

Helium Isotopes from Deuterium Absorbed in LaNi₅

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ABSTRACT

Helium isotopes (³He and ⁴He) from D₂ or H₂ gases absorbed in LaNi₅ were analyzed with a noble gas mass spectrometer. The reproducible increase in ³He, corresponding to a fusion probability of $>8.0 \times 10^{-24}$ d·d·s⁻¹, was observed on the D₂-experiment, whereas ³He was not formed by the reaction of H₂ and LaNi₅. ⁴He production was unreliable, because the reproducibility of the result has not been obtained.

1. Introduction

Helium is a suitable candidate in order to detect the cold fusion product, because helium generated can be accumulated and condensed in a closed system even if the nuclear reaction occurs instantaneously or discontinuously.

From this point of view, we have made a thorough study of the helium isotopes from the D₂ gas absorbed in LaNi₅ by using a noble gas mass spectrometer¹⁻³⁾ since Fleischmann and Pons⁴⁾ and Jones et al.⁵⁾ reported the occurrence of cold fusion.

It is necessary to measure not only ³He and ⁴He but also other noble gases, because the elemental abundance of these gases suggests the origin of the helium isotopes. The estimation of the increase in ⁴He needs the greatest care since the amount of ⁴He in the closed system is easy to be affected by the atmospheric ⁴He due to the air leakage. Such a work that only ⁴He has been measured is out of the question.

We present the results of the precision analysis for the helium isotopes and other noble gases in the D₂ gas

absorbed by LaNi_5 .

2. Experimental details

The reaction vessels (3.23×10^{-5}) sealed with a copper gasket, gas samplers and vacuum-gas connecting lines were made of stainless steel to exclude helium in air. The reaction vessels were previously degassed by heating at 1123 K for half a day in vacuo. After loading in the vessels, the LaNi_5 ingots (52.5 g) was heated at 1123 K for 18 h in the evacuated vessels, and then a portion of the resulting gas was collected in a sampler ("Degas" in Table 1). The 99.5 % pure D_2 gas or 99.999 % pure H_2 gas were applied to the LaNi_5 ingots at a pressure of 7.9×10^5 Pa. Since a large amount of helium was initially contained in the raw D_2 or H_2 gas, the helium gas was removed by evacuating while the applied hydrogen gas was absorbed in the LaNi_5 ingot. After removing the helium gas, a part of the residual gas was collected in a sampler ("Ante-reacted gas" in Table 1). The temperature fluctuation process ($77 \leftrightarrow 300$ K) to the vessel containing the hydrogenated LaNi_5 was repeated 135 times over a period of 130.0 days (2nd run; 131 times, 120.4 days), and a portion of the gas separated from the D_2 or H_2 gas was withdrawn into another sampler ("Post-reacted gas" in Table 1).

A VG5400 mass spectrometer (Operating in the "static" mode) and a noble gas purification system installed at Okayama University were used for helium isotopic analyses. The detection limit of helium with this apparatus was about 10^{-20} m³ STP (10^5 atoms). For helium isotopic analyses, 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, so that the high resolution (600) collector was used for the ^3He analyses and the low resolution (200) collector was used to separate ^4He and D_2 . The helium background of the system was about less than 1×10^{-16} m³ STP ($< 10^9$ atoms) and 1×10^{-20} m³ STP ($< 10^5$ atoms) for ^4He and ^3He respectively.

Nucleonic helium may be detected as an isotopic ratio different from the ratio of helium initially contained in the D_2 gas. The helium isotopic ratio, however, can be changed by air leakage and physical processes such as diffusional transport. Therefore, 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.

3. Results and Discussion

Elemental abundances of helium, neon, argon, krypton and xenon, and isotopic ratios of ^3He and ^4He are listed

Table 1. Contents and isotopic ratios of He, Ne, Ar, Kr and Xe.

	³ He (x10 ⁵)	⁴ He (x10 ¹⁰)	³ He/ ⁴ He (x10 ⁻⁶)	²⁰ Ne (x10 ⁹)	³⁶ Ar (x10 ¹⁰)	⁸⁴ Kr (x10 ⁸)	¹³² Xe (x10 ⁷)
[First run]							
D₂ Gas Experiment							
Post-reacted gas	27.6(7.53)	43.7(5.99)	6.3(1.9)	2.94(0.42)	489(69)	9.47(1.33)	25.0(3.5)
Ante-reacted gas	<u>0.20(0.72)</u>	<u>2.82(0.39)</u>	<u>0.71(2.6)</u>	<u>1.34(0.20)</u>	<u>106(15)</u>	<u>3.12(0.44)</u>	<u>4.08(0.59)</u>
Increment	27.4(8.3)	40.9(6.4)		1.60(0.62)	383(84)	6.35(1.77)	20.9(4.1)
H₂ Gas Experiment							
Post-reacted gas	<1	2.58(0.36)	<4	3.10(0.49)	0.734(0.104)	1.12(0.16)	0.883(0.125)
Ante-reacted gas	<u>0.83(0.67)</u>	<u>0.26(0.04)</u>	<u>32(26)</u>	<u>3.77(0.55)</u>	<u>1.10(0.15)</u>	<u>3.42(0.49)</u>	<u>2.32(0.33)</u>
Increment	—	2.32(0.40)		—	—	—	—
[Second run]							
D₂ Gas Experiment							
Post-reacted gas	30.8(11.2)	0.111(0.016)	2.78x10 ⁶ (1.09x10 ⁵)	56.1(7.8)	23.7(3.3)	29.0(4.1)	0.171(0.024)
Ante-reacted gas	<u>5.58(1.25)</u>	<u>0.286(0.040)</u>	<u>195(52)</u>	<u>29.9(4.2)</u>	<u>7.31(1.01)</u>	<u>21.4(3.0)</u>	<u>3.17(0.45)</u>
Increment	25.2(12.5)	—		26.2(12.0)	16.4(4.3)	7.6(7.1)	—
H₂ Gas Experiment							
Post-reacted gas	8.80(6.20)	1.46(0.21)	60(43)	0.577(0.0951)	1.71(0.24)	0.316(0.045)	0.171(0.024)
Ante-reacted gas	<u>0.80(1.26)</u>	<u>0.0849(0.0119)</u>	<u>94(149)</u>	<u>1.12(0.16)</u>	<u>0.306(0.043)</u>	<u>0.964(0.137)</u>	<u>0.571(0.086)</u>
Increment	8.00(7.46)	1.37(0.22)		—	1.40(0.29)	—	—
Degas(at 1123 K)	2(2)	14.6(2.5)	14(14)	47.7(8.2)	26.2(4.5)	26.6(4.6)	22.6(3.9)
Air			1.399(0.013)				

· The noble gas contents are given in number of atoms existing in the reaction vessel.

· The standard deviations are shown in brackets.

in Table 1. We assumed that the nuclear reaction occurs at the surface or near surface of LaNi₅ and that the generated helium is released from LaNi₅ with hydrogen. On the experiment using the D₂ gas, it is noteworthy that the amount of ³He(2.74(0.83)x10⁶ atoms) increased in the gas obtained after the repeated temperature cycling, while no enrichment of ³He was observed for the H₂-experiment. The phenomenon was reproduced in the second run. The increase in ⁴He(4.09(0.64)x10¹¹ atoms) was also observed in the first run. The ⁴He production, however, was unreliable, because the reproducibility of the phenomenon was not obtained in the second run.

The enhancement of ³He as well as ⁴He may be caused by only the air leakage. If we assume that the increment of ⁴He originated from the atmosphere, then it should be accompanied by 5.72(0.90)x10⁵ atoms of ³He which is derived by using the atmospheric ratio of 1.399x10⁻⁶. Thus, the excess of ³He is 2.17(0.92)x10⁶ atoms. Assuming that the ~2.2x10⁶ atoms ³He generated by the D₂ gas(2.5x 10²² molecules) absorbed in LaNi₅ during a period of the temperature fluctuation process for 1.1x10⁷ s, we obtained a lower limit of ~8.0x10⁻²⁴ fusion/d-d·s⁻¹ based on the reaction d + d → ³He + n. The fusion probability is comparable to that given by Jones et al.⁵⁾ and Menlove.⁶⁾

Figure 1 shows the noble gas elemental abundance relative to that in air, expressing the following equation,

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

where ^mX is the noble gas element with mass m and the subscript "atmosphere" means the atmospheric elemental

abundance.

The elemental abundance patterns provide significant information regarding the isotopic anomaly of helium observed in this work, such as the air leakage and the diffusional transport. If the leakage of air into the vessel occurred during the temperature fluctuation process, the abundance pattern of the post-reacted gas would be close to that of air ($\log F(m)=0$). The increase in ^3He was not significantly affected by the air contamination, because the abundance patterns for both ante- and post-reacted gases resembled each other, and were much different from those for air. The possibility of the diffusional transport was also denied due to the coincidence of the two patterns of the ante- and post-reacted gases.

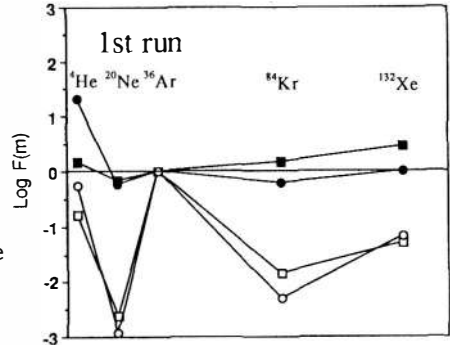


Figure 1. Elemental abundance Patterns.

■ H₂ Ante-reacted gas
 ● H₂ Post-reacted gas
 □ D₂ Ante-reacted gas
 ○ D₂ Post-reacted gas

4. Conclusions

The enrichment of ^3He was observed in the D_2 gas absorbed by LaNi_5 after the repeated temperature fluctuation cycling. The phenomenon, however, was not obtained from the H_2 -experiment. The elemental abundance patterns indicate that the increase of ^3He was not obviously caused by the air leakage. The fusion probability estimated by the ^3He production was $>8.0 \times 10^{-24}$ fusion/d-d \cdot s $^{-1}$ which is in agreement with the value determined by Jones et al. or Menlove.

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