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Unveiling Microplastic Leaching from Food Packaging Polyethylene Covers: A Preliminary Study

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Plastics became the preferred choice for food packaging due to their affordable cost, production efficiency, durability and application versatility. Small scale food beverage shops in metropolitan and urban areas in India frequently employ single-use plastic for packaging hot beverages and other food items, but the inadequately addressed issue of microplastic seepage, particularly from low-density polyethylene (LDPE) covers, remains a concern. The objective of the study was to identify the leachate of plastics from food packaging covers as a result of exposure to hot water at a temperature above 80 ºC. Microplastic sample treated with Fenton's reagent and filtered using Whatman® grade GF/C microfiber filter paper and visually inspected using a light microscope. The morphological identification and chemical composition of microplastics was done using scanning electron microscope-energy dispersive X-ray spectroscopy (SEM-EDS). Nature of microplastics was characterized using reflectance Fourier transform infrared (FT-IR) analysis. The study observed the presence of around 20-30 microplastics with an average size ranging from 40 to 200 µm. Further, the EDS analysis confirmed the presence of microplastic particles in the filtered leachate, as indicated by the carbon peak at 0.3 to 0.4 KeV. Furthermore, the FT-IR analysis showed the presence of polyethylene microplastics leached from the tested samples. Despite the ban on single-use plastics, their persistent use in food packaging necessitates this study to raise awareness about microplastics and their impact on human health.

Keywords: Food packaging, Leachate, Microplastics, Plastics, Toxicity.

INTRODUCTION

Plastic production has increased significantly since 1950, resulting in large amounts of plastic waste that end up in landfills or the environment. Disposed plastics are harmful pollutants in the environment and can be classified as macro or microplastics. Microplastics, particles having dimensions below 5 mm, have raised concerns due to their impact on environmental quality and ecological system [1]. These particles are found in various shapes and sizes, which can be originated from primary or secondary sources. The primary microplastics are globally manufactured and can be found in household items [2-4], milk products [5], personal hygiene products [6] and 3D printers [7]. Secondary microplastics result from the degradation of plastics that are disposed including synthetic fibres and packaging materials [8]. Microplastics have been detected in marine habitats and are of increasing concern due to their presence in food, air and water, although their exact consequences on human health are still under investigation [9-15]. The microplastics infiltrate agricultural soils through diffusion, contaminating crops and affecting the delicate balance of the agricultural ecosystem. Microplastic pollution in agriculture is an emerging complicated issue that requiring a multi-pronged approach [16]. More recently, microplastics have been detected in packaged food ice cubes from some commercial brands in Mexico City with concentrations ranging from 19 to 178 L^{-1} [17].

The present study focuses on the leaching of microplastics from food packaging polyethylene (PE) covers, particularly in the context of hot beverages such as tea and coffee. In India, many tea shops use low-quality polyethylene covers for packing hot beverages, which raises concerns about the potential leaching of microplastics into the packaged drinks. When packaging materials are exposed to high temperatures; primarily results in the contamination of food and beverages with plastic additives [18]. Food grade plastics may release toxic substances upon degradation when exposed to temperatures above 40 ºC [19-21]. The study aims to raise awareness about microplastic leaching in food packaging materials, shedding a light on risk of hot beverages packaging. Although the study analyses water rather than tea or coffee directly, it highlights the ease with whether microplastics can leach at 80 ºC or above. The findings aim to inform and educate the general public about the presence of microplastics and their potential health consequences.

EXPERIMENTAL

The plastic food packaging covers used in this study were purchased from a supermarket in Ambattur, Chennai, India (13.1186º N, 80.1574º E). All the glasswares and food packaging covers were immersed and rinsed twice with RO water before being used. The food covers had dimensions of 11 cm × 9 cm and could hold approximately 100 mL of beverages.

Preparation of food cover sample: A 1 L RO water taken in 2 L glass beaker was heated to temperature of above 80 ºC on the house hold induction stove to reach the stage of boiling. To this hot water nearly 10 food covers were immersed and kept for a period of 30 min. Usually, within a period of 15 min any hot beverages would cool down to ambient temperature at which it can be consumed. In order to observe for the leaching at prolonged soaking, the covers were immersed for 30 min. After this period, the covers were separated and the water extract was decanted into the glass beaker. The sample water was immediately covered using glass plate to prevent any possible contamination. This was repeated ten times and a total of ten litres of water samples was collected. The water was stored in two glass bottles of 5 L capacity each and analyzed for microplastics. Water heated at temperature of 95-100 ºC without immersing the food packaging covers served as the control.

Microplastic extraction and filtration: For the microplastic analysis, two methods were applied. In one method, the sample water was filtered directly using Whatmann® grade GF/C microfiber filter (1.2 µm pore and 47 mm diameter) paper using a vacuum pump. In another method, 2 L of decanted water was treated with Fenton's reagent [22] by preheating the decanted water to 75 ºC and to this 13.9 g iron(II) sulphate (0.05 M) was added followed by addition of 20 mL of 30 % H_2O_2 to ensure the removal of any dissolved or suspended organic matters. After cooling the water treated with Fenton's reagent was filtered through GF/C microfiber filter using vacuum pump. For the control, the water was directly filtered using GF/C microfiber filter.

After the filtration, the GF/C microfiber filters was dried in a petri-dish at room temperature. A total of four samples were analyzed. Sample 1 includes the GF/C microfiber filter, sample 2 consists of plastic decanted water without treatment with Fenton's reagent filtered through GF/C microfiber filter, sample 3 and sample 4-plastic decanted water following treatment with Fenton's reagent filtered through GF/C microfiber filter. All the filter papers were then visualized under light microscope and subjected for further SEM-EDS and FT-IR analysis. List of food cover samples collected is summarized in Table-1.

Identification of microplastics

Light microscopic observation: The dried microfilter samples (1-4) were directly observed under a low magnification (10X) and then under high magnification (100X) to locate the presence of microplastics. The observations were made using a binocular microscope (Model: BM-01, Brand: ESAW, India). The locations of the microplastics in the samples were marked and cut for further analysis.

Scanning electron microscope-energy dispersive spectroscopy (SEM-EDS): The GF/C microfiber filters of all the four samples were analyzed by SEM-EDS technique. Morphological changes, elemental mapping and elemental composition of the GF/C microfiber filters were observed and determined for the control (sample 1) and test samples (samples 2-4) using FE-SEM (Model: Apreo S, Thermo Fisher Scientific, Netherlands) connected with energy-dispersive spectroscopy (EDS) at an operating voltage of 20 kV. The SEM images were recorded at 100, 40, 20 and 5 µm magnification to identify the presence of various particles with variable sizes. The EDS studies showed the elemental mapping and composition of leachate present on the GF/C microfiber filters.

Fourier transform-infrared (FT-IR) analysis: FT-IR analysis was used to identify the functional groups of control (sample 1) and any leachates present on the experimental GF/C microfiber filters (samples 2-4). IR spectra of the all the four samples were recorded in reflectance mode on FT-IR (Model: IRTRACER 100, Shimadzu, Japan) in the spectral range of 4000-650 cm^{-1}.

RESULTS AND DISCUSSION

Preliminary screening using light microscopy: The GF/C microfiber filters of all the four samples were observed under the light microscope for visual inspection of microplastics. Fig. 1a shows the GF/C microfiber clear filter without any microplastic leachate (sample 1). However, the light brown coloured circle shaped microplastic leachate observed in sample 2-4 as shown in Figs. 1b-d, respectively. In order to confirm the microplastic leachate, the GF/C filter were analyzed using SEM-EDS and FT-IR for sample 1-3.

TABLE-1
LIST OF FOOD COVER SAMPLES

Sample No	Details
Sample 1 (Control)	RO water filtered GF/C microfiber
Sample 2	Food cover treated water sample filtered GF/C microfiber without Fenton's reagent treatment
Sample 3	Food cover treated water sample filtered GF/C microfiber with Fenton's reagent treatment
Sample 4	Food cover treated water sample filtered GF/C microfiber with Fenton's reagent treatment

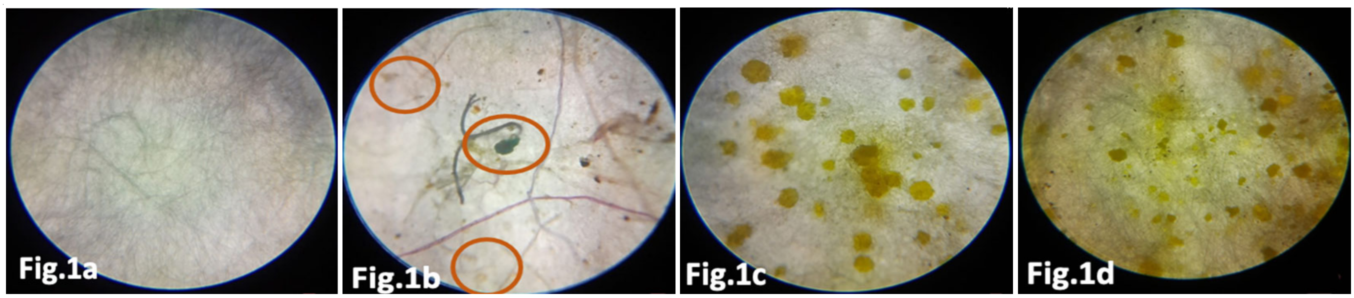

Fig. 1. Light microscopic observations (a) sample 1, (b) sample 2, (c) sample 3 and (d) sample 4

The Fenton reaction [22] refers to an advanced oxidation process in which Fe^{2+} initiates and catalyzes the decomposition of H_2O_2, leading to the *in situ* generation of hydroxyl and hydroperoxyl radicals. Normally, use of Fenton's reagent offers a simple, high speed and low-cost method for processing microplastics present within environmental samples [23]. The reagent is mainly used to isolate microplastics from organic rich wastewater. Thus, the use of Fenton's reagent brings about the lysis of any organic matter present and confirms the presence of microplastics alone. The light micrographs of samples 3 and 4 treated with Fenton's reagent clearly indicated the presence of microplastics alone.

SEM studies: Fig. 2a-c display SEM images taken at 200 µm magnification, comparing the control sample (sample 1) to the experimental water samples (samples 2 and 3) filtered through a GF/C microfiber filter. Fig. 2a shows a clear image of the GF/C microfiber filter, while Fig. 2b reveals the presence of microplastics in sample 2 without Fenton's reagent treatment. Fig. 2c-d depict SEM images of sample 3 and sample 4 after treatment with Fenton's reagent. These SEM images clearly demonstrate the presence of microplastic leachate ranging from 50 µm to 200 µm in size within an approximate area of 0.8 mm^2. The observations include 20-30 microplastic particles with sizes ranging from 40 µm to a maximum of 200 µm.

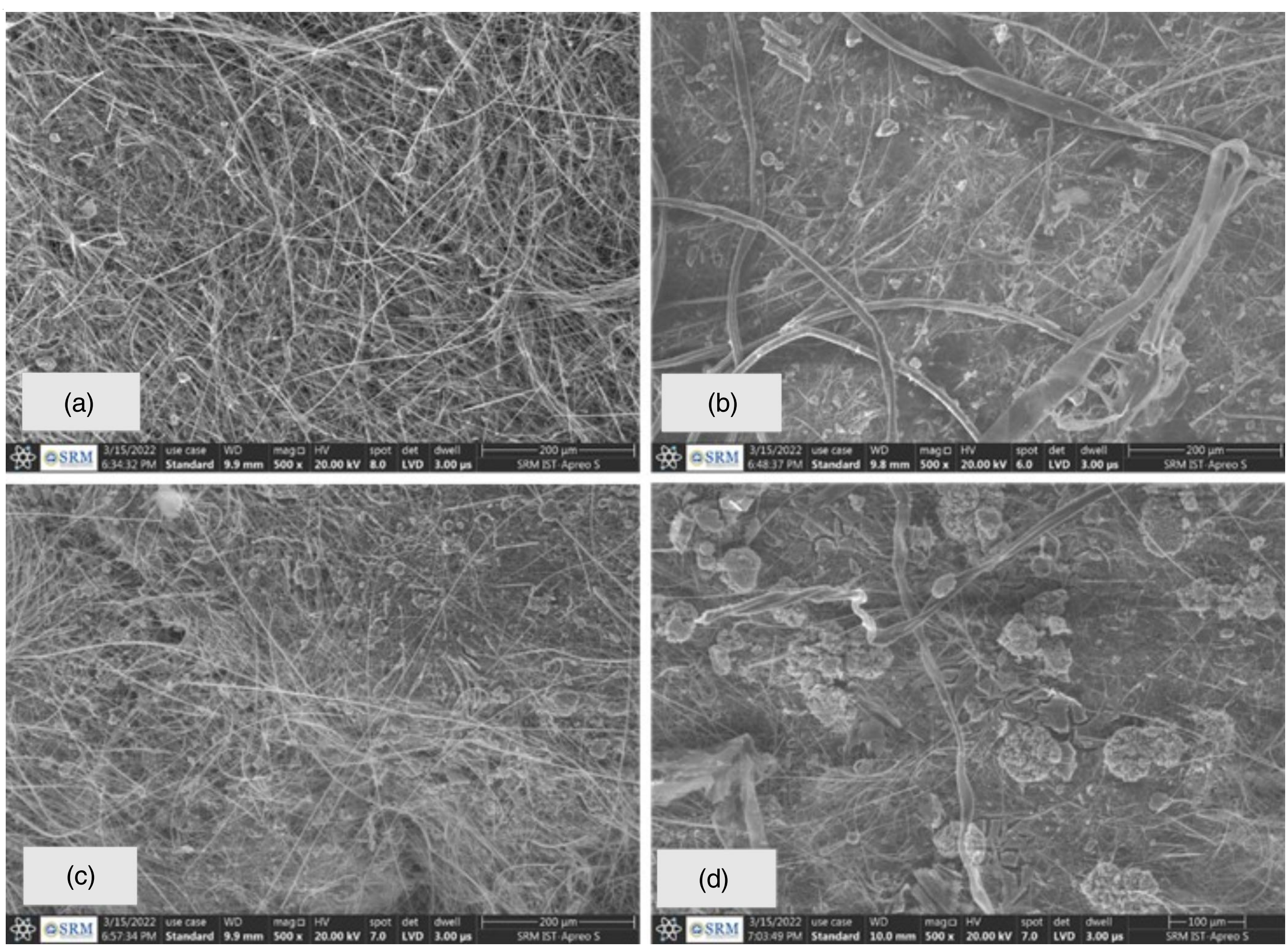

Fig. 2. Scanning electron micrographs at 200 µm (a) sample 1, (b) sample 2, (c) sample 3 and (d) sample 4 (recorded at 100 µm)

Fig. 3a-d present SEM images of all four samples captured at 40 μm magnification. Fig. 3a clearly displays clean fibres without any leachate, while Fig. 3b demonstrates the presence of microplastics with an average particle size of 5-40 μm, which were not clearly visible at 200 μm. Fig. 3c-d shows microplastic particles ranging in lowest size of 5 μm to maximum size of 40 μm following treatment with Fenton's reagent.

When observed at a higher magnification of 20 μm (Fig. 4b-d), the presence of microplastics with an average size of 2-10 μm becomes apparent. Sample 1 in Fig. 4a exhibits a clean glass fibres without any microplastics or any other particulate matter on the surface, while Fig. 4b (sample 2) reveals microplastics sized between 5-10 μm. In samples 3 and 4, after treatment with Fenton's reagent, a variety of smaller and larger microplastic fragments are observed (Fig. 4c-d).

At higher magnification, Fig. 5a displays a clear image showing glass microfibers, while Fig. 5b exhibits microplastics ranging from 5 to 7 μm in size. Treatment with Fenton's reagent effectively removes organic matter from samples 3 and 4, leaving only microplastics behind. Flake-shaped microplastics in samples 3 and 4 can be observed in Fig. 5c-d. The microplastics predominantly appear in oval-shaped particles and flakes. Overall, samples 3 and 4 exhibit the presence of microplastics ranging in size from 5 μm to 40 μm. Although these pieces of plastic are invisible to the naked eye, they can potentially enter the body through the consumption of hot beverages that are packaged in polyethylene covers. In nature, macroplastics can breakdown into microplastics due to physical, chemical or biological processes. These microplastics can take the form of filaments, granules, tubules and films [11,24].

EDS studies: Among the four samples, only samples 1 and 3 were subjected to EDS analysis for comparison. The EDX spectral results for sample 1 are presented in Fig. 6a and the elemental composition is summarized in Table-2. The major elements detected in the sample 1 are silicon and oxygen. The composition of silica and oxygen is significantly higher, indicating the presence of the GF/C microfiber filter (sample 1). Additionally, other elements such as magnesium, calcium, sulphur, iron, barium, zinc and indium were also identified. According to Table-2, the weight percentages of silicon and oxygen in the GF/C microfiber filter were 22.25% and 45.00%, respectively.

Table-2 displays the elemental composition of sample 3, which represents plastic water treated with Fenton's reagent. Fig. 6b clearly indicates the presence of microplastics in the filtered leachate, as confirmed by the carbon peak at 0.3 to 0.4 KeV. Most food packaging covers are made of polyethylene (PE) and carbon is a major component of PE plastics. Therefore, the study primarily detects carbon peaks associated with PE plastics. The EDS analysis of sample 3 confirmed the presence of carbon, with a weight and atomic percentage of 10.83%

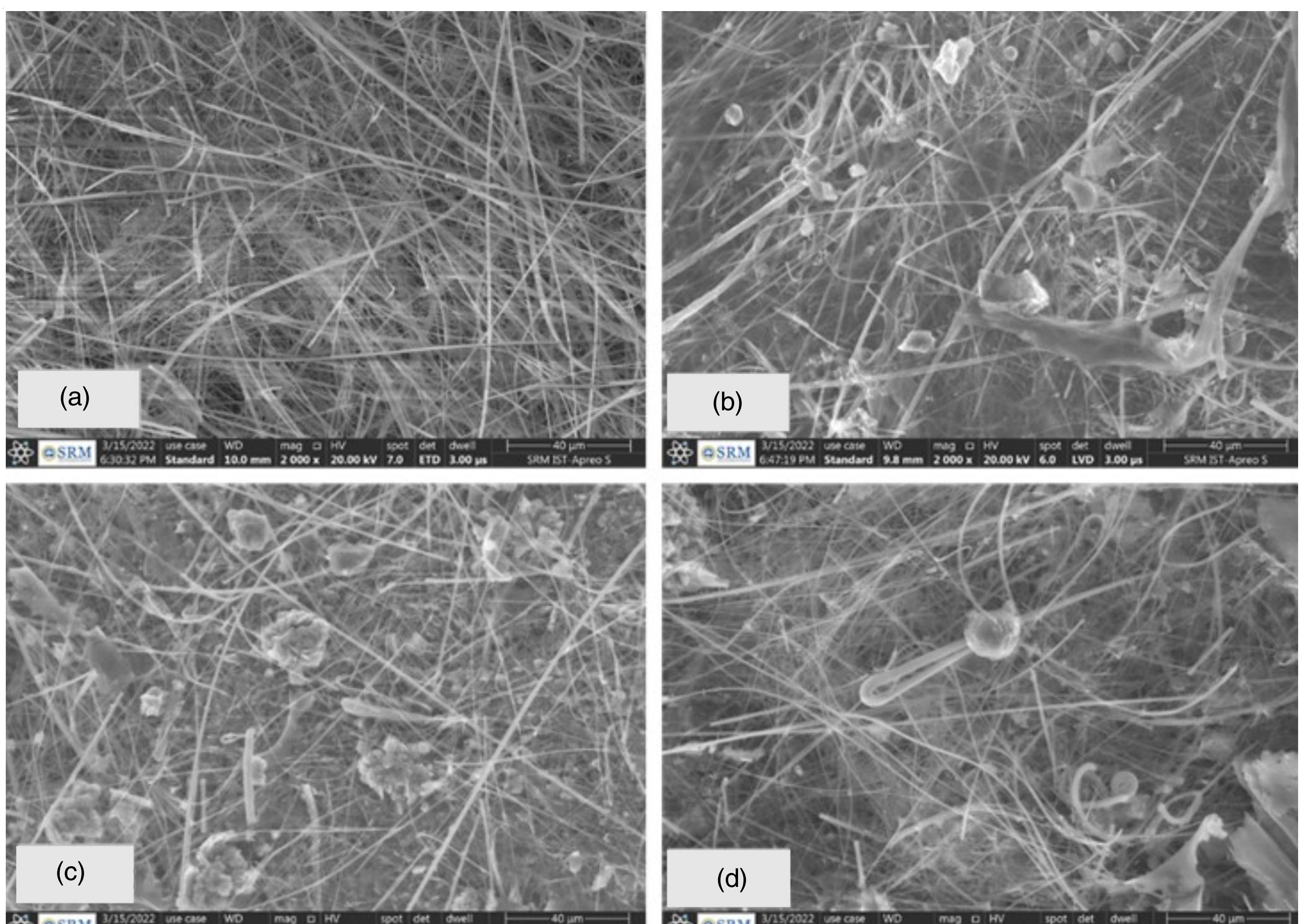

Fig. 3. Scanning electron micrographs at 40 μm (a) sample 1, (b) sample 2, (c) sample 3 and (d) sample 4

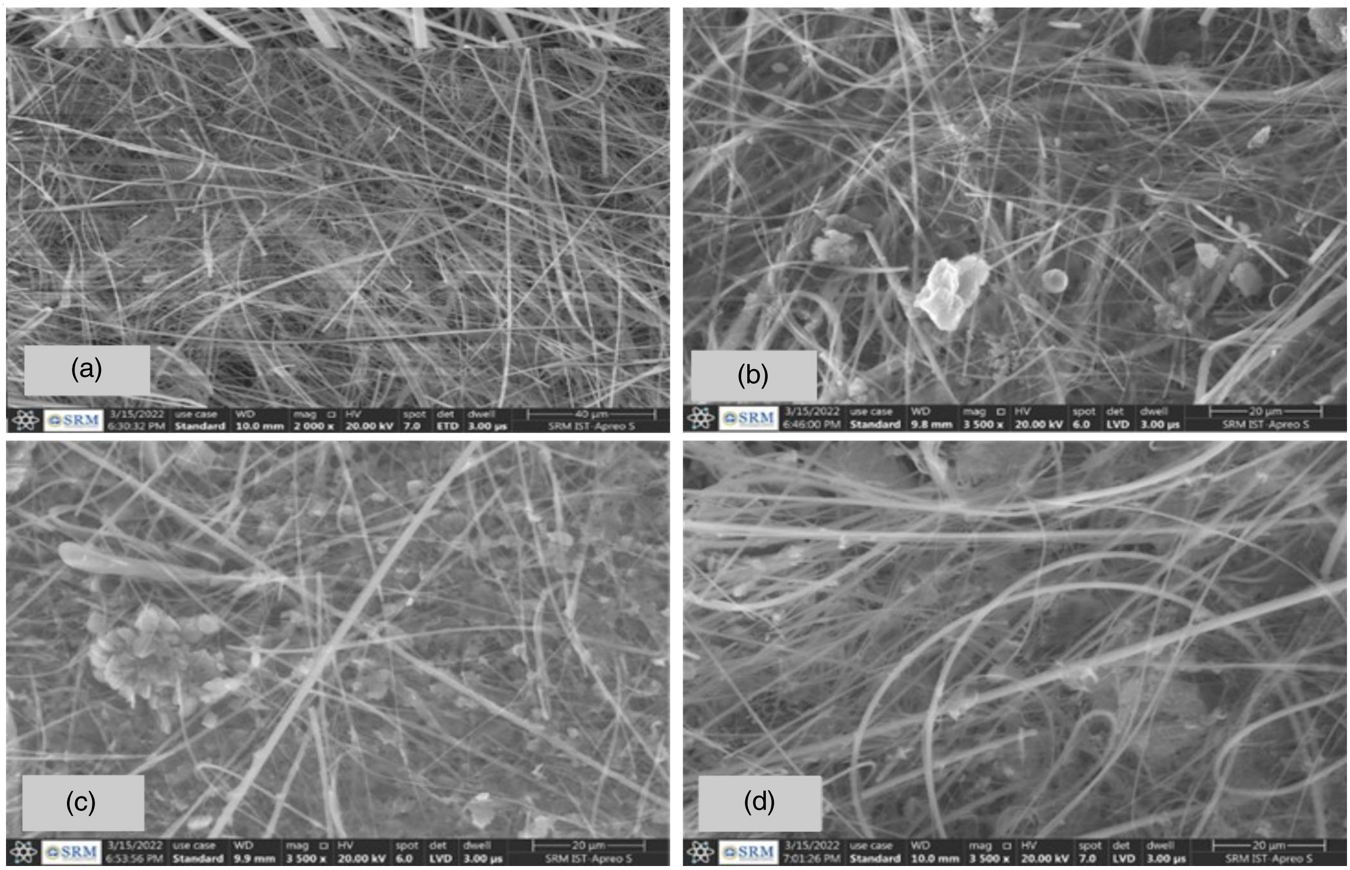

Fig. 4. Scanning electron micrographs at 20 μm (a) control, (b) sample 2, (c) sample 3 and (d) sample 4

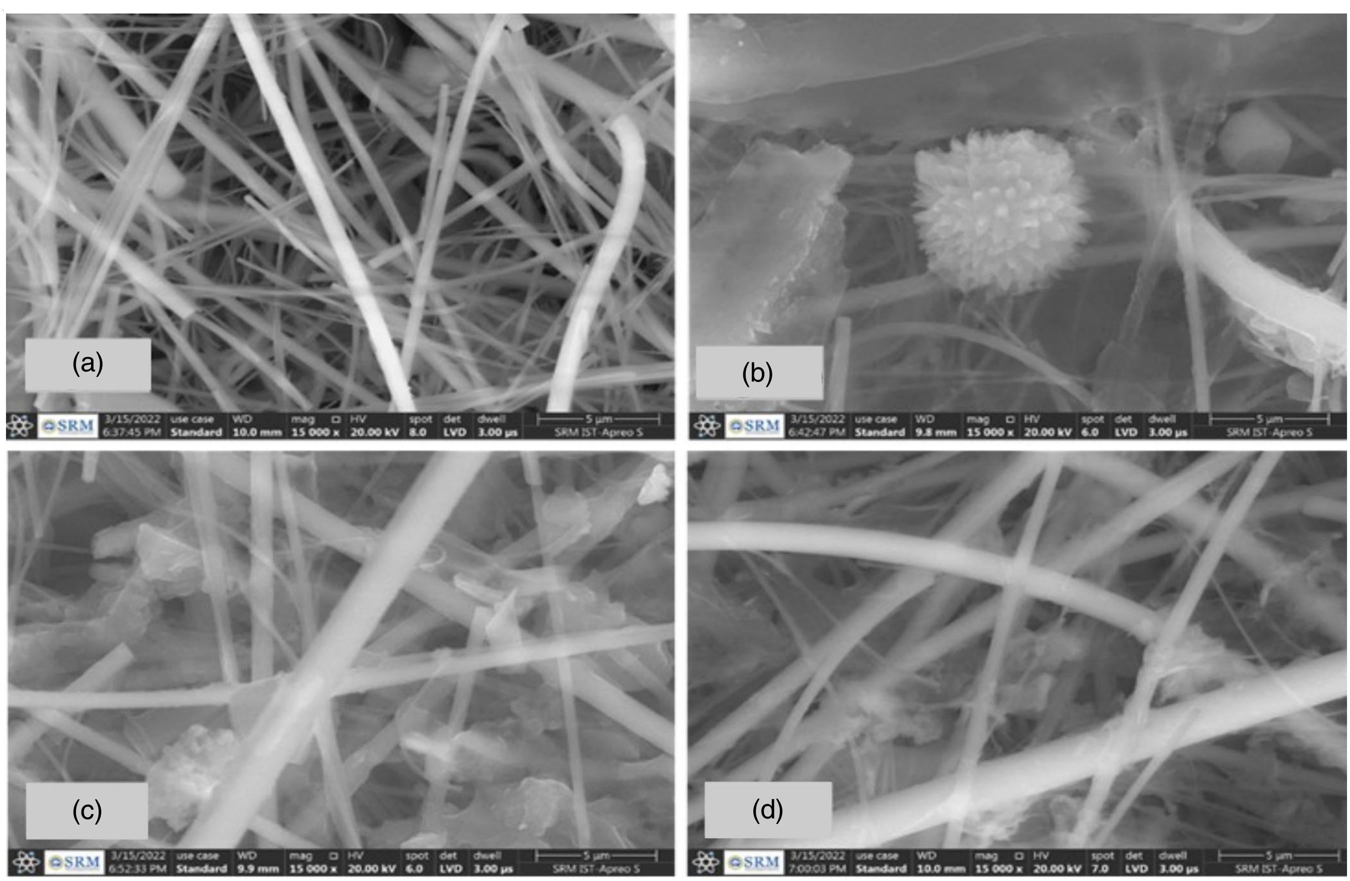

Fig. 5. Scanning electron micrographs at 5 μm (a) control, (b) sample 2, (c) sample 3 and (d) sample 4

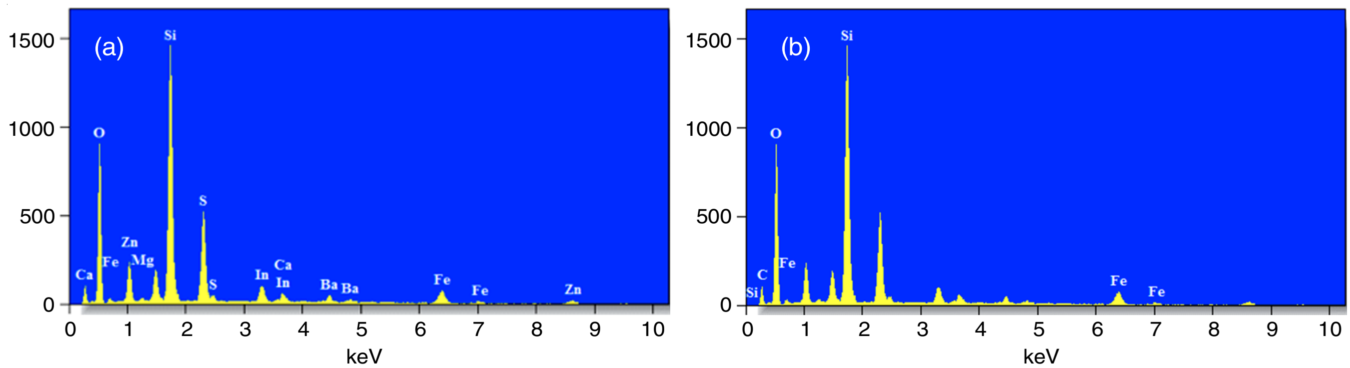

Fig. 6. EDX analysis of (a) sample 1 (GF/C), (b) sample 3 (GF/C with microplastics)

and 17.0%, respectively. The elemental composition results showed the presence of carbon-based material, which could be originated from PE plastic leachate. In addition, EDS analysis provided quantitative estimation of carbon, which is proportional to the amount of microplastic contamination.

FT-IR studies: Figs. 7a-d shows the FT-IR analyses of the samples 1-4. Invariably all the samples showed the characteristic peaks at 1060, 805 and 460 cm^{-1}, due to Si-O-Si symmetric stretching vibrations from silica GF/C microfiber filter.

In Fig. 7c-d, broad peaks at 3600-3200 cm^{-1} was observed which is due to -O-H stretching vibration and 1630 cm^{-1} due to -O-H bending vibration, arose from -OH functional group of polymers. Further, the broad peak observed between 3000 and 2900 cm^{-1} is due to -C-H stretching vibration of $-CH_2-$, alkyl chain of polyethylene polymers. Further, the peak 1060 cm^{-1} appeared as doublet in 1040-960 cm^{-1} region due to the C-O stretching vibration arising from polyethylene polymers. Thus, the FT-IR analyses strongly confirms the presence of polyethylene microplastics present in samples 3 and 4.

The findings of this study shed light on the significant issue of microplastic leaching from polyethylene food packaging covers commonly used in tea shops, food courts and mobile food corners. With the use of special Whatmann® grade GF/C microfiber filter (1.2 µm pore and 47 mm diameter), the microplastic leachate was filtered and filter paper was examined under the light microscope. The microfibers without any particles were seen in control filter paper. In sample 2 filter paper, that is without treatment with Fenton's reagent showed the presence of certain particulate matter. In order to confirm whether the leachate obtained in the current study is a microplastic or organic matter, Fenton's reagent was used to process the experimental water that is water immersed with polyethylene food packaging covers. Thus, it is evident that food packaging polyethylene covers causes leaching of fragments in the liquid, which it holds. As confirmed through SEM analysis where in the fragments of size 5- 40 µm were observed and EDS analysis showed the presence of carbon element only in the experimental sample and not in the control.

Conclusion

The study derives its conclusion based on water analyses rather than tea or coffee, with an idea that during brewing tea or coffee, the temperature is at or above 90 ºC and microplastics can easily leach at this temperature. It is concluded that low density polyethylene covers should be avoided for storing or for carrying the food samples, because the leaching or emission of microplastic from the plastic packaging is possible. This is particularly concerning because a large variety of foods are packaged or wrapped in plastic. Released microplastic particles could be directly ingested by the users. There is an urgent need to raise awareness among the people who has no knowledge of these microplastics. Additional research is needed to investigate the harmful mechanism and potential dangers of micro- and nano-plastics in higher concentrations in animals as well as in humans.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

TABLE-2
ELEMENTAL AND ATOMIC COMPOSITION

Element	Sample 1 (GF/C)			Element	Sample 3 (GF/C with microplastics)		
	Net counts	Weight (%)	Atom (%)		Net counts	Weight (%)	Atom (%)
O	5150	45.123	66.696	C	618	10.827	16.979
Mg	179	0.476	0.463	O	5292	51.423	60.540
Si	12974	22.247	18.732	Si	12992	29.242	19.611
S	5604	9.404	6.936	Fe	1201	8.508	2.870
Ca	991	2.136	1.260	Total		100.000	100.000
Fe	1201	5.851	2.478				
Zn	339	3.337	1.207				
In	2187	7.702	1.586				
Ba	803	3.725	0.641				
Total		100.000	100.000				

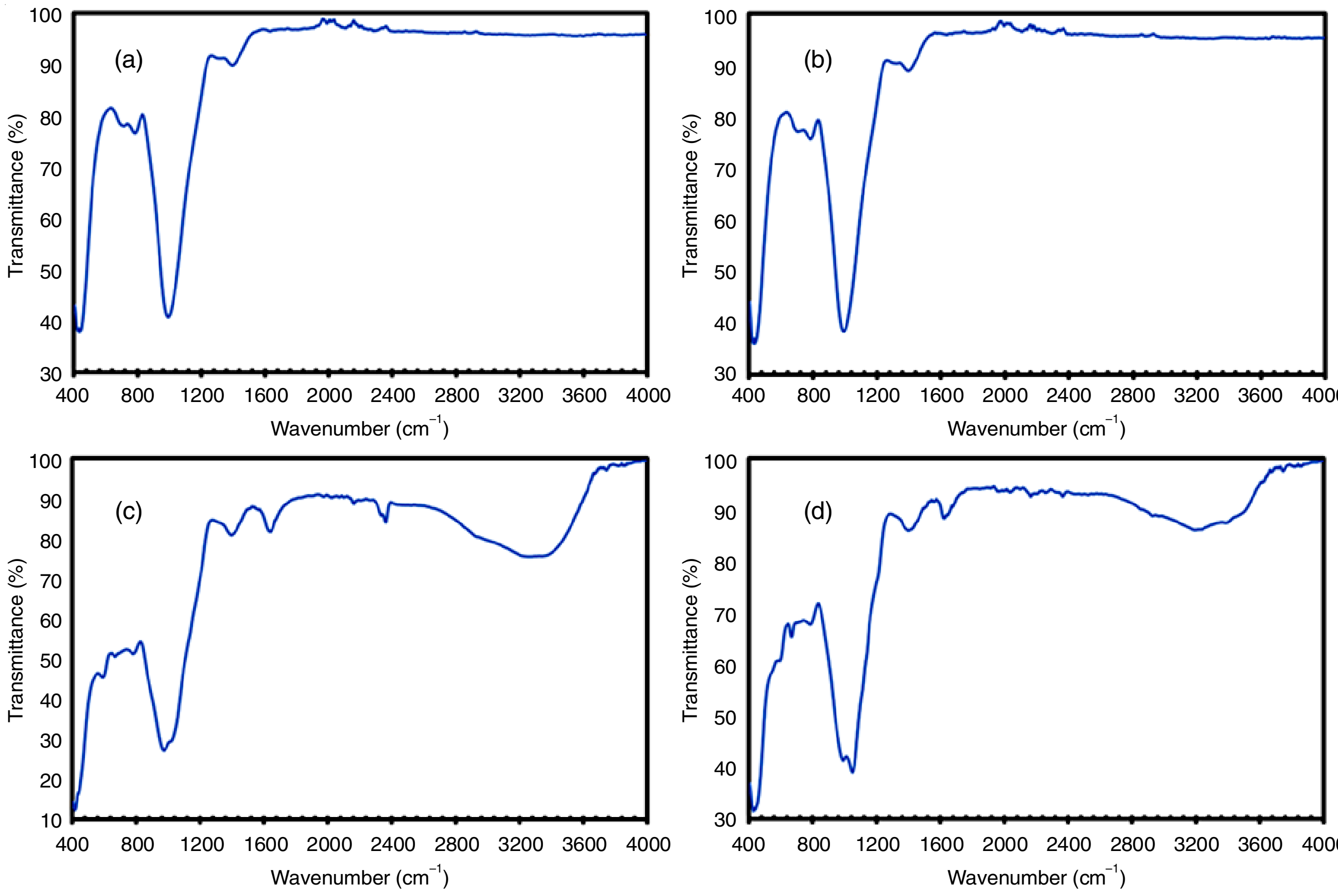

Fig. 7. FT-IR analysis (a) sample 1, (b) sample 2, (c) sample 3 and (d) sample 4

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