Vinyl Acetate Modified Scots Pine Reinforced HDPE Composites: Influence of Various Levels of Modification on Mechanical and Thermal Properties

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In order to compare the effect of vinyl acetate modification (VA) at different levels of weight percentage gain, Scots pine wood flour was modified with VA to three different wt% gains (10%, 16%, and 21% WPGs). Acetic anhydride (AA) modification at 24% WPG was also studied. Modified Scots pine wood flour reinforced HDPE composites (WPCs) were produced at 30 wt% wood flour loading by using extrusion-injection molding process and the mechanical properties of WPCs were determined. The thermal and morphological properties of WPG were characterised by using TGA and SEM techniques. The increase in tensile strength was significant for VA modified WPCs in comparison to the AA modified, unmodified or neat HDPE composites. Thermal stability was also significantly improved with increasing the WPG levels of VA modification.

Keywords: Vinyl acetate; Acetic anhydride; Acetylation; Wood plastic composites; High-density polyethylene; Scots pine

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INTRODUCTION

The popularity of wood-plastic composites (WPC) has rapidly increased over the last two decades. Wood-plastic composites refer to an extremely wide range of composite materials that use polymers ranging from PE to PVC and fillers ranging from wood flour to natural fibers (Rials *et al.* 2001). WPCs are commonly used in the many application areas in daily life such as a residential decking, automotive trim, window frames, *etc*. (Klyosov 2007).

The main drawback of WPCs is the lack of adhesion between the hydrophilic wood flour and the hydrophobic polymers; this leads to a decrease in some of the mechanical properties. Insufficient adhesion between hydrophilic wood flour and hydrophobic thermo-plastics results in poor mechanical properties of the wood-filled polymer composites. Wood flour shows a tendency of forming large aggregates, which cause poor dispersion of the flour in the polymer matrix.

In order to improve adhesion between lignocellulosic fibers and polymeric matrix, different approaches have been applied to change the fibers-matrix adhesive properties in lignocellulosic fibers-reinforced composites. These include chemical or physical modifications of the fiber such as reaction with acid compounds (Karnani *et al.* 1997; Zafeiropoulos *et al.* 2002), organosilane (Kokta *et al.* 1989; Karnani *et al.* 1997), and alkali treatments (George *et al.* 2001).

Another problem is the relatively low processing temperature that is required, since the rate of thermal decomposition of lignocellulosic fibers increases remarkably with an increase in temperature. The processing temperatures for most of the natural fibers are thus limited to about 200°C, although within the normal range of processing temperature (180 to 200°C), significant extents of decomposition are realized (Sanadi *et al.* 1996; Mohanty *et al.* 2000). It was reported that the use of acetylated wood flour in polystyrene matrix resulted in better thermal stability than a non-acetylated control sample (Lisperguer *et al.* 2007). It was also reported in our earlier study that a 19 wt% gain level of acetylation with vinyl acetate shifted the active decomposition temperature from 310°C to 370°C for wood flour in HDPE matrix (Özmen *et al.* 2013).

There are studies that focus on acetic anhydride-modified fiber reinforced polymer composites (Tserki *et al.* 2006; Lisperguer *et al.* 2007; Abdul Khalil *et al.* 2007; Luz *et al.* 2008; Mat Taib *et al.* 2010). Some authors claim that acetylation improves the mechanical and thermal properties of WPCs (Tserki *et al.* 2006; Lisperguer *et al.* 2007; Abdul Khalil *et al.* 2007; Özmen *et al.* 2013). In contradiction to these studies, it was also reported that acetylation reduces the mechanical properties of WPCs (Luz *et al.* 2008; Mat Taib *et al.* 2010).

In our previous study (Özmen *et al.* 2013), the effect of wood flour acetylation with vinyl acetate or acetic anhydride on the mechanical properties of WPC's was reported. The composites reinforced with VA acetylated wood flour at 20 wt% loading exhibited better mechanical properties than the composites with AA acetylated wood flour. The experimental results suggested that acetylation may also play a key role in determining the compatibility of WPC's. Therefore, further study is needed for a better understanding of the effect of acetylation at various modification levels on the mechanical properties of WPC's at higher wt% wood flour loading.

In this study, the influence of vinyl acetate modification levels on the mechanical and thermal properties of 30 wt% wood flour reinforced HDPE composites was determined. Scots pine wood flour samples were pretreated with vinyl acetate (VA) to achieve three different wt% gain levels (10%, 16%, and 21% WPGs) or with classical acetylation technique acetic anhydride (AA) at the level of 24 wt% gain then compounded with HDPE. The morphologies of both VA or AA modified and unmodified WPCs were observed by scanning electron microscopy (SEM). The effects of the various levels of acetylation on the mechanical properties of final composites material were determined by tensile, flexural, and impact tests. Thermal properties of the VA or AA modified WPCs were investigated by thermogravimetric analysis (TGA).

EXPERIMENTAL

Preparation of Wood Flour

Scots pine (*Pinus sylvestris*) sapwood was ground using a hammer mill and sieved to a size of 250 to 425 μm. Before modification, the samples were subjected to Soxhlet extraction with a mixture of toluene/acetone/ethanol (4/1/1, v/v) for 8 h in order to obtain extractive-free wood. Extracted wood flour was oven-dried overnight, at 103±2°C and allowed to cool to room temperature in a desiccator containing phosphorous pentoxide.

Acetylation of Scots Pine Wood Flour

The extractive-free Scots pine sapwood flour (500 g) was transferred to a 2-L glass reactor containing a dimethyl formamide (DMF) solution with acetic anhydride (AA) or vinyl acetate (VA). Reactions were carried out at 100°C for various reaction times (2, 4, and 6 h), with potassium carbonate (K_2CO_3) as a catalyst and DMF as a solvent. At the end of the reaction, the modified samples were Soxhlet extracted with deionised water for 6 h, then with a toluene/acetone/ethanol mixture (4/1/1, v/v) for 6 h and subsequently oven-dried overnight, at 103±2°C. The samples were transferred in a desiccator until cool and the weight percent gains (WPG) were calculated according to Equation 1,

$$WPG\ (\%) = \frac{Wt_1 - Wt_0}{Wt_0}\ x\ 100 \quad (1)$$

where W_{t0} and W_{t1} are oven dry weights of wood, before and after esterification, respectively.

Infrared Spectroscopy

Infrared absorption spectra of the unmodified and acetylated wood flour were obtained using the KBr (potassium bromide) technique, using a Shimatzu 8400s FT-IR spectrometer at a resolution of 4 cm^{-1} (40 scans). In each case, 1% w/w of oven dry wood flour was dispersed in a matrix of KBr and pressed to form pellets.

Thermogravimetric (TGA) Analysis

The thermogravimetric analyses of the produced composites were performed by thermal gravimetric analyzer (PerkinElmer TGA Analyzer Pyris 1 TGA). The sample weight was 20 mg. The measurement was carried out at a heating rate of 10°C/min in an inert nitrogen atmosphere with a flow rate of 20 mm/min. Weight loss versus temperature was recorded.

Production of Wood Plastic Composites (WPC)

High-density polyethylene was obtained from Petkim, Turkey (HDPE; Petilen® YYS0464). Six different compositions were used for production of composites which are shown in Table 1.

Table 1. Compositions used for Producing WPCs

ID	HDPE (wt%)	Wood flour (wt%)	Modifying agents	WPG (%)
HDPE	100	-	-	-
W	70	30	-	-
VA_{10}	70	30	Vinyl acetate	10
VA_{16}	70	30	Vinyl acetate	16
VA_{21}	70	30	Vinyl acetate	21
AA_{24}	70	30	Acetic anhydride	24

Scots pine wood flour and HDPE were pre-blended in a mixer and then extruded using a single-screw extruder with a length-to-diameter ratio (L/D) of 30. The temperature setting from the hopper to die was 150/170/170/177°C and the screw speed was 100 rpm. The pellets (wood/HDPE ratio of 30/70 by weight) were dried at 70°C for 24 h and then

injection-molded using a HDX Injection Molding Machine (pressure: 100 bar; injection speed: 80 mm/sec; screw speed: 40 rpm) to produce standard test samples.

All samples were conditioned at a relative humidity of 65±5% and a temperature of 23±2°C for 7 days, and then mechanical tests were performed in order to determine the effect of VA modification levels on the properties of WPCs. For each compositions, 10 specimens for tensile (dog bone shape (type III)) and flexural (5mm (t) x 13mm (w) x 150mm (l)) were tested with a crosshead speed of 5 mm/min and a crosshead speed of 2 mm/min in the three point of loading mode on a Zwick Roell Z010 Universal Testing machine, respectively. Notched impact specimens (5 mm (t) x 12.7 mm (w) x 64 mm (l)) were tested on a Zwick HIT5.5P Impact Testing machine. The notches were cut using a Polytest notching cutter by RayRan. The density, tensile, flexural, and impact properties of the manufactured samples were determined in accordance with ASTM D792 (2007), ASTM D638 (2001), ASTM D6109 (2005a), and ASTM256 (2005b), respectively.

Scanning Electron Microscope (SEM) Analysis

Samples from produced WPCs dipped into liquid nitrogen and then snapped into two to obtain a fractured surface for SEM analysis. The surfaces were analyzed directly with JEOL scanning electron microscope (Model NeoScope JCM-5000).

Statistical Analysis

Analysis of variance (ANOVA) was used to determine the effect of acetylation on the selected mechanical properties using OriginPro 8. The resulting F value was compared to the tabular F value at the 95% probability level. When F tests resulted in significant differences, comparisons between means were made by the Tukey test. Statistically different groups ($P<0.05$) are shown on the graphs with different capital letters.

RESULTS AND DISCUSSION

In order to understand the effect of the chemical modification levels on the properties of wood polymer composites, Scots pine samples were modified with VA with various reaction times (2, 4, and 6 h) and different wt% gain (WPG) levels obtained (10%, 16%, and 21% WPGs), respectively. Scots pine wood flour were also modified with AA under the same reaction conditions with VA, 24 % WPG level was achieved for 6 h of reaction time.

Figure 1 shows the FTIR spectra of AA-modified (24% WPG), VA-modified (10%, 16%, and 21% WPGs), and unmodified (W) Scots pine wood flour. As seen in Fig. 1, after acetylation with both chemicals (VA and AA), all modified samples showed a strong carbonyl stretching vibration (νC=O) peak, the methyl groups bending vibrations peak and C-O stretching vibration peak in the region of 1745 cm^{-1}, 1375 cm^{-1}, and 1242 cm^{-1}, respectively (Özmen 2012; Özmen *et al.* 2013). It was also observed that increasing with WPG levels of VA modified samples increased the intensity of these peaks.

The densities of neat HDPE, W, VA_{10}, VA_{16}, VA_{21}, and AA_{24} composites were measured as 0.94, 0.99, 0.98, 1.01, 1.02, and 1.13 g/cm^3, respectively. Statistical analysis showed no significant difference between the densities of the composites. Figure 2a shows the tensile properties for neat HDPE, W, VA_{10}, VA_{16}, VA_{21}, and AA_{24} composites at 30 wt% wood flour loading. Three conclusions can be drawn from this figure. Firstly,

addition of 30 wt% unmodified Scots pine wood flour had a negative effect on the tensile strength, which significantly dropped from 20.1 MPa to 18.2 MPa. This result might be due to the dissimilarities and lack of adhesion between the nonpolar HDPE matrix and polar wood flour filler. Similar results were also reported by others (Bledzki *et al.* 1998; Tserki *et al.* 2006). Another possible reason for this reduction might be the low thermal stability of lignocellulosic fibers. It was reported in Bledzki *et al.* (1998) that significant reduction was observed in the tensile strength values of the lignocellulosic fibers reinforced composites due to the fibers decomposition caused by high processing temperature. This decomposition can create voids inside the composite, causing a decrease in mechanical properties.

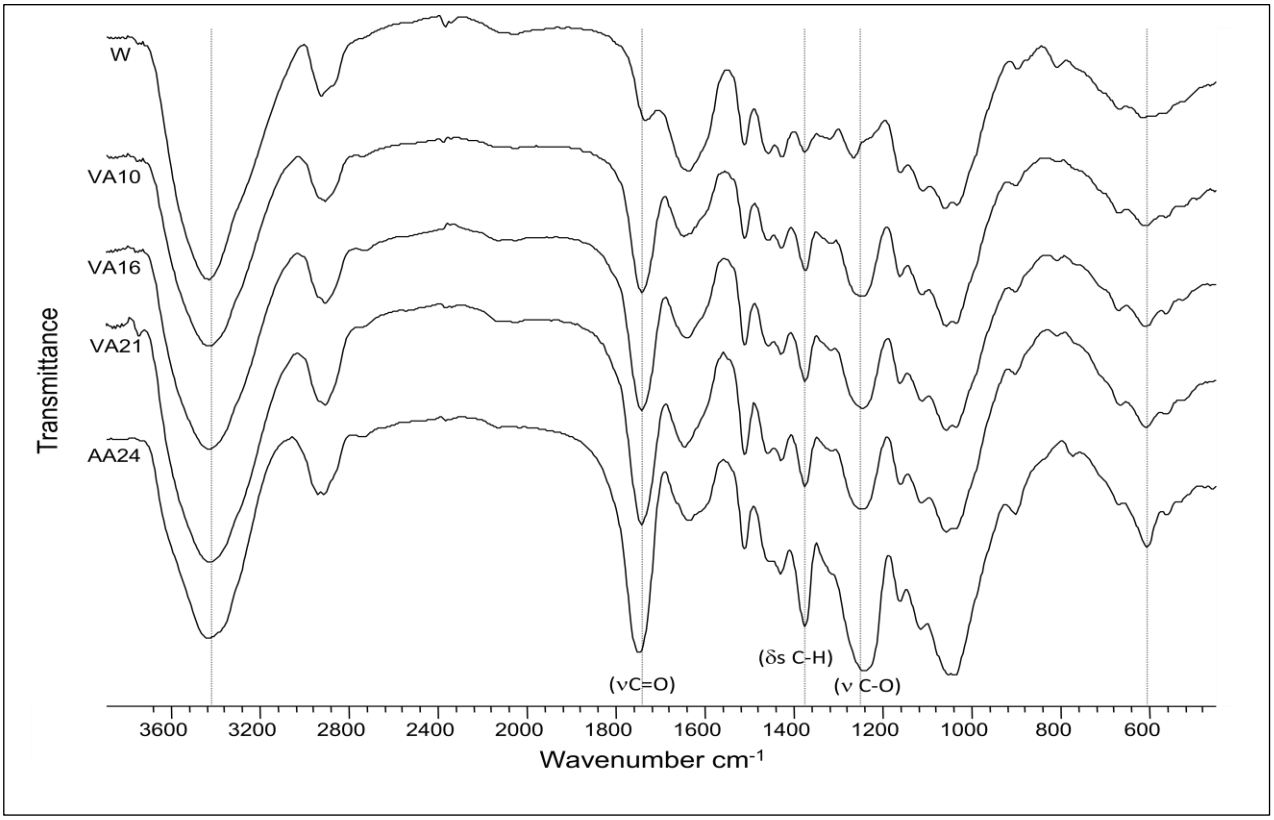

Fig. 1. FTIR spectra of untreated (W), acetic anhydride modified (24% WPG) (AA24), and vinyl acetate modified (VA10, VA16, VA21 represents 10%, 16%, and 21% WPGs, respectively) Scots pine wood flour.

Secondly, the ANOVA (α= 0.05) results showed that mean tensile strength values significantly increased with increasing modification levels. According to the Tukey test results, there was no significant difference between neat HDPE composites and low level modified (10% WPG) HDPE-wood composite (VA_{10}). An abrupt increase of tensile strength properties of HDPE-wood composites (VA_{21}) was achieved using high level acetylated wood flour (21% WPG level with VA modification) as a filler. Thirdly, a significant difference ($p \leq 0.05$) in the tensile strength was noted between two acetylation methods. The best tensile strength values were obtained with VA_{21} composites, which were 38% and 25% better than W (30 wt% unmodified wood-HDPE) and neat HDPE composites, respectively. The tensile strength properties of AA modified wood flour

reinforced HDPE composite (AA_{24}) were also significantly improved as compared to the unmodified one. It has been reported that the interface between fiber and matrix influences the mechanical properties of reinforced composites considerably (Bledzki *et al.* 1998; Tserki *et al.* 2006).

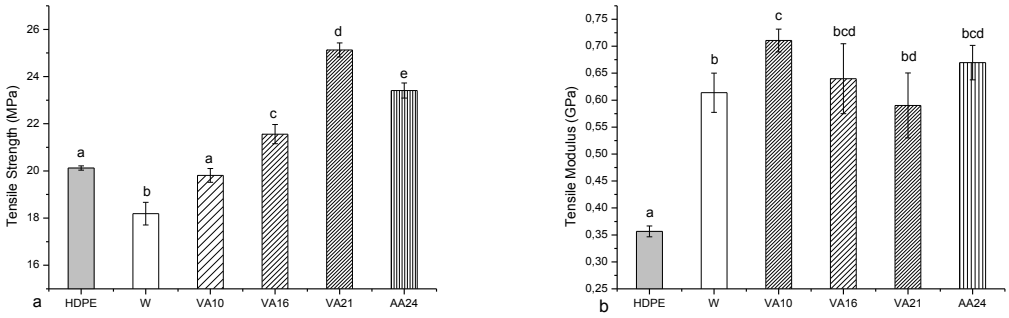

Fig. 2. Effect of acetylation levels on the tensile strength (TS) values (a) and the tensile modulus values (b) of WPCs (Bars with the same letter are not significantly different)

Mat Taib and coworkers (2010) reported that *Acacia mangium* fibers were modified with acetic anhydride to three different weight percent gain levels (4.9%, 13.7%, and 18.3% WPGs) and found that the acetylation of fibers significantly reduced the tensile strength of the HDPE composites. They stated that after modification, unreacted chemicals or by-products were removed by washing with water. However, in this study, at 30 wt% wood flour loading, AA or VA modified Scots pine wood flour reinforced HDPE composites gave better mechanical and thermal properties than unmodified wood flour-reinforced WPCs. Also, with the increase of acetylation levels, tensile strength values of the composites showed a significant increase (10%, 16%, and 21% WPGs levels, whereas VA-modified samples gave 19.8, 21.6, and 25.1 MPa tensile strength values, respectively). Hill reported (2006) that it is critical to remove unreacted chemicals, by-products, or solvents after a modification procedure, and this can greatly influence the properties of the chemically modified wood. It was not adequate to remove all unreacted chemicals or by-products by simple washing with water, and more effective extraction can be achieved with the use of Soxhlet apparatus (Çetin 2000; Hill 2006). This might be a reason for the high tensile strength values of VA or AA modified composites in this study due to effective Soxhlet cleaning procedure applied after acetylation.

Effect of acetylation levels on the tensile modulus of WPCs are shown in Fig. 2b. It was found that adding 30 wt% wood flour in matrix causes 72% increase on tensile modulus values compared to neat HDPE composites. This result is expected, since wood flour has higher modulus than HDPE matrix, a difference that is usually explained by the rule of mixtures. Low level acetylation (10% WPG) resulted in an additional 16% increase on the tensile modulus results. However, with further increase in the acetylation level, there was a slight decrease in the tensile modulus values compared to the low level VA modified composites (VA_{10}). This slight decrease on modulus can be explained by the reduction of wood flour amount in the matrix by increasing the acetylation degree of wood, since composites were produced through dry mixing of 70 wt% HDPE and 30 wt% wood flour.

Figure 3a shows the flexural strength values for the composites at 30 wt% wood flour loading. It is clear from the figure that adding wood flour in HDPE matrix caused

an approximate 51% increase of bending strength values. At low level acetylated samples, similar bending strength values were obtained compared to 30 wt% unmodified wood flour loading. After high level acetylation (VA_{21}), an additional 12% increase was observed on flexural strength compared to the W composite. In the case of flexural modulus (Fig. 3b), similar to the tensile modulus, it was increased with the addition of 30 wt% wood flour to the HDPE matrix. Acetylation of wood flour slightly reduced these values.

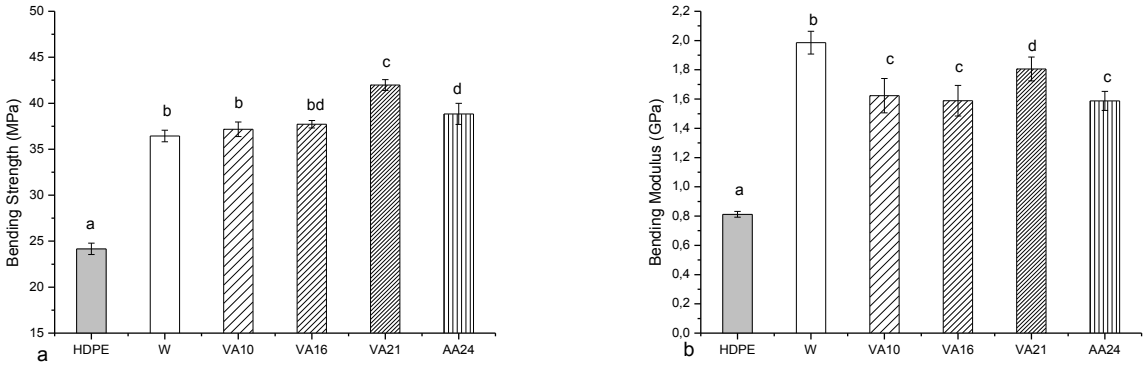

Fig. 3. Effect of acetylation levels on the flexural strength values (a) and on the flexural modulus values (b) of WPCs (Bars with the same letter are not significantly different)

Figure 4a shows elongation at break values of the neat HDPE, W, VA_{10}, VA_{16}, VA_{21}, and AA_{24} composites at 30 wt% wood flour loading. It is clear from the figure that addition of 30 wt% wood flour in the HDPE matrix reduced the elongation at break from 500% for neat HDPE to 5% for W composites. There was a reduction for elongation at break values with addition of wood flour into the polymer matrix (Tserki *et al.* 2006; Mathew *et al.* 2011). The elongation at break for composites with VA or AA modified wood flour (around 6%) was hardly higher than composites with untreated wood flour at similar loading.

The impact strength values of the WPCs are presented in Fig. 4b. Impact strength was significantly reduced with the wood flour addition. There was no significant difference in the impact strength values between W and acetylated (VA_{10}, VA_{16}, VA_{21}, and AA_{24}) composites.

Figure 5 shows an SEM micrograph of the freeze-fractured surface the unmodified wood flour composite (W). In the case of unmodified wood flour composites (W) fiber pull out took place, leaving holes and voids on the surface due to the weak interfacial adhesion between wood flour and HDPE matrix.

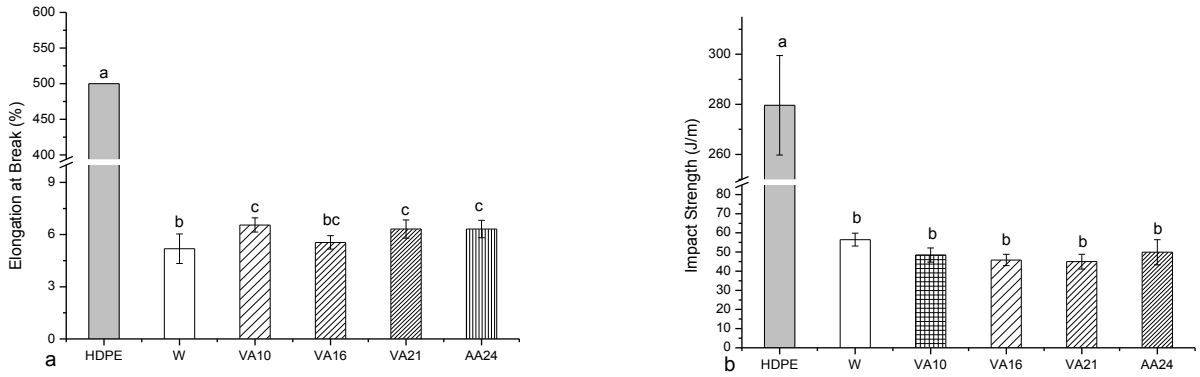

Fig. 4. Effect of acetylation levels on the elongation at break values (a) and the impact strength (IS) values (b) of WPCs (Bars with the same letter are not significantly different)

The SEM micrographs of the freeze-fractured surfaces of various WPG levels of VA modified (VA_{10}, VA_{16}, VA_{21}) and AA modified (AA_{24}) wood flour reinforced HDPE composites are shown in Fig. 6. As can be seen from Figs. 6a and b, low level VA-modified composites (10% and 16% WPGs levels) indicated the improvement of physical interaction between acetylated wood flour and HDPE matrix compared to the unmodified one, but there were some cavities and gaps between the wood flour and matrix. High level VA_{21} and AA_{24} modified composites showed better flour dispersion and an improved adhesion between flour and matrix (Fig. 6c, d), which resulted in significantly better mechanical properties of VA_{21} and AA_{24} composites.

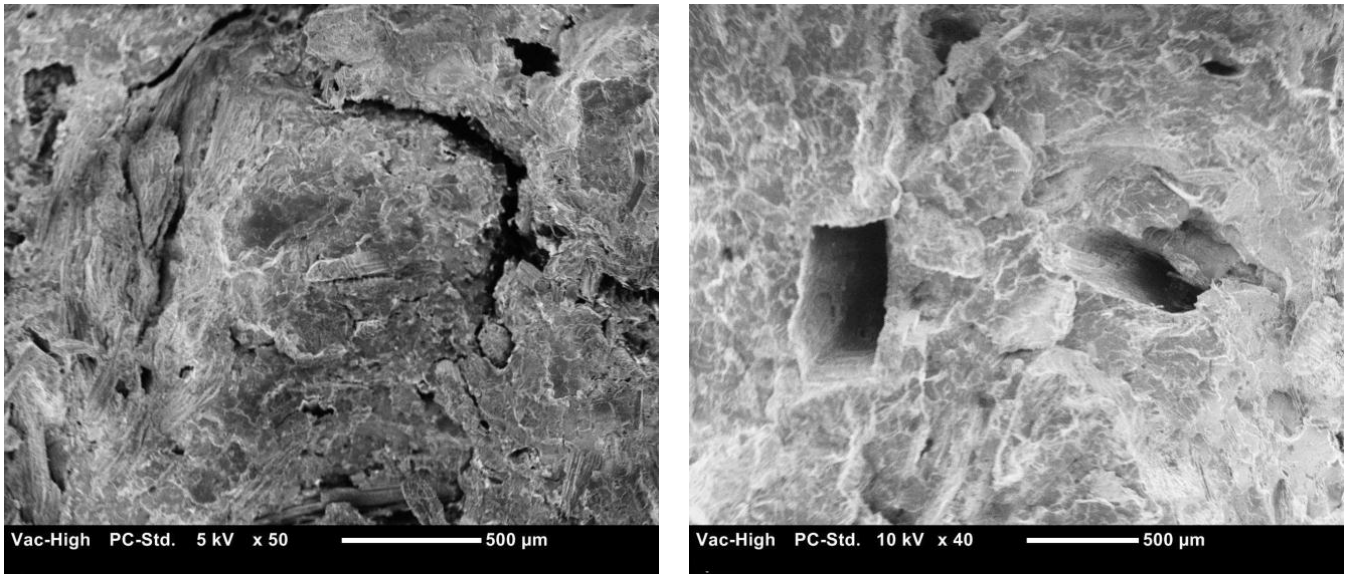

Fig. 5. SEM micrograph of freeze-fractured surface of W composite (30 wt% untreated wood flour reinforced HDPE) illustrating voids and fiber pullout

The composites, reinforced with untreated or VA-modified wood flour (at various WPG levels), were evaluated by thermogravimetric analysis (TGA) for verification of the thermal characteristic of the materials. Results are shown in Fig. 7. All WPCs showed a three-stages process. The initial weight lost due to moisture loss was located between 25 and 150°C (Stage I). The decomposition of wood flour occurred between 150 and 400°C in stage II. Finally, stage III was located between 400 and 525°C (revealing HDPE matrix decomposition). Complete decomposition of the composite occurred at 525°C.

On pyrolysis of the wood flour in HDPE matrix, the effect of acetylation levels on the active decomposition temperature was determined. Results are shown as a function of modification levels in Fig. 7f. It was observed that the main decomposition temperatures of wood flour in HDPE matrix showed a linear improvement with increasing the degree of VA modification (R-square = 0.987).

The main degradation temperatures of untreated wood flour, VA_{10} (10% WPG), VA_{16} (16% WPG), VA_{21} (21% WPG), or AA_{24} (24% WPG) were 343°C, 371°C, 381°C, 394°C, and 391°C, respectively. VA-modified Scots pine flour had the thermal stability to endure extruding temperature during thermoplastic composite manufacturing, resulting in higher tensile strength values compared to unmodified WPCs. W, VA_{10}, VA_{16}, VA_{21} or AA_{24} polymer composites gave around 8.8%, 7.6%, 6.7%, 5.9%, and 5.5% final ash contents, respectively. For VA modification, as WPG levels increased, the ash content gradually decreased and reached a minimum at 21% WPG.

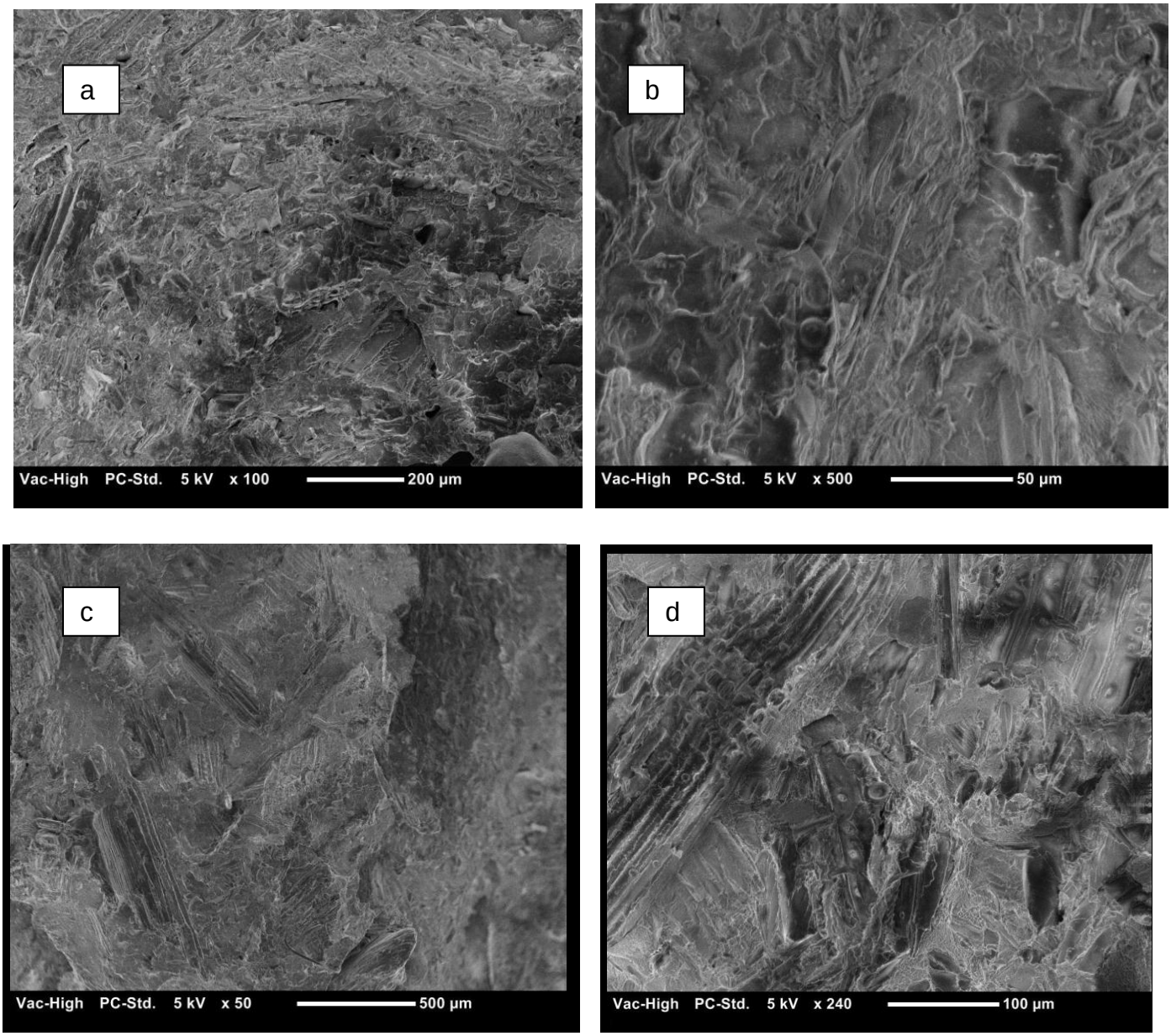

Fig. 6. SEM micrograph of freeze-fractured surface of VA_{10} (a), VA_{16} (b), VA_{21} (c), and AA_{24} (d) composites, illustrating more effective wood flour wetting by the matrix

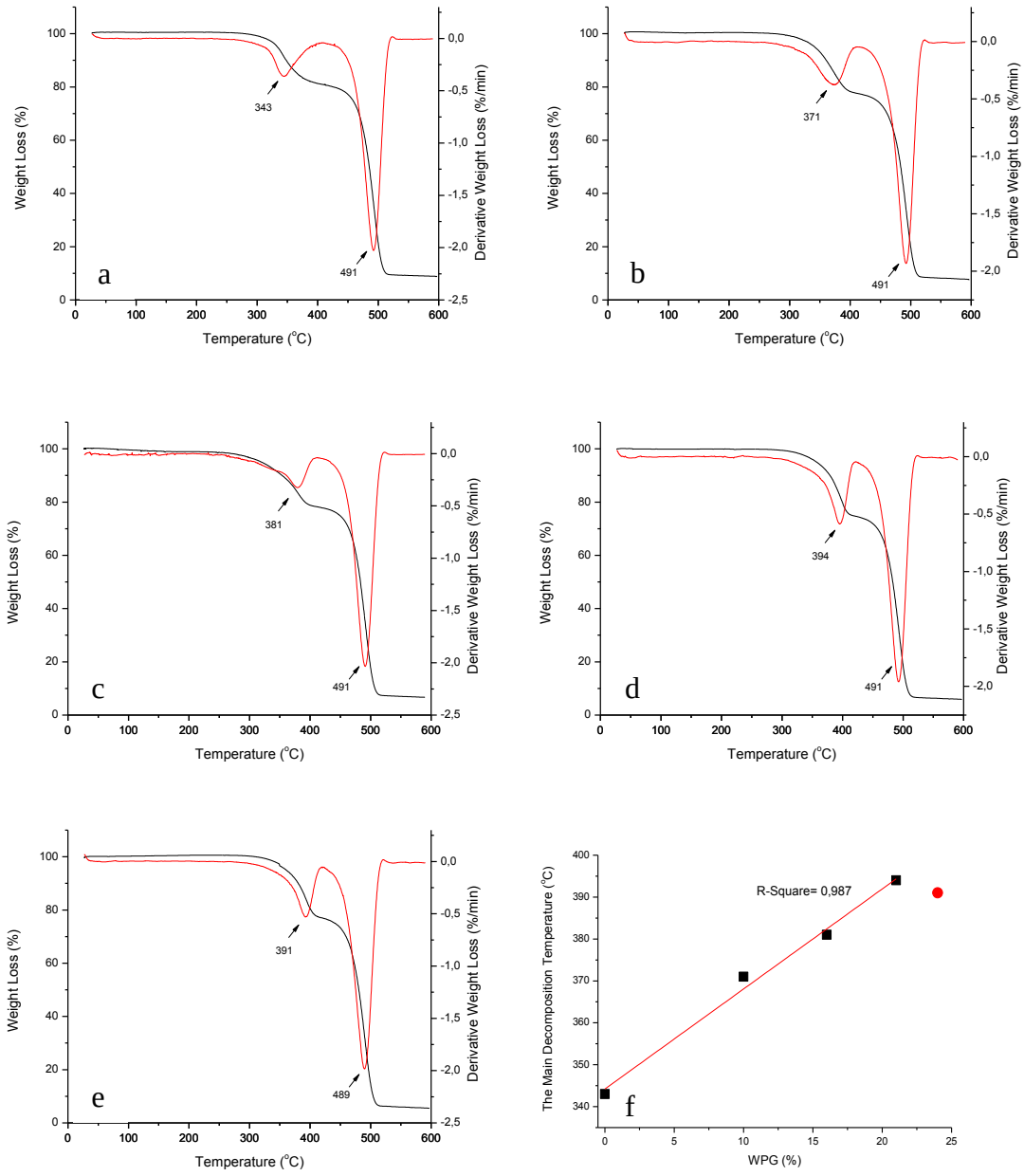

Fig. 7. TGA curves of (a) W, (b) VA_{10}, (c) VA_{16}, (d) VA_{21}, and (e) AA_{24} composites, (f) the effect of acetylation level on the main decomposition temperature of wood flour in HDPE matrix (squares: VA modification, circle: AA modification)

CONCLUSIONS

1. Scots pine wood flour samples were modified with acetic anhydride (AA) (24% WPG) and vinyl acetate (VA) at various WPG levels (10%, 16%, and 21% WPGs). AA- or VA-modified wood flour-reinforced HDPE composites showed higher tensile strength than unmodified wood flour-reinforced composites. Increasing the VA modification level improved the tensile strength and the flexural strength properties compared to neat and unmodified wood flour reinforced composites. With the increase of modification level with VA, tensile strength of the composites showed an

abrupt increase up to 25.1 MPa. Addition of 30 wt% of unmodified flour into the matrix significantly reduced the impact strength compared to neat composites. With modification of wood flour, there was no significant change in impact strength values of the composites compared with the unmodified wood flour.

2. SEM studies of the freeze-fractured surface of the composites gave evidence for the enhanced interfacial adhesion between the wood flour and the matrix on VA modification of wood flour. In the case of unmodified WPC (W), failure occurred at the weak wood flour/matrix interface, resulting in wood fiber pull-out, leaving voids and gaps on the surface. By contrast, in composites containing VA or AA modified wood flour, failure occurred at the wood flour due to strong adhesion between wood flour and matrix.
3. The VA modification showed a significant effect upon thermal properties of wood flour, making wood flour thermally more stable. The main decomposition temperature of acetylated wood was higher than that of unmodified wood flour. Increasing VA modification levels resulted in a linear increase in the main degradation temperatures and gradual decrease in the ash contents.

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