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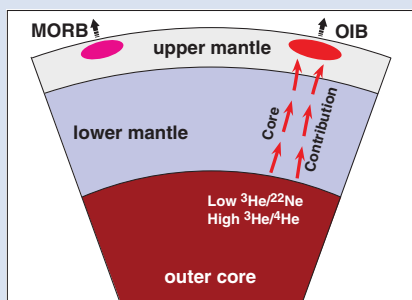
Potential of Earth's core as a reservoir for noble gases: Case for helium and neon

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Abstract



This study investigates metal–silicate partitioning of neon (D_{Ne}) under the likely conditions of early Earth's core formation: up to 16 GPa, ~3000 K and an oxygen fugacity near IW-2 (2 log units below the Iron–Wüstite buffer). We find that the D_{Ne} coefficients range between 10^{-2} and 10^{-1} . These partition coefficients are only one of the controlling factors of noble gas distributions within the early Earth: because, even if D_{He} and D_{Ne} are low ($\sim 10^{-4}$), there may have been sufficient noble gases present in the mantle to supply a significant quantity of He and Ne to the core. Assuming gas–melt equilibrium of the molten proto-Earth with a nebular gas composition and concomitant metal–silicate differentiation, the core would have inherited and maintained throughout Earth's history high $^3\text{He}/^4\text{He}$ ratios and low $^3\text{He}/^{22}\text{Ne}$ ratios (< 0.6), making the core a potential source of primordial light noble gases in mantle plumes.

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Letter

Earth's mantle contains a mixture of primordial noble gases, in particular solar-type helium and neon, and radiogenic noble gases (e.g., Marty, 2012; Moreira, 2013). The isotopic ratios of He and Ne in ocean island basalts (OIB) indicate the existence of a reservoir with high concentrations of primordial noble gases in the deep Earth. This reservoir appears to have been isolated from the MORB (Mid-Ocean Ridge Basalts) source reservoir 4.4 billion years ago (e.g., Pepin and Porcelli, 2006; Mukhopadhyay, 2012; and references therein).

The origin of high $^3\text{He}/^4\text{He}$ and low $^3\text{He}/^{22}\text{Ne}$ ratios in many OIBs is still much debated and there has long been a supposition that OIBs sample a primordial and undegassed reservoir located in the deep mantle. Increasing evidence for mantle convection processes has pushed the debated boundary for a primitive mantle source location ever deeper into the lower mantle, reaching the core–mantle boundary (e.g., Williams *et al.*, 2019). The Earth's core has also been suggested as an alternative source of noble gases (Porcelli and Halliday, 2001; Tieloff and Kunz, 2005; Armytage *et al.*, 2013; Bouhifd *et al.*, 2013; Roth *et al.*, 2019).

Extending our previous work on helium, here we investigate neon partitioning between molten silicates and Fe-rich alloy liquids up to 16 GPa and to a maximum temperature of ~3000 K. We also address the behaviour of He/Ne in an early magma ocean.

A CI-chondrite model composition was chosen for the starting silicate material (e.g., Bouhifd *et al.*, 2013). To simulate Earth's core formation under conditions of segregation from a deep magma ocean, we performed experiments with a starting mix of CI-chondrite glass (66 wt. %) with various Fe-rich alloys (34 wt. %).

We used a set-up described in previous studies (e.g., Jephcoat *et al.*, 2008) with all details reported in the Supplementary Information. Briefly, we used diamond anvils with 500 μm culets, and stainless steel gaskets pre-indented to a thickness of ~50 μm and drilled to a diameter of ~100 μm . Samples were mounted in the pressure chamber and neon was loaded with a high pressure gas-loading technique at 200 MPa. Pressures were measured using the ruby fluorescence method before and after each heating stage. The samples were heated by a 100 W air-cooled fibre laser with emission centred at 1070 nm for an average of about 15 minutes for each zone of the sample. During the heating stage the surrounding neon melted by conductive heating from the heated sample, allowing its dissolution into the sample. Temperatures were determined spectro-radiometrically with a fit to a grey-body Planck function. The temperatures of our runs were higher by at least 200 degrees than the liquidus temperature of peridotite (Zhang and Herzberg, 1994; Fig. S-2). Because our experiments are not routine the success of our LHDAC technique was demonstrated in our previous studies where we found a good agreement between

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our results and those provided from large volume presses for argon (Fig. S-3a,b).

The chemical compositions of the run products were analysed by an electron microprobe (JEOL JXA-8800R with 4 wavelength dispersive spectrometers) with an accelerating voltage of 15 kV and a beam current of 20 nA (see Table S-1). The chemical analyses for neon with the UVLAMP were carried out at the Open University, UK. A NewWave UP213 combined laser/microscope system uses a 10 Hz quintupled Nd-YAG laser with a wavelength of 213 nm yielding ~ 10 ns pulses (Heber *et al.*, 2007). The extracted gas was cleaned using three SAES AP-10 getters at 400 °C and room temperature to remove active gases before analysis with a MAP 215–50 noble gas mass spectrometer. Blanks were measured before and after each ablation and all the measured blanks were small with respect to the minimum neon concentrations measured in our samples. For both silicate and metal analyses, the measured ^{22}Ne blanks were $(0.6 \pm 0.1) \times 10^{-12} \text{ cm}^3 \text{ STP}$. To convert measured Ne abundances into concentrations, the volumes of all laser pits were determined by using a non-contact, vertical scanning white light interferometer (Zygo instrument), which has a vertical resolution of $\sim 1 \text{ \AA}$ and a horizontal resolution of $1 \text{ }\mu\text{m}$.

The experimental conditions, the neon contents of the run products and the resulting partition coefficients for neon are presented in Table 1. Each of the reported results is the average of several spot analyses made from the same phase. During Ne analysis, we ablated the first 2 microns from the surface before starting the quantitative determinations of Ne concentrations. This step is made to avoid the high near-surface concentrations of noble gases (*e.g.*, Bouhifd *et al.*, 2013). Therefore, the neon contents presented here correspond to neon dissolved in the bulk molten phases quenched from HP.

Here we found that the neon solubility in silicate melt dropped by a factor of ~ 3 (Fig. 1) from $\sim 450 \text{ ppm}$ Ne at 6 GPa to $\sim 140 \text{ ppm}$ Ne at 10 GPa. This drop of neon solubility can be attributed to the characteristic of silicate melts at high pressures as reported in our study for argon (Bouhifd and Jephcoat, 2006). We also found no evidence for a pressure dependence of Ne solubility in Fe-rich alloys. It is important to note here that the drop of Ne solubility in silicate melt by a factor of 3 will only vary the D_{Ne} coefficient from 3×10^{-2} at 6 GPa (before the drop of Ne solubility in the silicate melt) to 9.8×10^{-2} at 10 GPa (after the drop of Ne solubility). The

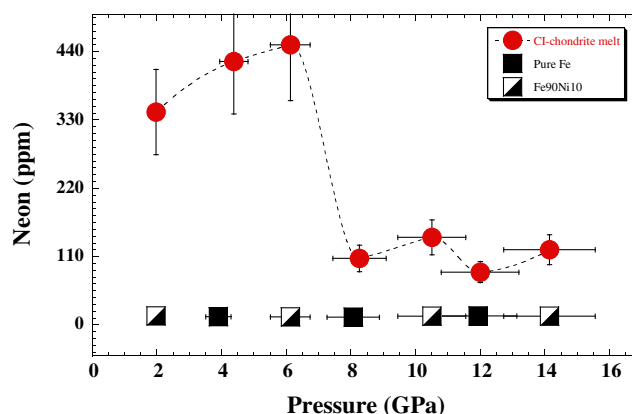


Figure 1 Neon concentrations in CI-chondrite melt, Fe and $\text{Fe}_{90}\text{Ni}_{10}$ alloy at high pressures. The reported Ne contents represent the average of 5 to 20 analyses. Error bars correspond to 1σ (ranging from 5 to 20 %) of the average of the chemical analyses and to 10 % of the nominal pressures reported. For the metallic phase no pressure dependence is observed and the average value of Ne concentrations is $13.5 \pm 0.8 \text{ ppm}$.

variation of D_{Ne} of less than one order of magnitude was taken into account in our calculations and was found not to affect the main conclusion of the present work. Previous studies show that the core can be a plausible source for light noble gases using much lower ($\sim 10^{-4}$) partition coefficients of He and Ne, meaning that the metal-silicate partition coefficients of noble gases are not the only limiting factor. The main controlling factor being the abundances of noble gases in the proto-Earth (Porcelli and Halliday, 2001; Tieloff and Kunz, 2005).

Measurements of Ne partitioning between metal and silicate have been reported by Matsuda *et al.* (1993) and Sudo *et al.* (1994). These studies indicate high D_{Ne} of $\sim 10^{-1}$ at $P \sim 0.5 \text{ GPa}$ that decreased with increasing pressure to near 10^{-3} at 6 GPa. At about 2 GPa, our D_{Ne} value is similar to that determined by Matsuda *et al.* (1993). However, at pressures $\geq 2 \text{ GPa}$, we do not observe the systematic decrease in the D_{Ne} coefficients, instead our results indicate higher D_{Ne} coefficients of $\sim 10^{-1}$ at pressures $> 8 \text{ GPa}$, which is little dependent on pressure (Fig. 2). This behaviour is similar to that of helium for which we found that D_{He} coefficients are constant over a wide range

Table 1 Neon concentrations (ppm) in molten CI-chondrite and Fe-rich alloys liquids. Values in bold (either for metallic or silicate phases) are linear interpolations between the two nearest experimental pressure data.

P (GPa)	T (K)	Ne contents (ppm)	Ne contents (ppm)	D_{Ne}
		CI-Chondrite melt	Fe-rich alloy liquids	
			$\text{Fe}_{90}\text{Ni}_{10}$ alloy	
2.0	2400	342 (± 25)	14.3 (± 1.3)	$4.2 (\pm 0.7) \times 10^{-2}$
4.4	2400	424 (± 36)	13.4 (± 1.0)	$3.2 (\pm 0.5) \times 10^{-2}$
6.1	2500	451 (± 32)	12.8 (± 1.0)	$2.8 (\pm 0.4) \times 10^{-2}$
8.3	2450	107 (± 9)	12.1 (± 1.0)	$1.1 (\pm 0.2) \times 10^{-1}$
10.5	2500	141 (± 15)	13.8 (± 1.5)	$9.8 (\pm 2.1) \times 10^{-2}$
12.0	2500	85 (± 8)	14.0 (± 1.5)	$1.6 (\pm 0.3) \times 10^{-1}$
14.1	2700	121 (± 14)	14.0 (± 2.0)	$1.2 (\pm 0.3) \times 10^{-1}$
			Pure Iron	
3.9	2200	420 (± 40)	13.4 (± 1.0)	$3.2 (\pm 0.5) \times 10^{-2}$
8.1	2200	110 (± 10)	12.1 (± 1.0)	$1.1 (\pm 0.2) \times 10^{-1}$
11.9	2300	85 (± 10)	14.4 (± 1.4)	$1.7 (\pm 0.3) \times 10^{-1}$

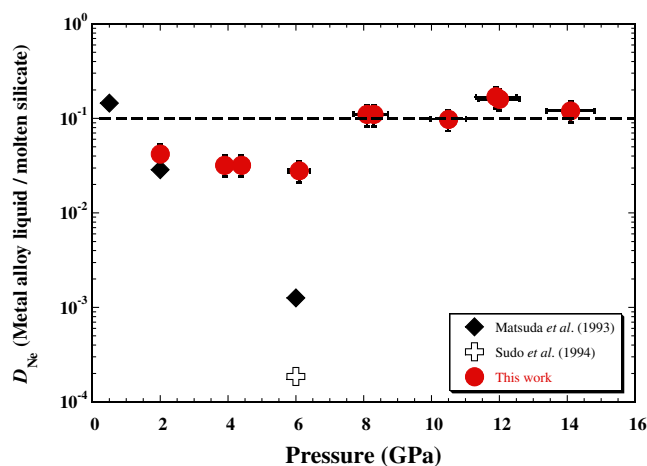


Figure 2 Partition coefficients of Neon, D_{Ne} (wt. % of Ne in metallic phase / wt. % of Ne in silicate) between molten silicate and iron-rich metal liquids as a function of pressure. The average value of our experiments at high pressure is represented by the dashed line. The previous data from Matsuda *et al.* (1993) and Sudo *et al.* (1994) are also plotted. At this stage the discrepancy between data sets at 6 GPa is unknown.

of pressure up to 40 GPa (Bouhifd *et al.*, 2013). We assume thus that the D_{Ne} is within 10^{-2} – 10^{-1} at the conditions of Earth's core formation that can reach conditions in the range of 30–70 GPa and 3500 K as determined from moderately siderophile elements.

Assuming a global magma ocean during core formation, our experimental solubilities of Ne and He into a chondritic melt show that under a hot, dense atmosphere from which volatiles are dissolved, such a magma ocean would have been enriched in He with respect to Ne. The reported [He/Ne] ratio (wt. % of He/wt. % of Ne in chondritic melt) shows a negligible dependence on pressure up to 17 GPa with an average of 6 ± 2 (Fig. 3). Because the solubility factor ratio of He by Ne is at most 2 (*e.g.*, Jambon *et al.*, 1986), an early magma ocean would have presented a ratio $^3\text{He}/^{22}\text{Ne}$ within 2.5–5, reflecting up to two global melting events during the first 100 million years of Earth's history (Tucker and Mukhopadhyay, 2014). For the estimate above, we consider that the initial $^3\text{He}/^{22}\text{Ne}$ ratio is that of the solar nebula or of implanted solar wind during the early

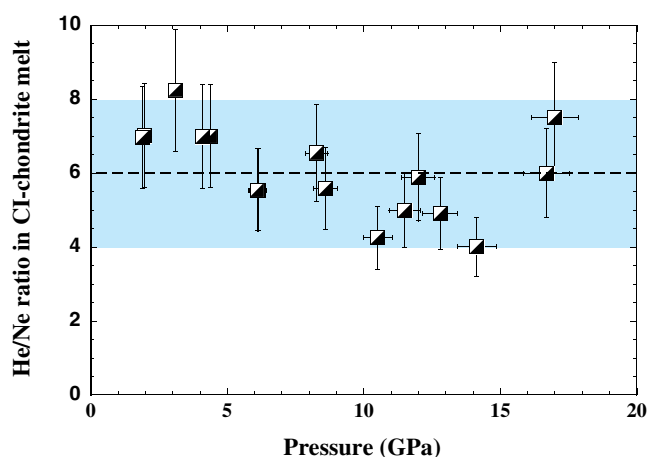


Figure 3 He/Ne ratio (wt. % of He in the melt / wt. % of Ne in the melt) in CI-chondrite melt at high pressures.

solar system, around 1.50 ± 0.06 , and 0.9 ± 0.1 , respectively (*e.g.*, Yokochi and Marty, 2004; Heber *et al.*, 2009; Raquin and Moreira, 2009; Tucker and Mukhopadhyay, 2014).

In order to evaluate the potential of He and Ne sequestration by the core, it is necessary to estimate the initial abundances of these noble gases on proto-Earth. In the early part of Earth's history, high concentrations of He and Ne that might have existed as Xe isotopes indicate extensive losses of volatile elements from the mantle within the first 100 Myr. Such losses appear to have depleted the noble gases by $\geq 97\%$, implying that their initial concentrations in Earth were two orders of magnitude higher than at present (Porcelli *et al.*, 2001; Yokochi and Marty, 2005). The amounts of primitive helium and neon can thus be estimated from the present day ^3He and ^{22}Ne concentrations in the mantle that range between 2.2 and $>14 \times 10^{-10}$ cc STP/g for ^3He and between 4 and $>58 \times 10^{-11}$ cc STP/g for ^{22}Ne (Yokochi and Marty, 2004; Moreira and Kurz, 2013). We can thus estimate that ^3He primitive mantle's concentrations are in the range of $\sim 10^{12}$ ^3He atoms/g. In addition, the initial Earth $^3\text{He}/^4\text{He}$ ratio was around $120 \times R_A$ (where $R_A = 1.39 \times 10^{-6}$ is the atmospheric ratio) if solar nebula was trapped in the mantle; or $330 \times R_A$ that represents the solar wind ratio, provided that terrestrial He was derived from solar wind implantation of accreting materials (Porcelli *et al.*, 2001).

By considering the values reported above, abundances of about 2.7×10^{12} ^3He atoms/g and 2.9×10^{12} ^{22}Ne atoms/g in the early Earth's mantle are very plausible. By taking these values as the initial ^3He and ^{22}Ne abundances at time of the core formation, and the measured D_{He} and D_{Ne} , ^3He and ^{22}Ne in the early core could have been between 1.3×10^{10} and 4.6×10^{10} ^3He atoms/g, and between 8.1×10^{10} and 4.9×10^{11} ^{22}Ne atoms/g, respectively. These estimates indicate that the Earth's early core might have had a $^3\text{He}/^{22}\text{Ne}$ ratio lower than 0.6. During core formation, the $^3\text{He}/^{22}\text{Ne}$ ratio of the mantle should be within the range 2.5–5, and the metal-silicate partition coefficient of the He/Ne ratio is about 5×10^{-2} (Fig. S-9) leading to a $^3\text{He}/^{22}\text{Ne}$ ratio within the core in the range of 0.12–0.25. We can thus conclude that the Earth's core must have inherited low $^3\text{He}/^{22}\text{Ne}$ ratio (< 0.6), and any contribution from the core to the OIBs source regions has the potential to lower their $^3\text{He}/^{22}\text{Ne}$ ratios.

The measured high $^3\text{He}/^4\text{He}$ and low $^3\text{He}/^{22}\text{Ne}$ ratios observed now in some OIBs (such as Hawaii) could be the result from mixing of He and Ne from a small core component with radiogenic ^4He and residual ^3He and ^{22}Ne from the mantle. Our new estimates, based on the ratio of the fluxes of noble gases in OIBs to their concentrations in the core, would require that less than 1 % of the mass of the core should have released its primordial light noble gases.

Since significant amounts of siderophile elements are incorporated in the core, additions of core material to the OIBs would impact these elements. For modern OIBs, tungsten isotopic data negatively correlate with $^3\text{He}/^4\text{He}$ ratios (Mundl *et al.*, 2017). This correlation can indicate core-mantle interaction with transfer of the high $^3\text{He}/^4\text{He}$ and low ^{182}W signature of the outer core to the source of the OIBs. A similar conclusion was reached in a more recent study by Rizo *et al.* (2019) where they found that core-mantle exchange might be facilitated by exsolution of W-rich, Si-Mg-Fe oxides from the core into the mantle. A correlation between the highest $^3\text{He}/^4\text{He}$ values and the highest $^{186}\text{Os}/^{188}\text{Os}$ ratios in Hawaii was also used as an evidence for a core contribution to He and Os (Walker *et al.*, 1995). However, subsequent studies in other OIBs show that this evidence in OIBs cannot be uniquely tied to the core (Luguet *et al.*, 2008). Clearly, the debate over core-mantle exchanges has some way to go, but the present study shows that the early Earth's core

could have incorporated helium and neon making it a potential supplier of light noble gases into deep-rooted mantle plumes. Also the Earth's core must have inherited high $^3\text{He}/^4\text{He}$ ratios (in the range 120–330) and low $^3\text{He}/^{22}\text{Ne}$ ratios (<0.6), and the core maintained high and low, $^3\text{He}/^4\text{He}$ and $^3\text{He}/^{22}\text{Ne}$ ratios, respectively, throughout Earth's history.

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Additional Information

Supplementary Information accompanies this letter at <http://www.geochemicalperspectivesletters.org/article2028>.



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