Desalination and Water Treatment
www.deswater.com

25 (2011) 170–175
January

1944-3994 / 1944-3986
doi: 10.5004/dwt.2011.1603

Recycle of alum sludge with PAC (RASP) for drinking water treatment

Lu Qi[a*], Rong Cheng[a], Hong-chen Wang[a], Xiang Zheng[a], Guang-ming Zhang[b], Gui-bai Li[b]

[a]*School of Environment & Natural Resources, Renmin University of China, Beijing 100872, China*
Tel. +86 (451) 86284452; Fax +86 (451) 86282732; email: qilu926@126.com
[b]*State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin 150090, China*

Received 20 October 2009; Accepted in revised form 19 July 2010

ABSTRACT

In this paper, a novel process of recycling alum sludge with powdered active carbon (PAC) was evaluated for drinking water treatment under various conditions. Results of this study indicated that the removal of turbidity, DOC and UV_{254} from simulated raw water by recycling alum sludge with PAC could reach up to 89.2%, 52.7% and 60.1%, respectively, which were better than that of recycling alum sludge alone, and it may be due to the adsorption of PAC which existed in mixed sludge. Turbidity of raw water had an important impact on the recycle of alum sludge with the PAC process, which is better to be applied in treating raw water with turbidity less than 100 NTU. In addition, the optimal pH for humic acid removal by recycling alum sludge with PAC was approximately 5. It was postulated that combination of adsorption and sweeping by hydroxide precipitates and the adsorption of PAC existing in mixed sludge played a key role in the enhancement of turbidity and organic matter removal.

Keywords: Recycle; Alum sludge; PAC; Adsorption; Organic matters removal; Drinking water treatment

1. Introduction

Large amounts of alum sludge are generated from water and wastewater treatment plants every day all over the world. The proper disposal, regeneration, or re-use of alum sludge (RAS) has become a significant environmental issue. Some investigators have shown that re-use of alum sludge from water treatment works may be feasible because the alum sludge contains a large portion of insoluble aluminum hydroxides that can be utilized as a coagulant in the primary sewage treatment. Recycle of the alum sludge could not only improve the particulate pollutant removal efficiency of a primary sewage treatment, but also ease the burden of water treatment works relating to sludge treatment and disposal [1].The use of RAS is a good way of removing heavy metal in wastewater and reducing the fresh alum dosage [2]. On the other hand, low turbidity source water has always been difficult to treat in the drinking water treatment. Bigger sized particles existing in alum sludge are easier to become the core of flocs in the low turbidity source water, which could improve the coagulation of low turbidity water and save a great deal of water resource simultaneously. Studies have also shown that alum sludge could be used to enhance the coagulation of low turbidity water, but the removal of organic matter is not as well as that of turbidity [3].

* Corresponding author.

For surface-derived freshwaters the removal of natural organic matters (NOM) has been proven much more difficult than that of mineral clays or other suspended particles [4,5]. Additional processes such as pre-oxidation and/or activated carbon adsorption have been applied to enhance NOM removal. Activated carbon was one of the principal means of NOM removal and its NOM adsorption mechanisms by activated carbon had also been investigated [6,7]. However, the retention time of PAC in a conventional process of water treatment plant is usually limited to only 10–20 min [8], which does not reach the adsorption equilibrium. As a result, it is not economical for the full use of the PAC adsorption.

Here a novel process of recycling alum sludge with PAC is presented and the turbidity and organic matters removal of raw water for drinking water treatment under various conditions is evaluated. The precipitated alum sludge with PAC was recycled to the flocculation tank in this process, which could reduce the dosage of fresh coagulant and the time required for floc formation, and used the adsorption capacity of PAC completely. The objectives of this study were 1) investigating the effect of recycling alum sludge with PAC (RASP) process on turbidity and organic matter (humic acid: HA) and the impact factors of this process, and 2) discussing the organic matter removal mechanism.

2. Materials and methods

2.1. Materials and reagents

HA (Sinopharm Chemical Reagent Co. Ltd, China) was obtained as a commercial reagent grade solid. Stock solution was prepared from 4 g of dry HA product, initially dissolved into 1 L of NaOH (pH 12.0) solution, and filtered by coarse and 0.45 μm membranes followed by the addition of HCl (0.1 M) to achieve pH of 7.0–7.2. The DOC of the stock solution was about 405 mg/L, measured by total organic carbon analyzer (TOC-VCPH, Shimadzu, Japan). The stock solution of natural colloids was prepared by putting clay (500 g), taken from the natural soil of 5 m below the ground, into 3000 mL tap water, followed by agitating well and taking the supernatant after settling for 1 d. The turbidity of this stock solution was about 1000 NTU. Turbidity was monitored by a turbidimeter (WGZ200, Shanghai Jinghe Inc., China). PAC (carbon size 98% < 200 mesh, Actview Carbon Technology Inc., China), which was made of wood, was prepared daily as a 1 g/L solution. Aluminum sulphate was obtained as extra pure grade (Kunshan Co., China) and prepared weekly as a 5 g/L solution and stored at 4°C.

2.2. Experimental procedure

The abbreviations of different materials and processes used in this work are listed in Table 1. A jar test apparatus (JJ-4 Scientific Flocculator, Wuxi Jianyi Co., China) was used, which had the facility to preset the stirring intensity (rpm) and time. The jar testing procedure involved a rapid mixing at 250 rpm for 30 s, followed by a slow mixing at 150 rpm for 10 min and at 50 rpm for 10 min consecutively and finally a 30 min settling. The sequence of addition of chemicals was as following: coagulant was initially added to each of the test solutions, followed by the addition of ASP and/or PAC at rapid mixing stage. Control tests without the ASP addition were also conducted in parallel. The operating conditions of different processes used in this paper are listed in Table 2. After each test, the supernatant was sampled for turbidity, DOC and UV absorbance. The UV absorbance of the samples was measured at wavelength of 254 nm (UV754, CANY, China), after being pre-filtered by 0.45 μm membrane.

Table 1
Abbreviation of different materials and processes

Material or process	Abbreviation
Powdered activated carbon	PAC
Alum sludge	AS
Alum sludge with PAC(mixed sludge)	ASP
Recycle of alum sludge	RAS
Recycle of alum sludge with PAC	RASP

2.3. Characterization of the simulated raw water and the alum sludge

The stock solutions of HA and natural colloids were added to the local (Suzhou, China) tap water to simulate a slightly polluted surface water. Table 3 summarizes the characteristics of the raw water used in this work. ASP and AS were taken from the jar test, in which a conventional dosage of PAC (10 mg/L) and aluminum sulphate (20 mg/L) were applied. Table 4 shows the characteristics of the two kinds of sludge used in this study.

All tests were carried out at a regulated room temperature such that solution temperatures were 25–26°C. Triplicate results were obtained for each experimental

Table 2
Operating conditions of different processes

Different processes	Coagulant dose (mg/L)	PAC dose (mg/L)	ASP/AS dose (mL)
RASP alone	0	0	0–120
RASP	20	0	0–120
RASP + PAC	20	10	0–120
RAS alone	0	0	0–120
RAS	20	0	0–120
RAS + PAC	20	10	0–120
Control tests	20	0 or 10	0

Table 3
Characteristics of the simulated raw water

Indexes	Raw water
Turbidity, NTU	15.21±1.00
DOC, mg/L	5.62±0.42
UV_{254}, cm^{-1}	0.185±0.011
pH	7.16±0.10

Table 4
Characteristics of the two kinds of sludge (AS and ASP)

Indexes	AS	ASP
Solid content, w/w%	0.226±0.021	0.242±0.013
Sludge content, g/L	2.276±0.230	2.452±0.212
pH	7.05±0.10	7.11±0.08
DOC, mg/L	9.83±0.30	9.67±0.19

condition and the mean result was taken, and variability within tests was typically 10% of the mean.

3. Results and discussion

3.1. Effect of ASP dosage on turbidity removal

Fig. 1 shows the effect of the RASP and RASP + PAC processes on the removal of turbidity. It was noted that RASP without PAC reduced raw water turbidity by 81.1–86.3%, compared to 76.8% of the control tests, and RASP+PAC reduced influent turbidity by 82.4–89.2%, compared to 79.1% of the control tests. The removal efficiency of turbidity reached the highest with the dose of ASP 80 mL (i.e. the proportion of recycle 8%) in both RASP with PAC and without PAC. It may be caused by the bigger sized particles existing in ASP, which increased the concentration of particles in raw water and promote the bridging and sweeping flocculation of particles. On the other hand, the removal of turbidity by RASP + PAC was better than that of RASP, which could be attributed to the fact that PAC could increase the concentration of particles and become the cores of flocculation itself to enhance coagulation. In addition, PAC was used as an effective adsorbent for organic compounds of concern in the water treatment processes due to its large surface area and high degree of surface activity [9], the adsorption of humic substances by PAC could improve the coagulation. It also could be seen from the results that the removal of turbidity became worse with the dose of PAS more than 80 ml, and this may be caused by the fact that particulates was too much to be treated by the coagulant (20 mg/L).

3.2. Effect of ASP dosage on DOC and UV_{254} removal

The dissolved organic matter removal in terms of DOC and UV_{254} by RASP and RASP + PAC processes are displayed in Fig. 2. The removal of DOC by RASP without PAC and with PAC was increased up to 27.2–42.4% and 38.5–52.7%, respectively, compared to 19.4% and 28.9% respectively in the control tests. The removal of UV_{254} was 46.2–52.7% and 53.3–60.1% respectively, which was improved by 11.3–17.8% and 6.3–13.1% compared to the control levels (34.9% and 47%). The curves of DOC and UV_{254} removal were similar as that of turbidity, which had a lowest point at the dose of ASP 80 mL (corresponding to the highest removal), and started to rise with the dose more than 80 mL. Combination of RASP and PAC could achieve a better pollution removal effect than that of RPAS alone.

All the results above indicated that RASP process could enhance coagulation and improve the removal of turbidity, DOC and UV_{254}, and there are several reasons contributed to these results. Aluminum-based coagulants

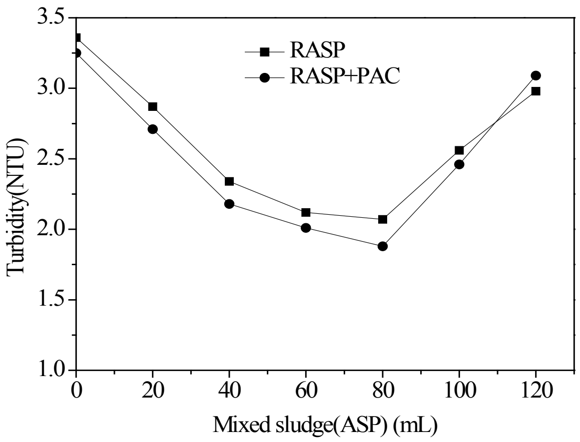

Fig. 1. Turbidity removal of RASP and RASP + PAC.

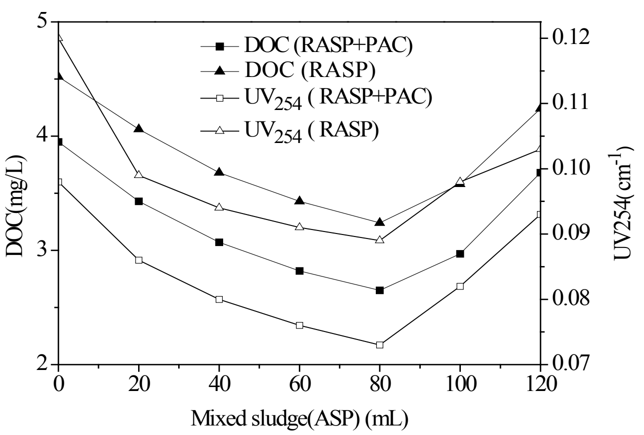

Fig. 2. DOC and UV_{254} removal of RASP and RASP + PAC.

are often used for removing NOM, especially those hydrophobic, charged and large-sized organic substances [9,10]. Under natural water treatment conditions (pH 6–8), $Al(OH)_3$ fraction dominates among the hydrolysis reaction products in water and the NOM removal efficiency is dependent on the adsorption of the humic substances on $Al(OH)_3$ crystals, and sweep flocculation plays the dominant role [11]. Hydroxide precipitates tend to have a rather open structure, so that even a small mass can give a large effective volume concentration and hence a high probability of capturing other particles [12]. It is also possible that binding of particles by precipitated hydroxide may give stronger aggregates. The removal of turbidity and DOC, enhanced by ASP was mainly due to a combination of floc sweeping and physical adsorption. ASP adsorbs and enmeshes particles (including some of the particles released from the ASP and those originally existing in the raw water) to form large floc with high settling rates. Thus, particles and organic matters could be removed more effectively by RASP than conventional process.

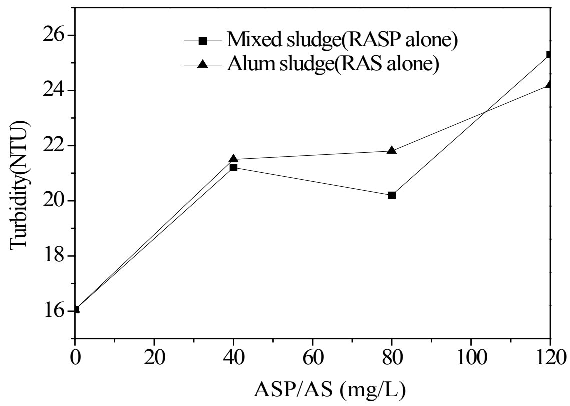

Fig. 3. Effect of RASP alone and RAS alone on turbidity.

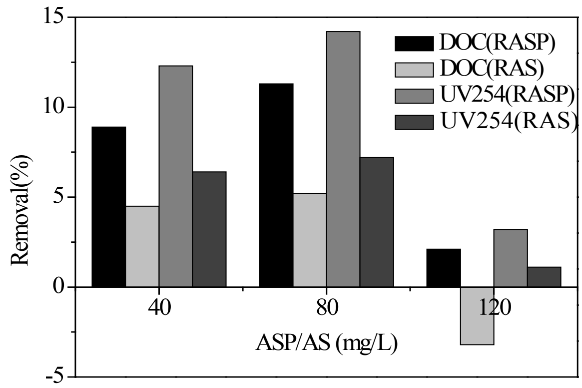

Fig. 4. Effect of RASP alone and RAS alone on DOC and UV_{254}. (ASP-mixed sludge, AS-alum sludge).

3.3. Removal of turbidity and organic matter by ASP as compared to AS

There have been many studies indicating that AS could also improve the removal of turbidity and organic matters in water and wastewater treatment [1,2], and comparison of effects on turbidity, DOC and UV_{254} between ASP and AS was investigated in the tests. The effect of RASP/RAS alone and RASP/RAS + PAC on turbidity, DOC and UV_{254} removal was compared.

3.3.1. Comparison without coagulant and PAC

The removal of turbidity of RASP alone and RAS alone is shown in Fig. 3, which indicates that the values of turbidity after jar tests became higher than the simulated raw water for both ASP and AS (40–120 mL). This was due to more hydroxide precipitates and particles than raw water existed in ASP/AS, which could not be coagulated themselves and precipitated well without the addition of coagulant.

Fig. 4 shows the removal of DOC and UV_{254} by RASP alone and RAS alone, which indicates that part of DOC and UV_{254} was removed for both two kinds of sludge. The removal of DOC and UV_{254} by ASP was better than that of AS with the same dose as ASP (40–120 mL). For instance, the removal efficiencies of DOC and UV_{254} by ASP were 11.3% and 14.2% respectively, while only 5.2% and 7.2% by AS in the dose of 80 mL. It may be the combination of adsorption and sweeping by hydroxide precipitates and the adsorption of PAC existing in ASP that make the effect of ASP on removing organic matters better than AS. There was a minus value of the DOC removal by AS (120 mL), which may be associated with the nature of the AS because some organic matters of the AS were released from the alum sludge.

3.3.2. Comparison with coagulant and PAC

The effect of RASP and RAS on turbidity, DOC and UV_{254} removal was compared in the tests (Fig. 5). The dose of ASP/AS was 80 mL, and two modes of comparison were used, i.e. RASP/RAS and RASP + PAC/RAS + PAC. As it can be seen from the results, all four treatment processes performed better than the conventional process, and there was little difference in the removal of turbidity, while there was moderate difference in DOC and UV_{254} removal. The removal efficiencies of DOC and UV_{254} by RASP were higher than those of RAS by 8.2% and 12.2% respectively, and by 9.5% and 5.9% in the condition of RASP + PAC and RAS + PAC. This implies that the effect of RASP on organic matter removal is better than RAS.

3.4. Influence of the raw water quality

In order to determine the scope of application of RASP, the effect of the raw water quality (turbidity and pH) was

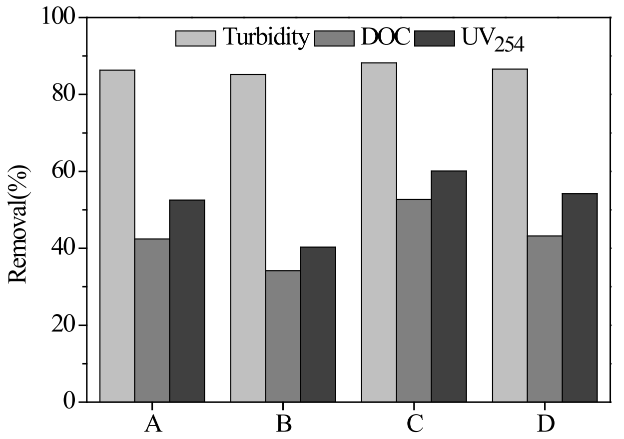

Fig. 5. Comparison of turbidity and organic matters removal by RASP, RAS, RASP+PAC and RAS + PAC (A – RASP, B – RAS, C – RASP +PAC, D – RAS + PAC).

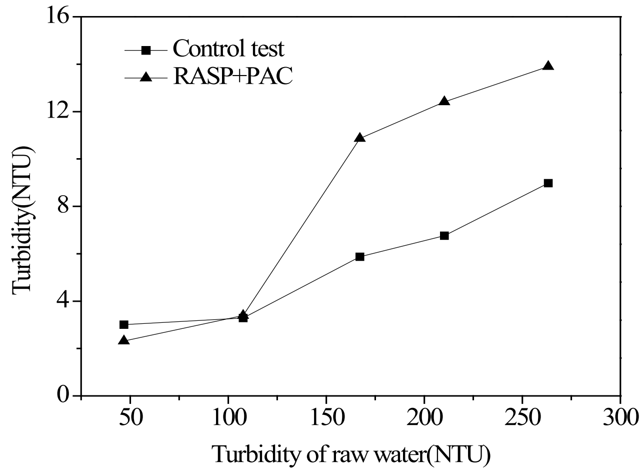

Fig. 6. Effect of the raw water turbidity on turbidity removal by RASP + PAC. (80 mL ASP was applied).

investigated. The raw waters with different turbidities were prepared by adding different dosages of the natural colloids stock solution, and the concentration of DOC kept constant with the former tests. Different pHs were achieved by the addition of HCl (0.1 M) with constant the turbidity and DOC. In these experiments, 10 mg/L PAC and 20 mg/L aluminum sulphate were applied.

3.4.1. *Effect of raw water turbidity*

The effect of raw water turbidity on the turbidity removal by RASP + PAC was examined (Fig. 6). The supernatant turbidity of the control test (conventional process + PAC) increased gradually with the increasing of the raw water turbidity, which was the same as that of the RASP test. The effect of RASP + PAC on turbidity removal was better than the conventional process + PAC when the turbidity of raw water was less than 107 NTU, and the turbidity removal would get worse than the control levels with the turbidity of raw water more than 107 NTU.

Fig. 7 shows the effect of the turbidity of raw water on DOC and UV_{254} removal by RASP + PAC. The increasing of the raw water turbidity could result in the increasing of DOC and UV_{254} of the supernatant, and the removal of DOC and UV_{254} was only better than the control levels with the turbidity of raw water less than 167 NTU. In view of this, RPAS had a better application in the treatment of the raw water with the turbidity less than 100 NTU.

3.4.2. *Effect of raw water pH*

The effects of solution pH on the removal of turbidity, DOC and UV_{254} were studied at a dose of ASP 80 mg/L, and at the following pH values: 3.1, 4.0, 5.1, 6.0, 7.1, and 9.2. The results of these tests are presented in Fig. 8. From the results, pH had little effect on the turbidity removal,

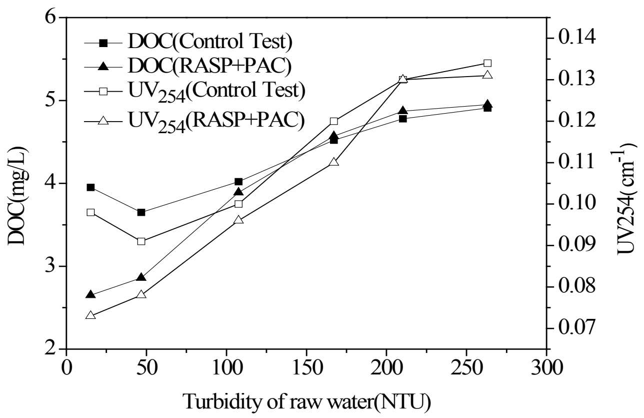

Fig. 7. Effect of raw water turbidity on DOC and UV_{254} removal by RASP + PAC. (80 mL ASP was applied).

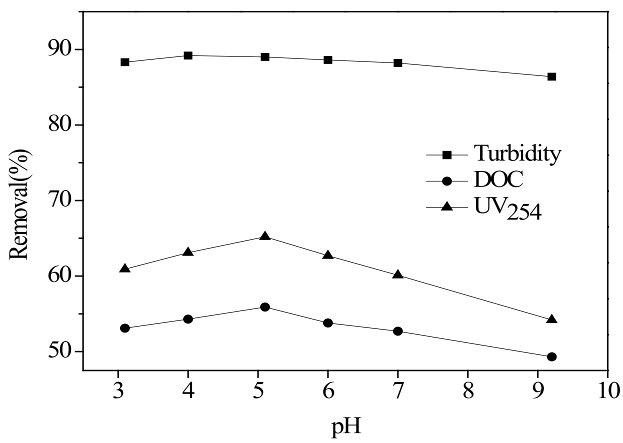

Fig. 8. Effect of pH of raw water on turbidity, DOC and UV_{254} removal by RASP + PAC.

but it was clear that the optimal pH for HA removal by ASP was approximately 5, the removal of DOC and UV_{254} was 55.9% and 65.2%, respectively. Besides, HA removal was slightly inferior at other pH values. This result is similar to some studies which have found that optimum pH for removal of dissolved organics is usually rather less (typically, pH 5–6) than that for removal of suspended particles [13]. At pH values around 5–6, the humic substances are negatively charged and Al hydroxides are positively charged, which would give strong adsorption and some charge neutralization. It is very likely that the removal of humic substances under pH 7.0 is entirely by adsorption on precipitated aluminum hydroxide. The reduction in absorbance is slightly greater at higher pH, especially at higher alum dosages [12].

This preliminary study mainly focused on the removal of turbidity, DOC and UV_{254}, and other water quality indexes were not involved. Further studies will be carried out in the pilot-scale test to investigate the removal of giardia cysts, cryptosporidium oocysts, organic matters with different molecular weight and ammonia nitrogen by RPAS in a long-term run.

4. Conclusions

Simulated raw water and ASP/AS were applied to the jar tests for evaluating the effect of RASP on the removal of turbidity, DOC and UV_{254} under various conditions. It was found that turbidity, DOC and UV_{254} removal was improved by RASP due to combination of adsorption and sweeping by hydroxide precipitates and the adsorption of PAC existing in ASP. The removal efficiency of DOC and UV_{254} by RASP without PAC was higher than RAS without PAC by 8.2% and 12.2% respectively, and by 9.5% and 5.9% with PAC, which could be related to the adsorption of PAC existing in ASP. Turbidity of raw water had an important effect on the RASP process, suggesting that RASP had a better application in the treatment of the raw water with the turbidity less than 100 NTU. In addition, the optimal pH for HA removal by ASP was approximately 5, which had slightly higher removal efficiency of turbidity, DOC and UV_{254} than that of the neutral pH.

Acknowledgements

This research is jointly supported by the Important Projects of Science and Technology for Water Pollution Control and Treatment (2008ZX07422-005), the National Science and Technology Supporting Project (No.2006BAJ08B06).

References

[1] X.H. Guan, Re-use of water treatment works sludge to enhance particulate pollutant removal from sewage, Wat. Res., 39 (2005) 3433–3440.
[2] W. Chu, Lead metal removal by recycled alum sludge, Wat. Res., 33 (1999) 3019–3025.
[3] D. Cornwell and R. Lee, Waste stream recycling: Its effect on water quality, J. AWWA, 86 (1994) 50–63.
[4] H. Selcuk, L. Rizzo, N. Nikolaou, S. Meric and M. Bekbolet, Coagulation/chlorination of surface water: a comparison between chitosan and metal salts. Separ. Purif. Technol., 62 (2007) 79–85.
[5] H. Selcuk, L. Rizzo, N. Nikolaou, S. Meric, V. Belgiorno and M. Bekbolet, DBPs formation and toxicity monitoring in different origin water treated by ozone-alum/PACl coagulation. Desalination, 210 (2007) 31–43.
[6] I.H. Suffet and M.J. McGuire, Activated Carbon Adsorption of Organics from the Aqueous Phase, Ann Arbor Science Publisher Inc., The Butterworth Group, 1, 1981.
[7] M.C. Lee, V.L Snoeyink and J.C. Crittenden, Activated carbon adsorption of humic substances, J. AWWA, 73 (1981) 440–446.
[8] AWWA, Water Quality and Treatment, A Handbook of Community Water Supplies, American Water Works Assoc., 4th ed., McGraw-Hill Inc., 1990.
[9] K.Y.J. Choi and B.A. Dempsey, In-line coagulation with low-pressure membrane filtration, Wat. Res., 38 (2004) 4271–4281.
[10] H. Humbert, H. Gallard, V. Jacquemet and J.P. Croué, Combination of coagulation and ion exchange for the reduction of UF fouling properties of a high DOC content surface water, Wat. Res., 41 (2007) 3803–3811.
[11] M. Kabsch-Korbutowicz, Impact of pre-coagulation on ultrafiltration process performance, Desalination, 194 (2006) 232–238.
[12] G. John and J.M. Duan. Hydrolyzing metal salts as coagulants, Pure Appl. Chem., 73 (2001) 2017–2026.
[13] J.J. Qin, M.H. Oo, K.A. Kekre, F. Knops and P. Miller, Impact of coagulation pH on enhanced removal of natural organic matter in treatment of reservoir water, Desalination, 149 (2006) 295–298.