

# Removal of bisphenol A from water by micellar enhanced ultrafiltration with nonionic-cationic surfactant mixtures

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#### ABSTRACT

The removal of bisphenol A (BPA) from water with micellar enhanced ultrafiltration (MEUF) by using a mixture of nonionic oxyethylated octylphenol TX100 and cationic hexadecylpyridinium bromide (HDPBr) surfactants was studied. It was shown that formation of mixed micelles promotes the BPA solubilization in the hydrophobic nucleus of the micelles and provide higher BPA removal from water compared to when the single surfactants were used for MEUF process. At a total surfactant concentration of 0.24 mM, the BPA rejection values were 12%, 86% and 95%, when using HDPBr, TX100 and TX100-HDPBr mixture, respectively. The higher BPA removal with the surfactants mixture is related to a larger size of the mixed surfactant micelles. It was found that the permeate flux was also notably higher when using the surfactants mixtures obviously due to formation of a more porous mixed micellar layer of lower hydrodynamic resistance on the membrane surface compared to the micellar layers for the singe surfactants. It was shown that the mixed TX100/HDPBr micelles are formed at lower critical micelle concentration values compared to ones for individual surfactants that reduce a total amount of the surfactants needed for MEUF process. Thus, employing of the surfactants mixture could improve both efficiency and cost-effectiveness of MEUF process of BPA removal from water.

*Keywords:* Micellar enhanced ultrafiltration; Bisphenol A; TX100; Hexadecylpyridinium bromide; Surfactants mixture; Water treatment

# 1. Introduction

Recently, a variety of chemicals that are extremely harmful to the human endocrine system, the so-called "endocrine disruptors", which include bisphenol A (BPA), are increasingly found in natural waters [1,2]. BPA is synthesized on a large scale with the yearly production exceeding 10 million tons [3]. BPA is widely employed for fabrication of polycarbonates, technical paper, herbicides, tanning agents and dyes [4,5]. Personal care products such as shampoo, lotions, soaps, laundry detergents and bowl cleaners also contain different amounts of BPA [6]. Given a broad range of applications, the BPA was found in ground water, soil and sediment samples [7] and even food [8,9]. BPA was spotted in urine in majority of population in different countries, indicating a typical exposure to BPA over the globe [10,11].

Because of similarity of chemical structure with estrogen, BPA, when entering into the human body, interacts with different physiological receptors and can negatively affect endocrine, immune, metabolic and cardiovascular systems [12–14]. As per the Environmental Protection Agency in USA the maximal allowable intake is 50  $\mu$ g of BPA per kg weight per day [15].

BPA has been found in effluents in waste water treatment plants (WWTP) [16] because of incomplete removal by common chemical and biological methods of water treatment [17]. The BPA removal efficiency in WWTP

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was reported from 37% to 94% [17,18]. The BPA content in WWTP effluents can vary from few to 370  $\mu$ g/L [19]. Given that the BPA still possess estrogenic activity at a very low concentrations less than 1  $\mu$ g/L [8], the employing of advanced water treatment is usually required to eliminate the BPA estrogen activity.

Different water treatment methods have been suggested for BPA removal from water including adsorption [15,20– 24], advanced oxidation [25,26] and membrane processes [27–29]. However, some common water treatment processes such as adsorption with activated carbon is lacking high removal efficiency due to low hydrophobicity of BPA [30], while formation of toxic by-products is a main concern for employing of oxidation methods of BPA removal from water [25].

As to the membrane methods, Yüksel et al. [27] used several nanofiltration (NF) and reverse osmosis (RO) membranes for BPA elimination from water. The authors reported high rejection of BPA by using polyamide RO membranes (XLE BWRO, BW-30 and AD SWRO types), while the cellulose acetate RO membrane CE BWRO showed a rather low BPA rejection of 10%–40%. The NF membranes (NF-270 and NF-90) provided BPA rejection of 80% and 98%, respectively [27]. Nghiem and Hawkes [31] reported 40% and 90% of BPA rejection by using NF-270 and NF-90 membranes in treatment of 0.75 mg/L feed solution, while Comerton et al. [32] showed only 4.6% removal of BPA with NF-270 membranes in 2 mg/L BPA solution.

It should be mentioned that RO membranes typically posses low permeate fluxes and need to be operated at high operating pressures. Hence, a large amount of energy is required to run the RO process [33]. On the other hand ultrafiltration (UF) membranes have higher permeate fluxes compared to RO and NF membranes and can be operated at lower pressures. However, UF membranes usually poorly remove low molecular weight organic pollutants such as BPA because of large pore sizes of UF membranes compared to a size of the pollutant molecules. Schäfer et al. [34] reported only 30%–45% of BPA rejection from grey water at BPA concentration of 1 mg/L when employing a "ZeeWeeed-1 UF module" with 0.04  $\mu$ m pore size membrane. The BPA removal was mainly related to BPA adsorption by the membrane matrix and by suspended solids.

Over the last decades the micellar enhanced ultrafiltration (MEUF), which can be run at mild operating conditions, has been suggested as an alternative approach to RO and NF processes for removal of low molecular weight organic pollutants and heavy metals from water [35-37]. MEUF process is based on spontaneous aggregation of the surfactant molecules and formation of the surfactant micelles in aqueous solutions when the surfactants concentration is above a critical concentration of micelle formation (CMC). The hydrophobic nucleus of the surfactants micelle is able to solubilize molecules of nonpolar hydrophobic organic substances from water [37]. Given a micellar size for different surfactants is in the range of 4-11 nm, the formed micelles can be efficiently removed from an aqueous solution by using high flux UF membranes [36,37]. The main features of MEUF include simple operation and notably lower energy consumption compared to RO and NF processes [35,37]. Over the last decades it was shown that MEUF might be employed for elimination of a variety of organic pollutants from water [34-37], however only a single paper related to BPA removal by MEUF was reported in the literature [38]. Urbanski et al. [38] used dodecyl sulfate (SDS), hexadecyltrimethyl ammonium bromide (CTAB) and alkyl polyglucoside Glucopon<sup>®</sup> (APG) to remove BPA from water with MEUF by employing the single solutions of the surfactants. It was shown that at optimal conditions SDS, CTAB and APG surfactants provided BPA rejection within the range of 89%-96%, 97%-99% and 60%-83%, respectively, however the surfactants dosage required was as high as 10 CMC that is 9.7, 0.92 and 1.1 mM, respectively. It should be mentioned that rather high concentration of surfactants to be added to the treated solution is one of the issues that impedes practical application of MEUF is water treatment applications.

Recently a so-called synergistic effect in the surfactants mixtures, when the formation of mixed surfactant micelles occurs at much lower concentrations than the formation of micelles of single surfactants in aqueous solutions, was reported [39–43]. These findings might be of interest for MEUF as a surfactant concentration in MEUF process should be as low as possible both from ecological and economical perspectives. The main objectives of this study is to evaluate the BPA removal from water with MEUF by employing a mixture of nonionic oxyethylated octylphenol TX100 and cationic hexadecylpyridinium bromide (HDPBr) surfactants. To the best of our knowledge, TX100, HDPBr surfactants as well as their mixtures were not studied yet for BPA removal from water by MEUF.

#### 2. Materials and methods

Bisphenol A (BPA) – the 2,2-(4,4'-dihydroxydiphenyl) propane with molecular weight of 228.29 g/mol, cationic HDPBr and nonionic oxyethylated octylphenol TX100 surfactants (Sigma-Aldrich, USA), which are shown in Fig. 1 were used in the study. Deionized water (Milli-Q Plus, Millipore) was used for preparation of the surfactants and BPA solutions.

The micelle formation in the single and mixed surfactant solutions was studied by the tensiometric method. The surface tension ( $\sigma$ ) values were evaluated by Wilhelm's approach [44] by using a BT-500 tensiometer (Analytprylad, Ukraine). Three surface tension measurements were recordered and the average  $\sigma$  value was reported.

The values of critical micelle concentration (CMC) were evaluated by using surface tension ( $\sigma$ ) data at different surfactant concentrations (*C*). The CMC corresponds to the surfactant concentration at which a break point happen at  $\sigma$  vs. ln*C* curve.

TX100 mole fraction in the binary surfactants mixture was evaluated as:

$$\alpha_{\rm TX100} = \frac{C_{\rm TX100}}{C_{\rm HDPBr} + C_{\rm TX100}}$$
(1)

where  $\alpha_{_{TX100}}$  is a mole fraction of TX100 in the mixture, while  $C_{_{TX100}}$  and  $C_{_{HDPBr}}$  are TX100 and HDPB concentrations in a mixture, mM.



Fig. 1. Structural formulas of TX100 (a), HDPBr (b), and BPA (c).

Mole fraction of HDPBr in the binary surfactants mixture was evaluated as:

$$a_{\rm HDPBr} = \frac{C_{\rm HDPBr}}{C_{\rm TX100} + C_{\rm HDPBr}}$$
(2)

where  $\alpha_{\text{HDPBr}}$  is a mole fraction of HDPBr in the mixture, while  $C_{\text{TX100}}$  and  $C_{\text{HDPBr}}$  are TX100 and HDPB concentrations in a mixture, mM.

The binary surfactant mixtures at different mole fractions were prepared by mixing the single surfactant solutions of the same molar concentration.

The surfactants concentration used in this study were in the range 0.1–1.3 mM.

The pH of the solutions was measured with a HQ40S pH meter (Hach, USA). pH values of the solutions were adjusted by using 0.1 M HCl and NaOH.

For MEUF experiments, an aqueous solution of the single surfactants or their mixture were added to 0.5–5 mg/L feed BPA solution, stirred with a magnetic stirrer for 30 min and after that filtered through a flat disk 10 kDa UP010 polyethersulfone (PES) UF membrane (Mann+Hummel GmbH, Germany) by using a 200 mL Amicon 8200 membrane cell (Millipore, USA) with the membrane surface area of 28.7 cm<sup>2</sup>. The operating pressure of 0.2–5 bar in the cell was created by compressed nitrogen. 200 mL of feed solution was placed in the membrane cell and the permeate recovery was maintained at 20%. The solution in the cell was stirred at a speed of 200 rpm. MEUF experiments were performed at room temperature.

The membrane permeate flux was evaluated as:

$$J = \frac{V}{A \cdot t} \tag{3}$$

where *V* is the permeate volume (L), which was filtrated through the membrane area A (m<sup>2</sup>) during filtration time *t* (h).

The BPA concentration in water samples was analysed by a liquid chromatograph Dionex (Chromeleon 6.1), Column C18 Luna (Phenomenex). A mixture of  $CH_3CN/H_2O$  (35:65 vol.%) was employed for elution at a pumping speed of 1.0 cm<sup>3</sup>/min.

The efficiency of the pollutant's removal from water was calculated as BPA rejection R (%):

$$R(\%) = \left(1 - \frac{c_p}{C_f}\right) \times 100 \tag{4}$$

where  $C_p$  and  $C_f$  represent the BPA contents in the permeate and feed, respectively.

A UV310 spectrophotometer (Shimadzu, Japan) was used for analysis of TX100 concentration in water samples by measuring a solution absorbance at 274 nm. HDPB concentration was evaluated by titration with sodium tetraphenylborate in the presence of tetrabromophenolphthalein ethyl ester [45]. The screening experiments performed showed that each single surfactant in the TX100/ HDPB mixtures did not disturb the analytical quantification of other constituent in the surfactant mixture.

The surfactants rejection in MEUF process was evaluated from Eq. (4) by using the surfactants concentrations in a permeate probe and in feed, respectively.

#### 3. Results and discussion

## 3.1. Micelle formation in HDPBr/TX100 mixtures

Fig. 2 displays the isotherms of the surface tension in single TX100 and HDPBr solutions as well as in the binary mixtures of the surfactants of different compositions. It is seen that an increase of a nonionic TX100 content in the mixture ( $\alpha_{TX100}$ ) lead to decrease of surface tension values in the surfactants mixture at a given total feed surfactants concentration. The lowest surface tension values were found for TX100/HDPBr/mixtures at  $\alpha_{TX100}$  of 0.6–0.8.

Based on the surface tension data shown in Fig. 2, the CMC values for TX100, HDPBr and TX100/HDPBr mixtures were evaluated. The experimentally obtained CMC values of 0.24 and 0.67 mM for TX100 and HDPBr, respectively, well correlate with the previously reported data: 0.25 mM [37] and 0.27 mM [46] for TX100 and 0.64 mM for HDPBr [47].

The CMC values for TX100/HDPBr mixtures evaluated from Fig. 2 were 0.51 ( $\alpha_{TX100} = 0.2$ ), 0.34 ( $\alpha_{TX100} = 0.4$ ), 0.26 ( $\alpha_{TX100} = 0.6$ ) and 0.22 mM ( $\alpha_{TX100} = 0.8$ ). As seen from the obtained data the lowest CMC value was found for the TX100/HDPBr/mixture at  $\alpha_{TX100} = 0.8$ .

Fig. 3 presents the variation of the surface tension with composition of HDPBr/TX100 mixture at different pH values of the feed solutions. As seen in this figure the lowest values of surface tension in the binary mixtures are found at feed pH of 6.7 and the surface tension increase when the solution pH was changed to 3.3 or 9.1. Obviously such findings might be related to altering of the strength of intermolecular interactions, which occur between the molecules of counterpart surfactants in their mixture [48], when feed pH changes. As was reported protonation of the oxyethylene chain of the nonionic surfactant in the acidic solution [49] on the one hand and chelation of polyoxyethylene chain with sodium ions in the alkaline solution on the other hand [50] occur, which might affect the intermolecular interactions between TX100 and HDPBr molecules.



Fig. 2. Dependences of surface tension vs. the surfactant concentrations (C, M) in single TX100 and HDPBr solutions as well as in their binary mixtures of different compositions.  $\alpha$  represents a molar fraction of TX100 in the mixture; pH 6.7.



Fig. 3. Effect of pH on surface tension in the binary mixtures of TX100 with HDPBr.  $\alpha$  represents a molar fraction of HDPBr. The initial concentration of single TX100 and HDPBr solutions for preparation of the binary mixture is 0.34 mM.

By using the experimentally found surface tension data and CMC values for binary surfactant mixtures, a  $\beta^m$  parameter, which represents a strength of intermolecular interaction in micelles, and composition of mixed micelles were calculated by employing the Ruben–Rosen model [51] and the data are shown in Table 1.

The  $\beta^m$  parameter is evaluated as per equation [52]:

$$\beta^{m} = \frac{\ln\left(\alpha_{1}CMC / X_{1}^{m}CMC_{1}\right)}{\left(1 - X_{1}^{m}\right)^{2}}$$
(5)

where  $\alpha_1$  and  $X_1^m$  are molar fractions of surfactant 1 (TX100 in this study) for a feed solution and a mixed micelle, while CMC<sub>1</sub> and CMC values correspond to surfactant 1 and a binary mixture, respectively.

It should be mentioned that the absolute value of  $\beta^m$  parameter indicates the strength of intermolecular interactions, while positive and negative  $\beta^m$  values specify the repulsive or attractive interactions between the surfactant molecules, respectively.

#### Table 1

Intermolecular micellar interaction parameter ( $\beta^m$ ), the micellar fraction of TX100 ( $X^m$ ) and CMC in HDPBr/TX100 systems at different feed pH

α	0.2	0.4	0.6	0.8
pH 3.3				
$\beta^m$	-2.2	-2.0	-2.5	-2.7
$X^m$	0.63	0.66	0.72	0.77
CMC (M) × 10 <sup>3</sup>	0.60	0.64	0.33	0.27
pH 6.7				
$\beta^m$	-2.5	-2.8	-2.4	-3.1
$X^m$	0.76	0.77	0.75	0.79
CMC (M) × 10 <sup>3</sup>	0.51	0.34	0.26	0.22
pH 9.1				
$\beta^m$	-1.6	-1.8	-2.1	-2.3
$X^m$	0.67	0.65	0.66	0.69
CMC (M) × 10 <sup>3</sup>	0.58	0.83	0.64	0.52

( $\alpha$ ) is a molar fraction of TX100 in the binary mixture.

The surfactant 1 (TX100 in this study) micellar molar fraction  $(X_i^m)$  was evaluated as [52]:

$$\left(X_1^m\right)^2 \ln\left(\frac{\alpha_1 \text{CMC}}{x_1^m \text{CMC}_1}\right) = \left(1 - X_1^m\right)^2 \ln\left(\frac{(1 - \alpha_1) \text{CMC}}{(1 - X_1^m) \text{CMC}_2}\right)$$
(6)

where  $X_1^m$  is the surfactant 1 micellar molar fraction,  $\alpha_1$  is the surfactant 1 bulk molar fraction, CMC<sub>1</sub>, CMC<sub>2</sub> and CMC data represents the singe surfactants and their binary mixture, respectively.

As seen in Table 1, regardless the feed pH, the values of  $\beta^m$  parameter are negative in HDPBr/TX100 mixtures. This indicate that attractive interactions occur between the surfactants molecules in their binary mixture. These interactions might be due to dispersion attraction forces between hydrophobic tails of the surfactants [39,53] as well as due to ion-dipole attraction of hydrophilic groups in TX100 and HDPBr molecules [54]. Unpaired electrons of oxygen atoms in polyoxyethylene groups of TX100 might electrostatically attract to the positively charged head groups of a cationic surfactant [55].

The data presented in Table 1 also show that absolute  $\beta^m$  values are higher at neutral pH of 6.7 compared to  $\beta^m$  calculated at acidic pH of 3.3. Previously it was reported that protonation of polyoxyethylene groups in the molecules if nonionic surfactants can occur at low pH [49]. As a result electrostatic repulsion between molecules of HDPBr and TX100 at these conditions possibly might be reduced. On the other hand,  $\beta^m$  values decline when the solution pH was increased to 9.1. Obviously this is due to decreasing of ion-dipole interactions between the TX100 and HDPBr molecules as a result of chelation of sodium, which was added at pH adjustment with sodium hydroxide, with polyoxyethylene groups of the nonionic surfactant [50]. As a result the ion-dipole TX100-HDPBr interactions decline at pH 9.1.

It should be highlighted that as seen in Table 1, the strengthening of intermolecular interactions between TX100 and HDPBr molecules lead to notable reducing of CMC values in the surfactant mixtures.

### 3.2. Removal of BPA from water by MEUF

Fig. 4a shows BPA rejection during MEUF by using single TX100 and HDPBr surfactants as well as their mixtures at different surfactants concentration in the feed solution. As seen in Fig. 4a BPA rejection with UP010 P membrane is as low as 6% without adding the surfactants. Obviously, the UP010 P membrane is not able to efficiently reject BPA molecules by the sieving mechanism because of the relatively high molecular weight cutoff (10 kDa) and the BPA rejection is mainly due to the solute sorption within PES membrane matrix. Fig. 4a displays that the observed BPA rejection increases when the single TX100 or HDPBr surfactants were added to feed solution. At the surfactants concentration of 0.12 and 0.33 mM for TX100 and HDPBr, respectively, which are below their CMC values of 0.24 mM for TX100 and 0.67 mM for HDPBr (cf. subsection 3.1), the BPA rejection were 45% and 26%, respectively. However when the concentration of added HDPBr and TX100 surfactants reached their CMC values, the BPA removal sharply increase to 76% and 80%. Such increase in BPA removal can be explained by formation of the surfactant micelles in the feed solution, which can solubilize BPA in their hydrophobic nuclei, similarly to solubilization of other organic pollutants such as phenols [46] and dyes [56] in the surfactants micelle. The formed surfactant micelles with solubilized BPA can be rejected with UF membrane. Higher values of BPA rejection at MEUF with TX100 compared to HDPBr surfactant could be explained by a bigger aggregation number (150 vs. 80) and larger hydrodynamic radius (4.7 vs. 2.5 nm) of TX100 micelles compared to HDPBr ones [37]. Some BPA rejection at the surfactants concentration below their CMC values could be due to formation the premicellar surfactant aggregates in the feed solution [39] and possible binding of BPA molecules with such aggregates presumably due to hydrophobic disperse interactions. Fig. 4a also shows that adding of the surfactants at concentration above the CMC values practically do not increase the BPA removal from feed solution and hence economically is not feasible due to higher cost of the surfactants used. Also, the flux during MEUF declines at higher surfactants concentration in treated water (Fig. 4b).

As seen in Fig. 4a, notably higher values of BPA removal of 95.0%–96.5% were obtained during MEUF when employing the TX100/HDPBr mixture at concentration above the CMC compared to the single surfactant solutions. This might be due the fact that the steric size of mixed micelles in surfactant mixtures is larger than the size of micelles in solutions of individual surfactants [37]. For example, it was reported that the size of cetylpyridinium micelle increased from 0.9 to 5 nm during solubilization of p-xylene in the micelle core [57]. The larger size of mixed surfactant micelles with solubilized BPA contributes to their higher rejection on the UF membrane. The schematic presentation of the mixed micelle with the solubilized BPA



Fig. 4. Dependences of the BPA rejection (a), permeate flux (b) and the surfactant rejection (c) vs. concentration of TX100, HDPBr and TX100/HDPBr mixture (a molar ratio is 0.8:0.2) in the feed during MEUF of 1 mg/L BPA solution. Operating pressure is 2 bar; pH = 6.7.

is shown in Fig. 5. Similar observation on higher removal of heavy metals and organic pollutants when using the mixed surfactants was also reported in other studies [37,41].

As seen in Fig. 4b, the permeate flux decreases with an increase of the feed surfactant concentrations and the most pronounced flux decline is found when the TX100 and HDPBr were added at concentrations, which correspond or higher their appropriative CMC values. This effect is obviously due to formation on the membrane surface of the deposited layer composed of the surfactant micelles, which possess additional resistance to the transmembrane flux. Using of high surfactant concentrations might also lead to higher viscosity of the feed solutions [58] and hence results in lower permeate fluxes. It should be highlighted that employing the HDPBr/ TX100 mixture the permeate flux is notably higher compared to permeate fluxes when single TX100 or HDPBr surfactants were used (Fig. 4b). This finding can be related to the larger size of the mixed HDPBr/TX100 micelles and as a result the mixed micellar layer deposited on the membrane surface might be more porous and less dense compared to deposited layers composed of single HDPBr or TX100 micelles of smaller size. Similarly, formation of denser and less porous layers made composed of inorganic particles of smaller size deposited on the membrane surface was reported previously [59].

One of the crucial parameters during MEUF is a residual concentration of the surfactant in the treated water. As seen in Fig. 4c, the rejection values for TX100 and HDPBr surfactant from their 0.01 M solutions with the UP010 UF membrane are 8% and 21%, respectively. The rejection is obviously due to the surfactants adsorption within the membrane matrix and higher removal of HDPBr could be because of stronger adsorption of this cationic surfactant on the PES membrane, which possess some negative zeta potential at neutral pH [60]. The TX100 and HDPBr removal values increase with the surfactants concentration and at the feed surfactants removal of the was found to be 92%–98% both in the single surfactant solutions and their mixture (Fig. 4c).

Fig. 6 shows the BPA removal by MEUF process at different BPA concentration in the aqueous solution over the range of 0.5–5.0 mg/L. As seen, the BPA rejection somewhat decreases from 91% to 78% and from 79% to 65% for MEUF process with TX100 and HDPBr surfactants, respectively, with an increase of feed BPA concentration. This effect is obviously due to saturation of the solubilization capacity of the surfactant micelles towards BPA at a higher BPA content in water. On the other hand, the BPA rejection decline slightly from 96% to 90% with 0.5 to 5 mg/L increment of BPA content in the feed. This effect can be related to higher solubilization capacity of the mixed TX100/HDPBr micelle towards BPA as a result of a larger size of the mixed micelle compared to the micelles of single surfactants.

# 4. Conclusions

It was shown that the introduction of a synergistic mixture of the nonionic TX100 and the cationic HDPBr surfactants in the amount that ensures the micelles formation promotes the BPA solubilization in the hydrophobic nucleus of the micelles and provide higher BPA removal from water compared to when the single surfactants were used for MEUF process. For example, at a total surfactant content of 0.24 mM, the BPA removal values are 12%, 80% and 95%, while 78%, 91% and 99% rejection was found at total a surfactant concentration of 0.75 mM when using HDPBr, TX100 and TX100-HDPBr mixture, respectively. The higher BPA removal values found with TX100/HDPBr micelles in MEUF process is obviously due to the larger size of the mixed micelle compared to the micelles of single surfactants.

It was found that the permeate flux during MEUF is notably increased when using the surfactants mixtures. For instance, at a total surfactant concentration of 0.24 mM, the

Ion-dipole interactions



Fig. 5. Immobilization of BPA molecules inside a mixed TX100/HDPBr micelle.



Fig. 6. BPA rejection vs. feed BPA concentration during MEUF with single TX100 (0.27 mM), HDPBr (0.70 mM) surfactants and TX100/HDPBr mixture (0.24 mM). Operating pressure is 2 bar; pH = 6.7.

permeate flux at filtration of 1.0 mg/L BPA solution with addition of TX100/HDPBr mixture is 86.4 L/m<sup>2</sup>·h, which is 15% higher than the permeate flux of 73.5 L/m<sup>2</sup>·h when employing the single HDPBr surfactant for MEUF. Higher permeate fluxes in MEUF with the surfactant mixtures compared to single surfactants is believe to be due to formation of a more porous TX100/HDPBr micellar layer with lower hydrodynamic resistance on the membrane surface.

It should be highlighted that formation of mixed micelles in synergistic surfactant mixtures, which occurs at lower CMC values compared to CMC concentrations for individual surfactants, reduces the total amount of the surfactants needed for MEUF process. Thus, employing of the surfactants mixture could improves both efficiency and costeffectiveness of MEUF process of BPA removal from water.

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