

^3He and ^4He from D_2 absorbed in LaNi_5

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Abstract

Helium isotopes from D_2 gas absorbed by LaNi_5 were analyzed with a noble gas mass spectrometer to search for evidence of cold fusion. The ratio of ^3He to ^4He in the sample gas obtained after repeated temperature cycling for about 30 days was different from that in the sample gas before treatment. Increases in ^3He were observed in two experiments using different reaction vessels, indicating that a nuclear reaction may have occurred.

1. Introduction

Numerous searches for “cold fusion” have been attempted since Fleischmann and Pons (FP) [1] and Jones *et al.* (J) [2] reported that fusion reactions took place in palladium after the addition of deuterium at ambient temperature.

There are four possible deuteron fusion reactions [3, 4]:



The fusion of two deuterium nuclei is dominated by reactions (1) and (2), whereas the reaction probabilities of (3) and (4) are much smaller than those of (1) and (2). In addition, the following fusion processes [5] were proposed to explain the significant gap between the FP [1] and J [2] results and to interpret the observations of an anomalously large tritium yield in comparison with the neutron yield [6]:



To confirm the nuclear fusion reaction, it is necessary to detect the reaction products, *i.e.* neutrons, helium, protons, tritium and excess heat. For the detection of neutrons, tritium and excess heat, many detailed

experiments have been performed [6–8]. However, these detecting methods must capture the products generated in the instantaneous reaction, or the results will be strongly influenced by the environment of the experimental site, for example, by cosmic ray irradiation. It should be easy to search for cold fusion reaction products if the reaction product can be accumulated in a closed system. Helium is the only suitable reaction product for this type of experiment.

We investigated helium from the D_2 gas absorbed in $LaNi_5$, because the resulting helium can be stored in a sealed vessel if the reactions, *i.e.* those of eqns. (1), (3) and (5), occur [9, 10]. If the accumulated helium isotopes are above the detection limit of helium, the number of helium atoms can be estimated. In other cases, nucleonic helium may be detected as an isotopic ratio different from the ratio of helium initially contained in the D_2 gas.

A mass spectrometer equipped with a noble gas purification system was used to analyze 3He and 4He in the D_2 gas obtained by gas pressurizing treatment in which D_2 was absorbed and desorbed by $LaNi_5$ in a sealed vessel at temperatures between less than 100 and 300 K. The helium detection limit for the apparatus is about $10^{-20} m^3$ (standard temperature and pressure, STP) which corresponds to 10^5 atoms, so that we could find evidence of nuclear fusion if the reaction continued for several tens of seconds and hours at the fusion rates given by De Ninno *et al.* [11] ($10^3 s^{-1}$) and Jones [2] ($10^{-1} s^{-1}$) respectively.

2. Experimental details

2.1. D_2 gas pressurizing experiment

Figure 1 shows the apparatus used for the reaction of $LaNi_5$ and D_2 gas. The reaction vessel ($6.46 \times 10^{-5} m^3$) sealed with a copper gasket, gas samplers and vacuum-gas connecting lines were made of stainless steel to exclude helium in air. Another reaction vessel ($3.23 \times 10^{-5} m^3$) was used for the second experiment, because the airtightness of this vessel was superior to that of the vessel used for the first experiment. The reaction vessels were previously degassed by heating at 1123 K for half a day under vacuum conditions (around $1.3 \times 10^{-3} Pa$). Before admitting the D_2 gas, the $LaNi_5$ ingot (52.2 g) was loaded in the vessel and then heated at 1123 K for 7 h (18 h for the second experiment) in the vacuum atmosphere ($6.6 \times 10^{-3} Pa$). A portion of the resulting gas was collected in sampler A (sample A in Table 1). The 99.5% pure D_2 gas was then applied to the ingot at a pressure of $7.9 \times 10^5 Pa$. The activation process, which is the repeated increasing and decreasing of temperature in the range 363–273 K was carried out so that the $LaNi_5$ ingot can sufficiently absorb and desorb deuterium. To remove the helium initially contained in the applied D_2 gas, the vessel was evacuated for $9 \times 10^2 s$, with $LaNi_5$ absorbed deuterium at about 100 K (77 K for the second experiment), and a part of the residual gas was collected in sampler

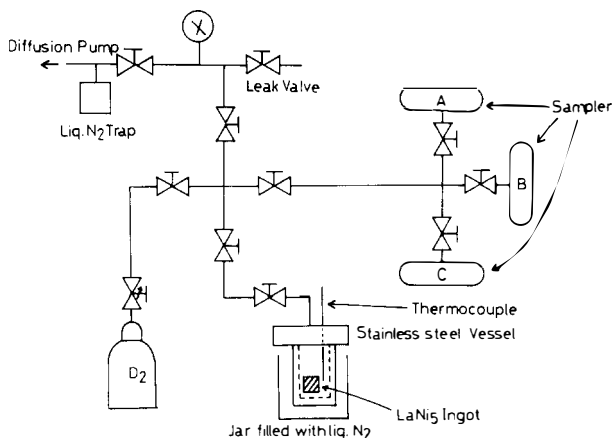


Fig. 1. Schematic diagram of the apparatus. Volumes are as follows: sampler A, $1.91 \times 10^{-4} \text{ m}^3$; sampler B, $1.89 \times 10^{-4} \text{ m}^3$; sampler C, $1.45 \times 10^{-4} \text{ m}^3$; vessel used for the first experiment, $6.46 \times 10^{-5} \text{ m}^3$; vessel used for the second experiment, $3.23 \times 10^{-5} \text{ m}^3$.

B (sample B in Table 1). The vessel containing the hydrogenated $LaNi_5$ was then removed from the vacuum line. Liquid nitrogen was placed around the vessel in order to cycle the vessel temperature between around 300 K and 100 K (77 K for the second experiment). After the temperature fluctuation process had been repeated 40 times over a period of 30 days (43 times and 28.2 days for the second experiment), a portion of the gas separated from the applied D_2 gas was withdrawn into sampler C (sample C in Table 1).

We analyzed the amount of helium isotopes in the three different gaseous samples previously mentioned, and in the gas extracted from the $LaNi_5$ ingot by heating at about 1123 and 1773 K.

2.2. Mass spectrometry of 3He and 4He

A VG5400 mass spectrometer and a noble gas purification system installed at Okayama University were used for helium as well as other noble gas isotopic analyses [12, 13]. All systems were made of stainless steel to avoid permeation of atmospheric helium into the system. The mass spectrometer is equipped with an electron bombardment ion source and with three types of ion collectors; two Faraday cups and a Daly-multiplier. The resolution of one Faraday cup was 200 and that of the other Faraday cup and the Daly-multiplier was 600. The Daly-multiplier system converts ions to secondary electrons with a Daly knob made of stainless steel, and to photons with a scintillator. The photons come from the collector system in a vacuum through a sapphire window and are amplified with a photomultiplier. The detection limit of helium with this Daly collector was about $10^{-20} \text{ m}^3 \text{ STP}$ (10^5 atoms). The mass spectrometer was evacuated with an ion pump and the pressure was about $7 \times 10^{-8} \text{ Pa}$. The noble gas purification system was pumped out with two turbomolecular pumps at a pressure of 10^{-7} Pa . The helium background of the whole system was about $1 \times 10^{-16} \text{ m}^3 \text{ STP}$ (10^9 atoms) and $1 \times 10^{-19} \text{ m}^3 \text{ STP}$ (10^6 atoms) for 4He and 3He respectively.

For helium isotopic analysis, the mass resolutions necessary to separate the hydrogen and deuterium peaks are 510, 400 and 100 for $^3\text{He-HD}$, $^3\text{He-H}_3$ and $^4\text{He-D}_2$ respectively. The high resolution (600) collector was used for the analysis, because it was sufficient to separate these doublets. The low resolution(200) collector was also used to separate ^4He and D_2 .

Sample gas reservoirs made of stainless steel were connected with the noble gas purification system and were heated overnight to decrease any air contamination. After heating, the pressure of the system was in the range of 10^{-7} Pa. Sample gas pipetted from gas reservoirs was introduced into the purification system and purified by Ti-Zr getters heated at about 1073 K. The purified noble gases were exposed to an activated charcoal trap kept at the temperature of liquid air to absorb argon, krypton and xenon. Helium and neon were admitted to the mass spectrometer to measure isotopic ratios and abundances. Most of the deuterium gas was removed by the Ti-Zr getters, and the residual deuterium admitted to the mass spectrometer was separated from the ^3He and ^4He peaks by the high resolving power already noted above and did not affect the helium peak recognition. After the measurements of helium and neon, argon, krypton and xenon were released from the charcoal trap, and were introduced into the mass spectrometer to measure the elemental abundances.

Although the fusion reaction of deuterium does not produce neon, argon, krypton and xenon, the elemental composition of these noble gases provides us with information about the origin of helium contained in the sample gases. For instance, if the elemental composition of these noble gases in the sample gases is similar to that in air, helium generated in the vessel would be caused by air leakage. Nuclear fusion is expected to be revealed as an isotopic anomaly of helium. However, the helium isotopic ratio can also be changed by physical processes such as diffusional transport [14]. During the introduction of gases from the gas sampler into the mass spectrometer, there are several gas diffusional transport processes because the concentration of the gases needs to be appropriate to the measurement. If the processes are carried out in a state of non-equilibrium, the diffusion rate would be larger for the lighter noble gases than for the heavier gases so that the composition of noble gases and isotopic ratios in the samples would change. To determine such an effect on the helium isotopic ratio, the elemental abundance of neon, argon, krypton and xenon was measured in addition to the helium isotopic ratios in some of the sample gases.

Sensitivities and mass discrimination correction factors of the mass spectrometer were determined by measuring a known amount of atmosphere according to the same procedures applied to the sample gases.

3. Results and discussion

Element abundances and isotopic composition of helium, neon, argon, krypton and xenon are listed in Table 1. The standard deviations are shown

TABLE 1

Contents and isotopic ratios of helium, neon, argon, krypton and xenon^a

Sample ^b	³ He	⁴ He	³ He/ ⁴ He	²⁰ Ne	³⁶ Ar	⁸⁴ Kr	¹³² Xe
First experiment							
	($\times 10^8$)	($\times 10^{14}$)	($\times 10^{-6}$)	($\times 10^{14}$)	($\times 10^{14}$)	($\times 10^{12}$)	($\times 10^{10}$)
A	1.57(0.08)	1.45(0.07)	1.08(0.08)	2.38(0.12)	111(5)	15.1(0.7)	99.5(5.1)
B	0.422(0.025)	0.561(0.027)	0.752(0.057)	0.336(0.017)	0.499(0.023)	0.479(0.023)	0.565(0.027)
C	10.8(0.5)	7.08(0.34)	1.53(0.10)	10.7(0.5)	15.3(0.8)	7.21(0.34)	5.16(0.26)
Air			1.399(0.013) ^c				
Second experiment							
	($\times 10^6$)	($\times 10^{11}$)	($\times 10^{-6}$)	($\times 10^{10}$)	($\times 10^{11}$)	($\times 10^9$)	($\times 10^8$)
A	1(4)	1.77(0.31)	6(23)	6.69(1.17)	4.77(0.82)	3.71(0.65)	3.01(0.52)
B	0.1(0.3)	0.214(0.038)	5(14)	1.75(0.30)	1.26(0.22)	0.0717(0.0126)	0.119(0.021)
C	3.4(1.4)	0.947(0.165)	36(16)	2.70(0.48)	5.95(1.03)	0.318(0.055)	0.587(0.103)

^aThe noble gas contents are given as the number of atoms existing in the reaction vessel. Values in parentheses in the body of table are standard deviations.

^bSample A was extracted from the LaNi₅ ingot by heating at 1123 K; Sample B was obtained from the applied D₂ gas; Sample C was obtained from the D₂ gas adsorbed in LaNi₅.

^cRef. 13.

in parentheses. We will discuss the results obtained in this work based on the two assumptions that the nuclear reaction occurs at the surface or near the surface of LaNi_5 and that the generated ^3He is released from LaNi_5 with deuterium. We initially confirmed that no ^3He was observed in the evacuated reaction vessel within the system's detection limits. On both the first and second experiments, it is noteworthy that the amount of ^3He increased in the gas (sample C) obtained after the repeated temperature cycling. The enhancement of ^3He as well as ^4He and other noble gases may have been caused by only the air leakage. However, it is very important to note that the isotopic ratio of ^3He to ^4He for sample C, $(1.53 \pm 0.10) \times 10^{-6}$, was larger than that for both sample B, $(0.752 \pm 0.057) \times 10^{-6}$, and the atmospheric ratio, $(1.399 \pm 0.013) \times 10^{-6}$ [15]. The ratio of ^3He to ^4He for sample C can never be derived by mixing that for sample B with the atmospheric ratio, if the ^3He to ^4He ratio is changed by the air leakage. Therefore, the isotopic anomaly for sample C appears not only to be caused by the leakage of air, but also by some process in the closed system. The amount of ^3He arising from any air leakage was estimated from the increments in ^4He , assuming that ^4He is not produced in this process, and it was then subtracted from the difference in ^3He between samples B and C. Consequently, for the first experiment, $(1.3 \pm 1.0) \times 10^8$ atoms of ^3He may be generated by the nuclear reaction. We did the experiment once again using another vessel with a high airtightness, since the amount of ^3He estimated from the air leakage, $(9.1 \pm 0.5) \times 10^8$, was substantial in the first experiment.

For the second experiment, the ^3He increment $((3.3 \pm 1.7) \times 10^6$ atoms) was observed in the gas obtained after the repeated temperature cycling. The amount of ^3He arising from the air leakage was $(1.03 \pm 0.29) \times 10^5$ atoms, which was very small in comparison with the amount of ^3He observed in the gas treated (sample C). Therefore, the enrichment of ^3He $((3.2 \pm 1.7) \times 10^6)$, which is given by subtracting the amount of ^3He caused by air leakage from the increment in ^3He observed, can be attributed to the occurrence of a nuclear reaction. We regard the second experiment as more reliable.

Table 2 shows the time required for the accumulation of $(3.2 \pm 1.7) \times 10^6$ atoms of ^3He , which is estimated from the reported fusion rates. It is very difficult to estimate the fusion rate for the ^3He production reaction, because

TABLE 2

The time required for ^3He accumulation

	Fusion rate (s^{-1})	The time necessary for ^3He accumulation (s)
FP [1]	4×10^4	80
J [2]	4×10^{-1}	8×10^6
De Ninno [11]	5×10^9	6×10^2
Menlove [7]	2×10^{-1}	2×10^7

This estimation was made for the results of the second experiment.

it cannot be judged which reaction predominates. Therefore, the calculation of the time necessary for ${}^3\text{He}$ accumulation was based on the fact that nuclear reactions (1) and (2) occur with equal probability. The value obtained corresponds to a nuclear reaction that continued for around 6×10^2 s at the fusion rate of 5×10^3 s^{-1} given by De Ninno *et al.* [11]. It took about 2.4×10^6 s (28.2 days) to prepare the sample, so there is no denying the possibility of fusion occurring. As for the comparison with results given by Jones *et al.* [2] and Menlove [7], the result obtained is beyond the realm of possibility.

The fusion rate may be regarded as around 1.3 s^{-1} , which is about 3–7 times as large as that of the results given by Jones and by Menlove, if 3.2×10^6 atoms of ${}^3\text{He}$ had accumulated over 2.4×10^6 s (28.2 days).

Figure 2 illustrates the rare gas elemental abundances relative to the rare gases in air.

$F(m)$ is given by:

$$F(m) = ({}^m\text{X}/{}^{36}\text{Ar})/({}^m\text{X}/{}^{36}\text{Ar})_{\text{atmosphere}}$$

where ${}^m\text{X}$ and the subscript “atmosphere” refer to rare gas element X with mass m and atmospheric respectively [16]. The elemental abundance patterns of all the rare gases for two samples were very different from that of air, and resembled each other. If the leakage of air into vessel during the repeated temperature cycling was large, the abundance pattern of sample C would be similar to that of air ($\log(F(m))=0$ for each rare gas). Therefore, it appears that the increase in ${}^3\text{He}$ was not significantly affected by air contamination. Although the isotopic anomaly of helium observed in this work may be caused by diffusional transport, its possibility was denied owing to the coincidence of the two abundance patterns.

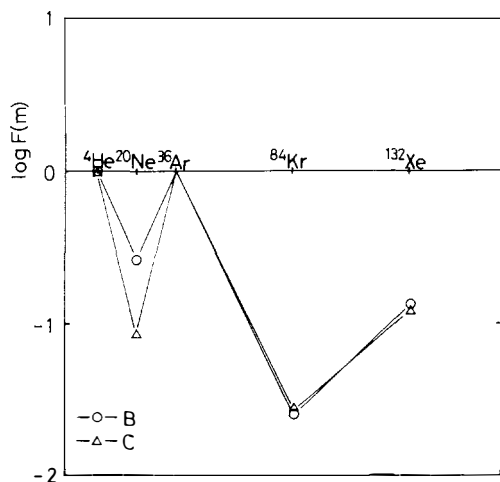


Fig. 2. Elemental abundance patterns of samples B and C for the second experiment. $F(m) = ({}^m\text{X}/{}^{36}\text{Ar})/({}^m\text{X}/{}^{36}\text{Ar})_{\text{atmosphere}}$ where ${}^m\text{X}$ and the subscript atmosphere refer to a rare gas element X with mass m and atmospheric respectively.

The amount of helium isotopes released from the LaNi₅ ingot by heating at 1123 and 1773 K was also measured before and after the repeated temperature cycling, because ³He may be generated in the interior of the LaNi₅ ingot. Although both ³He and ⁴He were detected in the sample before the treatment (sample A in Table 1), there were no helium isotopes in the treated sample within the detection limits. Therefore, it is assumed that the nuclear reaction does not occur in the inner part of LaNi₅, but at the surface or near the surface of LaNi₅.

Although the occurrence of cold fusion was suggested by the increase in ³He in the experiments, perfect reproducibility of data and the background measurements with protium (H₂) are needed to clarify the origin of ³He. These experiments are currently being prepared.

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