



Editor's choice paper

Synergetic decomposition performance and mechanism of perfluorooctanoic acid in dielectric barrier discharge plasma system with Fe₃O₄@SiO₂-BiOBr magnetic photocatalyst

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ABSTRACT

The Fe₃O₄@SiO₂-BiOBr (FSB) magnetic composite was prepared and introduced into the dielectric barrier discharge (DBD) system as a heterogeneous Fenton-like photocatalyst for synergistic degradation of perfluorooctanoic acid (PFOA). The FSB was characterized by X-ray diffraction, UV–vis diffuse reflectance spectra, energy dispersive X-ray spectroscopy and scanning electron microscope. Characterization results demonstrated that BiOBr was successfully coated on Fe₃O₄@SiO₂, forming FSB, and it exhibited good UV & visible response. The presence of FSB accelerated PFOA degradation: PFOA removal efficiency and total organic carbon removal from DBD-FSB system increased from 73.5% to 92.9% and 28.9% to 62.5%, respectively, within 60 min under reaction conditions of 20 mg L⁻¹ PFOA, initial pH 4.28, 100 mg L⁻¹ FSB and 22 kV peak voltage, as compared with DBD system. Accordingly, defluorination efficiency of PFOA and energy efficiency increased from 21% to 32.8% and 46.39 mg kW⁻¹ h⁻¹ to 72.47 mg kW⁻¹ h⁻¹, respectively. The synergetic mechanism was attributed to the combination of active species such as •OH, H₂O₂, O₃, directly generated from the DBD plasma and a great amount of •OH generated from Fenton-like reaction initiated by FSB photocatalyst under light irradiation emitted during discharge. The •OH and photogenerated holes played essential roles in the mineralization process. Based on the identified intermediates, possible degradation pathways of PFOA in DBD-FSB system were proposed, and PFOA degradation mainly occurred via the pathway of perfluoroalkyl radical pathway.

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1. Introduction

Perfluorooctanoic acid (PFOA) is one of the typical perfluorinated compounds, with characteristics in chemical and thermal stability due to the strong C-F bonds [1]. PFOA has been widely used in many industry fields as surfactants, emulsifying agent and fire retardants for several decades. As a consequence, PFOA has been frequently detected in environment media such as river [2], wildlife and human blood [3–5], and it presents potential hazard to human health and earth environment due to its bioaccumulation and biotoxicity. Thus, it's urgent to seek an effect technology for decomposing or converting PFOA to environmentally harmless compound.

Advanced oxidation processes (AOPs), such as photocatalysis [1,6,7], electro-Fenton [8] and photo-Fenton [9] technologies, which can produce active free radicals like •OH, have been widely employed to degrade PFOA in aqueous solution. Furthermore, Schroder and Meesters [10] reported that O₃ could not degrade perfluorooctane sulfonates (PFOS) at pH 11. However, Lin et al. [11] found that PFOA and perfluorooctane sulfonate (PFOS) could be decomposed by O₃ at pH 11 when the parent compounds were pretreated by 15 min of ozonation at ambient pH 4–5, which was attributed to the production of abundant of •OH. Thus, it's feasible for decomposition PFOA on the condition that the amount of •OH is large enough. In photocatalysis process, PFOA could be degraded due to the formation of hydroxyl radicals (•OH) under ultraviolet irradiation (UV); however, long reaction time is required [6]. For Fenton technology, •OH is also generated from H₂O₂ decomposition, where quantity of extra H₂O₂ is required. Therefore, a new technology that possesses both photocatalysis and Fenton reaction

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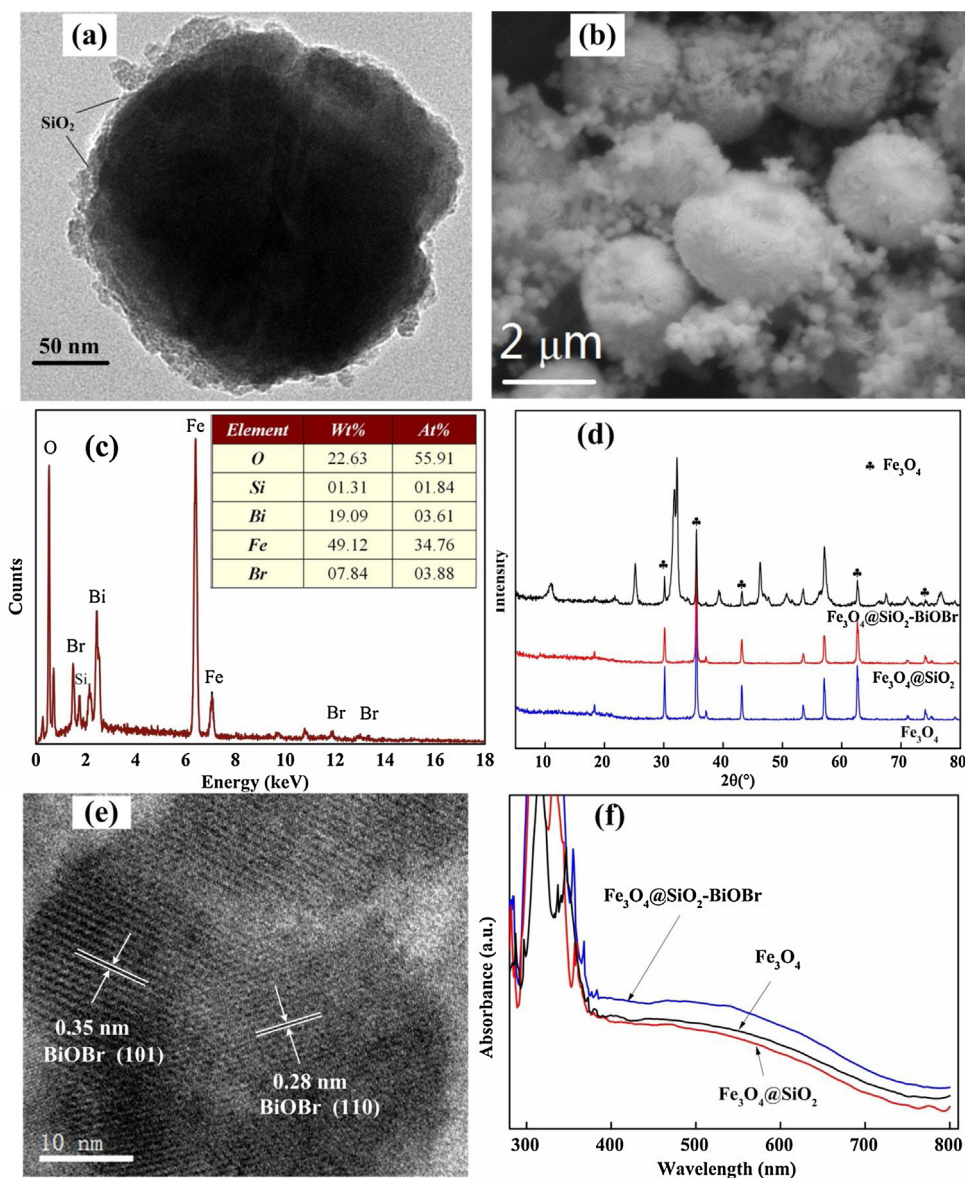


Fig. 1. TEM of $\text{Fe}_3\text{O}_4/\text{SiO}_2$ (a), SEM of FSB (b), EDS of FSB (c), XRD patterns of samples (d), HRTEM of BiOBr (e), and UV-vis DRS of samples (f).

should be an efficient way to degrade PFOA, especially requiring none of additional UV or H_2O_2 .

Dielectric barrier discharge (DBD) non-thermal plasma technology is an effect treatment method for decomposition of organic compounds, because a mass of active species (e.g. $\cdot\text{OH}$, $\cdot\text{OOH}$, $\cdot\text{O}$, H_2O_2 , O_3 , etc.) could be generated and ultraviolet light could be emitted via collisions between high energy electrons and neutral molecules [12–15]. DBD plasma has been employed to decompose organic compounds, such as 17 β -Estradiol [16], triclosan [15], 4-chlorophenol [17] and pentachlorophenol [18]. These previous reports suggested that it should be a feasible method for PFOA removal with DBD plasma discharge. In particularly, researchers reported that of UV-vis light (with the wavelengths between 200–500 nm) were emitted from plasma discharge [19–23]. It has been reported that with the presence of TiO_2 in the DBD system, phenol degradation was improved by utilization of UV light [23,24]. In our previous study, it was also confirmed that the removal efficiency and degradation yields of 17 β -Estradiol were obviously increased from 72.0% to 98.9% and 329.2×10^{-6} – 468.1×10^{-6} g/kWh by adding Pt- TiO_2 to DBD system due to the utilization of UV light [25]. All these revealed that UV light plays an important role in light

radiation emitted during plasma discharge process [20,23]. However, as is known, TiO_2 has no response to visible light irradiation; thus, BiOBr would be an appropriate semiconductor in DBD system since it is UV & visible-light-responsive [26–29]. Based on our previous study, it is noteworthy that the solution pH would decrease to below 4.0 quickly accompanied with H_2O_2 production during the DBD plasma process [30], which provides the perfect condition for Fenton reaction as long as iron was introduced. In this regard, Fe_3O_4 would be a suitable catalyst in view of recycling problem [31,32]. In consequence, the composite material of Fe_3O_4 and BiOBr would be an appropriate catalyst in DBD system. In addition, an “insulation” layer of SiO_2 would be necessary in order to prevent oxidation of Fe_3O_4 [33] and to avoid the recombination of e^- and h^+ from BiOBr in Fe_3O_4 [34,35]. Hence, $\text{Fe}_3\text{O}_4/\text{SiO}_2$ -BiOBr would be a desirable catalyst in DBD process.

In this study, a synergistic DBD system was set up to improve degradation efficiency and degradation yield of PFOA by adding the $\text{Fe}_3\text{O}_4/\text{SiO}_2$ -BiOBr to efficiently utilize UV & visible light and induce Fenton-like reaction. The effects of key parameters such as peak voltage, initial pH value, and $\text{Fe}_3\text{O}_4/\text{SiO}_2$ -BiOBr dosage on degradation performance were investigated. Furthermore, the

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