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The aim of this paper is to investigate the efficiency of a hybrid coagulation–microfiltration process for the removal of copper from a synthetic wastewater in comparison with the application of microfiltration alone. The wastewater samples contained various concentrations of humic acids, kaolin (clay), and Cu^{2+} ions in tap water. The effect of the concentrations of the constituents on the performance of microfiltration is examined. The efficiency of microfiltration was evaluated by measuring several parameters, including residual Cu^{2+} concentration, residual turbidity, residual humic acids concentration, pH, conductivity, and alkalinity. A conventional inorganic salt ($Fe_2(SO_4)^3$) and a novel composite inorganic-organic coagulant (PFS-PAA) were applied as coagulation agents to further enhance the microfiltration efficiency. The results show that membrane microfiltration is very efficient in treating such wastewaters, while the hybrid process exhibits even better performance, when compared to microfiltration.

Keywords coagulation; copper; humic acids; microfiltration; Natural Organic Matter

INTRODUCTION

Nowadays, liquid wastes from many industrial sectors, including mining, mineral processing, metal finishing, electroplating, and battery industries, contain a comparatively high concentration of heavy metals, which need to be separated before their discharge into surface water or sewage system. Water contamination by heavy metal ion is a major environmental hazard and its removal is highly essential. Heavy metals may usually behave as toxic elements; their accumulation in living tissues through the food chain may pose a serious health hazard not only for humans but also for many living forms (1). According to the World Health Organisation, the metals with the most immediate concern regarding their toxicity are—chromium, copper, zinc, lead, mercury, and iron. In addition to the necessity of removing metals from wastewaters due to environmental safety, the increasing demand for most of the heavy metals and the depletion of ores globally results in finding new and more efficient methods for recovering metal ions even at very low concentration in wastewater (2).

Copper is often regarded as a major pollution problem to the environment either from natural sources or from human industrial activities. The latter include wastewaters from industrial processing of paper, pigments, metal, fertilizers, and mining processing. Typical processes that are employed for removing copper from diluted aqueous solutions include sorption (3,4), biosorption (5–7), chemical precipitation (8–10), coagulation-flocculation (11–13), and flotation (14–16). Additionally, a variety of membrane processes have also been used, including microfiltration (MF), ultrafiltration (UF), reverse osmosis (RO), or emulsion liquid membranes (ELM) (2,17,18).

Microfiltration (MF) is the oldest of the four pressure–driven membrane technologies, which also include reverse osmosis, nanofiltration, and ultrafiltration. The pore size of membrane ranges from 0.05–10 μm, which classifies MF between ultrafiltration membranes and conventional filters (19). Due to its large pore size, the microfiltration process is primarily used for particle and microbial removal and can be operated under low pressure conditions. However, it is worth noting that MF does not always operate strictly based upon the pore size of membranes. A cake layer, which may be formed on the membrane surface due to materials contained in the feedwater, will provide additional removal capacity (20). Among the other available membrane processes, microfiltration is perhaps the most appealing due to economic reasons, i.e., lower membrane module costs and lower pressure drops (1). A major disadvantage of MF membranes is their sensitivity to fouling due to accumulation and deposition of contaminants on the surface, which compromises the membrane performance and increases the operating cost of the membrane filtration process (21). Therefore, for improving the membrane efficiency as well as reducing the membrane fouling, a combination of microfiltration with other physicochemical processes, including flotation (1,22,23), adsorption (24),

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complexation (25,26) and coagulation (27) have been investigated.

In particular, adding a coagulant prior to the MF process, which is called the coagulation-microfiltration hybrid process, is regarded as one of the most promising options for solving these problems (27). Coagulation of feed water enables the removal of Natural Organic Matter (NOM) that may act as a major fouling material, thus increasing the membrane permeability (28–30). It also improves the filtration characteristics of MF by reducing the cake resistance of the deposit on the membrane surface (31). However, it should be noted that the effect of coagulation on membrane performance is difficult to be predicted as a variety of parameters i.e., constituents and pH of feed water, membrane materials, type and concentration of coagulant, significantly affect the overall performance. Howe and Clark (32), state that previous research has been inconsistent, with some studies suggesting improved membrane efficiency and some others showing decreased performance.

Overall, there is still uncertainty regarding the efficiency of the hybrid–microfiltration process in removing heavy metal ions from wastewater. The aim of this paper is to investigate the efficiency of the hybrid process for the removal of copper from a synthetic wastewater in comparison with the application of microfiltration alone. The wastewater samples contained various concentrations of humic acids, kaolin (clay) and Cu^{2+} ions in tap water. The effect of the concentrations of the constituents on the performance of microfiltration is examined. The efficiency of microfiltration was evaluated by measuring several parameters, including residual Cu^{2+} concentration, residual turbidity, residual humic acids concentration, pH, conductivity, and alkalinity. A conventional inorganic salt ($Fe_2(SO_4)^3$) and a novel composite inorganic-organic coagulant (PFS-PAA) were applied as coagulation agents to further enhance the microfiltration efficiency.

MATERIALS AND METHODS

Composition of Synthetic Wastewater

The synthetic wastewater used in this experimental project consisted of kaolin (clay), humic acids, and Cu^{2+} ions (from $CuSO_4 \cdot 5H_2O$) in various concentrations. In particular, 6 different types of wastewater were employed:

A. Kaolin 10 mg/L, Humic acids 10 mg/L, 10 mg/L Cu^{2+}
B. Kaolin 10 mg/L, Humic acids 10 mg/L, 50 mg/L Cu^{2+}
C. Kaolin 10 mg/L, Humic acids 10 mg/L, 100 mg/L Cu^{2+}
D. Kaolin 50 mg/L, Humic acids 50 mg/L, 10 mg/L Cu^{2+}
E. Kaolin 50 mg/L, Humic acids 50 mg/L, 50 mg/L Cu^{2+}
F. Kaolin 50 mg/L, Humic acids 50 mg/L, 100 mg/L Cu^{2+}

The preparation protocol can be described as follows: First, kaolin is dispersed in tap water, and then humic acids is introduced under strong agitation. Finally, copper (as copper sulphate) is introduced into the mixture. The properties of each type of wastewater are summarized in Table 1.

As the composition of tap water is an essential parameter on the properties of synthetic wastewater, the analysis of tap water is presented in Table 2.

For conducting two additional experimental series, the pH of each wastewater type was adjusted to pH = 6–7 and pH = 3–4 with the addition of an appropriate volume of H_2SO_4 96%.

Experimental Setup

The experimental system consisted of a vessel reactor in which a flat sheet membrane module was directly submerged. The reactor was made of a cylindrical Plexiglass vessel (115 × 60 cm) with a working volume of 300 L. The module was operated in a dead-end mode. The synthetic wastewater was fed to the reactor with the aid of a peristaltic pump, while the treated effluent was removed by the use of a centrifugal pump. The TMP values were read on a screen of a manometer, located at the permeate line.

The membrane module used in this study was the flat sheet Maxipur® 4 M-ATB Umwelttechnologien GmbH. Its main characteristics are summarized in Table 3.

Microfiltration Experiments

The same experimental procedure for the treatment of each of the wastewater types (Table 1) was followed.

TABLE 1
Main properties of synthetic wastewater

Wastewater	UV_{254} (Abs)	Turbidity (NTU)	pH	Conductivity (μS/cm)	Alkalinity (mg/L $CaCO_3$)
A	0.279	10	8.1	554	284
B	0.341	14	7.8	562	238
C	0.289	34	7.9	724	254
D	0.580	27	8.1	555	300
E	0.574	31	8.4	721	230
F	0.425	46	7.8	742	140

TABLE 2
Main properties of tap water

Temperature (°C)	23–27
pH	7.1–7.3
Conductivity (μS/cm)	540–680
Turbidity (NTU)	1.5–2.5
Alkalinity (mg /L $CaCO_3$)	275–310
Hardness (mg/L $CaCO_3$)	250–270
Salinity (g/L)	0.5
Total dissolved solids (mg/L)	320–410

TABLE 3
Characteristics of the membrane module

Model	Maxipur® 4 M
Configuration	Flat sheet
Nominal pore size	0.4 μm
Nominal surface area	0.08 m^2
Total surface area	3.8 m^2
Typical operating TMP	0.05–0.40 bar
Operating pH range	1–10
Operating temperature range	1–60°C
Air consumption	10 m^3/h

The main experimental conditions applied in these experiments are summarized in Table 4. It is worth noting that after the end of each set of filtration time, a cleaning set of 5 min (total cleaning set time) was applied. In particular for each cleaning set the membrane backwashing lasted for 1.5 min and the membrane relaxation for 3.5 min. The MF filtration ran under constant permeate flux.

TABLE 4
Main experimental conditions for microfiltration experiments

Composition of feed water	1–6 wastewater type (Table 1)
Permeate flux	70–75 LMH
Total duration of filtration	20 min (4 sets of 5 min each)
Frequency of backwash cleaning	5 min
Duration of backwashing	1.5 min
Duration of membrane relaxation	3.5 min
Sampling frequency	2 min
Permeate volume consumed for backwashing	1.2 L

Coagulation–Microfiltration Experiments

Prior to microfiltration, a coagulation stage was applied as a means of pre-treatment of feed water. The coagulation stage was conducted within the same vessel reactor, where the membrane module was submerged. The fast mixing stage of coagulation was achieved by intense aeration of the vessel (duration of 3 min), while the slow mixing was achieved by use of a mechanical agitator (duration of 25 min). Then, the sedimentation stage lasted for 40 min. After the end of the sedimentation stage, the microfiltration process commenced, as described in the section titled "Microfiltration Experiments". Two coagulation reagents were used:

a) A conventional ferric sulphate coagulant ($Fe_2(SO_4)_3 \cdot 5H_2O$, FS). The working concentration of the coagulant was determined after preliminary experiments, while an anionic flocculant aid (Magnafloc LT 25, Ciba SC) was added at a concentration of 1/10th of the concentration of Fe in the coagulation reagents. The flocculant aid was added for about 30–45 s before the beginning of the slow mixing stage, in order to enable the complete mixing of the reagent.
b) A new composite coagulant PFS-PAA, PAA/Fe = 0.1, which was prepared in the laboratory, consisting of Fe and non–ionic organic polymer (Polyacrylamide, PAA). The concentration of the non-ionic polymer within the composition of the coagulant is 1/10th of respective concentration of Fe. The working concentration of the coagulant was determined after preliminary experiments. No flocculant aid was added, as it had already been added within the composition of the new coagulant. Further information regarding this new coagulant is presented elsewhere (33).

Methods for the Evaluation of Microfiltration and Coagulation-Microfiltration Processes

The efficiency of each process was assessed by measuring several parameters, including—residual Cu^{2+} concentration (Perkin Elmer, AAnalyst 400), residual turbidity (HACH RATIO/XR Turbimeter), residual humic acids concentration, as an expression of UV_{254nm} absorption (Hitachi UV-Vis spectrophotometer), pH (Metrohm Herisau pH-Meter), conductivity (Crison CM 35 conductivemeter), alkalinity (Methyl orange indicator), and transmembrane pressure alteration (Afriso-Euroidex DMU pressure sensor).

RESULTS AND DISCUSSION

Microfiltration Efficiency for Treating Each Type of Synthetic Wastewaters

Figure 1 illustrates the removal of copper in terms of residual concentration (Fig. 1). Overall, it is recorded by the experimental results that the microfiltration process

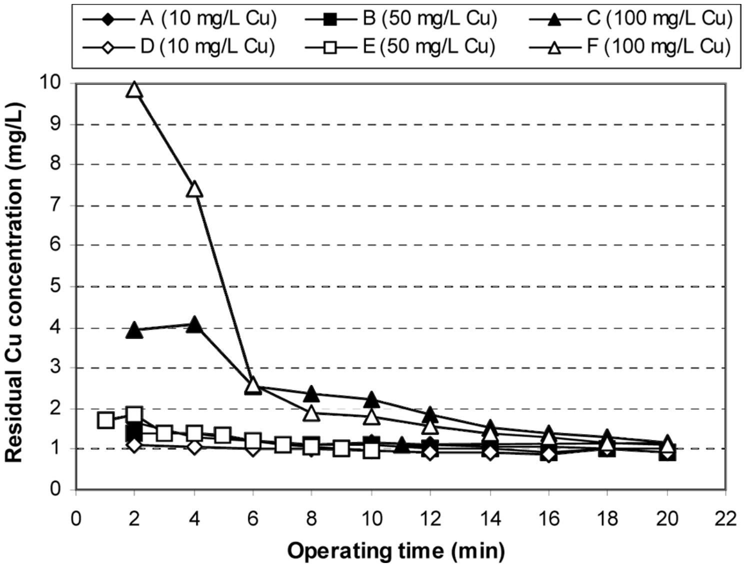


FIG. 1. Comparative microfiltration performance in copper removal.

TABLE 5
Comparative microfiltration efficiency in removal of copper

A/A	Synthetic wastewater	pH of wastewater	Permeate flux (LMH)	Final concentration of Cu^{2+} (mg/L)	Removal of Cu^{2+} (%)
1	A	7.5–8.5	70–75	1,01	90
2	B	7.5–8.5	70–75	0,92	98
3	C	7.5–8.5	70–75	1,08	99
4	D	7.5–8.5	70–75	0,94	91
5	E	7.5–8.5	70–75	0,97	98
6	F	7.5–8.5	70–75	1,12	99
7	A	6.0–7.0	70–75	0,63	94
8	B	6.0–7.0	70–75	0,87	98
9	C	6.0–7.0	70–75	2,90	97
10	D	6.0–7.0	70–75	0,52	95
11	E	6.0–7.0	70–75	1,15	98
12	F	6.0–7.0	70–75	3,75	96
13	A	3.0–4.0	70–75	$14{,}43^{A}$	–
14	B	3.0–4.0	70–75	$57{,}10^{A}$	–
15	D	3.0–4.0	70–75	$15{,}95^{A}$	–
16	E	3.0–4.0	70–75	$52{,}60^{A}$	–
17	10 mg/L Cu^{2+}	7.5–8.5	70–75	1,02	90
18	50 mg/L Cu^{2+}	7.5–8.5	70–75	0,93	98
19	100 mg/L Cu^{2+}	7.5–8.5	70–75	0,75	99
20	B	7.5–8.5	35–37	1,01 ($0{,}82^{B}$)	98
21	C	7.5–8.5	35–37	0,90 ($0{,}75^{B}$)	99
22	E	7.5–8.5	35–37	1,02 ($0{,}88^{B}$)	98
23	F	7.5–8.5	35–37	1,07 ($0{,}77^{B}$)	99

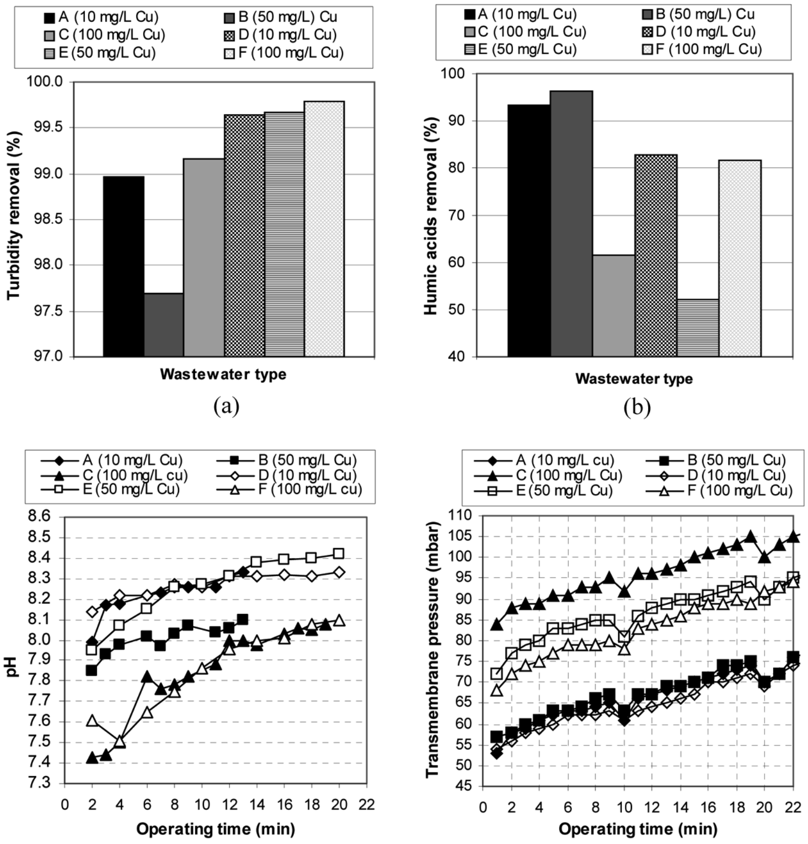
A: These values are not accurate, as in this pH region the copper remains soluble. However, it is evident that in this pH region the copper removal is practically zero.

B: The values within the brackets are the final concentration of copper when the duration of filtration was 40 min.

can successfully reduce copper concentration in each type of tested wastewaters, as the removal percentages are above 90% and particularly for wastewaters C and F are 99%. Additionally, it is shown that the initial concentration of kaolin and humic acids do not exert a significant effect on the performance of the process, although it is suggested that for kaolin and humic acids concentration equal to 50 mg/L the efficiency is slightly higher. Regarding the mechanism through which copper is removed from the wastewater, two different parameters should be taken into account. The first is the pH of the tested synthetic wastewater, which is within the range of 7.5–8.5. It is worth noting that the optimum pH for copper precipitation as a copper hydroxide is around 8.0. At the working pH copper mainly forms insoluble hydroxide, although some ionic species could also be present (34). Therefore, the reduction of copper concentration is not complete, as some ionic species can move through the MF membrane.

The second parameter is the presence of humic acids, which may facilitate the removal of copper via the formation of Cu-humic acids complexes. Then, the formed complexes can be adsorbed onto the surface of kaolin particles, forming larger agglomerates, which can be easily, removed through membrane filtration. However, it should be mentioned that at the working pH (pH = 7.5–8.5) the adsorption of humic acids onto the surface of kaolin is not highly favored (35), therefore it is suggested that probably the most dominant removal mechanism of copper is through the formation of insoluble hydroxide. The latter is partially confirmed by the observation that the increase of humic acids and kaolin concentration does not significantly affect the removal efficiency, along with the data of Table 5 (experiments 17–19). From the data of Fig. 1, it is observed that even from the first minutes of operation, the efficiency of microfiltration is relatively high, reaching the minimum value about 6 to 8 minutes after the beginning. This observation applies for the treatment of wastewaters with Cu concentration of 10 and 50 mg/L, while for the treatment of wastewaters with 100 mg/L Cu the minimum value is reached at 16 to 18 minutes.

Figure 2 shows the removal of turbidity (Fig. 2a) and humic acids concentration (Fig. 2b) that was achieved by the microfiltration performance for each tested synthetic wastewater. Additionally, Fig. 2c illustrates the pH changes during the operation of the microfiltration process, while Fig. 2d the respective alterations of transmembrane pressure.



(a) (b)

(c) (d)

FIG. 2. Comparative microfiltration performance in turbidity and humic acids removal, as well as in pH and TMP alterations.

Regarding the turbidity removal, the respective efficiencies for each wastewater treatment are very high, ranging from 97.5–99.5%, which was anticipated, taking into account the fact that the treated water was filtered through a membrane with a pore size equal to 0.4 μm. Regarding the humic acids reduction, it is observed that the efficiency of microfiltration is partially altered when treating the synthetic wastewaters with different kaolin and humic acids concentration, although the relative differences between the respective types of wastewaters are small. Figure 2c shows the evolution of pH values of the treated sample during the operation of microfiltration. It is evident that, in all cases, the pH values steadily increase about 0.2–0.5 pH units. The latter can be explained by the fact that a great amount of humic acids are held onto the membrane surface, thus reducing the acidity of the sample resulting in a subsequent increase in the pH of the permeate. Finally, the changes in transmembrane pressure are presented in Fig. 2d. It is evident that throughout the operation of the microfiltration process the pressure is steadily increasing, as it is expected. The small pressure drops recorded at certain time intervals are attributed to the backwash cleaning and the membrane relaxation that were respectively applied. Finally, regarding conductivity and alkalinity of the permeate samples, no significant variations were observed after measuring these properties throughout the operation of the process.

In order to thoroughly investigate the effect of pH of wastewater, of kaolin-humic acids concentration and of the duration of filtration upon the removal mechanism of copper, a series of microfiltration experiments were conducted by altering the appropriate parameters. In particular, the same experiments were conducted by adjusting the pH of each wastewater at pH = 6.0–7.0 and 3.0–4.0. Then, a series of blank experiments (without kaolin and humic acids) were performed at pH 7.5–8.5. Finally, some selected experiments were repeated by adjusting the duration of filtration at 40 min and simultaneously reducing the permeate flux at 35–37 LMH. Overall, the obtained results from all microfiltration experiments regarding the removal of copper are summarized in Table 5.

The data of Table 5 (experiments 1–12) demonstrate that no significant variations were recorded to the efficiency of copper removal regarding the different pH of each tested wastewater. The performance at pH 6.0–7.0 is practically the same as at pH 7.5–8.5, which is totally acceptable taking into account that both pH regions are very close to each other, therefore the removal mechanism remains the same. On the other hand, it is evident that at the pH region of 3.0–4.0 the removal of copper is not favored at all, as the final concentration of copper is at the same level as the initial concentration. The latter is attributed to the low working pH, which does not facilitate the formation of copper hydroxide, highlighting the fact that the formation of the insoluble copper hydroxide (which is favored at a pH value around 8) is the dominant removal mechanism during the microfiltration process.

This statement is further confirmed by the findings of experiments 17–19, according to which the tested samples contained no concentration of kaolin and humic acids. It is evident that the removal percentages are very similar to the ones achieved when the respective samples contained various concentrations of kaolin and humic acids. The reason for such a high efficiency is the working pH of 7.5–8.5, which favors the formation of insoluble copper hydroxide. However, some ionic species of copper could also be present (34), which justifies the fact that the reduction of copper concentration is not complete, as some ionic species can move through the MF membrane.

Finally, experiments 20–23 demonstrate that the increase of filtration time does not significantly improve the removal efficiency of microfiltration, as the removal values remains relatively steady (although slightly lower) in comparison with the respective experiments 2,3,5, and 6.

Overall, it is concluded that the critical parameter for removing copper through microfiltration is the pH of the tested wastewater. The pH range of 6.0–8.5 favors the formation of the insoluble copper hydroxide, which is the main mechanism through which copper is held on the microfiltration membranes.

Coagulation-Microfiltration Efficiency for Treating Synthetic Wastewaters

Coagulation-Microfiltration Experiments with (Ferric Sulphate, FS) and PFS-PAA

Coagulation-microfiltration experiments (with Ferric Sulphate, FS) were conducted for the following synthetic wastewaters: A, B, D, and E.

The working concentration of the coagulant was determined through preliminary experiments as follows: A – 2 mg/L as Fe, B – 2 mg/L as Fe, D – 20 mg/L as Fe, E – 10 mg/L as Fe. In all cases, an anionic polymer was added just at the end of the fast mixing stage, in a respective concentration of 1/10th of the Fe concentration.

Additionally, coagulation-microfiltration experiments (with PFS-PAA, PAA/Fe = 0.10) were conducted for the following synthetic wastewaters: A and B.

The working concentration of the coagulant PFS-PAA, PAA/Fe = 0.10 was determined through preliminary experiments as follows: A – 2 mg/L as Fe, B – 5 mg/L as Fe. The ratio PAA/Fe = 0.10 states that the concentration of the non-ionic polymer within the composition of the coagulant is the 1/10th of the respective concentration of Fe.

Figure 3 illustrates the removal of copper in terms of residual concentration (Fig. 3). It is evident that in all coagulation-microfiltration experiments, the removal of

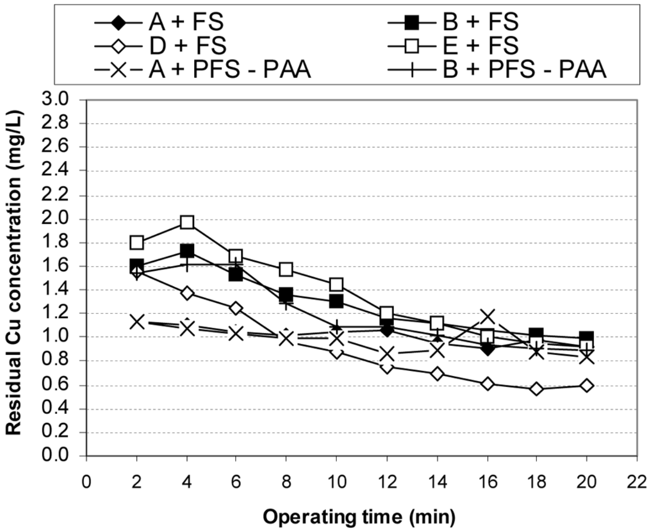
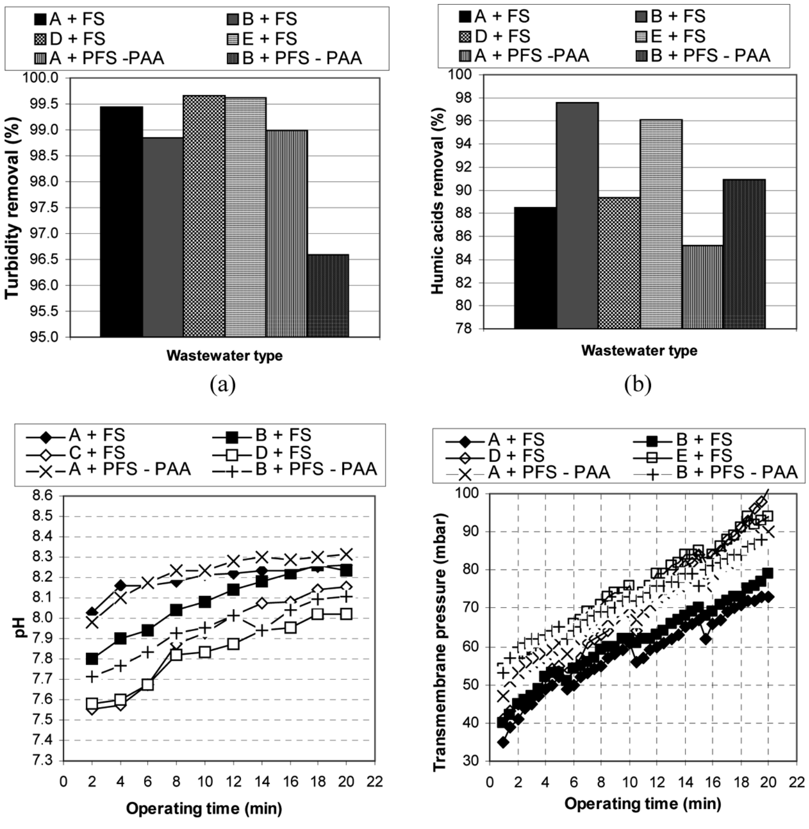


FIG. 3. Comparative coagulation–microfiltration performance in copper removal.

copper is relatively high, ranging from 91–98%. Comparing the two coagulation reagents, it is suggested that the new composite coagulant PFS-PAA is slightly more efficient than the conventional FS in treating the A and B type of synthetic wastewater. Similar to the conclusions drawn from Fig. 1, it is observed that high humic acids removal percentages are achieved when treating the synthetic wastewaters and that the differences in the initial concentration of humic acids and of kaolin particles do not significantly affect the removal of copper.

Figure 4 show the removal of turbidity (Fig. 4a) and humic acids concentration (Fig. 4b) that was achieved by the coagulation-microfiltration performance for each tested synthetic wastewater. Additionally, Fig. 4c illustrates the pH changes, while Fig. 4d illustrates the respective alterations of transmembrane pressure during the operation of the microfiltration process.

Regarding the turbidity and humic acids removal, the coagulation-microfiltration process with FS is more efficient than the respective process with PFS-PAA, although the recorded differences are relatively small. Again, it is recorded that high humic acids removal percentages are achieved when treating the synthetic wastewaters. Similar to the microfiltration process, the pH value of the permeate after the hybrid process is continuously increasing throughout the operation of the filtration. This reduction is attributed to the removal of humic acids, which takes place during the coagulation stage in the form



(a) (b)

(c) (d)

FIG. 4. Comparative coagulation–microfiltration performance in turbidity and humic acids removal, as well as in pH and TMP alterations.

of flocs and during the filtration stage, as the remaining amount of humic acids was held on the membrane surface.

Finally, the recorded values of transmembrane pressure indicate that the operation of the filtration is accompanied with a respective pressure increase. Again, the pressure drops are due to the backwash cleaning and membrane relaxation applied at specific time intervals.

Similarly, the recorded values of conductivity and alkalinity of the permeates show no significant variations.

Comparison of Microfiltration and Hybrid Coagulation–Microfiltration

For the comparison of the efficiency of the two processes, the respective performances in removing copper are evaluated and presented in Fig. 5.

Figure 5a shows the comparative performance for copper removal after treating A, B, D, and E type of wastewater, while Fig. 5b illustrates the respective performance when treating only D and E types of wastewater.

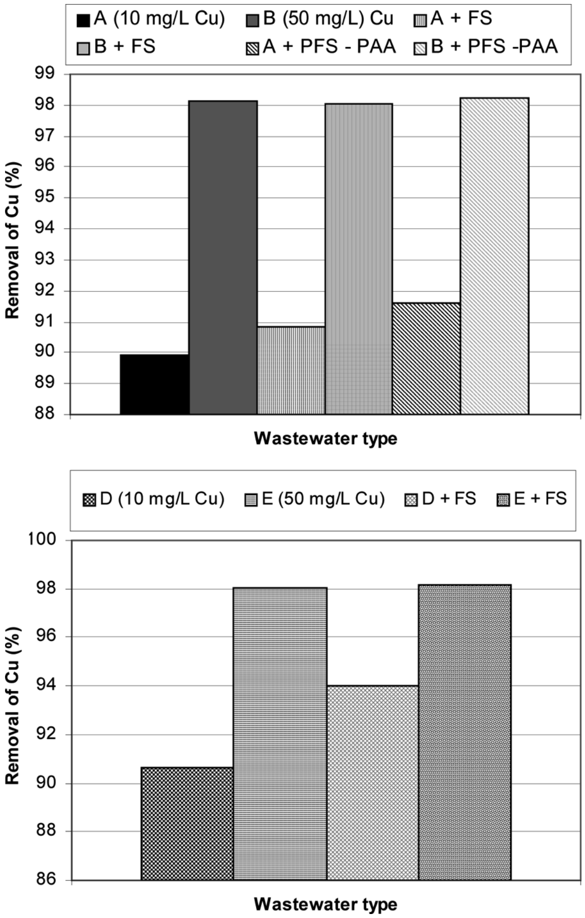


FIG. 5. Comparative performance of microfiltration and hybrid coagulation–microfiltration.

From the data of Fig. 5a, it is illustrated that the hybrid coagulation-microfiltration process is more efficient in removing copper than the simple microfiltration process, especially when treating the synthetic wastewater A. For wastewater B, the differences are relatively minors; however, the efficiency of the hybrid process with the novel composite coagulant PFS-PAA is slightly the highest. It is worth discussing about the effect that coagulation exerts upon the overall mechanism of copper removal. As it is suggested in previous studies (36,37), the negatively charged humic acids adsorb onto the surface of kaolin particles, thus increasing their negative charge. In general, the anionic character of the kaolin-humic acids suspensions increases with the concentration of humic acids added (36). However, the adsorption of humic acids is not always complete as it is strongly dependent on the pH of the suspension. At the working pH, ($pH = 7.5$–8.5) humic acids are not strongly adsorbed onto the kaolin particles and thus they do not have significant influence on their surface charge (34). In this case, the reaction mechanism can be described as follows. Once the coagulant is added into the synthetic wastewater, the iron species (i.e., Fe^{3+}, $Fe(OH)^{2+}$, $Fe_2(OH)_2^{4+}$, $Fe_3(OH)_4^{5+}$ etc.) reacted first with humic acids to form Fe-humic complexes which in turn were adsorbed onto the kaolin particles (or onto the kaolin–humic acids complexes that might exist to some extent) in order to destabilize them, thus enabling the formation of large settleable flocs. Simultaneously, the formation of copper hydroxide will enhance the formation of larger and denser flocs, as they might be adsorbed onto the surface of the flocs or they might be incorporated into the agglomerate. As a result of this, the subsequent removal of copper is enhanced.

Similar to the findings of Fig. 5a, the data of Fig. 5b demonstrate that again the hybrid coagulation–microfiltration process is more efficient to the simple microfiltration when treating the synthetic wastewaters with high concentration of kaolin and humic acids (D and E). Especially, for the initial concentration of copper equal to 10 mg/L the efficiency of the hybrid process is markedly better than the respective of microfiltration process alone.

CONCLUSIONS

Overall, it is concluded that the hybrid coagulation–microfiltration process is more efficient in removing copper than the simple microfiltration process, especially when treating the synthetic wastewater with copper concentration of 10 mg/L. The simultaneous formation of insoluble copper hydroxide and large agglomerates prior to the membrane filtration facilitates the enhanced removal of copper. Both the processes exhibit very satisfactory performance regarding copper, turbidity, and humic acids removal. Additionally, the pH values of the permeate are continuously increasing as the operation of filtration proceeds, due to the simultaneous removal of humic acids. Comparing the two coagulation reagents it is concluded that in terms of copper removal the new composite coagulant PFS–PAA is slightly more efficient than the conventional FS in treating the A and B type of synthetic wastewaters.

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