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STUDY THE ELECTRICAL PROPERTIES OF GRAPHENE OXIDE – NANOCELLULOSE COMPOSITE

Abstract

This study investigates the electrical properties of a graphene oxide (GO) and nanocellulose (NC) composite using impedance spectroscopy, complemented by thorough characterization through Fourier-transform infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS), Raman spectroscopy, and scanning electron microscopy (SEM). FTIR analysis revealed characteristic peaks corresponding to functional groups present in both GO and NC, providing insights into their chemical composition. XPS spectra exhibited distinctive peaks indicative of carbon and oxygen bonding states, elucidating the surface chemistry of the materials. Raman spectroscopy provided information on the structural order and defects within the samples, particularly highlighting the graphitic structure of GO. SEM images revealed the morphological features of the composite membrane, showcasing the distribution of NC particles and structural modifications induced by their incorporation. Impedance spectroscopy was utilized to investigate the electrical conductivity of the GO-NC composite. Results indicated a temperature-dependent behavior, with an increase in conductance observed as the temperature rose within the operational range of fuel cells. Remarkably, the addition of NC did not significantly alter the conductive behavior of the composite, suggesting compatibility and stability. In summary, this comprehensive characterization using multiple analytical techniques offers valuable insights into the electrical properties of the GO-NC composite. The findings suggest its potential for various applications requiring enhanced electrical conductivity, particularly in fuel cell technology.

Key words: Proton Exchange Membrane, Graphene Oxide, Nanocellulose, Membrane Electrode Assembly, Composite, Impedance Spectroscopy.

Introduction

In recent years, the scientific community has been captivated by the extraordinary properties of graphene, graphene oxide (GO), and other carbon-based materials. These materials exhibit a plethora of unique characteristics, including exceptional physical-mechanical attributes, electronic properties, high thermal conductivity, hardness, and strength, along with remarkable adsorption capabilities [1–3]. Particularly noteworthy is the widespread utilization of graphene oxide membranes in the fabrication of sensors and supercapacitors sensitive to environmental factors such as moisture, temperature, and organic gases.

Graphene oxide, derived from graphite, possesses a layered structure enriched with diverse oxygen-based functional groups, distinguishing it from pristine graphene. Historically, the synthesis of graphene oxide can be traced back to the 1859 work of Brodie, who chemically treated graphite to produce monolayer sheets of graphene oxide. Since then, various methods for synthesizing GO have been developed, with the modified Hammer method emerging as the most prevalent due to its efficiency [2–5].

Activated carbon serves as a pivotal raw material for the synthesis of graphene oxide, owing to its cost-effectiveness, high adsorption capacity, and ample surface area. Despite the universal appeal of GO, its susceptibility to structural instability in humid environments poses a significant challenge. Consequently, researchers have embarked on extensive modification efforts to enhance the stability and functionality of graphene oxide through chemical treatments with substances like hydrazine hydrate, dimethylhydrazine, hydroquinone, and NaBH_4 [4–9].

The recent surge in research endeavors has focused on the development of novel nanocomposites by integrating graphene oxide with nanocellulose. Nanocellulose, derived from natural cellulose, possesses remarkable physical, chemical, biological, magnetic, electrical, and optical properties at the nanoscale [7–14]. The synthesis of nanocellulose involves several stages, including the extraction of extractives/hemicellulose, separation of cellulose unit fibrils, and solvent removal, resulting in a material akin to graphene in terms of size, surface area, mechanical and chemical properties, crystallinity, biodegradability, and renewability.

The combination of nanocellulose with graphene oxide holds tremendous potential for the development of functional hybrid composites, offering enhanced mechanical reinforcement, biocompatibility, and environmental sustainability. Moreover, graphene oxide membranes have emerged as promising candidates for water purification applications, demonstrating superior sheet structure, efficient separation, and flexible fabrication methods [8–15]. Multilayer graphene oxide membranes have been shown to effectively trap water contaminants, facilitating rapid water molecule flow with minimal friction between layers.

The integration of carboxymethylcellulose (CMC) into graphene oxide matrices further expands the scope of potential applications, leveraging CMC's biocompatible and biodegradable nature, along with its excellent film-forming properties [13–24]. The synergistic combination of graphene oxide and carboxymethylcellulose offers opportunities for tailored functionalization, improved mechanical strength, flexibility, and stability, making it a promising candidate for various electronic devices and sensors.

In this work, we aim to investigate the electrical properties of the graphene oxide–carboxymethylcellulose composite using impedance spectroscopy. By elucidating the charge transport mechanisms, dielectric properties, and interfacial phenomena within the composite material, we seek to contribute to the advancement of graphene-based materials for diverse technological applications. Through interdisciplinary research at the intersection of nanotechnology, materials science, and electrical engineering, we aspire to unlock the full potential of graphene oxide composites for addressing pressing societal challenges and fostering sustainable technological innovation.

Materials and methodology

Two distinct samples, comprising graphene oxide (GO) and graphene oxide with nanocellulose (GO+NC), were synthesized utilizing the Hammer method, where thickness of GO is 18.03-23.5 micron, while the thickness of GO+NC (1/1) is 12.36 micron. The morphology of the synthesized samples was examined using a SEM Crossbeam 540 research instrument, with the results depicted in Figure 1A-D.

Further characterization of the samples involved infrared (IR) spectroscopy, conducted utilizing a Nicolet iS10 IR Fourier spectrometer (Figure 2A), and Raman spectroscopy (Figure 2B). These spectroscopic techniques provided insights into the chemical composition and structural properties of the synthesized materials.

Subsequently, X-ray photoelectron spectroscopy (XPS) was employed to analyze the spectra of the GO and GO+NC samples, revealing variations in binding energy, as illustrated in Figure 3 (GO: 280-290 eV, GO+NC: 290-300 eV).

To investigate the electrical characteristics of the samples, impedance spectroscopy was employed across various temperature ranges. The experimental setup utilized an HF₂ Impedance Spectroscopy system from Zurich Instruments. To ensure reliable electrical contact, a copper element was deposited onto the surface of each sample via magnetron sputtering, with a plasma power of 40 W and a deposition time of 30 minutes. Impedance measurements were conducted at a sinusoidal voltage amplitude of 50 mV, spanning a frequency range from 10 Hz to 1 MHz.

The obtained impedance spectroscopy data, depicted in Figure 4, provided valuable insights into the electrical properties of the graphene oxide-carboxymethylcellulose composite. The measured impedance spectra facilitated the analysis of charge transport mechanisms, dielectric properties, and interfacial phenomena within the composite material, thereby contributing to a comprehensive understanding of its electrical behavior.

Overall, this methodology encompassed a series of experimental techniques aimed at characterizing the synthesized graphene oxide-carboxymethylcellulose composite and elucidating its electrical properties through impedance spectroscopy. Through systematic analysis and interpretation of the obtained data, we aimed to advance the understanding of this composite material and its potential applications in various technological domains.

Results and discussion

Figure 1 presents cross-sectional scanning electron microscope (SEM) images (Figure 1A, B) showcasing the structural characteristics of both graphene oxide (GO) and graphene oxide with nanoclay (GO+NC) samples. In terms of thickness, the measurements indicate a range between 18.03 to 23.5 microns for GO and a slightly reduced thickness of 12.36 microns for GO+NC [25]. This reduction in thickness hints at a potential compaction effect induced by the incorporation of nanoclay into the composite membrane [25].

Analyzing the cross-SEM images of the GO/NC samples reveals distinct features, notably the presence of compact lamellar structures compared to the initial GO membrane. The inclusion of nanoclay seems to contribute to the densification of the membrane structure, leading to enhanced compactness. Furthermore, a comparative assessment between the surface morphologies of the original GO and GO/NC samples highlights significant differences.

In particular, the SEM images depict the presence of small nanoclay particles dispersed throughout the GO matrix in the GO/NC samples. Additionally, there are observable surface wrinkles in the GO/NC samples, suggesting some degree of aggregation. These surface features indicate a structural modification induced by the incorporation of nanoclay, altering the surface morphology of the composite membrane.

The presence of nanoclay particles and the observed surface alterations signify the successful integration of nanoclay into the GO matrix. The slight aggregation observed may be attributed to

the interaction between GO and nanoclay particles, leading to localized clustering. Overall, the SEM analysis provides valuable insights into the structural characteristics of GO/NC composite membranes, highlighting the impact of nanoclay incorporation on membrane morphology and structure.

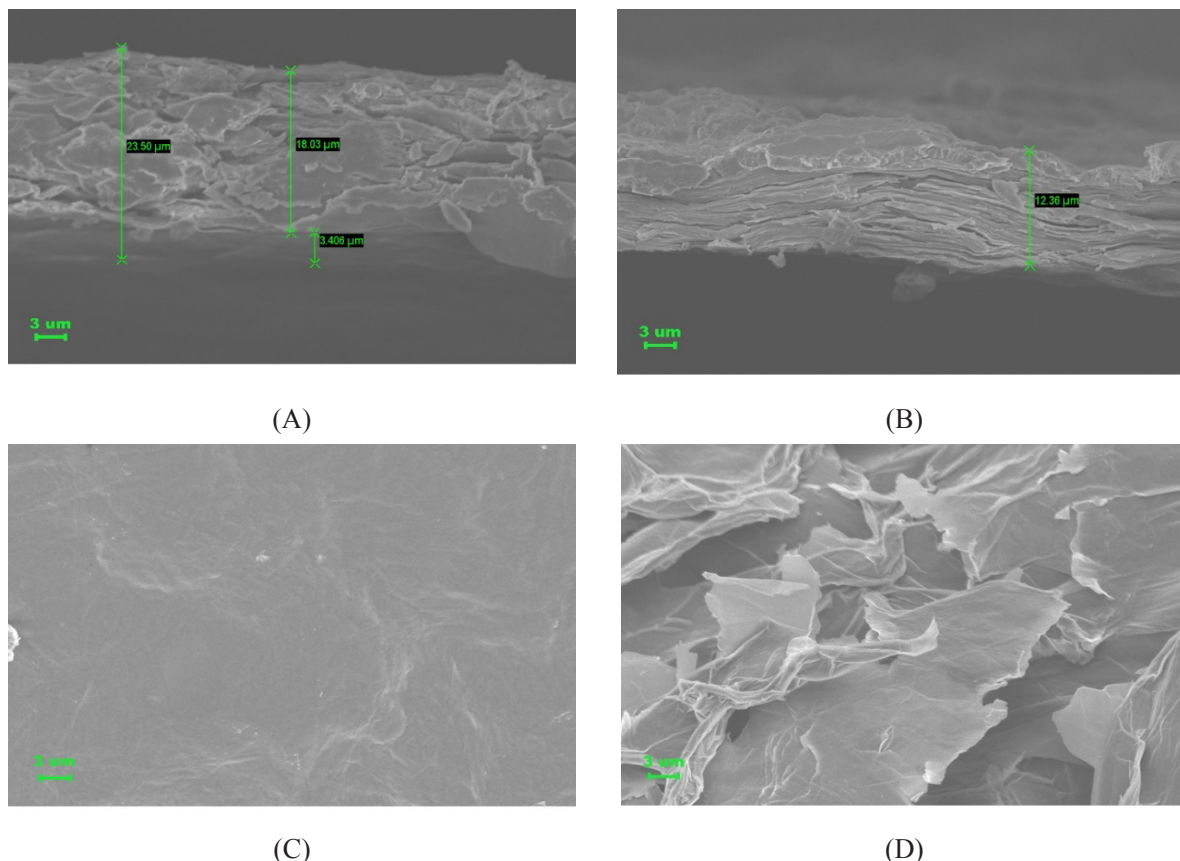


Figure 1 – Cross- and top-SEM images of GO (A, C) and GO+NC (B, D) samples

The chemical structure of the samples was meticulously examined through Fourier-transform infrared spectroscopy (FTIR), Raman spectroscopy, and X-ray photoelectron spectroscopy (XPS). Each technique provides unique insights into the molecular composition and bonding characteristics of the materials under study.

In the case of nanoclay (NC), FTIR analysis revealed characteristic peaks at specific wavenumbers: 3300 cm^{-1} corresponding to the stretching vibrations of O-H bonds, 1650 cm^{-1} representing asymmetric stretching vibrations of C=O bonds, 1411 cm^{-1} associated with the bending vibration of C-H bonds, 1344 cm^{-1} indicating the bending vibration of C-O-C bonds, and 1004 cm^{-1} attributed to specific structural features [26]. These peaks provide crucial information about the molecular structure and functional groups present in NC.

On the other hand, pure graphene oxide (GO) exhibited distinct spectral features in its FTIR spectrum. Strong bands were observed at 3338 cm^{-1} corresponding to O-H stretching vibrations, 1721 cm^{-1} indicative of C=O stretching vibrations, 1610 cm^{-1} representing C=C stretching vibrations, and 1004 cm^{-1} associated with C-O-C bending vibrations [27–29]. These characteristic peaks elucidate the chemical composition and bonding configuration of GO.

Upon combining GO with NC, FTIR analysis of the composite membrane unveiled a spectrum with additional peaks and altered intensities. In the wavelength range of $3600\text{--}3000\text{ cm}^{-1}$, prominent O-H stretching vibrations were observed, indicating the presence of hydroxyl groups. A peak at 1610 cm^{-1} suggested the existence of aromatic rings, while signals at 1249 cm^{-1} and 1054 cm^{-1} were

attributed to C-O bonds, including epoxy and alkoxy groups, respectively. Notably, new ether bonds (C=O) emerged at 1715 cm^{-1} , indicating a chemical interaction between GO and NC [30, 31].

Raman spectroscopy complemented the FTIR analysis by providing information about the structural order and defects within the samples. XPS, on the other hand, offered insights into the elemental composition and chemical states of the surface atoms. Through the combination of these analytical techniques, a comprehensive understanding of the chemical structure and bonding characteristics of the GO/NC composite membrane was achieved, laying the foundation for further elucidating its properties and potential applications.

In a typical Raman spectrum (Figure 2B) of graphene oxide (GO), a prominent G band is discernible. This G band manifests as a peak at 1605 cm^{-1} , representing the E_{2g} phonon of the sp^2 carbon atoms, indicative of the graphitic structure. Additionally, a characteristic D band appears at 1353 cm^{-1} , corresponding to the disorder-induced point phonon K absorption mode of A_{1g} symmetry. These spectral features are crucial for assessing the structural integrity and quality of GO materials.

Remarkably, upon comparison of the Raman spectra of GO and GO+NC samples, no noticeable difference in the Raman shift was observed. This lack of distinction implies that the incorporation of nanoclay (NC) did not induce significant alterations in the structural properties of GO as detected by Raman spectroscopy. The consistent Raman features suggest that the introduction of NC did not introduce detectable changes in the graphitic structure or the degree of disorder within the GO matrix.

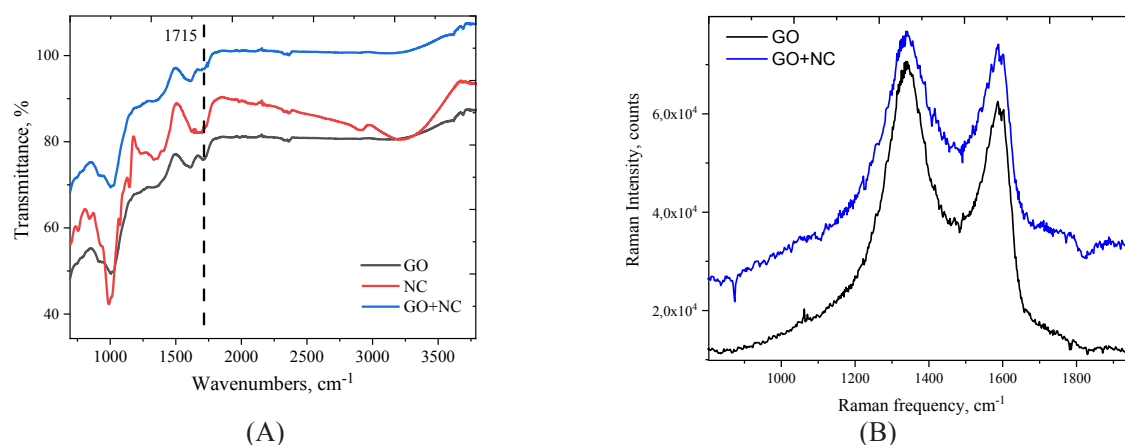


Figure 2 – FTIR (a) and Raman spectra (b) of GO, NC and NC+GO samples

The absence of discernible shifts in the Raman peaks between GO and GO+NC samples implies that the addition of nanoclay did not induce substantial structural modifications at the molecular level. This finding underscores the compatibility of nanoclay with the GO matrix, suggesting a stable composite formation without significant disturbance to the intrinsic properties of GO. Overall, Raman spectroscopy provides valuable insights into the structural characteristics and integrity of GO and its composites, facilitating a comprehensive understanding of their properties and potential applications.

Figure 3 presents both high and low-resolution X-ray photoelectron spectroscopy (XPS) spectra of nanoclay (NC), graphene oxide (GO), and their composite (NC-GO) samples, offering valuable insights into their elemental composition and chemical bonding states.

In the low-resolution XPS spectra (Figure 3A), characteristic signals of carbon (C 1s) at 284.6 eV and oxygen (O 1s) at 531.5 eV are evident across all samples. These signals serve as primary indicators of the presence of carbonaceous and oxygen-containing functional groups, respectively, in the analyzed materials.

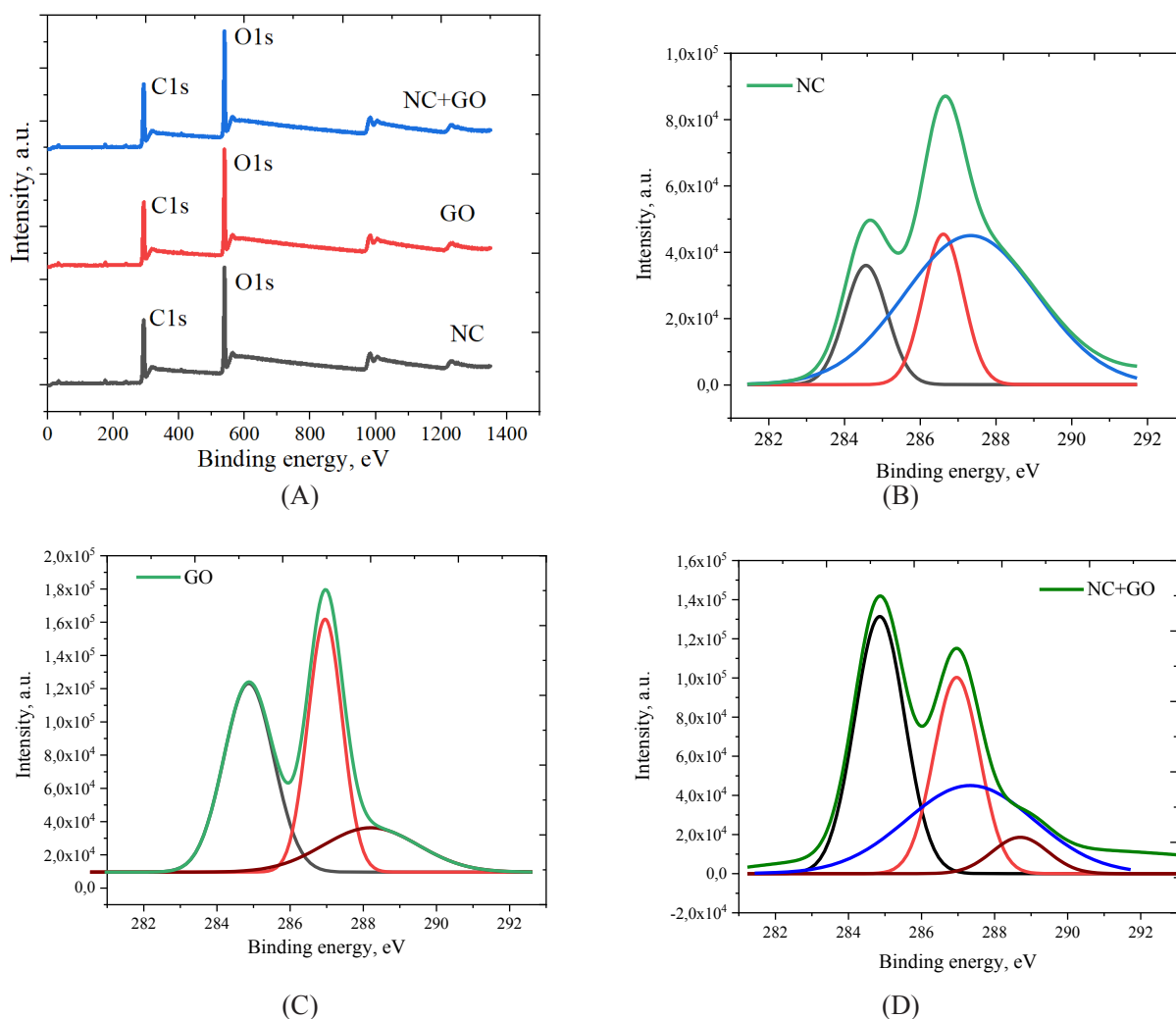


Figure 3 – General XPS spectra (A) and high-resolution C 1s spectra of NC(B), GO(C) and GO+NC(D) samples

Further analysis through high-resolution XPS spectra of NC reveals distinctive features in the C 1s region (Figure 3B). The spectrum exhibits Gaussian peaks fitted with three components centered at 284.6 eV, 286.3 eV, and 287.6 eV, corresponding to carbon-carbon (C-C/C-H), C-O, and C=O groups, respectively. These peaks elucidate the chemical environments and bonding configurations of carbon atoms within the NC structure.

Moving to the high-resolution XPS spectrum of GO (Figure 3C), the C 1s peak can be deconvoluted into three discernible components centered at 284.3 eV, 286.6 eV, and 288.2 eV. These peaks are attributed to sp^2 carbon (C_{sp^2}), C-O, and C=O bonds, respectively. The presence of these functional groups is consistent with the typical chemical composition of graphene oxide.

Upon analyzing the C 1s spectra of the GO-NC composite (Figure 3D), a complex pattern emerges. Three main chemical environments of carbon are observed: a dominant peak at 286.8 eV corresponding to C-O bonds, indicating the presence of oxygen functional groups; a peak at 284.7 eV attributed to C-C bonds, suggesting the presence of graphitic carbon; and a third peak around 287.7 eV indicative of O-C-O bonds, likely originating from carboxyl groups. Additionally, a minor fraction at approximately 289 eV is detected, suggesting the emergence of carboxyl groups following the modification process.

These XPS spectra provide detailed information about the chemical composition and bonding states of carbon and oxygen within the analyzed samples. The distinctive peaks and their relative

intensities offer insights into the surface chemistry and structural modifications induced by the incorporation of nanoclay into graphene oxide. Such characterization is crucial for understanding the interactions and compatibility between the constituents in composite materials, aiding in the optimization of their properties for various applications ranging from catalysis to energy storage.

Figure 4 illustrates the disparity in impedance spectroscopy between graphene oxide (GO) and GO+NC (graphene oxide with nanoclay) samples, shedding light on their electrical properties. Additionally, the temperature dependence of these research samples was investigated, unveiling intriguing insights into their conductive behavior under varying thermal conditions.

The impedance spectroscopy data reveals distinct characteristics between GO and GO+NC samples. Notably, as depicted in Figure 4, the conductance of both materials exhibits a temperature-dependent behavior. Interestingly, the conductance of graphene oxide appears comparable to that of GO+NC across different temperature regimes. With increasing temperature, the conductance of both samples demonstrates an upward trend. This observation suggests that temperature plays a significant role in enhancing the electrical conductivity of these materials.

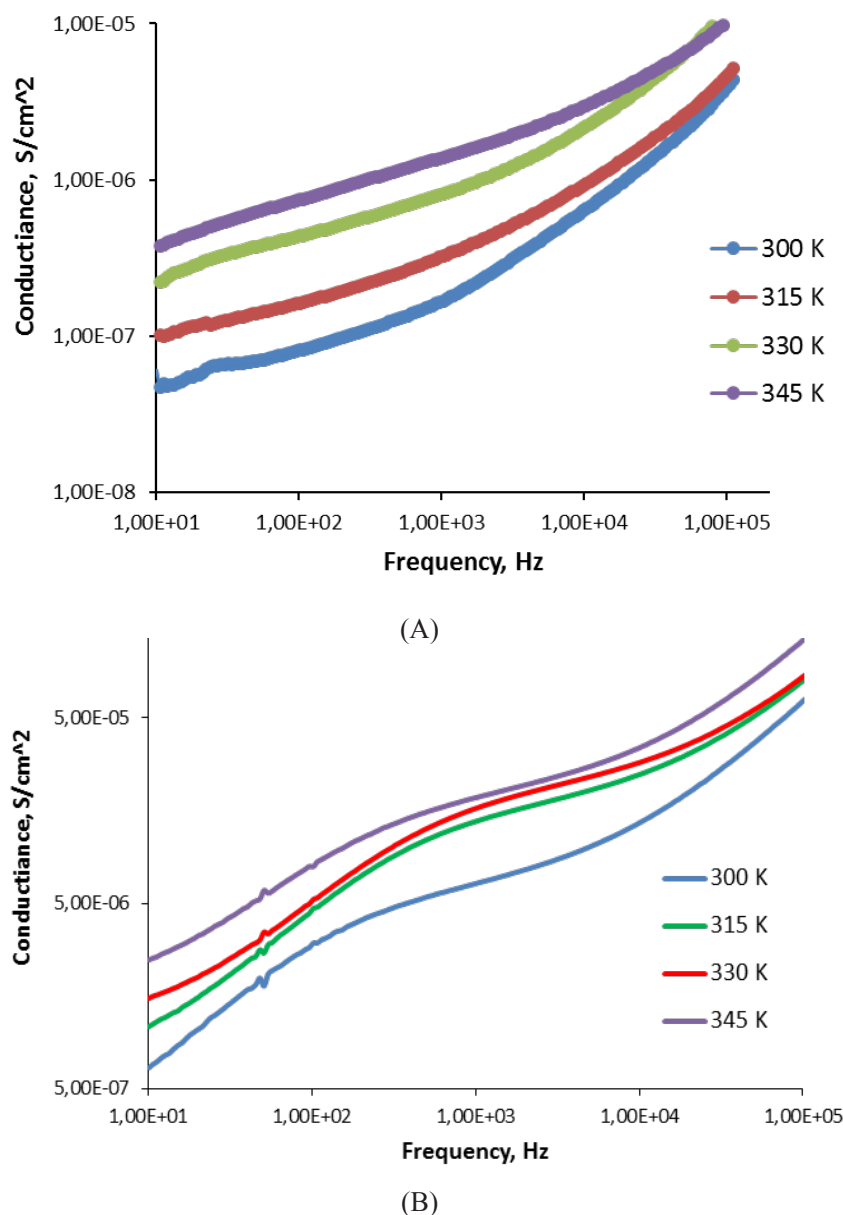


Figure 4 – Impedance Spectroscopy data for (A) GO, and (B) GO + NC

Furthermore, the relevance of these findings in the context of fuel cell applications becomes apparent through empirical studies. The operating temperature range of fuel cells typically falls within 300-345 K, with an optimal temperature of around 350 K. The conducted research reveals that the conductance of both graphene oxide and GO+NC samples experiences an increase within this temperature range. Specifically, the conductance elevates from 10^{-7} to 10^{-6} S/cm² as the temperature rises. This augmentation in conductance is particularly promising for fuel cell applications, as it suggests improved electrical conductivity at the operational temperatures of fuel cells.

It's important to note that the frequency range considered in these studies spans from 10 to 10^5 Hz. This frequency range is relevant for assessing the electrical properties of materials under varying conditions, providing valuable insights into their conductivity across different frequencies.

The implications of these findings are significant in the realm of fuel cell technology. Fuel cells rely on efficient ion conduction to facilitate electrochemical reactions, and materials exhibiting high conductance at operational temperatures are highly desirable. The observed increase in conductance with temperature suggests that both graphene oxide and GO+NC hold promise as potential candidates for fuel cell applications.

Moreover, the incorporation of nanoclay into graphene oxide does not appear to significantly alter the temperature-dependent conductive behavior, indicating the compatibility and stability of the composite material. This suggests that the addition of nanoclay may enhance certain properties without compromising the overall electrical conductivity of the composite.

The impedance spectroscopy data coupled with the temperature dependence studies provide valuable insights into the electrical properties of graphene oxide and its composite with nanoclay. The observed increase in conductance with temperature within the operating range of fuel cells highlights the potential suitability of these materials for fuel cell applications, paving the way for further exploration and optimization in this field.

Conclusion

The study focused on investigating the electrical properties of a graphene oxide (GO) and nanocellulose (NC) composite using impedance spectroscopy, complemented by an in-depth analysis of the material's chemical structure and morphology through Fourier-transform infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS), Raman spectroscopy, and scanning electron microscopy (SEM).

FTIR analysis revealed characteristic peaks corresponding to functional groups present in both GO and NC, providing insights into their chemical composition. Similarly, XPS spectra exhibited distinctive peaks indicative of carbon and oxygen bonding states, elucidating the surface chemistry of the materials. Raman spectroscopy provided information on the structural order and defects within the samples, particularly highlighting the graphitic structure of GO. SEM images revealed the morphological features of the composite membrane, showcasing the distribution of NC particles and structural modifications induced by their incorporation.

Impedance spectroscopy was employed to investigate the electrical conductivity of the GO-NC composite. The results indicated a temperature-dependent behavior, with an increase in conductance observed as the temperature rose within the operational range of fuel cells. Notably, the addition of NC did not significantly alter the conductive behavior of the composite, suggesting compatibility and stability.

Overall, the comprehensive characterization using multiple analytical techniques provided valuable insights into the electrical properties of the GO-NC composite. The findings suggest that the composite holds promise for various applications requiring enhanced electrical conductivity, particularly in the realm of fuel cell technology. Further optimization and exploration of these materials are warranted to unlock their full potential for practical applications.

In conclusion, the study offers a thorough investigation into the electrical properties of the GO-NC composite, providing a foundation for further research and development in this field. The combination of impedance spectroscopy with FTIR, XPS, Raman spectroscopy, and SEM analysis offers a comprehensive understanding of the material's structure, morphology, and electrical behavior. This work contributes to the ongoing efforts to harness the unique properties of graphene-based composites for advanced applications in various industries, including energy storage and conversion.

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REFERENCES

- 1 Yu W., Sisi L., Haiyan Y., Jie L. Progress in the functional modification of graphene/graphene oxide: A review. *RSC advances*, 2020, vol. 10, no. 26, pp.15328–15345.
- 2 Han B., Zou Y., Xu G., Hu S., Kang Y., Qian Y., Gu M. Additive stabilization of SEI on graphite observed using cryo-electron microscopy. *Energy & Environmental Science*, 2021, vol. 14, no. 9, pp. 4882–4889.
- 3 Li C., Chen X., Shen L., Bao N. Revisiting the oxidation of graphite: Reaction mechanism, chemical stability, and structure self-regulation. *ACS omega*, 2020, vol. 5, no. 7, pp. 3397–3404.
- 4 Sharma N., Swami S., Shrivastava V., Nair R., Shrivastava R. Graphene oxide and functionalized graphene oxide: Robust, 2D material as heterogeneous green catalyst for heterocyclic synthesis. *Materials today: proceedings*, 2021, vol. 43, pp. 3309–3317.
- 5 Alkhouzaam A., Qiblawey H., Khraisheh M., Atieh M., Al-Ghouti M. Synthesis of graphene oxides particle of high oxidation degree using a modified Hummers method. *Ceramics International*, 2020, vol. 46, no. 15, pp. 23997–24007.
- 6 Verma V., Gurnani L., Das A., Kumari N., Dasgupta T., Mukhopadhyay A. Reduced graphene oxides prepared via explosive and non-explosive thermal reduction: Structural evolution, functional properties and reinforcing efficacy. *Carbon*, 2023, vol. 209, p. 118007.
- 7 Liang K., Spiesz E.M., Schmieden D. T., Xu A.W., Meyer A.S., Aubin-Tam M.E. Bioproduced polymers self-assemble with graphene oxide into nanocomposite films with enhanced mechanical performance. *ACS nano*, 2020, vol.14, no.11, pp. 14731–14739.
- 8 Thomas R., Balachandran M. Luminescence and energy storage characteristics of coke-based graphite oxide. *Materials Chemistry and Physics*, 2021, vol. 257, p. 123854.
- 9 Han Z., Xiao X., Qu H., Hu M., Au C., Nashalian A., Chen J. Ultrafast and selective nanofiltration enabled by graphene oxide membranes with unzipped carbon nanotube networks. *ACS Applied Materials & Interfaces*, 2021, vol. 14, no. 1, pp. 1850–1860.
- 10 Cha M., Boo C., Song I.H., Park C. Investigating the potential of ammonium retention by graphene oxide ceramic nanofiltration membranes for the treatment of semiconductor wastewater. *Chemosphere*, 2022, vol. 286, p. 131745.
- 11 Yang K., Pan T., Hong S., Zhang K., Zhu X., Chen B. Ultrathin graphene oxide membrane with constructed tent-shaped structures for efficient and tunable molecular sieving. *Environmental Science: Nano*, 2020, vol. 7, no. 8, pp. 2373–2384.
- 12 Zhang W., Huang Q., Liu S., Zhang M., Liu G., Ma Z., Jin W. Graphene oxide membrane regulated by surface charges and interlayer channels for selective transport of monovalent ions over divalent ions. *Separation and Purification Technology*, 2022, vol. 291, p. 120938.
- 13 Keerthi A., Goutham S., You Y., Iamprasertkun P., Dryfe R.A., Geim A.K., Radha B. Water friction in nanofluidic channels made from two-dimensional crystals. *Nature communications*, 2021, vol. 12, no. 1, p. 3092.

- 14 Beroual M., Mehelli O., Boumaza L., Trache D., Tarchoun A.F., Derradji M., Khimeche K. Synthesis and characterization of microcrystalline cellulose from giant reed using different delignification processes. *Materials Research and Applications: Select Papers from JCH8-2019*, 2021, pp. 173–187.
- 15 Tshikovhi A., Mishra S.B., Mishra A.K. Nanocellulose-based composites for the removal of contaminants from wastewater. *International Journal of Biological Macromolecules*, 2020, vol. 152, pp. 616–632.
- 16 Fahma F., Febiyanti I., Lisdayana N., Arnata I.W., Sartika D. Nanocellulose as a new sustainable material for various applications: A review *Archives of Materials Science and Engineering*, 2021, vol. 109, no. 2, pp. 49–64.
- 17 Kawalerczyk J., Dziurka D., Mirski R., Szentner K. Properties of plywood produced with urea-formaldehyde adhesive modified with nanocellulose and microcellulose. *Drvna industrija*, 2020, vol. 71, no.1, pp. 61–67.
- 18 Wijesekara W.I., Gokila S., Gomathi T., Prasad S., Deepa M., & Sudha P.N. New Opportunities and Future Prospects. *Nanobiomaterials: Research Trends and Applications*, 2023, 314.
- 19 Zulaikha W., Hassan M.Z., & Ismail Z. Recent development of natural fibre for nanocellulose extraction and application. *Materials Today: Proceedings*, 2022, no. 66, pp. 2265–2273.
- 20 Xu T., Du H., Liu H., Liu W., Zhang X., Si C., ... & Zhang K. Advanced nanocellulose-based composites for flexible functional energy storage devices. *Advanced materials*, 2021, vol. 33, no. 48, p. 2101368.
- 21 Thomas P., Duolikun T., Rumjit N.P., Moosavi S., Lai C.W., Johan M.R.B., & Fen L.B. Comprehensive review on nanocellulose: Recent developments, challenges and future prospects. *Journal of the mechanical behavior of biomedical materials*, 2020, no. 110, p. 103884.
- 22 Ahmed A., Adak B., Faruk M. O., & Mukhopadhyay S. Nanocellulose coupled 2D graphene nanostructures: Emerging paradigm for sustainable functional applications. *Industrial & Engineering Chemistry Research*, 2021, vol. 60, no. 30, pp. 10882–10916.
- 23 Xu T., Du H., Liu H., Liu W., Zhang X., Si C. ... & Zhang, K. Advanced nanocellulose- based composites for flexible functional energy storage devices. *Advanced materials*, 2021, vol. 33, no. 48, p. 2101368.
- 24 Zhang K., Ketterle L., Järvinen T., Lorite G.S., Hong S., & Liimatainen H. (2020). Self-assembly of graphene oxide and cellulose nanocrystals into continuous filament via interfacial nanoparticle complexation. *Materials & Design*, 2020, no. 193, p. 108791.
- 25 Almasov N., Kurbanova B., Kuanyshbekov T., Akatan K., Kabdrakhmanova S. & Aimaganbetov K. Study of the structure and electrical properties of graphene oxide (GO) and graphene oxide+ nanocellulose (GO+ NC). *Kompleksnoe Ispolzovanie Mineralnogo Syra= Complex use of mineral resources*, 2024, vol. 329, no. 2, pp. 103–109.
- 26 Ge X., Shan Y., Wu L., Mu X., Peng H., Jiang Y. High-strength and morphology-controlled aerogel based on carboxymethyl cellulose and graphene oxide. *Carbohydr. Polym.*, 2018, no. 197, pp. 277–283.
- 27 Chen L., Li Y., Du Q., Wang Z., Xia Y., Yedinak E., Lou J., Ci L. High performance agar/graphene oxide composite aerogel for methylene blue removal. *Carbohydr. Polym.*, 2017, no. 155, pp. 345–353.
- 28 Kuanyshbekov T., Guseinov N., Tolepov Zh., Kurbanova B., Tulegenova M., Akatan K., Kantay N., and Zhasasynov E. Thermally Reduced Graphene Oxide Membranes From Local Kazakhstan Graphite “Ognevsky”. *ChemistrySelect* 2023, no. 8, e202301746.
- 29 Mengke Zhao, Sufeng Zhang, Guigan Fang, Chen Huang and Ting Wu. Directionally-Grown Carboxymethyl Cellulose/Reduced Graphene Oxide Aerogel with Excellent Structure Stability and Adsorption Capacity. *Polymers* 2020, no.12, p. 2219. <https://doi.org/10.3390/polym12102219>.
- 30 Kuanyshbekov T., Sagdollin Z., Zhasasynov E., Akatan K., Kurbanova B., Guseinov N., Tolepov Z., Kantay N., Beisebekov M. Composite Membrane Based on Graphene Oxide and Carboxymethylcellulose from Local Kazakh Raw Materials for Possible Applications in Electronic Devices. *J. Compos. Sci.*, 2023, no. 7, p. 342. <https://doi.org/10.3390/jcs7080342>
- 31 Kuanyshbekov T.K., Akatan K., Kabdrakhmanova S.K., Nemkaeva R., Aitzhanov M., Imasheva A., Kairatuly E. (2021). Synthesis of graphene oxide from graphite by the hummers method. *Oxidation Communications*, 2021, 44, no. 2.

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ГРАФЕН ОКСИДІНІҢ – НАНОЦЕЛЛЮЛОЗА КОМПОЗИЦИЯСЫНЫҢ ЭЛЕКТРЛІК ҚАСИЕТТЕРІН ЗЕРТТЕУ

Андатпа

Бұл зерттеу Фурье түрлендіруші инфрақызыл спектроскопия (FTIR), рентгендік фотоэлектрондық спектроскопия (XPS), Раман спектроскопиясы және сканерлеуші электронды микроскопия (SEM) әдістері арқылы мұқият сипатталған импеданс спектроскопиясын қолдана отырып, графен оксиді (ГО) және наноцеллюлоза (НЦ) композицияларының электрлік қасиеттерін зерттейді. FTIR талдауы ГО және НЦ құрамында болатын функционалдық топтарға сәйкес келетін сипаттамалық шыңдарды анықтады, бұл олардың химиялық құрамын түсінуге мүмкіндік берді. XPS спектрлері көміртегі мен оттегінің байланыс күйлерін көрсететін ерекше шыңдарды көрсетті, бұл материалдардың беттік химиясын сипаттауға мүмкіндік берді. Раман спектроскопиясы үлгілердің құрылымдық реті мен ақаулары туралы ақпарат беріп, әсіресе ГО-ның графиттік құрылымын ерекше атап өтті. SEM кескіндері композициялық мембрананың морфологиялық ерекшеліктерін ашып, НЦ бөлшектерінің таралуын және олардың қосылуынан туындаған құрылымдық модификацияларды көрсетті. ГО-НЦ композициясының электр өткізгіштігін зерттеу үшін импеданс спектроскопиясы қолданылды. Нәтижелер температураға тәуелді мінез-құлықты көрсетті: өткізгіштік отын элементтерінің жұмыс диапазонында температураның жоғарылауымен артты. Бір қызығы, НЦ қосу композицияның өткізгіштік қасиеттеріне айтарлықтай әсер етпеді, бұл материалдардың үйлесімділігі мен тұрақтылығын болжайды. Қорытындылай келе, көптеген аналитикалық әдістерді қолдана отырып жасалған бұл жан-жақты зерттеу ГО-НЦ композициясының электрлік қасиеттері туралы құнды мәліметтер береді. Нәтижелер бұл композицияның электр өткізгіштігін арттыруды қажет ететін әртүрлі қолданбаларда, әсіресе отын ұяшықтары технологиясында әлеуетті қолданылуын көрсетеді.

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ИЗУЧЕНИЕ ЭЛЕКТРИЧЕСКИХ СВОЙСТВ КОМПОЗИТА ОКСИД ГРАФЕНА – НАНОЦЕЛЛЮЛОЗА

Аннотация

В этом исследовании электрические свойства композита из оксида графена (ОГ) и наноцеллюлозы (НЦ) изучаются с помощью импедансной спектроскопии, дополненной тщательной характеристикой с помощью инфракрасной спектроскопии с преобразованием Фурье (FTIR), рентгеновской фотоэлектронной спектроскопии (XPS), рамановской спектроскопии и сканирующей электронной микроскопии (SEM). ИК-анализ выявил характерные пики, соответствующие функциональным группам, присутствующим как в ОГ, так и в НЦ, что позволило получить представление об их химическом составе. Спектры XPS показали характерные пики, указывающие на состояние углеродных и кислородных связей, что объясняет химический состав поверхности материалов. Спектроскопия комбинационного рассеяния позволила получить информацию о структурном порядке и дефектах в образцах, особенно выделив графитную структуру ОГ. СЭМ-изображения выявили морфологические особенности композитной мембраны, продемонстрировав распределение частиц НЦ и структурные изменения, вызванные их внедрением. Для исследования электропроводности композита ОГ-НЦ была использована импедансная спектроскопия. Результаты показали, что поведение композита зависит от температуры, при этом с повышением температуры в пределах рабочего диапазона топливных элементов наблюдалось увеличение электропроводности. Примечательно, что добавление НЦ существенно не изменило электропроводность композита, что свидетельствует о совместимости и стабильности. Таким образом, эта всесторонняя характеристика с использованием множества аналитических методов дает ценную информацию об электрических свойствах композита ОГ-НЦ. Полученные результаты свидетельствуют о его потенциале для различных применений, требующих повышенной электропроводности, особенно в технологии топливных элементов.

Ключевые слова: протонообменная мембрана, оксид графена, наноцеллюлоза, мембранно-электродный узел, композит, импедансная спектроскопия.

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