Atomistic simulations of hydrogen diffusion and point defects in diamond

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One of the major unresolved questions in planetary sciences is how and when Earth acquired its water. A major obstacle in addressing this question is that Earth has significantly changed since its formation ca. 4.56 Ga because it is an active planet. To explain the origin of Earth's water, we need to determine the amount and isotopic composition of H (D/H ratio) in Earth's interior, as this is used as a benchmark to compare with solar system sources. Most importantly, we need to constrain whether Earth's D/H evolved through geological time (Ga timescales) to understand the incorporation of H_2O during the formation and evolution of Earth. Diamonds from the mantle may be key as they contain trace amounts of hydrogen and are inert and robust time capsules capable of surviving over several billion years.

While diffusivities of light elements such as N and C (self-diffusion) in diamonds are significantly slow (1), H diffusion studies show contrasting results. In several studies (2-4), H was shown to be virtually immobile. In contrast, a recent study (5) indicated that diamonds would equilibrate with ambient H present in the surrounding mantle comparatively fast relative to most geological processes. However, this recent model does not consider that the behaviour of H in diamonds may be influenced by the presence of inclusions or by the interaction of H with N and complex nitrogen-related defects observed in many natural diamonds. In addition, previous diffusion studies were conducted over a very narrow range of temperatures (T), and most importantly, not at mantle pressures (P), which could significantly affect the diffusion rate. As the current knowledge of H diffusion reported in the literature may not be representative of the diffusion rate of H in natural diamonds, it is fundamental to understand the behaviour of H in diamonds at realistic mantle P-T conditions while considering the effects of N and the interactions of different point and extended defects with H that likely impede its diffusion.

This project aims to improve our fundamental understanding of the dynamic and static behavior of hydrogen in mantle diamonds at realistic T and P conditions at which diamonds form. This Ph.D. project will study various chemical and structural factors that control how interstitial H diffuses through diamond. With the help of atomistic simulations, the rates and mechanisms of H diffusion can be evaluated as a function of mechanical, thermodynamical, and thermochemical conditions. Molecular dynamics simulations will be performed using ab initio methods and machine learning interatomic potentials. The results will allow us to not only correctly interpret novel H isotopic data necessary to pinpoint the origin of Earth's water but also to improve the fundamental understanding of the properties of diamonds.

The project will be funded by the ERC starting grant INHERIT (grant n. 101041620). For more information about the INHERIT project visit https://www.marthapamato.com/inherit. The thesis work will involve several long-term visits to the computational groups at the *Institut de Physique du Globe de Paris* (IPGP, Paris, France) and the *Center for Planetary Habitability* at the University of Oslo (Norway). Computing and programming skills/experience are highly recommended. Proficiency in written and spoken English is mandatory.

References

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