



Functionalizing bottom ash from biomass power plant for removing methylene blue from aqueous solution



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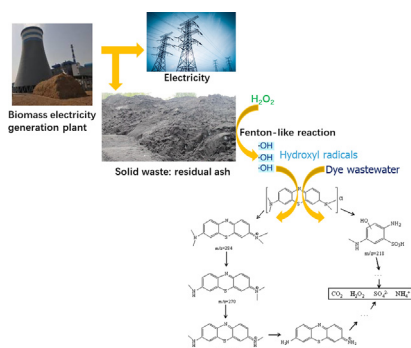
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HIGHLIGHTS

- H₂O₂ plus biomass ash was employed for removing methylene blue in aqueous solution.
- Fenton-like reaction was mainly responsible for the methylene blue degradation.
- Promoting temperature & time improved MB removal via H₂O₂ plus biomass ash.
- Maximal removal of 247.8 mg/g can be achieved at initial concentration of 2000 mg/L.
- Low pH (<2) was negative to MB removal, and higher pH (>5) intensified MB removal.

GRAPHICAL ABSTRACT



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ABSTRACT

In order to seek a possible path for utilization bottom ash, the solid waste from biomass incineration for power generation, its basic characteristics were investigated, and removing methylene blue (MB) from aqueous solution was attempted as well. Results indicated bottom ash dominantly contained Si and Ca-related minerals with >35 mineral elements, meanwhile, it was typically characterized by alkalinity (pH of 9.5) and low specific surface area (14.5 m²/g). As the only bottom ash was employed for removing MB, removal efficiency was lower than 44%, however, it was greatly improved to 100% as hydrogen peroxide was supplemented (final concentration of 4.0%). Based on the elucidated mechanisms, Fenton-like reaction was triggered by bottom ash, which was dominantly responsible for removing MB, rather than the adsorption by bottom ash. Besides, increasing reaction temperature and duration dramatically promoted MB removal by bottom ash with the aid of hydrogen peroxide (HBA). A great promotion on MB removal from 92.3 mg/g to 143.9 mg/g was achieved as pH was adjusted from 2.0 to 5.0, indicating MB removal was pH-dependent. The maximum removal of 260.9 mg/g was achieved by HBA at a relatively high initial MB concentration of 2000.0 mg/L, suggesting a great potential in treating dye wastewater by functionalizing bottom ash with the aid of hydrogen peroxide. Moreover, the released heavy metals and derived by-products from MB removal were acceptable in consideration of their potential environmental risks. Thus, the current work offers a new path to valorize the solid waste in biomass electricity generation plant.

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1. Introduction

With dual pressures from preventing environmental deterioration and promoting energy security, a low-carbon energy structure based on renewable energy is becoming increasingly important in the development of world's economy (Asgari et al., 2014). In contrast with the fossil energy, biomass is usually characterized to be “carbon-neutral” and can reduce pollutants (NO_x, SO₂) emission (Pang et al., 2017; Tan et al., 2017). Biomass power generation is a direct way to convert biomass via incineration, which plays an important role in utilizing waste biomass, especially, agriculture or forestry residues for electricity supply. Taking China as example, biomass power has experienced a booming growth since 2005, and electricity production from biomass power reached 35.6 billion kWh, which accounted for 0.68% of total electricity production in 2013 (Lin and He, 2017). Technically, biomass power generation, regardless of co-firing or direct combustion, now is mature enough and feasible in practice. However, 5% to 10% (dry basis), even higher ash content in biomass definitely resulted in large amounts of ash residues after biomass incineration (Shi et al., 2017). Lacking efficient paths for the utilization and commercialization makes the ash residues abandoned in piles, occupying a large amount of land, as well as resulting in the potential environment pollution to water and air in local area via rainfall and dustfall (Carrasco-Hurtado et al., 2014). Moreover, this situation will be specially worsened as the scale of biomass power generation was rapidly increased. Thus, seeking a valorization path for the ash residues, instead of the traditional disposal by landfilling, is extremely urgent to guarantee the sustainability of the biomass power industry.

Generally, biomass ash residues can be classified by fly ash and bottom ash, in which the bottom ash accounts for 60–90%, which are the solid residues from the complete or incomplete combustion of biomass, and generally produced on the grate in the primary combustion chamber of the boiler (Hinojosa et al., 2014). Overall, the bottom ash yield will be greatly related to many factors, such as biomass types, operating conditions, and the employed boiler system, and it is often mixed with the minerals of original biomass, unburned carbon, and impurities from the biomass, such as sand, stone, and dirt (James et al., 2013). In addition to the mentioned factors, the basic characteristics of biomass ash also depend on the transformations that occurs during the combustion process at different temperature (Vassilev et al., 2013a). Apparently, the bottom ash is a highly heterogeneous and complex product, and its chemical composition can be clarified roughly as inorganic matters and organic matters. As for the inorganic part, it is primarily consisted of silicates, oxides (or hydroxides), sulfur-containing salts (sulphates, sulphides, sulphosalts, sulphites and thiosulphates), phosphates, carbonates (or bicarbonates), chlorides, and nitrates, which mainly combined with various metal elements, such as K, Na, Ca, Mg, Fe, Al, Mn, and so on (Vassilev et al., 2013b). The organic part generally is incompletely combusted biomass and unburned carbon. Based on the basic chemical characteristic, bottom ash currently is mainly applied in soil fertilization due to its abundant elements, acid soil amendment due to its alkalinity, as well as cements and concretes, ceramic products and construction materials due to its chemical composition (Dahl et al., 2009; Maeda et al., 2017; Matalkah et al., 2016; Peys et al., 2016; Shi et al., 2017; Vassilev et al., 2013b). Recently, biomass ash as an adsorbent has been developed to be a new path for removing pollutants, such as, heavy metals, organic matters, and NO_x, SO_x, NH₃, and these work have been mostly investigated on fly ash due to its abundant porosity and high organic carbon content, and large specific surface area (Noonpui et al., 2010; Quirantes et al., 2017; Vassilev et al., 2013b). Unlike fly ash, bottom ash was characterized by relatively higher particle sizes, lower reactivity value and neutralizing value, which makes it hard to be utilized as an adsorbent directly (Dahl et al., 2009; James et al., 2012). However, recent studies have indicated that the ash-rich adsorbents, such biochar, coal ash and fly ash (from coal combustion), can greatly intensify its capacity on removing organic pollutants

through supplementing hydrogen peroxide, in which the triggered oxidation process by heterogeneous catalysis was mainly responsible for this intensified capacity (Fang et al., 2014; Fang et al., 2015; Gu et al., 2013; Liu et al., 2009; Song and Li, 2009). Theoretically, it is possible to functionalize bottom ash as a potential catalyst or inducer to construct such oxidation environment.

According to these scenarios, the aim of present work is to evaluate the potential utilization of the derived bottom ash from biomass power plant in dye wastewater treatment. Current work mainly focused on 4 aspects as displayed in the Supplementary Fig. 1, in which the basic characteristics of the employed bottom ash were clarified, and the feasibility of removing a typical model dye, methylene blue (MB), by HBA (bottom ash with the aid of hydrogen peroxide) was checked. Based on the efficient MB removal, the hidden mechanisms for the removal were discussed as well. Besides, the performances of methylene blue removal as affected by the main parameters, including reaction temperature, duration, and initial pH and MB initial concentration, were investigated to clarify the potentials in application.

2. Materials and method

2.1. Bottom ash

The employed bottom ash was taken from a biomass power plant in Qingdao, Shandong Province, China, and the plant mounted two of 15 MW circulating fluidized bed boilers (CFB). At present, various agricultural residues, including corn stover, wheat straw and peanut shell, and forestry residues are the main feedstocks for power generation. The bottom ash was taken from the boiler outlet after it was cooled down. In order to reduce the size of sintered parts and make bottom ash more homogeneous for the subsequent investigations, it was hammered and screened through a 40-mesh sieve, and the screened ash was stored in a sealed plastic bag for further investigations.

2.2. Chemicals

Methylene blue (Guaranteed reagent, purity ≥ 98%, Chron Chemicals Co., Ltd., China) was employed as the typical model pigment. Hydrogen peroxide (Analytical reagent, purity ≥ 35%, w/w, Chron Chemicals Co., Ltd., China) was employed for constructing the oxidation process. In order to check the released radicals, tert-butanol (Analytical reagent, purity ≥ 99.5%, w/w, Chron Chemicals Co., Ltd., China) was employed as scavenger. Besides, some frequently-used chemicals in this work, such as HCl, NaOH, were all purchased from Chron Chemicals Co., Ltd. in China.

2.3. Characterization of bottom ash

Ultimate analysis of the bottom ash was carried out by an Elemental Analyser (Vario MICRO cube, Quantachrome, USA). Moisture, ash and volatile matter content of the bottom ash was analyzed by a muffle furnace (OTL-1200, Boyuntong, China) according to the corresponding methods in ASTM D1762-84. Specific surface area (SA) was determined using N₂ sorption isotherms at 77 K (NOVA-2000E, Quantachrome, USA) following the Brunauer-Emmett-Teller (BET) method. pH of bottom ash was directly measured by a pH meter (PHS-3C, Leici, China) with the ratio of ash and deionized water of 1:20 (w/w). A Fourier transform infrared spectrometer (FT-IR) (Nicolet 6700, Thermo Fisher, USA) was employed to collect the spectral characteristic of bottom ash in the range of 400–4000 cm⁻¹ with the resolution of 4 cm⁻¹ and 64 scans per sample. X-ray diffraction (D8 Advance, Bruker, Germany) was carried out on bottom ash with Cu K_α radiation, and the generator voltage and tube current were 40 kV and 40 mA, respectively. The basic elemental composition of the bottom ash was analyzed by X-ray fluorescence spectrometry (XRF) (HD Prime, XOS, USA), and X-ray photoelectrons spectrometer (XPS)

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