Macromolecular Chemistry and Physics
www.mcp-journal.de

Effect of the Functional Groups of Polymers on Their Adsorption Behavior on Graphene Oxide Nanosheets

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Graphene-based polymer nanocomposites are emerging materials for both fundamental research and industrial applications. The influence of polymer functional groups on their adsorption behavior onto graphene oxide (GO) nanosheets is investigated, with poly(methyl methacrylate) (PMMA), poly(methyl methacrylate-co-methacrylic acid) (PMMA-co-MAA), and poly(methacrylic acid) (PMAA) having the same backbone but different functional side groups selected as the model polymers. Fourier transform infrared spectroscopy and X-ray diffraction results confirm the interfacial interaction between the polymer and GO. Thermogravimetric analysis reveals notable enhancements in the amount of the adsorbed polymer up to 30.6 wt.% for PMMA-co-MAA/GO and 49.7 wt.% PMAA/GO compared with 18.7 wt.% for PMMA/GO. The water contact angle decreases from 71.3° for PMMA/GO to 69.1° for PMMA-co-MAA/GO and to 61.2° for PMAA/GO. The further washing process reduces the adsorption amount for the polymer/GO hybrid. Overall, the polar functional groups of the polymer directly influence the polymer adsorption behavior onto GO.

1. Introduction

The unique structure and properties of graphene have attracted attention from researchers and engineers over the last two decades since the seminal work of Geim et al.[1] that isolated single graphene sheets from graphite. The single two-dimensional graphene sheet is one-atom-thick and has a high surface area of 2630 m^2 g^{-1}.[2] Graphene is the stiffest and strongest material ever measured, possessing exceptional thermal, mechanical, and electrical properties.[1,3] Graphene offers an extraordinary opportunity to improve the performance of polymers in comparison with microfillers,[4] and it has been studied for a wide range of applications, such as gas barrier,[5] mechanical performance,[6,7] hydrogen storage,[8] and supercapacitors.[9]

To achieve a stable and homogeneous nanofiller dispersion in the polymer matrix, their strong interfacial interaction is key, which may lead to property enhancements in polymer nanocomposites.[10,11] Therefore, a lot of investigations have been focused on enhancing the homogeneity of the filler dispersion and interfacial interactions by functionalizing nanofillers, such as silica nanoparticles,[12,13] and alumina powders[14] to improve the compatibility of nanofillers with polymers. This offers a possibility for the use of polymer nanocomposites in various applications, such as medical,[15] electrical,[16] optical,[17] and mechanical.[18] For instance, Sui et al.[12] investigated hairy silica nanoparticles (HSNs) modified with different functional groups as additives in polyalphaolefin (PAO). They found that the HSNs functionalized with $-NH_2$ groups showed the best friction resistance and anti-wear properties, yet the non-polar groups, such as $-C_6H_5$ and $-CH_3$, rendered HSNs with better dispersion in the PAO matrix.

Graphene lacks polar functional groups in the structure, resulting in poor compatibility with polar polymers.[19] So when mixing with a polymer, graphene sheets tend to undergo agglomeration due to the strong π–π and van der Waals interactions.[20] Compared to graphene, graphene oxide (GO) is abundant in functional groups, such as hydroxyl, carbonyl, epoxide, and carboxyl, which are important for improving interfacial interactions.[21,22] Different functionalization methods, including noncovalent and covalent modification,[23] have been applied to expand the applications of graphene and GO.[24] Noncovalent and covalent of functionalized graphene composites were reported to possess stable, good electrocatalytic activity and high conductivity.[22,25]

To the authors' knowledge, the previous work on polymer nanocomposites was mainly focused on the functionalization of nanoparticles such as silica,[12,13,26] graphene,[22,25,27] and GO,[24] with 3-aminopropyl triethoxysilane,[28] amine,[12,13,26] amide, thiourea, carbamate functional groups, etc.[29] However, the effect of the functional groups of polymers has not been systematically investigated, which is considered important in affecting the dispersion of nanofiller and the interfacial interactions[30] and so the macroscopic properties of polymer nanocomposites. This

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DOI: 10.1002/macp.202300101

