Microplastic release from damaged commercial teabags

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Abstract. The use of plastics in our everyday lives have been drastically increased during the last few decades. However with the usage of commercial plastic products there is a possibility of microplastic consumption, due to the fragmentation of the products. Additionally, the potential for microplastic ingestion may also be increased by using damaged products. Hence, the current study was conducted to understand the potential release of micro/nano plastics and organic matter from damaged teabags. To check the leakage tendency, the amount of damage to the tea bags from 1-10 cm were tested along with temperatures of $25-70^{\circ}$ C, and exposure times from 5 min to 1 hr was tested. Release of fibrous micro/nanoplastics, and organic leachate from the damaged teabags were observed to understand the outflow conditions. Results showed that with the increased degree of damage, temperature, and exposure time increased the release of fiberous matter, where the increase of temperature, and exposure time increased organic leachate. Additional analysis confirmed the leachate of nylon polymers into the heated water.

Keywords: microplastic; microplastic release; organic leachate; SEM analysis; teabags

1. Introduction

Plastic, widely used in our daily lives, due to its light weight, durability to mechanical stress, and cheap prices. However, plastic waste is emerging as a serious environmental problem due to its release into the environment. Plastic production has increased by approximately 42% worldwide in the last decade, reaching 359 million tons in 2018 (Kyu et al. 2018). Microplastics (MP), a small item of plastic waste, is becoming a concern due to the global distribution and long-term interaction between biota and the aquatic environment (Lu et al. 2016, Mato et al. 2001, Lagarde et al. 2016). MP does not have a universal definition of size, but generally a diameter smaller than 5mm is accepted as a standard. Examples of MPs include microbeads (primary sources) used in cosmetics or associated with industrial spills, or fragments separated from larger plastic waste due to ultraviolet radiation, oxidation, or mechanical wear (secondary sources) (Qin et al. 2020).

As concerns about MPs research increased, scholars reviewed the literature covering different aspects of the field. Readily, the number of publications was less than 100 per year in 2014 and this is when MPs research grew significantly. And as a result of the number of MP-related publications have increase during 1703 to 2020 (Qin *et al.* 2020). Hot spots in MPs studies include the source of MP formation, its spatial distribution, analysis methods, accumulation and potential effects on organisms. It has been reported that for the accumulation of MP in various

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Copyright © 2022 Techno-Press, Ltd. http://www.techno-press.org/?journal=mwt&subpage=7 organisms include mussels, fish, seafood, food nets, resulting in toxicity to human health (Qin *et al.* 2020). A study found that Chinese shellfish consumers could be exposed to 100,000 MPs a year because nine kinds of bivalves purchased from the Shanghai fish market are contaminated with MPs, and European shellfish consumers can consume up to 11,000 MPs a year based on the MP collected from Pacific oysters (Wright *et al.* 2020).

As such, MPs often occur not only in food but also in household goods and tap water. For example, MPs were found in take-out containers made of common polymer materials in five cities in China, especially in containers with rough PS surfaces (Du *et al.* 2020). It was also reported to be a large source of PP and PS, MPs found in WWTP, and white fiber from wet tissues and sanitary towels used in daily life (Briain *et al.* 2020). In the case of drinking water, about 75 percent of MPs, or 0.1 to 1mm in size, were reported in the free drinking zone of 42 stations of Mexico Metropolitan Subway (Shruti *et al.* 2020), and in the case of Hong Kong Metropolitan area, fiber-shaped MPs, or 1mm in size, were found in all samples of 110 tap water (Theresa *et al.* 2020).

Due to its abundancy in domestic environments, exposure of MP have been reported to potentially cause various toxic effects to humans. In a cell-based experiment using PP particles made by decomposition of solid PP, the presence of PP particles of less than 20 μ m increases ROS and causes toxicity to PBMC and Raw264.7 cells (Hwang *et al.* 2020). Other studies have reported that small PS particles with a diameter of 460 nm-1 μ m and PE particles up to 50 μ m adhere to each other by weak interactions such as van der waals forces and the affects of erythrocytes. The results showed that small particles were associated with an inflammatory response in surrounding tissues, including macrophage immune activation and cytokine production

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(Hwang *et al.* 2020). Thus, the potential health effects of MPs are largely dependent on the characteristics of the particles, and side effects on small size plastics can be inferred. Additives used to make plastics have also been reported to cause toxic effects. Additives used in making plastics include di-n-octyl phthalate (DnOP), di(2-ethylhexyl)phthalate (DEHP), Bisphenol A and styrene. DEHP, a chemical additive, is already known to cause reproductive toxicity (Wright *et al.* 2020). Bisphenol A and styrene are suspected endocrine disrupting chemicals (EDC). Indeed, it has been found in urine, blood, breast milk and tissue samples, and the common mechanism is intracellular transmission of MPs (Rist *et al.* 2020). MP particles accumulated in the human body can cause inflammation, and plastic additives can be found in human samples, causing endocrine disturbance.

The purpose of this study was to duplicate cases where tea bags are defective or damaged while opening the packaging, and to investigate the tendency of MPs to flow out due to the defect rate of plastic products using damaged plastic teabags. After exposing the damaged teabags under various environments, MP particles were examined using an optical microscope and an electron microscope (SEM). Additionally, TOC and MALDI-MS analysis was used to verify the amount of organic leachate from the teabags as well. The current research confirmed that damaged teabags generally tend to leak MPs and organic matter under exposed environments.

2. Materials and methods

2.1 Sample preperation

For the experiments, 6 different teas packages were purchased through commercial venders in Korea. The purchased teabags were composed of 5 different materials; polyethylene (PE), Nylon, polyethylene terephthalate (PET), polyamides (PA), and polylactic acid (PLA). For sample preparation, the teabags were cut with steel scissors to remove the tea leaves. The teabags were emptied and washed three times using deionized water to remove residual particles or tea leaves. Teabags were then dried in a clean bench for a minimum of 24 hr, and stored in a sterilized environment. To emulate damaged products, the teabags were cut with scissors under various different degrees.

2.2 Experimental procedure

To simulate the conditions of everyday usage, the damaged teabags were inserted into 40 mL glass vials and heated under various temperatures while shaked in a constant temperature bath. Three teabags were inserted into one vial each and was heated for five minutes to one hour and shaked at 70 rpm. The vials were sealed with teflon tape to prevent any leakage or contamination from the temperature bath. After the steeping process, the teabags were removed from the vials and residual water was filtered. Mixed Cellulose Ester filter paper (1.0 m pore size,

Advantec MFS Inc, USA) was used to filter the samples for residual microplastics. Prior to the filtration process, the filter paper was rinsed and filtered with deionized water to prevent contamination from the filters. After the samples were filtered, the filters were analyzed to verify the existence of micro or nano particles and fibers. The filter paper was dried at room temperature for 24 hr in a clean bench. Optical microscopy was used (Smartoy Touch Digital Microscope X1,000, Smartoy, China) to count microplastic particles and fibers on the dried filters. To count the fibers a counter was used, where the microscopy magnification was set to X1000. Image of the samples were taken with the microscope's digital imaging device. The water from the filtered samples were analyzed with a TOC analyzer and SEM to verify the amlunt and characteristics of the leachate from the teabags.

2.3 TOC and MALDI-MS analysis

A TOC Analyzer (Sievers 5310C Laboratory TOC Analyzer coupled with Sievers 900 Autosampler, General Electrical, Germany) was used to verify the amount of leached organic matter from the teabags. The flow rate for the acid was set to 1 L/min and the oxidizer was set to 2.5 μ L/min.

To verify the characteristics of the leached organic matter from the teabags, MALDI-MS analysis was conducted. 10 mg of 2,5-dihydroxybenzoic acid (DHB) was added to 1 mL of 50% CAN containing 1% phosphoric acid, or 1% trifluoroacetic acid (TFA) was added to distilled water to synthesize a DHB matrix solution. The teabag leachate solutions were mixed with the DHB matrix solution on a 1:1 (v:v) ratio. Mass spectrometric analyses of crystallized mixtures were performed using a MALDI-time-of-flight MS instrument (IDSys LT; ASTA, Korea, CNU chemistry core facility), which was operated under the following conditions: 30 laser shots of a 349 nm Nd:YLF UV laser, 2000 ns pulse width, 100 Hz pulse repetition rate, and positive ion linear mode.

2.4 SEM analysis

Electron microscopy was used to observe the samples that were not captured with the filtration process. Images were captured and analyzed using a Field Emission Scanning Electron Microscope SU8000 (Hitachi High-Tech, Japan) (FE-SEM), with a magnification range up to X30-X800,000. The plastic tea bags were fixed with carbon tape to analyze the shape, size, and length of the tea bag plastic fibers. For sample preparation, the filtered water was carefully dropped onto a silicon tape, and dried in a desiccator for SEM imaging. The samples were coated with Au using a sputter coater (208 HR High Resolution Sputter Coater, TED PELLA INC, USA) for conductivity. The acceleration voltage was set to 15kV and the beam current was set to 10uA, image were taken at X10.0K. To verify the elemental composition of the potential microplastics, EDS analysis was additionally conducted on the samples. The samples were observed for elements other than carbon (C), nitrogen (N), and oxygen (O), which are the basic elemental



Fig. 1 SEM image of the cross section of damaged tea bags of various chemical compositions

composition of plastic materials. After setting the working distance (WD) to 15mm, the image was sent to the EDS computer and was run using Aztec programming.

3. Result

3.1 Characteristics of plastic teabags

Fig. 1(a)-1(f) shows an SEM image of the teabag cross sections that were observed in the current research. As seen in the figure, the teabags composed of PLA, Nylon, and PA are fabricated in a woven fashion with an even thickness of 10 μ m for each fiber. Figs. 1(d) and 1(e) are images of teabags composed of PE and PET, which are made by non-woven fabric structure where the fiber aggregates are combined by physical or chemical means. Unlike the woven teabags, the non-woven structure showed plastic fibers of various thicknesses ranging from 10-30 μ m. The figures show that the non-woven structures have comparatively

uneven surfaces on the edge of the damaged areas. Fig. 1(f) shows the image of a teabag submerged in a 75° C water bath for 1 hr. Compared with the pristine teabags, the heated samples displayed cracks, and fragmentation of the surface when exposed to heat. Similar results have also been observed; where the BET specific surface of MPs are reported to have an approximate 50% increase after a 6-week exposure to heat and additional aging conditions, due to cracks on the surface of the plastic (Luo *et al.* 2020). Additional observation relating heated teabags have also indicated a higher amount of microplastic release when the teabags are exposed to temperatures temperatures of 95°C compared to samples exposed to 22° C (Hernandez *et al.* 2020).

3.2 Micro-fiber release from plastic teabags

3.2.1 Observation of microplastic fiber release

After filtering, the filter paper was dried to observe MPs fibers and particles under an optical microscope. As depicted



Fig. 2 Optical image of micro plastic fiber and particle release from the damaged teabags. The different color particles and fibers show the released fibers from the damaged teabags

in Fig. 2, it shows the MPs fibers and particles by using optical microscopy. The image shows tea bag fragments of various sizes larger than $1\mu m$ from the damaged areas of the tea bags. Under optical microscopy, plastic particles showed various colors, shapes and sizes, based on the origin of the teabags.

3.2.2 Fibers release from plastic tea bags

The release of MPs fibers was examined under different temperature, exposure time, and degreases of damage to the teabags (1, 2, 3, 5, 10cm). Figs. 3(a) and 3(b) present the amount of fibrous microplastic release with the degree of damage to the teabag, and the time of exposure to heated water (75°C). Results shows that with the increase of damage to the teabags, the MP fiber release also increase, with PA having the largest number of release. In average, based on the composition of the teabags PE may release 13-20 fibers, Nylon1 1,2 release 7-15 fibers, PA release 8-25 fibers, PET release 8-21 fibers and PLA release 14-19 fibers on average. Based on the results, it can be estimated that with a single tea bag it is possible to ingest approximately 10-19 fibers per sharing (>1 μ m in size).

The amount of MP fiber release, based on the time of water exposure was also verified. Fig. 3(b) shows the results of teabags with 3 cm damages, which were submerged and shaken in 75°C water for different time periods. Results show that the majority of fibers release, occurred during the first 30 min of exposure. After 30 min of exposure, the amount of fiber release reaches a plateau, resulting in a maximum amount of release. To be more specific, tea bag made of PE that was heated for 30 minute release 21 fibers, Nylon 1, 2 released 33 fibers, PA released 20 fibers, PET released 21 fibers and PLA released 19 fibers each. It was estimated in the current experiment that

when one plastic tea bag has a 3cm damage and the leaching time is 5min to 1hour, a person may ingest approximately 5-19 fibers (>1 μ m in size).

The amount of fiber release with time show similar trends with the experiment conducted with different degrees of teabag damage. In both experiments, PA displayed the highest amount of release compared to the other plastic materials. This is due to do the fact that structure between the polymers open and swell at temperatures above the glass transition temperatures, decreasing the strength and flexural modulus, and increasing the flexural stress. The decreases flexural modulus, and increased the flexural stress results in the higher degree of plastic fiber release (Ma et al. 2020). As displayed in Fig. 3(d), the glass transition temperature of PA is the lowest among the materials being approximately 35°C, making the material more flexible than the other materials, resulting in a higher possibility for the fibers to be released from the teabags. The trend between the temperature and fiber release are shown in Fig. 3(c). Teabags were exposed to temperatures of 25°C, and 75°C, which are above and below the glass transition temperatures of the plastic materials. Results show a high amount of fiber release of the teabags that were exposed to temperatures higher than their glass transition temperature, indicating that the glass transition temperature has a correlation with the micro plastic release of the teabags.

3.3 Organic leachate from plastic tea bags

3.3.1 TOC result of teabag leachate

When plastics are exposed to water for a long period of time it has been reported that the constituents of the plastic material can leach from the material. So to verify whether plastic teabags may leach constituents, experiments were



(a) The number of fibers released based on the size of damage (1, 2, 3, 5, 10 cm)



(c) The number of fibers released under various temperatures (25°C ,75°C)



(b) The number of fibers released by different exposure time (5,10,15,30 min,1 hr)



(d) Correlation between the glass transition temperature and fiber release

Fig. 3 Fiber release after exposure to various temperatures

20

15

(b)







(c) The organic leachate under various temperature (25°C, 50°C, 75°C) conditions

(b) Organic leachate under various exposure times $(5, 10, 15, 30 \min, 1 \ln)^{20}$



(d) Correlation between the glass transition temperature and organic leachate

Fig. 4 TOC result of filtered water for the 6 plastic tea bags based on various experimental conditions



Fig. 5 SEM imaging of filtered water for the tea bags; The numbers in (a), (b), and (c) show the thickness of the released fibrous microplastics

conducted under various conditions and analyzed for organic matter. Fig. 4(a) shows the amount of organic leachate for teabags with various amounts of damages. Results showed that the amount of damage did not influence the amount of leachate into water, indicating that organic leachate mainly occurs on the outer surface of the tea bag. Among the teabags, nylon and PA had the highest amount of leachate, due to the lower glass transition temperature.

Addition to the amount of damage, the exposure to various time durations to water was also tested. Fig. 4(b) shows the TOC results of organic leach ate from teabags submerged at temperatures of 75°C. Results show an increase after the first 10 minutes to exposure and a general increase after 15min. The amount of leachate was highest with nylon, and PA followed by PE, PLA, and PET. Results of organic leachate showed a high correlation with the glass transition temperature of the plastic materials. The relationship can be verified by comparing the amount of leachate in Fig. 2(b) with the glass transition temperature and organic leachate relationship in Fig. 4(d). The relationship shows an inverse correlation between the glass transition temperature and organic leachate. This relationship can again be proven with Fig. 4(c), where the amount of leachate is measured by plastic material at different temperature exposures. To verify the amount of leachate at temperatures above, bellow and approximate to the glass transition temperature, teabags were exposed to waters of 25, 50, and 75°C each. Result show that with PA the amount of leachate drastically increases when exposed to water of temperatures above the glass transition temperature (35°C), compared to exposure conditions with lower temperatures (25°C). With Nylon, the leachate experiments were each conducted at temperatures below, above and approximate to the glass transition temperature.

Results showed a gradual increase of the leachate, where the organic leaching increased at the glass transition temperature and further increased a higher temperatures. And with PLA, and PET the leachate increases at temperatures only higher than the glass transition temperature. This phenomena is due to the swelling effects of the polymers at temperatures higher than the glass transition temperatures. The swelling of the polymers result in space between the bonds making it possible for water to enter between the polymers or increase the diffusion of additives in the plastics or organic matter between the MPs fibers to leach out. The release of plastic monomers or additives is dictated by the Fick's model. Various studies have also verified the water adsorption, and diffusion behavior of composite materials (Ma et al. 2020, Hashin et al. 1980, Brahim et al. 2007). The diffusion behavior has been observed in higher degrees with aged MPs, where chromium diffusion from plastics with longer aging time leads to greater release. Additional research indicated that with freshwater, more leaching from MPs were observed, because of the weak- alkaline complexation at longer aging periods (Lou et al. 2020).

3.3.2 SEM imaging of teabag leachate

To verify the release of microplastics from the teabags, filtered water was observed with SEM imaging. A negative control of filtered deionized water was also observed with the SEM, to show no exertion of microfibers from the membranes. Fibers of various lengths were observed with thicknesses of 1-5 μ m, indicating the existence of microplastic exertion from the teabags. The thicker fibers observed may be due to inconsistencies of the membrane. This is consistent with previous studies where microplastics of up to 127 μ m was exerted from teabags (Hernandez *et al.* 2020).



Fig. 6 MALDI-MS analysis of filtered samples from teabags composed of nylon (Nylon 1 and Nylon 2)

To identify whether the fibers and the particles observed are of polymeric materials, EDS analyzer was used to evaluate the elemental composition. Results of the fibers in Fig. 5 showed a composition of only carbon, oxygen, and nitrogen. Fibers and particles were compared with the elements of tea bags to show a matching elemental composition.

4. Conclusions

In the current study, the environmental impact on teabag leachate was investigated. Results showed that the material property, and manufacturing processes effects the leachate and the release of MPs fibers. To verify the factors that affect teabag leaching, the degree of damage to the teabags, temperature, and exposure time to elevated temperatures were tested. Results showed that the release MPs fibers increases as the damage of the tea bag increased, and as the increasing of time exposed in hot water. Additionally, the temperature, and exposure time were found to be the major contributing factors for organic leachate from teabags as well. The increased release of micorplastic fibers and organic leachate was due to the glass transition temperature. The elevated temperature above the glass transition temperature transforms the polymer into an amorphous, flexible structure, with the addition of swelling of the polymer, increasing the fiber release and organic leachate. The results in the current research may aid in understanding microplastic release from daily consumer products, as well as their potential risks to human health.

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