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Diffusion of noble gases in silica polymorphs at Earth's mantle conditions

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Constraining the diffusional behaviour of noble gases in minerals is important for many geochemical studies (e.g. thermochronometry, cycle of volatiles). SiO_2 is a major component and phase in the crust and mantle, and represents up to 20% of basaltic mantle compositions. Its simple composition makes it easier to model as a large system than other more complex silicate minerals, becoming a good proxy for the investigation of the diffusion of noble gases from crustal conditions to the core-mantle boundary.

This work quantifies the diffusion rate of He, Ne, Ar and Kr in five polymorphs of SiO_2 (quartz, coesite with tetrahedrally coordinated Si; stishovite, $CaCl_2$-type, α-PbO_2-type with octahedrally coordinated Si). Diffusion coefficients are computed by long *ab initio* molecular dynamics runs using the VASP software. Diffusion rates are extracted from the mean square displacements. The activation energies of rare events (e.g. He and Ne diffusion in the α-PbO_2-type phase) are estimated using the nudged elastic band approach.

The diffusion is characterized by a sequence of hops between interstitial crystallographic sites. The simulations are sufficiently long and large to ensure that enough statistics are recorded. Results show that the diffusion rates and paths of noble gases depend on the structure of SiO_2, noble gas nature, temperature and pressure. The diffusion is anisotropic in almost all phases, especially when Si is octahedrally coordinated. Heavy noble gases seem to diffuse exclusively along the z-axis, whereas He and Ne diffuse along the x- and z-axes in quartz. The diffusion is often more isotropic in coesite. In the case of He, the denser the structure, the slower the diffusion. The diffusion rate globally decreases with increasing the size of noble gases. However, Ne may diffuse faster than He in denser phases, which is also shown by NEB calculations. At fixed composition, the transport of He and Ne is then very dependent on the local environment and the density of the crystal. From the core to the crust, the ratio between non-radiogenic noble gases may be impacted by SiO_2 phase transitions. Due to the abundance of SiO_2 phases, this behaviour is far from negligible.