

Synthesis and characterization of nylon-610 phase change materials with varied reduced graphene oxide filler ratios

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Abstract. This study delves into the synthesis and comprehensive characterization of Nylon-610 (PA610) phase change materials (PCMs), augmented with varied ratios of reduced graphene oxide (rGO) fillers, to assess their compatibility and integration within the polymer matrix. Utilizing condensation polymerization, PA610 was synthesized and incorporated with rGO fillers across a spectrum from 0 to 2.0 wt%, aiming to explore the interaction between the polymer and the fillers. The characterization of the synthesized composites was meticulously conducted through Fourier Transform Infrared Spectroscopy (FTIR) and X-Ray Diffraction (XRD) analyses. The FTIR results provided insights into the chemical bonding and interactions between PA610 and rGO, indicating successful integration of the fillers into the polymer matrix. Concurrently, XRD analysis revealed changes in the crystalline structure of PA610 with the introduction of rGO, suggesting an alteration in the material's microstructure due to filler addition. This research aims to illuminate the fundamental aspects of filler compatibility within polymer based PCMs, offering a novel perspective on the design and development of advanced materials for energy storage applications.

1 Introduction

In the pursuit of sustainable energy solutions, thermal energy storage systems have gained significant prominence, particularly for their ability to mitigate the intermittency challenges inherent in renewable energy sources [1, 2]. Among the various strategies explored, Phase Change Materials (PCMs) stand out for their remarkable capacity to harness and store energy through phase transitions, predominantly in the solid-liquid domain [3, 4]. These materials are celebrated for their high latent heat storage capabilities and minimal volume fluctuations during phase transitions, rendering them invaluable in the quest for energy efficiency across diverse industrial applications [5].

Despite their potential, the application of solid-liquid PCMs in polymer matrices often encounters the issue of material compatibility. This issue encompasses the molecular assimilation of fillers and the preservation of the polymer's structural integrity, aspects crucial

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for the operational viability of PCM composites [4]. In this regard, the incorporation of nano-fillers, particularly reduced graphene oxide (rGO), has garnered considerable attention [6]. Current research highlights the critical role of filler compatibility in sustaining the structural and functional integrity of polymer-based PCMs [7, 8]. The compatibility of rGO within the PCM matrix ensures uniform dispersion, which is crucial for effective heat transfer and storage. This uniformity prevents phase separation and maintains the material's latent heat capacity. Furthermore, the interaction between rGO and the polymer matrix can enhance the mechanical strength of the composite, making it more durable under repeated thermal cycling [9].

The significance of this work lies in its pioneering exploration of integrating reduced graphene oxide (rGO) into a Nylon-610 (PA610) polymer matrix for phase change materials (PCMs), a venture that merges the realms of advanced nanomaterials with thermal energy storage systems. By unveiling the intricate molecular and structural dynamics between PA610 and rGO through cutting-edge Fourier Transform Infrared Spectroscopy (FTIR) and X-Ray Diffraction (XRD) analyses. This study not only elucidates the chemical compatibility but also suggests the potential for rGO to enhance the thermal properties of PCMs, which can be further investigated in future work.

2 Materials and methods

The synthesis of PA610 PCM was achieved through condensation polymerization, with variations in monomer concentration and reduced graphene oxide (rGO) loading to explore their effects on the PCM's properties. Hexamethylenediamine (HMDA) was purchased from Merck with a purity of 98% and dissolved in distilled water. Sebacoyl chloride, purchased from Acros Organics with a purity of 95%, was dissolved in n-hexane, which was purchased from Merck with a purity of 95%. Reduced graphene oxide (rGO) was acquired from GO Advanced Solutions Sdn. Bh. Two sets of PA PCM with varying rGO loadings (0 – 2.0 wt.%) were prepared. One set was washed with distilled water, and the other set was washed with ethanol. Both sets were then dried in an oven at 80°C for 48 hours.

3 Results and discussion

3.1 Fourier transform infrared spectroscopy (FTIR)

The FTIR spectrum of pure reduced graphene oxide (rGO) as shown in Figure 1 establishes a foundational understanding of its chemical structure. The observed peak at 3449 cm^{-1} , indicative of the C–OH stretching band, points to the presence of hydroxyl groups typically associated with the binding water molecules in graphene oxide [10]. Concurrently, the distinctive peak at 1638 cm^{-1} represents the C=C stretching vibrations, reflecting the unoxidized graphitic domains that are quintessential to graphene oxide's unique properties [11]. This dual presence of structural water and graphitic domains provides insight into the reduced but not fully eliminated oxygenated functionalities within rGO, laying the groundwork for its interaction with the polymer matrix in the composites.

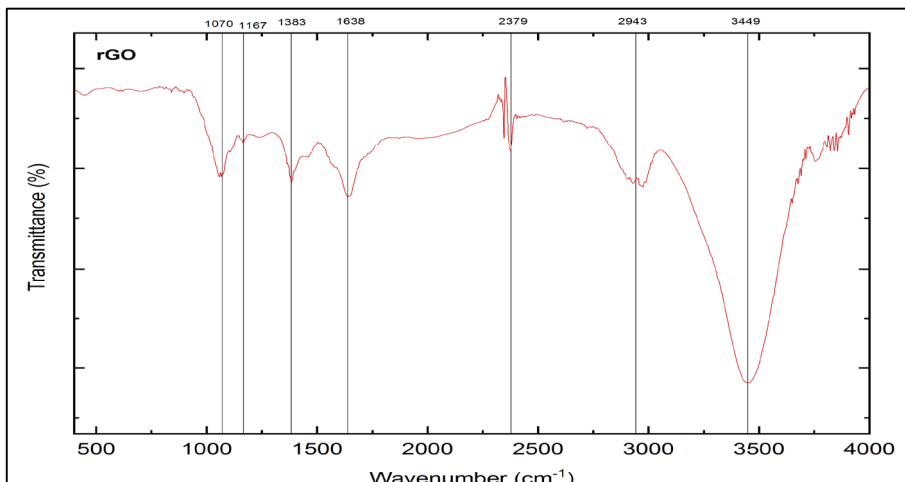


Fig. 1. FTIR spectra of reduced graphene oxide.

The FTIR spectra for PA PCMs washed with distilled water presented in Figure 2 which reveals the details of the polymer-filler interaction. The N–H stretching vibrations at 3307 cm^{-1} and the amide C=O stretch at 1698 cm^{-1} are consistent across the spectrum, suggesting a stable amide linkage throughout the composite [12]. However, the rGO introduction's most telling impact is the reduced intensity of key peaks, such as those at 1240 cm^{-1} related to the C–N stretch and at 932 cm^{-1} associated with O–H bending vibrations[13]. The diminished peak intensities are indicative of rGO's interaction with PA610 chains, likely through a reaction during polymerization that modifies the amide functional group, affirming the compatibility of rGO within the polymer matrix.

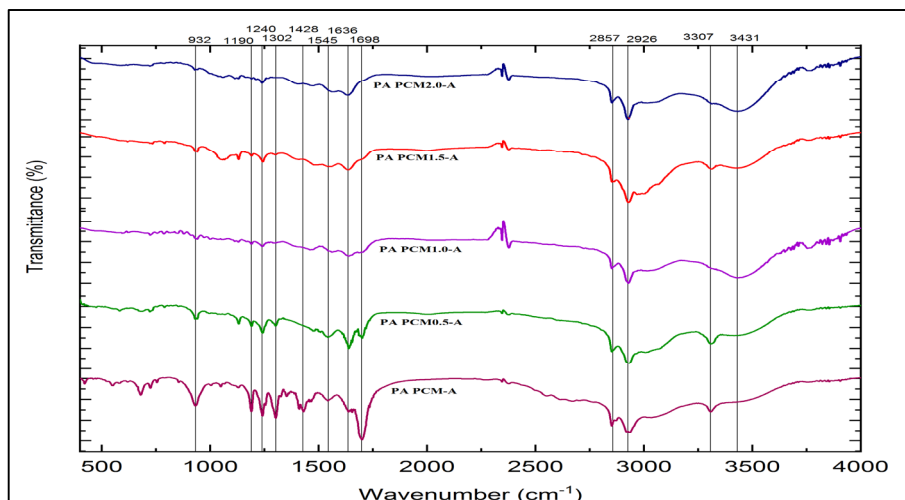


Fig. 2. FTIR spectra of polyamide PCMs washed with distilled water.

In contrast, Figure 3 illustrates the FTIR spectra of PA PCMs washed with ethanol with variations in peak intensities. Specifically, the O–H stretch at 3431 cm^{-1} shows increased intensity in all PA PCMs washed with ethanol, which can be attributed to the presence of ethanol in the samples. The presence of the peak at 1056 cm^{-1} , corresponding to the C–O stretch, further confirms the presence of ethanol [14]. While the intensity of the peak at 1240

cm^{-1} does not decrease with increased rGO loading in ethanol-washed samples, likely due to ethanol preventing interaction between rGO and the C-N functional group. This suggests that washing with ethanol hinders the interaction between rGO and the polyamide chains, affecting the overall chemical structure and interactions within the composite [15].

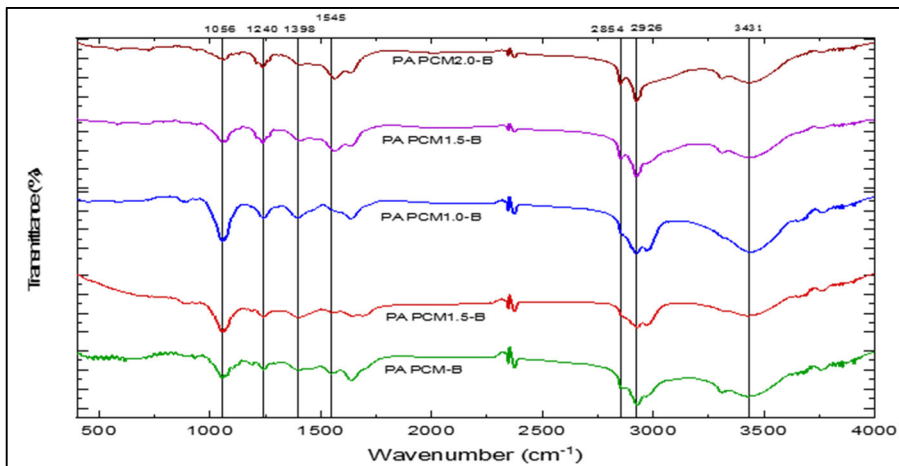


Fig. 3. FTIR Spectra of Polyamide PCMs washed with ethanol.

The FTIR spectral analysis of PA610 composites with reduced graphene oxide (rGO) confirms the chemical structures and interactions within the materials. Pure rGO spectra show hydroxyl groups and graphitic domains, indicating preserved oxygenated functionalities post-reduction. The PA PCMs washed with distilled water exhibit a clear amide bond presence alongside evidence of rGO-polymer interactions through altered peak intensities. In contrast, samples washed with ethanol show better preservation of functional groups, suggesting lesser interaction between rGO and the PA610 matrix due to the solvent's effects.

Additionally, the polymerization scheme presented in Figure 4 aligns with the FTIR findings, verifying the successful synthesis of PA PCMs and retention of key functional groups. This synthesis confirmation is crucial for optimizing the material's properties for specific applications, demonstrating the importance of integrating empirical FTIR data with theoretical models.

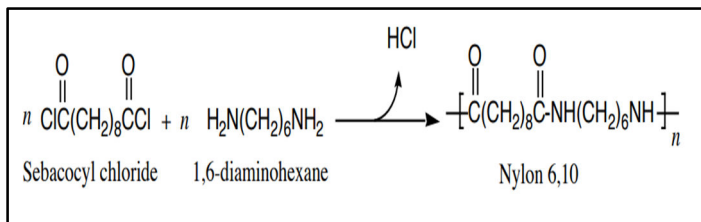


Fig. 4. Polymerization scheme of PA 610.

3.2 X-Ray diffraction (XRD)

X-ray diffraction (XRD) analysis is crucial for understanding the structural changes in Nylon-610 (PA610) phase change materials (PCMs) when combined with nano-fillers like reduced

graphene oxide (rGO). XRD helps assess crystallinity and phase composition, which are vital for material optimization for specific uses.

Figure 5 shows XRD patterns of PA610 PCMs washed with distilled water with clear peaks at 2θ values of 21.5° and 23.9° , representing the crystalline structure of PA610, specifically the (110) diffraction planes. The addition of rGO introduces a new peak at 26.6° , absent in pure PA610, which corresponds to the (002) diffraction plane of rGO [17]. This peak, which increases in intensity with higher rGO content, suggests significant interaction between the polymer chains and rGO, altering the composite's structural properties.

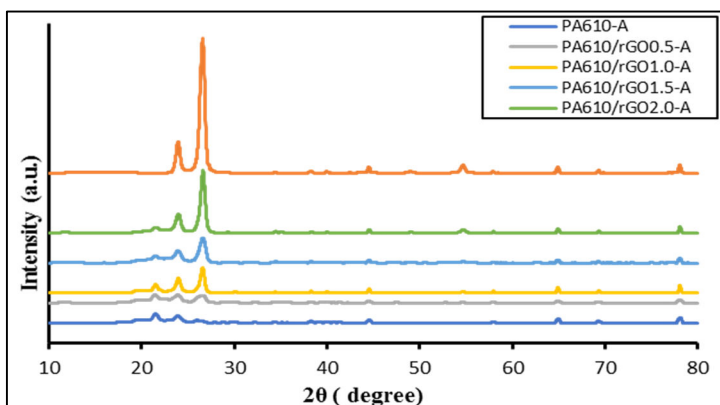


Fig. 5. XRD patterns of polyamide PCMs washed with distilled water.

Conversely, Ethanol-treated PA610 PCMs display distinct XRD characteristics compared to those washed with distilled water as shown in Figure 6. The presence of ethanol affects the crystallinity, as indicated by reduced intensity and broadened peaks, suggesting a more amorphous structure and altered interaction with the PA610 matrix. This modification potentially affects the dispersion and integration of rGO within the composite. Specifically, the broader, less-defined peaks at 21.5° and 23.9° highlight a shift towards an amorphous nature in these ethanol-washed samples, reflecting variations in the internal structure influenced by the solvent [18]. Additionally, the XRD patterns of pristine rGO show pronounced peaks at 23.9° and 26.6° , with the latter intensifying with increased rGO content, indicating its progressive integration within the PA610 matrix.

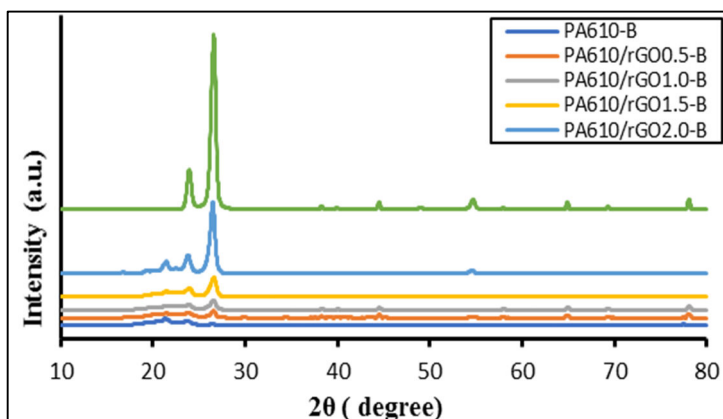


Fig. 6. XRD patterns of polyamide PCMs washed with ethanol.

Overall, the XRD analysis of PA610 PCMs, treated with both water and ethanol, reveals the complex effects of solvent treatment and rGO filler on the crystalline structure. The clear differences in XRD patterns between the two sample types highlight how rGO integration and solvent choice influence material crystallinity.

4 Conclusion

The study successfully synthesized Nylon-610 (PA610) phase change materials with varying concentrations of reduced graphene oxide (rGO) fillers, focusing on the compatibility and integration of rGO within the PA610 matrix. Characterization through FTIR and XRD analyses revealed significant molecular and structural interactions between PA610 and rGO. FTIR analysis confirmed specific interactions and chemical bonding, indicating a high degree of compatibility and homogeneous dispersion of rGO. XRD results supported this by showing changes in crystallinity and microstructure upon rGO addition, demonstrating effective filler integration. The study underscores the potential of rGO as a compatible filler in polymer based PCMs, particularly in enhancing the material properties essential for energy storage applications. The observed chemical and structural compatibility between PA610 and rGO pave the way for future research into optimizing these composites for enhanced thermal performance. This research contributes to the growing body of knowledge on the development of advanced materials for energy storage, highlighting the importance of filler compatibility in the design and synthesis of effective PCMs.

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