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Research article

Exploring temperature-dependent properties in advanced graphene oxide nanocomposites for enhanced energy applications

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Abstract. This article provides a comprehensive overview of recent advancements in the production of graphene-based biopolymer nanocomposites, which have garnered significant attention due to their remarkable properties. By adding graphene oxide (GO) to a biopolymer matrix (biopolymer composite [bioC]), we successfully synthesized high-performance bioC-GO nanocomposites and conducted a thorough investigation of their electrical and thermal characteristics. The nanocomposites were structurally characterized using analytical techniques such as transmission electron microscopy (TEM), scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS), X-ray diffraction (XRD), and thermogravimetric analysis (TGA). The XRD revealed the amorphous nature of bioC while bioC-GO nanocomposites exhibited distinctive peaks attributed to interactions among fish scales, GO, and the biopolymer matrix. Both SEM and EDS confirmed a uniform distribution of biopolymers on GO, with elemental analysis indicating slight variations in carbon and oxygen content due to functionalized GO. The TEM revealed nanostructured bioC-GO flakes with wrinkled morphology. The TGA demonstrated the enhanced thermal stability of bioC-GO nanocomposites withstanding temperatures up to 800 °C, attributed to covalent interactions between collagen and nanographene oxide. Likewise, we investigated the resistivity response of bioC-GO nanocomposites under forward and reverse bias at various bias voltages at room temperature. The resistivity values, which are inversely proportional to conductivity, were found to be very low for the bioC-GO nanocomposites when exposed to temperatures as high as 150 °C at a steady applied voltage of 5 V. These findings demonstrate the potential of bioC-GO nanocomposites for applications requiring superior thermal stability, electrical conductivity, and environmentally friendly materials.

Keywords. Graphene oxide, Biopolymers, Characterization, Electric properties, Energy.

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1. Introduction

Over the past two decades, nanoscience has undergone explosive growth with miniaturization becoming increasingly important in fields such as computing, sensing, and biomedicine. This tendency is expected to significantly improve the already excellent reputation of nanotechnology. To advance these domains, it is crucial to be able to successfully synthesize nanoparticles of different materials, sizes, and forms and to put them together into intricate structures [1–3]. The unique structural properties of nanoparticles have recently led to a wide range of applications. Meanwhile, scientists are concentrating on materials with superior physicochemical properties that make them more appropriate for nanoscience and technology. Graphene and its polymer nanocomposites are among these materials that have had a major influence on nanoscience and hence on contemporary technology and scientific study. Because of its numerous beneficial qualities, scientists and engineers have long been fascinated by carbon. Graphene, a unique form of carbon, consists of a two-dimensional arrangement of sp^2 -bonded carbon atoms in a hexagonal lattice [4]. The remarkable surface area, mechanical strength, photocatalytic qualities, electrical and thermal conductivity, and antibacterial properties of graphene oxide (GO) make it a very sought-after material [5]. Known for its honeycomb structure of nanoscale sheets of sp^2 carbon atoms, GO exhibits impressive molecular barrier capabilities, electrical conductivity, and mechanical strength among other advantageous features. Because of these characteristics, several research projects have focused on creating polymer-based nanocomposites that include graphene [6]. Graphene oxide's versatility extends to its role in composite materials, where it improves properties such as lubrication, thermal conductivity, and mechanical strength [7].

As environmental concerns grow, there is increasing interest in biopolymer mixtures and nanocomposites as sustainable alternatives [8]. In several industries such as packaging, agriculture, and medicine, biopolymers made from renewable resources have more advantages than their petroleum-based equivalents [9]. Recently, there has been a lot of interest in GO as a possible component for 2D nanomaterials that resemble graphene.

Graphene oxide typically has a carbon-to-oxygen atomic ratio of between 2:1 and 3:1 and comprises a carbon framework modified with residual oxygen-containing groups. These materials exhibit unique chemical and physical properties, including high electrical conductivity, a tunable degree of functionalization, and a large surface area, making them attractive for various applications [10,11]. The improvement of GO preparation methods and the exploration of its potential applications have been the focus of extensive experimental research.

Recently, there has been a surge in research on nanocomposites that integrate GO with polymers and inorganic nanoparticles [12]. For instance, red seaweed is the source of carrageenan, a biopolymer with special qualities for a range of uses. Kappa-carrageenan (KC) is a common thickening agent in the food industry because of its gelling properties [13]. Its counterpart iota-carrageenan (IC) is a water-soluble, biodegradable polymer, making it suitable for a variety of applications including as a host matrix in this study. The synergy between inorganic components and organic polymers is driving innovation in materials science. The mechanical, thermal, and electrical properties of graphene-based polymer nanocomposites are better than those of new polymers and traditional filler-based composites [14]. Because of their many uses, GO and polymer composites have attracted a lot of interest from a variety of industries, such as high-strength materials, catalysts, energy-related systems, and energy storage and conversion devices. Revolutionary breakthroughs in materials science and technology are anticipated because of the exceptional qualities of GO and the adaptability of polymer matrices [15].

This study aims to synthesize and evaluate novel biopolymer composite (bioC)–GO nanocomposites by incorporating a mixture of fish scale powder, KC, and IC into GO. The research explores the potential of these materials to develop advanced composites with enhanced thermal and electrical properties, such as resistivity, heat stability, and breakdown strength. To the best of our knowledge, this is the first time this unique combination of biopolymers and GO has been utilized, offering promising insights into the development of multifunctional materials for diverse applications.

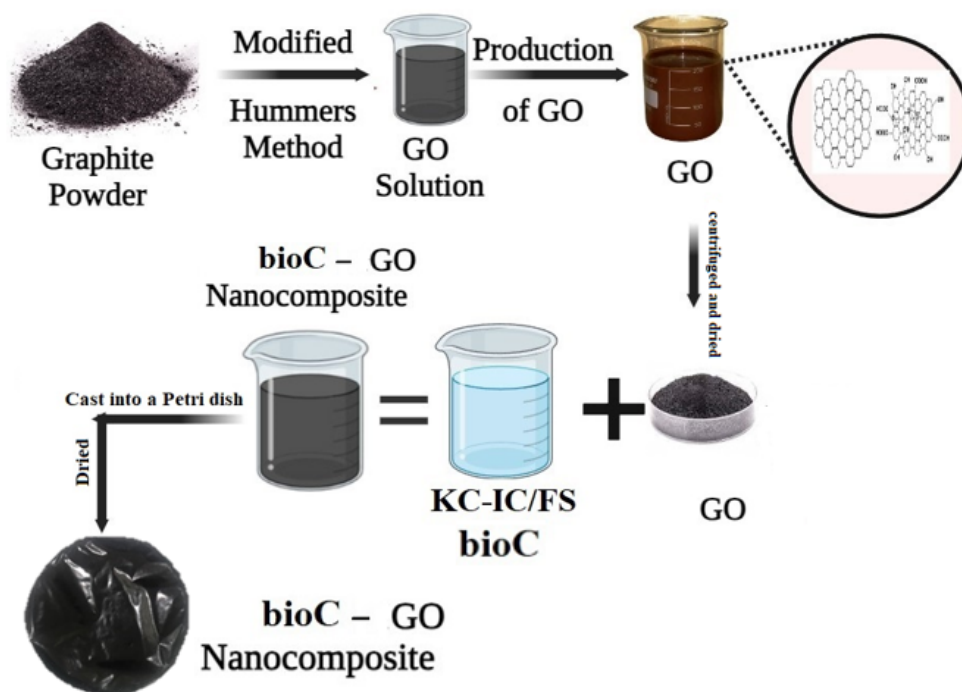


Figure 1. Schematic illustration of the synthesis process of bioC-GO nanocomposites.

2. Materials and methods

2.1. Preparation of fish scale powder

Biowaste Tilapia fish scales (FS) were sourced from the local fish market in Riyadh, Saudi Arabia. The scales were thoroughly cleaned, first being rinsed with tap water to get rid of any surface dirt and tissues that could have stuck. The cleanliness of the scales was then guaranteed by carefully removing any unnecessary elements. After that, the scales were carefully cleaned with deionized water to get rid of any remaining contaminants. The cleaned fish scales were sun-dried until they were crisp and brittle to maintain quality and avoid microbial contamination. To improve use, the scales were coarsely crushed into a powder after drying. To guarantee uniformity of the finished product, the powdered fish scales were then sieved to remove big particles and obtain consistent particle size. Then, 15–50 ml of a boiling mixture of ethanol and deionized water (1:9 v/v) was used to dissolve 30–75 mg of finely powdered fish scales. The mixture was stored at 4 °C until it was needed again, guaranteeing its preservation and preparedness for further research.

2.2. Preparation of bioC-GO nanocomposites

We procured GO, KC, and IC from Sigma-Aldrich for our experimental investigations. Fish scale powder (30 mg) from biowaste sources was dissolved in boiling ethanol and deionized water. Then, 100 mg of IC and 30 mg of KC were combined with 20 ml of the biowaste solution, and the combination of bioC was stirred constantly for 3 h at around 70 °C. A digital sonicator fitted with a 10 mm diameter probe was used to treat the final solution, known as bioC-GO, to guarantee complete homogeneity of the bioC solution and GO dispersions (150 mg). The sonicator was programmed to run in pulse mode at a frequency of 20 kHz and an ultrasonic output of 750 W. To keep the level of the solutions constant during the sonication procedure, the sample beakers were carefully positioned in a water bath. After sonicating the solutions, we dried them for 24 h at 35 °C using the evaporative-casting procedure. The mixes were then transferred to Petri dishes. This procedure made it easier for the solutions to form films (see Figure 1), which allowed for more research and testing.

2.3. Characterization techniques

The properties of the synthesized bioC and bioC–GO nanocomposites were investigated using a range of characterization techniques. Using a Bruker D8 Discover X-ray diffractometer (Billerica, Massachusetts, USA), the crystalline structure and phase purity were examined throughout a scanning range of 1°–100°. An energy-dispersive X-ray spectroscopy (EDS) spectrometer (JSM-2100F, JEOL, USA) and a scanning electron microscope (JSM-7600F, JEOL, USA) were used for morphological and elemental investigations. A drop of the sonicated bioC–GO solution was applied to a carbon-coated conductive tape for scanning electron microscopy (SEM) and EDS measurements, and it was let to dry under a light before examination. A thermogravimetric analysis (TGA) (TGA/DSC 1) analyzer (Mettler Toledo, Columbus, USA) was used to evaluate thermal stability and breakdown. Nitrogen flow at a rate of 10 °C/min was used to evaluate around 10 mg of the dried produced film throughout a temperature range of 0–800 °C. The resulting thermograms illustrate weight loss and heat flow as a function of temperature. The electrical resistance of the films was assessed by measuring temperature-dependent current–voltage (I–V) characteristics. A digital multimeter and waveform generator were used for these tests, which were carried out in a temperature-controlled setting.

3. Results and discussion

Figure 2A shows the X-ray diffraction (XRD) pattern of bioC characterized by broader and less intense peaks, indicative of its amorphous nature. In contrast, Figure 2B illustrates the distinctive XRD peaks of the bioC–GO nanocomposites observed at 2θ angles of 20°, 27°, 42°, 49°, 73°, and 89°. These peaks reflect the structural contributions of fish scales, GO, and the KC–IC/FS (bioC) biopolymer matrix. The XRD pattern of pure gelatin powder exhibits a broad peak at $2\theta \approx 20^\circ$, consistent with its semi-crystalline nature and triple-helical structure [16,17]. The reduced intensity of the peak at 42° suggests graphene exfoliation within the bioC matrix, significantly altering the crystalline structure of GO [18]. Peaks at 42° and 49°, previously attributed to pure KC and IC [18–20], show diminished intensity in the bioC–GO nanocomposites, indicating modifications in the

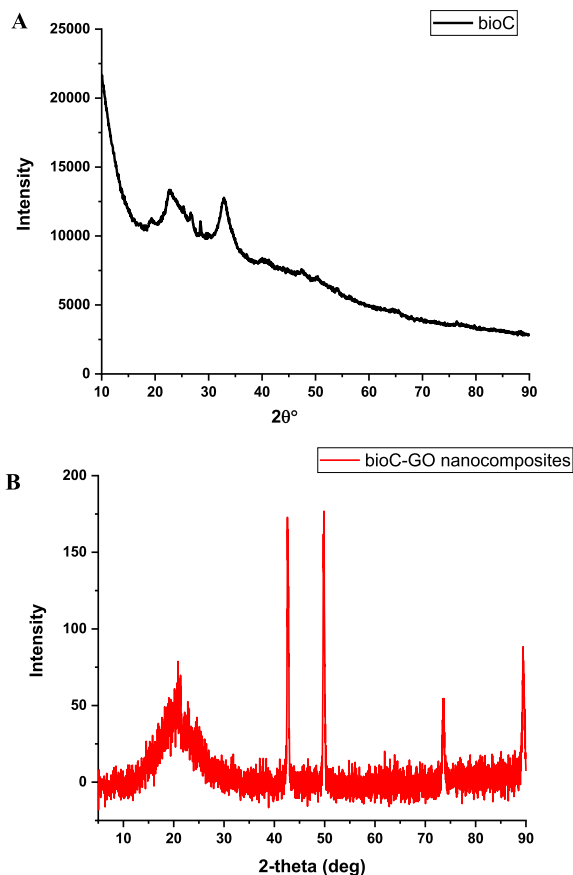


Figure 2. XRD patterns of (A) bioC film and (B) bioC–GO nanocomposite.

crystalline arrangement of these components. These findings provide valuable insights into the structural evolution of bioC–GO nanocomposites, underscoring the dynamic interactions between the biopolymer matrix and GO.

In Figure 3A, the SEM image shows that the bioC sample exhibits a compact and dense structure with a slightly flaky surface. The corresponding EDS spectrum (Figure 3B) confirms the presence of elements such as C, O, Na, Mg, S, Cl, K, Ca, and Fe, with the weight% of carbon measured as 30.44%. In contrast, Figure 3C reveals the morphology of the bioC–GO nanocomposites characterized by wrinkly surfaces resulting from the synthesis process. The SEM image demonstrates that the biopolymers uniformly cover the GO substrate. Figure 3D displays the EDS spectrum for the nanocomposite, identifying elements including C, O, Na, Mg, Si, S, Cl, K, and Ca, which are

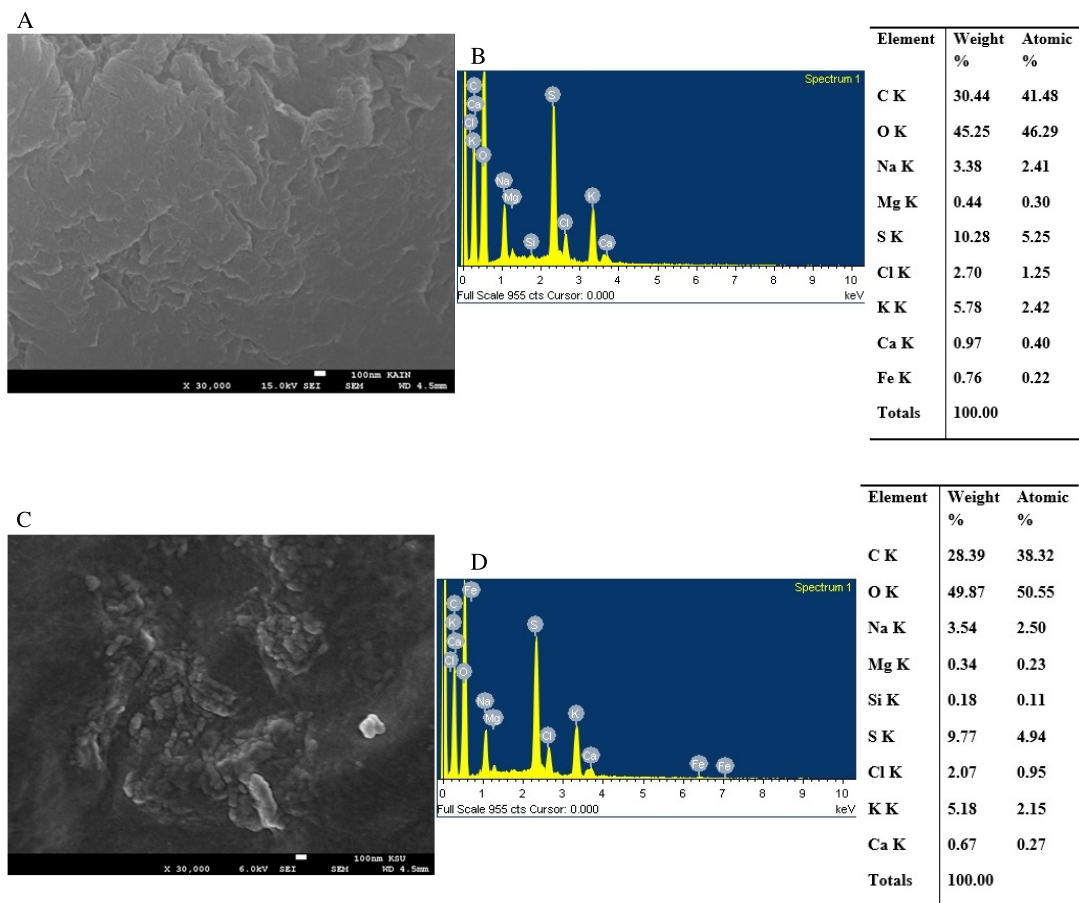


Figure 3. BioC film: (A) SEM image, (B) EDS patterns and a tabulation of the EDS spectrum; bioC-GO nanocomposites: (C) SEM image, (D) EDS patterns and a tabulation of the EDS spectrum.

attributed to the utilized substrate and incorporated materials [21]. From the tabulated data in Figure 3C, the carbon weight% in the bioC-GO nanocomposites is estimated as 28.34%, slightly lower than that of the bioC sample. Phiri *et al.* reported that functionalized GO can introduce oxygen-containing functional groups, which slightly increase the oxygen weight% in the composite [22]. These findings provide insights into the elemental composition and distribution within the bioC-GO nanocomposites, offering valuable information about their structural characteristics and potential applications [23,24].

Transmission electron microscopy (TEM) was used to assess the bioC-GO nanocomposite ink's morphology as shown in Figures 4A and B. Imaging using TEM revealed the presence of nanocomposite flakes with one or more layers. Although GO is

primarily clear, its surface frequently exhibits brightness changes due to elastic corrugations and scrolled or folded edges [25]. The TEM images revealed bioC-GO sheets that were extensively wrinkled, highlighting their distinct structural characteristics. After careful TEM investigation, it was determined that our samples' diameters fall within the nanometer range, highlighting their nanostructured nature [26]. Additionally, the samples' high level of disorder highlights how dynamic and complicated the bioC-GO nanocomposites are. Consequently, the other components in the composite help widen the interlayer gap between the GO sheets, preventing them from reassembling [27,28].

A thorough TGA of the bioC-GO nanocomposites was conducted in a nitrogen environment at a heating rate of 50 °C. The bioC and bioC-GO

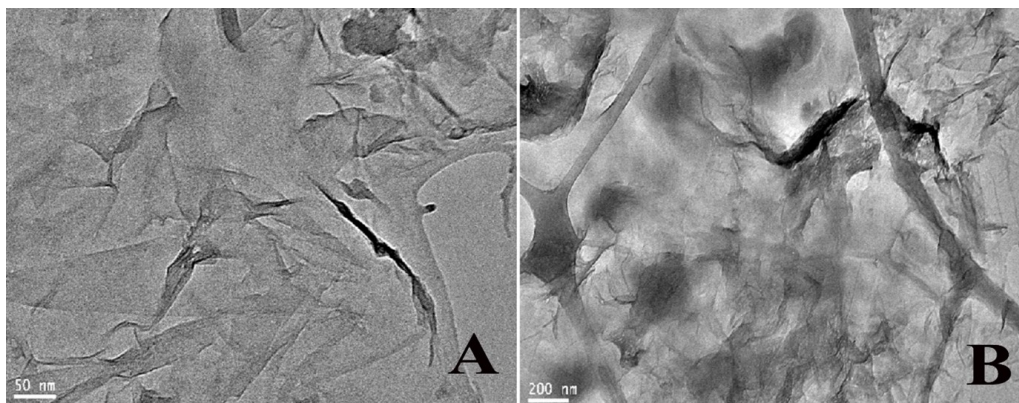


Figure 4. (A, B) TEM images with scale bars 50 nm and 200 nm, respectively.

nanocomposites were heated at a rate of 20 °C per minute from 100 °C to 800 °C. The TGA data displayed in Figures 5A and B provide a wealth of information on the thermal characteristics of the bioC and nanocomposites [29].

In Figure 5A, the TGA curve of a composite made of KC, IC, and fish scale powder reveals the main thermal decomposition stages of the bioC. Initially, weight loss occurs between approximately 50 and 200 °C, which is attributed to the evaporation of absorbed and bound water due to the hydrophilic nature of KC, IC, and fish scale powder. A subsequent weight loss is observed in the range of 200–300 °C, corresponding to the thermal degradation of carrageenan. This involves the breakdown of glycosidic bonds and the release of volatile products. Another significant weight loss occurs between 300 and 450 °C, primarily due to the thermal decomposition of collagen proteins in the fish scale powder along with the remaining organic matter from the carrageenan. Beyond 450 °C, additional weight loss is associated with partial demineralization or structural transformations of mineral components, such as hydroxyapatite, present in the fish scale powder. At temperatures exceeding 600 °C, the residual mass consists predominantly of inorganic material, primarily reflecting the mineral content of the composite [30].

In Figure 5B, approximately 15–20% of the total weight loss in the nanocomposites is attributed to the main chain breakdown, which begins at 200–250 °C. The bioC–GO nanocomposites are hypothesized to exhibit significantly higher thermal stabil-

ity, as they demonstrate only 45% weight loss at 800 °C, with complete combustion not occurring until around 800 °C [31]. Interestingly, weight loss due to solvent evaporation is observed at lower temperatures. Previous research by Pati *et al.* [32] suggests that the decomposition of graphene nanocomposites occurs in the temperature range of 200–280 °C. Meanwhile, polymer breakdown takes place between 350 and 480 °C. According to Tapas Mitra *et al.*, collagen-functionalized nanographene oxide (bioC–GO) scaffolds begin to degrade predominantly after 350 °C. Based on weight loss rates at 400 °C, 800 °C, and 900 °C, bioC–GO scaffolds demonstrate far greater thermal stability compared to native collagen. This enhanced stability is likely due to covalent interactions between nanographene oxide and collagen [33]. Furthermore, 47% of the amine groups in the bioC–GO scaffold are involved in intermolecular interactions [34]. These findings not only highlight the improved thermal stability of bioC–GO nanocomposites but also open avenues for further research and potential applications in various industries [35].

When fish scales are added to GO and KC or IC composites, the resulting composite's performance and thermal stability can be improved over earlier research outcomes [36–41]. According to earlier research studies, the following are some possible methods for enhancing thermal stability when fish scales are added. The hierarchical structure of collagen and mineral layers in fish scales contributes to their stiffness and flexibility. The resultant composite is more flexible and less prone to break or distort under

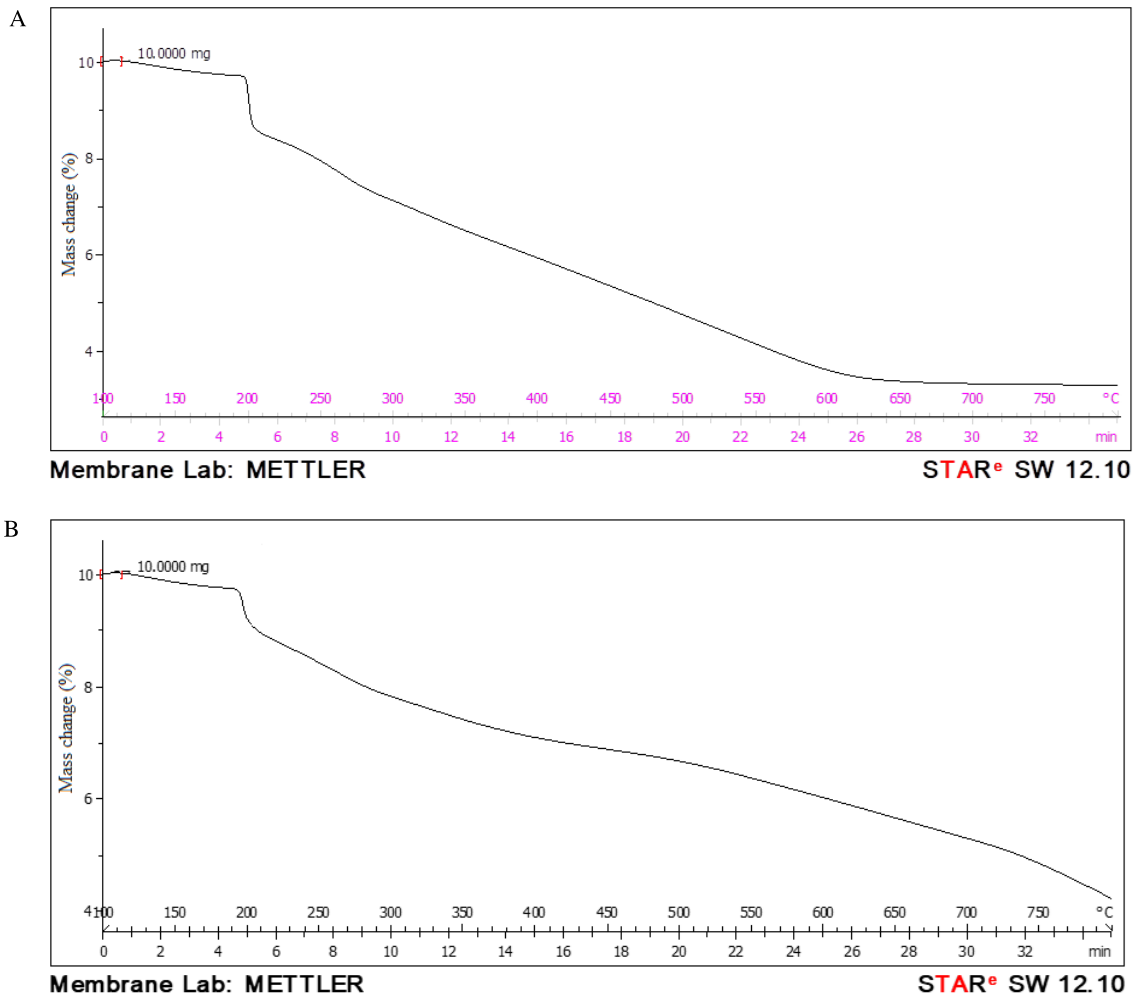


Figure 5. TGA curves of (A) bioC film and (B) bioC-GO nanocomposites.

stress when the mechanical rigidity of GO and the gel-like elasticity of carrageenan are combined. This synergy in mechanical properties also enhances thermal stability, as a well-integrated composite better resists thermal expansion and contraction. Likewise, when the material is subjected to mechanical or thermal stress, collagen from fish scales aids in energy dissipation. These characteristics delay structural collapse by allowing the composite to absorb and transfer energy more efficiently, resulting in increased thermal stability. By serving as a heat sink and dispersing thermal energy throughout the composite matrix, GO additionally supports this process [42–45].

At room temperature, the resistivity response of the bioC-GO nanocomposites under forward and reverse bias was examined at different bias voltages as shown in Figure 6A. Furthermore, the impact of temperature on the sample's DC conductivity was investigated using bias voltages of 5 V up to 150 °C as shown in Figure 6B. The resistivity measurements performed can be fitted by the one-dimensional variable range hopping model, using the following formula:

$$\rho_T = \rho_0 \exp \left[\left(\frac{T_0}{T} \right)^{\frac{1}{2}} \right]$$

where the sample T_0 stands for the energy barrier between localized states. The temperature-dependent

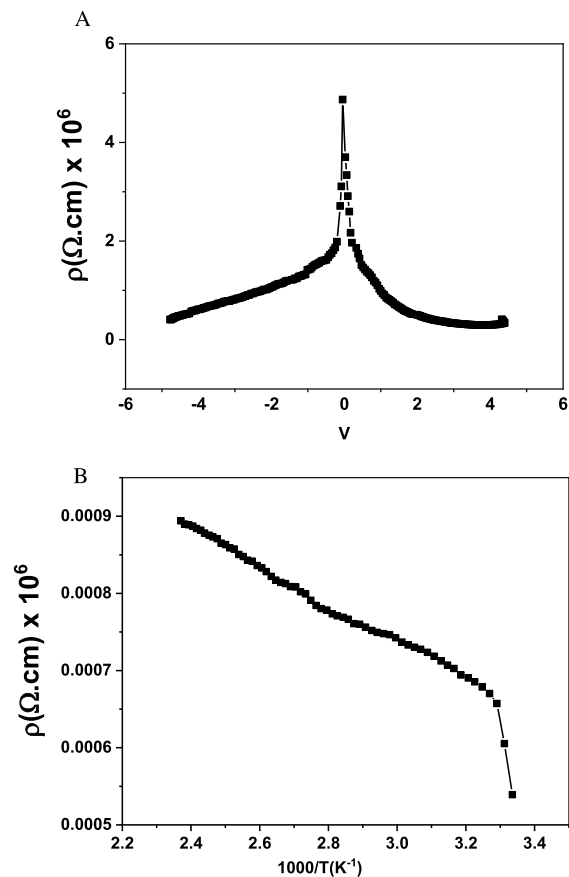


Figure 6. (A) Resistivity response under forward and reverse bias at room temperature and (B) resistivity change with respect to temperature for bioC–GO nanocomposites.

behavior of the produced bioC–GO nanocomposites yielded interesting results. The resistivity values, which are inversely proportional to conductivity, were found to be very low for the bioC–GO nanocomposites when exposed to temperatures as high as 150 °C at a steady applied voltage of 5 V. Consequently, the conductivity of the nanocomposites showed a significant increase with rising temperature, highlighting the dynamic nature of the material's conductivity. Furthermore, the temperature dependence of the resistivity measurements revealed several noteworthy patterns [46–48]. At lower temperatures, the localized electrons lacked sufficient thermal energy to overcome potential barriers, resulting in the lowest conductivity values. As the temperature increased, the conduction pro-

cess accelerated, reaching maximum efficiency at higher temperatures [49,50]. The promising properties of the bioC–GO nanocomposites emphasize their adaptability and potential for various technological applications, including their use as cathode materials in batteries and electronic devices [51].

4. Conclusion

This study presents the development and characterization of graphene-based bioC–GO nanocomposites specifically engineered for applications requiring enhanced thermal stability, electrical conductivity, and environmentally friendly properties. The structural and morphological analyses, including TGA, SEM, EDS, TEM, and XRD, confirmed the uniform dispersion and strong covalent interactions within the bioC–GO matrix. Our findings demonstrate that the resistivity values of bioC–GO nanocomposites remain very low even at temperatures as high as 150 °C at a steady applied voltage of 5 V. This indicates a significant increase in conductivity with rising temperature, highlighting the dynamic and thermally responsive nature of the material. These results indicate that bioC–GO nanocomposites hold promise as cathode materials for energy storage systems, such as batteries, and for use in electronic devices requiring stable conductivity under high-temperature conditions.

Declaration of interests

The authors do not work for, advise, own shares in, or receive funds from any organization that could benefit from this article, and have declared no affiliations other than their research organizations.

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