



# Observation of the degradation of three types of plastic pellets exposed to UV irradiation in three different environments

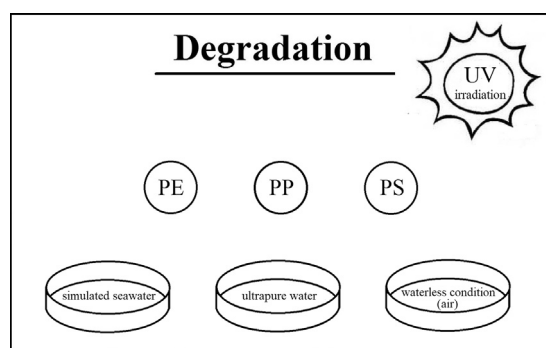
Liqi Cai, Jundong Wang, Jinping Peng\*, Ziqing Wu, Xiangling Tan

Faculty of Chemical Engineering and Light Industry, Guangdong University of Technology, Guangzhou 510006, China

## HIGHLIGHTS

- Degradation of plastics under UV irradiation in different environments was studied.
- Changes in chemical/physical properties were examined by FTIR, Raman, and SEM.
- Similarities and discrepancies in the degradation processes were investigated.
- Exposure time, oxygen content and the water chemistry are the important factors affecting the degradation degree.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 11 December 2017

Received in revised form 22 January 2018

Accepted 7 February 2018

Available online xxxx

Editor: D. Barcelo

### Keywords:

Plastic pellets

Plastic types

Different environments

UV irradiation

Degradation

## ABSTRACT

Plastic debris represents one of the most prevalent and persistent pollution problems in the marine environment. In particular, microplastics that are mainly degraded from larger plastic debris have become a growing environmental concern. However, studies on the degradation of plastics in the aquatic environment that hydrobionts reside in have been limited, while several studies regarding the degradation of plastics have been conducted under outdoor or accelerated weathering conditions. Thus, observation of the degradation of three types of virgin plastic pellets exposed to UV irradiation in three different environments (i.e., simulated seawater, ultrapure water, and a waterless (air) condition) was carried out. Data on the changes in physical and chemical properties were collected. The FTIR spectra showed that hydroxyl groups and carbonyl groups developed in three types of weathered plastic pellets under the air and ultrapure water environmental conditions after 3 months of UV irradiation, while only carbonyl groups were found in plastic pellets in the simulated seawater environment. In contrast, the Raman spectra showed no significant changes in the weathered plastic pellets, but there were different intensities of characteristic peaks after exposure to UV irradiation. In addition, SEM images illustrated that granular oxidation, cracks and flakes were common patterns during degradation, and the plastic pellets in the three different environments experienced different levels of chemical weathering. We suggest that further studies on the degradation processes of plastic debris are needed to predict the fate of plastic debris in the environment.

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## 1. Introduction

The annual production of plastic materials has grown exponentially over the past few decades, with approximately 322 million tons being produced in 2015 (PlasticsEurope, 2016). Plastics, due to their

\* Corresponding author.

E-mail address: [jppeng@gdut.edu.cn](mailto:jppeng@gdut.edu.cn) (J. Peng).

inexpensive, lightweight, and durable properties, are suitable for a large variety of consumer products used in everyday life (Thompson et al., 2009; Wang et al., 2016). However, the accumulation of plastic waste in the environment, especially in the ocean, has been of great concern leading to long-term environmental, economic, and pollution management problems (Singh and Sharma, 2008). Some studies indicated that there may be >5 trillion pieces of plastic in the oceans globally (Eriksen et al., 2014), and 4.8 to 12.7 million tons of plastic waste was estimated to enter the oceans in 2010 (Jambeck et al., 2015). In addition, it was reported that 60% to 80% of floating litter in the oceans consisted of plastic debris, and the amount of plastic debris in the environment increases each year (Moore, 2008; Gewert et al., 2015) because most plastic products are used in single-use applications that have a low recovery value and are difficult to degrade in the environment (Wang et al., 2016). Notably, it has been predicted that the total weight of plastic debris in the oceans could be heavier than the total weight of fish by 2050 unless effective measures are taken (Neufeld et al., 2016; Song et al., 2017).

Among the plastic debris in the ocean, microplastics (i.e., plastic particles <5 mm in diameter) have been reported to compose the majority of debris (Goldstein et al., 2013; Hidalgo-Ruz et al., 2012). Sources of microplastics include primary sources (i.e., plastics of microscopic size that are purposefully used) and secondary sources (i.e., fragmentation and degradation from larger plastic items). The continuous degradation of plastics could result in an amount of microplastics that would inevitably increase in the environment with time (Wang et al., 2016). In addition, because plastic debris of small size can be easily transported by ocean currents, wind, tides and tsunamis in the environment (Collignon et al., 2012; Wang et al., 2016), microplastics have become a prevalent component and can be found worldwide in the global environment, including seas (Van Cauwenberghe et al., 2013; Thompson et al., 2004), freshwater lakes (Fischer et al., 2016; Ballent et al., 2016), rivers (Peng et al., 2017; Morrill et al., 2014; Wang et al., 2017), terrestrial environments (Jambeck et al., 2015), and atmospheric fallout (Dris et al., 2016; Cai et al., 2017). Meanwhile, these environmental forces (mainly physical effects) could also contribute to the fragmentation and degradation of plastics in the environment to some extent. In addition, there is a particular concern that microplastics have been reported to be ingested by a wide variety of organisms (Gall and Thompson, 2015; Zhao et al., 2016; Li et al., 2016; Sanchez et al., 2014), which can be transferred through the food web (Setälä et al., 2014). More importantly, microplastics contain various chemicals, such as additives (Rochman et al., 2013; Lithner et al., 2011) and metals (Holmes et al., 2014; Turner and Holmes, 2015), which have also been widely reported. For example, some additives are added into plastics during the manufacturing process to obtain better stability. However, these additives could leach out from the plastics as they degrade in the environments because they are usually not covalently bonded to the polymers (Lenz et al., 2015; Gewert et al., 2015). Thus, the chemicals carried by plastics can bioaccumulate in organisms when the plastics are mistaken by organisms, which causes adverse effects on the normal functioning of organisms and even poses a potential health hazard to human beings (Eerkes-Medrano et al., 2015).

Plastic debris, including microplastics, could persist in the environment for hundreds or even thousands of years due to its stability and durability (Barnes et al., 2009; Gewert et al., 2015). In addition, larger plastics in the environment would continually degrade into microplastics (Andrady, 2011; Wang et al., 2016) as a result of physical, chemical and microbial effects, resulting in an annual increase in the amount of microplastics. Thus, it is essential to understand the degradation processes of plastic particles to increase our understanding of the source of microplastics, predict the fate of microplastics in the environment, and evaluate the potential environmental hazards to organisms. Currently, although some studies regarding the degradation of plastics have been conducted under outdoor conditions (Rajakumar et al., 2009; Azuma et al., 2009) or accelerated weathering conditions in laboratory experiments (Stark and Matuana, 2004; Song et al., 2017),

studies on the degradation of plastics in the aquatic environment that hydrobios reside in have been limited, especially for plastic particles of small size (<5 mm). In addition, knowing the degradation process of plastics in different environments is critical for calculating their residence times in different environments and providing a basis for evaluating the status of plastic pollution in these environments. This study focuses on the secondary source of microplastics (i.e., degradation) and intends to investigate similarities and discrepancies in the degradation process of plastics for different environments. Thus, observation of the degradation of three types of virgin plastic pellets exposed to UV irradiation in three different environments (i.e., simulated seawater, ultrapure water, and a waterless (air) condition) was carried out. Data on the changes in physical and chemical properties were collected, and comparisons of plastic pellets in three different environments and under different exposure times were also performed.

## 2. Materials and methods

### 2.1. Samples and experimental setting

Three types of commonly used plastics (i.e., polyethylene (PE), polypropylene (PP), and polystyrene (PS) virgin plastic pellets; 150 items, respectively) were obtained from DuPont Engineering Polymers, and three different simulation environments (i.e., simulated seawater (26.5 g/L NaCl + 24 g/L MgCl<sub>2</sub> + 0.73 g/L KCl + 3.3 g/L MgSO<sub>4</sub> + 0.2 g/L NaHCO<sub>3</sub> + 1.1 g/L CaCl<sub>2</sub> + 0.28 g/L NaBr), ultrapure water, and a waterless (air) condition) were established. In addition, UVA340 lamp was selected as simulated natural sunlight because it can give a good reproduction of natural sunlight (Brennan, 1987). First, the samples were placed evenly on glass dishes, and the samples were just submerged in the simulated seawater or ultrapure water solution environments (Fig. 1) using a dropper. Then, the glass dishes were transferred into homemade environmental chamber (Fig. 2; 60 cm × 30 cm × 30 cm for length, width, height, respectively) that was equipped with a UV lamp (UVA340).

This aging experiment, with a period of 3 months (one month was 30 days, which was defined as a cycle), were conducted with continuous UV irradiation. 15 of the plastic pellets in each solution environment were randomly removed in each cycle, and then the surface solutions of the removed samples were dried with absorbent papers. After that, these samples were dried at 60 °C and stored in glass containers in the dark (Brandon et al., 2016). Meanwhile, 15 of the plastic pellets in the air environment were also randomly selected out of each cycle for analysis.

### 2.2. FTIR analysis

For the FTIR analysis, 5 items of each type of virgin plastic pellet (PE, PP and PS) were analyzed using FTIR spectroscopy (Nicolet 6700, Thermo Fisher, USA) equipped with an attenuated total reflectance (ATR) diamond crystal attachment in order to establish their reference spectra. Ranges of the spectra were set from 4000 to 500 cm<sup>-1</sup>, and the collection time was 16 s for each measurement. In addition, 5 items of each type of weathered plastic pellets (treated by different UV irradiation times, i.e., 1 month, 2 months and 3 months) were randomly selected and analyzed by FTIR spectroscopy. Three different points (i.e., the central point and two different edge points) of each sample were examined. Among these resulting spectra, the representative resulting spectra were selected and compared with their reference spectra without post-processing or transformation after each experimental cycle.

### 2.3. Raman analysis

For the Raman analysis, 5 items of each type of virgin plastic pellet (PE, PP and PS) were examined using a Raman spectroscopy (LabRAM HR 800, HORIBA, France) with a laser wavelength of 632.8 nm in

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