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The Occurrence of He^3 in Natural Sources of Helium*

L. T. ALDRICH** AND ALFRED O. NIER
Department of Physics, University of Minnesota, Minneapolis 14, Minnesota

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An investigation of the He^3/He^4 abundance ratio in helium extracted from the atmosphere, gas wells and a number of minerals has been made. Wide variations in this ratio are observed indicating that a more comprehensive program would be well worth while. He^3 is observed in all sources of helium except radioactive minerals. This suggests the possibility of a primordial source as well as more recent sources for this isotope.

IT was found in 1939 by Alvarez and Cornog[1] that He^3 existed in nature and that its abundance in atmospheric helium was approximately ten times as great as that in well helium. In view of the great interest in the origin of helium and particularly He^3 it seemed worth while to examine the helium found in a variety of sources, both mineral and gaseous.

APPARATUS

The isotope analyses were made with a 60° mass spectrometer resembling in many respects one already described.[2] It differed in the following: (a) The ion source employed an additional focusing plate mounted between J_3 and G_1. G_2 and D were omitted, the spacing between G_1 and G_3 was 9 mm and the slits in G_1 and G_3 were 0.008 inch and 0.004 inch wide, respectively. (b) A single ion collector was used. The collector slit could be varied in width by means of a sylphon bellows adjustment. In most of the present work the slit width used was 0.007 inch. The usual secondary electron suppressor plate was present between the slit and the collector. Normally this plate would be operated at a negative potential, but since in this work high sensitivity was essential, the plate was operated at a positive potential of 45 volts above ground. This increased the sensitivity by using the collector plate as an electron multiplier. Since the multiplying effect depended slightly upon the mass of the ion, a correction was applied. (c) The feedback amplifier used a General Electric FP-54 electrometer tube in its first stage. By means of sylphon operated switches in the electrometer tube housing the grid resistor could readily be chosen to be 10^8, 10^{10} or 10^{12} ohms.

For normal operation of the spectrometer a sample of at least one standard cc was found desirable. If smaller samples were used the pressure decrease during a run was intolerable since the instrument consumes one-half standard cc per hour. For some of the gaseous samples as well as the spodumene the amount of helium available was considerably less than that required for normal operation. For these samples the mass spectrometer fore pump was replaced by a liquid nitrogen cooled charcoal filled trap. In this type of operation only 0.01 standard cc of helium is required since the sample is continuously circulated by connecting one end of the trap to the main diffusion pump and the other to the gas inlet system of the ion source. However, this type of operation did not permit as high an operating pressure in the spectrometer so that the sensitivity is somewhat reduced. Unquestionably, further alterations could be made to give the full sensitivity but time did not permit their development before the present study had to be concluded.

Figure 1 shows the helium gas extraction and purification system used. The vacuum furnace held up to 500 grams of mineral and could be heated to at least 1100°C. For radioactive minerals which contain large amounts of helium a direct fusion furnace similar to that described by Evans[3] could be employed. The same gas purification apparatus was used for mineral and gas well samples.

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* A preliminary report of this work was given at the 1948 Washington meeting, Phys. Rev. **74**, 1225 (1948).

** Present address, Department of Physics, University of Missouri, Columbia, Mo.

[1] L. W. Alvarez and R. Cornog, Phys. Rev. **56**, 613 (1939); **56**, 370 (1939).

[2] A. O. Nier, Rev. Sci. Inst. **18**, 398 (1947).

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RESULTS

Because of the large percentage mass difference of He^3 and He^4 the possibility of systematic error in the He^3/He^4 abundance ratio always exists in a mass spectrometer due to discrimination in the instrument. For this reason a certain atmospheric helium sample was used as a standard and all ratios given in this paper were computed assuming the He^3/He^4 ratio for the standard to be 12×10^{-7}. During the two years that elapsed since this study was begun the instrument gave values for this ratio varying from 10 to 17×10^{-7}. At present the absolute accuracy of the instrument is not well enough known to warrant choosing a better value for this ratio. It is believed that for He^3/He^4 ratios greater than 5×10^{-7} the precision of the spectrometer analyses is of the order of 10 percent. For ratios less than this the relative error may be as great as 30 percent.

In Fig. 2 are copies of recorder tracings showing typical spectra in the mass 3 region for well and atmospheric helium. Although the partial pressure of hydrogen in the instrument due to residual gas in the vacuum system or to impurity introduced with the sample was normally less than 10^{-8} mm, there was sufficient sensitivity to produce an HD$^+$ peak of the size shown. This ion differs in mass from He^3 by one part in 510. The resolving power was such that analyses were possible whenever the He^3 peak was at least 10 percent of the residual HD peak. Occasionally, as in the particular well helium spectrum shown, a small peak on the high mass side of HD appeared. Since it did not interfere with this study its origin has not been investigated thoroughly.

Table I gives the He^3/He^4 abundance ratio in helium from various minerals. No He^3 peak could be detected in helium extracted from radioactive ores and the upper limit set for its abundance was determined by the amplifier noise level at the time the work was done. For the beryls the mineral samples were divided in half and a complete run including gas extraction, purification and analysis made on each half. Excellent agreement was obtained for the two runs in each case indicating that the variations listed are real. While only one complete run was possible on each of the spodumene samples, the ease of extraction of the gas and the relatively large He^3 peak observed enables one to have real confidence in the results obtained.

Table II gives the He^3/He^4 abundance ratio in helium from several gaseous sources. Wide varia-

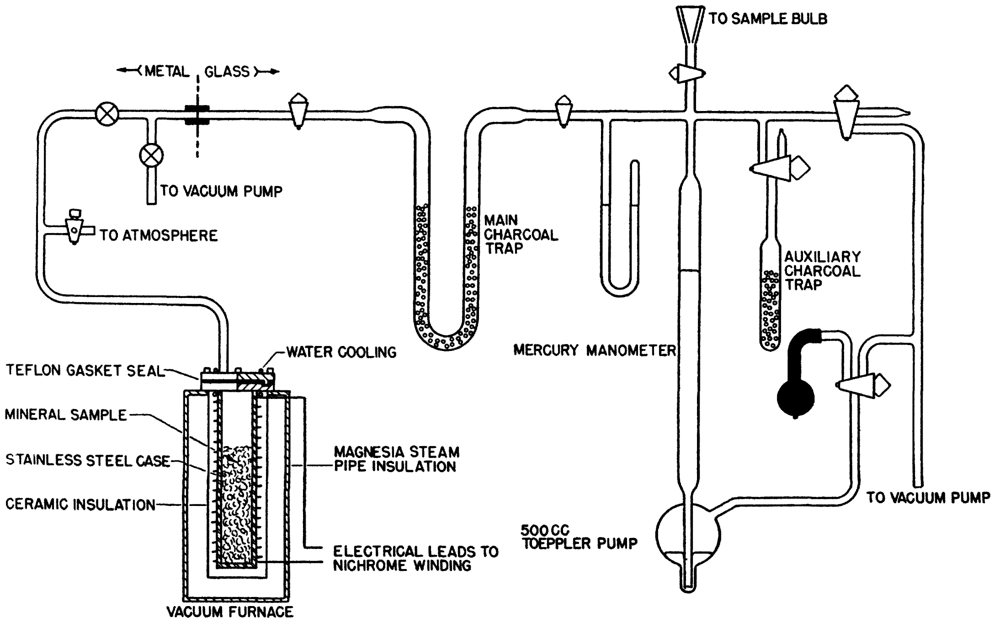

FIG. 1. Helium gas extraction and purification system. Main trap contained 50 grams of activated charcoal, auxiliary trap contained 10 grams. Toeppler pump and auxiliary trap were used to collect the helium in order that the pressure in the furnace and main trap could be kept below 1 mm Hg.

TABLE I. He^3/He^4 abundance ratios in helium from various minerals.

Mineral	Location	$He^3/He^4 \times 10^7$
	Radioactive ores	
Blomstrandine	Hittero, Norway	<0.2
Pitchblende	Great Bear Lake, Canada	<0.3
Monazite	Bahai, Brazil	<0.2
Monazite	Raode, Norway	<0.2
Uraninite	Joachimsthal, Czechoslovakia	<0.3
	Non-Radioactive ores	
Beryl	Eräpyhä, Eräjärvi, Finland	0.6
Beryl	Lemnäs, Kimito, Finland	0.5
Beryl	Keytone, South Dakota, U.S.A.	1.2
Beryl	Spruce Pine, South Carolina, U.S.A.	1.7
Beryl	Jokkmokk Parish, Lapland, Sweden	1.8
Beryl	Audabon, Maine, U.S.A.	3.0
Beryl	West Rumney, New Hampshire, U.S.A.	12.0
*Spodumene	Cat Lake, Manitoba, Canada	24.0
*Spodumene	Edison Mine, South Dakota, U.S.A.	120.0

* Analyzed by continous circulation method described in section on apparatus.

tions in this ratio are seen to exist. Since only small amounts of gas were immediately available at the time the work was done, the analyses of some of the samples could only be made using the continuous circulation method. The fact that He^3 was not observed in the last three samples was probably due to the lower sensitivity obtained when this method is used.

An examination of the mass 5 position for atmospheric helium and one of the well helium samples indicated a small peak having an abundance of the order of 1/1,000,000 that of the He^4 peak. An investigation of this peak indicated that in all likelihood it was due to HeH^+ formed in the spectrometer and that if He^5 should exist its abundance cannot exceed 1/5,000,000 that of He^4.

DISCUSSION

Since the discovery of helium, its origin has been a subject of considerable discussion. While the amount of helium occuring in the lithosphere is not so large that it could not be attributed entirely to radioactive decay, the possibility of a primordial source cannot be excluded.[4] The discovery that He^3 not only existed in nature but also that its abundance in atmospheric helium was considerably greater than in well helium[1] indicates that there are independent sources of the two isotopes.

TABLE II. He^3/He^4 abundance ratios for helium from gaseous sources.

Location	Gas	$He^3/He^4 \times 10^7$
Stamford, Connecticut	Air	12.0
Rattlesnake Well, New Mexico	Natural gas well	0.5
*Amerada State Well, Lea County, New Mexico	Natural gas well	3.0
*Gulf Oil, W. Grimes Well, Lea County, New Mexico	Natural gas well	5.0
Boggs *A*-2, Barber County, Kansas	Natural gas well	0.5
Miles *A*-7, Kingman County, Kansas	Natural gas well	0.5
Otis Field, Rush and Barton County, Kansas	Natural gas well	2.0
Excel Plant, West Panhandle, Texas	Natural gas well	1.5
Cliffside Well, Amarillo, Texas	Natural gas well	1.5
*Austin Storage Field, Michigan	Natural gas well	1.0
*Norwich Field, Newaygo County, Michigan	Natural gas well	<2.5
*Riverside Well, Kent County, Michigan	Natural gas well	<2.0
*Hatton Field, Clare County, Michigan	Natural gas well	<1.0

* Analyzed by continuous circulation method described in section on apparatus.

Hill,[5] in 1941, proposed that He^3 was the decay product of H^3 formed in the bombardment of Li^6 in the lithosphere by neutrons associated with cosmic rays in the reaction $Li^6(n, \alpha)H^3$. Similarly, Libby[6] has suggested the reactions $N^{14}(n, C^{12})H^3$, $N^{14}(n, 3\alpha)H^3$, as a possible explanation of the higher He^3 abundance in the atmosphere. Morrison[7] has suggested that the atmospheric He^3 does in fact originate as a result of such cosmic-ray processes, and that present data indicate that this source is adequate to supply the amount observed even though helium is in all likelihood continuously lost from the atmosphere. He ascribes the gas-well He^3 to the neutron capture by lithium in the rocks, with the neutron source

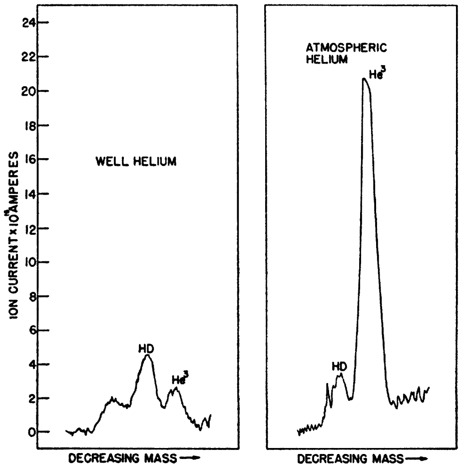

FIG. 2. Mass spectra in mass 3 region for well helium and atmospheric helium. He^4 ion current 2×10^{-8} amperes for both spectra.

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[4] G. S. Rodgers, Prof. Paper **121**, U.S. Geological Survey, (1921).

[5] R. D. Hill, Phys. Rev. **59**, 103 (1941).

[6] W. F. Libby, Phys. Rev. **69**, 671 (1946).

[7] P. Morrison, private discussion, April 29, 1948.

Table III. Data on gas from beryls.

Location	Scc He/g	He^3/He^4 $\times 10^7$	Scc He^3/g $\times 10^7$	Scc argon	$\frac{\text{Scc Radio argon*}}{\text{g beryl}} \times 10^4$	Age-year $\times 10^8$
Eräpyhä, Eräjärvi, Finland	0.018	0.6	0.01	0.13	2.0	14
Lemnäs, Kimito, Finland	0.011	0.5	0.005	0.33	<0.2	14
Keystone, South Dakota, U.S.A.	0.022	1.2	0.03	0.04	<0.2	14
Jokkmokk Parish, Lapland, Sweden	0.023	1.8	0.04	1.0	25	10
West Rumney, New Hampshire, U.S.A.	0.004	12.0	0.05	0.1	1.0	4

* Computed on basis that the "ordinary" argon present has the isotopic composition of present day atmospheric argon.

being the known (α, n) reactions in ordinary igneous rock.

The abnormally large amount of helium found in beryl[8] makes this mineral one of special interest for this study. Table I indicates a wide variation in the He^3/He^4 ratio depending upon the location from which the mineral was obtained. In order to learn more about the He^3 content of the minerals a more complete investigation of five of these samples was made. In this study an analysis of the total gas evolved from the beryl was made including a quantitative determination of the helium contained per gram of mineral. The analyses showed argon to be present in varying amounts in all five samples. In view of the recent discovery[9] that potassium minerals do indeed contain excess A^{40} due to the decay of K^{40} to A^{40} by K electron capture, it seemed worth while to make an isotopic analysis of the argon in these samples.

Table III lists some of the results of this investigation. It is seen that in spite of the fact that the He^3/He^4 ratio varies by a factor of over 20 in this group of samples, the absolute He^3 content per gram of mineral does not vary systematically with this ratio. Moreover the absolute He^3 content in minerals of the same age varies nearly as much as the total variation in all the samples. The absolute radiogenic A^{40} content does not appear to be correlated with any other factor. The Swedish beryl is of particular interest. Its radiogenic A^{40} content is greater than that found in any comparable sample of potassium mineral.[9] It contained a slight impurity of mica, a potassium mineral, but not nearly enough to account for the amount of A^{40} present. Without more specific information concerning the pegmatite from which the sample was obtained the data implies the presence of potassium in the magma from which the beryl crystallized. Moreover, this suggests that the helium like the argon in the beryl could have its origin in the magma.

The results on spodumene (Table I) are of special interest since the He^3/He^4 ratios in the two samples examined were the highest of any natural sources of helium examined, including atmospheric helium. This suggests the possibility that some of the He^3 found here could have its origin in the nuclear reaction $Li^6(n, \alpha)H^3$. However, if this is the case, no definite conclusion as to neutron source can be made. The Canadian sample, having the lower He^3/He^4 ratio, was taken fresh from the surface of the earth while that from South Dakota was mined 20 to 30 feet below the surface. The absolute He^3 content per gram of mineral in both samples was approximately 0.01×10^{-7} scc/g, a value comparable with that found for the beryls.

The data on the well helium samples are too incomplete at present to draw conclusions about the origin of either He^4 or He^3 in natural gas wells. It was observed that in every case where a thorough search was possible He^3 was detected. In four of the samples, 2, 7, 8 and 9 of Table II, the helium content was known from U. S. Bureau of Mines analyses. As in the case of the beryls the absolute He^3 content among these samples did not vary as much as the absolute He^4 content.

The present study can hardly be regarded as more than a preliminary exploration of a new and fascinating field of investigation. It is apparent that a far more comprehensive and systematic study will be required to definitely establish the natural sources of He^3 and He^4. Included in such a program should be an investigation of the helium from the upper atmosphere and that from meteorites.

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The Angular Distribution of 1 to 3.5 Mev Deuterons Scattered by Deuterons

J. Morris Blair,* George Freier, Eugene Lampi, William Sleator, Jr., and J. H. Williams
University of Minnesota, Minneapolis, Minnesota

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The differential cross section for the elastic scattering of deuterons by deuterons has been obtained in the energy range from 1 to 3.5 Mev. The observed yields were corrected for the counted particles produced in the D – D reactions and for those scattered by contaminants by using the results of other experiments. The resultant elastic cross sections are probably accurate to about 2.5 percent.

SCATTERING experiments have been a remarkably fruitful source of information about nuclear forces; and, in general, one might say that the importance of the information and the ease with which it can be obtained decreases as the complexity of the particles involved increases. From this point of view the deuteron-deuteron combination is the most promising one not previously investigated in the energy range available to the Minnesota Van de Graaff generator, since careful work has been done on proton-proton[1,2] and proton-deuteron[3] scattering. We have measured, as described in this paper, the deuteron-deuteron elastic scattering cross section as a function of angle and energy from 10° to 45° (in the laboratory system) and from 1 Mev to 3.5 Mev.

EXPERIMENTAL METHOD

These data on the scattering of deuterons by deuterons were obtained with the same apparatus and by the same method as the data on the scattering of protons by protons presented in a recent paper.[2] A well-collimated beam of deuterons, accelerated by the Minnesota electrostatic generator, was passed through a chamber containing deuterium gas at a pressure of about one centimeter of mercury. The intensity of this beam was measured by allowing it to pass from the gas through a 0.0002-inch Nylon window into an insulated cup in an evacuated region. A fraction of the deuterons scattered from a small, well-defined volume in the center of the chamber passed through a slit system and through a Nylon window into a proportional counter. The factors for obtaining the scattering

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* Present address: Argonne National Laboratory, Chicago, Illinois.

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