

Detection of Neutron and Tritium during Electrolysis of D₂SO₄ - D₂O Solution

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ABSTRACT

During the electrolysis of 0.5 M D₂SO₄-D₂O solution using Pd as a cathode material and Pt as an anode material, the emission of neutron was detected by means of the fission track method and the production of tritium was investigated with the liquid scintillation method.

The neutron emission rate was estimated to be comparable with natural abundance of neutrons at the surface of the earth which was 2 neutrons cm⁻² s⁻¹ estimated by fission track method comparing with the neutron radiation from the ²⁴¹Am-⁹Be neutron source. The tritium production rate was estimated to be 10⁴ T atoms cm⁻² s⁻¹ in the Pd metal used as the cathode materials by the liquid scintillation method. The branching ratio (T/n) was estimated to be 10⁴.

1. INTRODUCTION

Branching ratio of tritium production rate to neutron emission rate in the d-d nuclear reaction was estimated to be 10⁸ in the discharge of D₂ gas prepared using plasma focus device⁴⁾ and this was reported to be 10⁴ in the electrolysis of LiOD-D₂O solution³⁾. We have measured the neutron emission rate and tritium production rate using fission track method and liquid scintillation method, respectively, during the electrolysis of D₂SO₄-D₂O solution²⁾. In this case, it is necessary to obtain the correlation between the fission track number and the absolute neutron emission rate. The relation between the neutron fluence and fission track density was estimated using ²⁴¹Am-⁹Be neutron source to determine the absolute neutron emission rate.

In the present paper, some results obtained during the electrolysis are reported and the branching ratio (T/n) is briefly discussed.

2. EXPERIMENTAL

The electrolysis of 0.5 M D₂SO₄-D₂O solution was carried out for 5 h using Pd as a cathode material and Pt as an anode material. During the electrolysis, the emission of neutron was detected by means of fission track method. After the electrolysis, the β -ray radiation from the electrolyte and Pd used as the cathode material was measured by liquid scintillation method using scintillation counter (TRI-CARB 2500TR). The experimental procedures were determined in detail in the previous paper²).

3. RESULTS

In the neutron detection during the electrolysis of 0.5 M D₂SO₄-D₂O solution and 0.5 M H₂SO₄-H₂O solution using Pd as the cathode material, the fission track method was applied in order to investigate the effect of the electrolyte solution on the neutron emission. The results obtained are given in Table 1.

Table 1 Fission Track Density Observed in Fission Track Detectors in the Electrolysis

Electrolyte	Detector	Track density ^{a)} (cm ⁻²)	B/A
D ₂ SO ₄ -D ₂ O	Dummy	23 (A)	2.6
	Electrolysis	60 (B)	
H ₂ SO ₄ -H ₂ O	Dummy	12 (A)	1.3
	Electrolysis	16 (B)	

a) A fluctuation of neutron levels was observed in some dummy detectors. This may be due partly to slightly difference in treatment conditions. These conditions were the same in set of detectors in each experiment.

The number of fission tracks detected in the detector after electrolysis of D₂SO₄-D₂O solution was 2.6 times higher than that in the dummy detector, though the track density observed after the electrolysis of H₂SO₄-H₂O solution was 1.3 times larger than that in the dummy detector. The increased track number in the electrolysis of D₂SO₄-D₂O solution would be due to the (n,f) reaction of ²³⁵U by impinging of neutrons emitted during the electrolysis. The tracks observed in the dummy detector would correspond to the natural abundance of neutrons.

The relation between the neutron density and the track density was estimated by the determination of the relation between the neutron fluence from the $^{241}\text{Am}-^9\text{Be}$ neutron source and observed fission track density. The linear relation was obtained between the track density and the neutron fluence. One fission track corresponded to 10^7 neutron fluxes. The natural abundance of neutron was determined using fission track method and it was estimated to be $1.9 \text{ neutrons cm}^{-2} \text{ s}^{-1}$. This is approximately equal to that reported by Hess et al¹⁾. Comparing the results obtained during the electrolysis of $\text{D}_2\text{SO}_4\text{-D}_2\text{O}$ solution and $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ solution with the natural abundance of neutron, the neutron emission flux during the electrolysis of $\text{D}_2\text{SO}_4\text{-D}_2\text{O}$ solution is estimated to be $2 \text{ neutrons cm}^{-2} \text{ s}^{-1}$.

The β -ray radiation from the electrolytes and cathode materials after the electrolysis was measured by means of the liquid scintillation method. In the first case, the β -ray radiation from the electrolytes of $\text{D}_2\text{SO}_4\text{-D}_2\text{O}$ solution and $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ solution was measured before and after the electrolysis. The second one is the measurement of the β -ray radiation from the Pd plate used as the cathode materials in the electrolyses of $\text{D}_2\text{SO}_4\text{-D}_2\text{O}$ and $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ solutions. The results obtained are given in Table 2.

Table 2 β -ray Radiation due to Tritium Formation during the Electrolysis

Sample	cpm ^{a)}	dpm ^{a)}
$\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ (b) ¹⁾	0	0
(a) ²⁾	6.47	13.77
$\text{D}_2\text{SO}_4\text{-D}_2\text{O}$ (b)	117.92	235.84
(a)	193.82	387.64
Pd (b)	0	0
Pd ($\text{H}_2\text{SO}_4\text{-H}_2\text{O}$) ³⁾	3.85	9.17
Pd ($\text{D}_2\text{SO}_4\text{-D}_2\text{O}$) ⁴⁾	23.18	65.56

1) Before electrolysis, 2) After electrolysis, 3) After electrolysis in $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$, 4) After electrolysis in $\text{D}_2\text{SO}_4\text{-D}_2\text{O}$, a) Sampling 3 ml

From the results given in Table 2, the activity and the increment of tritium by the electrolysis of $\text{D}_2\text{SO}_4\text{-D}_2\text{O}$ solution were estimated as given in Table 3. The tritium production rate in the $\text{D}_2\text{SO}_4\text{-D}_2\text{O}$ solution during the electrolysis is estimated to be $10^4 \text{ T atoms ml}^{-1} \text{ s}^{-1}$. On the other hand, the production rate of tritium in the Pd electrode during the electrolysis of $\text{D}_2\text{SO}_4\text{-D}_2\text{O}$ solution is estimated to be $10^4 \text{ T atoms cm}^{-2} \text{ s}^{-1}$.

Table 3 Estimated Activity and Increment of Tritium during the Electrolysis

Sample	Activity (Bq ml ⁻¹)	Increment of Tritium
H ₂ SO ₄ -H ₂ O (b) ¹⁾	0	2.5x10 ³ atoms ml ⁻¹ s ⁻¹
H ₂ SO ₄ -H ₂ O (a) ²⁾	0.08	
D ₂ SO ₄ -D ₂ O (b)	1.31	2.7x10 ⁴ atoms ml ⁻¹ s ⁻¹
D ₂ SO ₄ -D ₂ O (a)	2.15	
Pd (b)	0	3.2x10 ³ atoms cm ⁻² s ⁻¹
Pd (H ₂ SO ₄ -H ₂ O) ³⁾	0.15	
Pd (b)	0	2.3x10 ⁴ atoms cm ⁻² s ⁻¹
Pd (D ₂ SO ₄ -D ₂ O) ⁴⁾	1.09	

1), 2), 3), and 4) are used as the same symbols in Table 2.

4. DISCUSSION AND CONCLUSION

The emission rate of the neutron during the electrolysis of D₂SO₄-D₂O solution was estimated to be 2 neutrons cm⁻² s⁻¹ by means of fission track method connecting with the neutron emission from the ²⁴¹Am-⁹Be neutron source. The tritium production rate in the Pd used as the cathode material during the electrolysis of D₂SO₄-D₂O solution was estimated to be 10⁴ T atoms cm⁻² s⁻¹.

The considerably large amounts of tritium observed in the Pd used as the cathode material in the electrolysis of D₂SO₄-D₂O solution would be formed by the d-d nuclear reaction, even if the increment of tritium in the electrolyte should be due to the enrichment of tritium during the electrolysis.

In conclusion, the branching ratio (T/n) estimated in the present experiment is 10⁴ like that in the electrolysis of LiOD-D₂O solution³⁾.

References

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