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Effect of air bubbling on atrazine adsorption in water by powdered activated carbons – competitive adsorption of impurities

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Abstract

The effect of air bubbling on trace organic (atrazine) removal from water by powdered activated carbon (PAC) adsorption was investigated by injecting air bubbles at different flow rates into the solution as a mixing method in batch kinetic tests. The factors introduced by the use of air bubbling, such as pH increase, N_2 and O_2 competitive adsorption, mixing efficiency and contaminant release from PVC tubing were examined to identify their contributions to the change of atrazine adsorption efficiency caused by the air bubbling.

The experiments revealed that the adsorption process could benefit from higher air bubbling rates. However, under some conditions, the uptake of atrazine was partially reversed after an initial rapid uptake. Several factors that could contribute to this undesirable effect have been examined. The evidence points to the detrimental effect of trace contaminants introduced by the plastic tubing delivering the air. All other factors examined had small to negligible impact. The leaked contaminants presented increasingly in the solution with operating time and competed with atrazine solutes for the active sites in the carbon particles and even displaced adsorbed atrazine over the 6-h kinetic test. As a result, atrazine adsorption efficiency was reduced. In contrast, when stainless steel tubing was used for air injection, no reversal phenomenon was observed in the bubbling batch kinetic adsorption tests. These observations may be important for hybrid adsorption-membrane processes which could use bubbling to provide mixing and fouling control. The common practice of using plastic piping for air delivery may need to be reconsidered especially for trace pollutants removal.

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Keywords: Air bubbling; Atrazine; Powdered activated carbon; Adsorption kinetics; Competitive adsorption

1. Introduction

Activated carbon has been widely used for water purification in both industrial and municipal applications due to its large surface area per unit volume, high degree of surface activity and strong affinity for a wide range of dissolved organics. Powdered activated carbon (PAC) has further advantages of requiring low additional capital expenditure as well as having the flexibility to be applied when needed [1]. Recently, with the development of membrane technology for water and wastewater treatment, PAC adsorption can be com-

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bined with membrane units for the removal of some specific organic compounds from water. In particular microsolutes (<1000 Da), which cannot be removed by low pressure micro-filtration (MF) or ultrafiltration (UF), can be removed by the hybrid PAC-membrane process [2].

In this hybrid membrane system, PAC adsorption converts microsolutes or dissolved organic matter to a particulate phase. Microporous MF/UF membranes can then separate these fine PAC particles, with adsorbed microsolutes in the pores, from the treated water. There is a potential cost saving compared with low cutoff, high-pressure membrane processes, such as reverse osmosis or nanofiltration. In the low-pressure hybrid process, the membranes may be located externally to an adsorption tank or submerged in the tank directly. In the latter case, air bubbling could be applied to the tank to provide mixing and introduce shear force

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at the membrane surface in order to enhance separation efficiency.

Previous studies have confirmed the competency of PAC-MF/UF hybrid external systems for dissolved organic compound removal [2-6]. The combination of PAC with submerged MF membranes was also reported [7] and was used as a polishing step following conventional clarification or a single step membrane process for natural organic matter (NOM) and synthetic organic chemicals (SOCs) elimination. A significant enhancement of NOM and SOCs removal and an excellent response to variations of the feed water quality were observed at relatively low flux and transmembrane pressure (TMP). It has also been reported that microbes could be removed more efficiently by adding PAC directly into an immersed MF reactor [8]. Further, the application of submerged UF membranes integrated with PAC alone or together with an enhanced coagulation process was shown to be a promising approach for organics removal at the pilot-scale [9]. This system operated well at high PAC concentrations and led to increased adsorption and higher efficiency of total organic carbon (TOC) removal. It was noted that the membrane performance may also be improved, as the organics are preferentially adsorbed onto the PAC rather than on the membrane and a dynamic PAC barrier on the membrane surface was formed that appeared to prevent the deposition of the organic compounds.

It is evident that the previous studies of PAC-MF/UF hybrid submerged membrane systems have mainly focused on evaluation of the overall contaminant removal efficiency. The effects of air bubbling on the PAC adsorption as well as on the whole system performance were rarely reported. Nevertheless, air bubbling has been proved to be effective for a single membrane filtration process by creating gas–liquid two-phase flow which increases local mixing and mass transfer and introduces secondary flow at the membrane surface and thereby improves membrane filtration by controlling concentration polarization and membrane fouling [10,11].

The PAC-MF/UF hybrid submerged membrane system is different from a single membrane process, as the carbon particles are suspended in the adsorption/filtration tank by vertical movement of injected air bubbles. Thus the gas–liquid twophase issue in a single membrane process has been converted to a complex gas–liquid–solid three-phase problem in the hybrid submerged membrane process. How the air bubbles interact with PAC and affect PAC adsorption, which ultimately associates with the overall removal efficiency, is a critical concern in order to gain a deep insight on the performance of such a system.

Therefore, this study aims to evaluate the impact of air bubbling on the adsorption efficiency of trace organic compounds by powdered activated carbon in water. The adsorption kinetics at different air bubbling rates were compared with each other along with the results of traditional mechanical stirring as a reference. In particular, factors introduced by air bubbling which are likely to influence the adsorption process such as pH, O_2/N_2 competitive adsorptions, contaminant release from the system as well as the mixing mechanism were investigated.

2. Experimental

2.1. Materials

Powdered activated carbon (Norit SA-2) with original mass median diameter (D_{50}) of 20.0 µm was kindly provided by Behn Meyer Chemical(s) Pte. Ltd. The specification provided by the supplier is presented in Table 1. To obtain carbon particles of smaller sizes, the original PAC was pulverized using a milling machine for different times. The particle sizes of pulverized powders were measured using a particle size analyzer (Malvern Mastersizer), which measures particle sizes based on laser diffraction in the range of 0.05–550 µm. The particle size distributions of PACs pulverized for different times are shown in Fig. 1. Following preliminary tests the smallest PAC, with 6.3 µm D_{50} , was chosen as a representative adsorbent in this study. The PACs used in the kinetic tests were oven dried at 105 °C before weighing.

Atrazine, one of the most widely used herbicides, was chosen as the target compound to represent trace organics frequently found in natural waters. The U.S. Environmental Protection Agency has set an enforceable level called the maximum contaminant level (MCL) of $3 \mu g/L$ for atrazine.

Table 1

Norit SA-2	general	characteristics	
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Property	Norit SA-2	
Molasses number (EUR)	340	
Methylene blue adsorption (g/100g)	15	
Iodine number	850	
Total surface area (BET) (m^2/g)	950	
Apparent density (tamped) (kg/m ³)	460	
Particle size D_{10} (µm)	3	
Particle size D_{50} (µm)	20	
Particle size D_{90} (µm)	140	
Ash content (mass%)	9	
Moisture (as packed) (mass%)	2	

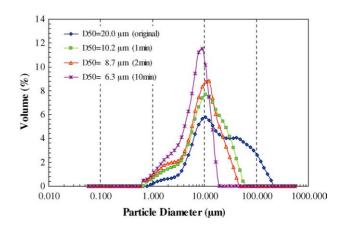


Fig. 1. Particle size distribution of PAC pulverized for different times.

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