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Treatment of vegetable oil refinery wastewater using alumina ceramic membrane: optimization using response surface methodology

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ABSTRACT

New regulations in environment protection and increasing market demands for “green” companies are forcing the industry to consider finding new and sustainable methods of wastewater treatment. The valuable information regarding treatment of the wastewater from the edible oil industry is presented in this research. The obtained results confirm a promising application of the third generation membranes, comprised of ceramic material (aluminium oxide), in treatment of wastewater from the edible oil industry. Positive results regarding chemical oxygen demand reduction and turbidity removal were noticed. Permeate flux values were used for process optimization in order to achieve a cost-effective process. Response surface methodology was used for the experimental design. The effects of wastewater temperature, transmembrane pressure, and feed-flow rate on the microfiltration model fit were studied. The experiments showed that microfiltration of this type of wastewater is a convenient technique for a possible large scale industrial application as a secondary step in treating wastewater from the oilseed processing facilities.

1. Introduction

The main task of wastewater treatment is to eliminate pollution to the extent that the treated wastewater can be discharged into the recipient without consequences or that it can be re-used (Kralj, 2015). The technological process of wastewater treatment can consist of many processing stages, depending on the characteristics of the raw wastewater and the required quality of the treated wastewater, and for each of these stages there are several options. A sufficient knowledge of the amount and degree of wastewater contamination can help us to design a simple and efficient plant for wastewater treatment (Joss et al., 2005; Kastelan-Macan et al., 2007). Numerous reports regarding the occurrence of non-regulated water contaminants, such as the emerging organic contaminants, have expressed concern about their possible undesirable effects in the environment (Bolong et al., 2009; Daughton, 2004; Daughton and Ternes, 1999). Satyawali and Balakrishnan (2008) concluded that physicochemical methods, like membrane filtration, are capable of organic load reduction and are therefore being widely field-tested.

According to the *OECD (The Organisation for Economic Co-operation and Development) report from 2013* vegetable oil production in Europe reaches 21,829,000.00 Mg per year. Therefore, the emission of poor quality effluents by the oilseed processing facilities is posing a dangerous threat to the fresh water resources (Saatci et al., 2001). The oilseed is usually processed in five stages: seed receiving and storage, seed preparation, solvent extraction unit, refining of crude oil and packaging of oil (Nucci et al., 2014). However, the harmful effluent is mostly discharged from degumming, deacidification and deodorisation stages, although the wastewater content and quality may significantly differ from one edible oil industry to another (Kale et al., 1999; Boyer, 1996).

Flocculation, coagulation, air floation, oil-skimming and biological processes (anaerobic and aerobic digestion) are typical techniques normally used for edible oil wastewater treatment (Pathe et al., 2000). However, most of these techniques separate only a part of undesired components or require an appropriate pretreatment of the wastewater.

Directive 91/271/EEC on urban wastewater treatment and Directive 96/61/EC on Integrated Pollution Prevention Control

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illustrate the current and future EU policy to encourage development of processes and standards to prevent negative effects on water, using the best available techniques (Schröder et al., 2007). The methods of wastewater treatment are numerous. That is illustrated in the *Best Available Techniques (BAT)* for the protection of the environment in the food industry and in Best Available Techniques in the treatment of wastewater and waste gases in the chemical industry, issued by the *Commission of the European Union*. One of the options recommended by these documents is the application of membrane separation techniques.

Membrane technology offers numerous advantages over traditional separation techniques providing higher efficiency, economical savings and faster processes. New membrane techniques are convenient for separation of suspended solids, colloids and high molecular weight materials that are present in treated wastewater (Cassini et al., 2010).

The driving force is the pressure difference which enables a continuous separation process (Noble and Stern, 1995). The idea is to microfilter the wastewater, after the separation of oil by the skimmer, where the permeate passes through the membrane and can be recycled and re-used in the vegetable oil refining process. Recent investigations showed promising results and possible large scale application (*Buecker, 2007*).

The main goal of this research was to investigate the possibility of microfiltration of wastewater from vegetable oil refinery through application of the new generation of ceramic membranes to reduce the chemical oxygen demand (COD) of edible oil refinery wastewater.

Wastewater was introduced into the laboratory plant for microfiltration treatment under different conditions of flow, transmembrane pressure (TMP) and feed temperature values. Besides COD, permeate flux was measured in order to obtain optimal filtration conditions. Statistical analysis of the data was conducted in order to obtain an appropriate mathematical model of the process. Finally, the model was analyzed and the influence of different factors on the permeate flux and COD was discussed.

2. Materials and methods

Treated wastewater was sampled from oilseed processing plants “Victoriaoil”, “Victoria group”, Serbia. Characteristics of this wastewater are: chemical oxygen demand in the range of 5000–18,000 mg O_2/L, turbidity in the range of 200–2500 NTU. Our research included pre-treatment of wastewater by filtration process through the filter cloth with pore size of 1 mm in order to separate larger particles that may interfere the microfiltration process.

The starting volume of feed mixtures for the cross-flow microfiltration was circa 3 L, which is optimal for such a laboratory plant (Fig. 1), given that this is the sufficient volume of feed mixture needed to fill the pipe system and apparatus to enable the undisturbed running of the pump. Batch cross-flow microfiltration runs were performed during 90 min, with permeate collected in vessel 5 and retentate in vessel 6.

A ceramic tubular membrane with a pore size of 200 nm, produced by GEA Westfalia, Germany, was used for a cross-flow microfiltration, under the transmembrane pressure in the range of 1–3 bar, temperature range from 40 to 60 °C and a flow rate of 100–300 L/h. During microfiltration, the permeate flux *J* was monitored, followed by the chemical oxygen demand and turbidity of wastewater (as a feed mixture), permeate and retentate. Chemical oxygen demand was determined by the titrimetric method JUS ISO 6060, Official Gazette of FRY, no. 45/94 (1994). Turbidity was determined by the device Turb 550 IR. The measurements were performed automatically.

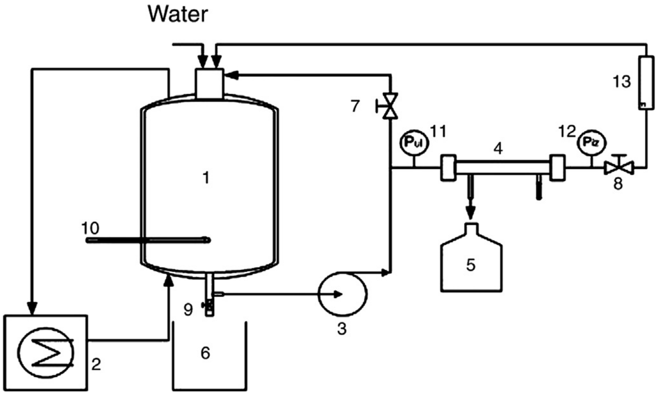

Fig. 1. Laboratory apparatus for cross-flow microfiltration (1 – feed tank, 2 – thermostat, 3 – rotary pump, 4 – module with membrane, 5 – vessel for permeate, 6 – vessel for retentate, 7, 8, 9 – valves, 10 – thermometer, 11, 12 – manometer, 13 – rotameter).

The RSM was applied to evaluate the effects of microfiltration parameters and optimize conditions for various responses. Box–Behnken experimental design (BBD) with three numeric factors on three different levels was used (Myers and Montgomery, 1995). Design included 13 randomized runs with one replicate at the central point. In Table 1 the experimental design is given.

Response surface methodology (RSM) is a statistical method of multifactorial analysis of experimental data which provides a better understanding of the process than the standard methods of experimentation, since it is able to predict how the inputs affect the outputs in a complex process where different factors can interact among themselves. All the coefficients of the different polynomial equations were tested for significance with an analysis of variance (ANOVA) (Martí-Calatayud et al., 2010).

In such work, the utility of statistical multifactorial analysis of experimental data for understanding the effects of different factors and their interactions in the process were confirmed.

For responses obtained after the experiments, a polynomial model of the second degree is established to evaluate and quantify the influence of the variables:

$$Y = b_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_{11}x_1^2 + b_{22}x_2^2 + b_{33}x_3^2 + b_{12}x_1x_2 + b_{13}x_1x_3 + b_{23}x_2x_3, \tag{1}$$

where Y is the predicted response, b_0 is an intercept; x_1, x_2 and x_3 are the coded levels of input factors; b_1 to b_{33} are regression coefficients; x_1x_2, x_1x_3 and x_2x_3 represent interactions of input factors, while x_1^2, x_2^2 and x_3^2 represent quadratic terms (Montgomery, 2001). The adequacy of the model was evaluated by the coefficient of determination (R^2) and model p-value. Statistical analysis was

Table 1
Variables and levels in the Box–Behnken experimental design.

	Factor levels		
	−1	0	1
Input factors			
Transmembrane pressure (bar)	1	2	3
Feed flow rate (L/h)	100	200	300
Temperature (°C)	20	40	60
Dependent responses			
Permeate flux (L/(m^2 h))			
Chemical oxygen demand reduction (%)			