Experimental determination of carbon diffusion in liquid iron at high pressure

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ABSTRACT

The transport properties of liquid iron alloys at high pressure (P) and temperature (T) are essential for understanding the formation, composition, and evolution of planetary cores. Light alloying elements (e.g., Si, O, S, C, N, H) in liquid iron are particularly relevant due to the density deficit of Earth’s core, yet high P-T experimental diffusion studies involving such alloys remain scarce, with large uncertainties on the P and T dependence required for extrapolation to core conditions. In this study, we measured the chemical diffusion of carbon in liquid iron over a P-T range of 3–15 GPa and 1700–2450 K using a multi-anvil apparatus. Diffusion couples consisting of pure Fe and Fe-2.5 wt% C cylinders were placed end-to-end in an MgO capsule in a vertical orientation. Carbon concentration profiles were measured by electron microprobe and modeled numerically to correct for non-isothermal diffusion that occurred prior to reaching the peak temperature. Carbon diffusion coefficients range from $6 \times 10^{-9}$ m$^2$ s$^{-1}$ to $2 \times 10^{-7}$ m$^2$ s$^{-1}$, with global Arrhenian fit parameters $D_0 = 1.4 \pm 0.5 \times 10^{-7}$ m$^2$ s$^{-1}$, $\Delta E = 43 \pm 6$ kJ/mol, and $\Delta V = -0.06 \pm 0.19$ cm$^3$ mol$^{-1}$. A negligible P effect is consistent with previous studies of oxygen diffusion in liquid iron and high-T calculations but differs from larger $\Delta V$ values previously reported from carbon self-diffusion experiments for liquid Fe$_x$C and simulations for an Fe-Ni-C alloy. Carbon diffusion coefficients determined here are approximately three times faster than those reported from Fe-Ni-C liquid simulations, which highlights the potential significance of compositional effects on mass transport properties of liquid iron alloys and the need for expanding the P-T experimental diffusion data set currently available in the literature to more complex and geologically relevant compositions.

Keywords: Carbon diffusion, core formation, outer core, liquid iron alloys, interstitial alloys

INTRODUCTION

The distribution of carbon in reservoirs of the Earth’s interior plays a critical role in a range of geological processes, including climate, habitability, and potentially the geodynamo (e.g., Hazen and Schiffries 2013). Carbon is a siderophile element (e.g., Dasgupta et al. 2013; Li et al. 2015, 2016; Fischer et al. 2020), which implies that the majority (80–90%) of the Earth’s carbon budget is expected to have partitioned into the core during planetary differentiation because of metal-silicate segregation. The extent of chemical equilibration between metal and silicate during core formation is largely controlled by diffusion kinetics of alloying elements in iron-rich liquid metal under relevant magma ocean pressure (P) and temperature (T) conditions (e.g., Rubie et al. 2003).

Most diffusion studies in liquid iron and its alloys have reported weak P-T dependences with small Arrhenian activation parameters (e.g., Poirier 1988; Ichikawa and Tsuchiya 2015; Umemoto and Hirose 2015; Posner et al. 2017a, 2017b) according to:

$$D_i(P,T) = D_{0i} \exp \left( \frac{-\Delta E + P \cdot \Delta V}{R \cdot T} \right)$$  \hspace{1cm} (1)

where $D_i$ is the diffusion coefficient of element $i$, $D_{0i}$, $\Delta E$, and $\Delta V$ are the pre-exponential diffusion coefficient, activation energy, and activation volume, respectively, and R is the gas constant. For isobaric fits, the numerator in the exponent is replaced by an enthalpy $\Delta H^*$ (e.g., Ni et al. 2015), and the pre-exponential factor is labeled as $D_{0i}^*$ in these cases. Carbon is incorporated interstitially into both solid (e.g., Goldschmidt 1967) and liquid iron (e.g., Sobolev and Mirzoev 2013; Ohmura et al. 2020) and is therefore expected to show relatively similar diffusion behavior to other interstitial elements (e.g., O and N) as predicted by density-functional molecular dynamics (DFT-MD) simulations (e.g., Posner and Steinle-Neumann 2019).

High P-T experiments involving oxygen revealed a negligible P effect on diffusivity in liquid iron between 3 and 18 GPa (Posner et al. 2017b). However, a high P-T experimental study of carbon self-diffusion in liquid Fe$_x$C (Dobson and Wiedenbeck 2002) and recent isothermal (T = 1675 K) computations for liquid Fe$_{0.73}$Ni$_{0.07}$C$_{0.20}$ (Wang et al. 2019) reported $\Delta V$ values of carbon self-diffusion that are substantially larger ($\Delta V \sim 1.2$ cm$^3$ mol$^{-1}$) than values reported for oxygen ($\Delta V = 0.1 \pm 0.1$ cm$^3$ mol$^{-1}$; Posner et al. 2017b) and hydrogen ($\Delta V = -0.2$ cm$^3$ mol$^{-1}$; Umemoto and Hirose 2015), and also for substitutionally incorporated silicon ($\Delta V = 0.41 \pm 0.06$ cm$^3$ mol$^{-1}$; Posner et al. 2017a) and chromium ($\Delta V = 0.34 \pm 0.11$ cm$^3$ mol$^{-1}$; Posner et al. 2017a).

Although Dobson and Wiedenbeck (2002) and Wang et al. (2019) reported similarly large $\Delta V$ values, their absolute $D_i$ values differ by nearly a factor of three. Combined with large errors on