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Effect of low fluence radiation on nanocomposite thin films of Cu nanoparticles embedded in fullerene C_{60}

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ABSTRACT

Metal-fullerene nanocomposite thin films with low concentration of Cu metal embedded in fullerene matrix have been synthesized using thermal co-deposition method. An ion irradiation with 120 MeV Ag ions beam was performed to assess the radiation effects for Cu (3%)-C_{60} nanocomposites at different fluences 1×10^{12}, 6×10^{12}, 1×10^{13} and 3×10^{13} ions/cm^2. Optical properties and surface morphology of nanocomposite thin films were studied using UV-visible spectroscopy and atomic force microscopy, respectively. Absorption spectra demonstrated that absorption intensity of nanocomposite thin film was increased and modes of fullerene were diminished with fluence. The thickness of the film and atomic metal fraction in matrix were estimated by Rutherford backscattering spectroscopy and found to be ~45 nm and 3%, respectively. Transmission electron microscopy was performed for structural and particle size evaluation of Cu nanoparticles (NPs) in matrix at higher fluence. Structural evolution of copper-fullerene C_{60} nanocomposite thin films with increasing fluence of 120 MeV Ag ion beam was studied by Raman Spectroscopy, which confirmed the amorphization of matrix (fullerene C_{60}) at higher fluence.

1. Introduction

The metal-matrix nanocomposites are interesting class of nanotechnology due to their bi-functional properties for applications in memory devices, solar cells and sensors [1–5]. The matrices based on carbon materials such as fullerenes (the widespread one is C_{60}) have preserved their position in industry due to their unique geometry and properties like high electron affinity and high resistivity [6–8]. The fullerene C_{60} forms soccer-ball type structure consisting of 12 pentagons and 20 hexagons. The metal-matrix nanocomposites are interesting class of materials embedded with metal NPs and supposed to have superior optical and electrical properties because of their high surface-to-volume ratio and physical properties related to small size of NPs in matrix [9]. However, the challenge for scientific community is to synthesize the noble metal particles/nanostructures with desired size and in different matrices with significant properties for particular applications [10]. The noble metal nanoparticles show unique optical properties different from bulk such as resonant absorption and scattering of light. The collective oscillations of the free electrons in the conduction band, known as surface plasmon resonance (SPR) are responsible for resonant absorption and scattering of light [11]. This unique property of noble metal nanoparticles can be utilized in light harvesting performance in solar cells. Our group has performed earlier the studies on Ag-C_{60} [12], Ag-C_{70} [13] and Ag/a-C [14] nanocomposites and irradiated these films with swift heavy ion beams for the tuning of optical property namely surface plasmon resonance in visible region. In the case of Ag-C_{60}, a tuning of ~50 nm was obtained with 120 MeV Ag ion irradiation, whereas for the case of Ag-C_{70}, SPR tuning of ~100 nm was obtained. With ion irradiation of fullerene thin film, it was gradually transformed into amorphous carbon with much lower refractive index than that of fullerene and this provided a way to tune the surface plasmon resonance wavelength of the nanocomposite thin film. The refractive index of fullerene C_{70} (~2.3) is higher than that of fullerene C_{60} (~2.1) and therefore giving higher range of tuning upon ion irradiation using same beam and energy. Since ion irradiation of metal-fullerene nanocomposite film at higher fluence results in the metal/

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carbon film, a separate study was also performed on Ag/a-C nanocomposite film with ion irradiation.

Among noble nanoparticles (Ag, Au and Cu), Cu NPs are attractive for applications in various fields of optics and electronics e.g. memory storage, catalysts [15–17], printable electronics, solar energy conservation, antimicrobial protections [18,19], sensors [20–22], heat transfer systems [23], lubricants because of mechanical properties, high electrical conductivity, catalytic properties, high stability, high natural abundance in nature and low cost over Ag and Au [24–26]. Copper has excellent electrical and thermal conductivity, adequate corrosion resistance and appropriate melting point and therefore, copper-based alloys and composites are being extensively used in static and sliding electrical contacts [27], thermal management applications covers from electronic packaging [28] to heat sinks for nuclear reactor technology [29]. Besides, the relatively low strength and limited thermal stability, in case of Ag and Au, make them confined for further use of these materials. This is an advantage in replacement of Cu over Ag in electronic circuitry. But the inherent instability of Cu NPs under atmospheric conditions is a major concern and make them prone to oxidation. Many efforts to develop the methods and supporting materials for enhancing the stability of Cu NPs have been made. When metal NPs are incorporated in a matrix, the particles are stabilized as well as they are prevented from agglomeration by vander Waals forces [30]. Therefore, one of the best economical ways to create the advanced Cu-based nanomaterials for catalysis and other purposes is to anchor Cu NPs on supports such as iron oxides, SiO_2, carbon-based materials, or polymers [31]. To alter the sensitivity of such small structures, metal-fullerene nanocomposites are of choice. These systems are said to be complex because of mutual solubility of copper and carbon and also the wettability of carbon on liquid copper is extremely low [32,33]. The size and shape of metal nanoparticles and surroundings dielectric media are influenced by various factors such as temperature, pressure and irradiation etc. [34]. Many groups have studied the metal-carbon nanocomposite and effect of temperature on them in order to understand the charge transfer of metal to matrix and changes in structural and mechanical properties [35–37].

From decades, Nuclear power plants are great source to produce electricity and consists of many metallic materials. Due to good corrosion resistance and boiling temperature, copper alloys and composites may be used as heat sinks, condenser tubes and balance plant heat exchangers in nuclear reactor technology. Since, the nuclear reactors operate at high moderate conditions such as high temperature, pressure and carries high radiation zone, there is a big issue of corrosion of nuclear metallic materials such as stress corrosion cracking, irradiation assisted stress corrosion cracking and environmental stress cracking etc [38]. This demands the detailed study on the radiation induced modifications in metal-fullerene nanocomposites.

Swift heavy ion irradiation has become of a great interest in order to engineer the physical, chemical and optical properties of materials [39–41]. When an energetic ion passes through the material, it loses its energy via (i) electronic energy loss which dominates at high energies (>1 MeV/nucleon) and (ii) nuclear energy loss which dominates at low energies (~few hundred keV). The effectiveness of the interaction of energetic ions with a particular type of target atoms or molecules depends on the mass, charge and energy of the incident ions. In nanocomposite thin films, the transfer of the energy by incoming ion can change the size and shape of NPs and also the dielectric properties of the matrix [13]. It creates the changes in the optical, electrical and thermal properties of materials by creating defects, track etc. On the other hand, the high boiling temperature of copper makes it significant for high temperature and pressure conditions. Due to the corrosion of nuclear metallic materials such as irradiation assisted stress corrosion cracking, it is worth to examine the irradiation induced modifications of such composites. To the best of our knowledge, no group has studied so far the irradiation effect on copper-fullerene nanocomposite thin films.

In the present study, Cu (3%)-C_{60} nanocomposite thin films are synthesized by thermal co-deposition method and irradiated with 120 MeV Ag ion beam at different fluences 1×10^{12}, 6×10^{12}, 1×10^{13} and 3×10^{13} ions/cm^2 to assess the high radiation effect on nanocomposite thin films. A beam of Ag ions was chosen for the ion irradiation as Ag being the high mass element, it will provide high electronic energy loss in the target material upon the ion irradiation. When an ion of Ag with 120 MeV energy passes through the film, it loses its energy in collision with the electrons of the target atom and this loss is termed as electronic energy loss. With the high electronic energy loss, it is expected to have substantial modifications in the structural and optical properties of Cu-C_{60} nanocomposite thin films. Also among fullerene C_{70} and C_{60}, fullerene C_{60} is easily available and low cost material compared to C_{70}, so fullerene C_{60} was preferred with Cu metal to make a nanocomposite film. Since with the low Cu concentration, it was difficult to excite intense SPR peak, the nanocomposite films are irradiated with the swift heavy ion beam. Furthermore, structural and optical properties of the pristine and irradiated films of nanocomposite were investigated using TEM, Raman spectroscopy and UV-visible spectroscopy. Raman spectra confirmed the amorphization of fullerene C_{60} with increasing ion fluence. The schematic of amorphization of fullerene C_{60} with ion beam irradiation is shown in Fig. 1. Surface morphology of films was studied using atomic force microscopy.

2. Experimental plan

The fullerene C_{60} thin films embedded with Cu NPs were synthesized by thermal co-deposition method. The fullerene C_{60} and Cu metal were placed in two different crucibles (tungsten) in a chamber which was evacuated to a high vacuum using turbo molecular pump backed by rotary pump. Before deposition, the base pressure in vacuum chamber was 5×10^{-6} mbar. Fullerene C_{60} and

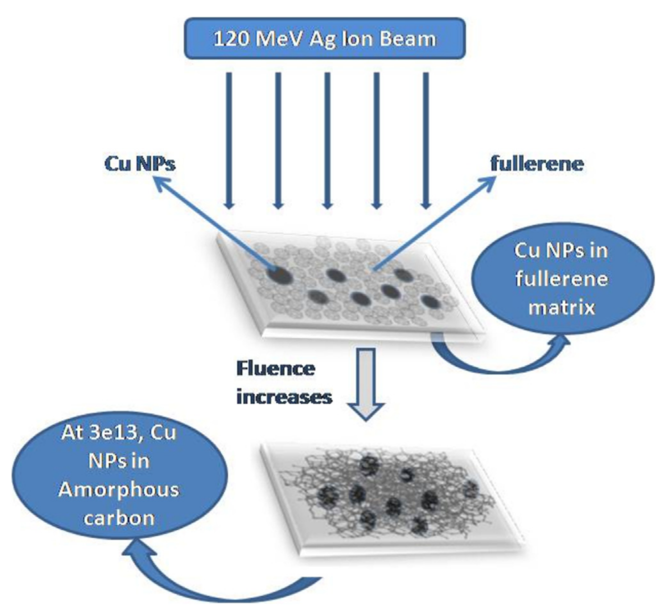

Fig. 1. Schematic of transformation of fullerene C_{60} into amorphous carbon with ion irradiation.