# Ceramic Water Filters Impregnated with Silver Nanoparticles for the Removal of Lead and Chromium Ions from Water

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***Authors' contributions***

*This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.*

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## ABSTRACT

The high cost and complexity of existing techniques used to remove heavy metal ions from water have necessitated developing and implementing cost-effective and socially acceptable options that can be applied at the household level. This work examined the ability of ceramic water filters impregnated with silver nanoparticles to remove Lead and Chromium ions from water. Ceramic water filters were fabricated using locally sourced materials to filter water samples spiked with known concentrations of Lead and Chromium ions. The filters were categorized into two categories; without silver nanoparticles (control filters) and filters impregnated with silver nanoparticles using two application methods; dip-soak method and paint-method. The study assessed the effects of the addition of silver nanoparticles, application method of silver nanoparticles, initial water temperature, and initial metal ion concentration on the performance of the different sets of filters. It was observed that the addition of silver nanoparticles improved the ability of ceramic water filters to remove Lead and Chromium ions from water. The relative amounts of Lead and Chromium ions removed in the dip-soaked filters were higher (12.58% and 15.43%) than in the control filters (5.64% and 7.40%). The study also showed that the application method of silver nanoparticles influenced the ability of the filters to remove Lead and Chromium ions. The adsorption of Lead was higher in the Paint-

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method filter (14.45%) compared to the Dip-Soaked filter (12.58%), whereas the adsorption of Chromium was higher in the Dip-Soaked filter (15.43%) than in the Paint-Method filter (14.22%). The effect of initial water temperature, over the range of 24 °C to 84 °C, was insignificant since the cumulative percentage for the three sets of filters was less than 1%. Further, the initial metal ion concentration had a significant impact on the adsorption capacities of the ceramic water filters. In the control filters (without AgNPs), the relative amounts of Lead and Chromium ions removed decreased with an increase in initial metal ion concentration. However, for the Dip-soaked and Paint-method filters, the relative amounts increase with an initial increase in initial metal ion concentration up to a saturation point (0.6g/l), after which it decreases. Surface morphology studied using a Scanning electron Microscope (SEM) showed an excellent surface binding between the clay particles and silver nanoparticles. Therefore, ceramic water filters impregnated with silver nanoparticles offer a practical, low-cost, and socially acceptable alternative for removing heavy metal ions from water at the household level.

*Keywords: Adsorption; ceramic water filters; chromium; heavy metal ions; lead; silver nanoparticles.*

## 1. INTRODUCTION

Access to safe drinking water is crucial for human survival and improving living standards. The term "safe drinking water" refers to water that does not pose any health risk to users over a lifetime of consumption. In addition to minimizing health risks, safe drinking water needs to be acceptable, adequate, reliable, sufficient, available, and accessible [1]. Access to safe drinking water forms the basis of goal 6 of the United Nations' Sustainable Development Goals (SDGs). Under the goal UN aims to achieve universal and equitable access to safe and affordable healthcare by 2030. To help achieve the target, the UN has commissioned multiple water projects worldwide. Additionally, humanitarian organizations such as the World Health Organization (WHO) have been on the frontline in championing public access to safe drinking water.

Over the years, remarkable progress has been made towards improving universal access to water. Despite the progress, billions of people from developing countries still lack access to safe drinking water sources. According to reports by WHO, as of 2020, more than 2 billion people still lacked access to drinking water [2]. Most people depend on water from unprotected and untreated water sources such as wells, springs, rivers, lakes, streams, and ponds. These sources are prone to contamination by different contaminants, including physical, chemical, and biological pollutants. Consumption of contaminated water poses significant health risks such as cholera, diarrhoea, dysentery, and typhoid. The health effects associated with different contaminants thus raise concerns about the overall health and wellness of the public.

There have also been growing concerns about high levels of heavy metal pollutants in water and their potential effects on human health. Most heavy metals occur naturally and may be essential in life, but they may become toxic if accumulated in the body. Heavy metals may get to drinking water through different avenues such as mining, industrial production, untreated sewage sludge, and diffuse sources like metal piping. Examples of heavy metals found in drinking water include Arsenic, Lead, Cadmium, Mercury, Chromium, Nickel, and Zinc. Each of these heavy metals is linked to specific health effects. For instance, Lead is a potent neurotoxin that can damage the brain. It is also linked to nervous system damage and delays in physical and mental development among children. Among adults, consumption of Lead-contaminated water is linked to memory loss, kidney damage, anaemia, and digestive problems [3]. Dinking Chromium-contaminated water is also linked to detrimental health effects such as gastrointestinal disorders, liver damage, kidney injury and cardiovascular collapse [4]. The health effects of different heavy metals thus highlight the need to implement different treatment methods to ensure the safety and wellness of the public.

Over the years, different water treatment methods have been developed and studied for their efficacy in removing heavy metal ions from water. These methods can be broadly classified into photocatalytic, electric-based, chemical-based, membrane-based, and adsorption-based techniques. Photocatalytic techniques are based on photocatalysis, which involves accelerating a photoreaction in the presence of a catalyst. In removing heavy metals from water, photocatalysis produces photo-excited electrons

that help reduce the metal ions. Photocatalytic techniques have proven effective in eliminating Chromium ions from water using different photocatalysts such as Zinc Sulphide, Cadmium Sulphide, and TiO$_2$ [5]. Photocatalytic techniques have also been proven effective in removing Arsenic, Mercury, Cadmium, Lead, and Copper ions from water [6-8]. The different studies show that photocatalytic techniques may effectively remove heavy metals from water. However, the methods are complex and may not be applicable at household levels.

Researchers have also explored a wide range of electric-based techniques to remove different types of heavy metals from water. The methods are based on electrochemical systems in which oxidation and reduction occur at the anode and cathode, respectively. Examples of electric-based water treatment methods include electroflotation, electrooxidation, electrocoagulation, and electrochemical reduction. These different techniques have proven effective in removing heavy metals from water. For instance, electrochemical reduction has shown promising results in removing Lead, Copper, Cadmium, and Mercury ions [9]. Electroflotation is effective in the removal of Cadmium, Nickel, and Zinc. Lead, and Iron [10]. The primary advantage of electric-based water treatment techniques is that they do not introduce additional chemicals into the water. However, they require high energy consumption and may thus be expensive to implement, especially in low- and middle-income households.

In addition to photocatalytic and electric-based techniques, researchers have also widely explored chemical-based techniques to remove heavy metals from water. The methods involve adding chemicals to remove organic and inorganic water contaminants. Chemical-based techniques include ion exchange, chemical precipitation, flotation, and coagulation-flocculation. The methods have been proven effective in the removal of heavy metals such as Lead, Copper, Zinc, Chromium, Nickel, and Arsenic [11-13]. The primary advantage of chemical-based techniques is that they are highly effective. However, they are characterized by high operational costs and may introduce chemical pollutants into the water.

Various studies have also focused on membrane-based techniques for removing heavy metals from water. These techniques are based on the concepts of transportation and the rejection of substances by membranes. They can remove pollutants of micro- to nanometer ranges. Examples of membrane-based techniques include forward osmosis, reverse osmosis, nanofiltration, ultrafiltration, and microfiltration [14]. Researchers often used modified or non-modified membranes in their studies. For instance, nanofiltration membranes prepared from cerium oxide nanoparticles have been applied to remove Copper, Cadmium, Cobalt, Aluminium, and Iron from water [15]. The reverse osmosis method has also been proven effective in removing Zinc, Arsenic, Antimony, Nickel, and Chromium [16]. Membrane-based techniques offer high efficiency; however, they may be complex and challenging to implement at household levels.

Recently, there has been an increased focus on applying adsorption-based techniques to remove heavy metals from water. The methods are based on the concept of adsorption, in which the adsorbate is adsorbed on to a surface of an adsorbent. A wide range of adsorbents has been applied in studies to remove different classes of heavy metals. The various classes of adsorbents used include carbon-based adsorbents, chitosan-based adsorbents, mineral, magnetic, and bio-sorbents. For instance, carbon-based adsorbents such as polyrhodanine-modified multi-walled carbon nanotubes have proven efficient in removing Lead. Other types, such as chitosan-based adsorbents and biosorbents, have also been applied in the removal of heavy metals such as Lead, Chromium, Copper, Nickel, Mercury, and Cadmium [17-19]. Researchers have also explored the potential of biosorbents such as watermelon rind [20] and coffee husks [21] to remove heavy metal ions like Copper, Lead, and Cadmium.

Apart from bio-sorbents, magnetic adsorbents and nano-materials have also been used to remove heavy metal ions from water. For example, nano zero-valent iron particles have been used to remove Cadmium, Copper, and Lead ions [22] and gold nanoparticles used to remove Mercury. Silver nanoparticles have also been widely studied in removal of heavy metals. The silver nanoparticles have been embedded on different media such as activated alumina, Benjamina leaves extract, and Schiff base to remove heavy metal ions like Cadmium, Copper, Mercury and Lead [23-25]. The studies thus prove the potential of silver nano-particles as an adsorbent material.

Despite milestones in research, the application of such techniques at the household level is still limited. Thus, there is the need to develop an affordable and socially available technique that can be used to remove heavy metal ions at the household level. Ceramic Water Filters (CWFs) have recently emerged as low-cost, effective, and socially-acceptable alternatives for drinking water treatment at household levels. CWFs are prepared by mixing clay with a burnable material such as sawdust or rice husks. They may be further modified using silver nanoparticles to improve their efficiency in removing microorganisms and silver nanoparticles (AgNPs). Preliminary studies also show that modification of CWFs with AgNPs may enhance their ability to remove heavy metals, but limited research supports the assertion. This research sought to investigate the ability of Ceramic Water Filters (CWFs) impregnated with silver nanoparticles to remove Lead and Chromium Ions from water. It focused on Lead and Chromium because they are listed among the top most harmful heavy metals linked to cancer and neuronal damage. It builds on the existing literature by investigating the effects of adding AgNPs, the method of application of AgNPs, initial water treatment, and initial metal ion concentration on the ability of CWFs to remove Lead and Chromium ions from water. It aimed to validate the use of CWFs impregnated with silver nanoparticles as a viable technique for removing heavy metals from water at the household level.

## 2. MATERIALS AND METHODS

### 2.1 Raw Materials

The raw materials used to fabricate the CWFs included clay, sawdust, *Corchorus olitorius* juice, and silver nanoparticles. Clay was sourced from the Migori-Mabera area in Western Kenya, a region popular for pottery practice. The clay was dried in an oven and ground into micron-sized particles using a ball mill grinder. After grinding, the clay was sieved through a 425 µm sieve and stored for later use. Saw dust was obtained by sanding different hardwoods. The sawdust was also sieved through a 425 µm sieve to ensure consistency with the clay particles. *Corchorus olitorius*, a type of vegetable locally referred to as *'Mrenda'* was used as the binder source. When boiled, the vegetable produces a highly viscous juice, which has been proven to improve the bulk density and sintered strength of clay ceramics [26]. *Mrenda* was locally sourced from Western Kenya regions, where it is widely planted and

consumed. The *Mrenda* was then dried and cleaned with distilled water. After cleaning, the *Mrenda* was boiled for thirty minutes, let to cool overnight, and then sieved to extract the binder juice. The juice was then packed in sterilized, clean plastic bottles and refrigerated to prevent decomposition. Commercial AgNPs (65-75% Ag basis) of approximately 10nm in size were purchased from Sigma Aldrich and used as received. An 800mg/L stock solution of silver nanoparticles was prepared by dissolving the nanoparticles in a litre of deionized water. The prepared solution was then stored in a sterilized and clean volumetric flask for later use.

### 2.2 Fabrication of Ceramic Water Filters

Fabrication of the CWFs involved a five-step process, as represented in Fig. 1. In the first step, the previously prepared clay and sawdust were mixed in a volume ratio of 50:50. The ratio has been shown to produce filters of sufficient porosity and flow rate without compromising their strength. The Hobart electric mixer (MA-152F) was used to mix the two at a revolution of 180 revolutions per minute for thirty minutes. After mixing, the homogenous clay-sawdust mixture was stored for the next step. In the second moulding step, the homogenous clay-sawdust was combined with the extracted *Mrenda* Juice at a clay-to-binder ratio of 1:1. The mixture was evenly kneaded to form a dough then wrapped in a plastic bag and left to age for 36 hours. After 36 hours, the dough was divided into small portions in readiness for pressing. The Retsch Pellet Press PP 25 manual press produced ceramic disc filters. The press has a maximum pressure of 25 tons (250kN) and a die set of 32 mm in diameter. To make uniform CWFs, small portions of the clay dough (5g) were placed between the die set and pressed using a force of 1 ton for one second. The average size of the produced disc filters was 32 mm in diameter and 3 mm in thickness. The disc membranes were then let to dry at room temperature for one week.

After a week of drying, the disc membranes were fired using the Naberthen electric furnace (Model LH 15/14), which has a maximum temperature value of $1400^{o}C$. The firing was done using a three-step ramping process. In the first step, the disc membranes were preheated to $100^{o}C$ at a rate of $50^{o}C$ per hour. In the second step, the temperature of the furnace was increased to $450^{o}C$ at a rate of $150^{o}C$ per hour, and the filters were soaked at the temperature for two hours to ensure complete combustion of the sawdust. In

the last step, the temperature was raised to 850°C at a rate of 150°C per hour and the filters were soaked at the temperature for four hours. The furnace was then switched off and let to cool down to room temperature before removing the disc filters. After firing, the filters were divided into two categories. The first category acted as the control filters (CF) and was not impregnated with silver nanoparticles. For the second category, two methods (dip-soak and paint) were used to impregnate the CWFs with silver nanoparticles. In the dip-soak process, the disc filters were dipped in portions of the previously prepared 800 mg/L stock solution and placed over a magnetic stirrer. In the paint method, portions of the AgNPs stock solution were painted on the edges, top surfaces, and bottom surfaces of the disc filters using a paint brush. The three sets of filters (Control Filters (CF), Dip-Soaked Filters (DS) and Paint Method Filters (PM)) were then safely stored for further tests.

## 2.3 Preparation of Lead and Chromium Spiked Water Solutions

Spiked water solutions containing Lead and Chromium ions were prepared by dissolving their metal nitrates in deionized water. Each standard solution was prepared by dissolving 1.6g of $Pb(NO_3)_2$ and 7.93g of $Cr(NO_3)_3.9H_2O$ respectively, in one litre of deionized water. The standard solutions were stored in clean and sterilized volumetric flasks for later use. The prepared standard solutions represented 1000 ppm of Lead and Chromium ions in terms of concentration.

## 2.4 Heavy Metal Removal Tests

The heavy removal tests included four independent tests aimed at investigating the effect of variation of specific parameters on the ability of the CWFs to remove Lead and Chromium ions. The first test sought to investigate the effect of the addition of silver nanoparticles, wherein 30 ml of 0.6g/l of the Lead and Chromium spiked water solutions were poured into separate filtration units fitted with the control filters and the dip-soaked filters. The second test aimed to investigate the effect of method of application of AgNPs. As in the first test, 30 ml of 0.6 g/l of Lead and Chromium solutions were poured into separate filtration units fitted with the dip-soaked and paint-method filters. In the third test, temperatures of 30 ml portions of 0.6 g/l of Lead and Chromium solutions were varied from 24 °C to 84 °C at intervals of 20 °C before pouring into separate filtration units fitted with the three filter sets (CF, DS, and PM). In the last test, different concentrations (0.2 g/l, 0.4 g/l, 0.6 g/l, and 0.8 g/l) of the Lead and Chromium ions were prepared through dilution of the initially prepared standard solutions. 30 ml portions of the different concentrations were poured into separate filtration units fitted with the three sets of filters (CF, DS, and PM).

## 2.5 EDXRF Measurement of Concentrations of Lead and Chromium Concentrations

The Energy Dispersive X-ray Fluorescence technique was used to quantify the Lead and Chromium ions concentrations before and after filtration. The NEX-CG Rigaku EDXRF machine was used in the study to quantify the concentrations of lead and chromium ions before and after filtration. 3ml of the representative samples were poured into double-ended cups sealed on one end with a 4μm thick polypropylene XRF film. The concentrations were quantified in parts per million (ppm). The values obtained were substituted into equation 1 to calculate the percentage (%) removal for each set of filters.

$$\%Removal = \frac{c_i - c_f}{c_i} \qquad (1)$$

Where $c_f$ is the ion concentration after filtration, and $c_i$ is the ion concentration before filtration in parts per million (ppm).
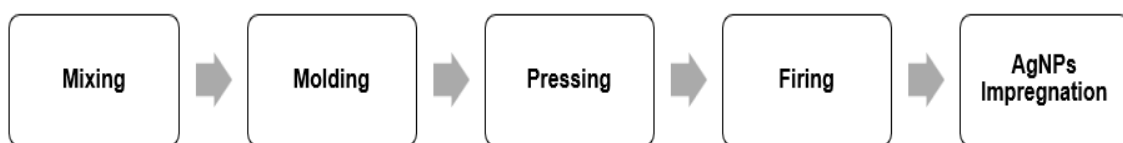


**Fig. 1. Schematic of fabrication process of the ceramic water filters**

## 2.6 Morphological Study of the CWFs Using a Scanning Electron Microscope (SEM)

Scanning Electron Microscopy technique was applied to study the fabricated ceramic filters' surface morphology to observe the silver nanoparticles' particle distribution. A Zeiss GeminiSEM system with a maximum of 30 keV acceleration energy was used in the current study. Representative samples were cut from the different sets of filters, adequately cleaned, and smoothened with emery paper. The samples were then coated with nanometers-thick layers of gold and palladium to enhance their optical reflectivity and conductivity. The coated samples were then mounted on the sample holder and analyzed using a 15 kV electron beam to obtain morphology images.

## 3. RESULTS AND DISCUSSION

The results of the effect of the addition of silver nanoparticles on the ability of ceramic filters to remove Lead and Chromium ions are presented in Fig. 2. It was observed that impregnation CWFs with silver nanoparticles improved their ability to remove Lead and Chromium ions. In the CWFs without AgNPs (CF), the percentage removal for Lead and Chromium ions was 5.64% and 7.40%, respectively. The values of the dip-soaked CWFs (DS) were 12.58% and 15.43% for

Lead and Chromium ions, respectively. The difference in performance of the two sets of filters can be attributed to the fact that the silver nanoparticles in the DS filters act as active sorption sites that adsorb the heavy metal ions from the water. As the spiked water sips through the ceramic filter, the Lead and Chromium ions get adsorbed by the AgNPs. It was also observed that the percentage removal of Chromium ions was higher than for Lead ions. This trend can be attributed to the smaller ionic radius of Chromium (52 picometers) compared to that of Lead (119 picometers). Studies have shown that the smaller the ionic radius, the higher the adsorption capacity [27].

The results of the effect application method of silver nanoparticles on the ability of the CWFs to remove Lead and Chromium ions are presented in Fig. 3. It was observed that in the dip-soaked filters (DS), the proportions of Lead and Chromium ions that were removed from the water samples were 12.58% and 15.43%, respectively. For the paint-method filters (PM), the relative removed quantities were 14.45% and 14.22% for Lead and Chromium ions, respectively. The results indicate that the adsorption capacity of Lead was higher in the PM filters. In comparison, the adsorption capacity of Chromium ions was higher in the DS filters than the PM filters.
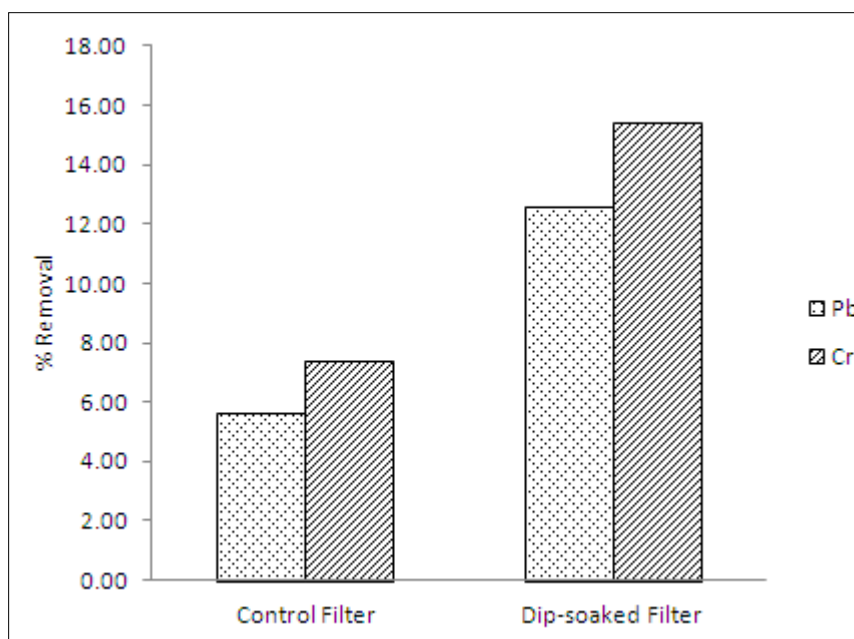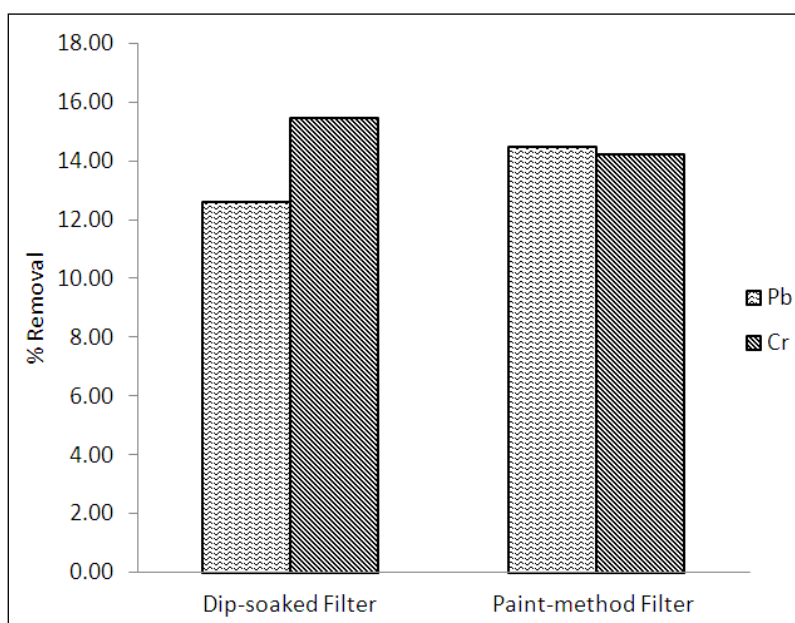


**Fig. 2. Comparison of removal of Cr and Pb ions by the control filters and dip-soaked filters**

**Fig. 3. Comparison of the removal of Lead and Chromium Ions by dip-soaked and paint-method filters**

Fig. 4 represents the results of the effect on initial water temperature on the ability of CWFs to remove Lead and Chromium ions. The relative amounts of Lead and Chromium removed reduced as the initial water temperature increased from 24°C to 84°C. The trend can be explained by comparing coefficients of linear thermal expansion of the CWFs, Lead, and Chromium ions. The coefficients of linear thermal expansions of CWFs, Lead and Chromium, are $5.5 \times 10^{-6}$ $K^{-1}$, $28.9 \times 10^{-6}$ $K^{-1}$ and $4.9 \times 10^{-6}$ $K^{-1}$, respectively. As the temperatures of spiked water samples are increased from 24°C to 84°C, the Lead and Chromium ions are likely to expand. Lead's coefficient of linear thermal expansion is about five times larger than that of the CWFs; therefore, the temperature rise has no significant effect on its adsorption capacity.

On the other hand, the coefficient of linear thermal expansion of Chromium is relatively lower than that of the CWFs; therefore, the temperature rise may have a minimal effect on the adsorption capacity of Chromium ions. Further computation of cumulative percentage change using equation 2 showed that the incremental percentage removal for all the sets of filters within the 24-84 °C temperature range was less than 1%. The results thus imply that the effect of temperature on the adsorption of Lead and Chromium ions is negligible within the specified temperature range.

The results of the effect of the initial ion concentration of Lead and Chromium on the adsorption capacity of the CWFs are shown in Fig. 5. For the CWFs without AgNPs (CF), it was observed that the percentage removal of Lead and Chromium ions decreased with an increase in initial ion concentration. The trend can be attributed to the fact that at higher concentrations, the pores get clogged by the heavy metal ions, thus decreasing the proportion of heavy metal ions filtered from the water. The ratios of Lead and Chromium ions removed for the DS and PM filters increase steadily for initial metal ion concentration between 0.2g/l to 0.6g/l. After 0.6g/l, the removed ratios decrease in both the DS and PM filters. The observed trends indicate that as the initial ion concentrations are initially increased from 0.2g/l to 0.6g/l, there is an increase in competition for the available binding sites, which are the AgNPs surfaces. However, adsorbents have a saturation limit. Thus, beyond an initial ion concentration of 0.6g/l, the percentage removal of Lead and Chromium ions decreases, indicating that 0.6g/l was the maximum saturation limit for the fabricated CWFs.

The SEM micrographs of the CWFs produced at different magnifications are presented in Fig. 6. Morphological analysis of the micrographs showed that the AgNPs were randomly distributed on the surface of the CWFs. It was observed that the AgNPs were firmly attached to

the membranes, indicating that they were successfully impregnated onto the CWFs, and played a role in the adsorption of Lead and Chromium ions.
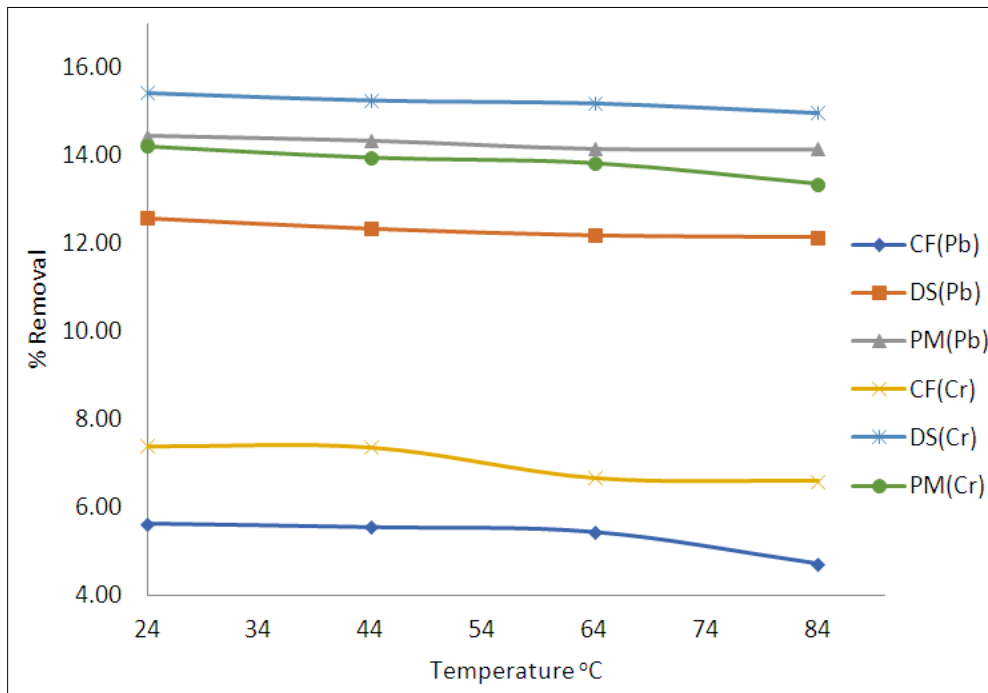


**Fig. 4. Representation of Removal of Pb and Cr ions at different initial water temperatures by Control (CF), Paint-Method (PM), and Dip-Soaked (DS) filters**
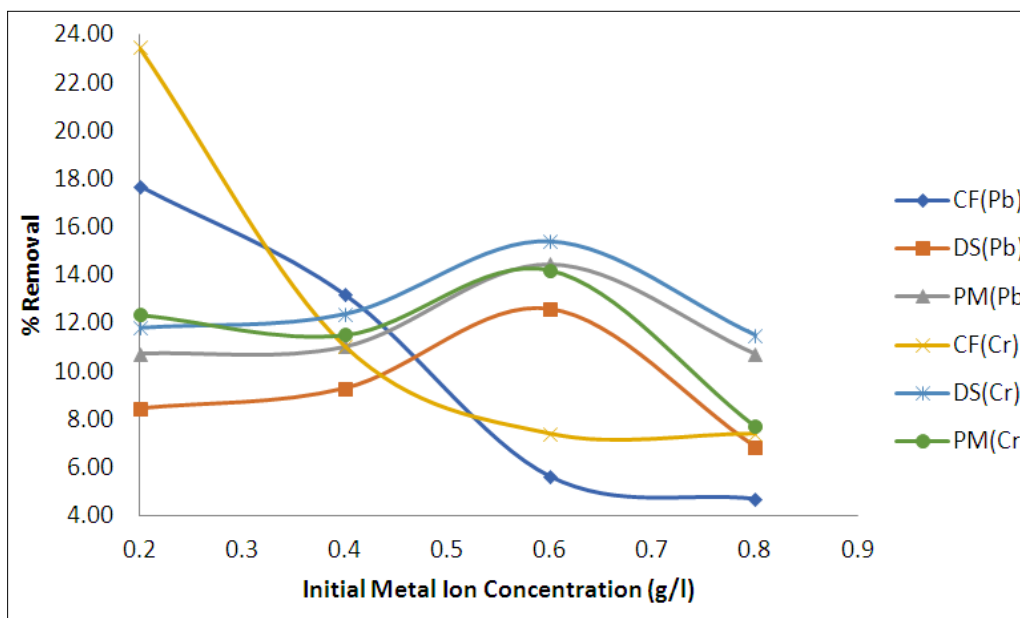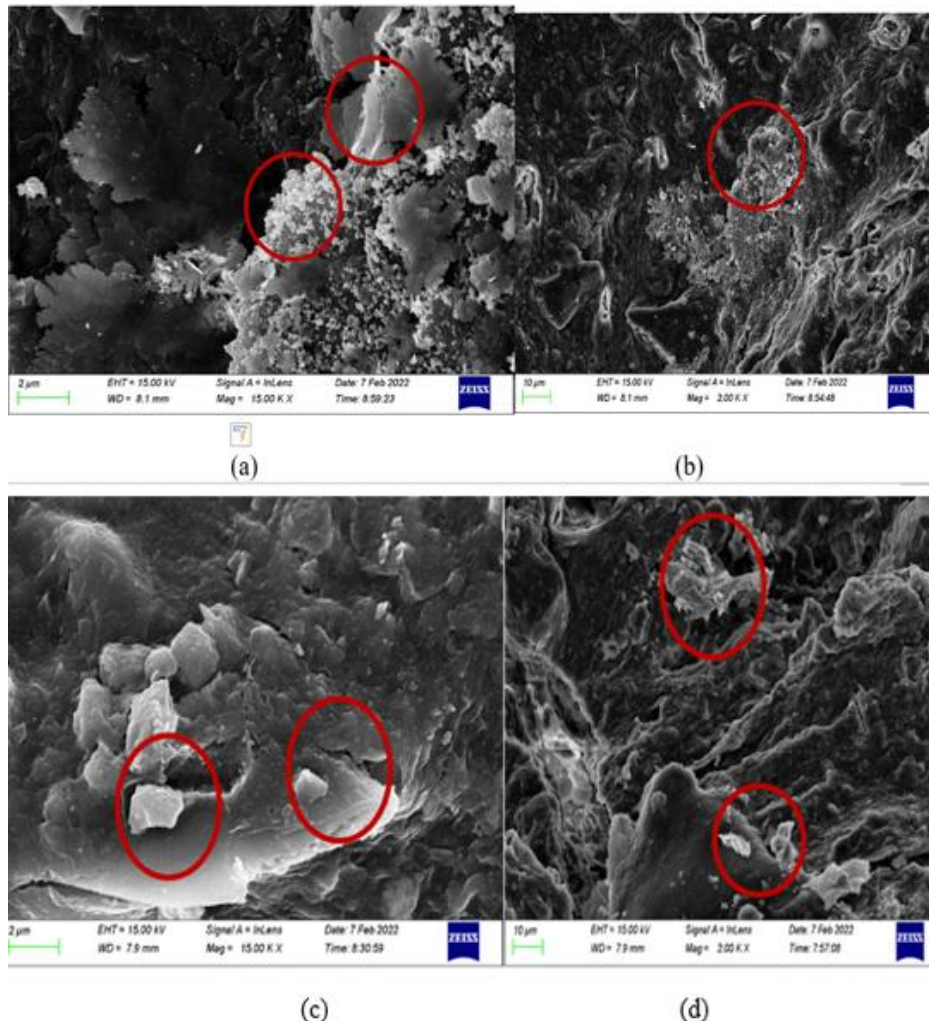


**Fig. 5. Comparative representation of the removal of Pb and Cr ion at varying initial ion concentrations by Control (CF), Paint-Method (PM), and Dip-Soaked (DS) filters**

**Fig. 1. SEM micrographs of two samples of CWFs indicating a random distribution of silver nanoparticles on the surface of the membrane (Circled parts)**

## 4. CONCLUSION AND RECOMMENDA-TIONS

This work has examined the ability of ceramic water filters impregnated with AgNPs to remove Lead and Chromium ions from water. The experiments show that impregnating CWFs with silver nanoparticles improves their ability to remove Lead and Chromium ions from water. The relative amounts of Lead and Chromium ions removed in the dip-soaked filters were higher (12.58% and 25.43%, respectively) than in the control filters (5.64% and 7.40%, respectively). The experiments also revealed that the application method of the AgNPs can influence the ability of the CWFs to remove Lead and Chromium ions. The adsorption of Lead was higher in the Paint-method filter (14.45%) compared to the Dip-Soaked filter (12.58%), whereas the adsorption of Chromium was higher

in the Dip-Soaked filter (15.43%) than in the Paint-Method filter (14.22%). It was further observed that the influence of initial water temperature on the ability of the CWFs to remove Lead and Chromium ions was limited over the 20 °C - 84 °C temperature range. The cumulative percentage removal over the range was less than 1% for all three filter types. However, further appropriate tests can be done to determine the effect of higher temperatures.

The experiments also revealed that the initial ion concentration impacts the ability of CWFs to remove Lead and Chromium ions. In the control filters (without AgNPs), the relative amounts of Lead and Chromium ions removed decreased with an increase in initial metal ion concentration. However, for the Dip-soaked and Paint-method filters, the relative amounts increase with an initial increase in initial metal ion concentration

up to a saturation point (0.6g/l), after which it decreases. The SEM measurements further reveal good surface binding between the CWFs and AgNPs. The study's findings show that CWFs impregnated with AgNPs are a convenient, low-cost, and effective household water treatment option that can remove heavy metal ions from drinking water. The limited effect of initial water temperature shows that the AgNPs-impregnated CWFs are suitable for use at all times and during different weather seasons. The findings of the current study show that AgNPs-impregnated CWFs could be used to remove heavy metal ions from water at household levels, thus reducing the health-related risks of drinking water contaminated by Lead or Chromium. It contributes to the field of materials science and water treatment industry by recommending a socially acceptable and affordable technique. Future research can build on the findings of the current study through inclusion of a wider range of heavy metal ions. Other experimental methods can also be applied to validate and improve on the current study.

## COMPETING INTERESTS

Authors have declared that no competing interests exist.

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