Combination of powdered activated carbon and powdered zeolite for enhancing ammonium removal in micro-polluted raw water

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HIGHLIGHTS

- We assessed the removal performance of the combination in coagulation.
- The efficient molecular weight removal range was studied upon GFC.
- Synergistic effect promotes ammonium removal efficiency.
- Antagonism blocks the due removal on COD_{Mn}, but not evident on UV_{254}.
- Early phase of flocculation shall be the optimized dosing point.

ARTICLE INFO

ABSTRACT

Even zeolite is promising in ammonia pollution disposing, its removal efficiency is frequently interfered by organics. As activated carbon has good removal efficiency on organic contaminants, combination of two adsorbents may allow their respective adsorption characteristics into full play. This paper provides a performance assessment of the combination for enhancing ammonium removal in micro-polluted raw water. Gel-filtration chromatography (GFC) was carried out to quantify the molecular weight (MW) range of organic contaminants that powdered activated carbon (PAC) and powdered zeolite (PZ) can remove. The polydispersity difference which also calculated from GFC may indicate the wider organic contaminants removal range of PAC and the relatively centralized removal range of PZ. The jar tests of combination dosing confirm a synergistic effect which promotes ammonium removing. Nevertheless, it also shows an antagonism hindering the due removal performance of the two adsorbents on COD_{Mn}, while it is not much evident on UV_{254}. Furthermore, a comparison study with simulated coagulation–sedimentation process was conducted to evaluate the optimum dosing points (spatial and temporal) of PAC and PZ among follows: suction well, pipeline mixer, early and middle phase of flocculation. We suggest to dose both two adsorbents into the early phase of flocculation to maximize the versatile removal efficiency on turbidity, ammonium and organic contaminants.

1. Introduction

Ammonia pollution in raw water has been a long-standing problem in south China cities. Frequently, it results from the domestic sewage overflow (Beler-Baykal et al., 2004). High concentrations of ammonia can adversely impact water quality (Cutrofello and Durant, 2007). Among the adsorbents proposed to reduce ammonium, zeolite is deemed to be promising (Beler-Baykal and Cinar-Engin, 2007; Du et al., 2005; Malekian et al., 2011; Yusof et al., 2010). Zeolite has strong selective adsorption on ammonium for its pore diameters are similar to that of ammonium particles, as well as the hydrophilic surface and good cationic exchange ability (Wang and Peng, 2010; Wang et al., 2006; Widiastuti et al., 2011). However, its ammonium removal efficiency is ubiquitously interfered by the organic contaminants in raw water to some extent (Bautitz and Nogueira, 2007; Ben et al., 2009).

General coagulation–sedimentation treatment is usually applied to remove the organic contaminants of which molecular weight (MW) are greater than 3 kDa, whereas, it barely has

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removal efficiency on the organic contaminants which are below 1 kDa (Li et al., 2003). Therefore, it is requisite to promote the ammonium removal efficiency of zeolite through utilizing other complementary adsorbents and optimized purification processes for decreasing the interference from organic contaminants in the purification of raw water.

Activated carbon is a widely used adsorbent which has good removal efficiency on organic contaminants in aqueous or gaseous phase (Gomez et al., 2007; Kristiana et al., 2011; Leboda, 1993). However, previous studies have reported that general activated carbon does not have sufficient adsorption capacity for ammonia because it usually possesses a non-polar surface (Halim et al., 2010; Park and Kim, 2005). Theoretically, combination of activated carbon and zeolite may bring their respective adsorption characteristics and advantages into full play. Nevertheless, combination of activated carbon and zeolite involves in their respective efficient adsorption MW range, and it needs to be quantified. Possible alteration of effluent quality which results from dosing both activated carbon and zeolite in coagulation process is also still indeterminate. Additionally, ascribe to the potential complicated chemical mechanism and dynamic conditions (Cooney et al., 1999; Sprynskyy et al., 2005), different dosing points of activated carbon or zeolite in treatment process may have various influences on the final effluent quality. To our knowledge, research about these issues still remain lacking.

In this study, the performance of combination of powdered activated carbon (PAC) and powdered zeolite (PZ), with a particular emphasis on enhancing ammonium removal, was assessed. Since PAC and PZ may contain the removal capacity targeting to different MW range of organic contaminants respectively, they were quantified based on Gel-Filtration Chromatography (GFC). The MW distribution polydispersity of organic contaminants in raw water before and after treatment were also calculated. A series of jar tests were employed to detect the removal efficiency of different combination group which added with different dosage of PAC and PZ. We further simulated a successive coagulation–sedimentation process, and used the parameters and schemes which were in consistent with the actual running purification schemes in water works of Shanghai, China. The different dosing points (spatial and temporal) of PAC and PZ, i.e., suction well, pipeline mixer, early and middle phase of flocculation, were set up for comparison. Turbidity, ammonium, chemical oxygen demand (COD_{Mn}) and ultraviolet absorbance at 254 nm (UV_{254}) were detected.

2. Materials and methods

2.1. Materials

The natural zeolites used in the study were obtained from Jinyun, Zhejiang, China. Zeolites were prepared by grinding to the grain size of 200 mesh. The resulting powdered zeolites were washed with de-ionized water and dried in oven at 105 °C for 24 h. PAC (grain size: 26–35 mesh; total surface area: 500–1000 m^2 g^{-1}; relative density: 1.9–2.1 g cm^{-3}) was purchased from Sinopharm. The coagulant used in this study was liquid Poly Aluminium Chloride (abbreviated as PAX, from Sinopharm), and its dosing concentration was 18 mg L^{-1} (converted into solid PAX).

Micro-polluted raw water was collected from Lingqiao water work in Shanghai, located near Yangtze River, which had original ammonium concentration of 2.70 mg L^{-1}. The stock samples were used in the GFC experiment only. The processed solutions of micro-polluted raw water ($C_{ammonium}$ = 4.13 mg L^{-1}) which added with NH_4Cl were used in the next two experiments. Particularly, extra adding of NH_4Cl is to magnify the distinction of effluent ammonium.

2.2. Methods

2.2.1. GFC analysis

MW distribution of organic contaminants was determined in Gel-Filtration Chromatography (Shimadzu, LC-20AD). Our standards were comprised of sodium polystyrene sulfonates (used as stationary phase, from Sigma–Aldrich Corp.) and Milli-Q water (used as mobile phase, from Millipore Corp.).

Batch jar tests were conducted in a series of glass beakers. Briefly, stock samples of micro-polluted raw water were divided into 3 groups, and 1000 mL each beaker. Group B was added with 1 g PAC, Group C was added with 1 g PZ, and Group A was set as blank. All the effluents were filtrated through 0.22 μm Whatman GF/C glass fiber filter and degased before chromatograph. Other conditional variables such as stirring time (20 min), stirring speed (200 rpm), settling time (30 min), temperature (25 °C) and initial pH (7) were kept constant. We operated each jar test three times and took the average, this was similar in the jar tests of combination dosing.

MW distribution polydispersity (ρ) can be obtained using the following equation (Chln et al., 1994; Zhou et al., 2000):

$$\rho = M_w/M_n \tag{1}$$

where M_n is the number-average molecular weight, M_w is the weight-average molecular weight, and ρ is the polydispersity.

M_n and M_w can be derived from the raw data-map of GFC directly. For a pure substance, $M_n = M_w$ and $\rho = 1$; whereas, for a mixture of molecules, $M_n < M_w$ and $\rho > 1$.

2.2.2. Jar test of combination dosing

7 groups of processed solutions of micro-polluted raw water were stirred and settled successively. These batch experiments were also completed under the optimum conditions of pertinent factors as described above, which aimed at maximizing the removal efficiency (Halim et al., 2010; Huang et al., 2010). Dosage of PAC and PZ in each group was listed in Table 1. PAC dosage more than 40 mg L^{-1} can result in excessive turbidity, and it can also impact the stability of effluent quality. Effluent ammonium concentration, COD_{Mn} and UV_{254} were analyzed.

2.2.3. Influences of dosing points on effluent quality

A sequence of jar tests were employed as simulated coagulation–sedimentation process (Zeng et al., 2007; Friedler et al., 2008). All the simulated phases and parameters referred to conventional treatment scheme, as depicted in Fig. 1. The dosing points (spatial and temporal) of PAC, PZ and PAX in each scheme are explained in Table 2, in which, the fixed dosage of PAC is 40 mg L^{-1}, PZ is 0.5 g L^{-1}, and PAX is 18 mg L^{-1}. The whole process was also performed under ambient temperature (25 °C) and neutral condition (pH = 7). Dosing coagulant (PAX) into pipeline mixer is an actual operation in many water treatment plants, and it is also the same in this work. The combination schemes in Table 2 is set, following a principle that is, dosing PAC before PZ.

2.2.4. Effluent quality determination

Turbidity was measured by HACH 2100P portable turbidity meter. Ammonium concentration was characterized through the

Table 1
Dosage of PAC and PZ in each group.

	Group No.						
	I	II	III	IV	V	VI	VII
PAC (mg L^{-1})	0	0	40	40	30	20	10
PZ (g L^{-1})	0	2	0	2	2	2	2