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COMPARISON OF EFFECTIVENESS
OF ADVANCED TREATMENT OF MUNICIPAL WASTEWATER
BY SORPTION AND NANOFILTRATION.
SEPARATE PROCESSES AND INTEGRATED SYSTEMS

The feasibility of effluent treatment in separate processes and in integrated systems has been compared. In the sorption process, carbon nanotubes were used in a pressure installation, enabling execution of the process of sorption with nanofiltration in a cross-flow system. Evaluation of treatment effects based on the measurement of typical pollution parameters (COD, TOC, phenolic index, forms of nitrogen and phosphorus) and concentration of bisphenol A. The results show poor feasibility of sorption as a process for advanced treatment of municipal effluent. Effluent polishing in integrated sorption-nanofiltration systems, however, guaranteed high removal rates of organic and inorganic pollutants.

1. INTRODUCTION

Municipal or industrial wastewater discharges are frequent contributors to the pollution of surface water. These streams contain organic and inorganic pollutants, trace amounts of micropollutants, heavy metals, viruses and bacteria. Conventional methods of wastewater treatment are insufficient for complete removal of micropollutants. Among anthropogenic micropollutants whose presence, even in trace amounts, is adverse for biological equilibrium in the aquatic environment and human health, are phenols, non-biodegradable organochloride compounds, pesticides, polycyclic aromatic hydrocarbons (PAHs), detergents, and heavy metals [1–4]. The effect of these

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micropollutants on the environment depend on the concentration, physicochemical properties of the compounds, their resistance to biodegradation, and bioaccumulation potential. In addition, products of the microbial degradation of some micropollutants can be more persistent and toxic than the original sources [5–7]. Some of them have their own biological activity and have toxicological effects on organisms in activated sludge. Effective removal of micropollutants during mechanical-biological wastewater treatment can also be limited by the hydrophobicity of chemicals, the result of degradation under aerobic conditions.

With these facts in view, the development of methods for purifying biologically treated municipal effluents enabling removal of non-biodegradable micropollutants otherwise released into the receiving water is significant. In modern wastewater treatment technologies, integrated systems are applied that couple with each other in individual (separate) processes, Most of these processes occur in multifunctional reactors. On the other hand, there are integrated systems where individual processes are carried out independently of each other, as in the following processes in separated reactors. Available data from the literature indicate that the effective removal of pollutants in separate physicochemical processes can be enhanced in hybrid systems [8]. A successful treatment of landfill leachate was obtained in a hybrid system comprising sorption on activated carbon with nanofiltration (NF). Reduction of COD reached 90% [9]. Heo et al. [10] indicated higher removal of bisphenol A and 17β-estradiole in a single-walled carbon nanotube–ultrafiltration membrane system than in ultrafiltration.

In integrated systems, more intensive fouling can occur than in separate membrane processes due to embedding small particles of nanotubes on the surface and pores of the membrane. The objective of this study was to investigate the feasibility and effectiveness of removal of pollutants from biologically treated wastewater by means of separate processes and various modifications of integrated systems.

2. EXPERIMENTAL

Materials. Bisphenol A (BPA) (purity > 99%) was purchased from Sigma Aldrich (Poland). The stock solution of BPA was prepared with methanol (analytical standard) supplied by POCH Inc. (Poland). Pure water was taken directly from a Milli-Q water purification system (Millipore LLC, Poland). Single-walled carbon nanotubes (SWCNT) were purchased from Chengdu Organic Chemistry Ltd., Chinese Academy of Science. These nanotubes were synthesized by the chemical vapor deposition method and the raw product was purified with a mixture of HNO_3 and H_2SO_4 (data from manufacturer). The commercial nanofiltration membrane TS40 (TriSep) was used in the present study. The detailed characteristics of the membrane, obtained from the manufacturer, are shown in Table 1.

Characteristics of the nanofiltration membrane

Manufacturer	TriSep
Symbol	TS40
Polymer	polypiperazine amide
Molecular weight cut off, Da	200
Retention NaCl, %	45
Retention MgSO$_4$, %	99
Deionised water flux ($\Delta P = 1$MPa), m^3/(m$^2 \cdot$s)	1.51×10^{-5}

Characteristics of wastewater. Effluent samples were collected from a mechanical-biological wastewater treatment plant located in the region of Silesia, Poland. This plant is designed for a wastewater volume of 51 000 m^3/day. Mechanical treatment is followed by biological treatment with activated sludge for degradation of nitrogen and phosphorus compounds. After final sedimentation, the effluent water is discharged into the receiving river. Effluent for this study was sampled directly from the settling tank. Biologically treated wastewater was immediately preserved in 5 l brown bottles and processed within five days. Its physicochemical characteristics are presented in Table 2. The values of typical parameters did not exceed the permissible norms prescribed by the Ministry of the Environment [11].

Characteristics of biologically treated wastewater

Parameter	Value in effluent	Maximum permissible value [11]
pH	7.60	6.5–9
Colour, mg Pt/dm^3	29.00	–
Turbidity, FAU	2.00	–
COD, mg/dm^3	31.00	125
TOC, mg/dm^3	19.83	30
Inorganic carbon (IC), mg/dm^3	56.10	–
Phenolic index (PI), mg/dm^3	0.16	0.1
Bisphenol A, µg/dm^3	3.12	–
N$_{tot.}$ mg/dm^3	6.00	10
N-NH$_4^+$, mg/dm^3	2.00	10
N-NO$_2^-$, mg/dm^3	0.036	1
N-NO$_3^-$, mg/dm^3	3.12	30
P-PO$_4^{3-}$, mg/dm^3	0.50	1
Cl$^-$, mg/dm^3	120.00	–

Sorption process. First, sorption of bisphenol A onto single-walled carbon nanotubes was conducted. Nanosorbent was obtained from the manufacturer (Chengdu Organic Chemistry Ltd., Chinese Acad. Sci.) in the form of black powder. A sufficient volume of BPA stock solution was added to achieve the concentration of 100 µg/dm^3 in biologically treated wastewater. This was done to ensure the analysis as precise as possible. In order to determine a sufficient amount of nanosorbent for high removal of micropollutants from a competitive system (real wastewater), the concentration of SWCNT was changed in the range of 15–100 mg/dm^3. Adsorption experiments were conducted in 100 cm^3 glass flasks equipped with a ground-in stopper using a batch system for 15 min [12] . The flasks were shaken at 150 rpm/min under ambient conditions (23±1 °C). Then the nanotubes were separated from the wastewater with a 0.2 µm PTFE filter. In the following stage of sorption studies, nanotubes were used for polishing the non-artificial biologically treated wastewater (without spiking with surrogate standard BPA). In this case, the concentration of sorbent was 75 mg/dm^3.

Nanofiltration. The nanofiltration process was carried out using cross-flow nanofiltration units (GE Osmonics) with flat sheet membranes TS40 (TriSep) under the transmembrane pressure of 1.5 MPa. The active surface area of the membrane was 144 cm^2. The installation was equipped with a feed container (30 dm^3) with a refrigerating system, rotameter, high pressure pump and manometer. The temperature of the wastewater was maintained at 19±1 °C. The process was conducted for 7 h in a closed system (without recirculation of permeate). The initial volume of wastewater was 30 dm^3. Prior to the first application, the membranes were conditioned by means of filtration of demineralized water. During filtration of wastewater, the volume of permeate was measured and then permeate flux (J_v) and feed recovery were calculated. After each filtration of wastewater, demineralized water was refiltrated, to evaluate the relative flux of demineralized water (α_w) and relative permeate fluxes (α_v)

$$J_v = \frac{V_p}{Ft} \tag{1}$$

$$S = \frac{V_p}{V_n} \times 100\% \tag{2}$$

$$\alpha_w = \frac{J_{w2}}{J_{w1}} \tag{3}$$

$$\alpha_v = \frac{J_v}{J_{w1}} \tag{4}$$

where: V_p is the volume of permeate, m^3, V_n – volume of feed, m^3, F – active surface area of membrane, m^2, t – time, s, J_{w1} – flux of deionized water before filtration of wastewater, m^3/(m^2·s), J_{w2} – flux of deionized water before filtration of wastewater, m^3/(m^2·s), J_v – flux of permeate , m^3/(m^2·s).

Integrated systems sorption–NF. Advanced treatment of wastewater in the integrated system was carried out at the concentrations of sorbents 30 and 75 mg/dm^3. The treatment process in this system was conducted in three major configurations:
- sorption 75 mg/dm^3–NF;
- sorption 30 mg/dm^3–NF "in line";
- sorption 75 mg/dm^3–NF "in line".

In the first configuration, adsorption and nanofiltration processes proceeded independently of each other. Nanosorbent was separated from the wastewater by means of filtration (filter disk). The configuration "in line" was a hybrid system, in which the nanosorbent was added directly to the recirculating system of the membrane installation.

Physicochemical analysis. The degree of wastewater polishing was determined as the difference in the values of typical parameters (COD, TOC, IC, color, phenolic index, concentrations of total nitrogen, nitrate, nitrite, chlorine) for biologically treated effluent and effluent after polishing. The concentration of BPA was determined according to the method described elsewhere [13]. COD and the phenolic index, the nitrogen species and phosphorus were determined with a Merck test kit. In COD analysis, the samples were oxidized with a hot sulfuric solution of potassium dichromate with silver sulfate as the catalyst. Color was measured by means of spectrophotometric analysis. Chlorine was analyzed by the Mohr method. The concentrations of forms of carbon were determined with a Multi N/C Analytik Jena carbon analyzer.

3. RESULTS

3.1. CHARACTERISTICS OF CARBON NANOTUBES

In order to confirm the presence of nanotubes in the studied material, transmission electron microscopy (TEM) and Raman spectroscopy examinations were performed. In TEM images (Fig. 1), the morphological structure of nanotubes was present. They were homogeneous. Trace amounts of cobalt and iron were also present in this sorbent. They were the residue of catalysts used during the production of nanotubes.
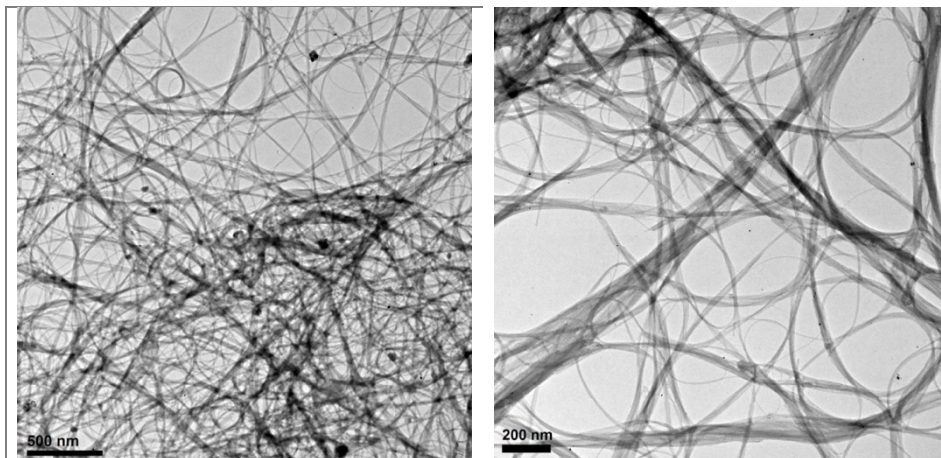
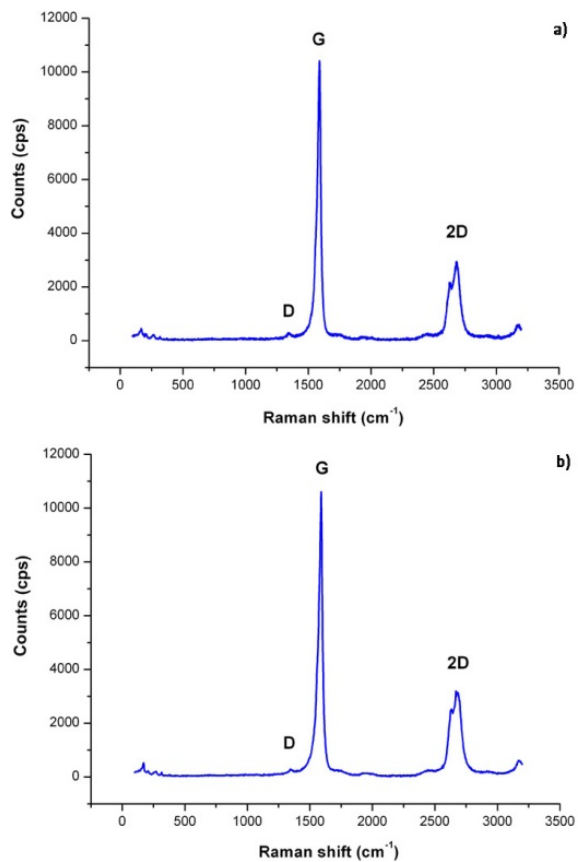Fig. 1. TEM images of carbon nanotubes in various magnifications

Fig. 2. Raman spectra of carbon nanotubes before (a) and after (b) sorption

In a typical Raman spectrum of carbon nanotubes, three main components are present, namely peaks D, G and 2D The D band, characterizes molecular deformation in carbon nanotubes resulting, for instance, from wrong treatment or chemical functionalization. The G band confirms that the material has graphitic structure, that is constructed solely of atoms of carbon p hybridisation sp^2. The peak 2D indicates stress on the graphene network. Generally the ratio between the intensity of the D and G peaks quantifies the disorder and defects of the structure. The lower the ratio D/G is, the less deformed the structure of nanotubes. When the intensity of the peaks is similar, the presence of graphene with a poor degree of crystallisation is indicated. These nanotubes can lack some of their typical properties.

The Raman spectrum was determined for nanotubes before (Fig. 2a) and after sorption (Fig. 2b). In both cases, the Raman line found at 1589 cm^{-1} was assigned as the G band. D peaks were very low. It confirmed the presence of properly constructed nanotubes with high structural stability. Moreover, narrow shape of G peak indicates that studied nanotubes were semiconductors. Mechanical stress in nanotubes can cause structural deformation, consequently leading to the appearance of a D band or the displacement of the G band in the Raman spectrum [14]. There was no shift of G and D peaks after sorption. Thus, sorption of pollutants contained in effluent did not influence their structural properties.

3.2. SORPTION OF BPA FROM BIOLOGICALLY TREATED WASTEWATER

It is obvious that the dose of nanosorbents had an important role in treatment effects. Figure 3 illustrates the dependence of the effectiveness of BPA removal from discharged wastewater on the dose of carbon nanotubes.
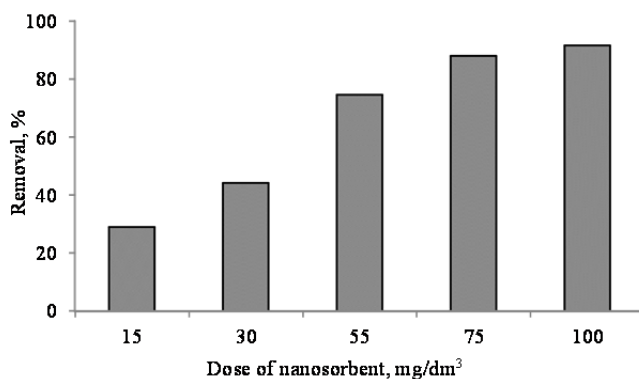

Fig. 3. Effectiveness of micropollutant removal from wastewater in function
of the dose of carbon nanotubes (initial concentration of BPA – 100 µg/dm^3)

The increased dose of nanotubes made micropollutant removal more effective due to existence of higher number of adsorption sites. The dose of nanotubes required to remove most of the BPA from the effluent was 75 mg/dm^3.

3.3. EFFLUENT POLISHING BY MEANS OF SORPTION

The effectiveness of effluent polishing in the sorption process is presented in Fig. 4. The compounds responsible for color in the discharged wastewater were well removed by sorption at the level of 34%, which corresponds to 10 Pt/dm^3. Removal of organic compounds responsible for COD and TOC did not exceed 25%. Similar results were obtained for inorganic compounds.
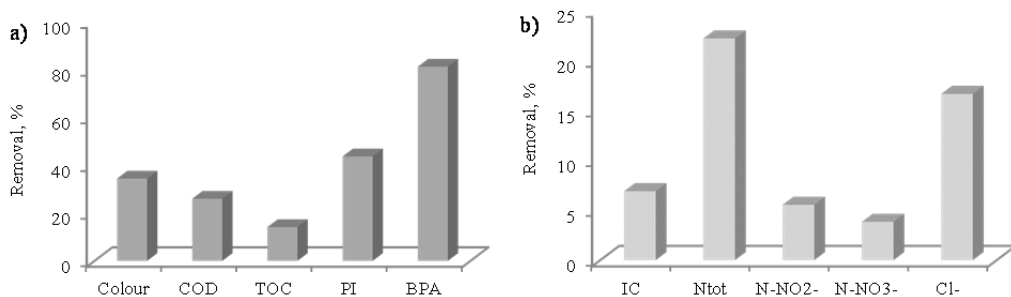

Fig. 4. Removal of organic (a) and inorganic (b) pollutants from wastewater
in sorption process with the use of nanotubes (sorbent dose – 75 mg/dm^3)

Reduction of nitrate and nitrite was not higher than 4% and 5.5%, respectively. Higher removal was observed for chlorine ions (16.6%) and total nitrogen (22%). Such a high removal of bisphenol A was a consequence of its very low concentration in the effluent (3.12 μg/dm^3). Generally, this study indicates that sorption as a separate process does not guarantee a high rate of polishing of discharges wastewater. Therefore, in the next step, sorption was examined in integrated systems.

3.4. EFFLUENT POLISHING BY MEANS OF NANOFILTRATION AND INTEGRATED SYSTEMS

Figures 5, 6 present the effectiveness of the polishing of discharged wastewater in three integrated systems (sorption 75 mg/dm^3–NF, sorption 30 mg/dm^3–NF "in line", sorption 75 mg/dm^3–NF "in line"). All systems showed good retention of color that occurred in traces in the treated filtrates. In terms of chemical parameters, high reduction degrees were obtained for COD (82–89%), TOC (70–92%) and phenolic index (50–69%). BPA was removed almost completely (71–90%). Thus all studied systems indicated very effective removal of organic pollutants. However, it seems that reduction of these organic pollutants was slightly lower in nanofiltration than in integrated

systems. A similar tendency was found in the removal of inorganic compounds. Thus, effluent polishing was enhanced owing to additional sorption before nanofiltration.
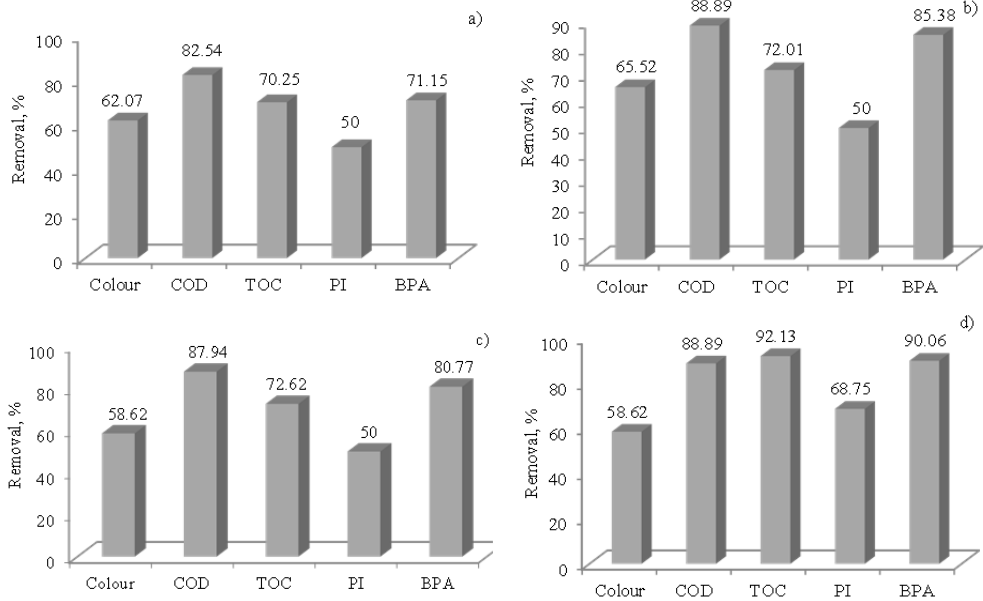

Fig. 5. Removal of organic pollutants in: a) NF, b) sorption 75 mg/dm^3–NF,
c) sorption 30 mg/dm^3–NF "in line", d) sorption 75 mg/dm^3–NF "in line"

The type of integrated system did not have a very significant impact on the treatment effects. The composition of permeates after integrated system treatment was similar.

The degradation of organic matter during mechanical-biological wastewater treatment leads to carbon dioxide, water and a removable fraction of biomass. Thus, this process is considered the main source of inorganic carbon. Independently of the system used, the retention of inorganic carbon was sufficient (80.1–94.4%). The retention of inorganic compounds in particular ions (N-NO$_2^-$, N-NO$_3^-$) and total nitrogen was higher for integrated systems in comparison with separate NF. Generally, the retention of monovalent ions in NF did not exceed 40%. In integrated systems, some of these ions were removed by means of sorption on carbon nanotubes and residue was retained on the NF membrane.

As regards the effectiveness of the polishing of discharged wastewater, the best system was sorption 75 mg/dm^3–NF "in line". On the one hand, the addition of over twice as much sorbent resulted in the sorption of a higher amounts of pollutants on carbon nanotubes. On the other hand, comparing systems with the same dose of nano-sorbent that differed in terms of the process used (sorption 75 mg/dm^3–NF and sorp-

tion 75 mg/dm^3–NF "in line"), greater effectiveness of polishing in the "in line" system followed the embedding of sorbents in the membrane surface. Thus, they created an additional separation layer on the membrane.
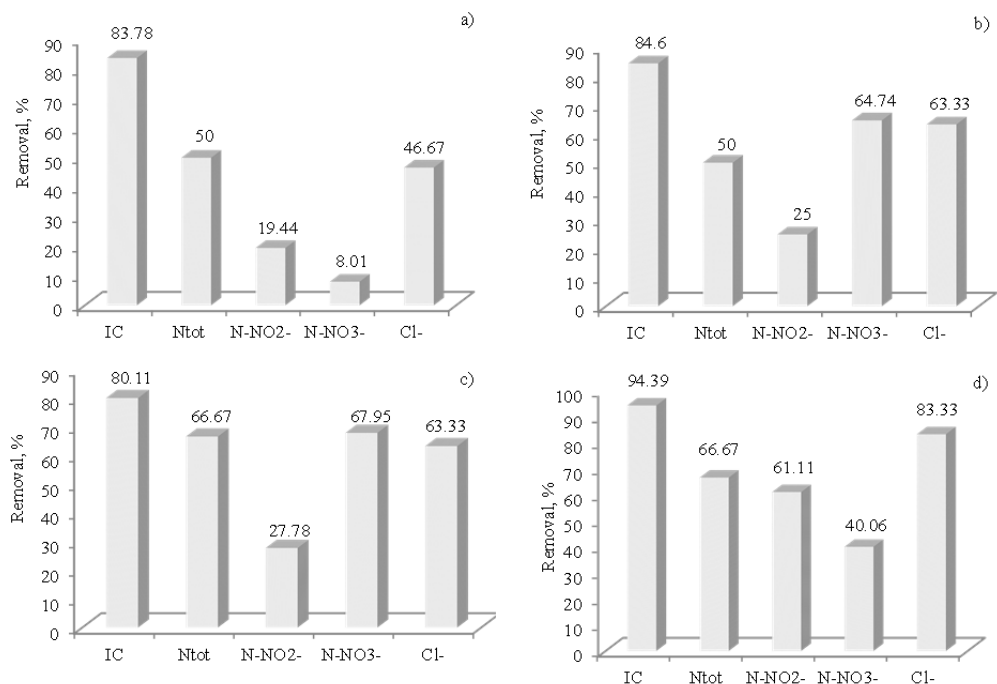

Fig. 6. Removal of inorganic pollutants in: a) NF; b) sorption75 mg/dm^3–NF,
c) sorption 30 mg/dm^3–NF "in line", d) sorption75 mg/dm^3–NF "in line"

Significantly, discharged wastewater treatment in an integrated sorption-nanofiltration system guaranteed high removal of bisphenol A and other micropollutants responsible for the phenolic index. Reduction of typical organic and inorganic parameters indicated the high feasibility of the treatment. The physicochemical composition of filtrates after polishing corresponded to basic standards for drinking water in accordance with the guidelines of the Ministry of Health [15].

3.5. PERMEATION AND FOULING OF NANOFILTRATION MEMBRANES

As shown in Figure 7, the lowest permeate flux was obtained for sorption 75 mg/dm^3–NF "in line" and the highest for sorption 75 mg/dm^3–NF. Permeability of membrane in the separate nanofiltration was lower than in the integrated system with independently conducted processes but higher than in integrated systems in "in line" mode. Thus, the sorption on carbon nanotubes before nanofiltration constituted the

pre-treatment process of feed before high pressure filtration and resulted not only in better retention of pollutants in comparison with separate sorption and nanofiltration but also in the enhancement of membrane permeability. The addition of nanosorbent to the membrane installation caused a considerable decrease in permeate flux, consequently leading to lower feed recovery. Generally, the higher permeability of the pressure process was connected with higher feed recovery (Fig. 8). In the system with the highest permeate flux, final feed recovery reached almost 30% and corresponded to a permeate volume of 8.7 dm^3, whereas in the system with the lowest flux the final volume of permeate was 6.9 dm^3 (Table 3).
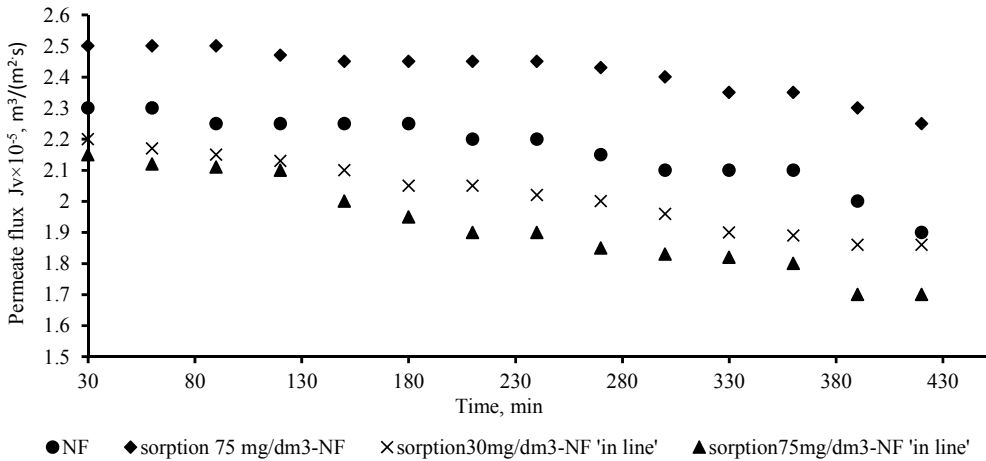

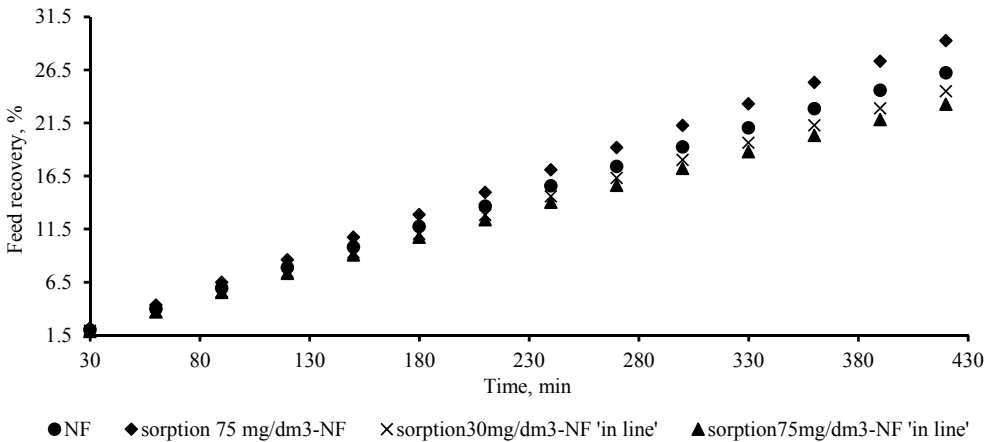Fig. 7. Time dependence of permeate flux in studied systems

Fig. 8. Time dependence of feed recovery in studied systems

The decrease of permeate flux over time was observed for all studied pressure filtrations, however (Fig. 7). Intensity of fouling depended on how effluent treatment was performed. The relative permeate flux (α_v) allowed determination of the susceptibility of the membrane surface for deposition of pollutants. Additionally, intensity of fouling was determined based on the relative fluxes of deionized water (α_w). The higher the values of these coefficients are, the less the fouling of the membrane. They are presented for all studied systems in Table 2. These coefficients confirmed previous observations in that membrane working in the system sorption 75 mg/dm^3–NF was responsible for the highest hydraulic performance and high fouling resistance. In this case, the values of relative permeate and fluxes of deionized water were at the level of 0.89 and 1.00, respectively. It means that 7 h filtration of feed in concentration conditions (without influent of fresh wastewater) did not cause significant fouling of membrane. Furthermore, after the treatment process, the permeability of the membrane for deionized water did not change.

<div align="right">T a b l e 3</div>

<div align="center">Relative fluxes of permeate and deionized water</div>

System	Average relative flux [m^3/(m^2·s)]		Total volume of permeate [cm^3]
	Permeate, α_v	Deionised water, α_w	
NF	0.80	0.98	7867
Sorption 75 mg/dm^3–NF	0.89	1.0	8774
Sorption 30 mg/dm^3–NF "in line"	0.75	0.92	7346
Sorption 75 mg/dm^3–NF "in line"	0.71	0.88	6980

Although pressure filtrations were carried out in the cross-flow system, it was impossible to avoid embedding of nanosorbent particles in the membrane surface. A nanoparticle layer was formed on the membrane surface, which increased the hydraulic resistance of filtration. Furthermore, because of a very small size of the nanosorbents, some of them migrated inside the membrane structure and blocked pores.

Based on the α_v and α_w values for the two integrated systems, the impact of adding carbon nanotubes to pressure installation on the permeability of the nanofiltration membrane was significant. The higher the sorbent concentration was, the more intensive the fouling. Taking into account the lower permeability of TS40 membrane in hybrid systems ("in line") in comparison with systems with independently conducted processes and also loss of initial permeability of membrane after the process, integrated systems in "in line" mode cannot be classified as an applicable solution.

4. CONCLUSIONS

The nanosorbent used in the study was made of very well crystallized carbon nanotubes. No molecular defects in the structure of nanotubes were found before and after sorption of pollutants.

The nanotube dose required to remove most of the BPA from biologically treated wastewater was 75 mg/dm^3.

Sorption as a separate process does not guarantee a high degree of effluent polishing. The high effectiveness of the removal of bisphenol A was a consequence of the very low concentration of this compound in effluent.

The degree of polishing of discharged wastewater was very good in separate NF and all integrated systems. Reduction of organic compounds responsible for COD and TOC was similar in all studied systems The retention of inorganic compounds, in particular ($N-NO_2^-$, $N-NO_3^-$) ions and total nitrogen was higher for integrated systems in comparison with separate NF. Taking into account only the effectiveness of polishing of discharged wastewater, the most advantageous system was sorption 75 mg/dm^3–NF "in line".

The lowest permeate flux was obtained for sorption 75 mg/dm^3–NF "in line" and the highest for sorption 75 mg/dm^3–NF. Sorption on carbon nanotubes before membrane filtration allowed partial removal of pollutants from the feed. This system (sorption 75 mg/dm^3–NF) offered the highest permeability and feed recovery.

Addition of carbon nanotubes to the membrane installation caused the embedding of nanoparticles on the surface and pores of the membrane. This nanolayer increased the hydraulic resistance of filtration. Nanofiltration membranes used in the integrated systems in "in line" did not recover initial deionised water permeability.

Taking into account the effectiveness and performance of effluent polishing, the best system was sorption 75 mg/dm^3–NF.

ACKNOWLEDGEMENTS

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