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Hydrogen content and isotopic composition in diamond by Secondary Ion Mass Spectrometry

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Numerous studies have aimed to quantify hydrogen content (H) in natural diamonds using various analytical techniques, including spectroscopy methods (FTIR), nuclear techniques (ERDA and PPS), and mass spectrometry techniques like Secondary Ion Mass Spectrometry (SIMS and NanoSIMS). Each of these methods, when used independently, has proven effective in detecting hydrogen in natural diamonds. However, comparing results from different techniques applied to the same sample often reveals discrepancies, which are frequently attributed to the presence of optically inactive hydrogen, leading to an underestimation of hydrogen content determined by FTIR.

In this presentation, we describe analytical developments of H-content and isotope composition measurements using large geometry secondary ion mass spectrometry (LG-SIMS) at the Nancy National facilities. By studying a compositionally and morphologically diverse set of lithospheric and sub-lithospheric diamonds (gem quality, cuboid, octahedral, and mixed habit morphologies, n = 20) from different geographic regions—we demonstrate that H-contents obtained by LG-SIMS qualitatively correlate with the intensity of C-H stretching peaks due to H-related defects observed in the IR. Our results suggest that IR-inactive hydrogen does not constitute a major proportion of H in diamonds. Inverse correlation between the N- and H-content and the lack of correlation between the N-aggregation state and H-content, indicate that N is neither the limiting factor nor the primary driver of H incorporation. This is confirmed by additional LG-SIMS measurements of nitrogen content and isotopic composition in the same samples. Finally, the preservation of spatial heterogeneities in H content in natural diamonds implies limited diffusion of H after its incorporation.