Evaluation of Sustainable Alternative Fillers in Polymer Systems by Dynamic Mechanical Thermal Analysis and the Payne Effect

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In this study, the conventional filler, carbon black N339, was substituted by alternative fillers, including glass sludge, wood flour, ash and carbon black derived from the pyrolysis of waste tires. In order to facilitate a more accurate comparison, all alternative fillers were prepared with the same particle size, ranging from 25 to 40 μm. The present study evaluates the influence of four different alternative fillers incorporated into an elastomeric polymer matrix, with the aim of identifying sustainable and high-performance substitutes for conventional reinforcing filler. Six polymer systems were prepared for the research, with the following labels: U, S1, W2, A3, C4 and N5 (Fig. 1). The incorporation of filler has a visible effect on the curve of mixing, where a decrease is noted caused by longer addition of more filler. Subsequently, during the 180-second mixing process, the curve demonstrates an upward trend, attributable to the augmentation in the viscosity of the compound and the friction within the mixing chamber resulting from the incorporation of the filler. The temperature increase is influenced by the type of filler used. The presence of a more active filler in the polymer sample is likely to result in a higher temperature rise during mixing. Depending on the temperature increase during the incorporation of the filler into the elastomeric polymer matrix, it can be written as follows: U< S1< A3< W2= C4< N5.

The highest tan δ peak (Fig. 2) was observed for the glass sludge-filled polymer sample (S1), followed by the pellet ash-filled polymer sample (A3).The unfilled polymer sample (U) also exhibited a relatively high tan δ peak, reflecting the greater mobility of polymer chains in the absence of reinforcing filler. A decline in tan δ peak values was observed for the polymer sample with the carbon black derived from the pyrolysis of waste tires (C4) and for the polymer sample with the conventional carbon black (N5). The lowest damping peak was observed in the wood flour-filled polymer sample (W2), indicating an increased elastic response and reduced viscous energy dissipation. The loss modulus curves (Fig. 3) further illustrate the influence of fillers on energy dissipation during dynamic loading. The maximum of E″ corresponds to the maximum viscous energy dissipation associated with the glass transition. The polymer sample N5 exhibited the highest E″ peak, suggesting robust filler–polymer interactions and substantial internal friction within the filled network. A comparable yet marginally diminished peak was detected for the polymer sample W2, indicating the potential involvement of lignocellulosic particles in mechanical energy dissipation. The storage modulus curves (Fig. 4) illustrate substantial variations in stiffness among the polymer samples under investigation within the glassy region. The polymer sample N5 demonstrated the highest values of E′, with the polymer sample W2 exhibiting closely related values. The high modulus values indicate strong reinforcing effects and reduced segmental mobility of the polymer chains.

In circumstances of minimal strain, the polymer samples exhibit a high storage modulus (G') due to the presence of filler-filler networks. As the magnitude of strain increases, these networks are subject to disruption, resulting in a rapid decrease in G'. It has been demonstrated that, upon being subjected to stress and strain, the rubber in question ceases to behave as an elastomer and becomes a rigid filler. This transition is accompanied by an increase in the G' modulus, indicative of an elevated degree of filler aggregation. As demonstrated in Figure 5, the storage modulus has been found to exhibit an increasing trend in response to the incorporation of various types of fillers. A close examination of the data reveals that the storage modulus (G') for both the uncured and cured NS and WS samples is higher. The observed increase can be attributed to enhanced filler-filler interactions (i.e., higher reinforcement) within the matrix. The Payne effect is characterized by a decrease in the storage modulus (stiffness) of filled elastomers. The Payne Effect (Tab. 1) will be amplified in proportion to the magnitude of the value of ΔG'. The polymer samples designated as WS and NS exhibited the highest values for the standard free energy change (ΔG') both prior to and following the application of the curing agent. The findings of the study demonstrate the potential of wood-based by-products and recycled pyrolytic residues as effective, environmentally beneficial fillers for polymer systems.