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Removal of volatile organic compounds by spark generated carbon aerosol particles

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Even though conventional activated carbon has been widely used for removal of volatile organic compounds (VOC), problems associated with its short and unpredictable life spans need to be solved. The present study first introduces an idea of removing benzene, toluene and xylene (BTX) gas in a controllable way through carbon aerosol particles produced by a spark generator made in our laboratory. A commercially available spark generator developed by Helsper et al. [1], Palas GFG 1000, has recently been used for production of the heterogeneous chemistry of reactive gases [2], and has been used for production of the role of adsorbed water in reactivity [3,4]. However, there have been no published studies of volatile organic compounds (VOC) gas removal by carbon airborne particles generated by any type of spark generator.

A diagram of our experimental setup is shown in Fig. 1. A cylindrical test duct with a volume of 1963 cm³ (5 cm (D) × 100 cm (L)) with duralumin body was used for this study. The carbon particles were stably generated from the spark generator. Two cylindrical graphite rods (fine extruded, 6 mm (D) × 100 mm (L) each; Graphite Store, NC001310, US) were used as electrodes, which were positioned in a glass chamber of 42.8 cm³ volume. The spark generator was operated at atmospheric temperature and pressure and dry condition (<1% R.H.). The applied voltage of −2.5 kV caused spark discharge across the electrode gap, vaporizing material from the receiving electrode and producing carbon particles by nucleation of the vapor. These particles carried by a flow of nitrogen (purity of 99.9999%) entered the test duct.

The BTX gas with a mixing ratio of 0.47:0.45:0.09 was used as the reactant gas and was acquired from a compressed gas cylinder at concentration of 981.6 ppmv, with air as the balanced gas. Flow rates of the BTX gas stream and the particle free air stream were controlled by two mass flow controllers, respectively, while the nitrogen flow rate was controlled by a mass flow meter with a needle valve. The nitrogen flow rate ranged from 0.1 to 0.5 L/min, and the total flow rate ranged from 2 to 10 L/min.

The overall removal efficiency of VOC gas (η) is defined as the difference in the BTX gas concentration between the inlet stream and the outlet stream:

$$\eta = (C_i - C_o)/C_i \qquad (1)$$

where C_i and C_o are the inlet and the outlet BTX gas concentrations (ppmv), respectively. Concentrations of BTX gas were continuously measured by a photoionization detection (PID) gas analyzer (Kinsco Inc., Sniffer II, lower detection limit of 0.01 ppmv), which monitors TVOC at a sampling flow rate of 0.5 L/min.

The ratio between the supply rate of pore volume and the supply rate of BTX gas volume (R) is

$$R = C_p Q V_p / C_i Q f_c \qquad (2)$$

where C_p (mg/m³) is the mass concentration of the generated carbon particles, Q (m³/min) is the total-flow rate,
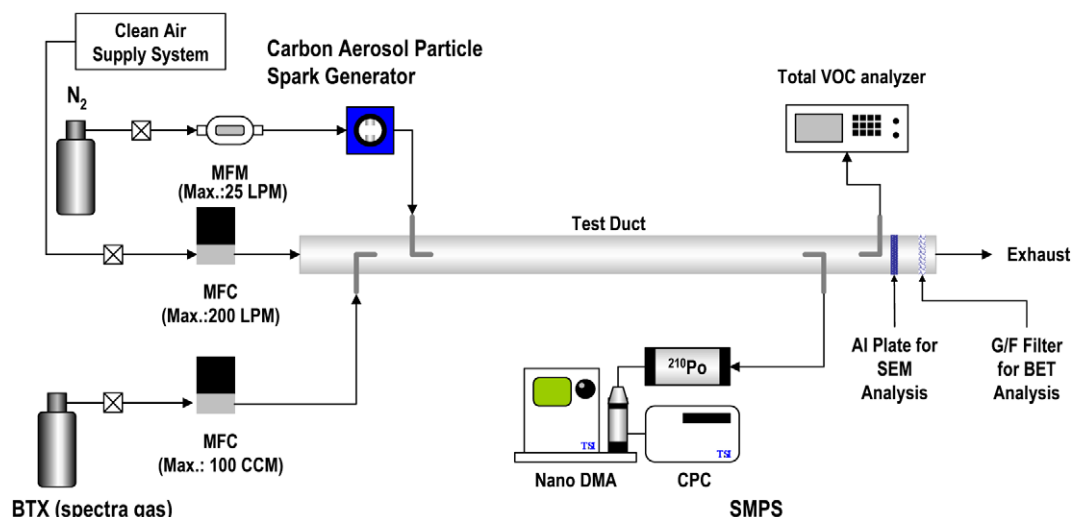
Fig. 1. Schematic of experimental set-up.

V_p is the pore volume (m^3/g) of the generated carbon particles, and f_c is the unit conversion factor (2.81×10^{-4} m^3-BTX mole/g-BTX mole). For complete removal of BTX gas ($\eta = 1$), R should be greater than 1, since all BTX gas molecules are supposed to be adsorbed on the pores of carbon particles.

BET surface area (m^2/g), micropore area (m^2/g), average pore diameter (Å), micropore volume (cm^3/g), and total pore volume (cm^3/g) of the particles were obtained by N_2 adsorption at 77 K by using a micromeritics ASAP 2010. To determine the mass concentration of the particles (C_p), size distribution and number concentration of the generated carbon particles were measured by a scanning mobility particle sizer (SMPS, TSI Custom) at a sampling flow rate of 0.3 L/min. Scanning electron micrographs (SEM) of the generated particles were obtained by using a SEM apparatus (Cambridge S250 MK2, operated at 20 kV).

The size distribution of the particles obtained by SMPS is shown in Fig. 2. The size ranged from several tens to hundreds of nanometers and their geometric mean diameter was about 95 nm, resulting in the total mass concentration of about 1.31 g/m^3. Fig. 2 also shows a SEM micrograph (20,000×) of carbon particles. It was expected that the aggregates, composed of primary nanoparticles, might contribute to the increase of the BET surface area and total pore volume, and thus, to higher chance of VOC adsorption onto the carbon particles.

Table 1 summarizes the characteristics of our carbon graphite electrode, spark generated carbon particles, and conventional activated carbon. Table 2 summarizes the data of η and R for BTX gas. The total pore volume (for <769 Å of pore size) of 1.62 cm^3/g and micropore volume (<20.9 Å) of 0.033 cm^3/g were used in the calculations of R_T and R_M (refer to Eq. (2)), respectively. The theoretical
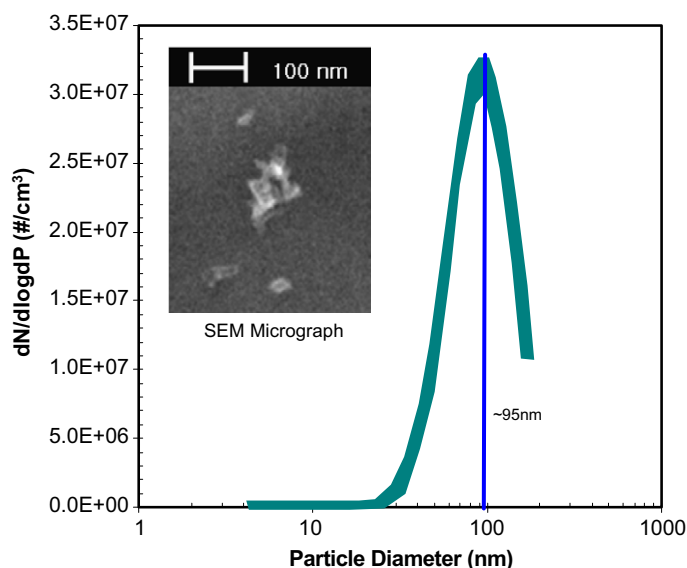

Fig. 2. Morphology and size distribution of generated carbon particles.

Table 1
Properties of the graphite rod, the generated carbon particles, and conventional activated carbon

Samples	BET surface area (m²/g)	Micropore area (m²/g, <20.9 Å)	Microporosity (%)	Average pore diameter (Å)	Micropore volume (cm³/g)	Total pore volume (cm³/g, <769 Å)
Carbon graphite electrodes	6.66	0.24	3.54	118	∼0	0.012
Spark generated carbon particles	639	29	4.51	104	0.033	1.62
Conventional activated carbon	800–1500	300–1000	20–60	∼20	0.3–1.4	0.5–2.0

Table 2
Data η and R based on pore volume

Residence time (min) [total-flow rate (L/min)]	0.5 ppmv			1.0 ppmv		
	η	R_T	R_M	η	R_T	R_M
0.983 [2]	0.04	4.24	0.086	ND	2.12	0.043
0.483 [4]	ND	4.09	0.083	ND	2.07	0.042
0.200 [10]	ND	4.03	0.082	ND	2.01	0.040

ND, not detected.

value of R required for the complete removal of BTX gas is 1.0 if all gas molecules are assumed to be adsorbed in all the pores of carbon particles. For R_T, however, the removal efficiency η for 0.983 min of residence time and 0.5 ppmv of BTX gas initial concentration was 4%, even though the supply rate of pore volume was 4.24 times higher than the supply rate of BTX gas volume. Such a low removal efficiency might be caused by inadequate BTX gas-to-particle collisions, which depend on the pore size distribution of the particles. When the residence time was 0.483 or 0.2 min for 0.5 ppmv of BTX gas concentration, the removal efficiency could not be obtained due to the relatively shorter particle residence time. With 1.0 ppmv BTX gas, the removal efficiency could not be obtained, regardless of residence time, due to detectable limit of the gas analyzer.

The values of R_M were obtained with Eq. (2) based on micropore volume, rather than the total pore volume, and are also listed in Table 2. All the values of R_M are much lower than 1.0. Therefore, such a low removal efficiency of 4% might be caused by the insufficient supply of micropore volume of carbon particles, even though the total pore volume and the BET surface area of the particles were similar to those of conventional activated carbon. One possible reason for this problem is the ineffective pore size distribution of the carbon particles for BTX gas adsorption as a result of ineffective interstitial behavior among the primary carbon particles.

The pore size distribution of the carbon particles that were collected on a glass fiber filter is shown in Fig. 3. As discussed with Table 1, the micropore volume of the carbon particles was very small compared to that of a conventional activated carbon. Much higher micropore volume of the particles would result in higher removal efficiency.

Conclusively, the use of the spark generator slightly enhanced the removal of the BTX gas (4% of removal efficiency) because the spark generator produced insufficient micropore volume of the generated carbon particles compared to that of conventional activated carbon. To increase
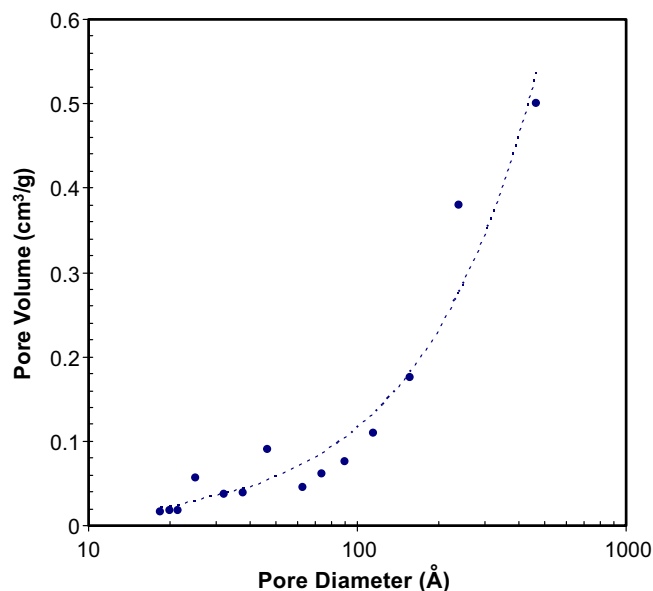

Fig. 3. Pore size distribution of generated carbon particles.

the micropore volume would require the change of characteristics of carbon aerosol particles, including size distribution and morphology, by modifying the spark generation system.

Acknowledgements

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