

Elemental Analysis of Pd Complexes: Effects of D₂ Gas Permeation

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Elemental analysis of Pd complexes, which consist of a thin Pd layer, alternating CaO and Pd layers and bulk Pd, is described, after subjecting the Pd complexes to D₂ gas permeation. The Pd complex was located in a vacuum chamber and the elemental analysis was performed using an X-ray photoelectron spectroscopy (XPS) apparatus mounted on the chamber. When Cs was added on the surface of a Pd complex, Pr emerged on the surface while Cs decreased after the Pd complex was subjected to D₂ gas permeation at 343 K and 1 atm for about one week. In the case of adding Sr on the surface, Mo emerged on the surface while the added Sr decreased after D₂ permeation for about two weeks. All the phenomena were reproduced qualitatively. The isotopic composition of the detected Mo exhibited characteristics indicating an isotopic abundance of Sr rather than the natural abundance of Mo. [DOI: 10.1143/JJAP.41.4642]

KEYWORDS: D₂ gas, permeation, Pd, Pd complex, thin film, Cs, Pr, Sr, Mo, isotopic composition, X-ray photoelectron spectroscopy

1. Introduction

The palladium–hydrogen system has been the most widely investigated system among the metal hydrides since it has the following features: (1) Pd exhibits high solubility and diffusivity of hydrogen; Pd and Pd alloys have been used as purification membranes of hydrogen.¹⁾ (2) Pd hydride is a super-conducting material²⁾ that attracted the attention of researchers in the 1970s–80s. However, interest in Pd hydride as a practical super-conducting material has declined because other materials that show higher transition temperatures have been discovered. (3) In the palladium–deuterium system, several researchers, mainly in recent years, have claimed low-energy transmutations,^{3–8)} although a definite theoretical explanation has not been provided.

We have been investigating the low-energy transmutations observed in the palladium–deuterium system. Our experimental results thus far have led to the assumption that necessary conditions which give rise to the phenomena should satisfy the following criteria:⁷⁾ (i) existence of a low work function material, typically CaO, near the Pd surface; (ii) sufficient diffusion flux of deuterium, (iii) sufficient D on the Pd surface. To check the validity of these assumptions, we have established an experimental method characterized by “permeation of D through Pd complex” as shown in Fig. 1. D₂ gas molecules cause a dissociative chemisorption on the D₂ gas side of the Pd complex, and then D atoms diffuse toward the vacuum side, where they combine and are released as D₂ gas. We assume that the above necessary conditions are satisfied on the near surface of D₂ gas side of the Pd complex.

The Pd complex is composed of a Pd thin film, alternating CaO and Pd layers and bulk Pd as shown in Fig. 2(a). This composition was determined based on condition (i) above since the work function of CaO is relatively low (1.6 eV–1.86 eV).⁹⁾ Permeation of deuterium through the Pd complex is attained by exposing one side of the Pd complex to D₂ gas while maintaining the other side under vacuum conditions. We assume that D₂ gas permeation satisfies the conditions (ii) and (iii) above. Although the definite physical implica-

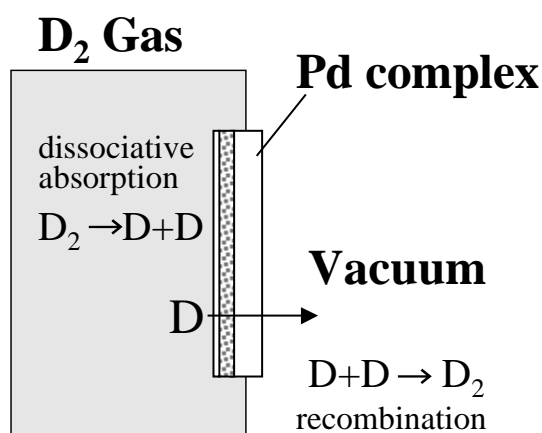


Fig. 1. D₂ gas permeation through the Pd complex.

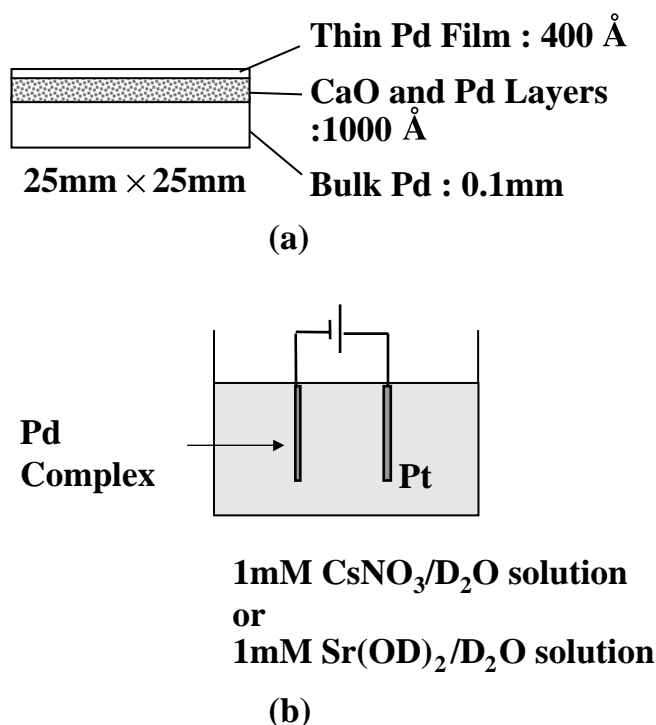


Fig. 2. (a) Structure of Pd complex, (b) Method of depositing Cs or Sr on the surface of Pd complex.

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tions of the above conditions are unclear at present, it is a fact that the experimental observations described in this paper are consistent with the assumptions.

Almost all the other researchers in this field are conducting experiments involving the electrolysis of D_2O or H_2O with metal as a cathode³⁻⁸⁾ and they usually analyze the transmuted products on the cathode metal by taking it out of the experimental apparatus after electrolysis. Our experimental method described above is superior in that it discriminates the products from contamination because we analyze the products in vacuum during the experiments without moving the products.

2. Experimental

Figure 2(a) shows a schematic view of the Pd complex cross section. The Pd complex is composed of a Pd thin film, alternating CaO and Pd layers and bulk Pd. Test pieces were prepared by the following procedures. A Pd plate (with purity up to 99.99%; Tanaka Kikinzoku Kogyo K.K.) was washed with acetone and annealed in vacuum ($<10^{-7}$ Torr) at 900°C for 10 h. It was then cooled to room temperature in furnace and washed with D-based aqua regia (with purity up to 99.9%), which consisted of a mixed liquid of DCl/D_2O and DNO_3/D_2O , to remove impurities on the surface of the Pd plate. The surface of the plate was covered by layers of CaO and Pd (1000 Å) which were obtained by five times alternately sputtering 20-Å-thick CaO and 180-Å-thick Pd layers. The thicknesses of the layers were measured by a crystal thickness monitor. Then a 400-Å-thick Pd layer was sputtered on the surface of the CaO and Pd layers. The thickness of a 400 Å is chosen because it is sufficient to form a bulk layer. These processes are performed by Ar ion beam sputtering, which is a process whereby material is sputtered from a target with an Ar ion beam and deposited onto a substrate. The samples are sputtered at room temperature and the voltage and current of the Ar beam are 1 kV and 20 mA, respectively. Films deposited by ion beam sputtering exhibit the property of low contamination due to the low background pressure that leads to reduced gas incorporation.

After forming a Pd complex, Cs or Sr was deposited on the surface of the thin Pd layer. Cs or Sr atom was deposited by applying an electric field to 1 mM $CsNO_3$ (with purity up to 99.9%) or $Sr(OD)_2$ (with purity up to 99%) solution as shown in Fig. 2(b). A Pt wire (with purity up to 99.9%) was utilized as a counter-electrode. A 1 V negative voltage was applied to the Pd complex for 10 s. The value of 1 V was chosen to suppress D_2 and O_2 gas generations by electrolyzing the $CsNO_3$ or $Sr(OD)_2$ solution.

Figure 3(a) shows a schematic of the experimental apparatus. One advantage of this newly developed apparatus is that we can analyze the surface of a Pd complex sample by X-ray photoelectron spectroscopy (XPS) without removing it from the vacuum chamber, thereby preventing contamination of the test sample. XPS is a surface analysis technique which is used to investigate the chemical composition of surfaces by irradiating a sample with monoenergetic soft X-rays and analyzing the energy of the photoelectrons. Another advantage is that we can detect the time dependence of the elements on the Pd complex surface. Repeating D_2 gas permeation and XPS analyses, we can obtain information on time variation of the detected elements.

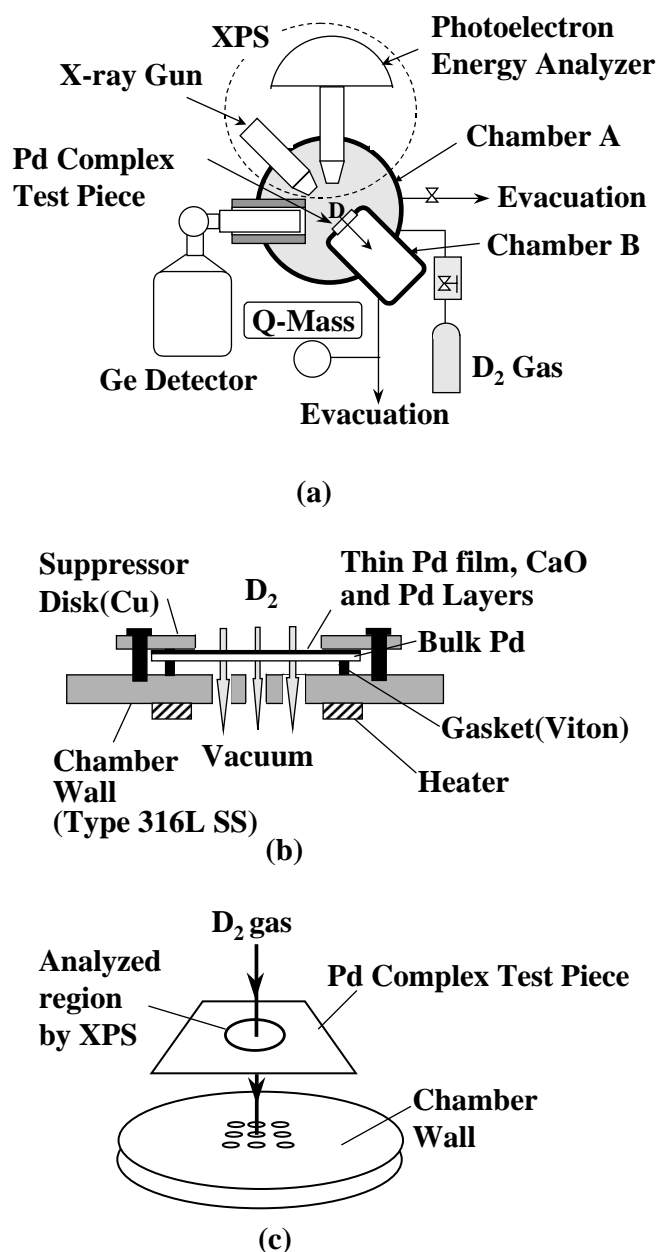


Fig. 3. (a) Experimental apparatus, (b) Schematic of test setup in the vicinity of Pd complex test piece, (c) Path of D_2 gas flowing through Pd complex test piece and chamber wall.

The apparatus consists of two chambers, A and B, an X-ray gun, an electrostatic analyzer for XPS, a mass spectrometer and a Ge semiconductor detector. The mass spectrometer is installed for analyzing the D_2 gas permeating the Pd complex. The Ge semiconductor detector is mounted for the detection of gamma rays. The X-ray gun is non-monochromatic and $Al-K \alpha$ is utilized. The power is 200 W, voltage is 10 kV and current is 20 mA. The electrostatic analyzer is equipped with a five-stage electrostatic lens and the pass energy is 100 eV. The X-ray gun and the electrostatic analyzer were made in our laboratory and the energy was calibrated using Au.

One chamber (Chamber A) is filled with D_2 gas, and the other chamber (Chamber B) is evacuated by a turbomolecular pump. These two chambers are divided by a Pd complex test piece. D_2 gas is supplied at 1 atm on the Pd film side of the test piece and dissolves in D atoms at the surface.

The D atoms intrude into the Pd thin film and diffuse through the Pd complex and then reach the surface of the bulk side, where they combine and are released as D₂ molecules.

Figures 3(b) and 3(c) illustrate magnified views of an installed test piece and D₂ gas flow. The thin Pd film (400 Å) side of the Pd complex test piece is exposed to D₂ gas and the test piece is fixed by a suppressor disk, a gasket (Viton) and a chamber wall made of Type 316L SS as shown in Fig. 3(b). The stainless steel chamber wall has several small holes that connect the D₂ gas side with the vacuum side. D₂ gas passes through the Pd complex test piece as shown in the Figs. 3(b) and 3(c). The total surface area of the holes is 1.0 cm². A heater is installed under the chamber wall and maintains the Pd temperature.

XPS analyses are performed with the following assumptions: 1) Intensities of incoming X-rays and the irradiated region are constant. 2) The analyzed region is a circle 5 mm in diameter and 20 Å in depth. The depth corresponds to the average path length of photoelectrons. 3) Atomic concentration of a detected element is calculated by the ratio of Pd peak intensity and the ionizing cross section of the element. The cross section at 1486 eV¹⁰⁾ is used because the target of the X-ray gun is Al.

The experimental procedure is as follows. The vacuum chambers are baked sufficiently before the experiments. A Pd complex test piece with added Cs or Sr is mounted on a flange in an air environment and then introduced into the vacuum chamber. Step 1: Both of the vacuum chambers are evacuated by a turbo-molecular pump and the test piece is heated to 70°C. The ultimate pressure in the vacuum chamber was about 3×10^{-8} Torr. Step 2: The surface of the test piece is analyzed by XPS to confirm that no other elements on the surface of the Pd test piece are detected except the given element (Cs or Sr), Pd and C. As is widely recognized in the field of surface science, a small amount of C is usually detected on a metal surface even in the vacuum chamber. Step 3: Chamber A is filled with D₂ gas up to 1 atm so that deuterium atoms permeate from this side to Chamber B behind the test piece. Step 4: After a certain period (from two days to one week) of deuterium diffusion through the Pd complex test piece, the D₂ is evacuated from Chamber A and the surface of the Pd test piece is analyzed by XPS in the chamber. New elements that did not exist on the test piece at the beginning of the experiment can be detected. Usually this process is repeated a few times to observe the time dependence of the given or newly generated elements. Step 5: The heater is turned off and all the chambers are filled with 1 atm N₂ gas. The test piece is removed from the chamber and its surface is analyzed by secondary ion mass spectrometry (SIMS). SIMS is a surface analysis technique in which energetic primary ions impact the surface and generate secondary ions, which are subsequently mass-separated and detected. SIMS is capable of analyzing all the elements with isotopic discrimination. In this study, we analyzed the Pd complex test piece after an experiment using SIMS to investigate the isotopic compositions of the detected elements. Only surface isotopic compositions were analyzed; we did not measure their depth profiles. SIMS analysis was performed by Probian Analysis, Inc. in France using the Physical Electronics 6600.

3. Results

Experimental results are shown in Figs. 4–9. During these experiments, we did not observe statistically significant gamma ray emission and gaseous products exceeding the detection limits (about 1ppm) of the mass spectrometer.

Experimental results obtained for test pieces deposited with Cs are shown in Fig. 4. Figure 4(a) shows the time dependence of the number of Cs and Pr atoms. Results for two runs are shown as examples. No Pr was detected at the beginning of the experiments. The number of Cs atoms decreased while that of Pr atoms increased with increasing experimental time. The amount of deuterium permeation was proportional to the elapsed time. At 120 h, the number of Pr atoms exceeded that of Cs atoms as shown in Fig. 4(a). Similar results were obtained in four other runs, although the initial number of Cs atoms was smaller than that in the Fig. 4(a). The reproducibility of these experiments is thought to be good.

The time variations in XPS spectra for the first run are shown in Figs. 4(b)–4(d) for Cs, Pr and Pd, respectively. All the elements are identified by two binding energy peaks¹¹⁾ of 3d_{5/2} and 3d_{3/2}. In Fig. 4(c), the peak ratio of 3d_{5/2} to 3d_{3/2} should be 3 : 2,¹¹⁾ however, it seems the two peaks are almost the same. The atomic ratio of Pr to Pd is a few percent and the major element is Pd. Therefore background signals derived from the photoelectrons of Pd–3d or 3p seem to influence the peak ratio of Pr. The error for Pr 3d_{5/2} and 3d_{3/2} is supposed to be a few cps. A peak between Pr 3d_{5/2} and 3d_{3/2} can be recognized around 939 eV. This is assumed to be derived from a shake-up or shake-off plasmon. It is evident that the two Cs peaks decrease corresponding to the increase in Pr peaks. On the other hand, XPS peaks for Pd do not change. This demonstrates that there is no problem with the stability of the obtained XPS spectra. Figure 4(e) shows a wide-range spectrum for the first run. Only Pd and C can be seen on the wide spectrum because the atomic ratios of Cs and Pr are a few percent. As mentioned earlier, C is usually detected on the metal surface even in the vacuum chamber.

In contrast, bulk Pd and the thin Pd film test piece with Cs deposition, which does not belong to a Pd complex, do not exhibit anomalous phenomena. All experimental conditions were the same as those of the Pd complex (Pd/CaO/Pd) experiments except for the formation of CaO and Pd layers. The annealing, etching by aqua regia, formation of a 400-Å-thick Pd thin film and deposition of Cs were performed in the same manner. Time variation of the Cs atom number, XPS spectra for Cs and Pr are given in Fig. 5. It is clear Cs did not change and Pr never appeared. This is quite reasonable in the sense that D₂ gas permeation usually does not affect the composition of the elements on the Pd surface. This experiment indicates that a CaO and Pd complex layer is essential for observing the decrease in Cs and the increase in Pr.

As a comparative study, we used H₂ gas instead of D₂ gas. The experimental results are shown in Fig. 6. In this case, only the gas is different; the other experimental conditions are all the same as Fig. 4. We also find that Cs did not change and Pr never appeared. This suggests that deuterium is a necessary factor for observing the anomalous behavior of Cs and Pr.

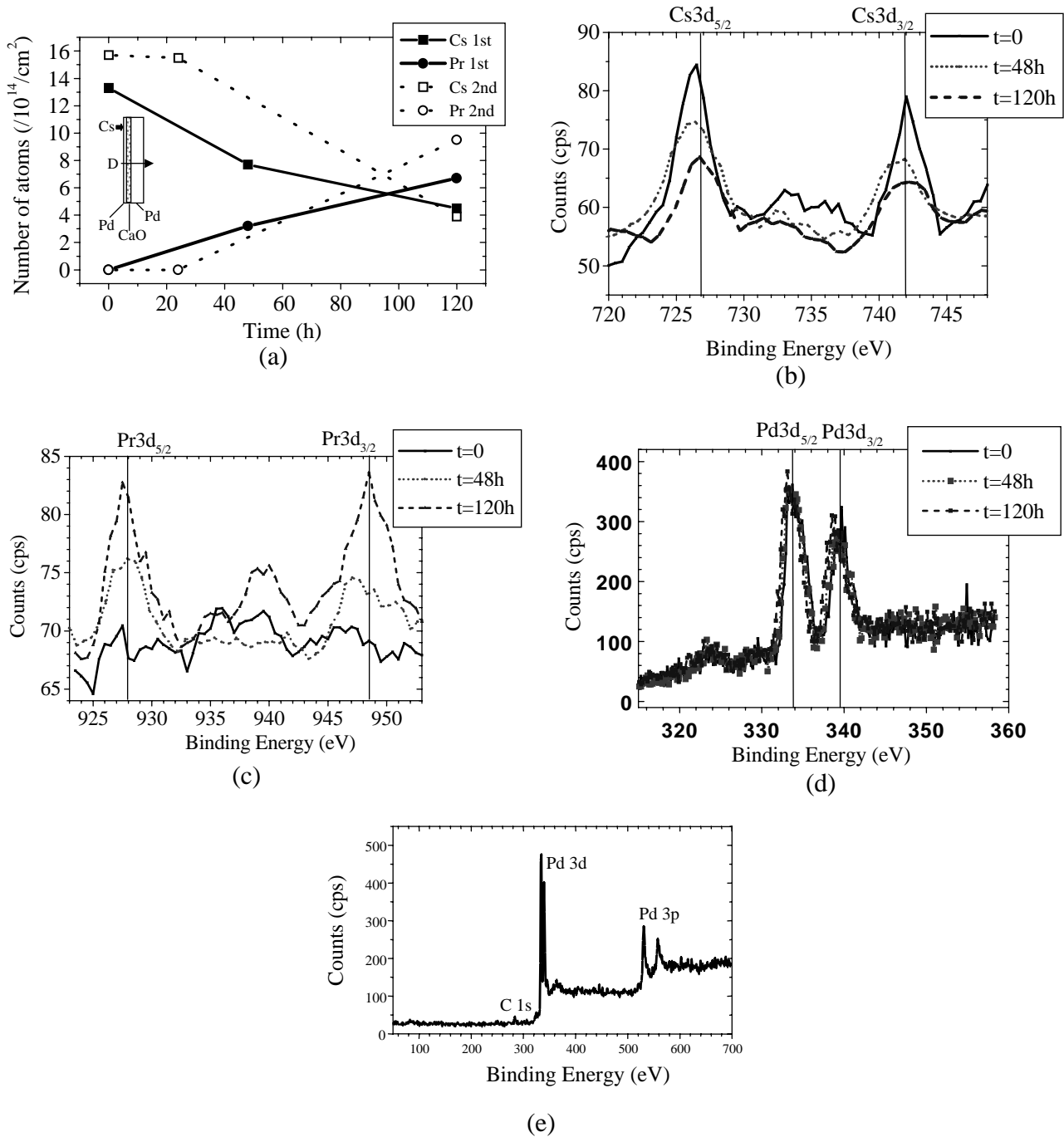


Fig. 4. Experimental results obtained by D₂ gas permeation through Pd complex (Pd/CaO/Pd) deposited with Cs: (a) Time variation in number of Cs and Pr atoms (number of atoms per cm²), (b) XPS spectrum of Cs for experiment run #1, (c) XPS spectrum of Pr for experiment run #1, (d) XPS spectrum of Pd for experiment run #1, (e) Wide-range XPS spectrum for experiment run #1.

The experimental results for Pd complex test pieces with added Sr are shown in Fig. 7. We observed that Sr decreased while Mo increased with increasing experimental time. Figure 7(a) shows the time variation of the number of Sr and Mo atoms. Experiments were performed three times and all the data are plotted in Fig. 7. At the beginning of the experiments, no Mo atoms were detected, however, Mo atoms increased gradually while Sr decreased correspondingly. It should be noted that runs for Sr are required to be longer to convert Sr to Mo than in the case of Cs to Pr. As can be seen in Fig. 7, qualitative reproducibility of is good. The time variations of XPS spectra for the first run are shown in Figs. 7(b)–7(d) for Sr, Mo and Pd, respectively. All

the elements are identified by two binding energy peaks¹¹⁾ in a similar way as for the Cs experiments. It is clear that the Sr peaks decrease corresponding to the increase in Mo peaks. The peaks of Pd did not change as shown in Fig. 7.

Figure 8 shows the results of the test piece devoid of CaO and Pd layers. All conditions of this comparative experiment were the same as those of the Pd complex (Pd/CaO/Pd) experiments except for the formation of CaO and Pd layers. We can see that Sr did not change and Mo was not detected. If we perform an experiment with a test sample devoid of CaO, we cannot observe any changes of elements on the surface of the sample. The CaO and Pd layers are considered to be an important factor for observing the decrease of Sr

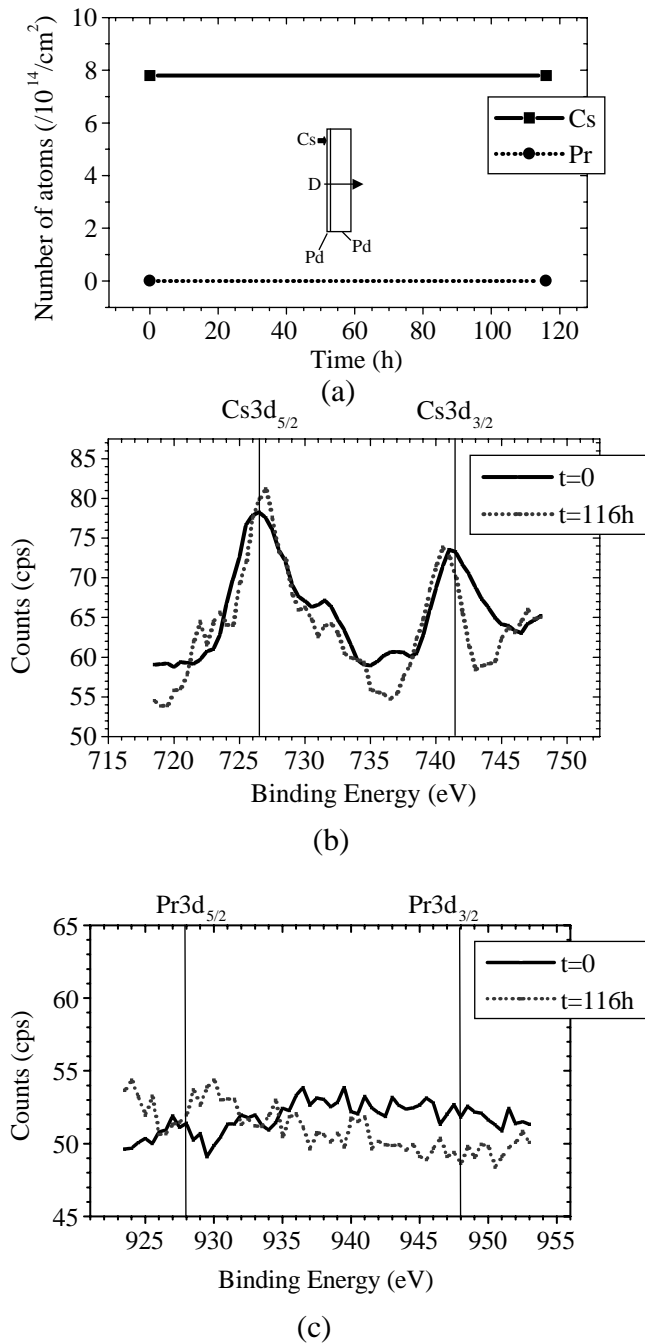


Fig. 5. Experimental results obtained by D_2 gas permeation through thin film and bulk Pd with added Cs: (a) Time variation in number of Cs and Pr atoms, (b) XPS spectrum of Cs, (c) XPS spectrum of Pr.

and the increase of Mo; this is the same as the case of Cs.

The authors performed a comparative experiment using H_2 gas for a Pd complex test piece with added Sr. The elements on the surface of the Pd complex test piece did not increase or decrease as expected. Since all the other experimental conditions except H_2 gas are the same as those of Fig. 7, it is suggested that deuterium is a necessary factor for observing the decrease in Sr and increase in Mo.

In order to investigate the isotopic composition, the samples after the experiment were analyzed by SIMS. Figures 9(a)–9(c) show the results of SIMS analysis for the three samples described in Fig. 7. The primary ions of the SIMS analysis were O_2^+ with the energy of 5.5 keV and the intensity of 30 nA. The area analyzed by SIMS was $150 \mu m$

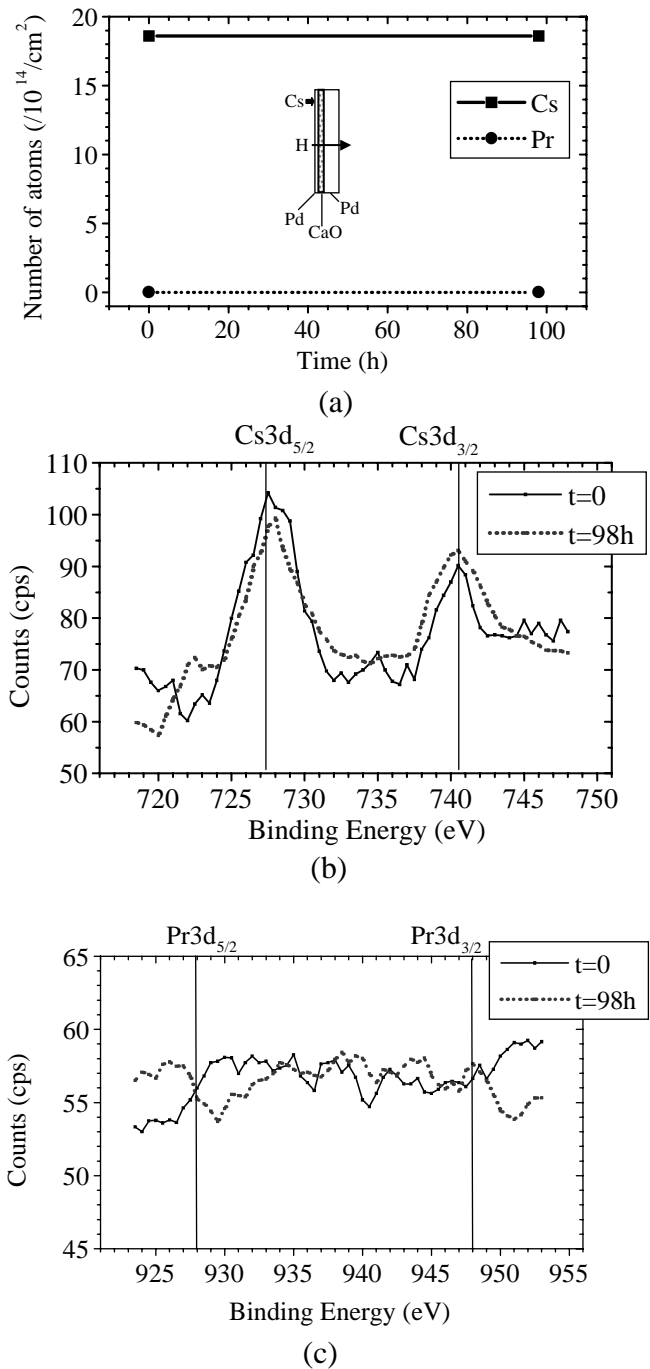


Fig. 6. Experimental results obtained by H_2 gas permeation through Pd complex (Pd/CaO/Pd) with added Cs: (a) Time variation in number of Cs and Pr atoms, (b) XPS spectrum of Cs, (c) XPS spectrum of Pr.

in diameter and mass resolution was 300. The intensities of mass number 96 were the largest for each sample, although the intensities were different. Figure 9(d) shows the results of SIMS analysis for a test piece with added Sr, however, no D_2 gas permeated through it. Signals arising from Mo impurity and organic compounds in Pd can be seen. The SIMS mass spectrum for a Mo layer (400 \AA thickness) which was deposited on a Pd disk is shown in Fig. 9(e). This spectrum reveals the natural abundance of Mo. Comparing these figures, we can easily recognize that the isotopic compositions of the detected Mo in Figs. 9(a)–9(c) are different from the natural isotopic abundance of Mo.

The difference in the ion intensities of the three samples is

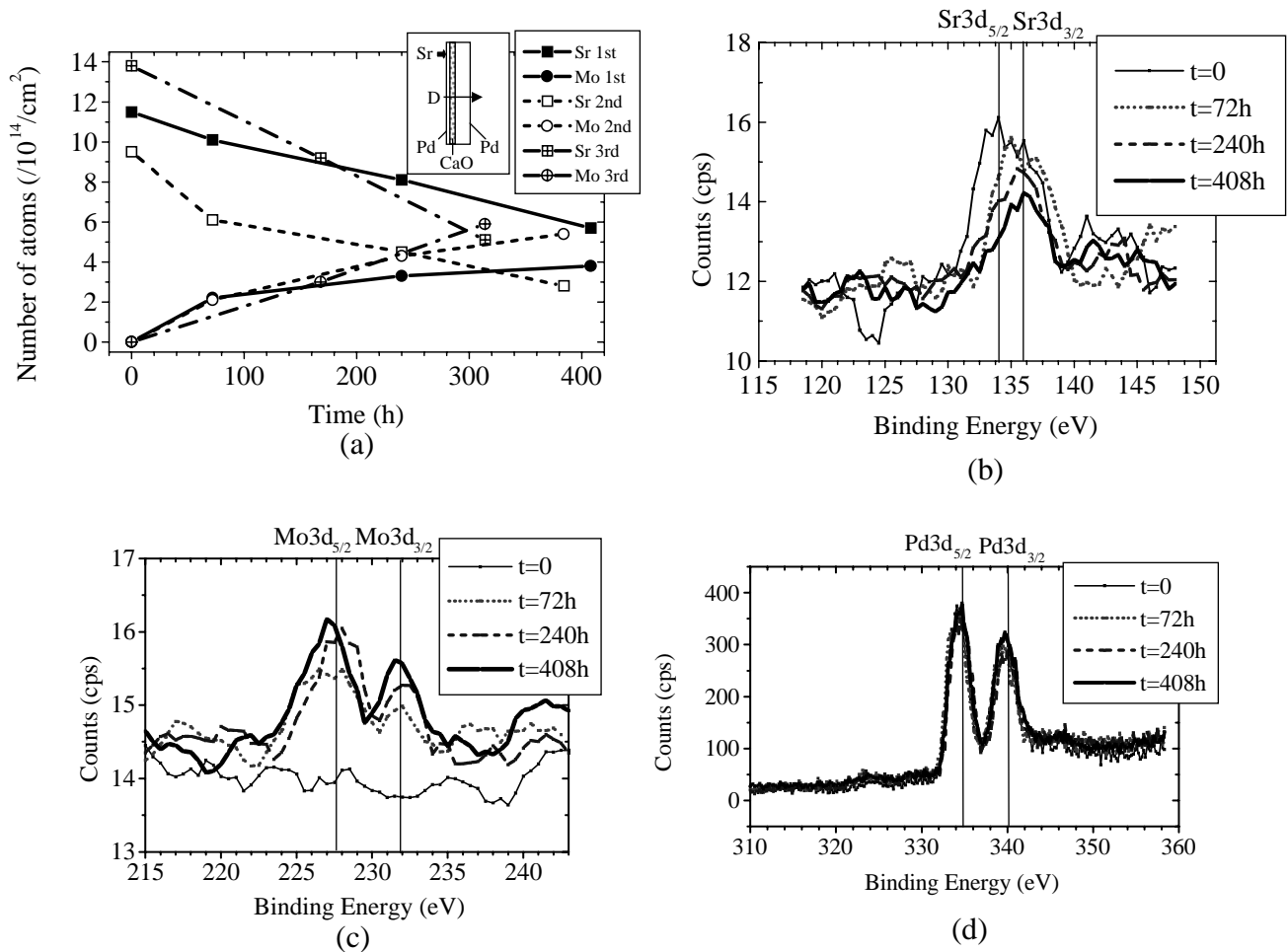


Fig. 7. Experimental results obtained by D_2 gas permeation through Pd complex (Pd/CaO/Pd) with added Sr: (a) Time variation in number of Sr and Mo atoms (number of atoms per cm^2), (b) XPS spectrum of Sr for experiment run #1, (c) XPS spectrum of Mo for experiment run #1, (d) XPS spectrum of Pd for experiment run #1.

presumed to exist that Sr and Mo did not distribute uniformly. The areas analyzed by SIMS and XPS are $150 \mu\text{m}$ and 5 mm in diameter, respectively. Therefore the signal analyzed by XPS is averaged over a large area, however the signal analyzed by SIMS is sensitive to the location of the analysis. If the distribution of Mo is not uniform, the result of SIMS analysis depends on the analyzed location. As we did not perform depth analysis of Mo, the dependence of isotopic compositions is not clear.

The SIMS analyses were performed by the "offset voltage technique"¹²⁾ to suppress the effects of molecular ions on mass spectra. The offset voltage technique is based on the fact that the kinetic energy distribution of atomic and molecular ions is entirely different and the distribution of atomic ion energy is broader than that of molecular ions. If we filter the central part of the distribution by applying a certain offset voltage, all signals decrease, however, the signal of molecular ions decreases much more than that of atomic ions. Therefore we can select only the signals of atomic ions. In our case, an 80 V offset voltage was applied. As for the formation of molecular ions, we should consider D, H, C, O, Pd and Sr; however, it is very difficult to form a mass number 96 by combining these elements. Moreover, Mo atoms were detected by XPS; it is natural to consider that almost all of the signal of mass number 96 can be

attributed to ^{96}Mo .

4. Discussion

The experimental results shown in Figs. 4–6 are summarized as follows: Cs atoms decrease and, correspondingly, Pr atoms emerge on the surface of the Pd complex by permeation of D_2 gas through it. These results were obtained six times and reproducibility was good. However, when we eliminate CaO and Pd complex layers from the Pd complex, or when we let H_2 gas permeate through the Pd complex, we observed no decrease in Cs and no emergence of Pr. Is it possible to interpret the phenomenon based on the established chemical or physical processes? The authors do not recall any processes other than nuclear transmutation; of course, there should be careful consideration of the possible contamination of test pieces and D_2 gas from the environment. The contamination issue will be discussed later in this paper. If the observed phenomenon is associated with nuclear transmutation, the following expression can be derived, as Cs and Pr each have only one isotope.



It should be noted that the mass number and the atomic number increased by 8 and 4, respectively, in this expression. It is natural to consider that deuterium plays an

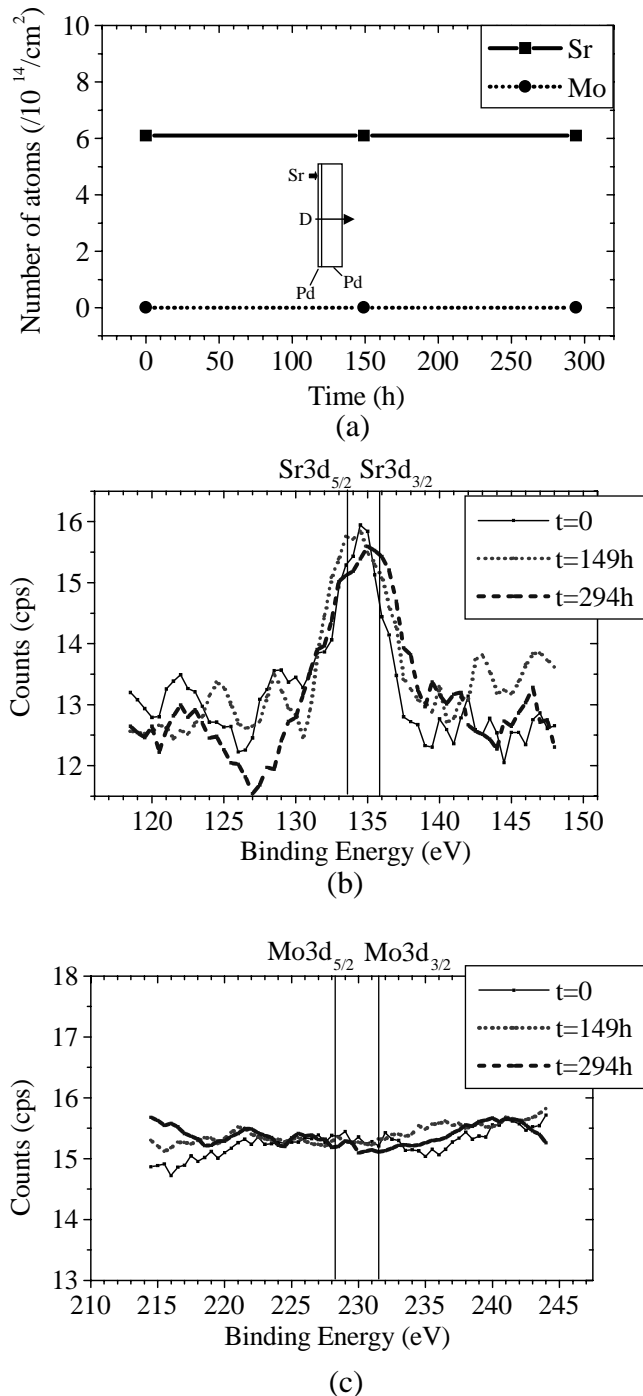


Fig. 8. Experimental results obtained by D₂ gas permeation through thin film and bulk Pd deposited with Sr: (a) Time variation in number of Sr and Mo atoms, (b) XPS spectrum of Sr, (c) XPS spectrum of Mo.

important role in this phenomenon. This is due to the fact that Cs atoms decrease and, correspondingly, Pr atoms emerge on the surface of the Pd complex as D₂ gas permeates through it.

The released energy for eq. (1) can be estimated by mass defects if we postulate that four deuterons react with Cs. About 50.5 MeV should be released per reaction. Figure 4(a) shows that there exist about 10^{14} atoms of Pr at the surface of the test piece because the reaction surface area is 1.0 cm^2 . Therefore released energy can be estimated to be 800 J. The average power is calculated to be 1.8 mW when the reaction time is assumed to be 120 h. Even if 1.8 mW were released

as heat during the experiment, we could not detect or notice the released heat.

We now discuss the Sr experiments. As indicated in Fig. 7, more permeating time is necessary to convert Sr into Mo than Cs experiments. In other words, Cs is easier to change than Sr. It might be presumed that the cross sections in this low-energy transmutation process depend on the type of nuclei. If we postulate the existence of this low-energy transmutation process it might be predictable. The observed results are expressed by the following expression because Sr and Mo have several isotopes.



Figures 10(a) and 10(b) show the relationship of mass numbers between given Sr and detected Mo. The characteristic feature of the detected Mo is that mass number 96 is the largest of the isotopes of Mo for all three cases of the Sr experiments. On the other hand, the major isotope of Sr is mass number 88. This fact suggests that the following expression can explain the results of SIMS analysis.



It is also noted that the mass number and the atomic number increased by 8 and 4, respectively, in this expression, as is the case with Cs. The released energy for eq. (3) is estimated to be 53.5 MeV per reaction. Therefore we cannot detect the released heat during experiments, as is the case with Cs.

Let us discuss the contamination issue. The first point is that the XPS analysis of the nuclear products is performed in the vacuum chamber. We do not expose the Pd complex test pieces to an air environment during experiments, and we monitor pressure in the chamber. Monitoring pressure enables us to detect any gaseous contaminants invading into the chamber, as the test apparatus is located in a clean room where temperature and humidity are kept constant.

The second point is that since the detected material, Pr, belongs to rare earth elements, it is difficult to assume that Pr accumulated on the Pd complex test pieces by an ordinary process. The purity of the used D₂ gas is over 99.6% and most of the impurity is H₂. The other impurities detected by a mass spectrometer are N₂, D₂O, O₂, CO₂, CO and hydrocarbons; their amounts are all under 10 ppm. We analyzed Pd complex test pieces deposited with Cs by glow discharge mass spectrometry and confirmed that the amount of Pr in the test pieces was under the detection limit (0.01 ppm). If we assume that all of the Pr at 0.01 ppm distributed in the Pd test piece (0.1 mm, 0.7 g) gathered in the analyzed area, a circle of 5 mm diameter and 20 Å depth from the surface of the test piece, the number of Pr atoms attained the same order (10^{13} atoms) of the detected Pr. However, it is impossible for all of the distributed Pr in the Pd test piece to gather in the narrow surface area against the flow of D₂ gas without the application of a specific force on Pr, because such a phenomenon breaks the law of thermodynamics.

The third point is that the isotope ratio of produced elements is anomalous. In this paper, we show the isotopic anomaly of Mo. It provides evidence that the detected material, Mo, was produced by certain nuclear processes. If the Mo were a contaminant, such efficient isotope separa-

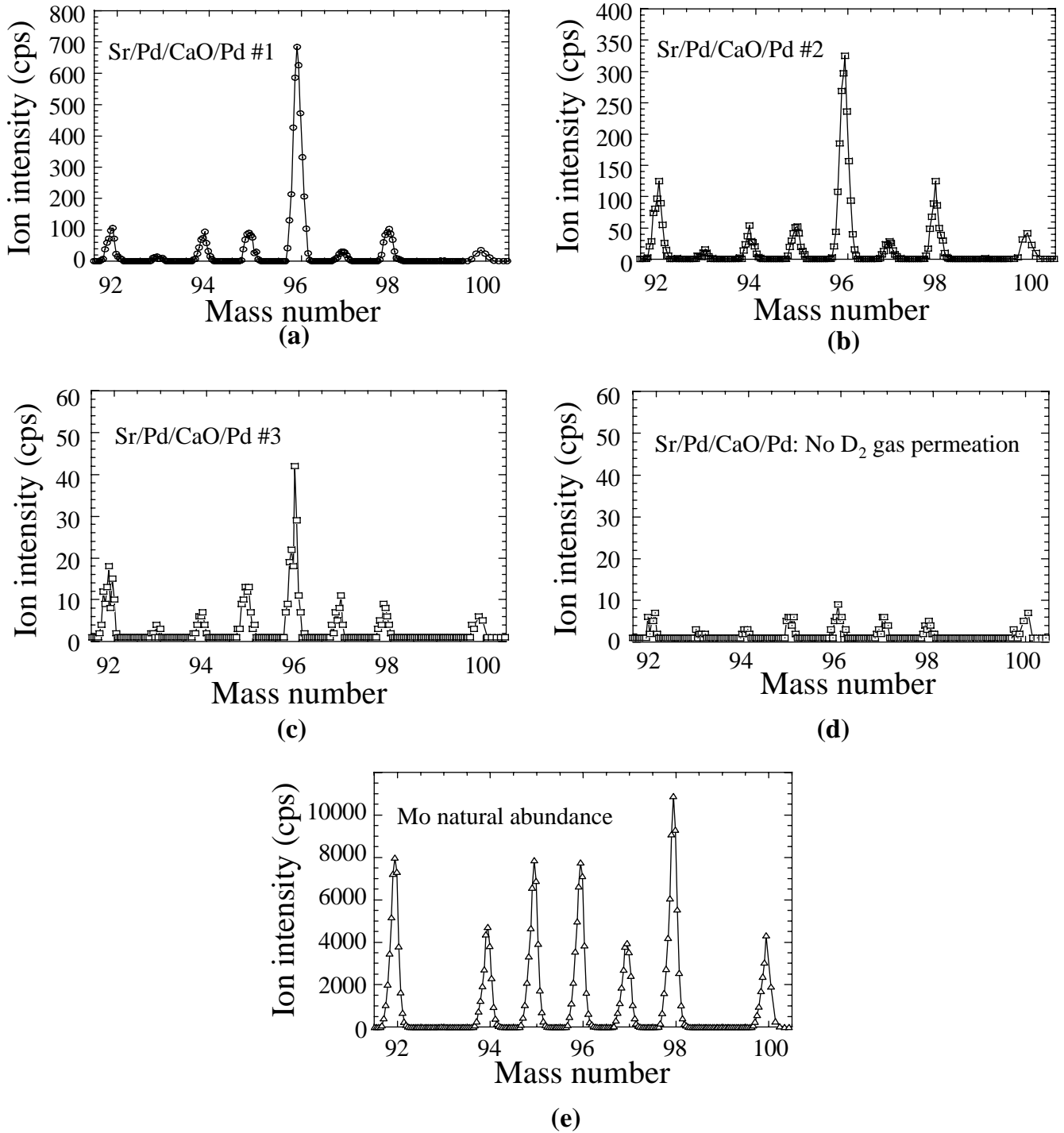


Fig. 9. Anomalous isotopic composition of detected Mo: (a) Isotopic composition of detected Mo for run #1, (b) Isotopic composition of detected Mo for run #2, (c) Isotopic composition of detected Mo for run #3, (d) SIMS analysis for Pd complex test piece with added Sr without D₂ gas permeation, (e) Natural abundance of Mo analyzed by SIMS.

tions would not be possible.

The last point is that the elements detected by the D₂ gas permeation vary depending on the given elements at the beginning of the experiments. In our experiments, Pr or Mo was detected when Cs or Sr was deposited on the surface of Pd complex test pieces, respectively. It is very difficult to assume that the detected elements change depending on the given elements by external contamination.

The above discussion strongly suggests the existence of low-energy nuclear transmutations induced by a simple method. We should stress, however, that impurity control of

Pd test pieces and the apparatus is an important factor to observe the transmutation process. In other words, we might not observe it if we perform experiments in a dirty environment.

We noticed that a certain rule exists between given and produced elements.¹³⁾ The increase in mass number is 8, and the increase in atomic number is 4. At present, we do not have a complete theory that can explain the obtained experimental results without a few assumptions. However, if several assumptions are accepted, they are basically explained by the EINR model,⁵⁾ which is one of the working

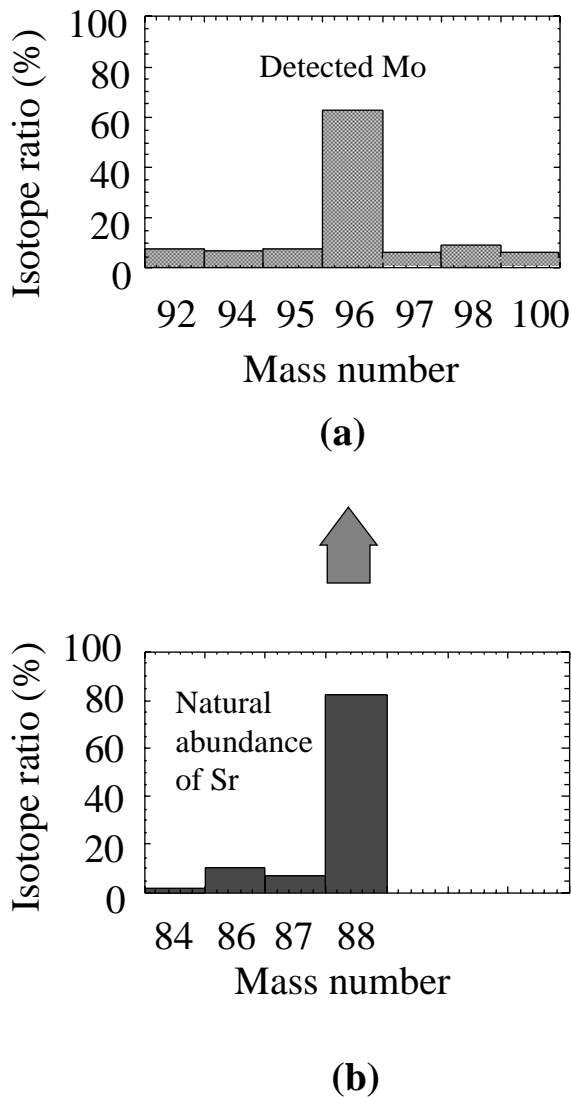


Fig. 10. Relationship of mass numbers between given Sr and detected Mo: (a) Isotopic composition of detected Mo, (b) Isotopic composition of given Sr.

hypotheses in the investigation of the nature of this phenomenon. It might be said that the observed transmutation processes belong to the new category of nuclear reactions on the metal surface associated with deuterium and electrons under low-energy conditions. However, there are many issues to be resolved, especially theoretical ones, and further investigations by other researchers are necessary.

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