J. Condensed Matter Nucl. Sci. 13 (2014) 422-431



Research Article

Method of Controlling a Chemically Induced Nuclear Reaction in Metal Nanoparticles

Tadahiko Mizuno*

Hydrogen Engineering Application and Development Company, Japan

Abstract

A nuclear reaction can occur when metal nanoparticles are exposed to hydrogen isotopes in the gas phase. When hydrogen isotopes (light hydrogen and deuterium) enter the nanoparticles and are exposed to electron irradiation, the hydrogen reacts inside the lattice, producing energy. The reaction also produces neutrons, gamma rays and transmutations. Protons and heavy electrons take part in the reaction. The method of control and the experimental results are reported here. © 2014 ISCMNS. All rights reserved. ISSN 2227-3123

Keywords: Gas loading, Heat, Nanoparticles

1. Experimental Method

Excess heat is observed when nanoparticles of nickel or palladium undergo glow discharge electrolysis in hydrogen or deuterium gas. After glow discharge ends, when additional gas is admitted to the cell, another burst of heat is sometimes observed.

This method produces nanoparticles by bombarding the electrodes with electrons during roughly 30 h of glow discharge. The nanoparticles are created in situ in electrodes [1–3]. The electrodes have already been cleaned and purified, and they are kept in a high-purity gas environment, so the nanoparticles have little contamination on the surface and they are highly absorbent. With other methods, the nanoparticle material is fabricated elsewhere and exposed to air and contamination before being placed in the cell.

The particle size and number of particles is controlled by varying the strength and duration of initial glow discharge. This, in turn, controls the reaction rate. In a typical run with a fine wire palladium electrode, the cell produced 4.4 kJ of anomalous heat at first. After polarity was reversed, making the electrode positive, 54 kJ more excess heat was produced. After glow discharge finished, 10 ml of additional deuterium gas was added to the cell, and an extra 15 kJ of anomalous heat was produced.

Excess heat was produced in most test runs (47 out of about 70 tests).

^{*}E-mail: head-mizuno@lake.ocn.ne.jp

^{© 2014} ISCMNS. All rights reserved. ISSN 2227-3123

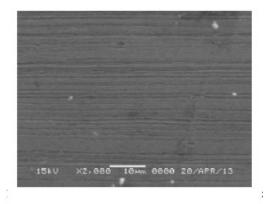


Figure 1. Palladium wire before use. The wire has been scored.

2. Materials and Equipment

2.1. Materials

The materials used in the deuterium–palladium experiments are described here. The deuterium gas (Japan Fine Products) purity is 99.99% for D, or for D₂, 99.8%, with 0.2% HD). Heavy water vapor (Across, 100.0 at %) is also used. Commercial grade nickel mesh made from 0.2 mm wire is used to cover the inside walls of the reactor, to prevent the glow discharge plasma from touching the steel reactor walls.

For the palladium tests, commercial grade palladium wire, 0.1-1.0 mm thick, 99.9% pure was used. The wires are scored before use. Figure 1 shows a palladium wire sample before exposure to glow discharge. This wire is 300 mm long. It is wrapped in a spiral around a rod, and then exposed to glow discharge which forms nanoparticles on the surface.

2.2. Nanoparticle size

The smaller the nanoparticles in a sample are, the more effectively they absorb hydrogen, and the faster the reaction will occur. I believe the reaction rate increases exponentially. On the other hand, the overall reaction size depends on the number of particles in the active site. As the metal particle size decreases, both the particle volume and the number of hydrogen atoms absorbed by the particle decrease by the third power (cubically).

We can calculate the optimum particle size taking into account these two factors. It comes out to be 2–5 nm, where the number of atoms per particle is 10^3-10^4 .

2.3. Reactor and electrodes

Two reactors were used in this study, a small one and a large one. This paper describes the large reactor and electrodes. The reactor is stainless steel. Grade 316 stainless steel is preferred. The vessel is about as wide as it is tall, (50 cm \times 50 cm) being cross-shaped with the gas inlet and window ports. Electrodes are introduced from the top, and there are a variety of connection terminals at the sides, including the gas inlet, pressure gauge and vacuum exhaust. The viewing window is made of Kovar glass. ICF flanges are used throughout, with oxygen-free copper gaskets. The reactor volume is 15 L, weight 50 kg.

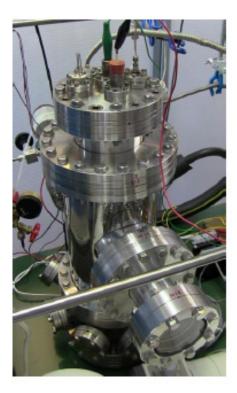


Figure 2. The reactor.

As shown in Fig. 3, the two electrodes enter the cell through the lid. Both are insulated. The polarity of the electrodes can be reversed. The entire reactor vessel is grounded for safety.

The core of the electrode is a square alumina ceramic holder, 30 mm per side, 2 mm thick, with palladium wire wrapped around it. The wire is 0.2 mm thick, 1000 mm long, and is coiled around the ceramic holder about 15 times. The other electrode is made with 300 mm of palladium wire wrapped in a tight spiral around a palladium tube, which is 50 mm long, 3 mm in diameter.

Counter-electrode: With nickel wire the counter-electrode is either 1 mm diameter 30 mm long wire, or thinner 0.1 mm diameter, 1000 mm long wire, both wrapped in a spiral around a rod. The wire is wrapped as tightly as possible. It is arranged so there are no protruding or pointed surfaces. Before use, the entire electrode assembly is washed in alcohol and acetone, in particular to eliminate contamination from fats and oils.

2.4. Nanoparticle size

The smaller the nanoparticles in a sample are, the more effectively they absorb hydrogen, and the faster the reaction will occur. I believe the reaction rate increases exponentially. On the other hand, the overall reaction size depends on the number of particles in the active site. As the metal particle size decreases, both the particle volume and the number of hydrogen atoms absorbed by the particle decrease by the third power (cubically). We can calculate the optimum



Figure 3. The reactor lid and electrodes.

particle size taking into account these two factors. It comes out to be 2-5 nm, where the number of atoms per particle is 10^3-10^4 . A cross-section of the reactor is shown in Fig. 4.

2.5. Equipment configuration

Figure 5 shows a block diagram of the reactor, the control system, the vacuum exhaust and mass spectroscopy system, and the measurement and control systems. Voltage is applied from the high voltage (HV) power supply (left-hand side of diagram). Also shown on the left is the computer, data logger, power analyzer and waveform analyzer. Along the top of the cell is a neutron detector and gamma detector, and a pressure gauge. In this diagram, the cell is shown in the lower portion.

The temperature of the electrodes is measured directly with a thermocouple (shown in the upper right pin in Fig. 4). This is a K-type thermocouple, 1.6 mm diameter, 300 mm long, in a stainless steel jacket. It touches the surface of the electrode wrapped around the ceramic holder.

On the right-hand side is shown one of the two K-type thermocouples measuring the reactor vessel wall temperature (T5 is shown; T6, at the base, is not). Another thermocouple is inside the cell, in direct contact with the electrode surface. The bottom right of the diagram shows the gas supply and quadrupole mass analyzer used to analyze the gas withdrawn from the cell. Temperature is measured in four locations total, including ambient temperature. Temperature, voltage, power, pressure, the neutron and gamma count, and the gas analysis is collected by the logger and recorded in the computer every few seconds.

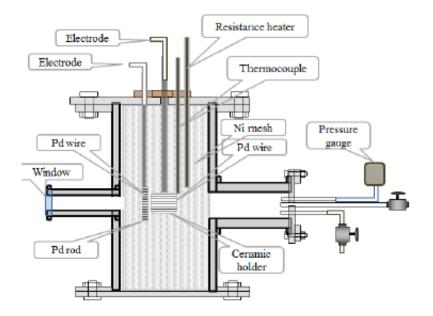


Figure 4. Cross section of the reactor.

3. Method

3.1. Heater calibration

Figure 6 shows the results of a calibration performed with the joule heater that is placed up against the electrode. The cell is filled with D_2 gas at 10 or 100 Pa, and the power is stepped from 0 to 50 W in 5 W increments. The results for 10

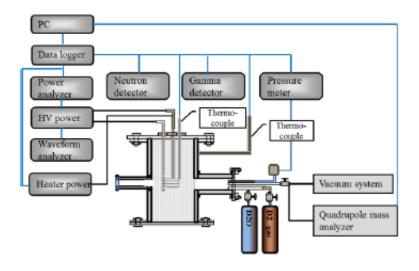


Figure 5. System.

and 100 Pa are close. The temperature is held at each step for about 18 h to ensure that it is stable. This graph shows the stable temperature minus ambient temperature for each step, recorded on the three K-type thermocouples listed above.

3.2. Electrode activation treatment

The electrode is treated by the following method to produce nanoparticles and activate the surface: The reactor vessel is evacuated down to several Pascals. After evacuating the reactor, the gas level is held at several Pascals, and electrons from the central electrode are used to bombard the counter-electrode. This activation treatment step exposes the metal, cleaning off impurities and oxides. Voltage and current are applied to the electrodes. At first the 0.1 mm thin wire palladium mesh is made the positive electrode at 600 V, and glow discharge at 20 mA is continued for 600 s. Then the mesh is switched to negative terminal, and glow discharge continues for 1200 s, again at 600 V and 20 mA.

This glow discharge cycle is repeated for about 15 h with electrodes made from thin 0.1 mm diameter wire, or for 30 h with the 1 mm wire. Glow discharge continues until many nanoparticles form on the surface, and the surface is activated. The vessel is then evacuated to a lower pressure while the electrodes are hot, to remove additional impurities. The electrodes are then activated by being heated with the resistance heater, to temperatures between 100° C and 200° C for about 3 h. This continues until light hydrogen, H₂O, and gaseous hydrocarbons are driven out of the electrodes and are no longer detected by the mass spectrometer at significant levels.

3.3. Glow discharge heating

Glow discharge is then performed to produce nanoparticles on the electrode surface. The thin wire palladium electrode is the positive terminal. Direct current glow discharge is maintained at about 20–30 mA and 600–800 V for about 10 ks (\sim 27 h). After this, D₂ gas is admitted into the cell, and it absorbs into the electrode surface.

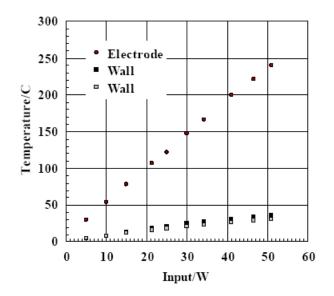


Figure 6. Heater calibration.

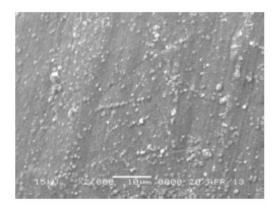


Figure 7. Palladium wire after activation.

Figure 6 shows a SEM photo of the palladium wire electrode surface after this treatment. Fine metal particles (nanoparticles) are formed on the metal surface. The magnification here is 2000 times, which is enough to reveal nanoparticles of less than 1 μ m. When the SEM magnification is increased, even smaller particles can be seen. Particles on the nanometer scale are ideal. Figure 7 shows a SEM photo of the nickel wire, with even more nanoparticles than the palladium sample.

This method produces nanoparticles in situ after cleaning and purifying the electrode material, in a high-purity gas environment. So the nanoparticles have little contamination on the surface and they are highly absorbent. With other methods, the nanoparticle material is fabricated elsewhere and exposed to air and contamination before being placed in the cell.

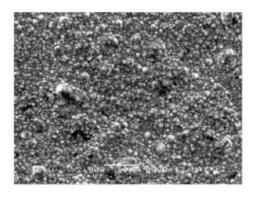


Figure 8. Nickel wire after activation.

4. Results

4.1. Anomalous exothermic reaction

Figure 9 shows an example experimental run. Input power is shown in the right axis, with the broken dashed line. It is roughly 46 W. The grey line indicates gas pressure, which begins with a vacuum and is increased to 320 Pa at 2000 s. When gas pressure is raised, the reactor electrode temperature (the difference between the electrode and room temperature) increases from 220° C to 300° C. Later, as shown in Fig. 10, the reactor wall temperature increases from 38° C to 44° C, and then stabilizes.

The amount of heat can be calculated based on the electrode temperature calibration, as follows. With 46 W of input, the calibration shows that the electrode temperature difference should reach 250°C, but it climbs to 300°C instead, which indicates 60 W of power. Therefore, excess power is 60-46 W = 14 W. This excess power can sometimes continue for over 50,000 s (14 h). Excess heat can also be computed based on the reactor wall temperature, and this also indicates ~ 14 W excess.

Figure 11 shows single points representing the stable (equilibrium) temperature differences from several tests. That is, the electrode minus room temperature, and cell wall minus room temperature. These temperatures were recorded in various experiments 2000–10,000 s after gas is admitted into the cell, after the temperature reaches equilibrium. The two straight lines in the figure indicate the calibration curves from control runs for the electrode (*top*) and cell wall (*bottom*), where the data was taken before the metal was activated. This shows that the temperatures are considerably higher for samples that have been activated.

In one example data point in Fig. 11, where input as 30 W, the average output temperature was 150° C during calibration. After the electrode was activated, the temperature difference rose as high as 280° C. In the control runs, 280° C indicates 50 W of input. In other words, 50-30 W = 20 W excess heat. During the control run, with 30 W input, the cell wall temperature is 17° C, but with activated material it reaches 26° C, which again indicates about 50 W of excess heat.

Excess heat first becomes apparent when the cathode temperature goes above 50°C. However, when the cathode temperature exceeds 300°C the metal is deactivated and the excess heat goes away, probably because of structural

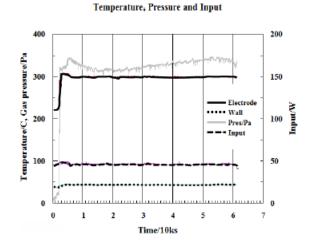
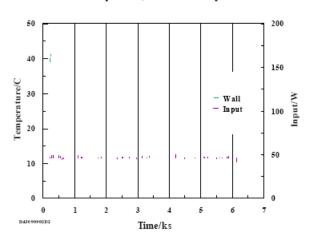


Figure 9. Electrode temperature, wall temperature, gas pressure (left axis) and input power (right axis).



Temperature, Pressure and Input

Figure 10. Detail showing reactor wall temperature and input power.

changes. At present the reason is unclear. Table 1 shows a summary of recent tests. These were all conducted with the large reactor. The electrodes were palladium, either a rod or a wire. D_2 gas or D_2O or H_2O vapor was used. The tests were performed after the electrode was activated with glow discharge for many hours. The electric power was either direct current or alternating current at 50 Hz. Output is calculated based on the calibration curves during the control

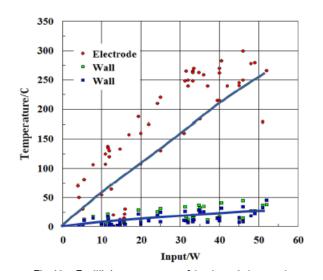


Figure 11. Equilibrium temperature of the electrode in several experimental runs.

run. The "Out/in" columns show the ratio of output heat to input power, estimated from the electrode temperature, and the cell wall temperature.

As shown in Fig. 9, after the sample has undergone heat treatment and activation with glow discharge, excess heat sometimes continues for 24 h or longer. In some cases output exceeds 10^6 kJ, and continues longer than 100,000 s. Heat has been observed with both heavy water vapor and D₂ gas. This indicates that palladium wire that has undergone surface treatment to produce nanoparticles can produce anomalous excess heat.

	Gas		Applied V		Power (W)		Heat out (W)		Time (ks)	$H_{\rm out}/H_{\rm in}$		Temp.(°C)
Cathode-	Component	Pressure (Pa)	Heater (W)	Plasma (V)	Heater input	Plasma input	By electrode temp.	By reactor temp.	()	By electrode temp.	By reactor temp.	Electrode
Wire	D_2O	70	30.7	780	30.7	13.9	61	48	2.8	1.37	1.08	260
Wire	D_2O	275	31	0	31	0	58	35	58	1.87	1.13	260
Wire	D_2O	50	31	0	31	0	46	34	78.3	1.48	1.07	200
Wire	D_2O	50	31	0	31	0	43	31	12.8	1.38	1.00	210
	D_2O	100	45.7	0	45.7	0	53	55	64.7	1.15	1.19	275
	D_2O	20	44.6	0	44.6	0	36	50	1.2	0.82	1.13	190
Rod	D_2O	50	45.8	790	45.8	16.6	62	62	2.48	0.99	1.2	320
Wire	D_2O	50	46.6	772	46.6	14.6	60.4	60	1.44	0.99	1.3	320
	D_2O	50	47	0	47	0	55	55	1.17	1.28	1.43	315
Wire	D_2O	145	45.9	0	45.9		124	114	75.2	2.7	2.48	280
Wire	D_2	330	45.8	0	45.8		64	63	62.5	1.4	1.37	325
Wire	H ₂ O	20	46	0	46	0	46	70	8.2	1	1.09	250
Rod	D_2	400	49	770	49	12	66	70	6.5	1.08	1.15	330
	D_2	200	49	0	49	0	61	60	1.3	1.24	1.22	300
	D_2	300	25	0	25	0	41	30	6.5	1.64	1.2	250
Rod	D2	300	36	0	36	0	54	48	1.5	1.5	1.33	300
	D2	330	36	0	36	0	54	44	2.8	1.5	1.22	285
	D2	330	51.2	0	51.2	0	58	80	8	1.13	1.56	298
	H_2O	20	35.5	0	35.5	0	42	42	60	1.18	1.18	230
Wire	H_2O	150	15.6	825	0	15.6	4.3	10	16	0.28	0.66	45
Rod	H_2O	145	16	800	0	16	3.1	10	9.4	0.19	0.66	44

Table 1. Summary of recent successful tests with large reactor, palladium electrodes

References

Tadahiko Mizuno, Tadashi Akimoto, Tadayoshi Ohmori, Akito Takahashi, Hiroshi Yamada and Hiroo Numata, Neutron evolution from a palladium electrode by alternate absorption treatment of deuterium and hydrogen, *Jpn. J. Appl. Phys.* 40 (2001) L989–L991

[2] Tadahiko Mizuno, Tadashi Akimoto, Kazuhisa Azumi, Tadayoshi Ohmori, Yoshiaki Aoki and Akito Takahashi, Hydrogen evolution by plasma electrolysis in aqueous solution, *Jpn. J. Appl. Phys.* **44**(1A) (2005) 396–401.

[3] Tadahiko Mizuno, *Transmutation Reaction in Condensed Matter, Low-Energy Nuclear Reactions Sourcebook*, American Chemical Society symposium series 998, Jan Marwan and Steven B. Krivit (Eds.), 2008, pp. 271–294.