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Recovery of recycled poly(ethylene terephthalate) via melt mixing with poly(butylene succinate) and ultrafine wollastonite

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Abstract

Recycled poly(ethylene terephthalate) (R-PET) flakes obtained from postconsumer bottles were melt mixed with poly(butylene succinate) (PBS) or with PBS plus ultrafine wollastonite (2000 mesh) at various compositions on a twin screw extruder, followed by injection molding. The tensile properties, morphology, thermal stability and limiting oxygen index (LOI) of the resulting blends and composites were evaluated. Among the investigated R-PET/PBS blend compositions (20, 30, 40, 50, and 60 wt% PBS), the 60/40 wt% blend exhibited the highest tensile strength and elongation at break, but a lower tensile modulus and thermal stability compared to those of the neat R-PET. This blend was subsequently blended with different loading levels of wollastonite (10–30 parts by weight per hundred (phr) of blend resin). According to the tensile testing, thermogravimetric analysis and LOI results, the tensile modulus, tensile strength, thermal stability and flame retardancy of the 60/40 wt% R-PET/PBS blend were improved by the incorporation of wollastonite (optimal at 30 phr), while the elongation at break exhibited no significant change. Moreover, wollastonite was effective in preventing melt dripping during combustion of the composites.

1. Introduction

As poly(ethylene terephthalate) (PET) waste is one of the major components of global municipal solid waste, the business of PET recycling has attracted investment capital. Not only the quantity but also the quality of PET wastes are high enough to support investment [1-3], where a large amount of PET wastes are principally obtained from a rigid postconsumer packaging. Recycled PET (R-PET) flakes are usually derived from bottles, which are commonly used for water, beverages, foods, cosmetics and household chemicals [4,5], since they can be easily sorted, collected and recovered from municipal solid wastes. With an efficient recycling system, discarded postconsumer PET can be recycled into value-added products [6,7]. Although the mechanical and thermal properties of the R-PET products are deteriorated during the mechanical recycling process from the thermo-mechanical and hydrolytic degradations induced by the heat, stress and the presence of retained moisture [8-11], this technique is still recommended due to its ecological and economical profits [2,6,12]. Embrittlement of the R-PET occurs with the reduction of its molecular weight. The addition of an appropriate toughening modifier and reinforcing filler into the R-PET is an effective method to improve certain properties with a consequential broadening of its utilization [1,9,10,13-15]. The melt blending of either virgin PET or R-PET with biodegradable polyesters has received considerable attention as a potential solution to solid waste management [4,10,16-19], among which poly(butylene succinate) (PBS) in particular has attracted an increasing level of commercial interest [19].

PBS is one of the biodegradable aliphatic polyesters with many attractive properties, such as a high flexibility, excellent impact strength and

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chemical resistance and easy processability [20-22]. These properties suggest PBS is an option for blending with R-PET to increase the toughness, biodegradability and applications of the resulting blended recycled products [19,20]. Biodegradation of PBS proceeds via hydrolytic degradation, due to its easily hydrolysable nature, whereas hydrolysis of PET is very slow under normal conditions [4,20,23]. In principal, after the biodegradable component in the blend has been removed by microorganisms that exist in the waste disposal environment, the remaining inert component should lose its integrity, disintegrate and disappear. In this study, the tensile properties of a series of R-PET/PBS blends with increasing PBS levels were investigated. Previous work has shown that, in general, the incorporation of PBS into brittle polymers results in a significantly improved ductility and toughness at the expense of a reduced strength and modulus [24,25]. However, polymer blends filled with inorganic rigid particles were shown to have an enhanced strength and modulus [26,27]. Therefore, the R-PET/PBS blend with the optimum tensile properties found in this study was subsequently filled with different amounts of wollastonite, an inorganic reinforcing agent, to try to improve its mechanical properties as well as its thermal stability and performance. A number of previous studies have reported the reinforcement of thermoplastics with wollastonite [28-31], but R-PET/PBS/wollastonite composites have not been studied previously.

Wollastonite is a calcium metasilicate ($CaSiO_3$) mineral that occurs naturally in an acicular (needle-shaped) crystal structure with a high aspect ratio (L/Dof 10-20). It is theoretically comprised of 48.25 wt% CaO and 51.75 wt% SiO_2 [28-30,32]. The wollastonite used in the polymer industries has a high chemical purity with a range of desirable properties including a high chemical and thermal stability, low coefficient of thermal expansion and water absorption, high level of whiteness and hardness (Moh's hardness 4.8), small health hazard compared to asbestos, and a very low cost [29,33]. In this study, ultrafine wollastonite (2000 mesh) was added at various loading levels (10, 15, 20, 25, and 30 parts by weight per hundred (phr) of R-PET/PBS blend) into the R-PET/PBS blend in order to produce composite materials with improved material properties. The tensile properties, morphology, thermal stability and flammability of the resulting composites were then investigated.

2. Experimental

2.1 Materials

R-PET flakes were obtained from grinding postconsumer bottles. PBS (GS Pla AZ91 TN) pellets with a melt flow index (MFI) of 4 g 10 min^{-1} (190°C, 2.16 kg) and a density of 1.26 g cm^{-3} were purchased from the Mitsubishi Chemical Corporation. The melting point of the R-PET and PBS, as determined from the differential scanning calorimetry (DSC) analysis, was 244°C and 111°C, respectively. Wollastonite (XYNFW-XA) powder with a particle size of 2000 mesh and a density of 2.85 g cm^{-1} was obtained from the Pacific Comma Trading Company.

2.2 Sample preparation and characterization

Prior to compounding, R-PET, PBS and wollastonite were oven-dried at 80°C for 24 h, 65°C for 12 h and 80°C for 12 h, respectively, to remove the residual moisture. Initially, R-PET flakes and PBS pellets were melt blended at five different weight compositions (80/10, 70/30, 60/40, 50/50 and 40/60 wt% PLA/PBS), using a laboratory-scale twin-screw extruder (LTE-26-40, Labtech Engineering Company). The extruder was operated under a temperature profile of 255, 260, 255, 250, 240, 235, 225, 215, 195, 185, and 180°C from the feed section to the die head at a fixed screw rotational speed of 50 rpm. After cooling, the extruded strands were pelletized and then injection molded into the standard tensile test specimens using the Toshiba Machine EC 130S injection-molding machine under a temperature profile of 260, 250, 245, and 240°C.

The R-PET/PBS/wollastonite composites were also prepared by the melt mixing and injection molding processes as described above but with the addition of one of five loadings of wollastonite (10, 15, 20, 25, and 30 phr of R-PET/PBS blend resin).

2.3 Characterization of the blends and composites

Each tensile test was performed on a dumbbell-shaped specimen according to ASTM D638 Type 4 standard using a universal testing machine (LLOYD LR 100K). The load cell capacity and crosshead speed were 10 kN and 50 mm min^{-1}, respectively. The values of the tensile strength, elongation at break and tensile modulus were averaged from at least five specimens.

The morphology of the wollastonite and the tensile fractured specimens was observed by scanning electron microscopy (SEM) using a JEOL JSM-5410 LV instrument under an accelerated voltage of 2.5 kV with a magnification of 1000×. The fractured surface was observed after sputter coating with a thin layer of gold to avoid electrostatic charges during examination.

The thermal stability of the samples was analyzed by thermogravimetric analysis (TGA) using a Mettler Toledo TGA/SDTA 851^e analyzer under a nitrogen (N_2) atmosphere at a heating rate of 20°C min^{-1} over a temperature range of 50–1000°C.

The limiting oxygen index (LOI) was measured on a Stanton-Redcroft Oxygen Indexer according to the standard 'oxygen index' test ASTM D-2863-91, with a thin sheet (140 mm × 52 mm × 1.5 mm). The test was carried out under a controlled N_2-oxygen (O_2) mixed atmosphere, where the gas mixture of N_2 and O_2 was allowed to pass through the burning sample at a carefully controlled rate. The minimum O_2 concentration in N_2 that was just sufficient to keep the sample burning was reported.

3. Results and discussion

3.1 Tensile properties

The results of the tensile test performed on the injection molded specimens, in terms of the tensile modulus, elongation at break and tensile strength, are summarized in Table 1. It is evident that the tensile modulus of all R-PET/PBS blends was lower than that of the neat R-PET and decreased continuously with increasing PBS contents from 2.1-fold lower at 20 wt% PBS to 3.6-fold lower at 60 wt% PBS, due to the inherent flexibility and low modulus of PBS that led to a reduction in the stiffness of the blends. Moreover, the neat R-PET was brittle and failed at an elongation at break of about 1.7%. With the inclusion of the far less brittle PBS, the elongation at break of the R-PET/PBS blends increased to a maximum value of 11.1% (6.5-fold more than that of the neat R-PET) at a 40 wt% PBS loading, which could be related to the ductility of PBS. However, it decreased slightly at higher PBS levels. In contrast, the tensile strength of the R-PET was decreased by about 1.2-fold with the inclusion of PBS at either 20 or 30 wt%, which may be due to the relatively lower strength of PBS than that of the R-PET and also to the insufficient level of dispersion of PBS in the R-PET matrix. However, the tensile strength of the R-PET/PBS blends then increased to a maximum value of 37.2 MPa at 40 wt% PBS, which was similar to that of the neat R-PET. This may be attributed to the better dispersion and miscibility between the R-PET and PBS. The further addition of PBS to 50 wt% and 60 wt% led to a 1.6- and 1.7-fold declined tensile strength, respectively, compared to that of the neat R-PET. This decrease in the tensile strength at high PBS levels is expected for a non-miscible blend as a result of the poor dispersion or agglomeration of PBS, providing less stress transfer across the phase of each polymer.

Based on the elongation at break and tesile strength data, the 40 wt% PBS containing R-PET/PBS blend was selected as the optimum and so was used for preparing the wollastonite containing (10 to 30 phr) composites.

As expected, the tensile modulus increased with the addition of wollastonite into the composites in a dose-dependent manner from 1.02-fold higher at 10 phr wollastonite to 1.3-fold higher at 30 phr wollastonite compared to that of the neat 60/40 R-PET/PBS blend (Table 1). This is a consequence of the stiff acicular wollastonite particles that restricted the mobility of the polymer chains. However, the elongation at break of the composites containing different levels of wollastonite was not improved, but rather was slightly lower (~1.1-fold) than that of the neat 60/40 R-PET/PBS blend and thus the incorporation of wollastonite provided only a slight effect on the elongation at break of the composite samples. Meanwhile, compared to that of the neat 60/40 R-PET/PBS blend, the tensile strength of the composites was increased with increasing wollastonite contents at 20–30 phr, but not with 10–15 phr, up to a 1.2-fold increase at 30 phr wollastonite. The increased tensile strength of the composites can be explained in terms of the reinforcing effect of the wollastonite with a needle-shaped structure (Figure 1), where the high aspect ratio of the particles enhanced the stress transfer from the matrix to the wollastonite particles. Thus, the incorporation of wollastonite in a well-dispersed manner could lead to an increased tensile strength of the polymer matrix. In conclusion, the addition of wollastonite increased not only the stiffness but also the tensile strength of the composites, whilst the elongation at break remained essentially unaffected.

Table 1. Tensile properties of the samples.

Sample	Young's modulus (MPa)	Elongation at break (%)	Tensile strength (MPa)
R-PET	1100 ± 21.3	1.7 ± 0.5	37.0 ± 0.7
PBS	350.1 ± 10.9	60.3 ± 17.6	32.6 ± 0.9
80/20 R-PET/PBS	537.0 ± 68.3	9.3 ± 0.8	32.0 ± 3.1
70/30 R-PET/PBS	462.5 ± 47.6	10.1 ± 0.9	32.0 ± 2.7
60/40 R-PET/PBS	452.2 ± 11.7	11.1 ± 0.4	37.2 ± 1.3
50/50 R-PET/PBS	325.5 ± 16.9	9.8 ± 0.7	23.5 ± 1.6
40/60 R-PET/PBS	305.7 ± 26.35	10.0 ± 1.0	21.5 ± 2.2
60/40/10 R-PET/PBS/Wollastonite	463.4 ± 15.9	10.2 ± 0.6	35.3 ± 0.8
60/40/15 R-PET/PBS/Wollastonite	532.6 ± 15.1	9.8 ± 0.2	36.7 ± 0.3
60/40/20 R-PET/PBS/Wollastonite	563.7 ± 13.8	9.9 ± 0.3	40.7 ± 0.5
60/40/25 R-PET/PBS/Wollastonite	589.8±12.0	10.0 ± 0.5	42.5 ± 1.0
60/40/30 R-PET/PBS/Wollastonite	607.2±13.0	9.8 ± 0.4	42.9 ± 0.5

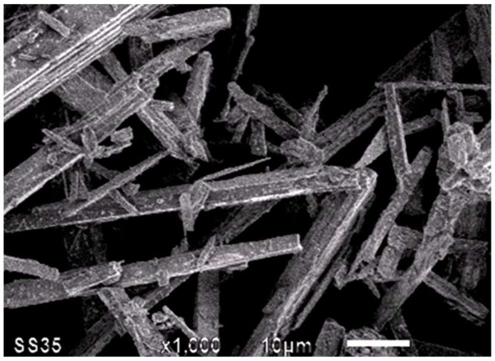

Figure 1. Representative SEM micrograph (1000× magnification) of the needle-like morphology of the wollastonite.

3.2 Morphology

The SEM micrograph of the tensile fractured surface of the neat R-PET revealed a relatively smooth surface with a brittle fractured behavior (Figure 2(a)), while the PLA/PBS blends showed much rougher fractured surfaces (Figures 2(b)-(f)), which was responsible for the increased fracture toughness of the blends. As can be seen in the R-PET/PBS blends with PBS as a minor phase (20-40 wt%), the PBS is dispersed in the form of small particles with the appearance of droplets and without noticeable agglomeration (Figures 2(b)-(d)). However, at 50 and 60 wt% PBS, larger dispersed PBS droplets were visible on the fractured surfaces as well as large voids, where PBS droplets were pulled out during the tensile testing. (Figures 2(e) and (f)). These could serve as local stress concentrators under tensile stress and so lower their tensile strength.

The morphology of the tensile fractured surface of the 60/40 R-PET/PBS blend composites with different wollastonite loading levels (10-30 phr) compared to the neat 60/40 R-PET/PBS blend is presented in Figure 3. The wollastonite particles were clearly dispersed and intimately embedded in the polymer matrix with no discernible bonding between the matrix and the particles. However, the needle-shaped wollastonite microparticles were well mixed and preferentially aligned along the melt flow direction during the injection molding of the test specimens, which would contribute to the enhanced tensile strength of the R-PET/PBS/wollastonite composites.

3.3 Thermal properties

TGA analysis was conducted to determine the thermal stability of the neat R-PET, PBS and 60/40 R-PET/PBS blend and the 60/40 R-PET/PBS blend composites with different amounts of wollastonite. Representative high temperature TGA thermograms are shown in Figure 4, whilst the derived temperatures for the onset (T_{onset}), end set ($T_{\text{end set}}$), 50% weight loss ($T_{50\%}$) and % char are listed in Table 2. The inclusion of PBS into the R-PET caused a dose-dependent reduction in the thermal stability of the blend, which was evident from the decrease in its T_{onset}, $T_{\text{end set}}$, $T_{50\%}$ and %char compared to those of the R-PET. This is because PBS has a much lower thermal stability than R-PET. Moreover, during the processing, the blends were melted twice under high shear rates (on extrusion and injection), resulting in a reduction in the molecular weight that, in turn, deteriorate the thermal stability. However,

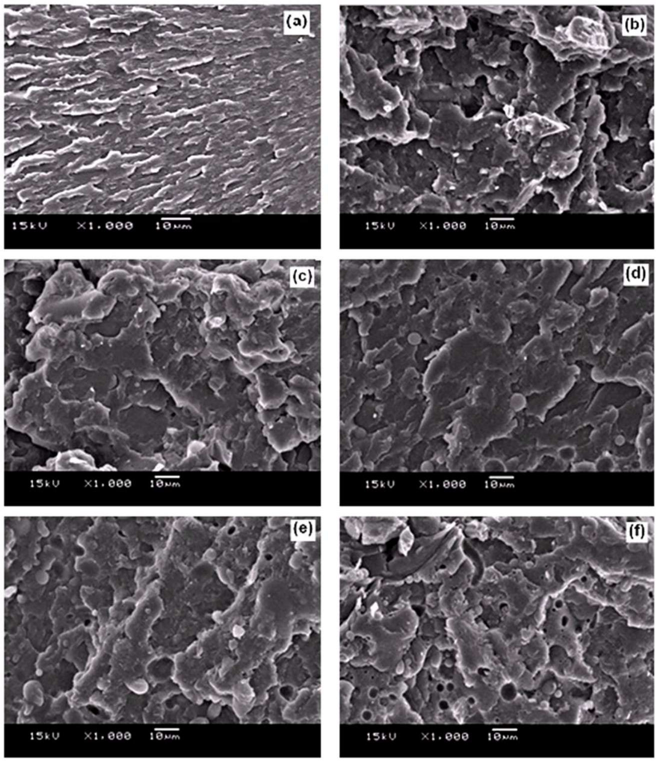

Figure 2. Representative SEM micrographs (1000× magnification) of the tensile fractured surface of the (a) R-PET and (b-f) R-PET/PBS blends with (b) 20 wt%, (c) 30 wt% (d) 40 wt% (e) 50 wt% and (f) 60 wt% PBS.

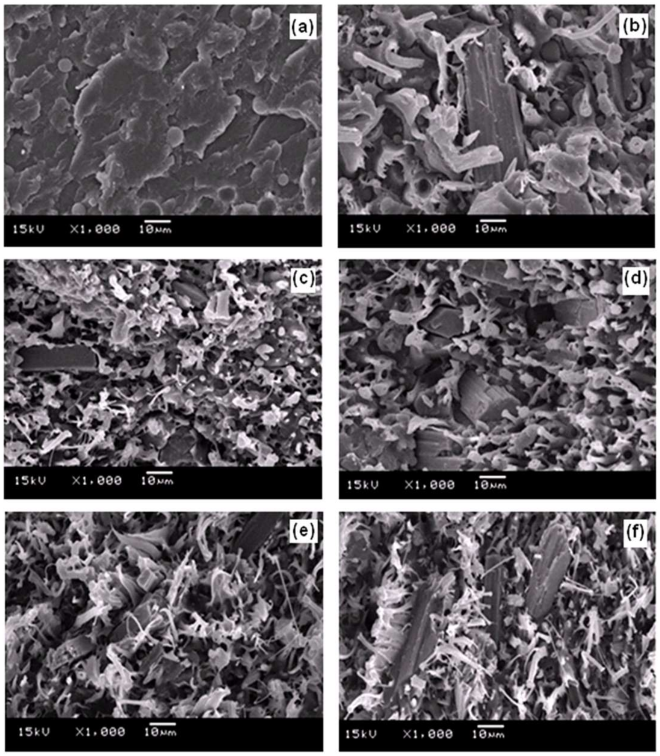

Figure 3. Representative SEM micrographs (1000× magnification) of the tensile fractured surface of the 60/40/x R-PET/PBS/wollastonite composites, where x = (a) 10 phr, (b) 15 phr, (c) 20 phr, (d) 25 phr and (e) 30 phr.

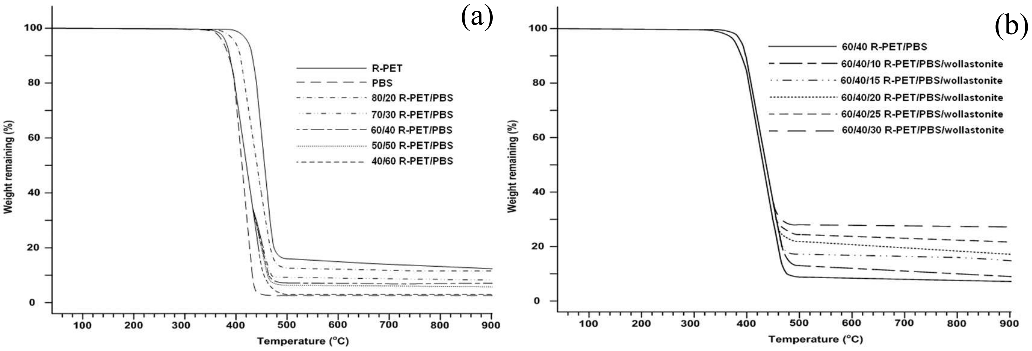

Figure 4. TGA thermograms of (a) R-PET, PBS and R-PET/PBS blends and (b) 60/40 R-PET/PBS blend and 60/40 R-PET/PBS blend composites with various loading levels of wollastonite (10, 15, 20, 25 and 30 phr).

Table 2. TGA-derived data for the samples.

Sample	T_{onset} (°C)	$T_{\text{end set}}$ (°C)	$T_{50\%}$ (°C)	Char (%)
R-PET	420	470	448	14.3
PBS	378	433	410	1.7
80/20 R-PET/PBS	390	466	436	10.8
70/30 R-PET/PBS	388	465	430	7.4
60/40 R-PET/PBS	387	464	428	7.0
50/50 R-PET/PBS	386	462	424	5.1
40/60 R-PET/PBS	384	461	416	3.4
60/40/10 R-PET/PBS/Wollastonite	390	466	428	11.2
60/40/15 R-PET/PBS/Wollastonite	392	467	428	16.0
60/40/20 R-PET/PBS/Wollastonite	392	468	428	19.1
60/40/25 R-PET/PBS/Wollastonite	394	468	428	23.2
60/40/30 R-PET/PBS/Wollastonite	394	469	428	28.8

the inclusion of wollastonite resulted in a dose-dependent enhancement of the thermal stability of the composites compared to that of the neat 60/40 R-PET/PBS blend. The T_{onset} and $T_{\text{end set}}$ values of the composites were shifted towards higher temperatures and the char residue level of the composites was increased with increasing wollastonite loading levels, while the $T_{50\%}$ remained unchanged. The char formation could effectively hinder the diffusivity and permeability of combustible gases into the polymer matrix and thus elevated the thermal stability of the composites. Consequently, wollastonite could be used as a heat-resistant modifier for PLA/PBS blends.

3.4 Flammability

The LOI value of the injection molded neat 60/40 R-PET/PBS blend was slightly increased in a dose-dependent manner with increasing wollastonite loading levels (Table 3). Nevertheless, all the specimens had sufficient LOI values above 21 and so were sufficient to render a degree of flame retardancy to the composite materials. Moreover, the samples with 0 and 10 phr wollastonite showed melt dripping of the materials during combustion, whereas the composites with higher wollastonite loadings (15–30 phr) showed a char residue on the surface of the specimens with no dripping of the materials (Figure 5). Therefore, wollastonite plays an important role in inducing char formation and hindering the melt driping during combustion.

Table 3. The LOI values of the samples.

Sample	LOI
60/40 R-PET/PBS	22.0
60/40/10 R-PET/PBS/Wollastonite	22.2
60/40/15 R-PET/PBS/Wollastonite	23.8
60/40/20 R-PET/PBS/Wollastonite	24.1
60/40/25 R-PET/PBS/Wollastonite	24.2
60/40/30 R-PET/PBS/Wollastonite	24.3

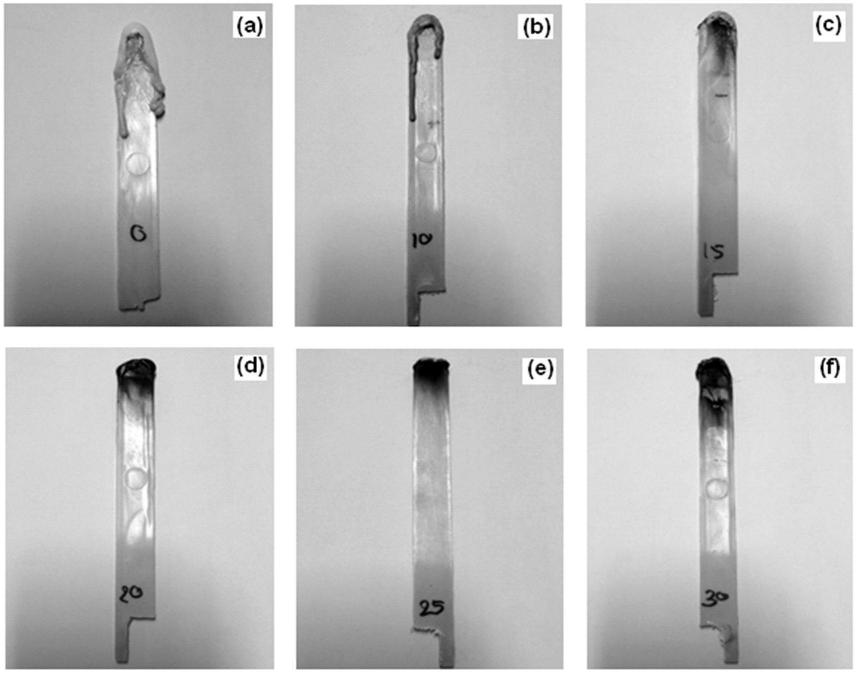

Figure 5. Representative images of the test specimens obtained from the flammability test of the 60/40/x R-PET/PBS/wollastonite composites, where x = (a) 0 phr, (b) 10 phr, (c)15 phr, (d) 20 phr, (e) 25 phr, and (f) 30 phr.

4. Conclusions

In this study, R-PET/PBS blends and composites of the 60/40 R-PET/PBS blend with ultrafine wollastonite were successfully prepared by melt blending on a twin screw extruder followed by injection molding. Among the studied R-PET/PBS blends, the incorporation of 40 wt% PBS was found to yield the highest tensile strength and elongation at break, but it had a lower tensile modulus and thermal stability than the neat R-PET. Regardless, based on having the highest tensile strength and elongation at break, the 60/40 R-PET/PBS blend was selected for preparing composites with five different loading levels of wollastonite (10-30 phr). The tensile strength and modulus of the composites increased to a maximum value of 42.9 MPa and 607.2 MPa, respectively, with the addition of 30 phr wollastonite, while the elongation at break was slightly decreased to 9.4%. Moreover, the thermal stability of the composites was also improved with the inclusion of the wollastonite, being optimal at 30 phr. Thus, it can be concluded that the strength, toughness, stiffness, thermal stability and flammability of R-PET were improved by melt blending with an appropriate amount of PBS (40 wt%) and wollastonite (30 phr). Moreover, wollastonite also depressed the level of melt dripping of the composites during combustion, which may offer a good opportunity to utilize R-PET in the engineering field.

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