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A small atom with big implications: formation, evolution and preservation of hydrogen defects in natural diamond

MAXWELL C DAY[1], ETIENNE BALAN[2], RAZVAN CARACAS[3], MICHAEL C JOLLANDS[4], FRANCESCA INNOCENZI[1], DAVIDE NOVELLA[1], PROF. FABRIZIO NESTOLA[1] AND MARTHA G. PAMATO[1]

[1]University of Padova
[2]IMPMC CNRS/Sorbonne Université/MNHN
[3]Institut de Physique du Globe de Paris, Universite Paris Cite, CNRS
[4]Gemological Institute of America

Natural diamonds represent the only means by which deep mantle fluids can be directly sampled. During crystallization, impurities, particularly nitrogen (N) and hydrogen (H), are incorporated into diamond from the parental fluid or melt. Therefore, the H content and isotopic composition of diamonds may reflect primordial (or recycled) H_2O reservoirs from which diamond-forming fluids are sourced and may help to elucidate Earth's deep H cycle. However, interpreting such data from diamonds requires a better understanding of the degree to which H re-equilibrates during mantle residence and kimberlite magmatism. Once trapped by N/vacancy(*V*)-related defects, H cannot re-equilibrate (diffuse out of the diamond) and thus one must determine if/how N/H-defects form during the *initial stages* of diamond crystallization. Until now, only a few N/H-defects (e.g., VN_3H and VN_4H) have been rigorously identified that represent the ***end-point*** of N/H-aggregation and mantle residence. For this reason, initial N/H-defect formation and aggregation remain poorly understood.

To address this problem, the *Gemmological Institute of America* (GIA) database was filtered to find FTIR spectra of extremely rare, H-bearing natural diamonds that reflect N/H-defect aggregation states shortly after diamond formation. This resulted in the discovery of two new N/H-defects, the identities of which were determined using *ab initio* simulations and the first machine-learning molecular dynamics simulation. Trends in the intensity of peaks assigned to these new defects suggest they may be the first H-bearing point defects that form in diamond defining a ***starting-point*** in N/H-aggregation sequences. Moreover, new formation mechanisms are proposed for the most common H-defects in diamond (e.g., VN_3H). We investigate how the early trapping of interstitial hydrogen by substitutional N-defects affects re-equilibration of H during mantle residence. These findings have major implications for relating the H content and isotopic composition of diamond to their parental fluids and thus to H_2O reservoirs in the deep mantle. These findings also have important technological implications for the controlled production N/*V*-defects in synthetic diamond with useful electromagnetic properties, such as NV^-, that are passivated by H.

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