Analysis of Tritium in Ti-^{3}H Thin Film Target by ERD-TOF and (d, α) Reaction

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Tritium in Ti-^{3}H thin film, which has been available for the target of mono energetic neutron source, was analyzed by ERD-TOF and (d, α) reaction. The areal density of ^{3}H up to the maximum accessible depth of 3770 Å was obtained to be 2.14±0.02 x10^{18} atoms/cm^2 by ERD-TOF. And the areal density of tritium of Ti whole depth was found to be 4.48±0.27 x 10^{18} atoms/cm^2 and the combination ratio of ^{3}H to Ti was estimated to be 1.33 by (d, α) reaction, respectively. The analytical result of tritium by (d, α) reaction is in good agreement with the given value by target manufacturer within 7 %. Assuming that the depth distribution of tritium is homogeneous in whole Ti layer, the result by (d, α) reaction is in agreement with that by ERD-TOF within 19 %.

INTRODUCTION

^{3}H(p,n)^{3}He and ^{3}H(d,n)^{4}He reactions are important in studying mono energetic neutron source due to their large cross sections [1]. Tritium gas target has been generally used as the target of these reactions. However, there is a disadvantage of thin entrance. This causes a small energy loss of the incident particles as well as the difficulties of treatment and transport. However, after the characteristics of the homogeneous impregnation of tritium in metal were identified [2], Ti-^{3}H thin film has been treated as neutron target more than tritium gas target.

Korea Institute of Geoscience and Mineral Resource (KIGAM) developed the ERD-TOF system using about 10 MeV Cl beam [3]. This method is well known to be an optimum method to analyze quantities of light elements in metal thin films. An accessible depth for this system was estimated to be about 3000Å for the ^{10}B, ^{11}B, C, N, and O in BN(C,O)/Si film with a depth resolution of about 60 Å. Also it was estimated that quantities as small as 10^{13} atoms/cm^2 could be measured without difficulty. However, the drawback of this method is the short penetration depth of incident Cl particle in spite of having a high energy, because of strong stopping power of Cl for backing material of thin film. However, this weakness can be overcome by (d, α) reaction. Especially the uncertainty of differential cross section for this reaction is estimated to be within 1.5 % for E_d < 400 keV, and increases to 4 % at high energies [4]. In this work, tritium quantity in Ti-^{3}H thin film is estimated by both ERD-TOF and (d, α) reaction.

EXPERIMENTAL

The thin film target used in this experiment was provided by the CEA Company in France 4 years ago. This company reported that the total areal densities of Ti and ^{3}H were about 280 μg/cm^2 and 5.32x10^{18} atoms/cm^2, respectively. Tritium quantity was measured by a calorimeter. The substrate of this target was Cu with a thickness of 1.2 mm.

The deuteron and Cl beams used in this experiment were extracted from the source of negative ions by cesium sputtering and were accelerated by 1.7 MV tandem pelletron accelerator. The beam energy calibration (<± 1keV) was done by

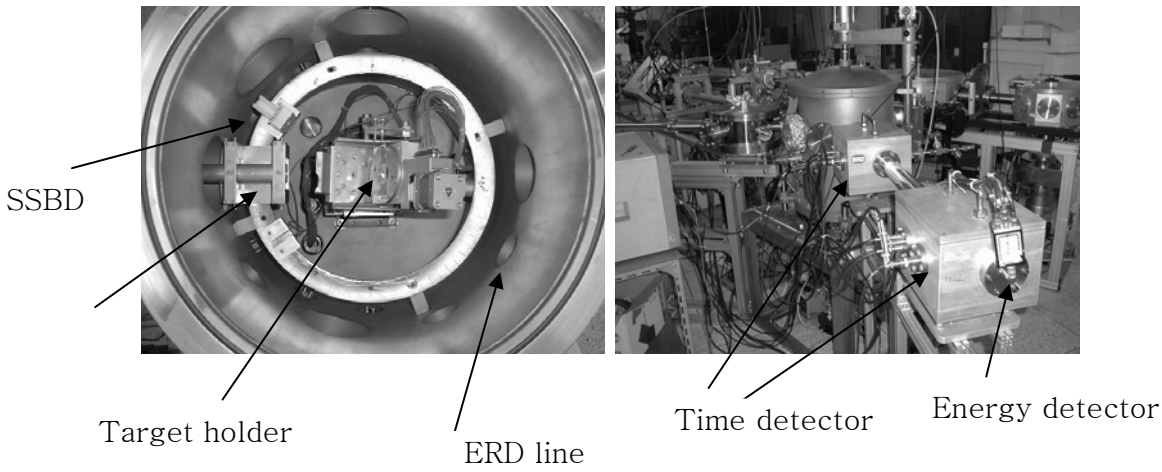

Fig. 1. ERD-TOF chamber and its detecting system.

using resonance reactions at 991±1 keV in ^{27}Al(p, γ) and at 3,047 ±1 keV in ^{16}O(α,α').

The detector for alpha particles emitted from (d, α) reaction was a silicon surface barrier detector (SSBD) , and was positioned at 140^{o} from incident deuteron direction with the solid angle of 0.1723 msr. The incident deuteron energy and the beam current of about 8 nA were determined by considering its penetration depth in Ti and target temperature. Ti-^{3}H thin film target had not a tilting angle in (d, α) reaction.

ERD-TOF could identify all the recoil particles by measuring their energies and velocities simultaneously. Their energies and velocities were determined by using the SSBD and TOF system, respectively. Two time detectors, which consisted of C foils with thickness of 5 μg/cm^{2} and micro-channel plate, were used in TOF system. The distance between the two time detectors was 0.745 m. The efficiency of the TOF spectrometer is defined as the ratio of the counts detected by the two time detectors to those of triple coincidence detected by the SSBD in coincidence with the two time detectors. The efficiency of this TOF system for proton with 1,000 keV was found to be 25 % and those of heavier elements than lithium with energy ranges from 2,000 keV to 5,000 keV were found to be better than 95 % by backscattering for ^{197}Au.

The target tilting angle in ERD-TOF experiment was 75^{o} and scattering angle for recoil particles was 30^{o} with respect to incident Cl beam with 9,628 keV. Figure 1 shows the ERD-TOF chamber and its detecting system.

RESULTS AND DISCUSSION

Two dimensional plot of recoil particles that were elastically scattered from a Ti-^{3}H target by Cl beams was shown in Figure 2. The abscissa denotes the recoil energy of the recoil particle and the ordinate means the flight time of the distance between two time detectors. Because mass resolution of ERD-TOF system proved to be about 1 amu for the atomic mass numbers below 50 [3, 5], Hydrogen, Tritium, ^{3}He, Oxygen and small amount of Nitrogen can be easily separated. Oxygen and Nitrogen seem to be included in a fabrication process. Observations of ^{3}He and ^{1}H may be remarkable. Ti-^{3}H target was fabricated a few years ago, and ^{3}He is a decay product of ^{3}H, and ^{1}H is normal surface contaminant. Although ^{3}H and ^{3}He have almost same mass, ^{3}He can be separated by differences of their stopping powers through carbon foil in time detector and dead layer of SSBD.

The time resolution of ERD-TOF time detector was better than energy resolution of SSBD in a few MeV range [3,5]. So, the time spectrum should be converted into an energy spectrum for the accurate analysis. The efficiencies of ERD-TOF system for tritium and ^{3}He were assumed to be 25 % and 30 %, respectively, by considering stopping power ratios of

Table 1. Areal densities of recoil particles by ERD-TOF ; Accessible depth was calculated assuming no variation of Ti density in the penetration of ^{3}H and ^{16}O into Ti.

	^{1}H (x 10^{15} atom/cm^{2})	^{3}H (x 10^{15} atom/cm^{2})	^{3}He (x 10^{15} atom/cm^{2})	^{16}O (x 10^{15} atom/cm^{2})
1 run	55.0	2124.0	330.9	403.1
2 run	59.4	2149.1	284.8	341.7
3 run	67.5	2161.2	315.7	381.8
total	60.6±6.3	2144.7±19.0	310.5±23.5	375.5±31.1
Accessible depth (Å)	106.6	3772	546.3	660.7

Table 2. ^{3}He energy for the incident deuteron energy at scattering angle of 140^{o} for ^{3}H(d, α)n reaction

Energy of deuteron [MeV]	Scattering angle of n [degree]	Scattered energy of n [MeV]	Scattering angle of αα [degree]	Scattered energy of αα [MeV]
0.200	33.90	14.3419	139.94	2.714
0.250	33.14	14.4681	140.00	2.638
0.300	32.51	14.5854	140.00	2.571
0.350	31.94	14.6966	140.00	2.509
0.400	31.42	14.8028	140.00	2.453
0.450	30.92	14.9053	140.00	2.401
0.500	30.45	15.0044	140.00	2.352

Table 3. Areal density of ^{3}H on Ti-^{3}H thin film by (d,α) reaction.

Energy of deuteron [keV]	Run number	Areal density [x10^{18} atoms/cm^{2}]	Average areal density [x10^{18} atoms/cm^{2}]	Differential cross section [4] [mb/sr]
350	1	4.82	4.53±0.30	84.90
	2	4.23		
	3	4.55		
550	1	4.31	4.31	51.33
Average areal density [x10^{18} atoms/cm^{2}]			4.48±0.27	

^{3}H and ^{3}He relative to that of ^{1}H for a carbon foil of time detector and the measured efficiency of ^{1}H for a carbon foil. Its efficiency for ^{16}O was measured to be 100 %.

Analysis of the converted energy spectra was performed by the simulation and evaluation code, SENRAS [6]. Figure 3 shows comparison of recoil spectra measured by ERD-TOF system with that simulated by SENRAS. The solid lines denote simulation results and symbols mean experimental data. Table 1 denotes areal densities during 3 runs corresponding to accessible depth of Ti, which were obtained assuming no variation of Ti density by the penetrations of ^{3}H and ^{16}O into Ti. By the depth profiling with ERD-TOF, ^{1}H is observed only on the near surface of Ti. By contrast, ^{3}H is found to be spread out all over Ti layer. However, since the accessible depth by ERD-TOF is about 3770 Å, the entire Ti layer can't be analyzed. To overcome this problem, the analysis by (d, α) reactions are performed on the relatively thick Ti-^{3}H target.

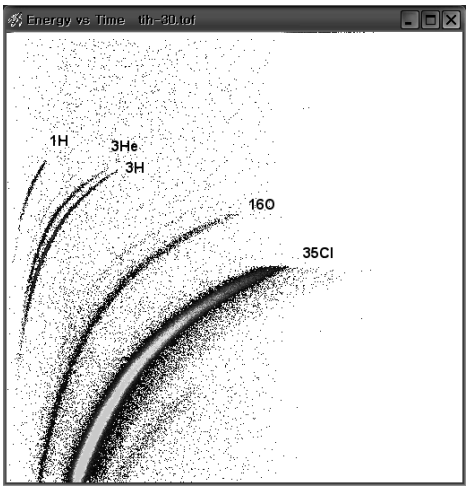

Fig. 2. Two dimensional spectrum of recoil particles elastically scattered from a Ti^{3}H target by Cl beams ; ^{1}H, ^{3}H, ^{3}He, ^{16}O, and ^{35}Cl.

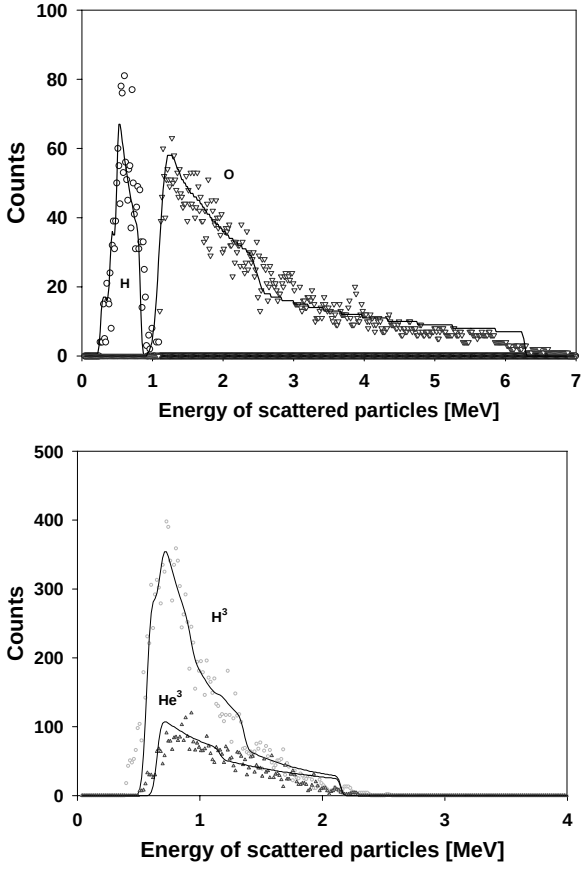

Fig. 3. Comparison of recoilarticle spectra measured by ERDTOF system (Symbol data) with that simulated by SENRAS (Solid line).

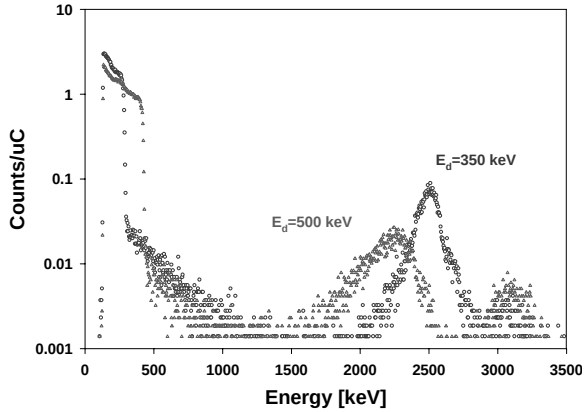

Fig. 4. Alpha spectra for ^{3}H(d, α) reaction at deuteron energies of 350 keV (○) and 500 keV(□).

The ^{3}He energy corresponding to the incident deuteron energy at the scattering angle of 140^{o} can be calculated by kinematics [7] and is shown in table 2. Also the evaluated total cross section and center of mass Legendre coefficients for ^{3}H(d,α,) reaction were taken from Ref. 4 ; the uncertainty was estimated to be 1.5% for E_d < 400 keV, and increases to 4 % at higher energies. And the α particle yield for this reaction was tested in energy range of 250 keV < E_d < 750 keV, and a good agreement of α yield with evaluated cross sections was found in Ref. 8.

Figure 4 shows spectrum of ^{3}H(d, α) reaction for deuteron energies of 350 keV and 500 keV. It is shown that peak widths for deuterons with the energy of 500 keV and 350 keV are different. It may be caused by that the energy broadening of α particles produced by 500 keV deuteron is larger than that by 350 keV deuteron. Since the penetration depth of 500 keV deuteron in Ti (3.78μm) is larger than that of 350 keV deuteron (2.19μm). A ghost peak above 3,000 keV is confirmed to be a internal conversion peak through blocking the scattered particles by putting the Al foil with thickness of 17μm before SSBD. Table 3 shows the areal densities of ^{3}H, it is obtained during 3 runs. Areal density of tritium is found to be 4.48±0.27 x10^{18} atoms/cm^{2}, and considering natural decay of tritium, it is in agreement with the given information of target manufacture within 7 %. In the measurement of ^{3}H areal density, charge by β^{-} decay of tritium was measured to correct the accumulated target charge.

The Ti width by this reaction is calculated to be 6,658 Å (alpha energy loss : 182 keV) using FWHM of the α peaks for deuteron energy of 350 keV and Ziegler's stopping power of α for Ti. So current combination ratio of ^{3}H to Ti is estimated to be 1.33 by comparing of the Ti width by (d, α) reaction with the areal density of tritium in same thickness.

CONCLUSIONS

The depth profiling up to about 3770 Å of Ti-^{3}H target was performed by ERD-TOF and total areal density of tritium on Ti-^{3}H target was measured by ^{3}H(d, α) reaction. It is found that tritium is spread out over all Ti layer by ERD-TOF. Total quantity of tritium on Ti-^{3}H thin film target is estimated to be 4.48±0.27 x10^{18} atoms/cm^2. The current combination ratio of ^{3}H to Ti ratio is determined to be 1.33 by comparing of the resolved Ti width by (d α,) reaction with the areal density of tritium in same thickness. Two thin film analysis methods, ERD-TOF and ^{3}H(d, α) reaction, are very useful for light element analysis than general methods such as RBS and other's nuclear reaction. The standard deviation of areal density for tritium by ERD-TOF is found to be smaller than 1 %. However, those of others recoil particles are determined to be about 10 %. And for ^{3}H(d, α) reaction, the standard deviation is estimated to be smaller than 6 %. And the analysis results of tritium by (d,α,) reaction is in good agreement with the given value by target manufacture within 7 %. Assuming that the distribution of tritium is homogeneous on Ti layer, the analysis result of (d, α) reaction is in agreement with that of ERD-TOF within 19 %.

REFERENCES

[1] C.R.Brune, K.I.Hahn, R.W. Kavanagh and P.R.Wrean, Phys. Rev. **C 60**, 15801 (1999).

[2] C.R.Brune, R.W. Kavanagh and P.R.Wrean, Nucl. Instr. and Meth. **A 343**, 415 (1994).

[3] G.D.Kim, J.K.Kim Y.S.Kim, H.W.Choi, S.Y.Cho, H.J.Woo, and C.N.Whang, Jour. of the Korean Phys. Soc. **32**, 739 (1998).

[4] M.Drosg and O.Schwerer, in Handbook on Nuclear Activation Data (International Atomic Energy Agency, Vienna, 1987) p 87.

[5] J.K.Kim Y.S.Kim,G.D.Kim, H.W.Choi, H.J.Woo, S.Y.Cho, and C.N.Whang, Nucl. Instr. and Meth. in Phys. Res. **B 140**, 380 (1994).

[6] G. Vizkelethy, Nucl. Instr. and Meth. in Phys. Res. **B 45**, 1 (1994).

[7] J.L.Fowler and J.E. Brolley, Rev. Mod. Phys. **28**, 103 (1956).

[8] C.R. Brune, R.W.Kavanagh, Nucl. Instr. and Meth. in Phys. Res. **A 343**, 415 (1994).