



## Regeneration of Rhodamine B saturated activated carbon by an electro-peroxone process



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### ABSTRACT

Due to the lack of feasible regeneration methods, spent powdered activated carbon (PAC) is conventionally disposed of by incineration or landfill after its adsorption capacity is exhausted in adsorption applications. In this study, an electro-peroxone (EP) process (a combined process of conventional ozonation and *in situ* cathodic hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) production) was proposed to regenerate PAC saturated with Rhodamine B (RhB) dye. Results show that the adsorption of RhB onto PAC involved primarily irreversible chemisorption. Therefore, the RhB-saturated PAC could not be regenerated upon dilution by water washing. In contrast, by oxidatively converting irreversibly sorbed RhB to more desorbable oxidation by-products (e.g., carboxylic acids), both ozonation and the EP regenerations could restore >90% of the adsorption capacity of the PAC for RhB reloading. However, ozonation regeneration could not effectively mineralize the desorbed pollutants due to the selective oxidation characteristics of ozone (O<sub>3</sub>). In contrast, due to the substantial generation of hydroxyl radicals (•OH) from the reaction of bubbled O<sub>3</sub> with electro-produced H<sub>2</sub>O<sub>2</sub>, the EP regeneration was capable of completely mineralizing the desorbed pollutants. During the EP regeneration, PAC was oxidized to some extents, thus gradually losing its adsorption capacity. By optimizing the applied current, ozone dose, and regeneration time, ~76% of the adsorption capacity of the virgin PAC could still be retained after five cycles of RhB adsorption and the EP regeneration. The results of this study indicate that the EP process may provide an environmentally-friendly and sustainable way to regenerate spent PAC for reuse.

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## 1. Introduction

Adsorption onto activated carbons (ACs) is an established technology to remove pollutants (e.g., organic contaminants (Danmaliki and Saleh, 2016; Saygili and Guzel, 2016; Saygili et al., 2015) and heavy metals (Saleh, 2015, 2016; Saleh et al., 2017b, c)) from aqueous media. Currently, the most commonly used ACs in water and wastewater treatment are granular activated carbon (GAC) and powdered activated carbon (PAC). Due to its smaller particle size, PAC usually exhibits higher adsorption kinetics than GAC. In addition, PAC can be dosed directly to water at various places in water treatment train (e.g., water intake and coagulation

facilities) (Clifford et al., 1983; Crittenden et al., 2012), whereas GAC needs specialized contactors to operate in a fixed-bed mode (Saleh et al., 2017d). Therefore, PAC usually requires lower capital costs and has greater flexibility in dosing (e.g., for seasonal and emergency applications) compared to GAC (Crittenden et al., 2012). However, the regeneration of spent PAC is much more difficult than that of GAC (Zanella et al., 2014). While thermal regeneration has been successfully applied to regenerate spent GAC at industrial scale, it is generally considered an infeasible way to regenerate PAC due to various reasons (Clifford et al., 1983; Salvador et al., 2015). For instance, fine PAC particles can be easily blown away by the heating gas during thermal regeneration, thus resulting in considerable PAC loss.

Due to the lack of feasible regeneration methods, PAC is conventionally used only once during water and wastewater treatment, then will be disposed of by incineration or landfill. It is well-known that PAC production is an energy intensive and

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polluting process, which involves high temperature operations and the uses of polluting chemicals (e.g., acids and bases) (Kundu et al., 2015; Saleh et al., 2017a; Saygili and Guzel, 2016; Saygili et al., 2015). Therefore, developing suitable technologies to regenerate spent PAC for reuse may offer an environmentally-friendly and sustainable way to save the valuable adsorbent material and reduce the carbon footprint related to PAC production.

Over the past several years, several research groups have tried to regenerate spent PAC by a variety of desorption- and decomposition-based technologies such as acid/alkali treatment (Li et al., 2015a), microwave regeneration (Ania et al., 2007; Li et al., 2014b), and catalytic oxidation (Do et al., 2011; Park et al., 2010; Yap and Lim, 2012). Some promising results have been obtained. For example, Ania et al. (2007) reported that high regeneration efficiencies (>95%) could be obtained for a salicylic acid-saturated PAC by microwave regeneration under 850 °C and CO<sub>2</sub> atmosphere. In addition, some researchers showed that by impregnating PAC with photocatalysis (e.g., titanium dioxide), visible-light and UV irradiation could restore up to ~50% (Yap and Lim, 2012) and ~90% (Park et al., 2010) of the adsorption capacity of bisphenol A-saturated PACs, respectively.

However, there are also some limitations that have yet to be overcome for PAC regeneration (Salvador et al., 2015; Zanella et al., 2014). For desorption-based regeneration processes (e.g., acid/alkali treatment and cathodic regeneration), they can usually effectively desorb weakly sorbed (physisorbed) pollutants from ACs, but are less efficient at desorbing irreversibly sorbed (chemisorbed) pollutants (Li et al., 2015a; Narbaitz and McEwen, 2012; Zanella et al., 2014, 2016). Moreover, the desorption-based regeneration processes cannot mineralize the desorbed pollutants, which therefore still require further treatment. On the other hand, while the sorbed pollutants can usually be degraded (or mineralized) during decomposition-based regenerations, ACs can also be oxidized to some extents (Ania et al., 2007; Clifford et al., 1983; Salvador et al., 2015; Zanella et al., 2014). The oxidation of AC would result in a decline in the adsorption capacity of ACs during reuse. Moreover, the leaching and deactivation of impregnated catalysts are problematic for the use of PAC/catalyst composites in practical water treatment (Do et al., 2011). These results indicate that more studies are needed to further improve the performance of PAC regeneration.

In a recent study (Zhan et al., 2016b), we have tested the regeneration of phenol-saturated activated carbon fiber (ACF) by an electro-peroxone (EP) process (Yuan et al., 2013), which couples the desorption-based cathodic regeneration with the decomposition-based ozone regeneration to enhance the ACF regeneration performance. For the EP regeneration, the phenol-saturated ACF was attached to a carbon-polytetrafluoroethylene (carbon-PTFE) cathode. A current was then applied to induce both the cathodic desorption of physically sorbed phenol and electro-generation of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) from excess oxygen (O<sub>2</sub>, Eq. (1)) in the bubbled ozone generator effluent (an O<sub>2</sub> and O<sub>3</sub> gas mixture) (Xia et al., 2017). The *in-situ* generated H<sub>2</sub>O<sub>2</sub> can then diffuse into the solution and react with the bubbled O<sub>3</sub> to produce the powerful oxidant hydroxyl radicals (•OH) (Eq. (2), i.e., the commonly referred peroxone reaction (von Sonntag and von Gunten, 2012)). Due to the oxidation with O<sub>3</sub> and •OH, irreversibly sorbed phenol (which cannot be desorbed by the cathodic desorption) can be converted to desorbable oxidation by-products (e.g. carboxylic acids). Consequently, the EP regeneration considerably improved the regeneration efficiency (RE) of phenol-saturated ACF compared to the cathodic regeneration (~92% vs. 68%).



In addition, due to the powerful oxidation capacity of •OH, the desorbed pollutants (phenol and its oxidation products) could be completely mineralized during the ACF regeneration. This eliminates the risks of the desorbed pollutants in the regeneration effluent.

Furthermore, the close contact between the ACF and the cathode can provide a cathodic protection to minimize the oxidation of ACF by O<sub>3</sub> and •OH during the EP regeneration process (Zhan et al., 2016a, 2016b). Consequently, the EP regenerated ACF retained still ~72% of the adsorption capacity of the virgin ACF after twelve cycles of phenol adsorption and the EP regeneration (compared to ~48% after only one cycle of ozonation regeneration (Zhan et al., 2016b)). Compared with the more conventional regeneration technologies, the EP process can thus considerably extend the lifetime of ACF for adsorption applications.

Based on the promising results obtained for ACF regeneration, this present study aimed to extend the application of the EP process to PAC regeneration. It should be noted that PAC has a very different morphology from ACF. ACF is highly microporous materials and its micropores are directly connected to the external surface (Dąbrowski et al., 2005). In contrast, PAC has a more complex pore structure that consists of interconnected macropores, mesopores, and micropores (Crittenden et al., 2012). Sorbates have to diffuse through macropores and mesopores of PAC before they can reach and be adsorbed in the micropores. Therefore, adsorption (and desorption) involves much higher mass-transfer resistance for PAC than for ACF (Crittenden et al., 2012; Dąbrowski et al., 2005). Moreover, unlike ACF cloth, which can be easily attached to the cathode, the fine particles of PAC cannot be kept in close contact with the cathode, but mainly suspend in water during the EP regeneration. Therefore, the benefits of cathodic desorption and cathodic protection, which play an important role in ACF regeneration (Zhan et al., 2016a), cannot be exploited during the EP regeneration of spent PAC. How these differences would affect the performance of PAC regeneration (e.g., the regeneration efficiency and oxidation of PAC) requires further evaluations.

The main objective of this study was to evaluate the feasibility of the EP process for the regeneration of PAC saturated with irreversibly sorbed organic pollutants. Rhodamine B (RhB) was selected as a model adsorbate because it is often detected at high concentrations in many industrial wastewaters (e.g., textile, leather, and food industries) (Gad and El-Sayed, 2009; Tian et al., 2016). RhB is known to have potential toxicity, mutagenicity, and carcinogenicity (Bai et al., 2011; Kadirvelu et al., 2005). Adsorption has often been employed to remove RhB from aqueous solutions, and the results indicate that adsorption of RhB onto ACs involves primarily strong chemisorption (Gad and El-Sayed, 2009; Kadirvelu et al., 2005; Zhuang et al., 2009). Therefore, RhB-saturated ACs cannot be effectively regenerated by simple desorption-based techniques (Gad and El-Sayed, 2009), but has to involve decomposition-based techniques. In this study, the regeneration of RhB-saturated PAC by water washing, electrolysis, ozonation, and the EP process were compared. The regeneration efficiencies (REs), textural and chemical changes of the PAC, and mineralization of desorbed pollutants were evaluated for the different processes. The effects of the main operational parameters (ozone dose, applied current, reaction time, and regeneration cycles) of the EP regeneration were evaluated systematically.

## 2. Materials and methods

### 2.1. Materials and reagents

A commercial PAC purchased from Tianjin Guang Fu Technology

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