

Antibacterial silver-diatomite nanocomposite ceramic with low silver release

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ABSTRACT

There is an urgent need for an effective and long-lasting ceramic filter for point-of-use water treatment. In this study, silver-diatomite nanocomposite ceramic filters were developed by an easy and effective method. The ceramic filters have a three-dimensional interconnected pore structure and porosity of 50.85%. Characterizations of the silver-diatomite nanocomposite ceramic filters were performed using scanning electron microscopy, transmission electron microscopy, X-ray diffraction, and X-ray photoelectron spectroscopy. Silver nanoparticles were confirmed to be formed *in situ* in the ceramic filter. The highest silver concentration in water was 0.24 µg/L and 2.1 µg/L in short- and long-term experiments, indicating very low silver-release properties of silver-diatomite nanocomposite ceramic filter. The nanocomposite ceramics show strong bactericidal activity. When contact time with *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*) of 10⁵ colony forming units (CFU)/mL exceeded 3 h, the bactericidal rates of the four different silver content ceramics against *E. coli* and *S. aureus* were all 100%. Strong bactericidal effect against *E. coli* with initial concentration of 10⁹ CFU/mL were also observed in ceramic newly obtained and ceramic immersed in water for 270 days, demonstrating its high stability. The silver-diatomite nanocomposite ceramic filters could be a promising candidate for point-of-use water treatment.

Key words | antibacterial effect, *in situ*, silver nanoparticles, silver release, water treatment

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INTRODUCTION

Homeowners are increasingly concerned about the presence of contaminants in their water supply that may affect health. The presence of bacteria is one main indication of water contamination (Lv *et al.* 2009). Untreated surface water or groundwater is often contaminated with pathogenic organisms. If not, it can become contaminated during transport and storage. Even water treated with a disinfectant often becomes contaminated when collected from a public standpipe and stored at home prior to consumption (Van *et al.* 2017).

Silver compounds are used extensively because of their medicinal and disinfectant properties exhibited even in

very small concentrations (Lalueza *et al.* 2011; Mirzajani *et al.* 2011; Islam *et al.* 2013). It has been demonstrated that silver ions bind to thiol groups (–SH) (Lalley *et al.* 2014) and produce reactive oxygen species by proxy (Park *et al.* 2009), prevent the replication of DNA, and affect the permeability and structure of the cell membrane (Choi & Hu 2008). It has been shown that the silver nanoparticles embedded in the ceramic porous media improve the removal and inactivation of *E. coli* cells in drinking water relative to filters without silver (Kallman *et al.* 2010; Dankovich & Gray 2011). Silver-impregnated ceramic filters were also used to provide point-of-use removal of *Cryptosporidium parvum* oocysts

from water (Abebe *et al.* 2015). Many researchers have prepared different filters containing silver for water treatment (Shen *et al.* 2011; Laan *et al.* 2014).

Both silver salts and nanoparticles are added to ceramic water filters in three different ways, i.e. by painting them onto the filter, by dipping the filter in a silver solution, and by mixing the silver with clay and water in a powder form (Oyanedel-Craver *et al.* 2014). According to a previous study (Ren & Smith 2013), for ceramic filters fabricated with silver nanoparticles by the paint-on and dipping methods (where the silver nanoparticles are applied to the filters after firing), significant release of nanoparticles into the filter disk effluent was observed relative to the fire-in method (where the nanoparticles are combined with clay and water before firing). This minimum release of silver nanoparticles from the prepared filters feature is important regarding their long-term usage for water purification. In addition, minimum release of silver nanoparticles is considered as a key environmental aspect, minimizing the risk associated with the ecotoxicity of silver nanoparticles released into the environment (Panáček *et al.* 2013).

Diatomite is one of the most commonly used filter aids in food industry because of its suitable properties such as high porosity, low density, and large surface area, which can be impregnated with silver allowing it to be used for water filtration (Panáček *et al.* 2013; Xia *et al.* 2017).

In this study, we report an easy and effective method for the preparation of silver-diatomite nanocomposite ceramic filter. The diatomite was impregnated with silver before firing, assuring the *in situ* formation of silver nanoparticles in ceramic. In order to compare the amount of silver release from ceramic filter, four different silver application methods were used: the conventional methods of painting a silver nitrate solution onto the filter (painting), dipping the filter into a silver nitrate (AgNO_3) solution (dipping), dipping the filter into a silver nitrate solution followed by calcination (dipping-fire), and a new preparation method of silver-diatomite nanocomposite ceramic filter (*in situ*). The amount of silver released by the ceramic filters was systematically investigated. Antimicrobial activity of ceramic filters containing silver was confirmed by using *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*) cells as indicators.

MATERIAL AND METHODS

Materials and preparation

Diatomite-based ceramic filters were fabricated in the shape of disks. Briefly, an amount of 20 g of 100-US mesh diatomite was immersed in 100 mL of a silver nitrate solution at concentrations of 0.05, 0.1, 0.15 and 0.2 mol/L. Then they were stirred for 2 h by magnetic stirrer, followed by vacuum filtration for 1 h. The diatomite containing silver was centrifugally separated and air-dried at 50 °C for 6 h in this study. The 15 g diatomite containing silver was mixed homogeneously with 3 g of 100-mesh pottery clay. Then the mixtures were combined with 2 mL 2% carboxymethyl cellulose solution (CMC) and again mixed until homogenous. The wet mixtures were transferred into a cylindrical steel mold with diameter of 50.0 mm and compressed for 30 s at 10 MPa. The pressed mixtures were then removed from the mold and air-dried for 1 day before they were calcined in a furnace. The resulting cylindrical-ceramic disks produced (*in situ* method) were 50.0 mm in diameter and 5.0 mm in thick with a mass of 17 ± 0.5 g. The ceramic filters without silver were prepared as control samples.

For the painting method, the ceramic filters were painted with a 0.1 mol/L silver nitrate solution on the inside and outside of the filters as it is commonly done. For the dipping method, the ceramic filters were immersed in a 0.1 mol/L silver nitrate solution, followed by vacuum infiltration for 2 h. Then the ceramic filters were air-dried for 24 h and oven-dried at 80 °C for 48 h. The dried ceramic filters were calcined in a furnace at 400 °C for 1 h in order to obtain the dipping-fire method specimens.

Filter characterization

The pore size distribution, average pore diameter, and porosity of silver-diatomite nanocomposite ceramic were measured with an automatic mercury intrusion porosimeter (AutoPore IV 9500). The morphology and microstructure of the specimens were observed with a field-emission scanning electron microscope (FESEM, FEI MLA650F). The elemental analysis of specimens was performed with energy

dispersive X-ray spectroscopy (EDS, Bruker). The formation of silver nanoparticles in the ceramic filters was analyzed with transmission electron microscopy (TEM, FEI Tecnai G2-20). The presence of silver was chemically verified using EDS elemental analysis in TEM mode. Silver content in the specimens was determined with an atomic-absorption spectrophotometer (AAS, PE FinAAcle 900F). The surface component and element quantivalence of the specimens were studied by using X-ray photoelectron spectroscopy (XPS). The XPS spectra were obtained with a VG MultiLab 2000 instrument using monochromatic AlK α .

Silver-release experiments

In order to test the binding strength between silver and the ceramic, 1.0 g of ceramic prepared by different silver application methods was immersed in 500 ml deionized water followed by ultrasonic treatment for 30 min at room temperature (25 °C). Then the amount of silver released from ceramic filter was determined.

In order to imitate the practical situation of ceramic filter, the silver release of ceramic filters with silver nitrate concentration of 0.1 mol/L was tested in short- and long-term experiments. In a short-term experiment, 1 g of ceramic filter prepared by dipping-fire method and *in situ* method was immersed in 500 mL deionized water followed by shaking at 50 revolutions per min at 25 °C. The tested shaking times were 15, 30, 60, 120, 180, 240, and 300 min, respectively. In a long-term experiment, 1 g of silver-diatomite nanocomposite ceramic was immersed in a plastic bottle filled with 500 mL deionized water for 30, 60, 90, 120, 150, 180, and 270 days at 25 °C. These bottles were shaken at 50 revolutions per min for 30 min every week. Then the quantity of silver released from ceramic filter was determined by inductively coupled plasma–mass spectroscopy (ICP–MS, Agilent 8800).

Microbiological experimentation

The bactericidal effect of ceramic filters with different silver nitrate concentrations was tested against *E. coli* (CMCC(B) 44102) and *S. aureus* (CMCC(B)26003).

Bactericidal immersion testing was studied by the spread plate method. For bactericidal immersion testing,

0.5 g of prepared ceramic filters were soaked individually in 10 mL of phosphate buffer solution (0.1 M, pH = 7.4, 25 °C), containing approximately 10⁵ CFU/mL of *E. coli* or *S. aureus* cells separately. Aliquots of 100 μ L of shaken ceramic-tablets solutions were inoculated into petri plates, spread with medium and dispersed evenly on their surface by using a coated stick. After inoculation, all plates were incubated at 37 °C for 24 h. All bactericidal tests were repeated three times. The colony formation was examined and photographed.

The bactericidal activity of a freshly obtained ceramic sample and a sample that was immersed in water for 270 days were tested against *E. coli* cells. For quantitative determination of the time-dependent lethal-effect of silver-diatomite nanocomposite ceramic against bacteria, cell suspension of *E. coli* with a concentration of 10⁹ CFU/mL was used. In this experiment 0.5 g tested-bulk ceramic samples were added into 10 mL of the *E. coli* suspension with a concentration of 10⁹ CFU/mL. Then 100- μ L were taken after 0.5, 1, 2, 3 and 6 h of contact time and plated using ten-fold dilutions on agar plates and incubated at 37 °C for 24 h until colonies could be enumerated. The experiments were carried out in triplicate. The bactericidal rates of tested ceramic samples were calculated from the equation:

$$X = \frac{A - B}{A} \times 100\% \quad (1)$$

where X is the bactericidal rate of samples, A is the average count of bacterial of control group without bactericide, and B is the average count of bacterial colonies of samples tested.

RESULTS AND DISCUSSION

Microstructure and morphology analysis

Scanning electron microscopy (SEM) images and an EDS spectrum of a silver-diatomite nanocomposite ceramic filter are shown in Figure 1. Surface and cross-section micrographs of ceramic filters show that macropores were formed after sintering at 1,000 °C. The micrograph depicted in

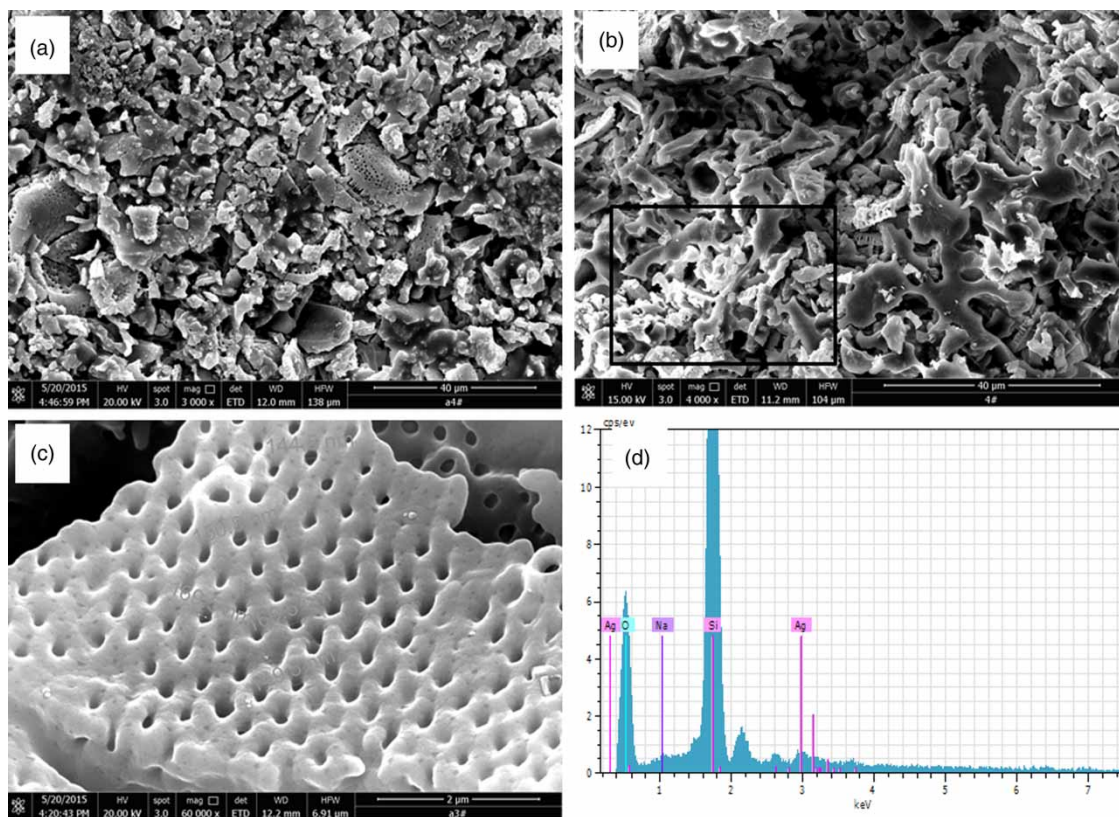


Figure 1 | SEM images and EDS spectrum of silver-diatomite nanocomposite ceramic filter, (a) surface photograph, (b) cross-section photograph, (c) micropores of diatomite, (d) EDS spectrum.

Figure 1(c) shows that the diatomite frustule was present in the ceramic in a disk-shape form and presented regularly spaced rows of fine pores within the disk walls. These pores were well-interconnected and formed a three-dimensional pore structure, which provides channels for water filtration. Meanwhile, the diatomite particles maintained their original micropores, which have a pore size in the range of 170–200 nm. These micropores are known to provide an excellent performance of adsorption and filtration.

EDS was performed on the surface marked with a black pane in Figure 1(b) and the results are shown in Figure 1(d). The surface of the silver-diatomite nanocomposite ceramic is mainly composed of silicon and oxygen, corresponding to the chemical components of diatomite, and silver as detected in the EDS spectrum, proving that silver had been deposited onto silver-diatomite nanocomposite ceramic.

TEM images and EDS spectrum of silver-diatomite nanocomposite ceramic are shown in Figure 2. Many

silver nanoparticles are observed through TEM as shown in Figure 2(a) and 2(b). The particle size of visible silver nanoparticles was in the range of 5–20 nm and their shape was confirmed as spherical according to TEM images. The arrows in Figure 2(b) indicate the silver nanoparticles formed *in situ* in diatomite-based ceramic filter. EDS analysis in TEM mode was performed on the substance marked with a circle in Figure 2(c) and the results are shown in Figure 2(d). The weight percentage of detected elements such as silicon, oxide, and silver was 44.5%, 36.7%, and 11.2%, respectively. Diatomite is known to adsorb silver, and therefore hydroxyl groups might bind the silver ions available from silver aqueous solutions. A calcination process could convert the silver ions into silver nanoparticles and the hydroxyl groups would function as stabilizing groups for the silver nanoparticles (Xia *et al.* 2017). From the SEM and TEM images, as well as from the corresponding EDS spectra, it is evident that silver had been impregnated on the surface of the ceramic filter and that

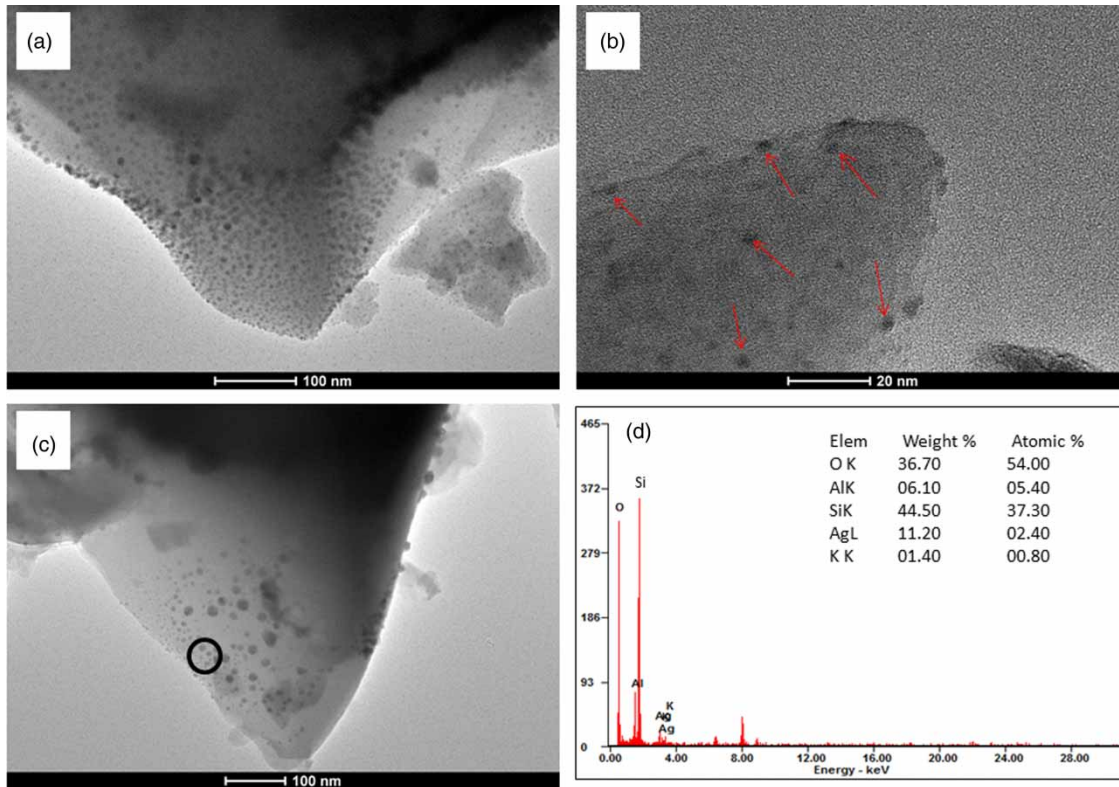


Figure 2 | TEM images (a) and (b) and EDS analysis (c) and (d) of silver-diatomite nanocomposite ceramic filter.

silver nanoparticles had been formed *in situ* within the filter structure.

The pore size distribution of the silver-diatomite nanocomposite ceramic was measured by the mercury intrusion method and the results are presented in Figure 3. The pore size of the ceramic filter was in the range of 1–3 μm and most of the pores were around 2 μm in size. The sample has a porosity of 50.85% as well as a median pore diameter and average pore diameter of 2.169 μm and 0.984 μm , respectively. The mercury intrusion results indicated that there were plenty of micropores in the ceramic, which was in accordance with SEM images.

X-ray diffraction (XRD) and XPS analysis

In order to further confirm the form of silver nanoparticles in the prepared silver-diatomite nanocomposite ceramic, XRD and XPS analyses were performed. Figure 4(a) shows the representative diffraction patterns of the silver-diatomite ceramic composites with different silver nitrate concentrations.

It is clear that the main phases are quartz (JCPDS, 46-1045) and cristobalite (JCPDS, 39-1425) for all ceramics. Moreover, the diffraction-peak positions for quartz and cristobalite of ceramic with silver does not shift significantly in comparison with the ceramic without silver, suggesting that the

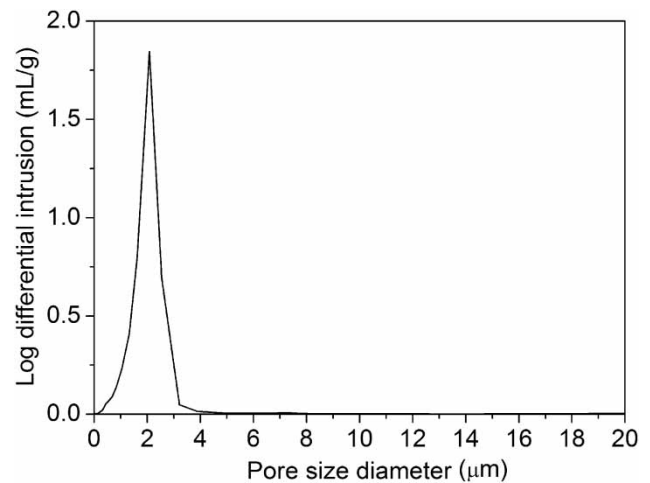


Figure 3 | Mercury intrusion porosimetry of prepared silver-diatomite nanocomposite ceramic.

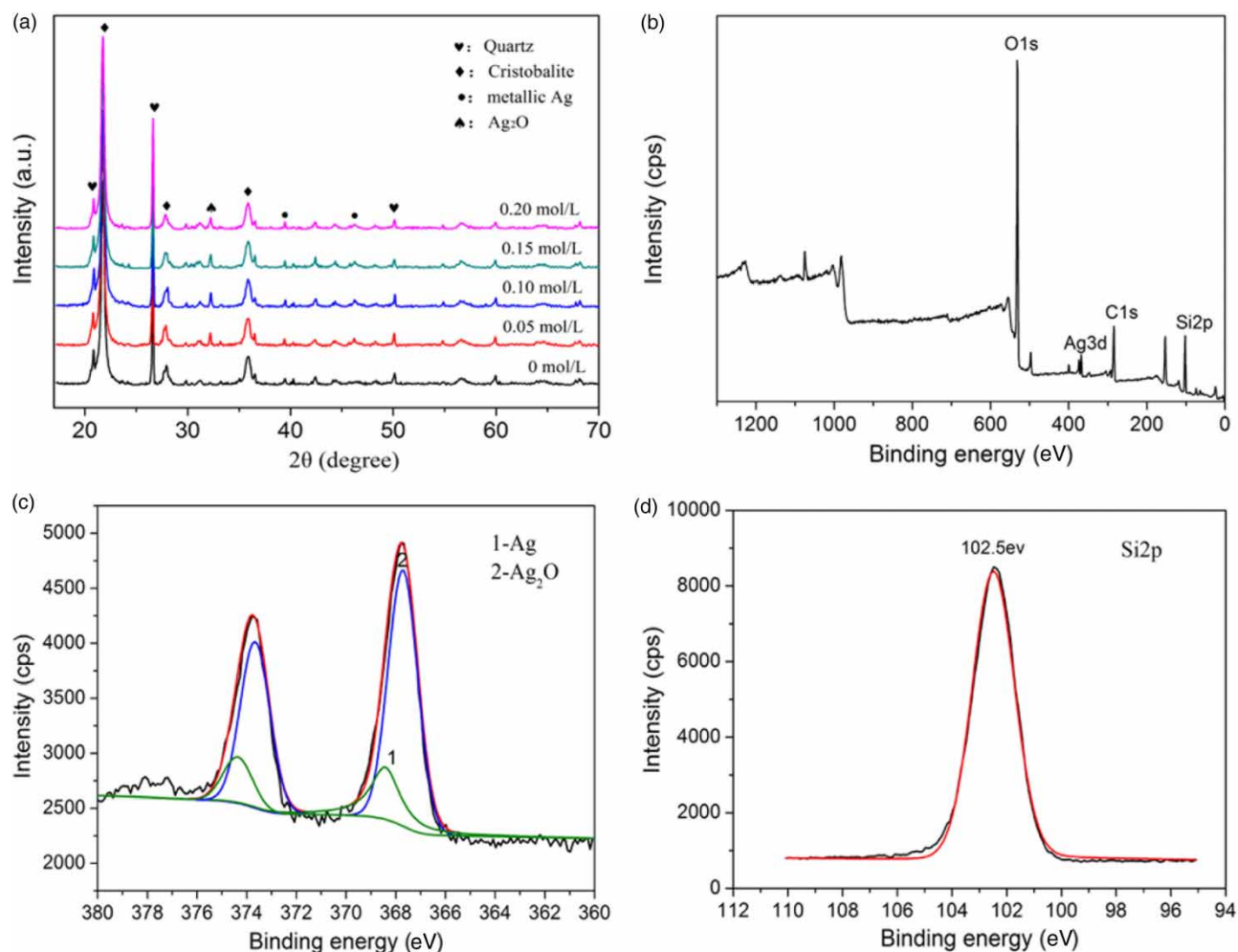


Figure 4 | XRD patterns of silver-diatomite nanocomposite ceramics prepared with (a) different silver concentrations; (b) XPS general spectrum of the prepared silver-diatomite nanocomposite ceramic; (c) narrow scan of Ag3d; and (d) Si2p.

deposition of silver did not change the phase composition or the crystal structure of diatomite. After being deposited with silver, the diffraction peaks of metallic silver and silver oxide appeared in the diffraction patterns of silver-diatomite nanocomposite ceramics. The diffraction peaks at 39.5° and 46.3° could be indexed to the (111) and (200) planes of metallic silver (JCPDS, 04-0783) (Lei *et al.* 2014) and the diffraction peak at 32.3° could be indexed to the (111) planes of silver oxide (JCPDS, 41-1104). However, the low intensity of the silver nanoparticles was probably due to the low concentration of silver in the silver-diatomite nanocomposite ceramic.

XPS measurement was performed in order to further investigate the valence of silver in the silver-diatomite

nanocomposite ceramic. Figure 4(b)–4(d) show the survey XPS scan spectra of ceramic composite and XPS narrow scan of silver. The binding energies of silicon, oxide, and carbon are distinct in the XPS spectrum and two peaks of silver were noted. As shown in Figure 4(c), the Ag3d_{5/2} and Ag3d_{3/2} are observed at binding energies of 367.8 and 373.8 eV, respectively. The high-resolution Ag3d_{5/2} spectrum could be fitted in terms of two individual component peaks indicating that silver exists in two forms. The peak at 367.8 eV is attributed to the presence of silver oxide (Hammond *et al.* 2002), while the peak at 368.4 eV is associated with metallic silver (Romand *et al.* 1978). This result is in agreement with the previous XRD analysis and confirms that silver in the silver-diatomite nanocomposite ceramic exist in

the form of silver oxide and metallic silver. Similarly, the peak at 102.5 eV in Figure 4(d) is attributed to SiO₂.

It is speculated that the silver ions adsorbed by and bound to diatomite in ceramics decompose into metallic silver during the calcination process (Ehdaie et al. 2014). The metallic silver obtained from the decomposition process presents a form of spherical nanoparticles. However, since the sintering atmosphere is air, the metallic silver nanoparticles are re-oxidized due to its high activity when the furnace temperature drops below 961 °C during the cooling process. In addition to that, when compared with the silver nanoparticles existing in the depth of diatomite pores, those absorbed on the surface have more chance to react with oxygen. As a consequence, only a portion of the metallic silver turns into silver oxide accompanied with crystal growth. This demonstrates the *in situ* preparation of the silver nanoparticles in the ceramic.

Silver release

The silver-release properties of ceramic filters at different conditions are demonstrated in Figure 5. The amount of silver released from the ceramic filters prepared by the four different methods after ultrasonic treatment are shown in Figure 5(a). For all ceramics, the same silver nitrate concentration of 0.1 mol/L was used. Total silver concentrations as released from ceramics were 1.54, 17.65, 49.27, and 1,118.13 µg/L for *in situ*, dipping-fire, dipping, and painting methods, respectively. As depicted in Figure 5(a), the *in situ* method retains most silver when compared to other methods. After undergoing ultrasonic treatment for 30 min, the silver concentration in the water was only 1.54 µg/L for the *in situ* method specimen, which was much lower than that observed in other specimens.

To further investigate the silver-release properties of ceramic filter, soaking test was also conducted for *in situ* and dipping-fire method specimen. The quantity of silver released from ceramic filter after immersion in deionized water followed by shaking for 15, 30, 60, 120, 180, 240, and 300 min are shown in Figure 5(b). The total silver concentration in the water was observed to increase with the immersion time. It can be seen that silver released from *in situ* method specimen was far below that observed for the dipping-fire method specimen. After immersion in water for 300 min, the total silver concentration in the

water was 0.24 µg/L for the *in situ* method specimen and 6.10 µg/L for the dipping-fire method specimen.

Long-term experiments were conducted in order to test the ceramic filters under real-world water-storage conditions. The silver released from *in situ* method specimens in a long-term experiment are shown in Figure 5(c). With the increase of immersion time from 30 to 60 days, the quantity of silver released from silver-diatomite ceramic nanocomposite increased rapidly at first, and then decreased in the later immersion time from 60 to 120 days. When the immersion time was increased to 150 days, the total silver concentration in the water was increased again up to 2.1 µg/L. With the immersion time further extended to 270 days, the total silver concentration was reduced to 1.5 µg/L. As observed in Figure 5(c) for long-term experiments, the total silver concentration in water did not increase continuously, but behaved erratically. This behavior may be due to the dynamic release and adsorption of silver from and into the ceramic filter. At short immersion times, silver released from the ceramic continuously led to an increase of the silver concentration in water. As the contact time was increased, some silver was re-adsorbed by the ceramic due to its microporous structure, and the silver concentration in water slightly decreased. With the extended immersion time, the silver adsorbed by the ceramic returned to water thereby increasing the silver concentration in water once again.

More importantly, the silver-diatomite nanocomposite ceramic prepared in this study displayed very low silver release in short- and long-term experiments. In short- and long-term experiments, the highest silver concentrations in water were found to be 0.24 µg/L and 2.1 µg/L, respectively. The maximum concentration of silver allowed in water limit is 0.1 mg/L (100 µg/L) according to the World Health Organization (WHO) and United States Environmental Protection Agency (US EPA) drinking-water standard (WHO 2011; US EPA 2011). After immersion in water for 300 min, the total silver concentration in water from the silver-diatomite nanocomposite ceramic was about 1/400 times the maximum concentration allowed for silver in drinking water. When silver-diatomite nanocomposite ceramic was soaked in water for 270 days, the highest silver concentration in water was about 1/50 value of drinking-water standard. This demonstrates that the *in situ* preparation of silver nanoparticles in ceramic filter improves silver retention and longevity of the water purifier.

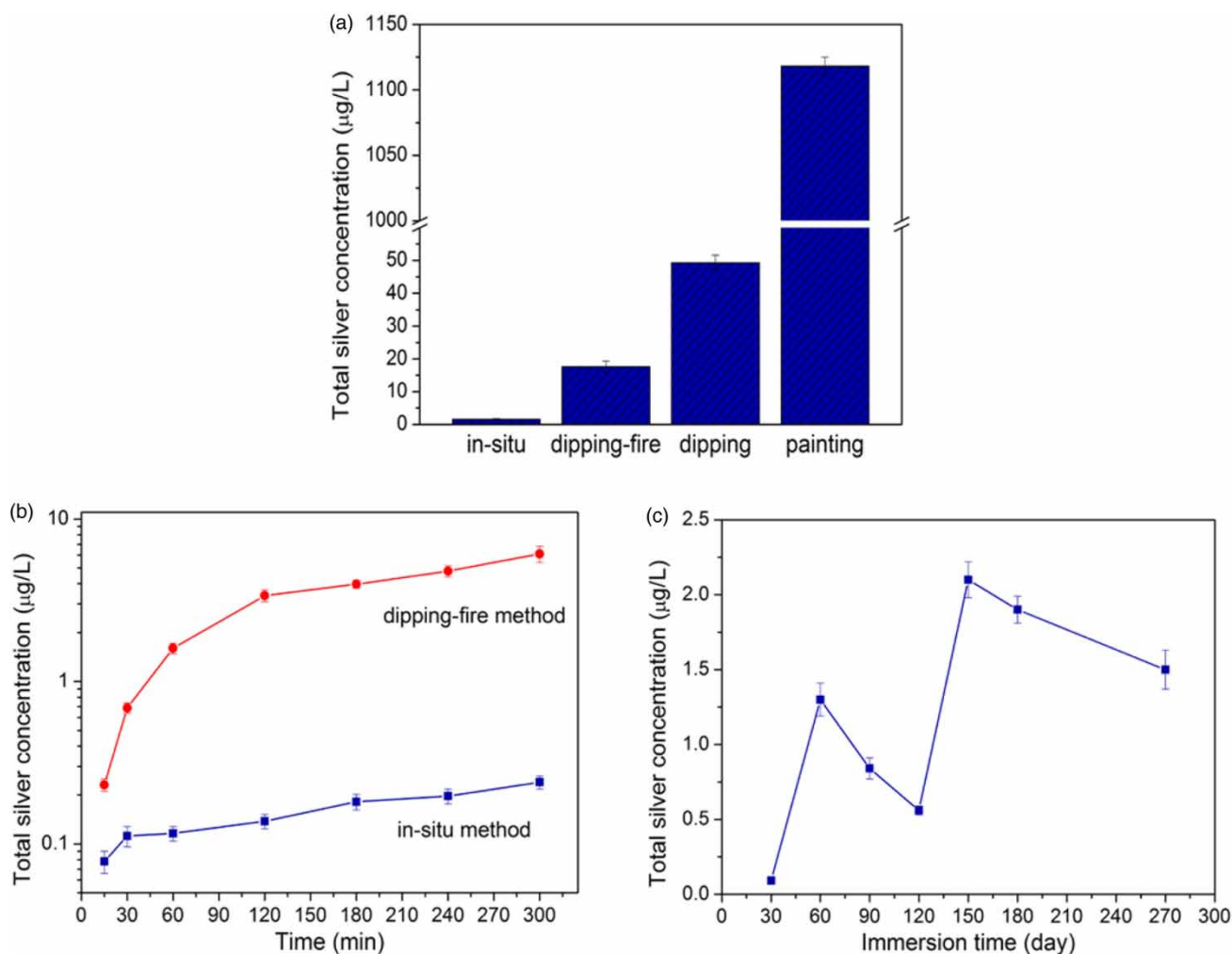


Figure 5 | (a) Amount of silver released from ceramic filters prepared by four different methods after ultrasonic treatment; (b) total silver concentration as a function of time for ceramic filters prepared by *in situ* and dipping-fire methods in short-term experiments; and (c) total silver concentration as a function of immersion time for silver-diatomite nanocomposite ceramic in long-term experiments.

This may contribute to the stable binding between nanosilver and diatomite. This minimum release of silver from the prepared ceramic filters feature is of great importance with respect to a long-term applicability of these filters while minimizing the potential risk associated with the ecotoxicity of silver (Mittelman *et al.* 2015; Praveena & Aris 2015).

Antibacterial efficiency

The bactericidal rate of ceramic filters with different silver nitrate concentration (0.05, 0.1, 0.15, and 0.2 mol/L) against *E. coli* and *S. aureus* are illustrated in Table 1. At 1 h, the bactericidal rates of the four different silver content

ceramics against *E. coli* and *S. aureus* all surpass 99%. This may be due to the amount of silver, silver-particle size, and distribution in the ceramic filter. As the contact time was extended to 3 h, the bactericidal rate was increased up to 100%, indicating that all *E. coli* and *S. aureus* cells were killed. When the contact time was further prolonged to 6, 12, and 24 h, the bactericidal rates for both *E. coli* and *S. aureus* remained at 100%. These results indicated that the prepared silver-diatomite nanocomposite ceramic have a good bactericidal activity.

Photographs of the antibacterial tests and a schematic of the proposed inactivation mechanisms of silver-diatomite nanocomposites are illustrated in Figure 6. Figure 6(a)–(f)

Table 1 | Bactericidal rate of ceramic filters with different AgNO₃ concentrations (0.05, 0.10, 0.15, and 0.20 mol/L) against *E. coli* and *S. aureus* cells

Time (h)	<i>E. coli</i>				<i>S. aureus</i>			
	0.05 mol/L	0.10 mol/L	0.15 mol/L	0.20 mol/L	0.05 mol/L	0.10 mol/L	0.15 mol/L	0.20 mol/L
1	99.91%	99.98%	99.79%	99.95%	99.92%	99.94%	99.00%	99.52%
3	100%	100%	100%	100%	100%	100%	100%	100%
6	100%	100%	100%	100%	100%	100%	100%	100%
12	100%	100%	100%	100%	100%	100%	100%	100%
24	100%	100%	100%	100%	100%	100%	100%	100%

show the viability assessment results obtained with the silver-diatomite nanocomposite ceramics against *E. coli* and *S. aureus*. The control-treated water shows a substantial growth of *E. coli* and *S. aureus* on the nutrient substrate (Figure 6(a) and 6(d)). After a contact time of 1 h with the silver-diatomite nanocomposite ceramic, the viable cells observed in the treated water were decreased dramatically (Figure 6(b) and 6(e)). When the contact time was further extended to 3 h, there were no viable cells detected in the treated water (Figure 6(c) and (f)).

Figure 6(g) shows the bactericidal effects of ceramic specimens freshly obtained and specimens immersed in water for 270 days against *E. coli* with an initial concentration of 10⁹ CFU/mL. The bactericidal rate against *E. coli* was 92.6% after 0.5 h followed by a gradual increase up to 99.8% after 6 h in the case of freshly obtained ceramic samples. In the case of the immersed specimen, the bactericidal rate against *E. coli* increased from 99.3% to 99.7% during the initial 3 h of contact time followed by a drop to 96.3% after 6 h. Nonetheless, both samples showed a strong bactericidal effect against *E. coli*. It is likely that while immersed in the water, the nanosilver or silver ion kept releasing from the ceramic sample until a dynamic balance of release-absorption of silver on the ceramic surface. As a result, the immersed sample lost mass of silver and a certain ratio of silver accumulated in the outer surface. Once added to the *E. coli* solution, the nanosilver or silver ion releases rapidly from the outer surface of immersed sample to liquid medium and easily restrained bacterial growth and inactivated *E. coli* during the first 3 h. The mass of silver accumulated in the outer surface of immersed sample is limited, while the process of inactivation and growth inhibition would trigger a consumption of silver. With that comes a shortage of silver to continue antibacterial activation and

E. coli cells multiplied after 6 h, whereas the samples immersed in water for 270 days still had a strong antibacterial effect against *E. coli*. The long-term preservation of the antibacterial properties of ceramics may be due to an uniform distribution and minimum release of silver from the prepared silver-diatomite nanocomposite ceramic filter.

Based on the above results, the antimicrobial mechanism of the silver-diatomite nanocomposite is discussed and is shown in Figure 6(h). First, cells are directly killed by the nanosilver and silver ions released from the nanocomposite ceramic, and the amount of silver released increased with increasing contact time. Nanosilver and silver ions might adhere to the negatively charged cell wall, changing the cell wall permeability. This action, coupled with protein denaturation, might induce cell lysis and death (bactericidal effect) (Samuel & Guggenbichler 2004). Second, bacteria cells contaminated with nanosilver and silver ions cannot grow into colonies on the culture medium because nanosilver and silver ions inhibit the replicative capacity of the cell and affect the growth of bacteria in a culture environment (bacteriostatic effect). Thus, the inactivation of bacteria might result from both the bactericidal and bacteriostatic effects produced by nanosilver and silver ions released from the silver-diatomite nanocomposite ceramic (Lv et al. 2009).

CONCLUSION

In summary, silver-diatomite nanocomposite ceramic filters were successfully prepared by an *in situ* method. Silver nanoparticles were formed *in situ* in the ceramic filter and silver exists in the form of silver oxide and metallic silver. More importantly, the silver-diatomite nanocomposite ceramic prepared in this study displayed very low silver release in

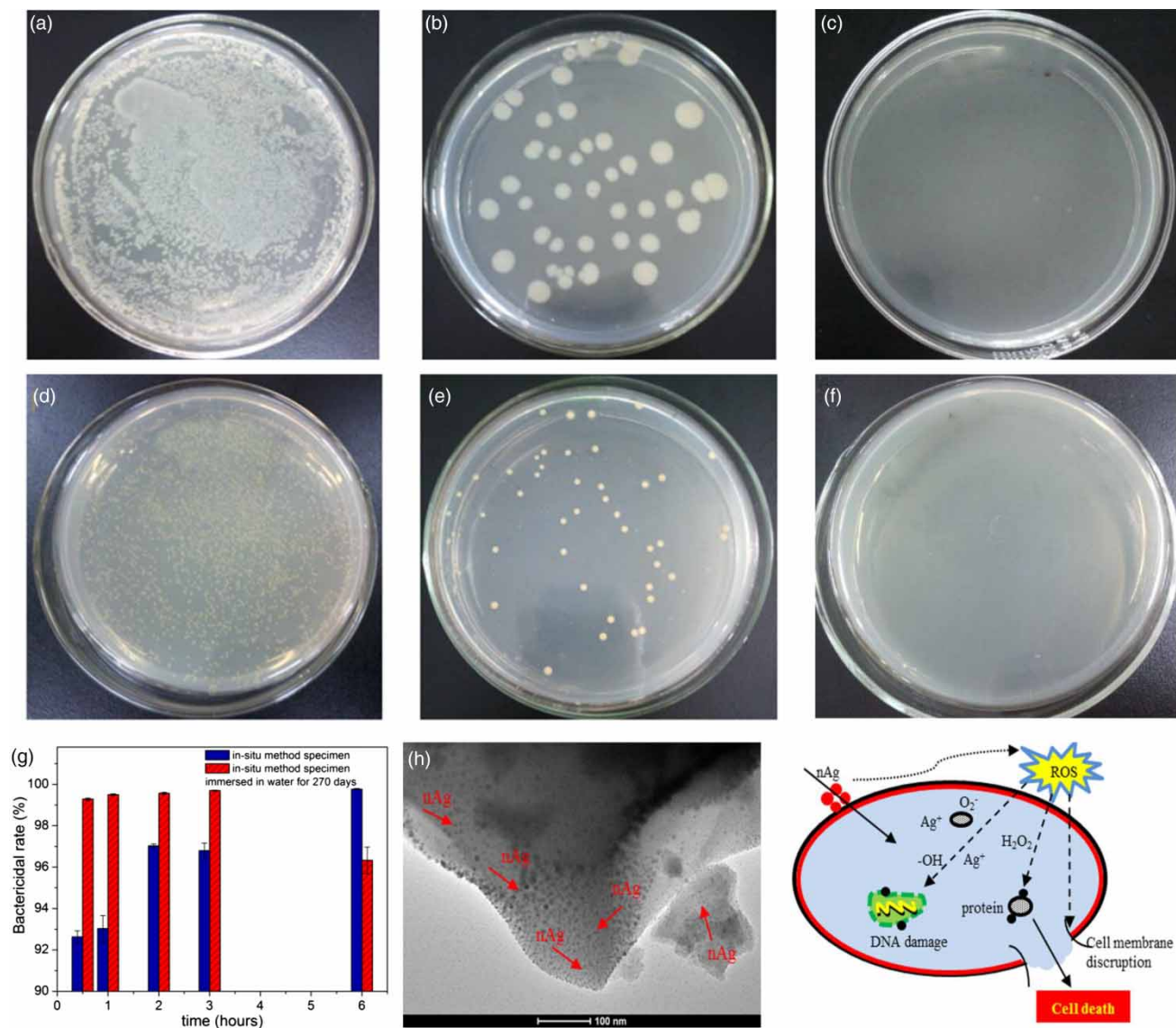


Figure 6 | Photographs of antimicrobial test results of silver-diatomite nanocomposite ceramic against *E. coli* and *S. aureus*. (a) Control sample, *E. coli*, 3 h; (b) 0.10 mol/L, *E. coli*, 1 h; (c) 0.10 mol/L, *E. coli*, 3 h; (d) control sample, *S. aureus*, 3 h; (e) 0.10 mol/L, *S. aureus*; (f) 0.10 mol/L, *S. aureus*, 3 h; (g) bactericidal rate of ceramic specimens newly obtained and specimens immersed in water for 270 days against *E. coli* with initial concentration of 10^9 CFU/mL; and (h) proposed antimicrobial mechanisms of silver-diatomite nanocomposite.

short- and long-term experiments. The ceramics showed strong bactericidal effect against *E. coli* and *S. aureus*. Moreover, ceramic filters immersed in water for 270 days still had a strong antibacterial effect. Therefore, the silver-diatomite nanocomposite ceramic filters could be a promising candidate for its practical applications in point-of-use water treatment due to its high antibacterial ability and stability.

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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