confidence (%)		68.3 (σ_n)	95.5 ($2\sigma_n$)	99.7 ($3\sigma_n$)
		$\sigma_n/X\times100$	$2\sigma_n/X\times100$	$3\sigma_n/X\times100$
Error (%)	^{60}Co	5.0	10	15
	^{85}Sr	7.7	15	23
	^{134}Cs	16	32	48

Table 5 Distribution ratio determined on both sorption and desorption process

Process	Distribution ratio (ml/g)		
	^{60}Co	^{85}Sr	^{137}Cs
R (on sorption process)	980	130	3100
R'(on desorption process)	2800	160	6900

Table 6 Fraction percent of radionuclides extracted by each reagent

Extracting reagent	Percent (%)		
	^{60}Co	^{85}Sr	^{137}Cs
$CaCl_2$	8.0	99.0	2.8
KCl	3.6	1.0	11.4
$NH_2OH \cdot HCl$	20.7	0.0	1.1
K-oxalate	27.6	0.0	2.0
H_2O_2	20.3	0.0	0.5
Residue	19.8	0.0	82.2

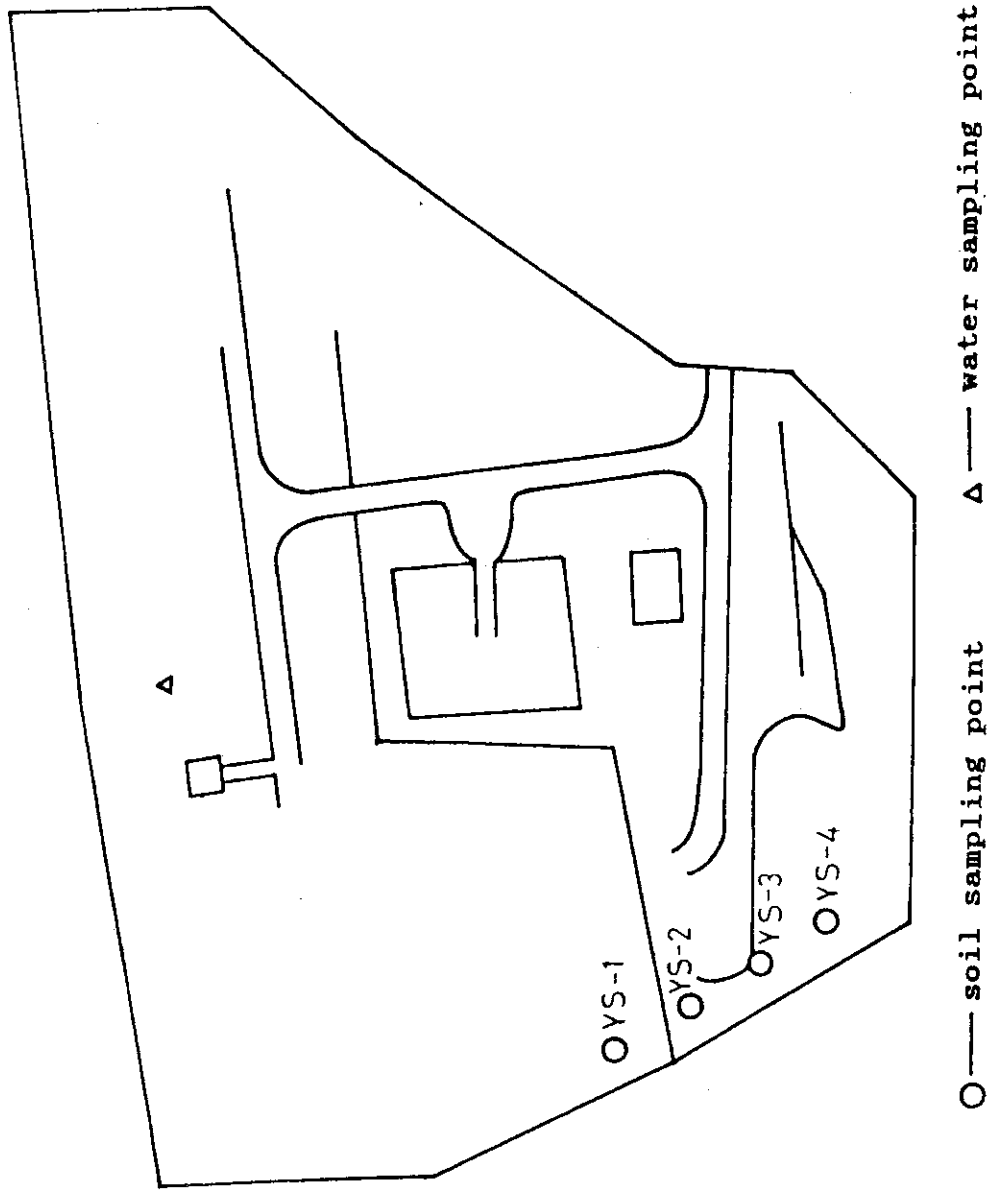


Fig.1 Sampling point of soil and ground water in field test site

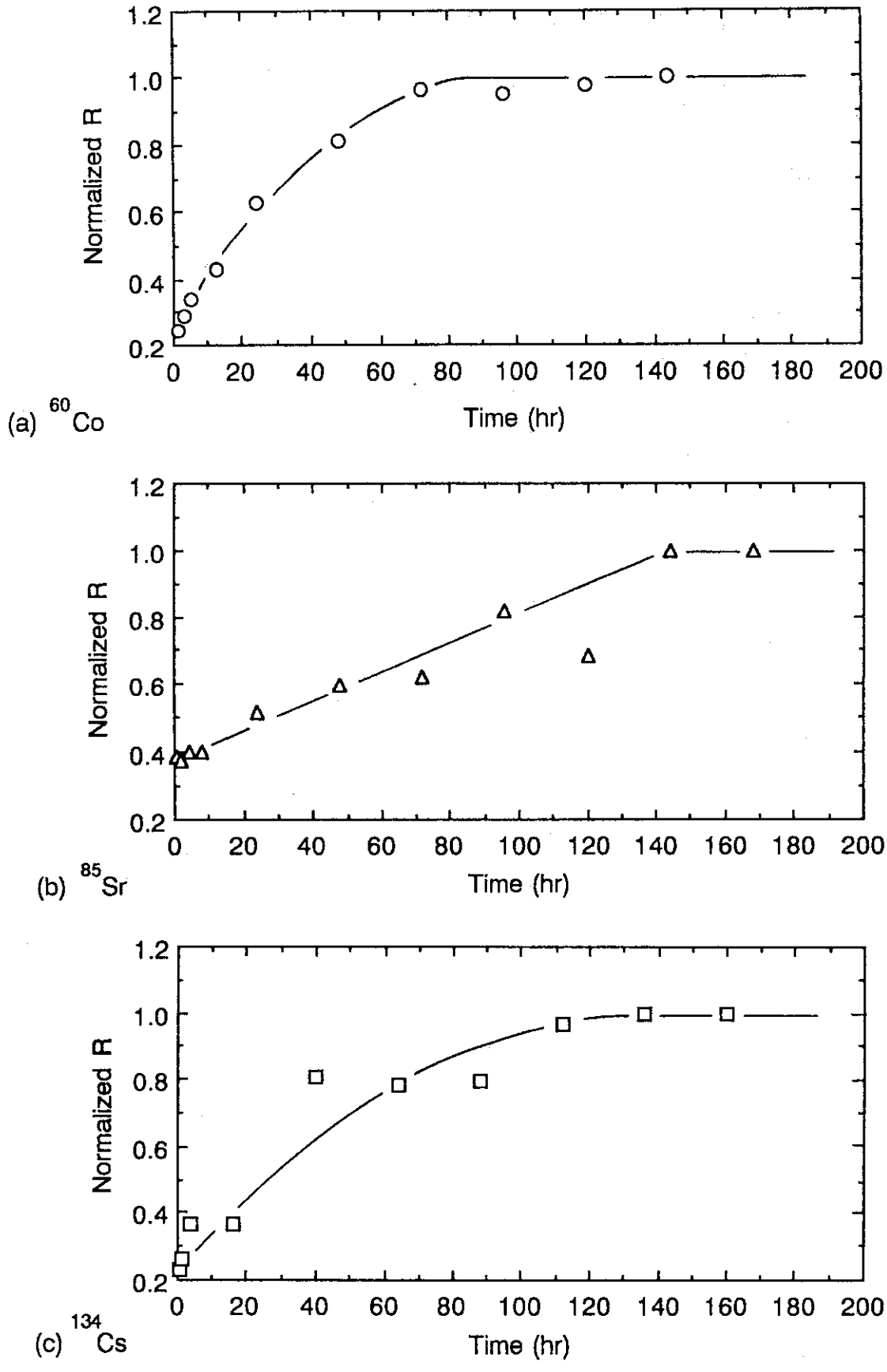
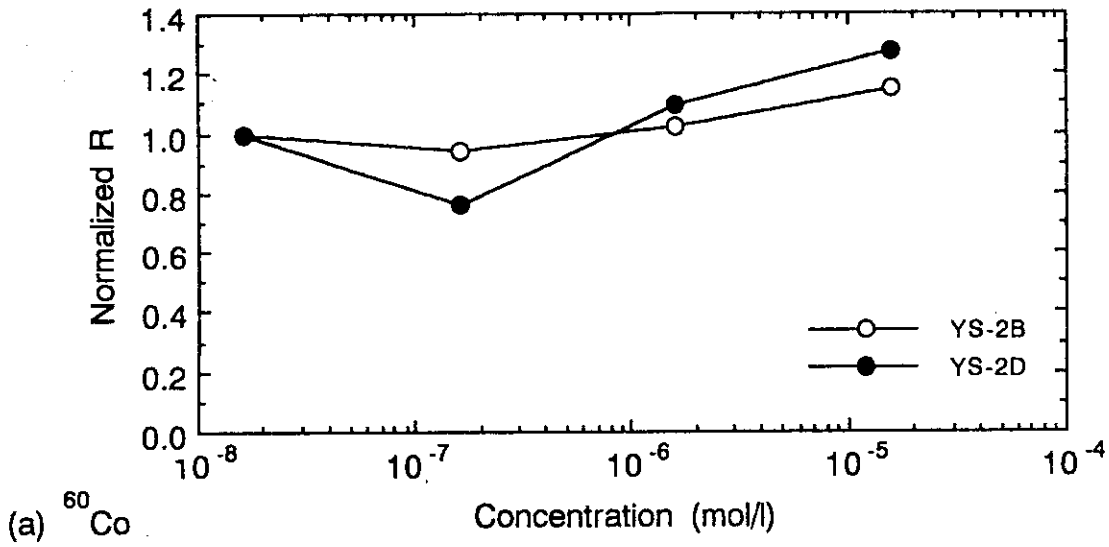
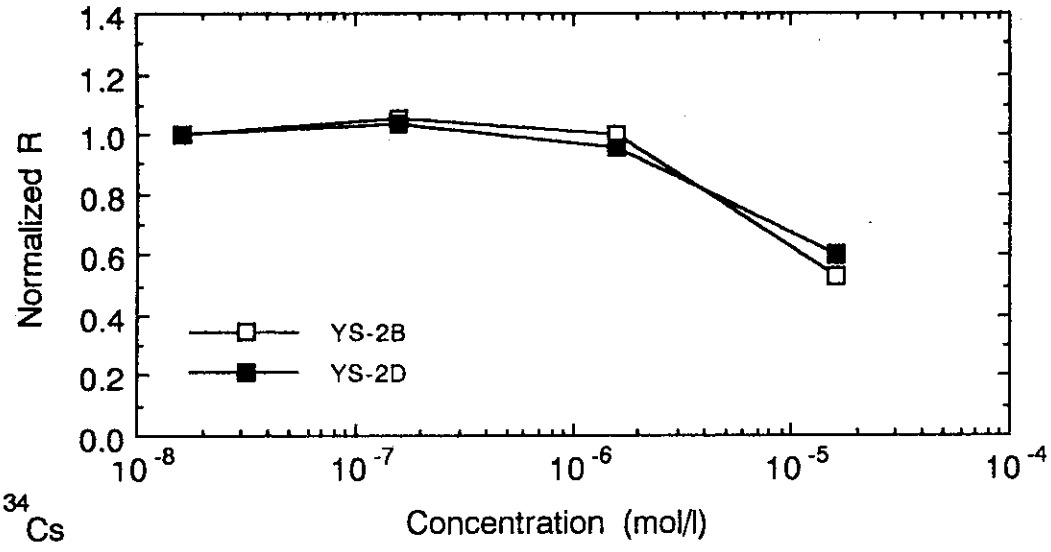


Fig.2 Effect of contact time on R



(a) ^{60}Co



(b) ^{134}Cs

Fig.3 Effect of concentration on R

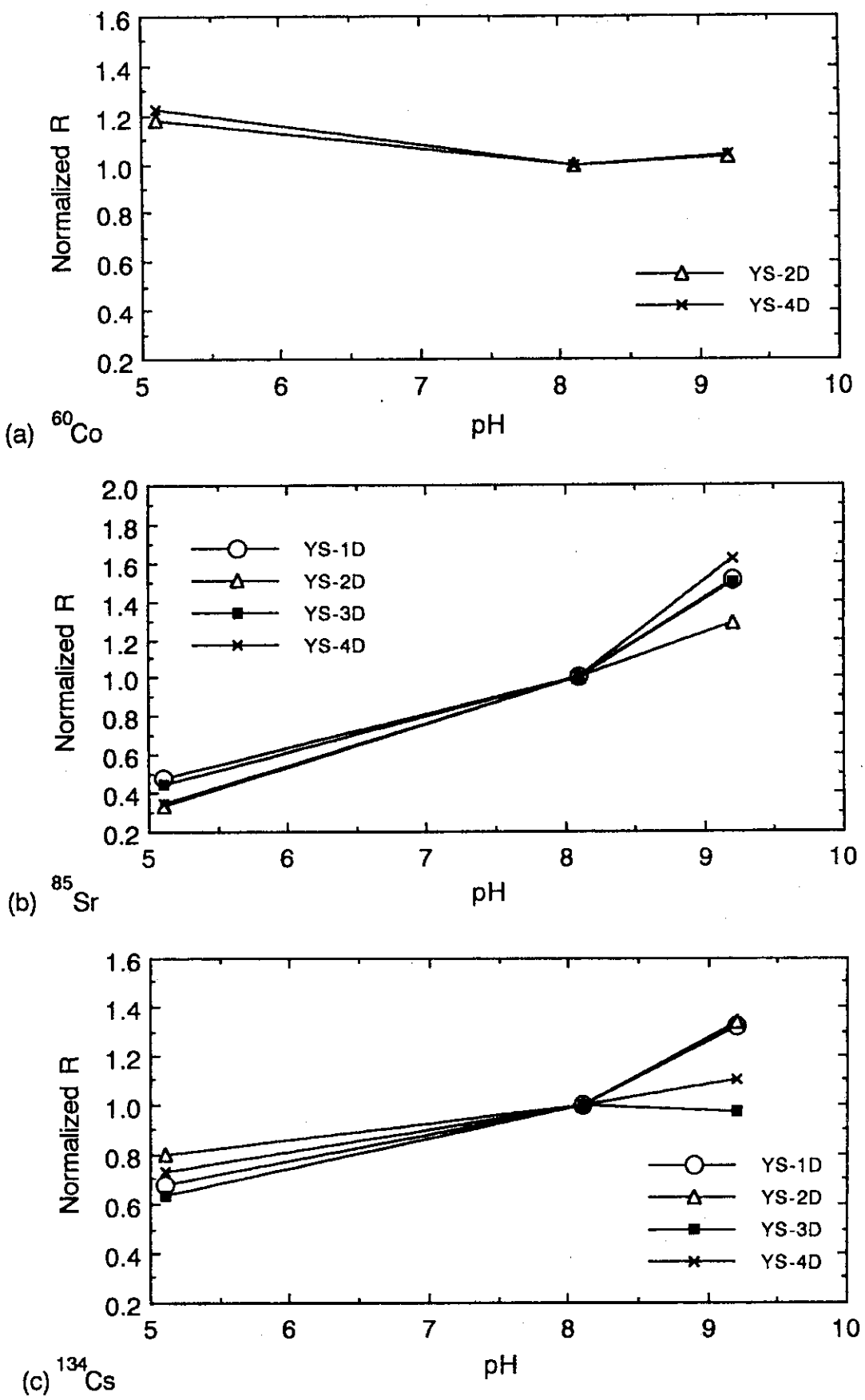


Fig.4 Effect of pH on R

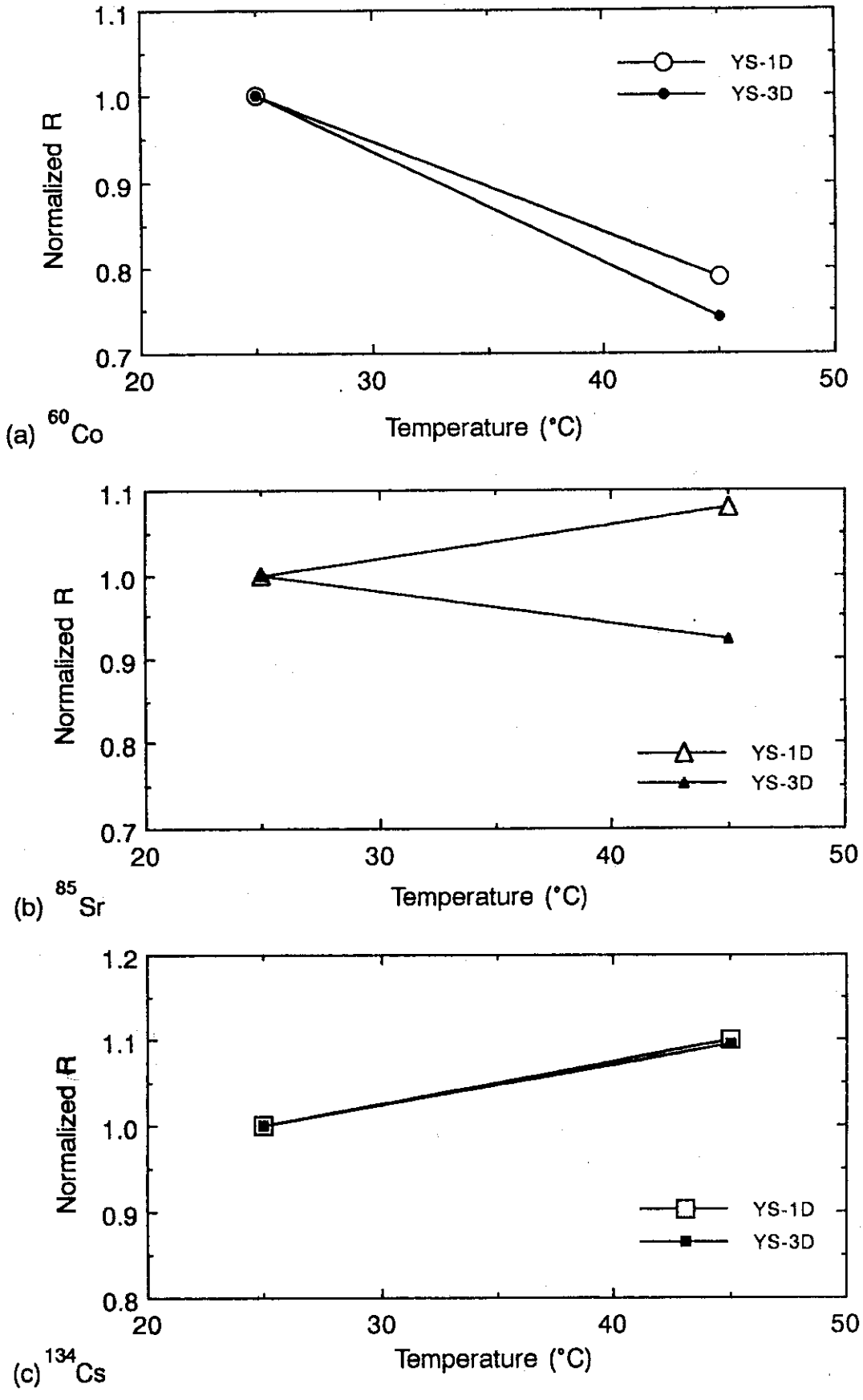


Fig.5 Effect of Temperature on R

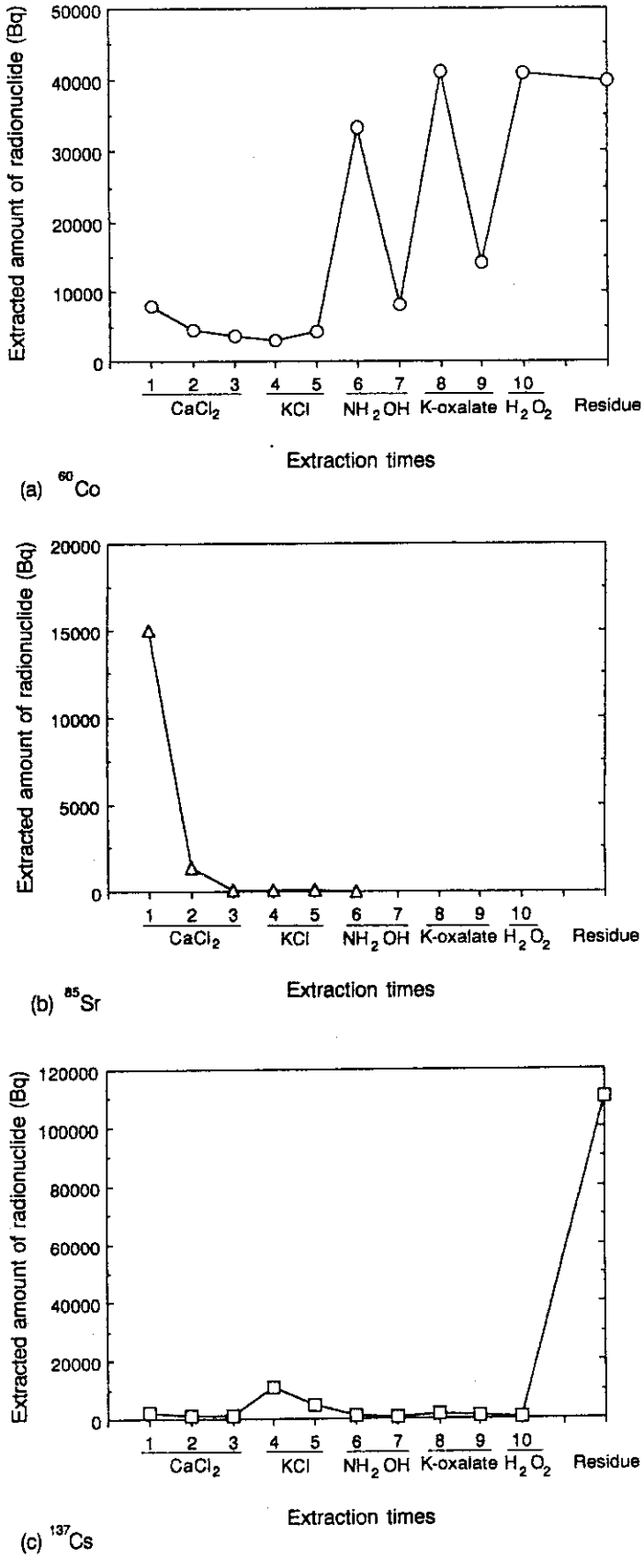


Fig. 6 Concentration of radionuclides desorbed by each extraction step