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An atomistic study of the incorporation of noble gas in silicate minerals: Lattice incorporation

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Olivine is one of the most common elements in the Earth's upper mantle, and knowledge of its physical properties is very important for understanding rheological processes in our planet[1]. In recent years, the behaviour of noble gases has been of particular importance for the development of degassing models and understanding the dynamics of the mantle[2]. In this work, we use computer simulations based on *ab-initio* methods to gain insights into the formation of point defects and the incorporation of noble gases into forsterite (Mg_2SiO_4), an end member of olivine. We calculate the enthalpies of incorporation of noble gases both at pre-existing vacancies and at interstitial positions. Our calculations show that most of the structural changes that occur at high pressure involve the octahedral Mg units and that the formation energies of defects involving Mg and Si vacancies (Schottky and Frenkel pairs) increase more sharply with pressure than those defects related to O sites. We use these calculated enthalpies to estimate the total uptake of noble gases into the bulk crystal as a function of temperature and pressure. For He and Ne, our calculated solubility points to noble gas atoms occupying mainly interstitial sites, in agreement with previous experimental results. However, Ar is located at Mg sites, because of its larger size and the deformation it causes cause within the crystal lattice. Electron density profiles suggest that the incorporation mechanism is mainly driven by the size mismatch between the host and the guest atom and that polarisation effects arising from the atomic size or the presence of a charged vacancy are minimal and do not contribute significantly to the uptake. Our work allows us to produce an estimate of the equilibrium concentration of noble gases in forsterite if only lattice point defects are present. Finally, the differences between our results for Ar and recent experimental work suggest that other incorporation mechanisms such as adsorption at internal and external interfaces and voids and grain boundaries must play a major role in noble gas storage and solubility.

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