



Full Length Article

TMA and H₂S gas removals using metal loaded on rice husk activated carbon for indoor air purification



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ABSTRACT

An excessive exposure to indoor toxic pollutant gases such as trimethylamine (TMA) and H₂S is fatal to humans especially in the confined workspace. A porous carbon pollutant filter was developed using rice husk as a biomass waste. The different carbonization temperature highly affected on the surface area of ARH (activated rice husk carbon) and their performance of pollutant gas adsorption. With copper impregnation on the activated carbon (Cu/ARH), much better toxic gas adsorption was achieved in an enclosed chamber. The best performance was obtained with a Cu/ARH filter carbonized at 450 °C as it eliminated the 400 ppm of TMA in 30 min and H₂S in 15 min. Isotherm and kinetic models were applied to understand the adsorption performance. The pseudo-second order kinetic model represented the best fit, followed by the intraparticle diffusion and Yoon-Nelson kinetic models. The H₂S adsorbed Cu/ARH filter was successfully regenerated whereas a TMA adsorbed Cu/ARH filter was not successfully regenerated. The current study is expected to expand the utilization of biomass waste for the indoor air purification.

1. Introduction

Rice is a representative biomass in the agricultural products of China and India, which annually produces tons of rice wastes even though there are many applications for the wastes including composting, animal feed, or bedding materials for cattle. Two major rice wastes, rice straw and rice husk, are normally left on the ground and burned in many underdeveloped and developing countries. The USDA announced that the estimated world rice production would be about 481 million metric tons in 2017 [1]. Considering a rice husk waste of 22% of all the rice [2], 106 million tons of rice husk is annually produced worldwide. One major management approach is rice husk combustion for heat and power, which incurs problems of corrosion, slagging, and fouling in the plant chamber due to its high alkali metals.

A thermochemical conversion method for biomass has been studied for last few decades, which handles solid wastes including agricultural, forest, municipal solid waste, and animal manure. Biomass pyrolysis as one of thermochemical conversion methods produces biochar, bio-oil and syngas at temperatures from 300 to 600 °C in the absence of oxygen. Major applications of bio-oil are biofuel, chemicals, and biopolymers whereas the applications of biochar include biofuel, soil amendments, and activated carbon. The production of activated carbon

even expands the applications of biochar to electrochemical material [3], catalyst support [4], hydrogen storage [5], and adsorption material [6]. The activated carbon, defined as a porous carbon with a surface area between 5 and 3000 m²/g, can be produced through chemical or physical methods. Due to its higher porosity and controllable properties, chemical activation has been widely used with activation agents including Na₂SO₄, H₃PO₄, ZnCl₂, KOH, and H₂SO₄ [7]. Among the activation agents, KOH has been frequently used for the production of activated carbon since it helps to produce a large fraction of micropores size distribution and a high surface area up to 3000 m²/g [3].

Trimethylamine (TMA) of N(CH₃)₃ is a colorless and flammable tertiary amine with a saline taste and a strong rotting fish ammonia malodor. This organic substance is soluble in water (8.9 × 10⁵ mg/L at 30 °C) and is easily vaporized from the surface of water [8], which is released from various wastes of decomposed animal, fish, and sewage. Even though the exposure of TMA concentrations from 0.1 to 10 ppm over an 8 h period are not considered toxic according to the U.S. EPA, TMA exposure to concentrations of 240 ppm for 10 min or 51 ppm for 8 h are regarded as thresholds for lung toxicity and neurotoxicity in rats [9]. Hydrogen sulfide (H₂S) is heavier than air and has a rotten egg smell, which is adsorbed on the lungs after inhalation. A continuous exposure to the gas by humans, even at a low concentration, leads to

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olfactory fatigue, which means the temporary inability to distinguish the odor. Extra caution or appropriate procedures to reduce the concentration of H₂S gas is required for working in poorly-ventilated confined areas such as basements, manholes, underground workspaces, and manure pits. The largest production of H₂S is from petroleum oil refineries during the hydrodesulfurization process, led by the reaction between sulfur and hydrogen. Other included syngas from the gasification process, Kraft paper mill processes, sewerage, and anywhere where sulfur reacts with organic material at a high temperature. According to NIOSH (The National Institute for Occupational Safety and Health, USA), the maximum H₂S concentration that can be endured for 1 h without serious consequences is between 170 and 300 ppm. A concentration between 400 and 700 ppm is dangerous after an exposure of 0.5 to 1 h, and can lead to unconsciousness and possible death. Thus, TMA and H₂S are possibly lethal pollutants that require a proper removal method in contaminated areas, especially in a poorly-ventilated enclosed space.

Many adsorption studies with activated carbons were focused on the removal of water contaminants (dye, heavy metal, and pharmaceutical compound) [10–14], while a relatively fewer number of studies were conducted for gas phase contaminants such as VOCs and H₂S using activated carbon [15–17]. Lee et al. [16] modified coconut-based activated carbon with an acid and a base solution for the removal of VOCs. Although the acidic nature of the carbon helped with TMA and mercaptan removals, no significant changes of the adsorption capacity was obtained. The equilibrium adsorption capacity of the TMA was obtained at around 100 mg_{TMA}/g_{carbon}. Li et al. [17] utilized rice husk as oxidized activated carbon to enhance the number of oxide functional groups. A comparable removal of VOCs was achieved with the largest surface area AC sample (350 m²/g). Also, they determined that a higher RH (relative humidity) of 90% reduced the efficiency of adsorption when it was compared to 55% RH due to the hydrogen bonds of water between the water molecules and oxygen groups of the AC. Raso et al. [15] demonstrated a two-step process for removing six different VOCs from indoor air using a CeO₂/TiO₂ oxidative catalyst combined with activated carbon. The in-situ thermal regeneration of the catalyst and subsequent oxidation could eliminate the adsorbed VOCs, which could be expected to reduce the costs of operating energy and maintenance. When it comes to H₂S removal, many studies used metal oxides such as Mn, Cu, Co, Fe and Zn at an elevated temperature (250–600 °C) mainly for H₂S gas removal from gasification syngas [18–20]. Sitthikhanhaew et al. [21] conducted H₂S removal experiments with KOH impregnated activated carbon at a higher temperature (30–550 °C). At the 550 °C adsorption temperature, the highest H₂S removal was achieved at 321 mg_{H₂S}/g_{carbon} with KOH impregnated on steam activated carbon.

To the best of our knowledge, no study has been reported on the TMA and H₂S gas removals in a confined space (at room temperature) using a metal loaded on agricultural waste carbon filter. The specific objectives of this study were a) to determine the effect of carbonized

temperatures on the properties of bio-chars (CRH) and activated carbon (ARH), b) to evaluate the effectiveness of adsorption of TMA and H₂S pollutant gases using CRHs, ARHs, and Cu-ARHs, c) to devise adsorption isotherms and kinetic models to understand and optimize the adsorption process, and d) to find the regeneration condition for the used filter catalyst. The current study is expected to help in the utilization of agricultural wastes for toxic gas removals, especially for indoor use.

2. Materials and methods

2.1. Sample preparation

The rice husk (RH) used in the experiment was harvested and stored in an open barn. The sample was dried for 10 h in a 110 °C oven after a half hour-long ultrasonic bath wash to remove possible impurities. The prepared RH was carbonized in a stainless steel reactor (53 mm inner diameter × 900 mm length) surrounded by an electric furnace (max. temperature 1500 °C). There were 14.3 g of RH used for each carbonization at conditions of 350, 450, and 550 °C for 30 min with 2 ml/min of inert argon gas. The temperature increased at a rate of 50 °C/min. Once each carbonization experiment was performed, the collected carbonized rice husk (CRH) was immersed in 2 M HCl acid for an hour at 100 °C to remove possible ash and impurities. Then the CRH was neutralized with distilled water before sample drying at 120 °C for 8 h. The CRHs carbonized at 350 °C, 450 °C, and 550 °C were identified as CRH-350, CRH-450, and CRH-550, respectively.

A KOH chemical activation was performed with dried CRH at three different temperatures to produce activated rice husk carbons (ARH). First, about 4 g of CRH was mixed with 100 ml of 7 M KOH solutions for 2 h using a stirrer to evenly mix the precursor. The KOH impregnated carbon was filtered from the solution, and dried overnight in 110 °C oven. Then, the same tube reactor with an electric furnace was used to activate the sample at 750 °C for 2 h with 2 ml/min argon gas. The residence time for the activation was imported from elsewhere [22]. After quenching the reactor, the sample was washed and neutralized with distilled water. Then it was dried for 12 h at 110 °C. The activated CRHs at 350 °C, 450 °C, and 550 °C were labelled as ARH-350, ARH-450, and ARH-550, respectively. The mass recovery after carbonization and activation is further discussed in the Section 3.1.

The 16% Cu on the carbon filter (Cu/CRH or Cu/ARH) was fabricated by mixing CuCl₂·H₂O, CRH/ARH, a polyester (PET) 2 × 2 mesh organic binder (DongJin Textile, Korea), and deionized water. After mixed for 10 min using a stirrer, the carbon slurry was evenly distributed on the PET filter, squeezed through a rolling presser, and naturally air dried for 24 h as shown in Fig. 1 (a) and (b). The air dried filter was cured in a 90 °C oven for 20 min. The optimized curing conditions of temperature and the curing time were determined from a previous work [23].

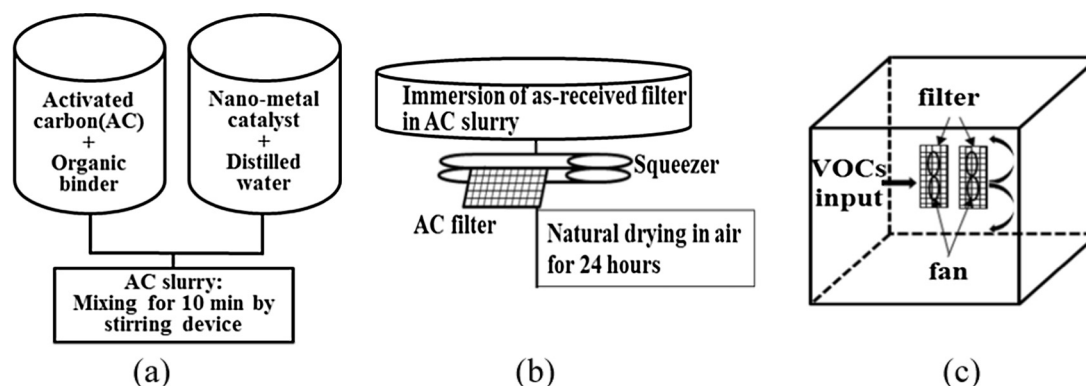


Fig. 1. Schematics of (a) carbonization and activation process, (b) PET ARH filter process, and (c) VOCs removal experimental setup.

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