

posed based on C₂ insertion into dimer rows of the reconstructed (100) surface. It turns out that the C₂ particle density is ~10⁸ cm⁻³ even in CH₄-1%H₂-99% mixtures, so that C₂ could in principle serve as an embryonic nucleus in H₂-rich plasmas as well. However secondary nucleation in such mixtures is likely to be impeded by the fact that C₂ does not lead to nucleation on hydrogen-terminated (100) surfaces, as elucidated by ongoing theoretical calculations. In contrast the much higher nucleation rates of 10¹⁰ cm⁻² s⁻¹ accompanying nanocrystalline diamond growth in CH₄-1%Ar-99% mixtures are due to the increase in C₂ particle densities from 10⁸ to 10¹² cm⁻³ and the likelihood that in such plasmas the diamond surface is largely not terminated by hydrogen. The embryonic C₂ nuclei are no longer regasified efficiently by the low concentrations of atomic hydrogen present in the plasma and grow to critical size so as to form new diamond crystallites. The reduction in growth rate by a factor of five that accompanies the transition from micron- to nanometer-sized crystallites may be due in part to the activation energy associated with forming a nucleus of critical size.

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