# Detection of Radioactive Emissions in the Electrolytic Deuteriding-Dedeuteriding Reactions of Pd and Ti

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ABSTRACT

This report demonstrates the occurrence of radioactive emissions detected by GM(Geiger-Mueller)counter in the electrolytic deuteriding-dedeuteriding reactions of Pd and Ti. For the Pd samples annealed or cold worked, the excess counts higher than BG(back ground levels=32 ±2 cpm) by factors 1.5 to 2 in average were measured almost continuously and reproducibly during the pules modulated electrolysis over 600 mA/cm². The excess counts were measured for a while even after electrolysis. For the Ti samples annealed or cold worked, the burst-like GM counts over 200 cpm were often measured at low current densities below 10 mA/cm². The much higher burst-like GM counts over 1500 cpm were measured after electrolysis.

## 1. Introduction

In previous investigations[1-3], we reported that the pulse modulated deuteriding-dedeuteriding reactions of Pd in a KOD cell exhibit distinct excess GM counts and that the cold working of Pd sample is effective to yield higher GM counts. The reactions of Pd deuteride formation-decomposition at the Pd surface seem to be crucial for the radioactive emissions. This work dealt with the similar measurement using Ti which forms more stable deuteride than Pd[4] and the results for Pd and Ti are compared and discussed.

## 2. Experimental Process[1]

The schematic arrangement of the cell, GM counter(ALOKA Basic Scaler TDC-105), NaI scintillation counter(CANBERRA series 20) and pulse generator used are shown in Fig. 1. BG fluctuations were monitored at real time before, during and after electrolysis using two additional GM counters set at at distances of 6 m(BG. 1) and 45 m(BG. 2) from the cell, respectively as shown in Fig. 2. Electric noises as well as the fluctuations of the GM counters used were strictly checked by switch-on and off of equipments in the laboratory

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building over one week before each experiment. Wire samples(1mmx1mmx50mm) of Pd and Ti and a Pt plate were used for electrolysis in 0.5 N-KOH or KOD solutions. The purity of $D_2O$ used was 99.8-99.9%.

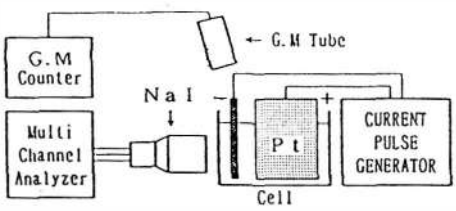


Fig.1 Schematic arrangement of the electrolysis.

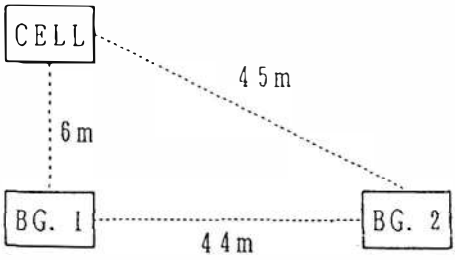


Fig.2 BG measurements at different sites:BG.1 and BG.2.

## 3. Results

The measured GM counts of BG at the cell are shown in Fig.3 as deflectionin %, Δ1 from BG.1 and Δ2 from BG.2 in Fig.3. Each point of deflection displayed was calculated from 10 min average counts. The GM counts at the cell exhibit around 20% steday deflections compared with BG.1 and BG.2. Fig.4 shows an example of the GM counts measured for a Pd sample

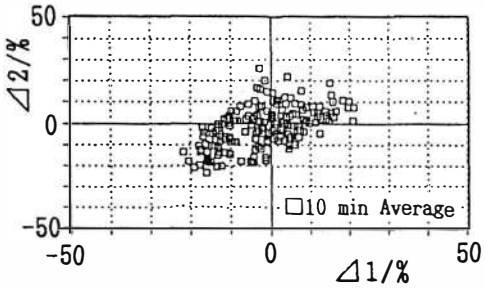


Fig.3 Fluctuation of BG at the cell in comparison with BG.1 and BG.2.

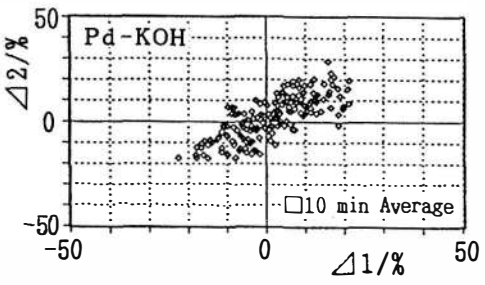


Fig.4 Deflections from BG.1 and BG.2 in pulse modulated hydriding-dehydridings of Pd cold worked 49.4 % in 0.5 N-KOH at 5 A/ $cm^2$

deformed by 49.4% in a pulse modulated electrolysis at 5000 mA/ $cm^2$ in a 0.1 N-KOH cell. The measured deflections exhibit an almost identical distribution with BG fluctuations in Fig.3, indicating no occurrence of meaningful phenomenon in the electrolysis of Pd in KOH. However, Fig.5 and Fig.6 show the deflections of GM counts at the cell in pulse modulated electrolysis of two different Pd samples deformed by 49.4% at 4000 mA/$cm^2$ and 600 mA/$cm^2$ in KOD cells, respectively. Much higher deflections than BG fluctuations can be seen in the first quadrant, meaning that these pulse modulated reactions yield distinct excess GM counts at the cell in 40-50% positive deflections. The distinct excess counts could be measured in 20 30 min after the initiation of

electrolysis and continued for a while after electrolysis.

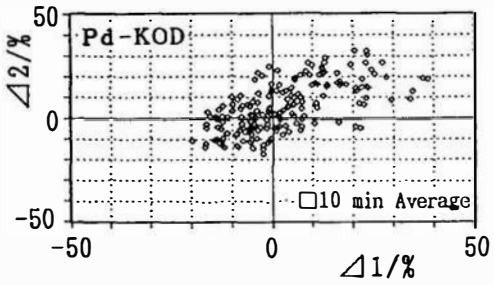


Fig.5 Excess GM counts as positive deflections in pulse modulated deuteriding-dedeuteridings of Pd cold worked by 49.4 % in 0.5 N-KOD at 4 A/ cm².

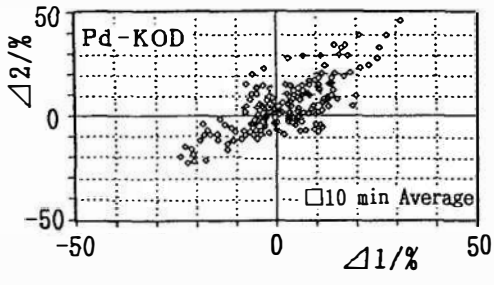


Fig.6 Excess GM counts as positive deflections in pulse modulated deuteriding-dedeuteridings of Pd cold worked by 49.4 % in 0.5 N-KOD at 0.6 A/cm².

The excess GM counts during and after electrolysis of Ti in KOD solution were much higher than those observed with Pd. Therefore, the measured GM counts for Ti are presented in cpm as a function of time t in Fig. 7 and Fig. 8.

Fig. 7 shows burst-like GM counts up to about 300 cpm for a Ti sample cold worked by 32.5% at t=100 to 130 min in an electrolysis at 6.0 mA/cm². Similar burst-like emissions took place also at t=500-540 min after electrolysis after the current had been cut off at t=400 min. At t=200 to 400 min, a pulse modulated current was applied at 0-4600 mA/cm², however, no excess GM count was measured. Fig. 8 shows much higher bursts in GM counts over 200 cpm for a Ti sample deformed by 7.1% at t=20-30 min in an electrolysis at 5 mA/cm² and also burst-like GM counts over 1500 cpm at t=450-460 min after electrolysis. Such burst-like emissions were measured three times in or after electrolysis using seven different Ti samples.

## 4. Discussion

The average compositions of Ti deuterides were $TiD_{0.6-0.8}$ for Fig. 7 and $TiD_{0.1}$ for Fig. 8 during electrolysis. However, the optical observations on the samples showed that only the surface regions down to a few $\mu$m were mainly deuterided, meaning the formations of deuterides with much higher D concentrations at the surface than the calculated average concentrations. Judging from thermodynamic properties of Ti hydrides[4], $\gamma$-$TiD_2$ phase may be formed in the electrolysis at the surface. The burst-like emissions observed seem to be related to the formation and decomposition of the di-deuteride phase in and after electrolysis, respectively, and which is similar to the excess emissions of the Pd-D system by a pulse modulated electrolysis[1-3]. The observations for Pd and Ti imply the occurrence of radioactive emissions like $\gamma$- or X-rays detectable by GM counters.

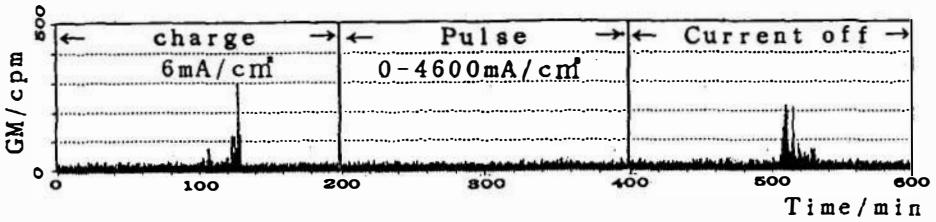


Fig.7 Burst-like GM counts in deuteriding of Ti cold worked by 32.5 % at 6 mA/cm² and in curret-off state after electrolysis.

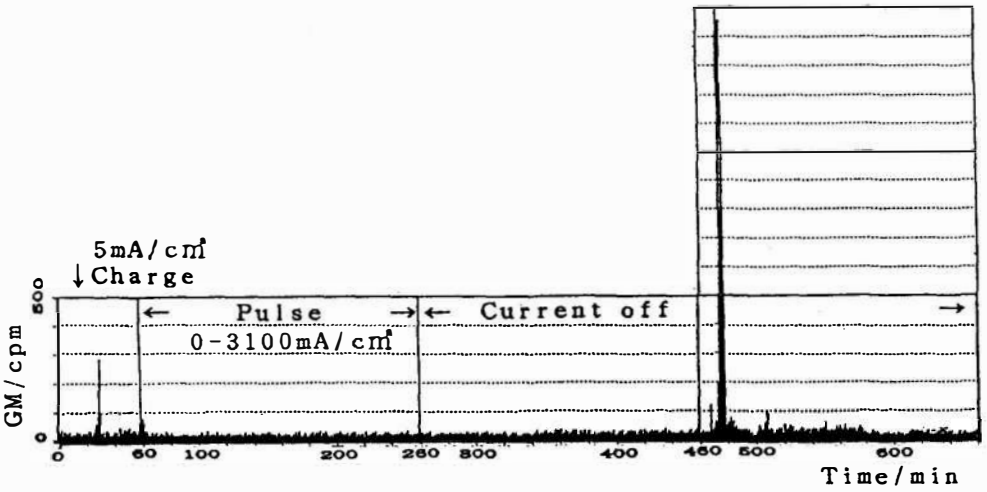


Fig.8 Burst-like GM counts in deuteriding of Ti cold worked by 7.1 % at 5 mA/cm² and in curret-off state after electrolysis.

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## 5. References

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