



Research Paper

Impact of bioplastic contamination on the mechanical recycling of conventional plastics

Michael J. Staplevan^a, Ashley J. Ansari^{a,b}, Aziz Ahmed^c, Faisal I. Hai^{a,*}^a Strategic Water Infrastructure Laboratory, School of Civil, Mining, Environmental and Architectural Engineering, University of Wollongong, Wollongong, NSW 2522, Australia^b Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, NSW 2007, Australia^c School of Civil, Mining, Environmental and Architectural Engineering, University of Wollongong, Wollongong, NSW 2522, Australia

ARTICLE INFO

Keywords:

Bioplastic
Plastic recycling
Contamination
Circular economy

ABSTRACT

Quality assurance of a recycled product is currently one of the biggest issues that the plastic recycling industry faces. The purity of the input plastic waste stream has significant influence over the quality of the recycled product. This research evaluated the impact of polylactic acid (PLA) contamination within the input waste stream of high-density polyethylene (HDPE) recycling. The ultimate tensile strength was noted to reduce by 50% when PLA contamination was at 10%. An investigation into the effect that UVA radiation (simulating solar radiation) has on HDPE contaminated with PLA was also performed to determine the long-term effect of the bioplastic contamination. After UVA treatment, the ultimate tensile strength was reported to reduce by 51% when PLA contamination was only at 2.5%. A water contact angle analysis indicated the PLA contamination increased the hydrophilic nature of the HDPE sheets, potentially creating issues if the intended use of the recycled product was to store liquids. Microscopic analysis of the HDPE sheets contaminated with PLA showed deformations, ridges, cracks, and holes appear on the surface due to the immiscibility of the two polymers that was confirmed by FTIR analysis. Colour changes were visibly noted, with UVA exposure increasing the rate of colour change. Based on the findings in this study, PLA contamination of even 1% in a HDPE waste stream would significantly reduce the quality of the recycled product.

1. Introduction

Plastic recycling is a crucial element for the transformation from a linear economy to a circular economy whereby the used plastics have potential economic value for a number of life cycles if disposed of properly. The dependency on fossil fuels can be reduced by keeping plastic material in the production market and out of landfills. Plastic recycling has also been found to result in a reduction in greenhouse gas emissions by reducing the need to manufacture virgin polymers (Chen et al., 2011). However, since the process of large-scale plastic recycling is still relatively new, associated problems with recovery rates and quality of the outputs are still frequent (Stapleton et al., 2023a). Research into optimising the plastic recycling process has become a topic of interest within the industry as many nations around the globe aim to increase their plastic recycling rates (d'Ambrières, 2019; Fehér et al., 2022; Staplevan and Hai, 2024; Vollmer et al., 2020).

Plastic waste can be recycled in three distinct ways, namely,

mechanical recycling, chemical recycling, and energy recycling (Al-Salem et al., 2010; Hahladakis and Iacovidou, 2019; Merrington, 2017). Mechanical recycling of plastics is currently the most common practice seen in industry due to its simplicity and environmental benefits (Maris et al., 2018). The research and development associated with chemical recycling (e.g., via pyrolysis or gasification) and energy recycling (Volk et al., 2021) has notably increased in the past decade to potentially fill the recovery gaps that are missed through the mechanical recycling process (Huang et al., 2022; Thiounn and Smith, 2020). However, this study is focused on mechanical recycling which is currently the most popular choice to recycle plastic waste.

A significant issue faced by the mechanical plastic recycling industry is contamination in the input waste streams, specifically contamination of different varieties of plastics (Pivnenko et al., 2015). Currently there are seven groups of plastics available, with the group number seven labelled as “All other plastics” (Shamsuyeva and Endres, 2021). Within this class alone, there are many different polymer types and polymer

* Corresponding author.

E-mail address: faisal@uow.edu.au (F.I. Hai).<https://doi.org/10.1016/j.wasman.2024.05.028>

Received 3 January 2024; Received in revised form 26 April 2024; Accepted 20 May 2024

Available online 29 May 2024

0956-053X/© 2024 The Author(s). Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

blends. With particular interest in mechanical recycling, a homogeneous input waste stream is important. Current research has focused largely on the effect of conventional plastic contamination in the waste stream, e.g., polypropylene (PP) contaminating a polyethylene terephthalate (PET) waste stream (Tim and Philip, 2015). Although bioplastics only currently make up about 1 % of the plastic manufactured globally, the global market for bioplastics is expected to double from 2018 to 2025, (Varghese et al., 2022). As there is a push to move towards the use of bioplastics in society, there is an ever-increasing risk of bioplastic contamination in the recycling of conventional plastics (Neves et al., 2020).

Although plastic separation techniques are improving significantly, there is still the potential for contamination to occur. A review of near-infrared spectrometry used to separate bottles made up of a type of bioplastic (polylactic acid, PLA) from a stream of waste PET bottles, found the technology to have a separation efficiency of 86–99.6 % (Alaerts et al., 2018). Researchers have begun to investigate the effect of bioplastic contamination on conventional plastic recycling (Åkesson et al., 2021; Aldas et al., 2021; Gere and Czigany, 2020; La Mantia et al., 2012; Samper et al., 2018; Van Velzen et al., 2022). PET contaminated with PLA has been found to have detrimental effects on the quality of the recycled material when contamination concentration was only at 1 wt% (Åkesson et al., 2021).

As high-density polyethylene (HDPE) is one of the most recycled plastics globally, it is vital to understand the impact that bioplastic contamination may have on recycled HDPE. The high use and popularity of PLA (Gere and Czigany, 2020) makes it the most plausible bioplastic that may contaminate the plastic waste stream. Current literature has focussed on the effect of thermoplastic starch blends contamination on HDPE recycling (Åkesson et al., 2021) or the effect of PLA contamination on the recycling of mixed polyethylene (MPE) films (Gere and Czigany, 2018). As far as the authors are aware, there is no current literature investigating the effects of PLA contamination on the quality of recycled HDPE. This study aims to fill this knowledge gap and provide relevant information to the plastic recycling industry about the implications of PLA in the input waste stream. Furthermore, the effect of UVA on any conventional plastic contaminated with a bioplastic has not been investigated within the current literature. This is a critical factor in assessing the implications of bioplastic contamination on the longevity of a recycled plastic product. Therefore, an investigation into the effect of UVA exposure on HDPE contaminated with PLA will be performed to determine the long-term impact of bioplastic contamination on plastic durability. The purpose of this research is not to disparage bioplastics but to highlight the importance of a homogenous waste stream on producing high quality recycled materials that support the transformation into a circular economy.

2. Materials and methods

2.1. Experimental protocol

The plastic recycling process was replicated to evaluate the impact of bioplastic contamination in the waste stream of conventional plastics. To do this, plastic material was purchased, shredded, dried, and remanufactured into sheets. The resultant material was analysed to determine

the effect that contamination has on the quality of the final product. The experimental protocol is outlined in Fig. 1.

2.1.1. Plastic processing

High-density polyethylene (HDPE) bottles and polylactic acid (PLA) cups were commercially purchased and used to simulate the input waste streams at a plastic recycling facility. The purity of the polymers was confirmed by FTIR analysis as discussed in Section 3.2. The plastic was shredded into flakes using a GP20 hybrid shredder (3devo, The Netherlands). All shredded material was oven dried at 60 °C for 12 h to ensure the moisture content was below 0.1 wt% (Shen and Worrell, 2014).

2.1.2. Laboratory scale hot press

HDPE plastic sheets were manufactured using a hot press (MIR Press, Australia) moulding technique, which is a common practice when making composite plastics (Adhikary et al., 2008; Liu et al., 2021). To determine the melting points of HDPE and PLA a single run differential scanning calorimetry (DSC) analysis was performed. The temperature of the heat press was set to 175 °C. A 1 mm stainless steel spacer was used to maintain a uniform thickness between the recycled sheets. Plastic sheets were fabricated at six different polymer blend concentrations by weight (HDPE_{100%}, HDPE_{99%} + PLA_{1%}, HDPE_{97.5%} + PLA_{2.5%}, HDPE_{95%} + PLA_{5%}, HDPE_{90%} + PLA_{10%}, PLA_{100%}).

2.1.3. Artificial ageing process

Artificial aging of the contaminated HDPE sheets was performed by simulating photodegradation. A UVA chamber (400 h × 600w × 300d (mm)) was fabricated using two 10 W UVA lights that simulated natural sunlight by emitting light at wavelength of 340 nm (Supplementary Figure S1). Plastic sheets were placed in the UVA chamber for 28 days and were flipped every seven days to evenly distribute the UVA irradiation across the plastic sheets. The samples were placed 150 mm away from the UVA lights within the chamber so the theoretical light irradiance on the surface of the plastics was 3.5 mW/cm² (Stapleton et al., 2023a). After the artificial ageing process had concluded, the plastic sheets were stored in a location that received no UVA radiation. All samples were analysed within two weeks of the conclusion of the artificial ageing process.

2.2. Analytical methods

2.2.1. Differential scanning calorimetry (DSC) analysis

A single run DSC analysis was performed using a DSC-214-Polyma (Netzsch, Germany) to determine the melting points of HDPE and PLA (Gill, Moghadam & Ranjbar, 2010). 10–20 mg samples were heated from 30–300 °C at the rate of 10 °C/min in the presence of nitrogen gas. The analysis was repeated in triplicates (n = 3) for repeatability.

2.2.2. Fourier transform infrared (FTIR) spectroscopy

FTIR analysis was undertaken on the fabricated composite plastic sheets (HDPE-PLA) to characterise the functional groups at different PLA concentrations (Solechan et al., 2022). FTIR analysis was completed using an IRAffinity-1S fitted with a MIRacle-10 ATR (Shimadzu, Japan). Using LabSolutions IR software, FTIR plots were generated and recorded

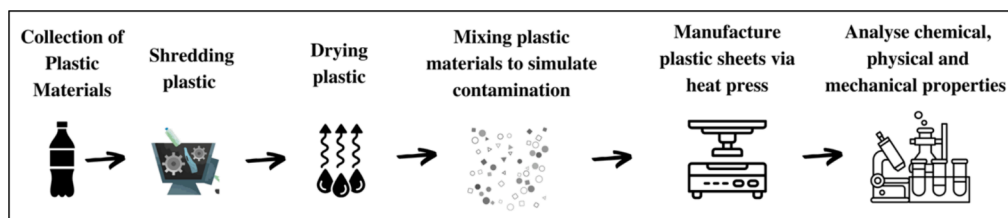


Fig. 1. Experimental protocol to investigate the impact of bioplastic contamination in the waste stream of conventional plastics.

in terms of absorbance values. Each sample was subjected to 16 scans at a resolution of 8 cm^{-1} across the targeted infrared spectrum range ($4000\text{--}600\text{ cm}^{-1}$). Three different locations ($n = 3$) were sampled on the plastic sheets to check the duplicability and consistency of the results.

2.2.3. Water contact angle analysis

A contact angle goniometer analysis (ramé-hart, USA) was performed to determine if PLA contamination affected the wettability of the HDPE sheets. 0.01 mL of water was dropped onto a cut out segment ($15 \times 15\text{ mm}$) of the plastic sheet. The water contact angle was measured using DROPimage Standard Software (ramé-hart, USA). Measurements were recorded in triplicates ($n = 3$).

2.2.4. Tensile testing

Plastic sheets were water jet cut into dog bone shape samples which replicated the profile of shape type IV from the ASTM D638 (19 mm wide and 115 mm long). The ultimate tensile strength of the plastic samples was determined through the use of an Instron 3367 universal tensile testing machine that was equipped with a 30 kN load cell. The samples were gripped to ensure that the gauge length was 25 mm. The loading rate was set to 10 mm/min for all specimens. Tensile tests were performed in triplicates ($n = 3$).

2.2.5. Stereo microscopic imagery

Characterisation of the surface for each contamination blend was performed using a Leica EZ4W stereo microscope (Wetzlar, Germany). The microscopic images were digitalised through the Leica Application Suite v4.12 (Stapleton et al., 2023b).

2.2.6. Colour change analysis

To determine the effect that PLA contamination has on the colour of HDPE sheets, a novel colour analysis was conducted (Stapleton et al., 2023a). A HP scanner (600 dpi) was used to generate digital colour images of the plastic sheets. The colour change analysis was performed by using a digital colour meter software (Apple, USA, version 5.22) which measured the L^* (lightness), a^* (redness) and b^* (yellowness) of the plastic sheets. Applying the values discovered (L^* , a^* , b^*) to the CIELab colour difference formula (Robertson, 1977), an overall colour difference value (ΔE^*) was established. The CIELab colour difference formula is as follows:

$$\Delta E^* = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2} \quad (1)$$

Where:

$$\Delta L^* = L_C^* - L_T^* \quad (2)$$

$$\Delta a^* = a_C^* - a_T^* \quad (3)$$

$$\Delta b^* = b_C^* - b_T^* \quad (4)$$

Note: C = control sample, T = test sample.

The L^* , a^* and b^* of five different locations on the plastics sheets were averaged and used to determine the overall average colour difference (ΔE^*) when compared to the reference sample (HDPE 100 %). Observable colour changes by the human eye are noted with a ΔE^* value greater than 1 (Ly et al., 2020) which is used within this study as the baseline to assess whether notable colour changes were observed due to PLA contamination. An investigation of the colour change after the samples were subjected to UVA irradiation was included to understand the effect that sunlight has on the recycled material.

3. Results and discussion

3.1. The impact of dissimilar melting temperatures

A DSC analysis was performed to understand the melting temperatures of 100 % HDPE and 100 % PLA. The purpose of this was to

determine if complications would arise during the plastic recycling process due to different melting temperatures. Fig. 2 depicts the results of a single run DSC analysis used to determine the melting temperatures of the two polymers (Tarani et al., 2023). From the results, HDPE_{100%} and PLA_{100%} melted at $134\text{ }^\circ\text{C}$ and $156\text{ }^\circ\text{C}$, respectively. The results discovered through the DSC analysis align with previous literature that has investigated the thermal properties of the two polymers (Aliotta et al., 2022; Blázquez-Blázquez et al., 2019; Gu et al. (2014); Suryanegara et al., 2009).

The mechanical recycling process involves a separation phase, a size reduction phase, a cleaning phase, and a remelting/extrusion phase (Hahladakis and Iacovidou, 2019). When investigating the effect that PLA contamination has on the recycling process of HDPE, DSC is an important tool for determining whether complications will arise in the remelting phase. If plastics have significantly different melting temperatures, there is a high risk of issues arising such as plastic flow being halted in the extruder and contaminants clumping due to not completely melting (Rosato, 2013). Although HDPE_{100%} and PLA_{100%} have peak melting temperatures within $25\text{ }^\circ\text{C}$ of each other (Fig. 2), to maintain a consistent melt during the extrusion phase, optimal operating temperatures are maintained at $50\text{--}60\text{ }^\circ\text{C}$ above the melting point of the polymer (Qenos, 2015). Therefore, for the polymers investigated, optimal operating temperatures would be $184\text{--}194\text{ }^\circ\text{C}$ and $206\text{--}216\text{ }^\circ\text{C}$ for HDPE and PLA, respectively. Based on this technical operations guide, there is a possibility that PLA contamination in a HDPE waste stream may impact the physical processes involved in the mechanical recycling of HDPE plastic.

To date investigations into the effect of bioplastics contamination on conventional *i.e.*, fossil fuel-based plastics have generally only assessed the effect that it has on the recycled product and not explored the effect it has on the recycling process (Åkesson et al., 2021; Aldas et al., 2021; Van Velzen et al., 2022). This research has highlighted that PLA contamination in a HDPE waste stream may impact the physical recycling process.

3.2. Change in chemical composition

FTIR analysis was performed to determine the chemical changes that occur to HDPE when it is blended with PLA material (Riaz and Ashraf, 2014). For the HDPE sample, there are four absorbance peaks (2916 cm^{-1} , 2846 cm^{-1} , 1462 cm^{-1} , and 716 cm^{-1}) that can be used as a reference point to evaluate any change that occurs when PLA is blended into the chemical structure (Fig. 3). The absorbance peaks seen within Fig. 3 correlate to atomic vibrations that occur within the chemical bonds of the sample material when it is exposed to the infrared spectrum (Torres-Rivero et al., 2021). Using FTIR, a polymer can be given a unique spectrum that can be used to investigate if any chemical changes have occurred due to a tested parameter. The C–H stretching region ($3000\text{--}2800\text{ cm}^{-1}$) in HDPE showcases two peaks, 2916 cm^{-1} and 2846

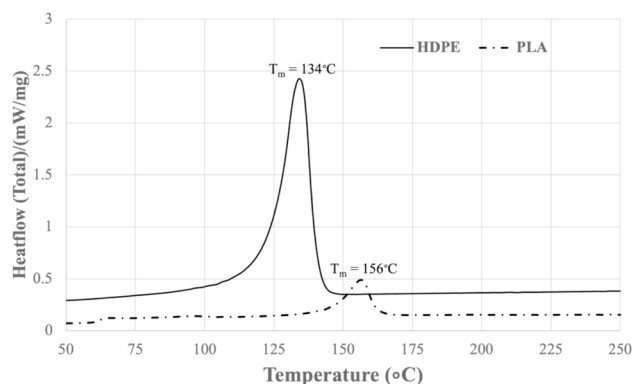


Fig. 2. Single Run DSC analysis of HDPE and PLA.

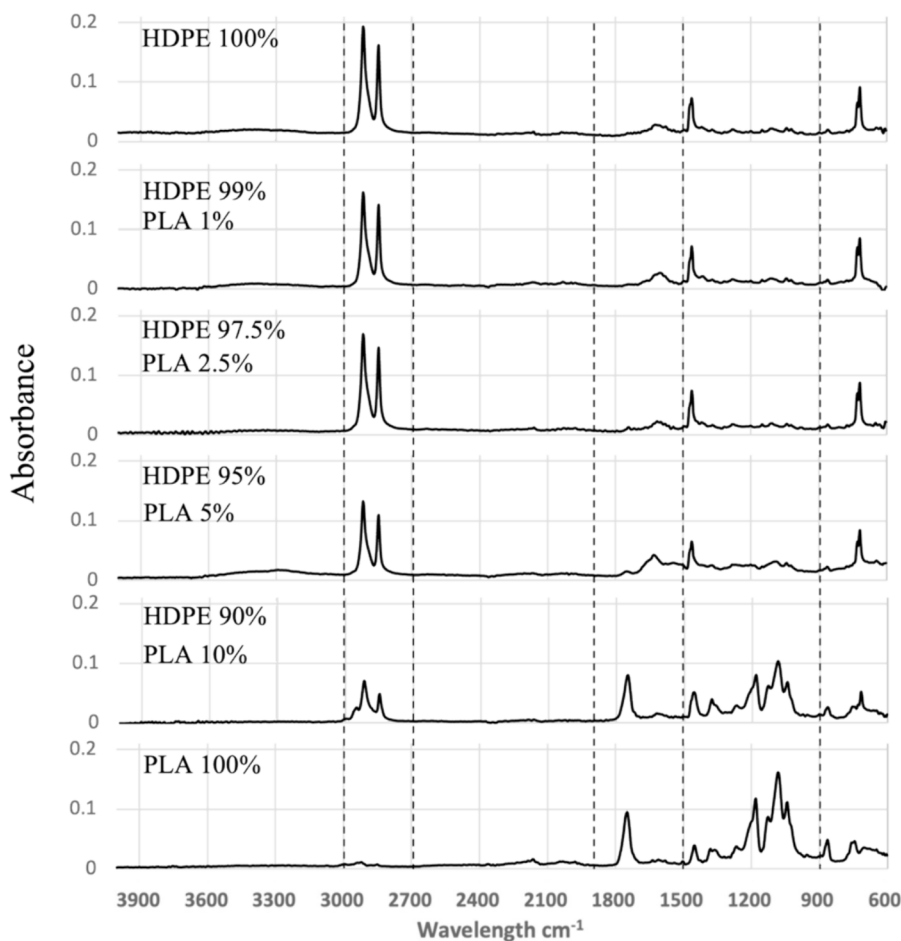


Fig. 3. FTIR analysis illustrating the generation of new absorbance bands, indicating chemical changes occurring within the HDPE chemical structure when it is contaminated with varying levels of PLA.

cm^{-1} , correlating to the CH_2 asymmetric and CH_2 symmetric vibrations, respectively (Smith, 2021). The peaks depicted at wavelength 1462 cm^{-1} and 716 cm^{-1} in HDPE corresponds to the CH_2 scissoring and CH_2 rocking vibrations, respectively (Charles, 2009). By contrast, the FTIR spectrum for PLA (Fig. 3) illustrates one distinct peak at 1744 cm^{-1} corresponding to C = O stretching vibrations (Stapleton et al., 2023a), and a cluster of peaks between 1500 cm^{-1} and 900 cm^{-1} which incorporates the C–H bending vibrations (1446 cm^{-1} and 1357 cm^{-1}) and the C–O stretching vibrations (1180 cm^{-1} and 1080 cm^{-1}) (Chiang et al., 2013). The FTIR spectrums of $\text{HDPE}_{100\%}$ and $\text{PLA}_{100\%}$ illustrated in Fig. 3 are consistent with results produced in previous literature (Chiang et al., 2013; Kai et al., 2019; Rocca-Smith et al., 2017; Stark and Matuana, 2004).

An investigation of the FTIR scans (Fig. 3) of the blended polymers depicted no change between the FTIR spectrums of $\text{HDPE}_{100\%}$, $\text{HDPE}_{99\%} + \text{PLA}_{1\%}$, and $\text{HDPE}_{97.5\%} + \text{PLA}_{2.5\%}$. The results indicate that the PLA contamination was in low enough concentrations that it was not detected in any of the three sample sites on the plastic sheets that were subjected to FTIR analysis. However, when examining the FTIR spectrum of $\text{HDPE}_{95\%} + \text{PLA}_{5\%}$ and $\text{HDPE}_{90\%} + \text{PLA}_{10\%}$, it is clear that the spectrum has peaks from both $\text{HDPE}_{100\%}$ and $\text{PLA}_{100\%}$. Within the FTIR spectrum of $\text{HDPE}_{95\%} + \text{PLA}_{5\%}$, the main peaks associated with $\text{HDPE}_{100\%}$ are still the dominant chemical structures present; however small peaks are noted to appear at 1744 cm^{-1} and scattered between 1500 cm^{-1} and 900 cm^{-1} , which is directly correlated to the fingerprint FTIR spectrum of PLA. The presence of PLA material within the HDPE chemical structure is more clearly seen within the $\text{HDPE}_{90\%} + \text{PLA}_{10\%}$ blend, where both $\text{HDPE}_{100\%}$ and $\text{PLA}_{100\%}$ spectrums are clearly defined

within the blended FTIR spectrum, indicating the significant presence of both polymers within the plastic sheet. Previous studies investigating polymer blends have illustrated similar findings through FTIR analysis. When PP was blended with PLA the resultant FTIR spectrums depicted peaks from both polymers (Samper et al., 2018), as seen within this study.

In general, if two polymers are immiscible, then the FTIR spectrum of the blended materials is the two spectrums of the pure polymers layered on top of each other (Sharma and Kimura, 2003). It was discovered within this study that when PLA contamination was greater than or equal to 5% by wt., the resultant FTIR spectrum was the addition of both homopolymer spectrums, therefore indicating the immiscibility of the two polymers.

3.3. Change in surface morphology

A microscopic analysis was applied to investigate the effect that PLA contamination has on the surface morphology of HDPE (Fig. 4). Both $\text{HDPE}_{100\%}$ and $\text{PLA}_{100\%}$ are seen to have smooth and uniform surfaces. It is noted that the pattern that is seen on the surface of $\text{HDPE}_{100\%}$ and $\text{PLA}_{100\%}$ is from the steel mould used in the hot press process. When 1% PLA is added to HDPE, morphological changes already become present, showcasing the immiscibility of the two polymers. Deformation of the surface morphology of immiscible plastics is stated to be caused by the limited interfacial adhesion between the two polymers, ultimately leading to the clear delineation between the two polymers (Chen et al., 2013). A microscopic analysis of the surfaces of $\text{HDPE}_{97.5\%} + \text{PLA}_{2.5\%}$, $\text{HDPE}_{95\%} + \text{PLA}_{5\%}$ and $\text{HDPE}_{90\%} + \text{PLA}_{10\%}$ indicates large amounts of

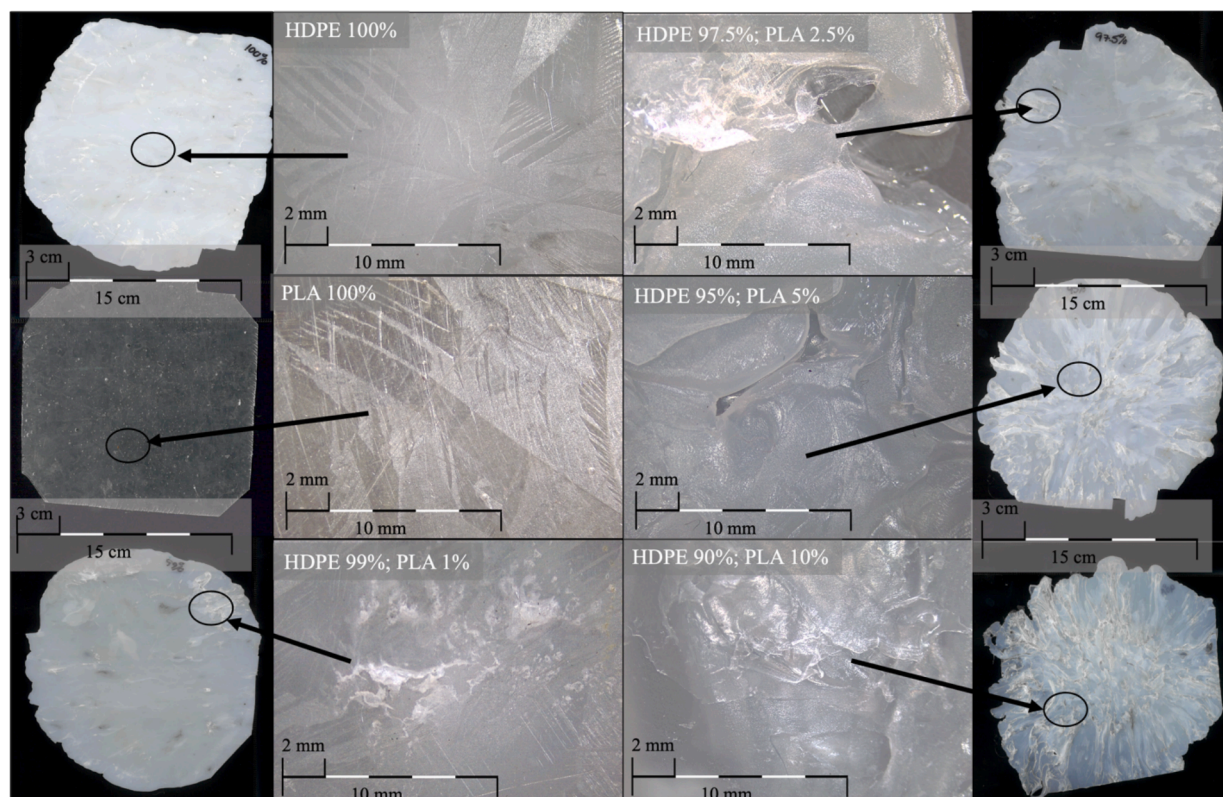


Fig. 4. Microscopic surface investigation to determine the physical effect that PLA contamination has on HDPE recycling.

deformation to the surface of the HDPE. Cracks, ridges, and holes are noticed even by the naked eye for HDPE_{95%} + PLA_{5%} and HDPE_{90%} + PLA_{10%}.

To be economically feasible, a plastic recycling process needs to ensure that the recycled output is of a constant quality and meets the customers specifications (Alassali et al., 2021). With the introduction of PLA into the HDPE waste stream, it is evident in Fig. 4 that at just 1 % PLA contamination the appearance of the recycled HDPE products has changed significantly. Previous research has employed more detailed techniques to investigate the effect of bioplastic contamination on conventional recycling such as scanning electron microscopy (Aldas et al., 2021; Samper et al., 2018), however, within this study, the physical surface changes were found to be so prominent, that it was not necessary to observe the surface morphology at such magnification.

3.4. Effect on tensile strength

Tensile testing was employed to understand the effect of PLA contamination on the mechanical properties of HDPE. The ultimate tensile strength, Young's modulus and toughness of the virgin and artificially aged samples (Section 2.1.3) are found in Fig. 5 and Table 1. The stress–strain curves yielded through the tensile test are shown in Supplementary Figure S2. Artificially aged samples were investigated to determine if PLA contamination had influence on the mechanical properties of HDPE over time.

From Fig. 5, a significant reduction in the ultimate tensile strength of HDPE was seen when PLA contamination was 2.5 % or higher. The reduction is likely due to the immiscibility of the two polymers having limited adhesion when mixed, allowing for deformations to easily form when subjected to external stresses (Ferri et al., 2020). As illustrated in Section 3.3, these deformations are notably seen on specimens that have 5 % and 10 % PLA contamination (Fig. 4), which directly corresponds to the reductions in tensile strength depicted in Fig. 5. PLA contamination in HDPE sheets had a greater effect on the ultimate

tensile strength when exposed to UVA radiation. HDPE_{100%} and PLA_{100%} both had a reduction in their ultimate tensile strength by 18 % after being artificially aged by UVA, which was taken as the baseline degradation due to UVA exposure. However, when PLA concentrations were at 1 %, 2.5 %, 5 % and 10 %, the tensile strength of the aged specimens reduced by 28 %, 40 %, 41 %, and 43 %, respectively when compared to their virgin counterparts. Our study shows that the addition of PLA to HDPE without any additive, e.g., compatibilizers, to help bind two immiscible polymers, increases its susceptibility to UVA irradiation and therefore has a negative effect on the material ultimate tensile strength.

To the best of the authors knowledge, this is the first study that has investigated the effect that UVA irradiation has on HDPE contaminated with PLA. It is important to understand the effect of UVA irradiation on recycled plastic materials as it can play a critical role in the degradation of the product (Doğan, 2021; Ouyang et al., 2021; Stapleton et al., 2023a). As seen within this study, if only the virgin samples were investigated, contamination levels of 2.5 % PLA and higher would be considered to have a negative impact on the ultimate tensile strength, however, when UVA ageing is also investigated, contamination levels of 1 % PLA are also discovered to have negative impacts. This highlights the importance of this work to also consider UVA ageing when investigating the effect of PLA contamination on HDPE recycling.

3.5. Change in wettability

A water contact analysis was performed to determine the effect that PLA contamination has on the wettability of HDPE (Fig. 6). Both HDPE_{100%} and PLA_{100%} sheets were concluded to be hydrophilic materials, i.e., water contact angle < 90° (Law, 2014). As the PLA concentration increased, the water contact angle decreased, making the contaminated sheets more hydrophilic. Previous literature has stated that an increase in the roughness of a hydrophilic surface results in the decrease of the material water contact angle (Li et al., 2021). This is evident within this study whereby the water contact angle decreased by

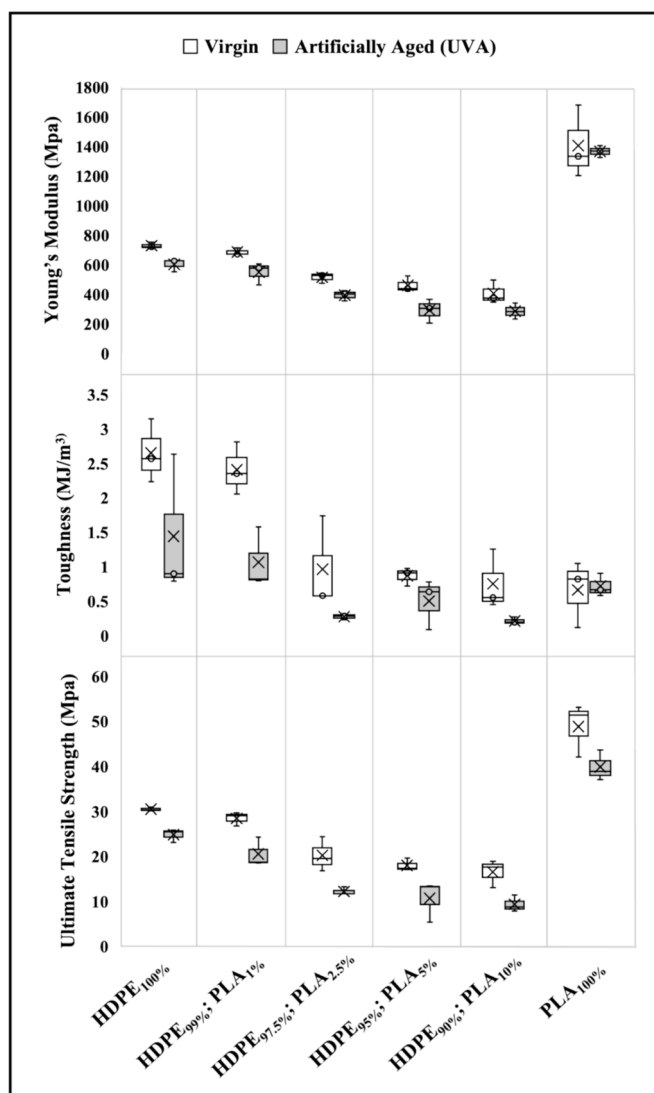


Fig. 5. Mechanical properties derived through the tensile test. A reduction in Young's modulus (top), toughness (middle) and ultimate tensile strength (bottom) were all reported when HDPE sheets were contaminated with PLA. Error bars are showing mean (symbolised by the x), median (symbolised by the line), and standard deviation of three samples.

5.8° between the HDPE_{100%} specimen and the HDPE_{90%} + PLA_{10%} specimen. The surface roughness notably changed when the contaminated sheets were visually inspected through the use of stereomicroscopes (Fig. 4).

The implications of the change in the wettability of a plastic material have been overlooked in previous studies investigating the effect of bioplastic contamination on conventional recycling. HDPE is primarily used in the form of a bottle that contains liquids such as milk, shampoos, conditioner, detergents, and motor oils (Huang et al., 2018; Papo and Corona, 2022). PLA has a water contact angle of $75.8 \pm 0.35^\circ$, indicating a greater hydrophilic surface compared to the HDPE. Previous research has illustrated PLA's ability to absorb liquid from its surrounding environment (Stapleton et al., 2023a). As the hydrophilicity of HDPE increases with the addition of PLA, it may be concluded that the contaminated mixed material may absorb moisture into the polymer matrix in a similar manner that was seen by pure PLA (Wahit et al., 2015). This would become problematic if the intended use of the plastic material is to be a container for storing liquids. This study has showcased how PLA contamination can significantly affect the wettability of a recycled product. This is an important factor to consider in the plastic

Table 1

Change in the ultimate tensile strength, Young's modulus and toughness of HDPE when contaminated with PLA. Artificially aged refers to plastic sheets being subjected to 28 days of UVA exposure. Values show average \pm standard deviation ($n = 3$).

Sample		Ultimate tensile strength (MPa)	Young's modulus (MPa)	Toughness (MJ/m ³)
Virgin	HDPE _{100%}	30.49 \pm 0.43	728.28 \pm 20.12	2.65 \pm 0.46
	HDPE _{99%} ; PLA _{1%}	28.44 \pm 1.52	685.38 \pm 24.95	2.41 \pm 0.38
	HDPE _{97.5%} ; PLA _{2.5%}	20.11 \pm 3.81	515.07 \pm 37.32	0.96 \pm 0.67
	HDPE _{95%} ; PLA _{5%}	17.96 \pm 1.41	461.15 \pm 54.57	0.87 \pm 0.13
	HDPE _{90%} ; PLA _{10%}	16.49 \pm 3.08	405.78 \pm 79.63	0.75 \pm 0.44
	PLA _{100%}	48.83 \pm 5.96	1404.80 \pm 247.34	0.66 \pm 0.48
	Artificially Aged (UVA)	HDPE _{100%}	24.80 \pm 1.48	602.54 \pm 43.81
HDPE _{99%} ; PLA _{1%}		20.51 \pm 3.28	548.84 \pm 74.65	1.06 \pm 0.44
HDPE _{97.5%} ; PLA _{2.5%}		12.16 \pm 0.90	393.51 \pm 35.38	0.27 \pm 0.36
HDPE _{95%} ; PLA _{5%}		10.65 \pm 4.59	292.10 \pm 81.73	0.50 \pm 0.36
HDPE _{90%} ; PLA _{10%}		9.33 \pm 1.81	285.62 \pm 53.92	0.21 \pm 0.05
PLA _{100%}		39.85 \pm 3.37	1365.13 \pm 41.08	0.72 \pm 0.17

recycling industry as the change in wettability may void the intended use of the recycled material.

3.6. Polymer discolouration

A colour change analysis (Section 2.2.6) was employed to determine if PLA contamination could affect the colour aesthetics of HDPE recycling (Fig. 7). Using the baseline value of $\Delta E^* = 1$ as observable colour changes by the human eye, it is evident that notable colour changes did not occur in the HDPE samples until PLA contamination was at 5%. As seen in Fig. 4, HDPE_{100%} was observed as being an off-white colour, whereas PLA_{100%} was transparent. The low contamination levels of 1% and 2.5% PLA appeared to have little influence on the colour of HDPE. However, when contamination levels increased to 5% and 10% PLA, notable colour differences were observed. Investigation of Fig. 4 shows distinctively visible colour change in the HDPE samples that have 5% and 10% PLA contamination. The immiscibility of the two materials, caused by the variations in their glass transition temperatures (Ramesh and Muthukrishnan, 2022), enables the deformations seen within the surface morphology of the blended materials. It is within these PLA blends that blemishes in the colour of HDPE are found causing the colour changes seen in Fig. 7.

Previous research has investigated the colour change in polymers that have been contaminated with PLA (Gere and Czigany, 2020; Van Velzen et al., 2022), however, the knowledge surrounding the effect of UVA on the colour of the polymer blends is currently limited. From Fig. 7 it is evident that visible colour changes are noted to occur at 1% PLA contamination, with a significant increase in the colour change from 2.5% PLA contamination and above. UVA makes up 95% of the UV radiation that reaches the surface of earth (Basen-Engquist et al., 2020), making it a critical factor to consider when analysing the longevity of a recycled polymer. UVA has been found to not only degrade a polymer mechanical properties but also significantly affect the optical behaviours of polymers (Stapleton et al., 2023a). Without considering the results of the artificially aged samples, this study would conclude that PLA

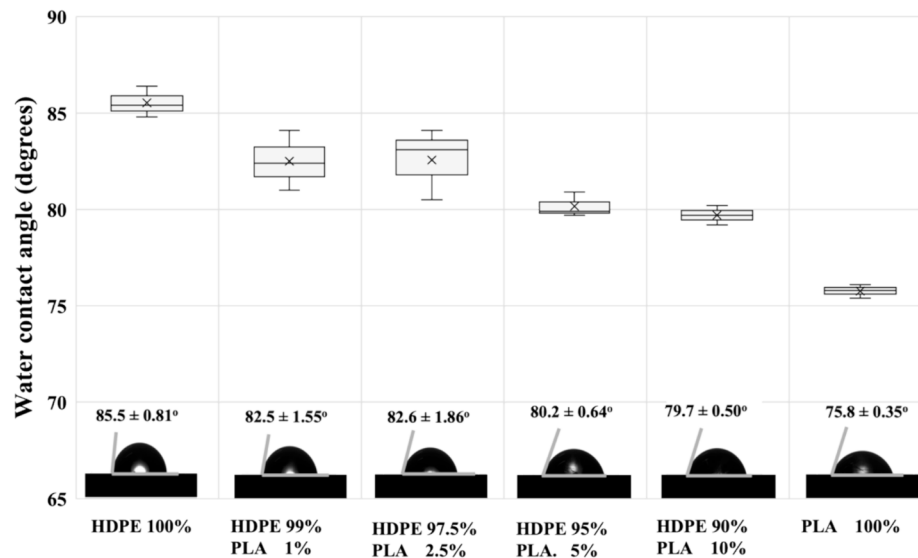


Fig. 6. Water contact angles of different HDPE and PLA blends. Error bars are showing mean (symbolised by the x), median (symbolised by the line), and standard deviation of three samples. Images obtained from DROP Image Standard Software are shown at the base of the graph.

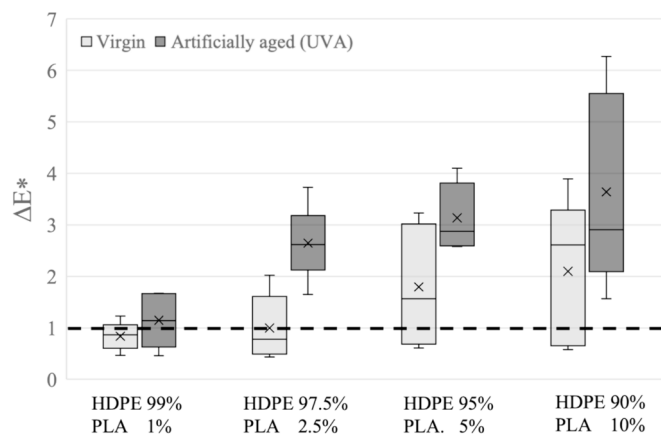


Fig. 7. Investigation of the surface colour change when HDPE recycling is contaminated with PLA. The CIELAB colour system was utilised to determine the change in colour between the samples, with ΔE^* symbolising a colour change value. Observable colour changes by the human eye are noted with a ΔE^* value greater than 1. Error bars are showing mean (symbolised by the x), median (symbolised by the line), and standard deviation of three samples.

contamination up to 2.5 % would have little effect colour of the recycled product. However, when UVA exposure is considered, it is clear that PLA contamination of 1 % or greater can influence the colour of the recycled product. Our data highlights the importance of including UVA exposure analysis into the investigation of the impact of bioplastics in conventional recycling.

3.7. Impact of PLA contamination on the mechanical recycling of HDPE

The popularity of PLA within society has continued to grow as it is derived from renewable sources instead of petroleum by-products. Production of PLA is expected to increase by 53 % over the five-year period from 2020 to 2025 (Sun et al., 2022). With the increased use and production of PLA, comes the potential for the polymer to become a contaminant in the recycling of conventional plastics. Although most thermoplastics are capable of being mechanically recycled, only six plastics (HDPE, LDPE, PET, PP, polyvinyl chloride (PVC), and polystyrene (PS)) make up the majority of the recycling market (Shamsuyeva

and Endres, 2021). A previous study that investigated the potential impact of bio-based plastics on current recycling processes found that PLA concentrations within a stream of PET packaging waste may be as high as 8 % (Alaerts et al., 2018). Although there are advancements in plastic separation technologies available (Wu et al., 2020), not all facilities may be equipped with such technologies. If the same contamination rates that were discovered within Alaerts, Augustinus and Van Acker (2018) study are applied to a HDPE waste stream, the recycled HDPE would fail to pass quality control measures, as in the current study PLA contamination of just 1 % was found to affect the physical properties of the recycled material. This research has highlighted the importance of efficient separation technologies within a plastic recycling facility to reduce the risk of PLA contamination entering a HDPE waste stream.

4. Recommendations

This research investigates the implication of PLA contamination in a HDPE waste stream. This is the first research investigating the effects of UVA radiation on HDPE contaminated with PLA. Future research can build onto this area of knowledge through the following recommendations:

1. This study illustrated that PLA contamination levels over 1 % can reduce the quality of recycled HDPE. An enquiry into the allowable limit of PLA contamination (<1%) in the HDPE waste stream should be performed to understand the necessary limit that separation technologies need to achieve.
2. Investigating the effect of other popular bioplastics contamination in HDPE recycling following the protocol proposed in this study is recommended.
3. If contamination rates are not reduced through efficient separation technologies, an investigation into potential compatibilizers that would improve the miscibility of HDPE and PLA should be explored.
4. Previous studies have shown that small amounts of PLA contamination were acceptable in the recycling of PET (Aldas et al., 2021), however, a UVA analysis was not performed. As in the current study, UVA exposure, which is very likely during the pre-processing storage, was seen to drastically effect the physical and mechanical properties of HDPE and PLA blends, this analysis should also be replicated for previous reports.

- Future research that investigates contamination in plastic recycling should not only look at the effect on the recycled product but also to the effect contamination has on the processes involved in generating the recycled product.

This research was not intended to portray PLA as a problematic plastic but was simply to show the effects that contamination has on the current recycling process. Creating homogenous stream of waste will reduce the risk of poor-quality recycled products and increase the recycling rates of plastics.

5. Conclusions

The effect of PLA contamination in the input waste stream on the HDPE recycling process was studied at different PLA concentrations (0 %, 1 %, 2.5 %, 5 % and 10 % by weight). It was discovered that PLA contamination of 2.5 % and higher affected the ultimate tensile strength, the colour and surface morphology of the recycled HDPE. To the best of the authors knowledge, this study was the first to investigate the UV durability of conventional plastics contaminated with bioplastics. When subjected to UVA exposure (simulating solar radiation), the quality of the HDPE sheets with just 1 % PLA contamination was notably affected. The results discovered within this study highlight the issues associated with inefficient separation of plastic types for plastic recycling.

CRedit authorship contribution statement

Michael J. Staplevan: Writing – review & editing, Writing – original draft, Visualization, Resources, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Ashley J. Ansari:** Writing – review & editing, Supervision, Conceptualization. **Aziz Ahmed:** Supervision, Resources, Methodology, Conceptualization. **Faisal I. Hai:** Writing – review & editing, Writing – original draft, Supervision, Resources, Project administration, Methodology, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

This research has been carried out with the support of the Australian Government Research Training Program Scholarship to Michael Staplevan, through the University of Wollongong. The authors acknowledge the use of facilities available within the School of Civil, Mining, Environmental and Architectural Engineering of UOW. We acknowledge the access to the FTIR and Stereo Microscopes at the School of Chemistry and Molecular Bioscience of UOW, Australia, and the DSC-214-Polyma at the School of Mechanical, Materials, Mechatronic and Biomedical Engineering.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.wasman.2024.05.028>.

References

- Adhikary, K.B., Pang, S., Staiger, M.P., 2008. Dimensional stability and mechanical behaviour of wood–plastic composites based on recycled and virgin high-density polyethylene (HDPE). *Compos. B: Eng.* 39 (5), 807–815.
- Akesson, D., Kuzhanthaivelu, G., Bohlén, M., 2021. Effect of a small amount of thermoplastic starch blend on the mechanical recycling of conventional plastics. *J. Polym. Environ* 29 (3), 985–991.
- Alaerts, L., Augustinus, M., Van Acker, K., 2018. Impact of bio-based plastics on current recycling of plastics. *Sustainability* 10 (5), 1487.
- Alassali, A., Picuno, C., Chong, Z.K., Guo, J., Maletz, R., Kuchta, K., 2021. Towards higher quality of recycled plastics: limitations from the Material’s perspective. *Sustainability* 13 (23), 13266.
- Aldas, M., Pavon, C., De La Rosa-Ramírez, H., Ferri, J.M., Bertomeu, D., Samper, M.D., López-Martínez, J., 2021. The impact of biodegradable plastics in the properties of recycled polyethylene terephthalate. *J. Polym. Environ* 29 (8), 2686–2700.
- Aliotta, L., Sciarra, L.M., Cinelli, P., Canesi, I., Lazzeri, A., 2022. Improvement of the PLA crystallinity and heat distortion temperature optimizing the content of nucleating agents and the injection molding cycle time. *Polymers (Basel)*, vol. 14, no. 5.
- Al-Salem, S.M., Lettieri, P., Baeyens, J., 2010. The valorization of plastic solid waste (PSW) by primary to quaternary routes: From re-use to energy and chemicals. *Prog. Energy. Combust. Sci* 36 (1), 103–129.
- Basen-Engquist, K., Brown, P., Coletta, A.M., Savage, M., Maresso, K.C., Hawk, E., 2020. Chapter 22 - Lifestyle and Cancer Prevention. In: Niederhuber, J.E., Armitage, J.O., Kastan, M.B., Doroshow, J.H., Tepper, J.E. (Eds.), *Abeloff’s Clinical Oncology*, sixth ed., Elsevier, Philadelphia. 337-74.e12.
- Blázquez-Blázquez, E., Pérez, E., Lorenzo, V., Cerrada, M.L., 2019. Crystalline characteristics and their influence in the mechanical performance in poly (ϵ -Caprolactone) / high density polyethylene blends. *Polymers* 11 (11), 1874.
- Charles, J., 2009. Qualitative analysis of high density polyethylene using FTIR spectroscopy. *Asian. J. Chem* 21 (6), 4477.
- Chen, J., Shi, Y.-y., Yang, J.-h., Zhang, N., Huang, T., Wang, Y., 2013. Improving interfacial adhesion between immiscible polymers by carbon nanotubes. *Polymer* 54 (1), 464–471.
- Chen, X., Xi, F., Geng, Y., Fujita, T., 2011. The potential environmental gains from recycling waste plastics: Simulation of transferring recycling and recovery technologies to Shenyang, China. *Waste. Manag* 31 (1), 168–179.
- Chieng, B.W., Ibrahim, N., Yunus, W., Hussein, M., 2013. Effects of Graphene Nanoplatelets on Poly(Lactic Acid)/Poly(Ethylene Glycol) Polymer Nanocomposites. *Polymers* 6, 93–104.
- d’Ambrières, W., 2019. Plastics recycling worldwide: current overview and desirable changes. *Field Actions Sci. Rep. J. Field Actions*, no. Special Issue 19, pp. 12-21.
- Doğan, M., 2021. Ultraviolet light accelerates the degradation of polyethylene plastics. *Microsc. Res. Tech* 84 (11), 2774–2783.
- Fehér, Z., Kiss, J., Kisszékelyi, P., Molnár, J., Huszthy, P., Kárpáti, L., Kupai, J., 2022. Optimisation of PET glycolysis by applying recyclable heterogeneous organocatalysts. *Green. Chem* 24 (21), 8447–8459.
- Ferri, J.M., Garcia-Garcia, D., Rayón, E., Samper, M.D., Balart, R., 2020. Compatibilization and Characterization of Poly(lactide and Biopolyethylene Binary Blends by Non-Reactive and Reactive Compatibilization Approaches. *Polymers (basel)* 12, no. 6.
- Gere, D., Czigany, T., 2018. Rheological and mechanical properties of recycled polyethylene films contaminated by biopolymer. *Waste. Manag* 76, 190–198.
- Gere, D., Czigany, T., 2020. Future trends of plastic bottle recycling: Compatibilization of PET and PLA. *Polym. Test* 81.
- Gill, P., Moghadam, T.T., Ranjbar, B., 2010. Differential scanning calorimetry techniques: applications in biology and nanoscience. *J. Biomol. Techniques: JBT* 21 (4), 167.
- Gu, J., Xu, H., Wu, C., 2014. Thermal and crystallization properties of HDPE and HDPE/PP blends modified with DCP. *Adv. Polymer Technol.*, vol. 33, no. 1.
- Hahladakis, J.N., Iacovidou, E., 2019. An overview of the challenges and trade-offs in closing the loop of post-consumer plastic waste (PCPW): Focus on recycling. *J. Hazard. Mater* 380.
- Huang, H.-H., Chen, L.-W., Lu, W.-H., Lin, W.-C., Chen, Y.-C., 2018. Design and simulation analysis of lightweight HDPE milk bottle. *Polym. Polym. Compos* 26 (1), 91–98.
- Huang, J., Veksha, A., Chan, W.P., Giannis, A., Lisak, G., 2022. Chemical recycling of plastic waste for sustainable material management: a prospective review on catalysts and processes. *Renew. Sustain. Energy. Rev* 154, 111866.
- Itim, B., Philip, M., 2015. Effect of multiple extrusions and influence of PP contamination on the thermal characteristics of bottle grade recycled PET. *Polym. Degrad. Stab* 117, 84–89.
- Kai, X., Yang, T., Shen, S., Li, R., 2019. TG-FTIR-MS study of synergistic effects during co-pyrolysis of corn stalk and high-density polyethylene (HDPE). *Energ. Convers. Manage* 181, 202–213.
- La Mantia, F.P., Botta, L., Morreale, M., Scaffaro, R., 2012. Effect of small amounts of poly(lactic acid) on the recycling of poly(ethylene terephthalate) bottles. *Polym. Degrad. Stab* 97 (1), 21–24.
- Law, K.-Y., 2014. Definitions for hydrophilicity, hydrophobicity, and superhydrophobicity: getting the basics right. *ACS. Publications* 4, 1948–7185.
- Li, C., Zhang, J., Han, J., Yao, B., 2021. A numerical solution to the effects of surface roughness on water–coal contact angle. *Sci. Rep* 11 (1), 459.
- Liu, H., Guo, L., Tao, S., Huang, Z., Qi, H., 2021. Freely moldable modified starch as a sustainable and recyclable plastic. *Biomacromolecules* 22 (6), 2676–2683.

- Ly, B.C.K., Dyer, E.B., Feig, J.L., Chien, A.L., Del Bino, S., 2020. Research techniques made simple: cutaneous colorimetry: a reliable technique for objective skin color measurement. *J. Invest. Dermatol* 140 (1), 3–12.e1.
- Maris, J., Bourdon, S., Brossard, J.-M., Cauret, L., Fontaine, L., Montembault, V., 2018. Mechanical recycling: Compatibilization of mixed thermoplastic wastes. *Polym. Degrad. Stab* 147, 245–266.
- Merrington, A., 2017. Recycling of Plastics. In: Kutz, M. (Ed.), *Applied Plastics Engineering Handbook*, second ed. William Andrew Publishing, pp. 167–189.
- Neves, A.C., Moyne, M.M., Eyre, C., Casey, B.P., 2020. Acceptability and societal impact of the introduction of bioplastics as novel environmentally friendly packaging materials in Ireland. *Clean Technol.* 2 (1), 9.
- Ouyang, Z., Yang, Y., Zhang, C., Zhu, S., Qin, L., Wang, W., He, D., Zhou, Y., Luo, H., Qin, F., 2021. Recent advances in photocatalytic degradation of plastics and plastic-derived chemicals. *J. Mater. Chem. A* 9 (23), 13402–13441.
- Papo, M., Corona, B., 2022. Life cycle sustainability assessment of non-beverage bottles made of recycled High Density Polyethylene. *J. Clean. Prod* 378, 134442.
- Pivnenko, K., Jakobsen, L., Eriksen, M.K., Damgaard, A., Astrup, T.F. 2015, Challenges in plastics recycling. In: *Proceedings of the Sardinia*.
- Qenos, 2015, *Extrusion - Technical Guide*, viewed 4th of August 2023, <<https://www.qenos.com/internet/home.nsf/web/PolyethyleneTechGuides>>.
- Ramesh, M., Muthukrishnan, M. 2022. 25 - Biodegradable polymer blends and composites for food-packaging applications. In: Mavinkere Rangappa, S., Parameswaranpillai, J., Siengchin, S., Ramesh, M. (Eds), *Biodegradable Polymers, Blends and Composites*, Woodhead Publishing, pp. 693-716.
- Riaz, U., and Ashraf, S. 2014, Characterization of Polymer Blends with FTIR Spectroscopy, in pp. 625-78.
- Robertson, A.R., 1977. The CIE 1976 color-difference formulae. *Color. Res. Appl* 2 (1), 7–11.
- Rocca-Smith, J., Lagorce-Tachon, A., Iaconelli, C., Bellat, J.-P., Marcuzzo, E., Sensidoni, A., Piasente, F., Debeaufort, F., Karbowiak, T., 2017. How high pressure CO2 impacts PLA film properties. *Express. Polym. Lett* 11, 320–333.
- Rosato, D.V., 2013. *Extruding plastics: a practical processing handbook*. Springer Science & Business Media.
- Samper, M.D., Bertomeu, D., Arrieta, M.P., Ferri, J.M., López-Martínez, J., 2018. Interference of biodegradable plastics in the polypropylene recycling process. *Materials* 11 (10).
- Shamsuyeva, M., Endres, H.-J., 2021. Plastics in the context of the circular economy and sustainable plastics recycling: Comprehensive review on research development, standardization and market. *Composites. Part C: Open. Access* 6, 100168.
- Sharma, L., Kimura, T., 2003. FT-IR Investigation into the miscible interactions in new materials for optical devices. *Polym. Adv. Technol* 14 (6), 392–399.
- Shen, L., Worrell, E., 2014. Chapter 13 - Plastic Recycling. In: Worrell, E., Reuter, M.A. (Eds.), *Handbook of Recycling*. Elsevier, Boston, pp. 179–190.
- Smith, B., 2021. The infrared spectra of polymers II: polyethylene. *Spectroscopy* 36 (9), 24–29.
- Solechan, S., Suprihanto, A., Widyanto, S.A., Triyono, J., Fitriyana, D.F., Siregar, J.P., Cionita, T., 2022. Investigating the Effect of PCL Concentrations on the Characterization of PLA Polymeric Blends for Biomaterial Applications. *Materials* 15 (20), 7396.
- Stapleton, M.J., Ansari, A.J., Ahmed, A., Hai, F.I., 2023a. Change in the chemical, mechanical and physical properties of plastics due to UVA degradation in different water matrices: A study on the recyclability of littered plastics. *Environ. Pollut* 334, 122226.
- Stapleton, M.J., Ansari, A.J., Ahmed, A., Hai, F.I., 2023b. Evaluating the generation of microplastics from an unlikely source: The unintentional consequence of the current plastic recycling process. *Sci. Total. Environ* 902, 166090.
- Staplevan, M.J., Hai, F.I., 2024. Recycling process produces microplastics. *Science* 383 (6686), pp. 958-.
- Stark, N.M., Matuana, L.M., 2004. Surface chemistry changes of weathered HDPE/wood-flour composites studied by XPS and FTIR spectroscopy. *Polym. Degrad. Stab* 86 (1), 1–9.
- Sun, C., Wei, S., Tan, H., Huang, Y., Zhang, Y., 2022. Progress in upcycling polylactic acid waste as an alternative carbon source: a review. *Chem. Eng. J* 446, 136881.
- Suryanegara, L., Nakagaito, A.N., Yano, H., 2009. The effect of crystallization of PLA on the thermal and mechanical properties of microfibrillated cellulose-reinforced PLA composites. *Compos. Sci. Technol* 69 (7), 1187–1192.
- Tarani, E., Arvanitidis, I., Christofilos, D., Bikiaris, D.N., Chrissafis, K., Vourlias, G., 2023. Calculation of the degree of crystallinity of HDPE/GNPs nanocomposites by using various experimental techniques: A comparative study. *J. Mater. Sci* 58 (4), 1621–1639.
- Thiounn, T., Smith, R.C., 2020. Advances and approaches for chemical recycling of plastic waste. *J. Polym. Sci* 58 (10), 1347–1364.
- Torres-Rivero, K., Bastos-Arrieta, J., Fiol, N., Florido, A. 2021. Chapter Ten - Metal and metal oxide nanoparticles: An integrated perspective of the green synthesis methods by natural products and waste valorization: applications and challenges. In: Verma, S.K., Das, A.K., (Eds.), *Comprehensive Analytical Chemistry*, Elsevier, vol. 94, pp. 433-69.
- Van Velzen, E.U.T., Chu, S., Molenveld, K., Jašo, V., 2022. Effect of poly lactic acid trays on the optical and thermal properties of recycled poly (ethylene terephthalate). *Packag. Technol. Sci.*
- Varghese, S., Dhanraj, N.D., Rebello, S., Sindhu, R., Binod, P., Pandey, A., Jisha, M.S., Awasthi, M.K., 2022. Leads and hurdles to sustainable microbial bioplastic production. *Chemosphere* 305, 135390.
- Volk, R., Stallkamp, C., Steins, J.J., Yogish, S.P., Müller, R.C., Stapf, D., Schultmann, F., 2021. Techno-economic assessment and comparison of different plastic recycling pathways: A German case study. *J. Ind. Ecol* 25 (5), 1318–1337.
- Vollmer, I., Jenks, M.J., Roelands, M.C., White, R.J., van Harmelen, T., de Wild, P., van Der Laan, G.P., Meirer, F., Keurentjes, J.T., Weckhuysen, B.M., 2020. Beyond mechanical recycling: Giving new life to plastic waste. *Angew. Chem. Int. Ed* 59 (36), 15402–15423.
- Wahit, M.U., Hassan, A., Ibrahim, A.N., Zawawi, N.A., Kunasegeran, K., 2015. Mechanical, thermal and chemical resistance of epoxidized natural rubber toughened polylactic acid blends. *Sains. Malaysiana* 44 (11), 1615–1623.
- Wu, X., Li, J., Yao, L., Xu, Z., 2020. Auto-sorting commonly recovered plastics from waste household appliances and electronics using near-infrared spectroscopy. *J. Clean. Prod* 246, 118732.