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Release of contaminants from polymer surfaces under condition of organized fluid flows

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ABSTRACT

The use of polymers for water storage or distribution is closely monitored, especially with regard to the possible contamination with substances coming from the material's surfaces. Different standards are practiced across countries according to type of applied materials and such test methods are prevalently based on constant temperature conditions. However, these polymers systems could be located in diverse environment which does not necessarily provide constant conditions. Experimental findings show that exposure of liquid inside polymeric materials to specific temperature gradients, and consequently to emerging organized flows, can result in an accelerated leaching of undesirable substances from the solid surface. In presented work model steady-state and organized flow conditions are used to compare release of contaminates from polyethylene by measuring of surface tension, UV–Vis spectroscopy, FTIR, scanning electron microscopy and elemental analysis of polymer surfaces and water leachates. The pilot study shows that convective flow generated via temperature gradient significantly affects contaminant release in comparison to a steady state and mixing flow conditions.

Introduction

Problems that may arise from insufficient water quality during transportation to the consumers represent potential risks to the health of living organisms. Apart from the well-established water treatment and quality control before distribution, the pipeline systems, which are used for the water delivery, represent a critical component that can significantly affect water quality. Currently, most newly installed and replaced pipes are made of polyethylene (PE). This material is favored for its numerous beneficial chemical and mechanical properties, making it a cost-effective solution widely used in both private and industrial water supply systems. These systems predominantly consist of polymers, comprising approximately 70–99 % of the materials, with the remaining percentage made up of various additives that enhance processability, mechanical properties, stability (e.g., antioxidants, flame retardants), appearance (e.g., dyes), and cost-efficiency (e.g., fillers) (Piergiovanni and Limbo, 2016). The growing emphasis on recycling plastic packaging has heightened quality demands. European Union regulations mandate recycling 50 % of plastic packaging by 2025 and 55 % by 2030 (European Commission, 2018). However, the sources of the recycled materials are very diverse, hence they can contain unknown additives which release is representing potential health risks.

A significant category of substances that can leach into drinking water are the reaction products of antioxidants, which are also known as stabilizers. These antioxidants are deliberately added to materials to prevent oxidative degradation during their production, processing, final use, and recycling. Ensuring the safety of used polymers is particularly crucial also in the food and pharmaceutical industries. One of the widely used additives is glycerol monostearate, commonly used as an antistatic agent as well as an emulsifier in food. However, this substance has been linked to potential health threat in recent years (Gao et al., 2019; Xia et al., 2021). Procedures for determining the extraction of additives from plastic used for both food or pharmaceutical purposes are standardized. For instance, European Standard EN 1420:2016 evaluates the influence of organic materials on the odor and flavor of water in piping systems (European Committee for Standardization, 2016) or European Standard EN 12,873–1:2014 examines the migration of materials into water intended for human consumption (European Committee for

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Standardization, 2014). These standards involve rinsing the test specimens with water for 60 min prior to testing, followed by immersion in water under static conditions at temperatures of 23 °C, 65 °C, or 85 °C for 24 h.

Using standardised tests, Kalweit et al. confirmed the migration and release of contaminants from plastic pipes affecting the water odour (Kalweit et al., 2019). Another study found that rubber particles can also be released from the seals between individual pipes, which is significant, because seals are not incorporated into standardized test methods (Diera et al., 2023). Zimmermann et al. found that more chemicals migrate from plastics than previously assumed, revealing shortcomings in current scientific and regulatory approaches (Zimmermann et al., 2021).

Beside the standardized measurements, there are researches which use non-standardized methods, involving static or dynamic extraction at various temperatures and durations in different media (Begley et al., 2008; Bertoldo and Ciardelli, 2004; Brocca et al., 2002; Lithner et al., 2012; Massey et al., 2007; Muhammad Ishaque et al., 2018; Skjevrak et al., 2003). Various contaminants were detected by these methods: non/volatile organic compounds (Muhammad Ishaque et al., 2018; Zhang et al., 2014), lead and phenolic compounds (Kowalska et al., 2011) or microplastics such as PVC, PA, PP, PE. Potential sources of microplastics could be polluted water bodies, but also mechanical abrasion of plastic equipment and pipes during water treatment and distribution (Lam et al., 2020). Importantly, Denberg et al. demonstrated that turbulent flows significantly affect the release rates of contaminants from polymer pipelines compared to laminar flows, where increased flow rates led to decreased release (Denberg et al., 2007).

To summarize the above mentioned information: the use of materials for water distribution is not strictly regulated and it relies primarily on water distributors (Craun and Calderon, 2001), who use materials apparently stable at static extraction conditions at constant temperatures.

However, to our knowledge, no studies have explored the influence of convective flows on contaminant release from polymer surfaces. Research has addressed energy and particle transfer under different flow modes and boundary layer conditions (Limm and Hollifield, 1995; Minarik et al., 2014; Minařík et al., 2011; Wan and Saito, 2018), but the direct link between flow characteristics and contaminant elution from plastics into liquids remains unexplored. Organized flows, such as Rayleigh-Bénard convection, can result from temperature gradients, a common environmental phenomenon (temperature gradient in soil (Aydin et al., 2015; Loupis et al., 2014; Singh and Sharma, 2017)). In case a certain vertical temperature gradient is exceeded, the heat conduction changes to the convective flow if no forcible fluid flow is induced.

This heat transfer mode is defined by the dimensionless Rayleigh number:

$$Ra = \frac{g\alpha\Delta T d^3}{\nu\kappa}, \tag{1}$$

where g is acceleration due to gravity, α is coefficient of thermal expansion of the fluid, ΔT is temperature gradient, d is thickness of a fluid layer, ν is kinematic viscosity and κ is thermal diffusivity of the fluid.

When the Rayleigh number exceeds a critical value, the heat transfer in the fluid changes from conduction to convection. The critical Rayleigh number for convective flow is about $\mathrm{Ra_c} = 1708$ (Akiyama et al., 1971; Pellew and Southwell, 1940; Volmar and Müller, 1997) which corresponds to very low temperature gradient of just about few tenths of degrees of Celsius for the fluid layer of units of centimeters thick. The formation of convective flow is controlled by a decrease in the density of the liquid on the warmer wall of the vessel. This hotter liquid rises upwards due to buoyancy, where it is cooled and flows downwards again, yielding in two or more counter-rotating liquid cylinders which allows very efficient energy transport (Colinet et al., 2005; Incropera, 2006; Rayleigh, 1916; du Puits and Willert, 2016).

This paper aims to investigate the influence of organized fluid flows, such as Rayleigh-Bénard convection caused by temperature gradients, on the release of contaminants from polymer surfaces in water. Specifically, the objectives of this research are to compare the contaminant release under commonly studied static and random fluid flow conditions, and convective flow which haven't been considered yet. We quantified the impact of these flow conditions on the release rate using surface tension measurements, UV–Vis spectroscopy, and elemental analysis. Our work seeks to provide a more comprehensive understanding of the factors affecting contaminant release in polymer-based water storage and distribution systems, ultimately leading to improved regulatory standards and practices.

Results and discussion

Analysis of polymeric foils

Two materials were tested. A high purity (pharmaceutical) PE used for storage of drugs and substances in healthcare and a foil made from recycled PE material were used. In the latter case, the material was not intended for food or potable water contact. Fig. 1 shows DSC curves for pharmaceutical and recycled PE-foils. The drop of the DSC curves of both materials between 40 °C and 50 °C may be associated with recrystallization and melting of the low-molecular fractions contained in the samples of the tested foils. Moreover, the foil from recycled PE has a wider endothermic peak with individual sub-peaks at 70, 117 and 121 °C, which possibly suggests multi-material composition, compared to the pharmaceutical foil. A sub-peak appearing around 70 °C could indicate the presence of glycerol monostearate (GMS), which is commonly used as an additive and acts as an antistatic agent (Butuc et al., 2017).

The DSC results (Fig. 1) correlate well with the changes in the surface activity (Fig. 3) of the respective aqueous extracts and suggest higher release of stabilizers from the recycled material. The characteristic peak, which is the proof of the presence of these substances in the compared foils, can be seen in the sample of recycled foil at about 190 °C and in the sample of pharmaceutical foil at 210 °C. These differences are caused by the dissimilarity in polymer composition of recycled and pharmaceutical foil.

The spectra of the inner sides of the foils (those exposed to the aqueous medium) and the GMS were measured (Fig. 2 and Fig. S1). Spectra of PE foils correlate with the DSC results confirming that both materials are polyethylene. In addition, the FTIR spectra of the recycled PE (Fig. 2b) shows peaks belonging to the additives. Correlations between the spectra of recycled PE and the spectrum of GMS (Fig. 2c) were

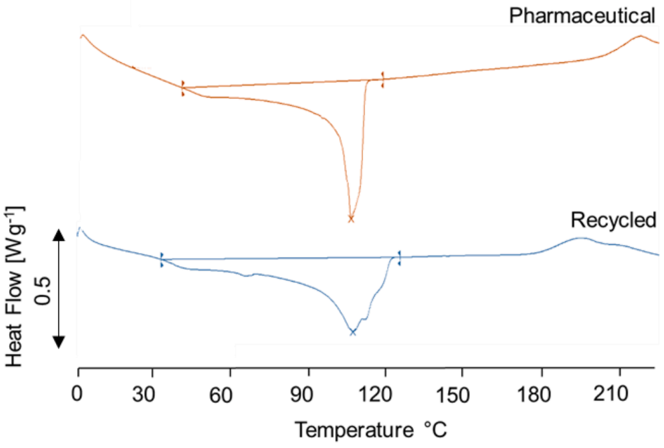

Fig. 1. DSC curves of pharmaceutical and recycled PE-foils.

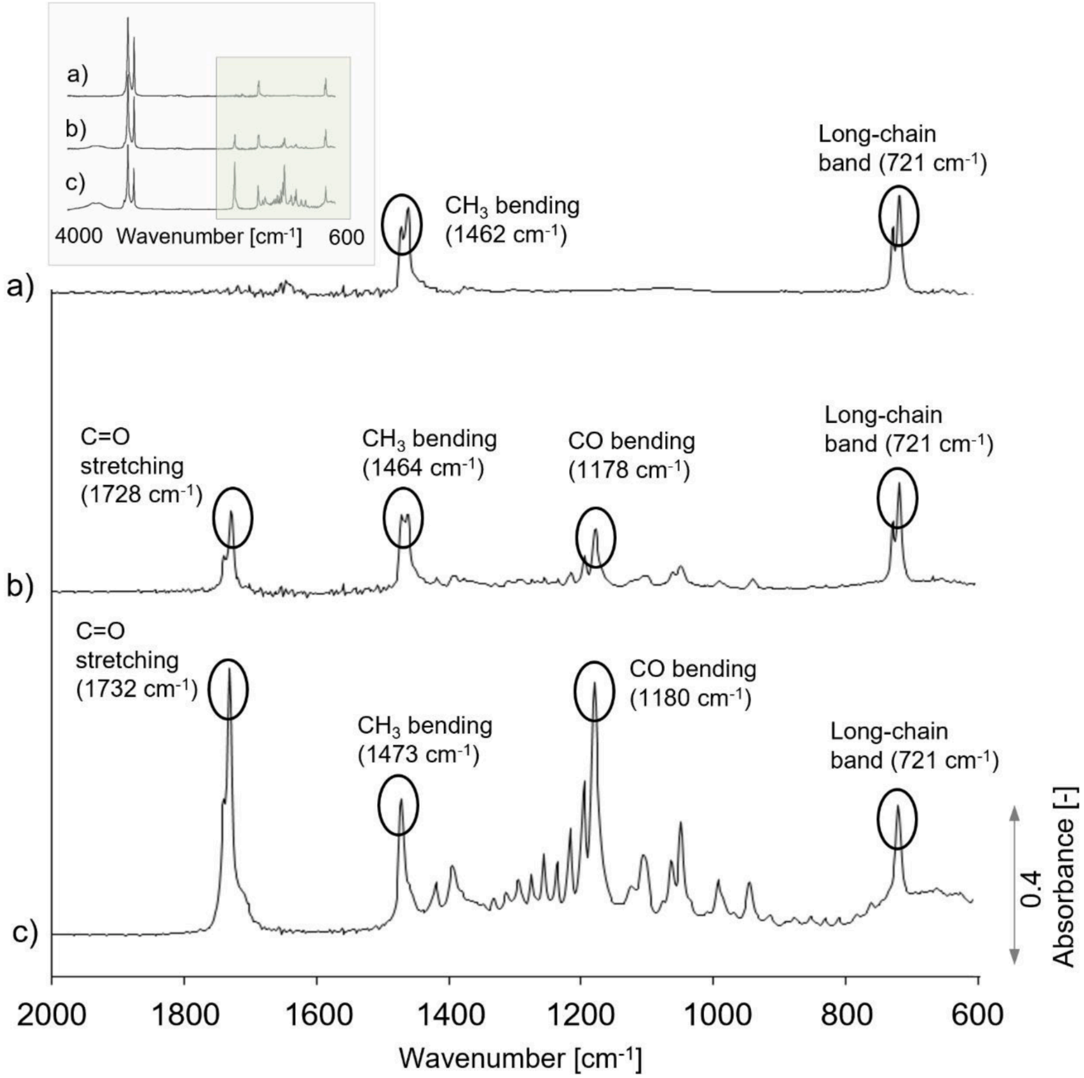

Fig. 2. FTIR spectra of a) inner side of pharmaceutical PE foil, b) inner side of recycled PE foil, c) glycerol monostearate powder.

noted. The typical infrared characteristic absorption bands observed in GMS and in recycled PE include: 1728–1732 cm^{-1}, stretching vibration of –C═O group present in ester link-ages; 1467 cm^{-1} bending vibration of the –CH_3 group; 1178–1180 cm^{-1}, bending vibration of –C–O band; 720–721 cm^{-1}, long chain band (Ng et al., 2018).

Scanning electron microscopy (SEM) images of the inner side of the PE foils (Fig. S2) clearly showed the presence of contaminants that are possibly released into the water. The elemental analysis performed by Energy-dispersive X-ray spectroscopy (EDX) during the SEM showing the presence of oxygen and nitrogen in the polyethylene foils are presented in the supporting information (Fig. S2). In addition, these measurements showed the presence of the alkali metals - sodium and potassium in the recycled polyethylene.

Surface tension analysis of water extract from polymer foils

Four types of experimental setup were designed, as depicted on the left side of Fig. 3a) – i)-iv). The mean water temperature was kept at 45 °C in all cases. The first case, Fig. 3a) – i), represents steady-state in PE foil pouch without mixing, corresponds to static conditions of heating. Fig. 3a) – ii) shows mixing of water by magnetic stirrer in a PE foil pouch for the simulation of random fluid flow over the polymer surface. Fig. 3a) – iii) PE pieces in water mixed vigorously by a magnetic stirrer to simulate a mixing flow around polymer surface. Finally, Fig. 3a) – iv) represents convective flow (mixing) of water in PE foil pouch caused by temperature gradient. The temperate conditions were controlled by the apparatus described in our previous study (Minařík et al., 2017). After 24 h, the leaching of contaminants from the PE surfaces into water was characterized using tensiometric measurements. The surface tension measurement method was chosen primarily for its high sensitivity (Kirk et al., 2019) and ability to detect surface-active species at very low concentrations. When the surface tension curves of aqueous extracts from PE-foils samples are compared, only a slight decrease in surface tension was observed in the pharmaceutically certified PE package in absence of a temperature gradient, Fig. 3b). Surprisingly, when the PE foil was exposed to a temperature gradient, significant decrease in the surface tension of water aliquots was noticed, which indicated the presence of contaminants. Since only a small temperature gradient (less than 1 °C) is required to exceed the critical Rayleigh number, it can be assumed that even with a gradient of just about $\Delta T = (T_H - T_C) = 2$ °C, convective flow can occur. In the case of recycled PE, a lower material purity resulted in a larger decrease in surface tension under all studied conditions (Fig. 3c). Also, in the case of the recycled PE, the highest water tension decrease was observed if the temperature gradient was applied. This implies that the convective flow could significantly affect the elution of contaminants from both compared PE materials if exposed to the temperature gradient, as reflected by the most significant drop of the surface tension, Fig. 3b) and c).

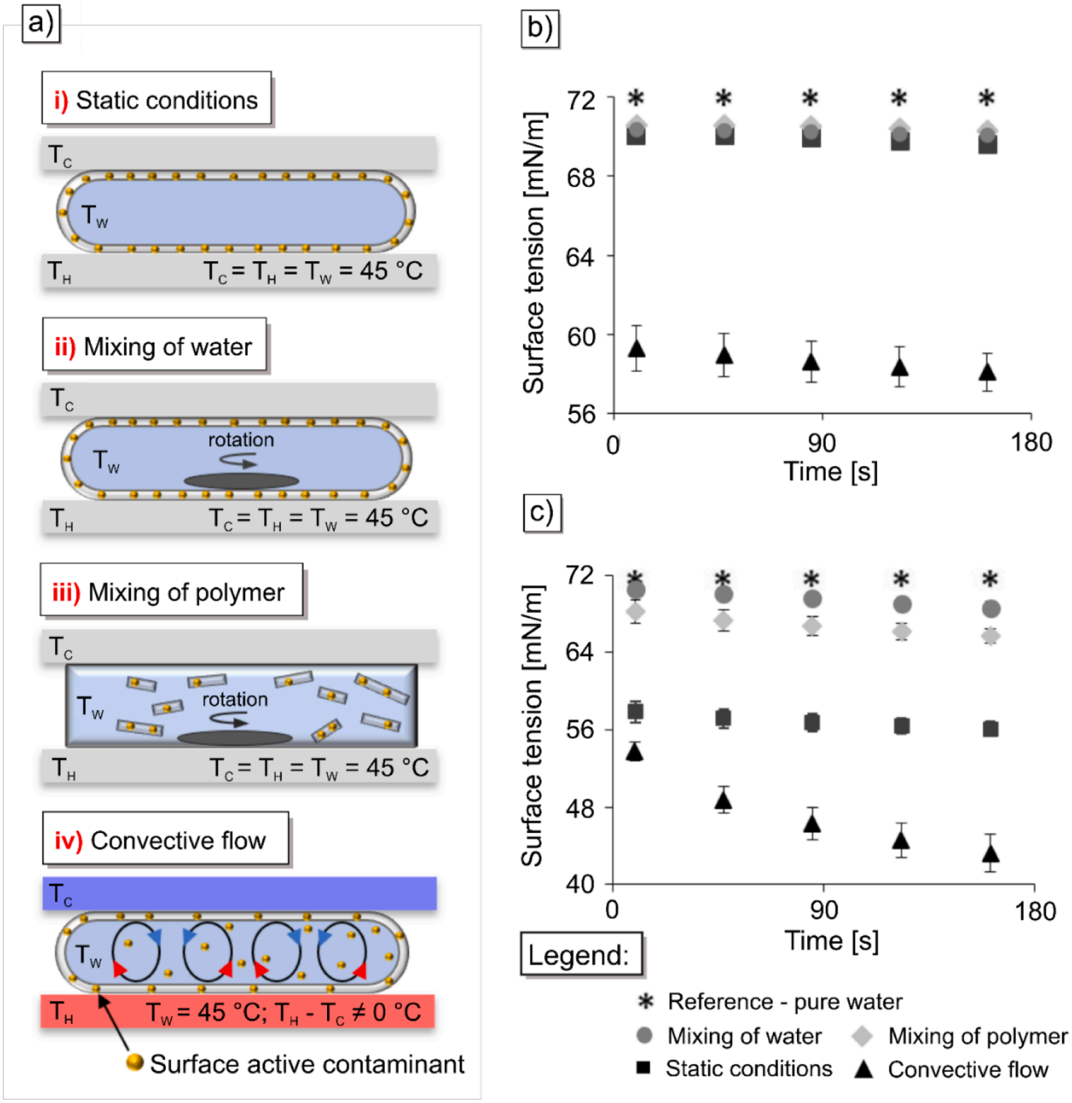

Fig. 3. a) schematic representation of experimental setups: i) water in the PE container without mixing; ii) mixing of water in a PE container; iii) mixing of PE pieces in water in a Petri dish; iv) water in the PE container with a temperature gradient. T_H –temperature of the heating plate; T_C – temperature of the cooling plate; T_W – mean water temperature. Yellow particles represent surface active contaminants in the polymer containers in the form of additives or impurities. b) surface tension of water extracts from PE with pharmaceutical grade, c) surface tension of water extracts from recycled PE.

For illustration, images of the extracts from polymer foils exposed to static conditions and convective flow (Fig. 4) at a mean temperature of 45 °C were taken from an experimental setup as illustrated in Fig. 3a i) and iv).

The images show the effect of the temperature gradient causing an organized flow apparently resulting in more efficient leaching of contaminants from the polymer surface, especially in case of recycled PE clearly showing opacity.

Analysis of water extracts from polymeric foils

The aqueous extracts from pharmaceutical as well as recycled PE-foils were characterized by UV–Vis spectrophotometry after the exposure to temperature gradients (case iv in Fig. 3a). The resulting spectra in the Fig. 5 proved clearly the presence of contaminants. To confirm this, elemental analysis was performed as well (Table S1, Supporting Information), revealing significantly increased amounts of carbon and nitrogen in both samples compared to the ultrapure water.

In the view of the DSC and FTIR analyses (Fig. 1 and 2), where the presence of GMS in recycled PE has been presumed, a solution of GMS in ultrapure water (5 mg/L) was compared using UV–Vis spectroscopy to the extracts obtained from PE foils exposed to the temperature gradients showing very similar maxima. Moreover, the baseline of the recycled extract is apparently shifted (Fig. 5) due to presence of the particles (Figs. 4 and 5). Hence the analyses further support our assumption that the GMS represents prevalent released additive. However, considering the results from the elemental analysis, which showed an increased nitrogen, sodium and potassium content, it is very likely that GMS is not the only contaminant released from recycled PE films under these conditions.

Observed differences in the release rate of surface-active species at the different flow conditions can be explained with the help of the boundary layers theory. Details can be found in the literature describing fluid mechanics, energy and mass transfer (Incropera, 2006; Schlichting and Gersten, 2017). Mathematical description of the problem was already presented by a number of authors (Benedict 1980; Cebeci and Cousteix, 2005; Oleinik and Samokhin, 1999; Schlichting and Gersten, 2017). Hence, the thickness of the boundary layer and the nature of the transport processes in this layer is determined by the flow velocity, species concentration and temperature gradients above the solid

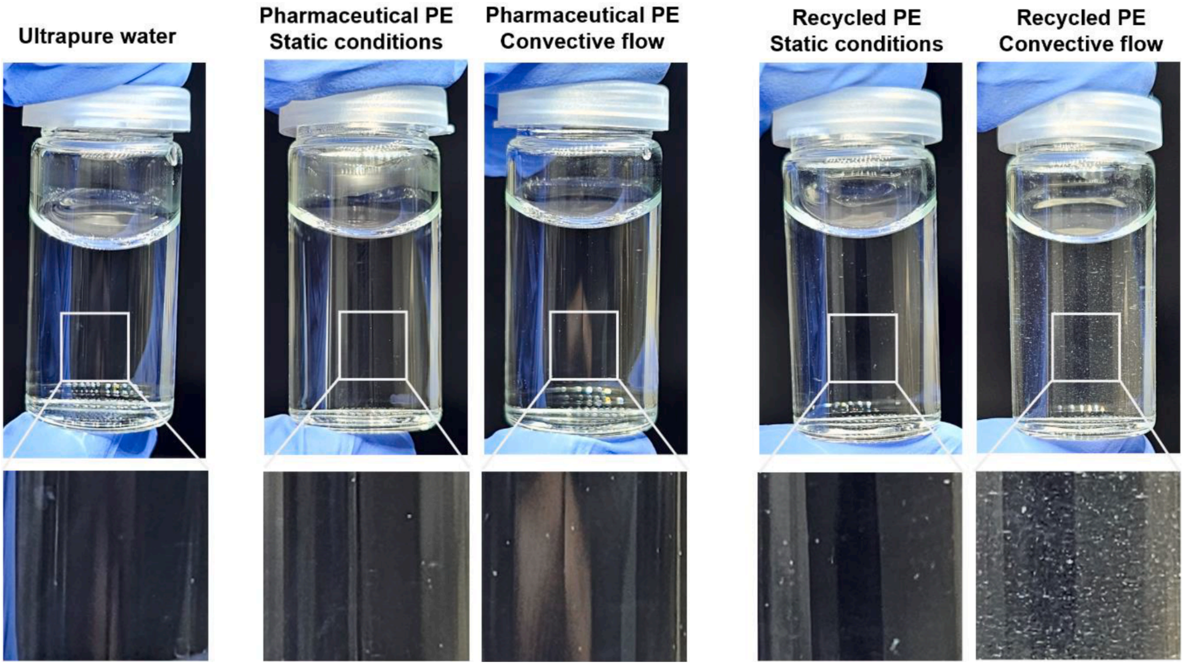

Fig. 4. photos of extracts from recycled and pharmaceutical grade PE in comparison to ultrapure water demonstrating the effect of the convective flow on contaminant release from the surface of recycled PE.

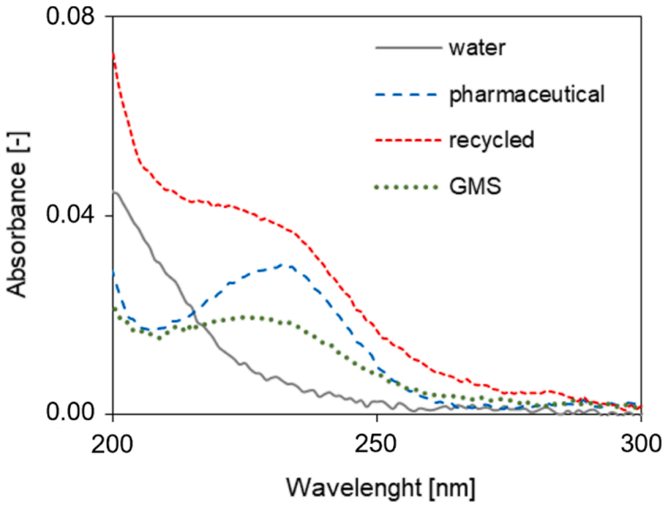

Fig. 5. UV–Vis spectroscopy of water and extracts from pharmaceutical and recycled PE foils exposed to the temperature gradient compared to pure GMS at 5 mg/L.

surface. The effect of surface topography, chemical composition of the released substances and their affinity to the liquid environment and the solid surface should be considered as well. From this perspective it is obvious that the applied experimental setups (Fig. 3a) will exhibit different extraction rate of the surface-active species into the aqueous media.

The different nature of the flows has a profound effect on convective mass and heat transfer rate (function *h(x)*, Fig. 6a). Various types of flows (laminar, transition, turbulent) reveal different convective mass and heat transfer rates. While experiment setups such as the static conditions (Fig. 6a) – i) and the mixing of water or polymer in container (Fig. 6a) – ii), iii) correspond to narrower intervals of the *h(x)* function (Fig. 6a), the convection flow (Fig. 3a) - iv) caused by temperature gradient ranges from laminar through transition to turbulent flow. This means that self-organised convective flow covers all areas with the highest rate of convective mass and heat transfer which resulted into the highest cumulative contamination release.

The flow velocity distribution determines the advective component of thermal energy or chemical species transfer within the boundary layer (black dashed line, Fig. 6b, c) as well. Similar to the laminar velocity boundary layer, the thermal and species boundary layer grows in the streamwise (increasing *x*) direction, whereas temperature and species concentration gradients in the fluid as well as the heat and mass transfer coefficients decrease with increasing *x* distance (transition from the I to II point for *h(x)* in the Fig. 6a) (Incropera, 2006; Schlichting and Gersten, 2017). Hence, under static conditions (I for *h(x)* in the Fig. 6a), no laminar flow nor temperature boundary layer preventing the release of surfactants into the media is formed above the surface. In such case, only the effect of particle concentration gradient above the polymer surface can be considered, as described in the film-layer theory (Denberg et al., 2007). This fact explains why under the static conditions, higher surfactant release was observed if compared to the simulated laminar and initial transition flow conditions (Fig. 3b) and c).

Turbulent mixing, characterized by typical large velocity gradients, also promotes large temperature and species concentration gradients adjacent to the solid surface as well as a corresponding increase in the heat and mass transfer coefficients across the transition region, going from II to III for *h(x)* in the Fig. 6a.

The effect of the organised flow in the form of Rayleigh-Bénard convective instability cannot be described with a single boundary layer type, as shown in the model described in the work of Q. Wang (Wang et al., 2018) as represented in the Fig. 6c). Both velocity and temperature boundary layers are formed above the solid surface. At the border of individual rotation liquid regions (Bénard cells) areas with a minimal viscous layer, sub-layer and higher shear stress are present. With growing turbulent flow rate (passing from III to IV for *h(x)* in the Fig. 6a), slight decrease in the heat and mass transfer rate occurs as a result of chaotic changes in flow velocity. The increase in the energy and mass transfer in the transition region (II – III for *h(x)*, Fig. 6a) is related to narrowing of the laminar viscous layer into the form of viscous sublayer (dashed white line in the Fig. 6b, c). Transport in this viscous sublayer is dominated by diffusion mechanism. Above this viscous sublayer is a buffer layer (white line in the Fig. 6b, c) in which diffusion and turbulent mixing are comparable. In the turbulent zone transport is dominated by turbulent mixing. With the narrowing of the viscous layer into the form of sublayer (the region below the dashed white line, II – IV, Fig. 6) the surface shear stress τ_s increases, while with growing laminar viscous layer (I – II, Fig. 6a) does the surface shear stress decrease. The action of the shear stress cannot be neglected in the case of polymer surfaces as it can play significant role in dragging polymer fragments and surfactants from the polymer surface into the liquid medium.

These phenomena, occurring in the boundary regions of the Bénard cells during convective flow contribute substantially to the accelerated release of surfactants into solution. It could be concluded that the release of surfactants into water (Fig. 3b) and c) is caused by the combination of

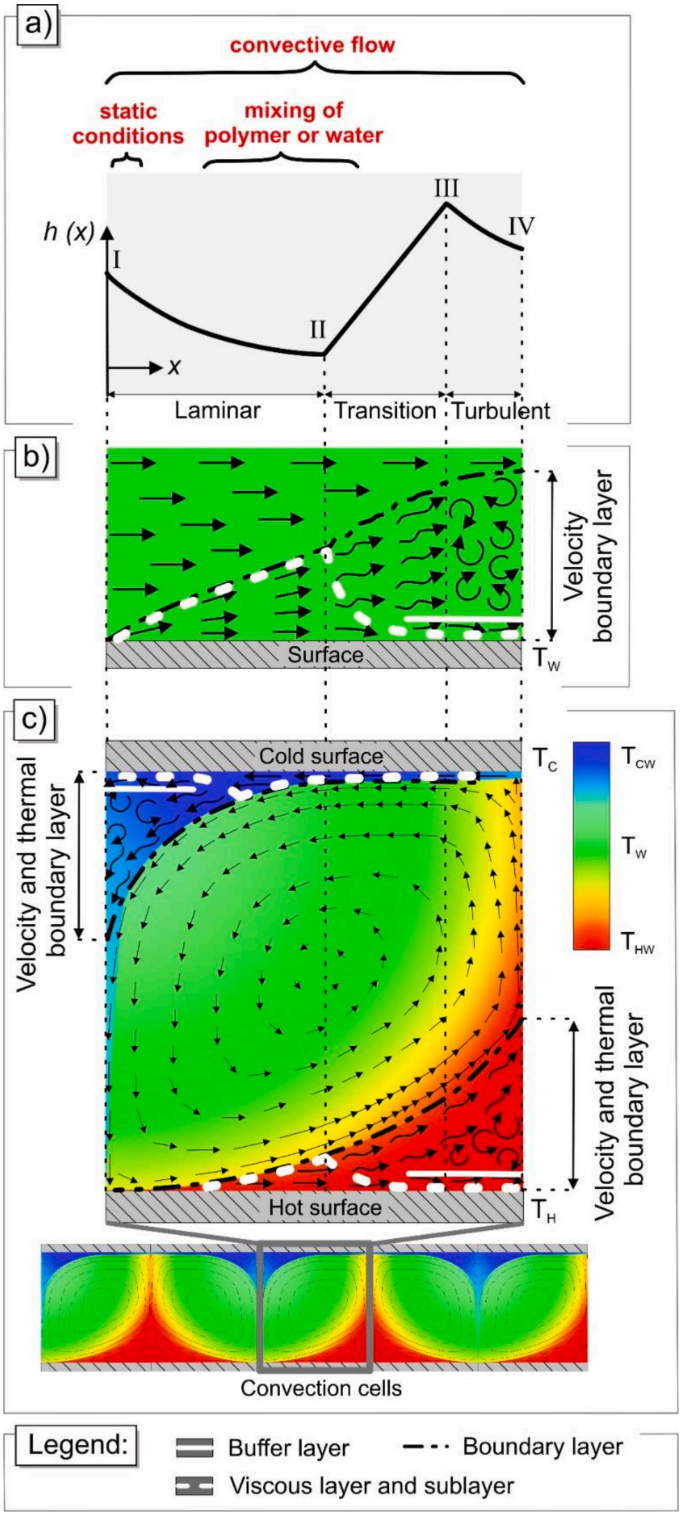

Fig. 6. Schematic representation and comparison of laminar and turbulent flow vs convective flow. a) local heat transfer coefficient (h(x)) for flow over isothermal flat plate. b) scheme of velocity boundary layer in laminar and turbulent flow. c) scheme of velocity and thermal boundary layer in convective flow. T_C – temperature of cold plate, T_H – temperature of hot plate, T_W – mean temperature of fluid (water), T_{CW} – temperature of water at the cold plate, T_{HW} – temperature of fluid (water) at the hot plate.

intense energy and mass transfer and shear stress acting on the polymer surface with minimal viscous layer (point I, Fig. 6a) and its narrowing to the form of viscous sub-layer (interval from II to IV, Fig. 6a).

Conclusion

The study has investigated the effect of organized fluid flow, generated by a temperature gradient, on the release of contaminants from PE surfaces in water. It has been found that the self-organized flow in the form of Rayleigh-Bénard convective instability, which combines intense energy and mass transfer with a shear stress acting on the polymer surface, has a significant influence on the release of additives from PE surface. The presence of contaminants was demonstrated by decrease in surface tension of 14 mN/m for the high purity PE and 29 mN/m for the recycled PE in comparison to the pure water. Based on DSC, FTIR and UV measurements, the presence of GMS in the extracts has been confirmed. However, elevated nitrogen, sodium and potassium values obtained by elemental analysis indicate the presence of other unidentified contaminants, especially in case of the recycled PE.

We are convinced, that the presented study has a direct implication for water storage in plastic containers and water transport in plastic pipes which are due to their underground placement commonly exposed to temperature gradients, i.e. convective flows. The enhanced release of the contaminants from PE surfaces under convective flow conditions indicates that the dynamic environmental conditions often encountered in real-world applications, such as varying temperatures in storage and transport systems, can lead to higher contamination levels than those predicted by commonly applied static testing methods. Our findings emphasise the need for more comprehensive testing protocols that simulate a real operational conditions for plastic water storage and distribution systems to ensure water safety and quality.

Moreover, the study shows the importance of considering the material composition and quality of polymers used in water systems. For instance, commonly used recycled PE showed in comparison to the pharmaceutical-grade PE considerably higher contaminant release under all tested conditions, especially under temperature gradients. This underlines the necessity for stringent quality control and material selection criteria for plastics intended for water contact applications.

The impact of convective flow on contaminant release from polymer surfaces warrants further investigation and consideration in regulatory standards. By enhancing understanding of these processes, water quality and public health could be better safeguarded, ensuring that both water storage and distribution systems are designed and maintained to minimize contaminant release under varying environmental conditions.

Materials and methods

Materials

As samples, pharmaceutical grade polyethylene (PE) foil and foil containing recycled material were used. Both materials - recycled and pharmaceutically pure polyethylene were used for the experiments as delivered by the manufacturers. The recycled polyethylene was in the form of a roll and the pharmaceutically pure polyethylene was supplied in the form of individual bags. In both cases the required piece of foil was cut of the delivered material and flushed before experiment with 100 mL ultrapure water to remove residual impurities or dust. Ultrapure water with the resistance of 18.2 MΩ.cm (Direct-Q® 3UV water purification system) was used as a solvent. Glycerol monostearate solution (Thermo Fisher Scientific) was prepared by mixing with ultrapure water at concentration at 5 mg/L and stirring on a magnetic stirrer at 80 °C for 2 h. All used glassware was cleaned to remove possible surfactants and contaminants by soaking it for 15 min in chromsulphuric acid, sulphuric acid and hydrochloric acid, rinsed with hydrogen peroxide and ultrapure water and finally sterilized at 125 °C for 20 min in an oven.

Sample preparation

Rectangular shape pouches (when filled with the medium) were made out of the PE foil with the aid of a standard pulse welder. The ultrapure water was always dispensed into the foils using a micropipette with a sterile tip. The samples were prepared according to experimental setups (Fig. 3a) as follows. i) static conditions: The PE pouch (10 × 8.5 × 0.7 cm) was filled with the water, welded and exposed to the constant temperature 45 °C. ii) mixing of water: The magnetic stirrer bar (20 × 6 mm) was put into the PE pouch (10 × 8.5 × 0.7 cm) filled with the water. Then the foil was welded preventing presence of air bubbles and exposed to mixing at 200 rpm and constant temperature of 45 °C. iii) mixing of polymer: PE foil (10 × 8.5 cm) was cut with scissors cleaned with water, acetone, water and ethanol into the pieces of roughly 0.5 × 0.5 cm and exposed to the action of mechanical stirring, in a water filled glass container (60 mL) at constant temperature of 45 °C. The size of the magnetic stirrer bar was 20 × 6 mm and the mixing velocity was set to 200 rpm. iv) convective flow: PE pouch (10 × 8.5 × 0.7 cm) was filled with water, welded and exposed to temperature gradient with help of special equipment described in the next chapter. Temperature of hot plate was set to $T_H = 46$ °C and the cold plate temperature was set to $T_C = 44$ °C. The experiments in each setup were repeated five times. The temperature of 45 °C was chosen because it does not exceed the temperatures used in standardised methods and is also more likely to be encountered in environmental conditions (warm substrate due to sunlight, etc.)

Equipment for the generation of temperature gradients

For the generation of defined temperature gradients a special equipment was developed at the Department of Physics and Materials Engineering, Tomas Bata University in Zlín (Minařík et al., 2017). The setup consists of a pair of horizontal plates that can be either heated or cooled. The construction of the plates allows for precise control of their absolute temperature with the aid of a feedback-loop temperature regulator and uniform heat distribution across the plate's surface. The samples were placed in intimate contact with the heating and cooling plates to avoid any obstruction to the heat transfer. The temperature was controlled by temperature sensors and recorded on PC throughout the sample exposure to a temperature gradient for 24 h.

Surface tension

The surface tension time dependence of the media prior to and after the exposure was measured using a Wilhelmy plate method with a Krűss K12 Processor Tensiometer (KRŰSS GmbH). Four mL of the extracts from each of the experimental setups A-D were taken by micropipette and put into 4 mL Petri dish. The measurements were performed at 24 °C for the period of 180 s. Five extracts were analyzed for each setup from which arithmetic mean and standard deviations were determined.

Differential scanning calorimetry, FTIR-ATR, UV–VIS spectrophotometry, scanning electron microscopy and elemental analysis

The differential scanning calorimeter (DSC) DSC1 Star System (Mettler Toledo) was used for thermal analysis. Weight of PE samples for DSC measurement was between 5 and 7 mg. The measurements were carried out under the nitrogen atmosphere at the flow rate of 40 ml/min and the heating rate of 10 °C/min.

The inner sides of the recycled PE and high purity PE and the powder of glycerol monostearate were analyzed using the FTIR spectrometer Nicolet iS5 (Thermo Fisher Scientific). The device was operated in attenuated total reflection (ATR) mode with a germanium crystal. During the measurement, 32 scans were recorded at the resolution of 4 cm^{-1} in the range of 600 to 4000 cm^{-1} with an air background.

The UV–VIS spectra of the aqueous PE-foil extracts were recorded using a CARY 300 Conc UV–Visible Spectrophotometer (Varian) in a standard rectangular quartz glass cuvettes with the pathlength of 10 mm. Ultrapure water with the resistance of 18.2 MΩ.cm was used for comparison. The measured absorbance range was from 190 to 800 nm.

The inner sides of the polyethylene foils were observed by scanning electron microscopy (SEM) with Phenom XL G2 (Thermo Fisher Scientific). Prior to SEM, the samples were sputter coated with a mixture of gold and palladium in a Quorum SCSC7620 Mini sputter coater (Quorum Technologies) (argon gas, 30 s sputter time) to increase sample stability for elemental analysis. Samples were analyzed at an acceleration voltage of 15 kV with backscattered and secondary electron detectors (50 % mix) with mapping mode. SEM elemental analysis was performed by Energy-dispersive X-ray spectroscopy (EDX) on each sample at a minimum of 6 points (3 points that contained a visible contaminant and 3 points that appeared to be pure polymer matrix) from which statistical analysis was performed and mean deviations were determined.

Elemental analysis of PE-foil extracts was performed with a carbon and nitrogen analyzer TOC-L (Shimadzu). From each sample the volume of 20 mL was taken and centrifuged (11 000 rpm/15 min) and the supernatants were analyzed. Three parallel measurements for each sample were performed from which arithmetic mean and standard deviations were determined.

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CRediT authorship contribution statement

Markéta Kadlečková: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis. **Karolína Kocourková:** Formal analysis. **Filip Mikulka:** Visualization, Writing – review & editing. **Petr Smolka:** Writing – review & editing, Writing – original draft, Investigation. **Aleš Mráček:** Writing – review & editing, Formal analysis. **Tomáš Sedláček:** Investigation, Formal analysis. **Lenka Musilová:** Formal analysis. **Martin Humeník:** Writing – review & editing, Writing – original draft, Visualization, Investigation. **Antonín Minařík:** Writing – review & editing, Writing – original draft, Visualization, Supervision, Project administration, Methodology, Investigation, Formal analysis, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.wroa.2024.100248.

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