

# Application of waste polypropylene bags as filter media in coalescers for oily water treatment

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## Abstract

The polypropylene fibers have been used in bed coalescers for separation of micro-sized oil droplets from water for a long time. Possibilities of applying different forms of polypropylene as filter beds are still being in the focus of many researches. The possibility of applying waste polypropylene bags used for packing vegetables (PPDJ) was investigated in this paper. The results are compared with results obtained by applying waste polypropylene fibers from carpet production (PP). It is well known that there are difficulties to separate the oils of low viscosity by polymer fiber beds. Due to the above mentioned, the presented research refers to separation of low viscosity mineral oil from water. The obtained experimental results confirm that the material PPDJ could be efficiently used as a bed material for coalescers. The critical velocity of 50 m h<sup>-1</sup> could be reached at using both polypropylene forms that is from bags PPDJ and from the carpet industry PP, when the adequate bulk density of materials is used.

**Keywords:** Oily water, waste polymers, fiber bed, bed coalescer, bed permeability

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## 1. INTRODUCTION

Oily wastewater is widespread in industry as well as in many services. It could be generated in solvent extraction processes, by heat-exchanges, during the crude oil production, in cooling of units or storage tanks [1,2]. Cleaning the workspace, washing and degreasing engine and heavy machinery parts also cause formation of oily wastewaters. The accidental oil spills may occur occasionally during transportation of crude oil and its products by tankers. This causes oil contamination of rivers, lakes and oceans.

There are many separation techniques for treatment of oily wastewater such as sedimentation, filtration, membrane separation, and adsorption. Selection of the appropriate technique depends on the wastewater quantity, concentration and form of the oil, as well as on required quality of the effluent, which has to be reached [2-5].

Bed coalescers are used for separation of micro-sized droplets of the dispersed phase. They are very successful in separation of both oil from water and water from oil. The fiber materials are the most frequently used as filter media in coalescers. The coalescers are usually consisted of a filter bed section and a settling section. The coalescer can operate in steady-state and non-steady state regimes [6-8]. The steady-state regime is reached, when a capillary-conducted disperse phase is formed inside the bed. This phase is spread from the inlet to the outlet of the bed. According to some authors, the capillary-conducted phase is formed very quickly, in 20 minutes maximum from the start of the emulsion flow [6-8]. Small droplets entering the bed are coalescing on the capillary-conducted surface, while large globules are detached from the surface and settled after leaving the bed [1,3, 6-8]. Coalescing filtration is a complex phenomenon due to involving a bed of high porosity, up to 98 %, and two immiscible liquids [9-15] leading to a large number of variables affecting occurrences in this complex system.

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For a long time, the polypropylene fibers have been used for separation of oil droplets from water. Even, Sareen *et al.* [16] investigated the separation efficiency of polypropylene fibers and compared it with efficiencies obtained by using different fibers like medical cotton, dynel, glass, polyethylene and Teflon. They used polypropylene fibers with a mean diameter of 45  $\mu\text{m}$ . The authors showed that the separation efficiency of a polypropylene fiber bed was in the range of separation efficiencies realized with other investigated materials [16]. Possibilities of applying different forms of polypropylene as a filter material are still being in the focus of many researches [17-23].

Beside polypropylene fibers with diameters of a few tens of microns, the other polypropylene forms such as wide strips, granules, hollow cylinders, and twisted tapes are investigated for oily wastewater treatment as well showing different efficiencies [19-22].

Over a long period of time, the research group of Šećerov Sokolović has been investigated possibilities for application of waste polymer fibers as filter media in coalescers for separation of mineral oil from water. In their investigations, the fluid velocity has been increased until the oil concentration in the effluent reached the value of 15  $\text{mg L}^{-1}$ . The acceptable effluent oil concentration is set to this value, as the often-recommended limit of oil concentration in wastewater [24,25]. In addition, the bulk density was varied as well. This parameter affects the bed permeability, porosity and pore size leading to a change in bed geometry. In their comprehensive research fiber diameter of investigated materials was close to 40  $\mu\text{m}$  [10,11,21]. In this way, the bed geometry for the same bulk density was maintained similar, while the authors investigated other phenomena in the bed coalescer.

The huge amounts of plastic bags are generated every day. For environmental protection and sustainable development, it would be very beneficial if part of these bags could be used again without any processing. Therefore, the scope of this paper is to investigate possibility of application waste polypropylene bags used for packing vegetables, PPDJ, as filter media in bed coalescers for oily water treatment. The obtained results for application of PPDJ are compared to previously published results [23], realized by applying waste polypropylene from carpet production (PP) as a filter bed material.

## 1. MATERIALS AND METHODS

### 1. 1. Experimental setup and operating conditions

Separation of mineral oil from water was performed on a pilot plant bed coalescer with the capacity of 100  $\text{L h}^{-1}$  in the steady-state regime, designed as described in detail in the previous paper [23]. Each experiment was repeated at least two times using fresh bed material for the same bulk density. The exact value of bed permeability was achieved by compressing the adequate mass of polymer fibers to the bed length of 5 cm. The model emulsion of mineral oil, at 20 °C temperature, marked as P1, was prepared in two tanks with the volume of 80 L, each. The oil was dispersed in water by continuous stirring by an impeller for 45 min at the rate of 650 rpm prior to the start of the experiment, and continuously during the experiment. In this way, the mean droplet diameter of 9-10  $\mu\text{m}$  (min. 0.9  $\mu\text{m}$ , max. 28  $\mu\text{m}$ ) was ensured during the experiment. The droplet size distribution was determined by the Elzone 280 PC particle counter (USA). The experiments were monitored by determination of the oil concentration in effluent,  $C_e$ , by FTIR spectrophotometer (Thermo Nicolet 5700, Netherlands). Samples of oily water were stabilized and adjusted to pH 2 by adding HCl. Oil from the sample was extracted with  $\text{CCl}_4$ . Fluid velocity was in the range from 16 up to 60  $\text{m h}^{-1}$ . The selected fluid velocity,  $v$ , was kept constant for 60 min. Composite samples of the effluent were taken during the last 15 min at 5 min intervals. The bed permeability,  $K_0$ , was widely varied by changing the bulk density amounting to the range of  $5.389 \cdot 10^{-9}$  to  $0.180 \cdot 10^{-9} \text{ m}^2$ .

### 2. 2. Fiber and bed properties

Waste polypropylene bags, obtained from marketplace, were cut in squares of 10×10 cm. Photographs of materials were taken by a digital camera (Fujifilm FinePix S5500, Japan). Surface morphology, microstructure, and size of the fibers and stripes were characterized by scanning electron microscopy by using a JEOL, JSM-6460 LV instrument (USA) and by optical microscopy (Olympus BH.2 RFCA, Netherlands). The density of the material and bed porosity was measured by

the weighing methods yielding the material density of  $900 \text{ kg m}^{-3}$ . Measurements of bed porosity were repeated three times with using a certain quantity of fresh polymer material over adequate bulk density. Bulk densities of both materials were in the range from 30 to  $150 \text{ kg m}^{-3}$ . The melting point of polypropylene of  $168.60 \text{ }^\circ\text{C}$  was determined by using a differential scanning calorimeter Q20 (TA Instruments, USA). The bed permeability was calculated from the measured pressure drop across the bed for tap water by applying the Darcy's law. Darcy experiments were repeated three times with a certain quantity of fresh bed material. The specific surface area of material was determined as the ratio of the fiber surface and fiber volume.

### 2. 3. Properties of dispersed mineral oil

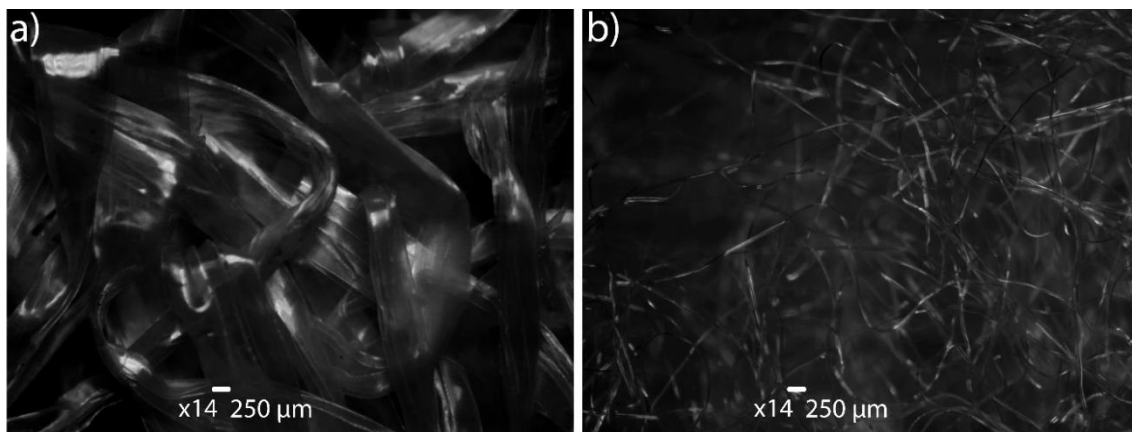
Main characteristics of the investigated mineral oil P1 (blended petroleum product with a high paraffinic content) were: density at  $20 \text{ }^\circ\text{C}$  was  $879.0 \text{ kg m}^{-3}$ , viscosity at  $40 \text{ }^\circ\text{C}$  was  $10.3 \text{ mPa s}$ , pour point was  $+3 \text{ }^\circ\text{C}$ , neutralisation number was  $0.13 \text{ mg KOH L}^{-1}$  and the interfacial tension was  $32.4 \text{ mN m}^{-1}$ . The oil density was determined according to the standard SRPS ISO 12185:2004, while the oil viscosity was determined according to the standard SRPS ISO 3104:2003. The pour point was estimated according to the standard SRPS ISO 3016:1997, while neutralisation number was measured according to the standard SRPS ISO 6619:1994. The standard ASTM D 971 was used to determine the interfacial tension.

## 2. RESULTS AND DISCUSSION

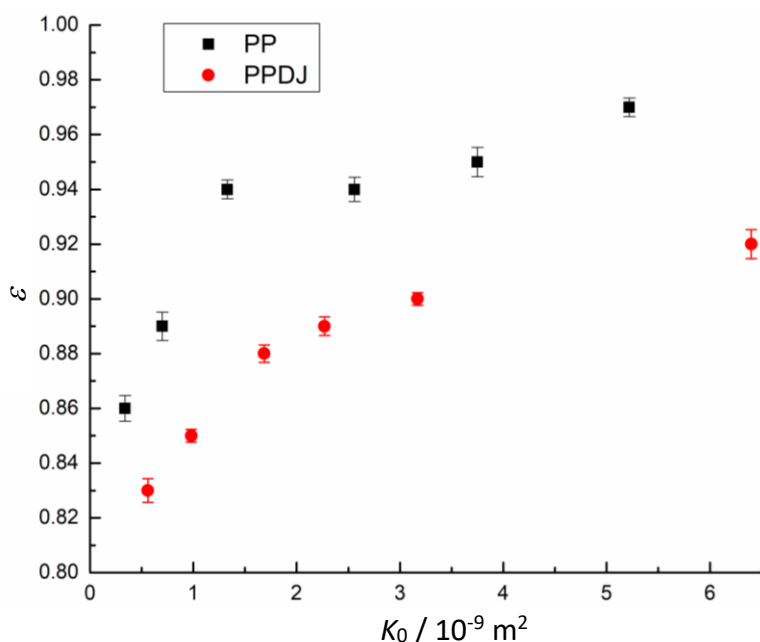
The waste polypropylene bags used for packing vegetables, PPDJ, are shown in Figure 1a, while the waste polypropylene fibers from carpet production, PP, that were described in the previous study [23] here are shown in Figure 1b for comparison. It could be seen that material PPDJ looks completely different as compared to the material PP. Therefore, it could be expected that PPDJ fibers would form a bed with different characteristics. Optical micrographs of packed beds composed of PPDJ and PP fibers are shown in Figure 2a and 2b, respectively. Cross-section of PPDJ stripes is rectangular with the width of  $1001 \pm 14 \text{ } \mu\text{m}$  and the thickness of  $20 \pm 4 \text{ } \mu\text{m}$ , while the cross-section of PP fibers is circular with the diameter of  $38 \pm 3 \text{ } \mu\text{m}$ . The question arises whether this fact will affect the separation efficiency of oil from wastewater. Relation of the bed porosity and the bed permeability obtained from experimental data is presented in Figure 3. From the obtained results it could be seen that the bed made of PPDJ has lower porosity than the bed of material PP over all investigated permeabilities. Porosity of the PPDJ bed is in the range from 83 to 93%, while that for the PP bed is in the range from 87 to 97 %.



**Figure 1.** Photographs of tested polypropylene **a** - waste bag PPDJ; **b** - waste fibers from carpet production, PP



**Figure 2.** Optical micrographs (14x) of polypropylene a. from waste bags PPDJ b. waste fibers from carpet production, PP; scale bar: 250  $\mu\text{m}$

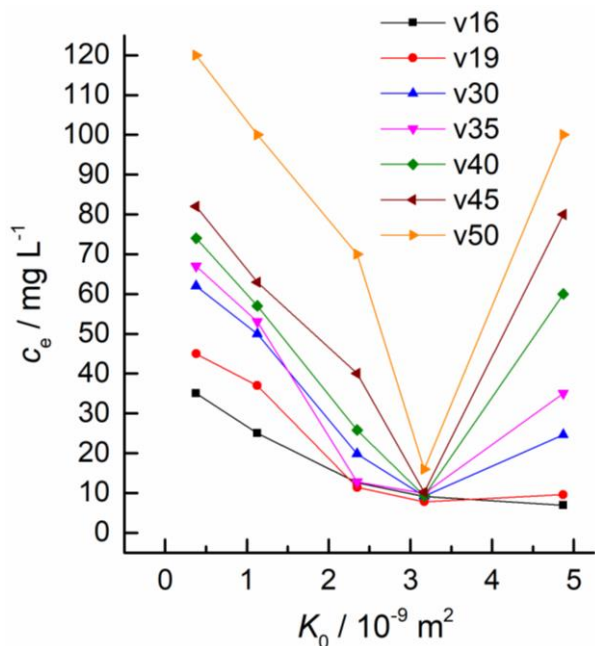


**Figure 3.** Experimental relation of the bed porosity,  $\epsilon$ , and bed permeability,  $K_0$ , for investigated polypropylene fibers

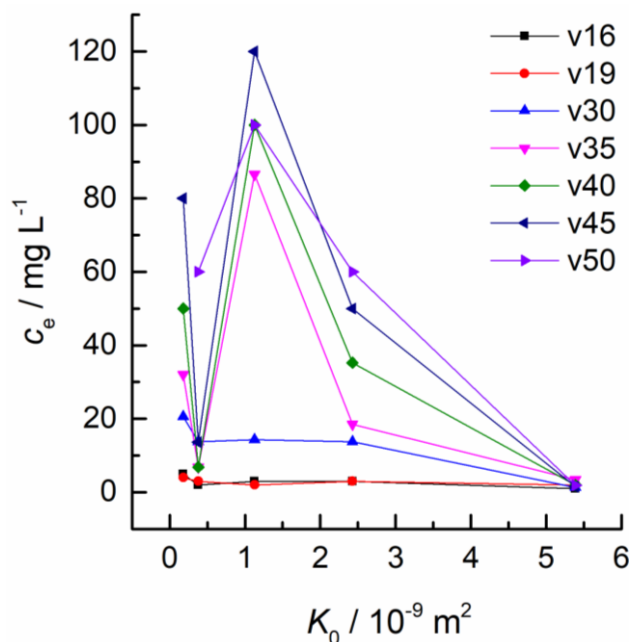
The other property that characterizes the material is the specific surface area. For the material PPDJ, the specific surface area is 2.2  $\text{m}^2 \text{m}^{-3}$ , while for the material PP it is significantly higher yielding 10.0  $\text{m}^2 \text{m}^{-3}$ . Since the selected materials form fiber beds of high porosities ranging from 83 to 97 %, it can be assumed that the bed of the PP material is filled with a significantly higher number of fibers as compared to that of the PPDJ material.

Dependence of the effluent oil concentration on the bed permeability over all applied fluid velocities could be presented in the form of a 2D diagram. Figure 4 shows such graphical dependence for material PPDJ, while Figure 5 shows the same dependence for material PP. The experimental results confirm that separation of the oil P1 from water by both materials is relatively difficult. Ranges of bed permeabilities and working velocities over which quality of the effluent is acceptable, are very narrow and unconnected. The recommended limit of oil concentration in the effluent (15  $\text{mg L}^{-1}$ ) over the whole range of investigated bed permeabilities is achieved for both materials at working velocities of 16 and 19  $\text{m h}^{-1}$ , only.

Coalescing filters are often used on ships and oil platforms where the space is limited. Therefore, it is important to minimize the coalescer dimensions. In order to minimize dimensions of the unit, it is necessary to experimentally find conditions at which the highest working velocity is realized at the acceptable effluent quality.



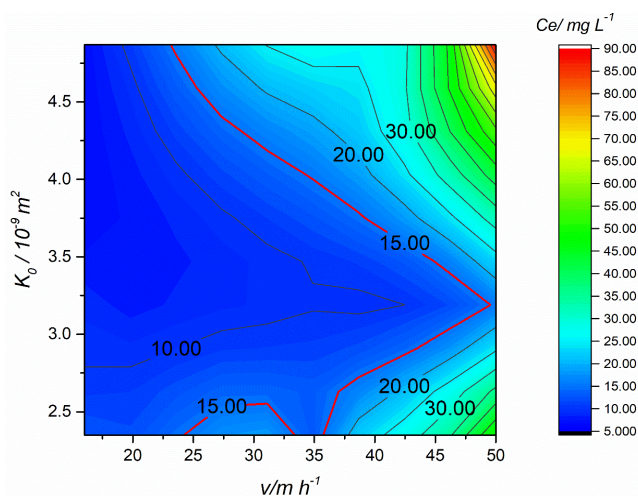
**Figure 4.** Effluent oil concentration,  $C_e$ , as a function of the bed permeability,  $K_0$ , for different superficial fluid velocities (v16 to v50 in  $m\ h^{-1}$ ) for the PPDJ material



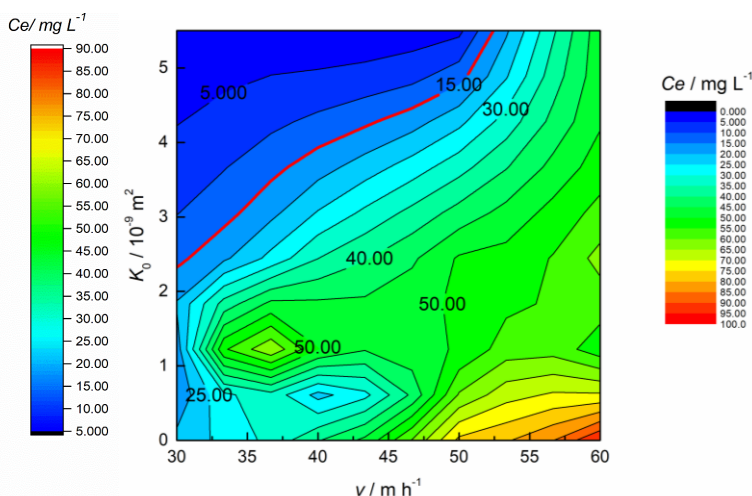
**Figure 5.** Effluent oil concentration,  $C_e$ , as a function of the bed permeability,  $K_0$ , for different superficial velocities (v16 to v50 in  $m\ h^{-1}$ ) for the PP material (adapted with permission from [23])

The material PPDJ at the working velocity of  $50\ m\ h^{-1}$  and the bed permeability of  $3.17 \cdot 10^{-9}\ m^2$  provided the effluent oil concentration lower than  $15\ mg\ L^{-1}$ . On the other hand, the material PP at the highest bed permeability provided the recommended water quality over all investigated velocities, including the fluid velocity of  $50\ m\ h^{-1}$ . The obtained improved separation of oil droplets at higher bed permeabilities for material PP, is due to resulting higher bed porosity, which causes lower interstitial velocity. According to literature, in these circumstances the highest amount of capillary-conducted phase is formed in pores of the bed [6-8], which favors effective coalescence of oil droplets and thus better droplet separation.

If the dependence of the effluent oil concentration on the bed permeability and fluid velocity is presented by contour diagrams (Figs. 6 and 7), the areas of the best operating conditions for coalescers are easier to be determined. Red bold contour lines separate suitable working areas for a coalescer from the unsuitable ones.



**Figure 6.** Contour diagram representing the interdependence of the effluent oil concentration, bed permeability and superficial velocity for oil P1 and material PPDJ



**Figure 7.** Contour diagram representing the interdependence of the effluent oil concentration, bed permeability and superficial velocity for oil P1 and material PP (adapted with permission from [23]).



The working velocity of  $25 \text{ m h}^{-1}$  is reached for the bed material PPDJ over permeabilities lower than  $3 \cdot 10^{-9}$  and higher than  $4 \cdot 10^{-9} \text{ m}^2$  (Fig. 6). However, it is possible to reach even a higher working velocity of  $45 \text{ m h}^{-1}$ , but just in a very narrow range of bed permeabilities, from  $(3 \text{ to } 3.5) \cdot 10^{-9} \text{ m}^2$ .

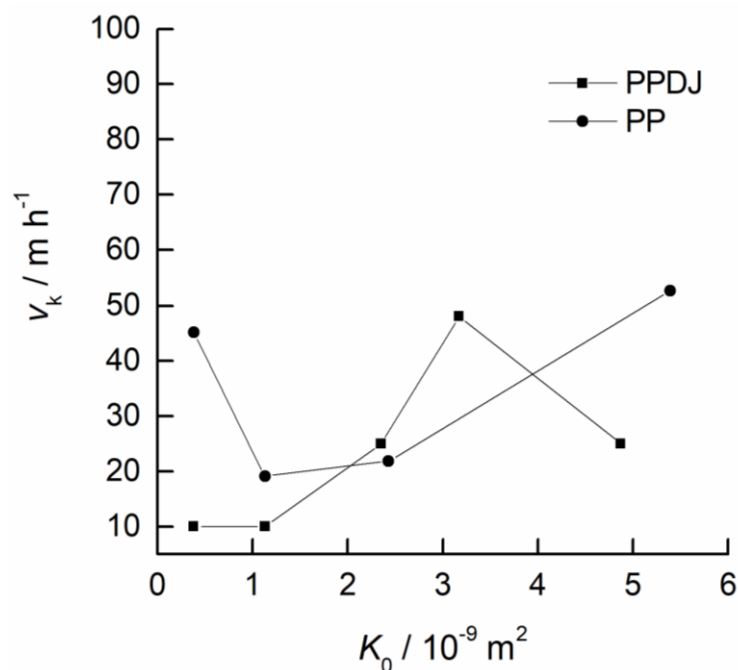
The bed material PP (Fig. 7) in the permeability range from  $(4 \text{ to } 5) \cdot 10^{-9} \text{ m}^2$  reaches working velocities in the range from 45 up to  $50 \text{ m h}^{-1}$ , while with a decrease in the bed permeability the working velocity decreases as well.

In their previously published papers [1,10,21], Šećerov Sokolović *et al.* suggested that the critical velocity is the best parameter for analysis of the results obtained from studies of fiber bed coalescence. The authors recommended that the critical velocity should denominate the velocity above which the effluent oil concentration exceeds the value of  $15 \text{ mg L}^{-1}$ . Dependence of the critical velocity on the bed permeability for both investigated materials is shown in Figure 8.

The bed of the PPDJ material reached the highest critical velocity of  $50 \text{ m h}^{-1}$  for the bed permeability of  $3.17 \cdot 10^{-9} \text{ m}^2$ . Such a high critical velocity could be also reached in the bed of PP material, but only at the highest bed permeability.

In the published literature [18,26-32] only the influence of a circular fiber diameter on separation efficiency was investigated. These investigations were realized over one or maximum two bed porosities, while the bed permeabilities were unknown. Based on these results, it was noticed that the use of fibers with larger diameters results in lower separation efficiencies. However, based on results presented in Figure 8, obtained over a wide range of bed permeabilities, the influence of fiber size and form on separation efficiency (critical velocity) could be noticed, showing a major dependence on the bed permeability. Under high range of bed permeability the material with a much larger cross-sectional area of fibers (*i.e.* PPDJ) results in the same separation efficiency as the material with a small fiber diameter and smaller cross-sectional area (*i.e.* PP; Fig. 8).

Based on the presented results, it is necessary to emphasize that the waste polypropylene bags used for packaging of vegetables, PPDJ, can be successfully used as a filter media in a bed coalescer for the treatment of oily water.



**Figure 8.** Critical velocity as a function of the bed permeability for investigated polypropylene materials

#### 4. CONCLUSION

The waste polypropylene bags used for packing vegetables, PPDJ, are fibers with rectangular cross-section, of the width of about 1 mm and the thickness of about  $20 \mu\text{m}$ . The specific surface area of this material is  $2.2 \text{ m}^2 \text{ m}^{-3}$ . Over the investigated range of bed permeabilities, from  $0.3 \text{ to } 6.0 \cdot 10^{-9} \text{ m}^2$  the obtained bed porosities are ranging from 83 up to 93 %. The presented experimental results confirmed that the material PPDJ could be efficiently used as a bed material

for coalescers applied for treatment of oily water. Critical velocity of  $50 \text{ m h}^{-1}$  is reached for PPDJ fibers as a filter media in bed coalescers. When comparing material PPDJ to the polypropylene fibers PP, that were previously examined, it can be noted that there are packing conditions under which the material PPDJ exhibits the same separation efficiency as the PP material. Therefore, waste bags PPDJ can be classified in the group of fiber bed materials with the high separation efficiencies of oil from water. Last but not the least, these bags do not present any cost, and the environmental pollution is reduced since they are not disposed anymore on landfills. In this way the concept of circular economy and sustainability could be reached as well.

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## SAŽETAK

### Primena otpadnih polipropilenskih džakova kao filtarski materijal u koalescentnim filterima za tretman zauljenih voda

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(Naučni rad)

U dužem vremenskom periodu polipropilenska vlakna su korišćena u koalescentnim filterima za odvajanje kapi ulja mikronske veličine iz vode. Mogućnosti primene različitih oblika polipropilena kao filatrskog materijala još uvek su u fokusu mnogih istraživanja. U ovom radu ispitivana je mogućnost primene otpadnih polypropilenskih džakova koji se koriste za pakovanje povrća PPDJ. Rezultati su upoređeni sa rezultatima dobijenim primenom otpadnih polypropilenskih vlakana iz proizvodnje tepiha PP. Dobro je poznato da postoje poteškoće u razdvajanju ulja niske viskoznosti slojem polimernih vlakana. Iz tog razloga predstavljena istraživanja odnose se na odvajanje mineralnog ulja niske viskoznosti iz vode. Dobijeni eksperimentalni rezultati potvrđuju da se materijal PPDJ može efikasno koristiti kao materijal u koalescentnom filter za separaciju ulja iz vode. Kritična brzina od 50 m h<sup>-1</sup> može se postići upotrebom polipropilenskih oblika, PPDJ i PP kada se koristi odgovarajuća nasipna gustina materijala.

*Ključne reči:* zauljena voda; otpadni polimeri; sloj vlakana; efikasnost koalescencije; permeabilnost sloja