Fast deposition of a-C:H and a-Si:H using an expanding thermal plasma beam

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Ar-H2:CH4 arcjet plasma was experimentally analyzed to obtain two-dimensional images of emissive species. The distributions of such emissive species as H (656 nm, 486 nm), Cz (516 nm) and CH (431 nm) were revealed to have a good correlation with the thickness and structure of diamond films. Though non-emissive radicals of CH and ground state H atoms have been reported to play an important role in diamond growth, the images of the emissive radicals were found to be useful for the prediction of the diamond growth. The distribution of emissive radicals in a high density plasma can be easily obtained compared with LIF or IR absorption spectroscopy which provides the non-emissive radical distribution following complicated experimental procedures.

The substrate was fixed at 2 cm downstream from the arcjet nozzle exit and kept at 1200 K during the plasma exposure. The arcjet was operated at 2 kW with the flow rates of 7 SLM for Ar, 2 SLM for H2 and 0.1 SLM for CH4. The pressure of a vacuum chamber was controlled from 0.1 to 20 Torr.

At 5 Torr, a diamond film of a crater-like surface with glassy carbon around a diamond rim was obtained. When this crater-like film profile was formed, intensity of the excited H atoms became stronger and Cz weaker over the jet column and emissive CH was rich in the outer region of the jet.

On the other hand, at 10-20 Torr diamond films of 1 cm diameter and of maximum thickness in the center of about 100 μm were obtained in the deposition time of 30 min. In this case the intense regions of Cz and CH emissions consist of a highly emissive core from the nozzle and a stagnation part in front of the substrate, while excited H atoms quickly decayed near the nozzle exit and the H emission was not observed near the substrate. However, a peak of Raman spectrum at 1334 cm⁻¹ for the deposited films suggests the existence of the ground state H atoms, which form a dense region in the vicinity of the substrate and work as effective etchants of graphitic carbons.

The increase of the pressure enhances the collisions between particles and the frequent collisions results in the energy transfer from hydrogen, argon and electrons to CH4 injected from outside of the jet region. At higher pressures the narrowed jet including the decomposites of CH4 makes the carbon rich region on the substrate surface, which was resulted from the strong emission of Cz.

Although the higher pressure is preferable to make a diamond film for the above-mentioned reasons, ball-like carbons covered with diamond facets were obtained even at 0.1 Torr. This means that CH4 was dissociated into some species including diamond precursors at such a low pressure.

In this experiment the concentration gradient of each reactive species had a greater influence on the radial variations of the film quality than the temperature gradient of the substrate surface did.

A recently developed fast deposition method, utilizing a thermal plasma which expands into a vacuum vessel, is used to deposit amorphous hydrogenated silicon and carbon layers (a-Si:H and a-C:H respectively). The deposited layers are produced by admixing silane and methane (or acetylene) to the argon carrier plasma. In contrast to the conventional Plasma Enhanced Chemical Vapour Deposition where the deposition is diffusion limited, in this deposition device the deposition mechanism is flow determined. As a result the deposition rates are large, typically 100 nm/s for a-C:H and 10 nm/s for a-Si:H. The a-Si:H layers are deposited on crystalline silicon and Corning glass substrates and the a-C:H layers on either steel, zinc or silicon substrates.

The produced layers are analyzed using in situ helium-neon ellipsometry to determine the growth rate, refractive index and absorption coefficient at λ = 632.8 nm, ex situ visible spectroscopic ellipsometry to determine the bandgap, the wavelength dependent absorption coefficient and refractive index, infrared transmission measurements to determine the bonding types in the layers, Rutherford Backscattering (RBS) and Elastic Recoil Detection (ERDA) to obtain the C/H and Si/H ratios and possible contaminations of oxygen and nitrogen. Furthermore for a-Si:H the dark to light conductivity ratio was measured. In case of a-C:H also Auger spectroscopy was performed to get depth profiling of species present in the layers.

From the results obtained for a-Si:H, it can be concluded that the layers have large oxygen contamination due to post oxidation. We believe that this observation is related to the morphology of the layers as determined from the in situ ellipsometry: the layers have voids and the refractive index is close to the substrate interface larger than at the layers surface. The infrared transmission measurements reveal that no SiH3 is present and that SiH is the dominant bonding form. The bandgap determined from the spectroscopic ellipsometric data is E_{gap} = 1.6 eV. The dark to light conductivity ratio is approximately 103. No correlation was found between the infrared transmission measurements and the RBS and ERDA results pointing again towards an interpretation in terms of morphology.

The a-C:H layers can be characterized from soft polymerlike to hard diamondlike with E_{gap} = 0.5 to 1.0 eV, a refractive index of 2.2 to 1.3, and a C/H ratio of 1.5 to 0.6. The bonding types change from dominant sp³ (both C-H and C-C, few CH₄) for diamondlike coatings, to sp² (C-C and C-H) for the more polymerlike coatings. From the depth profiles, obtained using Auger spectroscopy, it is found that for layers containing oxygen through the whole layer the corrosion wearness is detrimental, while for layers containing significant amounts of nitrogen the corrosion wearness seems to be improved considerably.