



Neon isotopic composition of the mantle constrained by single vesicle analyses



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ABSTRACT

The origin of volatiles on Earth is still a matter of debate. Noble gases are an efficient geochemical tool to constrain Earth formation processes due to their inertness. Several studies have focused on the neon isotopic composition of the lower mantle because the $^{20}\text{Ne}/^{22}\text{Ne}$ ratio is thought to reflect that of Earth's primordial components. Two models to explain the origin of light noble gases on Earth have been proposed: either solar wind implantation onto the Earth's solid precursors or dissolution into the mantle of a primordial atmosphere captured from solar nebula gas. In order to test these two models, we analyzed the noble gas compositions (helium, neon and argon) of two submarine oceanic island basalt glasses from Fernandina volcano (Galápagos archipelago), which have among the most primitive/unradiogenic terrestrial helium and neon isotopic compositions. Several sample pieces are studied both by step-crushing and by laser ablation analyses of single vesicles. Results of step-crushing are consistent with those of laser ablation analyses, but the latter results provide new insights into the origin of atmospheric contamination. The single-vesicle laser-ablation measurements overlap with the step crushing results, but have systematically higher $^{40}\text{Ar}/^{36}\text{Ar}$, and $^3\text{He}/^{36}\text{Ar}$, suggesting less atmospheric contamination using this method. The single vesicle data therefore suggest that atmospheric contamination is introduced by exposure to the modern atmosphere, after sample collection. $^3\text{He}/^4\text{He}$ values are about 23 times the atmospheric ratio (R/Ra) for the two Fernandina (Galápagos) samples, in agreement with previous studies. We obtain $^{20}\text{Ne}/^{22}\text{Ne}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ isotopic ratios as high as 12.91 and 9400, respectively, for the mantle source of the Galápagos hotspot. The new data show that step-crushing and laser ablation analyses are complementary methods that should be used together to derive the noble gas ratios in uncontaminated samples. The results of neon compositions are consistent with previous hotspot studies and support the model of solar wind implantation associated with sputtering to explain helium and neon origins on Earth.

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1. Introduction

The origin of volatiles on Earth is still a matter of intense research, but is fundamental to understanding Earth and atmosphere formation processes. Noble gases are important tools to address this problem due to their chemical inertness. In particular, neon gives precious information on that question. Neon has three iso-

topes of masses 20, 21 and 22, which can all be produced by nucleogenic reactions (e.g. $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$). However, the production rates of isotopes ^{20}Ne and ^{22}Ne are negligible in the terrestrial mantle (Yatsevich and Honda, 1997), so the $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of the mantle reflects the primitive neon composition. Many studies have put forward that the mantle $^{20}\text{Ne}/^{22}\text{Ne}$ ratio is higher than 12 and is therefore “solar-like” (Ballentine et al., 2005; Honda et al., 1993; Kurz et al., 2009; Moreira et al., 1998; Mukhopadhyay, 2012; Raquin and Moreira, 2009; Sarda et al., 1988; Tieloff et al., 2000). By comparison, the atmospheric $^{20}\text{Ne}/^{22}\text{Ne}$ ratio is 9.78 ± 0.03 (Sano et al., 2013) and the solar wind ratio, measured on the Gen-

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esis targets, is 13.78 ± 0.03 (1σ) according to Heber et al. (2009) or 14.001 ± 0.042 (1σ) according to Pepin et al. (2012). The solar wind is isotopically fractionated compared to the Sun, which, by calculation, has a $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of 13.34 using an inefficient Coulomb drag model (Heber et al., 2012).

It is essential to precisely measure the neon composition of the lower mantle. Indeed, the lower mantle must reflect a less degassed reservoir than the upper mantle and still contains primordial noble gas components, in particular primordial helium (^3He) and neon (Allègre et al., 1983; Kaneoka and Takaoka, 1980; Kurz et al., 2009; Valbracht et al., 1997). In contrast, the upper mantle is considered to be more degassed of its primordial noble gases and should be more sensitive to subduction of atmospheric noble gases. Only a few hotspots are well-suited to determine the lower mantle noble gas composition, namely Hawaii, Galápagos and Iceland, because fresh submarine glass samples, erupted at great depths, are required to obtain accurate neon analyses. Submarine glasses are among the best samples for recording the quenched magma (mantle) noble gas composition directly after eruption.

Several studies have focused on the lower mantle composition and have led to two opposing models for the origin of noble gases on Earth. Trielloff et al. (2000) found that oceanic island basalts (OIB) from Hawaii and Iceland showed a mean $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of 12.49 ± 0.06 (1σ), very close to that of the Neon B component observed in gas-rich meteorites and on the lunar soil (Black, 1972). They argued that since there were no extant terrestrial measurements higher than ~ 12.5 , Neon B was the likely terrestrial end-member, assumed to be derived from solar wind implantation (Black, 1972). Hence, Trielloff et al. (2000) suggested that planetesimals that accreted to form the Earth had suffered solar wind implantation and were then characterized by the Neon B component. However, Yokochi and Marty (2004) measured a $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of 13.04 ± 0.2 (1σ) in plume-related rocks from the Kola peninsula (Russia), which led them to propose that this ratio of 13 would represent a lower limit for the lower mantle neon composition and that terrestrial neon would come from the dissolution of solar nebula gas into the mantle. The latter scenario, also advocated by Mukhopadhyay (2012), implies that the solar nebula gas remained in the accretion disk long enough to permit the capture of a primordial atmosphere, an important temporal constraint for the early Earth. Raquin and Moreira (2009) developed a model that would explain implanted $^{20}\text{Ne}/^{22}\text{Ne}$ ratio, starting from solar wind values of ~ 13.8 , by isotopic fractionation during implantation and erosion on the grain surfaces. In this model, lighter isotopes are preferentially lost from accreting and eroding grains due to their lower energy, shorter implantation depth, and preferential loss by erosion. Raquin and Moreira (2009) were able to derive a steady state $^{20}\text{Ne}/^{22}\text{Ne}$ of ~ 12.5 for accreting particles, using reasonable values of exposure time and erosion rates. However, the timing and geometries (particle sizes) of accretion are poorly known, so higher steady state values (such as 12.7) are possible (Moreira, 2013). In all accretion models, whether gases are introduced by implantation on solid grains or by equilibration with nebular gas, the neon isotopic composition of the Earth's interior is one of very few constraints.

One of the major problems in noble gas geochemistry is the ubiquity of an air-like noble gas component in mantle-derived samples. Step-crushing experiments always consist of mixing between an air-like component and a mantle component as shown by many studies (e.g., Trielloff et al., 2000 and Yokochi and Marty, 2004). Ballentine and Barfod (2000) suggested that this air-like component is due to atmospheric contamination such that air fills the many cracks and microfractures within samples once they are brought to the surface. Conversely, Sarda (2004) suggested that this air-like component could correspond to atmospheric noble gas re-

cycling into the mantle and that the larger vesicles would carry this recycled component. Several studies (e.g., Holland and Ballentine, 2006, Parai and Mukhopadhyay, 2015 and Tucker et al., 2012) have now suggested that both processes (shallow level contamination and recycling through subduction) may occur for heavy noble gases (Ar, Kr, Xe) whereas light noble gases (He, Ne) would not be significantly recycled into the mantle (Holland and Ballentine, 2006). Burnard et al. (1997) and Burnard (1999) introduced laser ablation analyses of single vesicles and never found vesicles filled with air. Raquin et al. (2008) showed that laser ablation analyses can (partly) remove this shallow level atmospheric contaminant and therefore is a promising method to determine the true mantle isotopic composition.

In this study, we focus on OIB samples from one of the most primitive hotspots for helium, the Galápagos hotspot (Kurz et al., 2009; Kurz and Geist, 1999) in order to determine their helium, neon and argon compositions and to constrain which of the two models for the origin of noble gases on Earth is best supported by the data. In addition to step-crushing experiments, we conduct laser ablation analyses on single vesicles. Since OIB samples are not very vesicular, pieces of samples for laser ablation are characterized with X-ray microtomography, a powerful and non-destructive technique, which allows precise location of vesicles in samples.

2. Samples and methods

2.1. Sample selection

Two samples from Fernandina volcano (Galápagos hotspot) collected during the AHA-NEMO2 cruise are studied, samples AHA-NEMO2-D22A and AHA-NEMO2-D22B from a single dredge on the submarine western flank. Details about sample location can be found in Geist et al. (2006); helium and neon data for D22B and a number of other Fernandina samples can be found in Kurz et al. (2009).

These two samples were selected for their thick basaltic glassy pillow lava margins, which allowed the use of intact large (centimeter sized) glass chunks for the laser ablation and step crushing measurements. They are assumed to have recorded the quenched magma composition directly after eruption, without having suffered intense post-eruptive processes (such as slow cooling or crystallization). For such a study, it is best to choose samples collected at great water depth so that vesicle internal pressures are high when the magma is quenched (approximately equal to hydrostatic pressure (Colin et al., 2013)), increasing the likelihood of gas-rich vesicles. The two studied samples were dredged between 2210 and 2390 m deep.

2.2. X-ray microtomography

Before laser ablation analyses, pieces of samples were imaged via X-ray microtomography in order to locate the vesicles. X-ray computed microtomography is a powerful and non-destructive technique, which permits the reconstruction of the 3D volume of geological samples. The analyses were performed at the Institut des Sciences de la Terre at Orléans and at the Institute of Earth Sciences, University of Lausanne.

Sample pieces are first polished so as to make rectangular shapes of 5–8 mm high, 3–5 mm long and 2–5 mm wide. This ensures easy focusing of the laser beam. Details about X-ray microtomography principles and acquisition parameters can be found in Supplementary Information. Using the X-ray microtomography data, the internal 3D volume, and surfaces, of the sample were reconstructed with different softwares (CTvox, ImageJ, etc.) in order to locate vesicles. Fig. 1a shows one piece of the sample

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