

## ADVANCING ELECTROSPUN POLYMER-GRAPHITE COMPOSITES: STRATEGIES FOR IMPROVED PERFORMANCE

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### Abstract

Expanded graphite is considered a stepping stone to graphene because it can be easily exfoliated into graphene layers when forming composite materials. It therefore has the potential to significantly improve conductivity and mechanical properties in most composites, as shown in the literature. In its natural form, expanded graphite is unsuitable for electrospinning, so it was reduced in size through ultrasonication. AC electrospinning is preferred due to its high production rate and lower risk of particle settling, as it uses pumps instead of a slow-moving syringe. There was no significant improvement in electrical conductivity due to the dominant structural effects attributed to the spinning process, which resulted in increased moisture uptake. Enhancements can be achieved by adding a conductive polymer network that works in synergy with the expanded graphite, thereby overcoming the conductivity limitations attributed to moisture. This work also showed that at low loading, up to 10% uEG in PVA, there can be an improvement in mechanical properties; however, the addition of fillers generally resulted in a decrease in breaking strength and modulus of the materials.

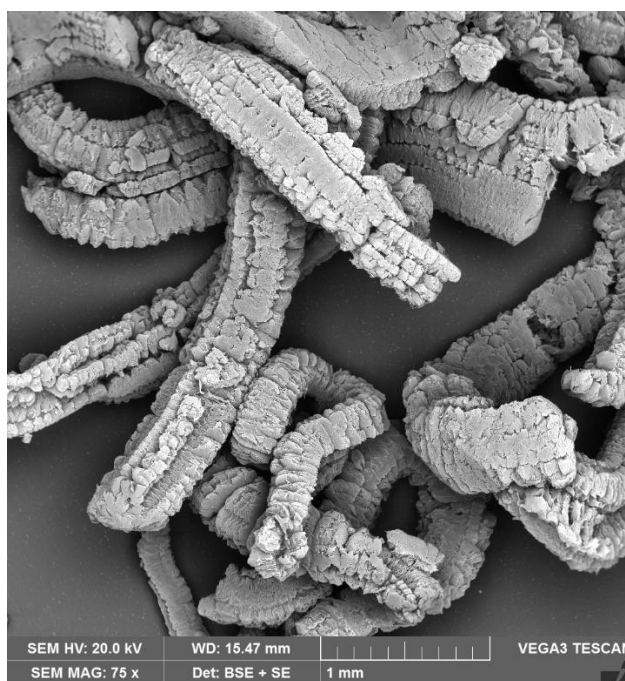
**Keywords:** Expanded graphite, ultrasonication, electrospinning, nanocomposite, conductive composite

### 1. INTRODUCTION

The emergence of electrospinning as a versatile nanofabrication technique has revolutionised the development of advanced functional materials, particularly in the field of polymer-based nanocomposites. This study focuses on the fabrication of AC electrospun nanofibres from three distinct polymer matrices—polyvinyl butyral (PVB), polycaprolactone (PCL), and polyvinyl alcohol (PVA)—combined with ultrasonicated expanded graphite (EG). The integration of expanded graphite into electrospun polymer nanofibres marks a significant advancement in creating multifunctional materials that combine the inherent properties of the polymer matrices with the exceptional thermal, electrical, and mechanical characteristics of graphite-based fillers. Due to the biocompatibility and low toxicity of the selected polymers, this research aims to enhance the understanding of conductive nanocomposite systems for biomedical applications and drug delivery systems [1,3,6,8,9].

Electrospinning, a process driven by alternating electrostatic forces, enables the production of continuous nanofibers with diameters ranging from nanometres to micrometres. This technique has gained substantial attention due to its ability to create fibrous structures that closely mimic the extracellular matrix (ECM) found in natural tissues, making it particularly valuable for biomedical applications. The AC electrospinning process involves the application of alternating current high voltage to a polymer solution or melt, creating a charged jet with periodically alternating positive and negative charge segments as it travels toward a grounded collector. Unlike traditional DC electrospinning, the alternating polarity in AC electrospinning suppresses the characteristic bending instability (whipping motion) due to decreased electrostatic repulsion between alternating charge segments. This results in a more stable jet formation with self-bundling behaviour, where the jet does not display significant attraction toward the grounded collector and can be easily deflected. The frequency of the AC voltage is critical for controlling the length of charged segments and ensuring proper charge transfer, with optimal frequencies enabling the formation of a visible thread downstream from the initial whipping cloud. AC electrospinning offers significant advantages, including higher productivity (10-20 times greater than DC electrospinning), reduced net charges on generated fibres enabling collection on non-conductive substrates, and improved process stability [2,7,10–12].

Expanded graphite constitutes a distinctive form of graphite that has undergone structural modification to develop a highly porous, accordion-like configuration with markedly increased volume and surface area, as illustrated in **Figure 1**. It is predominantly manufactured through the intercalation of natural flake graphite employing an oxidiser such as hydrogen peroxide, sulfuric acid, or nitric acid to produce expandable graphite. This product subsequently undergoes a thermal expansion process, wherein the expandable graphite is swiftly heated to approximately 900 °c. The heating induces the intercalating acid to rapidly decompose into a gas, leading to an expansion between the graphitic layers [4,5].



**Figure 1** Scanning electron microscope image of expanded graphite [own study]

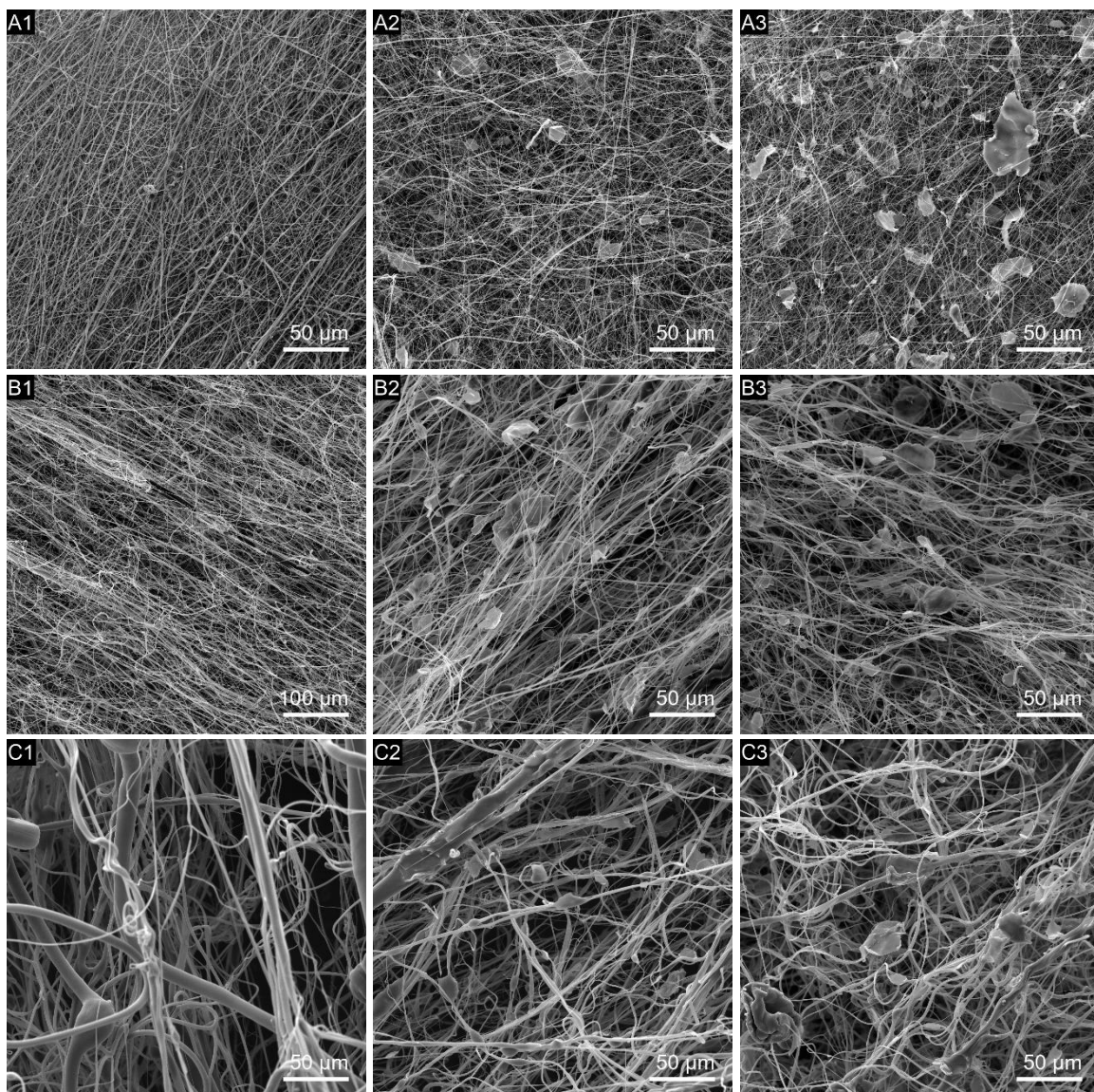
## 2. MATERIALS AND METHODS

Sulfuric acid intercalated expanded graphite was obtained from Sorbetin, Czech Republic, and incorporated into PVA (130000 Mw, 87-89% Hydrolysis rate from Sigma-Aldrich, 10% in distilled water), PVB (B60H, 55000 Mw from Penta Chemicals, 10.5% in absolute ethanol), and PCL (80000 Mw from Sigma-Aldrich, 10% in glacial

acetic acid, formic acid, and acetone in a 1:1:1 ratio). This mixture was electrospun using AC at 28, 31, and 34 kV for PVA, PVB, and PCL, respectively. The process was conducted at 23°C and 45% RH, employing a screw pump and a rotating drum electrode at a collector distance of 230 mm. Scanning electron microscopy was performed on various areas of the samples to evaluate filler dispersion and possible encapsulation. Surface resistivity was measured, along with mechanical properties through tensile testing. The tested samples were weighed to normalise the data.

### 3. RESULTS

#### 3.1 Microscopic analysis



**Figure 2** SEM images of planar materials: A1) PVA control. A2) PVA/uEG 90%/10%. A3) PVA/uEG 70%/30%. B1) PVB control. B2) PVB/uEG 90%/10%. B3) PVB/uEG 80%/20%. C1) PCL control. C2) PCL/uEG 95%/5%. C3) PCL/uEG 90%/10%.

The SEM micrographs displayed in **Figure 2** demonstrate that the uEG was successfully embedded into the nanofibers through the AC electrospinning process. For all the analysed polymers, higher graphite content

resulted in greater incorporation of thin, sheet-like graphite structures dispersed throughout the fibres. This is especially clear for the PVA samples (A1-A3) due to this material's ability to sustain a higher uEG concentration in the electrospinning solution. The control samples contained no graphite, while low concentrations showed initial presence of dispersed sheets. At the highest concentration, a significant amount of graphite sheets was observed, randomly distributed within the nanofibrous matrix. It appears as though the individual graphite sheets are completely encapsulated by the nanofibres. The fibres deform to cover each sheet, preventing sheet-to-sheet contact and thus disrupting conductive pathways, which explains the overall low conductivity observed in the samples. The addition of expanded graphite considerably reduced the fibre diameters across all polymers. For PVA, the fibre diameter decreased from  $889.6 \pm 556.9$  nm in the control sample to  $386.0 \pm 106.9$  nm at the highest uEG concentration. PVB fibres decreased from  $817.4 \pm 462.3$  nm to  $395.8 \pm 54.5$  nm, while PCL fibres decreased from  $1971.0 \pm 522.9$  nm to  $973.8 \pm 338.4$  nm. At the highest concentration, this corresponds to a 56.6% decrease in diameter for PVA, a 51.6% decrease for PVB, and a 50.6% decrease for PCL compared to their respective controls. This reduction can be attributed to the increased conductivity of the polymer solutions with graphite addition. Higher conductivity encourages greater stretching of the electrospinning jet under the applied electric field, leading to finer fibres. Furthermore, the standard deviations of fibre diameters decreased markedly for all three polymers, indicating improved uniformity and more stable jet formation during electrospinning. Therefore, the inclusion of expanded graphite not only enhances electrical conductivity and fibre thinning but also promotes more controlled and consistent fibre formation.

### 3.2 Electrical measurements

Electrical measurements were challenging because all polymer samples, regardless of filler type, exhibited unexpected resistivities in the megaohm range, which was primarily attributed to the presence of moisture. Although samples were dried and tested at 45% RH, high capillary pressure from the AC electrospinning process caused continued moisture uptake from the air, increasing conductivity. Thus, the material's structure, not the expanded graphite filler, was identified as the primary factor influencing conductivity.

### 3.3 Tensile measurements

The tensile properties of the composite nonwovens are provided in **Table 1**. Normalisation was carried out on the key factors, namely the breaking strength and modulus.

**Table 1** Tensile measurement data

Sample	Breaking Strength (N.m)	Modulus (MPa)	Maximum Elongation (mm)	Sample Weight	Norm. Breaking Strength (N.m/mg)	Norm. Modulus (MPa/mg)
<b>PVA</b>	1.02	101.90	10.19	4.57	0.22	22.30
StDev	0.20	19.75	0.74		0.04	4.32
<b>PVA/10%uEG</b>	0.31	30.86	27.59	1.19	0.26	25.93
StDev	0.04	4.04	4.36		0.03	3.40
<b>PVA/20%uEG</b>	0.16	16.38	29.02	0.88	0.19	18.65
StDev	0.02	1.64	2.70		0.02	1.87
<b>PVA/30%uEG</b>	0.08	8.24	26.56	0.62	0.13	13.29
StDev	0.01	0.91	6.31		0.02	1.47
<b>PVB</b>	1.07	106.52	10.66	4.75	0.22	22.44
StDev	0.17	16.62	0.57		0.04	3.50
<b>PVB/10%uEG</b>	0.73	73.01	9.34	6.35	0.12	11.50
StDev	0.03	2.80	0.55		0.00	0.44

<b>PVB/15%uEG</b>	0.22	22.42	9.34	2.68	0.08	8.37
StDev	0.08	7.92	1.58		0.03	2.96
<b>PVB/20%uEG</b>	0.25	24.94	10.26	2.61	0.10	9.55
StDev	0.04	3.78	1.25		0.01	1.45
<b>PCL</b>	1.00	100.13	33.59	8.76	0.11	11.44
StDev	0.15	15.30	6.59		0.02	1.75
<b>PCL/5%uEG</b>	0.24	23.91	37.87	2.53	0.09	9.44
StDev	0.03	3.31	4.78		0.01	1.31
<b>PCL10%uEG</b>	0.23	23.48	45.43	2.99	0.08	7.85
StDev	0.04	3.95	6.19		0.01	1.32

PVA samples exhibit initially enhanced properties at 10% uEG content, reaching the highest normalised breaking strength ( $0.259 \text{ N} \cdot \text{m}/\text{mg}$ ) and modulus ( $25.930 \text{ MPa}/\text{mg}$ ), but further increases in uEG concentration lead to a significant decline, culminating in a 40% reduction at 30% loading relative to pure PVA. In contrast, PVB and PCL show consistent decreases in both normalised strength and modulus as uEG content rises: for PVB, values fall from  $0.224$  to  $0.096 \text{ N} \cdot \text{m}/\text{mg}$  and from  $22.444$  to  $9.554 \text{ MPa}/\text{mg}$  (a drop of about 57%), while PCL drops from  $0.114$  to  $0.079 \text{ N} \cdot \text{m}/\text{mg}$  and from  $11.435$  to  $7.853 \text{ MPa}/\text{mg}$  (about 31%) with increasing uEG loading. This pattern suggests that low concentrations of uEG may enhance mechanical properties in certain matrices, such as PVA, likely due to improved dispersion and stress transfer at the polymer-filler interface. However, above a threshold, increased filler may induce agglomeration, poor dispersion, and defects that diminish mechanical integrity. PVB demonstrates the greatest susceptibility to these negative effects, while PCL displays the mildest decline. Normalisation by sample weight provides a robust basis for cross-comparison, highlighting compositional effects independent of sample size and confirming that optimal filler loading is polymer-specific and not universally beneficial.

#### 4. CONCLUSION

Expanded graphite composites demonstrate potential for use in electronic and mechanical applications. Expanded graphite was effectively integrated into AC electrospun PVA, PVB, and PCL nonwoven composites. SEM micrographs show that the fibre diameter reduces as the conductive particle load increases, as expected; however, upon drying, it is hypothesised that the micro- and nanocapillaries in the fibre structure develop high capillary pressure, which draws in moisture and affects the electrical conductivity. Evidence indicates that, at lower loadings, expanded graphite can reinforce PVA matrices; however, this benefit decreases after 10% loading. Therefore, it is advised to use a polymeric conductive network-forming agent, such as PEDOT:PSS, to counteract the insulating effects of the matrix and the porosity of the structure.

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#### REFERENCES

- [1] AZARI, A., GOLCHIN, A., MAYMAND, M. M., MANSOURI, F. and ARDESHIRYLAJIMI, A. Electrospun Polycaprolactone Nanofibers: Current Research and Applications in Biomedical Application. *Advanced Pharmaceutical Bulletin*. 2021, vol. 12, no. 4, pp. 658–671. ISSN 2251-7308.
- [2] BATKA, O., SKRIVANEK, J., HOLEC, P., BERAN, J., VALTERA, J. and BILEK, M. Methods for increasing productivity of AC-electrospinning using weir-electrode. *Scientific Reports*. 2024, vol. 14, no. 1, pp. 1–14. ISSN 2045-2322.

- [3] CHUI, C. Y., MOUTHUY, P. A. and YE, H. Direct electrospinning of poly(vinyl butyral) onto human dermal fibroblasts using a portable device. *Biotechnology Letters*. 2018, vol. 40, no. 4, pp. 737–745. ISSN 1572-977X.
- [4] COETZEE, D., MILITKÝ, J., WIENER, J. and VENKATARAMAN, M. Comparison of the Synthesis, Properties, and Applications of Graphite, Graphene, and Expanded Graphite. In: MILITKÝ, J. and VENKATARAMAN, M. (eds.). *Advanced Structured Materials*. Singapore: Springer, 2023, pp. 71–87. ISBN 978-981-19-6441-1.
- [5] COETZEE, D., ROJVIROON, T., NIAMLANG, S., MILITKÝ, J., WIENER, J., VEČERNÍK, J., MELICHERÍKOVÁ, J. and MÜLLEROVÁ, J. Effects of expanded graphite's structural and elemental characteristics on its oil and heavy metal sorption properties. *Scientific Reports*. 2024, vol. 14, no. 1, pp. 1–10. ISSN 2045-2322.
- [6] CUI, Z., ZHENG, Z., LIN, L., SI, J., WANG, Q., PENG, X. and CHEN, W. Electrospinning and crosslinking of polyvinyl alcohol/chitosan composite nanofiber for transdermal drug delivery. *Advances in Polymer Technology*. 2018, vol. 37, no. 6, pp. 1917–1928. ISSN 1520-6327.
- [7] KALOUS, T., HOLEC, P., JIRKOVEC, R., BATKA, O., ZABKA, P., POKORNY, P., STEPANOVA, P. and CHVOJKA, J. The effect of frequency change on the alternating current electrospinning of polyamide 6 and its productivity. *Journal of Environmental Chemical Engineering*. 2023, vol. 11, no. 3, pp. 109543. ISSN 2213-3437.
- [8] LU, L., YANG, B. and LIU, J. Flexible multifunctional graphite nanosheet/electrospun-polyamide 66 nanocomposite sensor for ECG, strain, temperature and gas measurements. *Chemical Engineering Journal*. 2020, vol. 400, pp. 125928. ISSN 1385-8947.
- [9] RAHIMKHOEI, V., PADERVAND, M., HEDAYAT, M., SEIDI, F., DAWI, E. A. and AKBARI, A. Biomedical applications of electrospun polycaprolactone-based carbohydrate polymers: A review. *International Journal of Biological Macromolecules*. 2023, vol. 253, pp. 126642. ISSN 0141-8130.
- [10] RAHMATI, M., MILLS, D. K., URBANSKA, A. M., SAEB, M. R., VENUGOPAL, J. R., RAMAKRISHNA, S. and MOZAFARI, M. Electrospinning for tissue engineering applications. *Progress in Materials Science*. 2021, vol. 117, pp. 100721. ISSN 0079-6425.
- [11] SILL, T. J. and VON RECUM, H. A. Electrospinning: Applications in drug delivery and tissue engineering. *Biomaterials*. 2008, vol. 29, no. 13, pp. 1989–2006. ISSN 0142-9612.
- [12] XUE, J., WU, T., DAI, Y. and XIA, Y. Electrospinning and Electrospun Nanofibers: Methods, Materials, and Applications. *Chemical Reviews*. 2019, vol. 119, no. 8, pp. 5298–5415. ISSN 1520-6890.