Synthesis of nanocrystalline diamond films in $Ar/H_2/CH_4$ microwave discharges

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In this work we report on the investigation of a Microwave Plasma Assisted CVD process used for nanocrystalline diamond (NCD) deposition. In the MPACVD system, $Ar/H_2/CH_4$ discharges are ignited in a low pressure vessel placed in a microwave cavity. The gas mixtures used are characterized by an argon amount greater than 90% and a CH_4 concentration of around 1 %.

The stability of $Ar/H_2/CH_4$ microwave discharges was first examined in order to determine the plasma conditions leading to an optimal power coupling without formation of soot particles. This point was of prime interest since plasma stability is required to obtain reproducible process characteristics and to avoid a long time variation of the plasma during deposition. It also enabled us to determine the range of experimental parameters appropriate for nanocrystalline diamond growth.

Various growth conditions were then studied by varying gas composition, input microwave power, pressure and substrate temperature. Film characteristics such as surface morphology and topography, structural properties and nanostructure were investigated using conventional characterization techniques such as SEM, AFM, visible and UV Raman spectroscopy, XRD and TEM. In-situ and real-time optical monitoring of the growing samples was systematically carried out using pyrometric interferometry technique with a bichromatic pyrometer. This technique, particularly suitable for smooth transparent films such as nanocrystalline diamond, enabled us to improve the process control and reproducibility. It also permitted the determination of some dynamic information related to the incubation time, the optical properties or the growth rate, using a monolayer or multilayers modelling of the growing film. Nanocrystalline diamond films of different thickness were deposited with the goal to realize surface acoustic wave (SAW) devices on AlN/NCD/Si layered structure. The results showed that the acoustic velocity obtained on devices based on as-grown nanocrystalline diamond is similar to that obtained with polished polycrystalline diamond films.

The $Ar/H_2/CH_4$ microwave discharges were investigated under growth conditions by spectroscopic measurements. Both emission and broadband absorption spectroscopy techniques were employed in order to estimate the gas temperature, that is a crucial parameter in plasma chemistry, and the density of C_2 dimer, that is supposed to be the key-species in the growth process. The results showed that, under the moderate power injected in the considered reactor, the microwave discharges are characterized by a relatively high gas temperature, above 3000 K, that favors the production of C_2 molecules up to 10^{14} cm^{-3}. These values were found to be in satisfactory agreement with those calculated by a thermochemical model developed in the frame of quasi-homogeneous plasma. Also, these results underlined an appreciable difference with the H_2/CH_4 microwave discharges commonly used for poly-crystalline diamond growth, that show lower gas temperature and where C_2H_2 is the most abundant carbon-containing species and the CH_3 radical is recognized to be the growth precursor. At least a part of the difference between the two processes comes from the quite different chemistry and transport phenomena that characterize $Ar/H_2/CH_4$ and H_2/CH_4 discharges.