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ISOTOPE COMPOSITION OF HELIUM IN ULTRABASIC XENOLITHS FROM VOLCANIC ROCKS OF KAMCHATKA

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The purpose of this work is to refine our knowledge about the nature of helium with a high abundance of the rare isotope ^3He ($^3He/^4He = 10^{-5}$) discovered in terrestrial volcanic gases in 1968.

We will discuss here the results of isotope analyses of helium released by step-wise heating of ultrabasic xenoliths and some volcanic rocks. On the basis of these results, possible sources of ^3He in the earth due to fission and nuclear reactions are considered critically. The most probable source of the high abundance of ^3He is shown to be due to the capture and trapping of primordial He by the earth during its formation (primordial helium $^3He/^4He = 3 \times 10^{-4}$), a small but significant fraction of which has been retained to the present time.

1. Introduction

Systematic studies of helium isotope composition from a wide variety of samples including natural gases, rocks, minerals and liquid–gas inclusions were initiated in 1968.
The helium isotope analyses were conducted in the magnetic resonance mass-spectrometer [21] designed especially for measurement of small isotope ratios of $^3He/^4He$ (up to 10^{-10}) characteristic of terrestrial helium [19].

One of the most interesting and unexpected results was a discovery of terrestrial helium with an extremely high isotope ratio of $^3He/^4He \sim 10^{-5}$ (i.e. 100–1000 times the usual ratio for the earth's crust) in gases of hot springs of the South Kuriles [18]. Later the same high ratio was determined for helium from volcanic gases in the North Kuriles, Kamchatka [34] and Iceland [20].

The source of volcanic helium seems quite certain to be the deep interior [34]*. A logical continuation of this work was the study of the isotopic composition of helium from ultrabasic xenoliths, which in the opinion of the majority of geologists represent mantle material. The first experiments gave promising results [35], viz. in xenoliths from Kamchatka and Antarctica which gave ratios of $^3He/^4He = (3.6–4.6) \times 10^{-6}$ which were much higher than that of the atmosphere, which has $^3He/^4He = 1.4 \times 10^{-6}$ [17], and similar to that characteristic of volcanic gases. To refine our knowledge about the nature of helium with such high $^3He/^4He$ ratios, we measured the isotopic composition of helium released by step-wise heating of xenoliths and also undertook experiments imitating nuclear processes. The present paper discusses the data obtained.

* It is necessary to note that in some particular cases helium with a high isotope ratio may form within the earth's crust, for example, in lithium minerals [1] and in some quartz samples [25, 36].

2. Experimental techniques

The extraction of helium from a sample and its purification were carried out in an all-steel high-vacuum apparatus joined to a mass-spectrometer. For this purpose we used a vacuum furnace with internal heating consisting of a steel housing with a crucible heated by a thick molybdenum coil, a thermocouple for measuring the crucible temperature and a tripper device for dropping samples into the crucible (suitable for the consecutive heating of five samples). Two cold traps with activated charcoal and a titanium getter were used to purify the helium. Large temperature steps (300°C), sample weights of about 10 g and decreases of experimental time when possible (to diminish the background of the apparatus) were used due to a low helium content.

Samples were put into the tripper device and the furnace was evacuated by a diffusion pump with the crucible heating up to 1150°C for 1 hour. Then, the apparatus background was measured, viz., evacuation was discontinued, and crucible heated to 1100°C for 10 min. The gas extracted was purified by means of activated charcoal cooled by liquid nitrogen and a titanium getter (at 400°C) and admitted into the chamber of the mass-spectrometer. The amount of ^{4}He in this blank was $(1-3) \times 10^{-8}$ cm^3 STP. Then the sample was introduced into the crucible and the heating was carried out in steps 300°, 600°, 900°, and 1100°C, each maintained for 10 min, with a subsequent purification of each fraction on the charcoal and titanium for 15 min (during the purification the crucible cooled down to a temperature not less than 300°C).

Helium isotope analyses were carried out for each temperature fraction; the mass-spectrometer was calibrated before and after the analysis by atmospheric helium or by a specially prepared standard mixture with isotope ratio of ^{3}He/^{4}He $= 1.29 \times 10^{-5}$. For calibration the chamber of the mass-spectrometer was filled with helium equal in amount to that extracted from a sample. The helium content in xenoliths was determined on the mass-spectrometer by the peak intensity method. The error of the isotope composition determination is (depending on helium amount measured) ± 5 to ± 20%; the error in the determination of the amount is ± 10 to ± 20%.

2.1. Specimen characteristics

The study was carried out on specimens of so-called cognate inclusions of ultrabasic composition found in rocks of different type in tectonically different regions; in high-alumina basalt-andesites of Kamchatka, i.e. a region similar in tectonic structure to island arcs, in tholeiitic basalts of Iceland close in structure to mid-oceanic ridges, and in basalts of the Sverre volcano (Spitsbergen) and Ross Island (Antarctica). Despite very different geological settings, the inclusions are homogeneous in composition and belong to a genetically common pyroxenite-peridotite-olivinite series. Their common features indicate that they belong to a spinel facies of ultrabasic rocks, with an absence of pyrope, and with gradual transitions between varieties similar in mineral composition. [7, 26].

Ultrabasic xenoliths which were found mainly in large, essentially basaltic, or more rarely in basalt-andesitic, stratovolcanoes of Kamchatka [24, 27, 22] and which are associated with high-alumina magma (Avacha, Kronotsky, Klyuchevsky and others) were studied in more detail. It is to be noted that xenoliths from basalts of young areal issues, Lower Quaternary

TABLE 1

Chemical composition of country rock and ultrabasic inclusions from Avacha and Kronotsky Sopka

	Avacha		Kronotsky Sopka	
	country rocks	peridotite inclusion No. 1	country rocks	peridotite inclusion No. 2
SiO_2	54.20	42.60	52.46	44.92
TiO_2	0.80	0.10	0.82	1.12
Al_2O_2	20.54	3.34	19.58	8.33
Fe_2O_3	1.67	0.71	5.54	7.81
FeO	4.86	7.60	3.65	5.02
MnO	–	0.14	0.02	0.15
MgO	3.72	41.78	3.76	15.25
CaO	9.44	0.84	9.72	14.96
Na_2O	3.42	0.16	3.47	1.35
K_2O	0.85	0.13	0.41	0.14
H_2O^-		–	0.10	
H_2O^+	0.47	–	0.16	0.09
P_2O_5	0.30	–	0.17	0.27
	100.27	97.40	99.86	99.42

plato-effusive complexes, basalt-andesitic volcanoes in areas of extensive acidic volcanism centers, and in thick acid pyroclastic sheets, have not been known as yet. The inclusions occur more often in pyroclastic flows and scoria, while in lavas they are not so common. The size varies from 1–2 to 50–80 cm, averaging from 3 to 8 cm; they are fragmentary and typically xenogenic in shape. The average size of grains is 1–3 mm; sometimes the rock has a coarse-grained aspect and grains 5–7 cm. The structure is massive, eutaxitic. Contacts with the country rock are usually abrupt, without signs of alteration or interaction. However, in some cases there is an amphibole rim 1–3 cm thick surrounding inclusions in rocks from the Kronotsky and Avacha volcanoes in which rimmed and unrimmed inclusions occur together rather often.

The country rocks of Avacha and Kronotsky are similar in chemical composition, while the peridotite inclusions are rather variable (Table 1); the inclusions from Avacha are characterized by high ratios of $MgO/(FeO + Fe_2O_3) = 9–10$ and $FeO/Fe_2O_3 = 6–13$ and lower Al_2O_3, TiO_2 and alkali contents; inclusions of the Kronotsky volcano are marked by very high CaO (15%), TiO_2, and Al_2O_3 contents, a FeO/Fe_2O_3 ratio enriched in Fe^{3+}, and a Mg content 2.5 times less than that in peridotites of Avacha. It is important that the minerals and the mineral compositions remain constant in different inclusions. In this case the differences are due to changes in the mineral proportions. Chrome-diopside occurs in particular in peridotites of Avacha only in single relict grains; the major part is enstatite. Chrome-diopside in peridotites of the Kronotsky volcano amounts to 30%.

Three mechanisms for the incorporation of ultrabasic inclusions are usually discussed: (a) accumulation of a crystalline phase at early stages of basaltic magma differentiation; (b) capture of upper crust xenogenic material; (c) capture of relicts of the original reservoir in which the magma was generated.

The regular relationship of these inclusions to basalt-andesite assemblage rocks, the absence of such inclusions in acid volcanics, the similarity of inclusion mineral assemblage, and the presence of chrome-diopside (which is not indicative of normal ultrabasic intrusions) are all arguments in favour of the third hypothesis.

The variety of mineral proportions in the inclusions implies the entrapment of inclusions at different stages of mantle matter transformation during magma generation. Inclusions rich in chrome-diopside appear to represent the mantle matter at early stages of basalt melting, and olivine and olivine-enstatite inclusions from Avacha were captured when basaltic components had been completely separated from the mantle matter

3. Results and discussion

3.1. All the samples (Table 2) of ultrabasic xenoliths studied are characterized by high isotope ratios of $^3He/^4He \simeq (3.2–10) \times 10^{-6}$. As the deep-seated origin of xenoliths is widely accepted, the high isotope ratio up to 10×10^{-6} found both is xenoliths and in volcanic gases appears to characterize the isotopic composition of deep-seated helium. It is important to note that the helium with high $^3He/^4He$ ratio is extracted from the xenoliths at high temperatures (for example, samples 3, 5 and 8 yield helium with a ratio of $^3He/^4He \gtrsim 10^{-5}$ which was extracted at temperatures above 1000°C). We infer from this observation that this helium could not get into the xenoliths due to surface low-temperature processes.

The results obtained by step-wise degassing of helium offer a possibility for samples to be arbitrary subdivided into two groups. The first group includes samples 1, 3, 5 and 6 characterized by a regular increase of $^3He/^4He$ ratio as the temperature steps increase. For example, for sample 5 there is a seven-fold increase of the isotope ratio in a high-temperature fraction.

The observed variations in the isotope ratio cannot be accounted for by differences in the diffusion coefficients of ^3He and ^4He, as the observed effect is too large [13] and is of opposite "sense" to that expected by diffusive fractionation (^3He should have been released preferentially in low-temperature fractions). These variations are rather due to differences in the sites of helium with a high $^3He/^4He$ isotope ratio and ^4He (or helium with low isotope ratio). In cases when the major loss of each "helium type" takes place at a different temperature, a well-defined change of isotope composition of helium occurs in the temperature fractions. Unfortunately, the large temperature intervals and the consequent low "resolution" of the method does not at present allow a determination of the nature of helium in xenoliths, viz. whether monoisotope ^3He or helium with high isotope ratio is characterized by high activation energy.

Samples 2, 4, 7 and 8, which are characterized by a

TABLE 2

Isotope composition of helium in xenoliths *

No.	In a sample as a whole			Temperature intervals (°C)					
				300–600		600–900		900–1200	
	$^3He/^4He$	^4He	^3He	$^3He/^4He$	^4He	$^3He/^4He$	^4He	$^3He/^4He$	^4He
1	3.0	6	18	1.6	3.5	3.5	1.5	7.6	1.0
2	3.9	–	–	4.1	–	–	–	3.8	
3	9.0	10	90	1.5	1.0	8.2	1.5	10	7.5
4	8.6	3.5	30	8.8	0.21	8.0	0.56	8.8	2.7
5	14	30	420	6.4	20	20	5.5	45	4.0
6	3.2	30	96	2.6	–	3.2	–	7.2	–
7	4	10	40	4.5	3.0	3.8	6.0	4.0	1.0
7a	2.8	10	28	2.5	6.0	2.9	3.2	6.3	0.8
8	9.5	1	9.5	1.8	0.25	12	0.50	12	0.25
13	9	3	27	–	0.10	8.3	0.8	9.8	2.1
14	2.2	0.8	1.8	2	0.62	2.8	0.16	–	0.02
15	1.5	0.6	0.9	1.5	0.35	–	0.2	–	0.05
16	1.6	3.5	5.6	1.6	3.0	–	0.4	–	0.01
19				6.7		2.6			
19a				7.1		2.7			
19b				8.7		2.9			
26	0.11	270	30						
27	0.15	200	30						
28	0.20	360	72						
A	1.4								

* $^3He/^4He$ ratio is to be multiplied by 10^{-6}; ^4He and ^3He abundances are given in $(cm^3\ g^{-1}) \times 10^{-8}$ and in $(cm^3\ g^{-1}) \times 10^{-14}$ respectively.

Xenolith samples: 1 = peridotite inclusion in andesites, Avacha; 2 = peridotite inclusion in basalts, Kronotsky Sopka; 3 = olivine inclusion in basalts, Avacha; 4 = the same for another sample (Kamchatka); 5 = dunite inclusion in basalts, Ross Island (Antarctica); 6 = peridotite inclusion in basalts, Sverre volcano; 7 = lherzolite inclusion; 7a = the same sample without pyroxene fraction (coll. by Yu.P. Burov, Spitsbergen); 8 = xenolith from Quaternary basalts (coll. by Jacobson, Iceland).

Samples of country rocks; 13 = peridotite; 14 = basalt; 15 = obsidian; 16 = pumice (Kamchatka). For sample 15 and 16, 0°–600°C interval is to be taken instead of 300°–600°C as shown in the Table.

Other samples: 19–19b = spodumene, repeated runs; 26–28 = helium from quartz crystalline lattice; chamber pegmatite from Korosten pluton (Volyn); A = earth's atmosphere.

constant isotope ratio in temperature fractions, may by assigned to a second group in which the activation energies of both "helium types" are similar, as may result, for example, from a scattered homogeneous distribution of uranium and thorium in the samples. It is important to note that the amounts of helium in separate temperature fractions in these samples varies by about an order of magnitude (sample 5), a factor significant for the problem of the origin of the helium (cf. *3.2.2.*).

3.2. Now we consider the origin of high-ratio helium in volcanic gases and ultrabasic xenoliths. The main working hypotheses to explain this phenomenon were proposed earlier by Mamyrin et al. [18].

3.2.1. First we will show that processes of radioactive decay and nuclear reactions cannot (on the basis of data available) give rise to helium with a high ratio of $^3He/^4He \sim 10^{-5}$. ^3H and ^3He are characterized by an exclusively low nuclear binding energy and so must not result from radioactive decay. Tritons and ^3He nuclei are known, however, to be emitted by heavy nuclei fission [14, 5]. The tritium yield (^3H_f) from uranium and transuranic isotope fission (up to $^{252}C_f$) is about 2.2×10^{-4}; fission also results in Xe isotopes, the yield being $^{136}Xe = (4.5–6.5) \times 10^{-2}$ [31, 29, 6]. These data allow an estimate of a minimum (as the loss of He occurs easier than that of Xe) ratio of $^{136}Xe_f/^3He_f \simeq 2.5 \times 10^2$. Considering the ^3He abundance in xenoliths of $30 \times 10^{-14} cm^3 g^{-1}$ (Table 2), the ^{136}Xe abundance must be not less than $10^{-10} cm^3 g^{-1}$. This value is at variance with results obtained by Shukolyukov and Tolstikhin [30] and Funkhouser and Noughton [8]; these authors have measured considerably lower Xe concentrations in ultrabasic rocks and xenoliths, i.e. $^{136}Xe < 10^{-12} cm^3 g^{-1}$. It is also possible to show that the absolute abundance of ^3He_f resulting from fission of ^{244}Pu is considerably less than that necessary for such an explanation. Thus ^3He production resulting from a known heavy nuclei fission cannot give the isotopic anomaly observed in xenoliths.

In the last few years many investigators have tried to find in nature a super-heavy transuranic element (SH). According to theoretical prerequisites, this element ($A \sim 300, Z \sim 110–114$) has to decay mainly by fission with a half-life of $T_{1/2} \sim 10^8$ yr. If one uses a super-heavy element fission process for an explanation of a high $^3He/^4He$ ratio in the earth's matter, then the tritium yield (^3He) with respect to xenon has to be at least 100 times that of known transuranic isotopes. The initial concentration of $C_{SH} \sim 10^{-3}$. $C_{^{244}Pu}$ shown by Schramm and Fowler [28] is inconsistent with the ^3He contents measured (with regard to losses) (cf. *3.2.3*), even if a triton forms in each act of fission.

Thus recent estimates for the properties and abundance of a hypothetical transuranic element do not provide a reasonable explanation of the helium isotope anomaly, but they are not sufficient to make a final decision on this problem.

Now let us consider the contribution of nuclear reactions which may take place under natural conditions to give the light helium isotope.

(a) *Neutron reactions.* In 1955 Morrison and Pine [23] showed that the isotope ratios of $^3He/^4He$ measured earlier by Aldrich and Nier [1] in some minerals (beryl, spodumene) and in natural gases may be explained as follows: (1) the radioactive decay of uranium and thorium yields α-particles, the majority of which are stopped by the matter of a mineral, they acquire two electrons, and turn into ^4He atoms; (2) a part of the α-particles react with nuclei in the minerals, mainly with nuclei of light elements, and some of these interactions are (α, n) reactions, giving rise to the appearance of a neutron flux in the rocks; neutrons also appear from the spontaneous and neutron-induced fission of uranium isotopes; (3) a part of the neutron flux reaches epithermal energies, and the reaction of these neutrons with a nucleus of the light isotope of lithium gives ^3He:

$$^6Li + n \rightarrow \alpha + T\, ; T \xrightarrow{\beta^-} {}^3He \quad (1)$$

This explanation was later developed in full detail in the book by Gorshkov et al. [11]; in particular, they refined the estimate of neutron yield from interaction of α-particles with light element nuclei.

The comparison of calculated results following this scheme and the measurements of helium isotope composition from particular rocks was made for the first time by Gerling et al. [10]. The authors showed: (1) the measured ratios of $^3He/^4He$ in rocks of granitic type are close in value to those calculated, and vary over the interval of $10^{-8} + 10^{-7}$; (2) the helium isotope composition of most natural gases varies over the same interval; (3) the experimental data as well as the neutron reactions suggested that helium with a high isotope ratio cannot be formed in the matter of ultrabasic rocks. We carried out an additional experimental test of this conclusion, viz. samples of ultrabasic rock were irradiated by a neutron flux, and after a year tritium contents and ^3He were measured. The results obtained (Table 3) imply the absence of neutron reactions leading to higher tritium (^3He) yields than in reaction 1.

It is possible to assume that in the earliest stages of the earth's evolution there was a neutron flux not related to (α, n) reactions, for example, a flux generated by the fission of a transuranium isotope. ^{244}Pu, traces of which were found in meteorites [37, 16] and recently in the earth's matter [12], is the most suitable for such an explanation. It is important to note that $\lambda_f/\lambda_\alpha = 3 \times 10^{-3}$ for ^{244}Pu, which is 5300 times higher than for ^{238}U. The contribution of ^{244}Pu to the process of ^3He formation may be estimated. For the calculation we take the abundance in the mantle of $U = 3.3 \times 10^8$

TABLE 3
Tritium (T) yield by neutron irradiation of ultrabasic rocks

Sample	Contents (atom g^{-1})			$Li/T_{meas.}$ $\times 10^7$	^3He contents (cm^3 g^{-1}) $\times 10^{-10}$		$^3He/^4H$*** $\times 10^{-6}$
	Li $\times 10^{17}$	T $\times 10^{10}$			calc.**	meas.***	
		calc.*	meas.				
Peridotite	2.85	20	6	0.47	4.2	–	–
Bronzite	2.25	16	4.8	0.47	0.98	1.1	7.4
Peridotite	3.35	28	9	0.37	1.7	2.0	4.0

* Column shows tritium content calculated from the formula;

$T_{calc.} = Li \cdot \sigma_{Li} \cdot \Phi_\Sigma$

where Li is the lithium content in atom g^{-1}; σ_{Li} is the cross-section of interaction between thermal neutrons and lithium, $\sigma_{Li} = 71 \times 10^{-24}$ cm^2; $\Phi_\Sigma = 10^{16}$ n cm^{-2} is the integral neutron flux. Systematic excess of calculated tritium concentrations above measured concentrations is apparently accounted for by the presence in the channel of epithermal neutrons.

** Column shows ^3He content that should have been formed in samples owing to $T_{meas.}$ decay in a time t = 1 yr, calculated from the formula:

$^3He_{calc.} = T_{meas.}(e^{\lambda t} - 1)$

where λ is the constant of the tritium decay, = 0.056 yr^{-1}.

*** Helium analyses were performed a year after irradiation of samples. Prior to irradiation ^3He content was low, about 10^{-12} cm^3 g^{-1}. It is essential that: (a) $T_{meas.}$ content is low; (b) the ratio $Li/T_{meas.}$ is nearly constant; and (c) $^3He_{meas.}$ content was accumulated due to tritium decay only.

g per gram and Th = 12×10^{-8} g per gram (Wasserburg et al. [38]; the ratio of $^{244}Pu/^{238}U = 0.03$ in the primary matter; an age of the earth of $t = 5 \times 10^9$ yr. The integral neutron flux $^{244}\Phi$ resulting from the fission of ^{244}Pu may be calculated from the formula:

$$^{244}\Phi = \frac{\bar{\nu} \cdot \lambda_f \cdot N_{Pu}}{(\lambda_f + \lambda_\alpha) \cdot \Sigma_i N_i \cdot \sigma_i} \quad (2)$$

where ν is the number of neutrons formed by the fission of ^{244}Pu, $\bar{\nu} = 2.5$; $\lambda_f/(\lambda_f + \lambda_\alpha)$ is the fraction of plutonium atoms which decay by fission, where $\lambda_f = 0.28 \times 10^{-10}$ yr^{-1} and $\lambda_\alpha = 0.1 \times 10^{-7}$ yr^{-1}; N_{Pu} is the content of ^{244}Pu, $N_{Pu} = 0.03 \times U = 2.5 \times 10^{12}$ atoms g^{-1}; N_i is the number of nuclei per gram and σ is the neutron capture cross-section in a nucleis; $\Sigma_i N_i \sigma_i \gtrsim 4 \times 10^{-3}$ cm^2 g^{-1}.

Substituting numerical values into this formula, we get $^{244}\Phi \lesssim 4.4 \times 10^{12}$ n cm^{-2}. Assuming that all the fission neutrons reached thermal energy, we get that the following amount of ^3He:

$$^3He = T = {}^{244}\Phi \cdot Li \cdot \sigma_{Li} = 3 \times 10^{-12}\ cm^3 g^{-1} \quad (3)$$

could be formed in matter of chondrite compostion (Li = 2.6×10^{17} atoms g^{-1}) over the time equal to the age of the earth. Concurrently, owing to α decay of uranium and thorium, there formed;

$$^4He = 68 \times 10^{-6}\ cm^3\ g^{-1} \quad (4)$$

Combining eqs. 3 and 4 we get (by "ideal" retention of helium) the ratio of $^3He/^4He = 4.4 \times 10^{-8}$ (100–1000 times less than that measured in the xenoliths).

(b) *α-particle reaction.* Calculations show that a single possible reaction under natural conditions [15];

$$^7Li + \alpha \rightarrow {}^8Be + T$$
$$T \xrightarrow{\beta^-} {}^3He \quad (5)$$

give rise to helium production with the isotope ratio of $^3He/^4He \sim 10^{-10}$[10].

As a check, three samples of ultrabasic rocks from Monchegorsk pluton ($^3He/^4He = 2.5 \times 10^{-7}$) were irradiated in a cyclotron by He^+ with the isotope ratio of $^3He/^4He = 2 \times 10^{-8}$, energy E_α = 10 MeV, and a flux value of $\Phi_\alpha \simeq 5 \times 10^{15}$ α cm^{-2}. Then the tritium content and isotope composition of helium were determined in the irradiated samples. The following ratios were obtained:

$$T/\alpha < 7 \times 10^{-9} \quad (6)$$
$$(^3He/^4He) \text{ before irrad.} > (^3He/^4He) \text{ after irrad.}$$

Thus α-particle reactions in matter close in composition to the xenoliths do not result in a marked appearance of tritium (^3He). A quantitative estimate of T yield resulting from α-particle reaction was not obtained as the flux was too low.

(c) *γ-quanta reaction.* One reaction is known [15] that may produce T under natural conditions:

$$^7Li + \gamma \rightarrow {^4He} + T \quad (7)$$

This reaction has a threshold $E_n = -2.5$ MeV and a quite low cross-section of $\sigma_\gamma < 0.02$ millibarn. It can proceed only on γ-quanta emitted by the isotope ThC'' ($E_\gamma = 2.62$ MeV).

It is possible to show that with the above (*3.2.1.* (a)) abundances of uranium, thorium and lithium, this reaction in matter similar in composition to ultrabasic rocks may in a time equal to the age of the earth lead to an accumulation of the following amount of ^3He:

$$^3He = T \simeq 10^{-17} \text{ cm}^3\text{g}^{-1} \quad (8)$$

Comparing eq. 8 with 4 we get:

$$^3He/^4He \sim 10^{-13} \quad (9)$$

which is negligible compared to the measured ratio in xenoliths.

(d) *Muon reactions.* Takagi [32, 33] tried to explain isotope anomalies of rare gases by the interaction of muons with the earth's matter. According to Takagi, a light isotope of helium is formed by the interaction of a muon with a rock building element with a cross-section of about 2.4×10^{-29} cm^2.

Experimental data on muon absorption in the earth's matter consistent with theoretical results [2] imply a rapid decrease of muon flux as depth increases. At depths 3–5 km the muon flux is about 10^{-8} times that on the surface. This factor may be used as a basis for testing Tagaki's assumption, viz. if ^3He resulted from muon interactions with the earth's matter, its content in rocks at depth should have been much (by many orders) less than in rocks of almost the same age occurring nearer to the earth's surface.

The experience available in the field of isotope geochemistry of helium suggests the absence of any relation between ^3He content and the depth of sample occurrence. Table 2 provides an example of ^3He content in the crystal lattice of quartz from chambered pegmatites of Volyn formed at a depth of about 3 km, while at present, owing to erosion, they are actually on the surface; the age of pegmatites is 1.8×10^9 yr. Thorough investigations of quartz from chambered pegmatites of Volyn's showed [36] that they were characterized by extremely well-retained helium; estimates of diffusion coefficient of He in crystalline quartz gave values of an order 10^{-19} cm^2 sec^{-1}. If we follow Takagi, then negligible (experimentally nomeasurable) contents of ^3He must occur in xenoliths formed in the mantle at depths of about 20 km and more, the (V + Th)/He age of which is only ~10^8 yr or less.

Actually almost the same concentrations of this isotope were found in xenoliths as in quartz crystals (and sometimes higher). Thus, it is possible to state that muon reactions have not contributed greatly to the production of ^3He, at any rate during the last 2–3 b.y.

Hence, the known nuclear processes (radioactive decay, fission, nuclear reactions) cannot explain the production of helium with the ratio found in xenoliths and volcanic gases.

3.2.2. We consider now the proposed earlier assumption about the origin of high-isotope ratios as due to differentiation of "radiogenic" helium isotopes by their migration under natural conditions. The experimental data obtained are at variance with this assumption and make it improbable.

Gerling et al. [10] showed that the expected isotope ratio of "radiogenic" helium in an ultrabasic rock was $(^3He/^4He)_{rad} = 1 \times 10^{-8}$; this estimate seems reasonable in the light of the above discussed (*3.2.1*) data. The maximum content of ^4He in the mantle matter must be $\sim 68 \times 10^{-6}$ cm^3 g^{-1}. Multiplying the two numbers, we get the maximum content of $^3He_{rad} = 68 \times 10^{-14}$ cm^3 g^{-1} that should be accumulated in the mantle matter in a time equal to the age of the earth. Conclusions following from the comparison of above calculated and the measured (Table 2) contents of ^4He and ^3He are highly improbably, viz. ^3He in xenoliths is actually retained completely (in some samples – 3, 5 and 6 – its content even higher than "the maximum possible")while there was almost a complete ^4He loss, more than 99% (in most samples about 99.9%); in other words xenoliths lost only ^4He. The estimates presented are true also for particular contents of uranium in samples studied (Table 3).

This conclusion is at variance with numerous data (those presented in Table 2 included) on migration of rare gases, viz. losses of ^4He must be accompanied and are accompanied by losses of ^3He (especially at the high temperatures in the mantle). So, the high ^3He content in ultrabasic xenoliths allows the conclusion that the isotope anomaly of helium cannot be related to isotope differentiation.

3.2.3. In the light of the experimental data available, the most attractive explanation for the nature of high-ratio helium in the mantle is that helium found in xenoliths (cf. *3.1.*) resulted from the mixture of radiogenic helium $(^3He/^4He)_{rad} = 10^{-7}-10^{-8}$ and primordial helium $(^3He/^4He)_{primord} = (2-4) \times 10^{-4}$ which was captured by the earth during its formation. Primordial helium was found for the first time by Gerling and Levskiy in 1956 [9] in the meteorite "Staroe Pesyanno" and later in other meteorites. The measured variations in isotope composition of the helium in xenoliths may be attributed to different addition of radiogenic helium or different thermal histories of samples resulting in different losses of rare gases. It is characteristic that high-ratio helium occurs not only in xenoliths of Kamchatka, but in Antarctica, Spitsbergen and Iceland, i.e. the earth's mantle within certain limits is "homogeneous" with respect to primordial helium*.

The isotope composition of helium in xenoliths is intermediate between radiogenic and primordial helium; the ratio of $^3He/^4He \simeq 10^{-5}$, most typical of volcanic gases and xenoliths, could have been produced by means of mixture of radiogenic and primordial helium in the ratio:

$$He_{rad}/He_{primord} \simeq 30/1 \qquad (10)$$

* An interesting anomaly in the isotope composition of helium dissolved in waters of the Pacific was found by American and Canadian investigators [3, 4]. However the origin of a relatively small excess of ^3He in oceanic water is now uncertain. Difficulties in obtaining an unambiguous interpretation of these data are attributed to the fact that in both "sections" in the northern and southern Pacific, the excess content of ^3He is confirmed to one and the same small depth at about 2 km.

The measured contents of helium in the xenoliths averages about 10×10^{-8} cm^3 g^{-1}, which is in good agreement with the results obtained by Funkhouser and Noughton [8]. Taking into consideration eq. 10, it is possible to estimate the primordial helium abundance in the mantle at present, i.e. $He_{primord} > 3 \times 10^{-9}$ cm^{3}g^{-1} If the values for uranium and thorium in the mantle proposed by Wasserburg et al. [38] are correct, then in the time of the earth's existence about 70×10^{-6} cm^3 g^{-1} of radiogenic helium should have appeared. Hence the minimum amount of primordial helium which should have been captured by the earth during its production was He $\simeq 10^{-5}-10^{-6}$ cm^3 g^{-1}. This value is not extremely large, as in some meteorites the primordial helium content reaches 0.02 cm^3 g^{-1} while in carbonaceous chondrites, the helium content varies usually from 10^{-5} to 10^{-4} cm^3 g^{-1}.

3.3. The (U + Th)/He age (cf. Table 4) was determined for three samples of xenoliths. Uranium was determined by the fission track method, the content of thorium was taken as equal to the trebled content of uranium. The values of the age obtained are higher than the time during which the sample was on the earth's surface (not more than 10^6 yr). It means that the sample contains mainly deep-seated helium retained in the period of abrupt drop of temperature and pressure at the moment of xenolith outburst. Similar and higher values of (U + Th)/He age were obtained by Funkhouser and Noughton. [8] for ultrabasic inclusions in lavas of Hawaiian volcanoes.

TABLE 4
Helium and uranium in xenoliths

No. of sample	He (cm^3 g^{-1}) $\times 10^{-8}$	U (g per g) $\times 10^{-9}$	Th (g per g) $\times 10^{-9}$	Age (10^6 yr)
3	10	5.9	18	71.5
5	30	53	150	27
7	10	48	150	10

4. Conclusion

In the earth sciences which deals with the reconstruction of the events occurred at a very distant time and place, under conditions that have no parallels in human experience, it is doubtful to speak about "exact and final" solutions of some questions or problems. It is reasonable to state only that our ideas correspond to the majority of the available observations, experimental and theoretical data, etc.

Taking into consideration the above mentioned, two main conclusions follow from our study:

(1) the earth's mantle is the source of helium with a high isotope ratio of $^3He/^4He \simeq 10^{-5}$.

(2) mantle helium was formed by the mixture of radiogenic helium ($^3He/^4He \simeq 10^{-8}$) and primordial helium ($^3He/^4He = 3 \times 10^{-4}$). The latter was captured by the earth in the time of its formation and was retained in the deep earth's interior.

The presence of helium with a high $^3He/^4He$ ratio in the earth's mantle may be of importance for the solution of some geochemical problems such as: (a) the early history of the earth; (b) the earth's degassing and the evolution of the atmosphere; (c) helium isotope dissipation into space; (d) the determination of mantle genesis of some natural gases and rocks; (e) the distinction of the tectonic dislocations providing matter transport from the mantle, etc.

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