Evidence of Gas Phase Nucleation of Nano Diamond Through the Analysis of Activation Energy

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The mechanism of ballas like nano diamond formation still remains elusive, and this work attempts to analyze its formation in the framework of gaseous chemistry. Nano crystalline diamond (NCD) thin films were grown from H_2/CH_4 plasma in a 2.45 GHz chemical vapor deposition system.[1] The activation energy (E_a) is calculated from the Arrhenius equation corresponding to the thickness growth rate, using substrate temperature (~1000−1300 K) in the calculations. While the calculated values ~10 kcal/mol match with the E_a for nano diamond formation throughout the literature, they are lower compared with 13.6 to 22.5 kcal/mol [2, 3] for standard single crystal diamond (SCD) formation as shown in **Fig. 1**, concluding thus far, that the energetics and processes involved in their formation are different.

To further investigate this, substrate preparation and the deposition setup of collecting the samples directly in the plasma on seeded substrates was modified, while keeping the growth parameters constant. Un-seeded substrates were physically separated from the plasma by a metal stub and were used for sample collection through a pinhole in the stub. Raman spectra and Scanning Electron micrographs of these samples show signatures of nanodiamond similar to the in-plasma collected samples, revealing that nano diamond self nucleates in the plasma and falls to the substrate which acts as a mere collection plate. This means that the location of formation of nanodiamond particles is in the gas phase as opposed to on the substarte surface, which warrants a revisit to the E_a calculation, where we propose to replace the substrate temperature with gas temperature.

A basic model of H_2/CH_4 plasma in the reactor geometry is simulated to obtain the gas temperature **(Fig. 2)** profile across the reactor volume. E_a is are now re-calculated using the gas temperature (~ 2000 K), giving values closer to SCD formation. It suggests that the formation process for nano diamond and SCD could be the same, but nucleation happens in the gas phase for nano diamond and directly on the substrate for SCD.

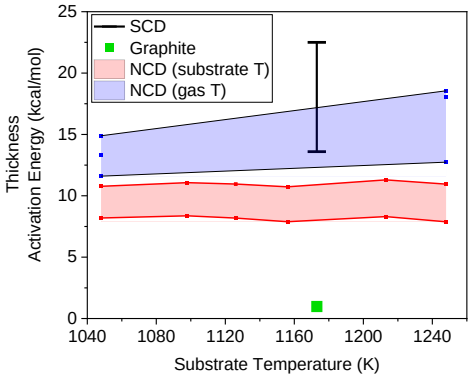

Fig. 1 Activation energies of NCD films calculated using substrate and gas temperature, compared to that of SCD and pure graphite [4].

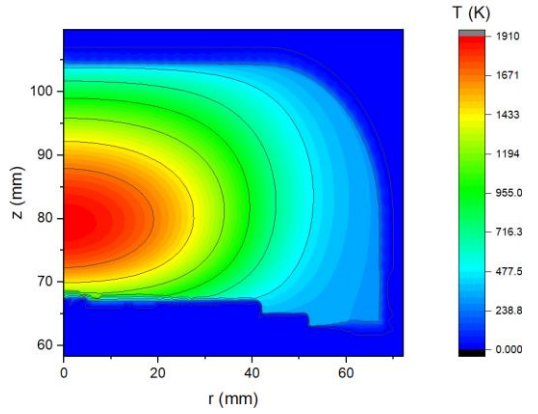

Fig. 2 Temperature profile simulated in an axi-symmetric reactor geometry for H_2/CH_4 plasma.

References

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