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METHODOLOGICAL NOTES

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On estimating the molecular viscosity of the Earth's outer core: comment on the paper by D E Smylie et al.

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Abstract. **The paper "Direct observations of the viscosity of Earth's outer core and extrapolation of measurements of the viscosity of liquid iron" by D E Smylie, V V Brazhkin, and A Palmer [*Phys. Usp.* 52 (1) 79 (2009)] is subject to critique for its proposed approach to estimating the viscosity of the Earth's outer core.**

1. In Ref. [1], the distribution of molecular viscosity over an outer liquid core of the Earth is estimated via extrapolation by starting with the authors' formula (45):

$$\eta \sim \exp \frac{E_{\text{act}_0} + PV_{\text{act}}}{kT},$$

where E_{act_0} is the activation energy at normal pressure, P is the pressure, V_{act} is the activation volume, k is the Boltzmann constant, and T is the absolute temperature.[1] In fact, the numerator in this expression should be the activation enthalpy $H^* = E^* + PV^*$, with both E^* and V^* being density- and temperature-dependent. It is clear that by setting E_{act_0} in Eqn (45) to a constant, it is an easy matter to obtain an exponential increase in viscosity with pressure — all the more so since, to quote from Ref. [1], "While the activation volume for liquid metals at atmospheric pressure is very small ..., it was supposed ... that, similar to the behavior of molecular melts, the activation volume in liquid metals increases under compression from $0.05\,V_{\text{at}}$ to $(0.2-0.4)\,V_{\text{at}}$ at pressures of ≈ 10 GPa and can increase further to $(0.5-1)\,V_{\text{at}}$ with a subsequent pressure rise to 100 GPa," (V_{at} being the atomic volume). For me, there is little or no justification for the transformations which the authors of Ref. [1] further perform on Eqn (45) and which lead them to a fantastic increase in the *molecular* viscosity in the outer liquid core of the Earth and to a very large molecular viscosity in the core near its boundary with the mantle.

2. In Ref. [2], the physics of the Earth's polycrystalline mantle was addressed by introducing the concept of diffusion viscosity and relating the flow of a polycrystal to thermal point defects in the form of vacancies. The vacancy formation enthalpy h has the same form as in Eqn (45), namely, $h = e + v_{\text{d}}P$, where e is the vacancy formation energy, and v_{d} is the activation volume equal to the change in the crystal volume due to the formation of a vacancy. The resulting problem was conceptually similar to that considered in Ref. [1] based on formula (45). At first sight it seemed that, because of the term $v_{\text{d}}P$, increasing pressure will rapidly increase the vacancy formation enthalpy, markedly decrease the vacancy concentration (the vacancies 'collapse'), and make the diffusion and self-diffusion coefficients tend to zero, thus sharply increasing the diffusion viscosity of the Earth's polycrystalline mantle. To clear the matter up was expressly the aim of the paper "Pressure effect on the diffusion coefficient in solids" [3]. In that paper, two problems were addressed.

At the time of the paper cited, the interaction potential between the Cl^- and Na^+ ions was well known, and the Mott – Littleton method [4] allowed a strict, indeed *ab initio*, calculation to be performed for NaCl to determine the formation enthalpy of the Schottky defect, a defect in which a vacancy in the lattice of Cl^- ions and a vacancy in the lattice of Na^+ ions form. It turned out that, contrary to what would at first sight be expected, increasing pressure does not destroy the Schottky defect. The reason for this is the large gain in overlap energy due to the formation of a Schottky type defect. The formation of a vacancy results in the disappearance of the repulsive interaction between a particle which occupied a lattice site and its neighbors. It is clear that qualitatively the same effect should occur in liquids.

The way in which the diffusion viscosity coefficient varies with temperature and pressure is determined by the self-diffusion coefficient

$$D = D_0 \exp\left\{-\frac{H^*(P)}{kT}\right\}. \tag{C1}$$

Because it was revealed empirically [2, 3, 5] that the enthalpy of activation $H^*(P)$ is proportional to the melting temperature $T_{\text{m}}(P)$, viz.

$$H^*(P) \sim T_{\text{m}}(P), \tag{C2}$$

the dependences of the coefficients D and η on pressure can be expressed in terms of the reduced temperature

$$\theta = \frac{T}{T_{\text{m}}}. \tag{C3}$$

It is then clear that both the self-diffusion coefficient and the diffusion viscosity are approximately constant along the

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[1] Henceforth, each numbered formula in this comment will have the letter C appended in front of the number.

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melting curve — a fact which is widely used when evaluating dependences in the physics of the Earth's interior.

3. The second question addressed in Refs [2, 5] was that of the derivation of simple interpolation formulas for estimating the effect of pressure on the self-diffusion and viscosity coefficients. For this purpose, the pressure dependences of the quantities of interest here can be written down [taking $H^*(P)$ as an example] in the form

$$H^* = H_0^* \exp\left\{-\int_{P_0}^{P} \frac{\partial \ln H^*}{\partial \ln V}\, \chi_{\mathrm{T}}\, \mathrm{d}P\right\}, \tag{C4}$$

where V is the (fairly) slowly varying volume, $\partial \ln H^*/\partial \ln V < 0$, and χ_{T} is the isothermal compressibility as determined from the equation of state or experimentally. For simple substances with exponential repulsion forces, the quantity $L = |\partial \ln H^*/\partial \ln V|$ at low pressures amounts to $\sim 3-2$ and decreases slowly with increasing pressure. (In this sense it is similar to the Grüneisen parameter, another well-known logarithmic derivative.) If the quantity L in formula (C4) is then replaced by its normal pressure value L_0, we arrive at the simple expression

$$H^* = H^*(\rho_0)\left(\frac{\rho}{\rho_0}\right)^{L_0} \tag{C5}$$

for estimating upper bounds, where ρ is the density, and ρ_0 is the density at normal pressure.

4. Let us utilize formula (C5) to estimate how the viscosity of liquid iron changes along the melting curve as we pass from normal pressure to that at the outer liquid core–mantle boundary. For this purpose we return to Eqn (45) of Ref. [1] and write down its analogue in the form

$$\eta \sim \exp\left\{\frac{H^*(\rho_0)}{kT_{\mathrm{m}_0}}\, N(\rho, T)\right\}, \tag{C6}$$

$$N(\rho, T) = \frac{T_{\mathrm{m}_0}}{T}\left(\frac{\rho}{\rho_0}\right)^{L_0}, \tag{C7}$$

where T_{m_0} and ρ_0 are the melting temperature and density of melted iron at normal pressure, while T and ρ are the temperature and density in the liquid core at the boundary with the mantle. Setting $T = T_{\mathrm{m}_0}$ and $\rho = \rho_0$ in formulas (C6) and (C7) corresponds to estimating the *molecular* viscosity, in our case that of iron at normal pressure. For $N > 1$, the viscosity along the melting curve increases, while for $N < 1$ it decreases.

Poirier is quite right in his book [6] when arguing that in solids the activation energy for diffusion is essentially the energy of formation of point defects, whereas in liquids (at normal pressure) the apparent 'activation energy' reflects only the temperature dependence of the mobility of the atoms because local defects need not be generated in this case. As is known, melting involves a marked increase in volume. For example, iron increases in volume by ~ 3% as it melts. Based on treating a large amount of empirical data, Poirier concludes that for a number of metals the effective activation energy for viscosity, Q_v, and the melting temperature at normal pressure, T_{m_0}, are related by

$$Q_v = 2.6RT_{m_0}, \tag{C8}$$

where R is the molar gas constant.

As reviewed in Ref. [7], the viscosity of liquid iron at normal pressure varies in the range $\sim (5-7.6) \times 10^{-2}$ P and slowly decreases from 7.60 to 5.22×10^{-2} P on heating from 1536 °C to 1850 °C. From the same reference, solutions of iron with Ni, S, O, and Si possess nearly the same viscosity as pure liquid iron. Liquid iron at normal pressure has a density of 7.25 g cm^{-3} and a melting temperature of 1808 K. At the core–mantle boundary, the pressure, density, and melting temperature are, respectively, 1.35 Mbar, 9.86 g cm^{-3} [1], and 3429 K [1]. An important point to remember when estimating the viscosity of liquid iron is that due to the presence of light element impurities, under the conditions considered the density of pure liquid iron is greater by about 10 wt.% and amounts approximately to 11 g cm^{-3}. Setting $L_0 \sim 3$ in formula (C7) and taking into account the above data we obtain the estimate $N \sim 1.84$, which yields for the molecular viscosity at the core–mantle boundary the following value

$$\eta_{\mathrm{cmb}} < 7.6 \times 10^{-2} \times 12 \text{ P} \sim 0.9 \text{ P}, \tag{C9}$$

which is a higher upper bound than would occur for a pressure of ~ 1.35 Mbar.

From Ref. [1], the extrapolation estimates of molecular viscosity at the core–mantle boundary and at the boundary with the inner core are $\sim 10^3$ and $\sim 10^{12}$ P, respectively. The same formulas (C6) through (C8) readily yield the upper bound for the molecular viscosity of the outer liquid core at the boundary with the inner core — clearly making 10^{12} P out of the question. In my opinion, Ref. [1] does not in any way shatter the idea that the *molecular* viscosity of liquid metals is low and approximately constant along the melting curve.

5. The interpretation of geophysical data led the authors of Ref. [1] to a value of 2.37×10^4 P for the core viscosity at the boundary with the mantle, and to a value of 1.2×10^{12} P for that at the boundary with the inner core, the former based on the decay of free core nutations (FCN), and the latter on the reduction in the rotational splitting of two equatorial translational modes of oscillations of the inner core.

The question naturally arises as to whether the interpretation used in Ref. [1] is unique.

It should be emphasized that the electromagnetic interaction is present at the core–mantle boundary in addition to the viscous interaction; that the boundary itself has not yet been sufficiently studied, and that the question of the FCN excitation and decay regimes is still under study.

The conclusion in Ref. [1] that there is "the close confirmation of the Ca18 Earth model" (a model of the inner structure of the Earth) is questionable. As can be seen from the parameter distribution for the model Ca18 as given in the well-known book [8], the parameters near the inner core boundary and in the core itself differ markedly from the parameters of the reference models PREM [9] and ak 135 [10] which are currently in use. In the inner core, the parameters of the two models are essentially the same. In the Ca18 model, the jump in density at the boundary between the outer liquid core and the inner solid core amounts to 1.17 g cm^{-3}, whereas for PREM and ak 135 models its respective values are 0.6 g cm^{-3} and 0.665 g cm^{-3}. The density jump at the outer/inner core boundary in Ca18 is ~ 8.8%, compared to ~ 3% for iron melting at normal pressure. In Ref. [11], the modes ${}_2S_3$, ${}_3S_2$, ${}_6S_3$, ${}_8S_5$, ${}_9S_3$, and 13 other overtone modes markedly dependent on the inner core parameters were studied in the spectrum of free oscillations of the Earth. In particular, the density jump at the inner core boundary was determined and found to be ~ 0.55 g cm^{-3}. A recent, more detailed study of the inner core properties using the method of free oscillations of the Earth [12] showed that the density range of 12.76 – 13.09 g cm^{-3} the PREM model predicts for the inner core is a good approximation for the density distribution. The corresponding range for the Ca18 model, 13.34 – 13.58 g cm^{-3}, lies considerably higher than in the PREM model. Table 4 of Ref. [1] presents the following values of rotation-unsplit periods T_0 (in hours): 3.7985 from observations; 3.82470 for Ca18, and 5.23826 for PREM. The authors of Ref. [1] note themselves that the period T_0 is strongly dependent on the density of the inner core.

That the Ca18 model with its unrealistically high inner core densities is consistent with the observed T_0 value, whereas the more substantiated models PREM and ak 135 are essentially not, is, in my view, due to the large error in the observed value of T_0 or, alternatively, the consistency of the Ca18-predicted and observed T_0 values is achieved due to the unrealistically high viscosity of the outer liquid core at the boundary with the inner core. In short, then, a physical — rather than a mathematical — inaccuracy must have been introduced at some point in the research.

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