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Activation of periodate by granular activated carbon for acid orange 7 decolorization

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ABSTRACT

This study investigated the activation of periodate (IO_4^-) by granular activated carbon (GAC) for acid orange 7 (AO7) decolorization at ambient temperature. It was found that there existed a remarkable synergistic effect in the GAC/ IO_4^- system. Some influential parameters, including periodate concentration, GAC dosage, initial solution pH and anions (Cl^- and CO_3^{2-}) were examined and analyzed. The results from radical scavenging experiments (with ethanol and *tert*-butyl alcohol) and electron spin resonance (ESR) analysis revealed that hydroxyl radical ($\cdot\text{OH}$) made a minor contribution to AO7 decolorization. Furthermore, the increasing formation of iodate (IO_3^-) as the reaction proceeded in the presence of excessive ethanol indirectly suggested that iodyl radical ($\text{IO}_3\cdot$) was likely to be the dominating reactive species that controlling the oxidation process. In the reuse experiments, the efficiencies of AO7 decolorization enhanced surprisingly and remained 100% when the dried-GAC (GAC collected from the former cycle experiment and dried at 105 °C for 24 h) was reused for over ten cycles, which indicated that the dried-GAC showed remarkable catalytic ability and sustainable reuse capability. Besides, a series of experiments were conducted to testify the main contributor to the strong enhancement of AO7 decolorization with the dried-GAC.

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1. Introduction

Periodate has been widely used for cellulose oxidation, in which periodate can selectively oxidize the hydroxyl groups on the cellulose chains to 2,3-dialdehyde groups by cleaving the cellulose's glucopyranoside rings between C2–C3 [1]. With this method, abundant dialdehyde groups are introduced into polysaccharides for crosslinking reaction or further modification, making the oxidized cellulose serve as intermediates for cellulose-based functional materials [2]. Due to their biodegradability and biocompatibility, these functionalized cellulosic products have a high utilization potential in papermaking [3], pharmacy [4,5], environmental protection [6] and so on.

Recently, periodate has been duly noted in advanced oxidation processes (AOPs), a technique that oxidants can be reductively converted into reactive radicals through particular reaction pathways and subsequently react with organic contaminants. Although periodate is a strong oxidant, whose oxidation-reduction potential is +1.60 V [7], it presents limited ability to oxidize organics independently. In this regard, particular interests are focused on catalyst-mediated activation of periodate and consider-

able progress has been made. Photoactivated periodate (UV/ IO_4^- system), whose mechanism was intramolecular electron-transfer reactions (Eqs. (1)–(3)) [8], turned out to be effective to generate reactive species (*i.e.* $\text{IO}_3\cdot$, $\cdot\text{OH}$, O^3P and $\text{IO}_4\cdot$) for contaminants degradation [7,9] and chemical oxygen demand decrease in industrial wastewater [10]. Subsequently, researchers attempted to introduce periodate to TiO_2/UV system and found that compared to TiO_2/UV system, there existed a significant rate enhancement in contaminant decomposition [11]. In the past decades, bimetallic nanoparticles have been employed to activate hydrogen peroxide (H_2O_2) [12] and persulfate (PS) [13] due to their excellent catalytic ability. Therefore, periodate activated by iron-based bimetallic nanoparticles (*e.g.*, $\text{nFe}^0\text{-Ni}$ and $\text{nFe}^0\text{-Cu}$) to degrade selected contaminants at neutral pH has also been investigated [14]. $\text{IO}_3\cdot$ was the predominant oxidant in the bimetallic $\text{nFe}^0/\text{IO}_4^-$ system and the formation of $\text{IO}_3\cdot$ was ascribed to one-electron reduction of periodate by $\text{nFe}^0\text{-Ni}$ or $\text{nFe}^0\text{-Cu}$ (Eqs. (4) and (5)). Study of ultrasound/periodate system showed that the degradation efficiency of perfluorooctanoic acid (PFOA) can be significantly enhanced and the possible mechanisms of PFOA degradation were both thermal decomposition and radical reactions [15]. More recently, it has been reported by Bokare and Choi [16] that singlet oxygen ($^1\text{O}_2$) instead of $\text{IO}_3\cdot$ and $\cdot\text{OH}$, was generated with a stoichiometric conversion of IO_4^- to IO_3^- in alkaline periodate solution ($\text{pH} > 8$) (Eqs. (6)–(8)). And compounds containing vicinal

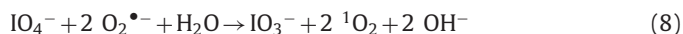
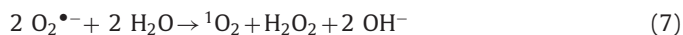
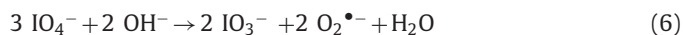
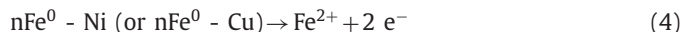
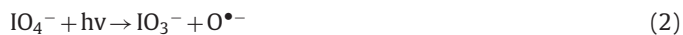
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hydroxyl groups could induce the additional generation of $^1\text{O}_2$ and further enhance the oxidation efficiency.



However, although the aforementioned activation methods enhance the reactivity, they also bring about some inevitable problems in terms of high cost, ineffectiveness, or secondary pollution. Therefore, development of less energy-consuming, efficient and environmentally friendly activators for periodate oxidation process is urgently needed.

Granular activated carbon (GAC), increasingly recognized as a multifunctional carbonaceous material, not only can be applied as an effective adsorbent for immobilization of organic pollutants in gas and water, but also as a promising catalyst or catalyst support in catalytic degradation of contaminants [17]. It is well known that its unique structure and intrinsic physicochemical properties (including surface area, porosity, surface oxygen groups and unpaired electrons) are crucial and have significant impact on its catalytic activity [18]. GACs coupled with a variety of oxidants, such as H_2O_2 and PS, have been used to degrade organic matters and regenerate the exhausted GACs simultaneously and the corresponding reaction mechanisms were proposed that oxidants could decompose over GAC surface to form highly reactive oxidizing species ($\bullet\text{OH}$ or $\text{SO}_4^{\bullet-}$) [19–21]. Comparatively, there have been no investigations focused on the catalytic activity of GAC toward periodate for the removal of organic contaminants.

Considering the oxidation capacity of periodate and the catalytic activity of GAC, we hypothesized that GAC may have potential to activate periodate. Therefore, the main objectives of this study were as follows: (1) to examine the feasibility of periodate activation by GAC and explore the effects of four parameters (periodate concentration, GAC dosage, initial solution pH and anions (Cl^- and CO_3^{2-})) on acid orange 7 (AO7) decolorization; (2) to identify primary reactive oxidants involving AO7 decolorization and conjecture the possible mechanism; (3) to testify main contributor to strong enhancement of AO7 decolorization in reuse experiments with the dried-GAC.

2. Experimental section

2.1. Materials

Sodium periodate (NaIO_4) and sodium iodate (NaIO_3) were supplied by Xilong Huagong Co., Ltd (Guangdong, China). AO7, sodium hydroxide (NaOH), sodium chloride (NaCl) and sodium carbonate (Na_2CO_3) were purchased from Sinopharm (Shanghai, China). Ethanol (EtOH) and *tert*-butyl alcohol (TBA) were obtained from J&K Scientific (Beijing, China). All chemicals were used as received without further purification. All solutions were prepared using ultrapure water with a resistance of $18.2\text{M}\Omega\cdot\text{cm}$ from a Milli-Q Integral 5 system (EMD Millipore, Billerica, MA, USA).

GAC was coal-based and manufactured by Tangshan Chemical Corporation, China. Before reaction, GAC was thoroughly immersed in 10% hydrochloric acid for 24 h and heated in boiling water for 2 h to remove fine particles and impurities, washed with ultrapure water in turn until the pH of supernatant tended to be stable, then dried at 105°C for 24 h and stored in a desiccator [22].

2.2. Experimental procedure

The experiments of GAC-activated periodate oxidation were carried out in magnetically stirred 250 ml conical flasks containing 200 ml of AO7 solution, whose temperature was kept at $25 \pm 0.5^\circ\text{C}$. Anions or scavengers (if necessary), sodium periodate, and GAC were added to 0.1 mM AO7 solution orderly and quickly at the beginning of each experiment. The initial solution pH was not adjusted and was 4.77 under the condition of periodate/AO7 molar ratio 100/1. Approximately 3 ml of sample was withdrawn at the given reaction time intervals, and the absorbance of sample was determined immediately. Control experiments without GAC or periodate were also conducted under the same reaction conditions. Note that the initial solution pH was adjusted by NaOH or H_2SO_4 in the experiments concerning the effect of initial pH on AO7 decolorization. In the reuse experiments, the dried-GAC means GAC was collected from the former cycle experiment and dried at 105°C for 24 h and then reused to the latter cycle experiment, while the wet-GAC was directly reused to the next cycle experiment. All experiments were conducted in duplicates or triplicates, and results were showed with corresponding average values and the standard deviations.

2.3. Sample analysis

AO7 concentration was immediately monitored by measuring the maximum absorbance at $\lambda = 484\text{ nm}$ with a DR6000 UV–vis spectrophotometer (HACH, USA) [23]. The filtered samples were analyzed by an ion chromatography (ICS-1100, Dionex, USA) equipped with a guard column and a separation column (Dionex-IonPacAS19 Analytical, $4\text{ mm} \times 250\text{ mm}$, USA) to measure the concentration of iodate (IO_3^-). A total organic carbon analyzer (TOC-L CPN, Shimadzu, Japan) was used to analyze total organic carbon (TOC) of the samples and further evaluate the degree of mineralization. Electron spin resonance (ESR) experiment was carried out with DMPO as a spin-trapping agent (JES-FA200, JEOL, Japan). The pH values were measured by a basic PB-10 pH meter (Sartorius Scientific Instruments, Germany).

3. Results and discussion

3.1. Decolorization of AO7 by the GAC/ IO_4^- system

AO7 was selected as a probe compound to explore the feasibility of periodate activation by GAC. Fig. 1 illustrates approximate 82% of 0.1 mM AO7 was removed by 10 mM periodate in the presence of 1.0 g/l GAC at initial pH 4.77 and 25°C within 4 h, while 35% loss of AO7 was found in control experiments without periodate owing to the adsorption on GAC, and no obvious loss of AO7 was observed in control experiments without GAC due to limited ability of periodate to oxidize AO7 independently. The simultaneous presence of GAC and periodate was far more efficient than the sole presence of either GAC or periodate means that both GAC and periodate are indispensable in the GAC/ IO_4^- system to AO7 decolorization. It can be observed in Fig. S1 that the removal of AO7 in the three systems (the sole periodate system, the sole GAC system and the GAC/ IO_4^- system) followed a first-order kinetics behavior and the rate constants were 0.0324 h^{-1} ($R^2 = 0.96$), 0.0993 h^{-1} ($R^2 = 0.95$) and 0.4217 h^{-1} ($R^2 = 0.98$), respectively. There existed

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