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Technical Note

Regeneration of activated carbon saturated with odors by non-thermal plasma

Jie Chen [a,*], Xinchao Pan [a], Jianwei Chen [b]

[a] Institute of Environment Science & Engineering, Hangzhou Dianzi University, Hangzhou 310018, PR China
[b] Max Planck Institute for Marine Microbiology, Celsiusstraße 1, D-28359 Bremen, Germany

HIGHLIGHTS

- It is a novel, efficient alternative process for odor-saturated AC regeneration.
- Sustainable high regeneration efficiency was obtained in successive regeneration cycles.
- DBD improved structural properties and surface chemistry of AC surface.

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ABSTRACT

The dielectric barrier discharge (DBD) regeneration process of an activated carbon (AC) saturated with dimethyl sulfide was studied on a laboratory scale. The results showed sustainable high regeneration efficiency (RE) (>90%) in successive regeneration cycles (10 cycles). Energy density, humidity and oxygen content were key factors for DBD system, with optimum conditions of 761 J L^{-1}, 0–1 vol% and 5%, respectively. The high efficiency was likely attributed to the improvement of structure and surface properties of AC by DBD. After the first regeneration, surface area, micropore volume and total pore volume of AC increased by 8%, 23% and 15% respectively, while average pore size decreased by 9.5%. The number of carboxylic groups doubled (from 0.22 to 0.48 mmol g^{-1}) while that of phenolic groups remarkably decreased (from 0.48 to 0.26 mmol g^{-1}) after successive regeneration cycles, which helped to maintain high RE. The results suggested DBD as a novel, efficient alternative process for odor-saturated AC regeneration.

1. Introduction

Odor pollution is one of the most serious public hazards, and should be removed before discarded into atmosphere. Activated carbon (AC) has been widely used as an efficient adsorbent in odor control due to its high surface area, tailored pore distribution and high degree of surface reactivity (Hsieh and Teng, 2000; Yenisoy et al., 2004). However, depending on the adsorption capacity, AC will become saturated after some time, and thus needs to be regenerated before reuse.

Traditional AC regeneration techniques include thermal regeneration (Suzuki et al., 1978; San et al., 2001), solvent regeneration (Chinn and King, 1999), direct oxidation and catalytic wet oxidation regeneration (Moshe et al., 1999; González et al., 2002) and biological regeneration (Scholz and Martin, 1998), which are normally limited either technically or economically.

The dielectric barrier discharge (DBD) technique has been efficiently used to modify/clean surfaces of materials, and may offer an interesting alternative for successive AC regeneration. It mainly comprises chemical and physical processes. The former includes formation of high energy electrons, short-lived reactive radicals (such as hydroxyl radicals, hydrogen radicals, oxygen free radicals) and long-lived molecular species (such as ozone). Physical processes simultaneously occur in air, i.e. UV light emission (Hao et al., 2009). These processes lead to decomposition and detoxification of organic pollutants in air/water with relatively low power consumption and high removal efficiency. Thanks to these advantages, DBD technique has been extensively applied in environmental protection (Ruan et al., 2005; Shi et al., 2005; Chen et al., 2009, 2010; Wan et al., 2011; Huang et al., 2012) and material modification (Kodama et al., 2002; Lee et al., 2005; Hao et al., 2009), and has gained great success. However, DBD has so far rarely been used for AC (saturated with odors) regeneration. The present work thus focused on the efficiency as well as the mechanism of AC regeneration by DBD. A wire-cylinder DBD reactor energized by a high voltage source with Blumlein pulse forming network (BPFN) was adopted. AC saturated with dimethyl sulfide (DMS) was used for regeneration. Different conditions such as humidity and oxygen contents were taken into account.

* Corresponding author. Tel.: +86 571 86919158; fax: +86 571 86913539.
E-mail address: chenjie_0002@126.com (J. Chen).

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2. Experimental

2.1. Materials

The AC from coconut shells with a size of 20–40 mesh (0.45–0.90 mm) was provided by Xushui Company, China. The AC sample was then pretreated at 523 K in N_2 atmosphere overnight to remove the moisture and other contaminants. Artificial air (Jingong Company, China) containing 80% N_2 and 20% O_2 was provided by mixing 99.999% liquid N_2 and 99.8% liquid O_2. DMS was provided by ACROS ORGANICS, Belgium, with a purity of +99%.

2.2. Reactor and power supply

The plasma reactor was a 30 (id) × 90 mm epoxy resin cylinder. It had a Ni–Cr alloy wire (the energized electrode) of 0.5 mm in diameter fixed along the axis. A ceramic tube made of alumina was used to form the dielectric barrier wall. The ground electrode was a film conductor made of aluminum and embedded between the epoxy resin tube and ceramic tube. A BPFN type of narrow pulse generator was used in the study. The maximum output power and the maximum peak voltage were 1 kW and 100 kV. The pulse frequency adopted in this study was 100 pps (pulse s^{-1}). The pulse voltage and current waveforms were measured using a four channel Tektronix TDS 2014B 350 MHz digital storage oscilloscope capable of sampling 1 GS s^{-1} (Giga Sample s^{-1}), a Texas HVP-3020 high voltage passive probe and a CT4 TCP202 current probe.

2.3. Experimental procedure

Fig. 1a shows the schematic diagram of DMS adsorption process. Air was introduced into the DMS generator to carry out DMS vapor. The mixed stream was further diluted by another air flow in a buffer tank to achieve a final DMS concentration of 500 mg m^{-3}. Afterwards it was continuously introduced to AC at a flow rate of 1000 mL min^{-1}. After 7 h, AC was saturated with DMS.

Fig. 1b shows the schematic diagram of AC regeneration after the discharging zone. The DMS-saturated AC was then used for regeneration. Dry air that acts as balance gas was first introduced into the discharging zone at a flow rate of 1000 mL min^{-1}, and then immediately led to the AC regeneration reactor. The outlet gas from the regeneration reactor was further analyzed by a gas chromatograph.

Fig. 1c shows a schematic diagram of AC regeneration in the discharging zone. In each experiment, 3 g AC samples were evenly spread on the ceramic wall with a thickness of 2 mm. The air with varied O_2 content was introduced into the reactor at a flow rate of 1000 mL min^{-1}. For humidity experiments, air was first bubbled in deionized water and then diluted with another air flow to achieve an appropriate humidity. All the streams were mixed in a buffer tank, where humidity was measured by a hygrometer (Rotronic A1H). At last, the mixed stream (total flow rate of 1000 mL min^{-1}) was introduced to the reactor. All the experiments above were performed at ambient temperature.

2.4. Chemical analyses

The concentration of DMS was measured by the gas chromatograph GC7890II (Tianmei corporation) which was equipped with 6-port gas sample valve. The concentration of sulfur dioxide was analyzed by a gas analyzer (KM9106, Kane International Limited) equipped with KMCLP20 standard probe. The concentration of ozone was measured by the gas indicator tubes (Sanhuan corporation), the minimum detectable limit of which was 0.2 ppm.

The BET surface area and micropores volume of AC samples were obtained by N_2 adsorption or desorption at 77 K, using a BEL-

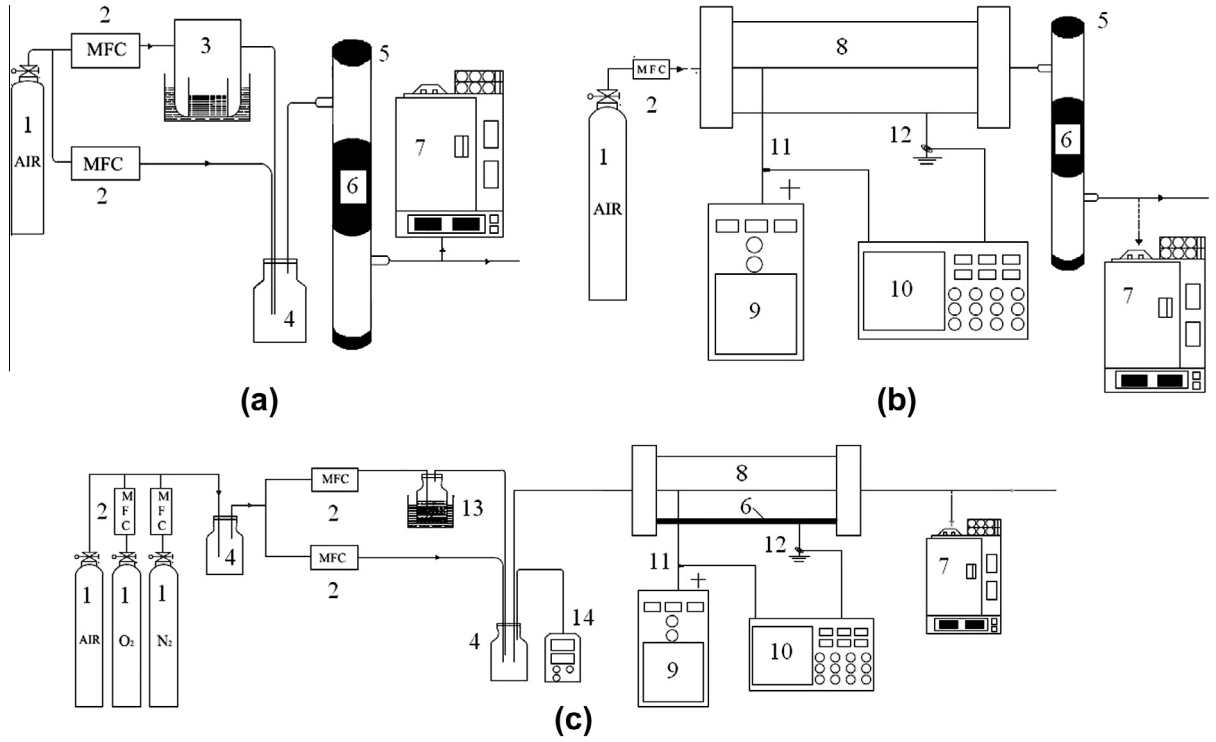

Fig. 1. Schematic diagram of the experimental setup: (a) DMS adsorption process; and (b) AC regeneration after the discharge zone (c) AC regeneration in the discharge zone. (1. Gas cylinder 2. mass flow control 3. DMS generator 4. buffer tank 5. adsorbent tube 6. AC 7. gas chromatograph 8. wire-cylinder DBD reactor 9. high pulse voltage source 10. digital storage oscilloscope 11. current probe 12. high voltage probe 13. water vapor generator 14. hygrometer.)