

Electrochemically Exfoliated Graphene Oxide for Thermoelectric Applications

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Introduction

- Graphene-based materials represents an excellent candidate for enhancing the thermoelectric properties of organic polymers such as PEDOT:PSS¹.
- The aim is to study the Seebeck coefficient for a range of temperature to identify the mode of transport, and electric conductivity to calculate the power factor.

Preparation

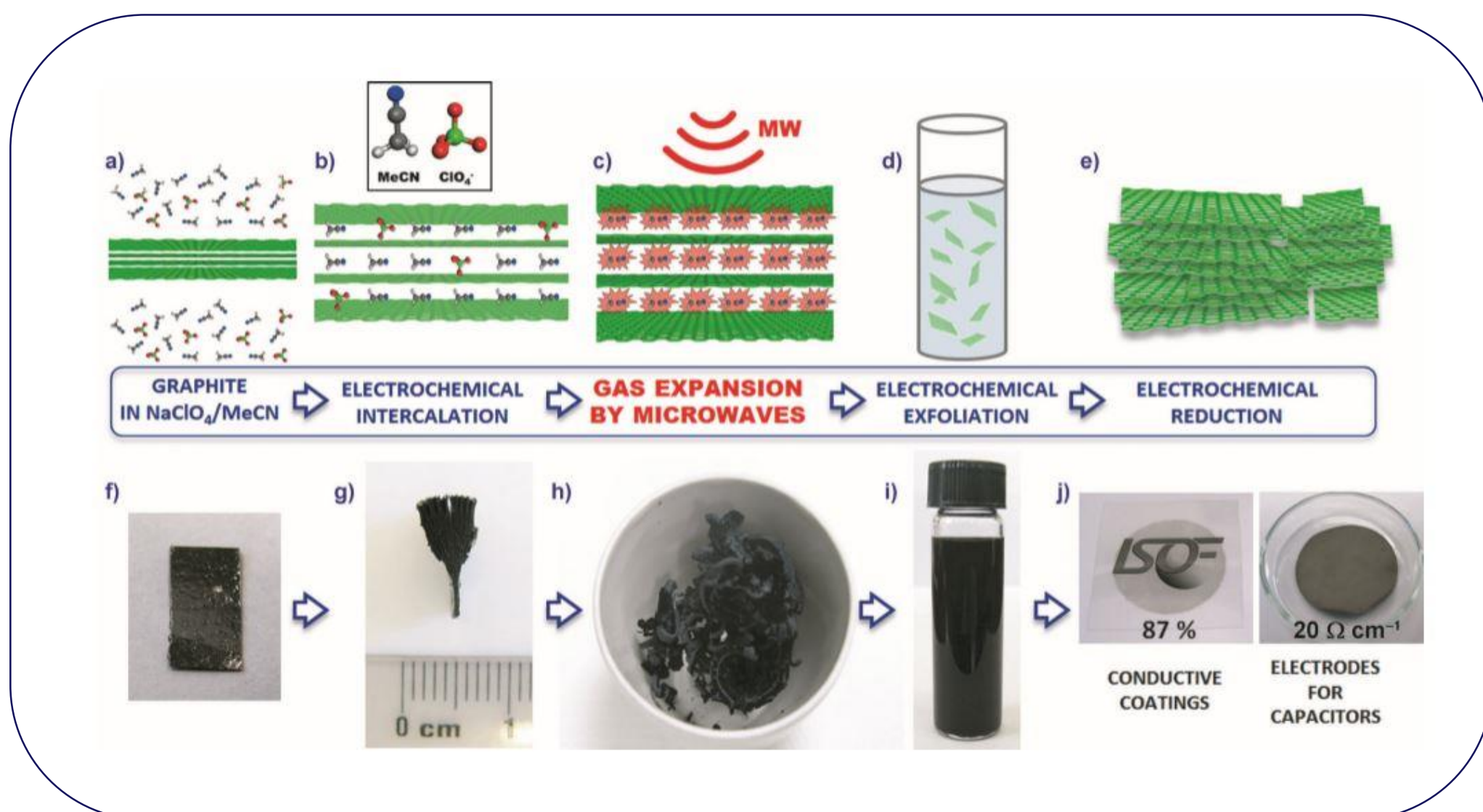


Figure 1. Schematic representation and photographs for the each step of the preparation process, intercalation, expansion via microwaves, electrochemical exfoliation and reduction from left to right².

The preparation process of the EGO samples consist on:

- Immersion of highly oriented pyrolytic graphite (HOPG) in NaClO₄/MeCN (Figure 1a).
- Electrochemical intercalation of ClO₄⁻ and MeCN in the HOPG by applying a +5V potential for 30 min (Figure 1b,g).
- Fully expansion by microwave irradiation yielding foam-like multi-layered graphite powder by gas evolution of MeCN (Figure 1c,h).
- Electrochemical exfoliation in 0.1 M H₂SO₄ applying a +10V potential for 30 min and collection of the electrochemically exfoliated graphene oxide (E-GO) by washing with water and dispersed in DMF with mild sonication (Figure 1d,f).
- Collection of exfoliated flakes by filtration on polytetrafluoroethene (PTFE) membrane and electrochemical reduction in 0.1M H₂SO₄ by applying a -2V potential for 10 min to yield electrochemically reduced graphene oxide (E-RGO) (Figure 1 e).
- The solution of E-GO and E-RGO in DMF and isopropanol was filtered onto thick nylon filters and transferred by roll-to-roll onto a polyethylene terephthalate (PET) substrate (Figure 1j).

Results

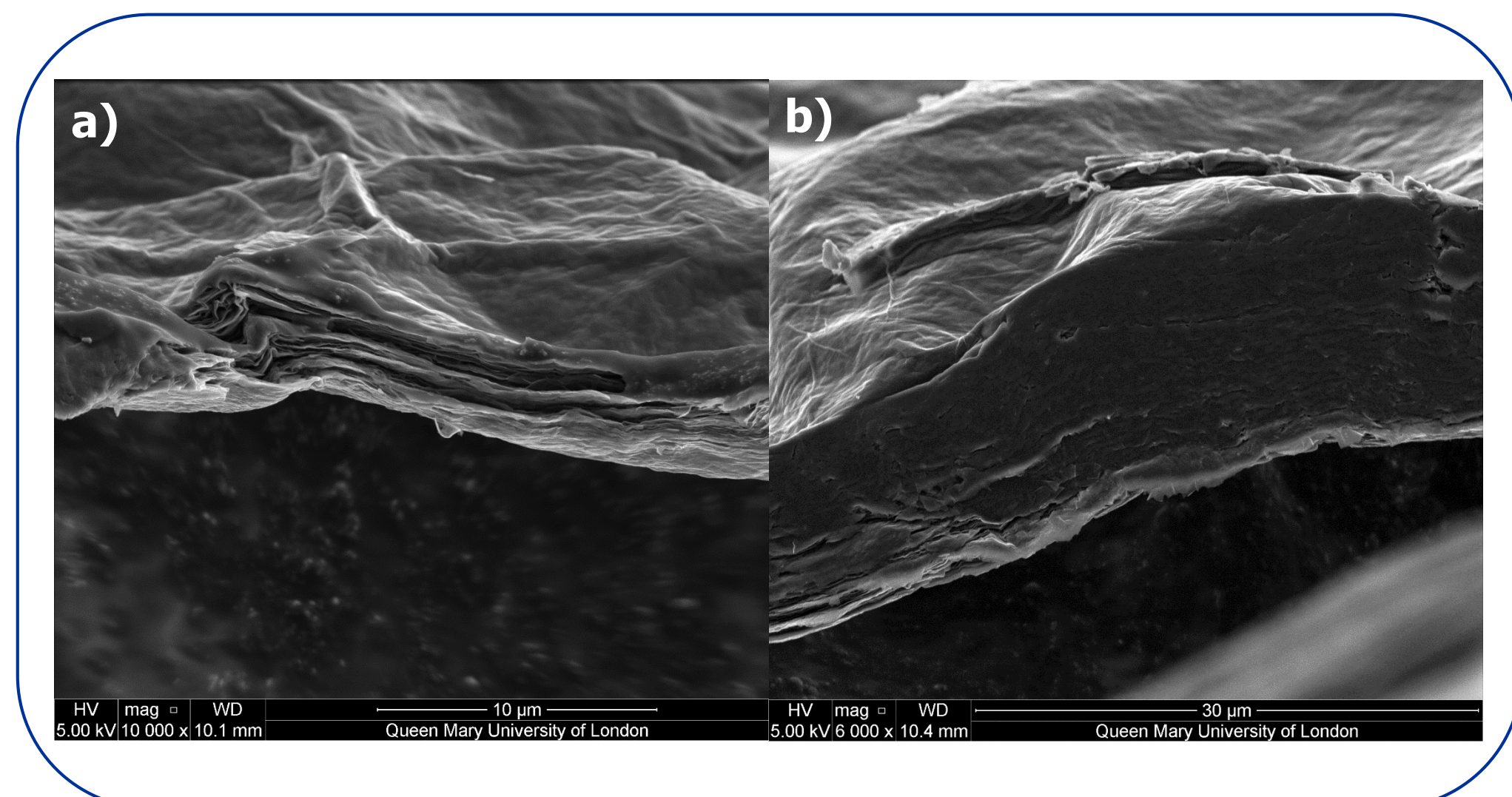


Figure 2. Cross-section SEM images of the a) 10mg and b) 20mg EGO samples.

In this study two multi-layered EGO samples were studied prepared by using the same precursor solution but differing in the loading amount by filtering approximately **10mg** and **20mg** of the suspended solids onto the substrate.

SEM images showed the exfoliated nature of the EGO samples. A thickness of the 10mg and 20mg EGO samples were measured as $2.4 \pm 0.2 \mu\text{m}$ and $13.8 \pm 3.2 \mu\text{m}$, respectively, see Figure 2.

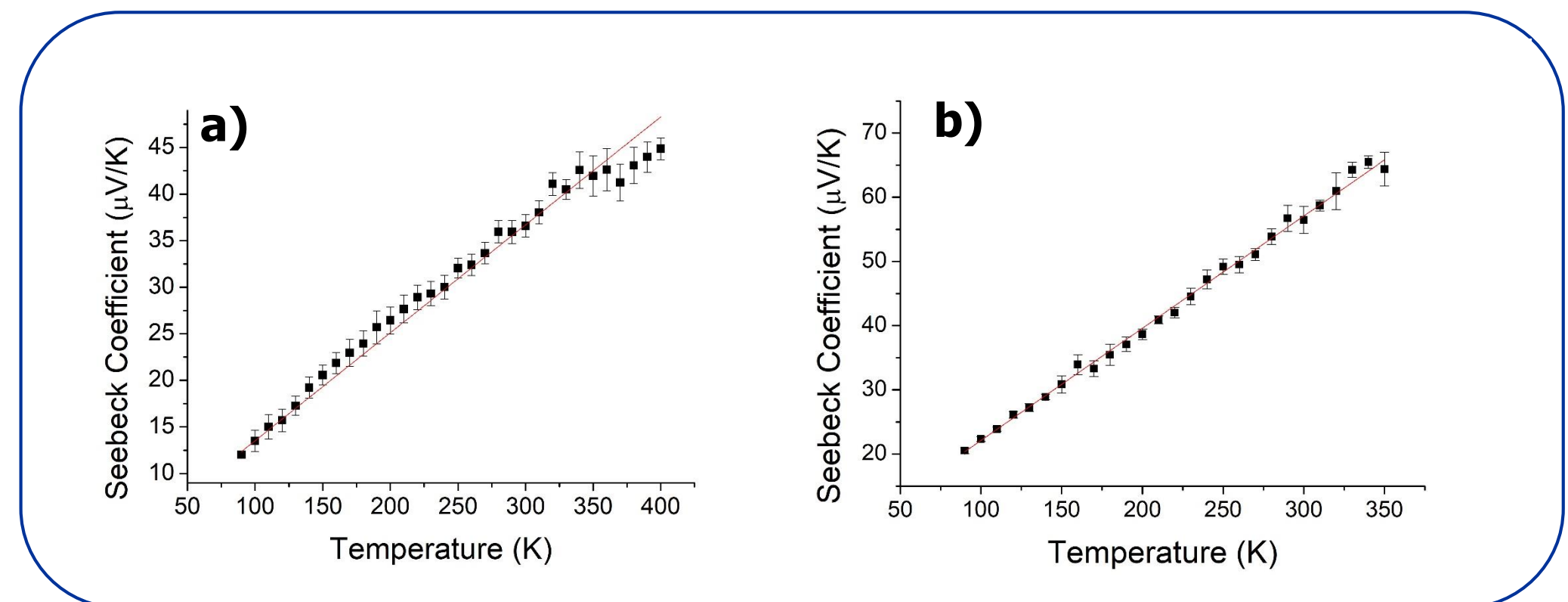


Figure 3. Seebeck coefficient versus temperature for the a) 10mg and b) 20mg EGO sample. The measurements represent one sample. The error bars represent one standard deviation for 10 measurements acquired at each temperature step.

Measurements using a MMR System yielded a linear proportionality between Seebeck coefficient and temperature, characteristic of metal charge transport, see Figure 3. Electric measurement were performed at room temperature using an Ossila Ltd linear four point probe and a Keithley 2680B.

The power factor was calculated as:

$$PF = \alpha^2 \sigma$$

the Seebeck coefficient being α and the electrical conductivity as σ .

10mg EGO @300K			20mg EGO @300K		
Seebeck Coefficient ($\mu\text{V}/\text{K}$)	Electric Conductivity (S/cm)	Power Factor ($\mu\text{W}/\text{mK}^2$)	Seebeck Coefficient ($\mu\text{V}/\text{K}$)	Electric Conductivity (S/cm)	Power Factor ($\mu\text{W}/\text{mK}^2$)
34.5 ± 2.7	200 ± 20	23.8 ± 3	56.5 ± 2.1	150 ± 35	48 ± 11.3

Figure 4. Seebeck coefficient, electrical conductivity and power factor of 10mg and 20mg EGO samples at room temperature.

The Seebeck coefficient and electrical conductivity showed a characteristic inversed proportionality when changing loading amount.

The power factor increased by a factor of 2 with increasing loading amount and it is higher than literature by an order of magnitude³.

The increased power factor with loading amount can be attributed to different unintentional oxidation states or defects within the samples increasing the Seebeck coefficient by changing the density of states near the Fermi level.

This method of electrochemical exfoliation is useful for thermoelectrics as it yielded large graphene flakes

Conclusions

- Seebeck coefficient measurement showed metallic charge transport for the EGO samples.
- Increased power factor with loading amount of EGO.
- Possible optimisation of power factor by tuning the loading amount of EGO.

References

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Acknowledgements

The authors would like to thank the Royal Society and the European Science Foundation (ESF) for financial support on this research.