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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^{\star}

Michael J. Stapleton, Ashley J. Ansari, Aziz Ahmed, Faisal I. Hai

Strategic Water Infrastructure Laboratory, School of Civil, Mining, Environmental and Architectural Engineering, University of Wollongong, Wollongong, NSW, 2522, Australia

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ABSTRACT

To move towards a circular society, the recyclability potential of littered plastics should be explored to provide potential value for a product that is typically destined for landfill or incineration. This study aims to understand the changes in physical, mechanical, and chemical properties of four types of plastics (polyethylene terephthalate (PET), polypropylene (PP), polycarbonate (PC) and polylactic acid (PLA) after simulated environmental degradation. Plastic samples were subjected to different water matrices (in an attempt to simulate terrestrial, ocean, and river environments) to understand the role the environment plays on plastic degradation. Significant physical, mechanical, and chemical changes were observed for the PET, PP and PLA samples. Flakes and cracks were noted during the scanning electron microscopy (SEM) analysis of PET, PP and PLA illustrating the surface degradation that had occurred. Colour scanning of the samples provided complementary information about their suitability for upcycling or downcycling. Both PET and PP had visual colour changes, making them unsuitable for upcycling purposes. PLA had a significant decrease in its tensile strength in all environmental conditions, alongside significant chemical and surface change as revealed by Fourier-transform infrared (FTIR) and SEM analysis, respectively. PC had little to no changes in its chemical, mechanical, and physical properties due to high resistance to solar (UVA) degradation in presence of salt and natural organic matter in the form of humic acid. Therefore, out of the four types of plastics tested, PC was the only plastic determined to have good upcycling potential if collected from the environment. However, PET and PP could still be recycled into lower value products (i.e., construction materials).

1. Introduction

Mismanagement of waste through unsatisfactory waste management infrastructures or through plastics being openly dumped, has led to an increasing amount of plastic pollution in the environment. It is reported that of an estimated 359 Mt of plastic that was produced in 2018, 14.5 Mt entered natural waterways and oceans (Wayman & Niemann, 2021). If left uncollected, plastics littered in natural ecosystems (terrestrial or aquatic) begin to break down into smaller fragments called microplastics (<5 mm in size). Microplastics have been found to enter the aquatic food chain and carry hazardous contaminants such as heavy metals, polycyclic aromatic hydrocarbons (PAHs), pharmaceuticals (Ateia et al., 2020; Atugoda et al., 2021; Llorca et al., 2018; Pyl et al., 2021; Singla et al., 2020; Sørensen et al., 2020; Tang et al., 2021). In terrestrial environments, microplastics have been found in soil environments, consequently effecting key processes in a plant's life cycle (Boots et al., 2019; Qi et al., 2018; Wang et al., 2020; Wu et al., 2021).

In the fight to reduce the ecological issues associated with plastic pollution, many companies and foundations are currently operating to remove plastics from the environment (e.g., Clean up Australia, The Ocean Clean Up, 4Ocean and The Ocean Conservancy). Plastics that are collected from the environment are generally sent to landfill or used as a fuel source if waste-to-energy facilities are available. To save resources and reduce the environmental impact of plastics, the potential to recycle littered plastics is becoming an interest to society. If a littered plastics is to be recovered and sent to be recycled it can either be up-cycled or

* Corresponding author.

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E-mail address: faisal@uow.edu.au (F.I. Hai).

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down-cycled. Upcycling is whereby the plastic material is converted back into a product of equal or greater value and downcycling is whereby the plastic is turned into something of lower value (Campbell-Johnston et al., 2020). Upcycling of plastics is ideally the preferred pathway for plastic waste, however, due to the demand for high quality recycled products most plastics are downcycled (La Mantia, 2004). The plastic guidelines for the recovery and recycling of plastic wastes (ISO 15720–2008) does not stipulate a specific level of quality that is required by a plastic to be recycled, only that the quality of the plastic waste should not impede the quality of the recycled material (ISO, 2008). Due to the environmental degradation that littered plastics are subjected to, it is expected that the quality of the littered plastic will be a contributing factor in the plastics recyclability (Hahladakis & Iacovidou, 2018).

Environmental degradation of plastics occurs through the following four mechanisms: photodegradation (reaction with light), thermooxidative degradation (reaction with oxygen), hydrolytic degradation (reaction with water), and biodegradation (action of microorganisms) (Andrady, 2011). Limited research has been reported on the recyclability of littered plastics, and it is unclear if environmental degradation varies among plastic types or the environment in which the plastic is littered. By answering these questions, potential economic value could be given to littered plastics if they are deemed suitable for the recycling process (Ranjan & Goel, 2021).

The recycling process can be defined as either mechanical recycling, chemical recycling, or energy recycling depending on how the plastic is being recycled (Al-Salem et al., 2010; Hahladakis & Iacovidou, 2019; Merrington, 2017). Since mechanical recycling is currently the most universally used and environmentally friendly method for recycling plastic waste (Maris et al., 2018), this study will focus primarily on the feasibility of mechanical recycling of plastics exposed to different environmental conditions based off the guidelines set out by ISO 15720-2008 (ISO, 2008). This study aims to fill the inadequacy of the current research surrounding the recyclability of littered plastic. By analysing the chemical, physical and mechanical changes that occurs to four commonly found plastics namely, Polyethylene terephthalate (PET), polypropylene (PP), polycarbonate (PC) and Polylactic acid (PLA) after being subjected to simulated environmental conditions, valuable information about the best recycling pathway for littered plastics can be made.

2. Materials and methods

2.1. Selection of plastics

The four thermoplastics used in this study are PET, PP, PC and PLA. PET and PP are two of the most widely used plastics and are highly prevalent in the environment as they are popular choices for single use plastics such as drink bottles and take away containers (Dahlbo et al., 2018; Kubowicz & Booth, 2017; Matias et al., 2020). PC's properties of high levels of toughness, good heat resistance and also being an electrical inductor has made it a favourable material for electronic application (Margolis, 2020). Proper management of e-waste is becoming a rising issue for many countries, with an estimated 20-50 million tonnes of e-waste being produced annually (Herat & Agamuthu, 2012). PLA is a biodegradable thermoplastic derived from lactic acid (Pang et al., 2010). Since the derivation of PLA is from a renewable source it has grown in popularity as an alternative to fossil-based plastics with an estimated 190,000 tonnes of PLA produced in 2019 (Jem & Tan, 2020). This study will focus on PET and PP due to their popularity as single use plastics, PC as it is an emerging source of environmental waste and PLA as it is the most widely used bioplastic alternative.

2.2. Sample preparation

All plastic samples used this study were 1 mm in thickness. PET, PP and PLA samples were 3D printed into two different shapes: dog bone

and rectangular (40 mm \times 20 mm). The dog bone shape was used for tensile testing and the shape profile was replicated from ASTM D638 shape type IV, measuring 115 mm long and 19 mm wide. The rectangular plastic samples were used for all other testing procedures. PC samples were jet cut from a 1 mm sheet obtained from RS Pro Components (Sydney, Australia) into the shapes stated above. Plastic samples were washed with Milli-Q water to remove any residues from the surfaces. Reagent grade humic acid and sodium chloride was purchased from Sigma-Aldrich (Sydney, Australia).

2.3. Preparation of simulated environmental conditions and experimental set up

Three environmental conditions (air, seawater, and surface water) were simulated in the laboratory. Seawater was simulated by dissolving 35 g/L of NaCl into Milli-Q water. 20 mg/L of humic acid was dissolved into Milli-Q water to simulate the dissolved organic material content in surface water (Basumallick & Santra, 2017). Milli-Q water was used as a controlled water condition and samples without water (exposed to air) was used to simulate polluted terrestrial plastics.

A UVA weathering chamber was constructed in the laboratory using two 10 W-UVA-340 lights which produced light at a wavelength of 340 nm to simulate natural sunlight (Fig. 1). Plastic samples (three dog bone shaped and three rectangular shaped) were placed into glass vessels separately under respective simulated conditions. This exposure experiment was conducted for 28 days, during which samples were flipped every seven days to evenly weather the plastics. Plastic samples were placed 15 cm from the light source, simulating an environment where the theoretical light irradiance on the surface of the plastics was 3.5 mW/cm². Simulated seawater, simulated surface water and Milli-Q water was replaced every seven days. A control sample was left in a cool, dark cupboard away from any UVA radiation. After 28 days, the samples were taken from the UVA chamber and were stored in a dark cupboard away from any further UVA radiation prior to analysis within two weeks.

2.4. Analytical method

2.4.1. Fourier transform infrared (FTIR) spectroscopy

To analyse chemical changes within the polymers due to UVA degradation, FTIR analysis was performed using a Shimadzu IRAffinty-1S fitted with a MIRacle-10 ATR. Absorbance was measured in the available infrared spectrum range of 4000–600 cm⁻¹. LabSolutions IR software was used to analyse the results and develop FTIR plots (Ranjan & Goel, 2021). Samples were measured in triplicates to assess and check the reproducibility of the results. One FTIR spectra was produced for each simulated environment and was used to compare against the control sample to determine if chemical changes had occurred.

2.4.2. Water absorbance

Water absorbance was calculated through the weight gains or losses after the samples had been subjected to 28 days of simulated environmental conditions. Immediately after being removed from the UVA chamber, the excess water on the surface of the samples were removed and the samples were weighed. Sample weights were recorded using a precision scale with a minimum range of 1.0×10^{-4} g. Samples were tested in triplicates and compared to control samples that had not been subject to UVA exposure. Results were recorded by a percentage change in weight compared to the average control sample weight.

2.4.3. Tensile testing

Tensile testing was performed on all plastic specimens to determine the change in mechanical parameters (Young's Modulus and tensile strength) after being exposed to different environmental conditions. Tensile tests were performed following the ASTM D638 standard. Using an Instron 3367 tensile testing machine equipped with a 30 kN load cell.



Fig. 1. UVA degradation experimental set up.

The specimens were gripped so that the gauge length was 25 mm. Testing for all specimens were performed in triplicates (N = 3).

2.4.4. Scanning electron microscopy (SEM) analysis

SEM (JEOL JSM-6490 series) was used to visually inspect the change in surface morphology of the plastic specimens. Small samples (5 \times 5 mm) were prepared for SEM analysis by coating in a thin layer of gold to create a conductive layer and improve secondary electron signal necessary for surface morphology examination. Each plastic specimen was measured in triplicate (N = 3) to ensure accuracy in recording.

2.4.5. Colour analysis

Colour analysis of digital images have been reported to have high accuracy and provides a novel approach for determining the change in colour of tested samples (Ayustaningwarno et al., 2021; León et al., 2006; Subhashree et al., 2017; Xu, 2010). Colour scanning was done with a HP scanner (600 dpi). The change in colour of the plastic samples was analysed following the CIELab system developed in 1976 by the International Commission on Illumination (Yang et al., 2015). Digital colour meter software (Apple, USA, version 5.22). Was used to determine. The software measured L* (lightness), a* (redness) and b*(yellowness) of the plastic samples to determine the ΔE^* value (overall colour difference) of each sample. ΔE^* was calculated using the following equation:

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

Where;

$$\Delta L^* = L_T^* - L_c^*, (T = \text{test sample, } c = \text{control sample})$$
(Eq. 2)

 $\Delta a^* = a_T^{*-} a_c^*, \tag{Eq. 3}$

$$\Delta b^{*} = b_{\rm T}^{*-} b_{\rm c}^{*}, \tag{Eq. 4}$$

Five locations were chosen at random on the plastic samples and the

average L*, a* and b* was used to determine the average overall colour difference.

3. Results and discussion

3.1. Fourier transform infrared (FTIR) spectroscopy

UV radiation can cause photooxidative degradation within a polymer, causing polymer chains to break and producing free radicals (Yousif & Haddad, 2013). FTIR spectrometry is a useful analysis in tracking changes in chemical structures and therefore ideal for analysing degradation caused by UVA radiation. The FTIR results of the four test environments (Air, Milli-Q, Milli-Q + NaCl and Milli-Q + humic acid) were compared to the control sample (sample exposed to air in dark) to determine if any changes to chemical structures had taken place. Peaks showing changes in the FTIR spectra for PET, PLA, PP and PC are seen in Fig. 2. The full FTIR spectra of PET, PLA, PP and PC can be found in Supplementary Figures S1 - S4.

PET and PLA exhibited two clear affected regions on the FTIR spectra (Fig. 2): the carbonyl stretching region, located from 1800 cm^{-1} to 1650 cm^{-1} and the hydroxyl group, found between 3700 cm^{-1} and 3000 cm^{-1} (Hoidy et al., 2010; Pereira et al., 2017). Carbonyl stretching relates to the carbonyl group (C=O) found in the chemical structure of PET and PLA (Mecozzi & Nisini, 2019). Significant changes in the absorbance levels within the carbonyl group in both PET and PLA samples were discovered. Photo-oxidation caused by UV radiation is expected to the primary cause of the chemical changes seen in the carbonyl group (Martínez-Romo et al., 2015). The absorbance of the hydroxyl group (3500 cm⁻¹ - 3100 cm⁻¹) was found to increase for the PET and PLA samples that were subjected to water environments. PET and PLA are hygroscopic plastics, meaning these can absorb moisture from environment (Jabarin & Lofgren, 1986; Yew et al., 2005). In both the PET and PLA samples, the Milli-Q + humic acid environment showed the greatest levels of water absorbance. Humic acid has been found to



Fig. 2. Investigation of absorbance peaks in the FTIR spectra to determine if simulated environmental exposure changed the chemical compositions of the plastics. Fig. 2.1 (PET) and Fig. 2.2 (PLA) investigated changes in the absorbance levels in the hydroxyl and carbonyl regions of the polymers. Fig. 2.3 (PP) and Fig. 2.4 (PC) investigated changes in the absorbance levels in the C–H stretching, C–H bending and carbonyl regions of the polymers. Changes in absorbance levels indicated chemical changes occurring. Full FTIR spectra for PET, PLA, PP and PC are found inSupplementary Figures S1 – S4.

increase water absorption in both soils and plants by altering their fixation properties (Belorkar, 2021; Ghabbour et al., 2017). From the FTIR spectra, humic acid may have the same effect on hygroscopic polymers as it does on soils and plants by also increasing their water retention capabilities.

The degraded PP samples have three absorbance regions of interest that can be seen in Fig. 2: the C–H bending region, located from 1500 cm⁻¹ to 1350 cm⁻¹, the carbonyl group, located from 1750 cm⁻¹ to

1550 cm⁻¹, and the C–H stretching region, located from 3000 cm⁻¹ to 2800 cm⁻¹. The C–H bending and C–H stretching regions on the FTIR spectra illustrate how the bonds between the C–H components of PP are behaving. Changes in absorbance seen in these regions demonstrate that the bonds have been changed due to the UVA radiation and the environments in which the samples were placed. Both the Milli-Q + NaCl sample and Milli-Q + humic acid sample had an increase in absorbance compared to the control sample, which can be attributed to the sodium

chloride and humic acid reacting under UVA radiation and increasing the C–H bond strengths of the PP (Ranjan & Goel, 2021). PP samples in Air and Milli-Q water both had a decrease in absorbance compared to the control. The absence of any ions (*i.e.*, NaCl or humic acid) allowed the PP chemical structure to solely react to the UVA radiation, which decreased the C–H bond strength. New absorbance peaks were found in the carbonyl group on the Milli-Q, Milli-Q + NaCl and Milli-Q + humic acid samples. The change in chemical structure seen in these samples can lead to discolouration and loss of mechanical properties over time (Ranjan & Goel, 2021). It is noted that in the PP samples the hydroxyl group was relatively unchanged compared to the PET and PLA samples. This can be attributed to the hydrophobic nature of PP (Santiagoo et al., 2018).

Due to its chemical composition, PC has three distinguished regions on the FTIR spectra. (Fig. 2). No significant change was seen in the C–H stretching region ($3100-2700 \text{ cm}^{-1}$), the carbonyl stretching region ($1800-1600 \text{ cm}^{-1}$), or in the C–H bending region ($1530-1470 \text{ cm}^{-1}$). The FTIR results indicate that little to no chemical changes occurred on any of the PC samples over the 28 days they were subjected to simulated environments.

3.2. Water absorbance

The change in the weights of PET, PP, PLA, and PC samples after they had been subjected to simulated environmental degradation were performed using precision scales $(1.000 \times 10^{-1} \text{ g})$ and are shown in Fig. 3. Negligible weight changes were seen by all four plastic types in the air environment. Minor changes were seen by PP and PC for all simulated water environments, illustrating their inability to absorb moisture from their environments due to their hydrophobicity. Both PET and PLA weights were found to increase when submerged in Milli-Q water environments, particularly in presence of salt or humic acid. The largest weight change for PET (5.27 \pm 1.58%) and PLA (8.62 \pm 2.29%) occurred in presence of humic acid and NaCl, respectively. It is worth reiterating that absorption of water was also confirmed by the change in hydroxyl group depicted in FTIR spectra (Fig. 2).

3.3. Tensile testing

The Young's modulus and ultimate tensile strength of PET, PP, PC and PLA were compared through tensile testing (Table 1). The PET control sample was found to have an ultimate tensile strength of 42.9 ± 1.5 MPa. Negligible changes were seen in the ultimate tensile strength of the air and Milli-Q samples, but for the Milli-Q + NaCl and Milli-Q + humic acid samples, the ultimate tensile strength was reduced by 16.6% and 25.4%, respectively. As there was no reduction in tensile strength in

Table 1

Mechanical properties of PET, PP, PLA and PC after subjected to different environmental conditions (mean and standard deviation of three replicates are reported).

		Ultimate Tensile Strength (MPa)	Young's Modulus (MPa)
PET	Control	42.9 ± 1.5	1270.0 ± 19.6
	Air	43.7 ± 1.4	1329.3 ± 15.5
	Milli-Q	41.7 ± 0.7	1262 ± 36.8
	Milli-Q + NaCl	35.8 ± 0.7	1297.3 ± 22.7
	Milli-Q + Humic	32.0 ± 1.3	1279.3 ± 23.2
	Acid		
РР	Control	18.9 ± 1.8	$\textbf{357.4} \pm \textbf{19.9}$
	Air	18.0 ± 1.0	330.5 ± 21.8
	Milli-Q	18.4 ± 1.2	325.8 ± 5.2
	Milli-Q + NaCl	19.1 ± 0.7	323.5 ± 7.6
	Milli-Q + Humic	18.9 ± 1.1	335.2 ± 10.4
	Acid		
PLA	Control	37.7 ± 1.6	1346.6 ± 54.4
	Air	29.0 ± 5.4	1149.3 ± 168.4
	Milli-Q	29.2 ± 8.4	1248.3 ± 168.1
	Milli-Q + NaCl	23.0 ± 1.9	1018.6 ± 58.8
	Milli-Q + Humic	22.3 ± 2.5	1082.9 ± 82.0
	Acid		
PC	Control	55.7 ± 1.1	1049.5 ± 49.4
	Air	51.0 ± 5.5	1090.2 ± 45.5
	Milli-Q	56.2 ± 4.4	1133.3 ± 18.9
	Milli-Q + NaCl	55.1 ± 0.7	1091.6 ± 33.4
	Milli-Q + Humic	53.8 ± 1.8	1109.5 ± 37.3
	Acid		

the Milli-Q sample, it can be concluded that the reduction seen by the Milli-Q + NaCl and Milli-Q + humic acid sample can be attributed to the ions present in the water samples. UV radiation is known to react with humic acid to create free radicals (Polewski et al., 2005), which ultimately increased the degradation of the PET samples. Another explanation for the reduction in tensile strength seen by the Milli-Q + NaCl and Milli-Q + humic acid samples can be due to PET being hygroscopic (Jabarin & Lofgren, 1986; Yew et al., 2005). A previous study reported the tensile strength of two plastics (polybutylene terephthalate and polyamide-6) to decrease when water absorption had increased (Mortazavian et al., 2015). As already discussed, the PET samples submerged in the Milli-Q + NaCl and Milli-Q + humic acid had increased peaks in the hydroxyl group in FTIR spectra (Fig. 2), indicating an increase in water absorption. Similar to the results seen in Mortazavian et al. (2015) study, in this study, the PET samples with higher water absorption had the most significant decrease in tensile strength. Significant change in ultimate tensile strength was seen for PLA samples from all four simulated environments (Table 1). Like PET, the largest reduction in the ultimate tensile strength (40.8%) of PLA was observed for samples



Fig. 3. Change in weight of plastic samples after being exposed to different simulated environmental conditions for 28 days. Error bars are showing mean, median and standard deviation of three samples.

submerged in Milli $\mathbf{Q}+\mathbf{humic}$ acid where higher water absorption was also observed.

Under the tested conditions, the change in the ultimate tensile strength of PP and PC was found to be insignificant (<10% for all samples). It was concluded that unlike PET and PLA, the strength of PP and PC was unaffected by the UVA exposure and the simulated environmental conditions the samples were placed in. Further insights into this can be found in Section 3.6.

3.4. Scanning electron microscopy (SEM) analysis

SEM analysis was used to visually inspect the change in the surface morphology of the plastic samples after being exposed to UVA radiation for 28 days in different simulated environments. The changes in surface morphology of PET, PP and PLA samples can be seen in Figs. 4-6, respectively. Results for PC samples can be found in Supplementary Figure S5 as they illustrated no noticeable changes to their surface morphology. It was apparent that the environment heavily influenced the degree of surface degradation that occurred, with samples subjected to the air environments showing the greatest change in surface morphology. Cracks, flakes, prominent ridges, and deformation were seen to occur on the surface of PET, PP, and PLA samples after subjected to the tested conditions. When PET and PLA samples were submerged in Milli-Q water environments, some level of protection to the surface morphology were seen. Both PET and PLA have been shown to absorb water from their environment which may potentially provide some level of surface protection from UVA irradiation. PP on the other hand is naturally a hydrophobic material (Santiagoo et al., 2018) and was seen to absorb no moisture from its surrounding. The lack of hydroxyl group in the PP samples may account for the degradation that was seen on the surface of all samples that were submerged in the Milli-Q environments.

The results found using SEM analysis not only shows how polymers degrade differently under the same tested conditions, but also how different polymers can have a higher potential to release microplastics into the environment. Comparing Fig. 4d, 5d, and 6d, which is the results of samples placed in a simulated ocean environment, shows the

surface of PP to have higher levels of degradation than PET and PLA. PP may pose a greater threat to oceanic life due to degrading into smaller fragments and being mistaking as food (Horton et al., 2017). Oceanic microplastic pollution has been found to affect the health and development of marine life (Barboza et al., 2020; Bertucci et al., 2022; Li et al., 2022) and potentially become a vector for resistant bacteria's and genes (Stenger et al., 2021).

Previous research that has used SEM analysis to examine the recyclability of littered plastics has yet to discuss the implications of degraded surface morphology and the potential to contaminate the recycled product with unknown contaminates (Barrington et al., 2018; Iñiguez et al., 2018; Ranjan & Goel, 2021). Cracks and holes that can be examined in Figs. 4–6 are all potential locations for contaminates to gather and accumulate (Stapleton et al., 2023). If collected and recycled, without proper and thorough cleaning, these contaminants can be transferred into the new recycled product. This is an important factor that has up until now been overlooked in the literature and needs to be considered when determining the viability of recycling littered plastics.

3.5. Colour analysis

A digital colour meter was used to analyse the change in colour of the plastic samples before and after being exposed to four different simulated environments for 28 days. The overall colour difference is expressed in terms of the ΔE^* value and can be seen in Fig. 7. A ΔE^* value of less than one indicates a change in colour that is not observable by the human eye (Ly et al., 2020). PLA and PC samples showed no visible colour change over the 28 days. Visible changes in colour were seen on all PET and PP samples, with the greatest change seen in the PET sample exposed to Milli-Q. The yellowing that is seen by both the PET and PP samples is due to the cross-linking and oxidation reactions that are occurring from the constant exposure of UVA radiation (Al-Sabagh et al., 2016). Both PLA and PC have also been reported to yellow after being subjected to UV radiation (Mistretta et al., 2020; Redjala et al., 2020), however, under the testing conditions in this experiment, it was not seen. The use of colour analysis has yet to be used by previous



Fig. 4. SEM images of the surface of PET samples after being exposed to different environmental conditions for 28 days. a) control; b) air; c) Milli-Q water; d) Milli-Q + NaCl; e) Milli-Q + humic acid. More prominent ridges were seen in all SEM images when compared to the control sample illustrating the surface degradation that occurred due to environmental degradation.



Fig. 5. SEM images of the surface of PP samples after being exposed to different environmental conditions for 28 days. a) control; b) air; c) Milli-Q water; d) Milli-Q + NaCl; e) Milli-Q + humic acid. Flakes, cracks, and separation were seen on PP samples that were exposed to 28 days of environmental conditions when compared to the control sample.



Fig. 6. SEM images of the surface of PLA samples after being exposed to different environmental conditions for 28 days. a) control; b) air; c) Milli-Q water; d) Milli-Q + NaCl; e) Milli-Q + humic acid. Image b) and d) shows degradation through surface deformation when compared to the control sample.

studies as a factor that influences the recyclability potential of littered plastics. Colour contamination can have negative effects on the aesthetics of a recycled product as the colour can disjointedly mix and create a dirty appearance (Faraca & Astrup, 2019). Further understandings of the effect of colour on recyclability potential is explained in Section 3.6.

3.6. Insight into recyclability potential

This study aimed to understand the chemical, physical and mechanical changes that occurred to four different types of plastics after being exposed to different simulated environmental conditions. By comparing the change in results to a control sample, the best pathway



Fig. 7. Change in colour between plastic samples subjected to different simulated environments for 28 days. Results based on the CIELAB colour system where the difference in colour is expressed as a ΔE^* value. Changes below a value of one are not visible by the human eye. Error bars are showing mean, median and standard deviation of three samples.

for recycling littered plastics was determined by following the guidelines set out by ISO 15720–2008 (ISO, 2008). Based on the guidelines, any plastic that did not have any significant changes to its chemical, physical, and mechanical properties would be considered for upcycling purposes. If significant changes were noticed, downcycling of the littered plastic would be recommended (Hopewell et al., 2009; Schwarz et al., 2021).

Within this experiment, PC was the only plastic that did not show any significant changes to the teste properties after being exposed to the tested conditions. PC is resistant to water, UVA and organic compounds, it has also been found to have good weathering, thermal and mechanical properties (Begum et al., 2020). The properties that PC exhibit, explain the results found within this study, whereby it was unaffected by the environments that it was exposed to. Previous studies have shown significant degradation to PC when exposed to UV radiation, however, understanding the type of UV radiation that it was exposed to rationalises the degradation. Redjala et al. (2020) for example, illustrated significant degradation of PC samples after they were exposed to UV radiation for only nine days. Severe yellowing was seen on the PC samples and significant changes to the FTIR spectra was also noted. However, the degradation of the PC was due to the UV source being in the UVC spectrum. Similarly, McKeen (2019) showed degradation of PC to occur when treated with UVB and UVC radiation. UV radiation is broken up into three counterparts: UVA, UVB and UVC. UVC's wavelengths are 100-280 nm and do not pass through our atmosphere. UVA and UVB both reach the surface of the earth but the concentrations are vastly different (95% UVA and only 5% UVB) (Basen-Engquist et al., 2020). The long wavelengths of UVA make it the least damaging of the three in the UV spectrum (Dasgupta & Klein, 2014). This highlights how previous studies using either UVB or UVC radiation could generate degradation in the PC samples. Since the most common UV radiation that will affect littered PC plastics is UVA, the degradation that would occur in the environment would match the results seen in this study rather than others mentioned prior. In this study, since no significant environmental degradation was found to occur under the tested conditions, it is concluded that PC litter that is collected from the environment, either from the land or aquatic environments, would likely be suitable for recycling into a product with equal or higher value.

PET, PP and PLA all exhibited levels of significant degradation after being exposed to various simulated environments for 28 days. The FTIR and weight analysis results confirmed PET and PLA as being hygroscopic plastics. Their susceptibility to absorbing water from their environment negatively affected their ultimate tensile strength, with a significant decrease seen for the samples that were exposed to Milli-Q environments. The presence of both inorganic and organic compounds in the water (NaCl and humic acid), further increased the degradation that occurred over the 28-day period. PP illustrated its hydrophobic properties in both the FTIR and weight analysis. The absence of water in the PP samples may be one of the explanations for why the ultimate tensile strength was unchanged after the 28-day exposure period. However, the tensile test results found within this study for PP varied significantly to the results reported in previous studies. Ranjan and Goel (2021) demonstrated that when PP was exposed to UVA degradation in an air environment, its tensile strength decreased by 63% when compared to the control sample. The discrepencies may be attributed to the thickness of the plastic sample being 65 μ m compared to this study where the plastic samples were 1 mm thick. Further investigations into whether thinner plastics are more succeptible to environmental degradation is required to make a clear correlation. PET and PP were found to be the only plastics that had a visibile change in colour after the 28-day exposure test. Discolouration was found in all simulated environments, indicating that the change in colour was due primarily to the UVA radiation. Although colour change is a significant factor in determing the recyclability of a plastic, it has not previously been investigated in other studies investigating the recyclability of littered plastics (Barrington et al., 2018; Iñiguez et al., 2018; Pelegrini et al., 2019; Ranjan & Goel, 2021). For PET, only clear/translucent plastic waste is currently accepted for upcycling purposes due to its marketability and flexibility to be transformed into new products of varying colours (Hahladakis & Iacovidou, 2018). This scenario is expected to be the case for most other plastic types indicating that any colour variation away from clear/transluscnet would not be fit for upcycling purposes. With the significant change in the colour that was seen by both PET and PP, this analysis on its own is satisfactory in deeming these plastics unsuitable for recycling into a product of equal and greater value and only useful for downcycling purposes. The degradation that PLA illustrated within this study was also expected due to biodegradable properties that it possess. Although, PLA will not completely degrade under regular environmental conditions (Chamas et al., 2020), it does degrade at a significant level that if it was to be recycled, the properties of the resultant material will be significantly less than required. From the results found, littered PET, PP, and PLA samples collected from any environment would not be suitable for upcycling purposes and only appropriate for downcycling.

Although only littered PC was found to be appropriate for upcycling purposes, downcycling of littered PET, PP and PLA is still an environmental benefitial pathway for the plastic waste compared to the traditional method of being landfilled or incinerated. Downcycling of littered PET bottles into textile fibres and littered PP containers into garderning items reduces oil usage and carbon dioxide emissions even if the downcycled item can no longer be recycled (Aubin et al., 2022; Hopewell et al., 2009). The use of plastic waste in construction materials has also exponentionally increased over the recent years providing a pathway for recycling littered plastics that are serverly degradaed (Nyika & Dinka, 2022; Suchithra et al., 2022; Yadav et al., 2022).

4. Conclusion

This study has shown that environmental conditions play a significant role on the level of plastic degradation. Dissolved organics in water alongside UVA exposure can generate notable change in the mechanical and chemical properties of plastics. The chemical, mechanical or physical properties of PET, PP and PLA degraded to a level where upcycling of the plastics were less suitable. By contrast, the chemical, mechanical and physical properties of PC were unaffected during the 28-day experimental period, indicating their potential for upcycling under similar exposure conditions.

Based on the changes that occurred to plastic samples that had been exposed to simulated environments, the findings of this study explored the possibility of giving value to plastics that were once considered wasted products. Future research within this area should focus on developing a grading system for plastic collected from the environment so they can be recycled accordingly and to develop standards for the allowable levels of degradation for a plastic to undergo before it is no longer acceptable for recycling.

Credit author Statement

Michael J. Stapleton: Conceptualization, Methodology, Investigation, Writing – original draft. Ashley J. Ansari: Supervision, Writing – Reviewing and Editing. Aziz Ahmed: Resources, Supervision, Faisal I. Hai: Conceptualization, Supervision, Project administration, Writing – Reviewing and Editing

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.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2023.122226.

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