

Comparative Simulation Study of Vacancy Diffusion in MgO and MgSiO₃ in Very High Pressures

Project Representative

Mitsuhiro Toriumi Japan Agency for Marine–Earth Science and Technology/ Graduate School of Frontier Science,
University of Tokyo

Authors

Mitsuhiro Toriumi Japan Agency for Marine–Earth Science and Technology/ Graduate School of Frontier Science,
University of Tokyo

Yosuke Ito Department of Sciences, University of Tokyo

MD simulations with pairwise potential on MgO and MgSiO₃ vacancies pair were conducted to obtain the Newtonian viscosity of lower mantle. The results of MgO are such that the viscosity of the upper half portion in the lower mantle shows uniform structure but in the lower half it rapidly decreases to the core mantle boundary by two order of magnitude. On the other hand, the viscosity of MgSiO₃ perovskite decreases gradually with increasing pressure in the whole lower mantle conditions.

To clarify mineral controlling the rheology of the deep lower mantle, we compare the diffusion coefficients of slowest atoms of MgO and MgSiO₃ in the lower mantle conditions. The results indicate that the viscosity of the whole lower mantle is governed by lattice diffusion of MgO. Thus, the actual lower mantle rheology should also show the viscosity lowering in the deepest lower mantle.

Keywords: vacancy diffusion, MgO, MgSiO₃, lower mantle, pairwise potential

1. Introduction

The aim of this research program is to obtain the mechanical behavior of earth's materials in the conditions ranging from shallow crust to deep mantle by means of the following simulations studies; (1) dislocation and vacancy structure and dynamics in MgO and MgSiO₃ in the lower mantle conditions using molecular dynamics method of several different type pairwise potentials, (2) water structure and the physical properties in earth's materials using ab initio simulation, (3) shear zone structure and dynamics involving mineral reaction, water migration, localization, crack formation using granular system simulation. This year, we intend to perform long term time studies of vacancy diffusion both in MgO and MgSiO₃ in very high pressure conditions for direct comparison of the Newtonian viscosity of them in the lower mantle.

2. Results

We investigated pair vacancies diffusion of SiO₂ by means of the molecular dynamic simulation of MgSiO₃ perovskite. After introduction of Si and 2O vacancies in MgSiO₃ perfect crystal, they move by random walk from the initial positions. The mean square distance of this random walk of the Si vacancy is equal to the temperature compensated diffusivity of MgSiO₃ because of slowest atom. The size of the basic cell reaches two thousands atoms. This type

of simulation required in the estimation of very high pressure viscosity of these solids is to consider the number density of vacancy, because equilibrium density depending on applied stress is very small at low stress conditions. To conduct the simulation, the relaxation time is needed to be several nanoseconds. Even using the ES, it is impossible to conduct the precise simulation of vacancy random walk for the relaxation process of vacancy migration need enough time.

Thus, the author (Ito) made a new algorithm utilizing table update method of near atomic position and velocities. As a result the n square calculation changes down to the n^{-1.5} power calculation, so that the times required for the 20 nanosecond relaxation experiments are possible in the evaluation of the Newtonian viscosity. In addition, the time studies for relaxation of vacancy are conducted and then it is concluded that the 20 ns experiments show enough narrow range of errors in diffusivity compared with 4 ns experiments. The potential parameters of MgSiO₃ used here are listed in Fig. 1 cited from Oganov et al. (2000).

The results of Si vacancy diffusion are shown in Fig. 2, indicating that the diffusion coefficients at 5000 K decrease from 60 to 120 Gpa. This means the increase of Newtonian viscosity governed by the vacancy diffusion. The increase of diffusion coefficients with increasing temperature at the constant pressure (80 Gpa) is shown as Fig. 3. These results

$$U_{ij} = \frac{Z_i Z_j e^2}{r_{ij}} + f_0 (b_i + b_j) \exp\left(\frac{a_i + a_j - r_{ij}}{b_i + b_j}\right) - \frac{c_i c_j}{r^6}$$

	z(charge)	a(Å)	b(Å)	c/[(KJ/mol) ^{1/2} Å ³]
Si	2.904	1.620	0.145	0.000
O	-1.605	1.614	0.138	36.532
Mg	1.910	1.635	0.149	0.000

MgSiO₃: Oganov et al. [2000]

Fig. 1 Parameters of pairwise potential in perovskite after Oganov et al.(2000).

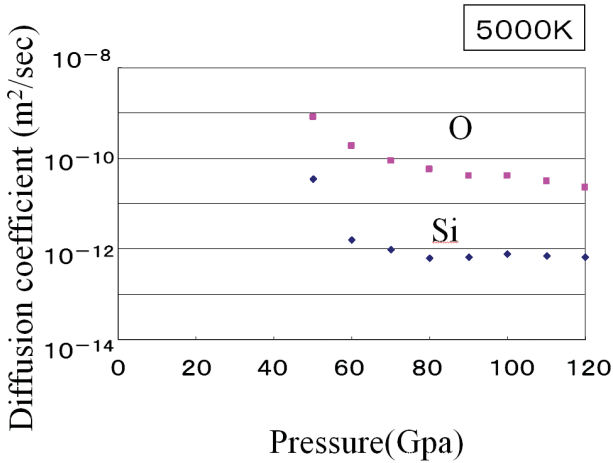


Fig. 2 Diffusion coefficients as a function of pressure of perovskite at 5000 K.

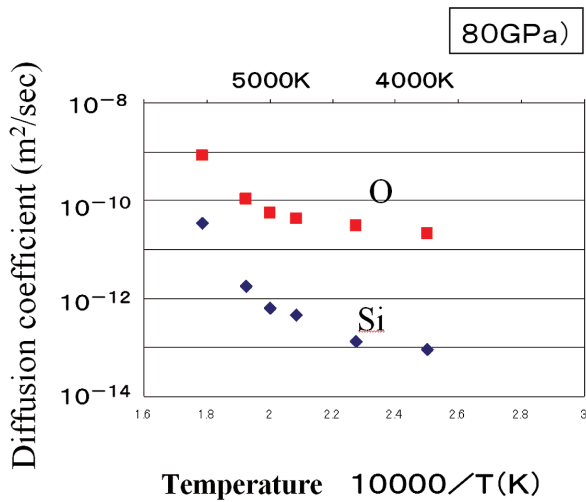


Fig. 3 Diffusion coefficients as a function of temperature of perovskite at 80 GPa.

indicate sharply that the activation volume of the vacancy diffusion in perovskite crystals decreases gradually in lower mantle conditions, but it does not change from positive to negative value.

The strain rate and stress relation is commonly called as the constitutive law, and it should be classified into the linear law and non-linear law. The latter is the power law creep in general. The linear type is Newtonian creep and representative model of the solid materials is the diffusion creep. This type

of creep is governed by the lattice diffusion along the paths of grain boundary or of volume. In this case, the flow law of the Nabarro-Herring creep can be formulated as follows;

$$d\epsilon/dt = AVD\sigma / RTd^2$$

where A = 13.3 and V = 1.8 × 10⁻⁵ m³/mol for SiO₂ vacancy in perovskite, and D and d are diffusion coefficient and grain size, respectively. In this case, the Newtonian viscosity of perovskite is controlled by the diffusion of Si vacancy as shown in the earlier sections.

3. Viscosity in the lower mantle

In order to evaluate the viscosity lowering of MgO in the deep mantle, the comparative study between the MgO (Ito and Toriumi, 2007) and MgSiO₃ perovskite (this study) will be carried out in the lower mantle pressure conditions. The ratio R of the viscosity between them holds as following relation;

$$R = (V_1/V_2) (D_1/D_2) (d_2/d_1)^2$$

in which 1 and 2 are MgO and perovskite, respectively. The value of V₁/V₂ is estimated from molecular volume ratio of them to be about 1.2 and the ratio of grain sizes is possibly assumed to be 1. Then, the difference of viscosity between these minerals is clear and that of MgO is smaller than MgSiO₃ by the magnitude of one to two orders.

On the other hand, the volume fractions of MgO in the lower mantle are considered to be the range from 0.3 to 0.4, judging from the upper mantle compositions (see Ito and Toriumi, 2007), and thus the rheology of the lower mantle should be governed by the MgO rheology. It concludes that the viscosity of the lower mantle should increase with increasing depth until 1500 km and then it should decrease rapidly with increasing depth to the core-mantle boundary. Consequently, we can conclude that viscosity lowering in the deep mantle predicted in the previous simulation studies is available for the real earth mantle as proposed in the previous paper (Ito and Toriumi 2007).

4. Conclusions

We conducted the MD simulations of vacancy pairs of MgO and MgSiO₃ perovskite in the lower mantle conditions. The results are such that the viscosity of perovskite is larger than that of MgO in the whole lower mantle conditions by the one to two order of magnitude. It indicates that the viscosity of the lower mantle is governed by that of MgO in the lower mantle and it decreases rapidly in lower half layer of the lower mantle by the order of magnitude.

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地球物質の数値計算科学的物性評価

プロジェクト責任者

鳥海 光弘 独立行政法人海洋研究開発機構・東京大学新領域創成科学研究科

著者

鳥海 光弘 独立行政法人海洋研究開発機構・東京大学新領域創成科学研究科

伊藤 洋介 東京大学地球惑星科学専攻

分子動力学法による格子拡散型の変形に対する直接計算は多体問題と精度の問題で長時間ステップの計算を必要とするために現在のところ地球シミュレータでしか実行可能ではない。下部マントルは MgSiO_3 と MgO との混合であるため、どちらがより小さい粘性率であるかによって下部マントルの粘性率が決まる。すでに MgO については著者らによって決定されたので(1)、ここでは MgSiO_3 について SiO_2 空孔の拡散係数を決定することによって流動則を比較しようと試みた。この計算には十分な精度の拡散係数を必要とするため、MD計算の時間を20ナノ秒まで行った。

計算結果は MgSiO_3 の拡散係数が MgO に比べて十分に遅く、粘性率に換算するとほぼ10分の1程度以下であることが判明した。したがって、下部マントルが MgO の組成を0.3から0.4と考えると、下部マントルの粘性率は MgO の粘性率に支配されることがはじめて明らかとなった。この結果、すでに発表されたように下部マントルの粘性率は1500kmから次第に減少し、2900km付近では1-2桁程度上部より低下していることが示された。

参考論文

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