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The effect of ultraviolet light stabilizers on color stability, melt properties and tensile properties of mixed waste plastics blends

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Abstract.

Plastic waste disposal is among the most challenging problems of the current era. Therefore, new methods and applications for the utilization of waste plastics are increasingly needed. To find them, it is essential to research and develop the material properties of recycled plastics. The effect of different ultraviolet light (UV) stabilizers on the color stability, melt properties and tensile properties of mixed waste plastics blends was studied in this paper. The mixed waste plastics collected from two different waste sources were prepared as specimens by injection molding, and studied with two different types and loading amounts of UV stabilizers. UV absorbers (UVAs) and hindered amine light stabilizers (HALS) were used as UV stabilizers. A specimen produced without the addition of a UV stabilizer was used as a reference specimen of both the blends. After the accelerated weathering, the addition of a UVA provided an improved, smaller change in color than the addition of HALS. Among the tensile properties, the addition of UV stabilizers clearly improved the tensile strength and tensile modulus for almost all the studied specimens. Additionally, the melt properties of both the studied plastic blends were found to be increased by the addition of UV stabilizers.

1. Introduction

Tightened environmental regulations and laws concerning waste materials have been implemented in several countries and companies [1], which has forced them to improve and encourage material efficiency and the recycling of wastes.. For example, the new plastic strategy of the EU outlines that all plastic packaging placed on the EU market shall be either reusable or recyclable by the year 2030 [2].Compared to other materials, such as metals, paper and glass, the recycling of end-of-life plastics is still quite low. In the EU, the latest data shows that 32.5% of plastic waste is recycled and 42.6% recovered through energy-from-waste [3]. The rate of plastic recycling is predicted to increase up to 55% by 2030 [2]. A large amount of various plastics accumulates from construction and demolition waste (CDW) and municipial solid waste (MSW) in which packaging is the principal source of plastic waste. Waste electric and electronic equipment (WEEE) is also a notable source of plastic waste.

Polymer blends are generally classified into either miscible (homogenous) and immiscible (heterogeneous) blends. Most of the polymers are incompatible by nature and, therefore, produce immiscible polymer blends that have separated phases and glass transition temperatures of the individual polymer components because the polymers are not mixed at the molecular level. Some examples of immiscible blend are polypropylene (PP)–polyethylene (PE) and polypropylene (PP)–polystyrene (PS) blends. By contrast, miscible blends, such as polystyrene (PS)–polyphenylene oxide (PPO) blend, are mixed at the molecular level, and they also have properties of the averages of the

individual polymer components, and a single glass transition temperature as well as a single phase [4,5].

Plastics or plastic products have a very wide variety of outdoor applications where polymers are susceptible to weathering factors, such as photodegradation caused by ultraviolet radiation, moisture and freeze–thaw action, which can significantly change the color/appearance and mechanical properties of polymers. The temperature has a great effect on the oxidation of plastics by accelerating the degradation process strongly with higher temperatures, whereas moisture does not have an equal role in the degradation of polymers [6]. UV radiation and heat are normally linked under natural circumstances [7]

The UV region of solar radiation has the greatest effect on the chemistry of polymers because they absorb UV radiation, which leads to a reduction in the molecular size of polymers and the build-up of reactive chemical groups [8]. The photodegradation of polyolefins, i.e., PE and PP, is caused mainly by the introduction of catalyst residues, hydroperoxide groups, carbonyl groups and double bonds during polymer manufacturing. Small amounts of these impurities can be sufficient to induce polymer degradation even without a significant amount of ultraviolet absorption [9]. To improve the resistance of polymers against photodegradation, UV stabilizers are typically incorporated in plastics or composites. The two most common groups of UV stabilizers are UV absorbers (UVAs) and hindered amine light stabilizers (HALS). The UVA absorbs the harmful UV radiation, and transforms it into harmless heat. HALS do not absorb UV radiation, but can inhibit the degradation of the polymer by slowing down the photochemically initiated degradation reactions [10,11].

In recent years, the potential of recycled plastics has increasingly studied to find new opportunities to meet tightened recycling targets. For example, Dorigato [12] reviewed the current state and challenges in the recycling of polymer blends. The study pointed that special attention should be given to recycled blends constituted by polyolefins as they generate the majority of the plastic waste. The UV durability of plastic waste from different sources has been object of several research articles, e.g., Campos de Bomfim et al. [13,14] studied plastic waste obtained from espresso coffee capsules and observed that recycled PP capsules had a good weathering resistance and, with the addition of natural reinforcement, also improved tensile strength without changes in elastic modulus and toughness values. Ronkay et al. [15] investigated plastic waste, i.e. poly(ethylene terephthalate) (PET) bottle material and high-density polyethylene (HDPE) cap material, from marine environment and found that after accelerated weathering of 2520 h the studied materials were damaged as a result of UV radiation. Great efforts, e.g., [16–18], have also been made to explore the potential of waste plastic blends as a raw material for polymer composites.

Polymer blending is a cost-effective and unique way to develop new polymeric materials or compounds compared to direct polymer synthesis [19]. During the recent decades, polymer blends has been widely studied by researchers and industries. The research has mainly focused on durability and degradation of polymer blends as well as improving the properties of polymer blends with compatibilizers. Regarding the recyclability of plastics, the effect of reprocessing on the performance of polymer blends has been investigated in several studies, e.g. [20–23]. However, the scientific literature does not report much data on studies about the effects of UV stabilizers on the properties of recycled polymer blends, while polymer blends containing various virgin polymers have been studied extensively, as reported by La Mantia et al [24]. Only a few studies published by Al-Salem et al. [25,26] have investigated UV stability of polymer blend containing recycled plastics. The modification or controlling of the properties of virgin polymer blends is clearly simpler than it is for recycled polymer blends. Recycled blends usually consist of several different types of plastic and their proportion in the blend can vary significantly. Also, plastics may have been contaminated during previous usage. This poses challenges to managing the properties of recycled blends.

More efficient utilization of recycled plastic blends, especially in outdoor applications, requires more information on their behavior under UV exposure. As known, UV radiation is detrimental to polymeric materials. The temperature has a great effect on the oxidation of plastics. A higher temperature accelerates the degradation process strongly. The photodegradation results in surface cracking and significant loss in strength and stiffness of plastic material [27]. UV resistance of virgin polymeric materials has been extensively studied and developed for several decades. Also, the UV behavior of plastic blends made of virgin polymer materials is well established. Today, plastic products are increasingly made of recycled plastics. Improving the UV behavior of a product containing one type of recycled polymer is much easier than that of a product made of recycled polymer blend. Therefore, the UV resistance of recycled polymer blends should be investigated more closely. Reliable and comprehensive research data on UV resistance can be used to ensure the safe use of products made of recycled plastic blend and to develop their properties to better meet the requirements of outdoor applications. Curtzwiler et al [28,29], for example, reported a remarkable increase in UV absorption potential for post-consumer recycled polyolefins compared to virgin polymers. This indicates that UV protection of recycled plastics should be carefully considered when using them in new and demanding outdoor applications. Determination of UV stabilizers in recycled polymers represents rather a difficult problem. Various analytical methods can be used to determine the type of stabilizers already present in plastics. For cost-effective utilization of recycled polymer blends without separate analysis, it would be important to find a stabilizer or the combination of stabilizers that could be used in blends of different compositions. Pospíšil et al. [30] reviewed upgrading of recycled plastics by restabilization and reported that without restabilization recycled PE showed a significant loss of crack resistance and tensile impact strength. Also, the addition of 0.1 % HALS resulted in mechanical performance compareble to that of the virgin polymer. For polymer blend consisting of polyolefins, styrenics and PVC, the combination of UVA and HALS was observed to ensure stability in outdoor applications.

The main objective of this paper is to provide novel information on the effect of UV stabilizers on recycled polymer blends and their different compositions. Even though many researchers have worked on polymer blends or waste plastics, very few researchers have reported about UV stability of recycled polymer blends. In addition, those papers have merely studied plastic blends that combined virgin and recycled plastics. In this present work, polymer blends made fully of recycled plastics are investigated. The research data are very useful in developing the performance of recycled plastic blends to meet growing demands of future applications. The study compares the performance of two UV stabilizers with different protection mechanism in two different plastic blends. In this study, mixed waste plastic blends were injection-molded with and without UV stabilizers, and exposed to accelerated weathering. The effects of UV exposure were investigated by analyzing color stability as well as melt and tensile properties.

2. Materials and Methods

Materials

The mixed waste plastics used were gathered from two different sources, named F and P, destined for further processing at a local recycling plant, located at Lappeenranta, Finland. Before arriving at the processing plant, the mixed plastics were mechanically sorted from CDW and MSW streams at a waste sorting plant. The plastic waste of the source F consisted mainly of household plastics, while the source P consisted mainly packaging waste. The studied waste plastic batches were collected randomly. Also, there was no information on the previous use or possible additives of the plastics.

The identification of plastic waste materials was performed manually with a portable NIR analyzer (Thermo Scientific microPHAZIR PC, Thermo Fisher Scientific, Waltham, MA, USA) that identifies material in less than three seconds in the spectral range of 1600–2400 nm without special specimen preparation. The share per component in the material stream was determined based on weight.

After separation, the plastic materials were reduced to approx. 4.0 mm flakes with a Shini SG-1635N low-speed granulator. After size reduction, the crushed fractions were mixed again to match the distribution of the original blend. The blends were named as a blend F and a blend P.

The compounding of the blends was performed using a BOY 30 injection-molding machine. Two different types of UV stabilizers were applied: a low molecular weight hindered amine light stabilizer (HALS) Tinuvin® 770 DF (BASF, Germany) and an ultraviolet light absorber (UVA) Chimassorb® 81 (BASF, Germany) with loadings of 3 and 5 wt%. The reference specimens were produced without the addition of UV stabilizers. The composition of the studied specimen is shown in Table 1. Except for reference specimens (FRef and PRef), the specimen names follow a formula in which the first character defines the blend type, the second character indicates the amount of UV stabilizer and the third character expresses the type of UV stabilizer used.

Table 1. Composition of specimens produced from the mixed plastic blends.

Blend F	UV stabilizer [wt%]
FRef	-
F3C	3% UVA
F5C	5% UVA
F3T	3% HALS
F5T	5% HALS

Blend P	UV stabilizer [wt%]
PRef	-
P3C	3% UVA
P5C	5% UVA
P3T	3% HALS
P5T	5% HALS

Analysis

The melt properties of the materials, melt mass-flow rate, melt volume-flow rate and melt density were determined according to the ISO 1133-1 standard. The equipment used for the analysis of melt properties was a Dynisco LMI500 series Melt Indexer. The test temperature was set to 230°C, and 5.0 kg load was used. The values reported are a mean of four results.

Accelerated weathering of the prepared specimens was performed according to the standard EN ISO 4892-2:2013 in a Q-Sun Xe-3 HS (Q-Lab Corporation, USA) test chamber using a xenon-arc lamp. The specimens were weathered for 500 h of exposure. The exposure cycle consisted of 102 min of light exposure followed by 18 min of simultaneous water spray and light exposure.

The surface color of the weathered and non-weathered materials was measured with a Minolta CM-2500d spectrophotometer (Konica Minolta Sensing Inc., Japan). The CIELAB color system was used to measure the surface color in $L*$, $a*$ and $b*$ coordinates. $L*$ represents the lightness coordinate and varies from 100 (white) to 0 (black); $a*$ represents the red ($+a*$) to green ($-a*$) coordinates; and $b*$ represents the yellow ($+b*$) to blue ($-b*$) coordinates. The color difference was calculated as outlined in ISO 7724 according to the following equation:

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}. \tag{1}$$

where ΔL^*, Δa^* and Δb^* represent the differences between the initial and final values of L^*, a^* and b^*, respectively. The surface color of the replicates was measured at three locations on each WPC test specimen.

The tensile properties, strength and modulus were measured in accordance with standard EN ISO 527-2/1A on a Zwick/Roell Z020 tester equipped with testXpert II testing software. All the mechanical and physical tests were carried out with 6 specimen replicates.

Statistical analysis

Statistical significance between the studied specimen data was tested by using Daniel's XL Toolbox Excel software. The data were analyzed by using the one-way ANOVA Bonferroni-Holm post hoc testing algorithm.

3. Results and Discussion

From both the studied sources, F and P, an amount of approximately 5 kg of mixed plastic waste was gathered and manually sorted with an NIR analyzer. Non-plastic materials were removed during the sorting. The sorting results of the studied blends are presented in Table 2. For both blends, the most significant proportion of plastics consisted of polyethylene (PE). In blend F, the distribution of different plastic types was more uniform than that in blend P. For blend F, most of the plastic fractions consisted of PE, approx. 40% of weight. Polyethylene terephthalate (PET), polypropylene (PP) and polystyrene (PS) had relatively similar shares. Additionally, the remarkable proportion of unidentified fractions should also be considered. In blend P, the proportion of PE dominated—approx. 88% of the total weight of the studied blend. The second largest proportion was PP—slightly over 5%. However, it should be noted that the material amount contained in both blends is very small compared to the total amount of wastes; therefore, even large variations in the composition of mixed plastic waste may be probable.

Table 2. Plastic material, polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), acrylonitrile-butadiene-styrene (ABS) and polyethylene/polyethylene terephthalate (PE/PET) shares (% of total weight) in the studied blends.

Plastic type	Blend F [wt-%]	Blend P [wt-%]
PE	40.70	87.85
PET	16.12	1.17
PP	10.33	5.37
PVC	2.07	1.17
PS	13.84	0.23
ABS	0.83	-
PE/PET	2.07	-
Unidentified	14.05	4.21

The analysis of melt properties provides significant information on the performance of a polymer or polymer blends in molten state. Both the melt flow rate (MFR) or melt volume flow rate (MVR) are essential characteristics describing the flow properties of polymeric materials [5]. The melt properties of the studied plastic blends were determined for the reference specimens produced without the addition of UV stabilizers, and for specimens that contained 5 wt% of UV stabilizers. The results,

shown in Table 3, revealed that the addition of a UV stabilizer has an impact on the melt properties of mixed waste plastics. Both the MFR and MVR of the studied blends increased with the 5 wt% addition of UV stabilizer. As the sorting results showed, PE was the predominant plastic type in both of the studied plastic blends. Thus, the impact of PE is also reflected in the melt properties. The measured density and melt flow values are in line with the values of virgin low-density polyethylene (LDPE) or high-density polyethylene (HDPE) given in the literature [31]. The addition of 5 wt% of UV stabilizers had a statistically significant effect on the MFR and MVR of blend P specimens at the 95% confidence level. By contrast, no statistically significant difference was found between the reference specimen of blend F and the specimens with the addition of UV stabilizers. From the processability point of view, the increase in melt indexes indicates a lower viscosity. Low-viscosity materials flow with lower resistance and, therefore, more quickly during melt-processing than high-viscosity materials. In injection molding, high-flow (low viscosity) materials are preferred because they can easily fill demanding flow paths in a mold. Conversely, low-flow (high viscosity) materials are preferred in extrusion because they enable the easier control of the shape of a complex profile.

Table 3. The melt properties of studied plastic blends: s and ns denote statistically significant and no significant changes with a 95% confidence level.

Specimen	Melt Density [g/cm^3]	MFR [g/10 min]	MVR [cm^3/10 min]
FRef	1.34 ± 0.20	4.88 ± 0.05	5.16 ± 0.18
F5C	0.99 ± 0.20	5.01 ± 0.33 [ns]	5.46 ± 0.71 [ns]
F5T	0.82 ± 0.03	6.41 ± 1.22 [ns]	8.54 ± 1.03 [ns]
PRef	0.73 ± 0.04	3.80 ± 0.15	5.25 ± 0.10
P5C	0.73 ± 0.01	5.18 ± 0.13 [s]	7.11 ± 0.13 [s]
P5T	0.74 ± 0.03	5.46 ± 0.14 [s]	7.48 ± 0.63 [s]

As presented in Fig.1 and Fig.2, the behavior of blend F and P during accelerated weathering was more similar for the specimens produced from blend F than from blend P. However, the total color change (ΔE) after 500 h of exposure was lower for the specimens of blend P than the specimens of blend F. For both material blends, the most notable change in color occurred before 50 h of exposure time. For both the blends, the lowest total color changes were observed for the specimens containing UVAs, i.e., specimens F3C, F5C, P3C and P5C. Thus, it can be stated that the addition of a UVA as a UV stabilizer achieves improved results for recycled plastics compared to the addition of HALS, and therefore, it could be considered as a superior UV stabilizer for mixed plastic blends. There was no clear indication from the results that the amount of UV stabilizer added had an observable effect on the total color change. It is essential to take into account that the studied recycled plastics supposedly contained different amounts of UV stabilizers or pigments. For specimens of blend P, the color change decreased by over 35% with the use of HALS, and approximately 70% with the use of a UVA as a UV stabilizer. The results for the specimens of blend F were deviating. Compared to reference specimen FRef, the addition of a UVA decreased the color change by about 20% compared to the addition of HALS with a loading of 3%, which increased the color change by a maximum of 20%. A presumable explanation for this kind of behavior might be the fact that the composition distribution of blend P was clearly more heterogeneous than with mixture F, and therefore, the effect of HALS on the color stability of studied specimens was negative. Investigation [32] into whether UVAs can provide improved UV protection compared to HALS when used individually has been carried out. However, the most effective protection against UV radiation is usually achieved by a combination of different UV stabilizers or protective agents.

It has been shown that the use of UV stabilizers for the restabilization of recycled plastics results in color stability and mechanical performance comparable to virgin polymers after accelerated weathering [30]. When examining the properties of a material containing recycled plastics, it should be considered that the plastics are likely to already contain different amounts of additives, such as

UV stabilizers, fire retardants and lubricants, depending on their previous use. Therefore, it might be difficult to find a clear trend for the change in color.
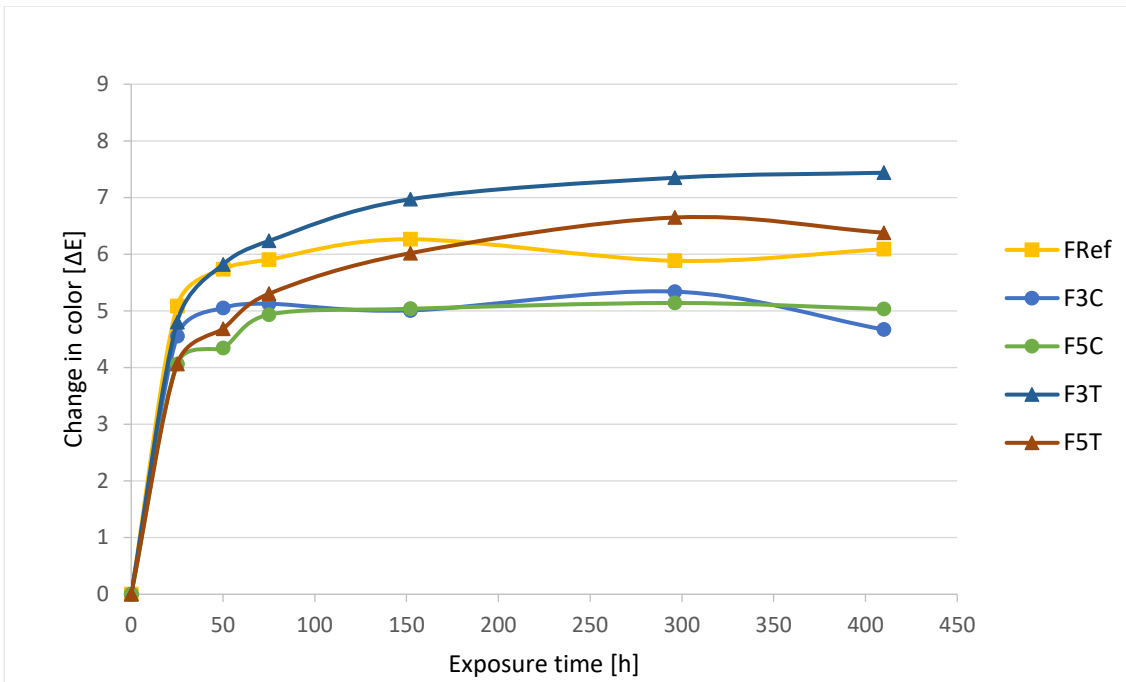

Fig. 1. Change in color (ΔE) in accelerated weathering for blend F.
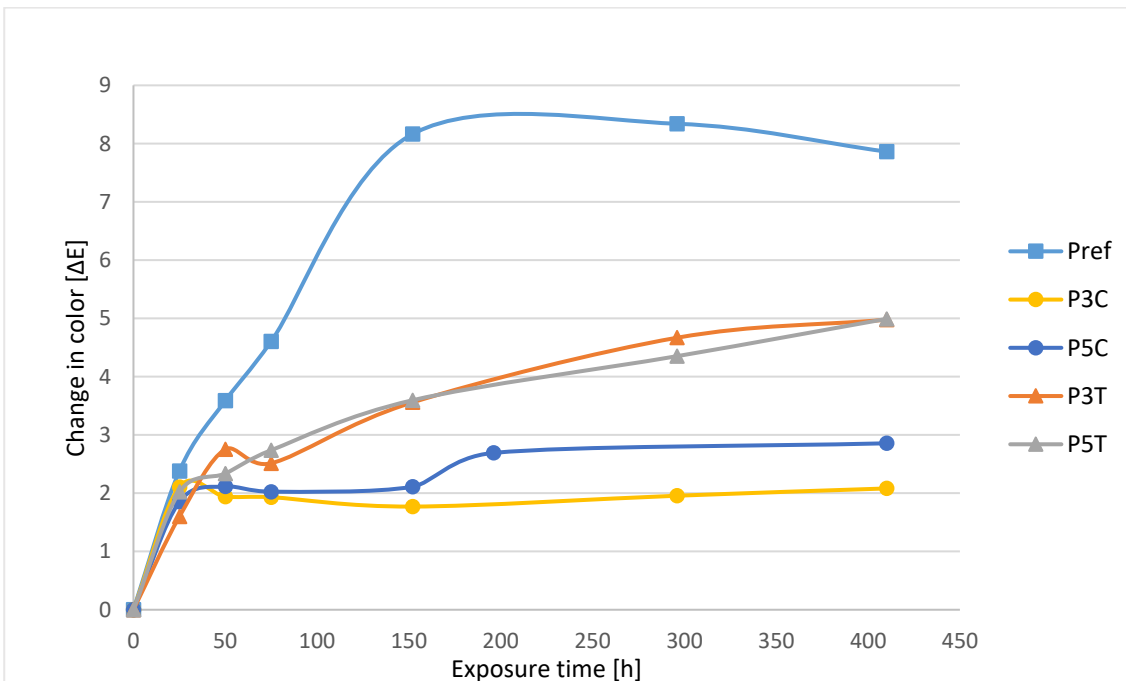

Fig. 2. Change in color (ΔE) in accelerated weathering for blend P.

Polymers are susceptible to weathering. The factor that exerts the greatest effect on weathering is the UV portion (295–400 nm) of solar radiation by changing the chemistry of polymer. This leads to changes in material properties, such as a deterioration in mechanical properties. The tensile properties of the studied mixed plastic blends are presented in Fig. 3, Fig. 4, and Fig. 5.
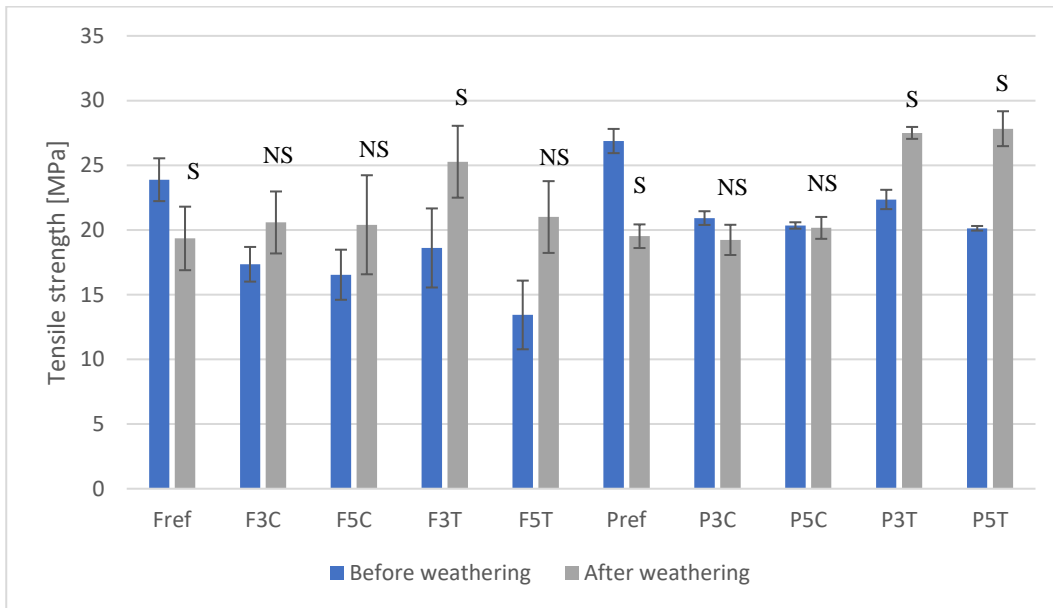
Fig. 3. Change in tensile strength after xenon-arc light accelerated weathering; N and NS denote statistically significant and non-significant changes, respectively.
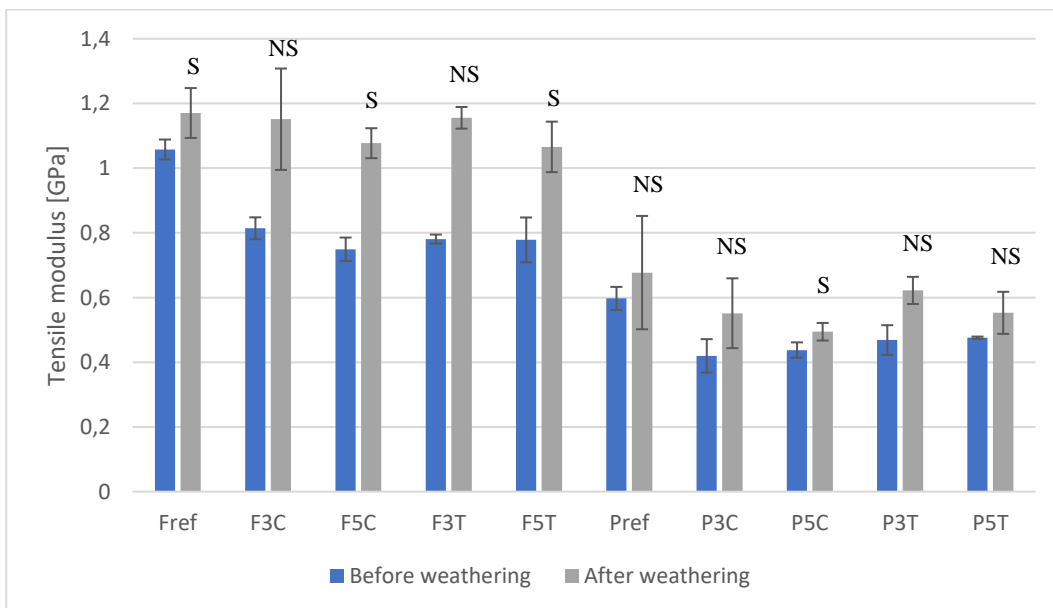

Fig. 4. Change in tensile modulus after xenon-arc light accelerated weathering; N and NS denote statistically significant and non-significant changes, respectively.
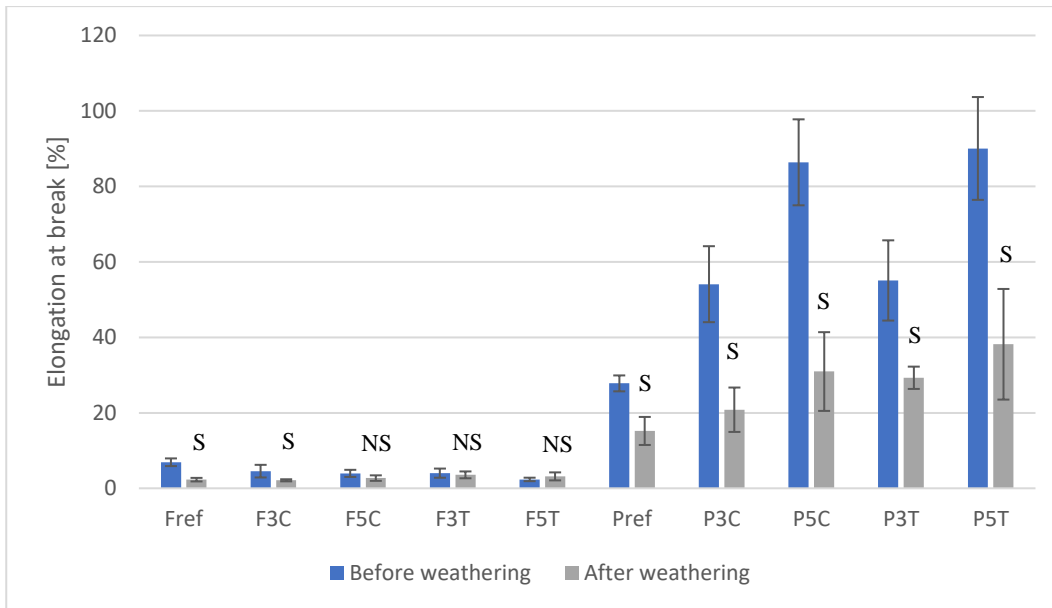
Fig. 5. Change in elongation at break after xenon-arc light accelerated weathering; N and NS denote statistically significant and non-significant changes, respectively.

The research results showed that the tensile properties were improved by the addition of a UV stabilizer after 500 h of accelerated weathering, except the tensile strength of P3C and P5C, which slightly decreased during UV exposure. A greater improvement in tensile properties was achieved by the addition of HALS than that of UVAs. The tensile strength and tensile modulus were observed to increase generally by over 30% with the addition of HALS. The effect of the loading amount of the UV stabilizer on tensile properties varied. The 5 wt% loading of the UV stabilizer provided a higher increase in tensile strength than the 3 wt% addition of the UV stabilizer. For the tensile modulus, the effect of loading amount was different. As shown in Fig. 3 and Fig. 4. the tensile strength and the tensile modulus were generally improved in most of the studied specimens with the addition of UV stabilizers. A significant improvement in tensile properties after accelerated weathering was slightly surprising as previous studies [27,28] has shown that mechanical properties tend to remain at their original level as a result of the use of UV stabilizers. But it has been also reported an increase of 40% in the modulus of elasticity for HDPE after 1300 h accelerated weathering [33]. Generally, the variations in protection ability of the studied UV stabilizers can be explained with the different protection mechanisms of the studied UV stabilizers. UVA absorbs harmful UV light and quickly transforms it into harmless heat energy as against HALS inhibits the degradation of the polymer by preventing the formation of free radicals. As the results revealed, the addition of HALS improved tensile strength more efficiently than the addition of UVA. However, both the UV stabilizers are well suitable for the UV protection of plastic blends. As with pure plastics [33,34], the most effective UV protection for plastic blends is probably achieved with a combination of different UV stabilizers.

Only the reference specimens showed a clear deterioration in tensile strength properties after the weathering. This behavior can be explained with higher crystallinity of waste plastics caused by several heat cycles during accelerated weathering. As a result of higher crystallinity, material became more brittle, leading to a reduction in the elongation at break, as shown in Fig. 5. The elongation at break is typically considered as a good parameter to represent the effect of aging on polymers [33]. That is a result of the scission of the polymer chain caused by UV radiation which reduces the molecular weight of the polymer material and leads to the decrease of the mechanical properties. The basic principle of degradation caused by UV radiation is the same for all the most commonly used polymers. As La Mantia et al. [24] stated the degradation behavior of a plastic blend is very difficult to predict on the basis of the properties of pure plastics as the degradation rate of a blend can result lower, intermediate or higher than that of the pure plastics.

The statistical significance between the specimens with the UV stabilizer and the corresponding reference specimen data was tested with Daniel's XL Toolbox Excel software. The data were analyzed by using the one-way ANOVA Bonferroni–Holm post hoc testing algorithm. Based on statistical analysis, no significant change in both studied tensile properties, i.e., the tensile strength and tensile modulus, was observed for any of the studied specimens with a 95% confidence level. A significant change in tensile strength was observed in specimens F3T, P3T and P5T. Correspondingly, a significant change in the tensile modulus was observed in specimens F5C, F5T and P5T. With one exception (i.e., F5C), all statistically significant changes occurred in specimens using HALS as the UV stabilizer.

The effect of UV exposure on the mechanical properties of recycled or waste plastics has been widely studied, especially for PET waste. For example, Attwood et al. [35] noticed that the tensile strength of polyolefin blends remained unaffected or slightly improved by 5000 h of UV exposure in accelerated weathering. Ronkay et al. [15] recorded a less than 1% decrease in the tensile strength and tensile modulus under 2520 h of accelerated weathering for poly(ethylene terephthalate) (PET) bottle waste material. However, Al-Salem et al. [36] studied the effect of accelerated weathering on the modulus for a linear low density polyethylene (LLDPE)-plastic waste blend at different exposure times, and found that higher loadings of waste plastics significantly increased the modulus.

For different applications, the properties of waste plastic blends can be tailored by mixing with different polymers or with the same virgin plastic in various loadings [18]. A large amount of research has been carried out to improve the properties of plastic blends consisting of virgin polymers and waste plastics. Navarro et al. [37] investigated the optimal ratio for blending virgin HDPE and PET waste without diminishing the final properties, and reported poor compatibility between HDPE and waste PET in the blend. They found that the tensile strength and elongation at break of blends remained at the level of virgin HDPE, as long as the proportion of HDPE did not exceed 5%. La Mantia et al. [38] studied the effect of the addition of small amounts of poly(lactic acid) (PLA) on the recycling properties of PET waste, and found that with an increase of less than 5% of PLA, the tensile modulus of recycled PET increased. Higher loadings of PLA resulted in poor compatibility between the components.

Conclusions

The effect of UV stabilizers on the color stability, melt properties and tensile properties of mixed waste plastics blends was studied. The findings revealed that the addition of a UV stabilizer decreased the change in color after 500 h of accelerated weathering. Of the studied types of UV stabilizers, the UVA showed improved color stability compared to the HALS for both the blends studied. The results also showed that the loading amount had not an observable effect on the total color change. Of tensile properties, tensile strength and modulus were found to be improved by both UV stabilizers studied. The most uniform increase was achieved with HALS. The effect of UV stabilizers on elongation at break was mainly negative. It was also observed a very significant difference in elongation between the blends. The melt properties were also found to improve with the addition of a UV stabilizer. However, it should be noted that the composition of the mixed plastic can vary significantly. To harmonize the effect of UV stabilizers on the material properties of mixed waste plastic blends, the composition of the plastic mixture should be homogenized to the greatest possible extent. Also, extensive research should be carried out with different types and amounts of UV stabilizers to find the most suitable combinations for effective UV protection of recycled plastic blends. The utilization of recycled plastics decreases the need for virgin plastics, as well as reduces the environmental impact of wastes.

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