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Neon diffusion in olivine and quartz

D.J. Cherniak *, J.B. Thomas, E.B. Watson

Department of Earth and Environmental Sciences, Rensselaer Polytechnic Institute, Troy, NY 12180, USA

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ABSTRACT

Lattice diffusion of neon in single, gem-quality crystals of quartz, forsterite, and natural iron-bearing olivine was determined from experiments using two different methods to introduce Ne. In the first set of experiments, ^{22}Ne was implanted into polished samples, which were then annealed for various times and temperatures in 1-atm furnaces. In the second group of experiments, referred to as soaking experiments, polished slabs of crystals were placed in open containers and inserted into cold-seal type pressure vessels where they were exposed to Ne gas at 340–1340 bars Ne pressure and elevated temperatures. Nuclear reaction analysis, using the reactions ^{22}Ne(p,γ)^{23}Na and ^{20}Ne(α,γ)^{24}Mg for the implantation and soaking experiments, respectively, was used to directly measure Ne distributions in the samples following diffusion anneals. The following Arrhenius relations were obtained for Ne diffusion parallel to c:

$$\text{Forsterite}: D_{\text{Fo}} = 1.4\text{x}10^{-13} \exp\left(-131 \pm 6 \text{ kJ mol}^{-1}/RT\right) \text{ m}^2\text{s}^{-1}$$

$$\text{Quartz}: D_{\text{Qtz}} = 1.6\text{x}10^{-14} \exp\left(-115 \pm 4 \text{ kJ mol}^{-1}/RT\right) \text{ m}^2\text{s}^{-1}$$

There are no significant differences in Ne diffusion along the investigated crystallographic axes in either quartz or olivine, and Ne diffusivities for synthetic forsterite and natural Fe-bearing olivine are similar. In addition, diffusivities determined from the soaking experiments and the implantation experiments are similar, indicating little effect on diffusion of any lattice damage that might be induced by the ^{22}Ne ion implantation. The lack of damage is also confirmed by imaging with high-resolution electron microscopy.

Ne diffusivities in quartz measured in this study are about 10 orders of magnitude slower than those determined from the bulk-release experiments of Shuster and Farley (2005). In the case of olivine, Ne diffusivities in olivine measured in the present study are 1–3 orders of magnitude slower than diffusivities determined by Futagami et al. (1993) and 2–5 orders of magnitude slower than Ne diffusivities determined by Gourbet et al. (2012) over the temperature range of overlap of the respective studies. In addition, activation energies for diffusion differ widely, with the value from of Futagami et al. (1993) significantly lower (87 kJ/mol) than that measured in the present work, and the preferred values of Gourbet et al. (2012) (360–370 kJ/mol) significantly higher than our findings. These differences may reflect the influence of factors that may disproportionately affect measurements of Ne diffusion determined through bulk-outgassing methods compared with direct profiling techniques measuring lattice diffusion such as those employed in this study.

Our data indicate that both quartz and olivine may be very retentive of Ne at temperature conditions typical near the surface of the earth, with 100 μm radius quartz and olivine grains experiencing less than 1% Ne loss over times on order of the age of the earth at 100 and 150 °C, respectively. Under mantle conditions, effective diffusion distances for Ne in olivine would be about 1 m and 0.2 m over 1 billion years at 1550 and 1100 °C, respectively. In contrast to Ne, helium is much more mobile, with effective diffusion distances of 1500 m over 1 billion years at 1500 °C, compared with 0.8 m for Ne under the same time–temperature conditions.

1. Introduction

Measurements of neon isotopes in minerals have applications in many areas of earth and planetary sciences, including thermochronometry, geomorphology, atmospheric formation, mantle geochemistry, and cosmochemistry.

In minerals, ^{21}Ne and ^{22}Ne can be produced in situ by nuclear reactions with cosmic rays. ^{21}Ne is created in surface rocks exposed to cosmic ray fluxes primarily through spallation reactions induced with ^{28}Si by high-energy neutrons, and also through the capture of muons, a component of primary cosmic radiation. Cosmogenically produced

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* Corresponding author. Tel.: +1 518 276 8827.
E-mail address: chernd@rpi.edu (D.J. Cherniak).

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^{21}Ne has been used to evaluate exposure ages of terrestrial materials (e.g., Bruno et al., 1997; Schäfer et al., 1999, 2000; Gosse and Phillips, 2001; Hetzel et al., 2002; Niedermann, 2002; Balco and Shuster, 2009; Kober et al., 2009; Davis et al., 2011) and of meteorites (e.g., Phillips et al., 1998; Wieler, 2002; Bhandari et al., 2005; Kober et al., 2007), date catastrophic events (e.g., Cerling et al., 1994) and determine erosion rates (e.g., Libarkin et al., 2002; Codilean et al., 2008).

^{21}Ne can also be produced from non-cosmogenic sources through alpha-capture, as in the reaction ^{18}O(α,n)^{21}Ne, and ^{22}Ne can be produced from ^{19}F through reactions induced by alpha particles from the decay of ^{238}U, ^{235}U, ^{232}Th and other alpha emitters (e.g., Yatsevich and Honda, 1997; Leya and Wieler, 1998). In turn, neutrons from these and other (α,n) reactions can produce isotopes of ^{21}Ne and ^{22}Ne through the reactions ^{24}Mg(n,α)^{21}Ne and ^{25}Mg(n,α)^{22}Ne, respectively. The vast majority of Ne produced in these alpha-induced reactions is ^{21}Ne, from the alpha capture reaction with ^{18}O. Production of ^{21}Ne through this reaction in accessory minerals and other phases bearing U and Th has potential for use as a chronometer (e.g., Gautheron et al., 2006).

Signatures of Ne in interplanetary dust particles, meteorites and lunar samples give clues to primordial noble-gas compositions (e.g., Ott, 2002; Wieler, 2002). Interplanetary dust particles reaching earth may contribute to the presence of solar-like Ne components in various mantle derived samples (e.g., Hiyagon, 1994). Ne evaluated from basalts (e.g., Nakai et al., 1997; Dixon et al., 2000; Graham, 2002; Doucet et al., 2006; Stroncik et al., 2007), and xenoliths (e.g., Gautheron et al., 2005) can provide information on mantle heterogeneities, offering insight into the processes of formation and evolution of the mantle, and the ratios of He and Ne isotopes can provide important clues to source composition prior to fractionation due to transport and eruption, given the links between ^{21}Ne and ^{4}He through their nucleogenic and radiogenic origins, respectively (e.g., Honda et al., 1993; Pepin and Porcelli, 2002; Jackson et al., 2009). Key to interpretation of these compositional signatures is determining whether Ne heterogeneities are likely retained during subduction and residence in the mantle. Diffusion kinetics are thus critical in influencing the behavior of noble gases in the mantle (e.g., Albarede, 2008), including the distribution and degassing of Ne (e.g., Porcelli and Ballentine, 2002).

Determinations of diffusivities of Ne in minerals provide constraints on whether Ne is retained in a particular mineral phase after its production or introduction, or lost due to heating events. In this study, we examine Ne diffusion in quartz, the mineral most commonly used in cosmic-ray exposure studies, and olivine, a mineral phase that may be critical in influencing noble-gas systematics in the upper mantle (e.g., Yokochi and Marty, 2004; Meibom et al., 2005). Extant diffusion data suggest modest retentivity of Ne in olivine, but activation energies for diffusion and Ne diffusivities differ widely among these studies (Futagami et al., 1993; Gourbet et al., 2012). Previous measurements of Ne diffusion in quartz indicate comparatively fast diffusivities (Shuster and Farley, 2005). To explore Ne diffusion in these minerals, we run experiments using two different approaches – (i) "soaking" experiments, in which samples are annealed in the presence of pressurized Ne gas; and (ii) implantation experiments, where ^{22}Ne is implanted into the minerals. In both cases, Ne distributions in samples following experiments are directly profiled with Nuclear Reaction Analysis (NRA). This analytical approach differs from previous measurements, in which bulk degassing was used to evaluate neon diffusivities. We measure Ne diffusion in both synthetic forsterite, and Fe-bearing natural olivine (Fo$_{90}$) to assess possible compositional effects, and examine the effects of crystallographic orientation on Ne diffusion in both olivine and quartz.

2. Methods

2.1. Experimental procedures

The minerals used in the experiments were clear, gem quality, inclusion-free specimens of natural and synthetic materials (Table 1). The synthetic forsterite (in the form of a boule ~2 × 6 cm) was from the Morion Corporation, and natural Fe-bearing olivine from San Carlos, Arizona (~Fo$_{90}$) (pieces 1–2 cm on a side). The quartz samples were from a synthetic crystal (~3 × 15 cm) and natural specimens from Herkimer, NY and Arkansas (1–2 cm in diameter).

Table 1
Composition of minerals used in experiments.

	Forsterite	Olivine	Quartz
SiO_2 (wt.%)	42.96 (0.27)	40.70 (0.26)	100.5 (0.50)
TiO_2	0.02 (0.02)	tr.	n.d.
Al_2O_3	n.d.	tr.	n.d.
FeO_t	n.d.	11.12 (0.23)	tr.
MnO	tr.	0.16 (0.05)	n.d.
MgO	56.70 (0.26)	47.78 (0.25)	tr.
CaO	n.d.	0.09 (0.02)	n.d.
NiO	n.d.	0.35 (0.08)	n.d.
Cr_2O_3	tr.	0.03(0.03)	tr.
K_2O	n.d	tr.	tr.
Total	99.68	100.22	100.50

Electron microprobe beam conditions used were 15 kV accelerating voltage, 15 nA beam current and 10 μm beam diameter. Numbers in parentheses are 1σ uncertainties based on counting statistics.
n.d. = not detected.
t.r. = <0.01 wt.%.

Oriented specimens were cut with a low-speed diamond saw into slabs measuring ~2–3 mm on a side and ~0.5 mm thick for use in individual experiments. The slabs were hand-polished to a smooth finish using standard SiC polishing paper, followed by polishing with an automated system in a slurry of 1 μm alumina powder in water. The final polishing step achieved an optically featureless finish by polishing with an automated polisher for ~2 h in a 0.06 μm colloidal silica suspension. Measurements taken during preparation showed that the final polishing steps using Al_2O_3 and colloidal silica removed ~90 μm of material to produce absolutely featureless, crack-free forsterite surfaces. The pyramid facets of the quartz crystals were simply cut from crystals by using a low-speed saw.

Two types of experiments were conducted, using different means to introduce neon into the samples: implantation experiments and soaking experiments. Ion implantation has been used in previous noble gas diffusion studies to introduce ^{3}He and investigate He diffusion in a variety of mineral phases, including zircon and apatite (Cherniak et al., 2009), rutile and titanite (Cherniak and Watson, 2011) and olivine (Cherniak and Watson, 2012). Soaking, in which samples are exposed to a pressurized gas, has been used in studies of Ar diffusion in quartz, olivine, enstatite and corundum (Watson and Cherniak, 2003; Watson et al., 2007; Thomas et al., 2008).

Soaking experiments were conducted in cold-seal pressure vessels made of René 41 alloy (buffering the system at ~Ni–NiO) which were run in a horizontal position. Experimental conditions are listed in Table 2. We utilized the thermal gradient along the length of the pressure vessel to heat multiple specimens of each mineral to different temperatures in the same experimental run with a single pressurization. Four boxes were milled into a filler rod made of René alloy. The 5 × 10 mm boxes were lined with a refractory ceramic to prevent mineral specimens from touching the metal; each box has space to accommodate multiple mineral slabs, depending on their dimensions. Mineral slabs prepared as above were loaded into the boxes so that the polished surfaces would be directly exposed to the Ne gas pressure medium, and the filler rod was then loaded into the pressure vessel. The position of each mineral slab was measured from the end of the filler rod. Five thermocouple wells were drilled into the side of the vessel, making it possible to monitor the temperature along the length of the vessel using type-K sheathed thermocouples, which were each connected to digital temperature readouts. Pressure was monitored with a Heise Bourdon tube gauge.