# Edible Oil Industry Wastewater Treatment by Microfiltration with Ceramic Membrane

Zita Šereš, Dragana Šoronja Simović, Ljubica Dokić, Lidietta Giorno, Biljana Pajin, Cecilia Hodur, Nikola Maravić

*Abstract*—Membrane technology is convenient for separation of suspended solids, colloids and high molecular weight materials that are present. The idea is that the waste stream from edible oil industry, after the separation of oil by using skimmers is subjected to microfiltration and the obtained permeate can be used again in the production process. The wastewater from edible oil industry was used for the microfiltration. For the microfiltration of this effluent a tubular membrane was used with a pore size of 200 nm at transmembrane pressure in range up to 3 bar and in range of flow rate up to 300 L/h. Box–Behnken design was selected for the experimental work and the responses considered were permeate flux and chemical oxygen demand (COD) reduction. The reduction of the permeate COD was in the range 40-60% according to the feed. The highest permeate flux achieved during the process of microfiltration was 160 L/m$^2$h.

*Keywords*—Ceramic membrane, edible oil, microfiltration, wastewater.

## I. Introduction

MEMBRANE separation is a filtration technique in which a feed stream is fractionized with a porous membrane. Some of the dissolved solids are held back because their molecular size is too large to allow them to pass through. The size range depends on the pore sizes of the used membrane. Fractionation of the feed stream occurs, with some molecules being concentrated on the upstream side of the membrane, which is known as the concentrate or retentate. The smaller molecules pass through the membrane into the permeate stream. The driving forces that cause mass transfer of solutes are usually difference in concentration, difference in electric potential and difference in pressure [1].

The emission of high organic effluent by the vegetable oil industry represents a serious threat to the ecosystem and fresh water resources. The OECD (The Organisation for Economic Co-operation and Development) reported vegetable oil production in Europe of 21,829,000.00 Mg per year in 2013 [2].

High organic load of the wastewater leads to lower COD removal using the hybrid biological reactor systems, and therefore a pre-treatment is required [3]. In such cases,

application of membrane technology proved to be a successful pre-treatment technique for both wastewater and drinking water [4]. However, the major hurdle in the extensive use of membranes is the continuous reduction of permeation flux caused by the concentration polarization and membrane fouling.

Concentration polarization causes deposition of retained compounds on the membrane surface. The pure water flux of micro- and ultrafiltration membranes is usually high, but when separation starts through the membrane, the permeate flux falls very quickly, which is caused by the gel formation on the membrane surface.

Membrane fouling is a sedimentation or accumulation of suspended or colloidal particles on the membrane surface, as well as crystallization, precipitation or adsorption of solute on the surface and / or in the pores of the membrane. There is no possibility for avoiding the membrane fouling, but it can be limited by applying a number of different techniques which enhance membrane flux. These techniques might be pre-treatment of feed stream, backflushing, fluidized bed, fluid instability, application of electric, magnetic and ultrasonic fields [1], [5].

Membrane technology has numerous advantages including high controlled and stable effluent quality, a compact construction providing the small area requirement. It is important to specify the absence of additional chemicals required for the membrane processes [6].

In the recent decades, several studies involving application of membrane separation in a wastewater treatment have been reported. However, most of these studies are related to the use of organic membranes in ultrafiltration and reverse osmosis in a wastewater treatment [7]-[10].

This paper will discuss the possibility of applying the new generation of ceramic membranes in order to reduce the COD of wastewater oil industry. The process would result in a permeate water with reduced COD and no turbidity, which can be recirculated into the process, and it should be possible to apply the retentate (concentrate) to saw dust, and thereby improve its energy value. During the production of edible oil, wastewater is produced in the amount of 10-25 m$^3$/t of the final product, with specific wastewater production of 3-5 m$^3$/t of raw material.

According to the regulation on limit values of emissions of industrial waste water, which is discharged into the public sewer, the limit, among other things, for the COD is 1000 mgO$_2$/L, biochemical oxygen demand (BOD) - 500 mg O$_2$/L [11].

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## II. EXPERIMENTAL

In this work, for the conducted experiments, an oil industry pooled effluent was used. The characteristics of this wastewater are: COD in the range of 5000-18000 mgO₂/L, turbidity in the range of 200-2500 NTU.

In order to remove coarse contaminants before the microfiltration, the wastewater was filtered through the cotton cloth. Installation for micro- and ultrafiltration, which is used in this paper, was designed at the Faculty of Technology in Novi Sad. Scheme of laboratory plant for micro- and ultrafiltration is shown in Fig. 1.
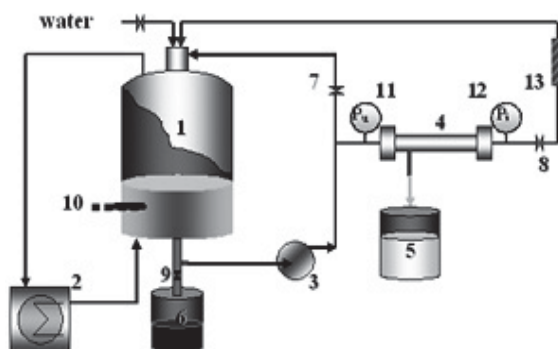


Fig. 1 Laboratory apparatus for cross-flow microfiltration: 1 – tank, 2 – thermostat, 3 – pump, 4 – membrane module, 5 –permeate tank, 6 – retentate tank, 7 – flow rate correction valve, 8 – pressure correction valve, 9 – retentate valve, 10 – thermometer, 11 – manometer, 12 – manometer, 13 – rotameter

For the purpose of this study, a ceramic membrane is used of the manufacturer GEA with pore opening size of 200 nm. Membrane is tubular shaped; length is 250 mm with an outside diameter of 10 mm and an internal diameter of 6.8 mm. The experiments of microfiltration of wastewater are planned according to the Box-Behnken's plan. Table I shows the varied values of independent parameters [12].

TABLE I
VARIED VALUES OF INPUT FACTORS AND DEPENDENT RESPONSES

| Input factors | Factor levels | | |
|---|---|---|---|
| | -1 | 0 | 1 |
| Transmembrane pressure (bar) | 1 | 2 | 3 |
| Feed flow rate (L/h) | 100 | 200 | 300 |
| Temperature (°C) | 20 | 50 | 60 |
| Dependent responses | | | |
| Permeate Flux (L/m²h) | | | |
| Turbidity (NTU) | | | |
| Chemical oxygen demand (mgO₂/L) | | | |

Dependent parameter which is constantly monitored during the microfiltration process is the permeate flux:

$$J = \frac{V_p}{A_m \cdot t} \tag{1}$$

where J represents permeate flux (L/m²h), $V_p$ volume of permeate (L), $A_m$ membrane surface (m²) and t time of microfiltration (h).

Physicochemical parameters, such as the COD and turbidity in the sample permeate and retentate were also measured. COD is determined by the titrimetric method SRPS ISO 6060 [13]. Turbidity is determined by the device Turb 550 IR. The measurements are performed automatically.

## III. RESULTS AND DISCUSSION

The obtained results are presented in Table II.

TABLE I
BOX–BEHNKEN EXPERIMENTAL DESIGN AND RESPONSES

| Run | Input factors[a] | | | Responses[b] | | |
|---|---|---|---|---|---|---|
| | TMP (bar) | Q (L/h) | T (°C) | J (L/(m²h)) | TU (NTU) | COD (mgO₂/g) |
| 1 | 2 | 100 | 20 | 29.23 | 0.3 | 1344 |
| 2 | 3 | 200 | 20 | 33.39 | 0.57 | 1420 |
| 3 | 1 | 200 | 20 | 35.48 | 0.89 | 1417 |
| 4 | 2 | 300 | 20 | 41.28 | 0.14 | 1574 |
| 5 | 1 | 200 | 60 | 3.35 | 1.27 | 1127 |
| 6 | 2 | 100 | 60 | 20.59 | 0.33 | 2030 |
| 7 | 3 | 200 | 60 | 45.66 | 1.11 | 2014 |
| 8 | 2 | 300 | 60 | 109.57 | 0.22 | 1483 |
| 9 | 1 | 100 | 50 | 28.91 | 0.28 | 1065 |
| 10 | 3 | 100 | 50 | 11.60 | 0.25 | 2112 |
| 11 | 3 | 300 | 50 | 144.54 | 0.85 | 1588 |
| 12 | 1 | 300 | 50 | 6.24 | 3.11 | 2170 |
| 13 | 2 | 200 | 50 | 41.31 | 1.67 | 1767 |

[a] TMP, transmembrane pressure; Q, feed flow rate; T, feed mixture temperature
[b] J, permeate flux; TU, permeate turbidity; COD, permeate chemical oxygen demand reduction
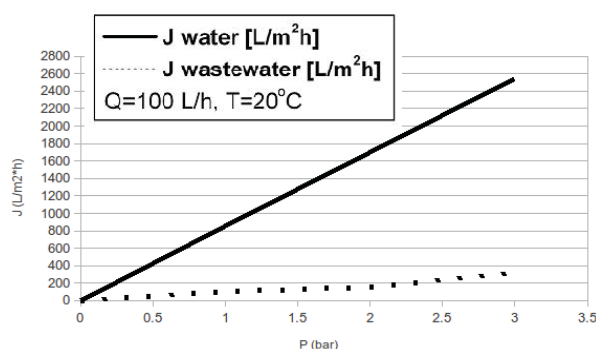


Fig. 2 Water flux and wastewater permeate flux, depending on the transmembrane pressure at microfiltration on ceramic membrane

Before examining the conditions of filtration of wastewater, it is necessary to determine the dependence of the flux of distilled water from the transmembrane pressure in order to be able to compare it to the results of permeate flux of wastewater. In Fig. 2, the dependence of the flux of distilled water and the permeate flux of wastewater are presented. The flux of water is the basis for comparison, because it plays the role of the solvent in wastewater. It shows that the flux of distilled water is from 5 to 8 times greater than the flux of permeate, indicating the additional resistance to the flow through the membrane pores during the microfiltration of wastewater.

On the basis of experimental values, graph of the two dependencies is drawn and by the program Statistica 12 the values of such get the equation that best describes the function of the flux dependence on pressure and flow. Fig. 3 presents the results obtained at the medium transmembrane pressure (2bar), the highest flow rate (300 L/h) and the medium tested wastewater temperature (50°C).
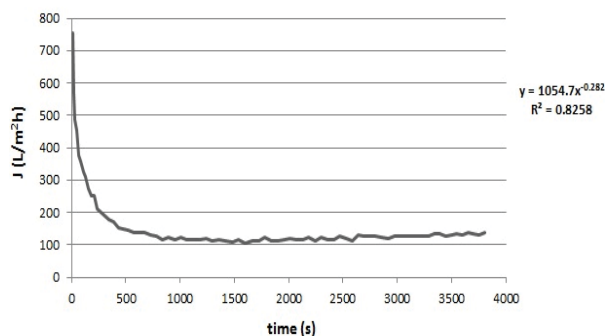


$y = 1054.7x^{-0.282}$
$R^2 = 0.8258$

Fig. 3 Wastewater permeate flux, depending on the time of microfiltration obtained at the highest feed flow rate

The highest values of flux, above 160 L/m²h, are obtained when the flow is maintained above 200 L/h and the pressure tested at medium values (above 2 bar). However, the further rise of the transmembrane pressure has not contributed to the significant rise of the permeate flux values. Due to the characteristics of a cake formed on the membrane surface, increasing pressure only changed the formed cake structure (reduced the thickness of the cake) and did not improve the accessibility of membrane pores to the water molecules. Nevertheless, the increase of the feed flow rate had a positive influence on the rise of permeate flux values. It is assumed that higher flow rates impact the removal of the particles from the upper layers of the cake, allowing the water molecules to pass through the membrane pores.

Fig. 4 presents the results obtained at the highest transmembrane pressure (3 bar), the lowest flow rate (100 L/h) and the medium tested wastewater temperature (50°C). It can be noticed that the permeate flux values obtained at the lowest feed flow rate are significantly lower during the microfiltration of wastewater.
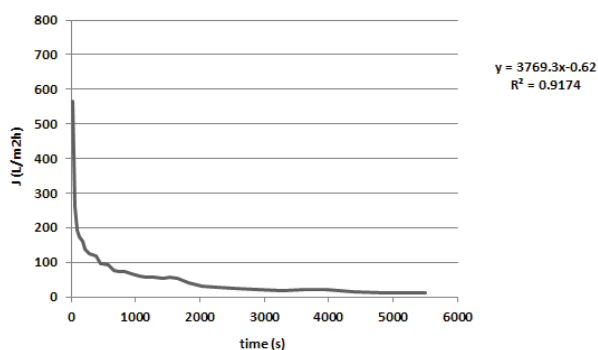


$y = 3769.3x-0.62$
$R^2 = 0.9174$

Fig. 4 Wastewater permeate flux, depending on the time of microfiltration obtained at the lowest feed flow rate

Fig. 5 gives the values of COD, and turbidity (turbidity) of wastewater before and after the microfiltration in permeate and retentate.

Fig. 5 shows that when using membranes of 200 nm, the COD is decreased by 85% and by 99% for turbidity.

Removal of the COD and turbidity from wastewater after the microfiltration is presented in Fig. 5. In this experiment, the obtained turbidity decrease was in the range of 50-99%. Ceramic membrane with 200nm pore size showed promising results, eliminating more than 80% of organic compounds responsible for the COD load. Since the permeate is clear of suspended solids and organic compounds, it could be expected that total amount of this particles had moved from the wastewater to retentate. However, a certain amount of particles from wastewater form a cake layer on the membrane surface and represent a membrane fouling effect. The cake layer represents an additional porous filter which decreases the original pore diameter of the membrane and enhances the microfiltration effect [14].
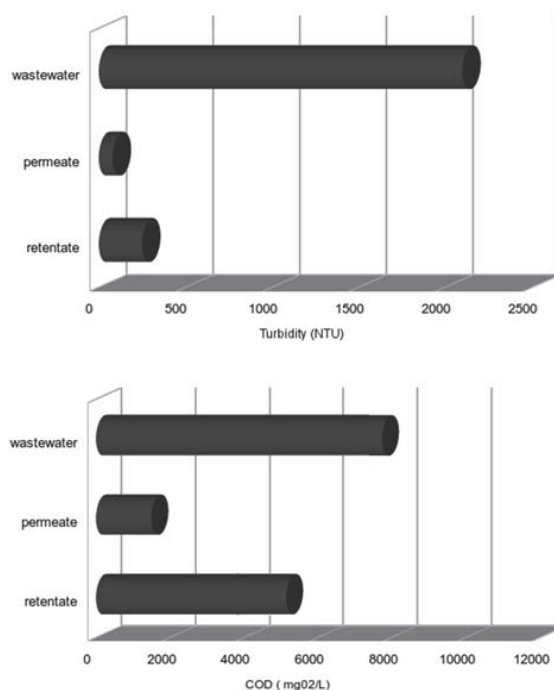


Fig. 5 COD and turbidity before microfiltration in wastewater and after microfiltration in permeate - purified wastewater, and retentate

During the experimental runs, it has been noticed that the lowest COD removal was achieved at the lowest wastewater temperature. It can be assumed that further increase of wastewater temperature stimulates the enhanced formation of aggregates of the present organics and chemical reactions between the wastewater components, which resulted in compounds of larger molecular masses, making them easier to separate from the wastewater by using the applied membrane pore size. This positive effect is highly prominent when comparing the COD removal after the microfiltration at 20 °C

(under 40 %) and the COD removal after microfiltration on 60 °C (over 70%).

## IV. CONCLUSION

Based on the effects of microfiltration of wastewater at a transmembrane pressure in the range of 1-3 bar, flow rate of 100 to 300 L/h and temperatures of 20-60°C, it can be concluded that the microfiltration can reduce the COD of wastewater, i.e. contamination of wastewater:

- Permeate flux wastewater (95 L/m$^2$h) at a transmembrane pressure of 1 bar is 8 times smaller than the flux of water, and the trend retains on the rising pressure. This leads to the formation of resistance during the microfiltration of wastewater on the surface membrane from the compounds present in the wastewater.
- At pressures above 2 bars, the permeate flux of 160 L/m$^2$h can be achieved.
- Using the membranes from 200 nm it is possible to reduce the COD by 85%, while the turbidity of the wastewater can be removed up to 99%.

## ACKNOWLEDGMENTS

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