

Effect Of Mangrove-Derived Nanoparticles On Water Disinfection

Asmathunisha. N¹ & Kathiresan. K²

^{1,2}CAS In Marine Biology, Annamalai University, Parangipettai, India

Abstract: Biological oriented nano-disinfection process is a rapidly expanding area of research. The present study investigated the effect of six different nanoparticles (silver, gold, zinc, copper, magnesium and calcium) in water disinfection process by polyurethane foam filtration. These nanoparticles were synthesized by a mangrove species *Xylocarpus mekongensis* Pierre. The nano-coated foam had significantly shown better antimicrobial filtration properties than the untreated foam filter. Nano treated filtered water showed the bacterial load in following order: calcium > magnesium > copper > zinc > gold > silver. Although the nano foam filter had large pores, the nano coated with polyurethane foam showed excellent antimicrobial performance within a very short contact time, indicating that it can be used as an alternative to chemical treatment in water sanitation and wastewater treatment applications.

1. Introduction

Water is the elixir of life. The deterioration of water quality is mainly indicated by the water-borne diseases. Microbial contamination is one of the greatest water-borne risks to human health, leading to disease outbreaks [1]. About 1.8 million People in the world die annually from diarrheal diseases (including cholera) of reported by WHO [2]. There is high escalating demands and pollution of the limited water sources, particularly in rural and developing communities [3]. The risks of waterborne diseases are prevented or reduced by water disinfection processes. Many scientific processes such as chlorination [4], ultraviolet light [5,6] reverse osmosis and use of silver catalyst [7] are well developed. Though, these disinfection processes also raise health, economic or technical issues. For instance, chemical disinfectants such as free chlorine, chloramines and ozone can react with various constituents in natural water to form disinfection byproducts which are harmful to health. The resistance of pathogens such as *Cryptosporidium* and *Giardia*, to conventional chemical disinfection needs large dosage of chemicals [8]. Methods like UV purification and reverse osmosis are not cost effective [9]. Because of the limitations of the conventional methods there is an urgent need to explore other innovative alternatives such as the use

of nanotechnology. Nanotechnology is considered as a new generation of technology [10]. And it can be cost effective and for water treatment systems [11]. The use of metal nanoparticles for water disinfection is being explored. Oligodynamic nanoparticles based disinfection includes the use of metals such as silver, gold, zinc, tin and copper due to their antimicrobial properties. Besides their oligodynamic nature, they also possess catalytic properties [12]. Still silver is recognized as a biocide, its application in purification of water is increasing. In nanoparticle form, silver becomes more reactive, has increased catalytic properties, large surface area to volume ratio, unusual crystal morphologies and thus toxic than silver ion [13,14]. To effectively apply silver nanoparticles in water disinfection, they should be impregnated in a substrate. Several studies have shown the use of silver nanoparticles coated on various substrates. For instance, silver nanoparticles are incorporated in materials such as montmorillonites [15], polysulfone [16] cellulose acetate [17], fibreglass [18], polyurethane foams [19], ceramic filters [20, 21], activated carbon [22]. In addition, nanoparticles loaded on some of the above substrates/media showed poor disinfection of water and are not structurally stable for use in water treatment systems. Therefore the present investigation was made here on the effect of silver nanoparticles incorporated filter system on water disinfection.

2. Materials and Methods

2.1. Collection of Samples

The polyurethane foams were purchased from local supermarket, in the present study, normal tap water was used for the treatment.

2.2. Synthesis of Nanoparticles

Mangrove extract of *Xylocarpus mekongensis* were prepared by taking 20 g of thoroughly washed and finely cut leaves in a 500-mL Erlenmeyer flask with 100 ml of sterile distilled water and then boiled the mixture for 2 min before finally decanting it. For reduction of Ag, Au, Zn, Mg, Cu, Ca ions, 5 ml of each leaf broth was added to 45 ml of 5 different

aqueous solutions such as silver nitrate (1 mM), chloroauric acid (0.5 mM), zinc nitrate (1 mM), magnesium nitrate (0.1 mM), copper nitrate (0.25 mM) and calcium nitrate (0.25 mM) solutions respectively as substrates. (Fig.1). The reduction of pure Ag, Au, Zn, Mg, Cu, Ca ions was monitored by measuring the absorbance of the solution at regular intervals after diluting a small aliquot (0.2 ml) of the sample 20 times. The absorption was measured at the range between 200-500 nm at a spectrophotometer (Elico, Chennai).[23,24].

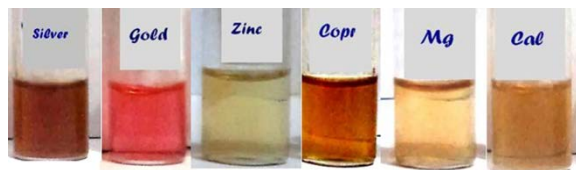
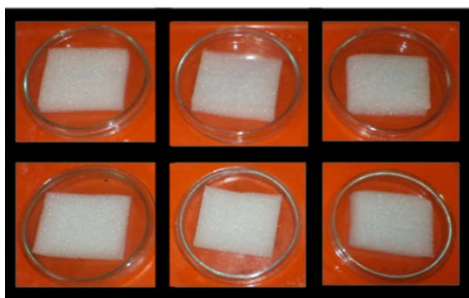


Fig. 1. Synthesized nanoparticles (A) Silver (B) Gold (C) Zinc (D) Copper (E) Magnesium (F) Calcium

2.2. Preparation of Nano-coated Foam Filters

The measurement of 10x10 cm and thickness of 6 mm Polyurethane foams were used in this experiment. Polyurethane foams were soaked in six different nanoparticles solution for overnight. For the saturated coating of foams, 250 ml of the nanoparticles solution was required. The sheets were washed repeatedly with water to remove any adsorbed ions and were air-dried. (Fig.2).

(A)



(B)

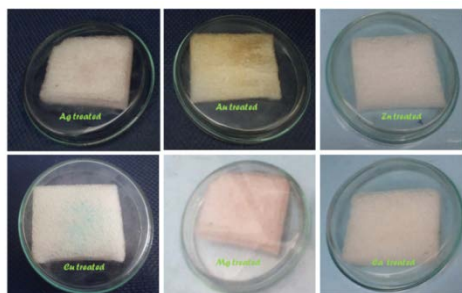


Fig. 2. (A) Foams before treatment with nanoparticles (B) Foams after treatment

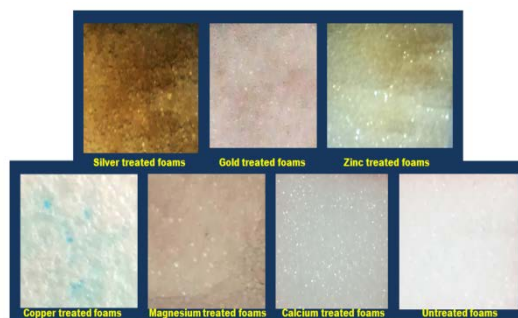


Fig. 3. Microscopical view of foams treated with nanoparticles

2.3. Filtration with Polyurethane Foams

Normal tap water was used in the filtration process. Filtration experiments were performed in a 500mL Erlen Meyer conical flasks. The polyurethane foams treated with six different nanoparticles (Silver, Gold, Zinc, Copper, Magnesium, and Calcium) were taken in a funnel. Each funnel was packed with Polyurethane foam coated with nanoparticles placed in the upper ends of each conical flask. The whole set up was tightly fixed to hold the water for proper filtration. The funnel was washed and sterilized under UV light for 30 min. The Conical flasks were autoclaved before use. The normal tap water was pumped vertically upwards through the funnel. And the filtered water was continuously collected in conical flasks. (Fig.4). The water was analysed for quality parameters as described below.



Fig. 4. Filtration with nano filter [(A) silver (B) Gold (C) Zinc (D) Copper (E) Magnesium (F) Calcium]

2.4. Assessment of Water Quality

The water quality of filtered and unfiltered water through nanofilter was assessed for colour visually, odour based on acceptability by involving five volunteers), pH (measured using a pen) salinity (by using refractometer) and microbial load by pour plate method

3. Results

The colour of polyurethane foam changed from white to brown with silver nanoparticle, white to pink with gold nanoparticle, white to straw yellow with zinc nanoparticle, white to light blue with copper nanoparticle, white to mahogany with magnesium nanoparticle, white to grey with calcium nanoparticles nanoparticle. The saturated binding of nanoparticles on polyurethane gave different colour with different nanoparticles solution as shown in fig. 2. There was less loss of nanoparticles after several (4-7 times) washing and drying operations and after keeping it for several months in a closed environment. Washing was done with distilled water for 20 min after it was air dried. Optical images of polyurethane foam coated with nanoparticles showed incorporation of nanoparticles as different colour contours in the film as shown in Fig. 3. The morphology of the film did not manifest any change as a result of nanoparticles incorporation. Odour, pH, salinity of the filtered and unfiltered water are shown in figure 5a and 5b and table 1. The bacterial load was accessed using plating technique, and it was shown in Fig. 6. The bacterial load was very high in unfiltered water compared to nanoparticles treated filter water. Nano treated filtered water showed the bacterial load in following order: calcium > magnesium > copper > zinc > gold > silver (Fig. 7).

Table 8.1. Assessment of physical parameters in treated and untreated filtered water

Nanoparticles treated with foams	Colour	Odour	pH	Salinity (ppt)
Untreatedwater	Slightly turbid	Acceptable	8.1	0.38
Silver	Clear	Acceptable	7.4	0.24
Gold	Clear	Acceptable	7.5	0.28
Zinc	Clear	Acceptable	7.8	0.31
Copper	Clear	Acceptable	7.8	0.32
Magnesium	Clear	Acceptable	7.9	0.34
Calcium	Clear	Acceptable	7.5	0.26

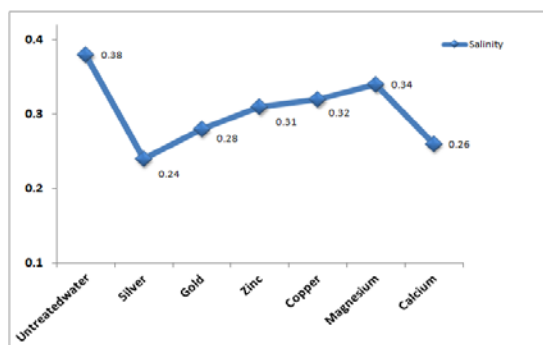
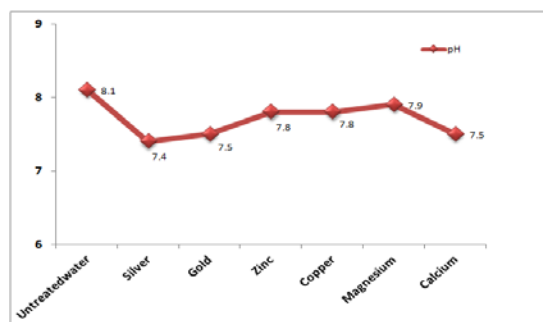


Fig. 5. (A) pH (B) salinity of the treated and untreated filtered water

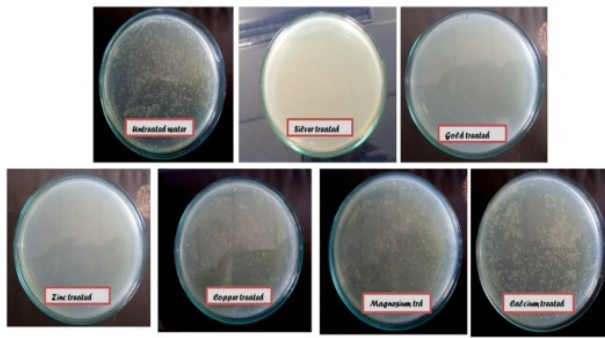


Fig. 6. Plates showing bacterial load in nano-treated and un treated filtered water

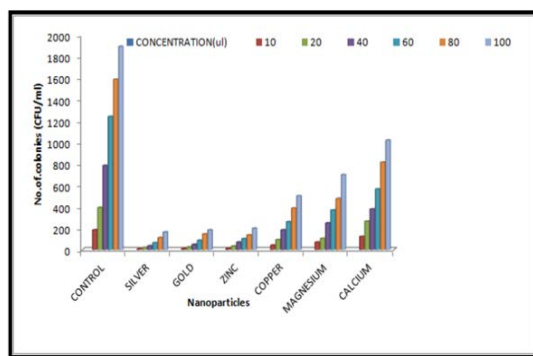


Fig. 7. Number of colonies present in the nano-treated and untreated filtered water

4. Discussion

In this study, nanoparticles were successfully deposited on foam filter for water disinfection process. Using a combination of polyurethane foam and nanoparticles, the water disinfection process was achieved. The bacterial load was very high in unfiltered water compared to nanoparticles treated filter water (Fig. 8.8). Nano treated filtered water showed the bacterial load in following order: calcium > magnesium > copper > zinc > gold > silver. Furthermore, filters coated with nanoparticles could be used for tap water purification, wastewater treatment, and bio-related applications. Nanoparticles have been extensively studied for its water disinfection properties [25] and as a antipathogenic agent [26], Similar results have been obtained by Jain and Pradeep in 2005 [19]. The nanoparticles on the filter surface have an important germicidal effect. According to Chick's report, disinfection by chlorine and its compounds requires long contact times or a high concentration of disinfectant for satisfactory germicidal efficiency [27]. In our study, although the water ran through the large pores of the foam filter coated with nano-composite film with little flow resistance, leading to a short contact time, the

increase in germicidal efficiency relative to the non coated foam filter was significant.

Silver nanoparticles on the matrix are probably ionized to silver ions during the filtration process. These silver ions can then interact with thiol groups in proteins, leading to the inactivation of proteins, including respiratory enzymes [28]. According to Feng *et al.*, (2000) [29] silver ions prevent DNA replication and affect the structure and permeability of the cell membrane. In previous research [30] the antibacterial performances of the specimens have been evaluated using the Gram-negative bacterium *E. coli*.

The nano-coated foam had significantly shown better antimicrobial filtration properties than the untreated foam filter. Although the nano foam filter had large pores, the nano coated with polyurethane foam showed excellent antimicrobial performance within a very short contact time, indicating that it can be used as an alternative to chemical treatment in water sanitation and wastewater treatment applications.

5. Acknowledgements

The authors are thankful to authorities of Annamalai University and to UGC, New Delhi for financial assistance (MANF).

6. References

- [1] World Health Organisation, Guidelines for Drinking Water Quality, third ed., USA, 2004.
- [2] World Health Organisation, Water, sanitation and hygiene links to health. Facts and figures, 2004.
- [3] Grabow, W.O.K. Waterborne diseases: update on water quality assessment and control, Water SA 22: 1996 , 193–202.
- [4] Son, H., Cho, M., Kim, J., Oh, B., Chung, H. and Yoon, J. Enhanced disinfection efficiency of mechanically mixed oxidants with free chlorine. Water Res. 39: 2005, 721–727.
- [5] Finch, G., Black, E. and Gyurek, L. Ozone and chlorine inactivation of Cryptosporidium, in: Water Quality Technology Conference, AWWA, 1994, 1303–1309.
- [6] Gujer, W. and von Gunten U. A stochastic model of an ozonation reactor. Water Res. 37: 2003, 1667–1677.
- [7] Hassinger, E., Thomas, A.D. and Paul, B.B. Reverse Osmosis Units Water Facts, 1994.
- [8] Shashikala, V., Siva Kumar, V., Padmasri, A.H., David Raju, B., Venkata Mohan, S., Nageswara Sarma, P. and

- Rama Rao K.S.. Advantages of nano-silver-carbon covered alumina catalyst prepared by electro-chemical method for drinking water purification, *J. Mol. Catal. A – Chem.* 268: 2007, 95–100.
- [9] Li, Q., Mahendra, S., Lyon, D.Y., Brunet, L., Michael, V.L., Li, D. and Alvarez P.J.J.. Antimicrobial nanomaterials for water disinfection and microbial control: potential applications and implications. *Water Res.* 42: 2008,4591–4602.
- [10] Albrecht, M.A., Evans, C.W., Raston, C.L. Green chemistry and the health implications of nanoparticles. *R. Soc. Chem.* 8 : 2006, 417–432.
- [11] Diallo, M.S. and Savage, N. Nanoparticles and water quality, *J. Nanopart. Res.* 7: 2005, 325–330.
- [12] Rodriguez Iznaga, V., Petranovskii, G., Rodriguez Fuentes, C., Mendoza, A. and Benitez Aguilar. Exchange and reduction of Cu²⁺ ions in clinoptilolite, *J. Colloid Interface Sci.* 316: 2005, 877–886.
- [13] Choi, O., Deng, K.K., Kim, N.J., Ross, L. J., Surampalli, R.Y. and Hu Z.. The effects of silver nanoparticles, silver ions, and silver chloride colloids on microbial growth. *Water Res.* 4 (2) 2008, 3066–3074.
- [14] Ren, G., Hu, D., Cheng, E.W.C., Vargas-Reus, M.A., Reip, P. and Allaker, R.P. Characterisation of copper oxide nanoparticles for antimicrobial applications, *Int. J. Antimicrob. Agents* 33: 2009, 587–590.
- [15] Magana, S.M., Quintana, P., Aguilar, D.H., Toledo, J.A., Angeles-Chavez, C., Cortes, M.A., Leon, L., Freile-Pelegri, Y., Lopez, T. and Sanchez R.M.T. Antibacterial activity of montmorillonites modified with silver, *J. Mol. Catal. A – Chem.* 281: 2008, 192–199.
- [16] Zodrow, K., Brunet, L., Mahendra, S., Li, D., Zhang A., Li, Q. and Alvarez, P.J.J. Polysulfone ultrafiltration membranes impregnated with silver nanoparticles show improved biofouling resistance and virus removal, *Water Res.* 43: 2009, 715–723.
- [17] Chou, W., Yu, D. and Yang, M. The preparation and characterization of silver-loading cellulose acetate hollow fiber membrane for water treatment. *Polym. Adv. Technol.* 16: 2005, 600–607.
- [18] Nangmenyi, G., Yue, Z., Mehrabi, S., Mintz, E. and Economy, J. Synthesis and characterization of silver-nanoparticle-impregnated fiberglass and utility in water disinfection, *Nanotechnology* 20: 2009, 495705.
- [19] Jain, P. and Pradeep, T. Potential of silver nanoparticle-coated polyurethane foam as an antibacterial water filter. *Biotechnol. Bioeng.* 90: 2005, 59–63.
- [20] Oyanedel-Craver, V. and Smith, J. Sustainable colloidal-silver-impregnated ceramic filter for point-of-use water treatment, *Environ. Sci. Technol.* 42: 2008, 927–933.
- [21] Yoon, K.Y., Byeon, J.H., Park, C.W. and Hwang, J. Antimicrobial effect of silver particles on bacterial contamination of activated carbon fibers. *Environ. Sci. Technol.* 42: 2008, 1251–1255.
- [22] Zhang, S., Fu, R., Wu, D., Xu, W., Ye, Q. and Chen, Z. Preparation and characterization of antibacterial silver-dispersed activated carbon aerogels. *Carbon* 42: 2004, 3209–3216.
- [23] N. Asmathunisha, K. Kathiresan, R. Anburaj and M.A. Nabeel, *Colloids and Surfaces B: Biointerfaces.* 79 :2010, 488.
- [24] Asmathunisha, N., and K. Kathiresan. "A review on biosynthesis of nanoparticles by marine organisms." *Colloids and Surfaces B: Biointerfaces* 103 (2013): 283-287.
- [25] Dharmendra, K., Tiwari, J., Behari and Prasenjit S. Application of Nanoparticles in Waste Water Treatment. *World Applied Sciences Journal* 3 : 2008, 417-433.
- [26] Nomcebo, H., Mthombeni, Lizzy Mpenyana-Monyatsib, Maurice, S., Onyango and Maggie N.B. Breakthrough analysis for water disinfection using silver nanoparticles coated resin beads in fixed-bed column, *Journal of Hazardous Materials* 133: 2012, 217– 218
- [27] Chick, H. [An Investigation of the Laws of Disinfection](#). *J. Hygiene* 8: 1908, 92 pp.
- [28] Matsumura, Y., Yoshikata, K., Kunisaki, S. and Tsuchido T. Mode of bactericidal action of silver zeolite and its comparison with that of silver nitrate *Appl. Environ. Microbiol.* 69: 2003, 4278- 4284.
- [29] Feng, Q.L., Wu, J., Chen, G.Q., Cui, F.Z., Kim, T.N. and Kim, J.O. A mechanistic study of the antibacterial effect of silver ions on *Escherichia coli* and *Staphylococcus*. *J. Biomed. Res.* 52: 2000, 662–668.
- [30] Seo, Y.I., Hong K.H., Kim, D.G. and Kim, Y.D. Ag/Al(OH)₃ mesoporous nanocomposite film as antibacterial agent *Colloids Surf. B: Biointerfaces* 81: 2010, 369-375.