

# Viscosity of Fe-Ni-C Liquids up to Core Pressures and Implications for Dynamics of Planetary Cores

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## Key Points:

- Experimental and computational determinations of viscosity of Fe-Ni-C liquids up to core pressures
- Viscosities of Fe-Ni-C liquids increase by a factor of ~2 at ~3-5 GPa at which a liquid structural transition occurs
- Fe-Ni-C liquid has higher viscosity than Fe and Fe-S liquid, causing difference in core dynamo and percolative core formation

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## Abstract

The viscosity of iron alloy liquids is the key for the core dynamo and core-mantle differentiation of terrestrial bodies. Here we measured the viscosity of Fe-Ni-C liquids up to 7 GPa using the floating sphere viscometry method and up to 330 GPa using first principle calculations. We found a viscosity increase at ~3-5 GPa, coincident with a structural transition in the liquids. After the transition, the viscosity reaches ~14-27 mPa·s, a factor of 2-4 higher than that of Fe and Fe-S liquids. Our computational results from 5 to 330 GPa also indicate a high viscosity of the Fe-Ni-C liquids. For a carbon-rich core in large terrestrial body, the level of turbulence in the outer core would be lessened as approaching the inner core boundary. It is also anticipated that Fe-Ni-C liquids would percolate in Earth's deep silicate mantle at a much slower speed than Fe and Fe-S liquids.

## Plain Language Summary

Liquid cores of Earth and other terrestrial planets are composed of Fe-Ni metal with certain amounts of light elements (LE) such as H, C, O, Si, and S. Located at the center of the planets, these cores are under extremely high-pressure high-temperature conditions. The viscosity of Fe-Ni-LE liquids is the key to understanding the formation and life duration of core dynamo. In the present study, we used experimental and theoretical methods to study the viscosity of Fe-Ni-C liquid, a candidate liquid core component, at high pressures and temperatures. Our results show that the viscosity of Fe-Ni-C liquids first increase quickly at ~3-5 gigapascal (GPa), and then increases slowly up to Earth's core conditions at 330 GPa and 5530 K. The fast growth of viscosity concurs with a liquid structure transition at around 5 GPa, around the core pressure ranges of the Moon, Mercury and several Jupiter's satellites, and might result in difference in viscous forces in core dynamos between those cores with pressures above and below transition pressure. The viscosity of Fe-Ni-C liquids at Earth's core conditions is higher than Fe and Fe-S liquids from previous studies, which provides new clues for the operation of geodynamo in a carbon-rich outer core.

## 1 Introduction

The liquid cores of Earth and other terrestrial planets or moons are composed of Fe-Ni metal, alloyed with a range of potential light elements (LE). The dynamics of these liquid cores determines the formation and the duration of the planetary dynamo and magnetic field, which are crucial for planetary habitability. Knowledge on the viscosity of candidate Fe-Ni-LE alloys at high pressures and high temperatures (HP-HT) are required to understand and model the dynamics of these liquid cores. Additionally, viscosity is one of the key properties that determines the percolation velocity of iron alloy through silicate/oxide rocks, which is crucial for studying the time scale of core formation in Earth and other terrestrial planets. Carbon is among the top candidate light elements in Earth's and planetary cores due to its high cosmic abundance, siderophile nature and ubiquity in iron meteorites. The phase diagram and physical properties of Fe-(Ni)-C solids and liquids at HP-HT have been widely investigated [Chen *et al.*, 2014; Lai *et al.*, 2017; Nakajima *et al.*, 2009; Nakajima *et al.*, 2015; Prescher *et al.*, 2015; Terasaki *et al.*, 2010; Wood, 1993], but studies on the viscosity of Fe-(Ni)-C liquid are limited due to the challenges of performing *in-situ* radiography observation and viscometry measurements on liquids at HP-HT conditions.

The development of *in-situ* X-ray radiography techniques enables the real-time monitoring of falling/floating sphere experiments at HP-HT conditions, which can be employed to study the viscosity of Fe-Ni-LE alloys melts (Table S1). Previous X-ray radiography studies have successfully measured the viscosity of Fe and Fe-S liquids [Dobson *et al.*, 2000; Kono *et al.*, 2015; LeBlanc & Secco, 1996; Perrillat *et al.*, 2010; Rutter *et al.*, 2002a; Rutter *et al.*, 2002b; Secco *et al.*, 2002; Terasaki *et al.*, 2006; Terasaki *et al.*, 2002; Terasaki *et al.*, 2001; Urakawa *et al.*, 2001], but the viscosity of other LE-bearing iron alloys at high pressure, including those with C, have only been sparsely studied to date. At ambient pressure, the viscosity of Fe-C was measured to be around 4-10 mPa·s at ~1600 K [Lucas, 1964], and the HP-HT viscosities of Fe-C liquids were only reported up to pressures of 4.5 GPa [Terasaki *et al.*, 2006]. The viscosity of Fe<sub>84</sub>C<sub>16</sub> scattered around 2-5 mPa·s at 3-4.5 GPa and 1843 K, which is similar to those of Fe and Fe-S liquids in this pressure/temperature range [Terasaki *et al.*, 2006]. However, only three data points were reported on this Fe-C alloy, and as such there are insufficient constraints for fitting or for extended extrapolations to higher pressures. In addition, recent liquid structure studies found a polyamorphic liquid structural transition in Fe-(Ni)-C alloy melts at ~5 GPa [Lai *et al.*, 2017; Shibazaki *et al.*, 2015]. With a similar liquid structure transition in liquid Fe around its  $\delta$ - $\gamma$ -liquid triple point [Sanloup *et al.*, 2000], Terasaki *et al.* [2002] reported a strong decrease in viscosity through the transition pressure around 5 GPa, while Kono *et al.* [2015] reported no discontinuous change. It remains unclear whether such liquid structural transition can have a significant influence on their viscosity. In this study, we measured the viscosity of two Fe-Ni-C alloys (Fe<sub>90</sub>Ni<sub>10</sub>-3wt.% C and Fe<sub>90</sub>Ni<sub>10</sub>-5wt.% C) up to 7 GPa. In addition, we carried out first principles molecular dynamics (FPMD) calculations for the viscosity of Fe-Ni-C alloys with 1wt.%, 3wt.%, and 5wt.% C up to the Earth's core conditions, to investigate the influence on the viscosity of iron alloy from the incorporation of carbon and from the liquid structural transition.

## 2 Materials and Methods

### 2.1 Paris-Edinburgh Press Experiments

The starting materials were mixtures of Fe (99.9+%, Aldrich), Ni (99.99%, Aldrich) and graphite (99.9995%, Alfa Aesar) powders. The mixtures were ground in acetone using an agate mortar for more than 1 h to achieve compositional homogeneity and then dried in a vacuum oven at high temperature of ~383 K overnight before being sealed in glass vials.

The viscosity measurement was conducted in a Paris-Edinburgh Press at 16-BM-B, Advanced Photon Source (APS), Argonne National Lab (ANL). A packed cylindrical sample of 1 mm diameter and 2 mm height was placed in a BN capsule, and a ruby sphere was placed near the bottom of the sample (Figure S1). The sample was first compressed to the target pressure and heated at 1073 K for 30-120 min to sinter the sample and capsule. The sample was then rapidly heated to 1973 K by applying a corresponding heating power, which is at least ~300-400 K above the melting temperature of Fe<sub>90</sub>Ni<sub>10</sub>-3wt.% C and Fe<sub>90</sub>Ni<sub>10</sub>-5wt.% C alloys at a given pressure for all the investigated pressure points, to ensure the sample was fully molten and the ruby can float freely [Nakajima *et al.*, 2009]. The movement of the ruby sphere upon melting was recorded by a high-speed X-ray camera at a frame rate of 500 frame/s for most experiments, except for two points at lower frame rate (Figure S2 and Figure S3). The viscosity of the molten sample was calculated by the Stokes' equation with correction factors for the effect of the wall (W) and the end (E) of a cylindrical sample container:

$$\eta = \frac{g d_s^2 (\rho_s - \rho_l) F}{18 \nu E} \quad (1)$$

$$F = 1 - 2.104 \left(\frac{d_s}{d_l}\right) + 2.09 \left(\frac{d_s}{d_l}\right)^3 - 0.95 \left(\frac{d_s}{d_l}\right)^5 \quad (2)$$

$$E = 1 + \frac{9}{8} \frac{d_s}{2Z} + \left(\frac{9}{8} \frac{d_s}{2Z}\right)^2 \quad (3)$$

where  $d$  and  $\rho$  are the diameter and density, and subscripts  $s$  and  $l$  stand for the solid and liquid.  $Z$  is the height of cylindrical sample and  $\nu$  is the terminal velocity of falling/floating sphere. The diameter of the ruby sphere and the dimension of the sample was determined by the X-ray image with 2  $\mu\text{m}$  resolution. The densities of ruby sphere [Fei *et al.*, 1995] and Fe-Ni-C liquid [Zhu *et al.*, 2021] are determined by their equation of states at corresponding pressure and temperature. The density error of the Fe-Ni-C liquid can be significant [Zhu *et al.*, 2021], but the density contrast between Fe-Ni-C and ruby of  $\sim 3 \text{ g/cm}^3$  is large enough to minimize the error introduced by the errors in density of each phase. The large density contrast results in a large terminal velocity, where the high-speed camera is essential to precisely determine the terminal velocity. The pressure was determined by measuring the unit-cell volume of MgO ring by energy dispersive X-ray diffraction after the floating sphere experiment before the temperature is quenched and then adopted a calibrated pressure correction between sample and MgO ring [Kono *et al.*, 2014]. The temperature was estimated from the power curve in calibration experiments with an estimated standard deviation of 50 K. The details of the experimental setup and viscometry method can be found in Kono *et al.* [2014].

## 2.2 First-principle Molecular Dynamics (FPMD) simulations

Details of the FPMD simulations were reported in previous publications and summarized here for completeness [Lai *et al.*, 2017; Wang *et al.*, 2019; Zhu *et al.*, 2021]. Density Functional Theory (DFT) with plane wave basis sets as implemented in Vienna Ab initio Simulation Package (VASP) [Kresse & Furthmüller, 1996]. The Projector-Augmented Wave (PAW) method and exchange-correlation with the Perdew–Burke–Ernzerhof (PBE) functional were utilized in the Generalized Gradient Approximation (GGA) [Blöchl, 1994; Perdew *et al.*, 1992]. The kinetic energy for the plane-wave basis was cut-off at 520.00 eV for equilibration simulation and 400.00 eV for equilibrium simulation. The Brillouin zone was sampled at  $\Gamma$  point. The FPMD simulations were carried out with the NVT ensemble and Nosé-thermostat for up to 3 ps for the system to relax in the equilibration run. After equilibration run, each of the FPMD simulations was run  $\sim 17$  ps (17,000 steps) for statistical analysis of the viscosity. The time step was set to be 1 fs. The details of viscosity calculation was summarized previously [Wang *et al.*, 2019]. The calculated pressures were benchmarked based on the experimental data on iron at similar conditions [Anderson & Ahrens, 1994]. The FPMD simulations were performed using a cubic computation supercell of 192 atoms. The ratio between Fe and Ni remains approximately 9:1 with C at 1%, 3%, and 5%, which is corresponding to 165 Fe + 18 Ni + 9 C, 151 Fe + 17 Ni + 24 C, and 139 Fe + 15 Ni + 38 C atoms respectively. A system of pure iron with 192 atoms was also simulated using the same method. The computational supercells were gradually adjusted to different volumes.

### 3 Results and Discussion

#### 3.1 Floating sphere experiments on the viscosity of Fe-Ni-C liquid

Figure 1a and Movie S1 show examples of the movement of the ruby sphere in Fe-Ni-C melts, while Figure 1b & 1c convert these results into velocity as a function of time. The ruby sphere sped up quickly after sample melting to reach the terminal velocity, and then kept the terminal velocity for a distance before it decelerated and approached the top of the sample capsule. Ideally, with only the drag force from liquid, the gravity, and the buoyancy, the sphere should either accelerate to or move uniformly at terminal velocity. The long deceleration distance in Figure 1b before reaching the sample end implies additional force on the sphere due to the wall and the end. Therefore equations (1-3) with correction factors for those effect from the wall and the end were used to calculate the viscosities. The terminal velocity was determined by fitting the slope of distance-time curve in the range where the single-step velocities inside reached a nearly constant highest value before deceleration of the probe sphere as demonstrated by Kono *et al.* [2015], e.g. between 718 and 750 ms in Figure 1b & 1c. This terminal velocity, together with the dimensions of sample and ruby sphere, was then used to calculate the viscosity by equations (1-3), which is shown in Figure 1c.

Figure 2a and Table S2 summarizes the viscosity of the two Fe-Ni-C compositions in the experimentally investigated pressure range. The viscosity of Fe<sub>90</sub>Ni<sub>10</sub>-3wt.% C was in the range of about 8-12 mPa·s up to 4.2 GPa, which is slightly higher but still consistent with the estimates of previous studies at ~4-6 mPa·s within the uncertainties [Terasaki *et al.*, 2006]. However, our results show marked increase of viscosity of Fe<sub>90</sub>Ni<sub>10</sub>-3wt.% C by a factor of ~2, reaching ~14-18 mPa·s at >4.8 GPa. Similar viscosity increase is also observed in the Fe<sub>90</sub>Ni<sub>10</sub>-5wt.% C liquid from 3.0 to 5.0 GPa. This substantial increase in viscosity at ~3-5 GPa coincides with the polyamorphous liquid structural transition in Fe-Ni-C liquids around 5 GPa [Lai *et al.*, 2017], suggesting that it is likely related to the transition. The increased fraction of the 3-atom shared polyhedral motifs was inferred as the main mechanism behind this transition. Unlike solid phase transition, this growth is a gradual change in a range of pressure from FPMD calculation [Wang *et al.*, 2019], indicating that the viscosity increase may also be gradual. It is difficult to precisely determine the initial transition pressure from our data: For Fe<sub>90</sub>Ni<sub>10</sub>-3wt.% C, it is possible that the viscosity increases steeply from 4.2 to 4.8 GPa; But if excluding the viscosity at 4.2 GPa, it is more likely that the viscosity undergoes an gradual growth from 2.7 to 4.8 GPa. Considering the nature of liquid structural transition, here we conclude that the major viscosity change occurs between ~3 to 5 GPa.

A previous study has reported an abrupt drop in viscosity of liquid Fe around 5 GPa near the  $\delta$ - $\gamma$ -liquid triple point in Fe phase diagram [Terasaki *et al.*, 2002]. The viscosity drop was considered to be associated with the liquid phase transition. However, a later study using a higher speed camera reported no obvious discontinuous change in the viscosity of liquid Fe [Kono *et al.*, 2015]. Thus, whether liquid structure transitions can induce substantial viscosity changes in iron alloys remains unclear. In this study, the viscosity increases by about 2 times through the polyamorphous transition around ~3-5 GPa [Lai *et al.*, 2017], and remains more or less constant after the transition, showing a different trend from the previous observations in liquid Fe [Kono *et al.*, 2015; Terasaki *et al.*, 2002]. FPMD calculation also shows the viscosity of Fe<sub>90</sub>Ni<sub>10</sub>-5wt.% C substantially increases through the transition around 5 GPa before it flattens out at >8 GPa [Wang *et al.*, 2019]. This trend agrees with the positive correlation we observed in



the experiments, although the absolute value of the computational viscosity was slightly lower than those observed in our experiments.

### 3.2 Viscosity of Fe-Ni-(C) liquids to Earth's core conditions by FPMD calculations

The experimental results show a plateau of viscosity for both Fe-Ni-C liquids after the transition at  $> \sim 5$  GPa, but the pressure range is limited. It is still unknown whether this trend will persist to higher pressures only based on experimental results at low pressures. The previous FPMD calculations calculated the viscosity of Fe-Ni-C liquids to 67 GPa, and found the viscosities are very similar between 8 and 48 GPa along an isothermal compression path [Wang *et al.*, 2019]. However, the viscosity showed another discontinuity at 67 GPa, which might result from the 1673 K temperature too low to keep Fe<sub>90</sub>Ni<sub>10</sub>-5wt.% C fully molten. Without considering both the core temperature and pressure, it is difficult to extrapolate these results at low temperatures and pressures to Earth's core conditions.

In this study, we directly calculated the viscosity of Fe, Fe<sub>90</sub>Ni<sub>10</sub>-1wt.% C, Fe<sub>90</sub>Ni<sub>10</sub>-3wt.% C, and Fe<sub>90</sub>Ni<sub>10</sub>-5wt.% C at the core mantle boundary (CMB) and inner core boundary (ICB) conditions, using the method as described in Wang *et al.* [2019] (Figure S4). The viscosity of Fe is consistent with the previous calculation of Fe and Fe<sub>90</sub>Ni<sub>10</sub> [Cao & Wang, 2017; de Wijs *et al.*, 1998]. Our results show that the viscosity of Fe<sub>90</sub>Ni<sub>10</sub>-3wt.% C and Fe<sub>90</sub>Ni<sub>10</sub>-5wt.% C are similar at both calculated conditions, and they are  $\sim 2$  mPa·s (30%) and  $\sim 7$  mPa·s (55%) higher than the viscosity of Fe at CMB and ICB conditions (Figure 3). But the increase is not linearly correlated with the amount of carbon incorporated. The viscosity of Fe<sub>90</sub>Ni<sub>10</sub>-1wt.% C is slightly lower than Fe at CMB condition, but it becomes higher at ICB conditions. The viscosity of Fe<sub>52</sub>S<sub>12</sub> from a previous FPMD calculation is lowest at ICB conditions among all Fe alloys [Alfè & Gillan, 1998]. Combined with the results from Wang *et al.* [2019], the viscosity of Fe-Ni-C liquids increases with pressure slightly faster than that of Fe-(Ni) liquid, but the increase rate is much lower compared with the substantial increase by a factor of 2 from 4.1 to 8.4 GPa. The viscosities at 8-48 GPa and 1673 K are only about 4-5 mPa·s lower than those at CMB conditions (135 GPa, 4050 K), and about 11-12 mPa·s lower than those at ICB conditions (330 GPa, 5530 K). The results suggest that either the pressure does not have a significant impact on the viscosity at  $> 5$  GPa after the polyamorphous transition, or its effect on elevating viscosity is largely mitigated by the elevated temperature towards the ICB, which lowers the viscosity.

Compared with the experimental results at  $< 10$  GPa, our calculation seems to systematically underestimate the viscosity by approximately a factor of 1.5-4.5. Due to the challenges in *ab initio* calculations, it is not uncommon that there exist discrepancies between computational and experimental results, especially for the liquid properties such as viscosity. However, our FPMD results are consistent with the experiments in terms of the pronounced viscosity increase around 5 GPa and the viscosity plateau after the transition (Figure 3). If the factor of 3 difference is uniformly applied to core pressures, the viscosities of Fe-Ni-C at Earth's core conditions are roughly estimated at  $\sim 33$  mPa·s at CMB and  $\sim 58$  mPa·s at ICB. Nevertheless, the incorporation of 1-5 wt.% C in liquid Fe could significantly enhance the rate at which the viscosity increases with pressures and Fe-Ni-C liquids generally have higher viscosity than Fe, FeNi, and Fe-S liquids.

### 3.3 Implications for the dynamics of planetary cores

The pressure of 4-5 GPa at which a substantial increase in the viscosities of the Fe-Ni-C liquids lies at a pressure close to core pressures within a number of small planets and moons, such as Moon, Mercury, Ganymede, Io, and Europa [Garcia *et al.*, 2012; Hauck *et al.*, 2013; Kronrod & Kuskov, 2006] (Figure 2b). The liquid cores of the Moon, Mercury, Ganymede and Io are estimated to be under pressures close to or above this transition pressure, while Europa has a central pressure of ~4 GPa. If carbon is one of the major light elements in the cores of some of these terrestrial bodies, their core viscosity may be distinctly different due to the liquid structural transition. These differences will result in distinct viscous forces within their cores and thus dynamos. The variation of viscosity due to the effect of pressure and/or liquid structural transition in Fe-C liquids as revealed in this study or some other liquid alloys such as Fe and Fe-S liquids may render the extrapolations to higher pressures unreliable. Therefore, the viscosity of these planetary liquid cores may be significantly different from those inferred based on low pressure (0-5 GPa) measurements, depending on the core pressure and the light element species.

The viscosity of Fe-Ni-C after the transition at ~4.5 GPa becomes higher than Fe-Ni in both experiments and calculations, and this relationship persists over the pressure range of Earth's core, as shown from the FPMD calculations. This indicates that a C-rich liquid core in planets or moons with core pressures higher than 4.5 GPa would be more viscous in conjunction with generating core dynamos than Fe-Ni cores. However, the magnitude of viscosity is still in the range of tens of mPa·s, which remains small. In particular, Coriolis and inertial forces will continue to dominate within the core, as illustrated by the low Ekman number of the outer core ( $O \sim 10^{-15}$ : [Christensen & Aubert, 2006]). Nevertheless, the transition to a higher viscosity near 4.5 GPa, and the positive increase in viscosity across the depth range of the outer core for carbon-bearing alloys implies that Earth's outer core may lie modestly closer to the Ekman number range of current core geodynamo simulations. These simulations typically have, because of computational limitations, artificially high viscosities — often ~9 orders of magnitude larger than the core itself [e.g. King & Buffett, 2013; Schwaiger *et al.*, 2019]. Our results suggest that the Earth's core might lie modestly closer to the domain in which current geodynamic simulations are conducted.

### 3.4 Implication for the percolative core formation and growth

The percolation of Fe-Ni-LE liquids through a solid silicate matrix is a possible mechanism for the formation of cores of terrestrial planets and moons in their early history. The segregation velocity,  $V_m$ , for a connected melt can be expressed as

$$V_m = \frac{k\Delta\rho g}{\phi\mu} \quad (4)$$

in which  $k$  is permeability,  $\Delta\rho$  is the density difference between solid and liquid,  $g$  is the acceleration of gravity,  $\phi$  is the melt fraction, and  $\mu$  is the viscosity of melt [Faul, 1997; McKenzie, 1989]. The viscosity has an inverse relation to the segregation velocity, which is vital in evaluating the time scale over which core-forming melts drained through early Earth's or protoplanetary silicate mantle. Considering the observed viscosity increase of Fe-Ni-C liquids at ~3-5 GPa, the segregation velocity could drop by several times at the corresponding depth. At higher pressures, the viscosity increases slowly and remains consistently higher than pure Fe-Ni, which means the segregation velocity of Fe-Ni-C will be consistently slower than Fe-Ni. A segregation velocity drop will delay the rate at which C-bearing Fe-Ni alloy enters the core

through percolation. Based on previous viscosity estimates ( $\sim 3\text{-}6\text{ mPa}\cdot\text{s}$ ), assuming the melt fraction  $\phi = \sim 0.1$ ,  $\Delta\rho = 3500\text{ kg/m}^3$  and  $k = \sim 10^{-15}\text{ m}^2$  and the viscosity remains constant in the entire mantle, the  $V_m$  is roughly estimated  $\sim 2\text{ m/yr}$  and melt would need 1.5 Myr to traverse through a 3000 km radius mantle, which is significantly faster than the 30 Myr core formation process estimated from mantle Hf-W isotopic ratios [Kleine *et al.*, 2002]. However, if the viscosity is increased to  $\sim 20\text{-}30\text{ mPa}\cdot\text{s}$ , the time to drain the melt increases to  $\sim 10\text{ Myr}$ . On the other hand, whether the enhanced viscosity of C-bearing alloys might also affect their wetting behavior is unknown, but scaling relations have been proposed between viscosity and surface energies [e.g. Egry, 1993]. Hence, it is anticipated that the viscosity change may influence the percolation velocity of the C-rich liquids in a silicate matrix.

#### 4 Conclusions

We have carried out viscosity measurements of Fe-Ni-C liquids up to 7 GPa and calculations up to 330 GPa. A pronounced viscosity increase was observed at  $\sim 3\text{-}5\text{ GPa}$ , concurring with a liquid structural transition in the Fe-Ni-C liquids. The transition pressure locates around the core pressures of many small planets and moons such as the Moon, Mercury and satellites of Jupiter. If their cores contain significant amount of carbon, the viscosity variation may result differences of the viscous forces within their cores at pressures above or below 4.5 GPa. Considering similar liquid structural transitions exist in other Fe alloys, whether the viscosity discontinuity through the structural transition is ubiquitous needs further investigation.

The viscosity after the transition is higher than the viscosity of Fe and Fe-S liquids from both experiments and calculations. The calculated viscosity at pressures greater than 5 GPa increases progressively and slowly up to inner core boundary conditions at 330 GPa and 5530 K. The high viscosity of Fe-Ni-C liquids at  $> 5\text{ GPa}$  implies that a C-rich core in any large terrestrial body would have both a larger Ekman number and smaller Reynolds number than generally inferred by about half an order of magnitude. As such, the level of turbulence in the outer core would be lessened as approaching the inner core boundary, and the higher viscosity of a C-rich core of the Earth would imply that current artificially high-viscosity geodynamo simulations are modestly closer to the potential conditions within Earth's core. Fe-Ni-C liquids would also be expected to percolate in Earth's deep silicate mantle at a much slower speed compared with Fe and Fe-S liquids. This effect will expand the timeframe over which late-descending C-rich iron alloys will reach the core, and will allow for more time for isotopic fractionation during the core-mantle differentiation.

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## Figure captions

**Figure 1.** A representative example of the movement of the ruby sphere upon sample melting. The sample was  $\text{Fe}_{90}\text{Ni}_{10}$ -3 wt.%C and the experimental conditions were 1.8 GPa and 1973 K. (a) The movement of the ruby sphere at a time interval of 75 ms between each frame, from 475 ms to 925 ms. The diameter of the  $\text{Al}_2\text{O}_3$  sphere is  $\sim 250$   $\mu\text{m}$ . (b) The distance-time relation was converted to a velocity-time relation to determine the terminal velocity. (c) The terminal velocity was used to calculate the liquid viscosity.

**Figure 2.** Viscosities of Fe and Fe alloys (a) Viscosities of  $\text{Fe}_{90}\text{Ni}_{10}$ -3 wt.%C and  $\text{Fe}_{90}\text{Ni}_{10}$ -5 wt.%C in comparison with previous results. The viscosities of both components increases from  $\sim 5$ -8  $\text{mPa}\cdot\text{s}$  at  $<3$  GPa to  $\sim 15$ -27  $\text{mPa}\cdot\text{s}$  at  $\sim 5$ -7 GPa, which may result from the polyamorphic transition around 5 GPa. Open red and blue symbols represent an upper limit of viscosities at those pressures, due to a possibility of not reaching terminal velocity. (b) The polyamorphic transition pressure of Fe (light gray bar) and FeNi-C (light red bar) liquids in comparison with the core pressure ranges of small planet and moons (black lines, arrow means central pressure higher than 7 GPa).

**Figure 3.** Viscosity of Fe-Ni-C liquids to Earth's core pressure from FPMD calculations. The viscosity increases quickly from 3.3 to 6.0  $\text{mPa}\cdot\text{s}$  at 4.1-8.4 GPa, corresponding to a polyamorphic transition in the liquid (light blue arrow), and then flattens at 8.4-48 GPa (light red arrow) [Wang *et al.*, 2019]. The viscosities from CMB (4050 K) to ICB (5530 K) conditions increase slightly for all Fe-Ni-C species (light red band) and remain similar for Fe liquid. The gray band shows the calculated viscosity of  $\text{Fe}_{90}\text{Ni}_{10}$  at 50-300 GPa from melting point ( $T_m$ , 2200 K at 50 GPa and 6500 K at 300 GPa) to 1.85 times of  $T_m$  [Cao & Wang, 2017]. Previous results of Fe [Alfè & Gillan, 1998; de Wijs *et al.*, 1998] and Fe-S [Alfè and Gillan, 1998] are also plotted for comparison. Some uncertainties for our calculated viscosities are within the symbols and can be found in Table S3.







