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Physical, Static and Dynamic Mechanical Properties of Waste Paper Reinforced Waste High Density Polyethylene Biocomposite

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Abstract: *This paper presents the physical, static, and dynamic mechanical properties of a biocomposite fabricated from wastepaper reinforced in waste high-density polyethylene. The produced composites had varying amounts of shredded wastepaper from 0 to 50 wt% at an interval of 10wt%. The size reduced paper was mixed with the waste high-density polyethylene in a two-roll mill set at 160 ^oC and 79 rev/min. The mixture was then compressed to 4 MPa at 150 ^oC and allowed to cure at 60 ^oC for 24 hrs. The results obtained indicated that water absorption increased with filler content due the hydrophilic nature of natural fibers, tensile stress and strain however reduced. Modulus of elasticity recorded the highest value at 40 wt% wastepaper in the composite. Dynamic mechanical analysis revealed that at 40 ^oC, the 40 wt% recorded the highest storage modulus, greater than unreinforced material by 40%. Higher filler content recorded increase in damping parameter of the materials. Increasing filler content also introduced a new glass transition behavior. The new glass transition (α) Tg was detected between 120 ^oC and 145 ^oC. Although elongation increased with temperature, it decreased with filler content. These properties contribute to establishing concept of waste reuse and recycling as a viable technique in sustainable engineering.*

Key Words: *Waste reuse, paper, high density polyethylene, composite, Dynamic mechanical properties, glass transition.*

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1.0 Introduction

The use of natural fibers or filers (or combination of both) in biocomposite application is gaining wider application and is currently receiving significant research interest especially in recent time. This is due to increasing output of plastic waste to the environment and the subsequent environmental threat (Iwata, 2015; Khan *et al*., 2017; Nasrollahzadeh *et al*., 2019). In 2015, it was reported that about 4900 Mt (equivalent to 60%) of all plastics ever produced are found in today's natural environment (Geyer *et al*., 2017). Plastics are not easily attacked by microorganism and degradation by other means such as incineration can lead to the release of one of the most toxic air pollutants (Kumar *et al*., 2020). Therefore, the present of plastic in the environment constitute a major burden to the ecosystem. They clog water drains, release poisonous gases when burned in open airs and disrupt land fertility when they get to farmlands (Kumar *et al*., 2020; Oosterhuis *et al*., 2014). Studies has revealed that plastic can influence climatic changes through their barrier to free evaporation and the release of gases that can interfere with the composition of the atmosphere (Fiore *et al*., 2015; Mohammed *et al*., 2015).

In view of their potential impact on the environment, several measures have been investigated on ways of reducing environmental menace caused by plastics. Some of the attempts are resource recovery, recycling and reuse (Ashton *et al*., 2016). However, studies reveal that due to their inherent content, some of these measures have not completely free their contamination potentials. Recent efforts by several regional and international Environmental protection agencies towards the reduction of greenhouse emissions have inspired polymer engineers and scientists to source for options in

natural materials (Lau *et al*., 2020). Natural materials can be incorporated in plastics to form biocomposites (Asasutjarit *et al*., 2007). Natural sources currently being studied in composites applications include wood, rice husk, sisal, luffa, paper, tamarind, palm trees etc (Ghori *et al*., 2018; Luchese *et al*., 2018). Natural fillers are environmentally friendly, low cost and are abundant in nature. They also show high stiffness, comparable specific strength, easy to process, renewable and biodegradable (Djafari Petroudy, 2017; Reale Batista *et al*., 2020; Tominaga *et al*., 2019).

Natural materials in plastic composites however have drawbacks, they are hydrophilic in nature and incompatible with hydrophobic matrices (plastics) (Herrera-Franco and Valadez-Gonzalez, 2005; Jawaid and Abdul Khalil, 2011; Stark and Rowlands, 2003). This leads to poor interfacial adhesion between the natural fiber and the plastic matrix (Govinda and Srinivasan, 2016; Rashid *et al*., 2011; Saba *et al*., 2015). The poor transfer of load from the matrix to the fiber interface can leads to low impact strength and low thermal stability of the resulting composites (Faisal and Salmah, 2012). These problems however have been confronted by fiber/filler modifications (Edeerozey *et al*., 2007; Kabir *et al*., 2012; Senthilkumar *et al*., 2019). Fiber or fiber surface modifications can provide wider applications of composites (Chun *et al*., 2015; Sahu and Gupta *et al*., 2018; Krishna and Kanny, 2016). Bio composites have found application in packaging, decorations, medicine, buildings and construction (Faisal and Salmah, 2012; Ge *et al*., 2019; Patil *et al*., 2019). They are also being tailored in automobiles, military and defense industries (Moliner *et al*., 2017; Pickering *et al*., 2016; Sanjay *et al*., 2017). Appropriate tests (such as mechanical, thermal and biodegradability) can provide information on possible applications of most fabricated composites (Leites *et al*., 2018; Sessini *et al*., 2019; Verma and Fortunati, 2019). Some materials possess excellent mechanical properties at room temperature but fail at higher temperatures. These problems have necessitated the shift from static mechanical tests to dynamic mechanical analysis (DMA). DMA analyses material's properties over a wider temperature range. It provides data on the glass transition temperatures (T_g) and damping properties, which are useful factors towards better understanding and improvement of operational conditions.

In view of the necessity of designing, testing and application of current technologies towards reducing environmental risk of plastics in the environment, this paper is aimed at using some waste material (such as polyethylene) to form useful composites, by incorporating waste papers as filler. This paper shows how waste materials such polyethylene considered as environmental nuisance can be taken advantage of to form composites. Waste paper serves as the filler. The Dynamic mechanical analysis coupled with physical and mechanical tests will help us to proffer a sustainable waste solution to waste papers and plastics in our environment.

2.0 Material and Methods

Waste paper was collected from used paper dumped site, within the premises of Ahmadu Bello University, Nigeria. The collected papers were-milled and sieved with Thomas Wiley, Laboratory Mill fitted with a 2 mm screen. Waste plastics were also collected from dumpsites in Zaria from which high-density polyethylene (HDPE) was sorted out using plastic identification code (2). They were thoroughly washed with deionized and double distilled water, dried and cut into smaller pieces of about 2 x 2 cm. The HDPE was then transferred into a two-roll mill (manufactured by reliable rubber and plastic machinery company, North Bergen, USA), operated at 160 ^{o}C and 79 rev/min. After 10 minutes, the milled paper was added. Silicon oil was also added at intervals into the mix to improve compatibility of the constituents. The composite was then transferred into the Carver compression machine (USA) hot press operated at 150 ^{o}C and 4 MPa. Reproducible tests of three (3) were carried out to ensured reliable and reproducible experiments. The same method was used to produce varying loading of fillers (10 wt% - 50 wt%) as shown in Table 1. Samples were then taken for physical, static, and DMA tests.

This was carried out according to ASTM D570 recommended method. The sample was cut to 50 mm diameter disc with 3 mm thickness, placed in water at 25 ^{o}C for 24 hrs. It was then removed dried and

weighed. Each test was repeated three times. Percentage water absorption was calculated using equation (1),

$$Water\ absorption\ (\%) = \frac{(Wet\ weight - dry\ weight)}{Dry\ weight} \times \frac{100}{1} \quad (1)$$

Table 1: Composition of Waste paper in HDPE Matrix.

Samples	Waste paper (%)
A	0
B	10
C	20
D	30
E	40
F	50

2.1 Water absorption

2.2 Tensile test

This was carried out according to the method recommended by ASTM D638. It was conducted at 25 ^{o}C and 50 % relative humidity. The tensometer was operated at cross-head speed of 50 mm/minute while the samples were respectively pressed cut to 40 x 10 x 3 mm^3 dimension, each. The samples were afterward placed horizontally at the grips of the tensometer, which was tightened and kept at gauge length of 45 mm. The increase in resistance was recorded by the load cell. The load value was recorded until the sample ruptured. Each test was repeated three times. The Load vs extension plot generated was converted to stress-strain curve and properties such as tensile strength (MPa), tensile strain (m/m), and modulus of elasticity (MPa) were evaluated.

2.3 *Dynamic mechanical analysis*

Test specimens of dimension 40 x 10 x 3 mm was cut according to ASTM D4065 recommendation in a 3-point bending deformation mode. It was operated from 34 ^{o}C to 150 ^{o}C at 5K/min. NETZSCH Proteus thermal analysis software was used to analyze and export the storage modulus (MPa), loss modulus (MPa), tan d (dimensionless) and elongation data (µm).

4.0 Results and Discussions

Fig. 1 shows plots for the trend observed for the water absorption capacity (%) of the produced composites. Water absorption increase with filler loading. This is because natural fillers consist of cellulose which contains hydroxyl bonds. The hydroxyl bonds react with water molecule to from hydrogen bonds, hence exhibiting hydrophilicity (Diallo *et al*., 2019; Faisal and Salmah, 2012; Gupta and Singh, 2018). The water absorption capacity of the unreinforced composite (sample A) was 1.70%. It increased to 6.52 % in the 50 wt% filler reinforced composite which is equivalent to 284% increase. High water absorption lowers mechanical property and dimensional stability. It however, favors the biodegradability of the composite (Faisal and Salmah, 2012).

When natural fillers are modified, the mechanical properties of their composites can be significantly improved (Ahmad *et al*., 2018; Lu and Oza, 2013). Figs. 2 to 5 show histograms for the respective mechanical properties of the composite formed. The tensile stress (Fig. 2) displayed a continuous drop as filler content increased. The tensile stress of unreinforced HDPE recorded 16.73 N/mm^2 while that of the 50 wt% reinforced HDPE recorded 8.89 N/mm^2. This value is 47 % of unreinforced HDPE. The tensile strain property (Fig. 3) was observed to decrease as filler content increases. This drop is 75% from 0.275 (unreinforced) to 0.068 (50 wt% reinforced) respectively. The Modulus of elasticity (MOE) shown in Fig. 4, increased from 60.81 N/mm^2 (unreinforced) to 168.68 N/mm^2 (40 wt% reinforcement). That equals a 177% increase before dropping by 17% to 140.21 N/mm^2 (50 wt%).

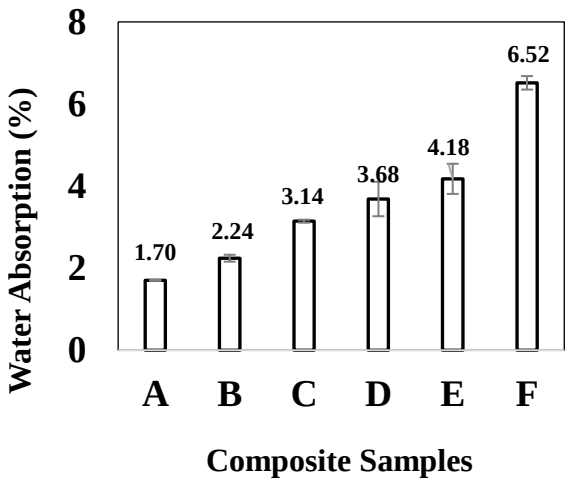

Fig.1:Effect of fiber loading on Water absorption capacity of the composites

The increase in mechanical property is due to ~~the~~ increased lignocellulose content of the fillers (Asasutjarit *et al*., 2007; Faruk and Sain, 2014). The drop between 40 wt% to 50 wt% may be due to fiber-fiber interaction caused by fiber overloading. Fiber overloading causes weak adhesion bond between constituents. This leads to lower property reliability(Leão *et al*., 2015; Santosha *et al*., 2018; Sarasini and Santulli, 2013).

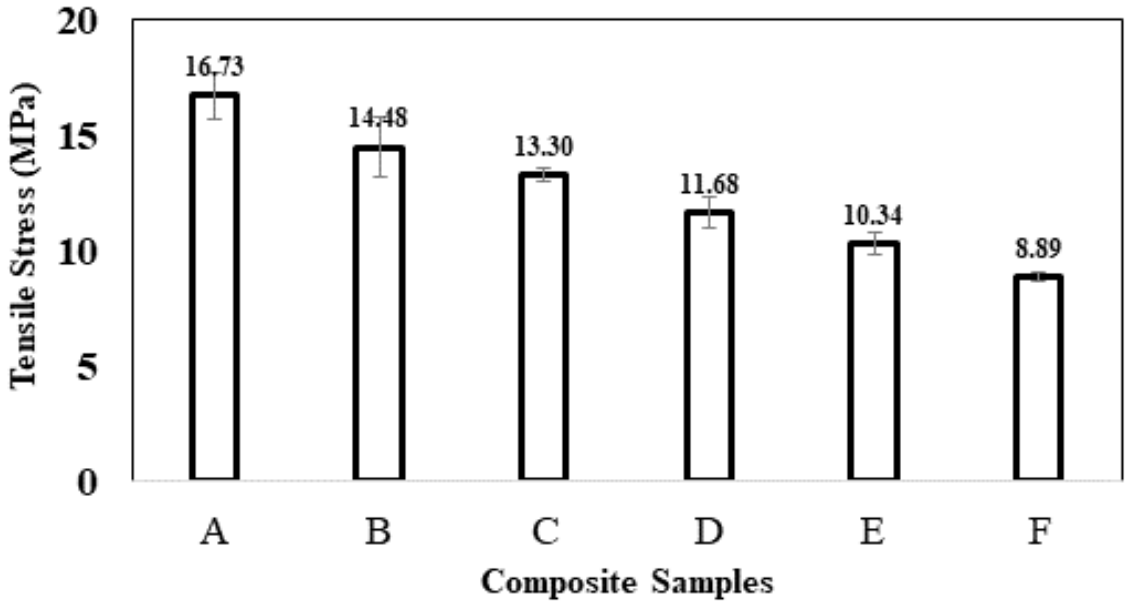

Fig. 2: Effect of fiber loading on Tensile Stress of the composites

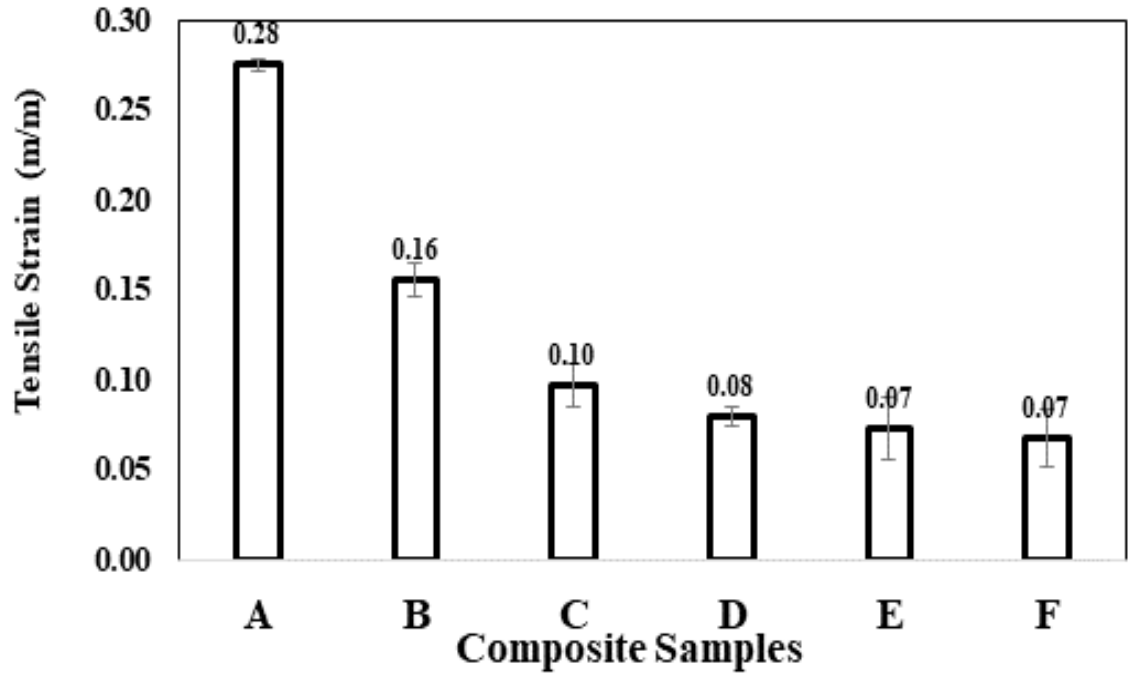

Fig..3:'Effect of fiber loading on Tensile Strain of the composites

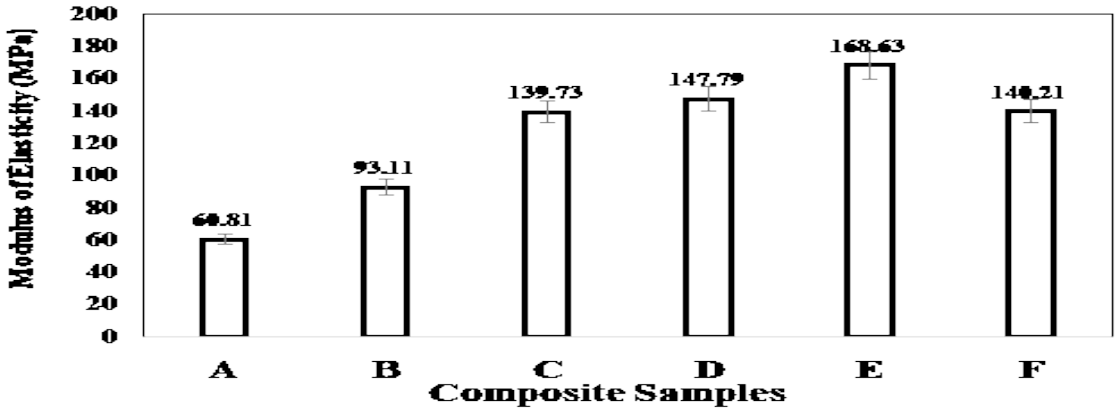

Fig. 4: Effect of fiber loading on Modulus of Elasticity the composites

Figs. 5 to 8 presents the dynamic mechanical analysis of the produced composites. Storage modulus as shown in Fig. 5 increased as the fiber content increases except for 10 wt%. The increase may be attributed to enhancement in physical crosslink bond created by fiber loading. This causes interfacial stress transfer leading to increased stiffness of the composite (Saba *et al*., 2016; Siakeng *et al*., 2019; Adam *et al*., 2008). The 10 wt% recorded lower storage modulus than the unreinforced waste HDPE. Low filler content can cause slippage of the methylene groups of the HDPE chains. The consequent pseudo-lubricating effect accounts for the behavior of 10wt% (Ahmad *et al*., 2018). At 40 ºC, the storage modulus of the unreinforced sample was 1700 MPa while that of 40 wt% reinforced material recorded 2500 MPa which indicates a difference of 800 MPa. This implies that 40 wt% reinforcement yielded a 47% storage modulus improvement and points toward better effectiveness and attractiveness of the filler. Increase in temperature was observed to initiate corresponding decrease in storage modulus of all the produced composite probably due to side-chain motion that has the tendency to facilitate the composites to lose it closed packed arrangements (Asim *et al*., 2015; Hameed *et al*., 2007; Jawaid and Abdul Khalil, 2011). Such changes have been reported to lead to decrease in viscosity (Sever *et al*., 2018). Also, filler content can favour increase in storage modulus. Sample E (40 wt%) recorded the highest value of storage and loss modulus (Fig. 6). The difference in storage modulus for the unreinforced composite and the 40 wt% reinforced composites at 40 ºC was 800 MPa (47% of the unreinforced composite) but with increasing temperature (up to 146 ºC), the value reduced to 100 MPa.

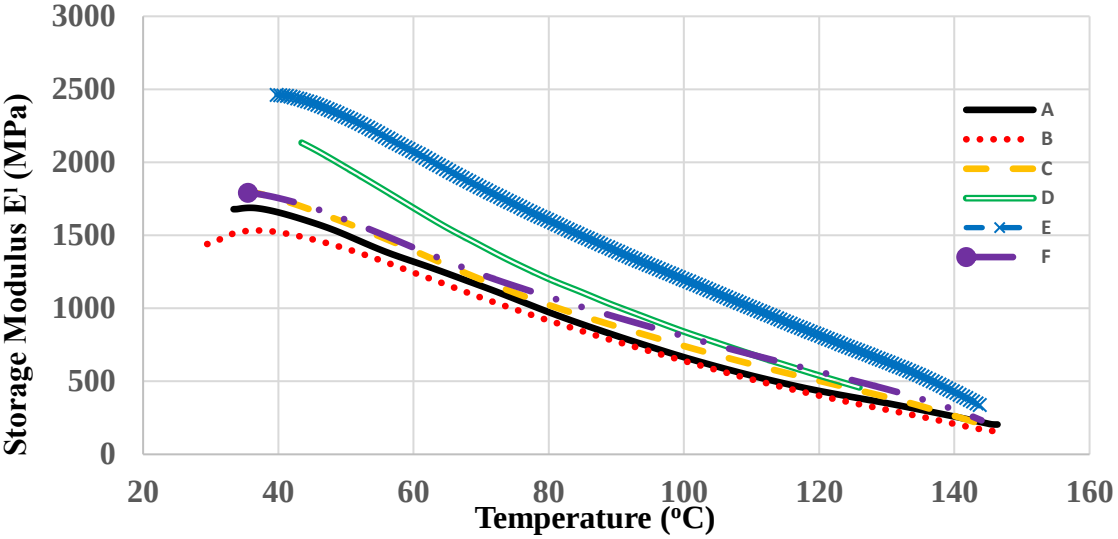

Fig. 5: Storage Modulus of paper reinforced High density Polyethylene

The loss modulus (Fig. 6) was observed to exhibit behaviour similar to that of storage modulus. The peaks tend to be broader as filler content increased. This is because fillers are responsible for energy absorption properties (Han *et al*., 2008; Pothan *et al*., 2003). Loss modulus peaks are associated with the glass transition temperatures (T_g) implying the composite samples recorded slight shift in T_g as the filler content increased(Han *et al*., 2008).

The slight shift in T_g is significantly evident as shown in the tan delta (δ) plots of Figure 7. Below 85 ^oC, the values of Tan delta decreased with filler loadings (Ahmad *et al*., 2018; B. Rashid *et al*., 2017; Sewda and Maiti, 2013). An outlier in 50 wt % recorded the highest tan δ. This increase is due to stiffness property introduced into the composite. Above 85 ^oC, the damping parameter increased with filler content to about 120 ^oC. The damping property improvement may be due to the improved filler interphase with the HDPE matrix leading to enhanced viscoelastic properties of the resulting composites(Abdo *et al*., 2019; Ilya *et al*., 2020; Zhao *et al*., 2020). and their interphase with the matrix. 50 wt% recorded the highest value until 145 ^oC when samples failed. High storage modulus and higher damping materials find applications in aerospace and automobiles industries (Harshavardhan *et al*., 2019; Pothan *et al*., 2003; Saba *et al*., 2016). The unreinforced waste HDPE recorded a Tg at 80 ^oC. HDPE exhibits gamma (γ) Tg between -140 ^oC and -100 ^oC, and beta (β) Tg between 0 ^oC – 65 ^oC. The Tg at 75 ^oC recorded by the waste HDPE matches beta Tg. The shift is due to the physical degradation of the plastic. This behavior had been previously reported in recycled materials (Alemdar and Sain, 2008; Tominaga *et al*., 2019). Increasing filler content led to the composite displaying alpha (α) Tg. This characteristic peak occurred at 120 ^oC -145 ^oC. This suggests bio-fillers causes segmental motions (Alemdar and Sain, 2008).

Elongation was found to increase with temperature but decreased with increase in the filler content of (except for 40 wt%) at temperature below the beta (β) Tg. This supports the tensile strain behavior (Figure 2) that the filler introduced a brittle property to the material.

Fig. 8 shows that the elongation at break of the studied thermoplastics decreased with increase in the natural filler content which is in agreement with literature (Chun *et al*., 2015; Salmah *et al*., 2013). Generally, increase in temperature led to a corresponding increase in elongation especially for those with filter content percentage weight of 10, 20 and 30 respectively. The observed behavior explains why the 10 wt% reinforcement composited had the lowest elongation after Tg while the 30 wt% which had the lowest Tg before Tg, had the highest. Such replacement of ranks with respect to elongation has been attributed to segmental motions originating from bio-fillers (Alemdar and Sain, 2008). Therefore, waste paper can help in tailoring properties of thermoplastic composites. The properties which may be physical, mechanical or thermal will be at low cost and in an environment friendly manner.

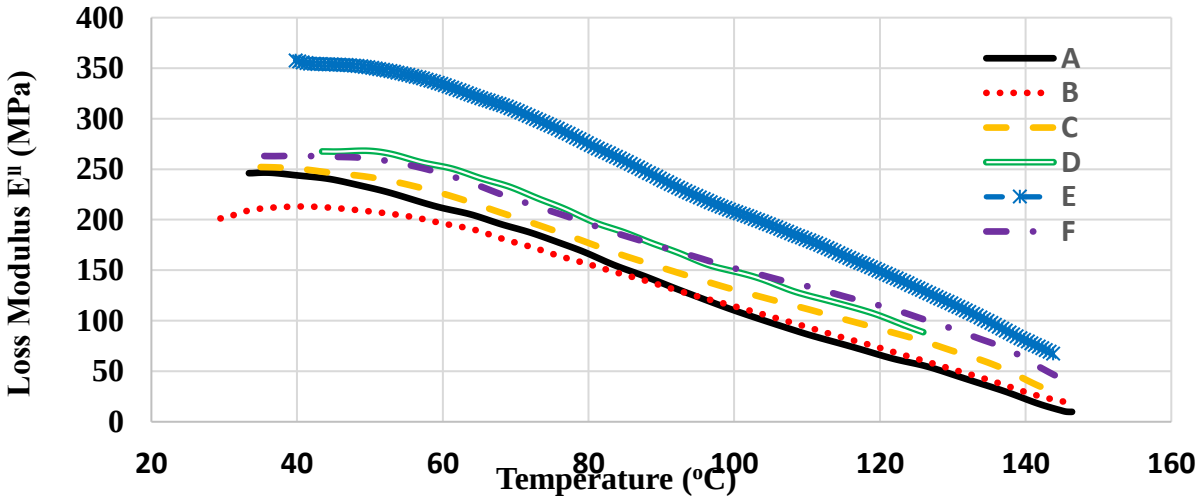

Fig. 6: Variation of loss modulus of paper reinforced high density polyethylene with temperature

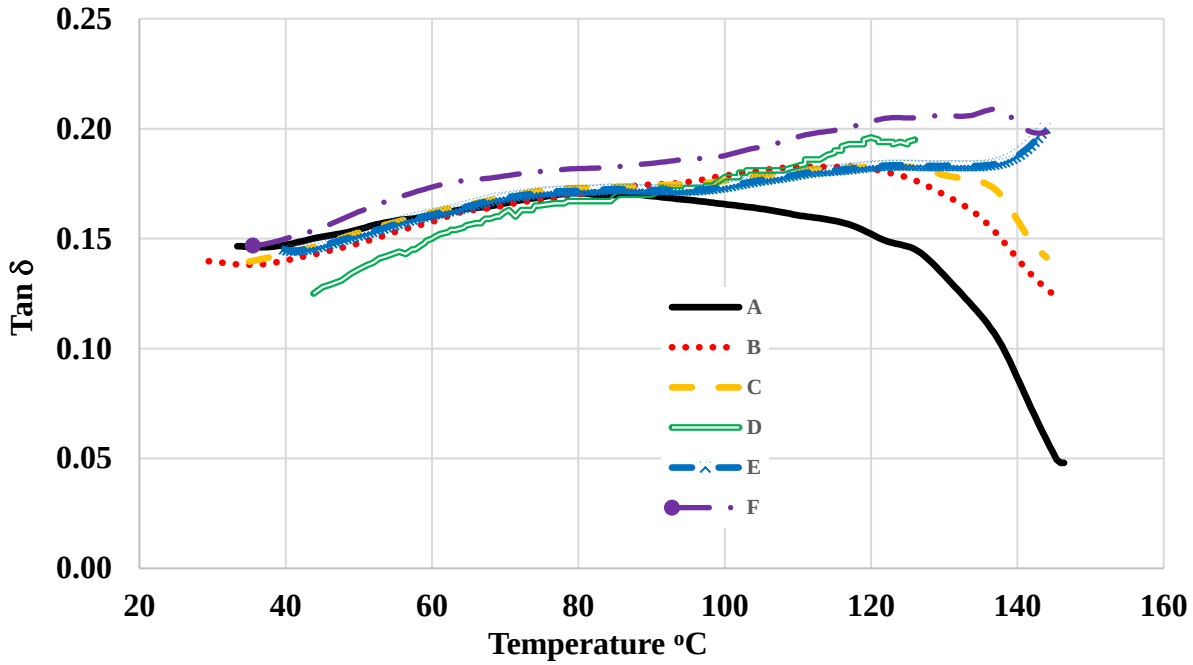

Fig. 7: Variation of damping parameter (tan δ) of paper reinforced high density polyethylene with temperature

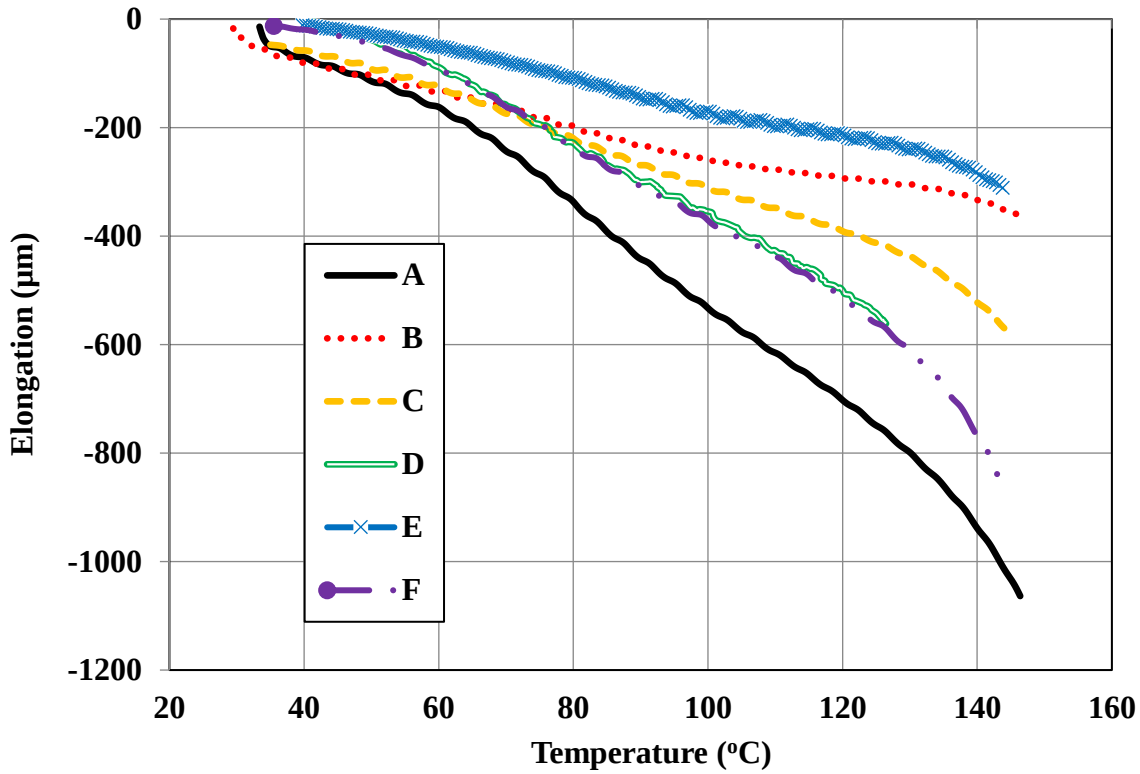

Fig. 8: Elongation of paper reinforced high density polyethylene

4.0 Conclusions

Waste paper (up to 50 wt%) increased water absorption by 284% while tensile stress and tensile strain dropped by 47% and 75% respectively. Modulus of elasticity improved by 177% at 40 wt%. Paper filled composite contributed an average 43.5

% increase in storage modulus over temperature range of 40 - 140 ^{o}C. Paper filler tend to slightly increase the damping behavior above the β glass transition and a new glass transition (α) between 120 ^{o}C and 145 ^{o}C. Paper shows a good prospect for increasing glass transition temperature and damping property of high density polyethylene. This also provides a green and cost effect way of tailoring the viscoelastic properties of plastic materials.

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Conflict of interest

The authors declared no conflict of interest