RESEARCH ARTICLE

Separation of water-oil emulsions with a dynamic membrane of gelatin on a nylon substrate

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Abstract

Introduction: According to the study of water-oil emulsion, chemical composition, and properties of oil-in-water type, they proposed the method of membrane separation for emulsion petroleum products (PPs). To increase the degree of PP separation from a water-oila emulsion, they offered the method of a dynamic layer application to the surface of a microfiltration membrane. **Research Tasks:** Nylon-gelatin dynamic membranes with a different content of the gelatin layer 1.6–6.3% (by weight) were obtained on the membrane surface by the filtration of colloidal gelatin solution through the nylon membrane with the particle sizes of the dispersed phase from 13 to 4153 nm. **Methods:** Using the wetting surface droplet method, it is determined that the initial nylon and the modified nylon-gelatin membrane have the properties of a hydrophilic surface, which effectively separates oil-in-water emulsions. **Results:** It was found that the specific productivity of the modified membrane decreased by 4.5 times after the application of a dynamic layer of gelatin. It was determined that the degree of PP separation from an emulsion with an initial nylon membrane makes no more than 46.7%. **Conclusion:** Moreover, after the modification, the degree of separation increased to 51%, depending on the amount of gelatin containing on the membrane surface. The maximum degree of WOE PP separation makes 97.4% with the gelatin content of 6.3% (by weight).

Key words: Cleaning, dynamic membranes, gelatin, nylon, petroleum products, specific productivity, ultrafiltration

INTRODUCTION

Currently, one of the most urgent problems is the increasing pollution of the world oceans with oil and the products of its processing. The main reason for the high level of fresh and salty waters is the discharge of sewage containing petroleum products (PPs) by the enterprises of various industries.

Despite the improvement of oil production and processing technologies, the type of its storage and transportation, the availability of a variety of technological schemes for cleaning aquatic environments from PP, the level of pollution by the latter remains quite high in general.

The removal of emulsified PP is a difficult task, which is associated with the need to destroy a stable structure of the developed emulsions, formed mainly as a result of lubricating cooling liquids (LCL) and detergent solutions application in engineering. Waste emulsion cleaning methods used in plants and local treatment facilities, such as flotation, sedimentation, coagulation, and filtration, do not provide the reduction of the main pollutant concentration to the established requirements which allow the use of purified water for the repreparation of emulsions, or for other industries, which leads to a significant pollution of environmental objects.

Membrane methods,[1-6] which have a high degree of retention and low energy costs in comparison with evaporation and vacuum distillation methods, are often used in the cleaning and the recycling of used emulsions. It is known that the initial LCL is a multicomponent emulsified system that contains mineral and synthetic oils, emulsifiers, corrosion inhibitors, biocides, and other components. In this regard, the

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separation of spent LCL allows the production of valuable components for subsequent processing and further use.

Hydrophobic membranes are used for membrane separation of inverse emulsions; hydrophilic membranes are used for direct emulsions, there are some attempts to change this rule.[7] They use various polymers, ceramics, and porous glass as the materials for membranes. They use flat, hollow fiber, and tubular membranes. To separate the emulsions, various membrane processes were used: Pervaporation,[8] membrane distillation,[9] reverse osmosis,[10] nanofiltration,[10,11] and microfiltration.[12-14] Ultrafiltration became the most popular one.[3,12,13]

Dynamic membranes have high porosity due to the layer of deposited material particles. Thus, water permeability is achieved, which becomes one or two orders higher than the permeability of polymeric membranes. Colloidal particles, neutral organic polymers, and organic and inorganic polyelectrolytes are suitable for the development of dynamic membranes. Dynamic membranes are obtained on the porous substrates of microfiltration, ultrafiltration, and reverse osmosis membranes with a pore size from 3–5 μm on a wide variety of materials (porous metals, ceramics, and polymer films).[14]

Semipermeable dynamic membranes from iron hydroxide compounds and polyvinyl alcohol were obtained in Rudepko *et al*.[15] for ultrafiltration purification of groundwater from radionuclides.

METHODS

In this paper, we describe the method for obtaining dynamic nylon-gelatin membranes to separate waste water-oil emulsions formed after the activity of petrochemical, oil-producing, and machine-building industries.

The microfiltration membrane made of nylon with an average pore size of 0.45 μm and the diameter of 47 mm was used as an initial substrate on which a dynamic layer was applied. The gelatin powder was dissolved in distilled water, heated to the temperature of 50°C, and mixed until a uniform consistency was obtained. The gelatin concentration was 2.5 g/dm^3. To form a dynamic layer of gelatin, the solution of gelatin with the volume of 50–200 ml was filtered through the membrane at the pressure of 100 KPa. The content of gelatin in the dynamic membrane was determined by the gravimetric method by membrane mass before and after the modification.

The particle size of the dispersed phase was determined by the method of dynamic light scattering, and ζ potential was determined by light scattering method with phase analysis using the "NanoBrook Omni" analyzer.

The study of the membrane sample wettability was carried out using "Kruss DSA 20E" apparatus, which makes it possible to determine the contact angle with a drop of distilled water on the membrane surface.

The specific productivity and the degree of water-oil emulsion separation, which was calculated as the ratio of PPs content in the emulsion before and after the separation, determined by the use of KH-3 concentrate meter, were considered as the main parameters of the emulsion membrane separation.

About 0.1% LCL emulsion of Inkam-1 grade was used as the model WOE for membrane separation. During the separation of distilled water and emulsions, the working pressure was applied from 0.1 to 1 MPa, the temperature of the liquid was 22°C.

RESULTS AND DISCUSSION

Figure 1 shows the distribution of the particle size during the disperse phase in gelatin solution.

Table 1 summarizes the gelatin particle size ranges in the solution and the zeta potential of the dispersed phase.

The particle size of the colloidal gelatin solution dispersed phase is in the range from 13.1 to 4153 nm, the ζ-potential of the dispersed phase has a negative sign.

Figure 2 shows the results of the studied membrane sample wettability measuring and the calculated values of the wetting contact angle.

According to the data presented in Figure 2a and b, it can be seen that the liquid forms meniscus drops on the surface of the initial and modified membrane. The area of liquid contact with the surface decreases. The droplets with a contact angle of no more than 45°C are formed on the surface of these membranes, which indicates the hydrophilicity of the membranes. It has been found that the application of a gelatin layer to the surface of a nylon membrane contributes to an insignificant decrease of wetting contact angle from 44.5°C to 42.3°C, which indicates the hydrophilicity increase of the nylon-gelatin membrane.

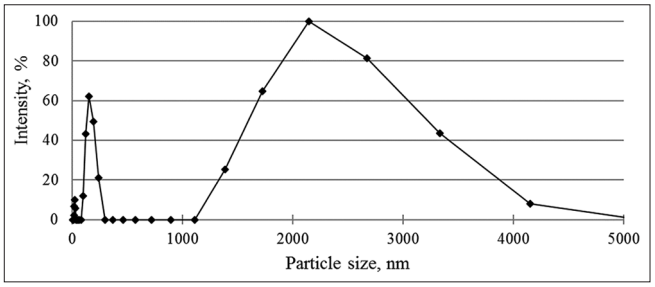

Figure 1: The graph of the particle size distribution during the disperse phase in the gelatin solution

After gelatin modification, the specific productivity by distilled water and by 0.1% water-oil emulsion was studied [Table 2].

After the application of the gelatin layer on the surface of the nylon membrane, the specific productivity of the latter decreases from 1.4 to 4.5 times, depending on the volume of the colloidal solution passed through an original membrane. The decrease of membrane specific productivity is conditioned by the accumulation of gelatin particles in the pores and on membrane surface, and thus, the average pore size of a membrane is reduced. The content of gelatin in a dynamic membrane makes 1.6–6.3%, depending on the volume of the gelatin solution used. The maximum productivity of the initial and dynamic membranes is observed during the filtration of distilled water.

To restore the initial productivity of the membranes after the filtering of 500 cm^3 of emulsion, the membranes were washed by backwashing with a 5% solution of sodium dodecyl sulfate, and then, they were rinsed with distilled water.

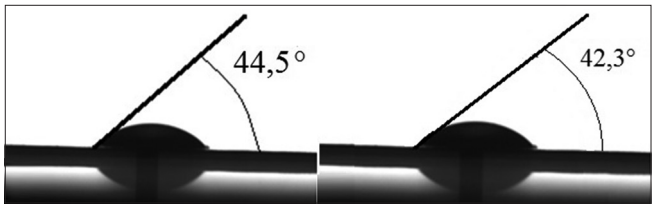

Figure 2: The images of membrane wettability with a drop of distilled water: (a) The original from nylon; (b) the dynamic nylon-gelatin

Table 1: Particle sizes and zeta potential of colloidal gelatin solution during the disperse phase

Colloidal gelatin	Particle size, nm	ζ-potential, Mv
Gelatin 2.5 g/dm^3	13.1–26.5; 99.2–239; 1384–4153;	−16±0.16

The results of the separation of WOE 0.1% separation from PP on the laboratory membrane plant are shown in Table 3.

According to Table 3 data, it is obvious that the efficiency of PP separation from the emulsion during an original nylon membrane use is significantly lower, as compared to the dynamic nylon-gelatin membranes. The purification degree from PP is increased with the increase of gelatin content on the surface of membranes. After the application of the dynamic gelatin layer, the efficiency of WOE separation into an aqueous and an organic phase increased by 41–51% as compared to the initial membrane.

CONCLUSIONS

By filtering through a microfiltration membrane of nylon colloidal gelatin solution with the particle sizes of the dispersed phase from 13 to 4153 nm, dynamic nylon-gelatin membranes are obtained with a different content of gelatin layer from 1.6% to 6.3% by weight on the membrane surface. Using the surface wetting droplet method, it is determined that the initial nylon and the modified nylon-gelatin membrane have the properties of a hydrophilic surface, which effectively separate oil-in-water emulsions. Furthermore, they determined the contact angle decrease between the drops of distilled water of the membrane from 44.5° to 42.3° after the application of gelatin dynamic layer. The application of a dynamic gelatin layer to the surface of an initial membrane results in specific productivity decrease of the latter by 1.4–4.5 times, depending on the volume of the colloidal solution passed through an initial membrane, by accumulating gelatin particles in the pores and on the membrane surface. It was found that the degree of PP

Table 2: Specific productivity of the initial and dynamic membrane "nylon-gelatin"

Membrane name	Gelatin content, % (by weight)	Specific productivity of membranes, $cm^3/cm^2 \cdot min$	
		By distilled water	By 0.1% of WOE
Nylon	-	9.3	1.18
Nylon-gelatin 50	1.6	6.5	0.54
Nylon-gelatin 100	2.7	4.0	0.35
Nylon-gelatin 200	6.3	2.1	0.12

Table 3: Oil product separation degree from 0.1% of WOE

Membrane name	Gelatin content, % (by weight)	Oil product concentration (mg/dm^3)		Purification degree (%)
		original	After purification	
Nylon	-	891±89.1	475±47.5	46.7
Nylon-gelatin 50	1.6		114±11.4	87.2
Nylon-gelatin 100	2.7		65.8±13.6	92.6
Nylon-gelatin 200	6.3		23.2±4.64	97.4

separation from WOE with an initial nylon membrane makes no more than 46.7%. Moreover, after the modification of the membrane by a gelatin layer application, the degree of separation increased by 41–51%, depending on the amount of gelatin containing on the membrane surface. The maximum degree of PP separation from WOE makes 97.4% with the gelatin content of 6.3% (by weight).

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