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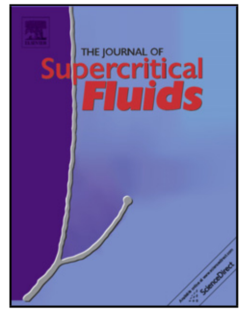

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Optimization of graphene production by exfoliation of graphite in supercritical ethanol: A response surface methodology approach

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Abstract: In the present study, graphene sheets were prepared via exfoliation of graphite in supercritical ethanol. Different characterization methods such as scanning electron microscopy (SEM), transmission electron microscopy (TEM), atomic force microscopy (AFM), X-ray diffraction (XRD) and Raman spectroscopy were used to identify the initial graphite and exfoliated samples. Characterization results revealed the successful exfoliation of graphite into single to few-layer graphene. The effects of main process parameters including temperature (325 – 425 °C), pressure (20 – 40 MPa) and mass percent of water as a co-solvent (0 – 50%) on the exfoliation yield were examined by using response surface methodology (RSM) in an experimental design. The maximum exfoliation yield of 18.5% was attained at the temperature of 325.1 °C, pressure of 39.8 MPa and water content of 28.9 wt%, as the optimum condition. In addition, results manifest the critical role of supercritical density in the graphite exfoliation. Hansen solubility parameters (HSPs) of supercritical fluid were calculated to investigate the exfoliation of graphite in terms of supercritical density. Results indicated that modification of HSPs of supercritical fluid by tuning the supercritical condition is an effective way of improving the exfoliation yield.

Key words: graphene; graphite; supercritical exfoliation; supercritical ethanol; Box-Behnken design; solubility parameter.

1. Introduction

Graphene is a wonder 2D material that has a planar structure and composed from completely sp^2 carbon atoms in a hexagonal lattice. This new member of carbon nanomaterial's family, possess many fascinating properties. Owing to its extraordinary features, There has been an extensive interest in graphene research in the past few years and significant portion of these studies have focused on preparation approaches [1]. Different methods were reported for graphene production that generally classified in two main approaches: bottom-up and top-down. Chemical vapor deposition (CVD) [2] and thermal decomposition of silicon carbide (SiC) which known as epitaxial growth [3], are the most studied approaches in bottom-up category. Although large area graphene can be obtained from these methods, but applying high temperature and usually vacuum, requires special equipment. In addition, applications of CVD grown graphene limited to post treatments for etching and transfer of graphene layer to desired substrates which can seriously damages the produced graphene. Also In the case of epitaxial growth, production of high quality graphene films needs single crystal SiC substrate which increases process costs.

On the other hand, top-down methods use graphite as a raw material. Chemical exfoliation route [4] is the best choice for bulk production of graphene, in which oxidation, exfoliation and reduction of graphite performed subsequently. However, there are some problems associated with this method, especially low quality of graphene and toxic chemical reagents that used in the oxidation and reduction steps. Other top-down methods like micromechanical cleavage [5] and liquid phase exfoliation [6], suffer from low production yield and cannot be used in commercial scale.