



Accelerated weathering of fire-retarded wood–polypropylene composites



Irina Turku*, Timo Kärki

School of Energy Systems, Lappeenranta University of Technology, P.O. Box 20, Lappeenranta 53851, Finland

ARTICLE INFO

Article history:

Received 9 August 2015

Received in revised form 29 October 2015

Accepted 17 November 2015

Available online 2 December 2015

Keywords:

Wood–plastic composites

Co-extrusion

Fire retardants

Weathering

ABSTRACT

In this study, the influence of fire retardants, namely aluminum trihydrate, zinc borate, melamine, graphite, titanium dioxide on the durability of polypropylene-based co-extruded wood–plastic composites is studied. The composites underwent accelerated weathering under a xenon-arc lamp source during 1000 h. FTIR analysis of the composite surface revealed a degradation process which was accompanied by chemical changes, including vinyl-like and carbonyl groups accumulation; fire retardants did not influence the photo-oxidation mechanism of the composite. Fire retardant-loaded samples had smaller color change compared to the unfilled one. The tensile properties of all composites declined after the weathering. Significant changes in the surface morphology of the weathered composites were observed with a scan electron microscope.

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1. Introduction

Wood–plastic composites (WPCs) are generally used for outdoor applications, and hence their overall performance are affected by ambient climate conditions. It has been shown in many studies that composites subjected to accelerated and natural weathering show loss of mechanical and physical properties [1–8]. The weathering process is usually cumulative action of different environmental factors, including sunlight, wetting, temperature changes and so on. High energy ultraviolet (UV) radiation in aerobic conditions acts as a powerful oxidant inducing chemical changes in both wood and polymer. The photo-oxidation of polyolefins is usually started from chromophoric functional groups, largely ketone carbonyl groups, which are capable of absorbing solar energy. Activated by solar energy, ketone groups initiate further reactions which result in polymer macromolecule degradation. The ketone photolysis of polyolefins proceeds through two major reactions, known as Norrish I and Norrish II (Fig. 1) [9]. A polymer macromolecule backbone with a mid-chain carbonyl group degrades via Norrish I to free radicals and chain scission or crosslinking. Chain scission via Norrish II results in vinyl-like terminal groups and carbonyl groups accumulation [9,10]. Chain scission and crosslinking are two opposite reactions; chain scission lowers the molecular weight of the polymer, whereas crosslinking results in increased molecular weight due to free radicals recombining and

creating new polymer chains. Crosslinking does not affect polymer crystallinity, while chain scission can increase crystallinity due to short chains being more mobile and crystallizing more readily [8]. The changes induced by UV radiation have a negative effect on the polymer properties. The crystallinity change in the polymer leads to spontaneous cracking of the surface and material embrittlement [8,11]. In addition, weathering can result in composite bleaching generally related to photodegradation of both the polymer and the wood [12,13]. Among the wood constituent polymers, lignin is the most sensitive to light, whereas pure cellulose is not a good light absorber. However, there are carbonyl groups or other chromophores present in the cellulose structure; they can be introduced during cellulose purification or isolation [14]. The photo-oxidation process of cellulose results in chain scission, dehydroxylation, dehydroxymethylation and dehydrogenation, which are responsible for the formation of different radicals [15]. The problem in the UV degradation of materials can be partly solved by the incorporation of UV-stabilizers [8,16–18], hindered amine light stabilizers (HALS) [17] and pigments [1,12,16].

Another important issue for WPCs are their thermal stability and fire retardancy properties. Today, different fire retardants (FRs) are applied in the composite technology. FRs are usually loaded in large amounts and they can influence the composite properties, including long-term durability [19,20]. Sinturel and co-workers have published a series of works where they examine the photo-oxidation of fire retarded polypropylene (PP) [21–23]. They have shown that aromatic halogenated FRs alone or in combination with Sb_2O_3 facilitate polymer photo-oxidation.

* Corresponding author. Tel.: +358 40 5283583; fax: +358 5 4321711.

E-mail address: irina.turku@lut.fi (I. Turku).

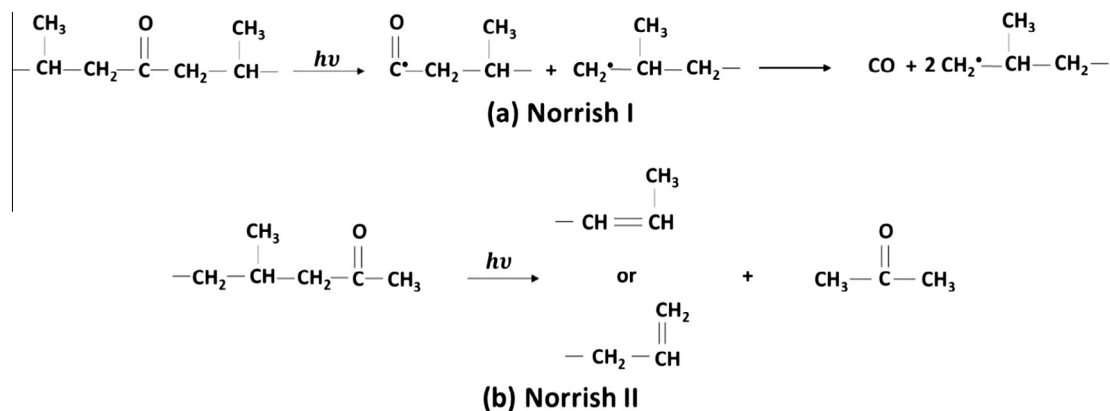


Fig. 1. Degradation of polypropylene backbone containing carbonyl groups.

The decreased photostability is attributed to the formation of reactive radicals due to FR decomposition under UV radiation. Mechanism of oxidation of PP was not change by the presence of FR. Sinturel et al. also report that the efficiency of HALS in improving the stability of PP is reduced due to the interaction of the FR with the stabilizer [24]. Microencapsulation of halogenated FRs has been proposed as an alternative to avoid the impact of brominated flame retardants on the stability of light [20]. Halogen-free FRs, such as metal hydroxides and nitrogen- and phosphate-based, can be classified as environmentally friendly due to the fact that they do not produce toxic gases during combustion like halogenated FRs do. It has been shown that melamine and ammonium polyphosphate (APP), also degrade under UV radiation [25–27]. They, however, do not produce free radicals and do not show antagonism with stabilizers [28]. APP has been reported to have no inductive effect on the oxidation of PP, and the stabilizing effect HALS is not reduced, either [27]. Garsia et al. [29] report that APP, melamine cyanurate and $\text{Al}(\text{OH})_3$ increased the sensitivity of polyethylene-based WPC to UV light, but in the combination with stabilizers the discoloration of the composites was decreased. Yet, the durability of FR is dependent on their solubility, as the FR can leach from the composite surface during the exposal of the composite in humid conditions [30].

The effect of FRs on the flammability and mechanical and physical properties are the main goals of the research of fire retarded composites. The effect of these fillers on durability is less studied, despite the obvious importance. The main objective of this study is to determine the effect of fillers, such as aluminum trihydrate (ATH), zinc borate (ZB), melamine, graphite (G) and titanium dioxide (TiO_2) on the durability of polypropylene (PP)-based co-extruded wood–plastic composites (WPCs). The composites were exposed to accelerated weathering by using a xenon-arc lamp source and water spray during 1000 h. Fourier transform infrared (FTIR) analyses, color change measurements with a spectrophotometer and mechanical tests were performed for the characterization of the composites. In addition, deterioration of the composite surface was studied by means of electron microscopy.

2. Experimental

2.1. Materials

A polypropylene (PP), Eltex P HY001P (Ineos), density 0.910 g/cm^3 and melt mass-flow rate 45 g/10 min ($230 \text{ }^\circ\text{C}/2.16 \text{ kg}$), was used in the experiments. The coupling agent was maleated anhydride PP (MAPP), Orevac[®]CA 100 (Arkema). Struktol TPW 113 was used as the lubricant. The recycled pulp cellulose (PC) was

delivered by UPM, Finland. Zinc Borate, $4\text{ZnO} \cdot 6\text{B}_2\text{O}_3 \cdot 7\text{H}_2\text{O}$, particle size $d_{\text{mean}} = 5 \text{ }\mu\text{m}$, 99% less than $25 \text{ }\mu\text{m}$, was obtained from Chemtura, Switzerland. Melamine, grade F 40, solubility 0.3 g/100 ml , was obtained from EcoChem Technologies, Belgium. Aluminum trihydrate (ATH) with particle size $d_{50} = 1.3\text{--}2.3 \text{ }\mu\text{m}$, $d_{90} \leq 4.5 \text{ }\mu\text{m}$; solubility $1.5 \cdot 10^{-3} \text{ g/1000 ml}$, was obtained from Albemarle Corporation, Germany. Graphite, Silvershine, was obtained from Skaland Graphite As, Norway; and TiO_2 masterbatch, Plastwhite 7038, TiO_2 :PE (polyethylene) = 50:50, was acquired from Cabot Clariant, Finland.

2.2. Processing

Six different composite materials having the same core material and differences in the shell layer were produced. The core layer was produced from a blend with the percentage ratio of WF (wood flour): PP:MAPP:UV-stab: lubricant:pigment:talc = 63:20:5:1:1.5:1.5:8. The materials and formulations used in the shell are listed in Table 1. A co-extrusion system, the Weber CE 7.2 counter-rotation conical twin-screw extruder, was used to produce the core layer. A fiberEX extruder was used to produce the shell layer. The processing temperatures in both extruders were between 174 and $202 \text{ }^\circ\text{C}$. The schematic co-extrusion profile is shown in Fig. 2.

2.3. Testing and analysis

2.3.1. Xenon-arc lamp accelerated weathering

Accelerated weathering tests were conducted in a Q-Lab Xenon Test Chamber, Model Xe-3-HS. The weathering procedure consisted of 102 min of UV irradiation (with average irradiance of 0.51 W/m^2 at 340 nm) at chamber temperature of $38 \text{ }^\circ\text{C}$ and $(50 \pm 10)\%$ relative humidity, followed by 18 min of water spraying, according to ISO 4892-2:2013 standard.

2.3.2. Tensile properties analysis

The tensile properties of the samples before and after aging were determined according to ISO 527-1:2012 in a Zwick/Roell Z020 testing machine. The xenon-arc light weathering test was carried out with 6 sample replicates.

2.3.3. FTIR spectral analysis

Spectrum 100 FTIR spectrometer (Perkin-Elmer, UK) equipped with an attenuated total reflection (ATR) accessory (Perkin Elmer) was used for the composite surface analysis. The spectra were collected by co-adding 20 scans at a resolution of 4 cm^{-1} in the range from 4000 to 400 cm^{-1} . All spectra were normalized by 2917 cm^{-1} (C–H band), the specific peak intensity of PP. This peak was

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