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A review: inhibition of Ag NPs on wastewater treatment

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ABSTRACT

This review summarizes and evaluates the present knowledge of the inhibition effect of silver nanoparticles (Ag NPs) on pollutants removal, activated sludge performance. The fate and behavior of Ag NPs in wastewater treatment plants (WWTPs) are also mentioned. Rapid progress in this area has been made over the last few years, but there are still some blanks in studies. Although some researches have been carried out on the inhibitory of Ag NPs on pollutants removal such as COD and nitrogen removal, most of them focus on microorganisms rather than treatment system. Few articles referred to impact of Ag NPs on phosphorus removal. Mechanisms of Ag NPs inhibition are still poorly understood although it seems clear that in some cases. As a well performance microcosmic ecosystem, the status of fungus, protozoa, and metazoan are also very important for activated sludge. However, little is known about the impact of Ag NPs on them. This review concludes with a set of recommendations for the advancement of understanding of the role of Ag NPs in WWTPs.

Keywords: Silver nanoparticles Ag NPs; Wastewater treatment plants WWTPs; Inhibition effect; Activated sludge

1. Introduction

Engineered nanoparticles (ENPs) are materials with at least one dimension of 100 nm [1,2]. In that size range, ENPs have physicochemical and electronic properties that greatly differ from the bulk counterpart, including much higher specific surface area, surface reactivity, and increased quantum effect [3]. Due to these unique properties, ENPs are being incorporated into numerous products and industrial processes such as cosmetics [4,5], health care products [6–8], food packing materials [9], clothing, and many other consumer goods [10]. The nanomaterials is growing rapidly, in 2007, 380 nano-enable consumer products were identified; three years later it exceeded 1,317 and the trend continues increasing [11].

However the special properties that make ENPs unique and useful may also cause some nanomaterials to pose hazards to humans and the environment under certain conditions. A number of authors have published literatures on characterization, fate, and toxicological information of nanomaterials and proposed research strategies for evaluation of safety of nanomaterials [12–20]. Not only the researchers but also the worldwide organizations are increasingly focused on the potential risk of ENPs. To help member countries efficiently and effectively address the safety challenges

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of nanomaterials, working party on manufactured nanomaterials (WPMN) was established by organisation for economic cooperation and development in 2006. WPMN is implementing its work through eight main areas [21], and put forward a list of priority nanomaterials for immediate testing [22].

Silver nanoparticles (Ag NPs) are one of the 13 engineered nanomaterials listed and they are currently the most widely used ENPs in consumer products with 259 out of 1,015 commercially available nanoparticle-based products containing nanosilver [23]. Ag NPs have distinctive physicochemical properties as electrical and thermal conductivity, surface-enhanced Raman scattering, chemical stability, catalytic activity, and non-linear optical behavior [24,25]. These properties make them of potential value in inks, microelectronics, and medical imaging [26]. Furthermore, with exceptional broad spectrum bacteriocidal activity [27–30] and relatively low cost of manufacturing [24], Ag NPs have been extremely popular used in a diverse range of consumer materials, including plastics, soaps, pastes, metals, and textiles [31].

With expanding use of nanosilver products, the release of Ag NPs to the environment cannot be completely avoided and has already been demonstrated. For example, textiles with nanosilver released Ag NPs during washing process [32,33] and emission from painted into rainwater that runs off a facade [34]. It has been predicted that the amount of silver released into wastewater from silver-containing products would reach a maximum of 410 tons per year for European countries alone in 2010 [36]. A significant fraction of Ag NPs can be expected to reach municipal and industrial wastewater treatment plants (WWTPs), since large fractions of Ag NPs are released to sewer systems [35–38]. Thus WWTPs plays an important role in controlling the release of nanoparticles into the environments, such as surface waters and land [39–41]. Given the antimicrobial activity of Ag NPs, the potential risk of Ag NPs effect on microorganisms leading to weaken treatment efficiency in wastewater treatment system cannot be overlooked. However, reports on these aspects are scarce [42–47].

The purpose of this review is to critically evaluate the existing knowledge of Ag NPs negative impact on WWTPs as a potential problem for environmental risk, taking into consideration the inhibition on organic pollutants removal, nitrification inhibition, and the fate, behavior of Ag NPs in effluent and sewage sludge products. From the current information on Ag NPs and their role in WWTPs, we then identify the current knowledge gaps.

2. Impact of Ag NPs on pollutants removal

2.1. Impact of Ag NPs on COD removal

Since healthy growth of heterotrophic and autotrophic microorganisms in activated sludge is crucial to the removal of organic matter from wastewater, the antibacterial properties of Ag NPs suggest its potential adverse impact on organic pollutants removal efficiency. Liang et al set up a modified ludzack–ettinger (MLE) process to evaluate the potential negative impact of Ag NPs on wastewater treatment and to determine the bacterial response to a shock load of Ag NPs (12 h of constant Ag NPs loading rate with final peak concentration of 0.75 mg/L)[48]. Their results indicate that there is no significant difference between the effluent COD concentrations before and after silver shock load. The average effluent COD concentrations before and after nanosilver shock load are 10.2 ± 4.4 and 10.2 ± 3.1 mg/L. The similar conclusions are obtained by Hou et al [49]. They use three simulated sequencing batch reactors (SBR) to compare the potential adverse effect of Ag NPs on COD removal efficiency among control dosage (without Ag NPs), low dosage (0.1 mg/L Ag NPs), and high (0.5 mg/L Ag NPs) dosage. The results show that average effluent COD in the dosed SBR reactors are not statistically different from control reactors. In some cases, COD in the treated reactors were even lower than in the controls. Y.F. Wang compare the effect of silver, titanium dioxide, and C_{60} (fullerene) nanoparticles on COD removal within SBR [50]. The similar conclusions are obtained that presence of nanoparticles do not adversely affect COD removal in the biological reactors.

These results strongly indicate that there are no evidences of low dosage Ag NPs would cause negative impact on organic pollutants. The phenomenon can be interpreted as two reasons. One is that low concentration Ag NPs could not inhibit heterotrophic activities due to the bacterial tolerance [48]. The other is that Ag NPs in biological environments (e.g. wastewater and sludge) may undergo rapid chemical changes within a relatively short period of time. Most Ag in sludge and effluent transform into other morphology or chemical compounds. This chemical change of Ag NPs may partly account for the lack of toxicity to bacteria responsible for COD removal.

However, very little information is currently available on impact of high concentration Ag NPs on wastewater treatment system and relevant studies are still at natal stage (Table 1).

2.2. Inhibitory effect of Ag NPs on nitrogen removal

2.2.1. Inhibitory effect of Ag NPs on microorganisms

With the use of Ag NPs in consumer products continuously on the rise, the accumulation of Ag NPs in WWTPs may lead to the disruption of important biological processes within the WWTP, including nitrogen removal [51]. The removal of nitrogen from wastewater is a two-step process in which ammonia (NH_3), the most common form of nitrogen in a WWTPs, is aerobically oxidized to nitrate (NO_3^-), via ammonia oxidizing bacteria (AOB) and nitrite oxidizing bacteria (NOB), before being anaerobically transformed nitrogen gas via denitrifying bacteria. AOB are widely considered to be the most sensitive microorganisms in WWTPs and are inhibited by a wide variety of contaminants, including Ag^+ and Ag NPs [52,53].

Choi and Hu use extant respirometry to evaluate the enriched nitrifying bacteria isolated from WWTPs nitrification inhibition of Ag NPs (mean diameter = 15 nm), Ag^+ ($AgNO_3$ solution) and AgCl (Colloid solution) [54]. In their experimental evidences, nitrification is significantly inhibited by nanosilver and Ag NPs are even more detrimental to nitrifying organisms than Ag^+ and AgCl at 1 mg/L Ag. Through cell membrane integrity test, they demonstrate the main reason of nitrification inhibition of Ag NPs is high intercellular reactive oxygen species induced by membrane-attached Ag NPs without compromising the cell membrane and Ag NPs are adsorbed to the microbial surfaces, probably causing cell wall pitting [55].

Radniecki et al choose *Nitrosomonas europaea* as model of AOB and compare its sensitivity to Ag^+ and Ag NPs (mean diameter = 20 nm and 80 nm) [56]. Unlike traditional aquatic toxicity studies [57,58], Ag NPs are not dissolved but rather suspended and their stability in suspension without organic stabilizing agents in their research. This research demonstrates that Ag^+ shown higher level of toxicity to *N. europaea* than Ag NPs (20 nm) and Ag NPs (80 nm). The increased nitrification inhibition on smaller Ag NPs is caused by their higher rates of dissolved silver release, via dissolution, due to a greater surface area to volume ratio. The inhibition mechanisms between Ag^+ and Ag NPs are similar with both causing decreases in ammonia monooxygenase enzyme AMO activity and destabilization of the outer-membrane of *N. europaea*.

Table 1
Experimental parameters of Ag NPs inhibition on wastewater treatment system

Process	MLE	SBR	
Parameter			
Mean diameter	1–29 nm	23 ± 10 nm	
Concentration	0.75 mg/L	0.1 mg/L	0.5 mg/L
Influent	Synthetic wastewater	Raw wastewater	

Ag NPs with the diameters of 9, 13, 15, and 23 nm and concentrations range from 0.12 to 4.82 mg/L are synthesized to test the inhibition effect of AOB and NOB cultivated from sludge of WWTPs in Zhang's research [59]. It is found that AOB is much more sensitive than NOB, since the NH_4^+–N removal efficiency is decreased by nearly 70% by the addition of nanoparticles with size of 23 nm when the silver concentration reached 4.82 mg/L, while NO_{2-}–N removal efficiency is merely affected by silver nanoparticles. Among four different species of Ag NPs, the smallest particles (9 nm) have the highest concentration as well as the most severe inhibition effect to nitrifier. However, the one with diameter of 9 nm also have the most severe toxicity when the concentration was the same.

With the scanning electron microscope imaging, it is observed that the nanoparticles are attached to microbial extracellular polymeric substances (EPS) and bacteria. It is assumed that the reaction would first happen in the cell membrane between silver nanoparticles and the substance composing membrane. After the reaction, cell membrane would be damaged and then large amounts of particles would then enter the cell, causing the death of cell.

2.2.2. Inhibitory effect of Ag NPs on activated sludge

The nitrogen removal inhibition degree of Ag NPs between activated sludge and microorganisms are significantly different. According to Hou's results, the slight initial reduction and subsequent quick recovery of NH_4 removal during the SBR process indicate impact of Ag NPs on NH_4 removal is minimal [49]. As high as 6 mg/L of sulfide concentrations in free ion form in the influent, Ag NPs added into raw wastewater may transform into form of silver sulfide (Ag_2S) rapidly which partly account for the lack of toxicity [60].

However Zhang presents different conclusions [59]. Ag NPs with different sizes and concentrations all have evident inhibition on nitrification process with both activated sludge for obligated nitrogen removal (decreasing 30–40%) and municipal wastewater treatment (decreasing 20%). He insisted the risk by

silver nanoparticles in municipal wastewater treatment should be taken into account. The results of Ag NPs shock load effect on activated sludge show a prolonged period of nitrification inhibition (>1 month, the highest degree of inhibition = 46.5%) and increase of ammonia/nitrite concentration in wastewater effluent [50]. The different conclusions above could be due to the different initial wastewater quality and also various diameters and concentration of nanoparticles.

2.3. Effect of Ag NPs on wastewater biofilms

Microbial biofilms are more tolerant to antimicrobial agents than are planktonic bacteria in wastewater treatment process [61–63]. Sheng and Liu compare the impact of Ag NPs (mean diameter = 15 nm) between original wastewater biofilms and the one with loosely bound EPS removal [64]. It is found that original wastewater biofilms are highly tolerant to the Ag NPs treatment. With an application of 200 mg Ag/L Ag NPs, the reduction of biofilms bacteria measured by heterotrophic plate counts is insignificant after 24 h. However, the viability of wastewater biofilms is reduced after the removal of loosely bound EPS when treated under the same conditions. Biofilms can provide physical protections for bacteria under Ag NPs treatment, and EPS may play an important role in this protection.

3. Impact of Ag NPs on sludge anaerobic digestion

Although anaerobic digestion is a key step for sludge stabilization and methane production which is widely used in WWTPs, there is very little research focus on antimicrobial activity of Ag NPs against anaerobic microorganisms. Yang et al investigated the impact of Ag NPs on anaerobic glucose degradation, sludge digestion, and methanogenic assemblages with Ag NPs at the concentrations up to 40 mg Ag/L (13.2 g silver/kg biomass COD) [65]. The results show that Ag NPs at moderate concentrations have negligible impact on methanogenic assemblages and anaerobic production. All the samples exhibit similar profiles of acetate and propionic acid as the only intermediate fermentation products, which last in a short time. More than 90% of Ag NPs are removed from the liquid phase and associated with the sludge, while almost no silver ions are released from Ag NPs under anaerobic conditions.

4. The fate and behavior of Ag NPs in WWTPs

WWTPs are considered to be key intermediate stations that control the most prominent flows of Ag between anthropogenic and environmental compartments [37,66,67]. During wastewater treatment process, Ag NPs maybe incorporated into the sewage sludge matrix through aggregation and/or sorption reactions and may be concentrated over time. A batch adsorption experiment with Ag NPs and wastewater biomass show that they would be likely accumulated in the active sewage sludge, in a manner similar to that of ionic Ag [51]. Sewage sludge samples are collected from WWTPs of Midwest region of the USA with high Ag content (856 mg/kg) to identify their perspectives on the presence of the particular Ag NPs in the final sewage sludge products. It is found that final existence of Ag NPs in the sewage sludge materials was α-Ag_2S nanoparticles. The α-Ag_2S nanocrystals were in the size range of 5–20 nm with ellipsoidal shape, and formed small loosely packed aggregates. Some of them have excess S on the surface under S-rich environments, resulting in a ratio of Ag to S close to Kaegi et al investigate the behavior of Ag NPs in a pilot wastewater treatment plant fed with municipal wastewater. The treatment plant consists of a nonaerated and an aerated tank and a secondary clarifier. Ag NPs are spiked into the nonaerated tank and samples were collected from the aerated tank and the effluent. The results show that most Ag in the sludge and effluent present as Ag_2S and the transformation from Ag NPs to Ag_2S nanoparticles occurs in the nonaerated tank within less than 2 h. The mass balance shows 5% of the added Ag left the WWTPs via the effluent, 85% end up in the excess sludge and 5% still remain in the WWTPs.

5. Conclusion and recommendation

The use of nanomaterials and their potential environmental and human health risks is of increasing concern and social debate and has been the subject of many government reports. This review has outlined the current knowledge and gaps on Ag NP as a potential problem for wastewater treatment. With the existing information we identify the current research gaps and needed areas of research:

(1) Although some researches have been carried out on the inhibitory of Ag NPs on pollutants removal such as COD and nitrogen removal, most of them focus on microorganisms rather than treatment system. In addition, few articles refer to impact of Ag NPs on phosphorus removal. Therefore, the effect of Ag NPs on treatment efficiency of WWTPs is largely remained unknown.

(2) As a well performance microcosmic ecosystem, the status of fungus, protozoa, and metazoan are also very important for activated sludge. However, little is known about the impact of Ag NPs on them in activated sludge.
(3) Most research results and conclusions mentioned above are obtained at low concentration according to current Ag NPs environmental concentration. Another pertinent issue that needs to be addressed is whether the high concentration Ag NPs in wastewater will cause abnormal operation of WWTPs.
(4) The nanoparticles may impact the performance of waste treatment processes by various mechanisms, including inhibition of microorganisms, increasing the turbidity, fouling of membranes, or affecting the efficiency of disinfection processes. Unfortunately, there are not sufficient research data to illustrate the exact mechanisms.

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