Abstract

Diamond - like carbon (DLC) is a unique material because of the excellent combination of properties such as high hardness, low friction coefficient and high wear resistant, electrical insulation and chemical inertness. Besides, DLC materials have tunable optical, electronic and electromechanical properties that depend largely on the ratio of sp^3/sp^2 carbon bonds. In this work, the roles of the sp^3, sp^2 bonds and hydrogen content are analyzed closely on DLC films prepared using RF and DC magnetron sputtering, with and without active biasing. Diamond-like carbon thin films were prepared using a graphite target in a CH_4/Ar atmosphere by RF and DC reactive magnetron sputtering. Variable sputter powers and CH_4 flow rates were used, whilst maintaining a fixed argon flow rate. As a first result, the optimum thin film growth condition comprised of 50 % of CH_4-Ar concentration for these room- temperature sputtered films, applicable to both RF and DC sputtered films were determined. The amorphous hydrogenated carbon films (RF sputtered) exhibit more diamond-like features at this composition, characterized by moderately high fraction of sp^3 bonds, coupled with higher Tauc gap values, (1 - 1.5 eV). There was also a notable increase in density (~ 2.5 g/cm^3) and mechanical strength (E ~ 51 GPa). It was evident that the mechanical and structural properties of the films depended on the degree of CH_4 dilution of the Ar ambient, due to the different chemistry occurring in the plasma. As the amount of methane increased, X-ray reflectivity (XRR) results showed structural changes from a more disordered polymer-like structure to a less disordered graphitic arrangement. In addition, at the optimum condition, the films had a higher resistivity owing to the increased proportion of sp^3 bonds as evidenced by the increased presence of CH_n (n=1-3) radicals. The accompanying observed changes of the phonon phase velocity were related to the microstructural changes of the films. It was demonstrated that the mechanical strength of the *as-deposited* films flattens out at 50 % composition, thus necessitating either an active biasing of the substrate or

annealing of the films in order to further improve the mechanical strength. The structural evolution of the DLC films as a function of the deposition conditions (such the substrate bias voltage, the pressure and power) studied using Raman and UV-Visible spectra showed that increasing substrate bias voltage leads to the reduction of the cluster size of sp^2 rings, whilst increasing the sp^2 chains and the sp^3 bonds. These observations were confirmed by the optical performance of the coatings, characterised by transmittance, which was observed to similarly increase as the sp^3 bonds increased. The results of Raman analysis suggested that the sp^3 phase that is related to the disordered sp^2 phase attained its maximum content at a bias voltage of - 100V on films grown on silicon. In addition, the results of electrical conductance, Fourier transform infrared (FTIR) and X-ray photoemission spectroscopy (XPS) analyses revealed that a bias voltage of -100V was more favorable to sp^3 phase formation under the current experimental conditions. The FTIR results further confirmed that these films were indeed more diamond-like carbon by virtue of the presence of absorption bands such as sp^3 C-H_2 symmetry and asymmetry (at 2850 and 2920 cm^{-1}) and sp^3 C-H_3 asymmetry (at 2950 cm^{-1}). In addition, it was postulated that it is likely that the main reason for the observed lower elastic moduli (~ 17 GPa - 20 GPa) for DC sputtered films, compared to that of RF sputtered films (~ 47 - 62 GPa) is likely directly linked to the increase of polymeric carbon content in the DC films as evidenced by the Rayleigh velocities ranging between 1860 - 2274 m/s, characteristic of polymer-like carbon films. Thus, in this study, the direct influences of growth conditions on the DLC microstructure and also construct a correlation between electrical, mechanical properties and electronic properties of these materials have been demonstrated.