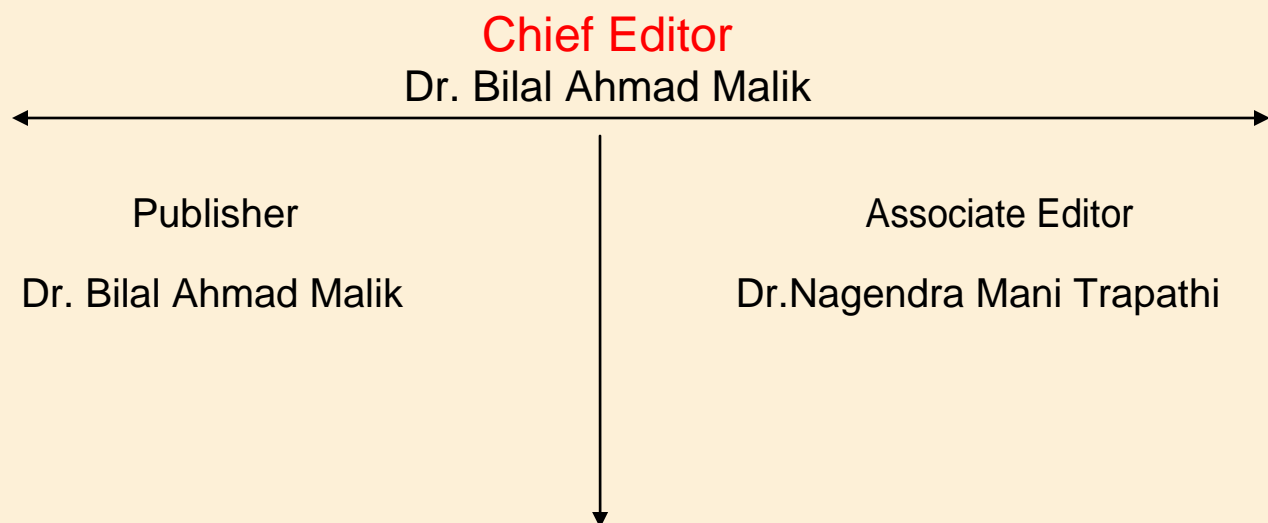


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A BRIEF REVIEW ON THE FULLERENES (C_{60} AND C_{70}) THIN FILM IRRADIATED BY DIFFERENT TYPES OF IONS BEAMS

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ABSTRACT

Ion beam irradiation of fullerenes (C_{60} and C_{70}) has attracted the interest of researchers due to its fascinating properties like high electron affinity, optical limiting property, unique molecular structure and high resistivity. The main objective of this paper to describe the fabrication of different ion beams irradiated fullerenes (C_{60} and C_{70}) thin film in different growth conditions by using Thermal Evaporation method as reported by various authors. The Change in the properties of Fullerenes near the Damage Cross Section after passing different ion beams in thin film is also reported.

Key words: Fullerenes C_{60} and C_{70} , Damage Cross Section, Thermal Evaporation, Ion beam.

INTRODUCTION:

The discovery of carbon clusters and new allotropes of carbon has emerged as a completely new field for physical chemistry and material research [1-2]. Oxidation is one of the basic process used to manipulate the carbon nanomaterials as for the preparation of very thin graphite film [3-4]. The increase of oxygen contents in irradiated film is not well understood, whether oxygen contents increases during ion irradiation or after exposure to the atmospheric oxygen [5]. Nowadays ion beam irradiation is known as a valuable and innovative tool for engineering and modification of materials at nano/atomic scale. Energetic ions (ions having velocities close to or higher than the orbital electron velocity) passing through materials lose energy along their path dominantly to the electronic sub-system, which leads to the excitation or ionization of atoms in the materials. The transfer of the electron energy to the atomic sub system is liable to modify the materials structure in remarkable fashion. The

reason for intensive research on irradiation effects in carbon nanostructure is the high technological importance of these systems owing to their unique properties which can be tailored by irradiation [6].

In the present study we have discussed the works of various researchers based on resistive heating method of ion beam irradiated fullerene (C₆₀ and C₇₀) and effect of ion beams on the properties of Fullerenes near the Damage Cross Section. The experimental works carried out by all researchers are in Nuclear Science Centre, New Delhi, India.

EXPERIMENTAL TECHNIQUE:

Nowadays, method of thin films deposition by resistive heating method in vacuum is highly widespread. The Figure of resistive heating method is shown in figure 1. The thermal evaporation is the process of transition from solid to the gaseous by means of break of interatomic bonds between atoms on the surface. The probability of evaporation of atoms on the surface of matter depends on the bond-breaking energy E_λ , and atomic specific evaporation rate (i.e. the number of atoms of matter evaporating per unit area of a second) is defined by

$$N_u = N^{2/3} \nu \exp \left[-\frac{E_\lambda}{kT_u} \right] \dots\dots(1)$$

where N_u - the concentration of atoms in solid material, $N^{2/3} = 10^{15} \text{ cm}^{-2}$ - the concentration of atoms at the surface, ν - the frequency of vibration the atoms in the lattice, kT_u - a value that proportional to the average kinetic energy of the atoms of the material. There is a probability that the instantaneous kinetic energy of the individual atoms on the surface exceed the bond-breaking energy E_λ , if the temperature is great. Evaporation of material takes place almost at any temperature. However, the evaporation temperature is conventionally called a temperature at which the vapor pressure of material above its own surface P_s reaches 1.33 Pa. The rate of evaporation G (amount of material evaporating in 1 sec from 1 cm²) depends on the saturated vapors pressure and temperature, and is described by the Langmuir’s equation:

$$N_\downarrow = 7,76 P_s \sqrt{\frac{M_u}{T_u}} \dots\dots\dots(2)$$

where PS - a saturated vapor pressure, Mu - the molecular weight of evaporated material, Tu - a temperature of the material.

If introduce the coefficient of evaporation α_1 (in the case of a clean surface $\alpha_1 = 1$) the rate of free evaporation N_u in vacuum will be defined by:

$$N_u = 4,66 \cdot 10^{24} \frac{\alpha_1 P_s}{\sqrt{M_u T_u}} \dots\dots\dots(3)$$

This formula is valid for the case, when atom after the evaporation adsorbs on the substrate without collisions and the vapor pressure of the metal does not exceed $1,33 \cdot 10^2 \text{ Pa}$. For high the vapor pressure the mean free path of the vapor atoms decreases because of accumulation of vapor above the melt. And the evaporation process begins to depend on the speed of "depressure" for this cloud that is the diffusion in the surrounding volume from interface layer.

Since the increase of temperature of the material over the evaporation temperature leads in almost ten times increasing in its vapor pressure, therefore it is convenient to conduct the deposition process in the forced mode. This lowers the harmful effect of residual gases in the working chamber volume, since continuously updated surface of the film does not have time to adsorb a significant amount of residual gas molecules.

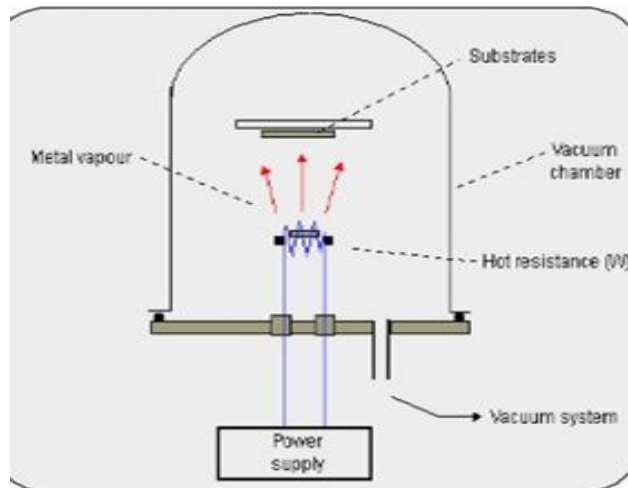


Figure 1: Schematic Diagram of resistive heating method

Forced mode also allows to condense on the substrate the initial ratio of the alloy components, because the cloud with increased of vapor pressure above the surface of the evaporator leads to the mixing of molecular flow. (The best mode is the explosive evaporation).

The atoms flying from evaporator to substrate have possibility of scattering and chemical interaction with residual gas molecules.

The evaporation rate of material depends on the presence of an oxide and other layers on the surface of evaporator. Depending on the thickness, thermal stability and other parameters of oxide film on metal it could decrease or increase the rate of evaporation. Most likely, evaporation of metal takes place through the crack in the oxide, and depends on the volatility of the oxide and the strength of its adhesion to the metal surface. In the presence of volatile oxides in a first approximation the evaporation mechanism can be represented by the sum of two processes: rising film and evaporation the formed compounds.

Main advantage of this method is universality because this device can obtain homogeneous metallic, semiconductor and dielectric layers of different thickness. Some more advantages of this method are (i) high speed of vaporization of the material and ability to control it in a wide range by changing power supplied to the evaporator (ii) high efficiency at group loading and processing of substrates (iii) the possibility to obtain required configuration of thin film elements of passive part of vacuum system simultaneously with deposition of films through the use of metal "free" masks; (iv) sterility of the process, which allows to obtain films almost free from pollution atoms (v) the possibility to guide the process as in high vacuum, since in oxidizing or regenerative environment of rarefied gas by issuing it in the working volume.

Process of film deposition in vacuum contains three main phases: (i) creating of atomic (molecular) flow of material from evaporator (ii) flight of atoms (molecule) from evaporator to the substrate (iii) condensation of atomic (molecular) flow on substrate [7].

PHENOMENOLOGICAL MODEL FOR DAMAGE CROSS SECTION

In this model an energetic ion passing through a solid film loses its energy, giving rise to a localized zone of high temperature and pressure. The energy (proportional to Se) is initially deposited in the electron system in a time 10-14s. It is then transferred to the lattice via electron phonon interaction. The ion beam makes a cylindrical zone with radius R_d in which the temperature exceeds. The damage cross section is given by

$$\sigma = \pi R_d^2 \dots\dots\dots(4)$$

Heat flow out of this zone by transient thermal processes. Due to this there will be annular zone around the cylinder R_d where the temperature is less than 4500 K but exceeds the melting temperature of C_{60} solid (~1700 K) and stay that way for duration longer than the time period for a lattice vibration (10-12s). This region has outer radius R_p . This zone will also have a high pressure because the molten material is prevented from expanding, being surrounded by relatively cold solid. This zone of high temperature and pressure is likely to contain dimer and polymer species once it has resolidified.

At low fluences, as long as the damaged cylindrical zones formed by the passage of individual ions do not overlap, the polymer content increases with fluence. With further increase in fluence these damaged cylindrical zones start to overlap.

Damage cross section (σ) is usually estimated using the relation

$$N = N_0 \exp(-\sigma\phi) \quad \dots\dots\dots(5)$$

Where N_0 is the total number of molecules in the unirradiated film and N is the number of remaining intact molecules after a fluence ϕ .

LITERATURE REVIEW

V. K. Jindal et al (2003) did work on C_{60} first time in Nuclear Science Centre, New Delhi. Here thin films of C_{60} were deposited on various substrates such as Si (100) and quartz using resistive heating method. Deposition was performed by sublimation of the pellets at a rate of 0.1nm/s and by passing a current of 75A in a Ta boat. The thicknesses of films were 160 and 230 nm. The films were irradiated by Ni ion beam of 110MeV energy, then electronic energy loss and nuclei energy loss were calculated and found electronic energy loss is dominant. The dimerization, polymerization and presence of nanocrystalline graphite embedded in a-C network formation have been revealed by Raman Spectroscopy and Optical Absorption Spectroscopy. In the damage area conductivity gets enhanced by 10^5 orders of magnitude. This reveals that the band gap of the materials decreases significantly [8].

A.Tripathi et al. (2006) deposited thin films on Si (1 0 0) substrate in vacuum of 1×10^{-6} Torr by resistive heating of C_{60} in a Ta boat. The thickness of films was 150nm. The samples were irradiated with 120MeV Au ion beam from NSC Pelletron with fluences of 2×10^{10} , 6×10^{10} and 2×10^{11} ions/cm². Raman studies revealed the

damage area of an ion to be 10nm. The formation of conducting tracks is attributed to breaking of fullerene molecules into carbon atoms, along the ion track and polymerization of fullerene molecules surrounding the ion path. The dispersed carbon atoms in insulating fullerene matrix serve as a hopping conductivity and polymerized fullerene molecules show graphite like metallic nature. The diameter of formed conducting zones varies from 40nm to 120nm and it is observed that conductivity of track increasing with increasing fluence [9].

Amit Kumar et al. (2006) deposited thin films on Si (1 0 0) substrate in a vacuum of 1×10^{-6} Torr by resistive heating of C_{60} in a Ta boat. The thickness of thin films was 500nm. The samples were irradiated with 200MeV Au ion beam from NSC Pelletron with fluences of 1×10^{12} , 3×10^{12} and 1×10^{13} ions/cm². The properties of fullerene films have been studied by Fourier Transform infrared spectroscopy. All the four fundamental IR active modes at 527, 577, 1183, 1428 cm⁻¹(T_{iu}) were observed. The sensitivity of damage for all the IR active modes are different under ion irradiation. It is found that 577cm⁻¹ T_{iu}(2) mode is most sensitive and damage cylindrical zone is around 1.35nm [10].

Films of C_{60} with a thickness in the range of 160-230 nm were deposited by Bajwa et al. (2008) on Si (1 0 0) float glass (FG) and quartz using resistive heating method. The film was irradiated by O, Ni, and Au ions using the 15UD pelletron accelerator at NSC IUAC. The films after exposure to the beams were characterized using Raman spectroscopy, optical spectroscopy, optical absorption spectroscopy and resistivity measurements. Raman study explained as the fluence of given specie of ion incident on a thin film of C_{60} is gradually increased, the polymer content as well as the a-C content increases and the C_{60} content decreases. In the conductivity measurement founds that increase in fluence causes further decrease in resistivity albeit at a slower rate. Finally the resistivity attains a value of $10^{-2} \Omega$ cm at fluence close to that causing complete destruction of C_{60} . Band gap of film reduces to zero over the same range of fluence. The C_{60} dominated material at low fluences as well as the a-C dominated material obtained after high fluences show the existence of a band gap. The damage cross section of films increases as fluence increases [11].

Rahul Singhal et al. (2008) developed C_{70} films with a thickness of 200nm. These films were deposited on quartz and Si (1 0 0) substrates by resistive heating of a C_{70} pellet. The base pressure in the chamber before and during the evaporation was 8×10^{-7} and 6×10^{-6} torr, respectively. The C_{70} thin films on the quartz substrates were irradiated with 120MeV Au ions at fluences of 1×10^{12} , 3×10^{12} , 6×10^{12} , 1×10^{13} and 3×10^{13} ions/cm². In the case of 120MeV Au ions, the electronic and nuclear energy losses in C_{70} are 1.4×10^3 and 16 eV/Å, respectively. The

damage cross section found here $2.73 \times 10^{-13} \text{cm}^2$ as deduced from Raman scattering. The radius of damaged cylindrical zone is about 2.9nm. UV-Vis absorption data show that optical band gap for pristine C_{70} is 1.98 eV and decreases with increasing ion fluence. The resistivity for pristine C_{70} is of the order of 10^5 Ohm-cm, which decreases with increasing ion fluence. Measurement show a steep increase in the conductivity of irradiated fullerenes beyond a fluence of 1×10^{12} ions/cm² which is ascribed to the transformation of C_{70} into a-C within each ion track, with a much higher conductivity than the surrounding fullerene matrix. At a fluence of 3×10^{13} ions/cm², the molecule is transformed into a-C [12].

Rahul Singhal et al. (2008) reported the synthesis of Ag NPs embedded in C_{70} matrix with the tunability of LSPR wavelength from 521 to 581 nm by varying the metal fraction from 4.5% to 28%. Unusually large LSPR wavelength and its large change with metal content were explained by Maxwell –Garnett theory considering the absorbing nature of the fullerene matrix. TEM and XRD results confirm the formation of Ag NPs in the fcc phase in C_{70} matrix. The appearance of hidden vibrational modes of C_{70} in Raman spectrum of this nanocomposite film indicates the possibility of application of this nanocomposite in surface enhanced Raman spectroscopy [13].

Amit Kumar et al. (2008) deposited fullerene thin film on Si(1 0 0) substrate in a vacuum of 1×10^{-6} Torr by resistive heating method. The thickness of film was 500nm and samples irradiated by 200MeV Au and 180 MeV Ag ion beams with fluences of 3×10^{10} to 1×10^{13} ions/cm². Here in ion irradiated fullerene films using on-line ERDA and off-line XPS and NRA techniques oxygen contents reported. Off-line techniques show that the oxygen contents increases with ion fluence, where as the on-line measurement revealed that the oxygen content decreases with ion fluence. These experiments findings show that the oxygen as well as hydrogen contents in the irradiated films increase after exposure to the atmosphere [14].

Rahul Singhal et al. (2009) deposited thin films of Ag- C_{60} nanocomposite by evaporation Ag and C_{60} simultaneously from two crucibles. Films were grown on different substrates such as glass Si and carbon coated Cu grids in a high vacuum chamber. In pristine sample the average size of Ag nanoparticles was 6nm and growth of Ag nanoparticles from 6 to 10 nm with fluence upto 3×10^{13} ions/cm² was observed with ion irradiation. The growth of nanoparticles is explained by dissolution and re-precipitation mechanism. UV-visible spectroscopy revealed a blue shift of 49nm with increasing ion fluence. The blue shift is explained in terms of transformation of fullerene matrix into amorphous carbon network with ion irradiation [15].

Rahul Single et al. (2009) deposited C₇₀-27% Ag nanocomposite thin films on different substrates by thermal co-deposition of silver and fullerene C₇₀. These films were irradiated with a beam of 120MeV Ag ions at different fluence ranging from 1×10^{12} to 1×10^{13} ions/cm². The growth of NPs is explained in the framework of the thermal spike model. The blue shift of 100nm is observed. The blue shift is explained in term of transformation of fullerene matrix into the a-C network with ion irradiation. This is confirmation of transformation of fullerene C₇₀ into amorphous carbon [16].

Rahul Singhal et al. (2009) deposited thin films of C₆₀ and C₇₀ fullerene on quartz substrate by resistive heating of their pellets. The thicknesses of films were 200nm. These films on quartz substrate were irradiated with 120MeV Ag ions at fullerenes of 1×10^{12} , 3×10^{12} , 6×10^{12} , 1×10^{13} and 3×10^{13} ions/cm². The radius of damaged cylindrical zone is about 3.5 and 2.5nm for C₆₀ and C₇₀ respectively. UV-vis absorption data shows that band gap decreases with increasing ion fullerene. In both cases at high fluence the fullerene structure completely transformed into a-C in both films. Although C₆₀ is known to be more stable in the family of carbon clusters, but C₇₀ is more stable against ion irradiation as compared with C₆₀ [17].

P.Sharma et al. (2016), grown C₇₀ thin film on double sided silicon substrates by resistive heating method. Ion track has been formed by irradiating the thin films with 90MeV Si ion beam. The damage cross section and radius of the track is approximately to be 0.32×10^{-13} cm² and 1.01 nm. The surface analysis revealed that as the fluence increases of 90MeV Si ion, the roughness and particle size also increases with increasing fluence, nature of fullerene surface transforms from hydrophobic to hydrophilic [18].

Pooja Sharma et al. (2016), grown C₇₀ thin film on glass substrate by resistive heating method. After 55 MeV Si ion beam irradiation of thin films, ion track formation take place. The radius of ion track is 1.41nm. UV-visible absorption spectroscopy results lead to infer that that the absorption bands declines with increasing fluences. The band gap decreases with increase in fluence from 1.75 to 1.68 eV. The roughness of the films decreases as a consequence of ion irradiation and agglomeration of the smaller particles also take place due to irradiation. The average particle size increases from 27 nm to 61 nm due to such particle coarsening effect of ion irradiation [19].

Pooja Sharma et al. (2016), grown thin films of C₇₀ on glass, quartz and silicon substrates by irradiated by Ni ion beam of 90MeV. The intensity of the Raman bands is reduced, which indicates the damage in the C₇₀ films

due to electronic energy deposition by the ion beam. The damage is more at higher fluence and the fullerene C₇₀ film is decreased from 1.87 to 1.80 eV with ion irradiation. Agglomeration in the particles has been observed after ion irradiation which results in increase of the particle size from 40 to 57 nm. The radius and cross section of ion track in C₇₀ found 2.3nm and $1.72 \times 10^{-13} \text{cm}^2$, respectively.

RESULTS AND DISCUSSION:

In the all papers [8-20] thin films of fullerenes (C₆₀ or C₇₀) deposited on the different substrates and thin films were irradiated by different ions (Ag, Au, Ni and Si) beam with different energy. The thickness of films generally varies from 150nm to 500nm. The results of all papers revealed that when ion beams passing inside the samples, a cylindrical zone or damage area developed inside the samples. The ions beams are very energetic, when a beam passes; the changes in the properties of sample take place. Carbon fullerene changes into amorphous carbon. In some cases agglomeration in the particles has been observed after ion irradiation which results in increase of the particle size from 40 to 57 nm. The roughness and particle size also increases with increasing fluence, nature of fullerene surface transforms from hydrophobic to hydrophilic. The band gap of the sample decreases with increase in ions fluence and it become zero at the some value of fluence. These studies also show that conductivity of ion irradiated films increases with increase in fluence.

CONCLUSION:

The study shows that when thin films of fullerene C₆₀/C₇₀ are irradiated by ions beam then its some properties changes. The change in the properties of fullerene is very much helpful in various fields i.e. (i) this study is very important for the design of new optical sensor active in a wide range of visible region within a single chip (ii) this study is helpful in the fabrication of nanowires using the cylindrical ion track zone (iii) the versatility of ion beam techniques in material modification and characterization; (iv) the increasingly important role that the ion beam techniques can continue to play in materials technology development;

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