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Effect of Hybrid Filler Loading on Some Physicomechanical Properties of Waste Polypropylene

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Abstract

This research work studied the potential of utilizing waste Polypropylene (wPP), Date Seeds Particles (DSP), and Clay Particles (CP) to fabricate a hybrid composite material of different hybrid filler loading combination. Compounding and Compression molding techniques where used to fabricate twelve samples with matrix/filler loading ratio of 90:10 that is 90 percent matrix (wPP) and 10 percent hybrid fillers (DSP/C) to give a 100 percent hybrid composite material except for the control sample which is 100 percent matrix. The 10 percent loading was distributed between DSP and CP by varying the loading from 0/10 DSP/CP and 10/0 DSP/CP that is as DSP is increasing the CP is decreasing in the first case and opposite in the second case. The research work also investigated some physical and mechanical and properties of the hybrid composites. For Physical properties, density and water absorption test were carried out, the result have shown that most of the composite have higher density than the control with 0.69g/cm^3 except the last two composite with 0.68 and 0.67g/cm^3. The highest density value was observed on the composite of 0DSP/10CP with 1.28g/cm^3 as the value continue to decrease progressively with increase in the DSP and decrease in CP. For water absorption capacity, the control (0DSP/0CP) and composite of 0DSP/10CP have shown 0.0% water absorption capacity. Within the composite, the water absorption is increasing progressively with increase in DSP and decreasing the CP as the composite of 10DSP/0CP absorbed much water with 0.54%. For tensile strength and elongation at break, the control sample displayed a higher result over composites with 25.9Mpa, 45.2% respectively. While for the tensile modulus, flexural modulus, and hardness, the composite of 10DSP/0CP displayed a highest result of 1.8Gpa and 89Hv respectively and the values are increasing with increase in DSP and decreasing the CP. The study validates that the balanced filler combination (such as 4DSP/6CP or 6DSP/4CP) can provide a synergic effect, improving both mechanical and physical properties.

Keywords: Polypropylene, Composite, Filler, Date seeds, and Clay.

INTRODUCTION

Composite materials have had a significant impact on the technology of design and construction. The commercial and industrial applications of composites are varied and widespread. The material applications include; transportation and infrastructure, sport, Aerospace, automotive industry, environmental engineering, and electrical distribution. The composites have attractive properties which make materials to replace the conventional ones. Composite material of natural fiber is being much used as they show good properties such as cost effective and ease of fabrication (Purnesh *et. al.*, 2022). The materials are made by mixing two dissimilar components together, physically or chemically to form composite (Santhanam,

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et. al. 2021). In Composite, there is continuous component known as matrix and there is a discrete or discontinuous component called filler. The matrix maybe a polymer, metal or ceramic, whiles the filler maybe a fiber or particles. Matrix and the filler come together to act as one called composite. Filler is the load bearing component of the composites while the matrix binds the filler together, which is the reinforcing material (Yuan *et. al*., 2018).

There are different types of composite. This include: ceramics matrix composite (CMC), Polymer matrix composite (PMC) and the metal matrix composites (MMC) (Jayaraman, 2013). Polymer matrix composites are of great interest around the world today with notable advantages that include lightweight, high strength, and ease of fabrication. (La-Mantia and Morreale 2011). Polymer composite have been reported to be in use for numerous years with a market share reported to have increased tremendously in the last decades and this has been attributed to the introduction of environmentally friendly natural additives (fibers) (Owonubi, *et. al.,* 2018). Date seeds (DSP) and clay particles (CP) are environmentally friendly natural additives and the used of both to produce a hybrid composite material is a good Idea. Hybrid composite use more than one kind of reinforcement material in a particular matrix, Its a material that consists of two or more type of filler embedded in a single matrix (Lin *et. al.*, 2019). Henceforth, the idea is that the hybrid composite material to have the synergic effect of the individual properties of the reinforcing materials (fillers). Polymeric hybrid composite material offers the designer to obtain the required characteristics in a controlled considerable extent by the choice of suitable fillers.The mechanical characteristics of a hybrid composite mainly depend upon the fiber content, length of individual fibers, filler orientation, and fille to matrix bonding and configuration of the filler. (EI-Wazery, 2017). Therefore, hybrid composite has currently received great attention from researchers due to their excellent potential when compared to the non-hybrid composite material.

The study of polymer nanocomposites has been focused on enhancing the conventional properties of the polymeric material, these properties changes drastically using a small fraction of nanofiller (Ciardelli et al., 2008). Kawasumi *et al.* (2011) reported a novel approach to prepare PP nanocomposite using a functional polyolefin oligomer (e.g. maleic anhydride grafted PP oligomer) as a compatibilizer between the matrix and filler, this approach has been well developed for polypropylene-based systems, the focus of most industrial research and development has been on formation of nanocomposites by melt-mixing or compounding processes because this is generally more attractive than in situ polymerization due to better commercial feasibility and lower cost.

Jamaludin N. *et al*., 2015) revealed that, the effect of maleic anhydride grafted styrene-ethylene-butylene-styrene (SEBS-g-MA) content on mechanical, thermal, and morphological properties of polyethylene terephthalate/polycarbonate/halloysite nanotubes (PET/PC/HNTs) nanocomposites and the nanocomposites of PET/PC (70:30) with 2 phr of HNTs compounded using the counter rotating twin screw extruder after some series of formulations were carried, they prepared composite by adding 5-20 phr SEBS-g-MA to the material. The Incorporation of 5 phr styrene/ethylene butadiene styrene grafted mialic anhydrate (SEBS-g-MA) into the nanocomposites resulted in the highest tensile and flexural strength. Maximum improvement in the impact strength which is 245% was achieved at 10 phr SEBS-g-MA content. The elongation at break increased proportionately with the SEBS-g-MA content. However, the tensile and flexural moduli decreased with increasing SEBS-g-MA content. Scanning electron microscopy revealed a transition from a brittle fracture to ductile fracture morphology with increasing amount of SEBS-g-MA.

Polypropylene (PP) is one of the commodity plastic produced in a very large scale and used in various applications. The material has many advantages, including excellent price/performance profile and versatility. It provides sufficient stiffness, strength, deformability, and process ability at a very reasonable price (Pregi *et. al*., 2022). Furthermore, PP can be modified to extend its properties; stiffness can increase with addition of fillers and hence increase its functional properties such as flammability and conductivity (Tolinski, 2016). Bio-degradability is one of the main drawbacks of PP because they are synthetic and produced from fossil fuels and their carbon footprint is highly significant. Recently, this issue has been compensated by using natural available additives to produce polymeric composite material that is cheaper and bio-degradable.

METHODOLOGY

Collected Date Seeds (DS) were washed to remove dirt and allowed to dry. The dried date seeds were then crushed using a jaw crusher and then sieved to 100um particle size. The same size was sieved for the clay particles also using standard sieved shaker. Waste polypropylene containers were washed and dried under sunlight before cutting to flakes. Eleven hybrid composites were prepared using 90% matrix (wPP) and various proportions of the fillers (DSP/CP) as presented in table 1 below. The control used was 100%wPP

Table 1.0 Composition of various Hybrid Composites

S/N	MATRIX WT%	HYBRID FILLERS WT%	
	PP	Date seed particles	Clay
1	100	00	00
2	90	10	00
3	90	09	01
4	90	08	02
5	90	07	03
6	90	06	04
7	90	05	05
8	90	04	06
9	90	03	07
10	90	02	08
11	90	01	09
12	90	00	10

Preparation of the Hybrid Composites

The hybrid composite fabrication was achieved using Compounding and Compression molding method as adopted by Nuhu, (2023). In this method, the flakes form of waste Polypropylene, date seeds, and clay particles were compounded using a two roll mill machine. The formulated mixture of PP/DS/CPs in wt.% were compounded in accordance to the Table 1.0 by first, introducing the flakes polypropylene to the two roll mill machine rolling at 25 RPM and 160^{o}C. The DSP and the CPs w% were added gently to the PP beyond its melting point. The mixture was then allowed to continuing rolling with proper monitoring to achieve a homogeneous mixture of the fillers and the matrix for five minutes.

The resultant mixture was then collected into a metal mold size 150 × 120 × 5 (mm) length, breath and thickness respectively. The mold containing the melted mixture was then compressed using compression molding machine at 160^{o}C for five minutes. Thereafter, it was the pressed at room temperature for three minutes and pressure of 4Pa to obtained a hybrid composite material with the shape of the mold. The achieved solidified composite sheet was then removedfrom the mold for finishing and cutting. The same process was applied to the remaining samples before characterization. This fabrication was conducted in the polymer processing laboratory of the Department of Polymer Technology, Nigerian Institute of Leather Science and Technology Zaria

Determination of density.
Measuring the density of a material is very important because the density measurement will help determine the characteristics of the material, for example, whether the material will float or sink.

Knowing the density of the material helps in determining the mass and volume of a material, all of which are extremely important measurement in industries.
The mass of each sample was determined using an analytical weighing balance, and the volume also determined using a displacement method. In this method water was added into a measuring cylinder and the level was recorded as initial volume (v_1) and then carefully the sample was placed into the cylinder and the new volume was recorded as final volume (v_2). The volume of the sample was calculated between the final and the initial volume, the difference of the water level was recorded as the volume of the sample, and its density were computed as the ratio of the mass to volume **$\sigma = m/v$ (g/m^3).**

Determination of water absorption capacity.
The water absorption data is important to understand the performance of the polymeric materials during processing, for example, to avoid premature moisture and failure in a humid environment.

Water absorption test was carried out according to the ASTM D570. The sample was positioned in an oven at 45 ^oC for 72 hours. After then, it was placed in a desiccator for 24 hours and finally weighted and recorded as (w_1) using weighing balance. The sample was then placed in water inside plastic container for 24 hours. The sample was then removed and dried using tissue paper and the resultant weight was then measured and recorded as (w_2). This procedure was repeated for 20 days, on the samples and the percent water absorption was calculated and recorded.

Mechanical test
A universal testing machine was used to calculate the tensile parameters (tensile strength, elongation at break, and tensile modulus). In this method the sample was held in the grips of the machine and tightened firmly to prevent any slippage as the test commenced. The resistance and elongation at break of the specimens were detected and recorded by load cell until a failure or rapture occurred. This procedure was repeated on each fabricated sample.
Tensile strength formula used was **$s = P / a$** where, **s** is the tensile strength **P** is the force required to break and **a** is the cross-sectional area.
Tensile modulus, the formula is **$E=\sigma/\varepsilon$** where, **E**=Young's, **σ** = uniaxial stress, and **ε**=strain or proportional deformation.
Elongation at break = (Final length-Original length) / Original length × 100%

Hardness test
The hardnessof the sample wasmeasured using VHT (Vinkers Hardness Tester) MV1-PC, Mh-Cm with a maximum load capacity of 0.3Kgf (100HV). The specimen dimension was 30×10×5, and the testing process was carried out on the surface of the sample byplacing it on the sample compartment focusing the indenter. The 0.3kgf load was applied on the material and the indentation on the sample was carefully observed through the lens of the machine, and the calculation was done using **$HR=N-(d/D)$** where HR is the Rockwell hardness value, N is the

load applied (in kgf), d is the depth of the indentation (in mm), and D is the diameter of the ball or the width of the diameter core (in mm).

RESULS AND DISCUSSION

Physical Properties

Density

The results obtained during this research work on the density of wPP / DSP/ CP hybrid composites are shown in Figure 1. From the graph obtained, the result shown the impact of the hybrid fillers loading in comparison with control as the density is decreasing by adding the DSP and decrease in CP. This behavior was due to the introduction of fillers into the matrix which may cause air to be trapped in the material causing micro voids in the composites along the individual fillers due to the fiber spacing and between the composite, which has adverse effect on the properties of composites hence, reduce the density of the material. Natural filler such as date seed powder has lower density of 0.68 g/cm3 than clay particles with 1.41 g/cm3 and polypropylene with 0.92 g/cm3 which explained why as the filler content increases the density of the composites decreased. These findings are in line with the results obtained by Danladi and Shu'aibu, (2014). The highest density value was obtained with 0 % DSP / 10 % CP as 1.21 g/cm^3 while the lowest at 10%DSP / 0% CP with 0.74 g/cm^3.

Water Absorption

The effects of filler loading on water absorption capacity of the hybrid composites are showed in Figure 2. The absorption of water is an important parameter while working with cellulose-based fillers, because high humidity worsens the mechanical properties of the material and change the dimensions. The effect of water absorption is an important consideration where the material has been designed for applications in contact with water. From the results the ability of the composites to absorb water after exposure at ambient temperature was studied during twenty (20) days. The results revealed that percentage (%) water absorption has no effect on the control, and the 0.0DSP/10CP sample. But for the remaining samples, the percent water absorption is increased with increase in immersion time. This increase is due to the water ability to saturate the surface of the material and diffused into the core of the material with the help of non-fully embedded hydrophilic filler.This is in conformity with results of similar works (Danladi and Tunde, 2013). This property is due an increase in the amount of hydrophilic filler in the composites. This is attributed to hydrophilicnature of the date seed particles (DSP) capable of absorbing water, this is to say that the higher the filler loading the higher the water absorption and similar trend was observed by Shehu *et. al.*, (2019).

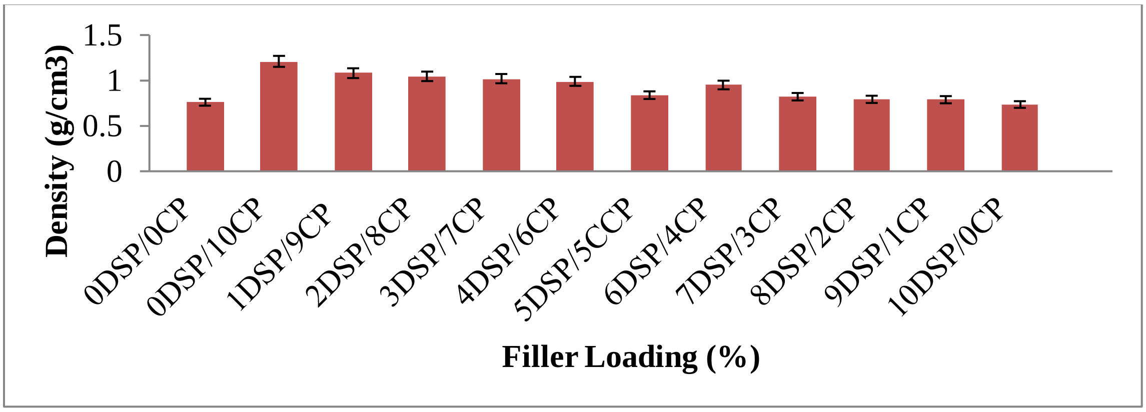

Figure: 1 Graphical presentation of density versus filler loading weight (%) for the Hybrid Composites

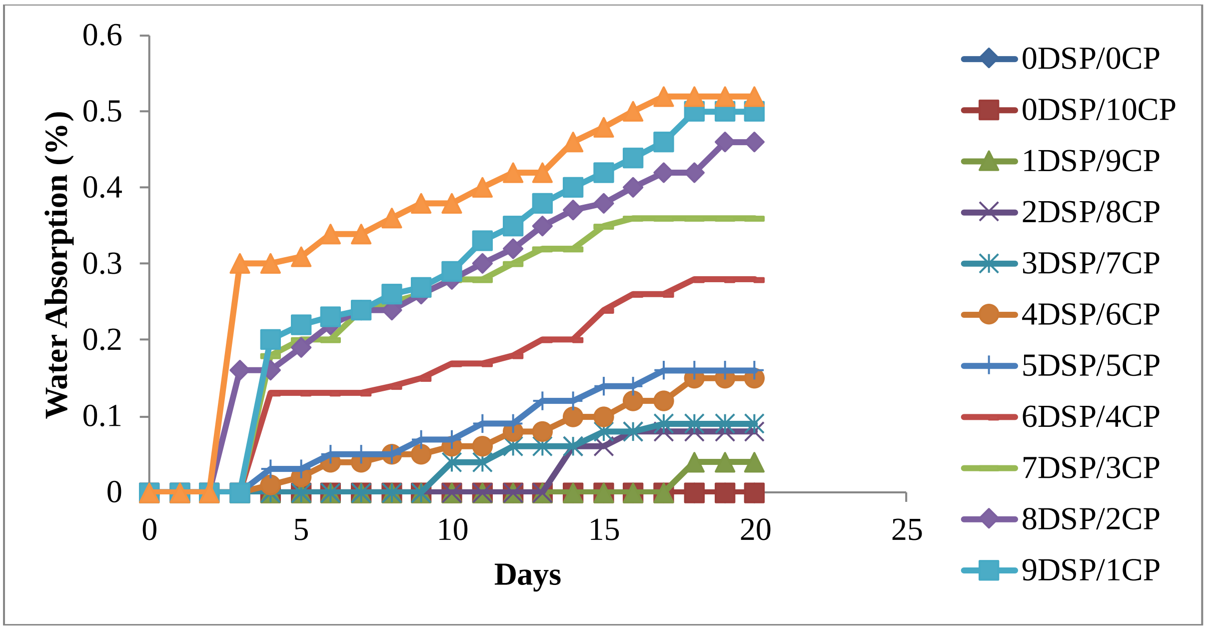

Figure 4.2: Water Absorption versus filler loading (%) for the Hybrid Composites.

MECHANICAL PROPERTIES

Tensile Strength

From the result obtained, the control sample (0.0% DSP / 0.0% CP/100%PP) shows the highest tensile strength values with 29.153 MPa compare to the hybrid composite samples, and this maybe a result of high polymer molecular chain mobility since there is no filler particles incorporated into the polymer matrix. It was observed among the hybrid composite samples 0.0%DSP/10%CP displayed a highest tensile strength value of 25.420Mpa followed by 4%DSP/6%CP with 25.031Mpa. This could be attributed to the extent of excellent inter mingling between the filler/fillers and the matrix interfacial interaction. This implies that the hybrid composites with 4% DSP / 6% CP can produce composites of adequate strength value for useful applications such as book shelf's, pharmaceutical shelf's, shoe horns, particle board and partition wall. Jamila *et al.,* (2016). Ahmad *et. al.,* (2021) studied the effect of hybrid materials configuration on the mechanical properties of composites. They observed that the palm and glass fibers in the polymer matrix continue to support each other when the tensile force is applied.

Elongation at Break

The effect of filler loading on elongation at breaks of the Hybrid composites are given in Figure 4. The elongation at break of the composites decreased with increase in filler loading but the control samples showed highest elongation at break of 46.467 MPa compared to the rest of the composites. This observation highlights the fact that the incorporation of much filler into polymer matrix improves the stiffness of the composites thus reducing toughness, that is to say the addition of the rigid filler decreased the ductility of the polymer matrix. This is because of the reduction in the bond strength at the filler / matrix interface. From the Figure 4 it can be seen that 4% DSP / 6% CP filler loading with 21.784 MPa is higher than the rest of the composite samples. This is attributed to the decrease in the polymer molecular chain mobility as reported by other researchers such as Muktari *et. al.,* (2019). They found that incorporation of rigid filler into the polymer matrix decreased the flexibility of low-density polyethylene.

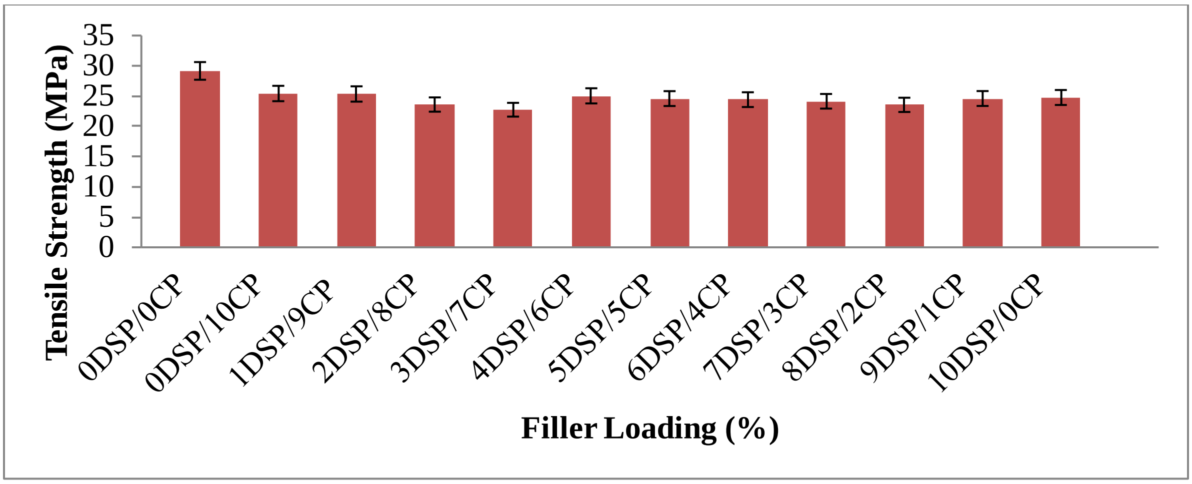

Figure 3: Tensile Strength versus filler loading (%) of the Hybrid Composites

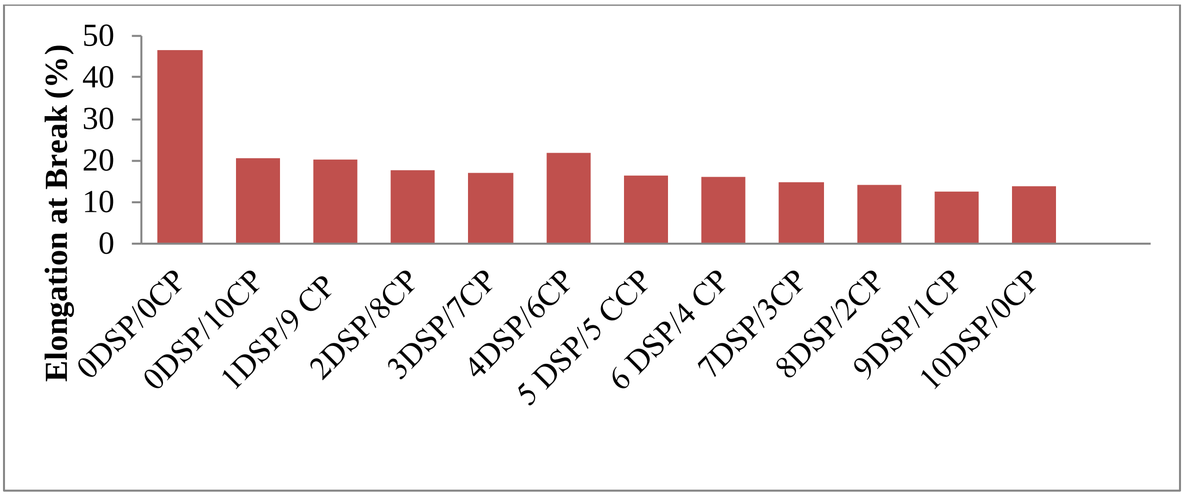

Figure 4: Elongation at Break versus filler loading (%) of the Hybrid Composite

Tensile Modulus

The results of tensile modulus of the hybrid Composites are presented in Figure 5. From the results obtained an increase was observed from 0%DSP / 0%CP, to 3%DSP / 7%CP with 0.627 and 1.489 GPa progressively. A shape decrease was then seen with 4%DSP / 6%CP at 1.169 GPa there after an increase from 5%DSP / 5%CP, to 10%DSP / 0%CP at 1.53, and 1.93 GPa respectively. This behavior is due to the reduction in the ductility / toughness of the polymer and increase in its rigidity as a result of incorporating rigid fillers (DSP and CP). This result is in line with the findings of Pai and Jagtap (2015). In the banana fiber and silica powder reinforced composite material that was developed by Singh *et. al.*, (2012), it was found that the adding of the fibers increases the modulus and decreases the ultimate tensile strength of the epoxy.

Hardness

The experimental result for hardness shown in Figure 6 suggests that the hardness increased after reinforcement with Date seed powder / Clay particles. The addition of 10% clay particles and 0.0% date seed particles to a 90% waste polypropylene matrix led to an increase in hardness to 31.73 HV for sample 0.0DSP / 10CP wt% and to 87.22 for sample 10DSP / 0CP wt%. The load was distributed on the filler, thus decreasing the penetration rate on the composite material's surface and increasing its hardness. The increase in the hardness in the composites is an indication of good bonding between polymer and fillers, reducing the movement of the polymer molecules. It has been reported in the literature by Karim, *et al.*,

(2018) that the hardness increased after reinforcement with carbon and glass fibres, the addition of 20% carbon and glass fibres to a 98% epoxy- 2% polysulfide rubber blend matrix led to an increase in hardness to 82.45 for sample A2 and to 82.66 for sample A3. In a hybrid composite, filler weight fraction significantly affects the hardness value of the hybrid composite material. It was found that hardness of neat polypropylene resin (0DSP / 0CP) is 18.47 HV. The hardness of the hybrid composite made of polypropylene resin and 10 wt% of date seed particles and 0 wt% of clay particles is maximum and is 87.22 HV. The hardness increases with the amount of date seed particles reflecting the reinforcement formed in the hybrid composite. The increased observed in hardness is due to the presence of enough rigid filler particles to absorb and share the stress as reported by researchers such as Singh, *et. al.*, (2012)

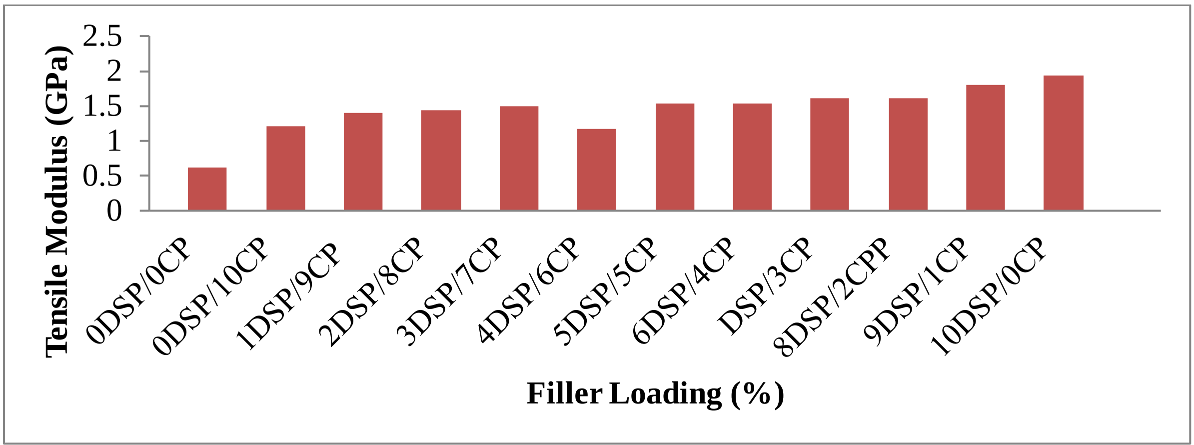

Figure 5: Tensile Modulus versus filler loading (%) of the Hybrid Composites.

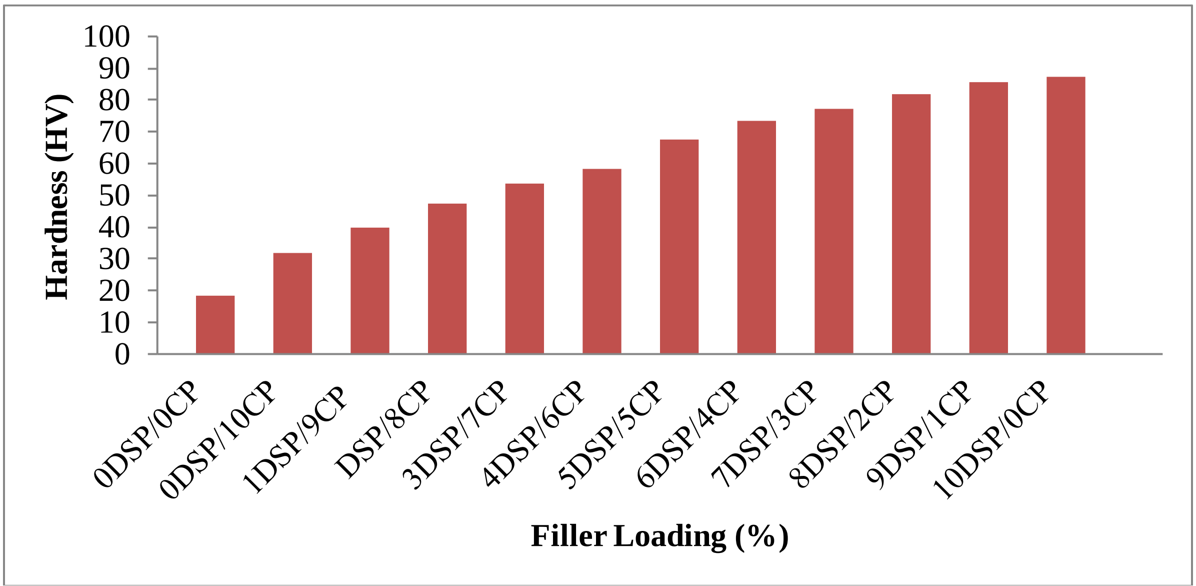

Figure 6: Hardness versus filler loading (%) for wPP / Date seed powder / Clay particles Hybrid Composites.

CONCLUSION

The research concludes that Hybrid Composite of wPP, DSP, and CP provide tunable properties depending on the filler loading combination. Mechanical Properties: Increasing DSP enhances tensile modulus, flexural modulus, and Hardness, making DSP enriched composites suitable for applications requiring rigidity. However, higher CP loading decreases mechanical properties but improves density and water resistance. Physical Properties, DSP lowers density but increases water absorption due to its hydrophilic nature, while CP improves water resistance and density due to its hydrophobicity and higher density. Flammability: Higher DSP content reduces flame resistivity, while CP Increases it, making higher CP loading more fire resistance. The study validates that the balanced filler combination (such as 4DSP/6CP or 6DSP/4CP) can provide a synergic effect, improving both mechanical and physical properties.

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