

The effects of pressure on immiscibility in metallic, core-forming liquids: Implications for protoplanetary differentiation

Geoffrey David BROMILEY^{1,2*}, Hidenori TERASAKI³, and Matthew VARNAM^{1,4}

¹School of GeoSciences, University of Edinburgh, Grant Institute, Edinburgh, UK

²Centre for Science at Extreme Conditions, University of Edinburgh, Edinburgh, UK

³Department of Earth Sciences, Okayama University, Okayama, Japan

⁴School of Earth Sciences, University of Bristol, Bristol, UK

*Correspondence

Geoffrey David Bromiley, School of GeoSciences, University of Edinburgh, Grant Institute, the King's Buildings, James Hutton Road, Edinburgh, EH9 3FE, UK.
Email: geoffrey.bromiley@ed.ac.uk

(Received 17 September 2025; revision accepted 11 March 2026)

Abstract—Mechanisms for metal core formation in rocky planetesimals and planetary embryos remain poorly constrained, in part due to complexities arising from immiscibility in core-forming liquids at low pressures. To assess the pressure dependence of immiscibility and its role in protoplanetary differentiation, we performed experiments at 3 and 5 GPa in the system Fe_{0.9}Ni_{0.1} + S, P, C. Immiscibility arises due to the highly non-ideal nature of light element mixing in Fe liquids, and results in separation of Fe-rich (S-depleted, C-rich, P-rich) and FeS-rich (S-rich, C-rich, P-depleted) liquids. For a broad range of core-forming liquid compositions, a miscibility gap is only present at pressures <5 GPa. With increasing pressure, the behavior of complex systems converges on that of the Fe-S-C ternary, although S-P-C interactions continue to influence metal liquid compositions and stability. Comparison with planetary core compositions derived from meteorite data suggests that immiscibility can play a significant role during melting in planetesimals, and up to medium-sized planetary embryos. In turn, the importance of immiscibility during differentiation depends on the extent of melting, mechanisms for core formation, and corollary degree of light element loss from planetary bodies.

INTRODUCTION

Metal-silicate differentiation, that is, the large-scale separation of liquid metal from solid, partially molten, or molten silicate, had a fundamental control on planetary evolution in the early solar system. Differentiation was a complex, multistage process, driven by heat from the decay of short-lived isotopes such as ²⁶Al, accretional energy, and by large impacts, and resulted in varying degrees of internal segregation of metal and silicate (e.g., Dodds et al., 2021; Hevey & Sanders, 2006; Kaminski et al., 2020; Keil, 2000; Lichtenberg et al., 2019, 2021; Maurel et al., 2020; Moskovitz & Gaidos, 2011; Neumann et al., 2014, 2018; Scheinberg et al., 2015; Sramek et al., 2012). High

temperatures required for segregation of core-forming, metallic liquids would have resulted in varying degrees of degassing, modifying the volatile element composition of differentiating bodies (Barrett et al., 2019; Grewal et al., 2022; Hirschmann et al., 2021).

Differentiated meteorites, and the existence of differentiated and metal-rich asteroids such as 4-Vesta and 16-Psyché, evidence metal core formation in planetary bodies a few hundred to a few thousand km in size, that is, in rocky planetesimals and planetary embryos (e.g., Elkins-Tanton et al., 2022; Greenwood et al., 2005; Russell et al., 2012; Wilson & Keil, 2012). Radiometric dating of magmatic iron meteorites suggests that core segregation within parent bodies occurred within a few Myr of the

onset of accretion in the solar system (Kleine et al., 2002, 2005, 2009; Kruijer et al., 2014). This is consistent with constraints from thermal models (e.g., Scheinberg et al., 2015), which suggest that only rocky planetesimals and planetary embryos which grew rapidly soon after the onset of accretion retained sufficient heat to undergo extensive melting. Although this has clear implications for understanding planetary formation, including constraining the composition of rocky planets, asteroids and meteorite parent bodies, the processes by which this period of early core formation occurred remain poorly constrained.

One complexity in exploring the nature of differentiation processes in small rocky planetary bodies is immiscibility in liquid metals (Bromiley, 2023). The metallic cores of planetary bodies are Fe-Ni alloys with variable proportions of lighter elements, including S, C, O, P, Si, and H (Hilton et al., 2022; Hirose et al., 2021; Komabayashi, 2021; McDonough, 2014; Steenstra & van Westrenen, 2018). Light element content has a significant influence on melting of the metallic component of planetary bodies, on the physical properties of liquid alloys, and on subsequent processes of core crystallization (e.g., Hirose et al., 2021; Komabayashi, 2014; Rückriemen et al., 2018; Terasaki et al., 2005). Under the relatively modest pressures of small rocky body interiors, mixing of light elements in Fe-rich liquids can be highly non-ideal (Morard & Katsura, 2010; Zhao et al., 2023). Electronic interactions between certain elements (e.g., S and C) are unfavorable, raising the energy of “mixed-atom” local configurations relative to “like-atom” configurations; correspondingly, interaction parameters between these elements are large and positive. For light element-rich compositions, this increases the enthalpy of mixing and the free energy of Fe liquids, and may result in liquid unmixing and immiscibility. At sufficiently high temperatures, the entropy of mixing offsets this enthalpy effect, stabilizing a single liquid. As such, immiscibility is characteristic of light element-rich compositions below a critical temperature, above which the immiscibility gap necessarily closes. With increasing pressure, light element mixing in Fe liquids becomes increasingly ideal, that is, interaction parameters approach zero, resulting in full miscibility in Fe liquids at sufficiently high pressure (Komabayashi, 2021).

Liquid immiscibility is observed toward lower pressures in some binary systems such as Fe-FeO (Komabayashi, 2014) and in many ternary systems, including in the systems Fe-S-C (Dasgupta et al., 2009; Deng et al., 2013), Fe-S-P (Chabot & Drake, 2000; Ulff-Møller, 1998), Fe-O-S (Tsuno et al., 2007; Urakawa et al., 1987), and Fe-S-Si (Morard et al., 2008; Morard & Katsura, 2010; Sanloup & Fei, 2004), as well as in compositionally complex metallic systems (Bromiley et al., 2024) and silicate/metal systems (e.g., Corgne et al., 2008; McCoy et al., 1999). For the range of conditions in small planetesimals and planetary

embryos, that is, pressures $\ll 10$ GPa, oxygen fugacities from 0 to 3 log units below the Fe-FeO buffer (Grewal et al., 2024; Zhang et al., 2024), and for core compositions inferred from magmatic iron meteorites (Goldstein et al., 2009; Hilton et al., 2022; Zhang et al., 2022, 2024), S, P, C, and O are probably the most important light elements controlling Fe-Ni liquid stability. However, constraints on the extent of immiscibility in compositionally complex systems and their control on liquid compositions are limited, with most previous experimental studies focused on ternary systems, under unrealistic C-rich or C-saturated conditions, and at higher pressures not applicable to differentiation in smaller rocky bodies/planetesimals (Bromiley et al., 2024). Furthermore, although previous studies have demonstrated full Fe liquid miscibility at sufficiently high pressure (Corgne et al., 2008; Dasgupta et al., 2009; Deng et al., 2013; Sanloup & Fei, 2004), differences in starting compositions used between experimental studies mean that the effective upper pressure limit of immiscibility for realistic planetary core-forming liquids remains unclear. For example, different experimental studies constrain closure of the immiscibility gap between S-rich/C-poor and S-poor/C-rich liquids at <5 GPa (Deng et al., 2013), between 5 and 6 GPa (Dasgupta et al., 2009), or at up to 8 GPa (Corgne et al., 2008).

Here, we present the results of superliquidus experiments conducted at 3.0 and 5.0 GPa which aim to constrain the extent of, and controls on, liquid immiscibility and liquid compositions in the system $\text{Fe}_{0.9}\text{Ni}_{0.1} + \text{S}, \text{P}, \text{C}$. We then synthesize results from a broad data set of experiments to assess the importance of immiscibility in compositionally complex core-forming liquids in rocky planetesimals and planetary embryos of varying size.

METHODS

Starting Materials

Starting mixes were prepared from high purity Fe (99.9%), Ni (99.9%), FeS (99.5%), Fe_3P (99.5%), and diamond powder, based on a bulk $\text{Fe}_{0.9}\text{Ni}_{0.1}$ ratio with variable, S, P, and C contents. This Fe:Ni ratio was chosen to allow comparison of results to lower pressure (0.5 and 1.0 GPa) experiments of Bromiley et al. (2024), and is representative of compositions of a significant proportion of inferred planetesimal core compositions based on iron meteorite data (Goldstein et al., 2009). Starting mixes did not contain O or N. However, the presence of small amounts of both elements in run products is expected due to sample/capsule/atmosphere interactions, as observed in previous studies (e.g., Bromiley et al., 2024; Corgne et al., 2008; Dasgupta

et al., 2009; Grewal & Asimow, 2023). Starting mixes were homogenized under ethanol for at least 1 h, and stored in a vacuum oven at 120°C prior to use. Compositions of starting mixes are listed in Table S1.

High-Pressure/Temperature Experiments

For all experiments, powders were loaded into hBN capsules with press fit lids. Capsules were machined from hot-pressed hBN ceramic rods (Goodfellow Advanced Materials) which were stored in a desiccator prior to use. Experiments at 3 GPa were performed using a 0.5" end-loaded piston-cylinder apparatus at the University of Edinburgh, and high-temperature talc-Pyrex-graphite assemblies with fired Al₂O₃ spacers (Potts et al., 2021). Experiments were performed using either one 5 × 4 o.d. mm, or two 2.5 × 4 o.d. mm capsules (Figure S1). Prior to experiments, loaded assemblies were stored in a drying oven. Short experimental durations of c. 1 h were used due to rapid attainment of chemical equilibrium in Fe-rich liquids under high temperatures used in superliquidus experiments (Bromiley et al., 2024; Corgne et al., 2008; Dasgupta et al., 2009), to minimize slow ingress of water into capsules during experiments (Dasgupta et al., 2009), and to minimize injection and percolation of metallic liquids into surrounding hBN capsules (Berg et al., 2017, 2018). Despite this, some ingress of liquids into capsules was noted, which affected sample recovery, as discussed below. Experiments were pressurized to 110% of run pressure at room temperature, and then heated at 100°C min⁻¹ to 800°C. Piston-cylinder experiments were then held at 800°C overnight to anneal hBN capsules and minimize volatile loss (S, P, and C) at higher temperatures (Bromiley et al., 2024; Grewal & Asimow, 2023). After annealing, experiments were heated at 200°C/min to the final run temperature, either 1673 or 1773 K, and pressure then lowered to the final target value. Temperature was continually monitored and controlled using an R-type thermocouple, placed close to the sample capsule but protected from Fe contamination by a 1 mm sintered Al₂O₃ disc. This disc was carefully examined after experiments were recovered to verify that thermocouple contamination had not occurred. Sample pressure was calibrated based on the albite = jadeite + quartz reaction (Holland, 1980) and quartz-coesite transition (Hemmingway et al., 1998). Pressure was monitored by measuring load applied to the piston, and controlled throughout experiments. Samples were rapidly, isobarically quenched by shutting off power to the heating circuit. Samples cooled to below 800°C, that is, below the solidus for all bulk compositions, in around 5–10s.

Higher pressure experiments were performed using a 500 tonne Kawai-type multi-anvil apparatus, with

Walker module, at Okayama University. Eight millimeter truncated edge length WC cubes were used with a 14/8 assembly, containing two 2.0 × 1.2 mm hBN capsules (Denka Co. Ltd.). Assemblies consisted of Cr-doped MgO octahedra with internal ZrO₂ insulators, internal graphite heaters and MgO spacers with Mo electrodes, with a central D-type thermocouple in Al₂O₃ sleeve to monitor and control temperature (Figure S2). Prior to experiments, the pressure calibration for this modified 14/8 assembly was verified using the quartz-coesite transition, based on Raman analysis of run products. All these experiments were performed at 5 GPa, and were pressurized at room temperature for c. 3 h until the final run pressure was reached. Experiments were then heated to 1873 K over approximately 1 h. A pre-annealing step was not used for multi-anvil experiments. Experiments were isobarically quenched by turning off power to the heating circuit, before being slowly depressurized and recovered. To minimize water ingress into sample volumes, loaded capsules and assemblies were kept in a desiccator prior to use, and ceramic components, except hBN, were fired and then stored in a vacuum drying oven prior to use.

Sample Preparation and Analysis

Due to the fragility of recovered hBN capsules, samples were fully removed from surrounding capsules before preparation where possible. Partial extrusion of run products into capsules in some experiments, especially those with abundant FeS-rich liquid, prevented the ready removal of capsule material, and in these instances, samples were prepared within recovered capsules. Samples were mounted using Crystalbond™, ground to reveal cross sections using 1200 grit SiC grinding paper, and then gently polished using 0.3 µm colloidal Al₂O₃ suspension on a polishing mat. Polished samples were examined optically and, to ensure accurate determination of C contents, removed from mounts by soaking in high-purity acetone and remounted by pressing into indium-filled 1 inch aluminum sample holders.

Samples were characterized at the University of Edinburgh using a Carl Zeiss SIGMA HD VP Field emission gun Scanning Electron Microscope (FEG-SEM) with dual Energy Dispersive Spectrometer (EDS) system, and using Oxford Instruments AZTec software for X-ray compositional analysis. Samples were mounted and analyzed with a C-blank (high-purity Fe, ECRM097) and carbon standards: carbon steel standards ECRM 090-1 (C) —1 wt% C— and ECRM 084-1(C) —0.4 wt% C, and Fe₃C, synthesized following the methodology of Dasgupta and Walker (2008). Immediately prior to analysis, surfaces of sample blocks were gently cleaned

using petroleum ether and then plasma cleaned under vacuum, using 2× 60s bursts. Cleaned sample surfaces were coated with 4 nm Al and immediately placed in the vacuum chamber of the FEG-SEM, to minimize oxidation of the coating. Samples were characterized using a 20 kV accelerating voltage at a fixed 7.0 mm working distance, and using a 70 µm aperture. Secondary electron and backscattered electron (BSE) images were used to characterize textures in run products. The presence of both primary—that is, immiscible liquids, crystalline phases, vesicles—and secondary—complex exsolution—textural heterogeneities with length scales up to 10s of microns (Figure 1) necessitated use of large area SEM-EDS analysis to determine representative phase compositions. SEM-EDS analysis is advantageous in enabling rapid sample characterization, minimizing issues with variable carbon backgrounds and sample contamination during analytical sessions, and enabling ready determination of any systematic changes in X-ray intensities during analysis (e.g., Bromiley et al., 2024). Uncertainties in in situ compositional data presented here, correspondingly, are largely due to textural heterogeneity within samples developed during quenching. For additional information on compositional analysis and calibration, see Section S1.

RESULTS

As detailed in Table 1, recovered samples consist of either a quenched miscible liquid (Figure 1A–D, G,H), or two quenched, immiscible liquids (Figure 1E–F). Complex exsolution textures are observed in all quenched liquids, consisting of intergrown S-poor and S-rich phases, Ni-poor and Ni-rich phases, and a C-rich phase (Figure 1). Exsolution textures are dependent on starting compositions (e.g., Figure 1A–C). SEM-EDS selected area analysis confirms that immiscible liquids are S-rich and S-poor, in accordance with previous experimental investigations (e.g., Chabot & Drake, 2000; Dasgupta et al., 2009; Deng et al., 2013; Zhang et al., 2018). Following Bromiley et al. (2024), we use the terms Fe-rich and FeS-rich liquid (Figure 1) to identify coexisting immiscible liquids. P and C are enriched in the immiscible Fe-rich (S-poor) liquid relative to the FeS-rich liquid. Miscible and immiscible liquids also contain minor O and N due to sample/capsule interaction (Section S2).

Small vesicles are noted in the majority of recovered samples (Figure 1), implying volatile loss during quenching. Bulk compositions of liquids in run products imply some S loss relative to target compositions (Table S1). However, there is no correlation between vesicle presence/volume proportion and liquid compositions. Instead, vesicles likely evidence minor H loss or O loss during quenching (see Section S3). In some

samples with immiscible liquids (e.g., Figure 1e), samples are partially injected into fractures in hBN capsules. Berg et al. (2018) noted that volume changes in high-pressure experiments can result in rapid injection of metallic liquids into hBN capsules, a process distinct from slower percolation of FeS-rich liquid along grain boundaries (Berg et al., 2017). Sample ingress into capsules does not appear to result from sample/capsule reaction (Section S2).

Presence of minor O in run products is consistent with previous studies (e.g., Bromiley et al., 2024; Corgne et al., 2008; Dasgupta et al., 2009). O contents of miscible liquids range from 1.3 to 1.8 wt%. Comparable O contents are noted in immiscible Fe-rich liquids, although ranging up to 2 wt%. In accordance with previous studies, we note preferential partitioning of O into immiscible FeS-rich liquid, which has O contents ranging up to 2.6 wt%. Run products also contain small amounts of N: 0.2–2.5 wt% in miscible liquids, 1–2 wt% in Fe-rich liquid, and 0.1–1.7 wt% in FeS-rich liquid. This is consistent with N contents from metal/silicate partitioning experiments, in which N is shown to be moderately siderophile in nature at oxygen fugacities near or below the Fe-FeO (IW) buffer (Grewal et al., 2019; Grewal & Asimow, 2023).

Some run products contain crystals of graphite, implying C saturation. Graphite in 3 GPa experiments is acicular (Figure 1d), while graphite in 5 GPa experiments is larger and more equigranular. Experiment HP-im-exp6a evidences additional crystallization of a P-rich phase. Run products for this experiment consist of immiscible Fe-rich and FeS-rich liquids, with a bulk composition significantly depleted in P and C relative to the target composition (Table S1). No C-rich or P-rich phases were noted in run products, although sample injection into the hBN capsule resulted in incomplete sample recovery. C content of this sample is consistent with the presence of graphite, that is, with C saturation of both the immiscible Fe-rich (at 5.6 wt% C) and FeS-rich (at 1.4 wt% C) liquids. A P-rich phase must also have been present, most likely Fe₃P based on comparison with Fe-S-P melting experiments by Zhai et al. (2022). Experiment HP-im-exp7b used the same bulk composition but was run at higher temperature (1773 K compared to 1673 K). Run products consisted of a single, miscible liquid and graphite. High P content of the miscible liquid is consistent with the absence of a P-rich phase in this higher temperature, C saturated experiment.

Figure 2 shows liquid compositions and position of the miscibility curve. Figure 2a shows compositions projected onto S versus P. Numbers correspond to C contents of miscible liquids and, for experiments with immiscible Fe-rich and FeS-rich liquids, calculated bulk liquid compositions. The gray line marks the position of

the miscibility curve for a C content of c. 2 wt% at 3 GPa, constrained from experimental data. In contrast to 3 GPa experiments, all experiments at 5 GPa produced a single, miscible liquid. Figure 2b shows the same data set on a plot of S + O versus C + P, a

projection based on element partitioning into immiscible liquids—O shows a slight preference for incorporation into immiscible FeS-rich liquid. The position of the projected 3 GPa miscibility curve is shown, based on bulk compositions for 1 liquid versus 2 liquid fields.

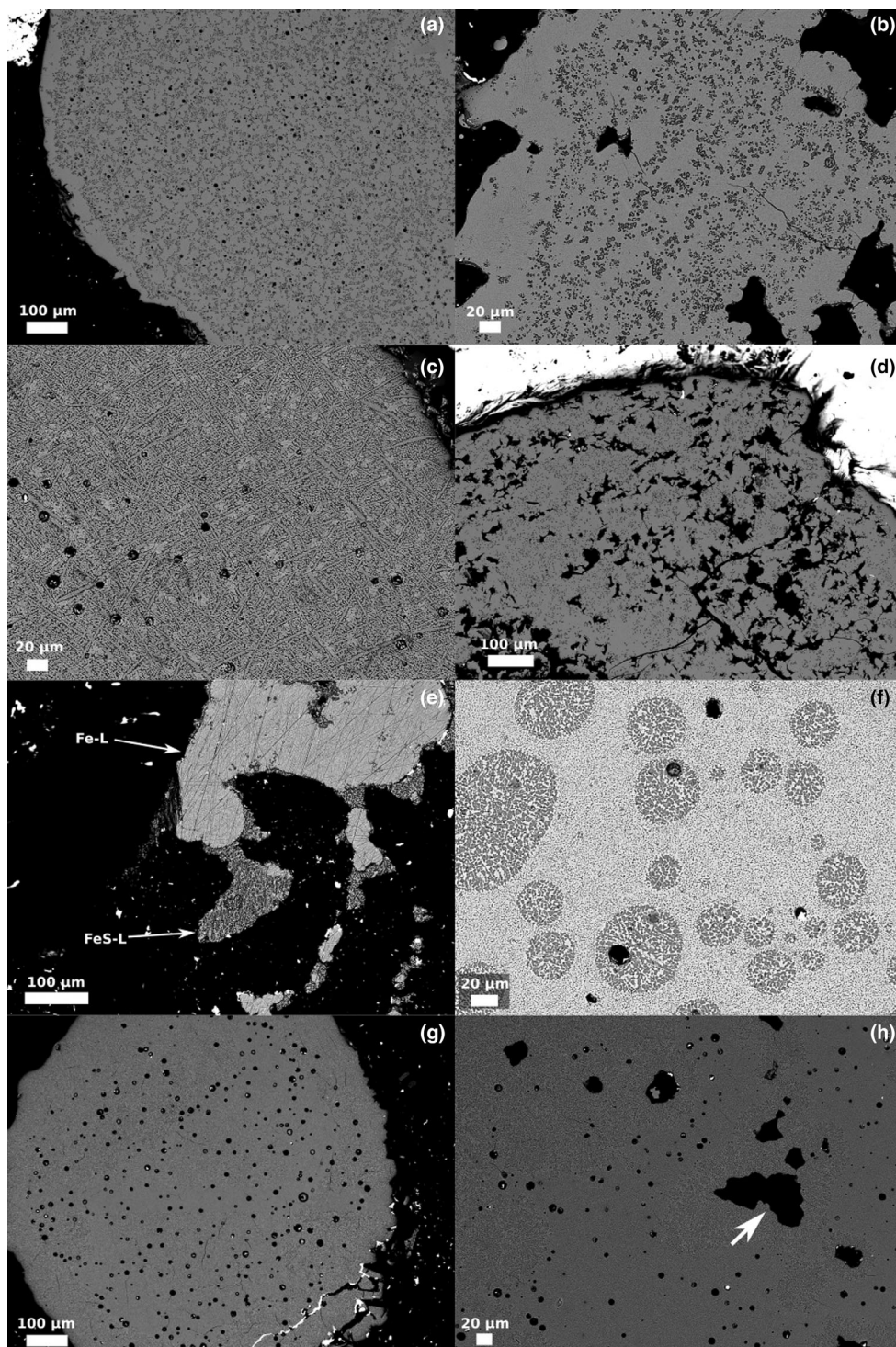


FIGURE 1. Backscattered electron images of run products. Unless indicated, black areas are surrounding polycrystalline hBN capsules, and white areas are indium mounts. (a) HP-im-3a, showing finer-scale exsolution textures in quenched miscible (single) liquids with lower S-contents (starting composition 5.8 wt% S, 3.9 wt% P, 3 wt% C). Sample also contains small vesicles (black). (b) HP-im-3b has a similar composition and quench texture, but higher bulk C content (5 wt% C) which results in stabilization of acicular graphite crystals (black). (c) HP-im-5a shows coarser exsolution textures typical of miscible liquids with higher bulk S contents (10 wt% S). Sample again contains vesicles. (d) HP-im-7b, showing textures typical of a lower-S quenched miscible liquid. High P content of sample results in reduced C solubility, and the formation of abundant acicular crystals of graphite. (e) HP-im-6a, showing immiscible Fe-rich (Fe-L) and FeS-rich (FeS-L) liquids. Sample only partially recovered due to injection of sample into fractures in the BN capsule (black). (f) HP-im-7a, showing immiscible FeS-rich liquid blobs within Fe-rich liquid, and representative exsolution textures for both liquids at higher magnification. (g) HP-OK-FeSPC6, showing typical variable fine/medium exsolution textures, and higher vesicularity, in miscible liquids from 5 GPa experiments. (h) HP-OK-FeSPC5, showing similar textures in miscible liquid at 5 GPa, with large graphite crystals (arrow).

DISCUSSION

Liquid Immiscibility and Element Partitioning in S, C and P-bearing systems

The extent of immiscibility in core-forming liquids is dependent on composition, temperature, and pressure. Experiments here constrain the extent of miscibility, and compositions of immiscible liquids, in a system with bulk composition $\text{Fe}_{0.9}\text{Ni}_{0.1}$ and variable S, P, and C, at 3.0 and 5.0 GPa, and 1673/1773 K—that is, at superliquidus temperatures. Bulk compositions are comparable to those in the 0.5/1.0 GPa experiments of Bromiley et al. (2024). S contents are variable and reflect considerable variation of S contents expected in core-forming liquids, as discussed in [Assessing the Importance of Immiscibility During Differentiation of Planetesimals and Planetary Embryos](#) Section. In contrast, bulk compositions have low to intermediate C (0–5 wt%) and P (1–5.7 wt%) contents. For run conditions, experiments vary from C undersaturated to saturated and are P undersaturated except for HP_im_exp6a. Over this compositional range, closure of the immiscibility gap is bracketed between 3.0 and 5.0 GPa (Figure 2), with all 5.0 GPa experiments producing a single, miscible liquid.

C solubility in Fe-Ni-S liquids has been shown to be markedly reduced with increasing S content and, additionally, modestly reduced with increasing Ni/Fe ratio (Zhang et al., 2018). Data from the present study also indicate a decrease in C solubility with increasing P content in S-poor liquids (Figure 3a). This is opposite to the trend which would occur if C solubility were only a function of S content of liquids. In contrast, C saturation in S-rich liquids is limited (Figure 3a) and possibly independent of P content.

Figure 3a–c highlights other compositional controls on light element behavior. Presence of minor N is unlikely to influence miscibility. However, N contents in immiscible Fe-rich and FeS-rich liquids do show

contrasting dependence on S content (Figure 3b; see also Section S2). Figure 3c shows Fe-rich liquid/FeS-rich liquid partition coefficients for P, C, and O, that is, $D_{\text{X}}^{\text{Fe-L/FeS-L}}$, as a function of the S partition coefficient. $D_{\text{P}}^{\text{Fe-L/FeS-L}}$ decreases markedly toward unity as $D_{\text{S}}^{\text{Fe-L/FeS-L}}$ increases toward unity, consistent with closure of the miscibility gap in Fe-S-P-rich systems. Trends are less clear for C and O partitioning. O is preferentially incorporated in FeS-rich liquid over Fe-rich liquid. $D_{\text{O}}^{\text{Fe-L/FeS-L}}$ increases from 0.7 ± 0.1 to 1.5 ± 0.4 as $D_{\text{S}}^{\text{Fe-L/FeS-L}}$ increases to unity. However, the observed trend is defined by one data point at higher $D_{\text{S}}^{\text{Fe-L/FeS-L}}$, with most values of $D_{\text{O}}^{\text{Fe-L/FeS-L}}$ at c. 0.8. $D_{\text{C}}^{\text{Fe-L/FeS-L}}$ remains approximately constant, in contrast to the Fe-S-C system, that is, to an expected decrease in the C partition coefficient with reduction in the extent of S partitioning. This again indicates complexity in light element behavior, that is, an effect of liquid P content on C solubility. Similar to Bromiley et al. (2024), we note that Ni is not preferentially partitioned between Fe-rich and FeS-rich liquids, despite previously noted controls of bulk Fe:Ni ratio on C solubility and the extent of miscibility (Zhang et al., 2018).

Pressure Dependence of Miscibility in Fe-Rich Liquids

Closure of the miscibility gap in the Fe-S-C ternary system has been bracketed between 5 and 8 GPa (Corgne et al., 2008; Dasgupta et al., 2009; Deng et al., 2013). The miscibility gap in the Fe-S-P ternary system closes at lower pressure, constrained by c. 1 bar experiments of Chabot and Drake (2000), which demonstrate immiscibility in P- and S-rich compositions, and the 3 GPa experiments of Zhai et al. (2022) in which full liquid miscibility is observed over a wide compositional range. Results from the present study are broadly consistent with these studies, as shown in Figures 4 and 5.

Figure 4 compares results from various studies projected onto S versus P. With modest increases in pressure, the miscibility gap in complex systems is

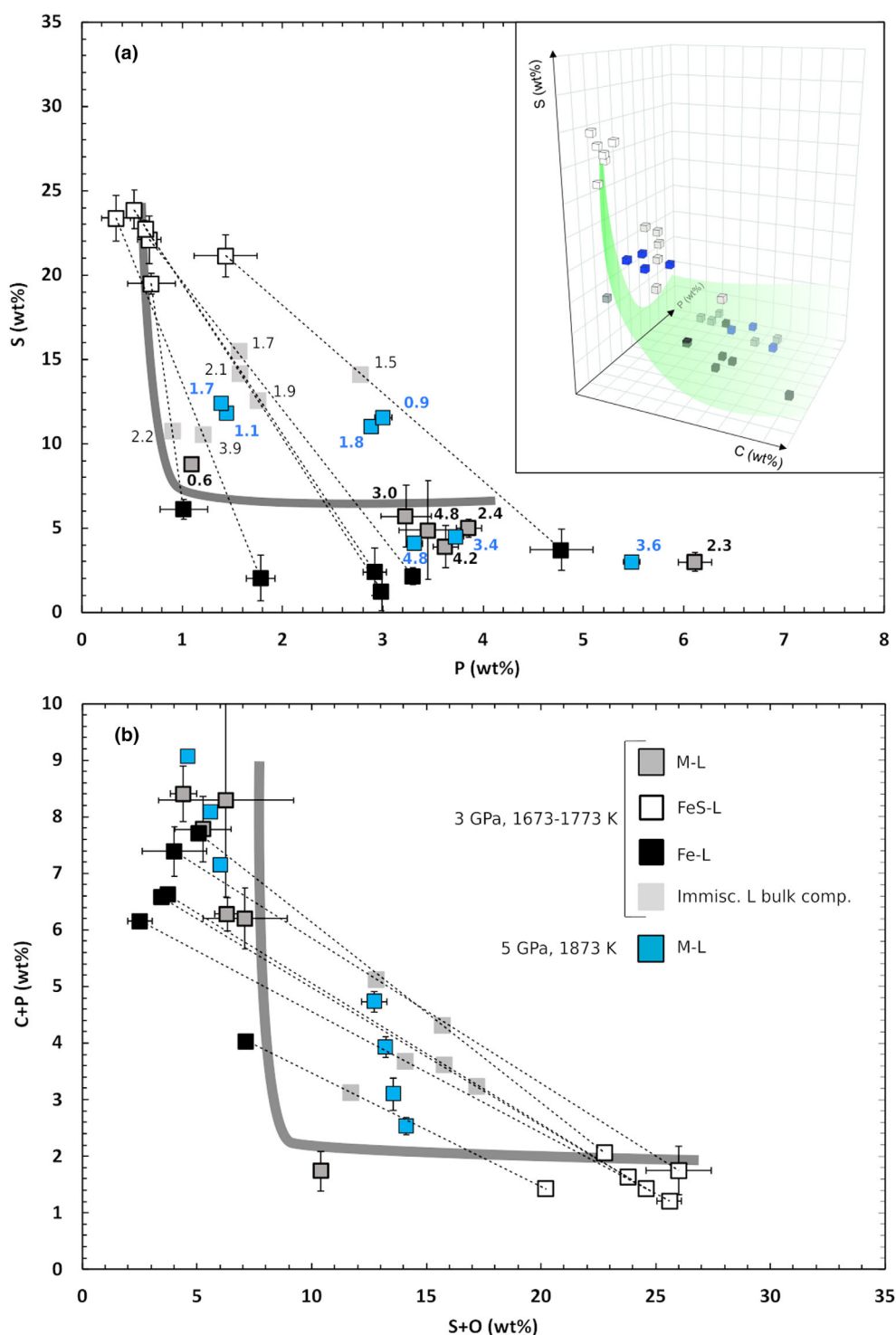


FIGURE 2. Compositions of quenched liquids from experiments in the system $\text{Fe}_{0.9}\text{Ni}_{0.1} + \text{S-P-C}$ at 3 (gray/black) and 5 GPa (blue). (a) S versus P plot. Tie lines join compositions of coexisting Fe-rich liquid and FeS-rich liquid, with light gray squares indicating bulk composition of immiscible liquids calculated based on estimated phase proportions from BSE SEM images. Numbers in text are carbon contents of miscible liquids (bold) and immiscible liquid bulk compositions (normal). Thick gray line is the miscibility curve for c. 2 wt% C at 3 GPa. Insert shows the same data set on a S-C-P scatter plot, with the miscibility surface shown in green. (b) Same data projected onto a plot of S + O versus C + P. (Color figure can be viewed at wileyonlinelibrary.com)

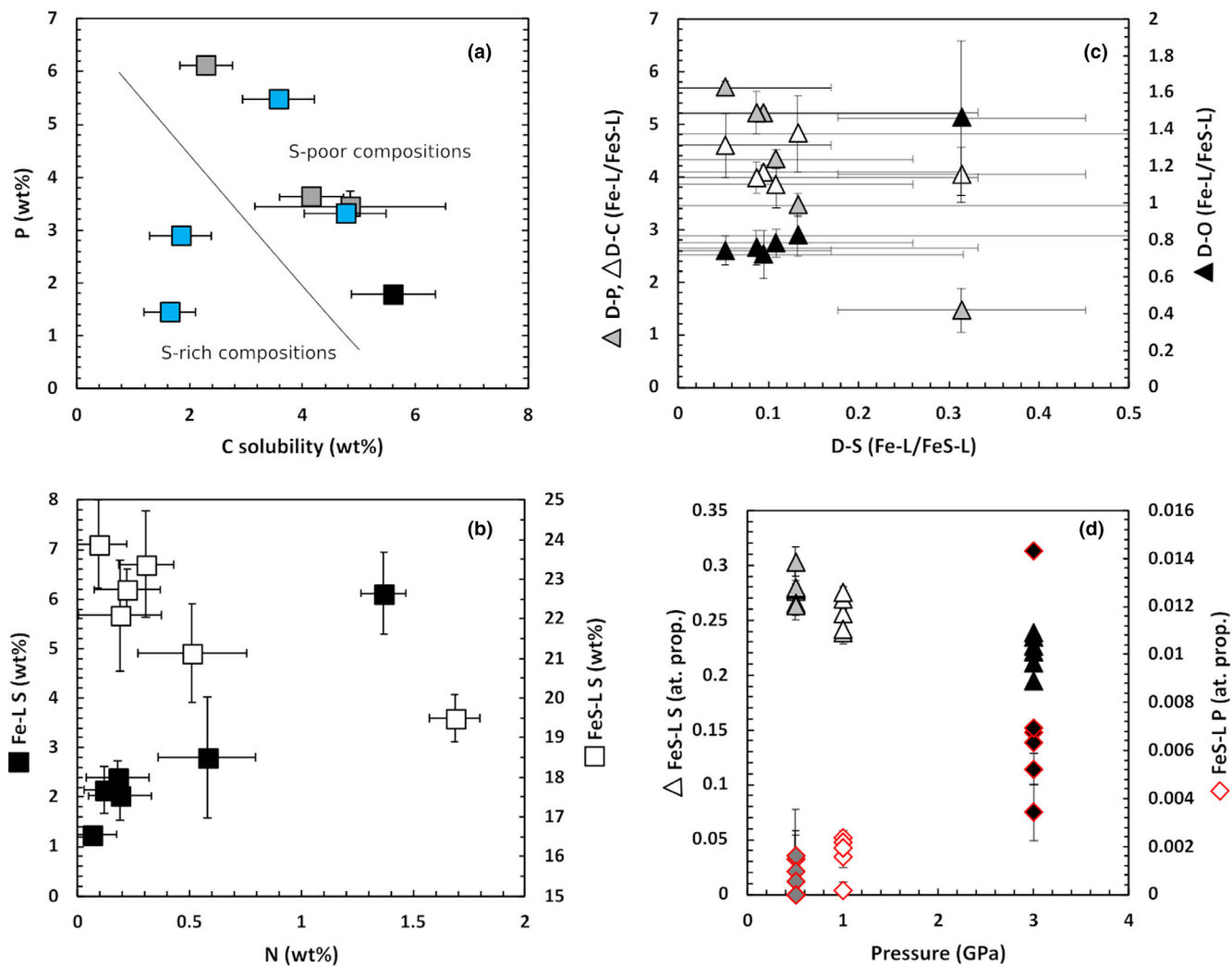


FIGURE 3. Compositional trends for miscible and immiscible Fe-rich liquids. (a) C saturation at 3 and 5 GPa as a function of P content of miscible liquid (gray/blue, for 3/5 GPa experiments, respectively) and Fe-rich liquid (black). (b) N content of Fe-rich liquid (black) and FeS-rich liquid (white) as a function of S content. (c) Relationships between partition coefficients, $D_X^{Fe-L/FeS-L}$ (wt%, Fe-rich liquid/FeS-rich liquid) at 3 GPa. Gray: decrease in $D_P^{Fe-L/FeS-L}$ as $D_S^{Fe-L/FeS-L}$ approaches 1, due to closure of the immiscibility gap; white: $D_C^{Fe-L/FeS-L}$ insensitive to changes in S content of immiscible liquids. Black: unclear relationship of $D_O^{Fe-L/FeS-L}$ with immiscible liquid S contents, with only moderate partitioning. (d) Pressure dependence of S content (triangles), and P content (diamonds) of immiscible FeS-rich liquid. Black: this study 3 GPa, white and gray: Bromiley et al. (2024), 1 and 0.5 GPa, respectively. (Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/maps.70135))

markedly reduced, similar to both ternary systems. There are corollary changes in immiscible FeS-rich liquid compositions (Figures 3d and 4c). The S versus P projection emphasizes the significant influence of small amounts of C in reducing the extent of liquid miscibility. This is evident not only from the 1 GPa experiments of Bromiley et al. (2024) (Figure 4a), but also from the comparison of results at 3 GPa (Figure 4b). Although Zhai et al. (2022) noted full liquid miscibility over a wide range of bulk P and S contents in C-free experiments, data here demonstrate that addition of c. 2 wt% C opens up a significant miscibility gap.

Figure 5 shows experimental data at 1, 3, and 5 GPa projected onto S + O versus C + P, which allows comparison with data from Fe-S-P and Fe-S-C ternary systems. Again, the effect of pressure in closing the miscibility gap for S-P-C bearing systems is evident. Changes in immiscible liquid composition as a function of pressure and bulk composition are also highlighted, for example, a decrease in FeS-rich liquid S content, and increase in C and P content. FeS-rich liquid compositions are similar to those in the Fe-S-C system (Dasgupta et al., 2009). In contrast, S contents of Fe-rich liquids are higher in P-bearing systems. Pressure dependence of the

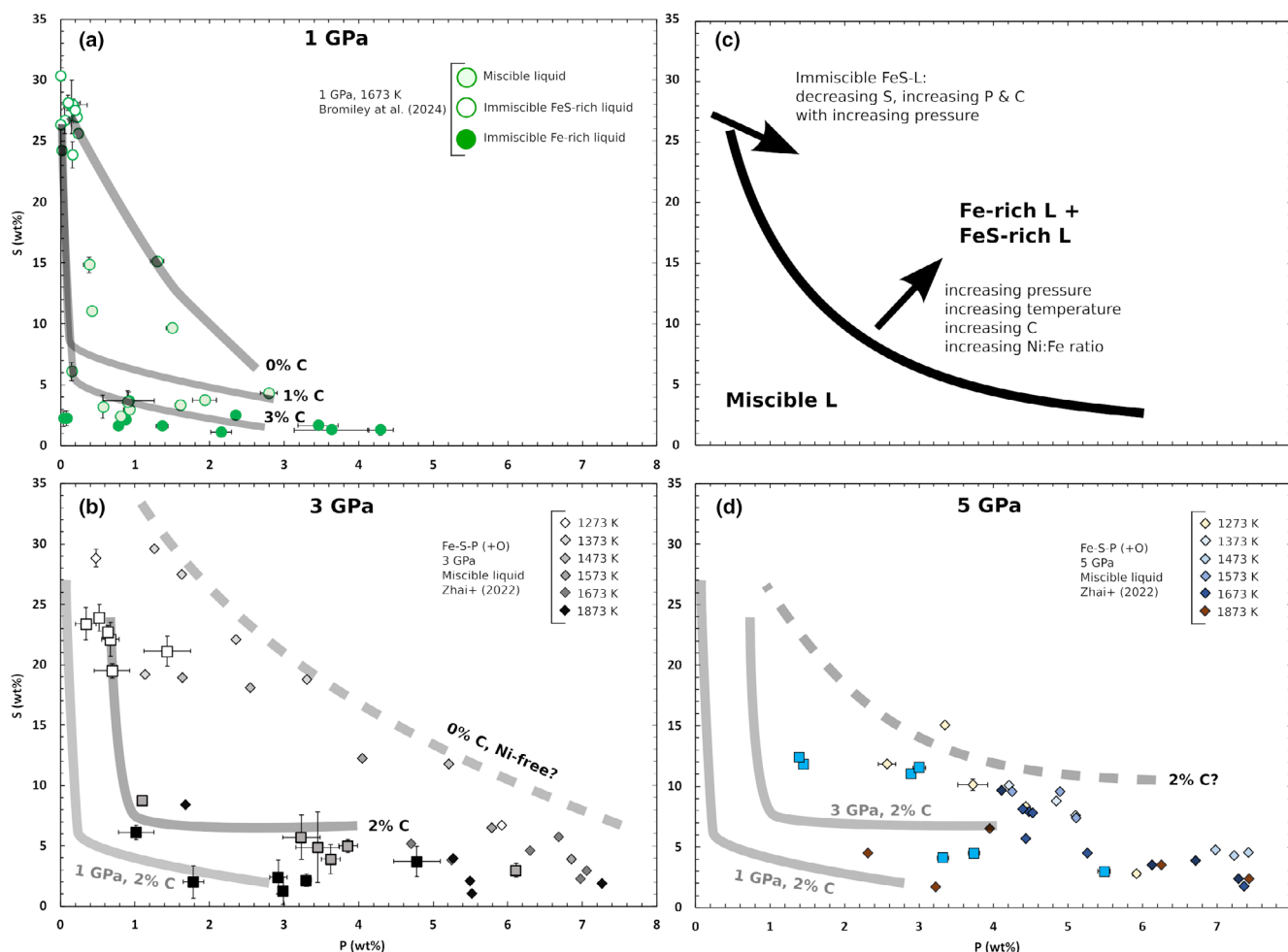


FIGURE 4. Summary of experimental data on the extent of miscibility in Fe liquids at (a) 1 GPa, (b) 3 GPa, and (c) 5 GPa, from this study and Bromiley et al. (2024), in the system $\text{Fe}_{0.9}\text{Ni}_{0.1} + \text{S}, \text{P}, \text{C} (+\text{O}, \text{N})$, and 3 and 5 GPa data from Zhai et al. (2022), for the system $\text{Fe} + \text{S} + \text{P} (+\text{O})$. (a) 1 GPa liquid compositions from Bromiley et al. (2024) with their estimated miscibility curves as a function of measured bulk C content; C contents of run products which constrain miscibility curves can be found in Bromiley et al. (2024). (b) 3 GPa data from this study and Zhai et al. (2022). Position of the miscibility curve at approximately 2 wt% C is shown (3 GPa: dark gray line; 1 GPa: light gray line). The C-free miscibility curve is not constrained, with all 3 GPa run products from Zhai et al. (2022) containing a single, miscible liquid; its (minimum) position is sketched here (dashed gray line). (c) Summary of effects of composition, pressure, and temperature on the extent of miscibility in Fe-rich liquids; systematic changes in FeS-rich liquid are noted with increasing pressure (Figure 3d). (d) 5 GPa data from this study and Zhai et al. (2022). Position of 2 wt% C miscibility curve is estimated (gray line), although not constrained. 1 and 3 GPa miscibility curves shown for comparison. (Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/maps.70135))

projected miscibility curve in Figure 5 is broadly consistent with experimental data in the Fe-S-C system, although our data imply effective closure of the miscibility gap at lower pressures, despite the presence of P. Data from Dasgupta et al. (2009) imply liquid miscibility in C-saturated systems persists to 5 GPa, although this is presumably close to the upper pressure limit of immiscibility due to the intermediate S compositions of immiscible liquids reported. However, Zhang et al. (2018) caution that C contents of these intermediate S-bearing liquids are inconsistent with their experimentally determined C solubility data, and should

be treated with caution. Data from Deng et al. (2013) constrain the upper pressure limit of immiscibility in the Fe-S-C ternary system between 3.5 and 5 GPa, although as shown in Figure 5, this is because of lower bulk S contents used in their study.

Contrasting pressure dependence of immiscibility in the systems Fe-S-C and Fe-S-P suggests that more complex systems behave closer to the Fe-S-C ternary at higher pressures. This is also apparent when comparing C contents required to drive liquid immiscibility in Fe/Ni-S-P liquids. At 0.5–1.0 GPa (Bromiley et al., 2024), immiscibility is noted in S-poor liquids (<5 wt% S) which

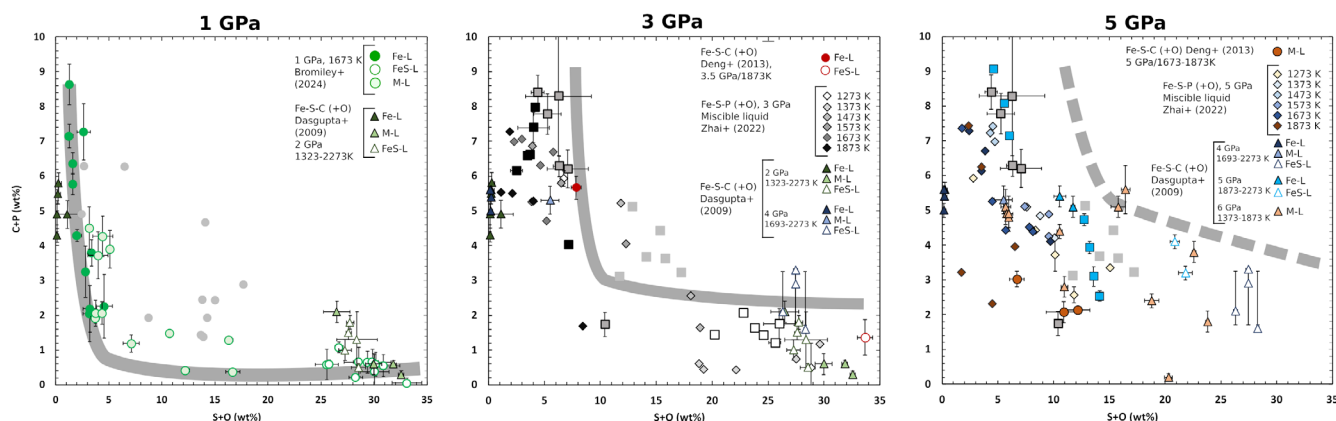


FIGURE 5. Effect of pressure on the extent of immiscibility in Fe-rich liquids, projected onto S + O versus C + P. Data (this study, squares) as in Figure 2, with additional liquid compositions from the Fe-S-C system at various pressures/temperatures from Deng et al. (2013) and Dasgupta et al. (2009). Note that the latter includes low fraction liquids coexisting with solid Fe-rich phases. Miscibility curve at 5 GPa is not constrained directly from data here, but its lower bound is estimated based on a compendium of data. (Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/maps.70135))

are C saturated (>4 wt% C), or P-rich. At 3 GPa, there is full miscibility at these low S contents, even at C saturation; immiscibility is noted in systems with c. 10 wt% S and at least 1–2 wt% C. Data from Zhang et al. (2018) suggest that for these compositions, C saturation is at slightly greater than 2 wt%, although the presence of P (and Ni) probably means that these compositions are close to C saturation. At 5 GPa, we note liquid miscibility at c. 15 wt% S, even in P-bearing, C-saturated systems. The only exceptions to this overall trend are P-rich experiments HP_im_exp6a and 7b, in which immiscibility occurs in low S, C-saturated systems at 3 GPa due, presumably, to P–C interactions.

Influence of S-C, S-P, and P-C Interactions

Liquid immiscibility in Fe-S-C and Fe-S-P ternary systems arises due to large, positive S-C and S-P interaction parameters. Closure of miscibility gaps in both systems with increasing pressure implies that (1) both interaction parameters approach zero with increasing pressure—that is, light element mixing becomes increasingly ideal—and (2) the S-P interaction parameter is more pressure sensitive. Our new data highlight additional complexity in S-P-C mixing. For example, experiments HP_im_exp6a and HP_im_exp7b had the starting composition 5.7 wt% P and 5 wt% C, and were both run at 3.0 GPa. HP_im_exp6a, run at 1673 K, produced immiscible Fe-rich and FeS-rich liquids with (inferred) coexisting graphite and Fe_3P . This is in contrast to the C-free experiments of Zhai et al. (2022) who reported a single, miscible liquid for the same run conditions. HP_im_exp6a, therefore, brackets the miscibility curve at 3.0 GPa, 1673 K between C-free

and C-saturated P-rich systems. The presence of Fe_3P in this experiment is consistent with observation that C reduces P solubility in Fe-S liquids (Figure 3a); Zhai et al. (2022) only noted Fe_3P stability up to 1273 K in C-free systems.

Experiment HP_im_exp7b was conducted at higher temperature. Run products are C saturated but contain a single, miscible liquid and no separate P-rich phase. As such, HP_im_exp6a and HP_im_exp7b bracket the miscibility curve between 1673 and 1773 K at 3 GPa for this P-rich, C-saturated composition. Miscible liquid in HP_im_exp7b is not P-saturated, presumably due to a lower C content ($2.3 \text{ wt.} \pm 0.5\%$ C). In contrast, P solubility in immiscible liquids in HP_im_exp6a is limited due to (1) the high C content in Fe-rich liquid ($5.6 \pm 0.7 \text{ wt\% C}$) and (2) the high S content of FeS-rich liquid, which results in P saturation and formation of a separate P-rich phase (Fe_3P).

S-P, S-C, and P-C interaction parameters in Fe-liquids can be rationalized by considering local atomic structure and bonding environments. C is an interstitial element in S-bearing Fe liquids (Zhao et al., 2023), and repulsive S-C interactions between interstitial C and S, arising from the more ionic nature of Fe-S bonding, limit the extent of C dissolution (Deng et al., 2013; Tafwidli & Kang, 2017). P incorporation in Fe-rich liquids is complex, and Fe-P bonding is mixed metallic-covalent in nature. At high P contents, P-rich clusters occur, although P is probably largely present as isolated substitutional defects for modest P contents in our experiments (Jiang et al., 2025). Positive S-P interaction parameters imply that S-P interactions are thermodynamically and structurally unfavorable. Our observations also indicate that the interaction parameter for interstitial C and

substitutional P is positive. This is consistent with observations of Fe-rich liquids at 1 bar, in which C has been shown to increase P activity, resulting in a mutual lowering of solubility (e.g., Do et al., 2017). However, as C and P both preferentially partition into immiscible Fe-rich (S-poor) liquids, P-C repulsion may have a less significant effect on liquid compositions in complex systems compared to S-C and S-P interaction.

Other Controls on Liquid Immiscibility in Core-Forming Liquids

Immiscibility in Fe liquids is only characteristic below a critical temperature, above which the entropic gain from light element mixing offsets the enthalpic penalty associated with S-P-C interactions. Although we note some effect of temperature on immiscible liquid compositions ([Influence of S-C, S-P and P-C Interactions](#) Section), it is likely that the role of temperature in enhancing miscibility in core-forming liquids at low pressure is modest due to the limited temperature range over which planetesimal differentiation occurred. For example, thermodynamic models in the Fe-S-C system (Tafwidli & Kang, 2017) predict that for a system with 2 wt% C, increasing temperature from 1673 to 1873 K shifts the miscibility curve by only c. 3 wt% S.

C has a marked influence on liquid stability in Fe-rich systems. However, the role of C is nuanced due to compositional controls of C solubility. C solubility in Fe liquids is systematically reduced with increasing S content. The large, positive S-C interaction parameter contributes to this decrease in C solubility, although C solubility is also dependent on Fe-C and Fe-S interactions. Experimental data from Zhang et al. (2018) suggest that pressure only has a modest effect on C solubility up to 8 GPa, in contrast to the strong control of pressure on the extent of miscibility (Dasgupta et al., 2009). This means that the interplay between C saturation, which results in the formation of C-rich phases such as graphite, and liquid unmixing varies with pressure. The role of components such as P in modifying C solubility (Figure 3a) may, by extension, be important in constraining compositional controls on immiscibility.

The presence of Ni also decreases C solubility in Fe-FeS liquids (Zhang et al., 2018), and enhances miscibility. However, although Bromiley et al. (2024) inferred a role of Ni in enhancing miscibility in $\text{Fe}_{0.9}\text{Ni}_{0.1}$ composition liquids at 0.5–1 GPa, the influence of Ni on liquid miscibility and C solubility in Ni-poor liquids is poorly constrained and extrapolated from Ni-rich systems. Similarly, the presence of O may also influence miscibility in Fe liquids. Tsuno et al. (2007) observed a miscibility gap between Fe-rich metallic and FeO-rich ionic

liquids in the binary Fe-O and ternary Fe-S-O systems up to 21 GPa, at temperatures exceeding 2173 K. Sample/capsule/atmosphere interactions in high-pressure experiments commonly result in minor O incorporation in Fe liquids, presumably dependent on oxygen fugacity. However, although studies consistently demonstrate partitioning of O into immiscible FeS-rich liquids, it is unclear whether incorporation of minor O—< 5 atom% O for all liquids in this study—has a significant influence on miscibility.

Assessing the Importance of Immiscibility During Differentiation of Planetesimals and Planetary Embryos

The role of liquid immiscibility during planetary differentiation is mainly defined by the composition of core-forming liquids and the size of planetary bodies, that is, interior pressure. We limit discussion here to core formation in less reduced bodies, that is, applicable to parent bodies for the majority of primitive and differentiated meteorites for which oxygen fugacity, $f\text{O}_2$, estimates range from $\log f\text{O}_2 = \text{IW}$ to $\text{IW}-4$ (i.e., from the Fe-FeO buffer to 4 log units below the Fe-FeO buffer). Under more reducing conditions, extending to $\log f\text{O}_2 = \text{IW}-10$, increasing solubility of Si in Fe-rich liquids results in a miscibility gap in Fe-S-Si-rich systems (Morard et al., 2008). These conditions, and the role of Si, are likely limited to reduced planetary bodies such as Mercury, with estimated $\log f\text{O}_2$ ranging from $\text{IW}-2.6$ to $\text{IW}-7.3$ (Edmund et al., 2022).

Core-Forming Liquid Compositions Constrained From Magmatic Iron Meteorites

Magmatic iron meteorites evidence metal segregation in at least 60 distinct rocky parent bodies, probably within a few Myr of the formation of Ca-Al rich inclusions (CAIs) (Goldstein et al., 2009; Jones, 2024; Kleine et al., 2002, 2005, 2009; Kleine & Wadhwa, 2017; Spitzer et al., 2021, 2025). Most iron meteorites appear to have crystallized from metallic bodies a few 100 km in diameter with variable thicknesses of surrounding silicate (Goldstein et al., 2009; Jones, 2024). The size of differentiated parent bodies from which these liquids ultimately derived could have ranged from a few 10 km to approximately 1000 km in diameter (Goldstein et al., 2009).

Figure 6a–c shows modeled iron meteorite parental liquid (IMPL) compositions compared to melt relations in the system Fe-Ni-S-P-C. S and P contents for IMPL are taken from Hilton et al. (2022) and C contents from Grewal and Asimow (2023). For reference, Figure 6d shows body center, that is, maximum internal, pressure versus radius fields for differentiated rocky planetary bodies. Radii were calculated as a function of pressure assuming hydrostatic equilibrium (e.g., Poirier, 2000) of a

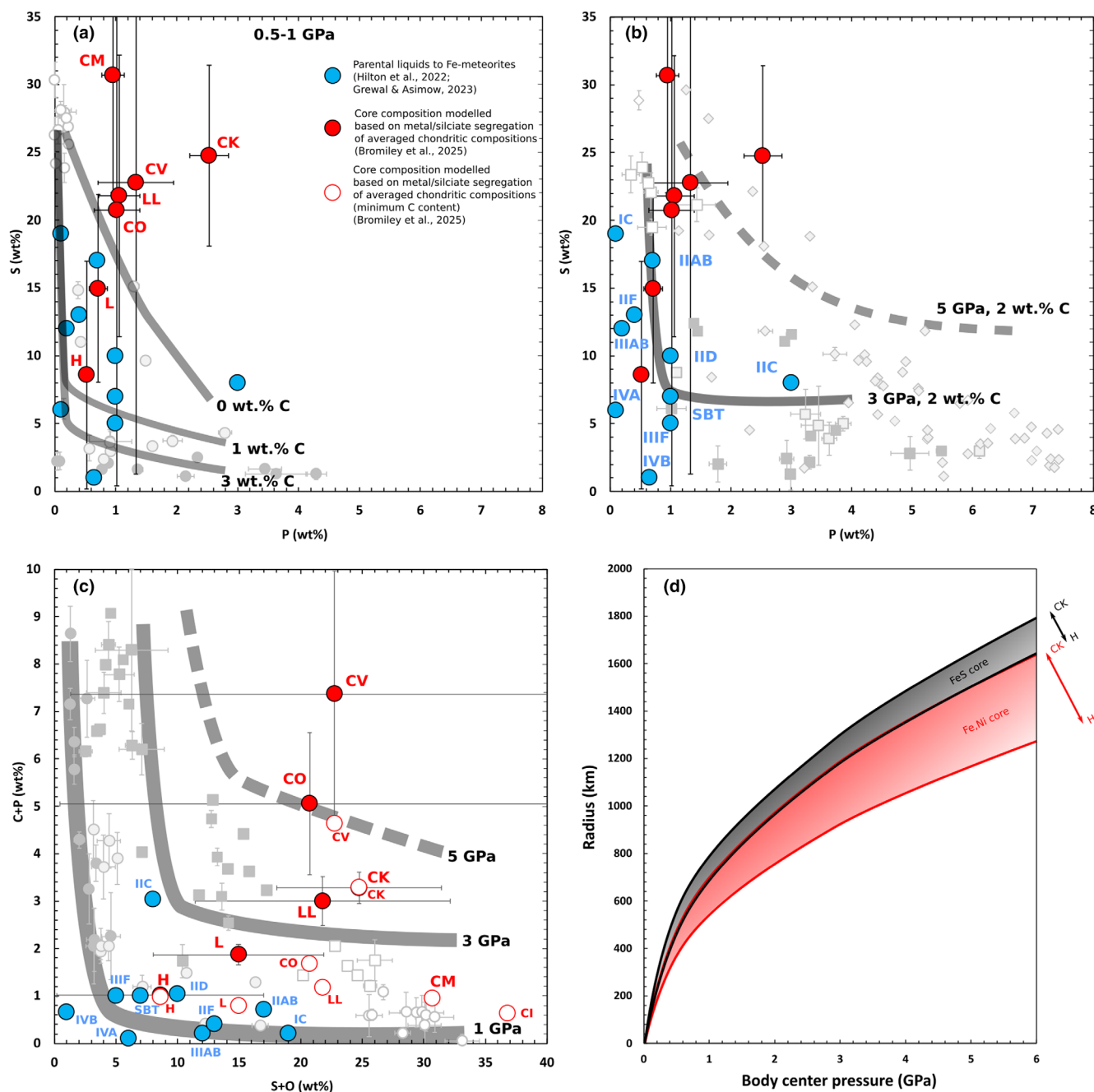


FIGURE 6. Estimates of core-forming liquid compositions. (a–c) show phase diagrams from Figures 4 and 5, with liquid and bulk compositions shown in light gray for clarity. Blue circles are compositions of modeled parental liquids for various groups of iron meteorites, with S and P contents from Hilton et al. (2022) and C contents from Grewal and Asimow (2023). Red open and filled circles are modeled core compositions for planetesimals assuming metal/silicate segregation for various averaged chondrites from Bromiley et al. (2024). Filled circles are core compositions calculated using average C contents. Open circles are compositions based on minimum projected C contents for each group. Red and blue text refers to specific groups of chondrite and iron meteorites. (d) Modeled body center pressures for 2-layer (metal core + silicate mantle) planetary bodies, based on core mass fractions from metal/silicate segregation of averaged chondrite compositions. Black: body center pressure as a function of planetesimal radius assuming a core density of troilite (FeS). Red: corresponding body center pressure assuming a core density of taenite (i.e., S-free). Fields shown encompass the range of pressure/range curves for all average chondrite compositions, delimited by curves for H and CK chondrites. (Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/maps.70135))

cooled two-layer, metal core + silicate mantle body. For this calculation, we adapted the method of Bercovici et al. (2022), who modeled core/mantle segregation for various average chondrite compositions.

Firstly, following Bercovici et al. (2022), we calculated silicate mantle density using an empirical P/T dependency of density (Kaus et al. 2005). We used a temperature of 298 K to mimic a cooled planetary body, and for a first approximation, a fixed body center pressure. This overestimates mantle density, although for small planetary bodies the error is small, and pressure versus radii estimates calculated by this method are in close agreement with those derived using a Vesta-like core mass fraction and fixed mantle density (Miura et al., 2025). Planetary radii were then calculated as a function of body center pressure for fixed values of core mass fractions and core densities. Core mass fractions for various average chondrite compositions were taken from Bromiley et al. (2024). To constrain a representative range of body center pressures for differentiated planetesimals, we calculated pressure versus radius curves for both (1) a fixed core density of 4.61 g cm^{-3} , that is, the density of troilite, FeS, and (2) a fixed core density of 7.87 g cm^{-3} , that is, the density of taenite, Fe,Ni. Figure 6d shows fields for pressure vs radius relationships assuming both S-rich and S-free metal cores, defined by the curves for average H—that is, the composition with the highest modeled core fraction—and average CK—lowest modeled core fraction. Full results for all chondrite groups are available in the associate data repository.

For an upper bound of iron meteorite parent bodies—IMPB, that is, the differentiated rocky planetesimals from which magmatic iron meteorites ultimately derived—of 1000 km diameter, maximum internal pressures are at or below the pressure range of experiments reported by Bromiley et al. (2024), that is, $<0.5\text{--}1.0 \text{ GPa}$. As estimated C contents of IMPL are typically $\ll 1 \text{ wt\%}$ (Grewal & Asimow, 2023), the 0 wt% C miscibility curve in Figure 6a is most suitable for assessing liquid stability. All IMPL compositions, except that for the IIC group, lie within the miscible liquid field. The IIC IMPL composition lies in the two-liquid field due to its higher P content. However, constraints on P and C contents of parental liquids are less accurate, with other studies suggesting a lower P content for IIC IMPL which lies within the miscible liquid field (Tornabene et al., 2020). As such, although modeled parental liquids for magmatic irons are variably S-rich to S-poor, low P and C contents imply that were single, miscible liquids, assuming that decreasing pressure below 0.5 GPa does not significantly expand the miscibility gap.

S, P, and C are variably volatile in silicate and metallic liquids. IMPL compositions may, therefore, partly reflect volatile loss before and during differentiation of parent

bodies, and following subsequent planetary disruption (Bromiley et al., 2024; Grewal & Asimow, 2023; Hirschmann et al., 2021). Element volatility in liquid metal can be estimated by comparing vapor pressures above liquid Fe alloys at high temperature. This implies that S is highly volatile, that P is somewhat less volatile, and that C is relatively refractory (Do et al., 2017; Wu et al., 2015), although the accuracy of this assessment is limited by the fact that literature values for vapor pressures are from Fe-rich (low S) liquids. Metallographic cooling rates and Pd-Ag isotope systematics in different groups of magmatic iron meteorites indicate disruption of parent bodies soon after differentiation, and rapid cooling of poorly shielded liquid metal (Hunt et al., 2022; Matthes et al., 2018; Yang et al., 2007, 2010). As such, partial degassing of S and P, and to a much lesser extent C, during and after impact disruption is possible.

Whether volatile-rich magmatic iron groups evidence pristine core compositions is difficult to ascertain. Evidence of post-disruption moderately volatile element (MVE) loss is recorded in some, but not all, groups of magmatic irons (Grewal et al., 2025; Rubin et al., 2022). C and N depletion in IIAB, IIIAB, IVA, and IVB groups, as well as a general pattern of C and N depletion in IMPB compared to chondrites, may also be indicative of volatile loss from parent body cores, possibly as a result of disruptive impact events (Grewal & Asimow, 2023). Conversely, there is no clear S isotope signature in magmatic irons for degassing, with variability in isotopic compositions instead reflective of heterogeneous distribution of S in the protoplanetary disc and of S volatility during accretion (Antonelli et al., 2014; Heiny et al., 2025). Similarly, MVEs such as Ge only provide clear evidence of isotopic fractionation due to evaporation in ungrouped iron meteorites, consistent with their formation by impact events on parent body surfaces (Wölfer et al., 2025).

However, regardless of the extent to which magmatic irons evidence pristine core-forming liquid compositions, it is unlikely that they represent the full range of compositions of differentiating planetary bodies. The limited size of IMPB and their rapid formation indicate that they sample a population of small, early formed differentiated planetesimals. As such, magmatic irons may represent bodies that underwent more substantial melting and degassing during differentiation. Differentiation processes and core compositions in latter-formed planetary bodies may have differed considerably (Kruijer et al., 2014).

Core-Forming Liquid Compositions Constrained From Metal/Silicate Segregation Models

Compositions of core-forming liquids can be constrained by modeling silicate/metal differentiation of

average chondrite compositions. Figure 6 shows estimated “chondrite” core compositions from Bromiley et al. (2024). Large error bars for these compositions reflect substantial compositional variability within each chondrite group, especially for S. Compositions are calculated after the method of Bercovici et al. (2022), based on (1) average C contents for each chondrite group, and (2) projected minimum C contents for each group. The latter calculation addresses the possibility that C contents of chondrites partly reflect secondary enrichment of C on parent bodies, and that highly volatile C may be more readily lost from silicate melts during planetary differentiation (Bromiley et al., 2024; Hirschmann et al., 2021). Modeled chondrite core compositions are substantially more S-rich than IMPL, and also variably C-rich, depending on methodology used, although they have a similar range of P contents. Compositions either imply an important role of liquid immiscibility during differentiation, or differentiation of monosulfide rather than Fe-rich liquid cores.

At 0.5–1.0 GPa (Figure 6a,c), S, P, and C contents of all modeled chondrite core compositions lie in the immiscible liquids field. At 3.0 GPa, modeled core compositions for H chondrite lie within the single, miscible liquid field (Figure 6b,c). Modeled L chondrite compositions also largely lie within the miscible liquid field, although close to the miscibility curve for “average C” calculations. Modeled LL core compositions either lie in miscible or immiscible liquids fields, depending on C content. A body center pressure of 3 GPa corresponds to differentiated bodies of c. 2×10^3 km diameter (Figure 6d). Low P contents suggest that the importance of immiscibility during differentiation of planetesimals with NC compositions depends mainly on S and C contents. However, unless significant S and C loss occurred prior to or during differentiation, that is, during melting at higher temperatures than those experienced by chondrite parent bodies, immiscibility would have controlled compositions of core-forming liquids in NC composition planetesimals and planetary embryos up to 10^3 km in diameter.

At 3 GPa, modeled CC core compositions lie within the immiscible liquids field (CV, CK), are single liquid sulfides (CM, CI), or only lie within/close to the miscible liquid field at minimum modeled C contents (CO). Immiscibility would, therefore, have an important influence on differentiation of planetesimals and up to moderate sized planetary embryos—bodies up to c. $2\text{--}3 \times 10^3$ km in diameter—with CC compositions, unless substantial volatile loss occurred prior to/during differentiation. Closure of the miscibility gap likely occurs between 4 and 5 GPa for the range of core compositions in differentiated bodies. This extends the role of core immiscibility up to core pressures in icy

moons such as Europa for bodies with broadly CC (C-rich, S-rich) bulk compositions.

Planetary Differentiation and Volatile Loss

Composition of core-forming liquids depends on bulk composition of planetary bodies, and the extent of volatile loss (S, P, C) both prior to and during melting and core segregation. The latter, in turn, is controlled by the nature of planetary melting and differentiation (e.g., Bromiley, 2023; Grewal et al., 2022; Hirschmann et al., 2021; Pathak & Dasgupta, 2024). Extensive melting of small planetary bodies, especially those with external magma oceans, would have resulted in significant volatile loss. In contrast, bodies which underwent lower degrees of melting, and had internal magma oceans insulated by thick silicate crusts, likely experienced limited volatile loss. This would have favored segregation of immiscible core-forming liquids, and possibly, formation of differentiated bodies with layered S-poor inner, and S-rich outer cores. Volatile loss would be minimized in bodies in which core-forming liquids segregated via percolation of metallic liquids through solid silicate, for example, in more oxidized planetesimals in which core segregation proceeded prior to the onset of silicate melting due to the wetting properties of O-bearing, S-rich liquids (e.g., Crossley et al., 2025; Miura et al., 2025; Terasaki et al., 2008). Liquid immiscibility during lower temperature, percolation-driven core formation is tentatively supported by melting experiments conducted using chondritic bulk compositions, in which coexisting S-rich and S-poor liquids are reported (Bromiley, 2023). Under such conditions, liquid immiscibility may aid percolation as it stabilizes immiscible S-rich (O-bearing) metallic liquids which are able to wet silicate grain boundaries (Terasaki et al., 2008) and segregate from both silicate and Fe-rich metal to form S-rich (monosulfide) metallic cores.

CONCLUSIONS

We performed superliquidus experiments at 3.0 and 5.0 GPa which constrain the effect of pressure on liquid immiscibility in the system $\text{Fe}_{0.9}\text{Ni}_{0.1} + \text{S}$, P, C. Run products contained additional minor N and O due to sample/capsule interaction. Comparison of results with those from previous studies provides insight into the role of liquid immiscibility during differentiation and metal core formation in rocky planetesimals and planetary embryos.

In complex systems, immiscibility results in separation of Fe-rich liquids (S poor, but enriched in P and C) and FeS-rich liquids (S rich, depleted in P and C). N and O are also partitioned between immiscible liquids, although low concentrations mean that they likely have a

minor role on liquid stability. For compositions applicable to differentiated rocky bodies, closure of the immiscibility gap occurs below, but probably close to, 5 GPa, corresponding to body center pressures in planetary bodies up to c. 3×10^3 km in diameter. At lower pressures, immiscibility arises due to the highly non-ideal nature of light element mixing in Fe liquids, that is, large, positive interaction parameters between light elements. Non-ideality of mixing decreases in extent with increasing pressure. However, the pressure sensitivity of interaction parameters is variable for different element pairs, with a miscibility gap extending up to at least 5 GPa in the Fe-S-C system, but to less than 3 GPa in the Fe-S-P system. As a consequence, compositionally complex systems behave closer to the Fe-S-C system at higher pressure, except in P-rich, C-rich compositions. However, although the Fe-S-C ternary is a useful analog system for modeling immiscibility in core-forming liquids, its applicability at lower pressures is limited, and liquid compositions under all conditions are controlled by complex S-P-C interactions.

Comparison with core compositions estimated from magmatic iron, CC, and NC meteorite data indicate that liquid immiscibility may have been important during core segregation in rocky planetesimals and in small to medium-sized planetary embryos. The extent of immiscibility in core-forming liquids largely depended on light element content, which was, in turn, dependent on parent body bulk composition and the extent of volatile (S, P, C) loss prior to and during differentiation. The latter is sensitive to differentiation mechanisms and the extent of silicate melting. Liquid immiscibility, in turn, could have influenced differentiation processes, and may have resulted in formation of differentiated bodies with layered S-poor inner and S-rich outer cores, and/or bodies with S-rich cores.

Acknowledgments—The authors acknowledge financial support of the Royal Society (International Exchange grant IEC\R3\213008) and Leverhulme Trust (RPG-2021-015), and thank Prof. Satoru Urakawa for his support and guidance in conducting multi-anvil experiments, and Dr. Nicola Cayzer for her support in conducting SEM analysis. The authors thank Prof. K. Righter for handling this manuscript, and two anonymous reviewers, who provided a number of useful and informed comments and suggestions.

Data Availability Statement—Original data associated with this publication are available at: <https://datashare.ed.ac.uk/handle/10283/9152>. The data that support the findings of this study are available from the corresponding author upon reasonable request. Upon acceptance, all data will be made available via the

University of Edinburgh Datashare repository (freely available, in perpetuity).

Editorial Handling—Dr. Kevin Righter

REFERENCES

- Antonelli, M. A., Kim, S.-T., Peters, M., Labidi, J., Cartigny, P., Walker, R. J., Lyons, J. R., Hoek, J., and Farquhar, J. 2014. Early Inner Solar System Origin for Anomalous Sulfur Isotopes in Differentiated Protoplanets. *Proceedings of the National Academy of Sciences of the United States of America* 111: 17749–54.
- Barrett, T. J., Barnes, J. J., Anand, M., Franchi, I. A., Greenwood, R. C., Charlier, B. L. A., Zhao, X., Moynier, F., and Grady, M. M. 2019. Investigating Magmatic Processes in the Early Solar System Using the Cl Isotopic Systematics of Eucrites. *Geochimica et Cosmochimica Acta* 266: 582–597.
- Bercovici, H. L., Elkins-Tanton, L. T., O'Rourke, J. G., and Schaefer, L. 2022. The Effects of Bulk Composition on Planetesimal Core Sulfur Content and Size. *Icarus* 380: 114976.
- Berg, M. T. L., Bromiley, G. D., Butler, I. B., Frost, M., Bradley, R., Carr, J., le Godec, Y., et al. 2017. Deformation-Aided Segregation of Fe-S Liquid from Olivine under Deep Earth Conditions: Implications for Core Formation in the Early Solar System. *Physics of the Earth and Planetary Interiors* 263: 38–54.
- Berg, M. T. L., Bromiley, G. D., Le Godec, Y., Philippe, J., Mezouar, M., Perrillat, J.-P., and Potts, N. J. 2018. Rapid Core Formation in Terrestrial Planets by Percolative Flow: In-Situ Imaging of Metallic Melt Migration under High Pressure/Temperature Conditions. *Frontiers in Earth Science* 6: 77.
- Bromiley, G. D. 2023. The Geochemical Legacy of Low-Temperature, Percolation-Driven Core Formation in Planetesimals. *Earth, Moon, and Planets* 127: 4.
- Bromiley, G. D., Varnam, M., Terasaki, H., Komabayashi, T., and Barosch, J. 2024. The Extent of Liquid Immiscibility in Planetesimal Cores. *Geochemical Perspectives Letters* 33: 7–12.
- Chabot, N. L., and Drake, M. J. 2000. Crystallization of Magmatic Iron Meteorites: The Effects of Phosphorus and Liquid Immiscibility. *Meteoritics & Planetary Science* 35: 807–816.
- Corgne, A., Wood, B. J., and Fei, Y. 2008. C- and S-Rich Molten Alloy Immiscibility and Core Formation of Planetesimals. *Geochimica et Cosmochimica Acta* 72: 2409–16.
- Crossley, S. D., Setera, J. B., Anzures, B. A., Iacovino, K., Buckley, W. P., Eckley, S. A., O'Neal, E. W., Maisano, J. A., Simon, J. I., and Righter, K. 2025. Percolative Sulfide Core Formation in Oxidized Planetary Bodies. *Nature Communications* 16: 3233.
- Dasgupta, R., Buono, A., Whelan, G., and Walker, D. 2009. High-Pressure Melting Relations in Fe-C-S Systems: Implications for Formation, Evolution, and Structure of Metallic Cores in Planetary Bodies. *Geochimica et Cosmochimica Acta* 73: 6678–91.
- Dasgupta, R., and Walker, D. 2008. Carbon Solubility in Core Melts in a Shallow Magma Ocean Environment and Distribution of Carbon between the Earth's Core and the Mantle. *Geochimica et Cosmochimica Acta* 72: 4627–41.

- Deng, L., Fei, Y., Liu, X., Gong, Z., and Shahar, A. 2013. Effect of Carbon, Sulfur and Silicon on Iron Melting at High Pressure: Implications for Composition and Evolution of the Planetary Terrestrial Cores. *Geochimica et Cosmochimica Acta* 114: 220–233.
- Do, K.-H., Nam, H.-S., Jang, J.-M., Kim, D.-S., and Pak, J.-J. 2017. Thermodynamic Interaction between Chromium and Phosphorus in High Cr Containing Liquid Iron. *ISIJ International* 57: 1334–39.
- Dodds, K., Bryson, J. F. J., Neufeld, J., and Harrison, R. J. 2021. The Thermal Evolution of Planetesimals during Accretion and Differentiation: Consequences for Dynamo Generation by Thermally-Driven Convection. *Journal of Geophysical Research: Planets* 126: e2020JE006704.
- Edmund, E., Morard, G., Baron, M. A., Rivoldini, A., Yokoo, S., Boccato, S., Hirose, K., Pakhomova, A., and Antonangeli, D. 2022. The Fe-FeSi Phase Diagram at Mercury's Core Conditions. *Nature Communications* 13: 387.
- Elkins-Tanton, L. T., Asphaug, E., Bell, J. F., Bierson, C. J., Bills, B. G., Bottke, W. F., Courville, S. W., et al. 2022. Distinguishing the Origin of Asteroid (16) Psyche. *Space Science Reviews* 218: 17.
- Goldstein, J. I., Scott, E. R. D., and Chabot, N. L. 2009. Iron Meteorites: Crystallization, Thermal History, Parent Bodies, and Origin. *Geochemistry* 69: 293–325.
- Greenwood, R. C., Franchi, I. A., Jambon, A., and Buchanan, P. C. 2005. Widespread Magma Oceans on Asteroidal Bodies in the Early Solar System. *Nature* 435: 916–18.
- Grewal, D. S., and Asimow, P. D. 2023. Origin of the Superchondritic Carbon/Nitrogen Ratio of the Bulk Silicate Earth – An Outlook from Iron Meteorites. *Geochimica et Cosmochimica Acta* 344: 146–159.
- Grewal, D. S., Bhattacharjee, S., Zhang, B., Nie, N. X., and Miyazaki, Y. 2025. Enrichment of Moderately Volatile Elements in First-Generation Planetesimals of the Inner Solar System. *Science Advances* 11: eadq7848.
- Grewal, D. S., Dasgupta, R., Sun, C., Tsuno, K., and Costin, G. 2019. Delivery of Carbon, Nitrogen, and Sulfur to the Silicate Earth by a Giant Impact. *Science Advances* 5: eaau3669.
- Grewal, D. S., Nie, N. X., Zhang, B., Izidoro, A., and Asimow, P. D. 2024. Accretion of the Earliest Inner Solar System Planetesimals beyond the Water Snowline. *Nature Astronomy* 8: 290–97.
- Grewal, D. S., Seales, J. D., and Dasgupta, R. 2022. Internal or External Magma Oceans in the Earliest Protoplanets – Perspectives from Nitrogen and Carbon Fractionation. *Earth and Planetary Science Letters* 598: 117847.
- Heiny, E. A., Stolper, E. M., and Eiler, J. M. 2025. Differentiated Planetesimals Record Differing Sources of Sulfur in Inner and Outer Solar System Materials. *Proceedings of the National Academy of Sciences of the United States of America* 122: e2418198122.
- Hemmingway, B. S., Bohlen, S. R., Hankins, W. B., Westrum, E. F., and Kuskov, O. L. 1998. Heat Capacity and Thermodynamic Properties for Coesite and Jadeite, re-examination of the Quartz–Coesite Equilibrium Boundary. *American Mineralogist* 83: 409–418.
- Hevey, P. J., and Sanders, I. S. 2006. A Model for Planetesimal Meltdown by Al-26 and its Implications for Meteorite Parent Bodies. *Meteoritics & Planetary Science* 41: 95–106.
- Hilton, C. D., Ash, R. D., and Walker, R. J. 2022. Chemical Characteristics of Iron Meteorite Parent Bodies. *Geochimica et Cosmochimica Acta* 318: 112–125.
- Hirose, K., Wood, B., and Vočadlo, L. 2021. Light Elements in the Earth's Core. *Nature Reviews Earth & Environment* 2: 645–658.
- Hirschmann, M. M., Bergin, E. A., Blake, G. A., Ciesla, F. J., and Li, J. 2021. Early Volatile Depletion on Planetesimals Inferred from C-S Systematics of Iron Meteorite Parent Bodies. *Proceedings of the National Academy of Sciences* 118: e2026779118.
- Holland, T. J. B. 1980. The Reaction Albite = Jadeite + Quartz Determined Experimentally in the Range 600–1200°C. *American Mineralogist* 65: 129–137.
- Hunt, A. C., Theis, K. J., Rehkämper, M., Benedix, G. K., Andreasen, R., and Schönbachler, M. 2022. The Dissipation of the Solar Nebula Constrained by Impacts and Core Cooling in Planetesimals. *Nature Astronomy* 6: 812–18.
- Jiang, C., Liu, B., Liang, W., Zhang, J., and Li, K. 2025. Microstructure Evolution and Properties of Liquid Fe-P with Different Phosphorus Content: Molecular Dynamic Investigation. *Journal of Molecular Modeling* 31: 137.
- Jones, R. H. 2024. Meteorites and Planet Formation. *Reviews in Mineralogy and Geochemistry* 90: 113–140.
- Kaminski, E., Limare, A., Kenda, B., and Chaussidon, M. 2020. Early Accretion of Planetesimals Unraveled by the Thermal Evolution of the Parent Bodies of Magmatic Iron Meteorites. *Earth and Planetary Science Letters* 548: 116469.
- Kaus, B. J. P., Connolly, J. A. D., Podladchikov, Y. Y., and Schmalholz, S. M. 2005. Effect of Mineral Phase Transitions on Sedimentary Basin Subsidence and Uplift. *Earth and Planetary Science Letters* 233: 213–228.
- Keil, K. 2000. Thermal Alteration of Asteroids: Evidence from Meteorites. *Planetary and Space Science* 48: 887–903.
- Kleine, T., Munker, C., Mezger, K., and Palme, H. 2002. Rapid Accretion and Early Core Formation on Asteroids and the Terrestrial Planets from Hf-W Chronometry. *Nature* 418: 952–55.
- Kleine, T., Palme, H., Mezger, K., and Halliday, A. N. 2005. Hf-W Chronometry of Lunar Metals and the Age and Early Differentiation of the Moon. *Science* 310: 1671–74.
- Kleine, T., Touboul, M., Bourdon, B., Nimmo, F., Mezger, K., Palme, H., Jacobsen, S. B., Yin, Q. Z., and Halliday, A. N. 2009. Hf-W Chronology of the Accretion and Early Evolution of Asteroids and Terrestrial Planets. *Geochimica et Cosmochimica Acta* 73: 5150–88.
- Kleine, T., and Wadhwa, M. 2017. Chronology of Planetesimal Differentiation. In *Planetesimals: Early Differentiation and Consequences for Planets*, edited by L. Elkins-Tanton, and B. Weiss, 224–244. Cambridge, UK: Cambridge University Press.
- Komabayashi, T. 2014. Thermodynamics of Melting Relations in the System Fe-FeO at High Pressure: Implications for Oxygen in the Earth's Core. *Journal of Geophysical Research: Solid Earth* 119: 4164–77.
- Komabayashi, T. 2021. Phase Relations of Earth's Core-Forming Materials. *Crystals* 11: 581.
- Kruijer, T. S., Touboul, M., Fischer-Godde, M., Birmingham, K. R., Walker, R. J., and Kleine, T. 2014. Protracted Core Formation and Rapid Accretion of Protoplanets. *Science* 344: 1150–54.

- Lichtenberg, T., Drazkowska, J., Schonbachler, M., Golabekt, G. J., and Hands, T. O. 2021. Bifurcation of Planetary Building Blocks during Solar System Formation. *Science* 371: 365.
- Lichtenberg, T., Keller, T., Katz, R. E., Golabek, G. J., and Gerya, T. V. 2019. Magma Ascent in Planetesimals: Control by Grain Size. *Earth and Planetary Science Letters* 507: 154–165.
- Mare, E. R., Chen, J., Buisman, I., Hayward, C., Burnham, A. D., Melai, C., Stüeken, E. E., Bromiley, G., and Mikhail, S. 2025. Problems and Solutions when Quantifying Nitrogen in Silicate and Oxide Minerals and Glasses Using Electron Probe Microanalysis. *Geostandards and Geoanalytical Research* 49: 569–590.
- Matthes, M., Fischer-Gödde, M., Kruijjer, T. S., and Kleine, T. 2018. Pd-Ag Chronometry of IVA Iron Meteorites and the Crystallization and Cooling of a Protoplanetary Core. *Geochimica et Cosmochimica Acta* 220: 82–95.
- Maurel, C., Bryson, J. F. J., Lyons, R. J., Ball, M. R., Chopdekar, V. R., Scholl, A., Ciesla, F. J., Bottke, W. F., and Weiss, B. P. 2020. Meteorite Evidence for Partial Differentiation and Protracted Accretion of Planetesimals. *Science Advances* 6: eaba1303.
- McCoy, T. J., Dickinson, T. L., and Lofgren, G. E. 1999. Partial Melting of the Indarch (EH4) Meteorite: A Textural, Chemical, and Phase Relations View of Melting and Melt Migration. *Meteoritics & Planetary Science* 34: 735–746.
- McDonough, W. F. 2014. 3.16 – Compositional Model for the Earth's Core. In *Treatise on Geochemistry*, edited by H. D. Holland, and K. K. Turekian, 2nd ed., 559–577. Oxford: Elsevier.
- Miura, T., Terasaki, H., Takaki, H., Kobayashi, K., Bromiley, G. D., and Yoshino, T. 2025. The Effect of Pressure on Dihedral Angle between Liquid Fe-S and Orthopyroxene: Implication for Percolative Core Formation in Planetesimals and Planetary Embryos. *Meteoritics & Planetary Science* 60: 2811–27.
- Morard, G., and Katsura, T. 2010. Pressure-Temperature Cartography of Fe-S-Si Immiscible System. *Geochimica et Cosmochimica Acta* 74: 3659–67.
- Morard, G., et al. 2008. In Situ Structural Investigation of Fe-S-Si Immiscible Liquid System and Evolution of Fe-S Bond Properties with Pressure. *Journal of Geophysical Research* 113: B10205.
- Moskovitz, N., and Gaidos, E. 2011. Differentiation of Planetesimals and the Thermal Consequences of Melt Migration. *Meteoritics & Planetary Science* 46: 903–918.
- Neumann, W., Breuer, D., and Spohn, T. 2014. Differentiation of Vesta: Implications for a Shallow Magma Ocean. *Earth and Planetary Science Letters* 395: 267–280.
- Neumann, W., Kruijjer, T. S., Breuer, D., and Kleine, T. 2018. Multistage Core Formation in Planetesimals Revealed by Numerical Modeling and Hf-W Chronometry of Iron Meteorites. *Journal of Geophysical Research, Planets* 123: 421–444.
- Pathak, D., and Dasgupta, R. 2024. Nitrogen Inventory of Iron Meteorite Parent Bodies Constrained by Nitrogen Partitioning between Fe-Rich Solid and Liquid Alloys. *Geochimica et Cosmochimica Acta* 371: 199–213.
- Poirier, J.-P. 2000. *Introduction to the Physics of the Earth's Interior*, 2nd ed. . Cambridge: Cambridge University Press. 326.
- Potts, N. J., Bromiley, G. D., and Brooker, R. A. 2021. An Experimental Investigation of F, Cl and H₂O Mineral-Melt Partitioning in a Reduced, Model Lunar System. *Geochimica et Cosmochimica Acta* 294: 232–254.
- Rubin, A. E., Zhang, B., and Chabot, N. L. 2022. IVA Iron Meteorites as Late-Stage Crystallization Products Affected by Multiple Collisional Events. *Geochimica et Cosmochimica Acta* 331: 1–17.
- Rückriemen, T., Breuer, D., and Spohn, T. 2018. Top-Down Freezing in a Fe-FeS Core and Ganymede's Present-Day Magnetic Field. *Icarus* 307: 172–196.
- Russell, C. T., Raymond, C. A., Coradini, A., McSween, H. Y., Zuber, M. T., Nathues, A., De Sanctis, M. C., et al. 2012. Dawn at Vesta: Testing the Protoplanetary Paradigm. *Science* 336: 684–86.
- Sanloup, C., and Fei, Y. 2004. Closure of the Fe-S-Si Liquid Miscibility Gap at High Pressure. *Physics of the Earth and Planetary Interiors* 147: 57–65.
- Scheinberg, A., Fu, R. R., Elkins-Tanton, L. T., and Weiss, B. P. 2015. Asteroid Differentiation: Melting and Large-Scale Structure. In *Asteroids IV*, edited by P. Michel, F. E. DeMeo, and W. F. Bottke, 533–552. Tuscon, AZ: University of Arizona.
- Spitzer, F., Burkhardt, C., Kruijjer, T. S., and Kleine, T. 2025. Comparison of the Earliest NC and CC Planetesimals: Evidence from Ungrouped Iron Meteorites. *Geochimica et Cosmochimica Acta* 397: 134–148.
- Spitzer, F., Burkhardt, C., Nimmo, F., and Kleine, T. 2021. Nucleosynthetic Pt Isotope Anomalies and the Hf-W Chronology of Core Formation in Inner and Outer Solar System Planetesimals. *Earth and Planetary Science Letters* 576: 117211.
- Sramek, O., Milelli, L., Ricard, Y., and Labrosse, S. 2012. Thermal Evolution and Differentiation of Planetesimals and Planetary Embryos. *Icarus* 217: 339–354.
- Steenstra, E. S., and van Westrenen, W. 2018. A Synthesis of Geochemical Constraints on the Inventory of Light Elements in the Core of Mars. *Icarus* 315: 69–78.
- Tafwidli, F., and Kang, Y. 2017. Thermodynamic Modeling of Fe-C-S Ternary System. *ISIJ International* 57: 782–790.
- Terasaki, H., Frost, D. J., Rubie, D. C., and Langenhorst, F. 2005. The Effect of Oxygen and Sulphur on the Dihedral Angle between Fe-O-S Melt and Silicate Minerals at High Pressure: Implications for Martian Core Formation. *Earth and Planetary Science Letters* 232: 379–392.
- Terasaki, H., Frost, D. J., Rubie, D. C., and Langenhorst, F. 2008. Percolative Core Formation in Planetesimals. *Earth and Planetary Science Letters* 273: 132–37.
- Tornabene, H. A., Hilton, C. D., Birmingham, K. R., Ash, R. D., and Walker, R. J. 2020. Genetics, Age and Crystallization History of Group IIC Iron Meteorites. *Geochimica et Cosmochimica Acta* 288: 36–50.
- Tsuno, K., Terasaki, H., Ohtani, E., Suzuki, A., Asahara, Y., Nishida, K., Sakamaki, T., Funakoshi, K., and Kikegawa, T. 2007. In Situ Observation and Determination of Liquid Immiscibility in the Fe-O-S Melt at 3 GPa Using a Synchrotron X-Ray Radiographic Technique. *Geophysical Research Letters* 34: L17303.
- Ulf-Møller, F. 1998. Effects of Liquid Immiscibility on Trace Element Fractionation in Magmatic Iron Meteorites: A Case Study of Group IIIAB. *Meteoritics & Planetary Science* 33: 207–220.

- Urakawa, S., Kato, M., and Kumazawa, M. 1987. Experimental Study on the Phase Relations in the System Fe-Ni-O-S up to 15 GPa. In *High-Pressure Research in Mineral Physics: A Volume in Honor of Syun-Iti Akimoto*, edited by M. H. Manghnani, and Y. Syono, 95–111. Washington D.C.: American Geophysical Union (AGU) Geophysical Monograph Series.
- Wilson, L., and Keil, K. 2012. Volcanic Activity on Differentiated Asteroids: A Review and Analysis. *Chemie der Erde-Geochemistry* 72: 289–321.
- Wölfer, E., Renggli, C. J., Burkhardt, C., and Kleine, T. 2025. Germanium Isotope Fractionation during Evaporation from Metal and Sulfide Melts: Implications for Volatile Element Loss from Planetary Materials. *Geochimica et Cosmochimica Acta* 401: 17–30.
- Wu, P., Yang, Y., Barati, M., and Mclean, A. 2015. Thermodynamic and Kinetic Aspects of Sulfur Evaporation from Fe-C Alloy Droplets. *ISIJ International* 55: 717–722.
- Yang, J., Goldstein, J. I., and Scott, E. R. D. 2007. Iron Meteorite Evidence for Early Formation and Catastrophic Disruption of Protoplanets. *Nature* 446: 888–891.
- Yang, J., Goldstein, J. I., and Scott, E. R. D. 2010. Main-Group Pallasites: Thermal History, Relationship to IIIAB Irons, and Origin. *Geochimica et Cosmochimica Acta* 74: 4471–92.
- Zhai, K., Yin, Y., and Zhai, S. 2022. New Constraints on the Abundances of Phosphorus and Sulfur in the Lunar Core: High-Pressure and High-Temperature Experimental Study of the FeSP Ternary System. *Geochimica et Cosmochimica Acta* 334: 1–13.
- Zhang, B., Chabot, N. L., and Rubin, A. E. 2022. Compositions of Carbonaceous-Type Asteroidal Cores in the Early Solar System. *Science Advances* 8: eabo5781.
- Zhang, B., Chabot, N. L., and Rubin, A. E. 2024. Compositions of Iron-Meteorite Parent Bodies Constrain the Structure of the Protoplanetary Disk. *Proceedings of the National Academy of Sciences of the United States of America* 121: e2306995121.
- Zhang, Z., Hastings, P., Von der Handt, A., and Hirschmann, M. M. 2018. Experimental Determination of Carbon Solubility in Fe-Ni-S Melts. *Geochimica et Cosmochimica Acta* 225: 66–79.
- Zhao, B., Morard, G., Boulard, E., Boccato, S., Siersch, N. C., Rivoldini, A., Guignot, N., et al. 2023. Local Structure and Density of Liquid Fe-C-S Alloys at Moon's Core Conditions. *Journal of Geophysical Research: Planets* 128: e2022JE007577.

SUPPORTING INFORMATION

Additional supporting information may be found in the online version of this article.

Figure S1. Assemblies used for high pressure/temperature experiments. (A) 0.5" Talc-pyrite-graphite assembly showing internal R-type thermocouple. Experiments used either x1 or x2 hBN (shown) 4 mm outer diameter capsules, with internal sample volumes of 3×2 mm or 0.75×2 mm, respectively. (B) 14/8 double hBN capsule multi-anvil assembly, with 1.2×0.2 mm sample volumes.

Figure S2. Carbon secondary standard calibrations used for EDS data. Data shown for C blank and C-bearing standards: with increasing C content, high purity, C-free Fe (ECRM097), carbon steel standards ECRM 084-1(C) (0.4 wt% C) and ECRM 090-1(C) (1.05 wt% C), and synthetic Fe_3C (6.67 wt% C). "Actual" C contents are quoted C contents for standards, and stoichiometric C for Fe_3C . "Measured" C contents are

background corrected C concentrations determined by full-profile EDS spectra fitting using AZTec software; that is, subtraction of C contents in the C-blank, which ranged from 0.4 to 1.1 wt%, from measured C concentrations in the standards. Three example calibrations curves (linear regressions) are shown from successive analytical sessions. Error bars are 2σ errors calculated from standard deviations on repeat analyses (typically 8–15 analyses for each standard). EDS data was obtained for selected area analysis, for area sizes comparable to those used for unknowns.

Table S1. Light element (volatile) loss from experiments, calculated as a percentage change in element content in runs products compared to target composition of starting mixes, and Fe-rich L/FeS-rich L partition coefficients for run products with immiscible liquids (by weight). "Bulk" compositions for immiscible liquids were calculated based on phase proportions estimated from SEM-BSE images. Numbers in parentheses are calculated standard deviation on last significant figure quoted.