

Optimal Design of Ceramic Water Filters Impregnated with Silver Nanoparticles for Point-of-Use Water Treatment

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Abstract

In the developing world, point-of-use (e.g. household-level) water treatment has been endorsed by the World Health Organization as an effective and potentially sustainable approach for treating water for domestic use. Ceramic water filters impregnated with silver nanoparticles (Ag-NPs) were studied because Ag-NPs have remarkable antimicrobial properties, and they function to significantly improve the microbiological quality of treated water when incorporated into ceramic water filters. Because there is little quantitative data on how the quantity and application method of the Ag-NPs affect filter performance, more experiments and data that quantitatively study and describe these design variables are needed. The objective of this research project is to optimize the amount and application method of the Ag-NPs to maximize pathogen disinfection while minimizing release of silver into the treated water. This study investigates the application of the silver prior to firing, which is expected to reduce the release of silver nanoparticles from the ceramic, provide a more uniform distribution of the Ag-NPs throughout the filter, and improve disinfection efficiency relative to the conventional design. To test these hypotheses, two types of ceramic filters were tested: (i) filters without Ag-NPs; and (ii) filters with Ag-NPs incorporated into the ceramic prior to firing. The technological performance of each filter type was quantified by passing an aqueous solution containing *E. coli* at 10^9 cfu/100 mL through the filter and sampling the effluent to determine the percent removal (log reduction). A conventional membrane filtration procedure detected approximately 400 cfu/100mL of *E. coli* passing through both types of filters suggesting that ceramic filters with and without Ag-NPs are highly effective at removing bacteria. While this is a promising result for filter manufacturers, more extensive testing will be necessary in order to determine whether silver treatment helps prevent bacteria growth in the filter, and thus, enhancing long-term performance. These findings will improve water quality and human health in some of the world's most impoverished communities.

Introduction

In the developing world, point-of-use (e.g. household-level) water treatment has been endorsed by the World Health Organization as an effective and potentially sustainable approach for treating water for domestic use. We propose to study ceramic water filters impregnated with silver nanoparticles (Ag-NPs). Ag-NPs are particles of metallic (zero-valent) silver typically fabricated by the reduction of ionic silver (Ag^+) in the presence of a capping agent such as citrate. Ag-NPs have remarkable antimicrobial properties, and when incorporated into ceramic water filters, they function to significantly improve the microbiological quality of the treated water. The objective of this research project is to optimize the amount and application method of the Ag-NPs to maximize pathogen disinfection while minimizing release of silver into the treated water.

Ceramic filters can be produced using local materials consisting of clay, water, and sawdust that are mixed, pressed into the shape of a pot and fired in kiln; the sawdust combusts producing micron-sized pores through which the water can flow. Turbidity and some microbial pathogens are removed by physical filtration. An Ag-NP solution is typically painted onto the inside and outside of the after firing.

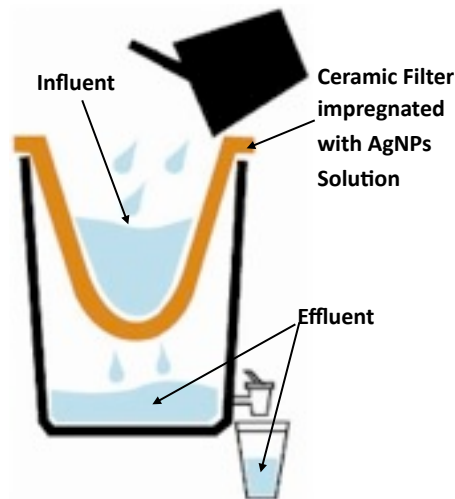


Figure 1: Ceramic water filter used to purify water at the household level³.

This study seeks to investigate application of the silver prior to firing. It was hypothesized that this application method would reduce the release of silver nanoparticles from the ceramic, provide a more uniform distribution of the Ag-NPs throughout the filter, and improve disinfection efficiency relative to the conventional design.

To test these hypotheses several mechanistic experiments in the laboratory were to be performed. Three types of ceramic filters were to be tested: (i) filters without Ag-NPs; (ii) filters with Ag-NPs incorporated into the ceramic prior to firing; and (iii) filters with Ag-NPs incorporated into the ceramic filter after firing. Due to time constraints and some unforeseen challenges that arose with the conditioning of the filters, only filters without Ag-NPs and filters with Ag-NPs incorporated before firing were tested for this work. The technological performance of each filter type was quantified by passing an aqueous solution containing *E. coli* at 10^9 cfu/100 mL through the filter and sampling the effluent to determine the percent removal (log reduction). *E. coli* was quantified by a conventional membrane filtration procedure. Effluent Ag^0 was to be quantified for each experiment by measuring the total Ag using atomic absorption spectroscopy with a silver lamp and then measuring Ag^+ using an Ag specific-ion electrode. Ag^0 was the difference between total Ag and Ag^+ . Unfortunately, due to mechanical problems with the atomic absorption spectrophotometer and some problems with the Ag^+ ion probe interface, total Ag^0 could not be quantified for the filters containing Ag-NPs.

Materials and Methods

*Cultivating E. coli Bacteria*¹

To cultivate a fresh batch of *E. coli* bacteria a broth was prepared consisting of 0.5 grams of yeast extract, 0.5 grams of sodium chloride, and 0.25 grams of tryptone were dissolved in 50-mL of distilled water in a 225-250-mL flask. This broth would serve as a source of carbon, nitrogen, and vitamins to feed the bacteria. Next, the flask was capped with a porous cap, and then autoclaved for 25 minutes at 14.7 psi at 240° F in order to sterilize the flask and mixture. Once cooled, 0.050 mL of thawed

¹ Adopted from Standard Methods written by graduate student Erin Kallman

concentrated *E. coli* was added and then the flask was put on a shaker and incubated for 12 hours at 35 °C at 200 rpm.

12 hours later, the solution was poured in a plastic 50-mL centrifuge tube and then centrifuged at 2500 rpm for 25 minutes. This was undertaken to separate the bacteria and leave them in a concentrated pile at the bottom of the mixture. The supernatant was poured out and 10 mL of random motility buffer, or RMB (10 mM ionic strength solution – see concentrations of RMB in tables 1 and 2) ,was added. This 10-mL solution is the concentrated *E. coli* stock solution.

Table 1: These tables provide the concentrations of the constituents of the RMB solutions used. To simulate groundwater conditions, the 10 mM IS RMB was used, typically diluted from the concentrated 228.4 mM solution. ($M_1V_1=M_2V_2$) Table 1: Random Motility Buffer Solution

	Concentration (g/L) IS = 228.4 mM	Concentration (g/L) IS = 10 mM
K ₂ HPO ₄	11.2	0.4949
KH ₂ PO ₄	4.8	0.212

Sterilizing the Bucket/Bottom Reservoir

First, the bucket that would serve as the bottom reservoir to the filter was given a cursory cleaning with hot tap water and occasionally some soap to scrub off some clay residue from a preceding experiment. Next, the bucket was rinsed with boiling water using a beaker that was submerged in boiling water for over 5 minutes. The inside of the bucket and bottom reservoir were then scrubbed using non-ionic detergent and a brush that had been boiling for over 5 minutes. After scrubbing the inside, the bucket was again rinsed with boiling water, this time allowing the boiling water and soapy mixture to pass through the spigot at the bottom of the reservoir. This was done to disinfect any residual *E. coli* or competing bacteria that may be lingering in the tube or spigot. Once completely rinsed out, the ceramic filter was placed on top of the bucket, and then the filter was ready for soaking/conditioning.

Conditioning the Ceramic Water Filters

There were a lot of unexpected problems and difficulties encountered when trying to condition and soak the filters in order to prepare them for testing. Originally, the standard procedure had been to initially fill up the filter with distilled water without opening the spigot until the filter was saturated. Then, the filter was to be left overnight to saturate overnight, and the next day, the spigot would be opened and the bottom reservoir would be emptied allowing the water in the filter to start passing through. Using a beaker, the water in the filter was also emptied. After this, the filter was typically ready to be tested with an influent solution with a known *E. coli* concentration.

When this procedure was performed in early June, the flow rates through the filter were found to be unacceptably low. Typically, flow rates for these filters should be between 1 and 2 liters per hour; however, on these filters, flow rates as low as 0.2 L/hour were observed. The manufacturer insisted that this may be due to some residual charcoal or other dust or dirt that may have been trapped in the pores during the firing process. They claimed that by passing water through the filter a few times, the flow

rate would eventually reach the recommended point. After obtaining this information from the manufacturers, a new conditioning procedure was adopted.

The new conditioning procedure aimed at performing two tasks: 1) removing any air that may be trapped in the pores, 2) flushing out the charcoal/dirt that may be clogging pores. First, the filters were soaked for 48-72 hours, and then they were flushed with 10-11 L of distilled, organic-free water and 4 L of 10 mM ionic strength RMB solution. Once this was completed, the filter was ready for use.

Preparing the Influent Solution

Because the objective of this study is to observe differences in percent-removal of bacteria between filters with and without Ag-NPs, a solution with a high concentration of *E. coli* was necessary. A 12-L influent solution was prepared consisting of 1 L of concentrated RMB (IS = 120 mM), 11 L of distilled water, and 6 mL of concentrated bacteria yielding a 12-L solution of influent with a concentration of 1.45×10^9 cfu/100-mL *E. coli* bacteria. Two different sets of three serial dilutions were made for the membrane filtration. These dilutions were 1/2000 followed by a 1/500 and a 1/100 dilution. Another dilution was also made: 1/2000 followed by 1/500 and 1/10.

Membrane Filtration

To quantify bacteria in the influent and effluent, a conventional membrane filtration procedure was conducted. The apparatus was sterilized in boiling water for over 5 minutes and then a paper membrane filter was carefully lowered in the apparatus using a pair of tweezers that had been soaking in Isopropanol and rinsed with DI water. Once the apparatus had cooled to a reasonable temperature the solution was passed through it and the filter was removed carefully and placed in a Petri dish with a growth medium and incubated for 22-24 hours. After 24 hours, the coliforms were counted and the concentration of the solution could be determined and adjusted if it had been diluted.

Simulating Use

Once ready, the influent solution containing *E. coli* was passed through the ceramic filter and for 12 to 18 hours (until it had almost completely passed through the filter) . Two 100-mL samples were taken and quantified using membrane filtration.

Preliminary Results/Discussion

Figure 2 presents the results of this experiment. The data show a 7-8 log reduction in the *E. coli* concentration of the filter effluent relative to the filter influent. The effluent of the filter without Ag-NPs had an *E. coli* concentration of 379 cfu/100-mL while the effluent of the filter with Ag-NPs (applied before firing) had an *E. coli* concentration of 437 cfu/100-mL . For two repetitions of the experiment conducted at the end of the summer, (summarized in table 2), we saw approximately 99% removal of bacteria as well. Influent and effluent turbidity data for these experiments is presented in Figure 3. Both filters lowered the turbidity to amounts acceptable by World Health Organization standards (<1 NTU).

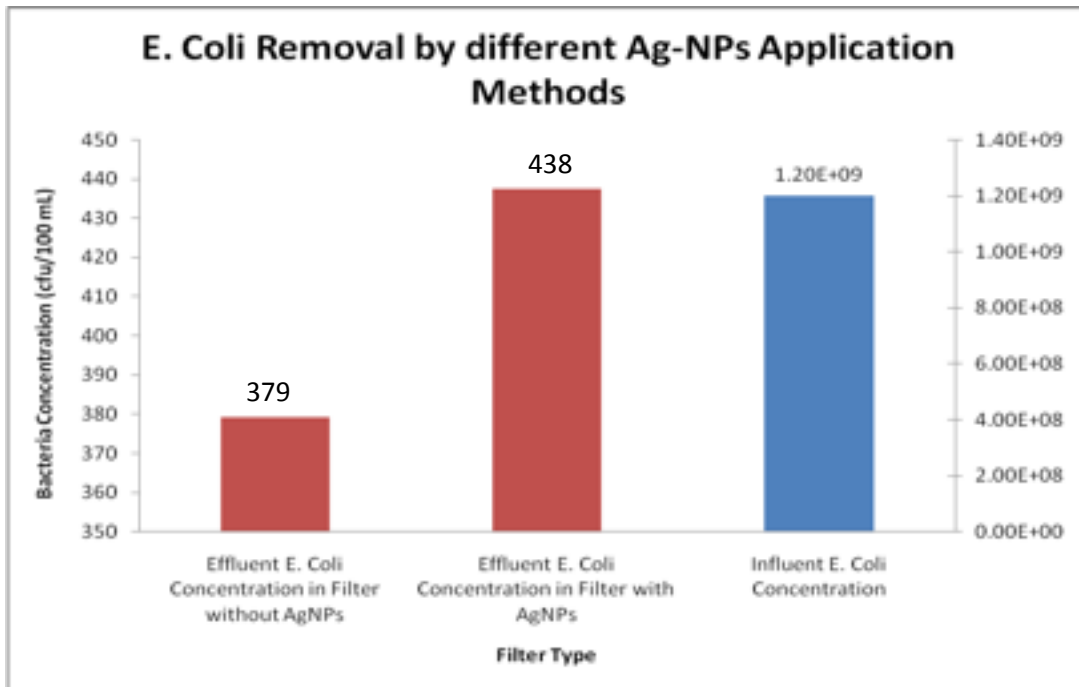


Figure 2: Influent and effluent E. coli concentrations for ceramic water filters without Ag-NPs and with Ag-NPs incorporated into the filter prior to firing. The concentration scale on the right-hand side of the graph applies to the influent concentration data.

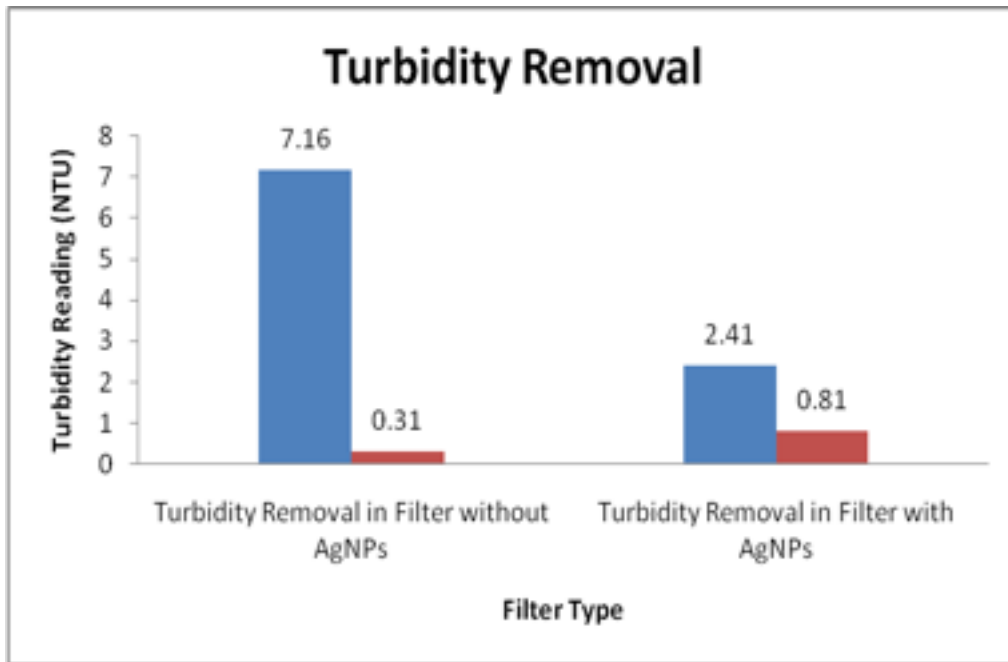


Figure 3: Comparison of influent and effluent turbidities for ceramic water filters with and without Ag-NPs.

	1		2		3	
	Influent (cfu/100 mL)	Effluent (cfu/100 mL)	Influent (cfu/100 mL)	Effluent (cfu/100 mL)	Influent (cfu/100 mL)	Effluent (cfu/100 mL)
Ag-NPs	3.4×10^7	1070	7.5×10^7	13.0	Contaminated	
Non Ag-NPs	3.4×10^7	19.1	7.5×10^7	174		

Table 2: Showing results from two repetitions of the previous work conducted towards the end of the summer with the measured influent and effluent concentrations.

While this is a promising and encouraging result for filter manufacturers, more work and study is still necessary in order to gain a more complete understanding of how the processing of Ag-NPs affects the long-term performance of these ceramic water filters. Filters with Ag-NPs painted on them still need to be tested, and some longer-term tests that go on for longer periods of time to simulate continued use are also critical. Even though both the filters with and without the Ag-NPs provide 7 to 8-log reductions in *E. coli*, that does not mean that the silver is unnecessary. Ag-NPs in the filter may still prevent bacteria growth within the filter and thereby increase its lifetime performance. Conducting breakthrough *E. coli* transport experiments using ceramic disks will also help provide a sampling of data over time which will

also inform decisions regarding the design variables mentioned before. Ultimately, it is the hope of the authors that these findings will one day help improve human health around the world.

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