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Combined membrane photocatalytic ozonation and wet absorption of elemental mercury

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ABSTRACT

Membrane photocatalytic ozonation coupled with wet absorption offers potential for elemental mercury (Hg⁰) removal. This study reports on a novel Fe–TiO₂-coated polyvinylidene fluoride (PVDF) wet photocatalytic membrane reactor (WPCMR) for mercury removal in flue gas. Hg⁰ removal efficiency in the WPCMR reached up to 93.3%. Ozone could enhance mercury oxidation in WPCMR. Wet absorption helps to increase mercury removal efficiency. Fe–TiO₂ catalyst was synthesized by sol-gel method and characterized by XRD, FTIR, UV–Vis, XPS and SEM. XPS analysis confirmed Hg⁰ oxidation to divalent mercury (Hg (II)). Elemental mercury was oxidized to mercuric oxide followed by wet absorption in the presence of OH free radical and ozone. Wet photocatalytic membrane reactor and photocatalytic membrane reactor (PCMR) of elemental mercury reaction with the Fe–TiO₂/PVDF catalyst all follow Langmuir–Hinshelwood kinetics.

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1. Introduction

Mercury emission has become a global issue due its toxicity, volatility, persistence and bioaccumulation (Driscoll et al., 2013), which generated from coal-fired power plants, industrial boilers, metal smelting and cement kiln (Pirrone et al., 2010). Mercury emission from coal-fired flue gas often presents in three main forms: elemental mercury (Hg⁰), oxidized mercury (Hg (II)), and particulate mercury (Hg^P) (Zygarlicke, 2000). Hg^P can be effectively removed by particle matter capture device, Hg (II) can be absorbed by aqueous solution (Helble, 2000). Hg^P and Hg (II) can be captured by wet flue gas desulfurization (WFGD). (Pavlish et al., 2003). However, Hg⁰, accounting for over 64–90% in flue gas, is the most difficult to be captured with the existing air pollution control devices (Wang et al., 2010). Thus, Hg⁰ oxidation to water soluble Hg (II) followed by flue gas desulfurization scrubber was a promising solution to control Hg⁰ emissions (Pavlish et al., 2003).

Extensive research have been undertaken using traditional catalytic oxidation (Zhao et al., 2015), membrane catalytic

oxidation (Guo et al., 2012) and photocatalytic oxidation to reduce Hg⁰ emission (Zhuang et al., 2014). Photocatalytic oxidation is a promising technology for at high removal efficiency, and low cost of maintenance (Zhou et al., 2017), which utilizes semiconductors like TiO₂, WO₃, FeTiO₃, and metallic ions-doped TiO₂ photocatalysts to carry out a photo-induced redox process (Kityakarn et al., 2013). Metal elements doping narrowed TiO₂ band gap and enhance its visible light activity (Ma et al., 2015). Catalytic membrane reactor combine chemical reaction and separation in a single unit, in which a porous membrane, rendered catalytic by impregnation with catalysts, the reactants flow convectively through the membrane pores where the catalyst is located, thus resulting in intensive contact between the reactants and the catalytic sites, and in high catalytic activity with negligible mass transport resistance (Kajama et al., 2016). Membrane functioned as selective barrier for mixed contaminants, carrier for catalyst and provided spacious area for reaction (Coronas and Santamar, 1999). A novel membrane delivery catalytic oxidation system was set up for Hg⁰ oxidation and shown phenomenal performance (Guo et al., 2011). Hg⁰ was oxidized to mercuric chloride oxidation by advanced oxidation process (AOPs) (Zhan et al., 2013). Ozonation can efficiently oxidize many recalcitrant organic compounds (Hassani et al., 2017).

The objective of this work is to study elemental mercury (Hg⁰) removal by coupling membrane photocatalytic oxidation and wet

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absorption with a novel Fe–TiO₂/PVDF wet photocatalytic membrane reactor (WPCMR). This study evaluates the influence of Hg⁰ inlet concentration, gas residence time (GRT), water spray rate, UV illuminance, ozone concentration, the coupling role of membrane and catalysis on mercury oxidation. XRD, FTIR, UV–Vis, XPS and SEM method are utilized in this study to figure out the formation of the intermediate products and their mechanistic involvement in membrane catalytic oxidation of Hg⁰, and the mechanistic and kinetic analysis of membrane catalytic oxidation were elicited, which is believed to provide impetus for the application of the membrane catalytic oxidation.

2. Materials and methods

2.1. Preparation of Fe–TiO₂/PVDF

Fe–TiO₂ catalyst was prepared by sol–gel method. 25 mL Ti(OC₄H₉)₄ and 10 mL acetic acid was dissolved in 350 mL absolute ethyl alcohol in a beaker. The mixture was stirred for 45 min and designated as solution A. 7.26 g ferric nitrate was dissolved in 30 mL deionized water, 150 mL absolute ethyl alcohol and 20 mL acetic acid. The mixture was stirred for 20 min and designated as solution B. Solution B was dropped to solution A by peristaltic pump at the speed of 1 drop per second. The mixture solution A and solution B

was stirred for 24 h at room temperature. The Fe–TiO₂/PVDF was synthesized by soaking the entire PVDF hollow fiber membrane in the mixed solution for 48 h then dehydrated for 2 d in thermostat at 80 °C.

2.2. Experimental procedure

The experimental schematic of wet photocatalytic membrane bioreactor (WPCMR) was shown in Fig. 1. PVDF hollow fiber membrane was purchased from Blue cross Company, Tianjin, China. The dimensional parameters of the PVDF hollow fiber membrane were as follow: 0.38 mm fiber internal diameter; 0.5 mm fiber external diameter; 2 400 fibers; 0.1–0.01 μm pore size; 60% porosity; 200 mm column length; 40 mm column diameter; 2.2 m² total area. Hg⁰ was generated by heating a mercury permeation tube (supplied by Qingan scientific instruments Co., Ltd) in a 50–70 °C water bath. Airstreams of Hg⁰ and compressed air were dehydrated in a glass jar loaded with calcium chloride granules, mixed (21% O₂ content) in a buffer jar then pumped into Fe–TiO₂/PVDF hollow fiber membrane reactor. Water was sprayed onto the membrane surface using a subaqueous pump in WPCMR. Outlet gas was absorbed by saturate potassium chloride solution and potassium permanganate solution (10%, m/V).

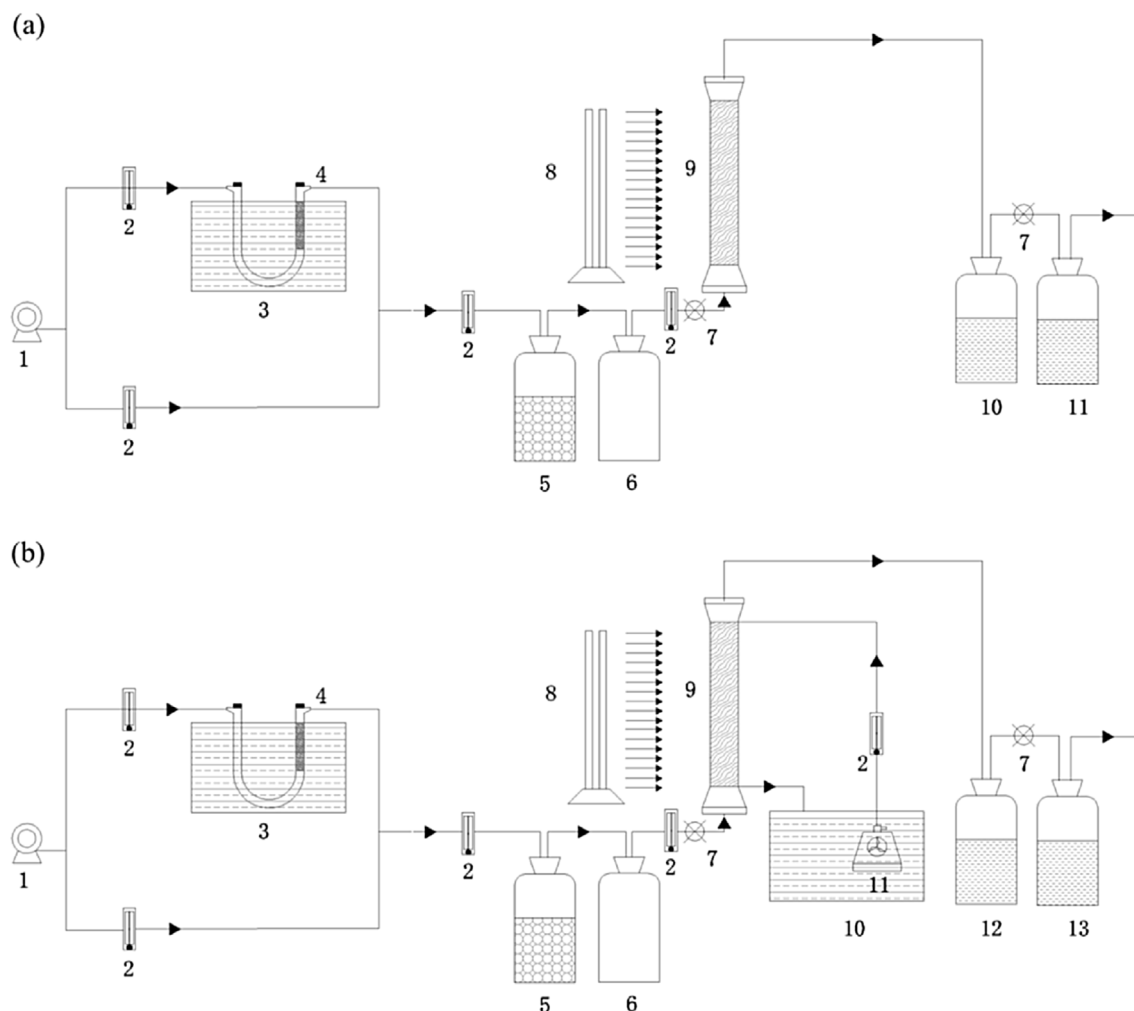


Fig. 1. The experimental schematic of wet photocatalytic membrane bioreactor (WPCMR): 1. Air compressor; 2. Flowmeter; 3. Water bath; 4. Elemental mercury permeation tube; 5. Gas drying jar; 6. Gas mixing jar; 7. Sampling point; 8. UV lamp; 9. Fe–TiO₂/PVDF; 10. Water tank; 11. Subaqueous pump; 12. Saturate KCl solution; 13. KMnO₄ solution.

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