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Research paper

Improving the combustion efficiency of diesel fuel and lowering PM2.5 using palygorskite-based nanocomposite and removing Cd²⁺ by the residue



Rongrong Li^{a,b}, Jingjing Tong^c, Guilong Zhang^{a,d}, Minguang Gao^c, Dongqing Cai^{a,d,*}, Zhengyan Wu^{a,d,*}

- a Key Laboratory of High Magnetic Field and Ion Beam Physical Biology, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031, China
- ^b University of Science and Technology of China, Hefei 230026, China
- ^c Key Laboratory of Environmental Optics and Technology, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031. China
- ^d Key Laboratory of Environmental Toxicology and Pollution Control Technology of Anhui Province, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031, China

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ABSTRACT

In this work, a nanocomposite (designated as HPAA) was fabricated using palygorskite clay (PC) modified by high energy electron beam (HEEB) irradiation, aminopropyltriethoxysilane (APTES), and amino siliconoil (ASO), and used as an additive for diesel fuel (DF) to enhance combustion efficiency (CE). HPAA possessed a micro/nano network structure and could adsorb DF molecules on the surface of HPAA nanorods, which promoted the contact of DF with oxygen and thus effectively enhanced the CE of DF. Additionally, this technology could enlarge the diameter of flying ash (FA), decrease FA amount, and reduce harmful gases emission during the combustion, which potentially favored the reduction of PM2.5. After combustion, the residue, carbon nanoparticles wrapping on the surface of HPAA nanorods, was obtained and displayed a high removal capacity on cadmium ions (Cd²⁺) from aqueous solution. This work provides a promising approach to improve the utilization efficiency of DF in boilers, lower PM2.5, and meanwhile decrease Cd²⁺ contamination, which might have a high application potential in heating and power generation.

1. Introduction

Along with the development of economy and society, more and more petroleum is demanded. Diesel fuel (DF), a typical petroleum product, is widely used with a global usage of 934 million tons annually (Kulkarni and Dalai, 2006). Unfortunately, petroleum is getting exhausted and might be depleted by the year of 2050 (Goyal et al., 2008). Because of the inadequate contact of liquid state DF with oxygen, it is difficult for DF to burn completely, leading to a low combustion efficiency (CE) and severe waste of source. Meanwhile, the incomplete combustion of DF brought about plenty of pollutants including NO_{x} , SO_{x} , CO, micro-nanoparticles, and volatile organic compounds, aggravating air pollution especially haze (Koberg and Gedanken, 2012). Therefore, it is urgent to improve CE of DF and reduce the related environmental contamination.

Up to date, several methods for improving CE of DF have been developed including mixing additives with DF, modifying engine system of machines and so on (Devan and Mahalakshmi, 2009). Therein, the development of additives for DF is attracting more and more attention

owing to their convenience in application. Currently, several additives have been reported including inorganic (Ulrich and Wichser, 2003; Zhu et al., 2012) and organic (Yang et al., 2013; Celik et al., 2015) nanomaterials. These additives could enhance the CE of DF to a certain extent, however they display drawbacks of high cost, secondary pollution, poor stability, etc., greatly limiting their large-scale applications. Hence, it is necessary to develop a novel, low-cost, efficient, and environmentally friendly nanomaterial used as the additive to enhance CE of DF and decrease pollution.

Palygorskite ((Mg,Al)₄(Si)₈(O,OH,H₂O)₂₆·nH₂O) clay (PC), a sort of natural crystalline hydrated magnesium aluminum silicate with unique three-dimensional structure and fibrous morphology, is made up of nano-scaled rods approximately 800-2000 nm in length and 30-40 nm in width (Chen and Wang, 2007; Li et al., 2011; Jiang et al., 2015). PC has a wide range of applications such as adsorbent (Zhou, 2016; Carazo et al., 2017; Yang et al., 2018), carrier (Frost et al., 2010; Wang et al., 2015b), construction materials and so on, due to its large specific surface area and high surface activity (Xiang et al., 2014). Nevertheless, PC nanorods tend to aggregate naturally because of the nano scale effect,

E-mail addresses: dqcai@ipp.ac.cn (D. Cai), zywu@ipp.ac.cn (Z. Wu).

^{*} Corresponding authors at: Key Laboratory of High Magnetic Field and Ion Beam Physical Biology, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031, China.

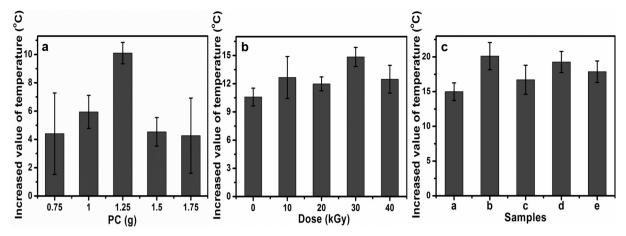


Fig. 1. (A) Influence of PC amount on the water IT treated with DF ($W_{DF} = 2.5 \, g$); (B) Effects of HP10, HP20, HP30, and HP40 on the water IT treated with DF ($W_{HP}:W_{DF} = 1.25 \, g$); (C) water IT of DF treated by HP30-APTES-ASO composites with $W_{HP30}:V_{APTES}:V_{ASO}$ of (a) 4 g:0 mL:0 mL, (b) 4 g:0.1 mL:1.0 mL, (c) 4 g:0.3 mL:1.0 mL, (d) 4 g:0.5 mL:1.0 mL, and (e) 4 g:0.7 mL:1.0 mL ($W_{(HP30-APTES-ASO)}:W_{DF} = 1.25 \, g$:2.5 g).

which causes a low dispersion of PC and greatly limits its applications (Cai et al., 2009; Zhang et al., 2013). Herein, high energy electron beam (HEEB) irradiation, a method of low-cost, high-efficiency, and ecofriendly, could transform the PC structure from nanorods to nano network and thus increase the dispersion and surface activity, which is favorable for its application as carrier in environment and energy fields.

In this work, the HEEB-treated PC (HP) was further modified with amino siliconoil (ASO) and aminopropyltriethoxysilane (APTES), and the resulting nanocomposite (the optimal one at dose of 30 kGy was designated as HPAA) was used as an additive to enhance utilization efficiency of DF. The result indicated that HPAA, with a micro/nano network structure, could adsorb DF molecules on the surface of nanorods, resulting in a larger contact area of DF with oxygen and thus a higher CE. Interestingly, it was found that this technology could effectively increase the diameter of flying ash (FA), which was beneficial for the deposition of FA and reduction of PM2.5. After combustion, the residue, carbon nanoparticles coating on the surface of HPAA nanorods, was obtained and could be used to efficiently remove cadmium ions (Cd²⁺) from aqueous solution. Cd²⁺ mainly originating from industrial activities including electroplating and mining is a typical highly toxic heavy metal ion. It can accumulate in vital organs and tissues of organisms, such as spleen, liver, testicles, immune system, central nervous system, blood and so on, causing a variety of diseases (Wu et al., 2009; Otomo and Reinecke, 2010; Paulino et al., 2011; Ge et al., 2012). In order to reveal the mechanism on CE enhancement, the interaction between HPAA and DF was investigated. This work provides a facile approach to improve the CE of DF in boilers and meanwhile reduce the discharge of contaminants, which could have a huge application prospect.

2. Materials and methods

2.1. Materials

PC (noncombustible powder, 200–300 mesh, 90% purity) was obtained from Mingmei Co., Ltd (Anhui, China). DF (0#, 0.83 g/mL) was bought from China Petrochemical Corporation (Anhui, China). Cadmium nitrate tetrahydrate ($Cd(NO_3)_2$ ·4H₂O) with purity of 99%, and other chemicals with analytical grade were provided by Sinopharm Chemical Reagent Co., Ltd (Shanghai, China).

2.2. HEEB irradiation of PC

PC (150 g) in a sealed plastic bag was irradiated using a HEEB accelerator (10 MeV and $10\,kW$) with doses of 10, 20, 30, and 40 kGy at

room temperature, and the resulting samples were named as HP10, HP20, HP30, and HP40, respectively.

2.3. Fabrication of HP30-APTES-ASO composites

Firstly, HP30 (4 g) was dispersed in 40 mL of ethanol to obtain a HP30 suspension. Subsequently, APTES with different amount was added to the suspension and the resulting system was stirred (500 rpm) for 5 min, then 1.0 mL of ASO (V_{ASO} : $V_{APTES} = 1.0$ mL:0.1 mL, 1.0 mL:0.3 mL, 1.0 mL:0.5 mL, and 1.0 mL:0.7 mL) was added under stirring (500 rpm) for 5 min at room temperature. Finally, the resulting system was dried at 60 °C overnight to remove ethanol and obtain HP30-APTES-ASO composite.

2.4. Combustion experiment

PC with different amount (0.75, 1.0, 1.25, 1.5, and 1.75 g), HP (1.25 g) with different doses, and HP30-APTES-ASO composites (W_{HP30} : V_{APTES} : $V_{ASO} = 4.0$ g:0 mL:0 mL, 4.0 g:0.1 mL:1.0 mL, 4.0 g:0.3 mL:1.0 mL, 4.0 g:0.5 mL:1.0 mL, and 4.0 g:0.7 mL:1.0 mL) were added to DF (2.5 g), respectively. Subsequently, these resulting systems were placed in a crucible lid (diameter of 9 cm) in a closed super clean bench and burnt respectively to heat 150 mL of water compared with DF alone, and the water temperature was measured after combustion. The increased temperature (IT) was calculated according to Eq. (1):

$$IT = T_{\text{sample}} - T_{\text{DF}} \tag{1}$$

where $T_{\rm DF}$ and $T_{\rm sample}$ were the resulting temperatures of water treated by DF and samples respectively. Herein, the optimal sample of HP30-APTES-ASO, which displayed the highest IT, was named as HPAA. All the experiments were performed in triplicate.

2.5. Effect of HPAA on gas emission

To investigate the influence of HPAA on the emission of gas generated during the combustion of DF, the component and concentration of discharged gas were measured. HPAA (1.25 g) was evenly added to DF (2.5 g), and the obtained HPAA-DF was put in a crucible lid. Then the resulting system was placed in a sealed cubic glass box (1 m \times 1 m). After combustion, the gas in the box was detected.

2.6. Effect of HPAA on FA amount

HPAA (2.5 g) was evenly added to DF (5.0 g) in a crucible lid. Then

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