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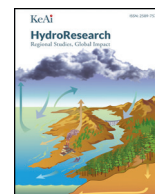
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Removal of arsenic in a sand filter coupled with zero valent iron

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ABSTRACT

Arsenic (As) in wastewater has negative effects on the environment and human health, hence As containing wastes must be handled properly. Given the accessibility of metallic iron, studies investigating into the potential application of zerovalent iron in the removal of arsenic are promising. In this study, the performance of sand filter blended with several kinds of zero valent iron (Fe⁰), such as iron wool, iron filings, and iron nails, were compared. These materials were combined in a sand filter column, and the efficiency was calculated using the As concentrations in the influent and effluent samples. Experiments were carried out in order to compare performance as a function of Fe⁰ dose and contact time. The outcome of this investigation showed that sand filter containing iron wool had a better removal efficiency of arsenic removal than iron filings and iron nails. The results in all columns showed that as dosage was increased, removal efficiency of arsenic increased significantly. In case of contact time the results revealed that arsenic can effectively be removed from water in the first 48 h. The early adsorption response is quick in all columns, but get slower as time goes on. The highest removal efficiency was 99.6% and the lowest removal efficiency was 82.7%.

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1. Introduction

Every human being has the right to safe drinking water at a reasonable cost (Fida et al., 2022). Population increase and climate change have impacted access to safe drinking water, putting public health at risk (Tang et al., 2019). One type of pollutants that pose a significant threat to human being is heavy metals including Arsenic (Richards et al., 2022; Shankar et al., 2014). Due to occupational or home exposure, Arsenic is considered as one of the hazardous metals of environmental significance and listed as one of the “Top 20 Hazardous Substances” by the US Agency for Toxic Substances and Disease Registry (ATSDR) (Zawierucha et al., 2022). Due to its toxicity and carcinogenic properties, arsenic contamination is a concern (Ahmad et al., 2018; Zhou et al., 2020). The most frequent side effects from prolonged exposure to arsenic are cancer, skin lesions, digestive problems, peripheral neuropathy, diabetes, renal impairment, and cardiovascular disease (Chowdhury et al., 2019). Maximum likelihood estimates of the risk of cancer for a 70-kg person consuming 2 L of water per day contaminated with 1 µg/L of arsenic range from 1×10^{-3} to 2×10^{-3} . The US EPA oral

slope factor of 1.5 per mg/kg bw per day is based at the middle of this range (Rashadul and Chowdhury, 2015).

Arsenic's (As) name originates from the Greek word arsenikon, which means yellow orpiment (Singh Thakur and Semil, 2013). Its sources include both anthropogenic and natural geochemical activities including mining, the release of industrial waste, and fertilizer use in agriculture (Huang et al., 2014). Both organic and inorganic forms of arsenic can be found in nature; inorganic arsenic is often found in natural water systems. Despite the fact that arsenic has multiple oxidation states, including 3, 0, +2, +3, and +5, arsenite (As(III)) and arsenate (As(V)) are the most prevalent oxidation states encountered in water. As(V) is found as oxyanions of arsenic acid, whereas As(III) is found as arsenious acid (Zawierucha et al., 2022). Typically, arsenite is the prevalent species in anaerobic environments, whereas arsenate occurs in aerobic environments (Huang et al., 2014). Even so, groundwaters are not always subject to this rule, it has been discovered that some groundwaters solely contain As(III), while others only contain As(V), and yet others have both As(III) and As(V) (Lekić et al., 2013).

In comparison to arsenate, arsenite is 20–60 times more toxic (Quino-Favero et al., 2021; Rodríguez-Martín et al., 2022). Arsenite is also more mobile than arsenate and thermodynamically more stable in reducing groundwater conditions (Chowdhury et al., 2019). The World Health Organization (WHO) recommends that arsenic levels in drinking water be less than 10 mg/L (Smith et al., 2017; Wan et al., 2012). However, even at concentrations below 10 mg/L, arsenic can

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have significant consequences for human health (Gude et al., 2018). The development of treatment technologies for the removal of arsenic at the household or small community level is therefore necessary. A good technique should not necessitate a considerable amount of manual labour or chemical usage.

Arsenic can be removed from aqueous solutions using a wide range of physicochemical techniques. The mobility of arsenical forms in water is strongly influenced by pH, redox conditions, and the presence of various chemical species. As a result, these physicochemical characteristics must be considered when developing removal techniques (Morgada et al., 2009). Among the methods used to remove arsenic includes coagulation and flocculation, membrane techniques, ion-exchange or chelation, chemical precipitation, and activated carbon adsorption (Chiban, 2012). However, these methods require a lot of energy and/or chemicals, which makes the treatment procedures expensive. They are also difficult to operate and maintain, necessitating regular part replacement, expensive aftermarket services, and specialized people who may not be available locally (Annaduzzaman et al., 2021). This makes them unsustainable over time in societies with limited resources. Filtration is the most reliable and effective method for removing arsenic ions at the household level due to its simplicity in designing, convenience of use, and cheap operating costs (Kim et al., 2022). For the adsorption of arsenic ions, a variety of materials have been employed, including rice polish, zeolite, red mud, activated alumina, surface-modified carbon black, iron hydroxides and oxides, open-celled cellulose sponge, and other adsorbents (Keerio et al., 2021). Systems using sand filters are frequently utilized to treat wastewater on-site and in small communities all over the world. It combines biological, chemical and physical processes. Compared to other natural technologies, it requires less land and doesn't need energy or chemicals (Yettefti et al., 2013).

Arsenic removal from aqueous solutions using iron, ferric oxide, and iron-based compounds as effective adsorbents has been well-documented (Kim et al., 2022; Smith et al., 2017). The process involves the corrosion of both Fe^0 and Fe^{2+} to produce $\text{Fe}^{3+}(\text{aq})$, H_2 , some precipitates, and green rusts (Deewan et al., 2022). These corrosion reactions and products are what cause exposed chemical species to undergo reductive transformation and/or be physically removed (Crane and Scott, 2012; Yang et al., 2015). Over the past decade, zero-valent iron (Fe^0) has drawn a lot of interest due to its several positive attributes, including non-toxicity, abundance, affordability, ease of production, and less maintenance on reduction process (Plessl et al., 2022; Simon et al., 2016). The removal mechanism entails the directional transfer of electrons from zero valent iron to the pollutant, which changes it into non-toxic or less toxic species. The processes involved includes adsorption, surface precipitation, surface complexation, reduction, oxidation, and possible co-precipitation (Feroze Ahmed, 2001; Fu et al., 2014). According to studies, iron corrosion plays an important role in arsenic removal due to the remarkable binding ability of Fe^0 corrosion products for arsenic (Zhao et al., 2021).

Zero valent iron is normally placed between the sand layers in a filter (Bretzler et al., 2020). It may take many different forms, such as composite iron matrix, iron fillings, iron nails, steel wool, sponge iron or cleaned scrap iron (Wenk et al., 2014). Each of these materials offers advantages for the intended application and they are also widely available and affordable (Hu et al., 2019; Makota et al., 2017). The type of iron material used, the physicochemical properties of the water, the starting arsenic concentration, and the arsenic speciation all affect how effective zero valent iron is at removing arsenic from the water (Biterna et al., 2010). Despite the fact that various studies have explored the effectiveness of various forms of zero valent iron in removing As, there is insufficient information comparing the efficiency of iron wool, iron fillings, and iron nails in removing As under the same physical conditions. Thus, the objective of this research was to compare the effectiveness of iron fillings, iron nails, and iron wool as zero valent iron for the removal of arsenic from simulated groundwater. The information from this work will help in the understanding and development of arsenic

removal techniques using readily accessible materials as zero valent iron. The adsorption of As(III) was investigated as a function of contact time, pH and adsorbent dosage.

2. Materials and methods

2.1. Chemicals and reagents

In this study, analytical grade chemicals were used without further purification. Arsenic trioxide (As_2O_3) weighing 12.00 g was dissolved in 5 L of di-ionized water to create 1000 mg/L stock solution of As (III). The stock solution was diluted to create solutions with 1000 $\mu\text{g}/\text{L}$ concentrations. A 0.1 M solution of sodium hydroxide (NaOH) and 0.1 M hydrochloric acid (HCl) were used to adjust the pH during the experiments. All glassware and plastic bottles were cleaned by soaking them in 10% nitric acid (HNO_3) and then rinsing them three times with deionized water.

2.2. Construction of the column filters

Three downscaled laboratory filters made of plastic columns (17 cm internal diameter and 50 cm height) were used to filter water with an initial concentration of 1000 $\mu\text{g}/\text{L}$ As (III). As illustrated in Fig. 1, the columns were filled with various vertical layers of gravel, zero valent iron material, and sand. Gravel (10 to 30 mm), coarse sand (1 mm to 2 mm) and medium to very fine sand (1/16 to 1 mm) was used. The columns were packed from the bottom to the top 10 cm gravel, 15 cm of a coarse sand, zero valent iron and 10 cm of fine sand. The sand utilized was a natural resource that had been sieved selectively with a set of sieves for sand analysis. Using tap water, the dirt was washed multiple times until the wash water became clear. The gravel utilized was a natural material obtained from local traders in Arusha town. The experimental setup was made at Arusha Technical College's water laboratory. After testing is complete, the device is supposed to be transferred for application in households but with appropriately designed housing.

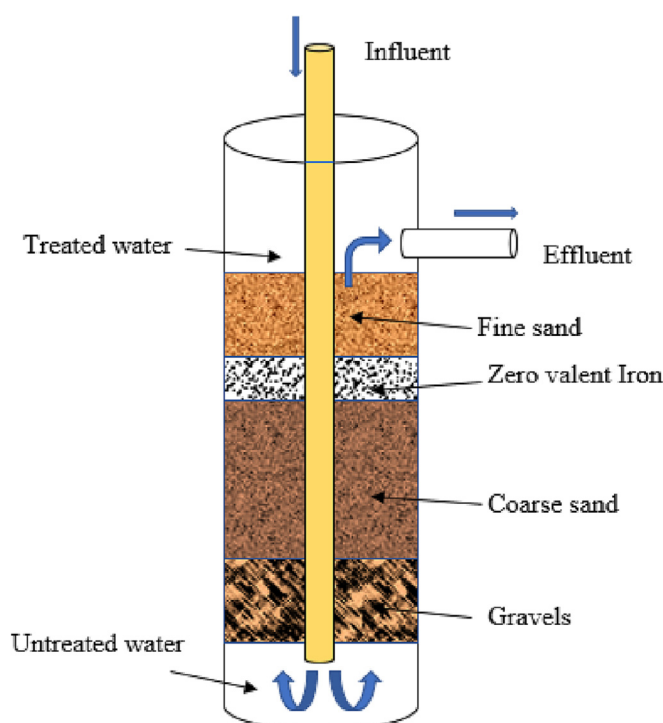


Fig. 1. Schematic diagram of the column filters with layers of substrates.



Fig. 2. Iron wool, iron fillings and iron nails used in experiments.

2.3. Batch experiments

Iron wool, iron fillings, and iron nails (Fig. 2) were all utilized as forms of zero valent iron. Iron wool and iron nails were obtained at local Arusha construction supplies shop. Iron fillings were gathered from welding shops in Arusha. These materials may be contaminated with grease and other pollutants from the manufacturing process. As a result, they were cleansed in an ultrasonic bath three times with acetone. Table 1 lists their elemental compositions. The synthetic ground water runs up the gravel bed and emerges at the top of the fine sand, where it is collected at the exit tap. Experiments were carried out with various material doses (200 g, 300 g, 400 g, 600 g, and 800 g) and contact times (6–72 h). After each contact period, triplet samples were collected and analysed.

2.4. Analysis of arsenic in samples

Samples (100 mL) were collected directly at the effluent outlet of each filter column. Arsenic in effluent samples was analysed using an Atomic Absorption Spectrometer at Arusha Technical College's water laboratory. All acid digestion and dilution work was done in a clean laminar flow bench equipped with an exhaustive system. The calibration of the instrument produced a linear calibration curve through zero and correlations above 0.9900.

2.5. Data analysis and interpretation

Excel software was used for statistical data analysis, including mean and visual analysis. The efficiency of arsenic removal was calculated using Eq. (1).

$$R = \frac{(C_{in} - C_{eff})}{C_{in}} \times 100 \quad (1)$$

Where;

R: removal efficiency (%).

C_{in} : the influent concentration of arsenic in the solution ($\mu\text{g/L}$).

C_{eff} : the effluent concentration of arsenic in the solution ($\mu\text{g/L}$).

Table 1
Elemental composition of used substrates.

Substrates	% Composition				
	Si	Ca	Al	Fe	Others
Fine sand	38	2.1	0.9	0.4	58.6
Gravel	18	8	6	13	55
Coarse sand	42	0.53	0.94	0.3	56.2
Iron nail	2.92	1.46	0.87	60.30	34.45
Iron wool	1.7	0.25	0.35	67.29	30.41
Iron filling	0.28	–	–	71.68	28.04

3. Results

The influence of dose and retention time on the performance of iron wool, iron filings and iron nails on removal of Arsenic was investigated. The initial concentration As (C_{in}) was set to be 1000 $\mu\text{g/L}$. The Table 2, summarises the effluent arsenic concentration (C_{eff}) and the removal efficiencies (% Removal) in all the columns.

3.1. Effect of contact time on arsenic removal efficiency

The contact time between the adsorbate and adsorbent is one of the most important design elements impacting the adsorption process's performance. The results in Figs. 3 and 4 demonstrate the time-dependent nature of As removal using 200 g and 800 g of zero-valent iron, respectively. According to the results, the rate of As removal increased continuously as the contact time was increased.

3.2. Effect of dose of zero valent iron on arsenic removal efficiency

The process of removing arsenic is significantly influenced by the adsorbent dosage. In order to reduce arsenic concentrations in an affordable way, the sorbent dosage must be optimized for practical applications. The removal of arsenic as a function of the dose of zero valent iron added in the filter was studied using 1000 $\mu\text{g/L}$ As (III). The results in all columns demonstrated that as dosage increased, removal efficiency increased. The effect of adsorbent dose on removal of arsenic with 6 h and 72 h contact time, are presented in Figs. 5 and 6 respectively.

3.3. Effect of type of zero valent iron

The results of this study demonstrate the variable performance of iron wool, iron filings, and iron nails in removing As at various dosages and contact times. Iron wool performed better than iron filings and iron nails with a fixed 200 g of the materials. With 800 g of the materials, iron filings performed better in a shorter contact time, while iron wool performed better in a longer contact time. When the contact time was set to 6 h, the results shows that iron wool performed better with a low dosage of materials, while iron filings performed better with a higher dose of materials. Iron filings and iron wool performed relatively similarly when the contact time was set to 72 h.

4. Discussion

4.1. Effect of contact time on arsenic removal efficiency

The removal efficiency increased significantly as contact time increased from 6 to 48 h, and then slowly as contact time increased. These findings indicate that arsenic was promptly removed, and that

Table 2
The removal of Arsenic at different dose and retention time.

Iron wool					Iron filings			Iron nails		
Dose of Fe (g)	Time (h)	Range (µg)	Average (µg)	SD	Range (µg)	Average (µg)	SD	Range (µg)	Average (µg)	SD
200	6	112.34–181.56	146.95	14.47	159.11–186.83	172.97	9.80	152.91–164.69	158.8	4.16
200	12	66.76–78.32	72.54	4.09	119.26–130.5	124.88	3.97	132.67–138.79	135.73	2.16
200	24	40.21–49.07	44.64	3.13	76.92–84.4	80.48	2.52	90.25–99.93	95.09	3.42
200	48	18.55–35.41	26.98	5.96	32.34–41.46	36.99	3.29	58.53–70.71	64.62	4.31
200	72	14.23–37.83	26.03	8.34	21.33–24.91	23.12	1.27	52.66–65.5	59.08	4.54
300	6	109.17–128.93	119.05	6.99	140.56–146.2	143.38	1.99	139.45–144.91	142.18	1.93
300	12	59.23–63.53	61.38	1.52	97.29–104.35	100.82	2.50	129.56–136.36	132.96	2.40
300	24	36.12–49.42	42.77	4.70	61.67–69.69	65.68	2.84	73.78–75.8	74.79	0.71
300	48	19.22–31.00	25.11	4.16	27.98–33.08	30.53	1.80	51.45–55.63	53.54	1.48
300	72	13.2–20.28	16.74	2.50	12.56–20.74	16.65	2.89	44.93–52.97	48.95	2.84
400	6	101.21–114.57	107.89	4.72	110.91–118.47	114.69	2.67	109.27–117.87	113.57	3.04
400	12	51.63–67.39	59.51	5.57	72.89–76.97	74.93	1.44	102.54–115.34	108.94	4.53
400	24	29.22–37.74	33.48	3.01	54.45–60.27	57.36	2.06	63.27–65.97	64.62	0.95
400	48	13.91–17.73	15.82	1.35	22.94–32.56	27.75	3.40	47.93–57.33	52.63	3.32
400	72	3.23–9.77	6.5	2.31	11.59–16.17	13.88	1.62	44.47–49.71	47.09	1.85
600	6	94.22–117.82	106.02	8.34	83.93–86.29	85.11	0.83	102.78–120.64	111.71	6.31
600	12	43.43–71.91	57.67	10.07	47.67–57.79	52.73	3.58	88.94–95.7	92.32	2.39
600	24	21.38–34.42	27.9	4.61	27.68–31.54	29.61	1.36	57.47–64.41	60.94	2.45
600	48	9.28–13.04	11.16	1.33	16.91–21.95	19.43	1.78	26.84–32.28	29.56	1.92
600	72	3.22–7.54	5.38	1.53	10.11–15.81	12.96	2.02	24.89–28.69	26.79	1.34
800	6	66.42–76.82	71.62	3.68	62.9–68.46	65.68	1.97	60.11–67.31	63.71	2.55
800	12	42.67–54.03	48.35	4.02	35.56–40.3	37.93	1.68	54.89–61.45	58.17	2.32
800	24	13.78–19.70	16.74	2.09	28.89–30.33	29.61	0.51	44.44–49.74	47.09	1.87
800	48	5.88–9.02	7.45	1.11	16.12–19.02	17.57	1.03	24.75–28.83	26.79	1.44
800	72	3.67–5.65	4.66	0.70	3.12–4.26	3.69	0.40	21.78–26.26	24.02	1.58

this removal increased quickly in the first 48 h. Arsenic removal increased over time due to holes and corrosion on metal surfaces, which increased the absorption cross section and active sites for arsenic adsorption (Biterna et al., 2010). As the contact time increases, the adsorbent has more time to absorb arsenic from the aqueous system (Azhdarpoor et al., 2015b; Khodabakhshi et al., 2021). The initial fast adsorption was caused by metal ions moving quickly to the surface of the adsorbent particles (Ahmed et al., 2021; Zhu et al., 2020). The subsequent slower phase was brought on by the reduction in empty sites, and the main mechanisms for the uptake of arsenic during this phase were intraparticle diffusion and surface precipitation (Rahdar et al., 2019; Wang et al., 2022; Wu et al., 2021).

4.2. Effect of dose of zero valent iron on arsenic removal efficiency

Adsorbent dosage has a considerable impact on how effectively pollutants are removed because it provides for cost-effective implementation of the process in the natural environment (Zou et al., 2016). The dependence of adsorbent dose on removal of As was studied using 1000 µg/L As (III) with varying dose of zero valent iron at 6 h and 72 h contact time. The results of the present study show that as dosage of zero valent iron increased, removal efficiency of As increased in all columns. This observation is based on the fact that increasing the amount of adsorbent increases the surface-active sites of adsorption and the probability of contact between arsenic and zero valent iron. This results

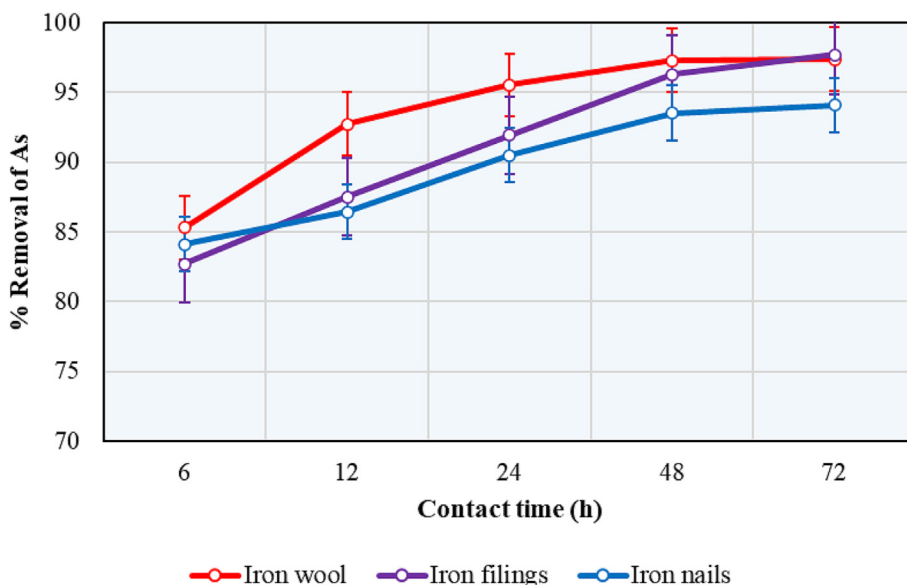


Fig. 3. Effect of contact time on removal efficiency of arsenic (200 mg of adsorbent).

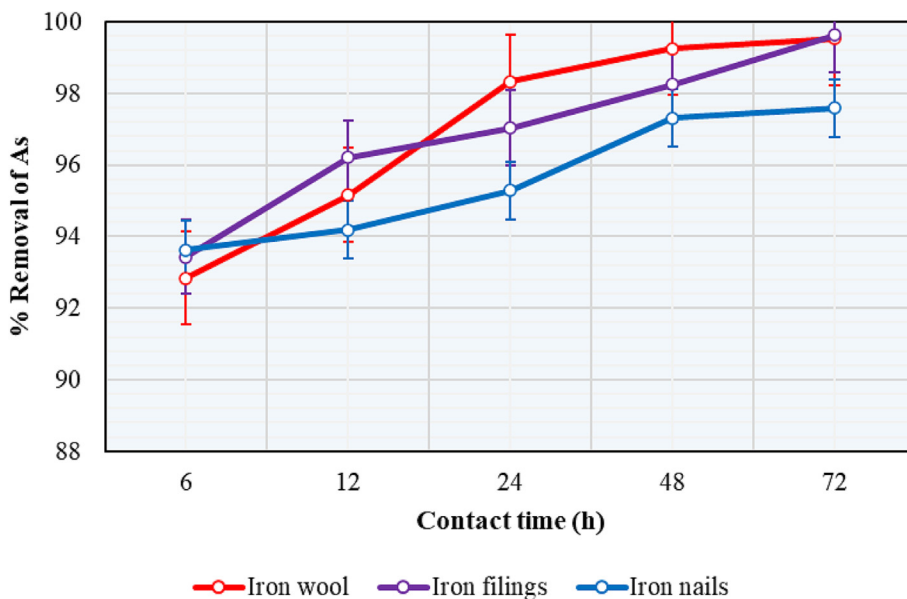


Fig. 4. Effect of contact time on removal efficiency of arsenic (800 mg of adsorbent).

in increased oxidation and reduction processes (Azhdarpoor et al., 2015b; Shih et al., 2011). Increasing the zero valent iron dosage increases the amount of free e- and unoccupied sites, which increases As removal (Abdel-Aziz et al., 2020).

4.3. Effect of type of zero valent iron

The findings of this study indicate that iron wool generally performs better than iron filings and iron nails at removing As. Different iron samples from various origins typically behave differently when it comes to removing As (Lackovic et al., 2000). It is generally considered that the intrinsic properties of iron, such as surface area, impurities, and oxide film coating, are factor which contributes to variations in in zero valent iron performance (Sun et al., 2016). A filter material's specific surface area is a crucial factor since it affects the number of adsorption sites and how accessible contaminants are to the adsorbent. In comparison to iron wool or iron nails, iron filings have a larger surface area.

Increased surface area of the materials indicates more iron particles are available for corrosion, and more corrosion products are available for As sorption (Bretzler et al., 2020). According to previous studies, as the surface area of iron increases, the removal rate of As increases (Gillham, 1994; Liang et al., 2022). The smaller the iron particle size, the more its reactivity with As, and hence the higher the removal rate. Thus, the particle size has a significant role in the adsorption and interaction with contaminants (Liang et al., 2022; Wenk et al., 2014). However, some other studies suggest that the removal of arsenic is not only dependent upon surface area but also takes into account the composition, surface characteristics, mechanical abrasion, and kind of oxide coating being generated (Sista et al., 2021). This supports what was observed in this study where the performance of iron wool and iron filings could not be defined based on surface area only.

In addition to the parameters mentioned above, environmental factors like temperature and pH may also have an impact on the removal of arsenic. For example, a study was carried out to explore the effect of

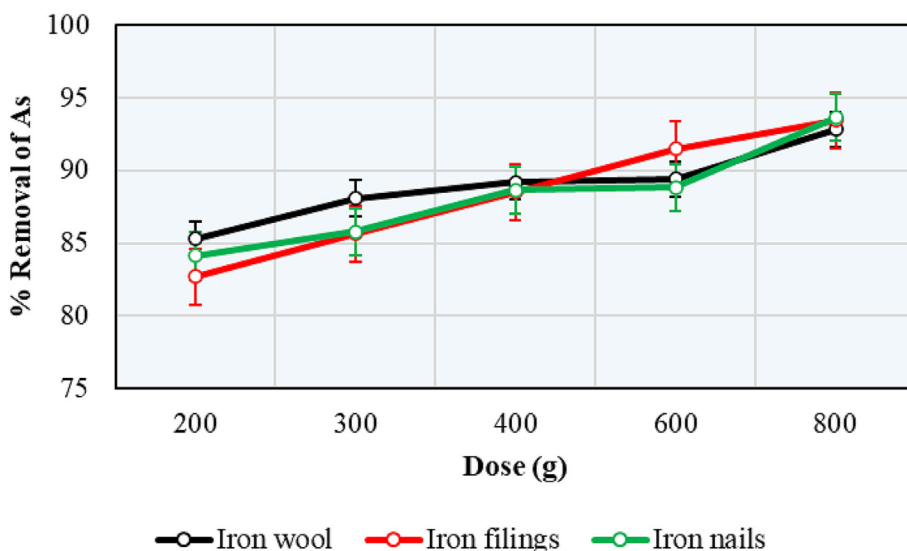


Fig. 5. Effect of adsorbent dose on removal of arsenic (6 h).

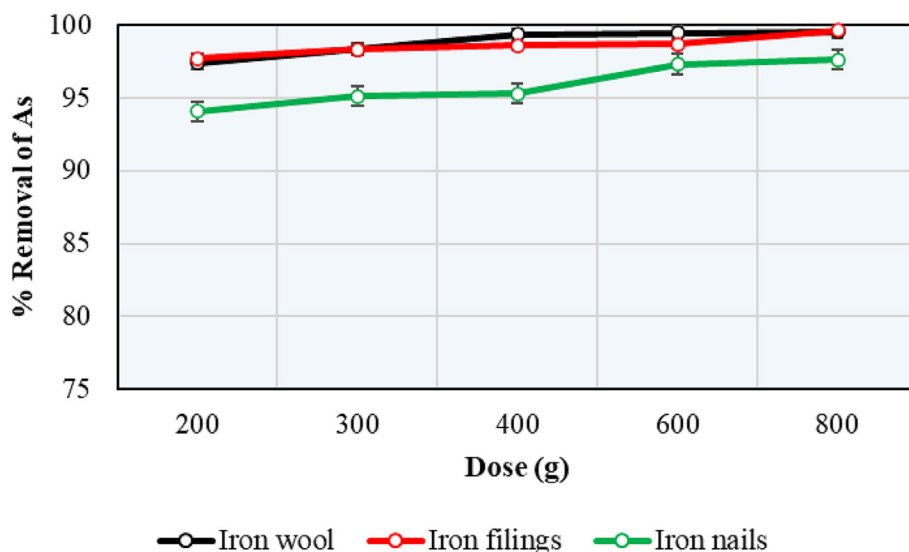


Fig. 6. Effect of adsorbent dose on removal of arsenic (72 h).

environmental factors such as temperature on the removal of arsenic from aqueous solution utilizing an ionic liquid and nano-iron (H/Fe) composite. The findings of this investigation indicated that as the temperature rises, the Arsenic removal efficiency of H/Fe rises while the temperature is below 40 °C and progressively drops as the temperature rises over 40 °C. This study proposed 40 °C as the optimal experimental adsorption setting for H/Fe (Wu et al., 2021). In another study the effect of pH on arsenic removal using a waste iron column with and without iron bacteria in continuous and batch phases was investigated. This study recommended the pH of 7 as the optimal pH for removal of Arsenic (Azhdarpoor et al., 2015a). The pH of 7 was also reported by other researchers as the optimal value for removal of Arsenic (Ahmed et al., 2021; Biterna et al., 2010; Farrell et al., 2001).

5. Conclusion

In this study, the performance of several kinds of zero valent iron (Fe⁰), such as iron wool, iron filings, and iron nails, was compared in terms of removing As from aqueous solution. The results of this experiment described that the polluted water containing arsenic can be remediated by using sand filters containing zero valent iron. The results shows that iron wool performed better in many experiments than iron filings and iron nails. Increasing the adsorbent dose increased the adsorption capacity of adsorbent and the removal efficiency of As. The early adsorption response is quick in all columns, but later reaction involves slow adsorption, producing a clear two-phase adsorption characteristic. The findings shows that arsenic can effectively be removed from water in the first 48 h. These findings demonstrate that locally available materials can be used as zero valent iron for arsenic remediation in polluted water. Because all of the materials employed demonstrated features such as high absorption capacity, effective removal efficiency, cost-effectiveness, and availability, the use of materials can be considered in application for removal of arsenic from aqueous solution.

CRediT authorship contribution statement

Daniel Edward: Conceptualization, Formal analysis, Resources, Investigation, Methodology, Writing – original draft. **Petro Karungamy:** Conceptualization, Formal analysis, Investigation, Methodology, Writing – review & editing. **Grite Nelson:** Conceptualization, Methodology, Writing – review & editing, Supervision. **Juma Selemani:** Conceptualization, Methodology, Resources, Writing – review & editing,

Supervision. **Karoli N. Njau:** Methodology, Resources, Writing – review & editing, Supervision.

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