Available online www.jsaer.com

***Journal of Scientific and Engineering Research*, 2018, 5(12):197-201**

Research Article

ISSN: 2394-2630
CODEN(USA): JSERBR

Removal of Pyridine with Peroxymonosulfate Oxidation Catalyzed by granular activated carbon

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Abstract Increasing attention has been paid to environmentally friendly activation methods of peroxymonosulfate (PMS) in advanced oxidation processes (AOPs) for organic pollutant elimination. In this study, granular activated carbon (GAC) was used as a catalyst to activate PMS to degrade pyridine in aqueous solution. The pyridine removal was much faster in the combined system than that in only GAC adsorption or PMS oxidation system. In subsequent experiments with pyridine, the removal rates for pyridine followed pseudo-first-order kinetics, with rate constant values ranging from 0.045 to 0.141 1/min depending on the operating parameters (initial PMS, pyridine concentrations and reaction temperature). Finally although GAC has almost lost its adsorption capacity, the GAC/PMS combined system could still significantly degrade AO7 as before.

Keywords Granular activated carbon; Peroxymonosulfate; pyridine; Reaction temperature

Introduction

The release of organic compounds wastewaters from the wastewater industry is a current problem encountered by developed and under developed countries over the world [1-3]. This release in natural environment, mainly in aqueous medium, is undesirable because of the potential transformation of these compounds to toxic and carcinogenic of species [4-6]. Organic compounds represent one of the large groups of these effluents. However, several treatment processes are available for the removal of this type of pollutants [7, 8].

The emission of high concentrations of organic pollutants and hazardous pollutants from the industrial production causes great harm to the environment [9, 10]. Especially, the nitrogenous heterocyclic compounds are difficult to be degraded and have inhibitive or toxic effects on biochemical reactions [11]. The presence of pyridine, as a typical nitrogenous heterocyclic compound, creates severe health hazards because pyridine is toxic, teratogenic, and higher concentration will result in weakness and ataxia [12-14].

Pyridine is often used to prepare a various products, such as pesticides, dyes, drugs, industrial solvents, rubber, etc [15]. It has many negative effects on the health of living organs, like liver, nerves, kidneys and vision system, causing headache, nausea, anorexia, insomnia and other disorders [16]. Also, it smells unpleasantly at an average concentration of 0.82 mg/L in water. Furthermore, the pyridine ring is the functioning part of many pesticides; the content of the pyridine in the atmosphere is very high [17, 18].

For instance, several decontamination methods, such as precipitation, biological treatment, coagulation, adsorption on various supports. Many advanced oxidation processes (AOPs) were also used for the oxidation of organic compounds in water [19]. These techniques (AOPs), which involve an in situ generation of highly oxidizing agents such as OH radical, have emerged as an important class of technologies to accelerate the non-selective oxidation [20, 21].

In the past years, persulfates such as peroxymonosulfate (PMS, HSO^{-}_{5}) and peroxydisulfate (PDS, $S_2O_8^{-2}$), have attracted increasing attention because they are much more stable than hydrogen peroxide [22]. Additionally, persulfates and their final product sulfate ion (SO_4^{-2}) have the least effect on native organism [23]. Recently, the system of AC combined with persulfate (PS), proposed that AC might enable the decomposition of PS with the release of organic radicals and SO^{-2}_{4} in the AC/PS system [24]. The current study presents an original approach of combining GAC and PMS to remove a model organic contaminant, pyridine, in aqueous solution. It was proved that a synergistic effect also existed in the GAC/PMS system.

Materials and Methods

All chemicals used were of analytical grade and were purchased from Sigma Aldrich Company.

Before reaction, GAC was washed with 10% dilute hydrochloric acid and distilled water by turns, and then dried at 105 °C for 24 h. The preparation process of pyridine-spent GAC was as follows: (1) GAC (1.0 g/L) was dipped into pyridine solution (200 mL, 100 mg/L) for 3-4 days, until the concentration of pyridine in the suspension stopped changing, (2) the treated GAC was then filtered from the suspension and dried at 105 °C for 24 h. All experiments were carried out using distilled water. Batch experiments were carried out in 500 mL flasks, shaken by a thermal oscillator tank, whose stirring speed was 300 rpm and temperature was kept at 25 °C. Pyridine solution (200 mL, 100 mg/L), PMS and GAC were added simultaneously at the beginning of each experiment. At pre-determined time intervals samples were withdrawn from the flask for analysis.

Results and Discussion

Pyridine can be absorbed by GAC or can be oxidized by PMS, as shown in Fig. 3. Within 120 min, only 66.9% pyridine is adsorbed by the sole GAC system and only 49.5 % is oxidized by the sole PMS system. A longer time and larger amount of GAC or PMS would be needed to completely remove pyridine by GAC adsorption or PMS oxidation [25, 26]. But nearly 98.9% pyridine is degraded by the GAC/PMS combined system in 120 min.

The reaction follows a pseudo-first-order kinetic (Fig 2). The value of k in the systems of GAC/PMS, GAC adsorption and PMS oxidation is 0.3412, 0.119 and 0.081 1/min, respectively. That is to say, there is also a remarkable synergistic effect in the GAC/PMS combined system [27]. Therefore, we can ensure that GAC is an excellent catalyst to activate PMS decomposition and the PMS decomposition reaction can induce the organic degradation at ambient temperature.

Fig. 3 shows the effect of GAC, PMS and GAC/PMS dosage on the removal of pyridine. It is clear that the rate of bleaching is enhanced as the GAC dosage increases in both the adsorption-only system and the oxidation/adsorption combined system [28, 29]. The reason may be that higher GAC surface increased the amount of active sites for adsorption and the catalytic reaction [30, 31].

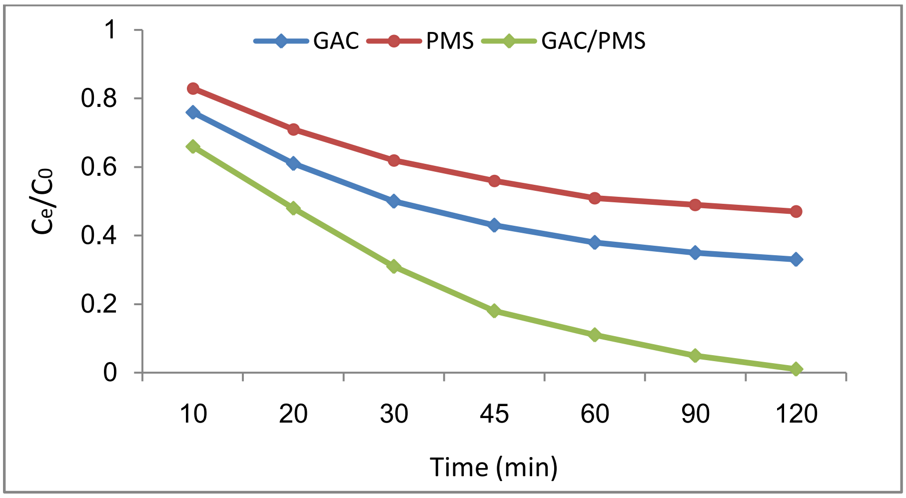

Figure 1: Removal of pyridine by only GAC and PMS and together GAC/PMS system

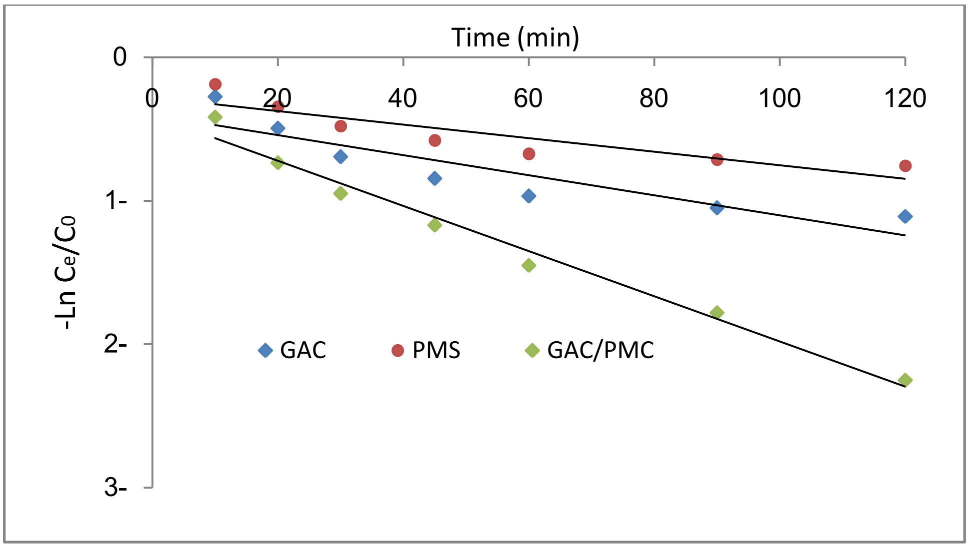

Figure 2: pseudo-first-order kinetic for pyridine removal in the GAC/PMS system

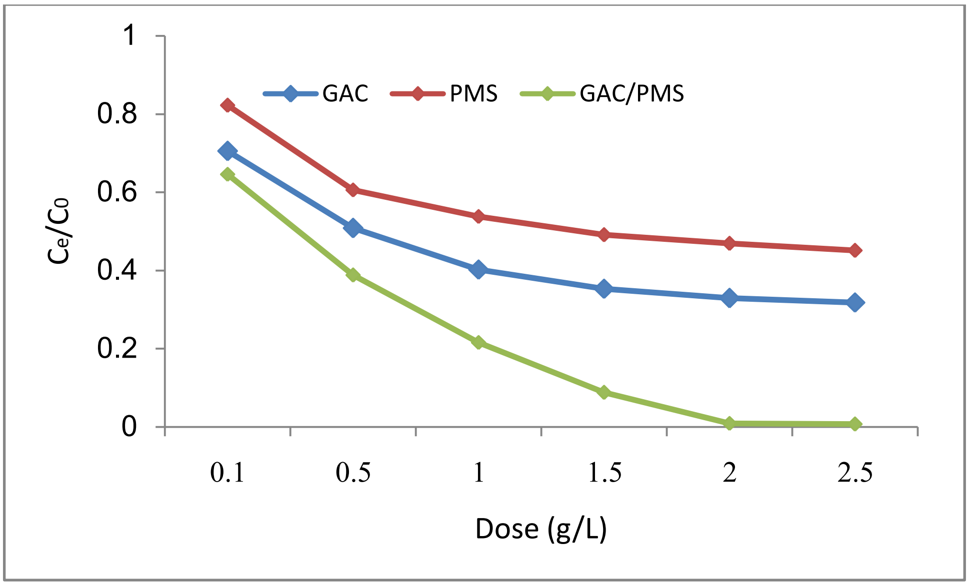

Figure 3: Effect of PMS and GAC dosage on the removal of pyridine in the GAC/PMS system

Conclusion

This study demonstrates that the combined process of PMS and GAC has much better effect on the removal of pyridine than the process of using only PMS or GAC. The reaction followed a pseudo-first-order model. The degradation efficiency increased as the PMS concentration, the GAC dosage increased. The combined system is easy to handle and GAC can sustain long-term operation, which demonstrate that GAC/PMS has a potential practical application.

Acknowledgement

The authors are grateful from Student research committee of Zahedan University of Medical Sciences because of supporting of this research.

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