



Review

Photocatalytic removal of perfluoroalkyl substances from water and wastewater: Mechanism, kinetics and controlling factors



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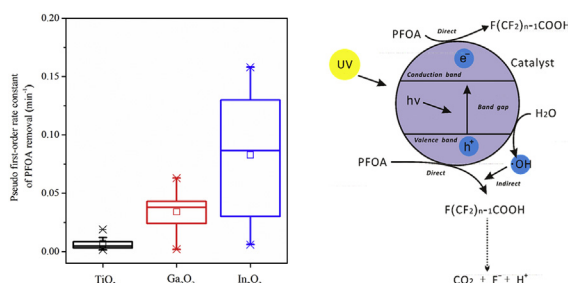
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HIGHLIGHTS

- Heterogeneous photocatalysis is an effective technology for PFAS degradation.
- Photodecomposition of PFAS in water follows the order: $\text{In}_2\text{O}_3 > \text{Ga}_2\text{O}_3 > \text{TiO}_2$.
- Photocatalysis performs well in acidic medium and increased with temperature.
- Dissolved organic matter reduces photocatalytic performance in wastewater.
- In_2O_3 nanoplates degrade 100% PFAS under UV light.

GRAPHICAL ABSTRACT



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ABSTRACT

This review focuses on heterogeneous photocatalysis of perfluoroalkyl substances (PFAS) which are of worldwide concern as emerging persistent organic contaminants. Heterogeneous photocatalysis is an effective and advanced technology for PFAS removal from water with relatively high efficacy. During photocatalysis, various short chain perfluorocarboxylic acids (PFCA) are produced as intermediates and the efficacy is related to the photo-generated hole (h^+) and photo-generated electron (e^-). PFAS photodegradation in water under UV irradiation is most effective by using In_2O_3 as the catalyst, followed by Ga_2O_3 and TiO_2 . Significantly, modifying the chemical composition or morphology of the catalyst can improve its efficacy for PFAS removal. In_2O_3 porous nanoplates were found to have the best performance of 100% PFAS decomposition under UV light with rate constant (k_t) and half-time ($\tau_{1/2}$) of 0.158 min^{-1} and 4.4 min, respectively. Catalysts perform well in acidic solution and increasing temperature to a certain extent. The photocatalytic performance is reduced when treating wastewater due to the presence of dissolved organic matter (DOM), with the catalysts following the order: needle-like $\text{Ga}_2\text{O}_3 > \text{In}_2\text{O}_3 > \text{TiO}_2$. Future studies should focus on the development of novel photocatalysts, and their immobilization and application for PFAS removal in wastewater.

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1. Introduction

Perfluoroalkyl substances (PFAS) are anthropogenic compounds that have been used in the industrial, military, and commercial applications since the 1950s (Houtz et al., 2013; Kotthoff et al., 2015). These chemicals are very stable as carbon-fluorine (C–F) bond is very strong with a bond energy of 544 kJ/mol, clearly indicating their persistent and bioaccumulative nature in the environment (Wang et al., 2008). PFAS have drawn increasing attention in recent years mainly because they are potential reproductive and developmental toxins, endocrine disrupters, and carcinogens (Ding and Peijnenburg, 2013; Gorrochategui et al., 2014; Lau et al., 2007), although the relationship between PFAS and particular human disease is not established yet. It is a fact that PFAS exist not only in different environmental media but also in human body (Table S1). Well-known PFAS such as perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) (molecular structure shown in Fig. S1) both commonly constitute high proportion of PFAS that have been detected in the aquatic environment (Estrellan et al., 2010; Yates et al., 2014). The source of PFAS in the environment can be divided into direct and indirect pathways (Wang et al., 2008). The direct route represents the emission from the manufacture and use of PFAS, or wastewater treatment plants (WWTP) (Armitage et al., 2006; Gallen et al., 2014). The indirect route refers to the long-range transport and degradation products from precursor compounds such as fluorotelomer alcohols and perfluorooctane sulfonyl fluoride (Armitage et al., 2006, 2009). Furthermore, it was reported that the direct route is accounting for more than 80% of the total source for PFAS in the environment (Pistocchi and Loos, 2009; Post et al., 2012). The transport of such compounds could also be separated as atmospheric and aqueous transport pathways (Pistocchi and Loos, 2009; Prevedouros et al., 2006). The atmospheric transport involves that PFAS diffuse in the atmosphere and then deposit on land or in water. The aqueous transport suggests the dissolution in water and transport by the currents to the other water matrices. The contamination of PFAS in the aquatic environment has been widely reported. In Spanish rivers, PFAS were found to be 43 ng/L (Flores et al., 2013). Zhang

et al. (2013) detected 16 PFAS in WWTP in China, with concentrations ranging from 0.04 to 91 ng/L in the influent. In addition, high PFOA concentration was detected in both the influent (2–91 ng/L) and effluent (3–107 ng/L) from WWTP. Moreover, serious PFAS water contamination was reported for the industrialized cities such as Dalian, China (Chen et al., 2017) and Tokyo, Japan (Murakami et al., 2013). Furthermore, as high as 68.6 ng/L of PFOA was detected in the Korean coastal water (Naile et al., 2010). Overall, water contamination by PFAS is widespread demonstrating a variety of sources and potential persistence.

Over the years, the technologies for PFAS removal from water have been developed with various decomposition efficacies due to the physical and chemical stability of C–F bonds (Cao et al., 2010; Chen et al., 2015). Treatment by sorption, filtration, biodegradation, and membrane bioreactor (MBR) were evaluated at bench scale (Appleman et al., 2014; Kwon et al., 2017; Yu et al., 2014). Nevertheless, sorption and filtration are the non-destruction methods that just transfer the contaminants from one medium to another and the waste still needs to be further managed (Urriaga et al., 2015). As for MBR and biodegradation, these methods have been proven to be inefficient for PFAS removal in many WWTP (Kwon et al., 2017; Yu et al., 2009, 2014). For example, when investigating MBR performance for the removal of PFOA and PFOS, Yu et al. (2014) reported that the removal efficiencies of these two compounds were less than 7%. This is probably because most microorganisms do not have the ability to decompose PFAS in water. Thus, technologies that cleavage C–F bonds are more suitable for PFAS removal. As an alternative, photolysis is recognized as an appealing option, which could degrade a variety of toxic substances e.g. agrochemicals in the presence of artificial or solar light, and is environmentally friendly and cost effective (Kitsiou et al., 2009; Konstantinou and Albanis, 2003; Toepfer et al., 2006). However, due to the strong bonding energy in C–F, direct photolysis for the removal of PFAS from water is still of low efficacy. Giri et al. (2011) used UV light (254 nm wavelength) for the photodegradation of PFOA in dilute aqueous solution, and observed no removal of PFOA within the reaction period of 300 min. This is mainly attributed to the fact that the bond dissociation energy of C–F bonds is up to

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