Compositional instability of Earth's solid inner core

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[1] All models that invoke convection to explain the observed seismic variations in Earth's inner core require unstable inner core stratification. Previous work has assumed that chemical effects are stabilizing and focused on thermal convection, but recent calculations indicate that the thermal conductivity at core temperatures and pressures is so large that the inner core must cool entirely by conduction. We examine partitioning of oxygen, sulfur, and silicon in binary iron alloys and show that inner core growth results in a variable light element concentration with time: oxygen concentration decreases, sulfur concentration decreases initially and increases later, and silicon produces a negligible effect to within the model errors. The result is a net destabilizing concentration gradient. Convective stability is measured by a Rayleigh number, which exceeds the critical value for reasonable estimates of the viscosity and diffusivity. Our results suggest that inner core convection models, including the recently proposed translational mode, can be viable candidates for explaining seismic results if the driving force is compositional. **Citation:** Gubbins, D., D. Alfè, and C. J. Davies (2013), Compositional instability of Earth's solid inner core, *Geophys. Res. Lett.*, *40*, 1084–1088, doi:10.1002/grl.50186.

1. Introduction

[2] Thermal history calculations show that the Earth's solid inner core has been growing as the liquid outer core freezes from the bottom up [see *Nimmo* 2007 for a review]. The density difference between solid and liquid cores is too large to be explained solely by freezing, and the liquid is supposed to contain more light elements than the solid. Cosmochemical arguments favor oxygen, sulfur, and silicon as candidate light elements, and ab initio calculations of binary mixtures (Fe-O, Fe-S, Fe-Si) show that O partitions almost completely into the liquid on freezing, S partitions almost equally with a slightly higher proportion in the liquid, and Si partitions equally between the two phases to the accuracy of the calculations [*Alfè et al.*, 2002]. This partitioning is crucial for powering the geodynamo in the outer core that gives rise to Earth's magnetic field; in this paper we show that it may also be crucial for the dynamics of the inner core.

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[3] Seismic studies have shown the inner core to be both heterogeneous and anisotropic, with a pronounced east-west hemispheric structure [*Souriau*, 2007; *Irving and Deuss*, 2011]. There is also evidence of a different structure in the innermost 300 km of the inner core [*Ishii and Dziewoński*, 2002] and a ~100 km thick isotropic layer below the inner core boundary (ICB) [*Waszek and Deuss*, 2011]. To explain the observations, much work has focused on inner core convection, driven either thermally [*Jeanloz and Wenk*, 1988; *Buffett*, 2009] or in combination with compositional gradients [*Deguen and Cardin*, 2011; *Cottaar and Buffett*, 2012]. The hemispheric structure has been proposed to arise from a translational convective mode involving a uniform drift of the inner core material [*Monnereau et al.*, 2010; *Alboussière et al.*, 2010], while termination of convection at an early stage has been suggested as the cause of the innermost inner core and the outermost isotropic layer [*Deguen and Cardin*, 2009]. While the results are encouraging, the proposition of inner core convection rests on one highly uncertain hypothesis: the inner core is assumed to be unstably stratified.

[4] The aforementioned models all assume a thermal origin for the unstable inner core stratification, implying that the inner core temperature gradient exceeds the adiabatic gradient at the relevant pressure-temperature conditions. Previous studies have found that this may be the case at present and was more likely in the past [*Buffett*, 2009; *Deguen and Cardin*, 2009, 2011], but the results depend critically on the thermal conductivity of inner core material. Recent work found the thermal conductivity at the base of the outer core to be over three times larger than previous estimates [*Pozzo et al.*, 2012] and this value must be further increased when applied to the solid to account for the lower concentration of light elements in the inner core. Such high values for the thermal conductivity make it highly unlikely that thermal convection can arise in the inner core [*Buffett*, 2012].

[5] When compositional effects have been incorporated into inner core convection models, they have been treated as neutral or stabilizing [*Deguen and Cardin*, 2011; *Cottaar and Buffett*, 2012]. This reasoning is based on the assumption that the partition coefficients (the solid-liquid concentration ratio) do not change with time. The concentration of light elements in the liquid increases as the outer core shrinks [*Stacey*, 1995]; a constant partition coefficient therefore implies that the concentration in the solid rises with that in the liquid causing a stabilizing density gradient. If this were the case, then inner core convection would not be viable because both thermal and compositional contributions to the inner core density gradient would be stabilizing.

[6] In this paper, we show that the partition coefficients for O, S, and Si are not constant but actually decrease with time. This important result is shown to arise as a direct consequence of the chemical potentials in the liquid being lower than in the solid and the drop in temperature at the ICB as

the inner core grows due to the decrease in melting temperature with falling pressure. The decrease in partition coefficients with time has the opposite effect to the rising concentration in the liquid. A destabilizing gradient results if the partitioning effect is the strongest. The theory and equations are given in section 2; they are solved with appropriate numerical values in section 3 in the form of a Rayleigh number as a function of inner core radius. Conclusions and discussion are presented in section 4.

2. Composition and Density Gradients

[7] The partition coefficient for a light element between solid and liquid is

$$P_{sl} = \bar{c}_s / \bar{c}_l, \tag{1}$$

where $\bar{c}$ is molar concentration (overbar denotes a molar, rather than a mass, concentration) and subscripts s and l denote solid and liquid, respectively. *Alfè et al.* [2002] express the chemical potential in the form

$$\bar{\mu} = \mu_0 + \lambda\bar{c} + kT\log\bar{c} = \tilde{\mu} + kT\log\bar{c}, \tag{2}$$

where k is Boltzmann's constant and λ is a linear correction to ideal solution theory obtained from ab initio calculations. The partition coefficient is calculated by assuming chemical equilibrium at the ICB and equating the chemical potentials on either side of the boundary:

$$P_{sl}(r) = \frac{\bar{c}_s(r)}{\bar{c}_l(r)} = \exp\left[\frac{\tilde{\mu}_l - \tilde{\mu}_s}{kT_m(r)}\right], \tag{3}$$

where r is the inner core radius and $T_m(r)$ the melting temperature. As time progresses, the inner core grows, r increases, and $T_m(r)$ decreases because of the lower pressure at larger radius. Thus, if $\tilde{\mu}_l - \tilde{\mu}_s < 0$, the partition coefficient will decrease with time, less of the light element will be incorporated into the inner core, and the density will increase with r.

[8] The melting temperature in earlier times is calculated by assuming a constant gradient with pressure P:

$$T_m(r) = T_m(r_i) - \int_r^{r_i} \frac{dT_m}{dP}\frac{dP}{dr}dr, \tag{4}$$

where r_i is the present-day inner core radius. The inner core is very close to hydrostatic pressure and uniform density. Acceleration due to gravity is therefore nearly proportional to r: $g = \gamma r$, where γ is the gravity gradient, and

$$\frac{dT_m}{dr} = \frac{dT_m}{dP}\frac{dP}{dr} = -\frac{dT_m}{dP}\rho_a\gamma r, \tag{5}$$

where ρ_a is the average inner core density. Equation (4) becomes

$$T_m(r) = T_m(r_i) + \frac{1}{2}\frac{dT_m}{dP}\rho_a\gamma\left(r_i^2 - r^2\right). \tag{6}$$

This expression for $T_m(r)$ will be used in equation (3) to determine the partition coefficient as a function of radius.

[9] *Alfè et al.* [2002] found oxygen to partition mainly into the liquid with very low concentrations in the solid, while substantial amounts of sulfur and silicon remained in the solid. The oxygen concentration therefore rises in inverse proportion to the outer core's volume:

$$\bar{c}_l(r) = \left(\frac{r_o^3 - r_i^3}{r_o^3 - r^3}\right)\bar{c}_l(r_i). \tag{7}$$

[10] *Alfè et al.* [2002] found virtually no difference in composition of silicon between solid and liquid, so there is no change with time and we do not consider it further. For sulfur the concentrations are $\bar{c}_l = 0.10$ and $\bar{c}_s = 0.08$ for model PREM [*Dziewonski and Anderson*, 1981], which is a big enough difference to be worth considering here. Estimating the change in $\bar{c}_l$ in this case is difficult and requires a few assumptions. We neglect the very small changes in $\bar{c}_s$ and assume a constant density inner core. The total number of atoms does not change and we can equate their number at a past time, when the inner core radius was r, with the present to give

$$\bar{c}_l(r) = \left(\frac{r_o^3 - r_i^3}{r_o^3 - r^3}\right)\bar{c}_l(r_i) + \left(\frac{r_i^3 - r^3}{r_o^3 - r^3}\right)\bar{c}_s. \tag{8}$$

Note that when $\bar{c}_s = 0$ equation (8) reduces to (7), and when $\bar{c}_l(r_i) = \bar{c}_s$ then $\bar{c}_l(r) = \bar{c}_l(r_i)$.

[11] Since $\tilde{\mu}_l - \tilde{\mu}_s < 0$ [*Alfè et al.*, 2002] and $T_m(r)$ decreases with r, equation (3) shows $P_{sl}(r)$ to decrease with r while (7) and (8) show $\bar{c}_l(r)$ to increase: the composition in the solid could either decrease or increase, i.e., the effect could be stabilizing or destabilizing, depending on which is the stronger effect.

[12] It remains to calculate the density in the inner core thus formed. We assume linear dependence on concentration,

$$\rho(r) = \rho(r_i) - \rho_a\alpha_{cs}[\bar{c}_s(r) - \bar{c}_s(r_i)], \tag{9}$$

where α_{cs} is the compositional expansion coefficient for the solid.

[13] The stability of the inner core to compositional convection is assessed by the Rayleigh number:

$$R_a = \frac{\rho'\gamma r^5}{D\eta} = \frac{\rho_a\bar{c}_s'\gamma\alpha_{cs}r^5}{D\eta}, \tag{10}$$

where the prime denotes differentiation with respect to r, D is the diffusion constant in the solid, and η the inner core viscosity. The first form applies to non-interacting mixtures of light elements with the same D, the second to a single species of atom.

[14] The Rayleigh number will be used in the next section to assess the stability of the inner core. The critical Rayleigh number for infinite Prandtl number thermal convection in a self-gravitating sphere of uniform density with a stress-free and isothermal boundary is 3091 [*Chandrasekhar* [1961], Chapter 6]. The critical Rayleigh number, Ra_c, for compositional convection with fixed concentration on the boundary is expected to be very similar and we take Ra_c to be on the order of a few thousand. We calculate Ra using (3) and (6) together with either (7) or (8) to calculate c_s from (1), which is then used in (9) and (10). In supercritical conditions ($Ra > Ra_c$) this assumes that the mixing timescale is long compared to the timescale of inner core growth. On the other hand, convection may be vigorous enough to mix the inner core to a broadly uniform concentration on a time scale that is short compared to the growth time scale. Compared to slow mixing (the former case), rapid mixing of the inner core to uniform concentration implies a higher concentration of light elements at the top of the inner core, higher

concentration gradients as new material, relatively depleted in light elements, is frozen on, and a higher value of Ra. Our goal is to demonstrate the destabilizing nature of compositional buoyancy independent of any assumptions about the convective processes and so we do not consider the case of rapid mixing further.

3. Numerical Results

[15] Numerical values for the calculations are given in Table 1. The values of $\mu_l - \mu_s$, which are needed to determine P_{sl} in (1), depend weakly on pressure, but we found that this made very little difference to the results. We use two seismic models for the density jump at the ICB, $\delta\rho$, which determines the concentrations of light elements in the outer core; they in turn determine the melting temperature in equation (6). The first, called *PREM*, uses $\delta\rho = 0.6$ g cc^{-1} [*Dziewonski and Anderson*, 1981]; the second, called *MG*, uses the higher value 0.8 g cc^{-1} [*Masters and Gubbins*, 2003]. The higher density jump requires a higher O concentration in the outer core; the concentration of the other light elements (S, Si) must be reduced in order to maintain the same total core mass. The higher concentration of O in the outer core carries with it a larger reduction in the melting point at the ICB. We calculate the effect of O and S separately and add them for a final density profile. A mix of O and Si gives the same result as for O alone because Si was found to partition equally between the solid and liquid phases.

[16] Estimating Ra requires values for the expansion coefficients, inner core viscosity, and mass diffusion coefficients. Expansion coefficients for the solid are calculated using the equations and partial volume data in *Alfè et al*. [2002] (see also the Appendix of *Deguen and Cardin* [2011]). Geodynamic constraints have been used to place bounds on η, but the range of possible values is still very broad. *Van Orman* [2004] explored the creep mechanism of iron under core conditions and obtained a low value of $\eta = 10^{11}$ Pas. *Greff-Lefftz et al.* [2000] used free-core nutation data to argue $\eta > 10^{16}$ Pas, a result supported by *Koot and Dumberry* [2011], who point out that this value is consistent with the seismologically determined Poisson's ratio for the inner core. *Buffett* [1997] gives bounds $\eta < 10^{16}$ or $\eta > 10^{20}$ for the inner core to rotate free of its gravitational coupling to the mantle. There are also significant uncertainties in estimates of the mass diffusion coefficients, but they must be smaller than the values for the liquid (10^{-8} m^2 s^{-1} for O and 5×10^{-9} m^2 s^{-1} for S) [*Alfè and Gillan*, 1998; *Alfè et al.*, 1999]. However, our calculations reveal that the instability criterion is very insensitive to the values of η and D (Figure 2); the most important factor is the net concentration gradient.
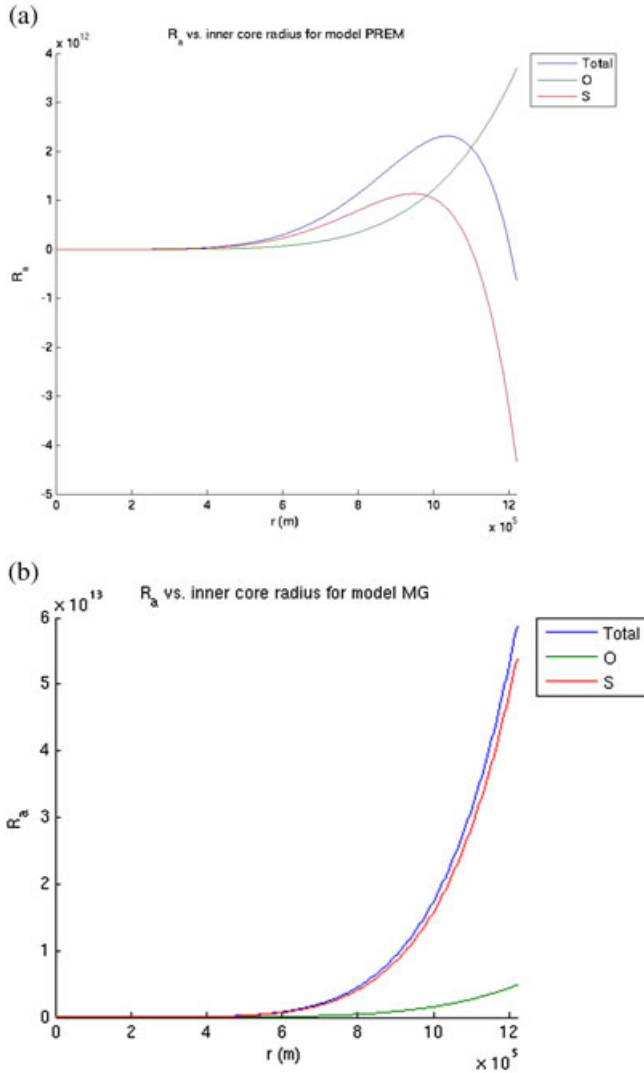
[17] We calculate compositions, densities, and Rayleigh numbers for O and S for the two seismic models. Results for O alone are appropriate for an Fe-O-Si core because Si has a partition coefficient of 1. Results for Fe-O-S are obtained by summing the separate density profiles. Each can be considered as end-members of a more complex Fe-O-S-Si mixture. For O, $\bar{\mu}_0$ is large in the solid: this dominates other terms in $\widetilde{\mu}$ (equation (2)) and we do not need to consider the correction terms λ when calculating P_{sl} in (3). Almost all the O remains in the liquid, making the calculation of the rise in $\bar{c}_l$ with time a simple matter of applying equation (7). A small but significant amount of S remains in the liquid core, 2% by mole compared with 8% O, double the proportion by mass because of sulfur's larger atomic weight. The increase in concentration with time requires the less certain equation (8) to calculate $\bar{c}_l$, and calculation of $\widetilde{\mu}$ in (2) requires values for λ and $\bar{c}_s$. Values for λ are obtained from *Alfè et al*. [2002]; values for $\bar{c}_s$ are obtained by solving equation (3) for present-day values, changes with time being completely insignificant for this purpose.

[18] Rayleigh numbers for the two models are shown in Figure 1. Values of the total Ra, calculated using the first form in equation (10), are both positive and therefore the compositional gradients are *unstable*: this is the substantive result, which follows directly from the negative argument of the exponential in (3), where $\widetilde{\mu}_l < \widetilde{\mu}_s$, which is typically the case because atoms of different size can find better arrangement in the liquid. The values of Ra are also many orders of magnitude greater than Ra_c, except for very small

Table 1. Numerical Values used in the Paper[a]

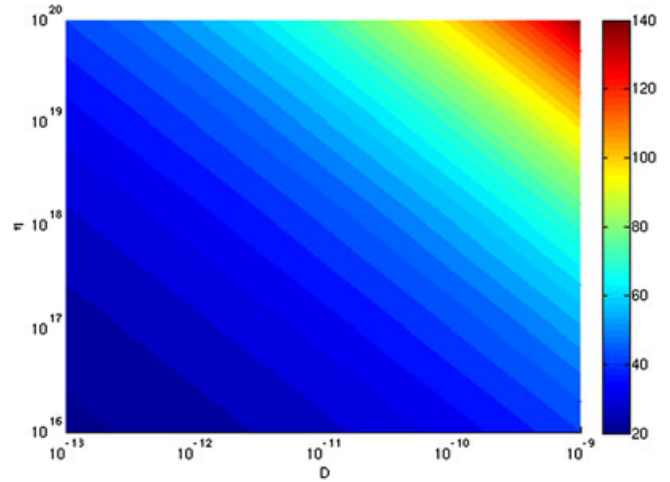| Variable	Symbol		
Boltzmann's constant	k	8.617$10^{-5}$ eV/atom	
Outer core radius	r_o	3485 km	
Present inner core radius	r_i	1221 km	
Mean IC density	ρ_a	12.9 g/cm^3	
Present ICB density	$\rho(r_i)$	12.76 g/cm^3	
Gravity gradient	γ	3.60 10^{-6} s^{-2}	
Melting gradient	dT_m/dP	9.0 K/GPa	
IC viscosity	η	$10^{16} - 10^{20}$ Pas	
Variable	**Symbol**	**O**	**S**
Difference in solute chemical potential between liquid and solid	$\mu_{0l} - \mu_{0s}$	-2.6 ± 0.2 eV/atom	0.25 ± 0.04 eV/Atom
Linear correction, solid	λ_s	-	5.9
Linear correction, liquid	λ_l	3.25	6.15
Expansion coefficients	α_{cs}	1.31	0.64
Diffusion constants	D	2×10^{-12} ms^{-2}	10^{-12} ms^{-2}
Variable	**Symbol**	**PREM**	**M and G**
Current IC temperature	$T(r_i)$	5700 (K)	5500 (K)
Concentration O	$\bar{c}_l^O(r_i)$	0.08	0.13
Concentration S	$\bar{c}_l^S(r_i)$	0.10	0.08

[a]The first group are well known and come mainly from seismology; the second group come from ab initio calculations of liquid iron mixtures; the third group depend on the density jump at the ICB. Entries in italics are very uncertain and have been estimated by methods described in the text.

Figure 1. Rayleigh numbers for (a) model PREM, 0.08% O, 0.10% S in outer core (b) model MG, 0.13% O, 0.08% S.

Figure 2. Regime diagram showing the radius in km (color scale) at which $Ra = Ra_c = 3091$, as a function of the inner core viscosity η and the mass diffusivity D. Other parameters correspond to model PREM.

4. Conclusions

[20] In this work, we have shown that the partition coefficients of O and S between the solid and liquid phases decrease with decreasing temperature (and therefore decrease as the inner core grows), leading to a lowering of the concentration of light elements with radius in the inner core. This produces an unstable density gradient that is very weak but easily strong enough to produce convection in the inner core because of the very low molecular diffusivities and viscosity: our estimated Rayleigh numbers are well above the critical value except near the center of the core. The destabilizing effect of the partition coefficient is offset by the increase in concentration of light elements in the liquid caused by the outer core's shrinking volume. Both of these effects are small, but for the most part the effect of the partition coefficient is larger, leaving an unstable density gradient. Si has no effect as it partitions equally between the solid and liquid.

[21] Our model invokes the common assumptions that the outer core is well-mixed and compositionally uniform. Some seismic observations [e.g., *Souriau and Poupinet*, 1991] suggest that P wave velocity in the bottom ~150 km of the outer core is slower than the neutrally stable model PREM [*Dziewonski and Anderson*, 1981], implying a density-stratified layer. *Gubbins et al.* [2008] proposed that this structure is caused by partial solidification in the liquid, resulting in a concentration profile in the layer that decreases exponentially from that in the main body of the outer core to the concentration in the inner core. We believe our calculations will still apply if a variable concentration layer exists at the base of the outer core because we expect the partitioning to be the same whether solidification takes place exactly at the inner core boundary or spread over a layer of fluid. In this case, the liquid concentration c_l should be taken to be the composition in the main part of the outer core.

[22] Our calculations suggest that the density gradient is stable for the first few tens of kilometers of growth in the core radius. This could relate to changes in seismic properties in the innermost inner core if our calculations have underestimated the concentration of light elements in the solid at early times; our results show that uncertainties in the inner core viscosity and mass diffusion coefficients are

r because of the factor r^5 in the definition of the Rayleigh number. For the cases shown in Figure 1, Ra_c is exceeded at $r_c = 30$ km in model PREM and $r_c = 35$ km for model MG. Figure 2 shows for model PREM the radius, r_c, where $Ra = Ra_c$ as a function of the two most uncertain parameters in the definition of Ra, D and η. We consider the ranges $10^{-13} \leq D \leq 10^{-9}$ and $10^{16} \leq \eta \leq 10^{20}$, which we believe to span the plausible range of values. Increasing η and D increases r_c up to a maximum of $r_c = 140$ km in the parameter regime considered, much smaller than the present-day inner core radius, r_i.

[19] The decrease in P_{sl} with time and inner core radius is larger than the opposite effect of increasing c_l in all cases except for S in model PREM at radii approaching the present-day value, $r = 1100$ km (Figure 1a). This corresponds to a decrease in Ra in the latter stages of inner core growth for this model, which falls below the critical value in the top ~20 km. The results suggest that the destabilizing compositional gradient can drive convection over most of the inner core history, except in the very early stages following inner core formation and possibly in the most recent times.

unlikely to delay compositional convection for long enough to account for the innermost inner core. The calculations for model PREM also predict a stable density gradient during the latter stages of inner core growth, where the acceleration of the concentration of S in the outer core overcomes the rather weak rate of decrease of the partition coefficient in the PREM model (Figure 1a). This calculation suggests stability at the top of the inner core, which may also cause a change in seismic properties.

[23] Thermal effects may be superimposed on compositional variations, but these are likely to be stabilizing based on recent estimates of the thermal conductivity [*Pozzo et al.*, 2012]. Estimating the combined effect is complicated because thermal effects must be converted to compositional effects or vice versa. The adiabatic gradient increases with radius, which may help create a stable outermost layer of the inner core; these effects may also increase the radius at which inner core convection begins to a value closer to the innermost inner core radius. Whether such a layered inner core is stable will ultimately be determined by numerical models of double-diffusive convection driven compositionally by the mechanism we have proposed. Such models are also needed to establish the dynamical regimes that are possible, although it seems that previous results for thermal convection [*Deguen and Cardin*, 2011; *Cottaar and Buffett*, 2012], including the recently proposed translational mode [*Monnereau et al.*, 2010; *Alboussière et al.*, 2010], will remain valid. Interpretations and implications for seismic structure for compositional convection will be the same: we end by emphasizing that the case for inner core convection is stronger than thought hitherto.

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