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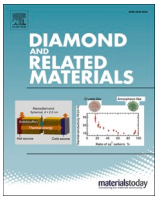

Combination of cellulose tissue paper and bleach-treated graphene in stiffness reinforcement of polyvinyl alcohol film

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ABSTRACT

A pre-treatment of graphene with bleach is considered one of the possible purification methods after liquid-phase exfoliation. However, the effect of this treatment on the mechanical reinforcement strategy for polymer film is yet to be investigated to date. In this full work, the influence of the C/O ratio, I_D/I_G, and volume of graphene after combination with cellulose tissue on the resulting stiffness of polyvinyl alcohol (PVA) composite film has been extensively studied. It is noticed that the incorporation of 30 ml graphene that had been pre-treated for 3 h into PVA had produced the best increment in elastic modulus (1.6 GPa against 0.4 GPa) while a shorter pre-treatment duration of graphene (1 h) would require more graphene volume (40 ml) to match the previous stiffness improvement level. By using the collected experimental data (90 samples), we further modeled the effect of tissue and PVA mass, C/O ratio, I_D/I_G, and graphene volume on modulus using machine learning (ML) algorithms.

1. Introduction

In designing graphene-based polymer composite film for various technological applications such as EMI shields [1], sensors [2], actuators [3], thermal management [4], and electronic components [5], the influence of mass, volume, flakes size, and interfacial compatibility between the graphene and host polymer is crucial for producing the targeted impactful mechanical reinforcement effect. Due to the attractive properties of graphene, whereby it not only possesses a strong theoretical elastic modulus [6] but is also able to conduct electricity [7] and is thermally stable [8], the prospect of successful incorporation of graphene into polymer has significantly motivated a lot of works focusing on the fabrication strategy [9,10], synthesis of modified graphene [11] and combination of multiple polymer [12] for the preparation of strong and flexible polymer composite.

One of the bio-friendly synthesis strategies for the preparation of polyvinyl alcohol (PVA)-compatible graphene is the liquid-phase exfoliation of graphite in polyphenols aqueous medium [13–15]. However, it is noticed that the purification of flakes against the adsorbed stabilizer after the exfoliation stage remains a challenge to date. While cascading centrifugation can be used for separation, size selection, and even graphene washing [16], the low removal efficiency of residue per centrifuge cycle greatly hinders the scalability of this process. To accelerate the washing step, a pre-treatment of graphene supernatant with bleach consisting of sodium hypochlorite (NaClO) has been recently reported with the maximum obtainable C/O of 8.1 and I_D/I_G of 1.0 in value [17]. Despite the progress, it is observed that graphene may lose its stability in water due to the polyphenol removal effect by bleach and this limitation certainly could limit its potential as a filler for various hydrophilic polymers such as PVA. In the presence of NaClO, the radical OH- from self-decomposition of NaClO at ambient will first oxidize the defect site (oxygen terminated regions) of graphene and lead to the decomposition of C-OH and C-O-C over time. Further hydrolyzation by radical OH- on free and unsaturated sites on graphene will create new C-OH terminals and further convert the sp^2 of graphene to sp^3 regions. The decomposition of oxygen site and hydrolyzation of graphene will repeat itself during the exposure to bleach and provided that the presence of radical OH- ion in the graphene supernatant is still active. On other hand,

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