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Removal of airborne microorganisms emitted from a wastewater treatment oxidation ditch by adsorption on activated carbon

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Abstract

Bioaerosol emissions from wastewater and wastewater treatment processes are a significant subgroup of atmospheric aerosols. Most previous work has focused on the evaluation of their biological risks. In this study, however, the adsorption method was applied to reduce airborne microorganisms generated from a pilot scale wastewater treatment facility with oxidation ditch. Results showed adsorption on granule activated carbon (GAC) was an efficient method for the purification of airborne microorganisms. The GAC itself had a maximum adsorption capacity of 2217 CFU/g for airborne bacteria and 225 CFU/g for fungi with a flow rate of 1.50 m^3/hr. Over 85% of airborne bacteria and fungi emitted from the oxidation ditch were adsorbed within 80 hr of continuous operation mode. Most of them had a particle size of 0.65–4.7 μm. Those airborne microorganisms with small particle size were apt to be adsorbed. The SEM/EDAX, BET and Boehm's titration methods were applied to analyse the physicochemical characteristics of the GAC. Relationships between GAC surface characteristics and its adsorption performance demonstrated that porous structure, large surface area, and hydrophobicity rendered GAC an effective absorber of airborne microorganisms. Two regenerate methods, ultraviolet irradiation and high pressure vapor, were compared for the regeneration of used activated carbon. High pressure vapor was an effective technique as it totally destroyed the microorganisms adhered to the activated carbon. Microscopic observation was also carried out to investigate original and used adsorbents.

Key words: activated carbon; adsorption; airborne microorganisms; oxidation ditch; wastewater treatment facility

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Introduction

The removal of odors generated in the processes of waste and wastewater treatment has gained attention in recent years. Airborne microorganisms, including fungi, bacteria, and yeasts are another kind of emission present in wastewater treatment plants (WWTP). They are correlated closely with air pollution and cause adverse health effects on workers (Fracchia et al., 2006). Many studies have been carried out to evaluate the biological risks of aerosols by determining the concentrations of viable microorganisms, using different sampling and detection methods (Fanni et al., 1985; Carducci et al., 2000; Brandi et al., 2000; Bauer et al., 2002; Pascual et al., 2003; Karra and Katsivela, 2007). In our previous investigation, bioaerosol samples were collected at a Beijing municipal wastewater treatment plant with orbal oxidation ditch. One of the main sources of bioaerosols at WWTP is the mechanical agitation of raw wastewater. High concentration of cultivable bacteria and cultivable fungi were detected from the sampling point located near the rotating brush.

As airborne microorganisms can be transported by wind over long distances, they might be hazardous not only for plant workers but also for the surrounding population. Many countries are currently regulating the impacts of bioaerosols and have proposed measures for their control. Air filtration and UV germicidal irradiation (UVGI) are two types of air purification systems routinely used for removing or inactivating microorganisms (Riley and Nardell, 1989; Green and Scarpino, 2002; Kujundzic et al., 2007). Air filtration systems must be changed periodically and require routine maintenance. Compared with air filtration, UVGI generally provides effective control for a lower cost, as well as easier installation and maintenance. Both systems, however, initially require adequate ventilation. Adsorption on activated carbon has undoubtedly been the most popular and widely used technology in the treatment of exhaust gases throughout the world (Bansode et al., 2003; Christian and Verga, 2004; Lillo-Ro'denas et al., 2005). Due to its suitable surface for microbial attachment, GAC itself has been used as a biofilter medium in the biological treatment of odors and volatile organic

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compounds (VOCs); however, little is known regarding the clean-up of airborne bioaerosols from the air by adsorption on activated carbon.

The objective of the present study was to explore a suitable method to reduce airborne microorganisms according to their emission and distribution characteristics in wastewater treatment processes. Airborne microorganisms generated from a pilot scale oxidation ditch for domestic wastewater treatment were diminished by adsorption on granule activated carbon prepared from coconut shell. Two regenerate methods, UV irradiation and high pressure vapor, were compared during adsorbent regeneration. The relationship between physicochemical characteristics of GAC and its adsorption performance were also investigated.

1 Materials and methods

1.1 Laboratory scale wastewater treatment facility description

A pilot scale wastewater treatment facility with an integrated oxidation ditch was operated. Each day, 1.0 m^3 of domestic wastewater with average concentrations of 267.5 mg/L for COD and 130 mg/L for BOD was treated, with an hydraulic retention time (HRT) of 10 hr. The oxidation ditch was covered by Plexiglas for airborne microorganism adsorption test. The speed of the rotating brush was 50 r/min.

The adsorption of airborne microorganisms was carried out in a glass adsorption tube with 5.0 cm in diameter and a working volume of 0.35 L (Fig. 1). Sterilized coconut-shell based activated carbon (Tianjin Activated Carbon Corporation, Tianjin, China) with particle size of 10–30 mesh was used as the adsorbent. Sampling ports were equipped at the bottom and the top of the adsorption tube for the determination of airborne microorganisms in untreated and treated gases. The flow rate was 1.50 m^3/hr.

1.2 Activated carbon regeneration

Sterilization via ultraviolet (UV) radiation and high pressure vapor were conducted to regenerate the activated carbon. Used activated carbon was spread evenly under a UV lamp (FL20S.BL/E 20W, Toshiba, Japan) on a sterilized bench or put into a pressure vapor sterilizer (VARIOKLAV, 300-EP, German) at appropriate temperature.

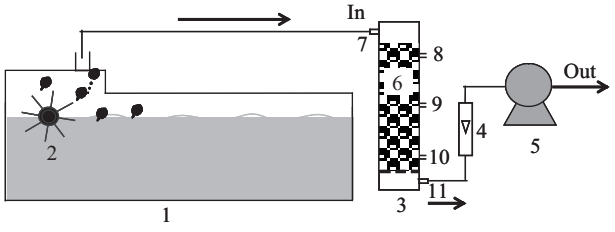

Fig. 1 Schematic diagram of treatment system. (1) oxidation ditch; (2) rotating brush; (3) adsorption tube; (4) flow meter; (5) vacuum; (6) granula activated carbon (GAC); (7–11): sampling ports 1–5.

1.3 Analysis methods

1.3.1 Characterization of GAC

The particle morphology of GAC was observed through a scanning electron microscope (Hitachi S-3000N /EDAX Inc., Japan) and the elements on the surface were determined by the same SEM together with energy dispersive X-ray spectroscopy.

The physical characteristics of the adsorbents, including specific surface area, pore volume distribution, and pore diameter, were determined by N_2 adsorption using Accelerated Surface Area and Porosimetry (ASAP 2000, Micromeritics, USA). Adsorbents were degassed at 350°C for 4 hr prior to the adsorption experiments. The BET surface area was obtained by applying the BET equation to the adsorption data. The pore-size was obtained using the BJH method.

Surface functional groups of GAC such as phenolic (–OH), carbonyl (C=O) and carboxylic (–COOH) groups were determined by Boehm titration. Approximately 25 ml of alkali solution (0.1 mol/L $NaHCO_3$, 0.05 mol/L Na_2CO_3, or 0.1 mol/L NaOH) was added to bottles containing 5 g of GAC and mixed constantly over a vibrator (100 r/min) at 25°C for 24 hr. Five milliliter supernatant was then drawn from the bottles and back titrated with HCl (0.1 mol/L) solution. The concentrations of various functional groups were determined by the residual bases after back titration. Number of acidic groups was calculated based on the assumption that NaOH neutralizes carboxylic, lactonic and phenolic groups, Na_2CO_3 neutralizes carboxylic and lactonic groups, and $NaHCO_3$ neutralizes the carboxylic group (Bohem, 1994).

1.3.2 Microbiological analysis

A six-stage Impacting Airborne Microorganisms Sampler (FA-1, China) was used to capture airborne microorganisms. Each stage included a plate with 400 holes of uniform diameter through which air was drawn at 28.3 L/min to impact Petri dishes containing agar media. Airborne particles were separated into six fractions, and the aerodynamic cut-size diameters in the six stages were: > 7.0 μm (stage 1), 4.7–7.0 μm (stage 2), 3.3–4.7 μm (stage 3), 2.1–3.3 μm (stage 4), 1.1–2.1 μm (stage 5), and 0.65–1.1 μm (stage 6) (Fang et al., 2005). Each sample was collected for 2 min. The sampler was sterilized by washing with a solution of 70% ethanol before the next sampling. Once the required volume of air had been drawn through, the plates were removed from the sampler, covered, inverted and incubated. The bacteria were incubated in Nutrient agar at 37°C for 48 hr. Fungi were cultivated in Rose Bengal Medium (BR, Aoboxing Biotech, Co., China) at 25°C (Karra and Katsivela, 2007) for 72 hr. The positive-hole correction method was used to determine colony count concentrations (Andersen, 1958; Macher, 1989). Results were calculated as the geometric mean of replicates and were expressed as colony forming units per cubic meter of air (CFU/m^3).

During the regeneration test, samples were collected at regular intervals to determine the residual of airborne

microorganisms. Approximately 0.05 g of used activated carbon was vibrated mechanically in 10 mL physiological saline solution for 5 min prior to dilution plating. Cultivation of bacteria and fungi were performed with the plate count method using cultivation media and incubation as described for airborne bacteria and fungi. Scanning electronic microscopy (FEI QUANTA 200, Japan) was used to observe microorganism characteristics on the used GAC.

2 Results and discussion

2.1 Adsorption of airborne microorganisms on activated carbon

Microbial samples were collected at sampling port 1 (Fig. 1) to investigate the concentration of airborne microorganisms in the inlet stream. As shown in Fig. 2, (2171 ± 242) CFU/m^3 of airborne bacteria and (207 ± 32) CFU/m^3 of airborne fungi were generated from the oxidation ditch. The concentration of bacteria was far greater than that of fungi. The ratio of fungi in total microorganisms was only 9.5%. In comparison to our previous WWTP investigations, airborne bacteria and fungi near the rotating brush of the oxidation ditch were (4726 ± 915) CFU/m^3 and (583 ± 37) CFU/m^3, respectively. Their ratio was almost the same as that obtained from the laboratory scale facility. However, the airborne microbes detected from the WWTP showed higher concentrations (over two times), which might be due to the large scale and high load. In addition, the emission of airborne microbes can also be influenced by environmental conditions, such as temperature, humidity, wind direction, season, and distance from the source of aerosol.

The GAC was employed to adsorb airborne microorganisms generated from an oxidation ditch. Figure 3 demonstrates the removal efficiencies of airborne bacteria and fungi at different contact time. As seen, 93.75% of airborne bacteria and 100% of fungi were adsorbed within 8 hr. The uptake of airborne microorganisms increased rapidly in the beginning, which was likely caused by strong attractive forces between the microorganisms and the adsorbents. The amount of airborne bacteria and fungi in the outlet stream were much lower than emissions from

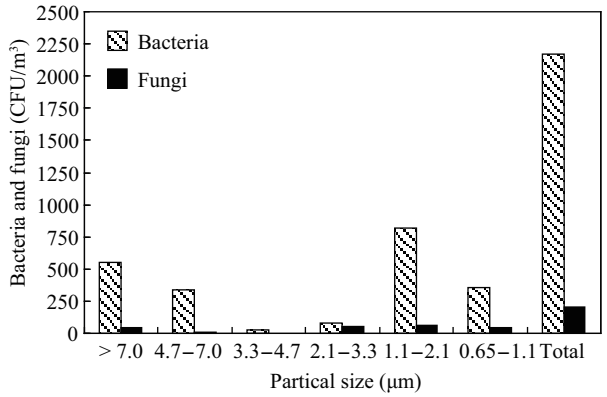

Fig. 2 Concentration and particle size distribution of airborne bacteria and fungi in inlet stream.

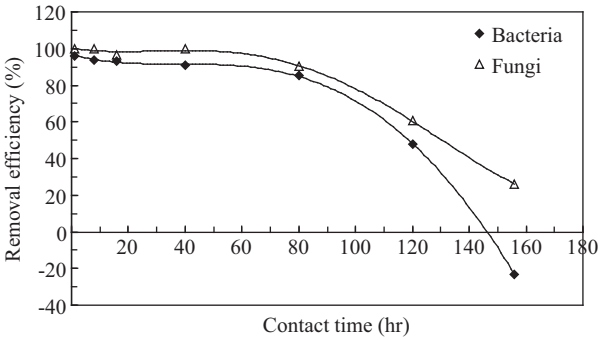

Fig. 3 Removal efficiencies of airborne bacteria and fungi.

the oxidation ditch. Only 136 CFU/m^3 of bacteria and no fungi emerged at the outlet of the adsorption tube. High removal efficiency (over 85%) was reached within 80 hr for adsorption of bacteria and fungi with the used flow rate of 1.50 m^3/hr. After this, however, they showed a gradual decrease with increase in contact time. Airborne bacteria (2677 CFU/m^3) were detected from the outlet of the adsorption tube at 156 hr, which exceeded the concentration in the inlet stream. Airborne microorganisms were difficult to capture as the adsorbent was saturated. Compared with the removal of airborne bacteria, fungi had higher removal efficiency.

The adsorption capacity of airborne bacteria and fungi on the GAC is presented in Fig. 4. It was evident that this parameter increased regularly with contact time. A gradual and linear increase in adsorption capacity occurred up to about 40 hr. Such adsorption capacity indicated that a linear relationship existed between removal rate and initial contact time, and airborne bacteria and fungi were almost completely removed. With further increases in contact time, the elimination rate increased more slowly until it reached a plateau, indicating that maximum adsorption capacity had been achieved. The GAC itself had a maximum adsorption capacity of 2217 CFU/g for airborne bacteria at 80 hr and 225 CFU/g for fungi at 120 hr. This threshold was obtained at a superficial gas velocity of 1.50 m^3/hr. The amount of bacteria captured by the GAC far exceeded that of fungi.

The GAC samples were collected from sampling ports 2, 3, and 4 to investigate the distribution of captured microorganisms in the adsorption tube. Results in Table 1 indicate that most bacteria and fungi accumulated in the middle part

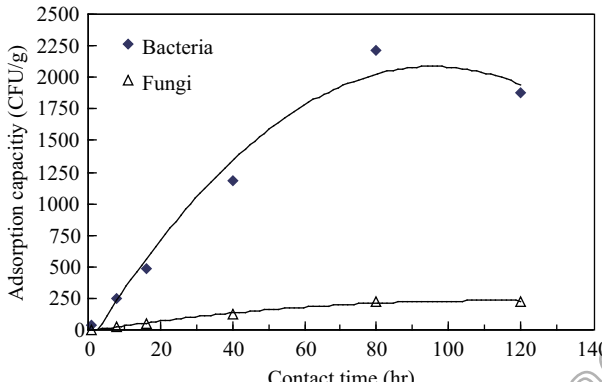

Fig. 4 Adsorption capacity of airborne bacteria and fungi on GAC.

Table 1 Distribution of captured microorganisms in the adsorption tube

Sampling position	Bacteria (CFU/g)	Fungi (CFU/g)
Sampling port 2	3.25×10^4	3.57×10^3
Sampling port 3	8.02×10^5	2.06×10^5
Sampling port 4	2.69×10^4	5.48×10^2

of the adsorption tube, while fewer microorganisms were immobilized on the adsorbent at the lower part.

2.2 Particle size distributions of airborne culturable bacteria and fungi

An Andersen sampler separates airborne particles by collecting them in stages using different hole sizes, which provides information on cell density and particle size (Andersen, 1958, Brandi et al., 2000). Large amounts of airborne bacteria and fungi were detected from particles larger than 7.0 μm or within the range of 1.1–2.1 μm (Fig. 2). Little bacteria or fungi were found in the particle size range of 3.3–4.7 μm. This was consistent with our previous results obtained from the WWTP. Bacteria are generally small and detection of bacteria with larger particle size suggests that airborne bacteria appear mainly as clusters or are attached to sludge particles or water drops when they are released from wastewater.

Figure 5 demonstrates the removal of airborne bacteria in each particle size. Particles in the range of 1.1–2.1 μm were adsorbed effectively and reached over 83.23%. Conversely, the removal of large particles (> 7.0 μm) declined dramatically. Particle size distributions of airborne bacteria in the inlet and outlet stream are shown in Fig. 6. The percentages of particles in individual sizes changed obviously after passing through the adsorption tube. Over 40% of bacteria on the particles were 1.1–2.1 μm in size, while nearly 80% of bacteria in the outlet stream were over 4.7 μm. A similar phenomenon occurred in the uptake of airborne fungi. These results indicated that airborne bacteria and fungi of large particle size were difficult to be adsorbed, while small particles were apt to be captured by the GAC.

The risks associated with exposure to aerosols may not only relate to composition (genera and species) and concentration but also size of aerosolized particles (Reponen et al., 1992; Brandi et al., 2000). Larger particles (>10 μm)

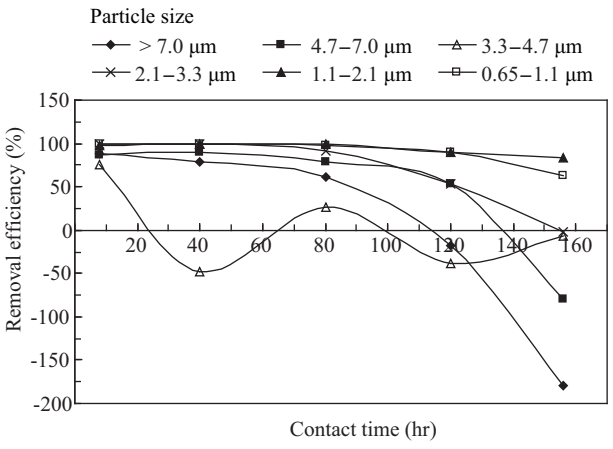

Fig. 5 Removal efficiencies of particles in each size.

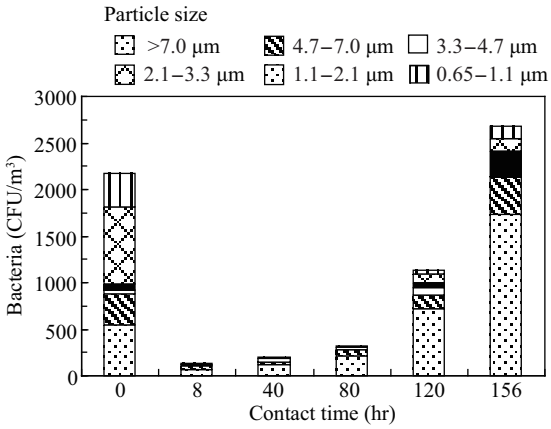

Fig. 6 Particle size distributions of airborne bacteria.

deposited in the upper airway of humans can result in fever symptoms, while smaller particles (especially < 5 μm) can penetrate the lower airways and lead to allergies or asthma. Additionally, bioaerosols within the ultra fine range (<1 μm) or submicrometer size can penetrate the deepest parts of the respiratory tract. Compared with large particles, small particles can be easily carried by the wind to distances ranging from a few hundred meters to several kilometers (Recer et al., 2001; Jones and Harrison, 2004). Such small bioaerosols from wastewater may therefore represent a health hazard not only to WWTP workers but also to nearby residents. The GAC used in this study was suitable for the capture of the small bioaerosols, which might cause greatest harm to human health.

2.3 Physicochemical characteristics of adsorbent and its adsorption capacity

Adsorption capacity was correlated with the surface physicochemical characteristics of the adsorbents. The GAC had a surface area, pore volume, and pore size of 701.90 m^2/g, 0.16 cm^3/g and 4.72 nm, respectively (Table 2). Higher surface area generally results in higher adsorption capacity. Micrographs demonstrated clearly that GAC had a rough surface (Fig. 7a) and various pore sizes were observed on the surface of the particle. Large numbers of small holes (1–2 μm) were distributed regularly in the large holes (>10 μm). Meso-macropore ranges are plotted in Fig. 8 with the curve showing a unimodal distribution, indicating that the GAC had a well-developed and uniform mesoporosity of around 10 nm in diameter. Small macrospores structure of GAC rendered it effective for the adsorption of small particles. As the size of bacteria or fungi were larger than 1.0 μm, bacteria cells or fungi spores adhered on the inner surface of large pores of GAC particle (Fig. 7b).

Surface roughness and hydrophobicity of the adsorbents are important surface characteristics for adsorption microbial cells. Microbial colonization increases with surface roughness (Rosenberg and Kjelleberg, 1986; Characklis, 1990), for example, which is mainly due to diminished shear forces and increased surface area. Most previous research has found that microorganisms attach more rapidly to hydrophobic, non-polar surfaces than to hydrophilic materials (Apilánez et al., 1998; Donlan, 2002). Hydrophobic

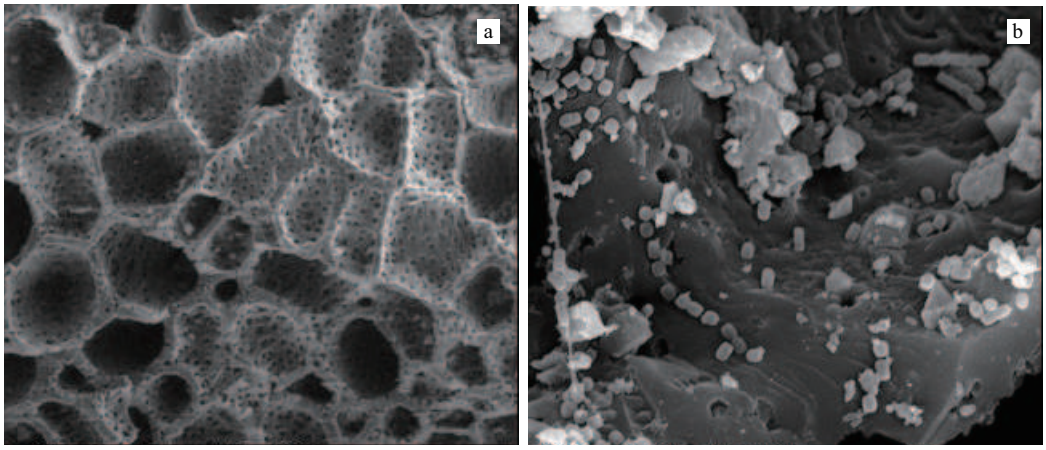

Fig. 7 Micrographs of GAC particle. (a) ×1000; (b) ×5000.

Table 2 Physicochemical characteristics of GAC

Texture		Surface functional group		Element			
Surface area	701.90 m^2/g	Carbonyl group (C=O)	0.12 mmol/g	C	82.90 wt.%	Cl	3.01 wt.%
Pore volume	0.16 cm^3/g	Phenolic group (–OH)	0.23 mmol/g	O	4.39 wt.%	Si	2.68 wt.%
Pore size	4.72 nm	Carboxylic group (–COOH)	0.18 mmol/g	P	4.88 wt.%	Ca	1.66 wt.%
						Al	0.48 wt.%

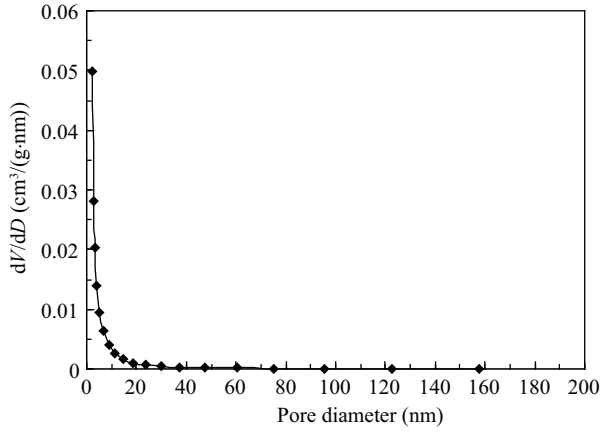

Fig. 8 Pore size distribution of GAC.

interaction between the cell's surface and the adsorbent's surface enables the cell to overcome the repulsive forces active within a certain distance from the adsorbent's surface and irreversibly attach. Microorganism adsorption on activated carbon surface increases its hydrophobicity, which benefits microorganism attachment on the surface of carbon. Porous structure, large surface area, and hydrophobic surface of the GAC used in this work favored effective attachment of airborne microorganisms.

The SEM/EDS analysis provided information on the distribution of elements in the adsorbent. Table 2 shows that GAC had elements of C, O, Ca, Al, P, Si, and Cl on the surface and inside the pore. Extracellular polymeric substances, produced by bacteria, have a binding capacity for metals, and form multiple complexes with metal ions. Metal elements, such as Al and Ca, on the surface may benefit for microbial cells adhesion.

Surface oxygenic functional groups, such as phenolic (–OH), carbonyl (C=O) and carboxylic (–COOH) groups were determined by Boehm titration. Total oxygen containing function group was 0.53 mmol/g with 22.6% (0.12 mmol/g) carbonyl group, 43.4% (0.23 mmol/g) phenolic group, and 34.0% (0.18 mmol/g) carboxylic groups. Previous research has demonstrated that most oxygen-containing groups are acidic, leading to an acidic surface. The enhancement of the acidic property results in improving the hydrophilicity of the carbon surface. Most bacteria are negatively charged, but still contain hydrophobic components on the surface (Hermansson et al., 1982). The interaction between adsorbate and adsorbents was favored by the similar chemical behavior of the surface and the adsorbate. Therefore, excess surface oxygenic functional groups, especially carboxylic group, hindered the adhesion of microbial cells.

2.4 Regeneration of activated carbon

Adsorbents should be regenerated after saturated adsorption. Two regeneration methods, UV irradiation and high pressure vapor, were compared in the present study. Figure 9 reveals an obvious decreasing trend in the amount of residue microorganisms accompanied by an increasing time of UV radiation. For fungi, high removal efficiency (91.53%) was obtained after 20 min radiation, while a similar eliminated rate (96.21%) of bacteria was reached at 60 min. Neither bacteria nor fungi were cultured from the activated carbon destroyed by high pressure vapor.

Because of the alteration in cells caused by UV rays, bacteria, viruses, moulds, and other microorganisms are unable to reproduce and may be considered inactive (Billmeyer, 1997). Some microorganisms hid in the inner holes of porous GAC (Fig. 7b), which prevented damage by UV radiation. Therefore, some microorganisms remained active and were consequently detected. Under high pressure vapor, the microorganisms on the activated

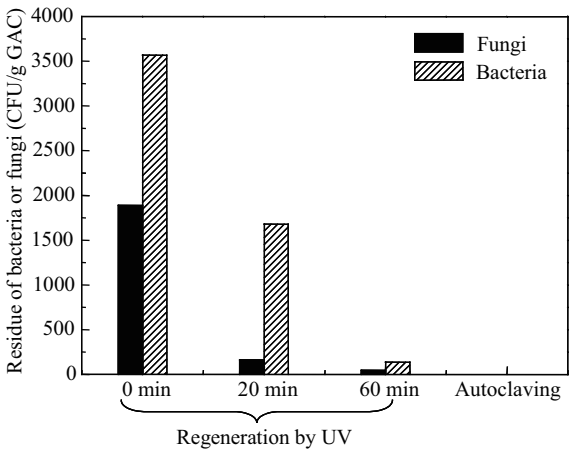

Fig. 9 Residue of bacteria and fungi on the activated carbon after regeneration.

carbon were completely destroyed, and could no longer be cultured. However, high pressure vapor for regeneration often requires special facilities and is difficult to carry out on site, which increases costs, especially for large applications.

The SEM exhibited the microorganisms on the activated carbon. As demonstrated, no microorganisms emerged on the new activated carbon (Fig. 10a). Bacillus, cocuus, and spores were found from the used activated carbon (Fig. 10b), with most adhered on the inner surface of the large holes. They were plump, smooth on the surface, and seemed vigorous. The cell well of the residue bacteria exhibited shrinkage and showed less activity after UV regeneration (Fig. 10c). No microorganisms were observed on the activated carbon regenerated by high pressure vapor (Fig. 10d), indicating it was a highly effective regeneration method.

3 Conclusions

Airborne microorganisms were adsorbed effectively on activated carbon. The GAC had a maximum adsorption capacity of 2217 CFU/g for airborne bacteria at 80 hr and 225 CFU/g for fungi at 120 hr with a flow rate of 1.50 m^3/hr. Over 85% of airborne bacteria and fungi emitted from the oxidation ditch were taken up within 80 hours of contact time, with most being 0.65–4.7 μm in size. The small macrospores structure of GAC renders it effective for the capture of small particles.

The relationships between physicochemical characteristics of GAC and its adsorption performance demonstrated that porous structure, large surface area, and hydrophobicity of the GAC rendered it an effective adsorbent of airborne microorganisms.

Both UV irradiation and high pressure vapor were

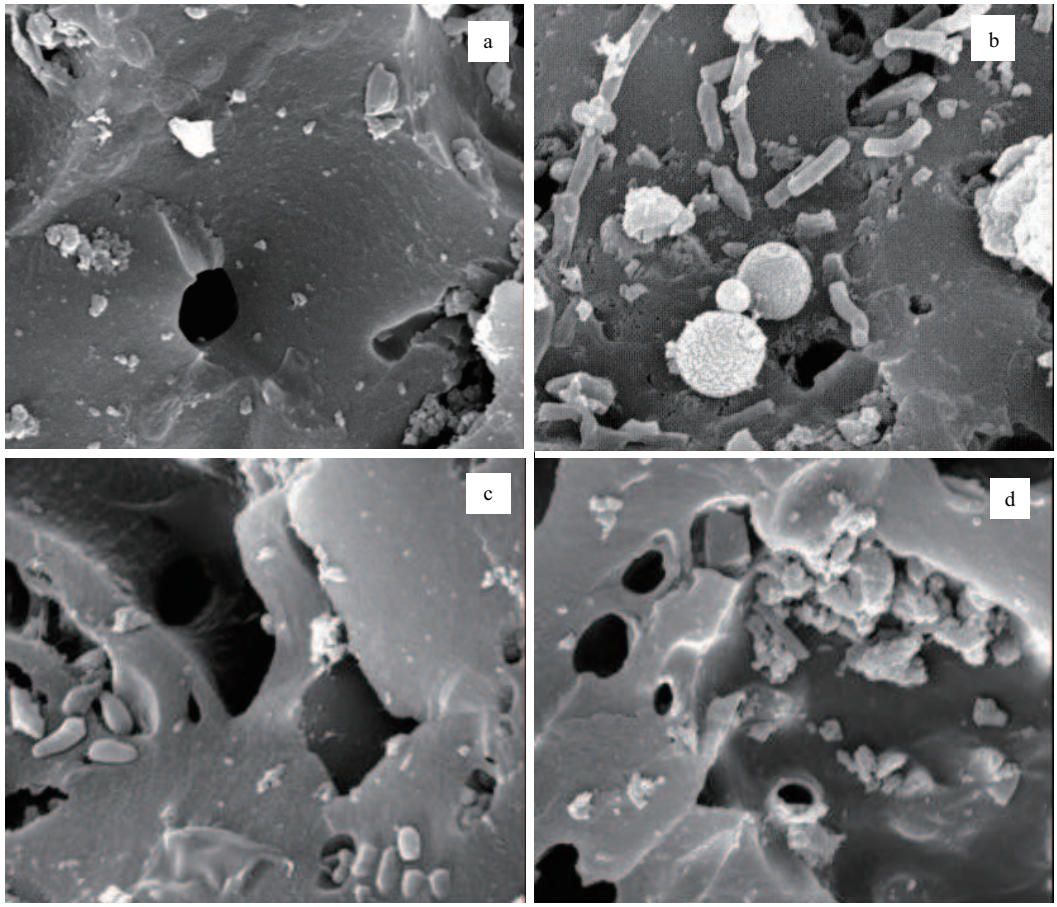

Fig. 10 SEM of the activated carbon (×10,000). (a) new activated carbon; (b) used activated carbon; (c) regenerated by UV; (d) regenerated by high pressure vapor.

compared for activated carbon regeneration. High pressure vapor proved to be an effective technique for the total destruction of microorganisms adhered on the activated carbon.

Acknowledgments

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