INS--1141
JP9608052

INS Report

INS-Rep.-1141
May 1996

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Abstract

The recoil properties in photonuclear (γ,n) reactions were measured by catcher foil method using natural copper and cesium chloride targets for the application to the transmutation of long-lived radioisotopes such as ^{137}Cs in high radioactive waste. The targets were prepared by a vacuum evaporation method. A stack consisting of target, catcher and beam monitor foils was placed in an evacuated chamber and irradiated with uncollimated electron-free bremsstrahlung. The recoil range was estimated from the radioactivities observed in the target and catcher foil, and the recoil energy calculated from a range-energy relation. As a result, it was found that the range caused by absorption of incident bremsstrahlung was one order of magnitude lower than the range caused by neutron emission from target nuclei in an excited state. This technique was applied to measurement of (γ,n) reaction cross section from radioisotope target of ^{137}Cs.

1. Introduction

Recently the handling of long-lived isotopes contained in high-level radioactive wastes (HLW) has become a serious problem because of their influence inflicted upon our environment over long period. The transmutation of these isotopes into short-lived and/or stable ones through nuclear reactions induced by high-energy particles or radiations is one of the promising ways to break this situation.

In this point of view, we have carried out the measurements of the transmutation rates of ^{137}Cs ($T_{1/2}$ = 30.17y), ^{90}Sr (29.1y) and transuranium elements among these long-lived isotopes in HLW into short-lived ones using photonuclear reactions with bremsstrahlung [1-3]. It is expected that (γ,n) reaction induced by giant dipole resonance process at energies of 10-30 MeV is very useful to transmute especially neutron rich nuclei ^{137}Cs and ^{90}Sr. Cross sections for stable isotopes in this energy region have already been measured by many groups and are summarized in the literature [4]. In general, reaction cross sections in this resonance region are more than 100mb, which is rather large in comparing with those for particle-induced reactions. In the transmutation by photonuclear reaction, production cross sections for (γ,n) and (γ,fission) reactions using radioisotope targets are indispensable as basic nuclear data. However, (γ,n) cross sections for ^{137}Cs and ^{90}Sr have never been measured. Since it is impossible to chemically separate the product from the target in (γ,n) reaction, it is very difficult to measure the product radioactivity in the existence of much amount of target radioactivity. Therefore, we have applied the nuclear recoil technique with catcher foil [5] to measure the cross section of (γ,n) reaction for ^{137}Cs target. By this recoil technique, we can separate the product from the target because some fractions of products are recoiled out by incident photons and neutrons emitted from (γ,n) reaction and collected in the catcher foil. Recoil yield is defined by the ratio of product radioactivity observed in the catcher to the total product one. From the obtained recoil yield, the (γ,n) cross section for radioactive ^{137}Cs target can be estimated in the assumption that recoil yield of the product ^{136}Cs from $^{137}Cs(\gamma,n)$ reaction is equal to that of ^{132}Cs from $^{133}Cs(\gamma,n)$. The detail of this estimation has been described in Ref.[2].

In this study, we first measured the recoil yield and range of the product ^{64}Cu in $^{65}Cu(\gamma,n)$ reaction using natural copper, and the reliability of this nuclear recoil method was confirmed. Then, natural cesium chloride targets were utilized to investigate the possibility of this technique for the application to the cross section measurements in the case of radioactive ^{137}Cs target.

2. Experimental procedure

2.1. Target preparation

Targets with various thicknesses up to about 100 $\mu g/cm^2$ were prepared by a vacuum evaporation method. Copper metal or cesium chloride mounted on a tantalum boat was evaporated on an aluminum foil by heating the boat in vacuum. The diameter of the evaporated area was adjusted to be 12.5 mm by considering the bremsstrahlung beam-size. Target thicknesses obtained are tabulated in Table 1. Target and catcher foil (0.1 mm thick aluminum) were stacked with a spacer ring (0.2 mm thick aluminum) placed inbetween. In order to measure forward and backward recoil ranges, catcher foil was set on downstream or on upstream for the beam direction as shown in Fig. 1.

2.2. Irradiation

The irradiation at the maximum bremsstrahlung energy (E_0) of 50 MeV was carried out by the 300 MeV electron linear accelerator at the Laboratory of Nuclear Science (LNS) of Tohoku University. The bremsstrahlung was obtained by the interaction of the electron beam with a 1 mm thick platinum converter placed on the beam axis. The average beam current measured by a core monitor placed in front of the converter was 70-80 μA. The electrons passed through the converter were bent down by the sweeping magnet. The stack containing beam monitor foil was put in the target chamber shown in Fig. 2. The chamber was evacuated and set at 60 cm downstream from the converter. Fig. 3 shows the schematic view of this experimental arrangement. Although we did not use a beam-collimator for the bremsstrahlung, the beam-size at the target position was measured to be about 10 mmΦ, smaller than the target-size. Therefore, the irradiation

was carried out with uncollimated electron-free bremsstrahlung. The typical irradiation time was 20 hr. The target chamber was cooled during irradiation by circulating water and air from a blower. The beam intensity was measured by the monitor reaction of $^{197}Au(\gamma,n)$ [4].

2.3. Chemical purification

In the recoil range calculation, the measurements of radioactivities produced in the target and recoiled into the catcher foil are required. The products of (γ,n) reactions for copper and cesium chloride, ^{64}Cu ($T_{1/2}$ = 12.7 hr) and ^{132}Cs (6.47 d), could not be measured quantitatively by non-destructive γ-ray countings of the catcher foil and the target, because of the low recoil yield and of interfering byproducts, e.g., ^{24}Na (15.02 hr) through $^{27}Al(\gamma,2pn)$ and $^{27}Al(n,\alpha)$ reactions. Therefore, the product isotopes in both catcher and target were chemically purified to remove these interfering byproducts. In the copper target, ^{62}Cu (9.74 m) was also produced through $^{63}Cu(\gamma,n)$ reaction besides ^{64}Cu through $^{65}Cu(\gamma,n)$. However, the yield measurement for ^{62}Cu was not performed because of its short half-life.

The copper targets with backing and the catcher foils were dissolved in aqua regia, and the copper was purified by an anion exchange with hydrochloric acid. The cesium chloride target and the catcher were also dissolved in aqua regia, and the cesium was purified by the precipitation of cesium tetra-phenyl borate. The schemes for these chemical purifications are shown in Figs. 4(a) and (b), respectively. The chemical recoveries in the purification process were measured to be 60-95% using atomic absorption spectrometry after the γ-ray counting. The chemical purification and the γ-ray measurement were performed at the Cyclotron and RadioIsotope Center (CYRIC) of Tohoku University.

2.4. γ-ray spectrometry

The γ-rays were measured with Ge detectors with an energy resolution of 1.9 keV full width at half maximum (FWHM) at 1332 keV, connected to multichannel pulse height analyzers. The detection efficiencies of the detectors were measured using the γ-ray reference source.

3. Analysis

3.1. Recoil range

Recoil range is estimated from the measurements of both radioactivities observed in the target and in the catcher foil. For simplification, we first consider the case that the product is recoiled out to the same direction as that of the incident beam. When the target thickness T is larger than the recoil range R of the product, the fraction of products recoiled from the target to the total products becomes constant. In another word, the product isotopes formed in the region from the surface to the depth R of the target are recoiled off the target, and the products produced in the deeper region than the recoil range, T-R, are stored in the target. From this consideration, the following equation is obtained,

$$A_T/A_C = (T-R)/R, \qquad (1)$$

or

$$R = \{A_C/(A_T+A_C)\}T, \qquad (2)$$

where A_T and A_C are radioactivities observed in the target and the catcher foil, respectively.

Secondly, we consider the isotropical recoil. If the target thickness equals to the recoil range, 1/2 of the total products are kept in the target and 1/4 are collected in forward or backward foil. Therefore, in the case of T>R, the recoil range of the product is defined to be

$$R = \{4A_C/(A_T+A_C)\}T. \qquad (3)$$

When the produced radioactivity in the unit thickness is k [Bq/(μg/cm^2)], the radioactivity A_C collected in the catcher foil is expressed as

$$k = (A_C+A_T)/T, \qquad (4)$$

or

$$A_C = kR/4. \qquad (5)$$

In photonuclear (γ,n) reaction, we presume the reaction process as following two steps; (i) target excitation with absorption of bremsstrahlung and the product recoils out to the same direction as that of the incident beam, and (ii) isotropical neutron emission from the target isotope at the excited state. When the recoil range by incident bremsstrahlung absorption is expressed as r, the radioactivity collected in the forward and backward catcher foils are given in the following equations,

$$A_C^f = A_C+k^f r = k^f(R/4+r), \qquad (6)$$

and

$$A_C^b = A_C-k^b r = k^b(R/4-r), \qquad (7)$$

where the superscripts f and b denote forward and backward, respectively. From these equations, the recoil ranges of r and R are obtained to be

$$R = 2(A_C^f/k^f + A_C^b/k^b), \qquad (8)$$

and

$$r = 1/2(A_C^f/k^f - A_C^b/k^b). \qquad (9)$$

3.2. Bremsstrahlung intensity

The saturated radioactivity A of the product is estimated by the following equation,

$$A = NC\int\sigma(E)\Phi(E)dE, \qquad (10)$$

where *C is number of incident electrons (e/s), N number of target atoms,* σ(E) reaction cross section (cm^2) and Φ(E) beam intensity of bremsstrahlung (cm^{-2}·MeV^{-1}·e^{-1}). The Φ(E) was calculated by the EGS-4 code [5] at E_0 = 50 MeV, and C was obtained using ^{197}Au(γ,n) monitor reaction. The detail of this calculation was already reported in Ref.[1].

4. Results and Discussion

The results of the saturated radioactivities observed in the targets and the catcher foils are indicated in Table 2. In the γ-ray countings, the determination of ^{64}Cu produced in the copper target and the catcher foil was carried out by the measurement of 511 keV γ-ray from positron annihilation instead of the 1346 keV γ-ray, because of its low γ-ray abundance. Annihilation γ-rays were also emitted from the byproducts of ^{61}Cu ($T_{1/2}$ = 3.35h) and ^{62}Cu (9.74m). We performed the chemical purification after several hours from the end of irradiation, and the produced ^{62}Cu already decayed out at that time due to the short half-life. Therefore, the yield of ^{64}Cu was estimated by the decay curve analysis of the 511 keV and the subtraction of the contribution from ^{61}Cu. The produced ^{132}Cs was determined by the measurement of its 668 keV γ-ray. The recoil yields obtained in the cesium target are shown as a function of target thickness in Fig. 5. The solid curve and the filled squares in Fig. 5 show the values theoretically calculated in the method described in the section 3.1 and experimental results, respectively. The experimental recoil yields agreed with the calculated ones in the target thickness more than 50 μg/cm^2, but were smaller than the calculated ones with the decrease of the target thickness. The similar result was also obtained for the copper target. One of the main causes for this difference in thinner targets than 50 μg/cm^2 seems to be non-uniformity of the target thickness prepared by vacuum evaporation.

The experimental recoil range was calculated using radioactivities observed in target and catcher foil. In this calculation, we adopted the data from the target with the thickness more than 60 μg/cm^2. As average values for these results, the followings were obtained,

$R = 17.4 \pm 1.9$ μg/cm^2 and $r = 1.70 \pm 0.22$ μg/cm^2 for Cu, and

$R = 6.27 \pm 0.70$ μg/cm^2 and $r = 0.623 \pm 0.081$ μg/cm^2 for CsCl.

The associated experimental uncertainties refer to those from counting statistics, detector efficiencies and chemical yields, which do not include the ambiguity in the target thickness estimation.

The recoil energies were estimated using the range-energy relation by Northcliffe and Schilling [7]. In the case of Cu, the recoil energies

were estimated from the interpolation of the range data of Cu ions in Si, and obtained to be

$$E_R = 68.8 \pm 7.6 \text{ keV} \quad \text{and} \quad E_r = 6.74 \pm 0.88 \text{ keV}.$$

In the case of Cs ions in CsCl, we first estimated the relative stopping powers of Cs ions in Cs and in Cl to the stopping power of Cs ions in Al according to Ref. [7], and obtained the stopping power of Cs in CsCl. The obtained stopping power was almost identical to that of Cs ions in Ag reported in Ref. [7]. Therefore, in the assumption that the stopping power of Cs in CsCl was approximately equal to that in Ag on the average, we estimated the recoil energy by interpolating the Cs range in Ag. The obtained energies were as follows,

$$E_R = 37.2 \pm 4.2 \text{ keV} \quad \text{and} \quad E_r = 3.70 \pm 0.48 \text{ keV}.$$

From the results for recoil energy E_r, the average energies of the incident photons were estimated to be 25.0 ± 3.3 MeV for Cu and 30.3 ± 3.9 MeV for CsCl, which agreed well with each other within the experimental uncertainty.

The experimentally obtained relation between recoil range and kinetic energy E was reported by Porll [8] for gallium ions in copper in the energy range up to 1.0 MeV as follows,

$$R \text{ [μg/cm}^2\text{]} = 0.193E \text{ [keV]}.$$

If this equation was approximately applied to the case of copper ions in copper, the recoil energies were estimated to be

$$E_R = 90.3 \pm 9.9 \text{ keV} \quad \text{and} \quad E_r = 8.81 \pm 1.14 \text{ keV}.$$

Although these recoil energies are somewhat higher than those estimated by the method according to Ref. [7], the consistency between both the results seems to be fairly good in considering the approximations in the calculation of the recoil energy.

5. Conclusion

We have measured the recoil range in photonuclear (γ,n) reaction using natural copper and cesium chloride target. From this result, it is concluded that the recoil range in the first step of target excitation with absorption of bremsstrahlung is one order of magnitude less than that in the second step of the isotropical neutron emission from the excited target isotopes. This nuclear recoil method can also be applied to the measurement of very weak radioactivity produced in the highly radioactive target.

Acknowledgements

The authors are indebted to Dr. K. Masumoto of the Laboratory of Nuclear Science (LNS) of Tohoku University for his kind arrangement in the irradiation experiment, and to the machine group of LNS for the accelerator operation.

This work is financially supported by the Power Reactor and Nuclear Fuel Development Corporation.

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Figure Captions

Fig. 1(a). Example of target stack.
(b). Stack holder.
Fig. 2. Target chamber for irradiation in vacuum.
Fig. 3. Schematic view of experimental arrangement for irradiation at LNS.
Fig. 4(a). Flow chart of chemical purification for copper.
(b). Flow chart of chemical purification for cesium.
Fig. 5. Relation between recoil yield, $A_C/(A_C+A_T)$, and target thickness in cesium chloride. Solid line shows calculated yield, and filled square experimental yield.

Table 1. Target thickness (ug/cm^2)

Target	Thickness
Cu1	106
Cu2	111
Cu3	95
Cu4	89
Cs1	70
Cs2	68
Cs3	60
Cs4	61

Table 2. Saturated radioactivity (Bq) observed in target and catcher foil

Target/ Catcher	^{64}Cu	Chemical Yield	Target/ Catcher	^{132}Cs	Chemical Yield
Cu1-T	12378 ± 50	0.908	Cs1-T	28848 ± 98	0.882
F	684 ± 13	0.840	F	800 ± 14	0.777
Cu2-T	13960 ± 53	0.897	Cs2-T	20605 ± 72	0.583
B	329 ± 9	0.852	B	394 ± 10	0.771
Cu3-T	10078 ± 50	0.837	Cs3-T	23828 ± 77	0.865
F	749 ± 14	0.933	F	869 ± 15	0.811
Cu4-T	12440 ± 56	0.944	Cs4-T	24786 ± 91	0.898
B	332 ± 9	0.846	B	315 ± 10	0.745

T: Target, F: Forward Catcher Foil, and B: Backward Catcher Foil.

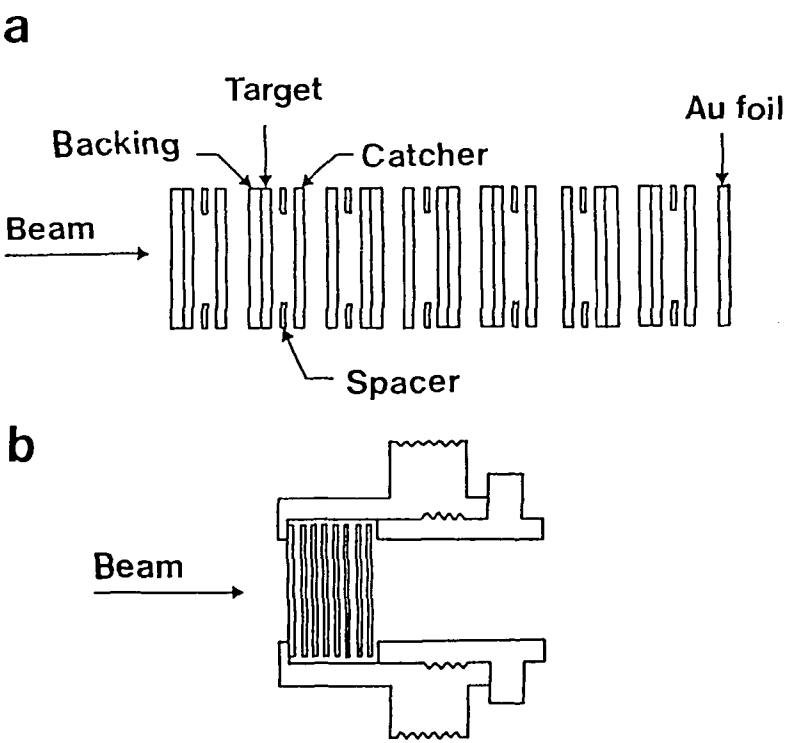

Fig. 1

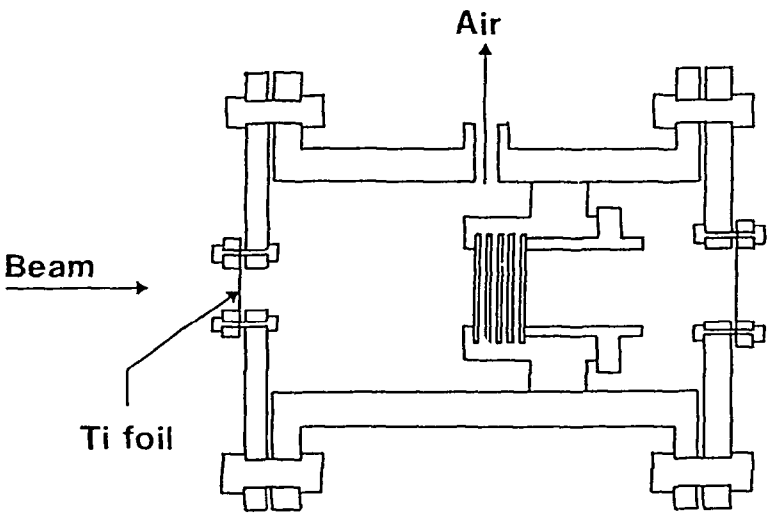
Air
Beam
Ti foil

Fig. 2

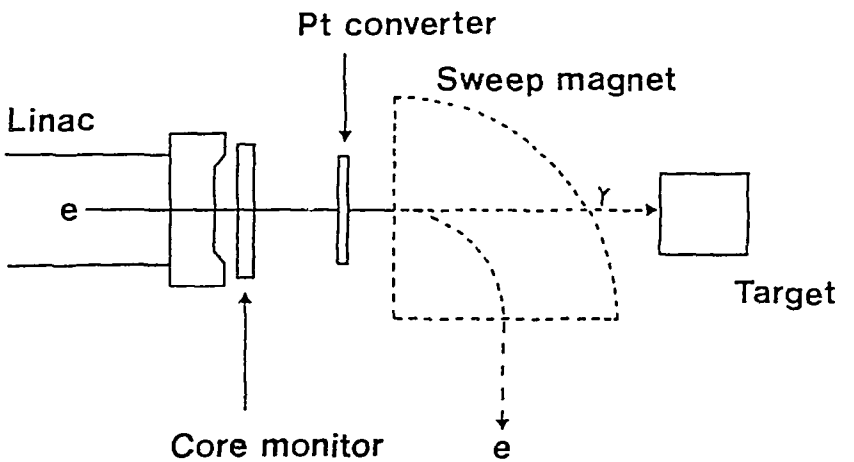
Pt converter
Sweep magnet
Linac
e
γ
Target
Core monitor
e

Fig. 3

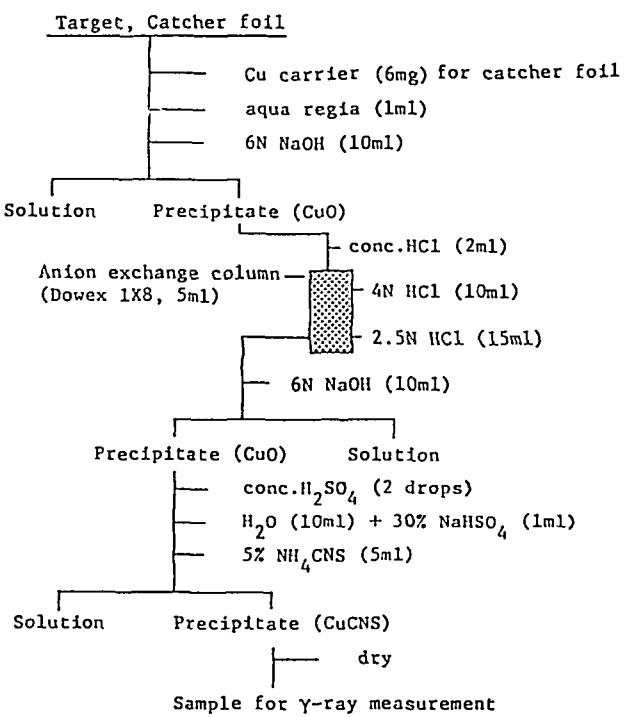
Target, Catcher foil
Cu carrier (6mg) for catcher foil
aqua regia (1ml)
6N NaOH (10ml)
Solution
Precipitate (CuO)
conc.HCl (2ml)
Anion exchange column
(Dowex 1X8, 5ml)
4N HCl (10ml)
2.5N HCl (15ml)
6N NaOH (10ml)
Precipitate (CuO)
Solution
conc.H_2SO_4 (2 drops)
H_2O (10ml) + 30% $NaHSO_4$ (1ml)
5% NH_4CNS (5ml)
Solution
Precipitate (CuCNS)
dry
Sample for γ-ray measurement

Fig. 4 a

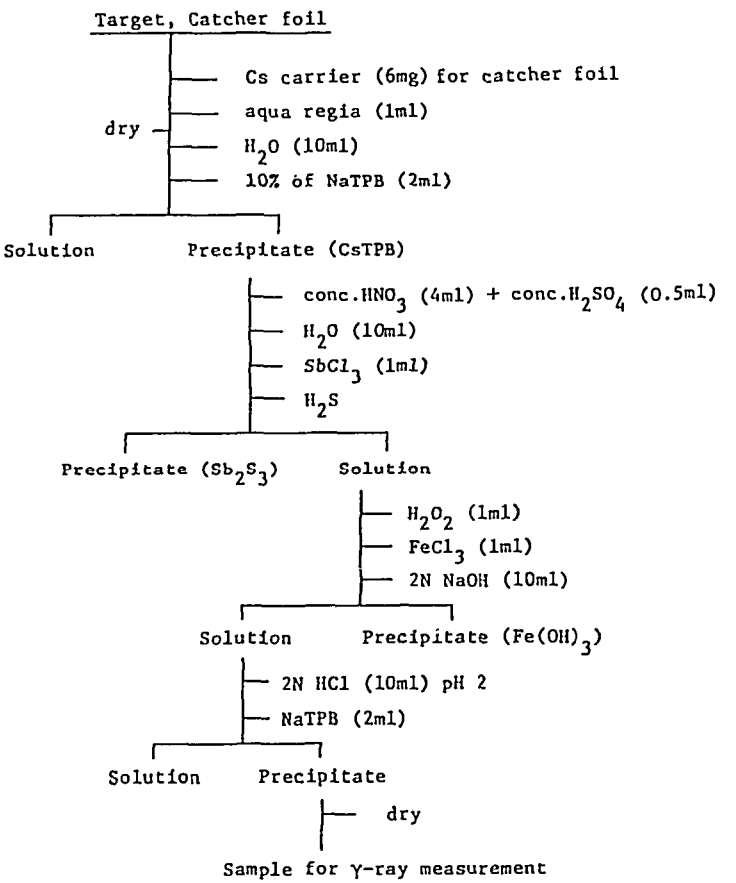
Target, Catcher foil
Cs carrier (6mg) for catcher foil
dry
aqua regia (1ml)
H_2O (10ml)
10% of NaTPB (2ml)
Solution
Precipitate (CsTPB)
conc.HNO_3 (4ml) + conc.H_2SO_4 (0.5ml)
H_2O (10ml)
$SbCl_3$ (1ml)
H_2S
Precipitate (Sb_2S_3)
Solution
H_2O_2 (1ml)
$FeCl_3$ (1ml)
2N NaOH (10ml)
Solution
Precipitate ($Fe(OH)_3$)
2N HCl (10ml) pH 2
NaTPB (2ml)
Solution
Precipitate
dry
Sample for γ-ray measurement

Fig. 4 b

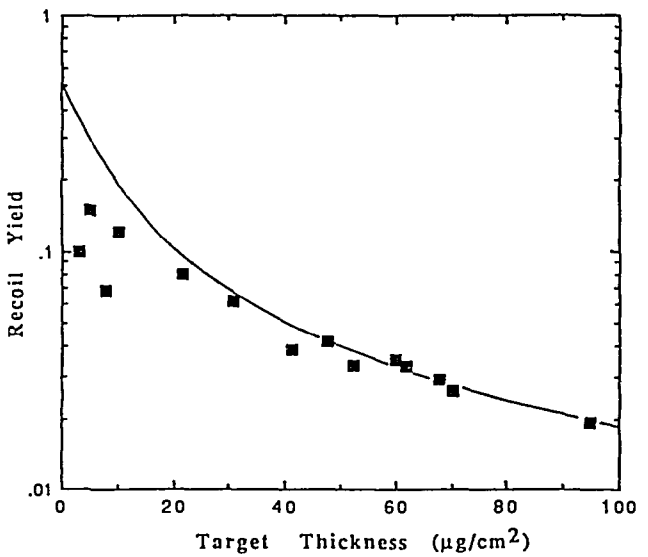

Fig. 5