Impact of Stone Ground 'V-fines' Dispersion and Compatibilization on Polyethylene Wood Plastic Composites

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Recent studies have suggested that blocky mechanical pulp fines (CTMP fines) and fibrillar fines (SMC fines) have a negative impact on biocomposite modulus of rupture (MoR) in compression molded biocomposites. In addition, it was suggested that CTMP fines also have a negative impact on biocomposite modulus of elasticity (MoE). This study investigated whether these findings transfer to other types of cellulose fines material and injection molding. The effect of 'V-fines' addition to sawdust- and TMP-based biocomposites was analyzed, with respect to fines concentration, dispersing agent, and compatibilizers. The results indicated that the addition of 'V-fines' increased the stiffness (MoE) of all the analyzed compositions, while reducing the elongation at break. The addition of 'V-fines' reduced the tensile and flexural strength of TMP biocomposites, while it was largely unaffected for sawdust biocomposites. Flexural strength for neat 'V-fines' composites showed an increase that was proportional to the remaining pulp fibers composition. The addition of a dispersant agent to the 'V-fines' increased tensile strength, suggesting that an increased dispersion of the 'V-fines' can be achieved and is beneficial to the composite. The effects of the analyzed compatibilizer (polyethyleneoxide) was negligible, except for a small indication of increased MoE for fines / sawdust biocomposites.

INTRODUCTION

Recent studies have suggested that blocky mechanical pulp fines (TMP and CTMP fines) (Peltola *et al*. 2011a,b, 2014; Miettinen 2016; Sandquist *et al*. 2020) and fibrillar fines (SMC fines) (Sandquist *et al*. 2020) have a negative impact on biocomposite modulus of rupture (MoR) in compression molded biocomposites. In addition, it was suggested that CTMP fines also have a negative impact on biocomposite modulus of elasticity (MoE). The proposed underlying mechanisms include reduced aspect ratio, increased surface area, and poor dispersion or agglomeration of the fines material. Fines aggregate strongly when processed under aqueous conditions (Ek 2009). However, for optimal reinforcement potential in composites, the reinforcing fibers should ideally be well dispersed in the matrix (Peltola *et al*. 2014).

In modeling polymer matrix wood fiber composites, fines are often disregarded (Miettinen 2016; Miettinen *et al*. 2012, 2015; Newman *et al*. 2014), as they have limited or no contribution to reinforcement of strength or stiffness of the composite (Miettinen 2016). Additionally, to achieve reinforcement, the fiber or particle needs to possess a

minimal aspect ratio, which relates to a critical fiber length. Thumm and Dickson (2013) showed that the critical fiber length for radiata pine pulp reinforcement in biocomposites is approximately 0.8 mm in length, which results in an approximate aspect ratio of 25 (length/width). Softwood pulp fibers possess a fiber length that is longer (~3 mm) (Dinwoodie 2000) than the critical fiber length (Thumm and Dickson 2013) prior to compounding, whereas sawdust and wood flour does not (Stark and Berger 1997b).

Fekete *et al*. (2018) observed substantial physical reinforcement with low aspect ratio cellulose fibers in thermoplastic starch, as did Toriz *et al*. (2002) with lignin particles in polypropylene (PP). However, in PP, polylactide (PLA), and in polyethylene (PE) Sandquist *et al*. (2020), Peltola *et al*. (2011a, 2014), and Gallagher and McDonald (2013), respectively, showed negative strength reinforcement of composites with very small wood particles or fines.

Previous studies on wood flour has indicated that the particle size distribution has a significant influence on the mechanical properties of wood plastic composites (Stark and Berger 1997a,b; Stark and Rowlands 2003). Wood flour content up to 40% by weight yielded an increase in physical properties, after which properties start to plateau (Stark and Berger 1997a). Smaller diameter wood flour particles gave an higher increase in strength, elongation, and stiffness compared to larger particles (Stark and Berger 1997b), especially comparing 60 µm with 500 µm fractions.

In sawdust-based wood-plastic composites (WPC), Islam and Islam (2011) showed that cetyltrimethylammonium bromide (CTAB)-modified SD improved almost all mechanical properties of PE-based biocomposites in the range of 20 to 35 wt% SD. This result indicates that there is significant room for improved compatibilization between SD and PE. Similarly, Godard *et al*. (2009) showed a significant increase in both strength and stiffness with the addition of MA-PE compatibilization agent to 40 wt% containing SD / PE composites. The highest effect was seen for fine SD particles (length average 0.57 mm), but also significant for intermediate (1.48 mm) and coarse SD particles (1.78 mm). Hillig *et al*. (2008) reported similar findings.

Viksne *et al*. (2004) analyzed the impact of a paraffin and a maleic anhydride (MA) containing modification on SD/PE composite. The paraffin modification significantly improved dispersion, with a subsequent improvement in the strength of the composite. Oppositely, the MA containing modifier improved the stiffness, but less prominently the strength, which corresponds to an increase in compatibilization.

The aims of this study were to investigate the effect of fines material addition to SD- and TMP-based biocomposites and to study what effect the addition of a dispersing agent and a compatibilizer may have on fines material in the SD ad TMP composites. The selected dispersion agent Arosurf PA 780V is a commercially blend of ionic and non-ionic surfactants used in tissue paper production. The selected compatibilization agent was polyethyleneoxide (POE). Lu *et al*. (2005) showed that POE is a less effective compatibilizer than a MA-PE based option. However, POE was selected because it is not chemically reactive, and it was feared that premature reactivity would limit dispersion and obscure the interpretation of the results. Other advantages of POE are that it is a thermoplastic, water soluble additive, easy to apply on cellulose fibre surface to provide improved fibre dispersion, and it is compatible with several thermoplastic polymers.

EXPERIMENTAL

Materials and Methods

Saw dust

The utilized wood sawdust (SD) was from a commercial lumber production facility (local sawmill, Southern Finland), and was predominantly made up of short shives (bundles of fibers) from Scots pine (*Pinus sylvestris*), with an average particle size of 450 μm and presented in Fig. 1. The fines material concentration of material that passed through a 200-mesh sieve was negligible.

Thermomechanical pulp

Commercial thermomechanical pulp (TMP) produced from spruce (*Picea abies*) from a Finish producer was utilized in the trial. The fines material content was stated by the suppler to be approximately 20% by weight.

Grinding stone preparation

The grinding stone for the custom laboratory grinder was a Norton A601 (Saint-Gobain Abrasives Canada Inc, Hamilton, Ontario, Canada) with a diameter of 300 mm, and was first milled flat, removing the original stone pattern. On the flat surface, a new profile was milled with 15º-angle chamfers as shown in Fig. 1.

Fig. 1. Dimensions of angles (the axis are not at the same scale in the figure) Separator.

Production of V-fines

Production of V-fines was made according to process described in Nurminen *et al.* (2018). The wood raw material used was never dried Spruce (*Picea abies*). Dry matter content of the wood was on average 44.8%. Wood logs were cut to blocks for grinding. Care was taken to utilize wood materials that was as similar as possible in appearance.

The previously described custom-built laboratory grinder (Nurminen *et al.* 2018) used a grinding area of 35 mm in both length and in width. Peripheral speed of the stone was 20 m/s. The shower water temperature varied from 60 °C to 70 °C, and the wood feeding rate was 0.5 mm/s. The pulp was screened through 4 mm hole sieve to remove larger, unground particles.

The fines concentration after grinding was low, approximately 1.2%. To increase the concentration, the suspension was concentrated *via* a WESTFALIA TSC 6-01-576 (GEA Westfalia Separator GmbH, Oelde, Germany) separator. The rotation speed was 12000 rpm; the outlet interval was 100 to 540 s and was increased while the consistency decreased. The feeding was 300 L/h, and the water phase was returned to feeding container.

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The original dry mater content of the V-fines suspension was 1.2%, the total volume of fines suspension processed for concentration was approximately 700 L, with a final dry matter content of the V-fines slurry of 4.5 to 5.3%.

V-fine treatment with debonder agent

The commercial debonder Arosurf PA 780V (Evonik Industries, Essen, Germany) was used as received and according to the manufacturer's instructions. Arosurf PA 780V was diluted with water, added into the concentrated V-fines slurry (consistency 2.5 to 3%), and stirred for 1 h at room temperature followed by spray drying. The debonder content after mixing was 1% by weight of the slurry. The Arosurf PA 780V debonder is a commercial fluff pulp debonder consisting of a blend of non-ionic and cationic compounds.

V-fine treatment with compatibilization agent

The non-chemically reactive compatibilizer polyethyleneoxide (PEO) (Polyox™ WSR N750, DOW Chemical Company, Texas City, USA) was mixed into water with Ystral disperser (Ystral GmbH, Ballrechten-Dottingen, Germany) and then mixed with the V-fines slurry, followed by spray drying. The amount of PEO was 17% of the weight of dry fines. It was assumed that a high amount of PEO in slurry will cover fines particles at least partially during drying and at the high temperature in extruder PEO melts and particles separates from aggregates at high shear of extruder.

Spray drying of V-fines and modified V-fines

Spray-drying of V-fines was performed at VTT Rajamäki with NIRO Spray Dryer P-6.3 (GEA NIRO A/S, Soeborg, Denmark). The fines slurry was fed on the atomizer at the consistency of 2.5% to 3%. The temperature of the coming airflow was 200 °C, which increased the temperature of the product to 85 to 90 °C. Feeding speed of slurry was 25.8 kg/h. The energy consumption was very high, 49.8 MWh/t, due to the large amount of water being vaporize.

The final product was a yellowish free-flowing flour (Fig. 2). The larger particles in Fig. 2 were weak agglomerates that formed when the flour was exposed to air moisture. When kept under dry conditions, these agglomerates were not present.

Fig. 2. Fines after spray-drying

Mixing of V-fines with sawdust or TMP

Both virgin and modified V-fines were dry-blended with SD and TMP *via* bag mixing 5 min prior to compounding.

Polymer matrix

The polymer used for the preparation of all the composite materials were Braskems bio-based polyethylene HDPE SHA7260 (Braskem, São Paulo, Brazil). This polymer is derived from sugar cane saccharose.

Composite compounding

The compounding of materials to total fibre content 40% in PE were made using co-rotating Berstorff ZE 25x33D compounder (Besrtorff GmbH, Hanover, Germany) and injection moulded to standard (ISO 527-2 2012) dog bone shaped test pieces with Engel ES 200/50 HL (Engel Maschinenbau GmbH, Schwefberg, Austria) injection moulding machine.

Mechanical testing

Tensile testing was performed according to ISO 527 and flexural test according to ISO 178 (2019) using Instron 4505 mechanical test equipment (Instron Corp., Canton, MA, USA). Samples were dog bone-shaped standard samples (ISO 527-2 (2012)), and the results were an average of minimum five replicates.

Charpy impact strength was tested according to ISO 179-2 (1997) using unnotched samples flatwise and a Charpy Ceast Resil 5.5 Impact Strength Machine (CEAST S.p.a. Torino, Italy). The sample size was 4 mm x 10 mm x 100 mm, and the average of 10 samples was utilized for further analysis. All the tested samples were conditioned in 23 °C and 50% relative moisture for minimum five days before testing.

Microscopic imaging for composites

The morphology of fibres and injection-moulded samples was studied by scanning electron microscopy (SEM). The sample surface was coated with gold particles. In injection moulded samples the scanning was made on cross-cut surfaces. Samples were observed on a Jeol JSM T100 with a voltage of 25 kV (Tokyo, Japan).

Microscopic imaging of V-fines

Liquid samples were suspended in water in order to improve the visibility of sample details by diluting. Samples were spread on a microscopy slide and covered with a cover slip. Samples were imaged using confocal laser scanning microscopy (CLSM) equipment consisting of a Zeiss LSM 710 attached to an Axio Imager Z microscope (Zeiss, Jena, Germany).

Each sample view was imaged without staining with transmission mode and utilising the autofluorescence properties of the sample structures. For the latter, a diode laser line of 405 nm was used for excitation, and emission was collected at 424 to 540 nm. Images were assembled of the optical sections taken using 10x, 20x and 40x objectives (Zeiss EC Epiplan-Neofluar, Cambridge, UK) and ZEN software (Zeiss, Jena, Germany).

RESULTS AND DISCUSSION

Particle Size Estimate of V-fines

With a grinding stone profile of angle of 15°, the amount of material that did not pass through a 200 mesh was initially 5%, but it increased and stabilized at 9%.
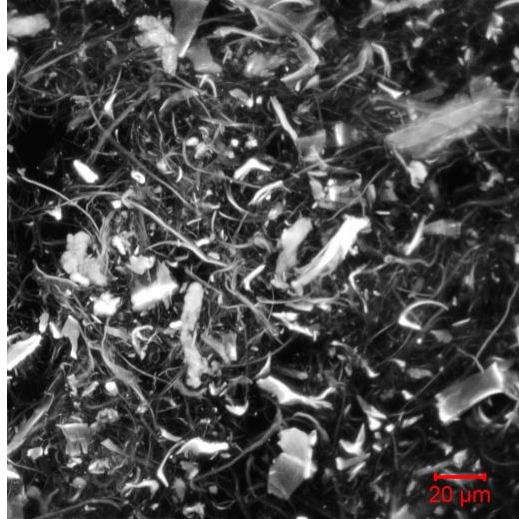

Fig. 3. CLSM microscopy autofluorescence image of V-fines particles, to show the dispersion of the fines material in the matrix. The matrix is dark as it does not possess autofluorescence.
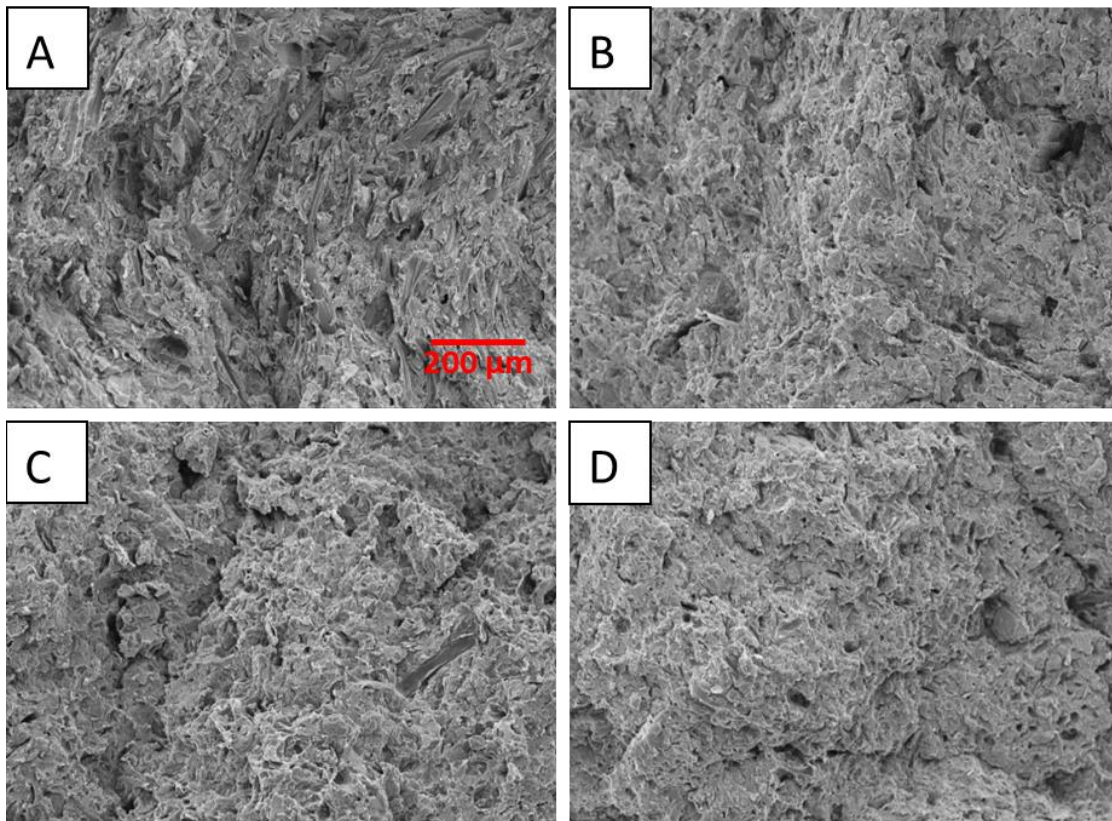

Fig. 4. SEM-pictures of injection molded PE-40% fibre composites fracture surfaces, 100x enlargedt. A) TMP, B) V-fine wood ref., C) Arosurf 780V mod. V-fines, D) PEO modified V-fines

The materials from both runs was mixed, and in the final mixture the amount of material that did not pass through a 200-mesh size was approximately 7%. A representative overview of the fines structures are shown in Fig. 3.

Dispersion and compatibilization of the cellulose material in the composite matrix

Wood material, fibers, and composite samples were examined with SEM to estimate the dispersion of the cellulose material and to investigate the interface between cellulose and matrix. As shown in Fig. 5A-D, the cellulose materials of TMP and V-fines were well dispersed in the matrix, with no apparent agglomeration. In the V-fines samples, some fibers were still present (approximately 7 wt%), as apparent in Fig. 4B-D.

Peltola *et al*. (2014, 2019) showed that for PP and PLA in particular, the compatibility between matrix and TMP is high. They postulated that this is related to the surface lignin on these fibers and the interaction with the OH-groups in the polymer matrix. However, PE lacks OH-functionality and is significantly more hydrophobic than PP or PLA.

As shown in Fig. 5A-B, uncompatibilized TMP and V-fines showed comparatively less strong interfacial bonding (arrows) compared with the compatibilized V-fines in Fig. 5C-D. This supports the earlier reports of improved compatibilization of PEO with cellulose and a PE matrix (Lu *et al*. 2005).
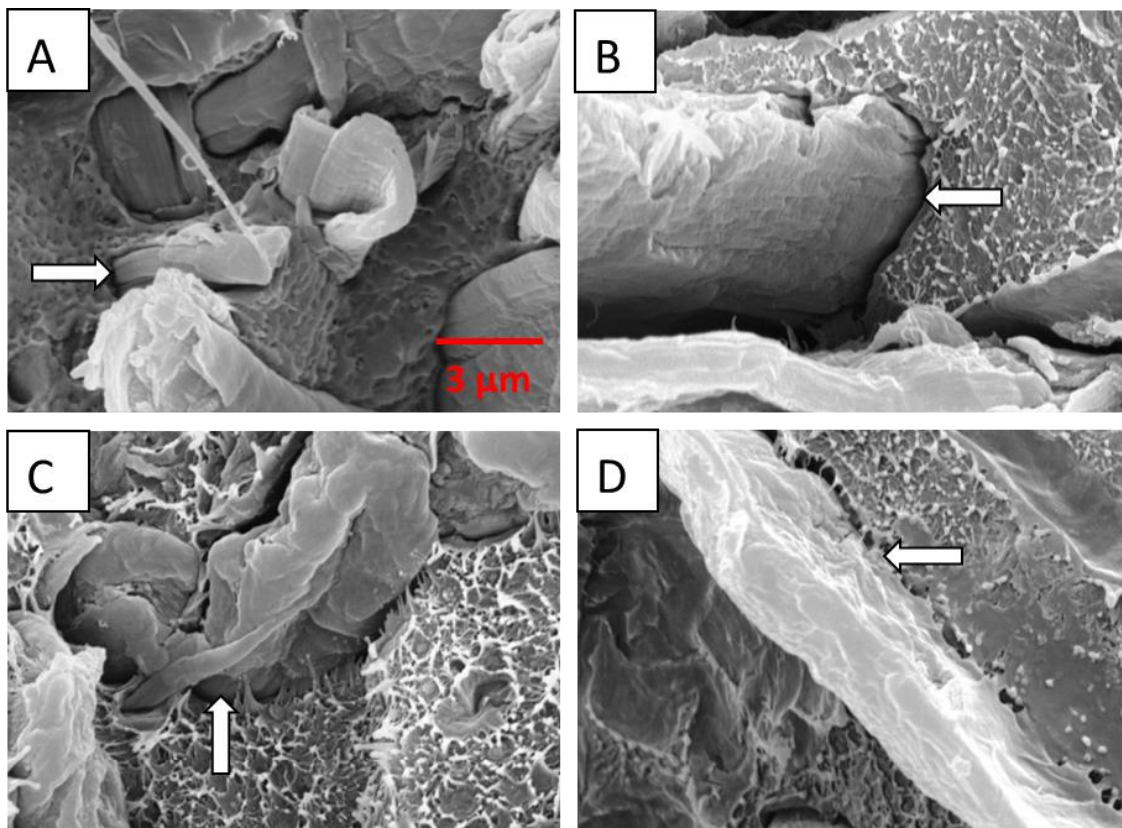

Fig. 5. SEM-pictures of injection moulded PE-40% fibre composites fracture surfaces with 7500x enlargement. A) TMP, B) V-fines, C) Arosurf 780V modified V-fines, D) PEO modified V-fines

Comparison of TMP, SD and V-fines composites

Figures 6 through 8 summarize the mechanical testing results of all the composite combinations. Overall, there was a large agreement between the tensional (Fig. 6) and flexural (Fig. 7) results.

Focusing on the TMP, SD, and V-fines results without any dispersing agent or compatibilizer, several interesting inferences can be drawn. Firstly, the addition of uncompatibilized SD to the HD-PE matrix increased the tensile and flexural strength and stiffness, while reducing impact strength, in agreement to earlier reports (Hillig *et al.* 2008; Godard *et al.* 2009; Islam and Islam 2011). The addition of TMP to the HD-PE increased the mechanical properties of the composite material, also in agreement with earlier reports (Mertens *et al.* 2017). The addition of V-fines also increased both the strength and stiffness of the composite compared to neat PE. It needs to be reiterated that the V-fines contained approximately 7 wt% material that does not pass through a 200 mesh and is mainly made up of fibers.

As a simplified hypothesis, assuming 80 wt% fiber content of the TMP pulp as stated by the producer; the tensile strength of a 100 wt-% TMP fiber composite was estimated at $(28.4/0.8) = 35.5$ MPa. The recorded tensile strength of the pure matrix was 12.3 MPa. Utilizing a linear regression, a composite containing 7 wt% fibers would be estimated to show a strength of $(12.3 + (35.5-12.3) * 0.07) = 13.92$ MPa. This shows a strong agreement with the recorded tensile strength of the fines reference composite at 13.90 MPa.
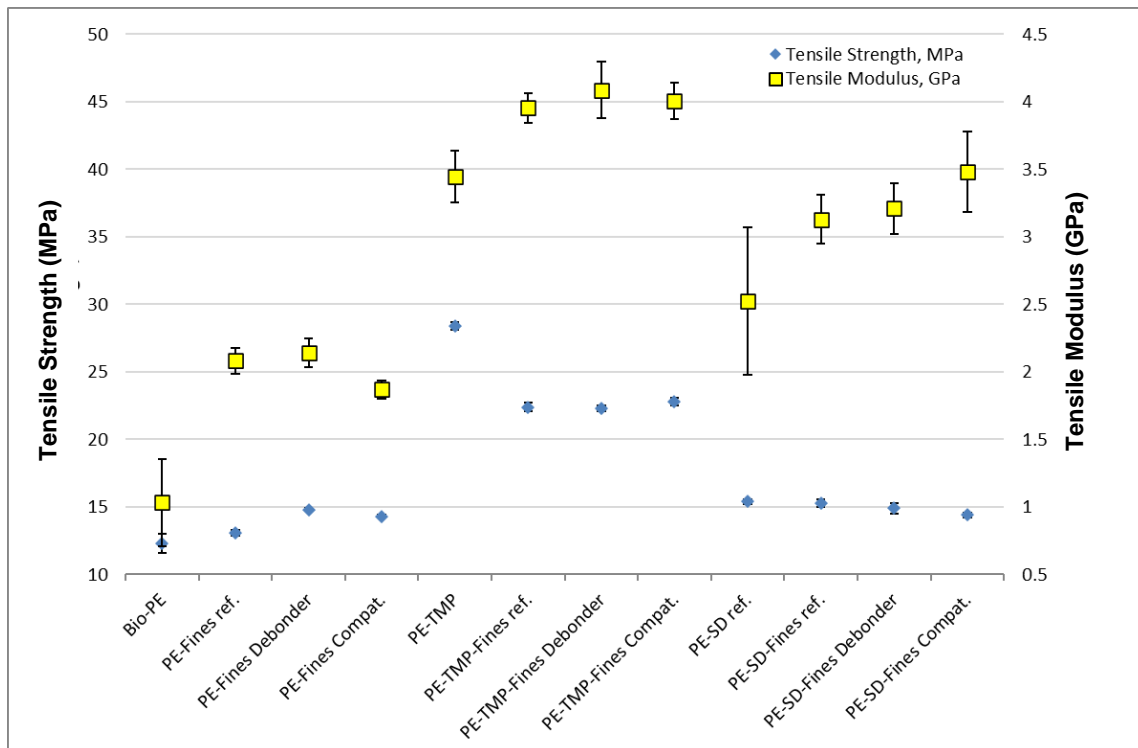

Fig. 6. Summary of tensile tests results for reference PE, 40/60 fines/PE, 40/60 TMP/PE and 40/60 SD/PE composites. Fines refers to V-fines, and was consistently added at a 5% concentration, independent of modification, to the TMP and SD composites. Abbreviations: Compat. Refers to the POE Compatibilizer Polyox WSR N750; Debonder refers to the Arosurf PA 780V debonder.
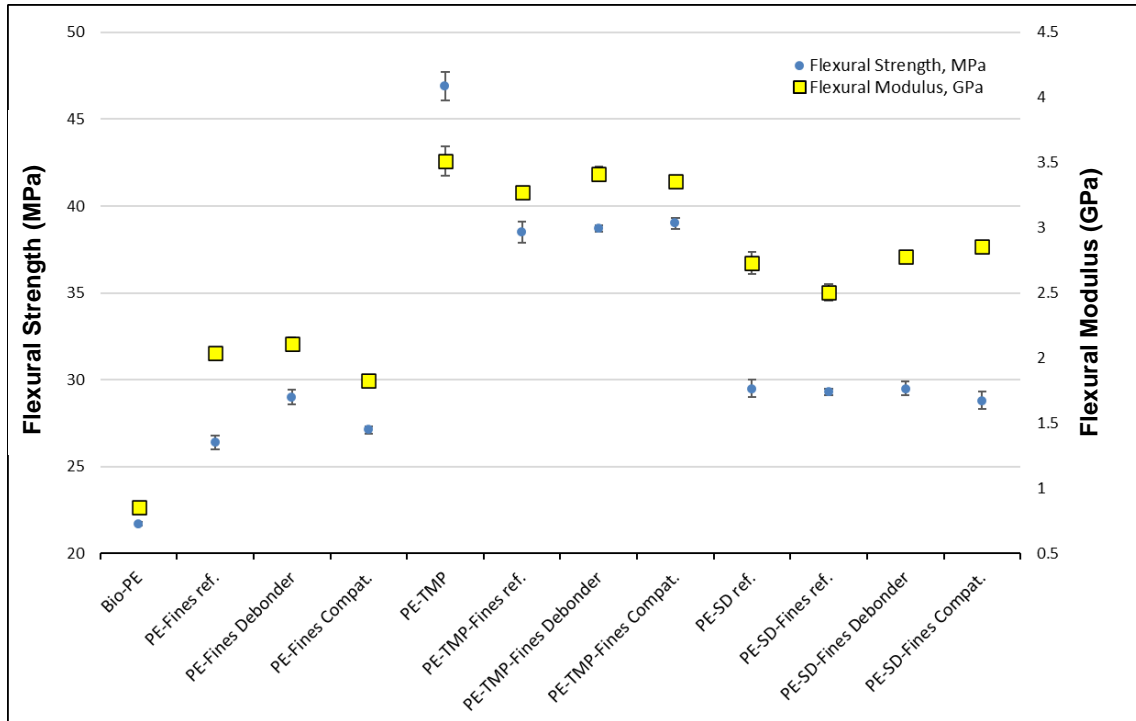
Fig. 7. Summary of flexural tests results for reference PE, 40/60 fines/PE, 40/60 TMP/PE and 40/60 SD/PE composites. Fines refers to V-fines, and was consistently added at a 5% concentration, independent of modification, to the TMP and SD composites. Abbreviations: Compat. Refers to the POE Compatibilizer Polyox WSR N750; Debonder refers to the Arosurf PA 780V debonder.

Fig. 8. Summary of Charpy impact strength tests results for reference PE, 40/60 V-fines/PE, 40/60 TMP/PE and 40/60 SD/PE composites. Fines refers to V-fines, and was consistently added at a 5% concentration, independent of modification, to the TMP and SD composites. Abbreviations: Compat. Refers to the POE Compatibilizer Polyox WSR N750; Debonder refers to the Arosurf PA 780V debonder.
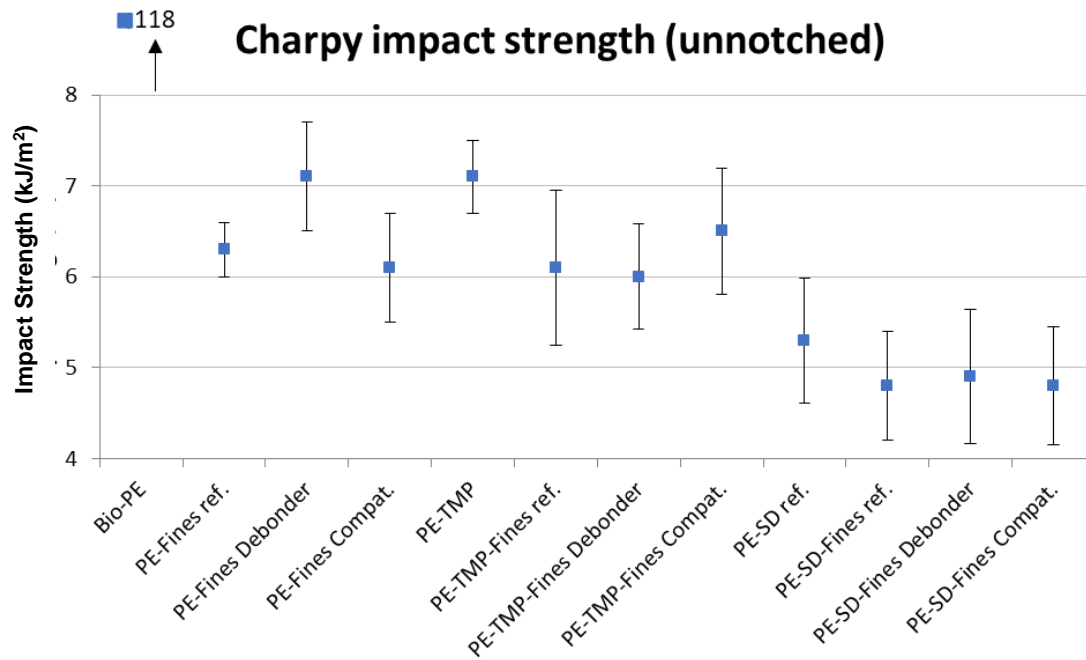
Effect of dispersant agent or compatibilizer to V-fines

The addition of a dispersing agent and compatibilization agent had only moderate effects on the mechanical properties of the composite material. Of note, in both tension and flexural properties there was a small increase in strength, which supports the postulation that the surfactant dispersion agent increases the dispersion of the fines material in the matrix. This is supported by a small, but not statistically significant, increase in impact strength.

Secondly, the addition of PEO compatibilization agent resulted in a small drop in stiffness for both tensional and flexural mechanical properties. A similar decrease was observed in impact strength. This points either towards an increased agglomeration of the fines material, or alternatively an overall reduced interfacial strength. This result is unexpected and counter-intuitive, as the PEO is a non-reactive compatibilization agent. Until a more complete analysis can be performed, this result was attributed to an increase in agglomeration of the fines prior to compounding.

Effect of addition of V-fines to TMP and SD composites

Focusing first on the unmodified V-fines, there was a clear effect in both TMP and SD composites in both tensile and flexural properties. For the TMP-based composites, there was a drop in strength, which was not recovered through any of the modifications. This drop was larger than what would be expected from a rule of mixture decrease of fibres. Simultaneously, there was a large increase in tensional stiffness (elastic modulus), which was not seen in the flexural results. There was a corresponding reduction in impact strength. Flexural testing of materials is a combination of a material's basic tensile, compressive, and shear properties. The interpretation of these results is that the introduction of the fines materials affects the shear and compression properties, alongside an overall reduction in strength and stiffness. The authors have not found any explanation for this behaviour, and postulate that a potential mechanism could be that the fines are agglomerating on the fiber surfaces, creating a 'slip layer'. The recorded increased increase in tensional stiffness may be due to increased surface area in total exposed to the polymer matrix.

For the SD-based composites, the effect was less dramatic than for the TMP composites. As there was less strength reinforcement from the SD particles, no real loss was recorded with the addition of V-fines. There was an indication of similar loss of strength and impact resistance as recorded for the TMP composites. Similarly, this result was interpreted as the increase in tensile stiffness for an increase in total surface area with the addition of V-fines, which is supported by the recorded increase in tensional stiffness.

CONCLUSIONS

1. The addition of a dispersant agent to V-fines increased mechanical properties of the wood-polyethylene composites, indicating that a better dispersion can be achieved.

2. Polyethyleneoxide (POE) provided little or no compatibilization at the analysed levels in a polyethylene-based wood plastic composite.

3. The addition of V-fines to a thermomechanical pulp (TMP)-based wood plastic composite reduced all mechanical properties apart from tensional stiffness. This result may be related to an increase in cellulose surface area.

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4. Similarly, the addition of V-fines to a saw dust (SD)-based wood plastic composite reduced most mechanical properties, while increasing tensional stiffness. This effect may be related to an increase in cellulose surface area.

ACKNOWLEDGMENTS

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Appendix

Table S1. Tensile Strength Results for Injection Moulded PE Composites with Total Fibre Content of 40%

Sample	Tensile strength at yield		Tensile strength at break		Modulus (Auto Young)		Elongation at break	
	MPa	s.d.	MPa	s.d.	MPa	s.d.	%	s.d.
Bio-PE	18.9	0.2	12.3	0.7	1033	325	103	85
PE-Fines ref.	13.9	0.1	13.1	0.2	2084	96	3.4	0.2
PE-Fines DA1	15.2	0.1	14.8	0.1	2143	107	3.3	0.2
PE-Fines PEO	14.7	0	14.3	0.1	1869	70	3.1	0.5
PE-TMP	28.6	0.4	28.4	0.3	3446	193	1.7	0.1
PE-TMP-Fines ref.	22.6	0.3	22.4	0.3	3957	110	1.5	0.1
PE-TMP-Fines DA1	22.4	0.2	22.3	0.2	4086	210	1.5	0.1
PE-TMP-Fines PEO	22.9	0.3	22.8	0.3	4008	136	1.5	0.1
PE-SD ref.	15.4	0.2	15.4	0.2	2525	545	2	0.2
PE-SD-Fines ref.	16	0.1	15.3	0.3	3130	183	2.2	0.2
PE-SD-Fines DA1	15.6	0	14.9	0.4	3210	189	2.2	0.2
PE-SD-Fines PEO	15.2	0.1	14.4	0.2	3482	297	2	0.2

Table S2. Flexural Strength Results for Injection Moulded PE Composites with Total Fibre Content of 40%

Sample	Flexural strength at yield		Modulus (Auto Young)		Elongation		Flexural load at yield	
	MPa	s.d.	MPa	s.d.	%	s.d.	N	s.d.
Bio-PE	21.7	0.1	855	11	6.2	0.1	36.6	0.1
PE-Fines ref.	26.4	0.4	2041	39	4	0.2	44.6	0.2
PE-Fines DA1	29	0.4	2111	49	4	0.1	48.2	0.1
PE-Fines PEO	27.1	0.2	1825	32	4.1	0.1	45.7	0.2
PE-TMP	46.9	0.8	3512	115	2.6	0.1	76.5	0.6
PE-TMP-Fines ref.	38.5	0.6	3269	34	2.5	0.1	66.3	0.9
PE-TMP-Fines DA1	38.7	0.2	3416	52	2.4	0.1	66.4	0.5
PE-TMP-Fines PEO	39	0.3	3354	16	2.6	0.1	67.1	0.7
PE-SD ref.	29.5	0.5	2729	84	3.3	0.2	53.4	0.9
PE-SD-Fines ref.	29.3	0.2	2501	64	3.9	0.1	51.1	0.3
PE-SD-Fines DA1	29.5	0.4	2782	10	3.6	0.2	51.6	0.6
PE-SD-Fines PEO	28.8	0.5	2854	40	3.3	0.1	49	0.8

Table S3. Charpy Impact Strength (Unnotched) Results for Injection Moulded PE Composites with Total Fibre Content of 40%

	Charpy impact strength	
Sample	kJ/m^2	s.d.
Bio-PE	118	15
PE-Fines ref.	6.3	0.3
PE-Fines DA1	7.1	0.6
PE-Fines PEO	6.1	0.6
PE-TMP	7.1	0.4
PE-TMP-Fines ref.	6.1	0.85
PE-TMP-Fines DA1	6	0.58
PE-TMP-Fines PEO	6.5	0.69
PE-SD ref.	5.3	0.69
PE-SD-Fines ref.	4.8	0.6
PE-SD-Fines DA1	4.9	0.74
PE-SD-Fines PEO	4.8	0.65