Direct Evidence for Nuclear Fusion Reactions in Deuterated Palladium

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ABSTRACT

Using our own "*in vacuo*" method with a heterostructure of deuterated Pd (Pd:D), we have succeeded in the first highly reproducible and "*in situ*" detection of ^{4}He production. The real time observation has been performed by high-resolution quadrupole mass spectroscopy (0.001 amu at 4 amu). The amount of ^{4}He gas produced was closely correlated with the evolution of excess heat, and it increased with the loading ratio of D to Pd. At the highest loading ratio of D to Pd, we have also observed T production by detecting HT. The amount of HT increased in the final stages of ^{4}He production. The system of H-loaded Pd (Pd:H) heterostructure, on the other hand, produced neither ^{4}He nor T. Furthermore, the energy spectrum of charged particles detected during these experiments has revealed that α particles with an energy of 4.5–6 MeV and protons with an energy of 3 MeV were emitted from the oxide surface of Pd. The amount, however, was extremely small in comparison with that of ^{4}He detected. These results indicate that a new class of nuclear fusion occurs in the Pd:D and Pd:H systems.
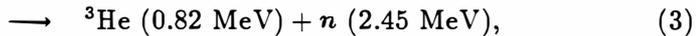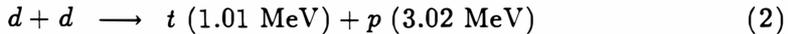
1. Introduction

After Fleischmann, Pons and Hawkins[1] claimed in 1989 that electrolyzing heavy water with a palladium cathode released energy from nuclear fusion at room temperature, much effort has been made to reinvestigate the possibility of the electrolytically induced nuclear fusion in condensed matter. Many researchers have recently reproduced the evolution of heat by electrolysis of D_2O, but none have yet proved that this heat is caused by nuclear fusion nor even obtained definite evidence for the so-called "cold fusion" in solids.

By the lack of neutrons consistent with the excess heat, Fleischmann *et al.* presumed that "the bulk energy release is due to a hitherto unknown nuclear process or processes," specificaly[2]

$$d + d \longrightarrow {}^4\text{He (76.0 keV)} + \text{photons/phonons (23.8 MeV)}. \qquad (1)$$

In comparison with the normal reactions

$$d + d \longrightarrow t \ (1.01 \ \text{MeV}) + p \ (3.02 \ \text{MeV}) \tag{2}$$

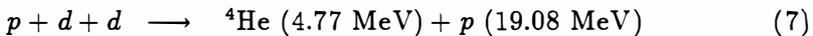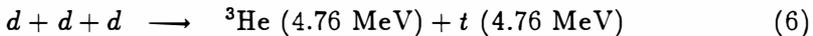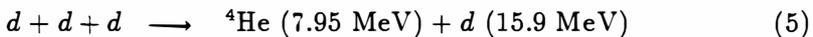$$\longrightarrow \ ^3\text{He} \ (0.82 \ \text{MeV}) + n \ (2.45 \ \text{MeV}), \tag{3}$$

this reaction has negligibly small probability (10^{-7}), and should be accompanied by an emission of 23.8-MeV γ-ray. Consequently, their suggestion of "cold fusion" has been profoundly controversial. Schwinger[3], however, supported their claim, and furthermore, suggested that, because of the decrease in the Coulomb barrier between hydrogen nuclei and because of the asymmetry of the $p + d$ situation in contrast to the symmetry of the $d + d$ situation, the reaction

$$p + d \longrightarrow \ ^3\text{He} \ (5.35 \ \text{keV}) + \text{photons/phonons} \ (5.49 \ \text{MeV}) \tag{4}$$
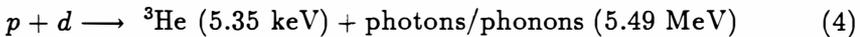
is more likely than that described by Eq. (1),

Other groups have suggested other mechanisms. Takahashi et al.[4] reinvestigated the experiment of Fleischmann et al., and not only found slight peaks near 2.45 MeV in the neutron energy spectra, but also found one-order-of-magnitude-weaker broad peaks at 4–6 MeV. They conjectured that multibody reactions such as

$$d + d + d \longrightarrow \ ^4\text{He} \ (7.95 \ \text{MeV}) + d \ (15.9 \ \text{MeV}) \tag{5}$$

$$d + d + d \longrightarrow \ ^3\text{He} \ (4.76 \ \text{MeV}) + t \ (4.76 \ \text{MeV}) \tag{6}$$

$$p + d + d \longrightarrow \ ^4\text{He} \ (4.77 \ \text{MeV}) + p \ (19.08 \ \text{MeV}) \tag{7}$$

etc. produced high energy deuterons (Eq. (5)), which then fused to produce neutrons with energies ranging from 4 to 6 MeV. Hagelstein suggested other multi-body mechanisms due to weak interactions[5].

Since the reality of cold fusion cannot be established simply by the observation that excess heat is released, it is necessary to determine the presence or absence of nuclear products. As shown in Eqs. (1)–(7), there are three kinds of possible nuclear products: (1) neutrons; (2) charged particles with high energies; and (3) ^4He, ^3He, or T compounds (such as T_2, HT or DT) in the gas phase.

With regard to the production of neutrons, a few groups have reported that neutron spectra show slight peaks at 2.45 MeV during the electrolysis of D_2O. These spectra, however, inevitably include background signals from space as well as from fissionable sources in the laboratories. The background contribution usually fluctuates so much that the observation of neutron emissions that are at most one order of magnitude greater than the averaged background has never given a definite answer. Anderson et al.[6], for example, found weak peaks near 2.45 MeV in background spectra without electrolysis.

Tritium measurement has encountered the same problem, because the heavy water used in the electrolysis experiments contains various amounts of contaminating T_2O. Although several groups have reported to have found large amounts of T when using radioactivity measurements to analyze the water after the electrolysis, their results have not yet persuaded sceptics.

Only the following two methods can produce evidence proving the presence or absence of cold fusion phenomena:

1. Mass spectroscopy for ^{4}He, ^{3}He and T in gas phase, with the background reduced to zero throughout all measurements. To investigate the correlation between the production of those gases and the excess heat, the measurement must be performed "*in situ*" and in real time. For instance, with a nominal mass 4 amu, there are 4 possible species: ^{4}He (4.00260 amu), HT (4.02388 amu) , D_2 (4.02820 amu), and DH_2 (4.02975 amu). It is therefore possible to distinguish between ^{4}He and the others by using a high-resolution mass spectrometer.

2. Energy spectroscopy of emitted charged particles, with the sensor under a pressure of less than 10^{-4} Torr. Background signals from cosmic rays can be zero within a tight chamber having sufficiently thick walls. The measurement must be also performed "*in situ*" in real time.

Even in the electrolysis measurements developed by Fleischmann *et al.*, mass spectroscopy can be used to analyze gas samples extracted from the closed electrolysis cell[7–9]. The existence of ^{4}He, ^{3}He, and T in the air and water, however, provides substantial background signals, so this analysis cannot give unambiguas evidence. Furthermore, real-time mass spectroscopic measurements are impossible in this kind of experiment. In fact, Morrey *et al.*[7] found that there was so much ^{4}He in the unused Pd that its production in the electrolyzed metal could not be established. The energy spectroscopy of charged particles is also difficult or impossible in the electrolysis type experiment.

We have therefore developed a new, "*in vacuo*," method[10–12] for inducing cold nuclear fusion in solids. This technique has enabled us to observe gigantic burst of neutrons and, at the same time, both the explosive release of D_2 gas and excess heat from deuterated Pd (Pd:D) plates and a biaxial plastic deformation of the samples[10]. We have for the first time reproduced the evolution of excess heat[11].

The key technology of this method is to form a layer of D accumulated at solid Pd:D surfaces by using hetrostructures, formed under He-free and T-free conditions, to control the out-diffusive transport of D atoms. One side of these Pd:D surfaces is covered with a thick Au-film that prevents leakage of D atoms from that side, and the other side of the surface is covered with an oxide (MnO_x, SiO_x or AlO_x) that provides a surface barrier to out-diffusing D atoms. A layer of D atoms therefore accumulates in the Pd near the Pd/oxide interface. The β phase formation at this interface expands the lattice two-dimensionally and then the Gorsky effect enhances the out-transport of D atoms toward the interface. As a result, a D-rich layer accumulates at the Pd surface.

The purpose of the present work is to demonstrate the results both for the high resolution mass spectroscopic analysis of released gases and for measuring the energy spectroscopy of emitted charged particles, using our "in vacuo" method. In Sec. 2, we explain the details of sample preparation and of the measuring technique. Section 3 presents experimental results showing the the first definite evidence of cold nuclear fusion. Section 4 summarizes our conclusions.

2. Experimental

Samples with a MnO_x/Pd:D(H)/Au heterostructure were prepared as follows.

First, after Pd plates ($3 \times 3 \times 0.1$ cm^3) with purity of 99.9 % were rinsed with a mixture of HCl and HNO_3 (3:1 by volume) for 10 seconds, an electron-beam evaporator was used to deposit a 200-Å-thick film of MnO_x on one surface of the Pd plates at room temperature.

Second, the samples were set in a stainless steel vacuum chamber (Vieetech Inc., 61.37 l), and annealed at 300–400 °C for 18–22 hours under vacuum (final pressure $< 5 \times 10^{-7}$ Torr) to completely degas the samples. With the heater kept on, 99.9% pure D_2 was introduced up to a pressure of 400 Torr. For some samples, H_2 gas or the mixture of D_2 and H_2 gases were used instead of pure D_2 gas. Then the temperature was decreased at -2°C/min to room temperature.

Third, after keeping the samples under the H_2, D_2, or D_2-H_2 mixture for 60 hours, the chamber was evacuated to less than 10^{-4} Torr and within 15 min., a 2000-Å-thick film of Au was deposited on the other surface of Pd plates. Note that these second and third procedures eliminate any chance of He or T being left in Pd lattices.

The samples were taken out of the vacuum chamber and weighted to the nearest 10 μg. The samples were also weighted after the measurement procedure, and since all D or H atoms in the Pd were completely out-diffused after the measurement, the average loading ratio of D or H to Pd could be estimated from the weight difference. This ratio varied between 30 to 60%. Two of the samples were then attached to a sample holder equipped with electrodes and Si-SSDs (solid state devices) and were set in the same vacuum chamber used for D loading. Measurements were started after reducing the chamber pressure to less than 10^{-5} Torr.

Measurement setup is shown schematically in Fig. 1. The chamber was evacuated by a turbomolecular pump (550 l/s). Part of gases around the samples was drawn through a 25-cm-long rambling stainless steel tube (1/8 inches in diameter) to the mass-sensing chamber equipped with one high-resolution quadrupole mass (Q-mass) sensor (Extrel: EXM25) and two independent low-resolution Q-mass sensors (Spectramass: Selector). This differential evacuation system made it possible to keep the pressure in the mass-sensing chamber one order of magnitude lower than that in the main chamber.

The signal from the high-resolution Q-mass sensor was analyzed with a controller (Extrel: C50): where the optimized resolution in the mass number at nominal mass 4 was 0.001 amu, and the sensitivity with which ion current was measured was 1 pA . Figure 2 shows the calibration data obtained after the measurements when the high-resolution Q-mass spectrometer was loaded with D_2 gas and with ^{4}He gas while no samples were in the main chamber. This figure shows that the D_2 gas used in this experiment did not contain detectable amounts of ^{4}He.

The system for measuring the energy spectrum of charged particles consisted of two independent sets made up of an Si-SSD (Canberra: PD-450-19-700-AM; active area= 4.5 cm^2, active thickness=700 μm), a preamplifier (EG&G Ortec: 142B), an amplifier (EG&G Ortec: 575A), and an AD converter (Canberra: 8713). Both sets were connected to a multichannel analyzer
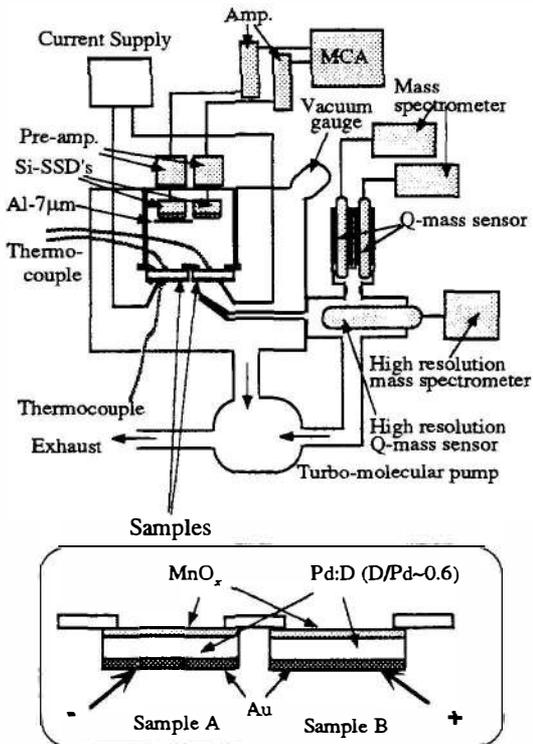
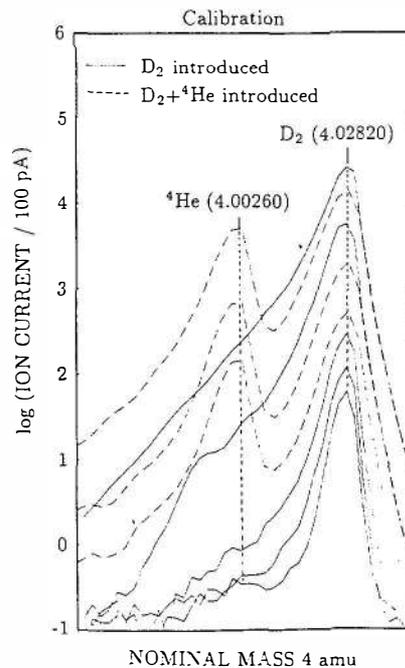Fig. 1: Schematic diagram of the measuring apparatus.

Fig. 2: Calibration data for high-resolution Q-mass spectrometer.

(Nuclear Data: 556-AIM), and both SSDs were 6.0 cm from the MnO_x surface of the Pd plates. One of the SSDs was covered by a 7.0-μm-thick piece of Al foil to identify the species of the charged particles. Time-resolved data from MCA was outputted to a workstation (DEC: VS3100-M76) running special software (Nuclear Data: GENIE system). The energy calibration was performed by using ^{241}Am (5.484 MeV) and ^{244}Cm (5.805 MeV). To avoid detecting background photons, the vacuum chamber was completely darkened throughout the measurement.

A constant current/voltage supplier (HP: 6032A) supplied electric current to the samples through tungsten needles. As shown in the inset of Fig. 1, two equivalent samples were reversely biased in order to find out whether the out-transport of D atoms in Pd was enhanced by electromigration. The sample surface temperature was measured by K-type thermocouples. Strain gauges were sometimes put on the Au surfaces, but never before attempts to detect ^{4}He production. This measurement setup thus provided a perfect system free of background ^{4}He.

3. Results and Discussion

Our previous papers[10,11] have shown that the following three events always occur chaotically within 1–3 hours of starting to pass electric current through the samples:

(1) Explosive release of gas from the samples, during which the ratio of partial pressure for molecules with mass 3 to that for molecules with mass 4 or 2 increases markedly.

(2) Evolution of excess heat (0.5–2 W for 10^3 seconds). This follows the explosive gas release.

(3) Uniform biaxial bending of the samples. This plastic deformation begins suddenly at the peak of gas release and it ends within 10 seconds.

We have also found that two equivalent samples (A and B) arranged as shown in the inset of Fig. 1 exhibit these phenomena. This means that the out-transport of D or H atoms in Pd lattices is due not to electromigration but to the temperature gradient perpendicular to the surface (heat of transport). Thus, the electric current turns out to work for heating the samples.

The present paper shows experimental results for four typical samples with $MnO_x/Pd:D(H)/Au$ heterostructures. Samples 1 and 4 were respectively made by immersing the Pd plates in D_2 and H_2 at 400 Torr; the loading ratio of D to Pd was 48 % for sample No. 1 and the loading ratio of H to Pd was 56 % for sample No. 4. Samples 2 and 3, on the other hand, were made by immersing plates in the mixtures of D_2 and H_2 gases ($D_2:H_2$ volume ratio=7:1 for No. 2; 1:1 for No. 3), each with a total pressure of 400 Torr. In each of the four measurements, two equivalent samples A and B configured as shown by the inset in Fig. 1 were heated by a continuous pulsed current (5 A/7 A at 50 Hz).

The results of time-resolved mass spectroscopic analysis of gases collected during experiment No. 1 (Pd:D) are shown in Fig. 3(a). This figure clearly shows that only D_2 is detected initially but a peak attributable to ^{4}He gradually appears. This is the first definite evidence for ^{4}He production because there is no background ^{4}He in this experimental method. In the final stages of ^{4}He production, a peak attributable to HT appears and increases with the time. Since there is also no background T in this system, this experiment also gives definite evidence for T production.

The mass spectra from experiments No. 2 (Pd:D/H) and No. 3 (Pd:H/D), Figs. 3(b) and 3(c), also show clear ^{4}He peaks at 4.00260. As shown in Fig. 4(a), this ^{4}He production is clearly correlated with the excess heat released by the sample B in experiment No. 3. The maximal intensity of the ^{4}He peak, however, as well as the total amount of ^{4}He produced, were lower for experiments No. 2 and No. 3 than for experiment No. 1. This indicates that the ^{4}He production decreases with the amount of D_2 loaded in the Pd.

The time-resolved mass spectra for experiment No. 4 (Pd:H), shown in Fig. 3(d), show only a single small peak at 4.02820 amu. This peak is due to D_2 contamination from the H_2 bottle or the inner wall of the chamber, or both. We can therefore conclude that Pd:H systems exhibit ^{4}He production nor T production. This figure also shows that there is no background ^{4}He in these experiments.
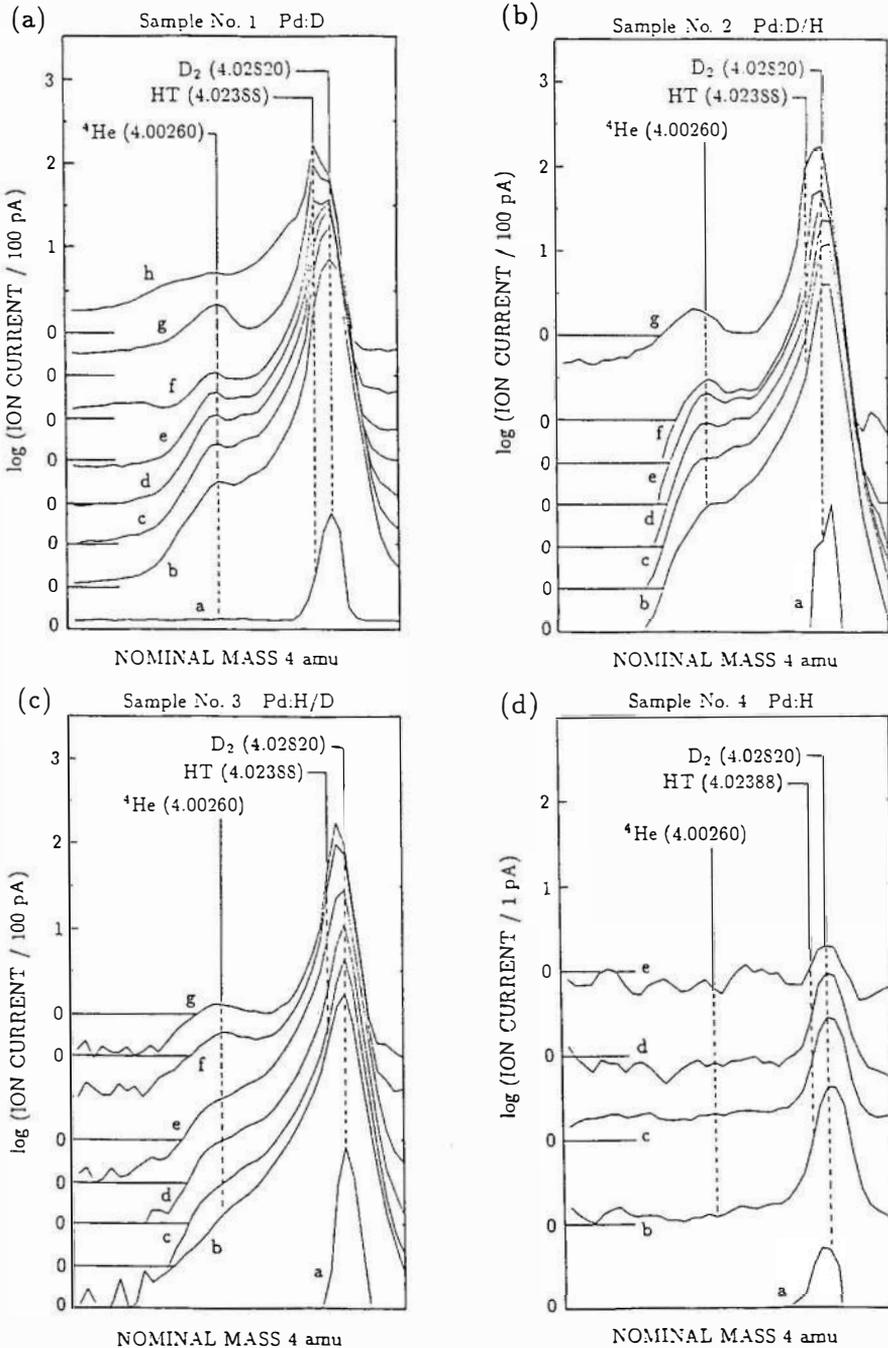
Fig. 3: The experimental results of mass spectroscopy at nominal mass 4 for released gases from (a) sample No. 1, (b) No. 2, (c) No. 3 and (d) No. 4, where each spectra from (a) to (h) were taken at every 72 min. for sample No. 1, 28 min. for No. 2 and 3, and 55 min. for No. 4.
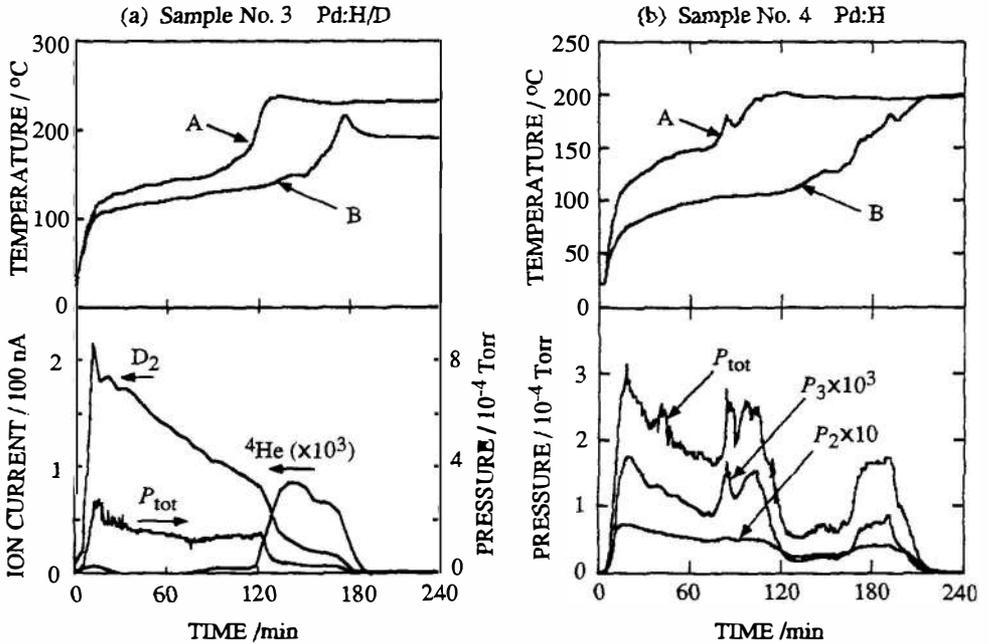
Fig. 4: Time dependence of the surface temperatures for the sample A and B, the total pressure, P_{tot}, of the main chamber, the ion current for D_2 and ^{4}He (or the pressure for mass 2, P_2, and mass 3, P_3) in the (a) measurement No. 3 and (b) measurement No. 4.

As shown in Fig. 4(b), however, the Pd:H system nevertheless chaotically produced excess heat in both the A and B samples. At those sudden increases in temperature, at 80–120 min. for sample A and at 160–200 min. for sample B, the ratio of P_3 to P_2 increases dramatically. Similar increases in P_3 have also been observed in experiments No. 1–3.

If the mass 3 gas were HD or H_3, the time dependence of the partial pressure P_3 should be proportional to P_2 and the pressure for D_2. Since the amount of T is negligible, as shown in Fig. 3(d), it is most likely that this dramatic increase in P_3 is due to ^{3}He production. To test Schwinger's conjecture, it will be necessary to use the high-resolution Q-mass system to observe the time-resolved mass spectra at nominal mass 3 amu.

While measuring the mass spectra described above, we simultaneously made two independent measurements of the energy spectrum of charged particles. It must be noted here that since the Si-SSD used in this experiment has a 700-μm-thick depletion layer, we can detect p, d, t, ^{3}He, and α emission at energies up to 10, 13, 14, 35, and 40 MeV, respectively. The amplifier and AD converter were set to detect, with a resolution of 7 keV, any charged particle of the energy up to 28 MeV.

Each sample spectrum was taken after starting the current injection, where the live time is 508 min. for No. 1 and 203 min. for No. 2–4. The sum is shown in Fig. 5. The spectrum shown in Fig. 5(a) has clear peaks at 3 MeV and a broad range of peaks from 4.5 to 6 MeV. These peaks probably correspond to

the peaks at 2.2 MeV and from 3.5 to 4.5 MeV in Fig. 5(b). From this energy loss resulting from the 7μm of Al foil, we can conclude that the emission at 3 MeV is attributable to protons and that the peaks around 4.5–6 MeV are attributable to α or ^{3}He particles. Our data shows that that there is no signal above 10 MeV.

The proton emission at 3 MeV is probably due to the reaction specified in Eq. (2). The origin of the higher energy emission, however, cannot be stated with certainty, because the background itself has an extremely low probability of containing α emissions at an energy of 5–6 MeV. Since January 1991, we have measured the backgound data of charged particle emissions 50 times, each with a live time of 810-812 min. Two of these sets of background data showed α particle emission with an energy of 5.7-5.9 MeV. This is perhaps due to fissionable sources in the stainless steel. The 4.5- to 6-MeV emission shown in Fig. 5, however, is almost ten times the highest background levels. More detailed and lengthy work will be needed to clarify the origin of this 4.5–6 MeV emission of α or ^{3}He.
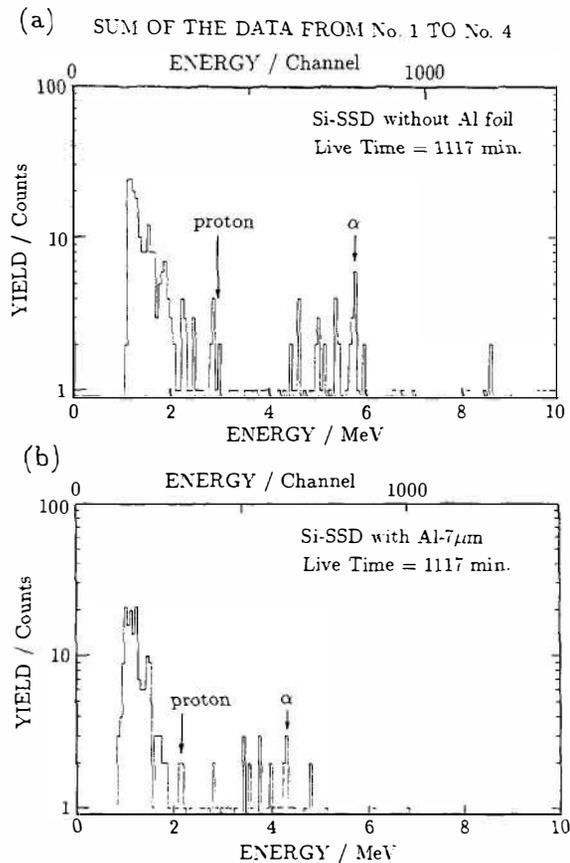

Fig. 5: The experimental results of energy spectroscopy for charged particles detected by Si-SSD (a) without Al-foil and (b) with Al-foil (thickness=7μm), where ths sum of the data from sample No. 1 to No. 4 are shown.

4. Conclusion

We have for the first time succeeded in detecting ^{4}He production "*in situ*" and with high reproducibility. Our "*in vacuo*" method gives the first definite evidence for the reality of "cold nuclear fusion" in solids. The real-time observation of fusion products has been performed by quadrupole mass spectroscopy with high resolution (0.001 amu at 4 amu). The amount of ^{4}He gas produced was closely correlated to the evolution of excess heat, and it increased with the loading ratio of D to Pd.

At the highest loading ratio of D to Pd, we also detected HT. The amount of HT produced increases in the final stages of ^{4}He production. The system of hydrogen loaded Pd (Pd:H), on the other hand, produces neither ^{4}He nor T.

This system nonetheless evolves the excess heat at the same time that 3-amu gas is explosively released.

Furthermore, the energy spectrum of charged particles emitted during these experiments has revealed that α particles with an energy of 4.5–6 MeV and protons with an energy of 3 MeV were released from the oxide surface of Pd. The amount of these particles, however, was extremely small in comparison with the amount of ^{4}He detected. These facts suggest that the reaction $d + d \longrightarrow\ ^4$He + photons/phonons is strongly enhanced in the system Pd:D, and that the reaction $p + d \longrightarrow\ ^3$He + photons/phonons is strongly enhanced in the system Pd:H/D.

Acknowledgments

References

1. Fleischmann, M.*et al.*, 1989, J. Electroanal. Chem., **261**, 301.
2. Walling, C. and Simons, J., 1989, J. Phys. Chem., **93**, 4693.
3. Schwinger, J., 1990, Z. Phys. D, **15**, 221.
4. Takahashi, A. *et al.*, 1991, AIP Conf. Proc., **228**, 323.
5. Hagelstein, P. L., 1991, AIP Conf. Proc., **228**, 734.
6. Anderson, R. E. *et al.*, 1991, AIP Conf. Proc., **228**, 43.
7. Morrey, J. R., 1990, Fusion Tech., **18**, 659.
8. Bush, B. F. *et al.*, 1991, J. Electroanal. Chem., **304**, 271.
9. Liaw, B. Y. *et al.*, 1992, J. Electroanal. Chem., to be published.
10. Yamaguchi, E. and Nishioka, T., 1990, Jpn. J. Appl. Phys., **29**, L666.
11. Yamaguchi, E. and Nishioka, T., 1991, AIP Conf. Proc., **228**, 354.
12. Yamaguchi, E. and Nishioka, T., 1991, Elsevier Studies in Applied Electromagnetic in Materials, **3**, 21.