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What is This?

Low combustible polypropylene/flax/ magnesium hydroxide composites: mechanical, flame retardation characterization and recycling effect

Ahmed El-sabbagh[1,2], Leif Steuernagel[1] and Gerhard Ziegmann[1]

Abstract
Natural fibre-reinforced polymer composites are nowadays receiving considerable attention, especially in the automobile and civil engineering industries. Reduction in flammability characteristics besides retention of their excellent mechanical properties will ensure wider market for these composites. Polypropylene matrix was reinforced with 30 and 50 wt% of flax fibres. Magnesium hydroxide was added at different levels of 0, 20 and 30 wt% in order to investigate the flammability resistance behaviour. Thermal and thermomechanical tests were carried out in order to evaluate new and recycled composites. Limited oxygen index was improved to more than 27 with 50 wt% flax/30% magnesium hydroxide, although very slow burning rate in the horizontal test condition was achieved with 30/30, 50/20 and 50/30 wt% for flax/magnesium hydroxides. Regarding the UL94 test, no sample passed vertical burn test. Recycling of these composites was investigated and showed 4 and 10% decrease in strength and stiffness, respectively. Recycling experiments on 50/30 system showed an effect of 5% limited oxygen index decrease after reprocessing for two cycles.

Keywords
Natural fibre composites, flax, magnesium hydroxide, flame retardance

Introduction

Finding new applications for natural fibres-reinforced polymers (NFRP) in the international market is increasingly becoming attractive due to the following factors: fibre price, availability[1,2] and high strength to weight ratios.[3,4] Besides, the current polymer production machines such as the extruder and the injection moulding machine are capable of handling these composites. The negative impact on the environment diminishes when compared with products made of glass and carbon fibres. The application of NFRP composites is mostly concentrated in the automobile industry and the civil engineering sectors.[2] However, NFRP composites will still look for technical solutions to problems that hinder the growth of their markets, namely, the limited window of the processing temperature for natural fibres, inconsistent mechanical properties[5] and hydrophilic nature of the fibre, which prevents perfect adhesion with the polymer matrix on one side[6] and from the other side, increases its affinity to water

absorption that results in dimensional instability.[5–7] Bacterial degradation[4] also presents a threatening problem.

Motivation and literature review

One of the most urgent problems, especially for the construction industry, is the insufficient data about the sensitivity of these composites to flame.[8] Even Brosius[9] reported that the use of NFRP has been excluded from most aircraft, rail and subway markets

[1]Institute of Polymer Materials & Plastics Engineering, Clausthal University of Technology, Germany
[2]Faculty of Engineering, Ain-Shams University, Egypt

Corresponding author:
Ahmed El-sabbagh, Institute of Polymer Materials & Plastics Engineering, Clausthal University of Technology, Agricola str. 6, D-38678, Germany.
Email: ahmed.sabbagh@tu-clausthal.de

until the issues of flame, smoke and toxicity are resolved. The use of these composites requires that they should pass regulatory safety tests such as IEC regulations for electrical appliances, ASTM E 108, UL 1256, ASTM D635-98 [UL94], EN 1187 for building, roofing and decorative materials as well as DIN 5510 for low toxic material in the confined areas, especially in transport applications.

The flammability behaviour of NFRP is governed by the polymeric matrix and the fibre type. This study deals with the degradation of thermoplastics, especially polypropylene (PP). PP with its aliphatic structure degrades normally by random scission due to the presence of hydrogen atoms on all carbon atoms,[10] hence resulting in flammability. Compatibilsers, known as maleic anhydride-grafted-polypropylene (MAPP), are normally used for linking PP and the natural fibres. MAPP affects the flame resistance positively, where the cyclic anhydride structures are prone to slow down the hydrolysis and thus result in intramolecular cross-linking of the polymer chains.[11]

Furthermore, the flammability of natural fibre is governed by a number of factors, namely fibre constituents (lignin, cellulose, hemicellulose, pectin, waxes, etc), the combustion characterisctics of the released organic volatiles during decomposition, degree of cellulose polymerisation and fibre orientation.[12] Lignin releases less volatiles and thermally decomposes at a temperature greater than those of hemicellulose and cellulose but it starts to decompose normally at earlier temperatures (i.e. 160°C for flax).[12] Higher amount of lignin lowers fibre decomposition temperature, as this is attributed to the decomposition of relatively weak bonds.[13] According to Manfredi et al.,[14] lower content of lignin is desirable in order to delay the ignition start time. However, the aromatic rings present in lignin resist fibre oxidation. Cellulose is also considered to be more flammable because it produces volatile flammables 'Levoglucosan,'[13,15] which consequently yield flammable and non-flammable volatiles and gases, tar and char. An increase in crystallinity results in more levoglucosan during pyrolysis, which requires more activation energy to imitate pyrolysis.[16] Finally, more fibre orientation results, assumedly, in less oxygen permeability in fibres and hence flammability resistance is improved.[17]

Therefore, flame retardant (FR) materials are necessary in order for them to be suitable for a wider range of applications. However, it should be considered that the implementation of such retardants might result in negative aspects that might compromise the mechanical properties.[8] Flame retardance and mechanical behaviour are necessary for composites. Flame retardance is revealed to be more effective if it can stop the ignition process during one of its steps, namely heating,

decomposition, ignition, combustion and propagation.[8,15,18] Kozlowski and Wladyka-Przybylak[19] reported that this objective can be achieved by impregnation of the fibres with FR before compounding, or by implementation of FR during manufacturing processes or by insulation of the composite product in the finishing stages with intumescent materials in order to prevent the penetration of heat.

Flame retardance, in general, can be employed either by chemical or physical mode of actions.[20] Physical mode flame retardation mechanism takes place by cooling resulting from the endothermic reactions accompanying the degradation of metallic hydroxides such as aluminium trihydrate or magnesium hydroxide $(Mg(OH)_2)$ due to its relatively higher thermal stability. The physical mode of action can also take place by the formation of a protective layer when the additive FR forms a shield with low thermal conductivity upon exposure to heat flux, i.e. phosphorous additive.[20] It is reported[20] that almost 75% of the flame retardancy industry for polyolefins uses the metallic hydroxide technique. This is attributed to the possibility of reaching the V-0 rating in UL94V test. Not only this, the smoke emission during burning is also reduced. Interestingly, it is reported[8] that the use of boric acid or zinc borate will have a synergetic effect with $Mg(OH)_2$ that can result in an excellent behaviour.[8] However, lower limited oxygen index (LOI) and horizontal flame speed are obtained in comparison with the $Mg(OH)_2$-treated composite without any other additives. Also, the tensile and the flexural properties attained by the fibres treated with $Mg(OH)_2$ are reduced by almost 15%.[8]

Finally, the recycling of these novel brands of composites presents a great question. Thus, will they retain their mechanical properties[21,22] and their flame resistance? Moran et al.[22] pointed out that the stiffness as well as the strength are increased. This was attributed to better dispersion of fibres after multi extrusion cycles despite the chain scission (melt flow, apparent viscosity). Hence, the impact toughness deteriorates. Moran et al.[22] concluded that more than three cycles could have remarkably negative effects on the overall performance of PP loaded with 20% flax. On the contrary, Duigou et al.[21] found that repeated injection moulding caused only a slight improvement of stiffness and about 30% drop in strength for the 30% flax was obtained. It was concluded that deteriorating behaviour was not noticed with regard to the rheology and glass transition temperature of the composites. However, few studies such as those of Duigou et al.[21] and Moran et al.[22] have been published regarding the effect of recycling on flame resistance.

This study aims to investigate the application of FR in one of the most widely produced natural fibre

Table 1. Composition and physical properties of the supplied flax.

Constituent	Cellulose	Hemicellulose	Pectin/lignin	Fats and waxes	Proteins/ash/minerals	Moisture
Composition (wt%)	65	17	4	1	4	12

thermoplastic composite systems, which is flax/PP. Flax is known for its high cellulosic content, high strength, low cost, low density and recyclability.[5–7,23] For flame retardance, only $Mg(OH)_2$ is considered in this study. Other types of FR are not considered, as published in previous literature.[24,25] The following points, namely the fibre content, FR content and number of injection cycles are considered in this study.

Experimental work

Retted flax fibres were supplied in bundle form by Sachsenleinen from Germany. The composition of flax fibres is shown in Table 1. Homopolymer PP developed for thin-wall high-speed injection moulding is used as the host matrix and is supplied by DOW GmbH (MFR = 52 at 230°C/2.16 kg and specific density of 0.9). MAPP and $Mg(OH)_2$ are supplied by Sigma-Aldrich. $Mg(OH)_2$, the FR powder CAS number 1309-42-8, is a purum p.a. grade.

Flax fibres were treated, as shown in the study of El-sabbagh et al.[26] by 5% NaOH solution for 1 h, then washed and dried overnight at 100°C in an oven. This alkalinisation process aims to remove possible impurities and increase the available area for coupling with the PP matrix. Fibres were manually cut to short fibres of on average 40 mm, then dried at 80°C for 2 h. Kneading takes place in a Haake kneader or roller mixer in a small mixing chamber of 50 cm³. Kneading proceeds as follows: PP was mixed with MAPP, $Mg(OH)_2$ and short flax fibres. To determine the appropriate kneading temperature and speed, preliminary experiments were performed in order to obtain homogeneous distribution of fibres within the matrix. Kneading speed was 50 rpm and was maintained for 15 min at 170°C. Then the speed was increased to 100 rpm for another 10 min. The stepped up speed was intended to help in breaking up the agglomerated fibres. The compound was cooled down in air and visually checked for any agglomerate formed during processing. The compound was shredded and dried overnight at 80°C before injection moulding. Figure 1(a) shows how the compound looks after kneading and whether it is homogeneous, Figure 1(b), or inhomogeneous, Figure 1(c). Different parameters, namely the fibre content at 30 and 50 wt% and the FR content at 0, 20 and 30 wt% were investigated in this study. Only six samples were compounded. Table 2

provides the data of the six experiments that were conducted and the composition corresponding to each one. Each sample is denoted with the governing percentages of fibre and FR. After shredding of the compound, the resulting granulates were injection-moulded at a temperature profile of 185–190–195–200°C using Allrounder 220C, Arburg, Lossburg, Germany. The injection-moulded samples were partially divided, where some of the samples were shredded, dried and re-injected once and twice, as shown in Table 2. Therefore, each sample is denoted by 0, 1 and 2 after the sample name to describe the number of cycles carried on the composites under study.

Samples of mechanical testing were conditioned at 23°C and 50% relative humidity for at least 88 h, according to ISO 291. Tension tests were made using Zwick 0.25 t tensile machine, according to DIN EN ISO 527-1. Test sample was injection-molded according to ISO 527-2. Test was conducted and evaluated according to ISO 527-1. Dynamic mechanical analysis (DMA), using Q 800 was done by TA instruments, was carried out on $17.5 \times 10 \times 3\ mm^3$ samples in order to determine the change in the composite characteristics (storage modulus and damping factor) at different fibre loadings and FR content. The cantilver loading was set at 1, 10 and 100 Hz frequency and 0.1% strain in temperature range of 40–160°C and heating rate of 2°C/min. Storage modulus (E′), loss modulus (E″) and mechanical loss factor (tan δ) were measured during the test and plotted as functions of temperature. Thermal gravimetric analysis (TGA) was carried up to 500°C at 10°C/min heating rate in non-inert gas atmosphere in order to monitor the decomposition of the prepared composites and the effect of the recycling.

Flammability tests were carried out as follows. Test of UL94 (Flame Chamber made by Atlas Material Testing Solutions), $130 \times 13 \times 4\ mm^3$ specimens, was according to ASTM D 635. LOI (Stanton Redcroft) test was carried out according to ASTM D 2863 with specimen dimensions of $120 \times 10 \times 4\ mm^3$. The specimen, in horizontal position UL94 test (which is always considered the lowest class of flame resistance in the UL94 test), was held at one end in a horizontal position with marks at 25 mm and 125 mm apart from the free end. Flame was applied to the free end and duration of combustion was timed between the two marks. Thus, the horizontal burning speed can be calculated.
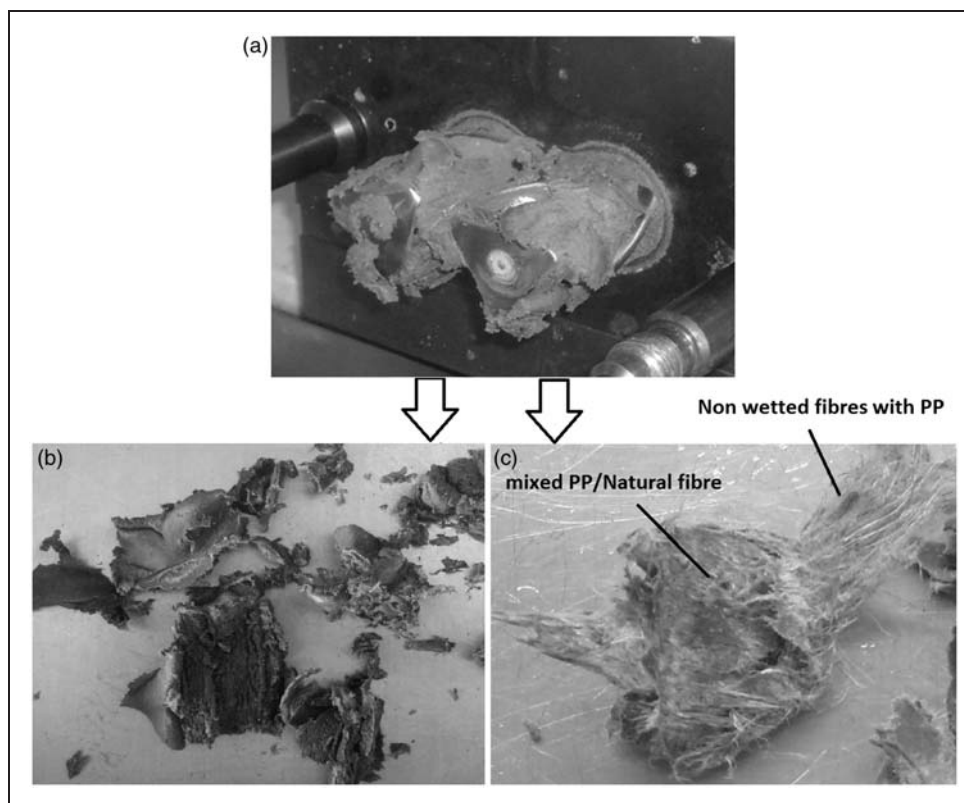
Figure 1. Kneaded compound: (a) in a kneader, (b) homogeneous and (c) inhomogeneous.
PP: polypropylene.

Table 2. List of the composition of each experiment.

Case (fibre%/Mg(OH)$_2$%)	Flax (wt%)	PP (wt%)	MAPP (wt%)	Mg(OH)$_2$ (wt%)	Recycling times
A (30/0)	30	67	3	0	0, 1, 2
B (30/20)	30	47	3	20	0, 1, 2
C (30/30)	30	37	3	30	0, 1,2
D (50/0)	50	45	5	0	0, 1, 2
E (50/20)	50	25	5	20	0, 1, 2
F (50/30)	50	15	5	30	0, 1, 2

PP: polypropylene; MAPP: maleic anhydride-grafted-polypropylene; Mg(OH)$_2$: magnesium hydroxide.

Results and discussion

Stiffness results

Figure 2 illustrates the stiffness results of the compounded samples. The two groups of 30 and 50% flax are presented in the same graph for comparison purpose. The addition of the FR powder shows a positive side effect of 45% stiffness rise, from 4766 to 6914 MPa at 0 and 30 wt% Mg(OH)$_2$, respectively, for the 30% flax group. On the other hand, in the 50% flax group, the stiffness of the 50/0 increased to 7755 MPa (65% compared with the 30/0 case); the stiffness then continued to improve to 14,448 Mpa, with the addition of 30% FR of Mg(OH)$_2$ (86% rise). It was previously reported[26] that 40% improvement in the stiffness of PP/Mg(OH)$_2$ (almost 30 wt% FR and without natural fibres) can be achieved. This stiffness improvement was because the fillers, FR particles, are stiffer than in the host polymer PP.[27] The effect of injection moulding cycles is also shown in Figure 2. A slight improvement took place in the 50/20 system. But in the case of the 30/0 composite, it was observed that stiffness decreased by 8%, whereas that of the 30/30 composite decreased by 15%. The stiffness of 50/0 and 50/30 systems decreased by 14% and 18%, respectively. The current findings are different from those discussed in the study
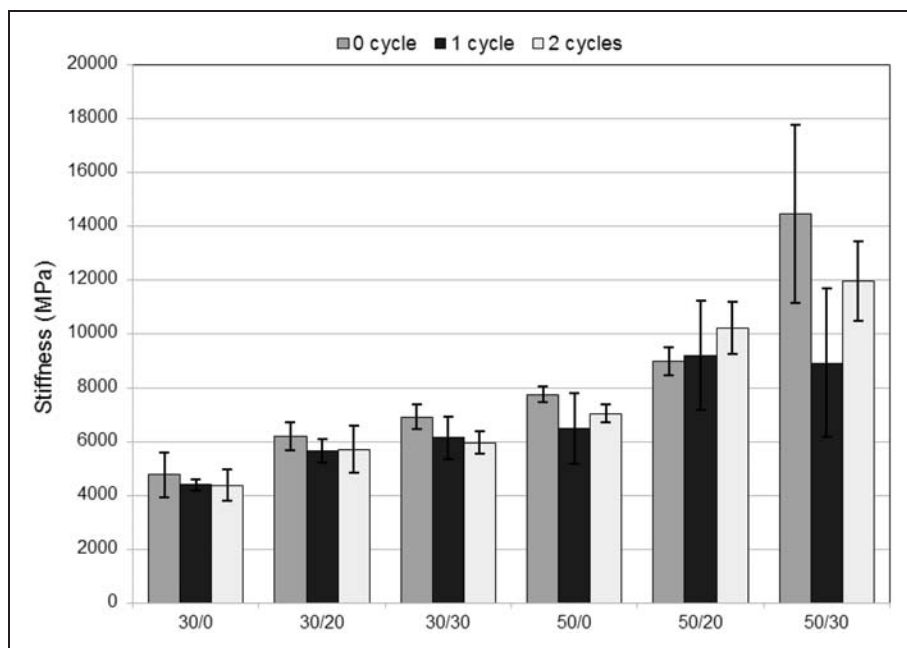
Figure 2. Effect of flax–FR on stiffness of the six investigated compounds at different cycle numbers.

of Duigou et al.[21] but similar to the observations of Moran et al.[22] As reported in the literature findings,[21] the stiffness of the biopolymer polylactic acid with 30 wt% flax decreased slightly from the first cycle. This can be attributed to the fact that non-polar polyolefin PP used in this study does not combine successfully with the flax fibres even in the presence of MAPP compatibiliser. Thus the mechanical recycling induces more sites for coupling between PP and the fibre and hence the mechanical stiffness is enhanced. Additionally, as in the study of Duigou et al.,[21] additives such as FR are not included in the composite. Therefore, the polymer ensured wettability and impregnation through the flax fibres. On the other hand, the experiments of Moran et al.[22] is similar to this study, where flax fibres are compounded with PP (non-polar) and the stiffness of the composite also increased up to the fourth cycle. The difference is mainly influenced by the compounding process, where extrusion is adopted to produce the composite in the work.[26] No compatibiliser was used to link the polymer and the fibres and thus the evolving bonding was mainly a mechanical one.

Similar trends for the stiffness results were found in the storage modulus using DMA. Temperature range of 40–160°C was investigated (i.e. around α-zone, where molecular motions in the crystalline phase are affected by the increasing flax content), as shown in Figure 3. The effect of fibre and the FR contents were studied and thus the glass transition and β-zone was not covered in this study. At high fibre contents, higher storage and loss moduli were observed, as

shown in Figure 3(a) and (b). This is attributed to the increasing frictional energy between the matrix and the filler fibres due to the increasing fibre content.[28] The addition of $Mg(OH)_2$ resulted in an increase in the stiffness of the composite, as shown in Figure 3(a). This matches with the results of the study by Velasco[29] upon the addition of untreated $Mg(OH)_2$ to PP. The loss modulus showed a maximum value around 60°C, as shown in Figure 3(b). The damping factor (tan δ) developed prominently at 40–80°C, as shown in Figure 3(c). This is explained by the presence of rigid amorphous molecular segments in the crystals.[28] After the α-zone peak, the damping factor (tan δ) developed strongly due to the sustaining elasticity of cellulose fibres.[30] The damping factor has greater values at low fibre content, indicating more damping. The effect of $Mg(OH)_2$ was more pronounced at 50% flax content but shows less damping at higher $Mg(OH)_2$ content. As presented in Figure 4, for the 30/0 system and the highly filled system 50/30, the effect of recycling is not significant at second recycle in this study (only the results of 0 cycle and 2 cycles are shown). But what is interesting is the effect of the forced oscillating frequency, where the induced input energy increases with an increase in the frequency. The position of the relaxation α-zone shifted to a higher temperature and the corresponding intensity decreases.[31] The damping factor develops in a positive trend at high temperature, regardless of the applied frequency, except for the 30/0 at 100 Hz. This indicates that the induced high frequency results in higher elastic energy with respect to the loss energy at a temperature range of 120–150°.
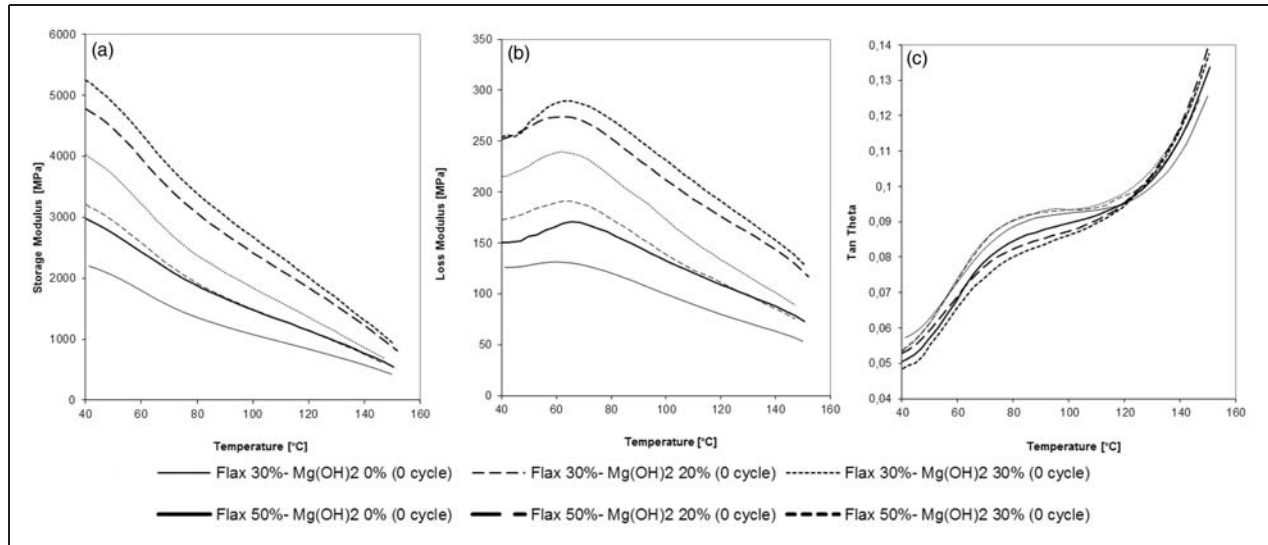
Figure 3. DMA of non-recycled composites at 1 Hz: (a) storage modulus (b) loss modulus and (c) tan θ.
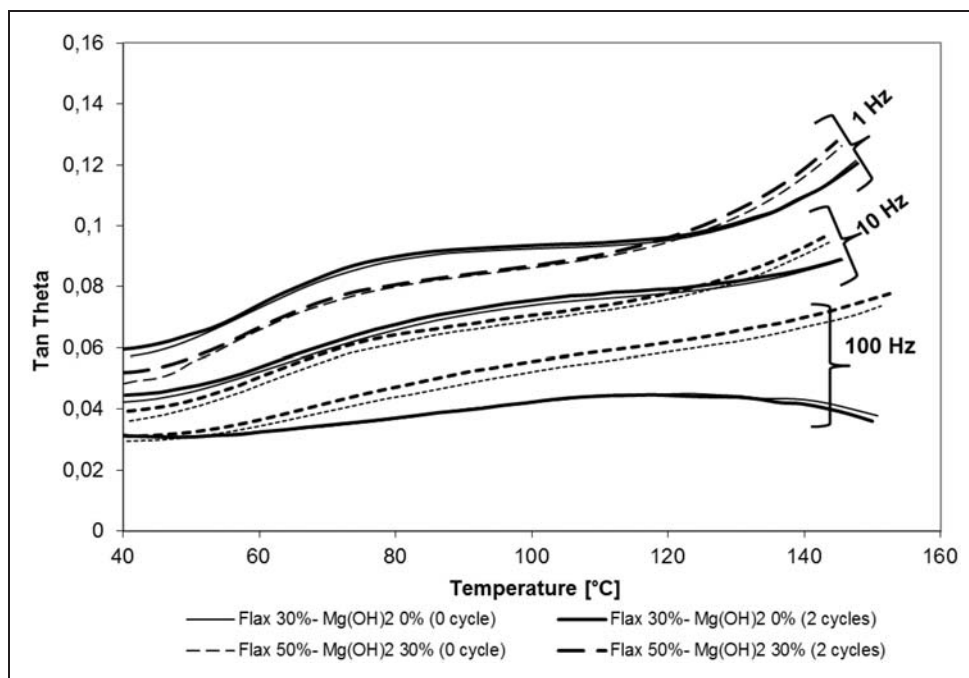DMA: dynamic mechanical analysis; Mg(OH)$_2$: magnesium hydroxide.

Figure 4. Effect of the applied frequency and the recycling number on the composites 30/0 and 50/30.

Tensile strength results

Figure 5 shows the tensile strength results. The ultimate tensile strength decreased in 30 and 50 wt% flax groups and also decreased after the addition of 30 wt% Mg(OH)$_2$ from 42.8 to 38.2 MPa and 51 to 35.1 MPa, respectively, (10.7% and 31.1% decrease). Comparing these results with data from the literature,[8] 15–17% drop in strength is found for 25% FR. The decrease in strength is due to the reduced effective load-bearing

cross-section[27] besides the lower interfacial adhesion strength between FR and PP, in contrast to the high adhesion between natural fibres and PP, which were treated with MAPP. This is evident in the scanning electron microscope micrographs illustrated in Figure 6. Figure 6(a) shows the original Mg(OH)$_2$ particles. The particles are broken and easily disintegrated under vacuum, as seen using scanning electron microscope. In Figure 6(b), the broken surface of the
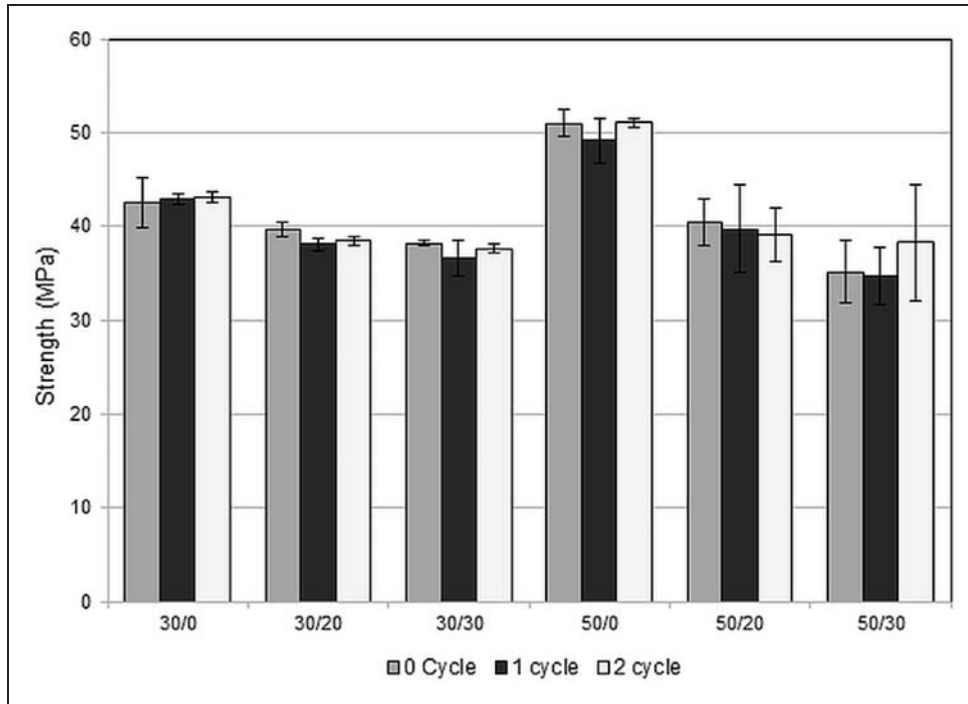
Figure 5. Effect of flax–FR on strength of the six investigated compounds at different cycle numbers.
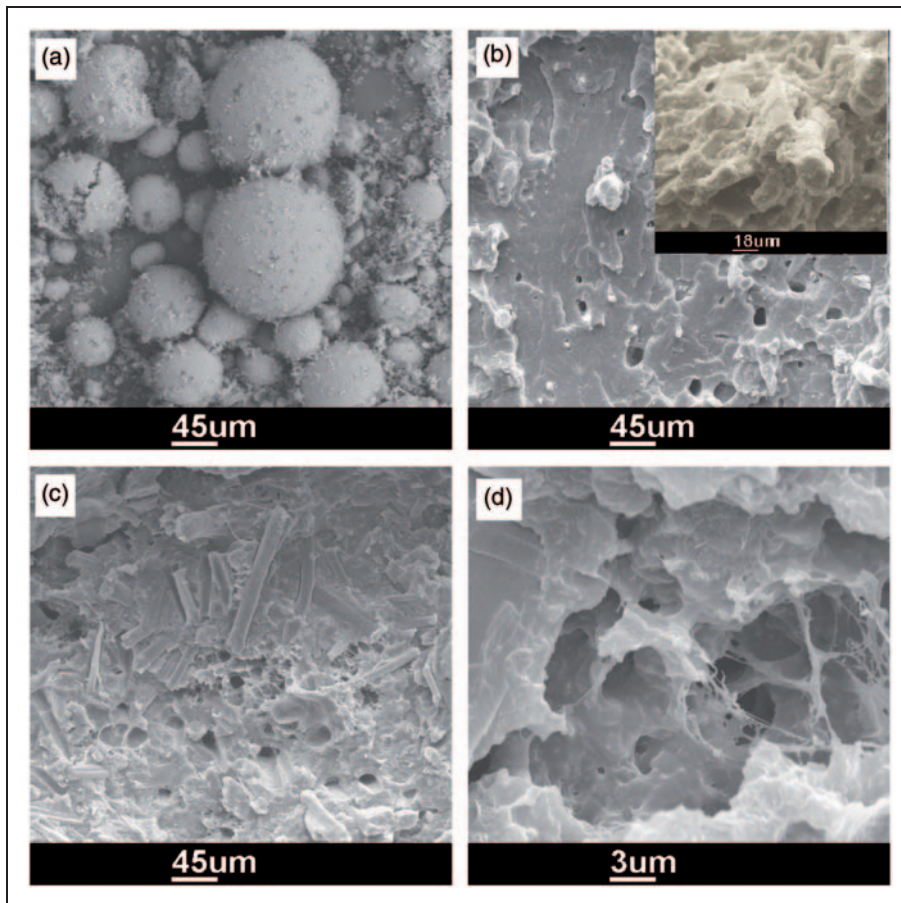

Figure 6. SEM micrographs of (a) $Mg(OH)_2$ particles and impact fracture surfaces for (b) 30/0 (c) and (d) 30/30 samples. SEM: scanning electron microscope; $Mg(OH)_2$: magnesium hydroxide.
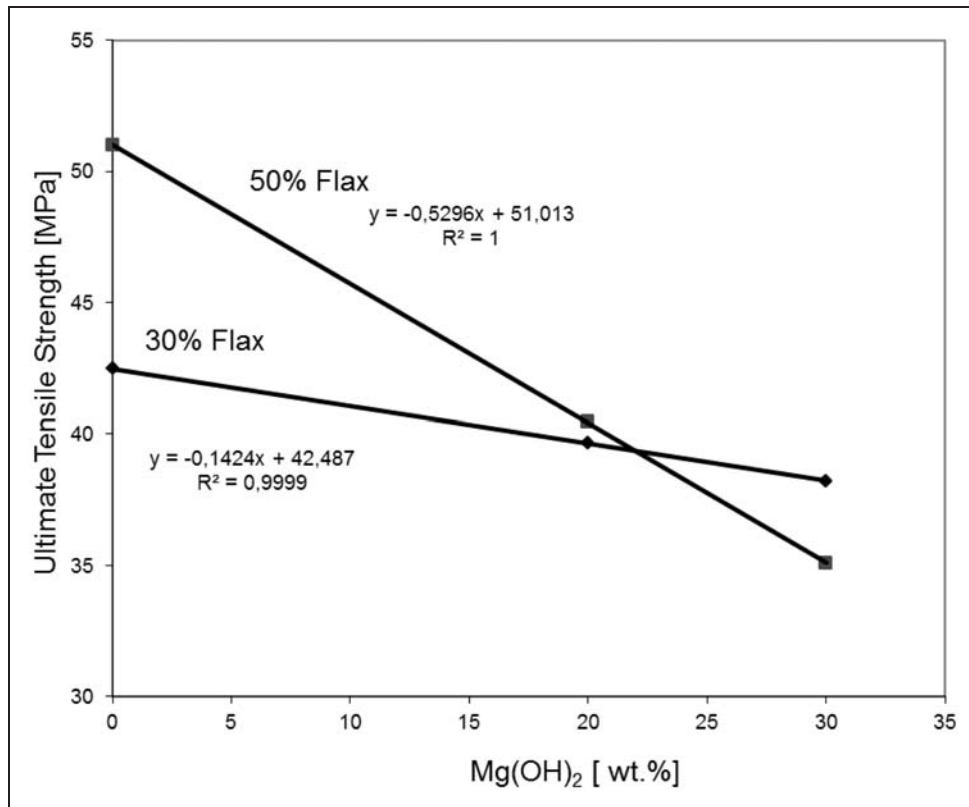
Figure 7. Effect of Mg(OH)$_2$ on the composite tensile strength of PP/30 wt% flax and PP/50 wt% flax at different Mg(OH)$_2$ contents. PP: polypropylene; Mg(OH)$_2$: magnesium hydroxide.

composite 30/0 (without FR) shows integrated matrix with good wetted fibres (the average diameter of elementary fibre after hot shear compounding is 16 μm and the length is 220 μm[32]). The failure mechanisms are either fibre pull-out or debonding or fibre tensile failure, as explained by Elsabbagh et al.[32] Figure 6(c) displays the effect of the Mg(OH)$_2$ fillers in which irregular edged pores are formed due to particle agglomerates. This shows the low adhesion of FR particles with the PP matrix. Figure 6(d) shows a high magnification micrograph of one of these surface areas in which the material suffers discontinuity and therefore strength decreases with the addition of FR fillers.

The linear relation of Mg(OH)$_2$% versus strength shown in Figure 5 displayed a great benefit for the mathematical description of the strength of these composites, as illustrated in Figure 7 for the samples of cycle 0. Coherent relations can be depicted from the recycled samples. The feasibility of strength semi-empirical relation versus FR content is due to either the rule of mixture or one of its modifications for these multi-phase composites.[19] These empirical trends do not precisely follow the rule of mixture due to the fibre length and orientation factors. In addition, there is the lack of data about the mutual interfacial

adhesion strengths between the FRs and the polymer matrix or those of polymer/natural fibre.

The recycling effect appeared to have a no significant effect on the strength of 30/0 system, while in the case of FR systems, a slight decrease in strength was observed. The 50/30 system showed an 8% improvement after two cycles. This might be due to the low PP content (which requires the session of the molecular chains into smaller ones after severe shearing). The several cycles ensure that the required shearing and impregnation of PP between the fibres is attained.

Tensile elongation at break and impact results

Figure 8 presents the elongation behaviour of the compounded samples under tension. A decrease in the elongation at breaking was evident with the addition of FR. This corresponds with the results of Yin et al.,[27] who reported that elongation decreases with the implementation of Mg(OH)$_2$. The recycling effect on elongation is positive. This finding was because the elongation value represents the summation of elastic and plastic zones. The elastic zone in these composites is not trivial but it represents a significant share of the whole elongation, depending on the amount of fibres
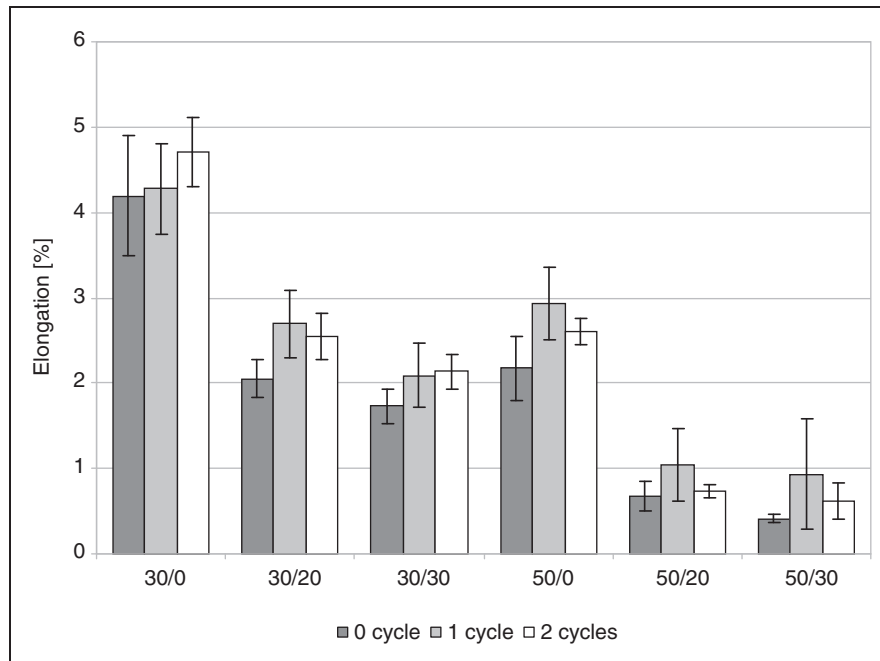
Figure 8. Effect of flax–FR-recycling on elongation at break.
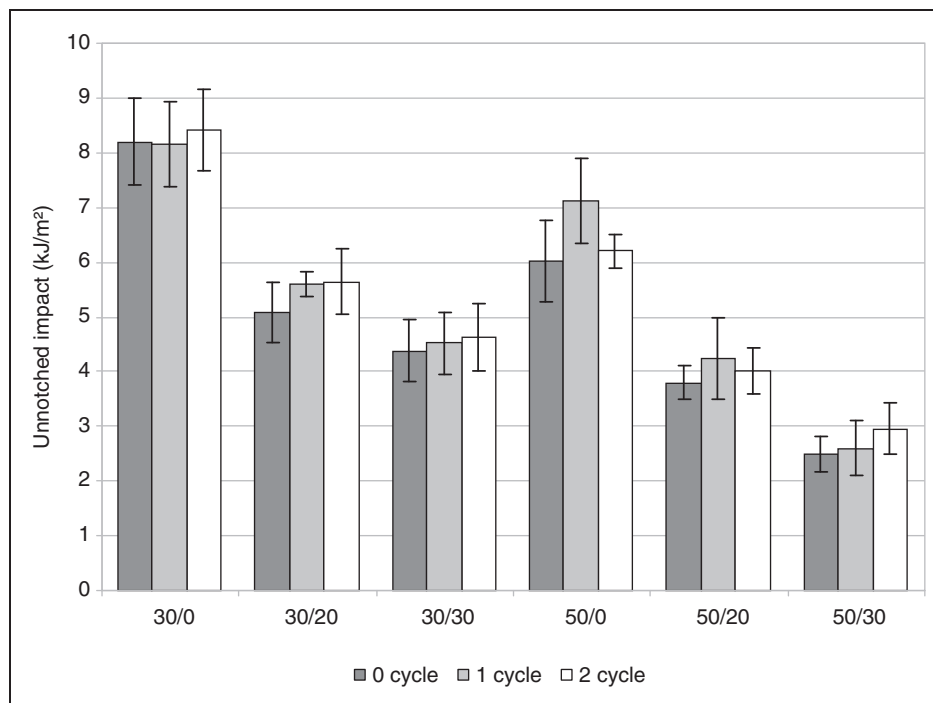FR: flame retardants.

Figure 9. Effect of flax–FR-recycling on impact strength.
FR: flame retardants.

and fillers. Therefore, the elastic elongation, which is presented by the quotient of strength to stiffness, is important in the analysis of these filled composites. So when the stiffness decreased and the strength showed less response to the cycles' repitions, the elongation improved by 12–40%.

Figure 9 illustrates the impact toughness of un-notched Charpy samples. The impact values

decreased with the addition of FR as a result of the decrease of strength and elongation. This is attributed to the reduced deformability of PP because of the effect of FR particles, which hinders the mobility of PP chains.[27] In the same context, it is reported that FR particles act as nucleating agents for PP crystallinity and decrease the amorphous share and hence the mobility is reduced.[29] The fracture surfaces shown in Figure 5 show that the failure mechanism is dominated by the debonding/craze damage and there are no significant signs of shear yielding, which is reflected in enhanced impact toughness.[27]

The decrease in impact of the 50% flax groups is more than that of the 30% group as the impact values decreased from 8 and 6 kJ/m^2 to 4.38 and 2.48

6 kJ/m^2 for the 30% and 50% flax groups, respectively. The recycling effect on impact toughness is directly proportional to the elongation improvement, where the toughness energy values increase to 2–20% of its original values.

Thermal test results of TGA

Figure 10 shows the TGA data of the compounds investigated (the effect of recycling is not shown). It is clear from the figure that the addition of MAPP as a compatibiliser was not very effective since all the 50% flax samples (50/0, 50/20 and 50/30) displayed two onset standard temperatures. The FR Mg(OH)$_2$ has a positive effect by increasing the thermal stability.
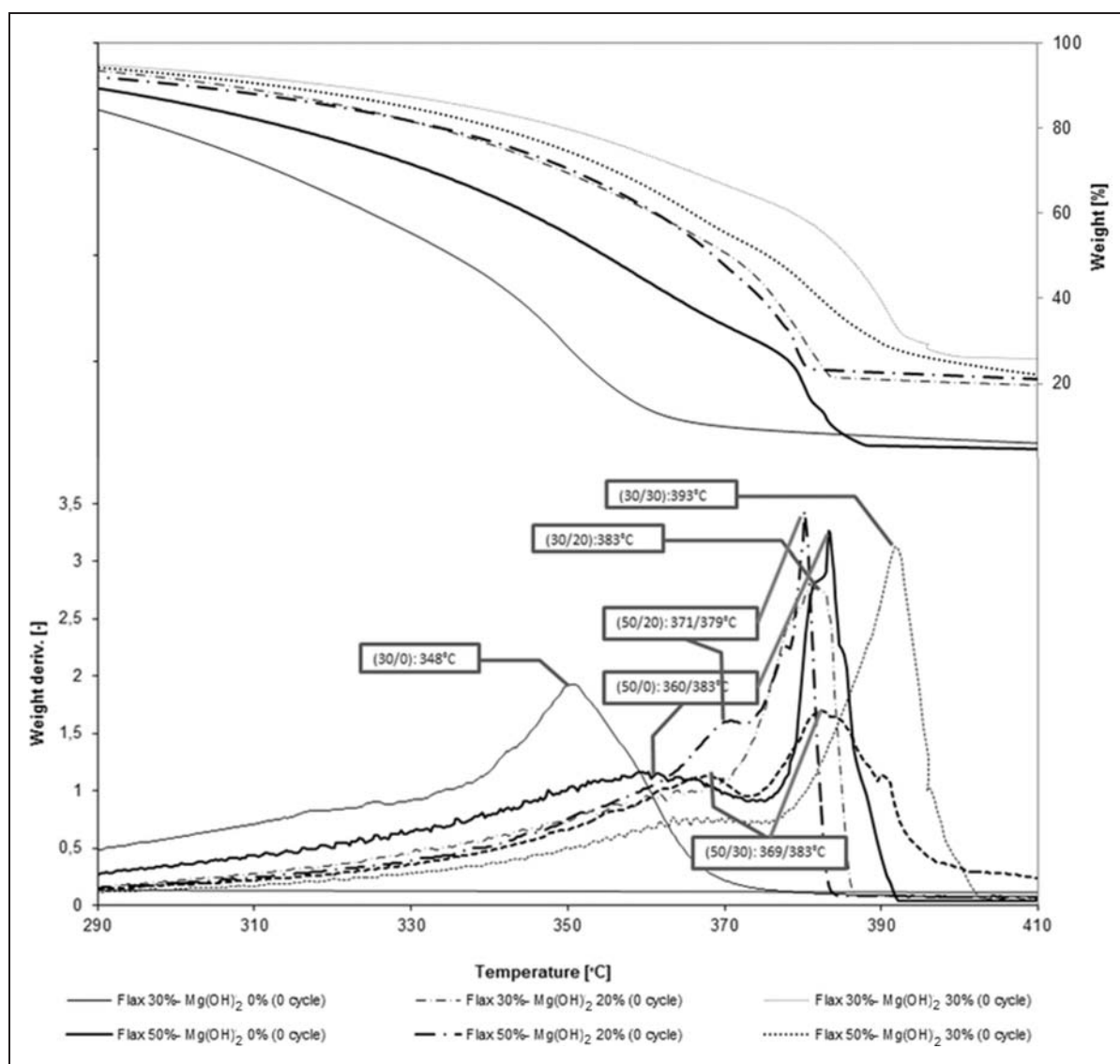

Figure 10. TGA and DTGA of the compounds investigated.
TGA: thermal gravimetric analysis; DTGA: differential thermal gravimetric analysis.
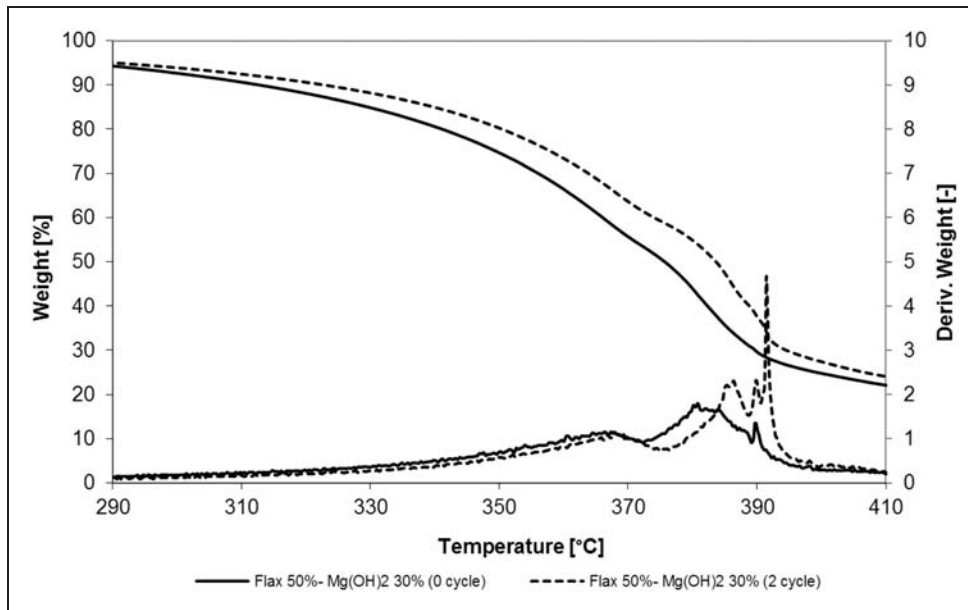
Figure 11. TGA and DTGA of the 50/30 composites (non-recycled and recycled).
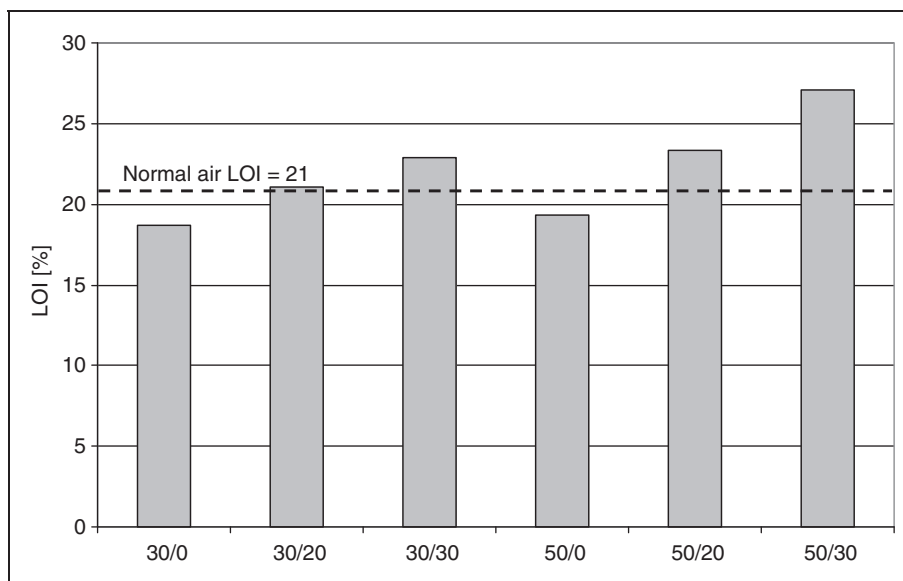TGA: thermal gravimetric analysis; DTGA: differential thermal gravimetric analysis.

Figure 12. Effect of flax–FR on LOI.
LOI: limited oxygen index; FR: flame retardants.

For instance, at 30/0 the onset temperature changes from 348°C to 383°C to 393°C for the 30/0, 30/20 and 30/30 composites, respectively. The onset decomposition temperature changes in a narrower range of 360–371°C in the case of the 50% flax composites. It is obvious that the weight derivative curve in Figure 10 shows a negligible change in the horizontal decomposition rate in the range of 365–375°C, which is attributed to the decomposition of $Mg(OH)_2$. This horizontal trend is greater in the case of 30/30 and 50/30 due to the greater amount of the FR, whereas the 30/20 and 50/20 have shorter horizontal trends.

The effect of recycling on the corresponding TGA is shown in Figure 11. High number of recycling does result in a better compatibilisation between the composite constituents, where the two peaks of the composite (0 cycles) in the range of 360–390°C come close to each other after two cycles. This is due to the increasing

Table 3. UL94V test observations and results.

Sample	Time of burn (s)	Dripping
A (30/0)	<60	Yes
B (30/20)	60–90	Yes
C (30/30)	180–240	No
D (50/0)	60–90	Yes
E (50/20)	180–240	No
F (50/30)	240–300	No
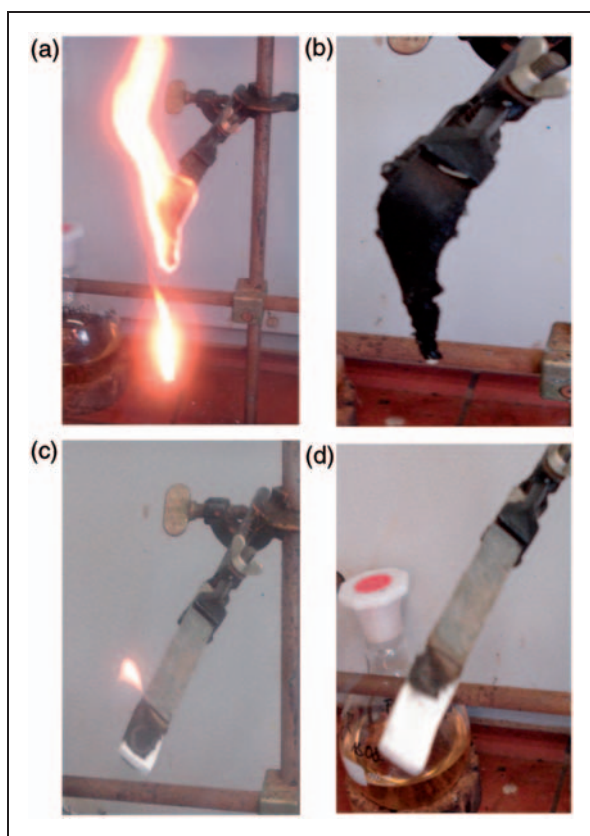
Figure 13. UL94H—(a) and (b): 30/0 and (c) and (d): 50/20 during and after test.

number of sites ready for linking between PP and flax upon increasing injection cycles. This result is in agreement with that of Tajvidi and Takemura,[33] who studied the effect of compatibiliser on TGA.

Results of flammability tests

Figure 12 illustrates the LOI values corresponding to the different compounding composite formulations. 30/0 and 50/0 composites have the lowest LOI values, which are below the normal oxygen percentage in the atmosphere, which is 21. That means that these systems support flame progression in air. Addition of $Mg(OH)_2$ increases the required oxygen needed to sustain flammability. At 20% level of FR in the 30 and 50% flax groups, LOI increases to 21.1 and 23.3, respectively. Although these values are higher than 21, they are not high enough to extinguish the active fire. At the 30% level of FR, LOI increased again to 22.9 and 27.1. The latter LOI value of 50/30 composite system is an indication of good flame resistance. This agrees with the results reported by Helwig and Paukszta,[23] where the cone calorimeter testing showed that 12.5 wt% flax in PP ensures the decrease of peak heat release rate by about 35%, compared with that of neat PP.

LOI test is an important quantitative test but UL94 test (specially the vertical test) is more relevant in practice. Results of UL94V are presented in Table 3. None of the compounded samples has succeeded to satisfy the required rating conditions of the least class V2, as mentioned in ASTM D 635 (briefly, self-extinguishing within 60 s after exposure to the direct burner twice). However, the observation of the test highlighted some important indications. Samples 30/30, 50/20 and 50/30 show glow combustion features. Sample 50/30 burns at a slow rate as it prolonged for 4–5 min. Also, these samples have successfully shown no signs of dripping. Figure 13 shows samples of 30/0 and 50/20 during and after combustion. As shown in Figure 13(a), the sample showed dripping during combustion. The sample, after complete burning, showed carbonaceous material (Figure 13(b)). The 50/20 sample showed a small flame in Figure 13(c) and stops, as shown in Figure 13(d); however, it is still glowing inside. Figure 14 shows the results of UL94H. As $Mg(OH)_2$ increases, the burning speed decreases. The 30% flax group shows 58% reduction in the horizontal burning speed, while the 50% flax group has 93% reduction. Therefore, it can be inferred that FR composites $PP/Flax/Mg(OH)_2$ (rated UL94V) cannot be attained with such low percentage of $Mg(OH)_2$. It was reported[8,20] that the minimum required $Mg(OH)_2$ level lies between 50% and 60%. This high amount of FR is not appropriate for natural fibre composites. In addition, the mechanical properties are expected to be very low. However, with 20–30% FR, a low combustible composite with an acceptable level of strength decrease and promising recycling results are obtained. Figure 15 shows the recycling effect on the best performance sample where a low decrease of almost 5% is recorded.

Conclusion

– Low combustible and reduced smoke of PP/flax composites can be obtained with the addition of $Mg(OH)_2$ FR.
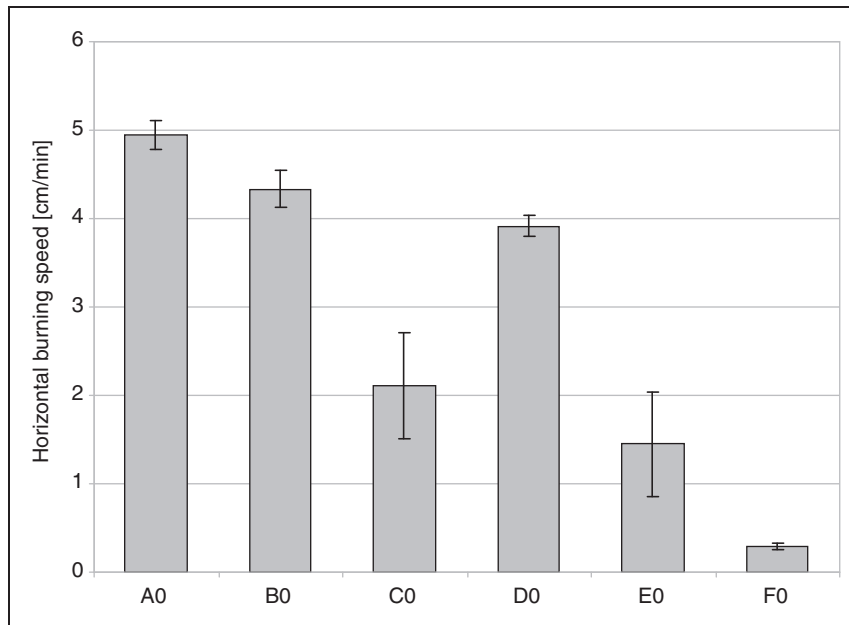
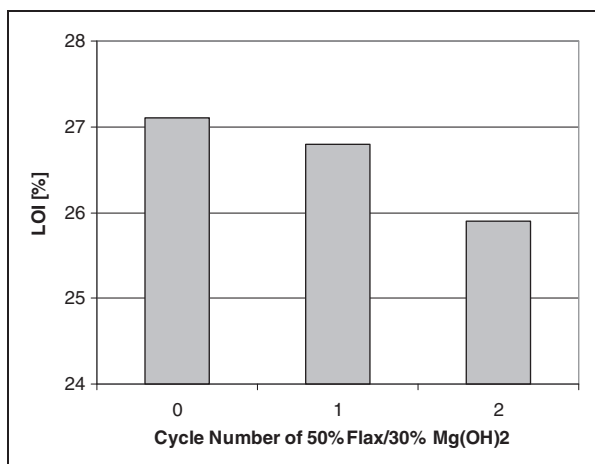**Figure 14.** Horizontal burning speed of non-recycled samples.

Figure 15. Recycling effect on LOI.
LOI: limited oxygen index; $Mg(OH)_2$: magnesium hydroxide.

– The 30 wt% flax/PP composite filled with 30 wt% $Mg(OH)_2$ is a promising system where a combination of slow combustion characteristics and good mechanical properties is attained. Horizontal burning speed is reduced by 52%, whereas the stiffness value is improved by 45% and the strength decreases by 10%.

– The group of 50 wt% flax/PP composites shows promising results concerning two systems, namely 50/20 and 50/30. Horizontal burning speeds are reduced by 71% and 93%, whereas the stiffness values are improved by 16% and 88% and the strength decreases by 20% and 31%, respectively. The group

of 50% flax/PP composites suffers principally from low impact strength. However, better mechanical performance can be attained by the control of fibre size to finer dimensions.

– Thermal and thermomechanical characterisation of the composite systems (new/recycled) show that the FR increases the thermal stability and the storage modulus but decreases the damping factor. Recycling affects the stiffness and strength only slightly, as they decrease by 18% and 8%, respectively. Flammability also decreases tolerably, whereas LOI decreases by 4.5% for the best 50/30 system.

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