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Enhanced mechanical, crystallisation and thermal properties of graphene flake-filled polyurethane nanocomposites: the impact of thermal treatment on the resulting microphase-separated structure

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Abstract

The present work investigates the combined effect of the addition of graphite nanoplatelets (xGNPs) to polyurethane copolymer (PUC) and thermal treatment was employed. The PUC reinforced by xGNP were synthesized by in-situ polymerisation, which leads to an effective performance of the PUC/xGNP system. Meanwhile, X-ray diffraction (XRD) and Raman spectroscopy tests displayed the inter-spacing planar quality of xGNP nanofillers. The thermal stability of PUC was seen to increase with addition of xGNP. Additionally, the dynamic storage modulus (E') showed better performance after thermal treatment than in the untreated samples. However, a relationship between the microphase-separated morphology of PUC induced by thermal treatment and the addition of xGNP has been observed. Consequently, the crystallinity percentage increased after thermal treatment @ 80 °C for 4 days, presuming a re-ordering of amorphous hard segments during the heating in a packed microphase conformation. On the other hand, better dispersion and interaction of xGNP can play a crucial role in enhancing the thermal and mechanical properties, and thus a significant reinforcement for PUC. The tensile properties such as modulus and tensile strength showed significant enhancement with xGNP incorporation, while the elongation steeply dropped. On the contrary, a deterioration in modulus and tensile strength resulted from thermal treatment, likely due to the restacking of xGNP during segmental movement and thus increasing the amorphous phase rather than the crystalline phase. A modified Halpin—Tsai model was utilised to predict the mismatch between the empirical and theoretical results. Consequently, the findings displayed the divergence of the nanocomposite modulus of PUC with greater amounts of xGNP nanofillers.

 $\textbf{Keywords} \ \ Polyure than e \ copolymer \cdot PUC \cdot Microphase-separated \ structure \cdot Thermal \ and \ mechanical \ properties \cdot Halpin-Tsai \ models$

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Introduction

Polyurethane copolymer (PUC), classified as a type of thermoplastic polyurethane, has attracted tremendous attention among the research community as well as in industrial fields. Commonly, polyurethane is considered one of the most versatile plastic materials derived from different kinds of raw chemicals such as MDI. The MDI is responsible for hard domains and polyether or polyester polyol which represents the soft domain and thus reflects the PUC flexibility that is needed for different applications [1–5]. Phase separation between hard and soft segments, as well as hydrogen bonding that exists between urethane bonds, have a high impact on the resultant mechanical properties of this polymer [6–8].

Polymer nanocomposites (PNCs) are a good candidate for use instead of virgin polymers. They have a massive interfacial area between the polymer matrix and the additives