

Graphite Exfoliation by an Alternative Mechano-Chemical Process Using Sulfur as Exfoliating Agent

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Exfoliated and expanded graphite (EG) are functional carbon-based materials with high porosity, flexibility and excellent thermal/chemical stability. They can be widely used in some fields such as: sewage treatment, gasket manufacturing, fire-resistant composite preparation, etc. [1]. Both are obtained through chemical oxidation or electrochemical method of graphite. However, the preparation involves too many steps and product washing consumes a lot of water and large amounts of energy during the thermal expansion step carried out at high temperature [2]. Due to the above, it is important to develop a simple and ecological route to synthesize EGs to use them for ecologic issues such as promissory candidates for pollutants reduction [3], etc. It is well known that mechanical milling is a simple route to prepare a wide variety of material with improved properties which does not generate wastewater.

The present work describes an alternative route based on a solid-state exfoliation of natural graphite based on mechanical milling and a selective lixiviation process. Graphite exfoliation was carried out following a mechanochemical method using pure graphite (Gr) flakes and sulphur (S) powder used as exfoliating agent. Eight grams of an equiatomic mixture of Gr-S were processed in a Spex 8000M high-energy mill using a ball to powder mass ratio of 5:1 for 4h. After milling, a lixiviation process was carried out using pure carbon disulphide (CS₂) as a selective solvent for sulphur (34.8 g of sulphur are dissolved in 100 g CS₂ at 25°C, whereas graphite is practically insoluble). The milled powders were refluxed in a Soxhlet extractor device with CS₂ for 20h, after extraction the solids were dried at 70°C for 1 h. Morphology and elemental distribution of samples was done through a SEM-JSM-7201F microscope.

Fig.1 shows SEM-EDS mapping (100X) of milled Gr-S samples where the bright green areas correspond to high S concentration regions (0h). After 4h of milling (4h), S was homogeneously dispersed in graphite, resulting in a uniform distribution. Fig. 2 displays the composition of samples before and after the extraction process. It can be observed the evident concentration reduction of S after the lixiviation treatment. After processing, CS₂ was distilled and pure sulphur was isolated, both chemical can be recycled for upcoming experimental. Fig. 3 exhibits some SEM micrographs of isolated graphite particles after processing (5 and 20 KX). It is evident the reduction of particle size and an increased level of defoliation reached with this process (in form of a huge number of isolated graphite layers and exposed surface).

References:

- [1] L. Zhongping et al., *Ceramics International* **44** (2018), p. 16256.
- [2] T.L. Ruijun Zhang et al., *Carbon* **119** (2017), p. 544.
- [3] Z. Hu et al., *Journal of Cleaner Production* **209** (2019), p. 1216.

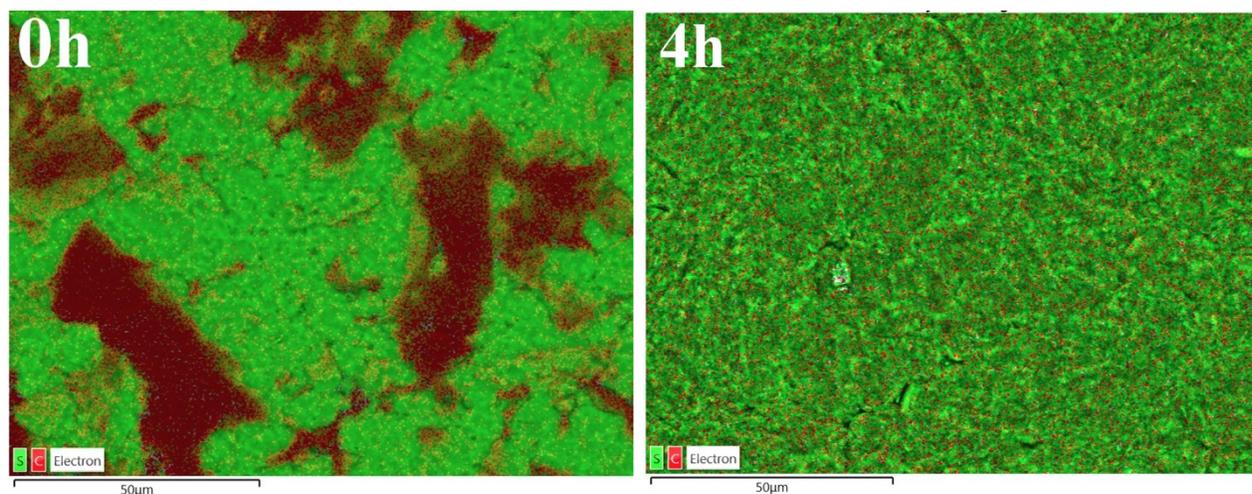


Figure 1. SEM mapping of un milled and 4h milled samples showing the distributions of S and C components.

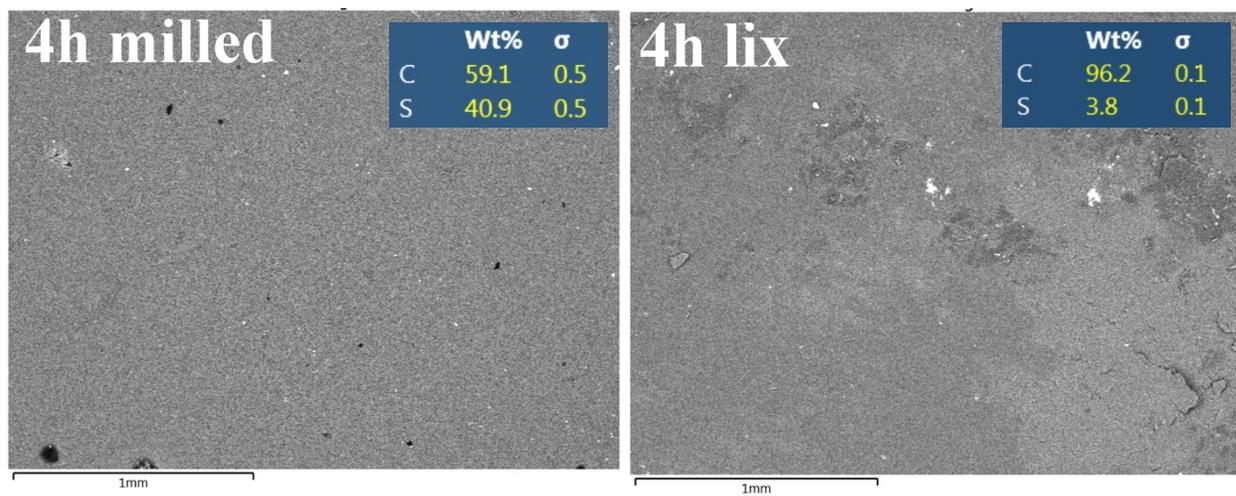


Figure 2. Low magnification SEM micrographs and EDS analyses of milled and extracted samples.

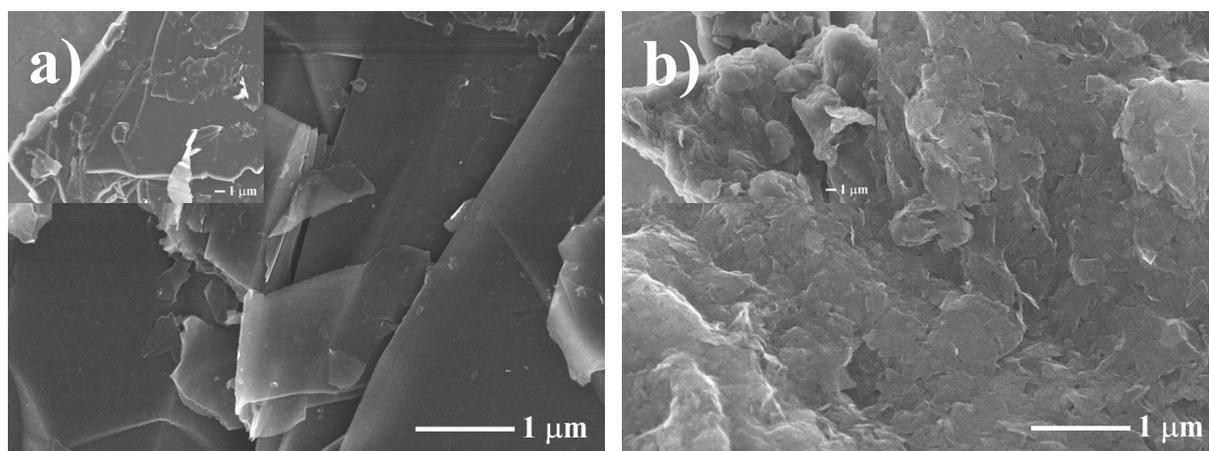


Figure 3. SEM micrographs of un milled (a) and 4h-extracted samples (b).