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Short Communication

A selective dielectrophoretic particle separator using flat electrodes covered with vertically aligned carbon nanotubes



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HIGHLIGHTS

• Carbon nanotubes were grown on flat electrodes for dielectrophoresis.

• TiO₂ nanoparticles were captured selectively from SiO₂ by CNT electrodes.

• Small particles resembling CNT diameters can be preferably captured.

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ABSTRACT

In this short communication, a dielectrophoretic (DEP) particle separator is proposed that is capable of selectively capturing extremely small particles, using flat electrodes onto which carbon nanotubes (CNTs) have been synthesized. CNTs can induce a strong DEP force at their surface, which allows extremely small particles, whose diameters are comparable to those of the CNTs, to be preferably captured by these electrodes. This study experimentally demonstrates that TiO₂ particles with diameters of approximately 10–20 nm are selectively captured from a mixture with SiO₂ particles with similar diameters by the separator using CNTs grown on the flat electrodes. For comparison, a conventional separator using CNT-grown mesh-stacked electrodes can not exhibit such selective capture from the same TiO₂–SiO₂ mixture. © 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Some conventional methods for selectively collecting particles from liquid dispersions containing multiple types of particles include filtering (Lorenzen et al., 2014; Ong et al., 2014; Iritani et al., 2015; Sugiyama et al., 2015; Sun et al., 2015), sedimentation (Durst and Raszillier, 1989; Tambun et al., 2012; Zhang et al., 2015), and centrifugation (Cao et al., 2014; Teduka and Nishioka, 2015; Zhu and Liow, 2014). However, these methods become less effective when there is no significant difference in sizes and densities of the particles, at which point dielectrophoretic (DEP) particle separation becomes a move attractive candidate for separation. In particular, DEP particle separation excels when used in the separation of particles whose dielectric properties are different. In addition, the particle size is also an important factor in determining the DEP forces present.

So far, many types of DEP particle separators have been proposed. Mostly, DEP separators using micro-patterned electrodes or

* Corresponding author. Tel.: +81 75 383 2684; fax: +81 75 381 2821. *E-mail address:* sano@cheme.kyoto-u.ac.jp (N. Sano). microfluidic channels (Kwon et al., 2008; Nam et al., 2013) have been investigated for developing particle separators on compact scales, with analytical applications as the end target. However, for the future scale-up of separators, it is important to develop electrodes for DEP separation which have suitable shapes; previouslydeveloped shapes include packing of dielectric beads in DEP separators (Suehiro et al., 2003), wetted-wall patterned-electrode DEP separators (Sano et al., 2011), and mesh-stacked DEP separators (Sano et al., 2012). Flat electrodes are also suitable for large-scale particle separation if they are subject to appropriate surface modification, because they can be expanded easily.

The development of DEP separators for the collection of extremely small particles is another challenge in this field. Because the DEP force is considered to be proportional to the cube of the particle diameter (Pohl, 1978; Jones, 1995; Hughes, 2002), collection of extremely small particles becomes very difficult when using this DEP force. To solve this problem, the use of electrodes onto which carbon nanotubes (CNTs) have been directly synthesized has been proposed (Sano et al., 2014) in a scheme where titanate nanoparticles were purified by a DEP particle separator using mesh stacked electrodes onto which CNTs were grown.

However, this electrode configuration was shown to have drawbacks in its selectivity. In this short communication, a new type of CNT-grown flat electrode is proposed for the assembly of DEP particle separators with increased selectivity capabilities. Experiments were conducted using this separator to demonstrate improved particle separation for the collection of small TiO₂ particles from a mixture with SiO₂ particles.

2. Experimental

Two types of DEP particle separators were prepared for comparison as schematically explained in Fig. 1, with one being the separator using mesh-stacked electrodes, and the other using flat electrodes placed in parallel. The electrodes were made of stainless steel (SUS316). On the surface of these electrodes, CNTs were directly synthesized to realize intensive DEP forces at the CNTs. The separator using the CNT-grown mesh-stacked electrode is referred to here as a CNT-MS separator, while the one using CNTgrown flat electrode is called a CNT-FE separator. Ethanol dispersed with anatase TiO₂ particles and SiO₂ particles was fed into these separators to evaluate their selectivity for particle capture.

The size of these particles was determined using a transmission electron microscope (TEM; JEOL, JEM1010), and the diameters of both of these particles were in the range of 10–20 nm, as seen in Fig. 2a and b. The initial TiO₂:SiO₂ weight ratio dispersed in ethanol was 1:1, and the weight concentration of these particles in ethanol was 1.2 g/L. The change of the component ratio of these particles due to capturing was analyzed by energy-dispersive X-ray (EDX; Technex Lab Co., Tiny-EDX(LE)- α) spectra.

The CNT-MS separator consisted of 10 stainless steel meshes (inter-wire aperture=0.5 mm, wire diameter=0.29 mm) stacked with acrylic spacers of 0.5 mm thickness. The total surface area of the stainless steel meshes used for the electrodes in the CNT-MS separator was estimated to be approximately 92 cm². The polarity of the voltage (200 V) applied to the adjacent mesh sheets was set to alternate so that DEP force could be generated in the zones between adjacent mesh sheets. The details about the operation of the CNT-MS separator can be found elsewhere (Sano et al., 2014). In brief, the CNT-MS separator was first submerged in ethanol dispersed with particle mixtures of anatase TiO₂ and SiO₂. After applying voltage to the CNT-MS separator in order to capture the

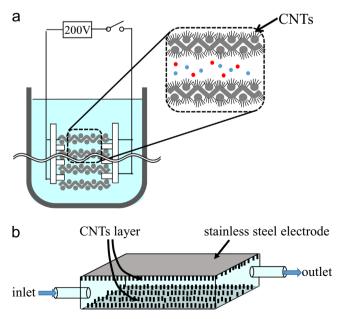


Fig. 1. Schematic of (a) the CNT-MS separator and (b) the CNT-FE separator.

particles, the separator was lifted up and transported into another ethanol reservoir to release captured particles through removal of the applied voltage. This cycle was repeated 10 times to accumulate particles.

The CNT-FE separator consisted of two flat stainless steel plates as electrodes, which were placed in parallel using acrylic spacers of 3 mm thickness. The total surface area of the electrodes facing the zone used for DEP particle capture was 18 cm², about one-fifth of the area used by the CNT-MS separator. An inlet and an outlet of liquid were placed on opposite sides of the electrodes to ensure a continuous flow of the particle-dispersed ethanol (1 mL/min) in DEP field. Because the inter-electrode distance in the CNT-FE separator was different from that in CNT-MS separator, the voltage used in the CNT-FE separator was set to be higher than in the CNT-MS separator so that the mean electric field at the electrode surface would be similar for the two separator types. As a result, a DC voltage of 1000 V was applied between the electrodes in the CNT-FE separator.

CNTs were synthesized directly on the surface of the stainless steel electrodes used for the separators via a previously-published surface modification technique (Sano et al., 2014). All steps for the CNT synthesis were carried out in a quartz tube (inner diameter=42 mm) placed in an electric furnace. The stainless steel electrodes were first oxidized in air at 800 °C for 30 min, and then reduced at 700 °C for 40 min under a H₂-Ar atmosphere (H₂ flow rate=95 cm³/min, Ar flow rate=290 cm³/min). By using this surface treatment, catalytic Fe nanoparticles were generated on the surface of stainless steel so that CNTs could be synthesized on the surface with addition of ethylene at 700 °C for 30 min. The images obtained using TEM and scanning electron microscopy (SEM; JEOL, JSM-6701F) are shown in Fig. 2c and d. From these images, it is clear that the diameters of most CNTs were similar to that of the particles fed to the separators, i.e., 10–20 nm.

3. Results and discussion

Before explaining the particle separation results, it is necessary to describe the unique particle-separation character of the CNT-FE separator. If an ordinary dielectrophoretic separator is used, larger particles can be more easily captured as shown by Eqs. (1) and (2) (Pohl, 1978; Jones, 1995; Hughes, 2002).

$$F = 2\pi r^3 \varepsilon_m \text{Real} [K] \nabla E^2 \tag{1}$$

$$K = (\varepsilon_{\rm p}^* - \varepsilon_{\rm m}^*) / (\varepsilon_{\rm p}^* + 2\varepsilon_{\rm m}^*) \tag{2}$$

where, F, r, $\varepsilon_{\rm m}$, E, and Real [K] are the DEP force, particle radius, permittivity of liquid medium, electric field strength, and real number in the complex number of K, respectively. The parameter K is called the Clausius-Mossotti factor, and is defined by Eq. (2), where $\varepsilon_{\rm p}^*$ and $\varepsilon_{\rm m}^*$ are the complex permittivity of the particles and liquid media, respectively. When the value of Real [K] is positive, particles tend to move toward high electric field zones (positive DEP force), while a negative value of Real [K] causes the particles to move in the opposite direction (negative DEP force). It has been previously reported that the values of Real [K] of SiO₂ and TiO₂ dispersed in ethanol are negative and positive, respectively (Sano et al, 2011). Furthermore, the calculated electric field profile around protruded electrode suggests that TiO₂ which is supposed to receive positive DEP force should be attracted into high electric field area at the protruded part of the electrode and inversely SiO₂ should be repulsed from there, and these results theoretically predicted can be observed experimentally (Sano et al., 2011). In addition, because F is proportional to r^3 in Eq. (1), it is expected that small particles (with size on the nanoscale) should be difficult to capture using DEP forces.

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