SEPARATION OF OIL DROPS FROM WATER USING STAINLESS STEEL FIBER BED

ARPAD KIRALJ1
TATJANA VULIĆ1
DUNJA SOKOLOVIĆ2
RADMILA ŠEČEROV SOKOLOVIĆ1
PERO DUGIĆ3

1University of Novi Sad, Faculty of Technology, Novi Sad, Serbia
2University of Novi Sad, Faculty of Technical Science, Novi Sad, Serbia
3University of Banja Luka, Faculty of Technology, Banja Luka, Bosnia and Herzegovina

SCIENTIFIC PAPER

UDC 544.7:66.06:628.3

Both oil-in-water and water-in-oil emulsions are widespread in industry and in nature. When it is necessary to carry out the separation of the dispersed phase, the emulsion stability, phase concentration and droplet size predominantly influence the separation technique. In the case of emulsions that have droplets smaller than 100 μm, one of the most economically acceptable separation techniques is coalescence filtration [1-4].

Coalescence filtration has a fiber bed through which the emulsion flows. If proper conditions are achieved, the saturated oil phase is formed in the pores, so-called capillary-conducted phase, that allows new incoming droplets to be coalesced in its volume, while large globules break off from this volume and exit the bed. Visually, the explained phenomenon involves small droplets entering the bed and larger droplets exiting the bed enabling separation by settling [1,5-9].

Many parameters influence the filtration and bed coalescence: bed thickness [10-15], nature of filter material [16-21], superficial flow velocity [2,3,13-15], orientation of the fluid flow [2,3,22], the properties of
both fluids [2,3,10,11,23] and the bed geometry [3,9,14,15,24].

The materials’ nature is defined by their chemical structure and roughness. According to the chemical structure, materials can have high surface energy, such as glass, metals, ceramics, or low surface energy as polymers [16-26]. This applies only to materials with smooth surfaces. If the surface roughness is increased then the surface energy of the polymer can also increase and become high. In literature, this double impact of surface energy is not emphasized enough in the liquid-liquid separation using bed coalescers.

Some opinions have been published noting that polymer materials are used for the separation of oil from water due to their oleophilic surface, and high surface energy materials are commonly used for the separation of water droplets from oil [14,15,18,19]. This approach shows the importance of surface wetting of fibers with dispersed phase. Accordingly, it should be noted that there are no reliable techniques for determining the wetting angle of curved surfaces, such as granules and fibers. It was found that the wetting angle depends on the surface geometry and not only on the roughness but also on the dimensions [27-32].

A significant number of authors studied the influence of oil properties on the separation of oil droplets from water [2,3,10,11,23]. It is important to underline whether pure chemicals or oils with complex structures are used. The authors agree that bed coalescence is most efficient for the separation of oil droplets with higher viscosity. Šećerov-Sokolović et al. pointed out that the polarity of mineral oil is an essential feature that significantly influences the separation efficiency [3,24].

In addition, Šećerov-Sokolović et al. think that the fiber bed geometry is very important and significantly contributes to the reduction or increase of separation efficiency in the filtration process but it is not enough investigated [1-3,8,23]. The properties that uniquely determine the bed geometry of fibers are not defined in the literature. Šećerov-Sokolović et al. revealed that the dependence of porosity on permeability, as well as constant fiber diameter determines the fiber bed geometry and underlined that it is necessary to maintain the same bed geometry during investigation of some other influences. It has been observed in some published research that the change of filter material and fiber diameter was not systematically investigated, which disabled the possibility to give conclusions about simultaneous impact of these two effects [33-37]. Davies, Austin and Jefreys [34,35] examined the influence of fibers nature on the separation of both, isooctane from the water, and water from isooctane. In the case where the water was dispersed phase the highest separation efficiency was achieved using glass fibers, slightly lower efficiency was obtained with stainless steel fibers, even lower with nylon and the lowest with teflon fibers. The authors found that the separation efficiency is inversely proportional to the value of surface energy of materials. However, when the water was the continuous phase and isooctane the dispersed phase, the dependence of the separation efficiency in relation to the surface energy did not change. It should be noted that the selected polymers, nylon and teflon, are not good representatives for polymer fibers, since these fibers are inelastic and sharp, and due to the liquid flow and the hydrodynamic forces influence they behave more like steel and glass fibers. This statement is confirmed by the fact that in today’s practice and research these polymers are no longer used. In addition, during these experiments, the porosity of the bed and the diameters of fibers were not constant. The authors argue that these properties are important in determining the overall efficiency of the system, but due to the little differences in the studied beds these properties were not crucial for the separation efficiency. The authors also concluded that the wettability of fibers with the dispersed phase that should be separated has no influence, but insist that roughness has a much greater impact on the separation efficiency.

Fahim and Akbar [14,15] used a fiber bed of combined glass and stainless steel fibers for the separation of jet fuel from water. They investigated the influence of the superficial velocity, bed thickness and inlet concentration of jet fuel on the pressure drop and separation efficiency. Glass fibers with 15 μm diameter and stainless steel fibers with 180 μm diameter were used. During the experiments, the oil content was monitored, as well as the droplet size distribution of the dispersed phase at the bed exit. The authors defined the separation efficiency through changes in the droplet diameters. As a result of visual inspection of the coalescence process inside the fiber bed, the authors observed that, when entering the bed, the droplets of the dispersed phase first attach to the surfaces of glass fibers. Thereafter, when the droplet diameters of dispersed phase are enlarged they transfer to the stainless steel fibers. In this way the capillary-conducted phase is formed. The authors considered that the surface of glass fibers has different wettability towards jet fuel compare to the wettability of steel fibers due to the earlier formation of
the oil membrane. The fact that was ignored by the authors was that the geometry of the glass fiber bed is completely different, due to the significantly smaller fiber diameter, by which the flow conditions, specific surface area of fibers bed and the size and shape of the pore space are significantly different when compared to the stainless steel fiber bed with much larger fiber diameter. Several authors varied fiber diameter in their studies and concluded that thinner fibers are more efficient for the droplet separation than thicker fibers [6,7,10,11,25,26].

Rebelein and Blass [36] examined the influence of the dispersed phase properties, the droplet diameter, the characteristics of fiber material, bed length and the superficial velocity on the separation of both, oil in water emulsions, and water in oil emulsions. The tested materials were glass and stainless steel fiber as well as polytetrafluoroethylene (PTFE). The diameters of fibers were: glass 12 μm, stainless steel 5 and 12 μm, and PTFE 40 μm. The authors concluded that the stainless steel fiber bed achieved the highest separation efficiency for the oil in water system. Simultaneous variation of both fiber nature and fiber diameter, made by these authors (also noticed in other published research) disables the possibility to obtain relevant conclusions. The PTFE fibers were about three to eight times thicker than steel fibers, which is not emphasized as an important factor in explanation of the results.

Magiera and Blass [37] examined the influence of the oil properties, droplet diameter, fiber diameter, fiber nature and bed length on the separation for both, oil in water and water in oil emulsions. Following fiber beds were investigated: glass fibers of 2.5 and 12 μm diameters, stainless steel fibers of 2.8 and 12 μm diameters, and teflon fibers of 60 μm diameter. The authors concluded that the high energy fibers were more efficient in the separation of dispersed droplets than the low energy fibers. Since in this study it was not specified which system was investigated oil-in-water or water-in-oil, it can be concluded that the findings relate to both systems. The same problem regarding the simultaneous variation of both fiber nature and fiber diameter, with the low energy fibers significantly thicker than the high energy fibers, leading to different bed geometry, was made in this study.

Painmanakul and colleagues [38] examined the influence of bed length, superficial velocity and the material nature on the separation efficiency of palm oil from the water. The authors have investigated commercial fibers of stainless steel and polymers. Material properties were determined based on the wetting angle measurements and critical surface tension of materials. The separation efficiency was monitored by determining the chemical oxygen demand (COD) in oily water before and after the flow through the fiber bed. The authors did not provide any information about the fiber properties, especially not the fiber diameters. Only photographs of the tested materials were presented. The efficiency of both fiber types as coalescence filter bed was tested. As a result, the polymer fibers exhibited extremely low separation efficiency of 44.4%. The stainless steel, which, according to the photographs, looks more like chips than fibers, showed separation efficiency of 40.2%. It can be assumed that the main reason for such low separation efficiency for both materials is badly selected bed geometry. In addition, the authors monitored the efficiency using COD. The values obtained in this way include also the dissolved organic part opening the question of the published research relevance.

Li and colleagues [39] investigated efficiency of commercial stainless steel fiber felt diameter of fiber 5 μm and modified fiber felt using different procedures for separation of oil droplets from water. Target of modifications were to change wettability or roughness and pore size of filter media. The dispersed oil phase was: n-hexadecane, n-octane, soybean and engine oil. Inlet concentration of the oil was 1000 mg/l. Mean droplet size was from 2 to 4 μm. Flow mode was vertical down. Fluid flow was 50 ml/min. They concluded that modified felts separate oil from water with high separation efficiency. The effects of pore size and surface wettability were investigated on model water with n-hexadecane. When the surface is amphiphobic, the separation is more sensitive to the change of pore size.

The aim of this study was to investigate the possibility of stainless steel fibers application maintaining constant diameter of this high surface energy, not wettable with the mineral oil droplets of different properties that have to be separated from the emulsion. In addition, the goal of this research was to determine the impact of the steel fiber bed geometry change, altered by the variation of the bed bulk density, which was achieved by compression of the fiber material.

**EXPERIMENTAL**

**Experimental setup of the bed coalescer and operating procedure**

The experiments were performed on a pilot plant bed coalescer capacity of 100 l/h with horizontal fluid flow orientation, the design of which has been described in detail in a previous paper [13,22]. Naphtenic
crude oil (A), its vacuum distillation fractions (A4), and petroleum semiproduct with a high paraffinic content without additives (P1) were used as the dispersed phase for bed coalescence experiments. All three dispersed phases were mineral oils of different properties containing natural emulsifiers such as asphaltenes. Oil droplets were dispersed in tap water by adding oil to the supply tank. The oil-in-water model emulsions with constant oil concentration (500 mg/L) were prepared in a two tanks (80 l each), by continuous agitation with a stainless steel impeller (650 rpm). In order to ensure the inlet mean droplet diameter of about 10 µm, each oily sample was continuously stirred 45 min prior to the experiment and onwards until the end of experiment. The mean inlet droplet size of the number distribution was dependent of the properties of the dispersed oil phase and it was determined by an Elzone 280 PC particle counter and Olympus BH.2 RFCA microscope:

- 9-10 µm (min. 0.8 µm, max. 31 µm) for oil A/water,
- 10-12 µm (min. 0.9 µm, max. 33 µm) for oil A4/water and
- 9-10 µm (min. 0.9 µm, max. 28 µm) for oil P1/water.

The steady-state regime of bed coalescence was achieved from the very beginning of the experiment by pre-oiling the fibers. The filter media were stainless steel fibers. The following parameters were kept constant in a coalescence experiment: bed length (5 cm), bed permeability, and working temperature (20 °C). Each oily water sample was tested for four bed permeabilities. The oil-in-water emulsion was pumped through a membrane dosage pump at superficial velocities ranging from 10 to 50 m/h. The selected velocity was kept constant for 1 h. Composite samples of oil-in-water emulsion were collected at the sampling point downstream of the bed and settling zone after 45 min of experiment start up at 5 min intervals.

Properties of dispersed oils

Three different kinds of dispersed oils were used with a wide range of physical and chemical properties. Their properties have been published previously [24]. Density was determined according to ISO 3675. Kinematic viscosity was measured using glass capillary viscometers according to the standard ISO 3104. Neutralization number was determined by potentiometric titration (ISO 6619). Mean molecular weight was estimated according to standard method ASTM d 2502-67 from kinematic viscosity measurements. Interfacial tension and surface tension measurements were done according the du Noüy ring method and stalagmometric method, respectively. Emulsivity was established using a centrifuge technique [40].

In this study mineral oils that are multicomponent mixtures of hydrocarbons with different structures were used. The selected oils significantly differ in all properties. Viscosity was in the range from 10 to 170 mP s, neutralization number from 0.10 to 1.70 mgKOH/l and emulsivity from 54 to 100%.

Properties of the bed

The stainless steel fibers were needle-punched and non-woven with random orientation. The surface morphology and size of the fibers were characterized by scanning electron microscopy (Figure 1).

Circular cross-section profile smooth fibers with average length of 30 mm and average diameter of 40 µm was used in the experiments. Due to compressibility of fibers, it was possible to vary the bed permeability over a wide range from 0.7 times 10 to 5.389 times 10 m² corresponding to bed porosity from 91 to 98%. The bed permeability was calculated from the measured pressure drop across the bed for tap water, because the data was complied with Darcy’s law.

Effluent oil concentration

Samples of oily water were stabilized and adjusted to pH 2 by adding HCl. Oil from the sample

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Figure 1. Surface morphology of used filter media. a) 100x; b) 20,000x.
was extracted with CCl₄. The effluent oil concentration was determined by FTIR spectrometry using a Thermo Nicolet 5700 spectrometer.

RESULTS AND DISCUSSION

Dependence of the effluent oil concentration on the superficial velocity, properties of oil and bed geometry

Due to the simultaneous analyses of the influence of three independent variables: the superficial velocity, the oil properties and bed geometry, it is more convenient to use 3D diagrams and contour diagrams that was already discussed in the published study of Šećerov Sokolović et al. [3,24].

The dependence of effluent oil concentration, superficial velocity and the bed permeability for oil A4 is given in Figure 2, in the form of a 3D diagram in Figure 2a, and contour diagram in Figure 2b.

The equivalent diagrams for oils A and P1 are given in Figures 3 and 4, respectively.

The 3D diagram noticeably reveals that the dependency of the effluent concentration, superficial velocity and bed permeability is drastically different in all the three oils investigated. Observing the oil A4, it was detected that there is an area bounded with velocity and permeability, where extremely low effluent oil concentrations are achieved. This area is located in a velocity range below 30 m/h and applies to all permeability values. For oil A such an area almost non-existent. At low permeability for almost all range of superficial velocity effluent concentration is relatively low, while all other areas are unfavorable for work. The higher the permeability is higher effluent oil concentration is achieved. Oil P1 has almost no area with low effluent concentration.

In order to observe the boundary of the recommended concentration values, it is necessary to analyze the contour diagrams. The lines within these diagrams are lines of equal concentration of oil in effluent and can be called iso-concentration. In these dia-

Figure 2. a) Three-dimensional diagram and b) contour diagram representing the interdependence of effluent oil concentration, superficial velocity and bed permeability for oil A4.

Figure 3. a) Three-dimensional diagram and b) contour diagram representing the interdependence of effluent oil concentration, superficial velocity and bed permeability for oil A.
grams are thickened lines correspond to the recommended concentration of 15 mg/l.

In the contour diagram for oil A4, Figure 2b, the precise area suitable for work is shown, as well as the numerical values of selected independent variables. For the whole range of bed permeability and superficial velocity, being even slightly higher than 30 m/h, the effluent oil concentration is below the recommended. For the area of low bed permeability (below $2.000 \times 10^{-9}$ m$^2$) it is possible to achieve the working velocity even at higher values of 45 m/h. The working velocity is the superficial velocity that obtains the required quality of effluent.

However, for the oil A, Figure 3b, the area of high bed permeability is completely unsuitable for the work. By reducing the bed permeability, the area of working velocity increases up to 40 m/h.

Based on the contour diagram for oil P1, Figure 4b, it is clear that the working area for this oil is incomparably smaller than for the previously discussed oils. For the full range of permeability, the effluent quality can be achieved at superficial velocity of slightly less than 20 m/h, while for this oil, low permeability is most favorable and provides maximum working velocity, which is slightly greater than 30 m/h.

Based on the presented results, two facts could be pointed out: first, low bed permeability for the selected working conditions and selected properties of stainless steel fibers is most favorable for work; and second, the separation of oil droplets using stainless steel fibers is extremely sensitive to changes in oil properties, which is bad for practice. In petroleum and petrochemical industry, the bed coalescer is often a central part of wastewater treatment plant, and must operate with different types of oily contaminants. If the selected filter material is sensitive to the change of oil properties, it would mean that the coalescer cannot successfully respond to such a task.

**Dependence of the separation efficiency on the superficial velocity, properties of oil and bed geometry**

If the analysis of oil droplet separation using stainless steel fiber bed is executed only based on separation efficiency, limitations are not visible and one could make completely different conclusions. This will be illustrated by the following analysis. In resulting figures contour diagram is given of the dependence of separation efficiency, superficial velocity and bed permeability of oil A4, Figure 5.

![Figure 5](image)

Figure 5. Contour diagram representing the interdependence of separation efficiency, superficial velocity and bed permeability for oil A4.
other authors, who achieved separation efficiency from 40 to 50% using stainless steel fibers, it can be noted that the selection of appropriate fiber properties and bed properties in the presented research initiated incomparable greater separation efficiency of oil droplets. Regarding the influence of bed permeability, it is evident that with low permeability higher separation efficiency is achieved for a wide range of superficial velocity. The range of efficiency in these circumstances is from 99 to 97%. The size of the area where the separation efficiency is 99% varies for different oils, and it is the highest for oils A4, but very low for the other two investigated oils.

From the results of separation efficiency, any limitations cannot be established regarding working conditions and effluent quality, since the value of efficiency is very high, over 90%.

**Influence of oil properties on separation using stainless steel fibers bed**

It has already been pointed out that the size of working area, limited between superficial velocity and bed permeability with satisfactory effluent concentration of 15 mg/l, is influenced by the oil properties, which was perceived as a disadvantage. Figure 6 shows the 3D plot of the dependence of the effluent oil concentration, the oil viscosity and bed permeability at constant superficial velocity of 30 m/h. It is clear that the current superficial velocity is suitable for work in a wide range of oil viscosity and bed permeability, and that the areas that are unfavorable are with low oil viscosity and high value of bed permeability. Figure 7 presents the dependence of separation efficiency, oil viscosity and bed permeability, where it is also noteworthy that the lowest values of efficiency is achieved in this area, as well as low oil viscosity and high bed permeability.

![Figure 6. Three-dimensional diagram representing the interdependence of effluent oil concentration, oil viscosity and bed permeability for superficial velocity of 30 m/h.](image)

When using stainless steel fiber beds, it could be pointed out that the low permeability has a positive effect on the coalescence of droplets and subsequent separation efficiency. At low bed permeability, the bed is the most compressed provoking smallest pores, and then the superficial velocity is the highest. During the visual monitoring of the experiment it was observed that a significant amount of oil does not form a capillary-conducted phase behind the bed, as presented in Figure 8.

![Figure 8. Mineral oil after separation of emulsion using stainless steel fiber bed.](image)

Therefore, it can be argued that in the pores of stainless steel fiber beds predominantly the coalescence mechanism between the droplets is present that are under these conditions, the closest to each other. Previous studies of most authors found that increasing the viscosity of the dispersed oil increases separation efficiency [3,10,11,24]. This was also confirmed with a stainless steel fiber bed in the horizontal...
flow mode. It is thought that viscous oil formed in the pores is difficult to push out of the bed by hydrodynamic forces, and their presence in the pores facilitates the coalescence in their volume.

CONCLUSION

Based on successful selection of bed geometry for stainless steel fibers, high separation efficiency of tested mineral oils of different properties was reached. The efficiency for all operating conditions were higher than 90%. If analysis of the results is directed towards the reached effluent oil concentration, then it can be concluded that the stainless steel fiber bed can achieve an effluent concentration of 15 mg/l for all investigated oils, but at different values of superficial velocity. Superficial velocity of 30 m/h at low bed permeability provides the recommended quality effluent. Low permeability of stainless steel fiber bed and selected working conditions are the most favorable for the high-efficiency separation of mineral oil droplet from water. Separation of oil droplets using stainless steel fibers is extremely sensitive to changes of oil properties. This can lead to operation problems in industrial application, where the composition of influent is not well defined. It can be assumed that at low bed permeability the coalescence between droplets is the predominant mechanism.

Acknowledgment

The work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia, Grant number 172022.

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U ovom radu ispitivana je separacija kapi ulja iz vode primenom vlaknastog sloja od nerđajućeg čelika. Efikasnost separacije je praćena preko koncentracije uljne faze nakon sloja. Ispitivana je efikasnost separacije tri mineralna ulja koja imaju širok opseg viskoznosti od 10 do 170 mP s, kao i neutralizacionog broja do 0.10 do 1.70 mg KOH/l. Karakteristike sloja su varirane promenom nasipne gustine filtarskog materijala što rezultira promenom permeabilnosti sloja u opsegu od $0.7 \times 10^{-9}$ do $5.389 \times 10^{-9}$ m² i promenom poroznosti sloja od 91 do 98%. Svi eksperimenti su realizovani u širokom opsegu radne brzine od 10 do 50 m/h. Na osnovu realizovanih eksperimenata očita se da se postižu visoke vrednosti efikasnosti, preko 90%, primenom vlaknastog sloja od nerđajućeg čelika. Niska permeabilnost sloja je pogodnija za rad kod ovog filtarskog materijala. Međutim, vlaknasti sloj od nerđajućeg čelika veoma je osetljiv na promene prirode ulja koje se separiše, što može predstavljati ozbiljan nedostatak u primeni ovog materijala za kolescentu filtraciju, sa obzirom na činjenicu da je priroda ulja u realnim otpadnim vodama izrazito promenljiva.

Ključne reči: tečno-tečna separacija, zauljene vode, koalescencija u sloju, vlaknasti materijal, nerđajući čelik.