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Selected unit processes in microplastics removal from water and wastewater

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ABSTRACT

The objective of the research study was to evaluate removal efficiencies of polyester fibers, polyvinyl chloride (PVC) granules, acetonitrile-polyester fibers, polypropylene fibers, foil fragments, and personal care products fragments during sedimentation and coagulation under laboratory conditions. The effect of oil and cellulose fibers presence was also evaluated. Most of the microplastics used during the studies did not settle down under the gravity force. Only a small part of polyester fibers (about 6%) settled down during 2 h of the experiment. Oil presence in samples had an important effect on the behavior of all kinds of microplastics particles. Oil spots have trapped microplastics fibers, powder, and fragments. The presence of cellulose fibers in the water had no significant effect on microplastics removal. The slightly higher removal efficiency was observed when the cellulose fibers were soaked with oil. The coagulation process was effective in the removal of two kinds of microplastic particles: polyester fibers (84%–89% removal efficiency) and PVC powder (20% removal efficiency). The main parameter which affected coagulation was the presence of oils. In the presence of oil huge part of microplastics was trapped in the spots and did not undergo coagulation.

Keywords: Microplastic; Sedimentation; Coagulation; Water treatment; Wastewater treatment; Cellulose fibers; Oils

1. Introduction

Microplastics are usually defined as small plastic particles of various shapes and sizes in the range of 0.001–5 mm. Particles smaller than 0.001 mm are called nano-plastics, whereas larger than 5 mm mesoplastics [1]. By some authors, as the upper end of the microplastic class, 1 mm is indicated [2]. Often, only microplastic particles of sizes 0.330–5 mm are analyzed, which is connected with the fact that samples are taken with plankton nets with a mesh size of 0.330 mm [1]. Material under 0.330 mm can be collected using such equipment as, for example, Niskin sampler [3].

Depending on the origin, microplastics can be grouped into primary (particles originally manufactured to be that size) and secondary (resulted from the decomposition of larger particles) [1]. Primary microplastics include mainly scrubbers, plastic powders, micro-beads for cosmetics. They are usually spherical or cylindrical in the shapes. Secondary microplastic is generated during the use or degradation of various products made of synthetics, and it can include, for example, fibers or foil fragments [1].

In the market there are 6 main classes of microplastics: polyethylene (PE, low and high density, density 0.92–0.97 g/mL), polypropylene (PP, density 0.90–0.91 g/mL), polyvinyl chloride (PVC, density 1.3–1.6 g/mL), polystyrene (PS, density 1.0–1.1 g/mL), polyurethane (PUR, density 1.25 g/mL) and polyethylene terephthalate (PET, density 1.3–1.4 g/mL) [1,4,5]. A density less than 1 means that the

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microplastic particle tends to float in water, however of course also other parameters affect flotation/sedimentation processes, for example, water flow velocity, presence of sediment particles and colloids, etc.

In wastewater, the dominant microplastic fractions are microfibers from the textiles. Common synthetic fibers are polyester/PET (e.g., Terylene™, Dacron™), acrylic fibers, polyamide (nylon), acetate and polyparaphenylene terephthalamide (PPT, e.g., Kevlar™) [6]. These fibers are released to wastewater during washing. It was estimated that annually total microplastic fibers releasing from the washing of synthetic clothing in Europe are 18,430–46,175 tonnes, which is equal to about 100 and 600 quadrillion fibers [6]. Experts point out that fibers have a greater propensity to be retained within organisms compared to sub-spherical forms [6]. Because of this research studies on their removal from wastewater are of great interest.

Also, cylindrical shape granules attract attention. Especially pre-plastics should be analyzed. They are small pellets, powders, and flakes which are of very small sizes. This makes it difficult to remove from aqueous solutions [6]. Such pellets have great potential for ingestion by living organisms and adsorption of hydrophobic micropollutants such as, for example, polycyclic aromatic hydrocarbons or polychlorinated biphenyls [6].

According to Hann et al. [6] pre-production of plastics and washing of clothing are among the three main sources of microplastics in surface water. The third biggest source is automotive tires. In raw wastewater concentration of microplastics may vary a lot and it is up to 900,000 particles in 1 m^3, however, the average concentration usually does not exceed several hundred particles in 1 m^3 [7,8]. In effluents concentration of microplastic particles is usually not higher than several dozen per one cubic meter; in some cases, it can however reach even several thousand [7–9]. In highly polluted rivers concentration of microplastic particles can reach even more than 1 million particles per 1 m^3, because of this not always effluents contribute to an increase in microplastics concentration in river water [10]. However, typically effluents significantly affect the level of microplastics in the receiving surface water, for example, Estahbanati and Fahrenfeld [11] have observed that downstream wastewater treatment plant (WWTP) microplastics concentration in the river has increased by 36%. Also Magnusson and Norén [12], Lechner et al. [13] and McCormick et al. [14] have stated that discharge of WWTPs effluent increased microplastics concentration in surface water by 77%, 89% and 64%, respectively. The results of Nocoń et al. [15] have indicated that WWTPs have an important effect on the concentration of microplastics in the river, even in highly urbanized areas. As it was stated by McCormick et al. [14] the dominant microplastics types in rivers, resulting from WWTPs discharges were pellets, fibers, and fragments. Taking into consideration types of polymers the ones the most frequently found in freshwaters are polyethylene = polypropylene > polystyrene > polyvinyl chloride > polyethylene terephthalate [16]. According to Everaert et al. [17], this pattern probably reflects the global plastics demand.

According to various sources during wastewater treatment up to 90%–99% of microplastic present in wastewater is removed [6,18,19]. As effective technologies for microplastic particles removal from wastewater sedimentation and tertiary treatment are indicated [5].

Talvitie et al. [20] have observed that most (>97%) of microplastics in wastewater treatment plants is removed during mechanical treatment (mainly sedimentation). Further biological treatment with activated sludge allowed for removal of about 7%–20% of remaining microplastic particles. Detailed studies that were done by Talvitie et al. [7] have shown that during preliminary (mechanical) treatment mainly larger microplastics particles (of sizes >0.330 mm) are removed, whereas the smaller ones are not removed so effectively. As a result, 0.02–0.1 mm fraction was dominant in the effluent. During wastewater treatment fibers are effectively removed, but micro-granules from personal care products are usually still present in effluent and are the main fractions of microplastics discharged to the receivers [7]. It was assumed that during preliminary sedimentation microplastic microfibers are "trapped" by toilet paper and plant fibers, which effectively removes them from the aqueous phase [21]. Microplastic particles can be also cumulated in foam collected on the surface of preliminary sedimentation tanks [22]. During mechanical treatment of wastewater particles of microplastics of 0.1 mm and larger could be also effectively removed by sieving on micro-sieves; fine bar screen also can remove some microplastic particles in an enabling environment [23,24].

Some authors have indicated that tertiary treatment can be effective in the removal of remaining microplastics in wastewater, for example, Talvitie et al. [20] have indicated that membrane reactors and rapid sand filtration could be effective for these purposes. It was stated that the filtration of wastewater is especially effective in the removal of microplastic particles of 0.300 mm and larger (according to other sources larger than 0.180 mm) [9,20,22]. It was also effective in the removal of fibers [25]. It is expected by some authors that microplastics can be removed from wastewater with oils and fats in grit chambers [26,27]. Research studies undoubtedly confirmed that membrane processes practically completely remove microplastics from wastewater. At present on market, only one commercial technology (VeSave) dedicated microplastic removal is available [28].

Also, water treatment plants are considered to be effective in microplastics removal from water [5]. It was confirmed that the conventional water treatment process (coagulation, flocculation, sedimentation, and filtration) is effective in the removal of microplastic particles of 0.001 mm and larger [5]. There is a lack of data on the effectiveness of individual processes on microplastics removal – enmeshment during floc aggregation within the coagulation process, flotation, sedimentation, filtration, and adsorption are identified as main removal processes [4]. It was however stated that coagulation and sedimentation are effective in the removal of fibers from treated water. Filters filled with granulated activated carbon removed microplastics in sizes in the range of 0.001–0.005 mm [29].

If membrane processes are involved, also particles of 0.001 μm can be removed, whereas ultrafiltration the particles of 0.01 μm or higher [5]. But these smaller particles should be indeed classified as nanoplastics. Despite the high efficiency of water treatment plants in microplastic removal from water, these pollutants can be present in drinking water [5]. They can be removed from plastic pipes in water distribution systems because of their corrosion or degradation [5]. Reported concentrations of microplastics concentrations in drinking water are in the range of 0 to over 10^4 particles/m^3 [5]. According to the report on microplastics presence in tap water [30] it was estimated that microplastic particles could be potentially present in 72% tap water samples in Europe.

Generally, sedimentation, trapping in oils, and coagulation is identified as the processes applicable for the removal of microplastics, however little detailed studies on the effectiveness of these processes under laboratory conditions have been done. One of the research studies concerning the coagulation effect on microplastics removal has been conducted by Ma et al. [31]. They have stated that contrary to the data presented in the literature after coagulation low microplastic removal efficiency has been observed. Coagulation with aluminum-based salts was more effective in the removal of microplastic particles than Fe-based ones. In the study by Ma et al. [31] only polyethylene microplastic behavior during coagulation has been examined. What was interesting the authors have observed that the sizes of PE particles were the smallest the higher removal efficiency was observed. In the whole experiment, the total efficiency of microplastic removal was, however not higher than 40%. If anionic coagulant aid (polyacrylamide) was added to the water removal efficiency of microplastics increased. It was because of the fact that PE has a positive charge and it is well attracted by negatively charged polyacrylamide. More detailed studies on the Fe-based salts coagulation efficiency on microplastic removal were conducted also by the team of Ma et al. [32]. The authors have observed that during conventional coagulation less than 15% of PE particles have been removed. This process was not significantly affected by physicochemical properties of water, for example, pH, but similarly, as during the previously mentioned experiment it was significantly affected by using anionic polyacrylamide. Detailed studies conducted by Lapointe et al. [33] on coagulation and flocculation of water containing polyethylene microspheres, polystyrene microspheres, and polyester fibers confirmed that coagulation efficiency in microplastics removal is affected by the charge of reagents used for coagulation, as well as by characteristics of the surface of these micropollutants. For example, weathered plastic debris of changes in surface chemistry was removed more effectively than not weathered ones. To date, no detailed studies on coagulation effectiveness in the presence of oil and cellulose fibers have been conducted.

The present study is aimed to evaluate removal efficiencies of six microplastics classes (polyester fibers, PVC granules, acetonitrile-polyester fibers, polypropylene fibers, foil fragments, personal care products fragments) during sedimentation and coagulation under laboratory conditions. The effect of oil and cellulose fibers presence was also evaluated.

2. Materials and methods

2.1. Microplastic particles used during the study and their analysis

Microplastic for the studies was obtained mainly by destroying fabrics or other plastic materials, with the exception of PVC powder which was a commercial product.

Polyester and acrylonitrile plus polyester (77% + 23%) fabrics were obtained by cutting them. Catted fibers were then separated into individual microfibers. The fibers were about 1 mm or smaller. Also, polypropylene fibers, foil fragments, and personal care product fragments were obtained by-products being cut up. In the case of polypropylene fibers by cutting plastic washer, whereas foil fragment by cutting plastic bags and personal product fragments by cutting plastic elements of diapers. Polypropylene fibers were about 1 mm in size, foil, and personal care products fragments about 1–2 mm. PVC was bought from the manufacturer as the powder used for the production of plastics.

The images of the microplastics used in the study are presented in Figs. 1–6.

The fibers (Fig. 1 – 100% polyester, Fig. 3 – 77% acrylonitrile and 23% polyester, Fig. 4 polypropylene fibers Fig. 6), powder (Fig. 2 PVC), or fragments (Fig. 5 – foil fragments) were spiked to tap water (clear water) or turbid water. The turbidity of water has been obtained by spiking to water kaolin grey clay, as a result, colloids present in the sample were only of mineral origin. The amount of clay was to obtain turbidity in the range of 30 NTU. The number of microfibers was estimated to obtain an initial concentration in 1 L of the sample at a level of about 100 particles. In the case of foil fragments and fragments of personal care products, the initial concentration per 1 L was at average 25–30 particles, which was enough to observe the processes which take place during sedimentation/coagulation. Because PVC powder was very fine obtained concentrations of its granules in 1 L were above 500 and less than 1,000 particles per 1 L. It was difficult to maintain the identical concentration of microplastics in each series and repetition, and because of this the particles

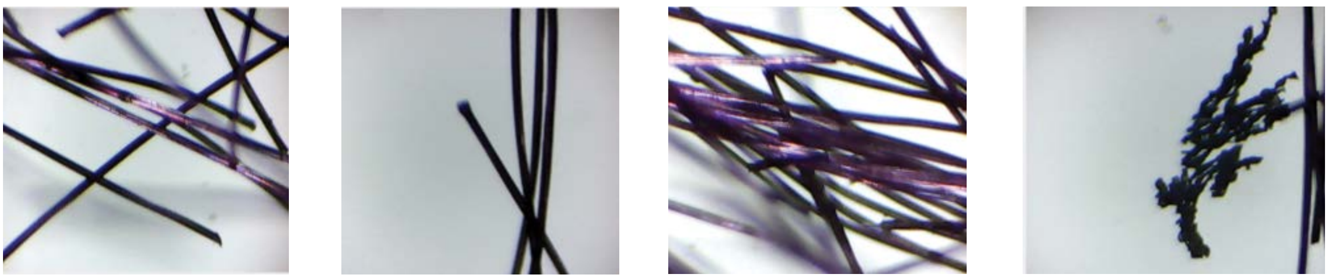

Fig. 1. Fibers used during the experiment (100% polyester), magnification 40×.

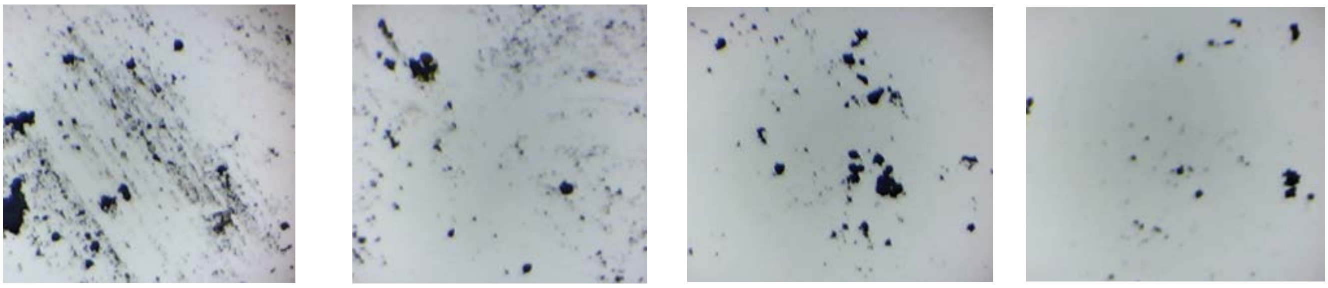

Fig. 2. PVC granules were used during the study (100% PVC), magnification 40×.

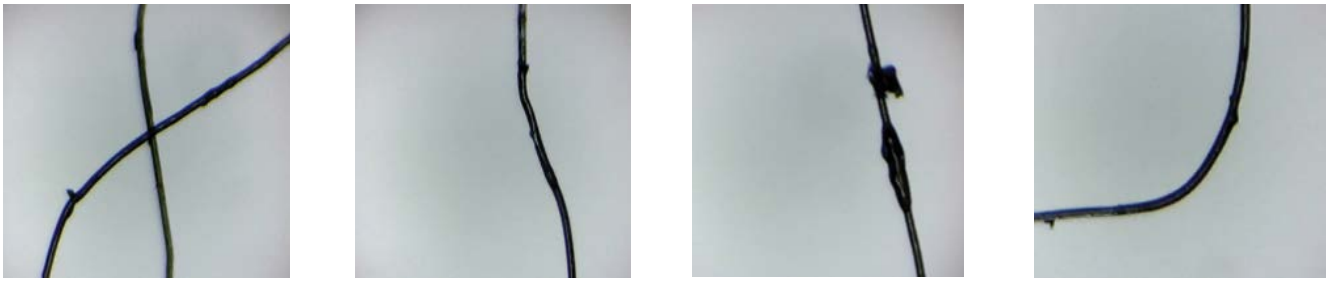

Fig. 3. Fibers used during the experiment (77% acrylonitrile and 23% polyester), magnification 40×.

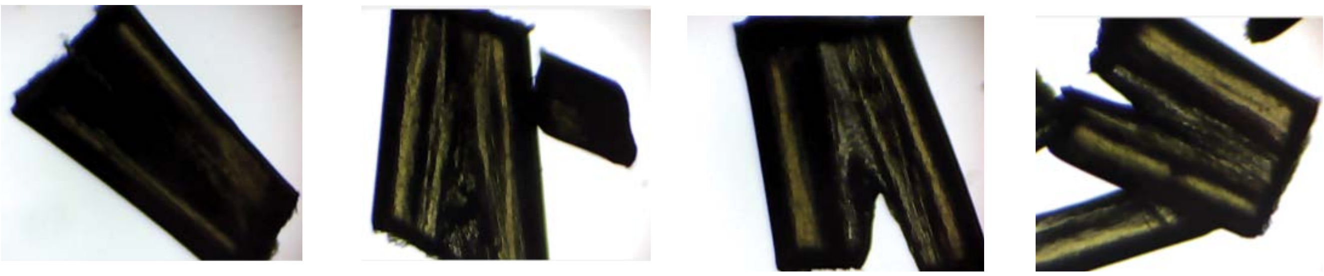

Fig. 4. Polypropylene fibers (100% PP), magnification 40×.

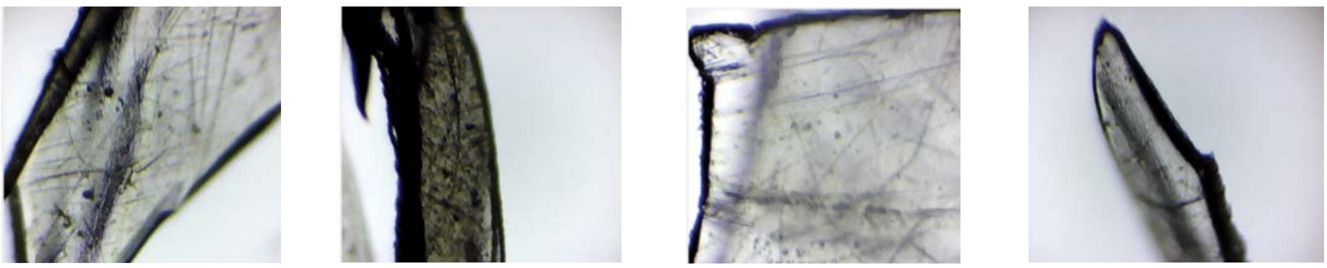

Fig. 5. Foil fragments were used during the experiments, magnification 40×.

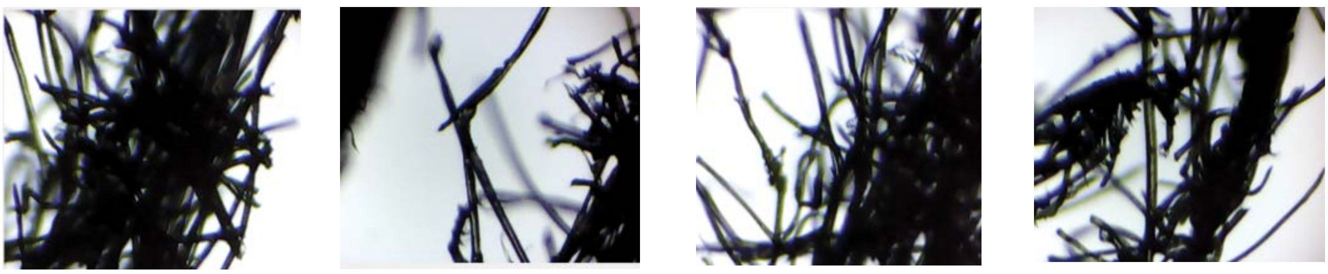

Fig. 6. Personal care products fragments used during the study, magnification 40×.

were each time counted and the result is the average of the 3 repetitions.

Particles were visualized under the Delta Optical SZH-650 B/T microscope and the exact number of particles in the precise volume of 1 mL of the sample. Sedgewick-Rafter's chamber was used to count microplastics particles. The samples were taken directly from the water body and counted under an optical microscope (magnification 40–100×). In most cases, magnification of 40 was enough to count all samples, in some cases of PVC granules it was necessary to use higher magnification. There was not necessary to identify a kind of microplastics because this parameter was prearranged. This assumption simplified the experiment. Microplastics particles were counted directly. Because we have used single kinds of microplastics of known origin and well visible under the optical microscope there was not necessary to digest samples with strong oxidants. The standard uncertainty of the results in all cases did not exceed 30%.

2.2. Sedimentation tests

The whole experiment has been done for the six categories of microplastics (Figs. 1–6). In the case of each fiber category the following experiments have been conducted:

- sedimentation in clear water,
- sedimentation in clear water with cellulose fibers (toilet paper),
- sedimentation in clear water with a small admixture of oil,
- sedimentation in clear water with greater oil admixture,
- sedimentation in turbid water (with clay amendment),
- sedimentation in turbid water with cellulose fibers (toilet paper),
- sedimentation in turbid water with little oil amendment,
- sedimentation in turbid water with a lot of oil amendment.

At the beginning of the experiment microplastic particles were spiked to the standing water, mixed and then left for 2 h. After that time observations of microplastic particles were conducted. The removal of microplastic particles was calculated as the difference between the initial and final concentrations of microplastics at the water table. All experiments were conducted in triplicates, in 1 L glass beakers.

2.3. Coagulation tests

Coagulation tests were also conducted in 1 L glass beakers. The beakers were placed at magnetic stirrers. The tests were done for turbid water, turbid water with cellulose fibers, and turbid water with oil. The pH of samples was in the range of 6.97–6.99. During the tests, $FeSO_4$ as a coagulating agent was added. The dose of coagulant was previously experimentally determined to remove turbidity from water. No polymers (coagulation aids were added) during the experiment. Mixing speed was maintained at 300 rpm for 2 min, and then 100 rpm for 20 min. with subsequent 1 h of sedimentation. Microplastics particles were counted before and after the coagulation process. The removal of microplastic particles was calculated as the difference between the initial and final concentrations of microplastics at the water table. All experiments were conducted in triplicates.

3. Results and discussion

3.1. Sedimentation process in the presence, or without presence, of oils and cellulose fibers

The results of the experiments on the sedimentation process have been collected in Tables 1 and 2. In Table 1 the results for clear water, without any colloidal substances are presented. After 2 h of sedimentation only in the case of one type of microplastics, sedimentation was observed – at average about 6% of polyester fibers have settled down

Table 1
The behavior of various microplastic particles during sedimentation under different conditions in a laboratory experiment – sedimentation in clearwater

Type of microplastics	Average concentration of microplastics, particles/L; expressed as initial/final that settled down					
	Sedimentation for 2 h in clearwater	Sedimentation for 2 h in clear water with oil amendment		Sedimentation for 2 h in clear water with cellulose fibers	Sedimentation for 2 h in clear water with oil amendment and cellulose fibers	
		Small oil amendment	Larger oil amendment		Small oil amendment	Larger oil amendment
Polyester fibers	68/4	99 (26)*/7	143/(94)*/9	126/11	78 (24)*/4	104 (68)*/8
PVC granules	640/0	720 (580)*/0	1,002 (980)*/0	780/0	540 (459)*/0	613 (599)*/0
Acrylonitryle/polyester fibers	84/0	82 (75)*/0	54 (49)*/0	68/0	72 (65)*/0	118 (111)*/0
Polypropylene fibers	120/0	180 (150)*/0	148 (130)*/0	220/0	120 (100)*/0	150 (146)**/0
Foil fragments	24/0	24 (21)*/0	16 (15)*/0	30/0	34 (31)*/0	24 (24)*/0
Personal care products fragments	25/0	26 (21)*/0	30 (24)*/0	25/0	27 (25)*/0	27 (24)*/0

*in brackets number of fibers/fragments captured in oil spots are placed;
**including the fibers trapped by the edge of toilet paper soaked in oil.

Table 2
The behavior of various microplastic particles during sedimentation under different conditions in a laboratory experiment – sedimentation in turbid water

Type of microplastics	Average concentration of microplastics, particles/L; expressed as initial/final					
	Sedimentation for 2 h in turbid water	Sedimentation for 2 h in turbid water with oil amendment		Sedimentation for 2 h in turbid water with toilet paper	Sedimentation for 2 h in turbid water with oil amendment and toilet paper	
		Small oil amendment	Larger oil amendment		Small oil amendment	Larger oil amendment
Polyester fibers	79/7	87 (30)*/8	102/(97)*/8	87/9	95 (41)*/7	86 (55)*/7
PVC granules	587/0	689 (402)*/0	865 (781)*/0	458/0	643 (242)*/0	704 (556)*/0
Acrylonitryle/polyester fibers	75/0	94 (69)*/0	43 (31)*/0	72/0	83 (61)*/0	89 (74)*/0
Polypropylene fibers	218/0	159 (146)*/0	205 (196)*/0	110/0	102 (90)*/0	184 (176)**/0
Foil fragments	25/0	23 (18)*/0	15 (14)*/0	33/0	23 (19)*/0	28 (26)*/0
Personal care products fragments	25/0	27 (20)*/0	25 (24)*/0	28/0	32 (26)*/0	27 (25)*/0

*in brackets number of fibers/fragments captured in oil spots are placed .

the beaker with clear water. It means that only on average 4 from 68 fibers settled down. Amendment of both small and larger oil amounts into the water did not affect significantly polyester fibers' behavior – on average 5 and 8 fibers were settled down in samples with the small and larger amendment of oil, respectively. It was equal to 5% and 4%.

Amendment of cellulose fibers (toilet paper) however slightly affected the fates of polyester microfibers. At average 3%–4% of more fibers were trapped by toilet paper in samples with clear water in the presence of cellulose fibers and with clear water with cellulose fibers and large amendment of oil. This phenomenon has not been observed in samples with small oil amendments in which on average 4 from 78 fibers settled. In samples collected from standing (clear) water cellulose fibers mainly trapped the microfibers which settle down, in their way down. It can be concluded that the admixture of oil has not significantly changed the sedimentation of polyester fibers.

It was simultaneously observed that oil spots attracted part of polyester fibers. The effect was especially visible in the samples were the concentration of oil was higher. In the case of polyester fibers, they were trapped at the periphery of a large spot (Fig. 7). In the case of smaller content of oil in the water, the same phenomenon was observed, however, the number of fibers attracted in this way was lower. Smaller spots of oils attracted on average about 26% of microplastics fibers form clear water whereas large spot about 66%. When cellulose fibers were present in the sample the percent shares were on average 31% and 65%, for smaller and larger oil amendment, respectively. In the samples with a lower concentration of oils no one large, but several smaller spots of grease were observed.

Observations concerning the sedimentation behavior of polyester fibers are important not only in terms of water treatment or wastewater treatment but also in the case of surface standing waters, for example, dam reservoirs. During water treatment sedimentation in clear water, sedimentation is probably not such important in some microplastics removal.

The different behavior of microplastic particles was observed in the other cases, it means in the case of polypropylene fibers, PVC granules, acetonitrile/polyester fibers, polypropylene fibers, foil fragments, and personal care products fragments. In all cases, no sedimentation of microplastic particles occurred during 2 h. All particles were floating on the table of water. They were however attracted by oil spots and contrary to polyester fibers, oil spots attracted them not only on the periphery but first in the area of the spot (Fig. 8). The figure presents polypropylene fiber behavior, however, the similar retention was observed in the case of all remaining microplastics, despite described above 100% polyester fibers. The fragments and fibers were mainly trapped inside the oil spots, not at their periphery.

Despite the acetonitrile/polyester fibers and PVC they were also no trapped by cellulose fibers (toilet paper). Cellulose fibers have trapped about 10% of PVC granules buy only in the sample with a large oil amendment in which the fibers of toilet paper were soaked with oil (data not presented in Table 1). Acetonitrile/polyester fibers were also slightly trapped by the cellulose fibers, but they were individual fibers only.

The presence of colloidal particles derived from kaolin did not affect sedimentation of the most types of microplastics (PVC granules, acrylonitrile/polyester fibers, polypropylene fibers, foil fragments, personal care fragments). They did not settle down both in clear water and turbid water samples. No microplastics particles at the bottom of the beaker were found. Oil spots, however, slightly better-trapped polypropylene fibers in the turbid samples. In turbid water percent shares were 92% for small oil amendment and 95% in the case of larger oil amendment. For the record in clear water, it was 83% and 87%, respectively. It was probably because the density of turbid water was higher than the clear one and buoyant force was higher. Polypropylene fibers which are made from light plastic and were rather wide and smooth compared, for example,

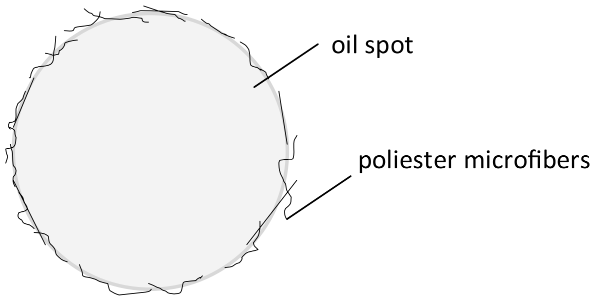

Fig. 7. Trapping of polyester fibers by oil spots.

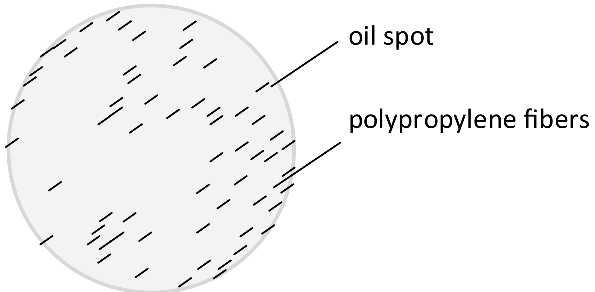

Fig. 8. Trapping of polypropylene fibers by oil spots.

polyester fibers tended to float at the surface of the water. As in the case of clear water also in the case of the turbid one cellulose fibers did not trap polypropylene fibers and others, except for the polyester fibers. Kaolin which made the water turbid slightly increased the percent share of polyester fibers which settled down compared to the clear water. For example, in turbid water, the percent share of the settled fibers was 8.8% compared to 6% in clear water. The amendment of oil did not significantly affect the sedimentation behavior of polyester fibers in turbid water compared to turbid water without the oil amendment. In the case of acetonitrile/polyester fibers which were of shape and constitution similar to cotton wool, part of the fibers tended to aggregate in larger flocks similar to wet cotton wool.

3.2. *Effect of coagulation on fibers content in the water body*

To evaluate the effectiveness of the coagulation process Fe(II)SO_4 solution was added into the samples. No coagulation aids were added because the aim of the process was rather to simulate the conditions in wastewater where ferric salts could be added to remove, for example, phosphates. The results of the experiments are given in Table 3.

In the case of polyester fibers coagulation process allowed for the effective removal of a significant part of fibers in samples without the oil amendment. It was on average 84% in the case of the sample without cellulose fibers and on average 89% in the presence of toilet paper in the sample. It means that in this case cellulose fibers slightly affected "trapping" of polyester fibers. Flocs which appeared after coagulation trapped polyethylene fibers. In the case of samples with oil part of the fibers were, however, remained in the spots of oil, but coagulation (probably as a result of mixing) caused the situation when part of the fibers was trapped in flocs and the part in oil spots. Percent shares of fibers that were trapped by oil spots were lower than in the case of water without coagulation. After coagulation in the sample with the presence of oil, but without cellulose fibers about 24% settled, and in the presence of oil and cellulose fibers, 31.6% were removed by sedimentation. If oil spots were present in the sample part of the fibers floated at the water table remained there. As a conclusion, it can be stated that coagulation effectiveness in the case of polyester fibers was affected by the presence of oils.

The coagulation process has not affected the removal of polypropylene fibers, foil fragments, and personal care products fragments, both in the presence of oil and without. The fibers still floated at the water table. Coagulation removed turbidity from water. During it, some flocks were generated. The effectiveness of coagulation could be increased by the use of polymers, however, it needs further experiments and has not been studied during the present research work. The coagulation process only slightly affected the fates of PVC powder. Small granules of PVC in 80% stayed on the surface of the water. In the case of the sample with oil practically no differences were observed in the granules' behavior before and after the coagulation process. They were mainly (>91%) trapped in oil spots. As it was stated by Ma et al. [31] using positively charged coagulants does not allow for the effective removal of positively charged microplastic particles. The process can be enhanced by adding negatively charged coagulant aids, which allow for effective aggregation of positively charged PVC particles. More detailed studies on this phenomenon are however necessary to determine the optimal conditions for both coagulation phosphorus removal and microplastics aggregation. Acrylonitrile/polyester fibers stayed at the surface of the water also. As a result of mixing they have been flocked on the surface of the water, similar to wet cotton wool. Some flocks which appeared during the coagulation have stuck to that large floc of microplastic fibers (Fig. 9), but they do not make them settled down.

The results obtained in the study allowed us to evaluate the effectiveness of microplastics separation from the water phase (both clear and turbid) under laboratory conditions. According to literature data, a large part of these pollutants are removed in wastewater treatment plants during the sedimentation process, or more accurately during preliminary (mechanical treatment), for example, Michielssen et al. [34] have confirmed that about 62%–82% of microplastics of sizes higher than 0.020 mm had been removed during preliminary sedimentation. As suggest Lusher et al. [35] by settling the microplastics of higher density could be mainly separated by sedimentation. The results obtained during our study suggest however that also the shape plays an important role, the fibers of polyester which were flexible and longer (Fig. 1) than polypropylene fibers have undergone sedimentation better than the mentioned polypropylene fibers (Fig. 4), however, the similar specific gravity values also could play an important role – specific gravity of polyester is 1.38 g/cm^3, whereas of polypropylene 0.9–0.92 g/cm^3 [1]. Also, Mitening et al. [25] higher density microplastics can be removed effectively during the sedimentation step and are expected to be present in sludge. The results reported by other authors indicated that in sewage sludge an average

Table 3
The behavior of various microplastic particles after coagulation under different conditions in the laboratory experiment

Type of microplastics	Average concentration of microplastics, particles/L; expressed as initial/final			
	Coagulation in samples without oil amendment		Coagulation in samples with oil amendment	
	With Fe(II) sulfate (sample without cellulose fibers)	With Fe(II) sulfate (sample with cellulose fibers)	With Fe(II) sulfate (sample without cellulose fibers)	With Fe(II) sulfate (sample with cellulose fibers)
Polyester fibers	69/58	76/68	84 (70)*/20	101 (95)*/32
PVC granules	587/117	479/86	312 (288)*/0	587 (540)*/0
Acrylonitryle/polyester fibers	88/0	67/0	64 (52)*/0	91 (85)*/0
Polypropylene fibers	107/0	87/0	74 (67)*/0	61 (55)*/0
Foil fragments	37/0	26/0	44 (30)*/0	31 (25)*/0
Personal care products fragments	27/0	32/0	24 (20)*/0	24 (23)*/0

*in brackets number of fibers/fragments captured in oil spots are placed.

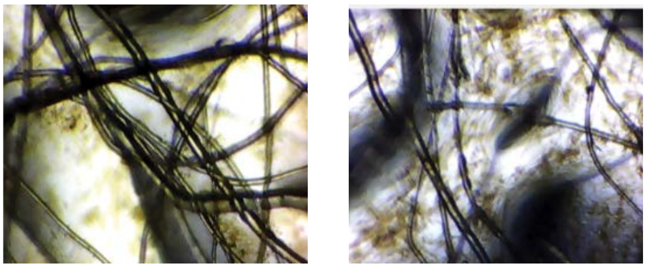

Fig. 9. Ferrous hydroxide flocks which occurred during coagulation trapped by acetonitrile/polyester fibers.

number of microplastic particles was in the range of about 500 to almost 6,000 particles/kg d.m. [35].

The results obtained during the study have indicated that sedimentation as a unit process alone did not affect the fates of the high part of microplastics. Only one class of microplastics – polyethylene microfibers have partially settled down during the experiment. The process which seems to have the most impact on microplastics fate in the water environment is trapping by oil, and also probably the grease. All microplastic particles showed a strong affinity for oil. Because of this content of microplastics in foam and grease in grease traps should be analyzed. Such authors such Lusher et al. [35] have indicated the importance of grease removal on the fates of microplastics, but no detailed studies have been done since now concerning this phenomenon. Lusher et al. [35] based on the results of Murphy et al. [26] and Carr et al. [22] that microplastic particles indeed have accumulated in grease skimmer and primary skimmer, but according to calculations only about 6% of the particles appeared to be contributed by the grease skimmer. The differences between the results obtained in our study and the studies by Murphy et al. [26] and Carr et al. [22] could have been caused by the conditions of the experiment. It is without a doubt difficult to obtain a representative sample of skimmer in WWTP. However, Lusher et al. [35] have noticed based on literature data that collected grease and foam could be important vectors for the transfer of low-density microplastics to the sludge treatment line. Also, Mintenig et al. [25] have indicated that buoyant microplastics may be removed in grease separating step. The results obtained in our study support this thesis.

Remy et al. [21] have indicated that toiled paper present in wastewater, and cellulose fibers are effective in microplastics removal from the water phase during preliminary sedimentation. Cellulose fibers can capture the synthetic microfibers. The results obtained in our study only partially confirmed this thesis. Cellulose fibers trapped some fibers but mainly when the cellulose was soaked with oils. Then it attracted some granules of PVC as well as some fibers of polyester. Fragments of foils and fragments of personal care products were not removed from wastewater in this way.

Literature data indicate that also coagulation may be effective in microplastic removal. Coagulation is concerned as one of the processes which play an important role in microplastics removal during water treatment. But more detailed studies do not always support this thesis. Research made by Ma et al. [31] on the coagulation process effectiveness on microplastics removal by using Al- and Fe-based salts in the presence of polyethylene showed removal efficiency of PE lower than 40%. Aluminum salts were more effective in microplastics removal than Fe salts. Other parameters, such as turbidity and ionic strength barely affected the effectiveness of the process. Our studies have confirmed that coagulation could be indeed effective in some classes of microplastic removal. Especially it concerns the flexible fibers and small granules of PVC powder. It was not effective in the removal of fragments of foils or personal care products. The coagulation process was affected by the presence of oils, which have trapped microplastic particles in the oil spots.

4. Conclusions

Results obtained during the study permit the following conclusions:

- Most of the microplastics used during the studies did not settle down under the gravity force. The only low part of polyester fibers (about 6%) settled down during 2 h of the experiment.

- Oil presence in the sample had an important effect on all kinds of microplastics particles. Oil spots have trapped microplastics fibers, powder, and fragments.
- The presence of cellulose fibers in wastewater had no significant effect on microplastics removal. The slightly higher removal efficiency was observed when the microplastics were soaked with oil.
- The coagulation process was effective in the removal of two kinds of microplastic particles polyester fibers (84%–89% removal efficiency) and PVC powder (20% removal efficiency). The coagulation process was not supported by coagulant aids. The main parameter which affected coagulation was the presence of oils. In the presence of oil huge parts of microplastics were trapped in their spots and did not undergo coagulation.

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