



Superiority of wet-milled over dry-milled superfine powdered activated carbon for adsorptive 2-methylisoborneol removal



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ABSTRACT

Superfine powdered activated carbon (SPAC), which is produced from conventionally sized powdered activated carbon (PAC) by wet milling in a bead mill, has attracted attention for its high adsorptive removal ability in both research and practice. In this study, the performance of dry-milled SPAC was investigated. 2-Methylisoborneol (MIB), an earthy-musty compound commonly targeted by water treatment systems, was used as the target adsorbate. Dry-milled SPAC exhibited lower adsorptive removal of MIB than wet-milled SPAC, even when both SPACs were produced from the same PAC and were composed of particles of the same size. One reason for the lower removal of MIB by the dry-milled SPAC was a higher degree of aggregation in the dry-milled SPAC after production; as a result the apparent particle size of dry-milled SPAC was larger than that of wet-milled SPAC. The dry-milled SPAC was also more negatively charged than the wet-milled SPAC, and, owing to its higher repulsion, it was more amenable to dispersion by ultrasonication. However, even after the dry-milled SPAC was ultrasonicated so that its apparent particle size was similar to or less than that of the wet-milled SPAC, the dry-milled SPAC was still inferior in adsorptive removal to the wet-milled SPAC. Therefore, another reason for the lower adsorptive removal of dry-milled SPAC was its lower equilibrium adsorption capacity due to the oxidation during the milling. The adsorption kinetics by SPACs with different degrees of particle aggregation were successfully simulated by a pore diffusion model and a fractal aggregation model.

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1. Introduction

Powdered activated carbon (PAC) is used widely in water treatment processes because of its ability to remove dissolved pollutants that cannot be removed by the conventional process of coagulation, sedimentation, and sand filtration (Cook et al., 2001; Li et al., 2003; Matsui et al., 2003; Shi et al., 2014). Recently, activated carbon with a much finer particle size than PAC (superfine powdered activated carbon, SPAC) has been intensively investigated, and SPAC is already being used in some water treatment plants (Bonvin et al., 2016; Ellerie et al., 2013; Heijman et al., 2009; Jiang et al., 2015; Matsui et al., 2007; Wang et al., 2011). Compared with PAC, which is composed of particles with diameters of tens of micrometers, SPAC, which consists of particles with diameters of no more than a few micrometers, has advantages with regard to both adsorption capacity and kinetics because of both its larger specific

external surface area per unit of mass and the shortened diffusion distance within each carbon particle (Ando et al., 2010; Matsui et al., 2009, 2015, 2013; Sontheimer et al., 1988).

In all scientific studies and actual field applications to date, wet-milling systems have been used to produce SPAC. In wet-milling systems, the PAC is suspended in carrier water to make a slurry, and then the slurry is introduced into a bead mill to produce SPAC (Partlan et al., 2016). However, the production efficiency of such small-sized particles by this wet-milling system is limited because the viscosity of the carrier fluid, and thus the slurry concentration, must be low (Knieke et al., 2010; Schmidt et al., 2012). The low concentration of the slurry also increases the bulk volume of the SPAC and thus the cost of its transport after production. Even if SPAC is produced onsite in water treatment plants, the SPAC slurry has a large storage footprint.

In addition to wet-milling systems, dry-milling systems have recently become available that can successfully produce submicrometer-sized particles of various materials, such as intermetallics (Suryanarayana, 2001), boron (Jung et al., 2015), and boron carbides (Ramos et al., 2006). Unlike wet-milling systems,

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dry-milling systems utilize gas or steam, rather than a liquid, as the carrier of the particles during comminution by the milling medium. Compared with wet milling, dry milling mitigates the high production volume problem, reducing both the size of the storage footprint and the transport volume. In addition to these advantages, the dry-milling process can reduce the size of the milled particles at a higher rate and the particles may be less likely to become contaminated by milling medium residues, compared with the wet-milling process (Jung et al., 2015).

Despite these merits of dry milling compared with wet milling for the production of fine milled particles, adsorption by dry-milled SPAC has not yet been investigated. Therefore, in this study, the adsorption performance of dry-milled SPAC was compared with that of wet-milled SPAC. To evaluate the adsorption ability of the SPACs, we chose 2-methylisoborneol (MIB) as a representative target compound for adsorptive removal. MIB, which is produced by the metabolism and biodegradation of actinomycetes and cyanobacteria (Watson et al., 2000; Westerhoff et al., 2005), is extremely refractory to removal by conventional water treatment processes (Srinivasan and Sorial, 2011), and it is a conventional target compound of activated carbon adsorption processes during water treatment (Cook et al., 2001; Gillogly et al., 1998; Matsui et al., 2008, 2013; Newcombe et al., 1997, Newcombe et al. 2002; Pendleton et al., 1997). The results of the comparison showed that the adsorption performance of the dry-milled SPAC was inferior to that of the wet-milled SPAC, so we further investigated the reasons for the different performances.

2. Materials and methods

2.1. Adsorbents

Two commercial wood-based PACs were obtained and designated PAC-S (Shirasagi, Osaka Gas Chemical Co., Osaka, Japan) and PAC-F (Futamura Chemical Co., Tokyo, Japan). Dry-milled SPACs for use in the experiments were produced by milling each PAC to the target size in a dry-milling system (SDA5, Ashizawa Finetech Ltd., Chiba, Japan; bead diameter, 1.5 mm; carrier, air), and wet-milled SPACs were separately produced in a wet-milling system (LMZ015, Ashizawa Finetech Ltd., Chiba, Japan; bead diameter, 0.1 mm; carrier, pure water). All activated carbons, including non-milled PACs, wet-milled SPACs, and dry-milled SPACs, were stored in a slurry (1%–5% w/v) made with pure water at 4 °C after vacuum conditioning to remove any air from the activated carbon pores. Aliquots of the dry-milled SPAC slurry were ultrasonicated (150 W, 19.5 kHz, 1 min for 50 mL carbon slurry) for dispersion. The SPACs were labeled according to the milling process used and the PAC (-S or -F) from which they were produced; for example, SPAC-S-D was produced by dry milling from PAC-S, and ultrasonicated dry-milled SPAC that was produced from SPAC-S-D was designated SPAC-S-DS (Table 1).

Particle size distributions of the SPACs and PACs were determined by using a laser-light scattering instrument (Microtrac MT3300EXII, Nikkiso Co., Tokyo, Japan). To measure the true particle size of the carbon samples, the samples were taken from the stock slurries and then pretreated by the addition of a dispersant (Triton X-100, Kanto Chemical Co., Tokyo, Japan; final concentration, 0.08% w/v) and subsequent ultrasonic dispersion before measurement with the Microtrac instrument. The apparent particle sizes were measured without this pretreatment. Elemental analysis to determine the oxygen content was performed on the SPACs by using an elemental analyzer (Vario EL Cube, Elementar Japan K.K., Yokohama, Japan) equipped with thermal conductivity and infrared detectors.

2.2. Adsorbate and working solutions

A stock solution of MIB (~900 µg/L) was prepared by dissolving pure MIB (Wako Pure Chemical Industries, Ltd., Osaka, Japan) in ultrapure water and then filtering the solution through a 0.2-µm pore size membrane filter (DISMIC-25HP; Toyo Roshi Kaisha, Ltd., Tokyo, Japan). Organic-free water was spiked with the MIB stock solution to obtain working solutions with MIB concentrations of around 1 and 4 µg/L, because MIB usually occurs naturally at concentrations lower than these concentrations. The working solutions were adjusted to pH 7.0 ± 0.1 with HCl or NaOH as required. The MIB concentration was detected by using deuterium-labeled geosmin (Wako Pure Chemical Industries, Ltd., Osaka, Japan) as an internal standard in a purge and trap concentrator (Aqua PT 5000 J; GL Sciences, Inc., Tokyo, Japan) coupled to a gas chromatograph–mass spectrometer (GCMS-QP2010 Plus; Shimadzu Co., Kyoto, Japan), where the *m/z* 95 and *m/z* 115 peaks were interpreted as corresponding to MIB and deuterium-labeled geosmin, respectively.

2.3. Batch adsorption tests

The adsorption kinetics was investigated by means of batch tests with efficient mixing. A working solution of water and MIB was placed in a beaker (1 L or 3 L). After the addition of a specified amount of SPAC or PAC slurry, aliquots were withdrawn at intervals and filtered immediately through 0.2-µm membrane filters (DISMIC-25HP, Toyo Roshi Kaisha, Tokyo, Japan) for analysis of the MIB concentration.

For the adsorption equilibrium tests, aliquots (110 mL) of the working solutions (hereafter, water samples) containing MIB (1 ± 0.2 µg/L) were transferred to 120-mL vials. Specified amounts of SPAC or PAC were immediately added (0.3–1.8 mg/L), and the vials were manually shaken and then agitated on a mechanical shaker for 1 week at a constant temperature of 20 °C. Preliminary experiments confirmed that the adsorption equilibrium of MIB was reached after 1 week of contact (Matsui et al. 2012, 2013). Control tests of water samples that did not contain carbon (i.e., blanks) were also conducted to confirm that changes in the blank MIB concentration during long-term mixing were negligible. After 1 week of shaking, the water samples were filtered through 0.2-µm pore size membrane filters, and then MIB concentrations in the aqueous phase were measured. Solid-phase concentrations of MIB were calculated from the mass balance.

2.4. Pore diffusion model for the batch adsorption process

These experimental data of adsorption kinetics were simulated with a pore diffusion model (PDM, see SI) that was modified to incorporate adsorbent particle aggregation. In the model, each adsorbent particle aggregate is treated as a single adsorbent unit; thus, for simplicity, diffusion in interparticle pores between the adsorbent particles composing the aggregate and diffusion in the pores of the individual adsorbent particles are merged. Although the shell adsorption model is used to simulate the different isotherms of PAC and wet-milled SPAC (Matsui et al., 2013, 2014), it is not yet possible to use this model to simulate the different isotherms of wet-milled and dry-milled SPACs, which have a similar particle size. Therefore, because of its wide application and simplicity, we decided to use the Freundlich model.

The apparent density (ρ) and porosity (ϵ) of an aggregate in PDM equation can be estimated by using fractal theory (Elimelech et al., 2013) as follows:

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