



Study of Thermal and Mechanical Characteristics of Polyacrylamide Composites: Carboxylate – Functionalized Multiwalled Carbon Nanotubes and Graphene Oxide Nanoparticles

Wadie Sultan^a, Taghreed Taqatqa^a, Laila Abuayyasha^a, Firas Fohely^b, Sami Makharza^{c,*}



^a Department of Chemistry, Faculty of Science and Technology, Al-Quds University, West bank, Palestine

^b Department of Medical Imaging, Faculty of Pharmacy and Medical Science, Hebron University, P.O. Box 40, Hebron, Palestine

^c College of Medicine, Hebron University, P.O. Box 40, Hebron, West Bank, Palestine

Abstract

The incorporation of multi-walled carbon nanotubes (MWCNT- COOH) and graphene oxide (GO) nanoparticles into poly(acrylamide-co-acrylic acid) poly(AAm-co-AA) have been investigated. FTIR demonstrated that GO, functionalized MWCNTs and poly(AAm-co-AA) polymer have good interfacial bonding. The nanocomposites were prepared using a simple casting method in separate experiments. The thermal properties like glass transition temperature (T_g) and melting point were elucidated by using Differential Scanning Calorimetric (DSC), the results reveal that the thermal properties were changed after incorporation of CNTs and GO into the poly(AAm-co-AA) matrix. The young's modulus was raised at 5% poly(AAm-co-AA) and 0.3% CNTs-COOH.

Keywords: Composites; Multiwalled Carbon Nanotubes; Graphene Oxide; Polyacrylamide

1. Introduction

The carbon allotropes like graphene oxide, carbon nanotubes, fullerenes, and carbon dots are used as nanofiller materials to produce nanocomposites with polymer matrices in different applications [1]–[7]. The implementation of nanofillers is employed to improve the thermal, electrical, and mechanical characteristics of polymeric materials [1], [3], [6]–[9]. Graphene oxide possesses a single-layered two-dimensional sp² hybridized structure, which can be presented as a one layer of carbon with several oxygen-containing functional groups including carbonyl, epoxide, hydroxyl, and carboxylic acid groups. These hydrophilic functional groups make the surface of the GO sheet forms a stable colloids in water which enhance the interfacial adhesion with the polymer matrix [9]–[11]. The structure of MWCNTs is composed of multiple layers of graphene rolled into tubes. Moreover, they possess exceptional mechanical properties which are stronger than steels in addition to their high thermal and electrical conductivity [12], [13]. The GO and MWCNTs can easily be mixed with different polymers such as

Poly(AAm-co-AA) forming a composite has attracted significant attention due to its remarkable properties, such as tensile strength, and excellent thermal and electrical characteristics [14]. Poly(AAm-co-AA) is a water-soluble synthetic polymer that possesses useful properties such as good adhesiveness, proper hygroscopicity, high hydrophilicity, and non-toxicity [7], [11], [15], [16]. It is utilized in many fields such as drug delivery vehicles, biomaterials in biomedical, tissue engineering and so forth. However, traditional hydrogels have poor mechanical properties that have hindered their development [7], [17]. By incorporating nanoparticles into the poly(AAm-co-AA) matrix forming composite material, the nanoparticles can reinforce the poly(AAm-co-AA) matrix causing an enhancement and improvement in mechanical, thermal, chemical, and electrical properties depending on the type of nanoparticles that is used [14], [16]. Therefore, improving the mechanical, electrical, and thermal properties of polymers has drawn the significant attention of many researchers around the world. In response to the rising need for the development of new materials, this

*Corresponding author e-mail: Makharza.sami@gmail.com.

EJCHEM use only: Received date 29 April 2024; revised date 15 July 2024; accepted date 26 July 2024

DOI: 10.21608/ejchem.2024.285410.9655

©2024 National Information and Documentation Center (NIDOC)

research was conducted to improve the thermal and mechanical properties of poly(AAm-co-AA) nanofillers [18]. Among the various nanofillers, GO and MWCNTs were chosen for this study due to their outstanding physical properties [6], [19], [20]. In this paper, graphene oxide (GO) and MWCNTs nanofillers were added into polyacrylamide hydrogels and the effect of these nanofillers on mechanical and thermal properties have been investigated. The properties and applications of such composites depend on several factors such as concentration, type of nanofiller and type of functional group. Many methods have been elucidated to fabricate polymer composite materials; simple casting method [21], In situ polymerization [22], [23], laser ablation [24], etc. In this study we used simple casting method at room temperature, the samples placed under vacuum to obtain thin films with homogenous distribution of nanofillers and thickness between 2 – 5 micrometers. These films are considered as thrilled candidates in many fields like pharmaceuticals [25], [26], drug delivery systems and biosensors [27]. The GO samples utilized as nanofillers in this study was obtained from our published protocol [28]. The scanning electron microscopy (SEM) images showed that the average size of GO nanoparticles is 200 nm.

2. Experiment

2.1 Materials

Poly(AAm-co-AA) powder, 80+%, Molecular weight = 150 – 520 kD and $D=0.75\text{g/ml}$. Multiwalled Carbon nanotube functionalized carboxylic acid, extent of labelling > 8% carboxylic acid. Graphite (99.99%, -200 mesh, Alfa Aesar). 98% H_2SO_4 , KMnO_4 , 3% H_2O_2 . All materials were purchased from Sigma Aldrich.

2.2 Methods

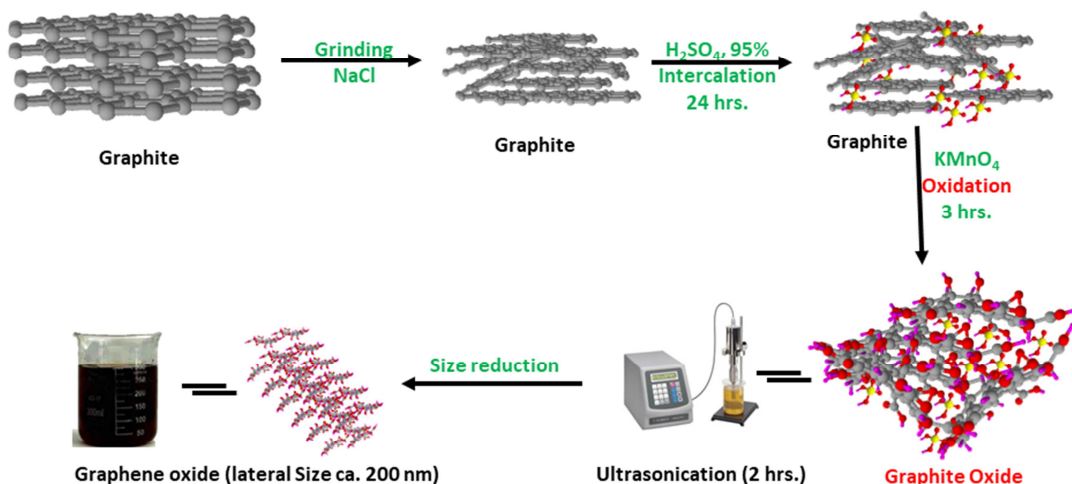
Fabrication of graphene oxide nanoparticles

The graphene oxide was synthesized as shown in Scheme 1. In summary, the oxidation process commenced with the introduction of graphite flakes into a sodium chloride (NaCl) solution. Subsequently, both components underwent grinding. To facilitate intercalation, a concentrated sulfuric acid (H_2SO_4) was then introduced into the mixture and stirred for a duration of 24 hours.

Following this interval, potassium permanganate (KMnO_4) was cautiously pace, while stirring continuing for 3 hours. To achieve further dilution, 200 milliliters of distilled water were incrementally incorporated into the mixture while maintaining continuous stirring. Ultrasonication was subsequently applied to the mixture for 2 hours to reduce particle size [28]. Ultimately, a sample of a graphene oxide (GO) solution featuring a lateral dimension of 200 nanometers was successfully obtained .

Synthesis of Poly(AAm-co-AA) thin film

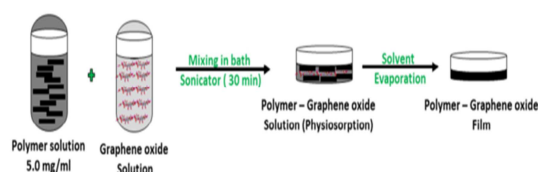
The production of the thin film involved the utilization of various weight percentages within the solution, specifically 0.9%, 1.25%, 2.5%, and 5%. These solutions were meticulously crafted by weighing the requisite quantities of polymer solutions, followed by a 30-minute stirring period at room temperature. Subsequently, the polymer films were fabricated via the spin coating technique, employing a range of rotation rates spanning from 200 to 550 round per minute (rpm) and differing application durations of 5, 15, 30, and 60 seconds. To complete the process, the samples were allowed to undergo natural drying within an incubator set at room temperature (RT). [29].



Scheme 1: Schematic showing graphene oxide preparation

Synthesis of Poly(AAm-co-AA)/GO nanocomposite film

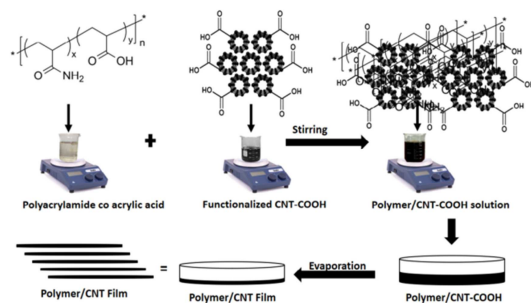
The process for producing nanocomposite films is detailed in Scheme 2. Initially, diverse concentrations of graphene oxide (GO) were prepared by dilution from a concentrated stock solution. Following dilution, the solution was partitioned into four segments to facilitate the preparation of polymer/GO nanocomposite solutions, each attaining concentrations of 0.9%, 1.25%, 2.5%, and 5% by weight of the polymer. Subsequently, these four different polymer/GO nanocomposite concentrations underwent ultrasonication within a water bath at room temperature for a duration of 30 minutes. Finally, the resulting samples were poured into Petri dishes to undergo solvent evaporation.



Scheme 2: Casting method fabrication of Poly(AAm-co-AA)/GO nanocomposite

Preparation of poly (acrylamide-co-acrylic acid) /MWCNT nanocomposite

Scheme 3 represents the fabrication of poly (AAm-co-AA)/MWCNT nanocomposite. Different concentrations of MWCNT – COOH were prepared using a dilution factor from a stock solution of 0.3mg/ml. The four concentrations of poly(AAm-co-AA)/MWCNT nanocomposite were sonicated using bath sonication at RT for 30 min. The samples were poured into Petri dishes for solvent evaporation.



Scheme 3: Fabrication of poly(AAm-co-AA)/CNTs composite

2.3 Equipment

The Perkin Elmer 4000 DSC instrument was employed in this study, utilizing a specific heating and cooling program. In the initial heating phase, the temperature was elevated from 25°C to 450.0°C at a

heating rate of 10°C/min, all conducted under a nitrogen atmosphere. Subsequently, the sample was cooled back to room temperature at the same rate of 10°C/min. The second heating phase followed the same protocol as the first. For the assessment of mechanical properties, a Mechanical Testing Machine was employed. To analyze chemical or physical interactions, a Bruker IFS 66/S mid-FTIR spectrometer was utilized, featuring a liquid nitrogen chamber, a cooled MCT detector, and a KBr beam splitter. For the production of graphene oxide nanoparticles, a VCX750 37 tip sonicator with a power output of 750 watts was employed. [17], [28], [30].

3. Results and Discussion

3.1 FTIR Spectroscopy

FTIR spectroscopy of poly(AAm-co-AA)

FTIR spectroscopy was used for scrutinizing the distinct shifts in the maximum absorbance bands associated with the C-N amide, C=O, and N-H groups. These spectral shifts offer critical insights into the intermolecular interactions that influence the vibrational behaviour of groups within polymer segments. By harnessing the capabilities of FTIR analysis, the study was able to glean essential information about these interactions. The utilization of FTIR spectra extended to the estimation of interaction phenomena between poly(AAm-co-AA) and the composite materials comprising both graphene oxide (GO) and multi-walled carbon nanotubes (MWCNT) nanofillers. As shown in figure 1, the spectrum of neat poly(AAm-co-AA) is characterized by several characteristic bands that clearly shows five regions. The first band is the broadest, observed at 3337 cm⁻¹ and corresponding to the stretching vibration of the hydroxyl (-OH) and amine (N-H) stretching in poly(AAm-co-AAc) structure.

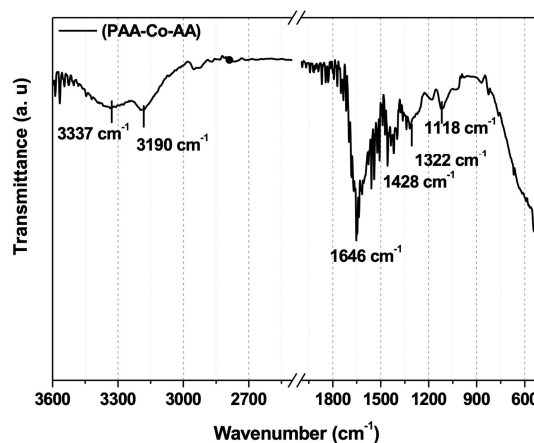


Figure 1: FTIR spectrum of Poly(AAm-co-AA)

The peak at 3190 cm^{-1} , corresponding to symmetric stretching vibration of NH_2 , 2850 cm^{-1} are attributed to the alkyl chain of the polymer, and 1646 cm^{-1} , 1428 cm^{-1} are the characteristics of the acrylamide unit for both $\text{C}=\text{O}$ and $\text{C}-\text{N}$ respectively. At 1118 cm^{-1} is another peak related to the amide group. The peak appearing at 1322 cm^{-1} is the characteristics ($\text{C}-\text{O}$) bending peak of $-\text{COOH}$ [5], [31], [32]

FTIR spectroscopy of GO

The FTIR spectrum depicting graphene oxide is presented in Figure 2. Specific functional groups associated with oxygen were identified as follows: The $\text{C}-\text{O}$ and $\text{C}=\text{O}$ groups were observed, represented by distinct peaks at 1062 cm^{-1} and 1730 cm^{-1} , respectively. The ($\text{O}-\text{H}$) group exhibited its characteristic presence at 3364 cm^{-1} .

Additionally, the $\text{C}=\text{C}$ group manifested itself distinctly at 1614 cm^{-1} . This FTIR analysis facilitated the identification and characterization of key oxygen-related functional groups within the graphene oxide material, as illustrated in Figure 2. [28], [33]

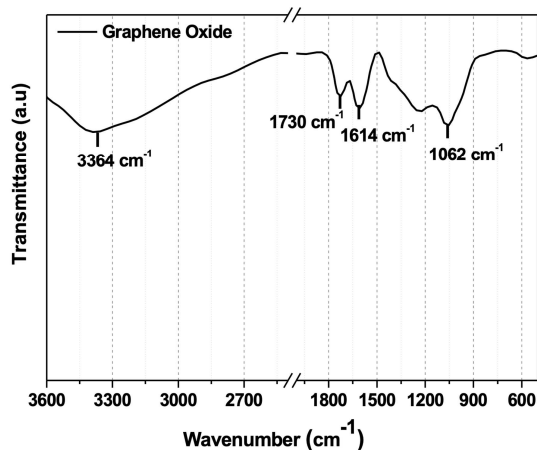


Figure 2: FTIR Spectrum of graphene oxide particles

FTIR spectroscopy of MWCNT

In Figure 3, the FTIR spectrum of multi-walled carbon nanotubes (MWCNTs) functionalized with carboxylic acid (COOH) is depicted. Notably, distinct oxygen functional groups were identified as follows: The $\text{C}=\text{O}$ group was clearly discernible, manifesting as a prominent peak at 1635 cm^{-1} . Additionally, the $\text{C}-\text{O}$ group was evident, with its characteristic presence marked by a peak at 1068 cm^{-1} . The hydroxyl group ($\text{O}-\text{H}$) was observed at 3300 cm^{-1} . Furthermore, the ($\text{C}=\text{C}$) group was distinctly visible, appearing at 1404 cm^{-1} . This FTIR analysis provided valuable insights into the oxygen-related functional groups within the COOH -functionalized MWCNTs, as shown in Figure 3.

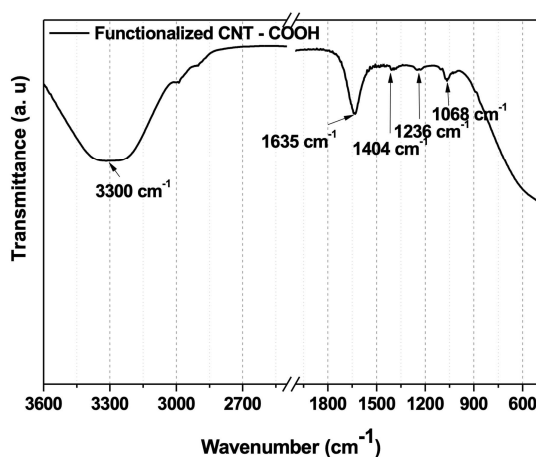


Figure 3: The FTIR spectrum of Functionalized CNT-COOH

FTIR spectroscopy of poly(AAm-co-AA) – GO and CNT nanocomposite.

The shift of the main bands of Poly(AAm-co-AA) from their original position is an indication of an interaction between polymer/GO and polymer/MWCNTs particles which is most probably due to the hydrogen bonding between the OH group of GO and COOH of MWCNT with the functional groups in polymer backbone as shown in figure 4 and 5.

The band at 1322 cm^{-1} corresponding to the bending vibration of COOH is highly shifted from the original position in poly(AAM-co-AA), as shown in figure 4, 95 cm^{-1} for GO, and 85 cm^{-1} for MWCNT (figure 5). This confirms some intermolecular noncovalent attractions between polymer and nanofillers. This interaction mainly corresponds to hydrogen bonding or electron transfer interaction. It was noticed that the magnitude of the shift in the case of GO was greater than MWCNT which could referred to the 2D structure of GO making closed packing easier than MWCNT cylindrical structure [33], [34]. Table 1 and Table 2 (See ESI) summarized the FTIR peak positions of poly(AAM-co-AA), GO, CNT-COOH, poly(AAM-co-AA)/GO and poly(AAM-co-AA)/CNT-COOH.

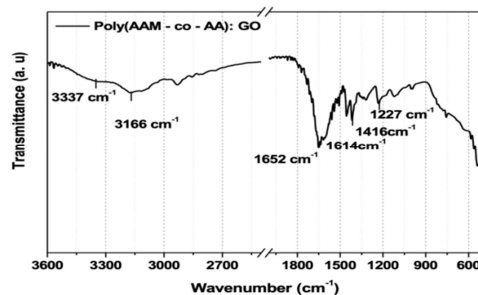


Figure 4: FTIR spectrum of Poly(AAM-co-AA): GO

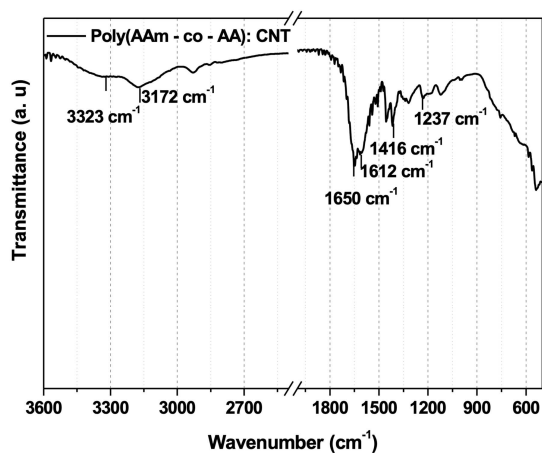


Figure 5: FTIR spectrum of Poly(AAm-co-AA): CNT

3.2 Thermal analysis

Thermal Analysis of Poly(AAm-co-AA) and composites

The thermal properties of poly(AAm-co-AA)/GO and poly(AAm-co-AA)/MWCNTs were studied using DSC. The glass transition temperature (T_g) of poly(AAm-co-AA) reveals several complications, one of these complications is that poly(AAm-co-AA) is reported to bind strongly with water (2-9%), which can act as a plasticizer lowering T_g of polymer. Another complication is that removal of the water required heating at a high temperature (above 150 °C) which will lead to degradation problem including imidization and crosslinking of poly(AAm-co-AA) and this could explain the presence of an endothermic peak between 280-290 °C is due to degradation [35].

Figures 6,7 and 8 exhibit the thermograms of poly(AAm-co-AA), poly(AAm-co-AA)/GO and poly(AAm-co-AA) /MWCNTs respectively. Graphene oxide and MWCNTs were added into poly(AAm-co-AA) hydrogels to modify their mechanical and thermal properties. When GO and MWCNT were added an improvement in glass transition temperature and heat capacity was observed. The T_g measurements evaluate the flexibility of polymer which is the temperature at which molecular mobility begins to take place. The shifts in T_g values were attributed to the improvement in the rigidity of polymer in the presence of nanofiller that interact with the polymer matrix and restrict the allowed mobility of the polymer matrix chain and raise the value of T_g . The magnitude of shift was higher in the case of GO nanofiller compared to MWCNTs which could be due to a high specific surface area which could be due to a high specific surface area and wrinkled GO, which can bind with the polymer chains and restricts the mobility of the matrix [36]. Table 3 (see ESI)

summarized the measured values of T_g , T_m and degradation properties of polymer and composite samples.

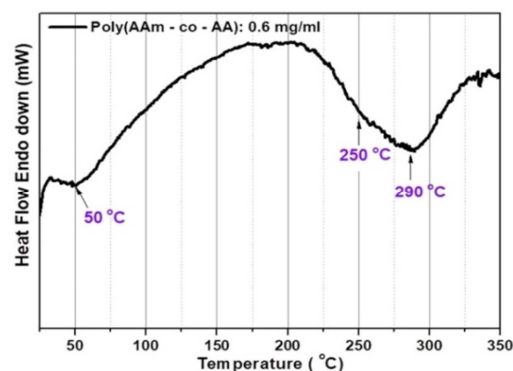


Figure 6: The DSC thermogram of neat Poly(AAm-co-AA)

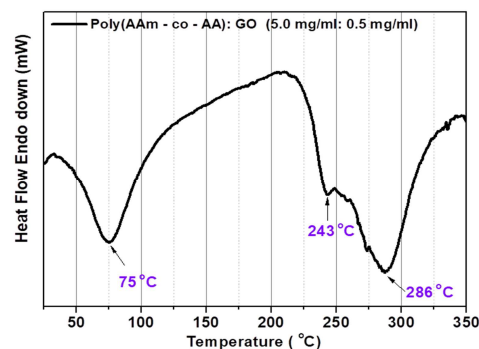


Figure 7: The DSC thermogram of Poly(AAm-co-AA)/GO

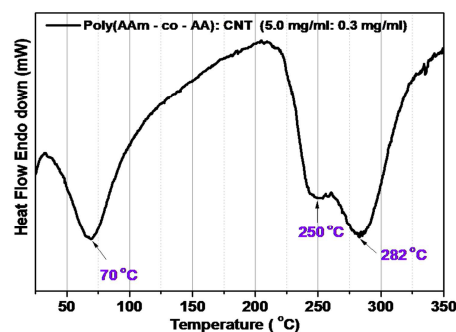


Figure 8: DSC thermogram of Poly(AAm-co-AA)/CNT-COOH

3.3 Mechanical analysis

The Figures 9 and 10 (a – b) represent the stress-strain behaviour of three distinct materials, Poly(AAm-co-AA), Poly(AAm-co-AA)/GO (5.0:0.5) mg/ml, and Poly(AAm-co-AA)/GO (5.0:1.0) mg/ml. The Poly(AAm-co-AA) film exhibits an elongation of approximately 97%. The Young's modulus for Poly(AAm-co-AA) measured at 125%, while it notably increased to 200% and 300% for Poly(AAm-co-AA)/GO (5.0:0.5) mg/ml and Poly(AAm-co-AA)/GO (5.0:1.0) mg/ml, respectively. It is a well-recognized fact that a higher modulus demands more stress to induce the same degree of strain. Our findings indicate a substantial enhancement in mechanical properties, wherein an increase in the concentration of GO within the polymer resulted in an elevated Young's modulus and improved

mechanical characteristics of the nanocomposite. Remarkably, the plastic strain region, typically observed in these materials, was absent in the nanocomposite. This phenomenon can be attributed to the nanocomposite's behaviour, which closely resembles that of fiber materials in terms of mechanical response, leading to a departure from typical polymer behaviour. Furthermore, it is worth noting that the tensile strength and elastic modulus of the nanocomposite hydrogel experienced a significant boost, increasing by 124% and 26%, respectively, with the inclusion of 3 wt.% GO loading. This comprehensive analysis sheds light on the mechanical properties and structural behaviour of the Poly(AAm-co-AA) and Poly(AAm-co-AA)/GO nanocomposites under various loading conditions, highlighting their potential for applications requiring enhanced mechanical performance.

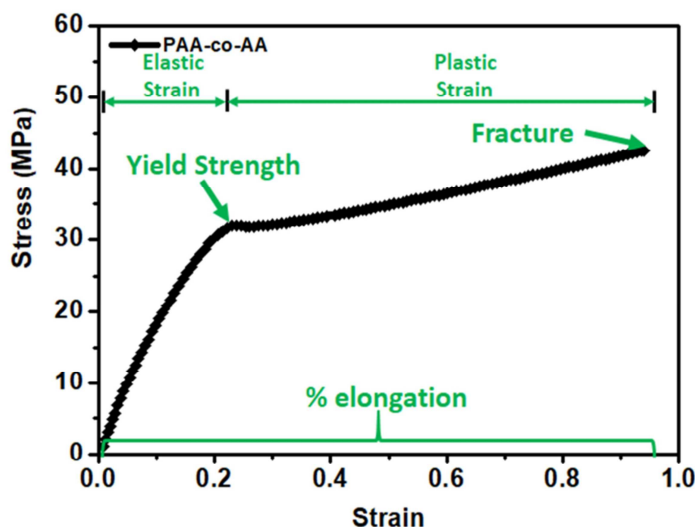


Figure 9. Stress-Strain curve of poly(AA-co-AA)

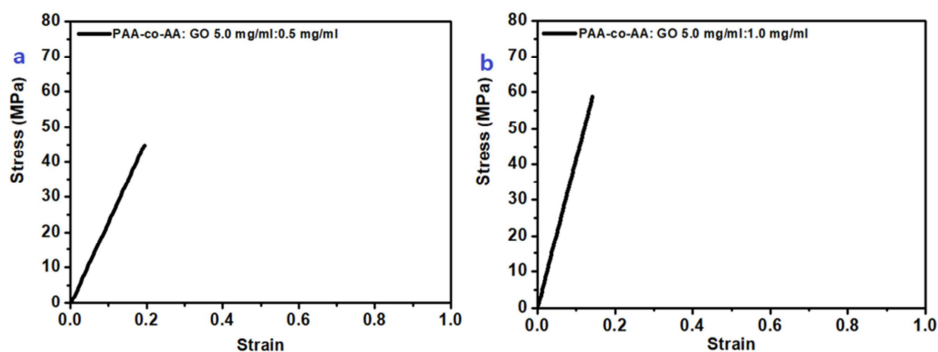


Figure 10. Stress-Strain curve of poly(AA-co-AA)/GO nanocomposite

Figure 11 reveals the stress-strain curvature of poly(AAm-co-AA)/MWCNT at different concentrations. It well appears that the mechanical properties of the nanocomposites are significantly influenced by interfacial interactions between

MWCNT – COOH and poly(AAm-co-AA) matrices. The effects of the aspect ratio of MWCNT - COOH reinforcements on the Young's modulus and yield strength, of the poly(AAm-co-AA)/MWCNT composites.

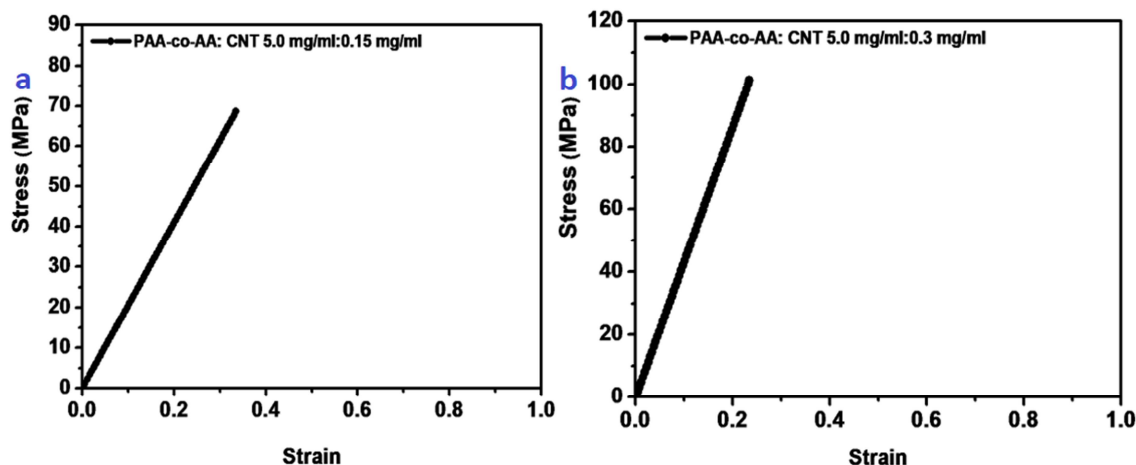


Figure 11: Stress-Strain curve of poly(AAm-co-AA)/CNT nanocomposite

4. Conclusion

In conclusion, using nanofillers in low concentrations to create polymer composite is an effective way to enhance their mechanical and thermal properties. These nanofillers act as reinforcements with the matrix, providing a significant effect on the properties. The nanofiller materials including Multiwalled carbon nanotube, graphene oxide are promising nanofillers for a poly(AA-co-AA) matrix. The enhancement in the properties depend on the type of nanofillers used.

5. Conflict of Interest

All authors declare no conflict of interest.

6. Acknowledgment

The authors would like to acknowledge the Nano lab at Al-Quds University, as well as the department of chemistry at Hebron University.

7. Data Availability

We confirm that the findings in this paper are available as part of master dissertations for Mrs. Taghreed Taqatqa and Laila Abuayyash at Al-Quds University – East Jerusalem.

Here are the links:

Mrs. Taghreed Taqatqa

<https://dspace.alquds.edu/items/b55bf26f-b6e9-4566-becd-3d674debbcb13>

Mrs. Laila Abuayyash

<https://dspace.alquds.edu/items/6553315e-3131-41c7-ae14-0252006c3b8f>

References

- [1] Z. Feng, K. H. Adolfsson, Y. Xu, H. Fang, M. Hakkarainen, and M. Wu, "Carbon dot/polymer nanocomposites: From green synthesis to energy, environmental and biomedical applications," *Sustainable Materials and Technologies*, vol. 29, 2021. doi: 10.1016/j.susmat.2021.e00304.
- [2] A. Kausar, "Polymer/carbon-based quantum dot nanocomposite: forthcoming materials for technical application," *Journal of Macromolecular Science, Part A: Pure and Applied Chemistry*, vol. 56, no. 4, pp. 341–356, 2019. doi: 10.1080/10601325.2019.1578614.
- [3] A. Kausar, "Advances in Polymer/Fullerene Nanocomposite: A Review on Essential Features and Applications," *Polymer - Plastics Technology and Engineering*, vol. 56, no. 6, pp. 594–605, 2017. doi: 10.1080/03602559.2016.1233278.
- [4] A. Kausar, I. Ahmad, M. Maaza, M. H. Eisa, and P. Bocchetta, "Polymer/Fullerene Nanocomposite for Optoelectronics—Moving toward Green Technology," *Journal of Composites Science*, vol. 6, no. 12, 2022. doi: 10.3390/jcs6120393.
- [5] Y. F. Zhang *et al.*, "Supramolecular ionic polymer/carbon nanotube composite hydrogels with enhanced electromechanical performance," *Nanotechnol. Rev.*, vol. 9, no. 1, 2020, doi: 10.1515/ntrev-2020-0039.
- [6] J. Gao, S. He, A. Nag, and J. W. C. Wong, "A review of the use of carbon nanotubes and

- graphene-based sensors for the detection of aflatoxin m1 compounds in milk,” *Sensors*, vol. 21, no. 11, 2021, doi: 10.3390/s21113602.
- [7] G. A. Evingür and Ö. Pekcan, “Mechanical properties of graphene oxide–polyacrylamide composites before and after swelling in water,” *Polym. Bull.*, vol. 75, no. 4, 2018, doi: 10.1007/s00289-017-2101-4.
- [8] A. Montazeri, J. Javadpour, A. Khavandi, A. Tcharkhtchi, and A. Mohajeri, “Mechanical properties of multi-walled carbon nanotube/epoxy composites,” *Mater. Des.*, vol. 31, no. 9, 2010, doi: 10.1016/j.matdes.2010.04.018.
- [9] S. C. Ray, “Application and Uses of Graphene Oxide and Reduced Graphene Oxide,” in *Applications of Graphene and Graphene-Oxide Based Nanomaterials*, 2015, pp. 39–55. doi: 10.1016/b978-0-323-37521-4.00002-9.
- [10] S. Kamel, M. El-Sakhawy, B. Anis, and H. A. S. Tohamy, “Graphene’s Structure, Synthesis and Characterization; A brief review,” *Egypt. J. Chem.*, vol. 63, pp. 593–608, 2020, doi: 10.21608/ejchem.2019.15173.1919.
- [11] S. Awasthi, T. P. Yadav, and K. Awasthi, “Notable electrical and mechanical properties of polyacrylamide (PAM) with graphene oxide (GO) and single-walled carbon nanotubes (SWCNTs),” *Int. Polym. Process.*, vol. 38, no. 3, pp. 290–299, 2023, doi: 10.1515/ipp-2022-4295.
- [12] J. H. Lehman, M. Terrones, E. Mansfield, K. E. Hurst, and V. Meunier, “Evaluating the characteristics of multiwall carbon nanotubes,” *Carbon*, vol. 49, no. 8, pp. 2581–2602, 2011. doi: 10.1016/j.carbon.2011.03.028.
- [13] C. H. Lau et al., “The effect of functionalization on structure and electrical conductivity of multi-walled carbon nanotubes,” *J. Nanoparticle Res.*, vol. 10, no. SUPPL. 1, pp. 77–88, 2008, doi: 10.1007/s11051-008-9376-1.
- [14] J. Shen, B. Yan, T. Li, Y. Long, N. Li, and M. Ye, “Study on graphene-oxide-based polyacrylamide composite hydrogels,” *Compos. Part A Appl. Sci. Manuf.*, vol. 43, no. 9, pp. 1476–1481, 2012, doi: 10.1016/j.compositesa.2012.04.006.
- [15] Y. L. Luo, C. H. Zhang, Y. S. Chen, and W. Yang, “Preparation and characterisation of polyacrylamide/MWCNTs nanohybrid hydrogels with microporous structures,” *Mater. Res. Innov.*, vol. 13, no. 1, 2009, doi: 10.1179/143307509X402138.
- [16] Z. Li, M. Tang, J. Dai, T. Wang, and R. Bai, “Effect of multiwalled carbon nanotube-grafted polymer brushes on the mechanical and swelling properties of polyacrylamide composite hydrogels,” *Polymer (Guildf)*, vol. 85, 2016, doi: 10.1016/j.polymer.2016.01.025.
- [17] R. Sengupta, M. Bhattacharya, S. Bandyopadhyay, and A. K. Bhowmick, “A review on the mechanical and electrical properties of graphite and modified graphite reinforced polymer composites,” *Progress in Polymer Science (Oxford)*. 2011. doi: 10.1016/j.progpolymsci.2010.11.003.
- [18] D. K. Lee, J. Yoo, H. Kim, B. H. Kang, and S. H. Park, “Electrical and Thermal Properties of Carbon Nanotube Polymer Composites with Various Aspect Ratios,” *Materials (Basel)*, vol. 15, no. 4, 2022, doi: 10.3390/ma15041356.
- [19] B. Arash, Q. Wang, and V. K. Varadan, “Mechanical properties of carbon nanotube/polymer composites,” *Sci. Rep.*, vol. 4, 2014, doi: 10.1038/srep06479.
- [20] V. D. Punetha et al., “Functionalization of carbon nanomaterials for advanced polymer nanocomposites: A comparison study between CNT and graphene,” *Prog. Polym. Sci.*, vol. 67, pp. 1–47, 2017, doi: 10.1016/j.progpolymsci.2016.12.010.
- [21] L. N. M. Dinh, B. N. Tran, V. Agarwal, and P. B. Zetterlund, “Synthesis of Highly Stretchable and Electrically Conductive Multiwalled Carbon Nanotube/Polymer Nanocomposite Films,” *ACS Appl. Polym. Mater.*, vol. 4, no. 3, 2022, doi: 10.1021/acspapm.1c01738.
- [22] D. Sharma, M. Kumar, V. P. Jain, S. Chaudhary, and G. Jaiswar, “Bio-based polyamide nanocomposites of nanoclay, carbon nanotubes and graphene: a review,” *Iranian Polymer Journal (English Edition)*. 2023. doi: 10.1007/s13726-023-01164-x.
- [23] M. Prosheva, M. Ehsani, Y. Joseph, R. Tomovska, and J. Blazhevskva Gilev, “Waterborne polymer composites containing hybrid graphene/carbon nanotube filler: Effect of graphene type on properties and performance,” *Polym. Compos.*, vol. 44, no. 8, 2023, doi: 10.1002/pc.27483.
- [24] J. Zhang et al., “Laser Ablation Mechanism and Performance of Carbon Fiber-Reinforced Poly Aryl Ether Ketone (PAEK) Composites,” *Polymers (Basel)*, vol. 14, no. 13, 2022, doi: 10.3390/polym14132676.
- [25] S. K. Ghosh et al., “Improved rheological, barrier, antibacterial, and electromagnetic interference shielding properties of graphene and graphene derivatives based linear low density polyethylene nanocomposites,” *Polym. Compos.*, vol. 44, no. 9, 2023, doi: 10.1002/pc.27520.
- [26] H. Samaraweera and A. H. Khan, “Recent trends in graphene-based materials for pharmaceuticals wastewater treatment,” in *The Treatment of Pharmaceutical Wastewater: Innovative Technologies and the Adaptation of Treatment Systems*, 2023. doi: 10.1016/B978-0-323-99160-5.00001-1.
- [27] G. Praveen and S. Rajkhowa, “Recent advances of economically synthesised polymers/composites consisting of graphene and silver nanoparticles to achieve sustainable existence,” *Polymer Bulletin*. 2024. doi: 10.1007/s00289-024-05199-9.
- [28] M. Atawneh, S. Makharza, S. Zahran, K. Titi, F. Takrori, and S. Hampel, “The cross-talk between lateral sheet dimensions of pristine graphene oxide nanoparticles and Ni²⁺ adsorption,” *RSC Adv.*, vol. 11, no. 19, pp. 11388–11397, 2021, doi: 10.1039/d1ra00400j.
- [29] T. Taqatqa, “Synthesis and Characterization of Polyacrylamide co acrylic acid Carbon Nanotube Nanocomposite for Pharmaceutical

- Packaging Applications Taghreed,” Al-Quds University, MSc thesis, 2022.
- [30] S. Zahran *et al.*, “Understanding the Adsorptive Removal of Bromophenol Blue Dye Based Graphene Oxide Nanoparticles as Adsorbent,” *Adv. Mater. Process. Technol.*, vol. 8, no. 4, pp. 4280–4296, 2022, doi: 10.1080/2374068X.2022.2061781.
- [31] H. Mittal, S. B. Mishra, A. K. Mishra, B. S. Kaith, R. Jindal, and S. Kalia, “Preparation of poly(acrylamide-co-acrylic acid)-grafted gum and its flocculation and biodegradation studies,” *Carbohydr. Polym.*, vol. 98, no. 1, 2013, doi: 10.1016/j.carbpol.2013.06.026.
- [32] S. Nesrinne and A. Djamel, “Synthesis, characterization and rheological behavior of pH sensitive poly(acrylamide-co-acrylic acid) hydrogels,” *Arab. J. Chem.*, vol. 10, no. 4, 2017, doi: 10.1016/j.arabjc.2013.11.027.
- [33] S. Makharza *et al.*, “Size-dependent nanographene oxide as a platform for efficient carboplatin release,” *J. Mater. Chem. B*, vol. 1, no. 44, p. 6107, 2013, doi: 10.1039/c3tb21090a.
- [34] A. K. Pathak, M. Borah, A. Gupta, T. Yokozeki, and S. R. Dhakate, “Improved mechanical properties of carbon fiber/graphene oxide-epoxy hybrid composites,” *Compos. Sci. Technol.*, vol. 135, pp. 28–38, 2016, doi: 10.1016/j.compscitech.2016.09.007.
- [35] M. E. S. R. E Silva, E. R. Dutra, V. Mano, and J. C. Machado, “Preparation and thermal study of polymers derived from acrylamide,” *Polym. Degrad. Stab.*, vol. 67, no. 3, 2000, doi: 10.1016/S0141-3910(99)00149-4.
- [36] A. K. Pathak and S. R. Dhakate, “Validation of experimental results for graphene oxide-epoxy polymer nanocomposite through computational analysis,” *J. Polym. Sci.*, vol. 59, no. 1, 2021, doi: 10.1002/pol.20200442.