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Use of filtration techniques to study environmental fate of engineered metallic nanoparticles: Factors affecting filter performance

Frederik Van Koetsem^{a,*}, Simon Verstraete^a, Elien Wallaert^b, Kim Verbeken^b, Paul Van der Meeren^c, Jörg Rinklebe^d, Gijs Du Laing^a

^a Laboratory of Analytical Chemistry and Applied Ecochemistry, Department of Applied Analytical and Physical Chemistry, Faculty of Bioscience Engineering, Ghent University, Coupure Links 653, Ghent, Belgium

^b Department of Materials Science and Engineering, Ghent University, Technologiepark 903, Zwijnaarde, Belgium

^c Particle and Interfacial Technology Group, Department of Applied Analytical and Physical Chemistry, Faculty of Bioscience Engineering, Ghent University, Coupure Links 653, Ghent, Belgium

^d Soil- and Groundwater-Management, Institute for Soil Engineering, Water and Wastewater Management, University of Wuppertal, Pauluskirchstraße 7, Wuppertal, Germany

HIGHLIGHTS

- Filters with pore sizes larger than the particle size could still retain the ENPs.
- Nanoparticle retention seemingly increased with decreasing initial concentration.
- Retention appeared to depend more on particle characteristics than membrane type.
- Centrifugal-UF using 10 kDa filters is recommended for removing ENPs from solution.
- 10 kDa UF might allow differentiation between particulate and dissolved species.

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ABSTRACT

We examined the filtration of aqueous suspensions of negatively charged (citrate-stabilized) Ag (14.5 ± 1.1 nm) and positively charged CeO₂ (7.3 ± 1.4 nm) engineered nanoparticles (ENPs) via different filtration techniques such as paper filtration, micro- and ultrafiltration, and evaluated the impact of initial concentration, matrix composition, and filter type and (pre-)treatment, on nanoparticle retention. Solutions of Ag⁺ and Ce³⁺ ions were tested in the same way. Significant retention of nanoparticles was observed even for filters having considerably larger pore sizes than the ENPs size. Retention also seemingly increased with decreasing initial concentration, but generally decreased upon preconditioning of the paper or membrane filters with diluted nitric acid or 0.1 M Cu(NO₃)₂, respectively. In ultrapure water, retention appeared to depend more on particle characteristics than on a membrane type. However, in 2 mM KNO₃, NaCl, or CaCl₂, more significant differences in recovery were observed between different membrane materials. Additionally, background electrolytes might reduce nanoparticle or ionic retention, but could also affect their (colloidal) stability, e.g., resulting in enhanced retention of Ag ENPs and Ag⁺ ions in chloride-containing matrices. Results from centrifugal ultrafiltration recommend using 10 kDa filters for nanoparticle removal from the solution, and suggest these filters might potentially be suitable to differentiate between (nano)particulate and dissolved species.

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1. Introduction

The rapid developments in nanoscience and nanotechnology during the last decades have allowed the extraordinary size-related properties of engineered nanoparticles (ENPs) (1–100 nm) to be implemented in a variety of commercial and industrial applications including cosmetics, personal care products, textiles, household appliances, food additives, food packaging, paints, coatings, cat-

* Corresponding author.

E-mail addresses: frederik.vankoetsem@gmail.com (F. Van Koetsem), Gijs.DuLaing@UGent.be (G. Du Laing).

alysts, fuel additives, fuel cells, electronics, and (waste)water treatment [41,32]. In October 2013, the Woodrow Wilson database had already listed 1628 nanotechnology-based market available consumer products in their online inventory compared to only 212 in March 2006, representing an increase by approximately 670% [36]. However, the increasing production and use of ENPs in such applications will also inevitably result in their (unintentional) discharge into aquatic environments (e.g., surface water or groundwater), where they could pose a risk to living organisms and human health (e.g., via drinking water consumption or transfer through the food chain) [27,41,32]. For instance, ENPs have already been demonstrated to be released from consumer goods [3], during washing of textiles [4,14,40], and via surface run-off from coated façades [20,19], and have been detected in effluents from wastewater treatment plants [21,39], indicating that conventional (waste)water treatment processes might be insufficient in removing ENPs [34,41]. Therefore, increasing concern has been raised within the scientific community regarding the occurrence, behaviour, and fate of these emerging contaminants in aquatic systems [28], in particular on their removal and fractionation methodologies [42].

Filtration processes are promising options when studying ENPs, not only for retaining and removing nanoparticles from solution, but also as fractionation techniques during characterization or sample preparation prior to further analysis [22,42]. Microfiltration (MF) using 0.45 μm cut-off filter membranes is commonly utilized to determine the dissolved metal fraction in e.g., surface waters or soil solutions [7,16], while 0.2 μm filtration is routinely used for sterilization of for instance nutrient media [38]. However, solely based on size, nanoparticles should be allowed to permeate during MF due to their extremely small dimensions, thereby ending up in the so-called “dissolved” fraction. On the other hand, nanoparticle-membrane interactions (e.g., sorption) or membrane fouling could lead to retention of particulates that are smaller than the filter pore size [22,41,18]. Ultrafiltration (UF) is already being extensively applied during drinking water treatment, and is anticipated to be an effective removal strategy of ENPs from drinking water sources as nanoparticles or their aggregates are often too large to pass through UF filters [27,42]. In contrast to MF, where the retention capability is basically expressed through size exclusion from a well-defined pore size of the filter membrane ($>0.1 \mu\text{m}$), UF membranes are characterized by their molecular weight cut-off (MWCO), which refers to the approximate molecular weight of a compound that is retained for 90% by the filter membrane [41]. Ultrafiltration has been regularly utilized in trace metal speciation studies [33,16], or in studies involving nanoparticles [8,7,30,34,9,42], primarily to investigate removal of (nano)particulate matter from solution, to examine dissolution behaviour by differentiating between (nano)particulate and “truly” dissolved compounds, or to characterize and purify nanoparticle dispersions.

The membrane-fouling phenomena and other filtration induced artefacts occurring during trace metal analysis have been investigated [13,38,29,16].

However, only few studies have investigated the removal and retention behaviour of metallic ENPs upon filtration of aqueous samples. Especially information regarding nanoparticle-filter interactions and how they are potentially influenced by membranes, solutions or particle properties are scarce.

Therefore, studying factors affecting the separation and removal efficiency, and the underlying mechanisms are important for human health and from environmental point of view because they could promote the potential use of membrane filtration in, for example, the removal of ENPs from water samples. For this study, two types of metallic ENPs, negatively charged citrate-stabilized Ag ENPs and positively charged CeO_2 , were chosen based on their environmental relevance as well as on particle characteristics. Cur-

rently, Ag ENPs are the most widely used engineered nanomaterials in consumer goods, mainly due to their antimicrobial properties [7,36], whereas the catalytic and UV-blocking properties of CeO_2 ENPs have led to their extensive use e.g., as diesel fuel additives or in cosmetics [7]. Both types have already been demonstrated to cause toxicological responses in aquatic organisms [37,23].

The aim of this work was to study the retention behaviour of aqueous suspensions of Ag and CeO_2 ENPs upon application of several different filtration techniques (i.e., paper filtration, syringe filtration (0.02 and 0.45 μm), and centrifugal micro- and ultrafiltration), in order to aid in providing useful information on for instance, the removal of nanoparticles from (waste)water streams, preventing their environmental release, or on their size fractionation and characterization in aqueous samples, and ultimately, in assessing their behaviour and fate in aquatic environments. In particular, the effects of nanoparticle concentration, matrix composition, and filter type, pore sizes and (pre-)treatments, on the recoveries of silver or cerium in the filtrates were examined, and compared to the recoveries of Ag^+ or Ce^{3+} ions treated in the same way, and potential interaction mechanisms were investigated. Additionally, several nanoparticle digestion methods were assessed for quantification of total silver or cerium content in solution.

2. Materials and methods

2.1. Nanoparticles and chemical reagents

Citrate-stabilized silver nanoparticles (Ag ENPs, ca. 10 nm, 100 mg L^{-1} , ζ -potential: $-50 \pm 5 \text{ mV}$, pH: 6–8) and cerium dioxide nanoparticles (CeO_2 ENPs, $4 \pm 2 \text{ nm}$, 50,000 mg L^{-1} , ζ -potential: $+45 \pm 5 \text{ mV}$) were obtained from PlasmaChem GmbH (Berlin, Germany) as aqueous dispersions. Ionic silver (as AgNO_3) and cerium (as $\text{Ce}(\text{NO}_3)_3$) reference standard solutions (Plasma HIQU, $1000 \pm 2 \mu\text{g Ag}^+$ or $\text{Ce}^{3+} \text{ mL}^{-1}$ in 2–5% HNO_3), and analytical grade nitric acid (HNO_3 , 65%, ρ : 1.39 g mL^{-1} , M : 63.01 g mol^{-1}), hydrochloric acid (HCl, 37%, ρ : 1.19 g mL^{-1} , M : 36.46 g mol^{-1}), hydrogen peroxide (H_2O_2 , 30 wt%, ρ : 1.11 g mL^{-1} , M : 34.01 g mol^{-1}), sodium chloride (NaCl, >99.8%, ρ : 2.17 g cm^{-3} , M : 58.44 g mol^{-1}), calcium chloride ($\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, >99.5%, ρ : 1.85 g cm^{-3} , M : 147.02 g mol^{-1}), potassium nitrate (KNO_3 , >99%, ρ : 2.11 g cm^{-3} , M : 101.10 g mol^{-1}), and copper nitrate ($\text{Cu}(\text{NO}_3)_2 \cdot 2\frac{1}{2}\text{H}_2\text{O}$, 99–102%, ρ : 2.05 g cm^{-3} , M : 232.59 g mol^{-1}) were all purchased from ChemLab NV (Zedelgem, Belgium).

2.2. Nanoparticle characterization

Photon correlation spectroscopy (PCS) (Malvern PCS-100, Malvern Instruments Ltd., Worcestershire, UK) was applied to determine the particle size distributions (PSDs) of the Ag and CeO_2 ENPs. Sample measurements were performed in quadruplicate, at 25 °C using a 633 nm HeNe laser positioned at a measuring angle of 150°. Cumulant and multi modal analysis were applied to obtain the so-called Z-average hydrodynamic particle diameter and PSDs, respectively. The surface charge of the nanoparticles was assessed through zeta (ζ) potential measurements (Zetasizer 3000 HSA, Malvern Instruments Ltd., Worcestershire, UK). Transmission electron microscopy (TEM) equipped with energy dispersive X-ray spectrometry (EDS) (JEM-2200FS, Jeol Ltd., Tokyo, Japan) was used to visualize the ENPs. Absorption spectra were determined via UV-vis spectroscopy (6400 Spectrophotometer, Jenway, Bibby Scientific Ltd., Staffordshire, UK).

2.3. Comparison of nanoparticle digestion methods

Different digestion media were tested for their applicability in the digestion of Ag and CeO_2 ENPs (Table 1). The ENPs (10 mL) were

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