



Removal characteristics of engineered nanoparticles by activated sludge



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HIGHLIGHTS

- Three engineered nanoparticles (Ag, TiO₂, SiO₂) were removed by activated sludge.
- This occurred in a time-dependent manner.
- The removal efficiencies depended on the type of nanoparticles.
- The presences of both ionic compounds and EPS enhanced the removal efficiency.

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ABSTRACT

Environmental release of engineered nanoparticles (NPs) has been on the rise due to the increased use of NPs in commercial products. In addition, the fate of NPs in sewage treatment processes may play an important role in determining the environmental release pathway of NPs. In this study, we investigated the removal of engineered NPs (AgNPs, TiO₂NPs, and SiO₂NPs) using activated sludge by evaluating the effects of several important factors of the NPs, including physicochemical properties, contact time between NPs and activated sludge, aquatic chemistry of sewage, and the presence of extracellular polymeric substances (EPS) in the activated sludge. For all three types of NPs tested, a considerable amount of NPs were removed after exposure to activated sludge in a time-dependent manner; nevertheless, the removal efficiencies depended on the type of NPs and seemed to be affected by the NP stability relative to the hydrodynamic diameter (HDD) and zeta potential. In addition, the presences of both ionic compounds and EPS significantly enhanced the NP removal efficiency, indicating that the instability of the NPs resulting from the ionic strength in sewage and entrapment of NP by EPS played an important role in NP removal by activated sludge. These results suggest that the removal efficiencies can be affected by the operating conditions of the activated sludge process and the conditions of the activated sludge; therefore, these factors should be considered when developing approaches to sufficiently remove NPs from sewage treatment plants.

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1. Introduction

Recently, commercial use of nanoparticles (NPs) in chemical products has dramatically increased with the development of nanotechnology due to their unique properties; extremely high surface area and reactivity (Nel et al., 2006; Zhang et al., 2011). According to a recent survey, NPs are widely used even in daily life products, including personal care, clothing, and cosmetics. In addition,

the number of these products in the commercial market has significantly increased in recent years (Project on Emerging Nanotechnologies, 2011). Therefore, the potential risk of releasing NPs into the natural environment threatens human health and ecosystems (Handy and Shaw, 2007; Bae et al., 2010; Akaighe et al., 2011; Dallas et al., 2011), suggesting that the fate and transport of NPs should be investigated thoroughly to minimize their negative environmental risk.

Sewage treatment processes may play an important role in determining the environmental pathways and disposal of NPs that are incorporated into different products (Jarvie et al., 2009; Brar et al., 2010). Typical domestic sewage treatment plants (STPs) mostly operate conventional or modified activated sludge processes and several previous studies have confirmed that a

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considerable amount of the NPs released into the activated sludge process can be absorbed by the activated sludge, resulting in a reduction of NPs in the effluent and an accumulation of biomass (Kiser et al., 2009; Liang et al., 2010; Kaegi et al., 2011). Even though several studies reported the fate of NPs in wastewater treatment plants, the removal characteristics of NPs by activated sludge are not well investigated. The particle instability by wastewater in the absence of activated sludge was just examined (Limbach et al., 2008; Jarvie et al., 2009) or the simple fate of nanoparticles in wastewater treatment plants (Kiser et al., 2009; Kaegi et al., 2011; Wang et al., 2012) was reported.

The aim of this study was to investigate the removal characteristics of engineered NP by activated sludge. Silver (Ag), titanium dioxide (TiO₂), and silica dioxide (SiO₂) NPs were selected as model NPs due to their high potential for environmental release, high quantities of these NPs in production and their wide spread use in commercial products (Bradford et al., 2009; Project on Emerging Nanotechnologies, 2011). The goal of this study was to better understand the removal characteristics, effects of physicochemical characteristics of NPs, contact time related to HRT, aquatic chemistry, and EPS on NP removal.

2. Materials and methods

2.1. Preparation and characterization of nanoparticle suspensions

Stock suspensions of AgNPs coated by citric acid were purchased from ABC Nanotech Co. (STU206011, Korea). The stock suspension containing 20 wt.% AgNPs was serially diluted in DI water (Millipore, France) to the desired concentration. Suspensions of TiO₂NPs and SiO₂NPs, TiO₂NPs and SiO₂NPs were respectively purchased from Evonik Co. (P25, Germany) and Sukgyung AT Co. (Korea) and were suspended in DI water by mixing at 200 rpm for 24 h. The NP sizes in the prepared suspension were determined by transmission electron microscopy (TEM, JEM-3010, JEOL, Japan) and electron light scattering spectrophotometry (ELS-8000, Otsuka, Japan). ELS was also used to evaluate the zeta potential (ζ), which is the magnitude of the electrical charge at the double layer. The pH of the NP suspension was measured using a pH meter F-54BW (Horiba, Japan). The pH of all NP suspensions was 7.0 ± 0.5, which would unlikely change the physical or chemical interactions between the activated sludge and NPs. Thus, no additional pH adjustments were performed.

2.2. Synthetic wastewater

Synthetic wastewater was prepared using glucose (Sigma-Aldrich, USA) with a target chemical oxygen demand (COD) concentration of 200 mg L⁻¹. The synthetic wastewater also contained micronutrients including 10 mg L⁻¹ yeast extract, 10 mg L⁻¹ bacto-peptone, 50 mg L⁻¹ (NH₄)₂SO₄, 30 mg L⁻¹ K₂HPO₄, 30 mg L⁻¹ KH₂PO₄, 1.8 mg L⁻¹ MgSO₄, 0.04 mg L⁻¹ FeCl₃, 1.4 mg L⁻¹ NaCl, 0.04 mg L⁻¹ CaCl₂, 0.48 mg L⁻¹ CoCl₂, and 30 mg L⁻¹ NaHCO₃, which was consistent with the composition of synthetic wastewater previously reported (Yeon et al., 2008) with a slight modification. The ionic strength of the synthetic wastewater was ca. 3 mM.

2.3. Activated sludge preparation and EPS removal

The activated sludge was prepared from a lab-scale continuous bioreactor with a total reactor volume of 3.5 L. The bioreactor with ca. 3.0 L of mixed liquor at the mixed liquor suspended solids (MLSS) concentration of ca. 3000 mg L⁻¹ was operated at a hydraulic retention time (HRT) of 8 h and a solids retention time (SRT) of 10 d. Fresh activated sludge obtained from the aeration tank of the Recopark sewage treatment plants (Gimpo, Korea), was acclimated by feeding the synthetic wastewater for more than two months in

the lab-scale continuous bioreactor. An aliquot (ca. 20 mL) of the acclimated activated sludge was collected from the bioreactor and centrifuged at 1000g for 10 min to separate activated sludge from the synthetic wastewater. The pellet was resuspended in DI water or synthetic wastewater, and then the adsorption experiment was conducted.

The EPS of the sampled activated sludge was extracted using ethylenediaminetetraacetate (EDTA) as described previously (Cao et al., 2011). Briefly, the sludge was exposed to 25 mM of EDTA containing a 0.9% NaCl solution (pH 7) at 4 °C for 3 h. The microorganisms and EPS of the sludge were then separated by centrifugation three times at 5000g with DI water. The polysaccharide concentration of the EPS solution was quantified using the phenol/sulfuric method as described elsewhere (Dubois et al., 1956). Two milliliters of the extracted EPS solution, 0.025 mL of phenol, and 2.5 mL of sulfuric acid were mixed for 30 min, and then the absorbance of the solution was measured at 490 nm using an Agilent 8453 UV/vis spectrophotometer (Agilent, Germany).

2.4. Batch adsorption experimental with activated sludge

Approximately 3000 mg L⁻¹ MLSS of the activated sludge was exposed to the desired concentration of the NP suspensions at 100 rpm and 25 °C. After the indicated contact time, the activated sludge was centrifuged at 500×g for 20 min, and the supernatant was withdrawn and the NP concentration quantified. The concentration of NPs was quantified by measuring the elements (Ag, Ti, and Si) using inductively coupled plasma (ICP) spectrometry and by taking into account the effluent concentration of the sewage treatment process. For Ag and Ti quantification, and ICP-Mass Spectrometer (Elan 6100, Perkin-Elmer SCIEX, Canada) was used. The Si content was measured using an ICP-Optical Emission Spectrometer (ICP-730 ES, Varian, Australia). The detection limits of both ICPs were approximately 1 µg L⁻¹. In addition, the percent removal efficiency was calculated using the following equation:

$$\text{NP removal}(\%) = \frac{C_{\text{influent}} - C_{\text{effluent}}}{C_{\text{influent}}} \times 100(\%)$$

where C_{effluent} is the NP concentration in the effluent after the contact time and C_{influent} is the initial NP concentration in the influent. All experiments were conducted under dark conditions to prevent the photocatalytic activity of TiO₂. The results are presented as the means and standard deviations from duplicate experiments.

2.5. TEM analysis of nanoparticles in activated sludge

All chemicals used in the TEM analysis were purchased from Electron Microscopy Sciences Co. (Hatfield, USA). The TEM specimens obtained from the sludge exposed to the NPs were prepared as described previously (Lee et al., 2008). The sludge was concentrated by centrifugation at 2500g for 2 min to remove the supernatant. The collected sludge was fixed in 2% glutaraldehyde and 0.05 M sodium cacodylate buffer (pH 7.2) for 2 h and washed three times with 0.05 M sodium cacodylate buffer. The samples were then exposed to 1% osmium tetroxide in 0.05 M sodium cacodylate buffer for 2 h and stained with 2% uranyl acetate for 18 h. Next, with the samples were dehydrated through sequential treatment of 30%, 50%, 70%, 80%, 90%, and 100% ethanol for 10 min. The samples were infiltrated in propylene oxide, embedded in Spurr's resin, and polymerized at 70 °C for 24 h. The prepared polymerized blocks were sectioned using an ultramicrotome (MT-X, RMC, USA) and TEM (JEM-3010, JEOL, Japan) analysis was conducted.

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