NUCLEAR REACTION CAUSED BY ELECTROLYSIS IN LIGHT AND HEAVY WATER SOLUTIONS

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ABSTRACT

A series of analyses of the products of some nuclear reactions caused by electrolysis was performed by a gamma-ray or liquid scintillation spectroscopy. The electrolysis was carried out by use of the so-called thermally open cell which was equipped with the cathode made of porous nickel or platinized platinum, in 0.1 ~ 0.5 mole/liter Li_2C0_3, Na_2CO_3, K_2CO_3, Rb_2SO_4 and Cs_2SO_4 light and heavy water solutions. The result obtained by the analysis and that of our previous works indicated that;

1. gamma peaks due to ^{22}Na and ^{24}Na, ^{40}K, ^{89}Rb and ^{92}Sr, or ^{134}Cs and ^{135}Xe occur during each electrolysis of Na^+, K^+, Rb^+ or Cs^+ solution, respectively,
2. gamma peaks due to ^{56}Co, ^{64}Cu and ^{65}Zn were shown in the cases of all electrolytes including even Li^+ solutions,
3. a gamma peak due to the positron annihilation was also observed in every solution, at 511 keV,
4. liquid scintillation spectra showed the increment of tritium produced by electrolysis in all light and heavy solutions except for Rb^+, and
5. some nuclear reactions were caused by electrolysis and occurred as the branching reactions of the hydrogen evolution reaction.

INTRODUCTION

The cold fusion occurring in the deuterium-palladium system becomes gradually not-so curious owing to many scientists' efforts in the world, since Fleischmann and Pons proposed it on the basis of finding anomalous large heat evolution during electrolysis in a heavy water solution of Li^+ [1]. On the other hand, by use of the so-called low over-voltage metals for hydrogen electrode reaction, large heat evolution and some amount of tritium production were observed during electrolysis, even in light water solutions of various alkali-metallic ions M^+ [2]. Furthermore, a few methods of chemical analysis of the electrolytes used for electrolysis provides some evidence of nuclear reactions, for example, Ca in K^+ [2][2] or 132-140X in Cs^+ solution [3], and so forth. For the last a few years, evidence of the nuclear reactions caused by electrolysis has rapidly increased owing to accumulation of works, experimental and theoretical. In particular, the simultaneous measurements of the excess heat and the radioactivity in an electrolytic cell are notable during electrolysis. In these systems, the reaction mechanism was determined as follows on the basis of the result of our studies using the galvanostatic transient method [4]:

$$M^+ + e^- \Rightarrow M(I) \quad (1)$$

$$M(I) + H(a) \Rightarrow M(I) + H(a) + OH^- \quad (2)$$

$$H(a) + H(a) \rightarrow H_2 , \quad (3)$$

where M(I), H(a), $\Rightarrow$ and $\rightarrow$ mean the inter-metallic compound between M and the electrode material, an adsorbed hydrogen, the rate non-determining and the rate determining steps, respectively.

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[2] First done by J.O'M. Bockris. –Ed.

The aim of this work is to find the evidence for the nuclear reactions of M(I), which must be occurring even in light water solutions. The data concerning the positron annihilation is reported in another place [5]. Therefore, it will be mentioned summarily in this paper.

2. Experimental Part

The electrolytic cell and electrodes were the same as described in the previous papers [2,3]. Namely, we used porous nickel or platinized platinum as a cathode (1.0 x 0.5 cm in size, ≤ 0.1 cm in thickness). The same cathode was usually used a few times for electrolysis in the same kind of electrolyte. Twenty or thirty milliliters of 0.5 mole/l LiOH, Na_2CO_3, K_2CO_3, or 0.3 mole/l Rb_2SO_4, or 0.1 mole/1 Cs_2SO_4 solution with light or heavy, or mixed water, was used as the electrolyte. Some series of observations of excess heat and γ-ray spectra were performed simultaneously during electrolysis of 6~100 hours in a dark chamber made of lead walls 10 cm in thickness. After electrolysis, the electrolytes were analyzed by use of a liquid scintillation spectrometer. The procedure was the same as described in a previous paper [6].

3. Results and Discussion

3.1. γ-ray Spectra

Fig. 1 shows the typical spectrum observed in K^+ solution by a γ-ray spectrometer equipped with a pure germanium detector (made by Nippon Atomic Industry Group Co. LTD).

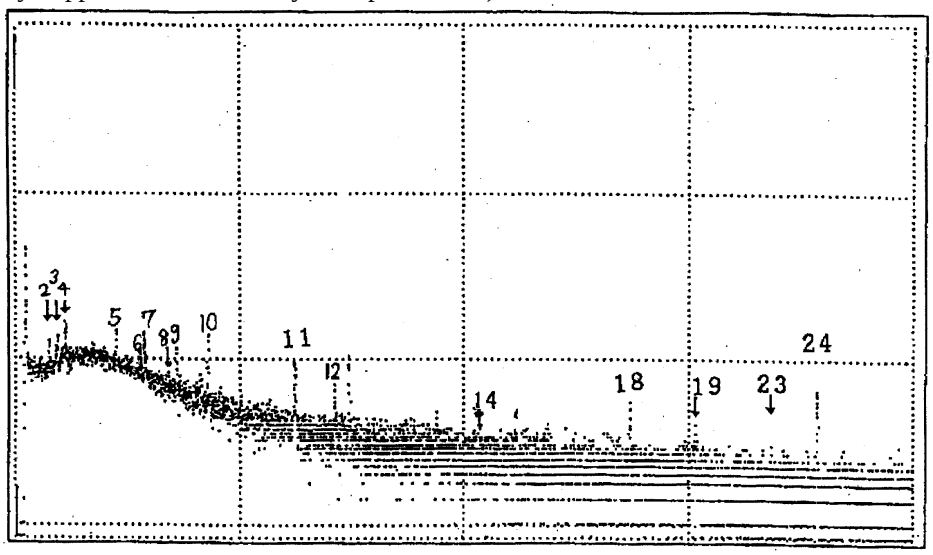


Fig. 1. γ-ray spectrum observed in K^+ solution on Ni: energy region, 0~1.6 M eV, duration and input power of electrolysis, 50 hour, 0.65 w. Peaks are numbered the same way in Table 1.

Table 1. Identified Nuclear Species and Their Amounts.

Nuclear Species	Energy (keV)	Amount (Bq)	Limit of Detected Value (Bq)	Peak No.
Th-234	63.33	2.11E-1 ± 1.21E-1	〈 3.65E-1	2
Hg-197	77.11	1.22E-2 ± 6.51E-3	〈 1.96E-2	3
Th-234	92.48	2.34E-1 ± 6.04E-2	1.81E-1	4
U -235	185.79	7.41E-3 ± 2.54E-3	〈 7.64E-3	5
Ra-225	185.79	1.03E-1 ± 3.52E-2	〈 1.06E-1	5
Pb-212	238.65	9.11E-3 ± 3.51E-3	〈 1.05E-2	6

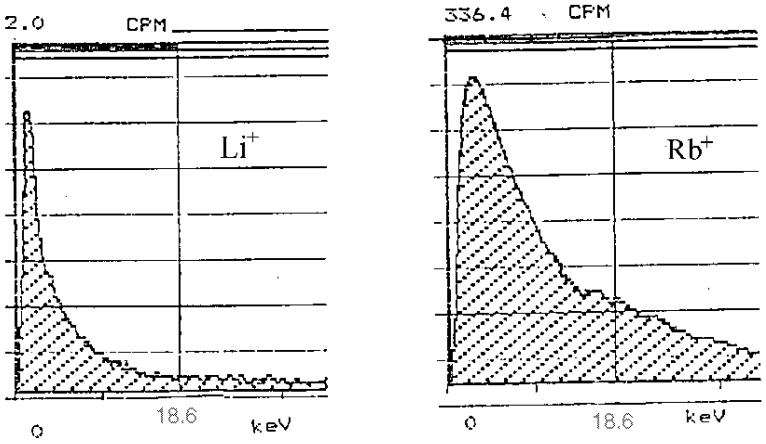
Pb-214	241.94	3.04E-2 ± 1.19E-2	〈 3.60E-2	7
Tl-208	277.48	1.05E-2 ± 1.01E-2	〈 3.08E-2	8
Pb-214	295.31	1.51E-2 ± 6.43E-3	〈 1.93E-2	9
Pb-214	351.98	1.51E-2 ± 2.54E-3	7.59E-3	10
Cu- 64	511.26	2.53E-2 ± 6.70E-3	2.00E-2	11
Zn- 65	511.26	1.04E-1 ± 2.75E-2	8.23E-2	11
Co- 56	511.26	6.76E-3 ± 1.78E-3	5.35E-3	11
N - 13	511.26	3.35E-1 ± 8.86E-2	2.65E-1	11
Tl-208	582.96	1.46E-3 ± 8.14E-4	〈 2.45E-3	12
Co- 56	846.53	4.93E-4 ± 4.82E-4	〈 1.46E-3	14
Zn- 65	1119.84	1.95E-3 ± 1.92E-3	〈 5.80E-3	18
Co- 56	1238.00	8.87E-4 ± 3.90E-4	〈 1.19E-3	19
Na- 24	1368.63	1.35E-3 ± 1.28E-3	〈 3.91E-3	23
K- 40	1458.72	1.89E-2 ± 3.73E-3	1.11E-2	24

The amount of each nuclear species estimated from the peak value shown in Fig. 1 was listed in Table 1, respectively.

These quantities estimated from the observation of γ-ray spectra are found in the region of 0.001 ~ 0.1 Bq. In Li^+ solution, merely the peaks due to the positron annihilation, ^{64}Cu and ^{65}Zn were increased by electrolysis. In the case of Na^+, ^{22}Na (1,276 keV) was often detected together with ^{24}Na (1,366 keV).

In K^+ solution, ^{40}K (1,461 keV) is increased always twice from 0.02 Bq to 0.04 Bq in 24 hours electrolysis with 0.5 ~ 0.6 w.

In the case of Rb^+ solution, ^{89}Rb (1,032 and 1,250 keV) was produced by electrolysis and sometimes ^{92}Sr. In the case of Cs^+, a small amount of ^{134}Cs (609 and 794 keV) and ^{135}Xe (25l keV) were detected. But in Cs^+ solution, the peak due to positron annihilation showed the highest. The reason is not clear at this time.



Figs. 2a and 2b Liquid scintillation spectra observed by use of Li^+ and Rb^+ samples.

3.2. Liquid Scintillation Spectra

A liquid scintillation spectro-analyzer (Packard CA 2550) was used for determination of tritium concentration in electrolyte after electrolysis. Two types of a liquid scintillation spectra were exemplified in Fig. 2a and

Fig. 2b, observed by use of the samples of Li^+ and Rb^+ solutions. The maximum position shown in Fig. 2, and those of the spectra in the samples of Na^+, K^+ and Cs^+ solutions, agree with that of the authentic sample of tritium. But, the maximum point of spectrum shown in Fig. 2b moved towards higher energy, that was strongly affected by the presence of ^{87}Rb (the natural abundance: 27.835%). The rates of generation of tritium by electrolysis were determined in all kinds of electrolytes of light and heavy water except for Rb^+, which were listed in Table 2.

Table 2. Generation Rate of Tritium in Various Electrolytes on Porous Nickel.

Electrolyte		W_{input} [a]	Excess Heat	^3T [b]
Ion	Water	w	%	Bq/20ml · 24 hr.
1. Li^+	H_2O	0.75	53	0.25
2. "	"	0.73	52	0.032
3. Na^+	H_2O	0.61	134	1.92
4. "	"	0.77	59	0.222
5. "	H_2O+ D_2O	- - -	- - -	2.006
6. K^+	H_2O	0.47	97	0.89
7. "	H_2O+ D_2O	- - -	- - -	3.42
8. "	D_2O	0.65	110	393.0
9. Cs^+	H_2O	0.71	62	32.0
10. "	"	0.72	61	72.0
11. "	"	0.41	- - -	7.0

a: Input power
b: Bq denotes tritium amount generated in 20 ml electrolyte during 24 hrs. electrolysis.

3.3. Nuclear Reactions Caused by Electrolysis

A clear peak was found at 511 keV, which must be due to the positron annihilation. In particular, it was remarkable that the peak appeared on all spectra which were observed during the electrolysis of all electrolytes used for this work. The increase of the peaks characterized by ^{64}Cu, ^{65}Zn and ^{56}Co with the increase of duration of electrolysis were common to all solutions. Besides, the increase of the products of some nuclear reactions described above can be estimated on the basis of γ-ray spectral data. These radioactive species are well known to be produced easily in a nuclear furnace, for example. $^{23}Na\ (n,\gamma)\ ^{24}Na$, $^{23}Na\ (\gamma,n)^{22}Na$, $^{39}K(n,\gamma)\ ^{40}K$, ^{56}Fe (p,n or d,2n)^{56}Co, $^{63}Cu\ (n,\gamma)^{64}Cu$, $^{64}Zn\ (n,\gamma)^{65}Zn$,and $^{133}Cs\ (n,\gamma)^{134}Cs$. ^{89}Rb, ^{92}Sr and ^{135}Xe may be produced through a series of some elementary reactions. Ca and the elements of the mass number from 132 to 140 were produced in the same way, which had been detected in the electrolysis of K^+ and Cs^+ ions.

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