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INVESTIGATION INTO MECHANICAL, SURFACE AND ADHESIVE PROPERTIES OF DATE PALM WOOD-POLYOLEFIN MICRO COMPOSITES

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ABSTRACT

Wood-plastic composites are composite materials made of wood fibre or other ligno-cellulosic materials and thermoplastic(s). Date palms are one of the potential replacements of insufficient timber sources in the Middle East and the Horn of Africa countries. Low density polyethylene (LDPE) was blended with date palm wood (DPW) (*Phoenix dactylifera*) powder to prepare composites with the concentrations of filler ranging from 10 to 70 wt. %. The Young´s modulus of the composites significantly increased with an increase in the filler content in the entire concentration range. The maximum value of 1933 MPa for the composite filled with 70 wt.% of the filler is approximately 13 times higher than that for the neat LDPE. The incorporation of DPW into the LDPE matrix led to a significant increase in the polarity of composites and to an increase in their adhesion to polar substrates.

Key words: date palm wood, polyethylene, wood-plastic composite, adhesion, shear strength.

INTRODUCTION

The date palm (*Phoenix dactylifera*) consists of six different potential sources of natural fibres, namely bunches, mesh, petiole, fruit (pits), leaves and palm trunk (Keledi *et al.* 2012). Mechanical characteristics of palmyra palm (*Borassus aethiopum* Mart.) were determined and results of this study showed that mechanical properties of the palmyra are very influenced by the number and the mechanical characteristics of the fibres (Kimtangar *et al.* 2019). Both date palm fibres and wood themselves and their composites have been recently investigated (Agoudjil *et al.* 2012).

Alsewailem *et al.* (2010) studied high density polyethylene (HDPE) and polystyrene (PS) matrices reinforced with powder from date palm pits and their mechanical and thermal properties. Polyethylene based matters were successfully used for a dimensional stabilization of wood (Repák and Reinprecht 2020) or bonding of veneer (Bekhta and Sedliačik 2019). Alsaeed *et al.* (2013) investigated epoxy resins reinforced with long date palm fibres. The authors searched for the optimum length of embedded fibres that have controlled interfacial adhesion properties and determined that 10 mm was the optimum length. Similar research focused on the effect of diameters and alkali treatments on the tensile properties of date palm fibre reinforced epoxy composites was performed by Abdal-Hay *et al.* (2012). These authors determined that the ultimate tensile strength and percentage elongation of a single fibre after alkali treatment increased by 57% and 24.7%, respectively. Because the alkali treatment of date palm fibres was able to provide good adhesion within the matrix, the tensile strength, elastic modulus and the fibre-matrix interaction of the composite were improved. Date palm wood powder/glass fibres reinforced hybrid composites of recycled polypropylene were investigated by many researchers (WOLCOTT and ADCOCK 2000, LYUTYY *et al.* 2018, MIRSKI *et al.* 2019, AL-OTAIBI *et al.* 2020). The influence of date palm fibres from different parts of the date palm plant (the trunk, rachis, and the petiole) on the mechanical properties of HDPE-based composites was studied by MAHDAVI *et al.* (2010). The highest strengths were achieved in composites with 30 and 40% fibre content, and these gains were dependent on what parts of the original tree were used.

Wetting of wood based materials by standard liquids is a complex process controlled by chemical composition of the liquids used, properties of the substrate, interactions among unsaturated force fields across the phase boundary between wood and liquid, as well as by secondary effects of a range of factors implied by specific properties of the wood and the liquids used (KÚDELA 2014).

The aim of this contribution is determination of selected physical and mechanical properties of composites based on low-density polyethylene (LDPE) and date palm wood powder.

MATERIAL AND METHODS

Linear low density polyethylene was used as the matrix (melting point = 110.6 ± 0.1 °C and specific enthalpy of melting = 118 ± 5 J/g) and ground date palm wood powder (DPW) was used as the filler.

Large pieces of DPW were ground using a high energy mill. The obtained filler had a linear fibrous shape with the moisture content of 7%. The average length and the standard deviation were calculated from at least 20 measurements. The majority of the filler particles had the length ranging from 1 to 3 mm. The composites were prepared by mixing both components in the 30 ml mixing chamber of a Brabender Plasticorder PLE 331 at 140 °C for 10 minutes at a mixing speed of 35 rpm. 1-mm thick slabs were prepared by compression moulding of the mixed composites using a Fontijne SRA 100 laboratory press at 140 °C for 1 minute. Dog-bone shaped specimens with a working area of 30×4×1 mm were cut from the slabs. The mechanical properties were measured at room temperature using an Instron 3365 universal testing machine at a deformation rate of 50 mm.min^{-1}.

For adhesive tests, Epoxy resin CHS-Epoxy 531 and polyamine hardener Telalit 410 (Spolchemie), mixing ratio of epoxy resin to hardener = 4 : 1 weight parts, dichloromethane (Merck), have been used.

The surface properties of the LDPE/DPW composites were determined by measuring the contact angles of re-distilled water using a Surface Energy Evaluation system coupled with a web camera (Advex) and PC software. The drops of water, which was used as a testing liquid (V = 3 μl), were placed on the investigated surface with a micropipette (0−5 μl, Biohit), and the contact angle of the testing liquid was measured just after drop deposition.

The shear strength of the adhesive joints (P_{shear}) was measured by tensile testing of the single overlapped adhesive joints. The adhesive joints were prepared using LDPE/wood filler slabs with dimensions of 60 × 10 × 2 mm. The thickness of the epoxy/based adhesive layer between slabs was 0.1 mm, the length of the overlap was 15 mm, and the bonded area was 150 mm^2. The LDPE/wood filler slabs were bonded together at laboratory temperature in a hand press using an epoxy/based adhesive. The adhesive joints were tested using an Instron 4301 universal testing machine at a constant crosshead speed of 10 mm.min^{-1}.

RESULTS AND DISCUSSION

Surface and adhesive properties

The surface and adhesive properties of the LDPE/DPW composites were investigated. The dependence of the water contact angle on composite surface *vs*. the filler content in LDPE is shown in Figure 1. The increase in the filler content results in a more polar nature of the composite material. The dependence of the water contact angle *vs*. the wood content decreases nonlinearly. The water contact angle on the LDPE/DPW composite surface significantly decreases with the DPW concentration from 93.2 deg (unfilled polyethylene) to 87.8 deg (30 wt. % of wood in composite), and to 78.9 deg (70 wt. %. of the filler).

The results of the shear strength in the adhesive joint LDPE/DPW composite – epoxy *vs*. filler content are shown in Figure 2. The shear strength of the adhesive joint between the LDPE/DPW composite and the epoxy resin significantly increased from 0.62 MPa (unfilled PE) to 1.37 MPa (LDPE/DPW composite with 70 wt.% of the filler).

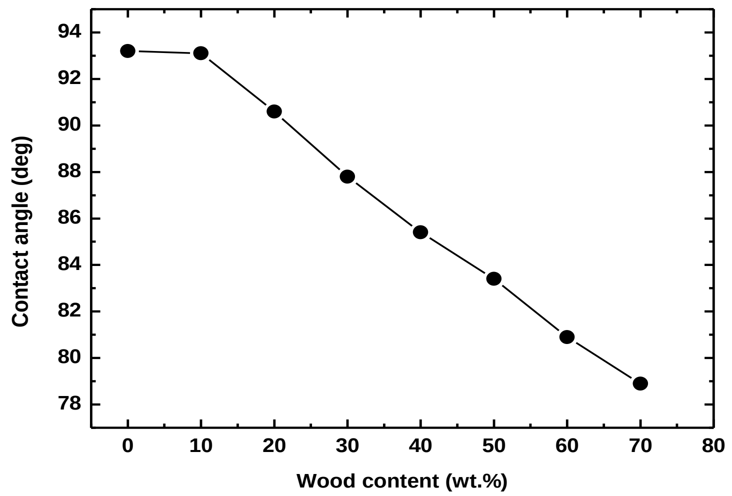

Fig. 1 The dependence of the water contact angle on the LDPE/DPW surface vs. filler content.

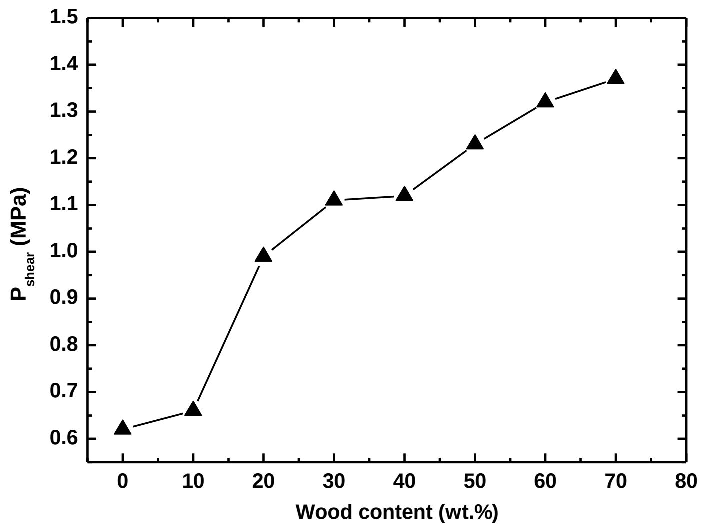

Fig. 2 Shear strength of adhesive joint LDPE/DPW composite – epoxy adhesive vs. filler content.

Mechanical properties

The mechanical properties of the composites tested in the tensile mode at room temperature (25 °C) were characterised. The static mechanical properties evaluated from the stress-strain curves included the yield point, stress and elongation at break and Young's modulus. The stress-strain diagram of LDPE is shown in Figure 3.

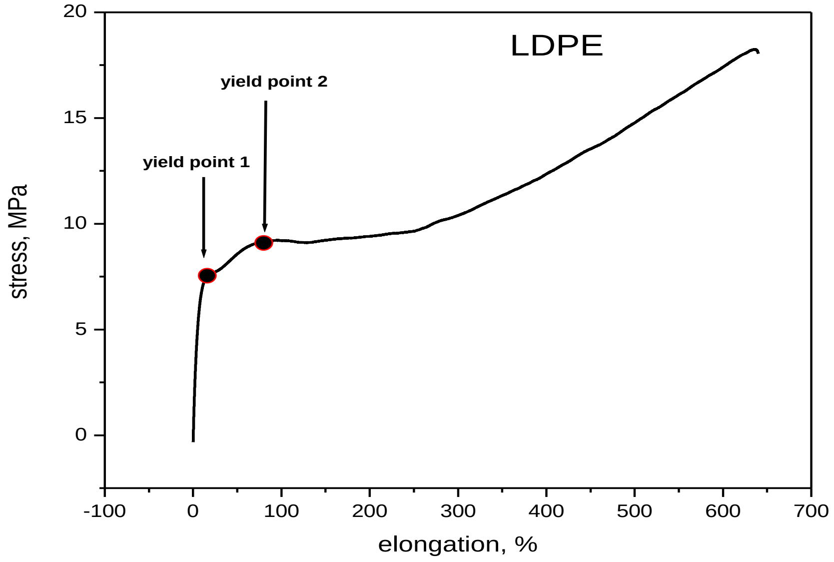

Fig. 3 The tensile stress-strain diagram of neat LDPE.

The LDPE behaviour is common for polyolefins. Fig. 3 reveals significant cold drawing and good deformability up 650%. The materials also undergo strain (orientation) hardening, which results in a high strength at break. There are also two distinguished yield points in the curve, which is a common behaviour of polyolefins that have a broad size distribution of crystallites. A yield point in polymers is conventionally accepted as being the point where the stress-strain curve exhibits a local maximum. For samples that initially deform homogeneously, this maximum occurs as a result of the internal plastic strain rate of the material increasing to a point where it becomes equal to the applied strain rate. In some cases, a maximum in the force also relates to the onset of necking, where the strain hardening of the necked materials is not sufficient to counteract the reduction of the cross-sectional area, which leads to a reduction in load. This maximum may become less defined as the testing temperature is increased or as the strain rate is decreased, until it disappears. The temperature where the local maximum disappears is lowest for the most branched material and highest for the unbranched, high-density material. The yielding phenomenon of semi-crystalline polymers is associated with a change in the morphology of the material, where a spherulitic structure transforms into a fibrillar one. During stretching, this change occurs through shearing and fragmentation of the crystalline lamellae into blocks that rearrange into the form of parallel microfibrils.

The stress-strain diagram of composite consisting of 10 wt.% of DPW is shown in Figure 4.

A dramatic decrease in drawability is observed, even at this very low filler content. The filler particles represent defects and stress concentrators and significantly reduce the drawability of the matrix. The orientation hardening is completely suppressed; however, the material exhibits some extent of plastic deformation. In this case, the rupture is not brittle.

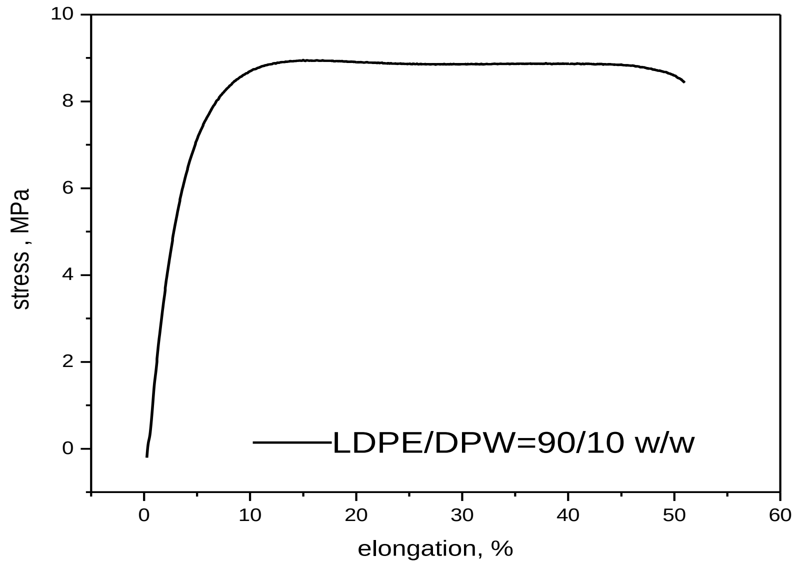

Fig. 4 The tensile stress-strain diagram of LDPE/DPW=90/10 w/w.

The behaviours of the composites filled with 20 and 30 wt.% of wood are similar. However, when the matrix is filled with 40 wt.% of the filler and greater, the material becomes brittle. The plastic deformation is fully suppressed, and the material is broken after the yield point. This behaviour is shown in Figure 5.

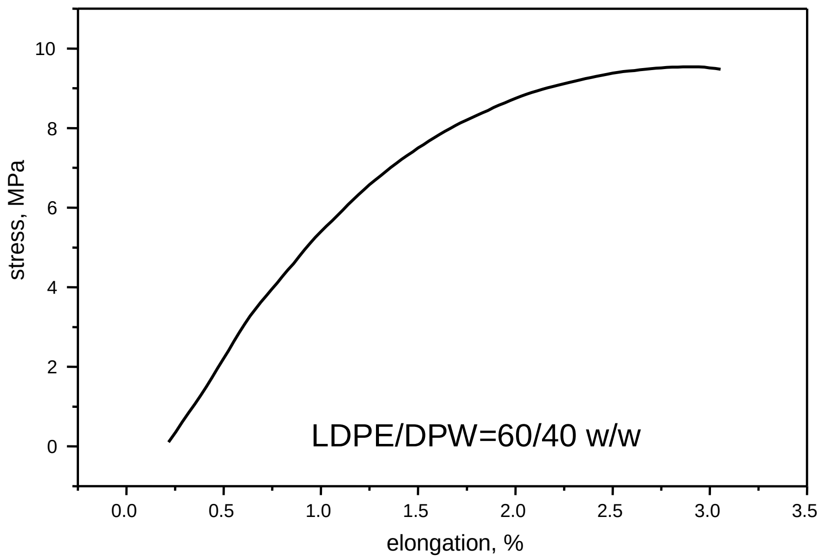

Fig. 5. The tensile stress-strain diagram of LDPE/DPW=60/40 w/w.

The mechanical properties of the composites determined at 25 °C are summarised in Table 1. The stiffness of the composites, which is characterised by the Young´s modulus, significantly increased with an increase in the filler content in the entire concentration region. The maximum value of 1933 MPa for the specimen filled with 70 wt.% of the filler is approximately 13 times greater than the one of the LDPE. This result indicates that the filler has a very strong reinforcing effect. Similar results were achieved by AMMAR et al. (2019) who obtained results showed that the sample made from date palm fibres and bio-matrix from lignin-glyoxal-resin thermo pressed in the ratio of 50/50 wt.% has the best mechanical properties.

Tab. 1 Basic statistical variables for the stress and strain characteristics of the composites loaded in tension. The x/y notation represents the LDPE/DPW w/w ratio.

Sample	φ	ε_y ($S\varepsilon_y$) (%)	σ_y ($S\sigma_y$) (MPa)	ε_b ($S\varepsilon_b$) (%)	σ_b ($S\sigma_b$) (MPa)	E (S_E) (MPa)
LDPE	0	15.5[a] (0.3) 96.0[b] (0.4)	8.0 (0.2) 9.4 (0.1)	633 (20)	18.5 (0.7)	150 (7)
90/10	0.069	15.2 (0.3)	9.2 (0.2)	22.0 (1.9)	9.0 (0.3)	285(22)
80/20	0.142	br	br	8.8 (0.6)	9.2 (0.4)	376 (22)
70/30	0.221	br	br	4.8 (0.6)	9.4 (0.2)	562 (71)
60/40	0.306	br	br	3.2 (0.3)	9.3 (0.5)	800 (42)
50/50	0.398	br	br	2.1 (0.1)	9.7 (0.5)	1064 (83)
40/60	0.498	br	br	1.4 (0.1)	10.2 (0.4)	1457 (122)
30/70	0.608	br	br	1.1 (0.1)	11.1 (0.6)	1933 (124)

Where:

ε_y, σ_y, ε_b, σ_b, E – elongation at yield, yield stress, elongation at break, stress at break, and Young's modulus of elasticity,

$S\varepsilon_y$, $S\sigma_y$, $S\varepsilon_b$, $S\sigma_b$, S_E – standard deviations,

φ – the volume portion of the filler,

br – refers to the brittle rupture,

[a,b] – two yield points are observed, as shown in Figure 3.

The stress at the break of the composites and the dependence on the filler fraction varies nonlinearly. We have considered two influences of the filler on the stress at the break. On the one hand, we have to consider that the reinforcing effect of the filler leads to an increase in the tensile stress values with an increase in the filler fraction and, on the other hand, the orientation strengthening occurs for semi-crystalline polymers at high deformation. The latter effect is indirectly negatively influenced by the filler presence and by a steep decrease in the deformation such that orientation of the matrix cannot occur. At low filler fractions, the deformation is sufficiently low to prevent the orientation, but the reinforcing effect of the filler presence is marginal. The particles of the filler represent defects and stress concentrators. The behaviour of the stress-strain curve is changed; orientation hardening and cold flow are suppressed. The samples break close to the yield point. Therefore, an initial dramatic decrease of tensile strength was observed. The initial stress at the break of LDPE (18.5 ± 0.7 MPa) decreased to a value of 9.0 ± 0.3 when it was filled with 10 wt. % of the filler. However, in this case, only the orientation hardening was suppressed. The cold flow occurs only up to 10 wt.% of the filler. After this level, material becomes brittle if the filler content increases. The slight increase in the stress at break at higher filler contents is caused by the reinforcing effect of the filler.

The Young's modulus is often a property of particular interest. The improvement in the Young's modulus is an expected outcome because of the reinforcement effect of the filler particles. It is generally known that the improvement of the tensile modulus is caused by the good dispersion of particles and good interfacial adhesion between the particles and the matrix; therefore, the mobility of polymer chains is restricted under loading. The effect of different polypropylene (PP) matrices on the mechanical, morphological, and thermal properties of date palm fiber (DPF)-reinforced PP composites was investigated by AL-OTAIBI et al. (2020). They concluded the same tendency, the tensile modulus is slightly increasing with increasing the weight percent ratio of added amount of date palm fibres starting at 1850 MPa and growing to 2600 MPa.

CONCLUSION

A fine powder with a fibrous shape was prepared from date palm wood by grinding in a high energy mill. The prepared fibres have a broad size distribution; the majority of the fibres have a length between 1 and 3 mm. Low density polyethylene was used as the matrix for preparing LDPE/DPW composites. The filler concentration ranged from 10 to 70 wt.%. The stiffness of the composites, which were characterised by the Young´s modulus, significantly increased with an increase in the filler content in the entire concentration range. The maximum value of 1933 MPa for the composite filled with 70 wt.% of the filler is approximately 13 times greater than that for the LDPE. This result indicates that the filler has a strong reinforcing effect and that there is a good distribution of the filler. Furthermore, the strength limit of the composites and its dependence on the filler fraction varies nonlinearly. The material becomes brittle if filled with more than 10 wt. % of the filler. The incorporation of DPW into the LDPE matrix led to a significant increase in the polarity of composites and to an increase in their adhesion to polar substrates.

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