



Study of the structure and electrical properties of graphene oxide (GO) and graphene oxide+nanocellulose (GO+NC)

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ABSTRACT

Proton exchange membranes (PEMs) that function at elevated temperatures surpassing 100°C and exhibit exceptional mechanical, chemical, and thermochemical stability have garnered significant interest. This is primarily due to their practical utility in proton exchange membrane fuel cells (PEMFCs). In the present era, an extensive array of polymers and polymer-blended membranes have been scrutinized for their applicability in this domain. Each of these materials presents a set of advantages and disadvantages. However, the realm of PEMFCs is still in search of the perfect membrane endowed with distinct properties. Graphene oxide, a two-dimensional substance arising from the oxidation of graphite, has manifested itself as a promising candidate. Oxygen (O) functional groups are incorporated within the sp² carbon (C) plane of the oxidized graphite, forming graphene oxide. This material can be synthesized by exfoliating graphite oxide, a three-dimensional carbon-based compound, into layered sheets using ultrasonic or mechanical agitation. The presence of multiple reactive oxygen functional groups renders graphene oxide suitable for a diverse array of applications, such as composite polymers, energy conversion materials, environmental safeguards, sensors, transistors, and optical components. This versatility is attributable to its outstanding electrical, mechanical, and thermal properties. Among the various methodologies for graphene oxide synthesis, the modified Hammer method stands out for its simplicity, cost-effectiveness, and high yield. This research delves into the structural analysis of graphene oxide obtained through the Hammer method, utilizing commercially available graphite. The study involves the creation of membranes based on carboxymethylcellulose (NC) that integrate dispersed graphene oxide (GO) sheets. These novel membranes, as well as pristine graphene oxide, were subjected to a comprehensive array of analytical techniques including XRD, XPS, Raman, FTIR, and SEM microscopy. Additionally, electrophysical characterizations were undertaken employing electrochemical impedance spectroscopy (EIS) measurements. The investigation uncovered that the introduction of NC into the graphene oxide matrix significantly enhances the electron conductivity of the composite membrane. Simultaneously, the presence of graphene oxide contributes to the mechanical robustness and thermomechanical stability of the membrane structure. The principal impetus behind this article lies in furnishing vital insights into the physical and structural attributes of graphene oxide membranes relevant to their deployment in hydrogen energy applications.

Keywords: Hammers method, graphene oxide, nanocellulose, XRD, XPS, IR Fourier spectroscopy, impedance spectroscopy (EIS)

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Introduction

Graphene oxide (GO) originates from the exfoliation of graphite oxide, resulting in a layered structure containing one or multiple carbon atom layers, accomplished through ultrasonic or mechanical processes [1]. These GO sheets are mainly classified as chemically generated graphene, displaying analogous characteristics to pristine graphene. However, a fundamental distinction exists between graphene and GO: while graphene solely comprises sp^2 hybridized carbon atoms, GO encompasses a carbon framework featuring diverse oxygen based functional groups.

Back in 1859, Brodie labeled graphene oxide as either graphite oxide or graphitic acid [2]. This was achieved through the chemical treatment of graphite using $KClO_3$ and HNO_3 . The subsequent step involves the transformation of graphite oxide into monolayer sheets of GO, a process that can be accomplished using various thermal and mechanical techniques [3]. In the current context, the monatomic carbon stratum within graphite oxide is recognized as graphene oxide (GO).

Four methods of GO synthesis are well known, such as Brody's method [2], Staudenmaier's method [4], Hammer and their modifications [[5], [6]], and Tur's method [7]. At present, the synthesis of GO by the modified Hammer method is considered to be the most common method.

GO contains various functional groups of active oxygen, and its polymer composites enable the development of materials for energy conversion and environmental protection [8].

In addition, GO can be used to produce membranes for obtaining pure water. In recent years, multilayer two-dimensional films for purification and chemical separation of water have been obtained from bilayer hydroxides (LDHs), transition metal dichalcogenides (MoS_2 , WS_2), transition metal carbides (MXene), and many other materials based on graphene [[9], [10], [11], [12]].

Compared with other 2D membranes, graphene oxide (GO) membranes have better sheet structure, efficient separation, and flexible fabrication methods. Studies have shown that GO multilayer membranes can effectively trap water contaminants, and water molecules can quickly flow between layers with virtually no friction [[12], [13]].

Because cellulose is partially crystalline, it comprises regions that are both crystalline and non-crystalline [14]. The polymer's molecular and intermolecular chemical constituents possess distinctive characteristics, including hydrophilicity, limited solubility in certain aqueous environments, and a capacity for straightforward chemical modification. Cellulose can be categorized into different structural arrangements and transformed from one configuration to another through either chemical or thermal processes [15].

Nanocellulose (NC) exhibits superior physical, chemical, biological, magnetic, electrical and optical characteristics compared to some materials in their nanoscale form [16]. Conceptually, NCs can be obtained at different stages using hydrolysis technique, i.e. (1) extraction of extractives/hemicellulose using physicochemical, biological or a combination of two or more treatments, (3) separation of cellulose unit fibrils or microfibrils to obtain nanofibers using different substances and (4) solvent removal, ultrasonic treatment, centrifugation, stabilization and drying [[17], [18], [19], [20]].

As a natural nanomaterial, plant-derived nanocellulose and natural cellulose have the same properties as graphene, such as large size, specific surface area, mechanical and chemical properties, high crystallinity, biodegradability, and renewable resource intensity [21]. Recently, many researchers have been working on combining NCs with graphene to create functional hybrid composites of nanocellulose and graphene [[22], [23], [24]].

Experimental part

Two different graphene oxide (GO, GO+NC) based samples were fabricated by the Hammers method (Table 1) [[25], [26]].

Table 1 - Samples of graphene oxide

Name	Thickness
1. GO	18.03 – 23.5 micron
2. GO+NC(1/1)	12.36 micron

The morphological structure of the samples was determined using the SEM Crossbeam 540 research instrument. The results obtained are shown in Figure 1.

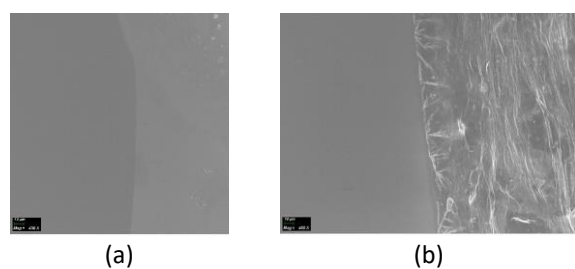


Figure 1 - SEM images of samples of graphene oxide (a) (GO) and b) GO + nanocellulose (NC)

The spectra of GO, GO+NC were studied by X-ray photoelectron spectroscopy. The results obtained are presented in fig. 2. The spectra of the two samples in the figure show the difference in binding energy (GO = 280-290 eV, GO+NC = 290-300 eV).

IR spectra were determined using a Nicolet iS10 IR Fourier spectrometer (Fig. 3-a) and the Raman spectra were studied (Fig. 3-b).

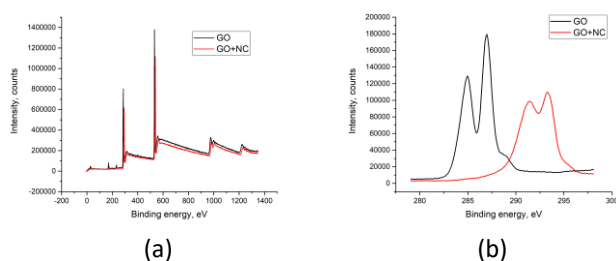


Figure 2 - X-ray photoelectron spectrum (XPS) of GO + NC samples

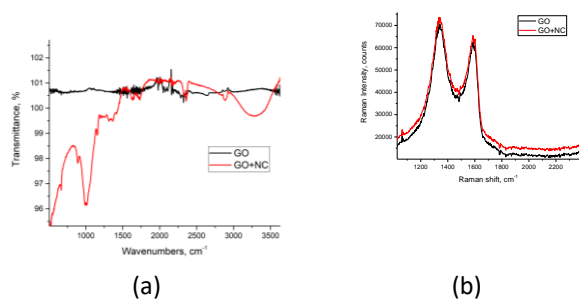


Figure 3 - a) FTIR spectra and

b) Raman spectra of GO and GO + NC samples

The electrical characteristics of the samples were studied in various temperature ranges by the method of impedance spectroscopy (HF2 Impedance spectroscopy, Zurich). The copper element was deposited on the surface of the sample by magnetron sputtering to create an electrical ohmic contact. Plasma power 40 W, deposition time 30 min.

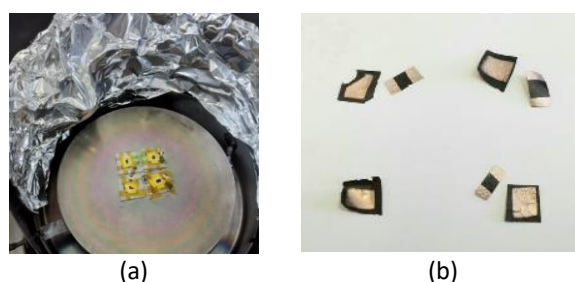


Figure 4 - a) the process of magnetron deposition and b) the type of samples obtained

Impedance measurements were carried out at a sinusoidal voltage amplitude of 50 mV in the frequency range from 10 Hz to 1 MHz. On fig. 5 shows the measurement results.

Results and Discussion

As can be seen from the SEM images (Fig. 1), the addition of nanocellulose to graphene oxide leads to the formation of a composite material with pores and folds.

On fig. 2 shows the C 1s XPS spectra for GO and GO + NC. It can be seen that the spectrum of C 1s GO has two peaks at 284.6 and 286.6 eV, corresponding to the sp^2 and C-O carbon functional groups, respectively. However, the addition of nanocellulose led to an upward shift in the binding energy and a twofold decrease in the overall intensity. The peak shift is related to a change of oxidation state of GO after adding NC, where the higher binding energy shows higher oxidation state [27]. The intensity is directly connected to the number of atoms with respective oxidation state. Thus, there is a certain degree on the amount of oxidized atoms in GO+NC composite which is less than the amount of non-oxidized atoms in pristine GO without any addition. Also, the XPS spectrum of graphene oxide showed there isn't any elements other than C and O, indicating the absence of foreign impurities.

A typical Raman spectrum (Fig. 3b) of GO is characterized by an approximate G band. The Raman spectrum has a 1605 cm^{-1} band corresponding to the E_{2g} phonon of the sp^2 C atoms and a 1353 cm^{-1} D band corresponding to the point phonon K absorption mode of A_{1g} symmetry. Here, the graphene G band was observed at 1600 cm^{-1} , which was slightly shifted from the GO position. There was no difference in the Raman spectra of GO and GO + NC models.

In the FTIR GO spectrum (Fig. 3a), there is a strong and broad elongated O-H vibrational band

which occurs at 3410 cm^{-1} from - due to oxidation. In addition, the carboxyl at 1721 cm^{-1} has a C = O stretching band, an O-H deformation vibrational band at 1404 cm^{-1} and a C - O stretching band at 1087 cm^{-1} . The addition of nanocellulose resulted in an increase in the total FTIR conductivity signal and additional peaks.

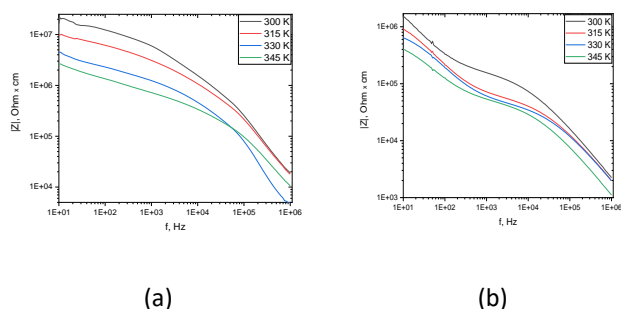


Figure 5 - frequency dependence of electrical resistance with temperature change, a) GO, B) GO + NC

In Figure 5, it can be seen the difference in the electrical resistance of graphene oxide and GO + NC samples. Temperature dependences of the research samples were also revealed. The obtained results show that at room temperature the electrical resistance of graphene oxide is one order of magnitude higher than GO+NC. As the temperature increases, the resistance of both samples decreases and the conductive property increases. Whether these materials can be used as a fuel cell is evidenced by actual studies.

Conclusions

Graphene oxide and GO + NC results obtained by simple chemical wet process, XRD, FTIR, Raman, SEM showed successful results. It was investigated that the conductivity of the samples increased with increasing temperature during operation using electrochemical impedance spectroscopy (EIS) measurements. XPS results showed that the addition of NC to the GO leads to the increase of oxidation degree of the whole membrane. GO with elevated levels of oxygen content could make the membrane significantly more hydrophilic. This enhancement in hydrophilicity would lead to improved membrane properties, such as a decrease in biofouling processes and an increase in water flux. Also, GO can be easily dispersed in water and exfoliated in a wide range of solvents due to the hydrophilicity. Graphene oxide plays an important role in the development of science and technology today. Studies show that graphene oxide can be used for not only making battery and supercapacitor cathodes, electrical sensors, photovoltaic devices, electrochromic devices and optical devices, but also for making membranes in hydrogen energy. Many studies provide evidence for the future utilization of graphene oxide, which is an advanced technology based on graphene oxide.

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Графен оксиді (GO) және графен оксиді + наноцеллюлоза (GO+NC) құрылымын және электрлік қасиеттерін зерттеу

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<p>Мақала келді: 5 маусым 2023 Сараптамадан өтті: 14 тамыз 2023 Қабылданды: 24 тамыз 2023</p>	<p>ТҮЙІНДЕМЕ</p> <p>Жоғары температурада жұмыс істейтін және тамаша механикалық, химиялық және термохимиялық тұрақтылыққа ие протон алмасу мембраналары (PEM) протонды мембраналық отын элементтерінде (PEMFC) практикалық қолданылуына байланысты көп назар аударуда. Қазіргі уақытта осы мақсаттар үшін полимерлермен араласқан көптеген полимерлі және әртүрлі мембраналар зерттелуде, олардың барлығында артықшылығы және кемшіліктері бар. Дегенмен, PEMFC бірегей қасиеттері бар идеалды мембраналар әлі де жоқ. Графен оксидінің құрамында әртүрлі реактивті оттегі функционалды топтары бар, бұл оның тамаша қасиеттеріне байланысты оны полимер композиттері, энергияны түрлендіруге арналған материалдар, қоршаған ортаны қорғау қолданбалары, сенсорлар, транзисторлары және оптикалық қолданбалар сияқты көптеген қолданбаларда электрлік, механикалық және жылулық қасиеттері негіз ретінде қолданылады. Графен оксидін синтездеу үшін кеңінен қолданылатын әдіс қарапайым процесс, құны төмен және жоғары кірістілігіне байланысты модификацияланған Хаммер әдісі болып табылады. Бұл жұмыста біз коммерциялық графитін пайдалану арқылы Хаммерс әдісімен алынған графен оксидінің құрылымдық зерттеулері туралы баяндаймыз. Бұл графен оксидінің құрылымдық сипаттамаларын беретін өлшеу құрылғыларымен әр түрлі спектрометрлердің көмегімен қарастырылады. Дисперсті графен оксиді (GO) парақтары бар карбоксиметилцеллюлоза (NC) негізіндегі мембраналар PEMFC қолданбалары үшін жасалып, зерттелді. Бұл мембраналар мен графен оксиді XRD, XPS, Raman, FTIR және SEM микроскопиясының комбинациясы арқылы зерттелді, ал электрофизикалық сипаттау электрохимиялық импеданс спектроскопиясы (EIS) өлшемдері арқылы орындалды. GO-ға СМС қосу бүкіл мембрананың электр өткізгіштігін жоғарылататыны, ал GO мембрананың жақсы механикалық және термомеханикалық тұрақтылығын қамтамасыз ететіні анықталды. Бұл мақаланы жазудың негізгі мотивациясы графен оксидінің сутегі энергетикасында қолданылатын мембраналар үшін қажетті физикалық құрылымдық сипаттамалар беру болып табылады.</p>
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Исследование структуры и электрических свойств оксида графена (GO) и оксид графен+наноцеллюлоза (GO+NC)

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АННОТАЦИЯ

Протонообменные мембраны (PEM), работающие при высоких температурах выше 100 °С, с превосходной механической, химической и термохимической стабильностью, привлекли большое внимание в связи с их практическим применением в топливных элементах с протонообменными мембранами (PEMFC). В настоящее время для этого применения исследовано огромное количество полимеров и различных полимерных мембран, все из которых имеют как плюсы, так и минусы. Однако идеальных мембран с уникальными свойствами для PEMFC все еще нет. Оксид графена представляет собой двумерный материал, образованный из окисленного графита с функциональными группами кислорода (O), занимающими плоскость sp^2 углерода (C). Оксид графена можно получить путем

	<p>расплавления оксида графита (окисленного трехмерного материала на основе углерода) на ламинированные листы с использованием ультразвукового или механического перемешивания. Оксид графена содержит различные функциональные группы активного кислорода, что делает его основой для многих приложений, таких как полимерные композиты, материалы для преобразования энергии, приложения для защиты окружающей среды, датчики, транзисторы и оптические приложения благодаря его превосходным электрическим, механическим и термическим свойствам. Широко используемым методом синтеза оксида графена является модифицированный метод Хаммера из-за его простого процесса, низкой стоимости и высокого выхода. В этой работе мы сообщаем о структурных исследованиях оксида графена, полученного методом Хаммера с использованием технического графита. Мембраны на основе карбоксиметилцеллюлозы (NC) с листами дисперсионного оксида графена (GO) были изготовлены и исследованы для применения PEMFC. Эти мембраны и первичный GO были изучены с помощью комбинации XRD, XPS, Raman, FTIR и SEM-микроскопии, в то время как электрофизические характеристики были выполнены с использованием измерений электрохимической импедансной спектроскопии (EIS). Выявлено, что добавление NC к GO повышает электрическую проводимость всей мембраны, а GO обеспечивает хорошую механическую и термомеханическую стабильность мембраны. Основной мотивацией написания этой статьи является обеспечение необходимых физических структурных характеристик для мембран из оксида графена, используемых в водородной энергетике.</p>
	<p>Ключевые слова: метод Хаммера, оксид графена, наноцеллюлоза, XRD, XPS, ИК Фурье-спектроскопия, импедансной спектроскопии (EIS)</p>
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