Metal-containing diamond-like nanocomposite temperature sensors

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Metal-carbon nanocomposites are characterized by a number of unique properties, perspective for the various-type sensor applications. Presented experimental data of the conductivity of amorphous tungsten- and niobium-containing carbon-silicon nanocomposite films show the possibility to design the advanced wide-range temperature sensors, which are expected to possess the chemical stability, biocompatibility, mechanical, and other properties typical for this class of materials. The investigated films were deposited onto polycrystalline substrates using combination of Plasma Enhanced Chemical Vapor Deposition (PECVD) of siloxane vapors and DC magnetron co-sputtering of metal target. The conductivity σ of the films, measured using standard 4-probe technique in the temperature range 80-400K, is characterized by the gradual decrease with temperature. The experimental $\sigma(T)$ dependences are well fitted by the power-law expression, $\sigma(T) = \sigma_0 + aT^n$, where σ_0 , a , and n are the fitting parameters dependent on the type of metal and the value of metal concentration. The electron transport mechanism in the investigated amorphous metal-containing carbon-silicon nanocomposite films is discussed in the framework of the model of inelastic tunneling of electrons in amorphous insulators in the presence of the structural transformation in the carbon-silicon host matrix. The evolution of the structure of the carbon phase by metal concentration increase was studied by Raman spectroscopy.

Keywords : Temperature sensor, diamond-like, metal grain, inelastic tunneling effect

I. INTRODUCTION

Diamond-like carbon (DLC) films are widely used as a coating material because of these attractive properties such as high hardness, low friction coefficient, biocompatibility and so on.¹ These properties are originated from the structure of carbon sp^3 and sp^2 bonding state. In contrast to these benefit of DLC it has been thought that there are mainly two disadvantages of it; low adhesion strength and

difficulty to use as high temperature applications. In order to improve them additional element was doped into DLC films. Diamond-like amorphous carbon-silicon nanocomposite films or metal-containing diamond-like carbon silicon nanocomposite films are thought to be one of the most perspective material for the wide temperature range applications. This class of materials are called diamond-like nanocomposites.²

In this paper we investigated W- and Nb-DLN films with the aim of advanced wide range temperature sensors. These materials are hoped to posses possess the two functionality; mechanical properties and sensor property.

II. EXPERIMENTAL DETAILS

W- and Nb-containing DLN films were deposited onto polycrystalline substrates with two process. Amorphous carbon-silicon matrix were fabricated by DC discharged of siloxane vapor. During the deposition DC magnetron sputtering was used for doping metals. After the deposition samples were prepared by Ar plasma for four-terminal geometry. Au sublayer were also deposited onto the films in order to decrease the contact resistance.

Electrical conductivity were measured by AC lock-in technique in the temperature range of 80 – 400K under vacuum condition. Atomic concentration measurement was carried out by electron probe microanalyser (EPMA).

Structural analysis was performed by Raman spectroscopy with He-Ne laser (632.8nm wavelength) and 20mW power. We controlled input power for 2mW.

III. RESULTS AND DISCUSSIONS

Fig.1 shows the plot of electrical conductivity at 300 K against W and Nb concentration. As it was seen that conductivity at 300 K increased with metal concentration increase. Me-DLN structure was considered that about metal grains are dispersed in amorphous insulating DLN matrix. The grain size was determined by the concentration of metal. From the percolation theory this conductivity vs. concentration behavior was well explained by following relationship,

$$\sigma \sim |X - X_c| \quad (1)$$

here, σ is conductivity, X concentration and X_c concentration of percolation threshold.

Fig.2 shows that temperature dependence on normalized resistivity ($\rho(T)/\rho(300\text{ K})$) in Nb-DLN films. Normalized resistivity decreases with temperature increases. These are typical ones of metal-insulator composites.

Fig.1. Plot of electrical conductivity at 300 K against metal concentration.

Fig.2. Temperature dependence on conductivity in Nb-DLN with different Nb concentration.

The nanometer-sized metal clusters are dispersed in amorphous insulating matrix. The electron transport in the films is well explained in the framework of the inelastic tunneling model of the electrons in amorphous insulators, which predicts the power-law temperature dependence of the conductivity. The power-law expression is following way,

$$\sigma(T) = \sigma_0 + AT^p \quad (2)$$

here $\sigma(T)$ is conductivity, T temperature and , are parameters that were determined by metal concentration, deposition parameters and so on. Eq.(2) was well fitted for the all experimental results.

It was considered that electrons go from one metal grain to other through a potential barrier with the existence of localized states. From inelastic tunneling

model the average number of localized states in an intercluster potential barrier can be calculated from the value of p .

$$\langle n \rangle = (1/2)(p-1+(p^2+2p+9)^{1/2}) \quad (3)$$

here $\langle n \rangle$ is average number of localized states in an intercluster potential barrier. Fig.3 shows calculated $\langle n \rangle$ depend on metal concentration. The investigated films are characterized by the two different types of metal concentration dependences of $\langle n \rangle$. The W-, and Nb-DLN films demonstrate the clear minimum of it at approximately 22-25 at. % of metal.

In order to analyze the evolution of the carbon phase structure with metal concentration increase a Raman spectra measurements were carried out. The observed Raman spectra of Me-DLN films are similar to those typical for DLC films as shown in Fig.4. The metal concentration dependence of peaks intensity ratio $I(D)/I(G)$ ($I(D)$ and $I(G)$ are the intensities of D and G peaks respectively) which is proportional to the typical size of sp^2 cluster³ well correlates with concentration dependence of $\langle n \rangle$.

Thus in W-, and Nb-DLN films the growth of sp^2 cluster size is observed starting from 22-25 at. % of metal.

Finally let us consider the both films as a temperature sensors. There are some method to evaluate the sensitivity of temperature sensors. As one of the possible method dimensionless temperature sensativity (S_T) was used. S_T was determined from,

$$S_T = d(\log(V))/d(\log(T)) \quad (4)$$

From eq.(4)

$$S_T = (T/V)(dV/dT) \quad (5)$$

Here, V is measured potential and T is temperature. What this value means is that the if S_T ranges from 0.1 to 10 in certain temerature range, this temperature sensor can be used with high sensitivity against the temperature in this range. The caluculated dimensionless temperature sensitivity was shown in Fig.5. in this figure S_T of Nb-DLN were present.

In Fig.5 the value of S_T was within torelance in the investigated temperature range. We also obtained the same results for W-DLN films.

Fig.3. Average number of localized states in W-DLN films.

Fig.4. Raman spectra of W-DLN films with different metal concentration.

Fig.5. Dimensionless temperature sensitivity in Nb-DLN films. Nb concentration is 12.8 at.%(upper) and 22.2 at.%(down).

IV. CONCLUSION

The W- and Nb-containing diamond-like nanocomposite films were studied. From the experimental results we obtained;

- Conductivity at room temperature depend on the amount of metal.
- Temperature dependence on conductivity were typical one of metal-insulator composites.
- Average number of localized states was calculated and affected by the structural transition due to the amount of metal.
- From the calculated dimensionless temperature sensitivity, W- and Nb-DLN films can be used as a temperature sensors.

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