High-pressure elasticity of alumina studied by first principles

WENHUI DUAN, BIJAYA B. KARKI, AND RENATA M. WENTZCOVITCH*

Department of Chemical Engineering and Materials Science, and Minnesota Supercomputing Institute, University of Minnesota, Minneapolis, Minnesota 55455, U.S.A.

ABSTRACT

We investigate by first principles the elastic behavior of Al₂O₃-alumina under pressure (up to 300 GPa) in the corundum and Rh₂O₃ (II) phase. The results are in excellent agreement with available low pressure (<1 GPa) experimental data. The anisotropy in elasticity for corundum decreases up to 50 GPa and then increases slowly with pressure whereas for the Rh₂O₃ (II) phase the anisotropy increases monotonically with compression. Strong shear wave anisotropy in the Rh₂O₃ (II) phase is found to be associated with the relatively small $c_{55}$ modulus, and its softening at high pressures. Unlike corundum, the directions of the fastest and slowest wave propagation, and the maximum polarization anisotropy of Rh₂O₃ (II) phase remain unchanged with pressure. At the corundum to Rh₂O₃ (II) phase transition pressure (78 GPa at 0 K), the anisotropy increases by more than 100% but the density and wave velocities increase only by 2%. The calculated (0 K) densities and wave velocities at lower mantle pressures are slightly larger (by 5%) than the corresponding seismic profiles. Our results suggest that the presence of free Al₂O₃ in small amounts in the lower mantle may not be detected in seismic density and velocity profile. However, its anisotropy may produce a detectable signal, particularly, at pressure conditions typical of the D" region.

INTRODUCTION

Al₂O₃ (alumina) is of great interest due to its diverse applications in high-pressure science. The pressure-dependence of the $R$ fluorescence lines of Cr³⁺-doped Al₂O₃-corundum (ruby) provides a convenient and accurate technique for pressure calibration in diamond-anvil cell experiments. It is also used as a window material in dynamic shock-wave measurements and is a useful ceramic material. Finally, free Al₂O₃ may exist in the lower mantle in small amounts, making up about 3.6% (in mass) of the mineralogical composition of this region (Anderson 1989; Brown and Mussett 1993).

Recent X-ray diffraction experiment observed that ruby undergoes a transformation to the Rh₂O₃ (II) structure when heated to 1000 K and pressurized to ~100 GPa (Funamori and Jeanloz 1997). At room temperature, experiments have shown a wide stability field of corundum up to pressure of 180 GPa (Richet et al. 1988; Jephcoat et al. 1988). However, several theoretical investigations have suggested the possibility of structural transformation of corundum to Rh₂O₃ (II) phase at much lower pressure at zero temperature (Cynn et al. 1990; Marton and Cohen 1994; Thomson et al. 1996; Duan et al. 1998). The latest study of six alumina polymorphs (Duan et al. 1998), namely, corundum, Rh₂O₃ (II), $Pbnm$ perovskite, $R3c$-perovskite, A-type rare-earth sesquioxide and B-type rare-earth sesquioxide, confirmed previous prediction of two phase transitions: corundum $\rightarrow$ Rh₂O₃ (II) at 78 ± 4 GPa and Rh₂O₃ (II) $\rightarrow$ $Pbnm$ perovskite at 223 ± 10 GPa. The first one is in good agreement with the all electron linearized augmented plane wave (LAPW) value of 90 GPa (Marton and Cohen 1994).

Measurements of individual elastic constants of Al₂O₃ exist up to 1 GPa (Gieske and Barsch 1968) whereas their temperature dependences have been measured up to 1825 K (Goto et al. 1989). The pressure dependence of isotropic wave velocities have been studied up to 61.6 GPa by Zhang and Chopelas (1994) from side-band fluorescence measurements. Cohen (1987) has studied the pressure dependence of elastic constants of corundum using the potential-induced breathing (PIB) model. Because the PIB model makes some crucial approximations to the nature of bonding such as full ionization, spherical ions, and use of pairwise interactions, the high pressure elasticity should ultimately be reinvestigated by a first-principles method. This paper reports the elastic constants ($c_{ij}$) of two alumina polymorphs, corundum and Rh₂O₃ (II), as a function of pressure up to 300 GPa. Using the calculated elastic constants, we study the pressure dependence of the elastic anisotropy and wave velocities of the mineral and speculate on the geophysical implications of these results.

CALCULATED ELASTIC CONSTANTS

Full structural optimization is performed using first principles variable cell shape molecular dynamics (Wentzcovitch et al. 1993) within the local density approximation (LDA). Norm-conserving Troullier and Martins (1991) pseudopotentials are used with plane-wave energy cut-offs of 70 Ry for electronic wavefunctions. Brillouin Zone sampling is performed using spe-