
DYNAMIC MECHANICAL ANALYSIS OF RECYCLED LDPE/HDPE POLYMER BLENDS

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ABSTRACT

This study investigated the dynamic mechanical behaviour of blended low-density polyethylene (LDPE) and high-density polyethylene (HDPE) waste plastics using Dynamic Mechanical Analysis (DMA). The aim was to evaluate the effect of HDPE loading on the viscoelastic and damping properties of LDPE/HDPE blends. Samples with varying HDPE loadings (0–50 wt.%) were prepared by melt compounding on a two-roll mill machine, followed by compression moulding. DMA was conducted at a frequency of 1 Hz over a temperature range of 30–120 °C at a heating rate of 6 K/min. The results showed that increasing HDPE content significantly improved the mechanical performance of the blends. Relative to pure LDPE, the storage modulus increased by approximately 573.4%, while the loss modulus increased by approximately 426% at 50 wt.% HDPE loading. The $\tan \delta$ values ranged from 0.13 to 0.27 across all blends within the investigated temperature range of 30–120 °C, indicating good damping characteristics. The findings demonstrate that HDPE incorporation enhances the stiffness and mechanical strength of LDPE-based blends, indicating the potential of recycled LDPE/HDPE waste plastics for engineering and structural material applications.

Keywords: High-density polyethylene (HDPE); low-density polyethylene (LDPE); plastics; polymer blend; Dynamic mechanical Analysis (DMA)

1. INTRODUCTION

Polyethylene-based plastics have become highly valued in modern engineering and industrial systems owing to their lightweight, chemical inertness, ease of processing, and low production cost. Among the various polyolefins currently in circulation, Low-density polyethylene (LDPE) and high-density polyethylene (HDPE) contribute to a much larger share of global plastic production and waste than most other plastics (Vannessa, 2004).

In 2023, global plastic production was estimated at around 415 million metric tons, roughly double the amount produced at the beginning of the 21st century, yet only approximately 9% of all plastic waste generated to date has been successfully recycled (Surendarnath et al., 2025). This is a challenging situation in Nigeria, where limited waste sorting and recycling infrastructure results in LDPE and HDPE being dumped together with other waste materials, worsening environmental pollution (Akan et al., 2021). A cost-effective method for recovering value from mixed polyethylene waste is heat blending, where LDPE and HDPE are processed together in the molten state to give a material with properties that can be tailored by adjusting the blend ratio.

Evaluating the mechanical behaviour of polymer blends under real service conditions requires more than conventional tensile or flexural tests, since polymers exhibit viscoelastic behaviour influenced by temperature, loading frequency, and the duration of deformation. Dynamic Mechanical Analysis (DMA) is widely used to investigate this behaviour, providing key parameters including the storage modulus (E'), the loss modulus (E''), and the damping factor, $\tan \delta$, expressed as the ratio of E'' to E' (Chartoff et al., 2009).

Although interest in recycled polyolefin blends has increased in recent years, several important gaps still exist in the available literature. Most studies on LDPE/HDPE blends have used virgin polymers with controlled and well-defined properties (Schyns & Shaver, 2021). In contrast, recycled plastics are often more complex, containing impurities and variations in composition and quality (Nagy & Weltsch, 2023). Previous research has also focused largely on the tensile, thermal, and rheological

properties of LDPE/HDPE blends, with comparatively few studies examining their dynamic mechanical behaviour, particularly for materials derived from post-consumer plastic waste, and information is especially limited over the temperature range commonly encountered in service conditions (30–120 °C).

To help address these gaps, the present study investigates the dynamic mechanical behaviour of binary LDPE/HDPE blends prepared from post-consumer plastic waste collected in Zaria, Nigeria. The blends were produced with HDPE contents of 0wt.%, 10wt.%, 20wt.%, 30wt.%, 40wt.%, and 50 wt.% using a melt-compounding process. The specific objectives of the study are to:

1. Examine the effect of increasing HDPE content on the storage modulus (E'), loss modulus (E''), and damping factor ($\tan \delta$) of recycled LDPE/HDPE blends over the temperature range of 30–120 °C at a frequency of 1 Hz and a heating rate of 6 K/min.
2. Determine how blend composition affects key thermomechanical transition temperatures, including the onset, peak, and offset temperatures.
3. Evaluate whether the dynamic mechanical properties of the blends make them suitable for engineering and structural applications.

2. METHODOLOGY

2.1 Materials

Post-consumer low-density polyethylene (LDPE) waste used in this study was obtained from discarded sachet water packaging films collected within Samaru, Zaria, Kaduna State, Nigeria. High-density polyethylene (HDPE) waste was sourced from used yoghurt packaging containers (“San yoghurt” plastic) collected from the same location.

It is acknowledged that the use of post-consumer waste plastics introduces inherent variability that virgin polymer studies do not face. In this study, material identity was inferred from the known source of the plastics, such as sachet water films (LDPE) and yoghurt containers (HDPE). These are commonly used product categories with well-documented polymer types in the Nigerian context. However, formal characterisation of the raw materials using techniques such as Fourier-transform infrared spectroscopy (FTIR), differential scanning calorimetry (DSC), or melt flow index (MFI) testing was not conducted in this study. This represents a limitation, as residual contamination from labels, adhesives, or mixed plastics cannot be completely ruled out despite the washing procedure employed. Future work should incorporate material characterisation of the recycled feedstock before blending to quantify contamination levels and confirm polymer identity, allowing a more definitive interpretation of the dynamic mechanical results.

2.2 Collection and Preparation of Waste Plastics

LDPE and HDPE post-consumer waste plastics were collected from designated waste bins and disposal locations. The collected plastics were sorted manually to separate LDPE from HDPE contaminants and other foreign materials. The sorted plastic materials were thoroughly washed with water and liquid soap to remove dirt, grease, labels, and adhered impurities that could interfere with the blending and thermal analysis processes.

After washing, the plastic materials were not dried under direct sunlight to avoid potential degradation and distortion of the LDPE sachet water films, which are highly sensitive to prolonged solar exposure. Instead, the samples were dried in a well-ventilated room under continuous fan-assisted airflow for approximately 4 days. This approach was adopted to ensure gradual and uniform removal of moisture while preserving the physical integrity and intrinsic properties of the polymers prior to further processing. Complete drying was necessary to prevent moisture-induced defects during melt blending and compression moulding operations. Drying was considered complete when no visible moisture was observed, and the flakes remained free-flowing and non-adherent upon handling.

The dried plastic materials were subjected to size reduction using a locally fabricated shredding machine to obtain relatively uniform polymer particle chips of about 10mm for effective blending.

2.3 Preparation of LDPE/HDPE Blends

Mass measurements of the polymer samples were conducted using a digital weighing balance. The polymer blends were prepared using a two-roll mill compounding machine (Reliable Rubber and Plastic Company, New Jersey, U.S.A., Model 5189), operated at a processing temperature of 180°C for 10 - 15 minutes. The two-roll mill compounding step used a higher temperature (180°C) to ensure adequate melt flow, shear mixing, and homogenization of the LDPE/HDPE blend across the open rollers, where heat loss to the environment is greater. The shredded polymer materials were introduced into the heated rollers and blended thoroughly to ensure proper dispersion and homogenization of the polymer phases.

The blends were prepared using different LDPE/HDPE formulations as follows:

- 200 g/0 g, 180 g/20 g, 160 g/40 g, 140 g/60 g, 120 g/80 g, 100 g/100 g

These formulations were prepared to evaluate the influence of HDPE loading on the dynamic mechanical properties of the polymer blends.

2.4 Compression Moulding of Polymer Blends

Compression moulding of the blended samples was carried out using a Carver compression moulding machine (Carver Inc., Wabash, U.S.A., Model 5019).

The compounded polymer blends obtained from the two-roll mill were processed using the compression moulding technique to produce sheet samples suitable for dynamic mechanical analysis (DMA). The mould platens were electrically heated to a temperature of 150 °C and allowed to stabilise before processing. Compression moulding, by contrast, was carried out on the already-compounded (pre-homogenized) blend in a closed, pressurized system; a moulding temperature of 150°C, above the melting points of both LDPE (~105–115°C) and HDPE (~130–137°C) was sufficient to ensure both polymer phases were fully molten and could consolidate into a uniform sheet, while minimizing additional thermal exposure and reducing the risk of oxidative degradation during the moulding dwell time.

The mould was then preheated to the same temperature, after which the plastic paste was placed into the mould cavity. Compression moulding was carried out at a pressure of 3 MPa for 5 min to produce sheets with dimensions of 240 mm × 220 mm × 4 mm. Before moulding, the mould surfaces were coated with used engine oil to facilitate easy removal of the cured samples. After curing, the moulded samples were removed and allowed to cool to room temperature. The moulded sheets were subsequently cut into smaller specimens with dimensions of 60 mm × 12 mm × 4 mm for DMA characterisation. The prepared samples were then stored under ambient laboratory conditions before testing.

2.5 Dynamic Mechanical Analysis (DMA)

Dynamic mechanical analysis (DMA) of the prepared samples was performed using a DMA 242E NETZSCH Artemis dynamic mechanical analyzer located at the Department of Mechanical Engineering, Ahmadu Bello University, Zaria.

The DMA test was performed under controlled experimental conditions using a heating rate of 6 K/min and an oscillation amplitude of 60 µm. The temperature range employed during the analysis was 30°C to 120°C. Frequency-dependent measurements were conducted at a frequency of 1 Hz.

The key parameters measured by DMA are defined as follows. The storage modulus (E') represents the energy stored and recovered per cycle of deformation, and is given by:

$$E' = (\sigma_0/\epsilon_0) \cos \delta \quad (1)$$

where σ_0 is the stress amplitude, ϵ_0 is the strain amplitude, and δ is the phase angle between stress and strain. The loss modulus (E'') represents the energy dissipated as heat per cycle, and is expressed as:

$$E'' = (\sigma_0/\epsilon_0) \sin \delta \quad (2)$$

The damping factor, $\tan \delta$, is defined as the ratio of the loss modulus to the storage modulus:

$$\tan \delta = E''/E' \quad (3)$$

A high $\tan \delta$ value indicates a material that dissipates more energy relative to what it stores, reflecting dominant viscous behaviour, while a low $\tan \delta$ value indicates more elastic, stiffness-dominated behaviour.

Each blend composition was tested once under the specified frequency and heating rate conditions. Formal replicate testing (i.e., triplicate runs with reported means and standard deviations) was not conducted in this study due to material and equipment constraints; this limitation is discussed further in the Results and Discussion section.

3. RESULTS AND DISCUSSION

3.1 Dynamic Mechanical Analysis of LDPE/HDPE Blends

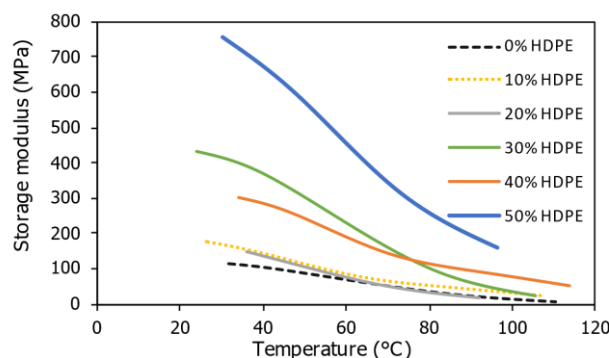


Figure 1.0: Effect of temperature on storage modulus of LDPE/HDPE blend

The variation of storage modulus with temperature for the LDPE/HDPE blends is presented in Figure 1. The results showed that the storage modulus decreased progressively with increasing temperature for all blend compositions due to increased molecular chain mobility within the polymer matrix.

At lower temperatures of approximately 35°C, the incorporation of HDPE significantly improved the stiffness of the blends. The sample containing 50 wt.% HDPE exhibited the highest storage modulus, while the control sample containing 0 wt.% HDPE showed the lowest storage modulus. The 50 wt.% HDPE blend demonstrated an increase of approximately 573% in storage modulus compared to the unfilled control sample.

The increase in storage modulus with HDPE loading can be attributed to the higher crystallinity and rigidity of HDPE compared to LDPE. Consequently, blends containing higher HDPE concentrations exhibited greater resistance to elastic deformation. This behaviour is consistent with findings demonstrating that HDPE forms well-organised spherulitic structures with high crystallinity (81.5%) and rigid amorphous layers between lamellae. In contrast, LDPE forms irregular aggregates with considerably lower crystallinity (27.5%) and mobile amorphous layers, which are structural differences that directly govern the stiffness of their blends (Fu et al., 2022). Furthermore, DMA measurements confirmed that the rigid, confined amorphous phase in HDPE suppresses molecular chain mobility, while the mobile amorphous phase in LDPE introduces greater viscoelastic compliance, explaining the progressive reduction in storage modulus observed as LDPE concentration increases across the measured temperature range of 30°C to 120°C in the present study (Fu et al., 2022).

The storage modulus decreased continuously as the temperature increased from 30°C to approximately 120°C. This reduction is associated with softening of the polymer matrix and decreased resistance to molecular motion at elevated temperatures. Similar observations have been reported for polyethylene-based polymer blends subjected to dynamic mechanical analysis.

It should be noted that due to material and equipment constraints, formal replicate testing with statistical error bars was not conducted in this study. This represents a limitation of the present work, and future studies should incorporate, at a minimum triplicate testing per composition with reported standard deviations to allow full assessment of data reproducibility and statistical significance.

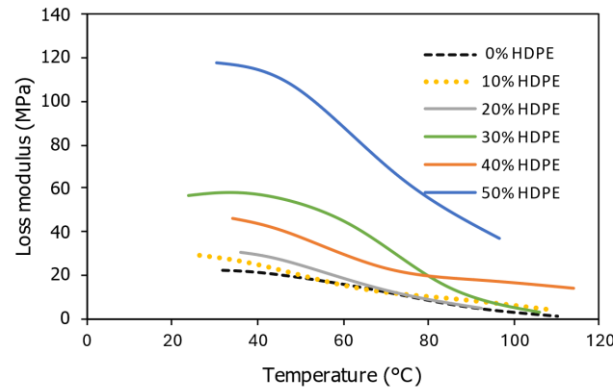


Figure 2.0: Effect of temperature on loss modulus of LDPE/HDPE blend

The loss modulus defines the viscous response of the material, reflecting its capacity to dissipate mechanical energy as heat during deformation, a behaviour governed by internal molecular friction and chain mobility within the polymer network (Satya & Sreekanth, 2020).

The results indicated that the loss modulus decreased with increasing temperature for all blend compositions. At approximately 35°C, the 50 wt.% HDPE sample exhibited the highest loss modulus, whereas the control sample without HDPE loading showed the lowest value. The increase in loss modulus for the 50 wt.% HDPE sample was approximately 426% higher than the control sample, with a modulus difference of about 96.4 MPa.

As the temperature increased, all samples exhibited gradual reductions in loss modulus and eventually converged near 110°C, except for the 50 wt.% HDPE blend, which maintained relatively higher values. This behaviour indicates thermal softening and reduced energy dissipation capability at elevated temperatures due to increased polymer chain mobility. Similar trends have been reported in thermoplastic polymer blends where elevated temperatures reduce viscoelastic resistance (Techawinyutham et al., 2021).

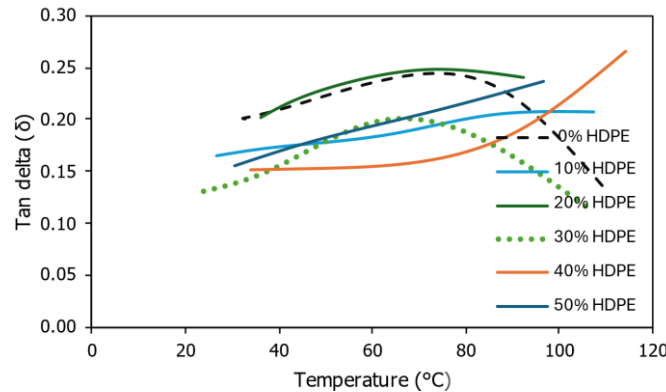


Figure 3.0: Effect of temperature on tan delta (δ) of LDPE/HDPE blend

Tan δ is the ratio of loss modulus (E'') to storage modulus (E'). It represents the damping factor, essentially how much mechanical energy is dissipated as heat versus stored elastically (Poh et al., 2022)

The tan δ curves showed varied behaviour across the HDPE Loadings with an increase in temperature. The polymer matrix for 0 wt.% HDPE blend exhibited a damping value of about 0.20 at room temperature, which constantly declined throughout the entire temperature range, reaching approximately 0.13 at 110 °C. This shows a progressive reduction in energy dissipation capacity with increasing temperature. The 20 wt.% and 30 wt.% HDPE blends displayed a characteristic rise and fall pattern, with their tan δ peak values of about 0.245 and 0.20 at around 70–75°C and 75–80°C, respectively, before declining sharply at higher temperatures. This peak behaviour is associated with the glass-to-rubber transition zone, where maximum molecular mobility and energy dissipation coincide.

In contrast, the 10 wt.% and 50 wt.% HDPE blends exhibited a gradual and continuous increase in damping factor with temperature, plateauing at elevated temperatures without a well-defined peak within the tested range. Most notably, the 40 wt.% HDPE blend demonstrated a distinctly different response, showing a continuous and steep rise in $\tan \delta$ from approximately 0.15 at room temperature to 0.27 at 120°C, with no observable peak or decline. This behaviour suggests that at this composition, the viscoelastic transition is either significantly broadened or shifted beyond the measurement window, likely due to a unique phase morphology and interactions between the HDPE crystalline domains and the matrix at this specific blend ratio (Sha et al., 1991). These divergent behaviours suggest that higher HDPE content fundamentally alters the damping characteristics of the blends, possibly due to increased crystalline phase constraints and enhanced polymer chain interactions that shift or suppress the peak damping temperature beyond the tested range (Munaro & Akcelrud, 2008).

3.2 Thermal Transition Behaviour

Table 1: Thermal transition temperatures of LDPE/HDPE blends at 1 Hz

LDPE (g)	HDPE (g)	wt.% HDPE	Onset (°C)	Peak (°C)	Offset (°C)
200	0	0	47.9	73.9	92.5
180	20	10	36.8	99.7	108.0
160	40	20	54.1	74.1	92.0
140	60	30	53.5	104.0	114.0
120	80	40	48.4	114.4	116.0
100	100	50	47.7	96.2	Increasing trend

The thermal transition temperatures of the LDPE/HDPE blends obtained from DMA analysis are presented in Table 1. The onset, peak, and offset temperatures varied with blend composition, indicating that HDPE loading influenced the relaxation behaviour and thermal stability of the polymer blends.

The 40 wt.% HDPE blends exhibited a higher peak transition temperature than the 50 wt.% blend, which may suggest a non-linear composition-property relationship. One possible explanation is that HDPE crystalline domains may be more favourably dispersed within the LDPE matrix at 40 wt.% loading, which could promote greater interfacial co-crystallisation and chain entanglement (Sha et al., 1991). Beyond this composition, a shift toward a phase-inversion-type morphology may disrupt the crystalline network, potentially reducing the effective thermal transition temperature despite the higher overall HDPE content. However, this interpretation remains speculative, as no direct morphological evidence (e.g., SEM) or complementary thermal analysis (e.g., DSC) was obtained in this study to confirm the proposed dispersion or phase-inversion mechanisms; such characterisation is recommended in future work to validate this hypothesis (Rana, 2002).

Lastly, the 50 wt.% HDPE composition shows an anomalous result in Table 1, where the recorded peak temperature is 96.2°C, and the offset showed an increasing trend. This is physically inconsistent, as the offset temperature should always exceed the peak temperature in a thermal transition. This discrepancy is likely attributable to instrument resolution limitations or data-processing artefacts at the boundary of the transition and should be interpreted with caution. The values for this composition should be revisited with additional testing, if available, or explicitly acknowledged as a measurement uncertainty.

4. CONCLUSION

The dynamic mechanical study of recycled LDPE/HDPE polymer blends indicated that compositional variation has a considerable impact on the material system's viscoelastic and

thermomechanical performance. The storage modulus gradually increased from 112.0 MPa at 0 wt.% HDPE to 754.2 MPa at 50 wt.% HDPE, indicating a significant improvement in stiffness brought about by the addition of HDPE to the LDPE matrix. This improvement suggests that because HDPE has a stronger crystallinity and less molecular chain mobility than LDPE, it encourages greater elastic energy storage capacity.

The loss modulus increased with higher HDPE loading, rising from 22.6 MPa to 119.0 MPa at approximately 31°C. This increase indicates an enhanced ability of the blends to dissipate mechanical energy through viscous deformation. As the temperature increased, all samples transitioned toward rubbery behaviour, which confirms the thermally activated relaxation of polymer chains and the gradual reduction in mechanical integrity at elevated temperatures.

The damping factor ($\tan \delta$) showed peak behaviour depending on the composition, with well-defined peaks observed for the 20 wt.% and 30 wt.% HDPE blends ($\tan \delta \approx 0.245$ at 70–75°C and ≈ 0.20 at 75–80°C, respectively), while the 0, 10, 40, and 50 wt.% HDPE blends did not exhibit a clearly defined peak within the tested temperature range. The presence of separate $\tan \delta$ peaks demonstrates the viscoelastic nature of the blends and indicates the effect of the interfacial interactions between the LDPE and HDPE phases on the energy dissipation characteristics. The results overall show that LDPE blending with HDPE is a feasible way to achieve mechanical stiffness and damping performance through controlled compositions.

This study contributes to the existing knowledge by establishing a quantitative correlation between the content of HDPE and the dynamic mechanical performance in recycled polyethylene systems. It further demonstrates that waste polyethylene streams can be engineered into value-added materials with tunable mechanical and damping properties instead of being limited to disposal or low-grade recycling applications. The effect of compatibilizers on phase interaction and interfacial adhesion in LDPE/HDPE blends should be studied in future work. Furthermore, morphological studies using SEM and thermal analysis techniques like DSC and TGA are suggested to correlate microstructural changes with the observed viscoelastic behaviour. Tests of long-term fatigue and cyclic loading would also give more insight into the durability and real-world applicability of these recycled polymer systems.

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