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Cosmogenic helium and neon in 11 Myr old ultramafic xenoliths: Consequences for mantle signatures in old samples

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[1] The helium and neon isotopic compositions of olivines coming from a 11 Myr old xenolith sampled at Mt. Hampton (West Antarctica) were analyzed by crushing. The helium isotopic ratio varies between 1340 and 6300 (R/Ra between 115 and 539) with ^{4}He content around 3–8 10^{-10} ccSTP/g, confirming that cosmogenic helium can be extracted by crushing. The neon also shows a clear cosmogenic origin (^{20}Ne/^{22}Ne down to 7.7 and ^{21}Ne/^{22}Ne > 0.32), indicating that the cosmogenic neon can also be extracted by crushing out of the olivines. Melting of the powder left after the crushing experiment gives a ^{4}He/^{3}He ratio as low as 42 ± 8 (R/Ra = 17,300) and ^{21}Ne/^{22}Ne as high as 0.78, close to the cosmogenic production end-member. This study shows that up to ~0.5% of cosmogenic helium and neon can be extracted by crushing. In this way, for samples that had been exposed to cosmic rays for a long time (e.g., a few Myr), a crushing procedure may not give the mantle ratios without ambiguity, and measurement of neon can help to discriminate between cosmogenic and mantle origin of the ^{3}He.

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1. Introduction

[2] Interpretation of helium isotopic ratios in subaerial lavas has always been the object of suspicion when the samples are aged (>1 Myr) [*Bernstein et al.*, 1998; *Hilton et al.*, 1999; *Ikeda et al.*, 2001; *Kirstein and Timmerman*, 2000; *Matsumoto et al.*, 2002; *Richard et al.*, 1996; *Stuart et al.*, 2003]. For noble gas measurements, the most reliable phase in lavas that erupt at the surface is olivine because of its low U and Th contents and because it contains magmatic helium within fluid or melt inclusions [*Kurz et al.*, 1982]. However, the helium content is small (<5 10^{-12} mol/g ^{4}He), and therefore olivine is sensitive to posteruption processes such as alpha particles implantation from the host lava or ^{3}He production by interactions of cosmic-related neutrons with Mg, Fe, Si and O atoms [*Kurz*, 1986]. Moreover, helium diffusivity in minerals may limit the study in old samples. One technical solution to avoid these posteruption effects is to crush the samples in vacuum in order to release the gas from

melt or fluid inclusions (mantle derived). This procedure generally assumes that the cosmogenic atoms (e.g., ^{3}He, ^{21}Ne), that are located within the olivine matrix, are not extracted during crushing. Only melting of the sample is believed to release the cosmogenic component. This has been checked for relatively young samples (<100,000 years) and careful sampling (e.g., road cut or samples from 1 m deep) usually allows the correct attribution of a measured helium isotopic ratio to a mantle-derived ratio. However, this may not be the case for samples with possible long exposure history (>1 Myr) even for samples picked at more than one meter in depth because of the muon-induced production [*Gosse and Phillips*, 2001]. In fact, despite the absence of available data for noble gases, this effect is expected to look similar to that observed for other cosmogenic nuclides as described by *Heisinger and Nolte* [2000]. Moreover, as demonstrated by *Hilton et al.* [1993] and *Scarsi* [2000], damages in mineral structure produced by crushing may allow the extraction of cosmogenic helium. To overcome this problem, the authors suggested that step-crushing is the correct method to extract the mantle-derived helium when one considers only the first step of crushing. More recently, *Yokochi et al.* [2005] have shown that samples from Japan (olivines from ultramafic xenoliths), aged ~1 Myr, release a large proportion of cosmogenic helium (up to 25%) during crushing, modifying the initial (mantle) ^{4}He/^{3}He ratio from 90,000 to 9,000 (R/Ra from 8 to 78). The present note confirms the results of *Hilton et al.* [1993], *Scarsi* [2000], and *Yokochi et al.* [2005] for helium, and also extends this phenomenon to the cosmogenic neon. Our results on neon confirm that diffusion is not the process that extracts helium during crushing.

2. Sample Location and Analytical Procedure

[3] The sample analyzed in this note (PK 91005) comes from Mt Hampton, located in the Executive Committee Range in West Antarctica (76°30S 126°W). It was given to us by Stanley Hart from the Woods Hole Oceanographic Institution. Mount Hampton is the northernmost of the five major volcanoes in the Executive Committee Range and is ~11 Myr old [*Handler et al.*, 2003]. The altitude of its summit is 3323 m. The exact location of the sample (altitude) is not known. PK91005 is a lherzolite xenolith with green olivines that do not show the presence of fluid or melt inclusions. Olivines of 1–2 mm were picked under a binocular and either leached with HF (30′ with 5N) or just cleaned with ethanol and acetone. Olivines were introduced in the crusher and baked at 100°C over one night. For one experiment, the crusher with the sample was not baked the night before analysis (indicated by a * in Table 1). Crushing is performed manually using a magnetic ball moved using a magnet. The number of strokes varies between 150 and 700; 700 strokes allow a perfect crushing (grain size ~ micron), whereas for 150 strokes some grains are a few micrometers large. Released gas was then purified using either one hot Ti getter (ARESIBO I) or two (ARESIBO II) and a SAES getter at room temperature. Noble gases were trapped on charcoal at a temperature of ~10K. Helium and neon were analyzed successively by desorbing the helium and neon at 25K and 70K respectively and introduced in our mass spectrometers ARESIBO I or II [*Staudacher and Allègre*, 1982]. ^{4}He was analyzed on a faraday cup whereas ^{3}He and neon isotopes were measured with an electron multiplier equipped with an ion counting system. Corrections of HD^{+}, $^{40}Ar^{++}$ and CO_2^{++} were applied on ^{3}He, ^{20}Ne and ^{22}Ne, using the HD^{+}/H_2, $^{40}Ar^{++}/^{40}Ar^{+}$, CO_2^{++}/CO_2^{+}ratios of 2 10^{-4}, 0.15 and 0.001 for ARESIBO I and 0, 0.11 and 0.001 for ARESIBO II. H_2 in the two mass spectrometers were around 10,000 cps, which implies HD^{+} around 2–3 cps at mass 3.016. For ARESIBO II, a special procedure was applied with the ^{3}He measured at mass 3.014 rather than 3.016, which implies no HD^{+} correction. $^{40}Ar^{+}$ at the time of the measurement were 100 cps for ARESIBO I and 10,000 for ARESIBO II during neon analyses. CO_2^{+} was ~1000 cps for ARESIBO I and ~4000 for ARESIBO II. Crusher blanks were ~2 10^{-10} ccSTP ^{4}He and ~10^{-13} ccSTP of ^{22}Ne for ARESIBO I with the new fully automated line dedicated to helium and neon measurements. For ARESIBO II, blanks were ~2 10^{-9} ccSTP ^{4}He and 3 10^{-13} ccSTP of ^{22}Ne.

[4] The powder of two samples (one that was leached with HF and another that was not), packed in an aluminum foil of ~0.06 g, was heated at 1500°C for 5 min in a glass furnace with Mo crucible and analyzed for helium and neon. Helium blank at this temperature was 2 10^{-9} ccSTP. The ^{22}Ne blank was 5 10^{-12} ccSTP with isotopic ratios close to air. Postfractions were performed to check

Table 1. Helium and Neon Results[a]

Sample	^{4}He	^{3}He	^{22}Ne	^{4}He/^{3}He	R/Ra	^{20}Ne/^{22}Ne	^{21}Ne/^{22}Ne	^{21}Nec
				Crushing				
Aresibo I								
(HF, 1.2686g 150x)	0.3	0.05	0.14	6280 ± 550	115 ± 10	9.32 ± 0.14	0.068 ± 0.005	0.01
(0.9904g 700x)	0.5	0.25	1.1	1990 ± 90	363 ± 17	9.14 ± 0.07	0.114 ± 0.003	0.08
(HF, 0.7899g 550x)	0.4	0.30	0.29	1340 ± 80	539 ± 33	7.74 ± 0.22	0.323 ± 0.028	0.07
Aresibo II								
(0.9513g, 300x)* step1	0.8	0.20	0.95	3990 ± 350	181 ± 16	10.11 ± 0.15	0.061 ± 0.003	
(0.9513g, 300x)* step2	0.3	0.12	0.19	2500 ± 440	289 ± 51	8.76 ± 0.49	0.159 ± 0.029	0.02
(HF, 0.4461g, 200x)	<0.06	0.08	0.49	-	-	9.34 ± 0.40	0.092 ± 0.016	0.03
				Heating				
(0.600g 1500°C)	8.3	72 ± 8	na	116 ± 12	6231 ± 660			
(0.205g 1500°C)	4.5	79 ± 20	27.1	57 ± 14	12700 ± 3000	2.89 ± 0.37	0.74 ± 0.08	21
(HF 0.733g 1500°C)	3.0	59 ± 5	na	51 ± 3	14210 ± 880			
(HF 0.395g 1500°C)	3.0	71 ± 14	18.8	42 ± 8	17300 ± 3200	2.54 ± 1.15	0.78 ± 0.12	15

[a] Here, ^{4}He is in nccSTP/g, and ^{3}He, ^{21}Ne$_c$, and ^{22}Ne are in pccSTP/g. Star indicates a sample that was not baked during the night. When HF is indicated, that means samples were leached during 30 min at 7N HF. ^{21}Ne$_c$ was obtained by assuming a ^{21}Ne/^{22}Ne ratio of 1 for the cosmogenic end-member.

the complete extraction of helium and neon at this temperature.

3. Results and Discussion

[5] Helium and neon results are given in Table 1 and are represented in Figures 1 and 2. The helium concentration obtained by crushing is very low compared to typical xenolith samples (3 to 11 10^{-10} ccSTP/g) (when two crushing steps were performed, the sum of concentrations was considered), whereas the isotopic ratio is among the lowest measured on Earth (^{4}He/^{3}He between 1340 and 6300 (R/Ra between 115 and 539). Such values cannot be attributed to a mantle ratio (^{4}He/^{3}He > 15,000; R/Ra < 50) but rather to a cosmogenic component that is extracted by the crushing experiment. This is demonstrated by neon isotope signature, that clearly shows a non-mantle component [*Honda et al.*, 1991; *Moreira and Allègre*, 1998; *Moreira et al.*, 1995, 2001; *Sarda et al.*, 1988] (Figure 2). Such neon isotopic ratios (below the air ^{20}Ne/^{22}Ne ratios) are either representative of a nucleogenic or a cosmogenic component. Since it was analyzed in olivines, which typically have low U and Th concentrations (being therefore a low alpha particle producer), we exclude the nucleogenic origin and rather prefer the cosmogenic origin (as also illustrated by the high ^{3}He concentration). Moreover, the neutron flux, produced in the surrounding basalt, that may induce ^{24}Mg(n, α) reactions, will give a maximum nucleogenic ^{21}Ne content of 5 10^{-15} ccSTP/g in 11 Myr [*Yatsevich and Honda*, 1997], negligible compared to the ^{21}Ne in excess measured in our sample (>10^{-13} ccSTP/g). This interpretation is confirmed by the measurements proceeded by heating where the ^{4}He/^{3}He ratio is as low as 42 ± 8, close to the cosmogenic production ratio (~2 in meteorites [*Hoffman and Nier*, 1958]). The neon isotopic ratios obtained by heating are also very close to the cosmogenic production ratios of 0.8 in terrestrial quartz [*Niedermann et al.*, 1993].

[6] Our study on this mantle-free sample shows that the cosmogenic helium extracted by crushing may represent up to 0.5% of the total cosmogenic helium located in the matrix (Table 1, Figure 3).

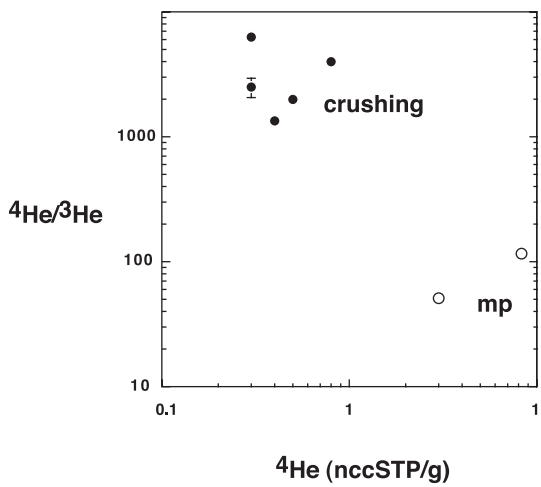

Figure 1. The ^{4}He/^{3}He ratios (and R/Ra) versus [^{4}He] in nccSTP/g for the different duplicates of PK91005 and crushing steps as well as for the melting of the powder (mp).

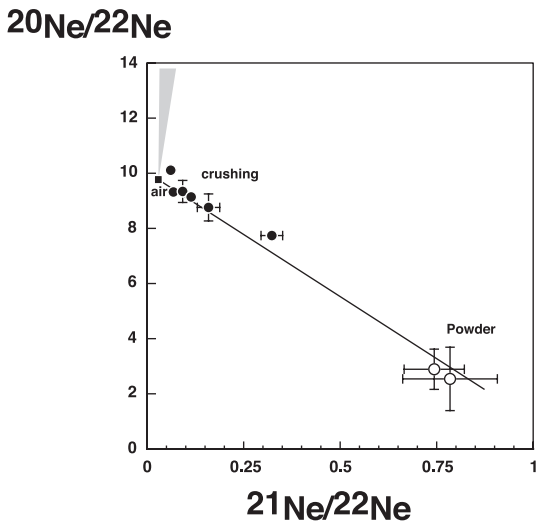

Figure 2. Three isotope neon diagram showing the results for PK91005 sample. Black dots, crushing; white dots, melting of the powder. The mantle domain is represented by the gray area.

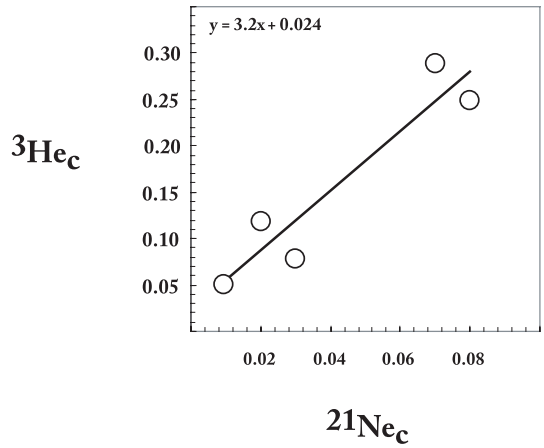

Figure 4. Correlation between ^{3}He and ^{21}Nec. This figure suggests that the two cosmogenic nuclides are extracted in the same way, suggesting that diffusion is not the extraction process. Moreover, the slope of the correlation gives a constant $(^3He/^{21}Ne)_c$ ratio of 3.2 during crushing, similar to the same ratio in powder.

Moreover, the number of strokes (e.g., quality of crushing) appears to control the extraction of cosmogenic nuclides (Figure 3). The higher the number of strokes, the higher the number of cosmogenic atoms released by crushing. One should also note that the helium isotopic ratio and concentration are lower when the sample was leached with HF (51 and 42 vs. 57 and 116; Table 1). This suggests that the posteruption radiogenic component (from grain boundaries or host lava), certainly located at the first microns of the olivine surface (alpha implantation), is at least partly removed by leaching.

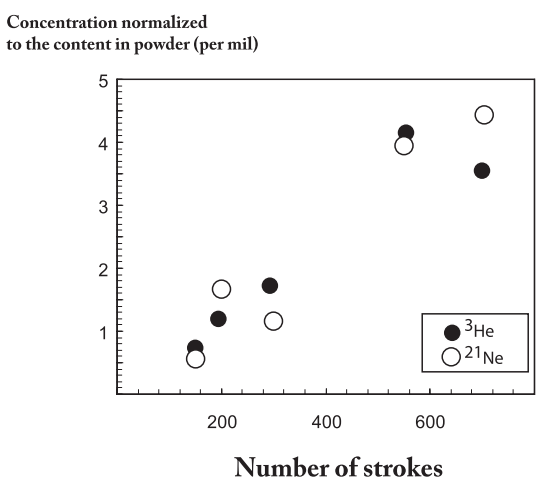

Figure 3. The cosmogenic ^3He_c and $^{21}Ne_c$ as a function of the number of strokes during crushing (normalized by the content obtained by melting of the powder).

[7] The main result of this study is that both cosmogenic helium and neon are extracted by crushing from olivines. As suggested by *Yokochi et al.* [2005] for helium, diffusion during crushing cannot be the process that extracts the helium and neon from olivine (the temperature is lower than 70°C in the crusher during crushing; this temperature was checked by C. Gautheron by using a paint that changes color with temperature on a magnetic ball and on a sample). Therefore another process has to be found. *Yokochi et al.* [2005] proposed a model of fracture-related extraction to explain how cosmogenic helium can be extracted by crushing. We propose here that this model can be extended to cosmogenic neon. Indeed, as illustrated in Figure 4, ^{3}He and $^{21}Ne_c$ are correlated. Moreover, the $^3He/^{21}Ne_c$ ratio (~3.2) obtained during crushing is similar to the ratio within the matrix of the olivine (3.8–4.7), excluding a diffusion process to extract ^{3}He. This rather suggests a gas extraction by destruction of the structure of the olivine, as suggested by *Yokochi et al.* [2005].

[8] As a remark, the mean ^{3}He concentration in the powder is 70 ± 8 pccSTP/g (1sigma), that represents 1.9 ± 0.2 10^9 atoms/g. Using a minimum production rate of 130 at/g/an in olivine at high latitude and sea level [*Ackert et al.*, 2003], the maximum exposure age would be ~15 Myr, consistent with the eruption age (11 Myr) if we consider the sample was not sampled at sea level.

Assuming that the exposure age is 11 Myr, we can derive a possible sampling altitude of 900 m.

[9] A consequence of this study is that the helium and neon measurements in samples with a long exposure history (even at few meters deep) can be delicate to interpret in case of a high measured $^3He/^4He$. This was recently pointed out by *Yokochi et al.* [2005] for samples from Japan that showed relatively low $^4He/^3He$ ratios, that may be attributed to the presence of a mantle plume. In fact, careful measurements by step crushing have proven a cosmogenic origin for these low ratios [*Yokochi et al.*, 2005]. Even melting the powder cannot be conclusive if an important radiogenic component is implanted in the olivines. We have shown in this study that the analysis of neon isotopes can be useful to discriminate between a mantle origin and a cosmogenic origin for helium.

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