

A Role of Lithium for the Neutron Emission in Heavy Water Electrolysis

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ABSTRACT

The depth profile analysis of Pd, Li, and D has been performed by means of SIMS to clarify the roles of lithium in D_2O electrolysis for cold fusion research. Very clear differences between the depth profiles of Li and D in the Pd electrode surfaces with the neutron emission and without it. The depth profiles were also found to depend on the mode of the electric current employed. Based on the above findings, it is discussed that the anomalous deuterium accumulation in the surface region of the Pd with the neutron emission attributed to the formation of Pd-Li layer in the surface region and to the Low/High pulse mode electrolysis of heavy water with $LiOD$.

1. Introduction

Lithium has been recognized as a key element not only for the neutron emission but also for the excess heat in the heavy water electrolysis method. There has been no clear evidence to discuss the role of lithium in the cold fusion cells.

In the present work, the depth profiles of lithium and deuterium have been deduced from SIMS analysis data of these two elements in the surface region of the Pd electrodes used in the electrolysis of heavy water with $LiOD$. The depth profiles of the two elements could be enable us to discuss the roles of lithium to accumulate deuterium in the surface region of the Pd cathodes.

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2. Experimental

The characteristics of Pd test pieces are listed in Table 1. The test pieces were selected by the following criteria.

- (1) Pd electrodes gave the appreciable neutron emission in the Low/High mode electrolysis, Pd-1, Pd-2, Pd-3.
- (2) Pd electrode did not give the neutron emission in Low/High mode electrolysis, Pd-4.
- (3) Pd electrode did not give the neutron emission in a constant current electrolysis, Pd-5.

The details of the electrolysis experiment and the neutron detection will be described in the proceedings of the present conference [Frontiers of Cold Fusion].

Table 1. The characteristics of the test pieces Pd

Test Piece No.	Electrolysis mode	Current density Low/High	Neutron emission
Pd - 1	L / H	10/200 mA/cm ²	yes
Pd - 2	L / H	10/200 mA/cm ²	yes
Pd - 3	L / H	10/200 mA/cm ²	yes
Pd - 4	L / H	2/40 mA/cm ²	no
Pd - 5	Constant current	40 mA/cm ²	no

The SIMS analysis was performed on a Secondary Ion Mass Spectrometer (IMS-4S, CAMECA/France) at Interdisciplinary Graduate School of Science and Technology, Tokyo Institute of Technology.

3. Results and discussion

As the typical examples of the depth profiles obtained by SIMS analysis are shown in Fig.1 (a) and (b) for the test sample Pd-2 and Pd-5, respectively. In this case the former is a typical sample which gave neutron emission and the latter is a typical sample without neutron emission. Comparing these two profiles, we can find the clear differences in the profiles of lithium and also deuterium between the two graphs. The depth profiles of the two elements at the surface region in Pd-2 have some structures, while the profiles in Pd-5 are monotonous. As will be shown later, the same feature is confirmed in other samples. This point is the first significant difference in the depth profiles of Li and D

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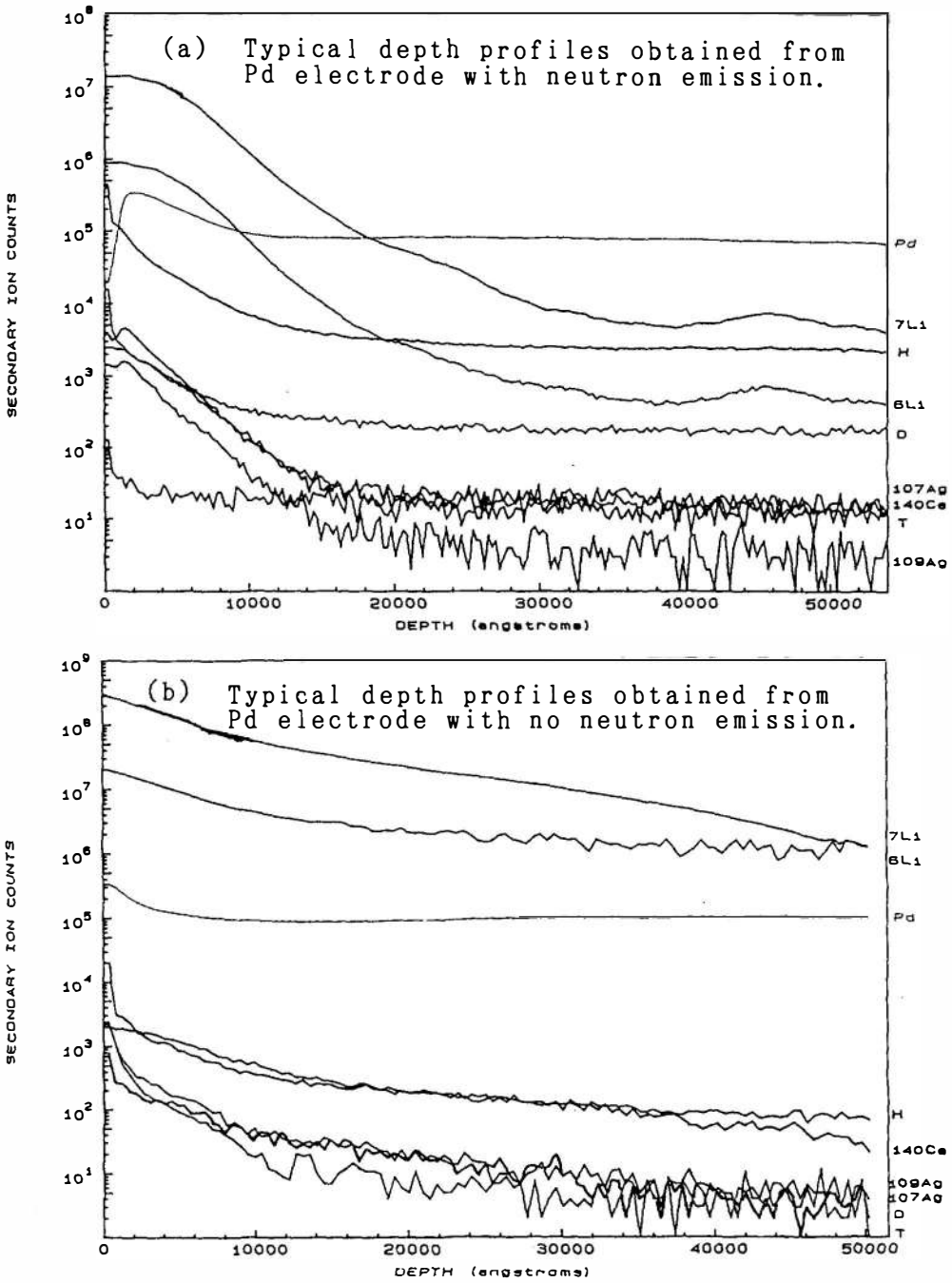


Fig. 1 The typical examples of depth profiles observed by SIMS analysis.

between the Pd sample with the neutron emission and that without the neutron emission.

The sensitivities for the individual elements in SIMS analysis are very different each other, so the absolute concentration of each elements can not be evaluated from the intensities obtained. The intensity of Pd ions is almost same in the two graphs, we can compare the relative concentration in each element. The lithium concentration in the surface region in Pd-5 is higher than that of Pd-2, while the concentration of D in the same region in Pd-5 is lower than that of Pd-2. This big difference of deuterium concentration seems to be the key factor for the occurrence of the neutron emission. This fact is the second significant difference found in the depth profiles in the two typical Pd samples.

In Fig.2, the depth profiles of Pd, Li and D are illustrated for the surface region upto 30 μm . The first significant difference mentioned above can be evidently found between the samples with the neutron emission (Pd-1, Pd-2, Pd-3; positive group) and those without the neutron emission (Pd-4, Pd-5; negative group). The second point is also clearly confirmed between the positive group and the negative group. We can find that the deuterium concentration in the positive group is much higher than those in the negative group. The anomalous high accumulation of deuterium may have a significant contribution to the neutron emission.

The next discussion point is also very important to elucidate the role of lithium for the neutron emission. The depth profiles of lithium have very similar feature to the profiles of deuterium. It means that the deuterium accumulation in the surface region may attribute to the accumulation of lithium in the region. The profiles of Pd also have similar feature to that of lithium. This fact indicates that Pd-Li compound formation takes place in the surface region.¹ The higher intensities of Pd and Li in the near surface region indicate that the atomic density of Pd and Li in the compound should be larger than that in the bulk of Pd electrode. The layer of the Pd-Li compound works as a barrier to escape the D in the stage of the low current density electrolysis.

Based on the above discussion and the high desorption rate of D from Pd reported by Mizuno² a model can be imaged for the mechanism of the anomalous accumulation of deuterium in the surface region.

- 1) By high current density electrolysis, lithium and deuterium absorbed into the Pd electrode.
- 2) Absorbed lithium formed Pd-Li compound at the surface region.
- 3) By reducing the current density to the low current density electrolysis, the deuterium moves to the surface with very high migration rate to attain to a

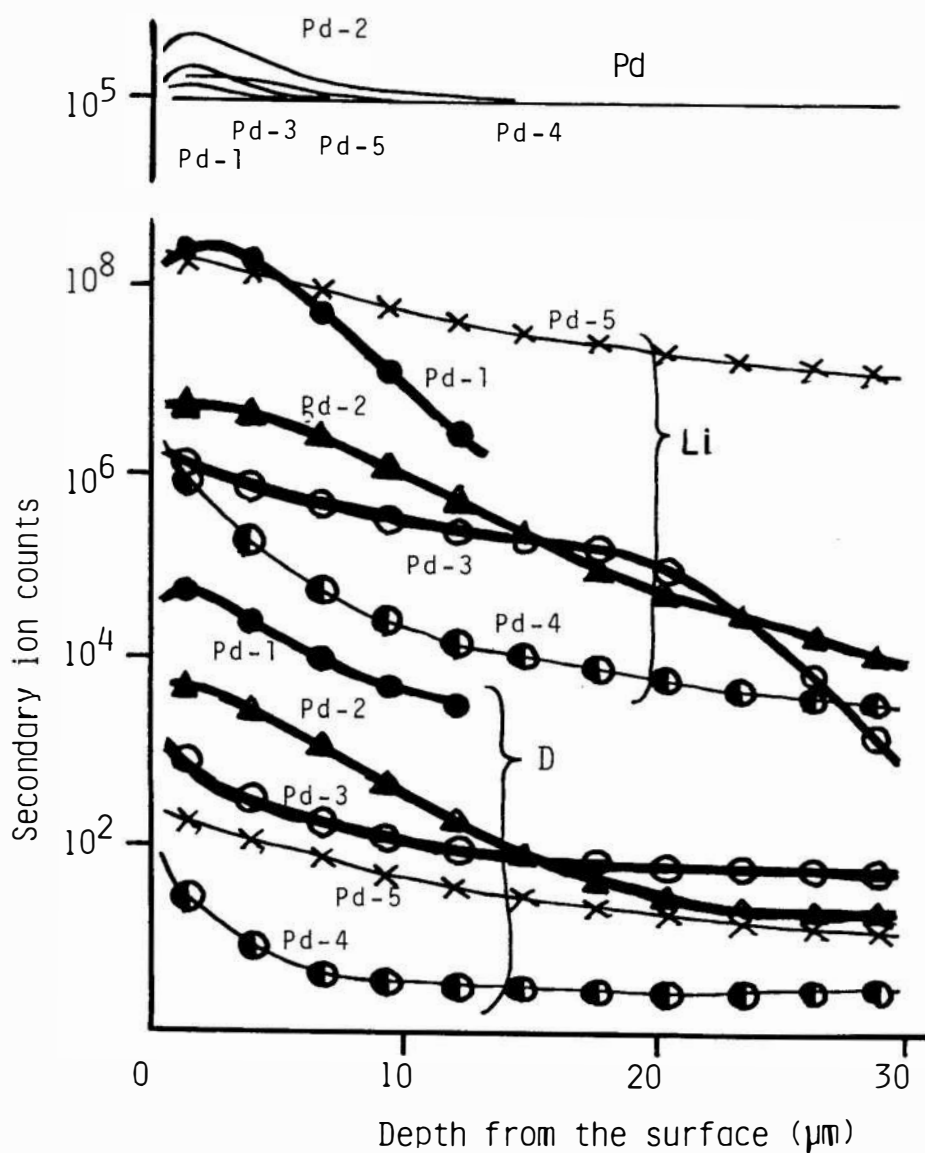


Fig.2 The depth profiles of Li, Pd and D in the surface regio upto 30μm.

In the figure, the bolt lines represent the depth profiles observed from the positive Pd samples and the thin lines from the negative Pd samples.

concentration equilibrated to the low current density.

- 4) The escape of deuterium to the heavy water was disturbed by the Pd-Li compound layer.
- 5) Thus the deuterium is accumulated anomalously in the surface region. As shown in Fig.2, the concentration of deuterium in the surface region become higher one to two order than that in the bulk.

The SIMS analysis was carried out after the rather long time storage of the electrodes in a sealed glass wear, still then such a depth profile was well kept. It indicates that the Pd-Li-D phase has to be very stable after its formation in the L/H pulse mode electrolysis. In the present work, the neutron emission was found in only the cases of L/H pulse mode electrolysis and the anomalous accumulation of deuterium in the surface region.

4. Conclusion

The depth profiles of Li, D and Pd were observed by the SIMS technique for several Pd test pieces cut from the Pd electrodes employed in the study of the neutron emission from the heavy water electrolysis using Pd-D₂O-LiOD system. The depth profiles can be classified into two types; one is the profile with a structure and the other is the monotonous profile. The former was observed on the Pd test piece from the Pd electrode with neutron emission, while the latter was from the Pd electrode without neutron emission.

The structure attributes to the formation of Pd-Li compound in the surface region of the Pd electrode. The compound works as the barrier to the migration of deuterium from the bulk to the surface at the stage of the low current density electrolysis in L/H pulse mode operation, resulting the anomalous accumulation of deuterium at the surface region of the Pd electrode.

It can be concluded that lithium forms Pd-Li compound and the formation of the compound causes the high accumulation of deuterium. The anomalous accumulation of deuterium may have an important effect to the neutron emission in Pd-D₂O-LiOD electrolysis.

References

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2. Mizuno T. Akimoto T. Azumi K. and Enyo M., 1992, DENKI KAGAKU, 60, No.5, 405.