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An Advanced Carbon Nanotube-Graphene Hybrid Nanocomposite Prepared by Arc Plasma Technique for Various Critical Applications

This study presents a hybrid nanocomposite consisting of carbon nanotubes combined with graphene, which was synthesized using arc plasma-treated graphite discs for durations of 10 and 15 minutes. This material is intended for a range of significant future applications, including hydrogen storage, sensors, supercapacitors, lithium-ion batteries, spintronics, and biomedical technologies, among others. The X-ray diffraction (XRD) peak intensity of C(002) is found to be significantly reduced for plasma-treated graphite because of the change of flaky microstructure of graphite into a composite form of carbon nanotube and graphene. The Raman result shows that 15-minute plasma-treated graphite has a bilayer form of graphene. Transmission electron microscopy (TEM) analysis reveals bunch of multiwall carbon nanotubes incorporated with a graphene structure produced after 10-15 minutes' arc plasma treatment of graphite. These structures in bunches look like a spheroid in field-emission scanning electron microscopy (FE-SEM), when they are observed from the top view. The plasma treatment of graphite for 15 minutes has resulted significant increase in Brunauer–Emmett–Teller (BET) specific surface area and electrical conductivity, achieving 1401 m^2/g ± 38 m^2/g and (52.2 ± 2.1) $\times 10^3$ S/cm, respectively.

Keywords: Carbon nanotubes; Graphene; Microstructure; Hybrid nanocomposites
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1. Introduction

Nanotubes, nanorods, and nanowires, typical nanostructures in 1D, are widely reported in current studies across the globe to achieve different desired properties in materials. The unique behavior of nanostructured materials makes them valuable in material research. Nanotubes have potential applications in electronic device, gas sensor, catalyst, hydrogen storage, high-strength composite, etc. Nanotube form of carbon is one of the most important nanostructure materials, employed in various applications, including composites and vital roles in cancer diagnosis and treatment [1-2].

Graphene has been recognized as one of the most promising emerging nanomaterials because of its distinctive combination of exceptional characteristics. It is incredibly strong and rigid, exceptionally thin and nearly entirely transparent, extremely lightweight, and exhibits extraordinary conductivity for heat and electricity, holding the potential to transform numerous industries [3-4]. Recently, a cutting-edge graphene-derived nanomaterial, which includes stacked graphene platelet nanofibers and nanotubes [5], was developed via a bottom-up growth approach. Despite their dimensional similarity to carbon nanotubes, they have a fundamentally different internal structure and exhibit completely different electrochemical characteristics. Simply stacked graphene nanofiber (SGNF) is a newly developed microstructural integrated composite of ID carbon nanotube (CNT) and 2D graphene. It is a new challenging potential material for various critical applications, including hydrogen storage, sensors, supercapacitors, Li-ion batteries, spintronics, and biomedical applications, etc. [6-8]

The production of high-quality CNT/graphene hybrid nanocomposites/nanostructures (SGNF) is extremely interesting due to their unique properties and significant potential applications [5, 9]. Several synthesis routes have been reported for CNT/graphene hybrid structures, such as laser ablation, chemical vapor deposition (CVD), and electrochemical deposition [8–10]. However, these techniques often involve complex procedures, high costs, and limited scalability. For instance, laser ablation needs sophisticated equipment and high energy input, whereas hybrid structures produced via CVD may experience structural imperfections and contamination.

In contrast, the arc plasma technique can be considered as a simple, rapid, and highly energetic environment capable of achieving localized

temperatures exceeding 3500–4000 °C, thereby enabling direct vaporization and condensation of carbon in a controlled manner. This facilitates the simultaneous growth of CNTs and graphene as the hybrid nanocomposite (SGNF) within a single-step process. Moreover, the arc plasma process allows large-scale (~500 g) synthesis within 15 minutes of treatment of graphite, providing a practical route for producing high-quality CNT/graphene hybrid nanocomposite (SGNF) with minimal structural defects.

The present work systematically focuses on the arc plasma-assisted transformation of graphite into an advanced CNT/graphene hybrid nanocomposite (SGNF). By systematically varying the plasma treatment duration, we report how the microstructure evolves from graphite to a well-integrated CNT–graphene network, confirmed through property evaluation of materials. The results reveal that the arc plasma process offers a scalable, energy-efficient pathway to synthesize high-quality hybrid carbon nanostructures suitable for multiple critical applications.

2. Experimental

The starting material was taken as graphite of 99.999% purity (electrode grade) in the shape of disc of 4-5 cm. Transferred arc type plasma discharge (DC) (50 kW) is used for the treatment of graphite. Argon (Ar) is used for forming plasma. Ar was passed at a flow rate of 2 l/min. via the central axial aperture of the upper vertical graphite cathode electrode. Samples were plasma treated at voltage: 70-100 V and current: 320-360 A. A gap of 20 mm was maintained between the cathode and anode during plasma reaction time. The plasma treatment of graphite samples was carried out for 10-15 minutes. After plasma treatment was over electrical power was switched off. The *in-situ* cooling of the reactor / furnace was carried out for around 3-4 hours to attain room temperature. The plasma treated graphite samples were collected from the reactor / furnace at room temperature and taken for various characterization studies.

X-ray diffraction (XRD) study of samples was done by employing a PANalytical X'Pert Pro diffractometer. The microstructure and nanostructure of the samples were carried out employing a field emission scanning electron microscope (FESEM) (model ZEISS SUPRA 55) and transmission electron microscope (TEM) (model: TECNAI G2, FEI (Netherland)), respectively. Elemental analysis was performed by energy dispersive spectroscopy (EDS) facility available with FESEM. Spectroscopic property was evaluated by a micro Raman spectrometer (Renishaw inVia Reflex (UK)). Model: ASAP 2020, micrometrics was used for Brunauer–Emmett–Teller (BET) specific surface area analysis. Using the Keithley 6221 multimeter, the electrical conductivity of the samples was evaluated.

3. Results and Discussion

The graphite disc samples were arc plasma-treated for 10-15 minutes under an argon atmosphere. The crystalline and phases structure of the material was studied by XRD shown in Fig. (1). Various diffracted planes of carbon were identified in the untreated and plasma treated graphite based on their XRD patterns, by matching the observed *d* values against the 1999 JCPDS data file C: 41-1487. A sharp peak is seen at around 26.4º angle in all samples, which shows the presence of C (002). It is indicated that all the samples reveal various low-intensity peaks of C (100), C (101), C (004), and C (110). It found that the intensity of carbon peaks, particularly i.e. C (002), is significantly decreased after plasma treatment of graphite. The decrease in intensity is attributed to the change in crystallinity, structural changes, lattice position alteration, or a new kind of microstructure developed. This XRD result can be corroborated by TEM findings (discussed in the next sections), which shows microstructural changes occurred in plasma treated graphite. It has been revealed that the *d*-spacing of plasma-treated graphite is larger than that of high-purity graphite, which can be due to lattice expansion (this expansion takes place in the AB stacking order of the graphite lattice, as demonstrated by the disordered structure observed in the micro-Raman investigation). This lattice expansion occurred due to the processing of material under plasma conditions.

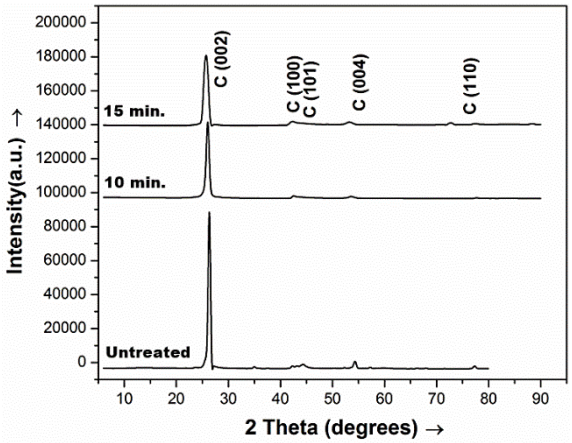

Fig. (1) XRD patterns of the untreated and plasma-treated graphite disc

Experimentally we have determined *d* spacing values for high intense C(002) peak as 3.3586 Å, 3.3767 Å and 3.3878 Å for untreated, 10 min and 15 min plasma treated graphite samples, respectively i.e., a small, measurable increase in interlayer spacing after plasma treatment. Using the Scherrer's equation [11], the crystallite size for untreated, 10 min and15 min plasma treated graphite samples, was measured as

~85.5 nm, ~28.2 nm, and ~18.0 nm respectively, by the following equation:

$$D = \frac{k\lambda}{\beta cos\theta} \quad (1)$$

where, k = 0.9, λ is the X-ray wavelength (Cu Kα λ = 1.5406 Å), θ is the Bragg's angle (in radians), β is the FWHM (in radians)

The broadening and shifting of peaks towards a lower diffracting angle along with reduction of crystallite size indicate the strong bonding present within the graphene sheets and CNTs. The deviation of crystalline and structural modification confirms the formation of CNT/graphene hybrid nanocomposite (SGNF) in plasma treated graphite. Non-carbon phases are not seen in XRD spectra, which exhibits high quality nature of the prepared novel composite SGNF by arc plasma treatment.

The simultaneous formation of CNTs and bilayer graphene in our arc plasma process results from a controlled balance of carbon vaporization, nucleation, and condensation governed by plasma parameters. Under DC transferred arc plasma conditions (70-100 V, 320–360 A, Ar flow rate 2 L/min, arc length 20 mm), temperatures exceed 3500–4000°C, causing rapid graphite sublimation into a dense carbon vapor of atoms and small clusters. The inert argon atmosphere prevents oxidation and enables controlled supersaturation near the cooler plasma periphery. During cooling, carbon species follow two concurrent growth routes: (i) at intermediate supersaturation and strong electric fields, tubular sp^2 networks form as CNTs; and (ii) in regions of slower cooling and higher carbon density, planar stacking yields few-layer or bilayer graphene. The interplay between plasma energy density (voltage/current) and gas dynamics (Ar flow) creates overlapping CNT and graphene growth zones, producing an integrated composite SGNF (CNT/graphene hybrid nanocomposite).

Figure (2) illustrates the comparison of the micro Raman spectra of untreated and plasma-treated graphite. Different carbonaceous phases such as G, D and 2D were identified. The G, D, and 2D peaks correspond to the first-order scattering of the E2g phonon from sp^2 carbon (graphite lattice), the first-order disorder in the graphite lattice, and the second-order disorder in the graphite lattice, respectively [3, 12]. In untreated graphite D peak is found to be absent, which indicates its perfect crystallinity with the least defect in the C-C arrangement in the skeleton. The intensity of the D peak is found to increase with increasing plasma treatment duration, indicating a rise in defect density and the formation of additional edge sites within the carbon framework. The G peak to 2D peak intensity ratio (I_G/I_{2D}) was evaluated for untreated graphite and those treated with arc plasma for durations of 10 and 15 minutes, with the findings indicating values of 2.1, 1.6, and 0.7, respectively. As shown in Fig. (2), the following number of layer were identified in plasma-treated graphite: few layers (4-5) of graphene in 10-minute treatment and bilayer of graphene in 15-minute treatment, referring to the study of Ferrari et al. [13].

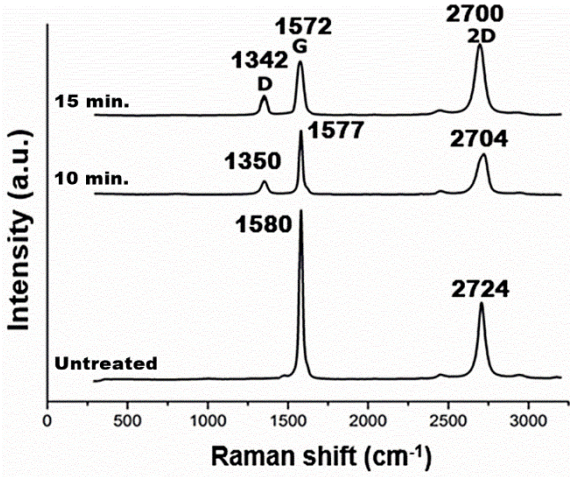

Fig. (2) Micro Raman analysis of the untreated and plasma-treated graphite disc

The average size of the crystallites in the sp^2 domains (La (nm)) was determined for the graphite samples treated with plasma by utilizing the equation presented by Lucchese et al. [14], which links the I_D/I_G ratio to the fourth power of the laser line wavelength (λ_l in nm units). 10 and 15-minute plasma-treated graphite samples show La values as 78.6 ± 2.7 nm and 62.3 ± 3.5 nm, respectively. The La (nm) measurements obtained for samples subjected to plasma treatment reveal that the average size decreases as the plasma treatment duration increases. This correlation associates the fragmentation of crystallites with the enhanced exfoliation of graphene layers, resulting in the creation of defects, disorder, sp^3 hybridization, and modifications in crystalline structure. This finding is supported by our XRD results, which show a reduction in the intensity of the primary peak of C (002), as illustrated in the XRD spectra (Fig. 1) of the plasma-treated samples.

$$L_a(nm) = (2.4x10^{-10})\ \lambda_l^4\ (I_D/I_G)^{-1} \quad (2)$$

Figure (3) illustrates surface morphological analysis by FESEM of both untreated graphite and graphite that has undergone typical plasma treatment for 15 minutes. The untreated graphite displays an irregular and flaky grain structure. In contrast, when the graphite disc is treated with arc plasma for 15 minutes, the morphology of the irregular flaky grains is modified, resulting in a rounded shape/spheroid due to the formation of a CNT/graphene nanocomposite (SGNF). The typical round type grained microstructures visible in FESEM are clearly recognized as a nanocomposite of carbon nanotubes (CNT) and graphene when analyzed under TEM (Fig. 4a and Fig. 4b). The TEM micrograph, as shown in Fig. (4b), was observed after 15 minutes of plasma treatment on graphite and shows a high

magnification and low contrast image of two folded, thin flat layers of graphene that are integrated with a nanotube structure. FE-SEM and TEM analysis results can be corroborated as follows. The as observed surface morphological spheroidal features by FESEM for plasma-treated graphite correspond to 3D agglomerates composed of entangled CNTs intertwined with few-layer graphene sheets. TEM reveals 2D structure at nano level that these spheroids consist of a dense CNT core surrounded and interconnected by folded graphene layers, forming a porous, networked architecture. In this 3D structure, CNTs provide a tubular backbone, while graphene sheets bridge and wrap around the CNT bundles, ensuring strong interfacial contact and enhanced electrical connectivity within the stacked graphene nanofiber (SGNF) framework. The nanotubes exhibit an external diameter in the range of about 5-15 nm.

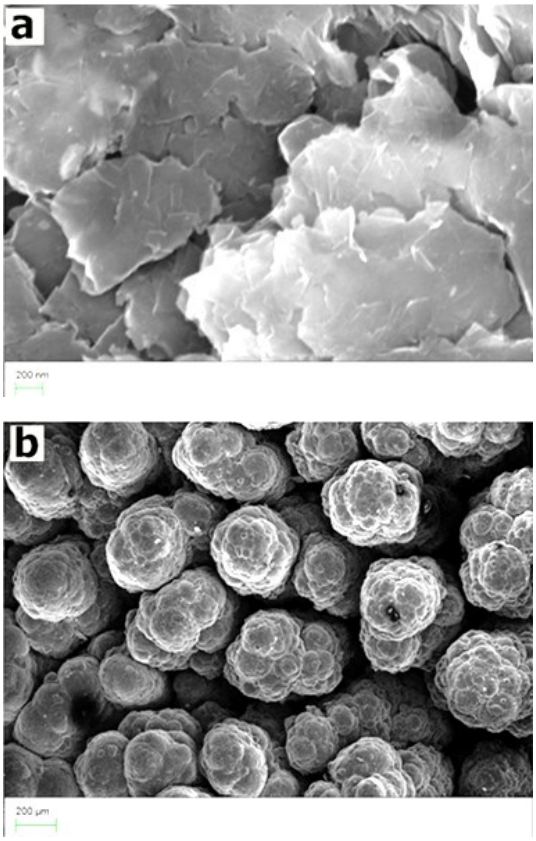

Fig. (3) FE-SEM image of graphite disc surface: (a) untreated sample, (b) 15 min. plasma- treated typical sample

The BET surface area of the untreated graphite was measured at 5 m^2/g, whereas the plasma-treated graphite powder for 10 and 15 minutes exhibited surface areas of 987±22 m^2/g and 1401±38 m^2/g, respectively. This observation aligns with the findings by Amiri et al. [15], which indicated a BET surface area of 1559 m^2/g for bilayer graphene. The BET surface area of such a structure has a particularly potential application for the storage of hydrogen because of having large surface area. The above measured BET specific surface value further confirms bilayer graphene formation in the nanocomposite of CNT/graphene after 15 minutes of plasma treatment of the graphite disc. The resistivity/conductivity (dc) of graphite disc was measured by treated and untreated graphite discs by four-point probe method.

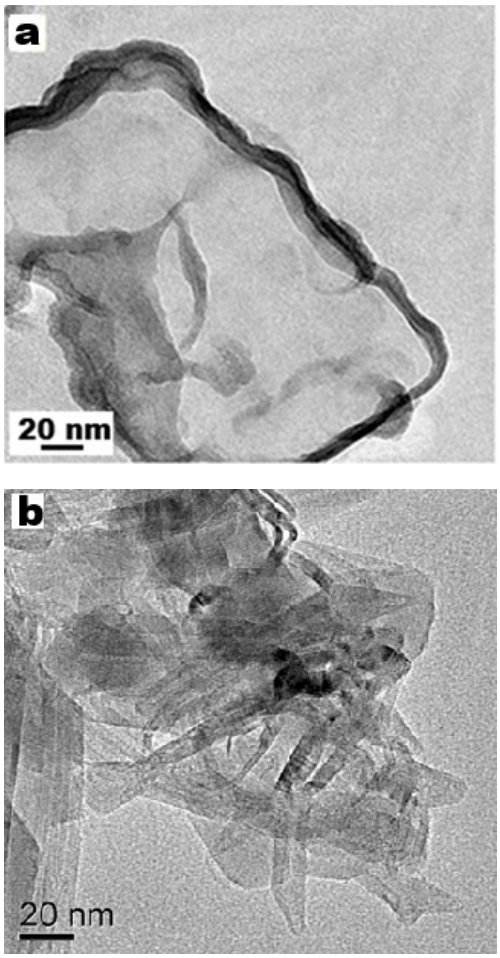

Fig. (4) TEM images of graphite disc shows CNT/graphene hybrid nanocomposite (SGNF): (a) 10 min. plasma treated sample; (b) 15 min. plasma- treated sample

Untreated and 10 minutes' plasma-treated graphite show electrical conductivity values as (0.65±0.05) $\times 10^3$ S/cm and (3.8±0.6) $\times 10^3$ S/cm, respectively. It is evident that the highest electrical conductivity (52.2±2.1) $\times 10^3$ S/cm ≈ (5.22±0.21) $\times 10^6$ S/m is observed in the case of 15-minute plasma-treated graphite. This value is found significantly higher than typical conductivities reported for bulk CNT/graphene hybrid composites and comparable to (or above) many high-quality CVD graphene reports. For comparison, monolayer graphene produced by CVD has been measured near 1.46×10^6 S/m [16], while bulk CNT/graphene hybrid films and porous composites commonly show conductivities in the 10^2-10^3 S/cm

range (i.e., 10^4-10^5 S/m) [17] depending on fabrication and density. However, exceptionally highly aligned CNT fibres or specially engineered graphitic assemblies show conductivities approaching the highest metallic-like values (up to ~10^7 S/m) [18], but these materials are specialized, highly densified structures rather than porous and bulk hybrids. In our study the unusually high conductivity observed in situ arc plasma-grown SGNF can be attributed to the arc plasma technique which provides an ultrafast and high-temperature environment (3500-4000°C) that facilitates simultaneous nucleation and growth from a common carbon vapor phase, extensive interconnection of sp^2 domains, high intrinsic mobility of few-layer/bilayer graphene domains and improved interfacial/graphitic contact.

4. Conclusions

In this work, an advanced nanocomposite of CNT/graphene (SGNF) was prepared in an arc plasma treated (10-15 min.) graphite disc. XRD indicates a decrease in the intensity of the primary C (002) peak following plasma treatment. The Raman analysis shows three significant peaks, namely D, G, and 2D, in the sample that has undergone plasma treatment. By calculating the intensity ratio of G to 2D for plasma-treated samples, it was found that the typical 15-minute plasma-treated sample shows a value of 0.7, indicating a bilayer form of graphene in the nanocomposite. The 15-minute plasma-treated graphite sample exhibited a high BET specific surface area of 1401 ± 38 m^2/g and an electrical conductivity of (52.2 ± 2.1) × 10^3 S/cm. These values represent a significant enhancement compared to untreated high-purity graphite, confirming that arc plasma treatment significantly improves both the surface and electronic properties through the formation of the CNT/graphene hybrid nanocomposite (SGNF). The applications of the nanocomposite developed in this study hold significant potential for future applications in hydrogen storage, sensors, supercapacitors, lithium-ion batteries, spintronics, and biomedical technologies, among others. These applications are progressive and are integral to our current and ongoing research.

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