Measurement of Protons and Observation of the Change of Electrolysis Parameters in the Galvanostatic Electrolysis of the 0.1M-LiOD/D_2O Solution

Shinya.MIYAMOTO, Keisuke.SUEKI, Hidekazu.IWAI, Masatoshi.FUJII, Toshiaki.SHIRAKAWA, Hiroaki. MIURA, Toshihiro.WATANABE, Hideyuki.TORIUMI, Torahiko.UEHARA, Yuki.NAKAMITSU, Masami.CHIBA, Tachishige.HIROSE, and Hiromichi.NAKAHARA
Faculty of Science,Tokyo Metropolitan University
Minami-Ohsawa, Hachioji-shi, Tokyo 1-1, JAPAN

Abstract

In order to confirm the cold fusion phenomena, measurements of protons in the galvanostatic electrolysis of the 0.1M-LiOD/D_2O solution have been carried out. The upper limit of fusion rates was deduced to be 1.35×10^{-24} fusion/d-d/sec with an assumption of the atomic ratio D/Pd of unity. No charged particles predicted by Takahashi[1] for d-d-d fusions were observed. The Li content in the electrolyte was measured by ICP-AES after the electrolysis and found to be appreciably reduced in the electrolyte; the rest being found mostly in the 0.5%-Pd alumina catalyst used for recombination of D_2 and O_2 and partly in the Pd cathode.

1.Introduction

In march 1989, M.Fleishmann et al.[2] and S.Jones et al.[3] first reported the cold fusion phenomena which they claimed proceeded through next reactions:

$$d + d \longrightarrow {}^3He\ (0.82MeV) + n\ (2.45MeV) \quad (1)$$
$$d + d \longrightarrow {}^3H\ (1.01MeV) + p\ (3.02MeV) \quad (2)$$
$$d + d \longrightarrow {}^4He + \gamma + 23.4MeV \quad (3)$$

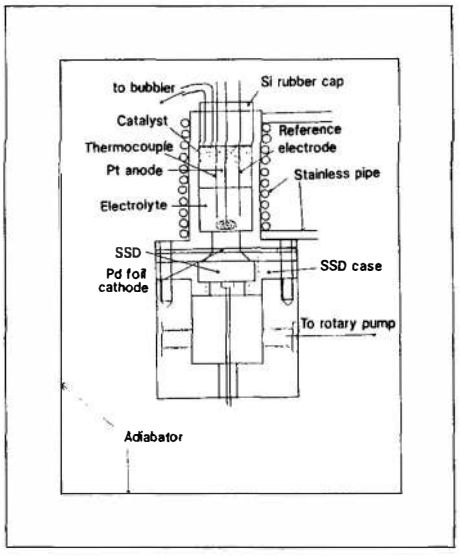
According to the current knowledge of nuclear physics, reactions (1) and (2) occur with the similar probability. A few paper reported to measure charged particles.[4],[5] In the present work, protons were measured to confirm the cold fusion phenomena through the reaction (2). The detection of protons is generally more sensitive than that of neutrons due to the lower background and the higher detection efficiency. But, as the range of 3.02MeV protons is 30μm in palladium, the

FRONTIERS OF COLD FUSION

maximum allowed thickness of a Pd foil is only about 25μm.

2.Methods

Galvanostatic electrolysis of the 0.1M-$LiOD/D_2O$ solution was carried out under a constant current of 200mA ($100mA/cm^2$) with a 25μm thick Pd foil (Nilaco co., PD343228,) as a cathode whose effective diameter was 1.6cm. (0.0604g as Pd) Six runs were performed until now. In RUNs 001-004, the back side of the Pd foil was coated with about 1.1μm thick SiO_x for prevention of leakage of deuterium. Figure shows the cell especially fabricated for the experiment. In RUN006, the whole system was thermally insulated for the measurement. A $4.5cm^2$ surface barrier Si detector (SSD) (EG&G ORTEC, Model No.; BA-21-450-500, Serial No.; 30-531C) was used for detection of protons. The void space in between the Pd foil and the SSD was evacuated with a rotary pump for reducing the background level due to ^{222}Rn and also for avoiding the degradation of proton energy. The 0.5%-Pd alumina pellets (NE Chemcat co. Lot No.: 256-18130) were suspended above the electrolyte solution as a catalyst for recombination of D_2 and O_2 gas into D_2O. The current, input voltage, reference voltage, and the temperature of the electrolyte and the room were monitored. After electrolysis, Li contents in the electrolyte, in the catalyst, and in the Pd foil were measured by ICP-AES. The expected energy spectra of the p, d, ^3H ^3He, and ^4He have been calculated by the Monte Carlo method for various sites of their origin within the Pd foil.



< Figure >
The cell for the experiment.

3.Results

Measurements of protons were performed three times for the $LiOD_{aq}$ (RUNs 001, 002, and 006) and once for $LiOH_{aq}$ (RUN003). No protons were observed above the background level which was 9 counts in 74.39hrs with the effective counting efficiency of 12.4%. From 3σ of the

background counts, the upper limit for the reaction (2) was deduced to 1.59×10^{-24} fusion/d-d/sec. No charged particles predicted by Takahashi were detected who proposed the d-d-d fusion in order to explain the T/n ratio of 10^5-10^6 reported the many investigators. Detailed conditions and results of each run are summarized in table 1 and 2.

In RUN001, the electrolyte temperature reached above 353K after 6days. But unfortunately, the input voltage and the room temperature were not measured in this run, and no definitive conclusion can be made as to the excess heat. For observing the excess heat, both input voltage and room temperature were monitored in RUN004, 005. In RUN004, a sudden increase of the input power was observed after 14days, although the electrolyte temperature decreased unsteadily. In RUN005, the space in between the Pd foil and the SSD was not evacuated and then, no such an increase of the input power and a decrease of the electrolyte temperature was observed. In RUN006, the whole electrolysis system was thermally insulated from the environment, and the cell was cooled by the forced flow of water through a stainless pipe. From the difference of the water temperature between IN and OUT, no excess heat was observed.

In RUN001 and 005, atomic ratio of D/Pd was measured. These were 0.652 and 0.705, respectively. From our previous measurement of D/Pd by weight and TPD of Pd rods, D was found to escape from the Pd rod in two stage. The percentage of D that escape quickly after the end of electrolysis is about 30% while that of the slow component is about 70%. The above cited values of about 0.7 is only the amount of the slow component, and so, the total D/Pd ratio is estimated nearly 1.0 in our

Table 1 : Detailed conditions and results of each run.

RUN	Electrolytic time(h)	Measurement of protons(cph) 1501-2400ch	701-1000ch	301-4096ch•	Fusion rate♣ (f/d-d/s)
001	165.5	0.103±0.025	1.01±0.082	31.6±0.437	1.35×10^{-24}
002	120	0.125±0.032	1.29±0.104	36.1±0.548	1.64×10^{-24}
003♥	74.39	0.121±0.040	1.01±0.117	35.6±0.692	1.59×10^{-24}
004♦	503	———	———	———	———
005♦	956.7	———	———	———	———
006♠	247	0.181±0.027	1.61±0.083	74.2±0.562	2.24×10^{-24}

♥ : 0.1M-LiOH/H_2O solution was used for electrolyte.
♦ : Protons were not measured.
♣ : For the calculation of fusion rate, D/Pd was assumed To be 1.0.
♠ : In RUN006, the background level is to be higher.
• : 2400ch = 3.02MeV

experiment.
Li contents before and after the electrolysis were measured by ICP-AES. While the volume of electrolyte solution decreased typically from 20ml to 6ml in 22days, the Li concentration reduced to less than half of its initial concentration. In RUN004, presence of a white substance was recognized on the back of the Pd foil and in the vessel of vacuum system. This white substance was found to contain Li. In all run, most of the Li lost from the electrolyte was found in the catalyst, but a small amount of Li was always found also in the Pd cathode. Therefore, it is probable that Li plating on Pd becomes possible after Pd attaining a certain amount of D in the cathode.

Table 2 : Li content in Pd cathode, Li/Pd, D/Pd

RUN	Li content in Pd cathode	Li/Pd	D/Pd
001	$7.914X10^{-6}g$	$2.00X10^{-3}$	0.652
002	$1.970X10^{-6}g$	$5.00X10^{-4}$	———
003	$1.205X10^{-6}g$	$3.07X10^{-4}$	———
004	$1.251X10^{-5}g$	$3.15X10^{-3}$	———
005	$1.293X10^{-6}g$	$3.24X10^{-4}$	0.705

4.Discussion

Measurements of protons and excess heat in the galvanostatic electrolysis in the 0.1M-$LiOD/D_2O$ were carried out, and no protons above the background level and no excess heat were observed. A longer run is being planned for making our conclusion more solid. We found the Li content in the Pd foil increased after electrolysis. The finding may be related to the report[6] that if Pd metal contains Li, deuterium solubility increases. More careful chemical investigations have to be made before ascribing the observed anomalous phenomena to more unrealistic nuclear fusion.

5.Reference

1. A.Takahashi et al., 1991, THE SCIENCE OF COLD FUSION (PROCEEDINGS OF THE II ANNUAL CONFERENCE ON COLD FUSION, Societa Italiana di Fisica), 93
2. M.Fleischmann, S.Pons and M.J.Hawkins, 1989, J.Electroanal. Chem., 261, 301
3. S.E.Jones et al., 1989, Nature, 338, 737
4. P.B.Price et al., 1989, Phys. Rev. Lett., 63, 1926
5. J.F.Ziegler et al., 1989, Phys. Rev. Lett., 62, 2929
6. Y.Sakamoto et al., 1991, Zeitschrift für Physikalische Chemie, 173, 235