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Conditions for segregation of a crystal-rich layer within a convective magma ocean

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9 Abstract

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The vigor of the thermal convection in a terrestrial magma ocean tends to 10 prevent the sedimentation of the solid grains. Understanding of the overall dy-11 namical behavior of this solid phase segregation is required to anticipate the 12 solidification mechanisms in the early Earth mantle. We develop numerical 13 models using COMSOL Multiphysics to monitor the crystal fraction evolution 14 of a convecting magmatic reservoir. Our models show that the ability of the 15 crystal fraction to disperse within the domain strongly depends on the crystal 16 size, the density difference and the magma viscosity. Two regimes can be identi-17 fied: sedimentation or suspension regime. We show that the critical value of the 18 convection/buoyancy stress ratio separating these two regimes is smaller than 19 proposed by Solomatov et al. (1993). Hence, during the early crystallization of 20 a magma ocean, suspension should be the dominant process. 21

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We then investigate the implications of our models on the mechanism of magma ocean solidification after a major melting event on an Earth-like planet. We define the relevant set of parameters; including the P-V-T equations of state of coexisting melt and bridgmanite in the mushy MO. We observe that bridgmanite grains are unlikely to segregate in a mantle of pyrolite composition.

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However, bridgmanite segregation is more likely to occur at the bottom of a MO enriched in SiO₂, compared to pyrolite. When a solidifying layer contains 60% of bridgmanite and 40% of melt, we observe a significant SiO₂ enrichment with increasing mantle depth in a primitive mantle compatible with seismic and geochemical observations.

³³ Keywords: Early Earth, thermal evolution, magma ocean, numerical

³⁴ modeling, two-phase flow, convection

35 1. Introduction

During its early history, the Earth has experienced several episodes where 36 a significant volume fraction of its mantle was molten. These melting events 37 were enhanced by radiogenic heating (Yoshino et al., 2003), viscous dissipation 38 during core formation (Monteux et al., 2009; Samuel et al., 2010) and kinetic 39 energy converted into heat during large impacts (Tonks and Melosh, 1993). The 40 giant impact leading to the Earth/Moon system could even be responsible for 41 the complete melting of the Earth mantle (Nakajima and Stevenson, 2015) and 42 the subsequent formation of a nearly 3000 km thick magma ocean. 43

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Extremely vigorous convection is likely to occur within such a molten reser-45 voir (Solomatov, 2015; Monteux et al., 2016). The variations in temperature 46 are strong enough to generate a significant natural convection flow. Moreover 47 the potentially large thickness of a molten-silicate reservoir coupled with low 48 viscosity material (Karki and Stixrude, 2010; Xie et al., 2021) leads to Rayleigh 49 numbers (comparing advective and diffusive timescales) ranging between 10^{20} 50 to 10^{30} (*Patočka et al.*, 2020) while the current Rayleigh number of the Earth 51 mantle is ranging between 10^6 and 10^8 (*Ricard*, 2007). The cooling and solidifi-52 cation dynamics are difficult to monitor within such an environment. However, 53 recent numerical models agree on the characteristic cooling timescales that is 54

⁵⁵ constrained to several thousands of years (*Lebrun et al.*, 2013; *Monteux et al.*,
⁵⁶ 2016).

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The study of particle sedimentation in vigorously convecting fluids and, by 58 extension, the study of crystal behaviour in convecting magmas is a debated 59 subject in fluid dynamics (Lavorel and Le Bars, 2009) and in Earth Sciences 60 (Jaupart and Tait, 1995). A large number of study has been dedicated to the 61 crystal settling in magma chambers using laboratory experiments (Martin and 62 Nokes, 1988; Sturtz et al., 2021), numerical modelling (Höink et al., 2005; Ver-63 hoeven and Schmalzl, 2009) or both (Weinstein et al., 1988). All these studies 64 point out the importance of particle deposition and the subsequent differenti-65 ation mechanisms that may occur within large plutonic reservoirs. In magma 66 oceans, the physical context is quite different with potentially more vigorous 67 convection related to much thicker reservoirs. Recent numerical models from 68 Patočka et al. (2020) constrained the settling of inertial particles in turbulent 69 Rayleigh-Bénard convection without considering the re-entrainement process. 70 In particular, Patočka et al. (2020) identified different settling regimes and de-71 rived characteristic residence timescales. For their range of particule sizes and 72 densities, their results show that the settling should be rapid, much shorter 73 than the typical time scales for the solidification of a whole-mantle terrestrial 74 magma ocean supporting the idea of a fully fractional crystallisation. However, 75 using analog models, Solomatov et al. (1993) introduced the conditions for par-76 ticle re-entrainment after sedimentation at the bottom of a convecting reservoir. 77 For sufficiently high Rayleigh numbers, their models illustrate that the parti-78 cles might be re-entrained by the viscous stress produced by thermal plumes 79 emphasising that entrainement of crystals cumulated within dunes was likely at 80 the bottom magma oceans. Such a re-entrainment process could hence favour 81

equilibrium crystallisation. More recent laboratory experiments from (*Lavorel* and *Le Bars*, 2009) confirmed the founding from *Solomatov et al.* (1993) and characterized the influence of the density ratio between fluid and crystals and the temperature difference driving thermal convection.

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Several parameters likely influence the settling dynamics: (1) the density 87 difference between the settling crystal and the surrounding convecting magma 88 ocean, (2) the crystal size that is a strong function of the magma ocean cooling 89 rate and (3) the viscosity of the magma ocean which value may cover several 90 orders of magnitude depending on the pressure/temperature conditions within 91 the early magma ocean. Recently, Caracas et al. (2019) proposed that the ac-92 cumulation of crystals might occur near the depth of neutral buoyancy between 93 crystals and the coexisting melt. They localised this neutral buoyancy depth for 94 different partition coefficients iron partitioning and solid fractions. They show 95 that the crystals may cumulate and form a mushy layer inside the magma ocean 96 separating a shallow MO and a basal MO. Here, we monitor the stability of a 97 thin mushy layer cumulated within a magma ocean. We consider grain sizes, 98 the density differences between the crystals and the surrounding magma ocean 90 and viscosities of the magma ocean derived from petrological experiments. In 100 particular, we aim at determining if the crystal layer will be sedimented at the 101 bottom of the reservoir or will be efficiently mixed in the convective reservoir. 102 103

The hypothesis of a fully molten mantle is difficult to reconcile with geochemical data and the need for hidden reservoirs (*Boyet and Carlson*, 2005). However, the cooling and solidification processes occurring within large partially molten silicate reservoirs may lead to important chemical differentiation events and to chemical segregation. The depth at which solidification initiates is governed

by the crossing between the early geotherm and the melting curves. Crystal 109 settling could then occur towards neutral buoyancy depths where crystals are 110 paradoxically more easily entrained by the flow (*Caracas et al.*, 2019). Both 111 chemical composition and temperature contribute to the buoyancy of a crystal 112 layer within a convecting reservoir. The chemical differentiation occurring after 113 a large melting event is strongly related to the ability of the crystal fraction to 114 sink or float within the convective reservoir. This ability is itself governed by 115 the propension of the crystallised minerals to integrate in their structure heavy 116 elements such as Fe (Nomura et al., 2011; Andrault et al., 2012). Depending 117 on the crystal buoyancy relative to the molten magma, different scenarios may 118 emerge with a bottom/up solidification (Monteux et al., 2016) or a mid-mantle 119 solidification making a basal magma ocean possible (Labrosse et al., 2007). In 120 our study, we will discuss the implications of our model on the mechanism of 121 magma ocean solidification after a major Moon forming impact on an Earth-like 122 planet. 123

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In the following study, we have developed a numerical model to monitor 125 the cooling, segregation and chemical evolution of the early Earth mantle. Our 126 study is organised as follows. In section 2, we present our physical and numerical 127 models. In section 3, we present the main numerical results from our systematic 128 parametric study emphasising the influence of the melt viscosity, the crystal size 129 and the density difference between crystals and melt. Section 4 details a criterion 130 for the stability of a crystallising layer. Finally section 5 describes the evolution 131 of mushy material in the context of a progressive magma ocean crystallisation. 132 We discuss our results and conclude in section 6. 133

¹³⁴ 2. Physical and numerical models

135 2.1. Governing equations

We monitor the stability of a horizontal mushy layer composed of solid crys-136 tals within an initially convecting fully molten magma reservoir. For this pur-137 pose, we consider the Euler-Euler model which is an accurate dispersed mul-138 tiphase flow model describing both the crystal and the liquid phases. We dis-139 tinguish two non-miscible phases in our models: the continuous phase (with 140 c subscript) that represents the liquid magma ocean and the dispersed phase 141 (with d subscript) that represents the crystal phase. Both phases behave as 142 Newtonian and incompressible fluids. The local volume fraction of the fluid 143 continuous phase ϕ_c and the dispersed solid phase ϕ_d are the local average vol-144 ume of melt and crystals respectively ($\phi_c = 1 - \phi_d$). 145

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The dynamical and thermal evolution of the mushy layer and ambient liquid are governed by the Navier-Stokes and heat transfer equations. We initially solve the equations for the continuous phase heat in 2D rectangular geometry using the Rayleigh-Bénard system. Once it reaches a steady state, we then solve the dynamical evolution of the mushy layer within the convective reservoir. The Euler-Euler flow model defines one set of Navier-Stokes equations for each phase:

• The mass conservation:

$$\nabla (\phi_c \boldsymbol{u_c} + \phi_d \boldsymbol{u_d}) = 0, \tag{1}$$

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$$\nabla .(\phi_d \boldsymbol{u_d}) = 0 \tag{2}$$

with u_d and u_c represent of the dispersed phase and the continuous phases fluid velocity vector respectively (m.s⁻¹).

• The momentum conservation:

$$\rho_c(\frac{\partial \boldsymbol{u}_c}{\partial t} + \boldsymbol{u}_c.\nabla \boldsymbol{u}_c) = \boldsymbol{\nabla}.[-P\boldsymbol{I} + \eta_c(\nabla \boldsymbol{u}_c + (\nabla \boldsymbol{u}_c)^T)] + \rho_c \boldsymbol{g} + \frac{\boldsymbol{F}_{\boldsymbol{m},\boldsymbol{c}}}{\phi_c} \quad (3)$$

$$\rho_d(\frac{\partial \boldsymbol{u}_d}{\partial t} + \boldsymbol{u}_d, \nabla \boldsymbol{u}_d) = \boldsymbol{\nabla} \cdot [-P\boldsymbol{I} + \eta_d (\nabla \boldsymbol{u}_d + (\nabla \boldsymbol{u}_d)^T)] + \rho_d \boldsymbol{g} + \frac{\boldsymbol{F}_{\boldsymbol{m},\boldsymbol{d}}}{\phi_d} \quad (4)$$

with ρ_c the melt density (kg.m⁻³), ρ_d the solid phase density, η_c the melt viscosity (Pa.s), η_d the solid phase viscosity (Pa.s), t the time (s), P the fluid pressure (Pa), g the gravitational acceleration (m.s⁻²) and I the identity matrix. $F_{m,c}$ and $F_{m,d}$ are the drag forces (N/m^3)

- 162
- The heat conservation:

$$\rho_c C_p \left[\frac{\partial T}{\partial t} + \boldsymbol{u} \cdot \nabla T \right] + \boldsymbol{\nabla} \cdot \left[-k \boldsymbol{\nabla} T \right] = 0$$
(5)

with C_p the heat capacity at constant pressure (J.kg⁻¹.K⁻¹), T the fluid temperature (K), u the fluid velocity vector (m.s⁻¹), k the thermal conductivity (W.m⁻¹.K⁻¹). No heat source is considered in our models (e.g. radiogenic, tidal or latent heat). For simplicity, we consider that both the continuous and dispersed phases have the same heat capacities and thermal conductivities.

For each fluid parcel, the viscosity ranges between the viscosity of the fully molten phase η_c that is imposed for each simulation (see Tab. 1 for values) and the viscosity of the solid dispersed phase. In between the mixture viscosity η depends on the dispersed solid phase volume fraction ϕ_d . For the mixture, we consider a Krieger-type dynamic viscosity:

$$\eta = \eta_c (1 - MIN(\frac{\phi_d}{\phi_{d,max}}, 0.999))^{-2.5\phi_{d,max}}$$
(6)

where $\phi_{d,max}$ is the maximum packing concentration. $\phi_{d,max}$ depends on the geometry of the crystals within the system and on the shear rate. For monodispersed systems with spheres and low shear rates, $\phi_{d,max}$ ranges between 0.5 and 0.7 (*Dörr et al.*, 2013). We consider in our models that $\phi_{d,max}$ is fixed to 0.6 meaning that ϕ_c can locally range between 40 and 100% while ϕ_d can locally range between 0 and 60%.

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To compute the drag force F_m , we consider the Schiller-Naumann drag model (Schiller and Naumann, 1935; Ibrahim and Meguid, 2020) that is relevant for dispersed rigid spheres and for the range of Reynolds number (comparing inertial and viscous forces) values in our study (lower than 10^4). F_m is defined as:

$$\boldsymbol{F}_{\boldsymbol{m},\boldsymbol{c}} = -\boldsymbol{F}_{\boldsymbol{m},\boldsymbol{d}} = \beta(\boldsymbol{u}_{\boldsymbol{d}} - \boldsymbol{u}_{\boldsymbol{c}}) \tag{7}$$

with β the momentum transfer coefficient (in kg m⁻³ s⁻¹) between fluid and solid particles (*Ibrahim and Meguid*, 2020)

$$\beta = \frac{3}{4} \frac{\rho_c \phi_d C_D (\boldsymbol{u_d} - \boldsymbol{u_c})}{D_d} \tag{8}$$

¹⁸⁸ D_d is the particle diameter (m) and C_D is the drag coefficient for a single ¹⁸⁹ particle. For Re < 1000:

$$C_D = \frac{24(1+0.15Re^{0.687})}{Re} \tag{9}$$

For Re > 1000, $C_D = 0.44$ (Wen and Yu, 1966).

The density of the magma ocean is supposed to vary linearly with temperature in the buoyancy term of the momentum equation following the Boussinesq approximation. The density calculation accounts for the contrast related to the thermal effects and the phase influence. It is calculated as:

$$\rho_c = \rho_{c,0} (1 - \alpha (T - T_0)) \tag{10}$$

where T_0 is the reference temperature (in K) and α is the thermal expansion coefficient (K^{-1}). The initial reference temperature of the reservoir is highly influenced by the full thermal history of the early earth as well as its depth within the Earth mantle. Here we fix $T_0 = 4000$ K. The particle density is calculated as:

$$\rho_d = \rho_c + \Delta \rho \tag{11}$$

with $\Delta \rho$ the intrinsic density difference between the molten silicates and the solid crystals.

202 2.2. Non dimensional parameters

Before considering the stability of a crystal layer within the molten reser-203 voir, we first solve only the equations relative to thermal convection within the 204 reservoir. In our geophysical investigation, the main parameter is the magma 205 ocean viscosity which ranges widely in value. Here we use viscosities for the 206 continuous phase $(\eta_{c,0})$ ranging between 0.1 and 10 Pa.s (Karki and Stixrude, 207 2010; Xie et al., 2021). The convection flow induced within the magma ocean 208 is characterised by the thermal Rayleigh number (Ra) describing the vigor of 209 convection as a consequence of basal heating: 210

$$Ra = \frac{\alpha \rho_c g \Delta T H^3}{\eta_c \kappa} \tag{12}$$

with κ the heat diffusivity of the convective fluid (see Tab. 1 for value). 211 This leads to Ra values ranging between 1.8×10^6 and 1.8×10^8 . From our 212 purely thermally convective models, we monitor the surface heat flux F which 213 is proportional to the average value of the temperature gradient at the surface 214 of the reservoir. We obtain F as a function of time for the range of Ra values 215 considered in our study. We stop our thermally convective simulations once 216 the surface heat flux has converged to a constant value and use this value to 217 calculate the corresponding Nusselt number Nu: 218

$$Nu = \frac{HF}{k\Delta T} \tag{13}$$

Fig. 1 shows the Nu-Ra relation obtained from our models and compares our numerical results with the scalings obtained from Turcotte and Schubert (1982) and Wolstencroft et al. (2009). Our results show an agreement with these 2 scalings and we obtain $Nu = 0.14Ra^{0.34}$. For each Ra value used in our models we obtain a temperature field that will be used as the initial temperature field for the Euler-Euler model in order to study the stability of a thin mushy layer with different characteristics (D_d and $\Delta \rho$).

226 2.3. Numerical model

We used the commercial software COMSOL Multiphysics (version 5.4) previously validated for two phase flow applications (*Qaddah et al.* (2019, 2020)). We consider a 2D domain with a thickness H and a width of 4H (See Fig. 2). As a first attempt to characterise the stability of a thin newly formed mushy layer within a convective magma ocean, we arbitrarily consider that the thickness of this mushy layer is h = H/10. The mush thickness is set constant in all our

models. Within the mushy layer, the crystals are considered as spheres with a 233 fixed diameter D_d . In our models, this diameter ranges between 10^{-4} and 10^{-2} 234 m. The initial volume fraction of crystal $\phi_{d,0}$ is also set to a constant value 235 $(\phi_{d,0} = 0.1)$. For the dynamics, we use free-slip conditions at all the system 236 boundaries for both the dispersed and continuous phases. For the temperature, 237 we consider no flux conditions at lateral boundaries. We fix the temperature 238 to 4100 K at the bottom boundary and to 4000 K at the top boundary (i.e. 230 $\Delta T = 100K).$ 240

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The COMSOL Multiphysics software employs the finite-element method on 242 a grid mesh to solve the set of equations (Eqs. 1 to 5). In order to study 243 the sensitivity analysis of mesh size on the main results of this paper, we run 244 several models varying the size of the mesh (between 1.6 mm and 50 mm). In 245 these resolution tests we consider the overturn of a mushy layer with h = H/10, 246 $\phi_{d,0} = 0.5, \ \eta_c = 10 \ (Ra = 1.8 \times 10^6) \ \text{and} \ \Delta \rho = 30 \ \text{kg.m}^{-3}$. Fig. 3 shows 247 the average value $\phi_{d,average}$ along a horizontal profile located at the bottom of 248 the reservoir at t = 600s when the mushy layer is fully sedimented. $\phi_{d,average}$ 249 increases when decreasing the mesh size from 0.05 m to 0.01 m. Then, below a 250 mesh size value of 0.01 m, $\phi_{d,average}$ converges to a constant value of 0.45. In 251 order to save computational time, we consider a mesh size of 0.08 which leads to 252 an error of 7.7% relative to the converged value obtained for the smallest mesh 253 size. In addition, we decrease the size of the mesh by a factor 2 in the lowermost 254 part of the reservoir where crystal-rich patches are likely to settle or within the 255 whole reservoir for the most vigorously convective contexts. Therefore, the 256 calculation domain is composed of 100 000 to 180 000 cells. We list the details 257 of our models in Tab. 2. Each run presented in this study represents (for the 258 Euler-Euler flow model only) 12 to 72 hours computation time on a bi-processor, 259

²⁶⁰ 8-cores, 3.2-GHz workstation.

261 2.4. Reference case

In this subsection, we detail the case corresponding to simulation no. 47 in 262 Tab. 2. For this reference case, we consider the case with $Ra = 4.4 \times 10^6$ (cor-263 responding to $\eta_c=4$ Pa.s), $\Delta\rho=50$ kg.m^{-3} ($\Delta\rho/\rho_{c,0}=1.7\%$) and $D_d=10^{-3}$ 264 m. Fig. 4 illustrates the thermal and dynamical time evolutions of the convec-265 tive magma ocean and the mushy crystal layer. Because of negative buoyancy, 266 the crystal layer rapidly sinks. Fig. 4 shows that an overturn rapidly occurs 267 concentrating the crystals in the lower part of the reservoir during the first hun-268 dreds of seconds after the layer formation. The crystal fraction is first diluted 269 in the lower reservoir then starts to organise in patches where the concentration 270 is much higher than the initial concentration. This early stage limits the heat 271 transfer from the bottom surface to the reservoir leading to a transient cooling 272 of the magma. 273

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To monitor the evolution of the average concentration C (particles/m³) in crystals in the upper mid reservoir for the reference case, we define:

$$e = \frac{\int_{H/2}^{H} \int_{0}^{4H} C \mathrm{d}x \mathrm{d}z}{\int_{0}^{H} \int_{0}^{4H} C \mathrm{d}x \mathrm{d}z}$$
(14)

Höink et al. (2005) refers to this parameter as the relative entrainment. Fig. 5 illustrates the time evolution of e for our reference case and shows that during 279 2000 s, the upper reservoir is depleted in crystals. After nearly 10 000 s, large hot plumes formed at the bottom of the reservoir (see Fig. 4) transport the crystals back in the upper reservoir. Hence the concentration in crystals in the upper reservoir increases. This second phases leads to an efficient mixing in the whole reservoir and the crystal concentration gets back and slightly oscillates around its initial value. Fig. 5 shows that the initial concentration in the upper reservoir is slightly larger than its theoretical value of 0.01. This is due to the mesh properties that encompasses a larger volume of crystals leading to an initial value of e = 0.0105.

288 3. Parametric study

In the following section, we monitor the influence of the density difference between the crystals and the magma, the crystal size, and the magma viscosity on the settling dynamics within a convecting magma ocean.

²⁹² 3.1. Influence of the density difference $\Delta \rho$

During the solidification of the magma ocean, the density of the crystal phase 293 potentially increases (or decreases) relative to the molten phase depending on 294 its ability to integrate dense chemical elements in its structure. Within the fully 295 molten mantle, the first crystal to form is a crystal of bridgmanite. This first 296 crystal is Fe-poor, and becomes chemically neutrally buoyant close to the core 297 mantle boundary (Caracas et al., 2019). Then, the relative Fe content increases 298 in the melt potentially leading to the formation of a basal magma ocean overlaid 299 by a thick layer of bridgmanite. The front of neutral buoyancy moves towards 300 the shallow mantle as the solidification proceeds (Solomatov, 2000; Boukaré 301 et al., 2015; Caracas et al., 2019). This scenario potentially leads to a fractional 302 crystallisation of the early mantle (Patočka et al., 2020). However, when ac-303 counting for re-entrainment enhanced by vigorous convection, crystal dispersion 304 might be an efficient process that should favor batch (or equilibrium) crystalli-305 sation of the early mantle (Solomatov et al., 1993). The melt-crystal density 306 crossover in both these scenarios is discussed by Caracas et al. (2019). Here, we 307 monitor the influence of the chemical density difference on the settling dynamics 308 considering relatively small chemical density differences $(10-100 \text{ kg}.\text{m}^{-3})$ which 309

is relevant in the early stages of mantle solidification. The chemical density difference is fixed in each simulation and is superimposed to the thermal density difference which can evolve during the cooling. We note here that in our models, the density difference related to temperature differences ($\alpha\rho\Delta T = 3 \text{ kg.m}^{-3}$) is smaller that the density difference between the magma and the crystals.

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In Fig. 6, we considered the reference case illustrated in Fig. 4 in which 316 we increased the density difference from $\Delta \rho = 50 \text{ kg.m}^{-3}$ to $\Delta \rho = 100 \text{ kg.m}^{-3}$. 317 As already illustrated in Fig. 4, Fig. 6 shows that because of negative buoy-318 ancy, the crystal layer rapidly sinks. After a rapid settling at the bottom of the 319 reservoir, particles entrainment occurs from the dunes (10 000 s after the initi-320 ation of the settling). However, because of a larger density difference between 321 the dispersed phase and the continuous phase, efficient mixing is prevented. 322 Particle entrainment nearly vanishes and the layer is fully sedimented at the 323 bottom of the reservoir (28 000 s after the initiation of the settling). Again, 324 the sedimented layer acts as an insulating layer by limiting the heat transfer 325 from the bottom of the reservoir. As a consequence, the mean temperature is 326 significantly lower due to bottom insulation when full sedimentation is achieved. 327 328

Fig. 5 compares the time evolution of the concentration of particles in the 329 upper part of the reservoir for the two density differences illustrated in Fig. 4 and 330 6. During all the simulation, the concentration for the case with $\Delta \rho = 50 \text{ kg.m}^{-3}$ 331 remains larger than the concentration for the case with $\Delta \rho = 100 \text{ kg.m}^{-3}$. This 332 feature is in agreement with Lavorel and Le Bars (2009) who monitored the 333 relative number of particles in suspension as a function of the fluid/particle 334 density difference during the first 1000 s after being uniformly distributed within 335 the reservoir. Fig. 5 also shows that increasing the density difference decreases 336

³³⁷ both the minimum value of the particle concentration in the upper reservoir and ³³⁸ the time at which this minimum is reached. This feature is in agreement with ³³⁹ the theoretical overturn timescale derived for a viscous layer of thickness δ from ³⁴⁰ Hess and Parmentier (1995) and modified to account for the thermal buoyancy ³⁴¹ contribution:

$$t_{overturn} = \frac{4\pi^2 \eta_{lay,0}}{(\phi_{d,0}\Delta\rho - \alpha\rho_{lay,0}\Delta T)g\delta}$$
(15)

where $\eta_{lay,0}$ is the initial viscosity of the crystal rich layer (see Eq.6) and $\rho_{lay,0}$ is the initial density of the crystal rich layer (= $\phi_{d,0}\rho_d + (1 - \phi_{d,0})\rho_c$). The overturn timescales decreases with increasing density differences as shown in Fig. 5.

346 3.2. Influence of the crystal size

Upon solidifying, crystals form following the nucleation and growth mecha-347 nisms (Solomatov, 2015). The crystal size reached at equilibrium depends on 348 the nature of the mineral and on the cooling rate within the convecting reservoir. 349 The grain size may range between 10^{-4} m and 10^{-2} m in typical magma ocean 350 conditions (Solomatov, 2015). Here, we monitor the influence of the crystal size 351 in our models. To that aim, we compare the reference case (with $D_d = 10^{-3}$ m) 352 to cases with crystals larger $(D_d = 10^{-2} \text{m})$ and smaller $(D_d = 10^{-4} \text{m})$. Fig. 7 353 shows the time evolution of the crystal fraction in the upper part of the reservoir 354 for the three particle diameters D_d considered. Fig. 7 illustrates that, even for 355 the wide range of crystal sizes considered (2 orders of magnitude), the overturn 356 timescale is poorly sensitive to the particle diameter D_d (factor 3 in difference). 357 This result is in agreement in agreement with Eq. 15. 358

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However, Fig. 7 shows that the final behaviour of the reservoir in terms of sedimentation/suspension is dependent on the crystal size. Indeed, for the parameters chosen for the reference case ($\eta_c = 4 \text{ Pa.s} \text{ and } \Delta \rho = 50 \text{ kg.m}^{-3}$), Fig. 7 shows that when the particle size is equal to 10^{-2} m, the suspension regime is not reached after the overturn. For smaller particle diameters ($D_d = 10^{-4}$ and $D_d = 10^{-3}$), the overturn process is rapidly followed by a suspension regime where the particles are efficiently mixed within the convecting reservoir.

367 3.3. Influence of the viscosity

The viscosity of the molten phase (η_c) is a key parameter governing the cool-368 ing dynamics of the magma ocean and in particular the solidification timescales 369 (Monteux et al., 2016). According to Karki and Stixrude (2010), η_c for liquid 370 anhydrous MgSiO₃ may decrease down to 0.048 Pa.s at mid-mantle conditions 371 (P = 70 GPa and T = 4000 K along the magma ocean isentrope). For molten 372 peridotite, Xie et al. (2021) obtained a viscosity ranging between 0.017 and 373 0.038 Pa.s. To maintain a laminar flow, (i.e. with low Reynolds number), we 374 consider $0.1 \le \eta_c \le 10$ Pa.s. 375

376

Fig. 8 shows that the continuous phase viscosity governs the ability of the 377 solid phase to mix or sediment at the bottom of the reservoir. The interesting 378 feature arises from the non linearity of the process. Indeed Fig. 8 shows that for 379 the lowest viscosity, the crystal layer rapidly mix with the convecting reservoir. 380 Then for $0.4 \leq \eta_c \leq 1$, the rapid overturn process is not followed by a mixing 381 process and the final concentration within the upper part of the reservoir does 382 not reach the typical value synonymous of efficient mixing. Finally, for $\eta_c > 1$ 383 Pa.s, the overturn process is followed by a vigorous mixing process similarly to 384 the case with $\eta_c = 0.1$ Pa.s. 385

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Fig. 8 shows that the molten phase viscosity also governs the early overturn timescale (i.e. the time needed for the particle concentration in the upper part

of the reservoir to reach the minimal value). This overturn time increases from 389 nearly 40s (for $\eta_c = 0.1$ Pa.s) up to 2000s (for $\eta_c = 10$ Pa.s). We represent 390 these overturn timescales in Fig. 9 and compare them with the scaling law from 391 Eq. 15. Fig. 9 shows that the overturn timescales from our models is 5 to 392 40 times larger than the overturn timescale from Eq. 15. We attribute this 393 discrepancy to two points: (1) Eq. 15 is related to an overturn timesecale while 394 Fig. 9 represents the time from which the concentration in the upper part of 395 the reservoir re-increases after the overturn which is closer to a re-entrainement 396 timescale and (2) the mixing dynamics tends to dilute the concentration in 397 particles within the overturning layer during its sinking. Initially the particle 398 concentration in the layer is $\phi_{d,0}$. As illustrated in Figs. 4 and 6 (second 399 line), the vigorous convection redistribute the particles within the reservoir even 400 during the early overturn. As a consequence the density of the mushy layer 401 relative to the ambient fluid decreases from its initial value. The mushy layer 402 settles with a timescale that is larger than if the layer could maintain its initial 403 structure. 404

405 4. A criterion for the stability of a crystallising layer

We have illustrated the separated influence of the density difference between 406 crystals and melt, grain size and melt viscosity on the behaviour of a crystallising 407 layer within a magma ocean. We now aim at anticipating the ability of the flow 408 generated from the convection within magma ocean to initiate the motion and 409 suspension of sedimented crystals and to lead to efficient mixing within the 410 magma ocean. We introduce the Shields number Sh that is the ratio between 411 the tangential stress from convective flow over the buoyancy stress relative to 412 density difference between the crystals and the melt (Solomatov et al., 1993): 413

$$Sh = \left(\frac{\eta_c \alpha gF}{C_p}\right)^{1/2} \frac{1}{\Delta \rho g D_d} \tag{16}$$

Large Sh values favor a suspension regime while small Sh values favor a 414 sedimentation regime. The two regimes are separated by a critical Shields num-415 ber value Sh_c . From their analog experiments, Solomatov et al. (1993) have 416 estimated a critical value $Sh_c = 0.1 - 0.2$. We have run a total of 76 numer-417 ical models (with values listed in Tab. 2) considering different sets of D_d , $\Delta \rho$ 418 and η_c with value ranges in Tab. 1. Each model corresponds to a Sh value 419 calculated considering the combination of Eq. 12, Eq. 13 and the expression 420 $Nu = 0.14 Ra^{0.34}$ obtained from Fig. 1 to compute the heat flow F (See Tab. 2 421 for values). Monitoring the concentration of grains in the upper reservoir (e(t))422 in each models enables to decipher if the models end up in the suspension or 423 sedimentation regime. 424

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In Fig. 10, we illustrate the Sh values as a function of the molten phase 426 viscosity η_c . A case where the grain concentration in the upper part of the 427 reservoir (e) never reaches back its initial value is considered in the sedimen-428 tation regime. Oppositely, a case where the grain concentration in the upper 429 part of the reservoir gets back to its initial value is considered in the suspen-430 sion regime. As shown in Fig. 8, some cases may end up in an intermediate 431 regime where the final concentration is 0.005 < e < 0.01. We consider that these 432 intermediate cases fall out in the sedimentation regime. Sedimentation and sus-433 pension regimes are separated by the critical Shields number value (Sh_c) . 434

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According to Fig. 10, the Sh_c value is a non-linear function of the continuous molten phase viscosity. For $\eta_c > 1$, Sh_c seems to be constant and independent of the η_c value. However, when $\eta_c < 1$, the Sh_c value decreases with decreasing values of the viscosity following $Sh_c = 0.1\eta_c^{0.45}$. As a consequence Sh_c is not influenced by the increase of Rayleigh number up to $Ra = 1.8 \times 10^7$. For Ravalues larger than 1.8×10^7 the increase of the kinetic energy induced by the vigorous convection in the system results in a decrease of Sh_c following:

$$Sh_c = 184.Ra^{-0.45}$$
 (17)

The non-linear dependence of Sh_c on the viscosity explains why the crystal 443 settling behaviour may vary back and forth between sedimentation and suspen-444 sion regimes when the viscosity increases. Indeed Fig. 10 shows with the red 445 dotted line the path followed for the models detailed in Fig. 8. Moreover, Fig. 446 10 shows that Sh_c is smaller than the range of values proposed by Solomatov 447 et al. (1993) (0.1 - 0.2). This feature is particularly visible for the lowest range 448 of viscosities used in our models. For a magma ocean viscosity value of 0.1 Pa.s, 449 the Sh_c value may decrease down to 0.04 which means that the crystal settling 450 is difficult to envision in such a convecting environment especially regarding the 451 typical viscosity values for a magma ocean (Karki and Stixrude, 2010; Xie et al., 452 2021). Decreasing the viscosity down to the lowest values proposed by Xie et al. 453 (2021) may enhance this feature. 454

455 5. Fate of mushy material in the context of a progressive magma 456 ocean crystallisation

457 5.1. Parameters controlling the solidification of a magma ocean on a Earth-like 458 planet

We now investigate the implication of our model on the mechanism of magma ocean (MO) solidification on Earth. More precisely, we investigate the possibility of Bridgmanite (Bg) segregation depending on its buoyancy (positive or negative). To that aim, we compare the density difference between the MO and the solid crystal $\Delta \rho_{MO}$ calculated for different compositions and the critical density contrast $\Delta \rho_{crit}$ above which crystal segregation is likely to occur. $\Delta \rho_{crit}$ is derived from Eq. 16 by calculating:

$$\Delta \rho_{crit} = \left(\frac{\eta_c \alpha gF}{C_p}\right)^{1/2} \frac{1}{Sh_c gD_d} \tag{18}$$

and considering $Sh_c = 0.1$ which is a conservative value regarding a magma ocean viscosity of 0.1 Pa.s or less (*Xie et al.*, 2021). At a given depth (z), a $\Delta \rho_{MO}$ larger than the critical value $\Delta \rho_{crit}$ would yield an efficient Bg sedimentation to the bottom of the MO. By extension, we also consider that Bg would efficiently segregate into a solid layer above a basal magma ocean if $\Delta \rho_{MO}/\Delta \rho_{crit}$ is smaller than -1. Calculating $\Delta \rho_{MO}$ requires a number of assumptions:

1. We use pyrolite as a model of reference for the mantle composition. Py-472 rolite is obtained by adding some crust component to the present-day 473 depleted (peridotitic) upper mantle (*Ringwood*, 1975). To implement flex-474 ibility on the possible mantle compositions, we add or subtract a compo-475 nent of bridgmanite (Bg) to pyrolite; it yields less or more mafic mantle 476 material, respectively. Later, we will argue that the composition of the 477 primitive upper mantle (PUM) should be close to pyrolite, as this is widely 478 479 accepted.

2. The composition of the bulk silicate Earth (BSE) is still not well estab-480 lished, because a number of deep mantle reservoirs could have a com-481 position different than pyrolite: The large low seismic velocity provinces 482 (LLSVP) seem to present a different mineralogy (e.g. Vilella et al. (2021)), 483 the deep lower mantle could be more bridgmanitic than pyrolite (Mu-484 rakami et al., 2012) and there could be relics above the core-mantle bound-485 ary (CMB) of a basal magma ocean (Labrosse et al., 2007) or of early 486 mantle overturns (e.g. Boukaré et al. (2015)). In our models, we give a 487

large freedom of size of the possible Bg-rich or mafic reservoirs that could have segregated in the deep mantle during the MO solidification. The flexibility of BSE composition is achieved by adding or subtracting up to 40% of Bg to the pyrolite mantle reference. This is a much broader range of chemical variation than the uncertainty remaining today on the major elements concentration in BSE.

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3. The Bg composition is calculated by equilibrating it with the melt in 494 the mushy MO, using partitioning coefficients $K(Al)^{Bg/Melt} = 0.5$ and 495 $K(Fe)^{Bg/Melt} = 0.1$ or 0.25. The value of $K(Al)^{Bg/Melt}$ does not impact 496 our calculations significantly. $K(Fe)^{Bg/Melt}$ remains controversial as it is 497 reported between ~ 0.07 to ~ 0.5 (Andrault et al., 2012; Nomura et al., 498 2011). The former value could be underestimated as it was measured in a 499 sample presenting extreme chemical segregation in a temperature gradient, 500 which artificially promotes the Fe-depletion of Bg at the highest temper-501 ature. In the latter study, the experimental conditions corresponded to 502 solidus-like temperatures, at which Fe is more compatible in Bg. Relevant 503 temperatures for MO solidification evolve from liquidus-like to intermedi-504 ate between liquidus and solidus. Therefore, we perform our calculations 505 for $K(Fe)^{Bg/Melt}$ values of 0.1 or 0.25. 506

4. As a reference model for the mushy MO, we use Bg grain diameter (D_{Bq}) 507 of 0.1 to 10 mm, viscosity (η) of 0.1 Pa.s (Xie et al., 2020), intrinsic 508 conductivity (k) of 5 W/m/K (Hofmeister, 1999) and heat capacity (Cp) 509 of 1800 J/Kg/K (Asimow, 2018). We consider that the melt has the same 510 composition at all depths because of efficient mixing in the turbulent MO. 511 5. We consider that the term ΔT used in F calculation from Eq. 18 is the 512 temperature difference between the Earth's surface and the bottom of the 513 MO, after correcting for the adiabatic increase of temperature with mantle 514 depth. We first fix the surface temperature (T_{surf}) to 500 K (Lebrun et al., 515

2013; Monteux et al., 2020), however, we will investigate later T_{surf} of 1000 516 and 1500 K. We consider that the bottom of the MO (z_{MO}) is the depth 517 where the mantle solidification is achieved for a mixture of 60% of Bg and 518 40% of melt. During the mantle solidification, z_{MO} evolves from the CMB 519 depth towards the surface. We assume that the temperature at a depth 520 z_{MO} follows the solidus temperature profile of pyrolite $(T_{sol}(z_{MO}))$ (Pierru 521 et al., subm.). We note that using the solidus profile yields a conservative 522 ΔT , compared to the true temperature that should be between solidus 523 and liquidus. Then, we calculate $\Delta T(z_{MO})$ of a MO extending from a 524 depth z_{MO} to the Earth surface using the following equation: 525

$$\Delta T(z_{MO}) = T_{sol}(z_{MO}) - T_{surf} - \Delta T_{ad}(z_{MO}) \tag{19}$$

where $\Delta T_{ad}(z_{MO})$ is the temperature difference between $T_{sol}(z_{MO})$ and the corresponding potential surface temperature extrapotated from $T_{sol}(z_{MO})$ to the surface along the MO adiabatic temperature profile. The adiabatic gradient in the MO is calculated using the equations of state available for different silicate melt compositions (*Asimow*, 2018). $\Delta T(z_{MO})$ is found to range between ~ 1300 K and ~ 2000 K for MO extending from 660 km (24 GPa) to 2800 km (135 GPa), respectively (see Fig. 11).

6. We calculate the density of Bg (ρ_{Bq}) along the mantle solidus, using the 533 equations of state (EoS) of MgSiO₃, FeSiO₃ and Al₂O₃ end-members 534 (Matas et al., 2007). The decomposition in end-members assumes Bg 535 of $(Mg_{1-x-y}Fe_xAl_y)(Si_{1-y}Al_y)O_3$ stoichiometry with a dominant fraction 536 of Fe^{2+} over Fe^{3+} at lower mantle conditions, in agreement with pre-537 vious works (e.g. Andrault et al. (2018)). This assumption has little 538 impact on the Bg density; the dominating parameter is the global Fe con-539 tent in Bg, itself controlled by $K(Fe)^{Bg/Melt}$ (discussed above). At the 540

same P-T conditions, we calculate the MO density (ρ_{Melt}) and thermal 541 expansion (α_{Melt}) using the EoS of the end-member molten MgSiO₃ en-542 statite, Mg₂SiO₄ forsterite, Fe₂SiO₄ fayalite, CaMgSi₂O₆ diopside, and 543 $CaAl_2Si_2O_8$ anorthite (Asimow, 2018). These calculations yield the Bg-544 to-melt density contrast ($\Delta \rho_{MO} = \rho_{Bg} - \rho_{Melt}$, Fig. 12) along the mantle 545 solidus: $\Delta \rho$ decreases with increasing pressure whatever is the composi-546 tion of the mushy mantle, because the melt is more compressible than 547 Bg. 548



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7. We do not attempt to model the latest stages of MO crystallization, when the bottom of the MO is shallower 660 km depth, because the phase relations rely on the melting diagrams at upper mantle P-T conditions.

⁵⁵² 5.2. Conditions for segregation of Bg in the mushy magma ocean

553 5.2.1. Effect of melt composition

We now calculate the ratio of density differences $\chi_{\rho} = \Delta \rho_{MO} / \Delta \rho_{crit}$ at all 554 lower mantle depths using the parameters defined above. First, we fix the grain 555 size of Bg to 1 mm, $K(Fe)^{Bg/Melt}$ to 0.1 or 0.25, and vary the MO composition 556 from pyrolite -40% of Bg to pyrolite +40% of Bg (i.e. 70\% of pyrolite and 30\%) 557 of Bg). χ_{ρ} is found to be positive or negative (Fig. 13), depending on the solid-558 melt density difference (Fig. 12). A $\Delta \rho_{MO}$ value larger than $\Delta \rho_{crit}$ (i.e. $\chi_{\rho} > 1$) 559 indicates an efficient Bg segregation to the bottom of the MO. Our calculations 560 show that Bg segregation happens in shallow MOs for all chemical compositions 561 investigated. For pyrolite, this is in agreement with a previous work dedicated 562 to the determination of the MO viscosity (Xie et al., 2020). The higher is the Bg 563 content in the MO, the earlier (the deeper) Bg segregation occurs during the MO 564 solidification; this is because $\Delta \rho_{MO}$ is larger when the bulk Fe-content is lower. 565 As expected, lower $K(Fe)^{Bg/Melt}$ favors a denser melt, and thus lower $\Delta \rho_{MO}$. 566 Still, our calculations show a moderate impact on the results of $K(Fe)^{Bg/Melt}$ 567

variations from 0.1 to 0.25. Finally, efficient Bg segregation could occur above a dense MO for a MO composition highly depleted in Bg; this is suggested by $\chi_{\rho} < -1$ for pyrolite -40%Bg at pressures larger than ~ 120 GPa, for a Bg grain size of 1 mm (Fig. 13).

572 5.2.2. Effect of grain size

We now investigate the role of the Bg grain size, for five different mantle 573 compositions (Fig. 14). We first focus on a melt of pyrolite composition. χ_{ρ} 574 is found positive at most of the mantle depths, because $\Delta \rho_{MO}$ is also positive 575 (Fig. 12), except in the lowermost mantle for $K(Fe)^{Bg/Melt} = 0.1$. Still, χ_{ρ} 576 can only be larger than 1 for D_{Bg} larger than 0.5 mm; Bg-grains of e.g. 2 mm 577 segregate when pressure (depth) is larger than 70 GPa (1650 km). Grain size 578 in a crystallizing MO remains a complicated issue (Solomatov, 2015): (i) as 579 long as the MO is fully liquid close to the planetary surface, grains are often 580 remolten within the large scale convection movements and, therefore, grain size 581 is controlled by nucleation processes; (ii) When the crystallization zone reaches 582 the surface, it enables some grains to grow indefinitely. According to the shape 583 of mantle solidus and liquidus, this change of mechanism could happen when 584 the bottom of the MO reaches a depth of 1000 km (40 GPa) (see Xie et al. 585 (2020)). In this article, we aim at modelling the chemical segregation in the 586 first stages of the MO solidification, when grain size is controlled by nucleation 587 processes. D_{Bg} in these conditions depends largely on the cooling rate of the 588 MO, with estimated values of 0.1 to 1 mm (Solomatov, 2015). For 1 mm grain 589 size, χ_{ρ} is larger than unity at pressures (depths) larger than 45GPa (1100 590 km) (Fig. 13). As a result, it appears unlikely that Bg grains could segregate 591 in a crystallizing MO of pyrolite composition for a depth larger than ~ 1100 km. 592 593

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A BSE composition enriched in Bg yields a lighter melt on the first stages

of the MO solidification, and thus a larger $\Delta \rho_{MO}$. It makes it easier the segregation of Bg at the bottom of the mushy MO. For a melt composition enriched with 20% or 40% of Bg, Bg grains of 1 mm could efficiently segregate at pressures (depths) of 55 GPa (1350 km) or 65 GPa (1550 km), respectively (Fig. 14 a and b). From a geochemical point of view, an early segregation of Bg to the CMB requires a BSE composition enriched in Bg if the late shallow MO should be of pyrolite composition; we will discuss this matter later in the article.

Alternatively, a BSE composition depleted in Bg yields a denser melt in the 603 mushy MO, as discussed in several works (Boukaré et al., 2015; Ballmer et al., 604 2017a; Caracas et al., 2019). Our calculations of χ_{ρ} for melt compositions of 605 pyrolite -20% and -40% of Bg show that Bg is indeed less dense than the melts in 606 a large fraction of the lower mantle (Fig. 12). Bg remains always denser at low 607 pressures, due to its very high bulk modulus. For Bg grain size of 1 mm, values 608 of χ_{ρ} are found lower than -1 at pressures (depths) larger than 120 GPa (2600 609 km) for a melt composition of pyrolite -40% of Bg (Fig. 14 e). The pressure 610 (depth) at which Bg could segregate upwards decreases with increasing the Bg 611 grain size. From a geochemical point of view, an early segregation close to the 612 CMB of a Bg-depleted melt requires a BSE composition depleted in Bg, if the 613 shallow MO were to be pyrolitic. 614

⁶¹⁵ 5.2.3. Effect of temperature

Another important parameter in $\Delta \rho_{crit}$ is ΔT (Eq. 19), which is intrinsically related to the rate of MO cooling. During the mushy stage of a MO solidification, the surface temperature is ≈ 500 K, which yields to final MO solidification in 100-200 thousands years after the Moon forming impact (*Lebrun et al.*, 2013; *Monteux et al.*, 2020). Presence of a blanketing atmosphere atop the MO would induce a larger surface temperature and a lower rate of cooling (*Sleep et al.*,

2014; Salvador et al., 2017). We note that the formation of a blanketing atmo-622 sphere is unlikely to affect the first stages of the MO solidification for different 623 reasons: (i) H₂O (a most efficient blanketing compound) is efficiently vaporized 624 to the atmosphere only after a high level of MO solidification (Elkins-Tanton, 625 2008; Salvador et al., 2017). (ii) a surface temperature larger than 1500K yields 626 an atmosphere dominated by rock-vapor, which is inefficient to stop the surface 627 cooling by thermal radiation (Sleep et al., 2014). For these reasons, we also per-628 form calculations of $\Delta \rho_{MO}$ for surface temperatures of 500K, 1000K and 1500 629 K (Fig. 15). 630

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⁶³² Calculations show that larger surface temperatures yield more extreme pos-⁶³³ itive and negative values of χ_{ρ} and therefore easier Bg segregation. This is be-⁶³⁴ cause larger T_{surf} corresponds to smaller temperature difference ΔT (Eq. 19) ⁶³⁵ and thus a less turbulent MO. Still, the impact remains modest; for a pyrolite ⁶³⁶ mantle, variation of T_{surf} from 500 to 1500 K yields Bg segregation at pressures ⁶³⁷ (depths) lower than 45 GPa (1100 km) and 67 GPa (1600 km), respectively.

5.3. Possible scenarios of MO solidification

⁶³⁹ 5.3.1. Conditions for Bg segregation at the bottom of the MO

Geochemical scenarios of the Earth's formation involve building blocks of 640 chondritic composition. Uncertainties remain about the composition of the 641 primitive "chondritic-type" mantle after the core-mantle segregation. Still, it 642 is very likely that it would be enriched in SiO_2 , compared to pyrolite (Allègre 643 et al., 1995; Javoy, 1995; McDonough and Sun, 1995). Such enrichment raised 644 long-term discussions about how much the core could contain some Si and how 645 much the deep mantle could be enriched in SiO_2 , compared to the present day 646 upper mantle. A high $MgSiO_3/MgO$ ratio in the lower mantle is compatible 647 with results from inversions of seismic profiles based on elastic parameters of 648

⁶⁴⁹ minerals (Samuel et al., 2005; Matas et al., 2007; Murakami et al., 2012).

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Our model shows that increasing the Bg content in BSE favors the segre-651 gation of Bg in a deep MO (Fig. 14 a and b). Bg grains of 1mm diameter 652 segregate efficiently (i.e. $\chi_{\rho}>1)$ at pressures (depths) lower than 45GPa (1100 653 km), 55 GPa (1350 km) and 65 GPa (1550 km) for pyrolite, pyrolite with ad-654 dition of +20%Bg and +40%Bg, respectively. Still, Bg grains would segregate 655 at the early stages of the solidification if the grain size would be 5 mm or more. 656 Such big grains are not expected in a very thick MO (Solomatov, 2015). Bg 657 segregation becomes possible when the MO is shallower and the greater the 658 Bg enrichment, the earlier (the deeper) the Bg-segregation. For a grain size of 659 1mm, a Bg layer could form at MO depths of 1100 to 1550 km, depending on 660 the BSE composition. Such thin MO could also favor grain growth, because 661 the MO is partially crystallized up to the Earth's surface (Solomatov, 2015; Xie 662 et al., 2020). Such Bg-layer could yield silica-enriched domains (Ballmer et al., 663 2017b). Unfortunately, our models are not adapted to address the thickness of 664 layer, because of too many adjustable parameters. Segregation of some Bg in 665 the lower mantle would drive the MO toward a pyrolite mantle, for a BSE is 666 enriched in Bg. Decreasing the Bg-content makes the segregation of Bg grains 667 less efficient (Fig. 13). 668

5.3.2. Solidification of a pyrolite-type bulk silicate Earth

A pyrolite-like BSE would solidify with no Bg segregation from the melt, as long as the Bg grain size always remains lower than 0.5 mm (Fig. 14 c). Bg segregation could still happen in the uppermost lower mantle if the Bg grain size gets larger. We note, however, that the grain growth of olivine and other minerals of the upper mantle may have a moderate impact on the grain size of Bg in the lower mantle. Modelling the mechanism of MO solidification in the ⁶⁷⁶ upper mantle is beyond the scope of this article.

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Interestingly, absence of Bg segregation does not imply a chemically homo-678 geneous mantle once it is entirely solidified. The origin of this effect is that the 679 solidified mantle contains $\sim 60\%$ of Bg and $\sim 40\%$ of melt, while the overlying 680 mushy MO contains a different fraction of the two phases. Here we assume 681 the rate of mushy-mantle compaction (with extraction of the melt) significantly 682 slower than the rate of MO solidification, following a previous work (Soloma-683 tov, 2015). To model the chemical stratification resulting from homogeneous 684 solidification, we proceed as follows: (i) we choose a BSE composition close to 685 pyrolite. Still, we keep its Bg content adjustable, to enable a final shallow MO 686 (thickness of ~ 660 km) composition as close as possible from pyrolite. (ii) At 687 each time step of the progressive MO solidification, we estimate the fraction of 688 Bg grains in the MO above the bottom of the MO (ϕ_{MO}) based on our previ-689 ous model of progressive MO solidification (Fig. 3 of Monteux et al. (2016)). 690 (iii) We calculate the composition of the coexisting melt and Bg grains, fixing 691 $K(Fe)^{Bg/Melt} = 0.25$. Anyway, the value of $K(Fe)^{Bg/Melt}$ has no significant 692 impact on this calculation. (iv) Starting from the CMB, we cool the mantle and 693 calculate the composition of a mixture of 60% of Bg grains and 40% of melt 694 which solidifies at the bottom of the MO. The chemical content of this bottom 695 layer is then subtracted to the composition of the overlying mushy MO, before 696 a next step of the solidification is performed within a thinner MO. 697

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This calculation shows significant changes of the SiO_2 , FeO and CaO contents with depth in the solidified mantle (Fig. 16). They occur due to Bg presenting higher SiO_2 and lower FeO and CaO contents, compared to the melt. The shallow MO presents a chemical composition close to pyrolite, when the

BSE composition is adjusted to 80% of pyrolite and 20% of Bg. The SiO₂ con-703 tent in the deep mantle is found to decrease from ~ 46 to 39%, which translate 704 into a decrease from ~ 90 to 70% of the Bg-content, with decreasing depth from 705 2900 to 660 km, respectively. The Fe-content also increases with decreasing 706 depth, because of its incompatible character; this effect is found even larger 707 when using $K(Fe)^{Bg/Melt} = 0.1$, instead of $K(Fe)^{Bg/Melt} = 0.25$. Still, the 708 overlying mushy MO does not become denser than the solidified layers, because 700 Bg is denser than the melt at all mantle depth for Bg-enriched BSE composition 710 (Fig. 12). 711

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Such a chemical layering would only survive as such in a mantle that would 713 be stagnant after its solidification. Instead, one could expect important ver-714 tical mixing in mushy mantle presenting a relatively low viscosity and with a 715 major heat flux from the core to the surface (Monteux et al., 2016). Another 716 effect that can moderate the chemical stratification is a simultaneous mantle 717 solidification in a large range of mantle depths. Upon cooling, the tempera-718 ture profile becomes almost parallel to the solidus (Miller et al., 1991; Stixrude 719 et al., 2009; Monteux et al., 2016), which could yield to a bulk solidification. 720 Still, these effects may not suppress entirely the chemical gradients, in partic-721 ular between a shallow MO and the lower mantle. The progressive enrichment 722 of Bg with mantle depth is in good agreement with the mineralogical models 723 (Samuel et al., 2005; Matas et al., 2007; Murakami et al., 2012), as well as with 724 the difference between shallow the mantle composition today (pyrolite) and a 725 "chondritic-type" BSE (see above). 726

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We note that the scenario of Bg-segregation in the lowermost mantle does not help explaining the LLSVPs atop the CMB. Several origins were proposed

for the LLSVP, in particular some primitive mantle that did not melted upon 730 the Moon forming impact. Primitive mantle compositions imply a relatively low 731 proportion of ferropericlase, which is compatible with the comparison between 732 the LLSVP seismic signature and the elastic parameters of the minerals (Vilella 733 et al., 2021). Still, LLSVPs represent a quite small fraction of the mantle vol-734 ume, typically 1.5 to 2.5 % (Burke et al., 2008). Therefore, our models would 735 not be much different if some primitive mantle would have survived to melting 736 at the Moon Forming impact. 737

738 5.3.3. Conditions for the formation of a primary basal magma ocean

The segregation of large Bg grains to the bottom of a pyrolite mantle would 739 yield a mafic Fe-enriched overlying MO. Following this scenario, it has been pro-740 posed that the melt could become denser than the underlying mantle and induce 741 early mantle overturns (Boukaré et al., 2015; Ballmer et al., 2017a; Caracas 742 et al., 2019). Such a reversal could result in a secondary MO with a composi-743 tion typical of a shallow MO. Our results do not strongly support this scenario, 744 because the lower the Bg-content in the mushy MO, the less efficient the Bg 745 segregation (Fig. 14). Therefore, bulk solidification should rapidly be favored, 746 after some Bg-segregation may have occurred in a thick MO. Mantle overturns 747 may still occur due to the crystallization processes in the upper mantle; this 748 should be investigated in further works based on analyses of the melting dia-749 grams in the upper mantle. 750

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Formation of a basal MO during the solidification process (i.e. a primary basal-MO) would require the efficient segregation of Bg in an overlying solid layer (*Labrosse et al.*, 2007). This conditions translates into $\chi_{\rho} < -1$ in our model. Such condition is reached for mafic BSE composition, which present a lower Bg content, compared to pyrolite (Fig. 14 c and d). Still, our calculations

show that Bg segregation upwards is only expected (i) in the lowermost part of 757 a MO (ii) for a BSE composition depleted of more than -20% of Bg, compared 758 to pyrolite and (iii) for Bg grain size larger than 0.5 mm. We note that low 759 $K(Fe)^{Bg/Melt}$ value at these conditions help making the melt denser than Bg 760 (Fig. 13, Nomura et al. (2011)). For a MO composed of pyrolite -40%Bg, Bg 761 grains of 1 mm diameter would aggregate efficiently to a Bg-rich layer that 762 would be located pressure (depth) of 120 GPa (2600 km). For a MO composed 763 of pyrolite -20% Bg, only Bg grains of 5 mm diameter could segregate efficiently 764 upwards. The simultaneous formation of a basal MO and a solid layer of Bg may 765 maintain mafic the composition of the overlying MO. No further Bg segregation 766 should be expected above ~ 2500 km (i.e. 100-110 GPa) in a mafic MO, for 767 a reasonable Bg grain size. In case the MO would be further depleted in Bg 768 due to the mechanism of "at equilibrium fractionation" described above (Fig. 769 16), it would make it even more mafic. At this point, it is difficult to imagine 770 which geological process could lead to the disappearance of such a major volume 771 of early mafic MO. The path to pyrolite-type upper mantle seems difficult to 772 achieve. It makes the formation of a primary basal MO simultaneous to MO 773 solidification relatively unlikely. 774

775 6. Conclusion

The solidification within a deep convecting magma ocean (MO) is a key process that significantly shaped the Earth's interior (*Ballmer et al.*, 2017a) and likely the early mantle evolution for several planetary contexts : Mars (*Elkins-Tanton et al.*, 2005), Mercury (*Mouser et al.*, 2021) or the Moon (*Elkins-Tanton et al.*, 2011; *Dygert et al.*, 2017). This process is responsible for the separation between compatible elements that prefer the solid phase (e.g., Mg, Si) and incompatible elements which prefer the liquid phase (e.g., Al, Na, Fe). The solidification depth is governed by the geotherm and the melting curves. With density profiles of the melt and bridgmanite crystals crossing in the lower mantle, crystal settling could then occur towards neutral buoyancy depths where crystals are paradoxically more easily entrained by the flow. Both chemical composition and temperature contribute to the buoyancy of a crystal layer within a convecting reservoir. Hence different scenarios may emerge: Bottom/up solidification or mid-mantle solidification making a basal magma ocean possible.

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In our study we have shown that the ability of the crystal fraction to sed-791 iment or to disperse within the domain is the result of a combination of the 792 influence of the crystal size, the density difference between the crystals and the 793 magma and the magma viscosity. We have shown that this ability is a fonc-794 tion of the ratio of the convection over the buoyancy stress (expressed as the 795 Sh dimensionless number). From a relevant range of parameters we observe 796 that the critical Sh_c separating suspension and sedimentation regimes exhibits 797 values lower than proposed by Solomatov et al. (1993). A a consequence, our 798 results strengthen the idea that suspension mechanism should be the dominant 799 process during the magma ocean solidification compared to sedimentation and, 800 as a consequence, equilibrium crystallisation should be favoured compared to 801 fractional crystallisation. 802

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In our models, we consider a particule size range $(10^{-4} - 10^{-2} \text{ m})$ and a density difference range between the magma and the particles $(10 - 100 \text{ kg.m}^{-3})$, which are probably closer to the relevant MO conditions (see e.g. $\Delta \rho = -200$ to 200 kg/m³ in Fig. 12) than the range of parameters proposed by *Patočka et al.* (2020) $(0.5 \times 10^{-3} - 10^{-2} \text{ m} \text{ and } 10 - 3000 \text{ kg.m}^{-3})$. Moreover, their models neglect the re-entrainement process by eliminating the particles that reach the

bottom of the reservoir. Hence, it is not surprising that their models favour a 810 rapid crystal settling and a a fractional crystallisation. Patočka et al. (2020) 811 identified three settling regimes: "stone-like", bi-linear and "dust-like" regimes 812 depending on the ratio between the crystal terminal velocity and the average 813 vertical velocity of the flow. It appears that the models we run mostly fall out 814 in the "dust-like regime" where the terminal velocity of a particle is smaller 815 than the average velocity within the convective reservoir. In the "dust-like" 816 regime the particles from the models developed by $Patočka \ et \ al. (2020)$ appear 817 uniformly distributed in the reservoir in agreement with our models. 818

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We have also considered in our models different end-members for the chem-820 ical composition of the early mantle adding or subtracting up to 40% of Bg to a 821 pyrolite mantle reference. Exploring the early mantle composition within such 822 range leads to a wide range of density difference between the molten and the 823 solid phases. Hence the chemical composition also influences the ability of the 824 crystals to cumulate at the bottom or in the middle of the magma ocean. Our 825 models emphasize that (1) Bg grains are unlikely to segregate in a mantle of 826 pyrolite composition, except in a shallow MO when the grain size may increase 827 to 1 mm and more, (2) Bg grains are likely to segregate at the bottom of a MO 828 enriched in SiO_2 , compared to pyrolite and (3) the formation of a basal MO 829 simultaneous to mantle solidification could be possible for mafic BSE composi-830 tions (difficult to reconcile with the Earth's geochemical models). We also note 831 that a homogeneous bottom-up solidification of the MO is expected to produce 832 a significant SiO₂ enrichment with increasing mantle depth in a primitive man-833 tle (compatible with seismic observations) and a higher SiO_2 content in BSE 834 compared to pyrolite (compatible with Earth's geochemical models). 835

836

This study is a supplementary step toward a more realistic approach that 837 should account for several other major processes. In our models we have not 838 consider the time evolution of the crystal size, of the crystal fraction and of the 839 density of the crystals during their growth. If crystal settling is likely to occur 840 close to mechanical boundaries (CMB or solidified wall) where the velocity is 841 lower than in the middle of the magma ocean (within "calm" conditions), future 842 models should explore parameters compatible with the center of the magma 843 ocean (i.e. larger Rayleigh numbers). This would imply much higher Reynolds 844 number and would require to consider the inertial forces and the turbulence 845 equations in our model. The likeliness of a thick magma ocean and the presence 846 of a large Moon much closer to the Earth than today should also lead us to 847 consider te influence of the Coriolis forces (Solomatov, 2000). 848

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855 Data availability

The data that support the findings of this study were obtained using the commercial software COMSOL Multiphysics (version 5.4). COMSOL Multiphysics is a simulation platform that provides fully coupled multiphysics and singlephysics modeling capabilities. COMSOL Multiphysics (version 5.4) has been previously validated for two phase flow applications (*Qaddah et al.* (2019, 2020)).

- ⁸⁶¹ To compute our simulations we used the Heat Transfer (https://www.comsol.com/heat-
- transfer-module) and Computational Flow Dynamics (https://www.comsol.com/cfd-
- module) modules in addition to the main Multiphysics platform (https://www.comsol.com/comsol-

multiphysics). All the parameters used in our simulation are listed and described
in the manuscript. The open source software used for data visualisation was
Xmgrace (https://plasma-gate.weizmann.ac.il/Grace/). Data obtained can be
dowloaded on the following link: https://drive.uca.fr/d/2f9684aae96c4947b3cf/

⁸⁶⁸ Code availability

The software used for this study is the commercial software COMSOL Multiphysics (version 5.4) previously validated for two phase flow applications (*Qaddah et al.* (2019, 2020)). A trial version of COMSOL Multiphysics may be requested (see https://www.comsol.com).

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Figure 1: Nusselt number as a function of the Rayleigh number during the phase the first phases where only the thermal convection is considered (no settling of particles). Results from our models are represented with black circles. For comparison, we plot the scaling laws proposed by *Wolstencroft et al.* (2009) and *Turcotte and Schubert* (1982) with blue and green dashed lines respectively. The red dashed line represents the result from a power-law fitting of our numerical results. Corresponding scaling laws are detailed in the legend of the figure.

Figure 2: Schematic representation of the computational domain of our study. Blue material represents the continuous phase (molten silicates) and green material represent the initially mixed continuous and dispersed phases. A close-up view is illustrated in the right panel.

Figure 3: Dispersed fraction averaged along an horizontal profile at the bottom of the computational domain as a function of the mesh size. In these models, h = H/10, $\phi_{d,0} = 0.5$, $\eta_c = 10$ Pa.s and $\Delta \rho = 30$ kg.m⁻³. The red dashed line represents the asymptotic value at which $\phi_{d,average}$ is converging for high-resolutions (i.e. small mesh size) models.

Figure 4: Time evolution (from top to bottom) of the dispersed volume concentration (left) and the temperature (right) for the reference case (with $Ra = 4.4 \times 10^6$ (corresponding to $\eta_c = 4$ Pa.s), $\Delta \rho = 50$ kg.m⁻³ and $D_d = 10^{-3}$ m).

Figure 5: Time evolution of the particles concentration in the upper part of the reservoir with $Ra = 4.4 \times 10^6$ (corresponding to $\eta_c = 4$ Pa.s) and $D_d = 10^{-3}$ m. The red line illustrates the case where $\Delta \rho = 50$ kg.m⁻³ and the green line the case where $\Delta \rho = 100$ kg.m⁻³. Vertical dashed lines illustrate the time when the minimum value of e is obtained, which is referred in the text as the overturn time.

Figure 6: Time evolution (from top to bottom) of the dispersed volume concentration (left) and the temperature (right) for the case with $Ra = 4.4 \times 10^6$ (corresponding to $\eta_c = 4$ Pa.s), $\Delta \rho = 100$ kg.m⁻³ and $D_d = 10^{-3}$ m.

Figure 7: Time evolution of the particles concentration in the upper part of the reservoir with $Ra = 4.4 \times 10^6$ (corresponding to $\eta_c = 4$ Pa.s) and $\Delta \rho = 50$ kg.m⁻³. The red line illustrates the case with $D_d = 10^{-3}$ m, the green line the case with $D_d = 10^{-4}$ m and the blue line the case with $D_d = 10^{-2}$ m. Vertical dashed lines illustrate the time when the minimum value of e is obtained, which is referred in the text as the overturn time.

Figure 8: Time evolution of the particles concentration in the upper part of the reservoir with $\Delta \rho = 50$ kg.m⁻³ and $D_d = 10^{-3}$ m. The different color lines illustrate with cases with different viscosities of the continuous phase ranging from 0.1 to 10 Pa.s. Vertical dashed lines illustrate the time when the minimum value of e is obtained, which is referred in the text as the overturn time. The corresponding values of the overturn times are plotted in Fig. 9. We note we have tested the case with $\eta_c = 0.1$ Pa.s with two different meshes (a 6 and 8 mm mesh) and the difference between the two curves is not significative.

Figure 9: Overturn timescale as a function of the molten phase viscosity. Black symbols illustrate the results from our numerical models. The dashes lines show the overturn timescale obtained from Eq. 15 (with 1 to 100 $t_{overturn}$). We note we have tested the case with $\eta_c = 0.4$ Pa.s with two different meshes (a 4 and 8 mm mesh) and the difference between the two timescales is not significative.

Figure 10: Shields number value (Sh) as a function of the molten continuous phase viscosity (η_c) (or Ra for the top x-axis). Square symbols correspond to the models run in this study (see Tab. 2 for values). Empty black symbols represent cases where the suspension regime is reached. Filled green symbols represent cases where the sedimentation regime is reached. The two regimes are separated by dashed lines (green for $\eta_c \leq 1$, blue for $\eta_c \geq 1$) illustrating the critical Shields number (Sh_c) . Red symbols and dotted lines illustrate the values for the cases represented in Fig. 5, Fig. 7 and Fig. 8 respectively. We calculate the Sh values considering the combination of Eq. 12, Eq. 13 and the expression $Nu = 0.14Ra^{0.34}$ obtained from Fig. 1 to compute the heat flow F within Eq. 16.

Figure 11: Temperature profiles corresponding to (grey thick line) the solidus of the pyrolite mantle (*Pierru et al.*, subm.) and (colored lines) adiabatic gradients of MO for potential surface temperature from 1800 to 2500 K.

Figure 12: Solid-melt density difference $(\Delta \rho_{MO} = \rho_{Bg} - \rho_{melt})$ calculated using the P-V-T equation of state of Bg (*Matas et al.*, 2007) and MO (*Asimow*, 2018), after decomposition in end-members. Calculations are performed for several MO compositions by adding or subtracting a fraction of Bg to a pyrolite-based mantle reference, and for Fe partition coefficient between Bg and MO ($K(Fe)^{Bg/Melt}$) of 0.1 or 0.25. Positive value indicates lighter melt.

Figure 13: Density contrast ratio $\Delta \rho_{MO} / \Delta \rho_{crit}$ in the MO. Values larger than 1, or smaller than -1, indicate efficient Bg segregation below, or above, a mushy MO, respectively.

Figure 14: Density contrast ratio $\Delta \rho_{MO} / \Delta \rho_{crit}$ in the MO calculated as a function of the Bg grain size, from 0.1 to 5 mm, for 5 different melt compositions, from pyrolite -40%Bg to pyrolite +40%Bg. Values larger than 1 or lower than -1 indicate efficient Bg segregation below, or above, the mushy MO, respectively. $\chi_{\rho} = 0$ is found for $\Delta \rho_{MO} = 0$ (Fig. 12). It indicates infinite value of *Sh* and perfect crystal suspension (Fig. 10). Continuous and dashed lines indicate $K(Fe)^{Bg/Melt}$ of 0.1 or 0.25, respectively.

Figure 15: Density contrast ratio $\Delta \rho_{MO} / \Delta \rho_{crit}$ calculated for various surface temperatures (Eq. 19). Thick, intermediate and thin lines correspond to 500, 1000 and 1500 K, respectively. The calculations are also performed for various mantle compositions and $K(Fe)^{Bg/Melt}$ values.

Figure 16: Chemical profiles obtained in the case of no Bg segregation from the melt. The chemical gradient comes from the solidification of $\sim 60\%$ of Bg and 40% of the melt at the bottom of the MO. These profiles assume no mantle mixing once the mantle layers are eventually solidified. "BSE" and "s-MO" are bulk silicate Earth and a shallow MO extending up to 660 km depth, respectively. Here, the BSE composition is adjusted to 80% pyrolite and 20% of Bg.

| | Symbol | Value or range |
|-------------------------------|---------------------------------|--|
| Magma ocean density | $ ho_{c,0}$ | $3000 \mathrm{~kg/m^3}$ |
| Crystals density | $ ho_d$ | $\rho_{c,0} + \Delta \rho$ |
| Density difference | $\Delta \rho = \rho_d - \rho_c$ | $5-100 \text{ kg/m}^3$ |
| Magma ocean viscosity | η_c | 0.1- 10 Pa.s |
| Crystal diameter | D_d | 10^{-4} - 10^{-2} m |
| Heat capacity | C_p | $1000 \text{ J.kg}^{-1}.\text{K}^{-1}$ |
| Thermal conductivity | k | $5 \text{ W.m}^{-1}.\text{K}^{-1}$ |
| Thermal diffusivity | $\kappa = k / (\rho_{c,0} C_p)$ | $1.7 \times 10^{-6} \text{ m}^2.\text{s}^{-1}$ |
| Thermal expansion coefficient | α | 10^{-5} K^{-1} |
| Reference temperature | T_0 | 4000 K |
| Temperature difference | ΔT | 100 K |

Table 1: Symbol definitions and values of the physical parameters used in this study.

| | $\eta_c(Pa.s)$ | Ra | D_d (m) | $\Delta \rho ~(\mathrm{kg/m^3})$ | Sh |
|-----|----------------|--------------------|-----------|----------------------------------|-------|
| #1 | 0.1 | 1.8×10^8 | 10^{-4} | 5 | 4.16 |
| #2 | 0.1 | 1.8×10^8 | 10^{-4} | 10 | 2.08 |
| #3 | 0.1 | 1.8×10^8 | 10^{-4} | 20 | 1.04 |
| #4 | 0.1 | 1.8×10^8 | 10^{-4} | 30 | 0.69 |
| #5 | 0.1 | 1.8×10^8 | 10^{-4} | 40 | 0.52 |
| #6 | 0.1 | $1.8 	imes 10^8$ | 10^{-4} | 50 | 0.42 |
| #7 | 0.1 | $1.8 	imes 10^8$ | 10^{-3} | 5 | 0.41 |
| #8 | 0.1 | $1.8 	imes 10^8$ | 10^{-3} | 10 | 0.2 |
| #9 | 0.1 | 1.8×10^8 | 10^{-3} | 20 | 0.1 |
| #10 | 0.1 | 1.8×10^8 | 10^{-3} | 30 | 0.07 |
| #11 | 0.1 | 1.8×10^8 | 10^{-3} | 40 | 0.052 |
| #12 | 0.1 | 1.8×10^8 | 10^{-3} | 50 | 0.042 |
| #13 | 0.1 | 1.8×10^8 | 10^{-3} | 80 | 0.026 |
| #14 | 0.1 | $1.8 	imes 10^8$ | 10^{-3} | 100 | 0.02 |
| #15 | 0.1 | $1.8 	imes 10^8$ | 10^{-2} | 5 | 0.041 |
| #16 | 0.1 | $1.8 	imes 10^8$ | 10^{-2} | 10 | 0.02 |
| #17 | 0.1 | 1.8×10^8 | 10^{-2} | 20 | 0.01 |
| #18 | 0.1 | 1.8×10^8 | 10^{-2} | 30 | 0.007 |
| #19 | 0.2 | $9. \times 10^{7}$ | 10^{-3} | 50 | 0.052 |
| #20 | 0.2 | $9. \times 10^7$ | 10^{-3} | 60 | 0.044 |
| #21 | 0.4 | 4.4×10^7 | 10^{-3} | 5 | 0.65 |
| #22 | 0.4 | 4.4×10^7 | 10^{-3} | 10 | 0.33 |
| #23 | 0.4 | 4.4×10^7 | 10^{-3} | 20 | 0.16 |
| #24 | 0.4 | $4.4 	imes 10^7$ | 10^{-3} | 30 | 0.11 |
| #25 | 0.4 | $4.4 	imes 10^7$ | 10^{-3} | 40 | 0.082 |
| #26 | 0.4 | $4.4 	imes 10^7$ | 10^{-3} | 50 | 0.065 |
| #27 | 0.7 | $3. \times 10^{7}$ | 10^{-3} | 40 | 0.1 |
| #28 | 0.7 | $3. \times 10^7$ | 10^{-3} | 50 | 0.081 |
| #29 | 1. | 1.8×10^7 | 10^{-4} | 5 | 8.9 |
| #30 | 1. | 1.8×10^7 | 10^{-4} | 10 | 4.45 |
| #31 | 1. | 1.8×10^7 | 10^{-4} | 20 | 2.22 |
| #32 | 1. | 1.8×10^7 | 10^{-4} | 30 | 1.48 |
| #33 | 1. | $1.8 	imes 10^7$ | 10^{-4} | 40 | 1.1 |
| #34 | 1. | $1.8 	imes 10^7$ | 10^{-4} | 50 | 0.089 |
| #35 | 1. | $1.8 	imes 10^7$ | 10^{-3} | 10 | 0.44 |
| #36 | 1. | $1.8 	imes 10^7$ | 10^{-3} | 20 | 0.22 |
| #37 | 1. | $1.8 	imes 10^7$ | 10^{-3} | 30 | 0.148 |

Table 2: Parameters for all performed simulations used in this study. We calculate the Sh value from Eq. 16 considering the combination of Eq. 12, Eq. 13 and the expression $Nu = 0.14 Ra^{0.34}$ obtained from Fig. 1 to compute the heat flow F.

| | $\eta_c(Pa.s)$ | Ra | D_d (m) | $\Delta \rho ~(\mathrm{kg/m^3})$ | Sh |
|-----|----------------|-------------------|-----------|----------------------------------|--------|
| #38 | 1. | $1.8 	imes 10^7$ | 10^{-3} | 35 | 0.127 |
| #39 | 1. | $1.8 	imes 10^7$ | 10^{-3} | 40 | 0.111 |
| #40 | 1. | $1.8 	imes 10^7$ | 10^{-3} | 50 | 0.09 |
| #41 | 1. | $1.8 	imes 10^7$ | 10^{-3} | 100 | 0.044 |
| #42 | 1. | $1.8 	imes 10^7$ | 10^{-2} | 5 | 0.089 |
| #43 | 1. | $1.8 	imes 10^7$ | 10^{-2} | 10 | 0.044 |
| #44 | 1. | $1.8 	imes 10^7$ | 10^{-2} | 20 | 0.022 |
| #45 | 1. | $1.8 	imes 10^7$ | 10^{-2} | 30 | 0.015 |
| #46 | 4. | $4.4 	imes 10^6$ | 10^{-4} | 50 | 1.4 |
| #47 | 4. | $4.4 	imes 10^6$ | 10^{-3} | 50 | 0.14 |
| #48 | 4. | $4.4 	imes 10^6$ | 10^{-3} | 70 | 0.1 |
| #49 | 4. | $4.4 	imes 10^6$ | 10^{-3} | 80 | 0.0875 |
| #50 | 4. | $4.4 	imes 10^6$ | 10^{-3} | 100 | 0.07 |
| #51 | 4. | 4.4×10^6 | 10^{-2} | 50 | 0.014 |
| #52 | 10 | 1.8×10^6 | 10^{-4} | 5 | 19 |
| #53 | 10 | $1.8 	imes 10^6$ | 10^{-4} | 10 | 9.51 |
| #54 | 10 | $1.8 	imes 10^6$ | 10^{-4} | 20 | 4.76 |
| #55 | 10 | $1.8 	imes 10^6$ | 10^{-4} | 30 | 3.17 |
| #56 | 10 | $1.8 	imes 10^6$ | 10^{-4} | 40 | 2.37 |
| #57 | 10 | $1.8 	imes 10^6$ | 10^{-4} | 50 | 1.9 |
| #58 | 10 | $1.8 	imes 10^6$ | 10^{-4} | 100 | 0.95 |
| #59 | 10 | $1.8 	imes 10^6$ | 10^{-3} | 5 | 1.9 |
| #60 | 10 | $1.8 	imes 10^6$ | 10^{-3} | 10 | 0.95 |
| #61 | 10 | $1.8 	imes 10^6$ | 10^{-3} | 20 | 0.47 |
| #62 | 10 | $1.8 	imes 10^6$ | 10^{-3} | 30 | 0.31 |
| #63 | 10 | $1.8 	imes 10^6$ | 10^{-3} | 40 | 0.24 |
| #64 | 10 | $1.8 	imes 10^6$ | 10^{-3} | 50 | 0.19 |
| #65 | 10 | $1.8 	imes 10^6$ | 10^{-3} | 100 | 0.095 |
| #66 | 10 | $1.8 	imes 10^6$ | 10^{-2} | 5 | 0.19 |
| #67 | 10 | 1.8×10^6 | 10^{-2} | 10 | 0.095 |
| #68 | 10 | $1.8 	imes 10^6$ | 10^{-2} | 15 | 0.063 |
| #69 | 10 | $1.8 	imes 10^6$ | 10^{-2} | 20 | 0.047 |
| #70 | 10 | $1.8 	imes 10^6$ | 10^{-2} | 30 | 0.032 |
| #71 | 10 | $1.8 	imes 10^6$ | 10^{-2} | 50 | 0.019 |
| #72 | 10 | $1.8 	imes 10^6$ | 10^{-2} | 100 | 0.0095 |

Table 2: Part 2

Figure 1.



Figure 2.



Figure 3.



Figure 4.





m

Figure 5.



Figure 6.





Figure 7.



Figure 8.



Figure 9.



Figure 10.



Figure 11.



Figure 12.


Figure 13.



 $\Delta \rho_{MO} \ I \ \Delta \rho_{crit}$

Figure 14.



Figure 15.



Figure 16.

