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The removal of arsenic from synthetic solution using a sand filter coupled with zero valent iron

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THE REMOVAL OF ARSENIC FROM SYNTHETIC SOLUTION USING A SAND FILTER COUPLED WITH ZERO VALENT IRON

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A Dissertation Submitted in Partial Fulfillment of the Requirements for the Degree of Master of Science in Environmental Science and Engineering of the Nelson Mandela African Institution of Science and Technology

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ABSTRACT

Contamination of groundwater with arsenic (As) poses a serious risk to public health, demanding the development of effective remediation technology. This study investigated the use of sand filters coupled with of zero-valent iron (ZVI) for removal of arsenic from synthetic solutions as the function of ZVI dosage and contact time. ZVI materials, including iron wool, iron fillings, and iron nails, were investigated and compared for their efficacy in removing As from synthetic solutions. The experiment employed synthetic solutions spiked with As compounds (set to be 1000 µg/L) to simulate contaminated groundwater scenarios. The concentrations of As in the influent and effluent samples were used to calculate the efficiency. The findings indicate that all three forms of ZVI tested exhibited significant As removal capabilities. Iron wool, iron fillings, and iron nails demonstrated varying efficiencies, likely influenced by their surface area, reactivity, and specific surface chemistry. According to the findings, iron wool-containing sand filter was more effective at removing As than iron filings and iron nails. As removal efficiency increased with increase in ZVI dose. The results show that As can effectively be removed from water in the first 48h. The highest removal efficiency was 99.6% and the lowest removal efficiency was 82.7%. These results suggest that ZVI materials could be promising candidates for practical applications in As-contaminated groundwater treatment systems. This study contributes valuable insights into the use of inexpensive and widely available ZVI materials for As remediation, highlighting their potential as sustainable solutions for addressing water quality challenges.

DECLARATION

I, Daniel Edward, do hereby declare to the Senate of Nelson Mandela African Institution of Science and Technology that this dissertation is my original work and it has neither been submitted nor being concurrently submitted for degree award in any other institution.

Daniel Edward **Date 14.08.2024**

Name and signature of the candidate

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Prof. Karoli N. Njau **Date 14.08.2024**

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Selemani

Dr. Juma Rajabu Selemani **Date 14.08.2024**

Name and signature of supervisor 2

Dr. Grite Nelson **Date 14.08.2024**

Name and signature of supervisor 3

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CERTIFICATION

The undersigned certify that they have read and found that the dissertation conforms to the standard and format acceptable for submission. Therefore, do hereby recommend for acceptance of dissertation entitled “***The removal of arsenic from synthetic solution using a sand filter coupled with zero valent iron***”, in partial fulfillment of the requirements for the award of the Degree of Master’s in Environmental Science and Engineering of the Nelson Mandela African Institution of Science and Technology.

Prof. Karoli N. Njau **Date 14.08.2024**

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DEDICATION

I would like to dedicate my work to my wife Ms. Getruda Nicholaus Kidesheni, my children Daniella Daniel, Daniani Daniel, Dagna Daniel, Danikiel Daniel and Damiani Daniel. They imparted in me a wish to learn and made ransoms so I would have access to Master degree. Also, this is dedicated to my close friends Atugonza Majura, Aziza Konyo, Alpha Mangula, Erick Edward, Petro Karungamye, Jumanne Shimba, JohnBosco Karungamye and Munira Kaoneka who always discussed together and supported each other throughout my years of studies.

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LIST OF ABBREVIATIONS

AAS	: Atomic Absorption Spectroscopy
As	: Arsenic
ATSDR	: Agency for Toxic Substances and Disease Registry
DNA	: Deoxyribonucleic Acid
MCL	: Maximum Contaminant Level
NM-AIST	: Nelson Mandela African Institution of Science and Technology
SD	: Standard deviation
SDG	: Sustainable Development Goals
US EPA	: United States Environmental Protection Agency
WHO	: World Health Organization
XRF	: X-ray Fluorescence
ZVI	: Zero Valent Iron

CHAPTER ONE
INTRODUCTION

1.1 General 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 (As) (Richards et al., 2022; Shankar et al., 2014). Due to occupational or domestic exposure, As 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 which need proper handling (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 (Costa, 2019).

The name "arsenic" is derived from the Greek word "arsenikon" (ἄρσενοκόν), which means yellow orpiment. Orpiment itself is a yellow arsenic sulfide mineral (As_2S_3), and it was known and used in ancient times as a pigment and for medicinal purposes (Ahmed, 2022; Paul et al., 2023). Its sources include both anthropogenic and natural geochemical activities including mining, the release of industrial waste, and fertilizer use in agriculture (Patel et al., 2023; Selinus et al., 2013). 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 (Zheng et al., 2017). As(V) is found as oxyanions of arsenic acid, whereas As(III) is found as arsenious acid (Zawierucha et al., 2022).

In comparison arsenite is 20-60 times more toxic than arsenate, with toxicity depending on oxidation-reduction states and methylation levels during metabolism (Chen et al., 2018; Vromman et al., 2018). Arsenite is also more mobile than arsenate and thermodynamically more stable in reducing groundwater conditions (Kumari & Jagadevan, 2016; Li et al., 2020). The World Health Organization (WHO) recommends that As levels in drinking water be less than 10mg/L (WHO, 2011). However, even at concentrations below 10 mg/L, As can have significant consequences for human health (Gude et al., 2018). The development of treatment technologies for the removal of As at the household or small community level is therefore necessary.

As can be removed from aqueous solutions using a wide range of physicochemical techniques (Morgada et al., 2009) includeing coagulation and flocculation, membrane techniques, ion-exchange or chelation, chemical precipitation, and activated carbon adsorption (Fu & Wang, 2011; Qasem et al., 2021). 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 As ions at the household level due to its simplicity in designing, convenience of use, and cheap operating costs (Kim et al., 2022). Different materials have been investigated regarding adsorption of As including rice polish, zeolite, red mud, activated alumina, surface-modified carbon black, iron hydroxides and oxides, open-celled cellulose sponge (Keerio et al., 2021). Systems using sand filters are frequently utilized to treat wastewater on-site and in small communities all over the world. Compared to other natural technologies, it requires less land and doesn't need energy or chemicals (Yettefti et al., 2013). As 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 $Fe^{3+}(aq)$, 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 & Scott, 2012; Yang et al., 2015).

Over the past decade, zero-valent iron (ZVI) 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). ZVI 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). Despite the fact that various studies have explored the effectiveness of various forms of ZVI 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 aim of this research was to compare the effectiveness of iron filings, iron nails, and iron wool as ZVI for the removal of As from synthetic solution. This research is in line with Sustainable Development Goals (SDGs) related to reduction of As from drinking water. These includes SDG 3: Good Health and Well-being, SDG 6: Clean Water and Sanitation and SDG 15: Life on Land.

1.2 Statement of the Problem

The presence of As in drinking water poses a severe health risk to millions of people globally. In Tanzania for instance, high concentrations of As above the WHO guideline (10µg/L) has been reported in Lake Victoria Gold fields (Ijumulana et al., 2018; Irunde et al., 2016). Around 86.53% agricultural soils in Usangu agroecosystem, have As concentration above maximum allowable limit (Mng'ong'o et al., 2021). As contamination in groundwater primarily originates from natural geological sources, exacerbated by human activities such as mining and industrial processes. In an attempt to minimize the level of As contamination, a variety of methods including coagulation/filtration, reverse osmosis, ion exchange, adsorption, and biological treatment have been investigated. While most of these methods are effective on removal of As, their application in poor communities especially in developing countries, is affected by high initial and operational costs, technological feasibility, low local adaptability, skills requirements, energy requirements, maintenance requirements, community acceptance and awareness (Alka et al., 2021; German et al., 2019). ZVI has become an effective remediation tool for As contamination due to its chemical reduction capabilities. This involves converting soluble As species into less toxic precipitates, thereby reducing their mobility and bioavailability in the environment. Various forms of ZVI, including iron fillings, iron nails, and iron wool, have been studied for their efficiency in As removal, with each having unique characteristics and performance under different conditions. This makes it essential to understand the performance of different forms of ZVI under similar environments in order to optimize its application in actual As remediation efforts. Therefore, the aim of this research is to compare the effectiveness of iron wool, iron filings, and iron nails as ZVI for the removal of As from synthetic solution. It is expected that the results of this study will provide significant insights on the use of ZVI for As remediation, especially in resource-constrained places where accessible and costs-effective technologies are essential. This study intends to provide practical solutions for reducing As pollution in drinking water by comparing the performance of wool, nails, and iron fillings. This will improve public health and environmental sustainability.

1.3 Justification of the study

As-related water contamination has been addressed through various kinds of established and recommended interventions which may be classified broadly into two categories. The first approach is to remove As from groundwater in order to make water with little or no As available. The second approach is to provide access to a sustainable and easily available alternative clean water source. The majority of As removal techniques are expensive to execute complicated, and impractical particularly in developing countries like Tanzania. Over the past 20 years, zero valent iron has been used for groundwater As removal because of its affordability, ease of availability, and strong affinity for As. It is worthwhile to do this research comparing the performance of iron fillings, iron nails, and iron wool as zero valent iron for removal of As from synthetic solution. These materials were chosen as a Fe-source because they are affordable, accessible worldwide, including in Tanzania, and do not require extensive cleaning.

1.4 Research Objectives

1.4.1 General Objective

The general objective of this research is to compare the effectiveness of iron filings, iron nails, and iron wool as ZVI for the removal of As from synthetic solution.

1.4.2 Specific Objectives

i) To characterize the composition of different forms of ZVI such as iron fillings, iron nails, and iron wool for removal of As from synthetic solution.

ii) To assess the influence of iron fillings, iron nails, and iron wool as forms of ZVI incorporated in sand filter in the removal of As from synthetic solution.

1.5 Research Questions

i) What are the characteristics of different forms of ZVI such as iron fillings, iron nails, and iron wool?

ii) How does different forms of ZVI incorporated in sand filter influence the removal of As from synthetic solution?

1.6 Significance of the Study

This study aims to improve the understanding and application of sustainable methods for removing arsenic from contaminated groundwater, which poses a severe risk to human health. The findings are particularly relevant to Tanzanian communities in the Lake Victoria basin, where over 30% of water sources have arsenic levels exceeding the WHO guideline of 10 µg/L. The contamination is particularly prevalent in regions surrounding the North Mara gold mine and Geita mining areas. The study identifies the most effective form of ZVI for arsenic removal, providing insights into cost-effective and sustainable remediation strategies. This knowledge can inform policy-making, guide community-level interventions, and support efforts to provide safe and clean water to vulnerable populations in Tanzania and beyond.

CHAPTER TWO

LITERATURE REVIEW

2.1 Chemistry and occurrence of arsenic

Arsenic is an interesting element because of its complex behavior and potential negative health effects (Bagherifam et al., 2019). Its mobility and bioavailability in diverse environments are greatly influenced by its existence in a variety of chemical forms as well as its interactions with environmental variables (Kumari et al., 2017). This variation further emphasizes how crucial it is to investigate As from the viewpoints of the ecosystem and human health. As can enter the environment through both natural and anthropogenic sources as summarised in **Figure 1**.

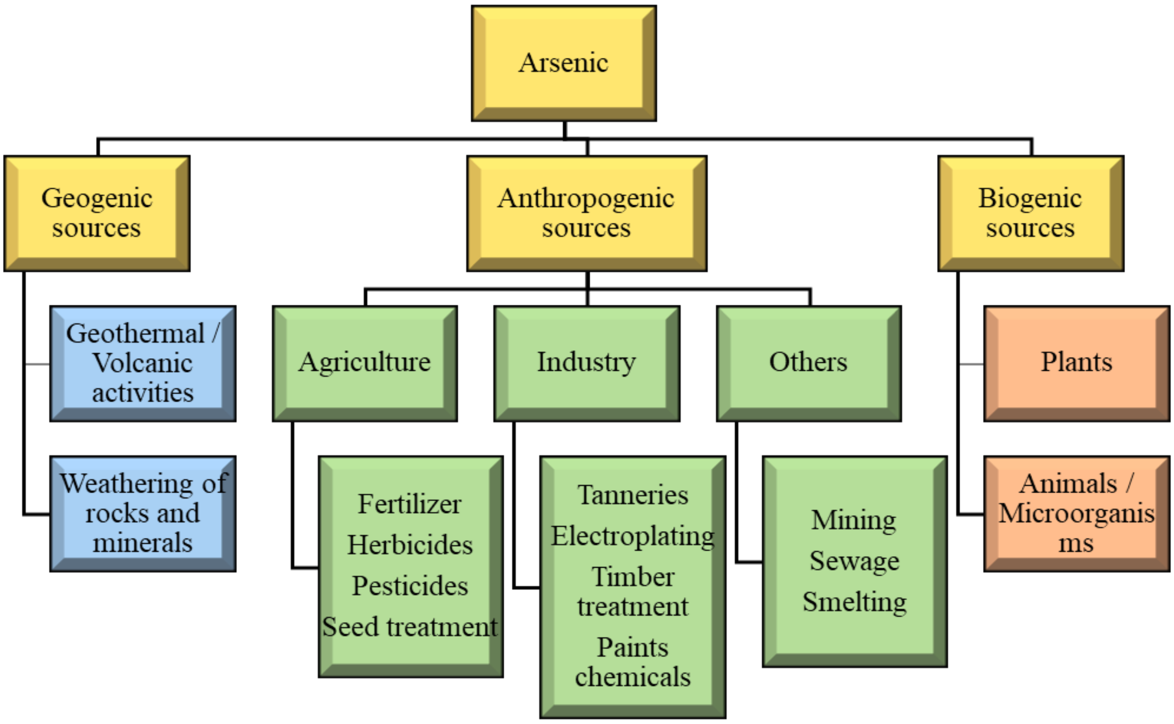


Figure 1: Natural and anthropogenic sources of arsenic (Hare et al., 2018)

2.1.1 Physical and Chemical Properties

As is a distinctive silver-grey crystalline solid with an atomic number of 33 and an atomic weight of 74.9 (Mahamallik & Swain, 2023). It is particularly dense, with a specific gravity of 5.73. Its boiling point is 613 °C, and it has a vapor pressure of 1 mmHg at 372 °C (Fatoki et al., 2013; Fu et al., 2014; Quino-Favero et al., 2021). Under extreme pressure (28 atm), As melts at 817 °C, yet it typically sublimes at 616 °C under normal atmospheric pressure, bypassing the liquid state (Fowler et al., 2015; Zheng et al., 2017). In the absence of oxygen, As undergoes sublimation to yield a yellow gas; when heated in an oxygen-rich environment, it transforms into As trioxide (As_2O_3) (Hassan & Westerhoff, 2024; Paul et al., 2023). Positioned in Group VA of the Periodic Table, As is recognized as a metalloid, bridging characteristics between metals and nonmetals (Jang et al., 2016; Raju, 2022). Its chemical behavior is influenced by this intermediate nature, allowing it to form a range of compounds with diverse properties (Chellan & Sadler, 2015). These properties have been presented in **Table 1**.

Table 1: Physical and Chemical Properties of Arsenic and Selected Inorganic Arsenic Compounds (Agency for Toxic Substance and Disease Registry (ATSDR), 2007)

Property	**Arsenic**	**Arsenic acid**	**Arsenic pentoxide**	**Arsenic trioxide**
Molecular weight	74.9216	141.944	229.840	197.841
Color	Silver-gray or tin-white	White	White	White
Physical state	Solid	Solid	Solid	Solid
Melting point	817 °C (triple point)	35 °C	Decomposes at ~300 °C	313 °C (claudetite) 274 °C (arsenolite)
Boiling point	614 °C sublimes	Loses H_2O at 160 °C	No data	460 °C
Density	5.778 g/cm^3 at 25 °C	~2.2 g/cm^3	4.32 g/cm^3	3.865 g/cm^3 (cubes), 4.15 g/cm^3 (rhombic crystals)
Solubility in Water	Insoluble	302 g/L at 12.5 °C	2,300 g/L at 20 °C	17 g/L at 16 °C
Vapor pressure	7.5×10^{-3} mmHg at 280 °C	No data	No data	2.47×10^{-4} mmHg at 25 °C

2.1.2 Environmental Mobility and Redox Chemistry

The mobility of As in the environment is governed by pH, redox potential (Eh), and the presence of competing ions. Its behavior is closely linked to redox processes involving iron and sulfur (Al-Abed et al., 2007; Bertin et al., 2022; Demergasso et al., 2007). As exhibits various oxidation states, including -3, 0, +3, and +5 (ALSamman et al., 2023; Kapp, 2018). Some of the common arsenic compounds found in speciation analysis are presented in **Table 2**.

Table 2: Some of the common arsenic compounds found in speciation analysis (Mana & Fatt, 2017)

Name	Abbreviation	Chemical formula
Inorganic compounds		
Arsenite (arsenous acid)	As(III)	$As(OH)_3$
Arsenate (arsenic acid)	As(V)	$AsO(OH)_3$
Organic compounds		
Monomethylarsonous acid	MMAIII	$CH_3As(OH)_2$
Monomethylarsonic acid	MMAV	$CH_3AsO(OH)_2$
Dimethylarsinous acid	DMAIII	$(CH_3)_2AsOH$
Dimethylarsinic acid	DMAV	$(CH_3)_2AsO(OH)$
Arsenonetaine	AsB	$(CH_3)_3As^+CH_2COO^-$
Arsenocholine	AsC	$(CH_3)_3As+CH_2COO^-$

In aquatic environments, arsenite (As(III)) and arsenate (As(V)) are indeed the most common forms of As (Rodríguez-Martín et al., 2022). These two oxidation states, As(III) and As(V), play critical roles in the biogeochemical cycling and toxicity of As in natural waters (Izaditame et al., 2024; Muzaffar et al., 2023). Arsenite (As(III)) exists predominantly in reducing environments such as groundwater and wetlands (Stuyfzand & Bonte, 2023). It is highly toxic and more mobile than arsenate, making it a significant concern for human health when present in drinking water (Bhagat et al., 2022; Etesami et al., 2023). In its trivalent form, arsenic tends to form complexes with sulfhydryl groups of proteins, interfering with cellular functions and causing oxidative stress (Sevak & Pushkar, 2024; Virk et al., 2023). On the other hand, arsenate (As(V)) is more prevalent in oxidizing conditions, such as surface waters and aerobic soils (Fathi-Gerdelidani et al., 2022; Mawia et al., 2021). It is less toxic than arsenite but still poses health risks (Fathi-Gerdelidani et al., 2022). Arsenate mimics phosphate structurally, allowing it to be taken up by organisms through phosphate transport mechanisms (Garbinski et al., 2019). This similarity can lead to arsenic accumulation in aquatic organisms, posing risks to ecosystems and human consumption through the food chain (Bridges & Zalups, 2005; Urrialde et al., 2017; W. Zhang et al., 2022).

The interconversion between arsenite and arsenate is influenced by redox conditions in the environment (Deewan et al., 2022; Vromman et al., 2018). Under reducing conditions, arsenate can be reduced to arsenite, increasing its mobility and toxicity (Bhagat et al., 2022). Conversely, under oxidizing conditions, arsenite can be oxidized to arsenate, reducing its mobility but maintaining its potential health risks (Fowler et al., 2015; Pintor et al., 2018;

Rodríguez-Martín et al., 2022). To determine environmental impacts of As and establish effective mitigation strategies, it is essential to understand its speciation and dispersion in aquatic environments.

2.1.3 Arsenic-Microorganisms Interactions

Microorganisms play a crucial role in the biogeochemical cycling of As (Izaditame et al., 2024; Xie et al., 2024). They have developed various mechanisms to cope with high concentrations of As, including sorption, mobilization, precipitation, redox reactions, and methylation (Sevak & Pushkar, 2024; Soares et al., 2024). The microbial resistance to As involves a detoxification system where arsenate (As(V)) is reduced to arsenite (As(III)) by cytoplasmic arsenate reductase and subsequently extruded from the cell via a membrane-bound As(III) efflux pump (ars system) (Crognale et al., 2017; Kumari & Jagadevan, 2016). This microbial activity significantly influences the speciation, mobility, and toxicity of As in natural environments (Ding et al., 2024; Izaditame et al., 2024; Luo et al., 2024). The mechanisms of microbial transformations of As in the environment are shown in **Figure 2**.

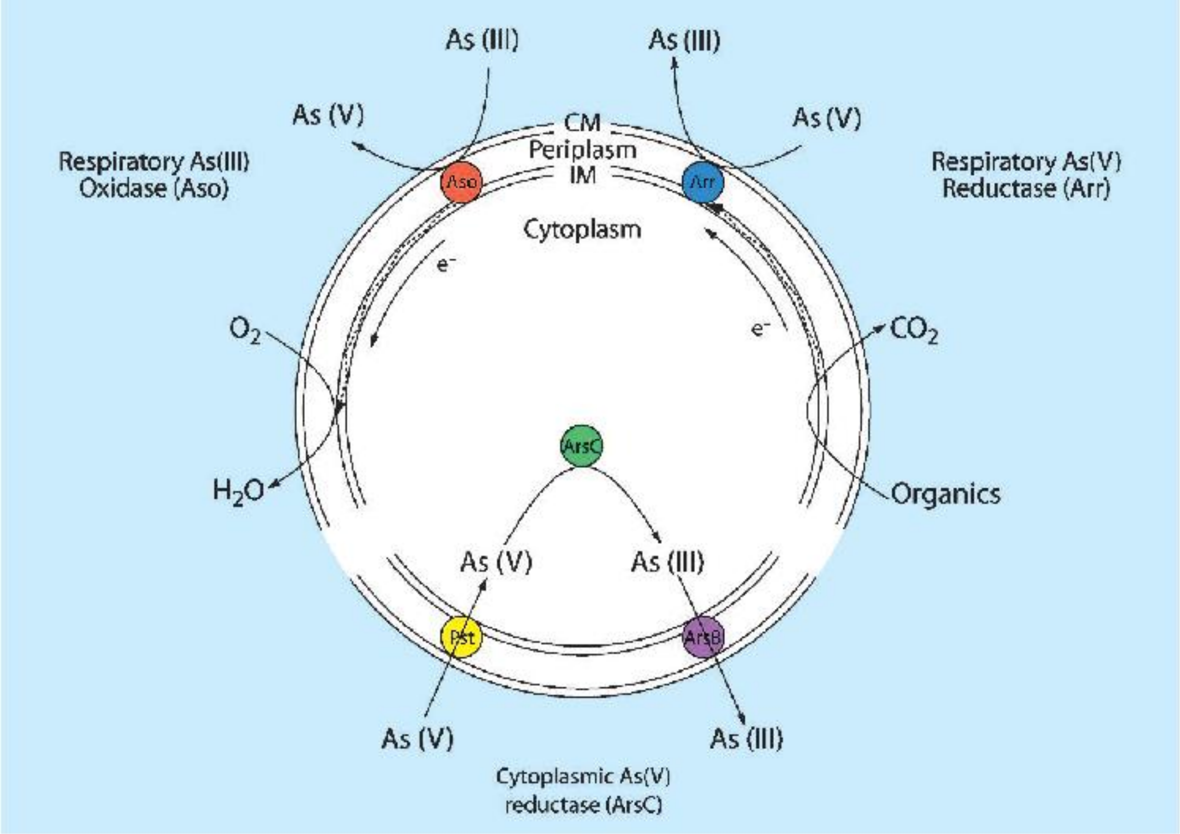


Figure 2: Mechanisms of microbial transformations of As in the environment (Lloyd & Oremland, 2006).

2.2 Arsenic induced toxicity

Humans can become exposed to As through inhaling, consuming, or absorbing through their skin (Ramasamy & Lee, 2015). However, exposure to As primarily occurs from drinking water that has been contaminated with the arsenic (Huang et al., 2015). As in drinking water has been identified as a major public health issue in a number of countries throughout the world (Mazumder, 2015). As a result, the WHO considers that As contamination in the drinking water poses the most significant hazard to human health (Quino-Favero et al., 2021). The WHO has set the maximum allowable limit for As in drinking water at 10 µg/L (Muzaffar et al., 2023; WHO, 2011). The maximum contamination level (MCL) is the highest level of a contaminant that is permitted in drinking water based on a cost-benefit analysis and is enforceable. The maximum contaminant level goal (MCLG) is a non-enforceable level at which harmful effects on human health are unknown or not anticipated to occur while yet providing an appropriate margin of safety. On January 22, 2001, the US Environmental Protection Agency (EPA) published an MCL for arsenic of 10 µg/L and an MCLG of Zero (EPA, 2001). The MCL for As was established at a level where the advantages of reducing lung and bladder cancer were maximized at a cost that was justified by the benefits (Ramasamy & Lee, 2015). The maximum allowable contaminant level (MCL) for total As of different regulatory authorities are presented in **Table 3**.

Table 3: Maximum allowable contaminant level (MCL) for total As of different regulatory authorities

Authority/Country	MCL, µg/L	Reference
WHO	10	(WHO, 2011)
Australia	7	(NHMRC, 2011)
US EPA	10	(EPA, 2001)
European Community (EC)	10	(EU, 2020)
Canada	10	(HC, 1984)
Bangladesh, China, Mexico, Taiwan, Vietnam, etc.	50	(Nordstrom, 2002)

As can enter the food chain through agricultural practices, water sources, and environmental factors like overexploitation of arsenic-contaminated groundwater and mining activities (Majumder & Banik, 2019; Mitra et al., 2017). Organic arsenic which is the form mostly found in foods is generally considered less toxic than their inorganic counterparts (Guillod-magnin et al., 2018). As in seafood forinstance, mainly occurs as organic As with low toxicity, and it does not pose any risk to human health (Akter et al., 2006; Taylor et al., 2017).

Inorganic As can enter the food chain from sources like groundwater contamination, industrial effluent, and drainage problems, causing wide spread distribution throughout plant and animals (Upadhyay et al., 2019; Zhang et al., 2021). The bioavailability and health impact of As in food depend on several factors including Chemical Form of As, food Source, cooking and processing methods, individual characteristics (age, nutritional status, and overall health), geographical and environmental factors (Khan et al., 2022; S. Singh et al., 2023). It has been projected that 94 million to 220 million individuals globally may be exposed to unacceptable higher levels of groundwater arsenic (Irshad et al., 2021; Podgorski & Berg, 2020). **Figure 3** shows human exposure to As from food chain

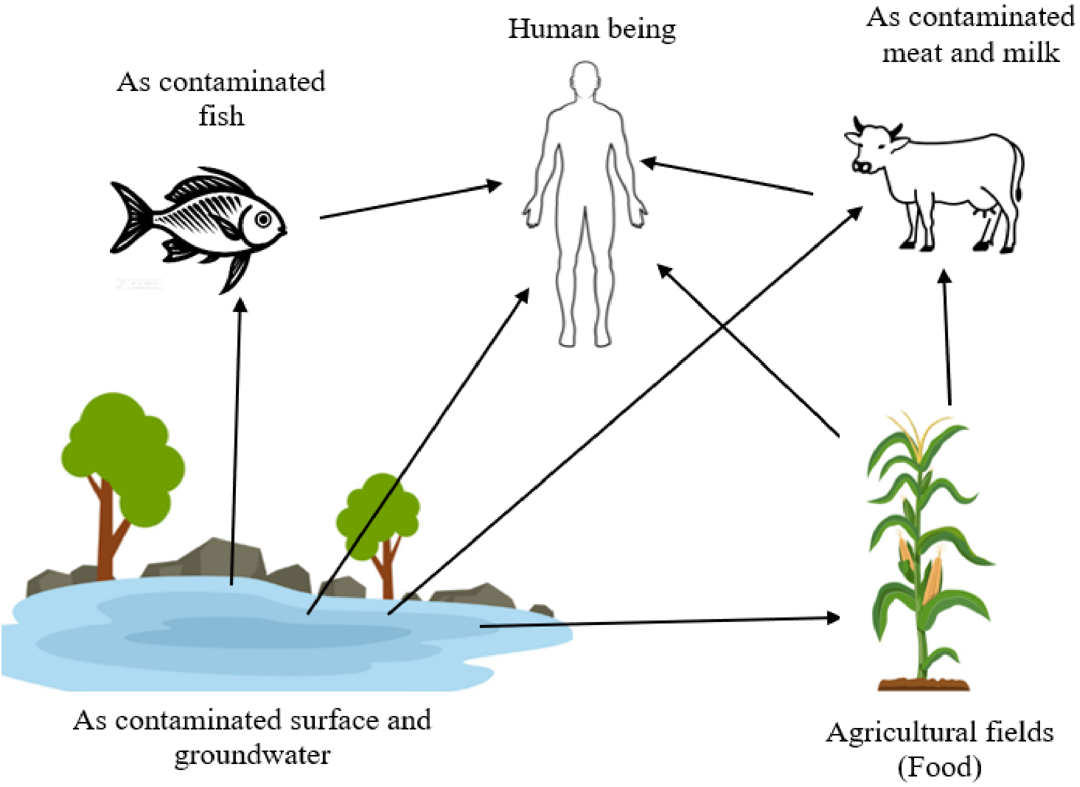


Figure 3: Human exposure to As from food chain (Source: Author)

The toxicity of arsenic is determined by its chemical speciation, with inorganic arsenic being more toxic than organo-arsenicals (Thakur et al., 2021). The toxicity of As compounds causes the production of free radicals, which can induce DNA single-strand breaking. A considerable decrease in antioxidants caused by As exposure causes oxidative stress in various organs (Banu et al., 2018; Ghosh & Sil, 2015). As toxicity, both acute and chronic, can have detrimental consequences in organs and tissues, such as peripheral neuropathy, cardiovascular disorders, pulmonary illnesses, hyperkeratosis, and changes in skin color. It can also affect cognitive and developmental processes (Sanyal et al., 2020; Singh et al., 2021).

In humans, severe acute As toxicity can cause gastrointestinal distress, vomiting, diarrhea, bloody urine, anuria, shock, seizures, coma, and potentially death (Ratnaike, 2003). Chronic As exposure through drinking water has been linked to a number of diseases, including diabetes mellitus, peripheral vascular disease, cardiovascular disease, cerebrovascular illness, nervous system disease, and different types of cancer (Huang et al., 2015). Given that the toxicity of As has been linked to its speciation (such as inorganic trivalent/pentavalent forms), more research on As speciation needs to be done than on general As. The major human organs and systems affected by chronic As exposure are shown in **Figure 4.**

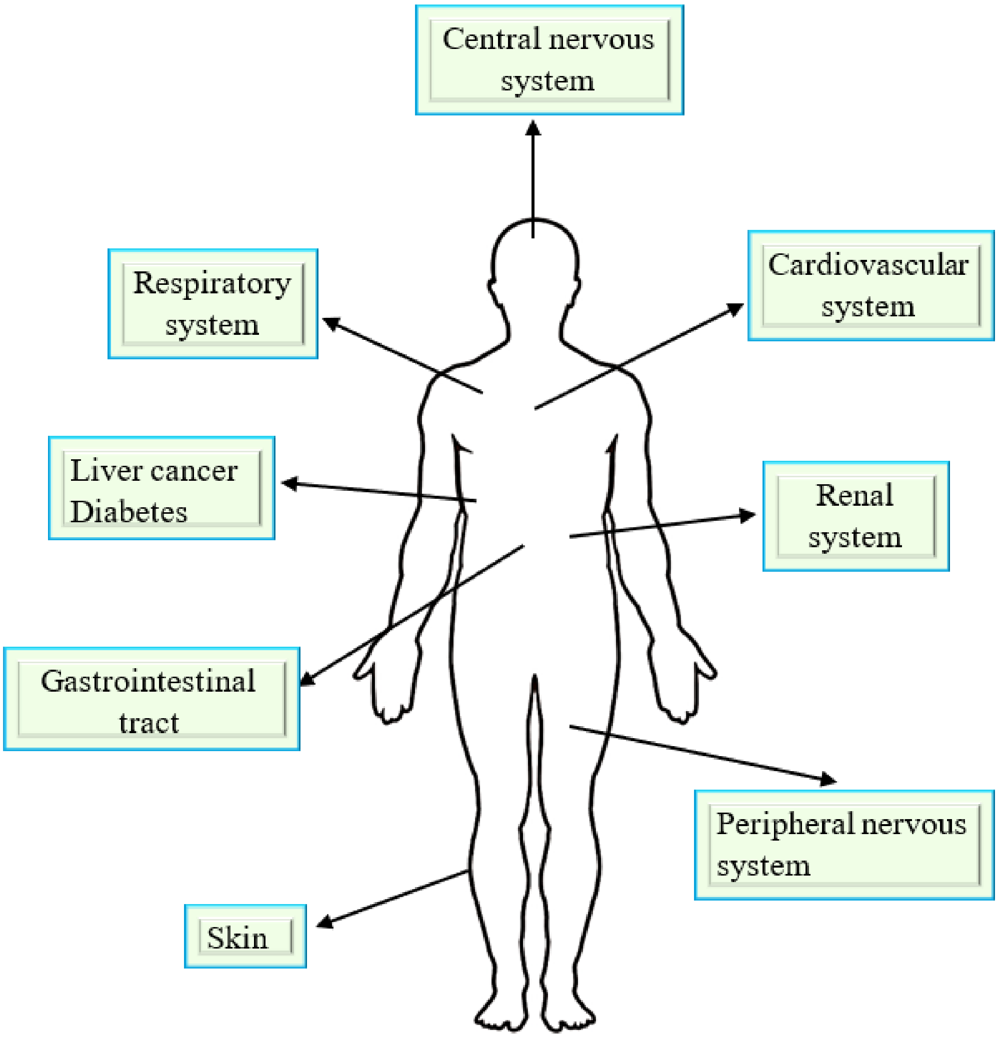


Figure 4: Human organs and systems affected by chronic arsenic poisoning. Modified from (Rahaman et al., 2021).

The signs and symptoms of As exposure appear to vary between individuals, demographic groupings, and geographic regions (Folesani et al., 2022; Khosravi-Darani et al., 2022). Therefore, there is no accepted definition of the disease caused by As. This complicates determining the extent to which As is harmful to human health. Moreover, there is no method to distinguish tumors induced by As from those produced by other causes in internal instances of cancer (Rahidul, 2023). As toxicity has no recognized effective medication for treatment, however, drinking water free of arsenic can help those who are affected by As get rid of its symptoms (Evens et al., 2004; Khosravi-Darani et al., 2022; Nurchi et al., 2020). As a result, As should be considered a top priority for toxicological research, including toxicokinetics, toxicodynamics, and mode of toxic action. The legislative bodies need to prioritize this matter and allocate sufficient funding for this kind of research.

2.3 Technologies for arsenic removal

As removal from aqueous solutions has been made possible by a wide range of physicochemical techniques (Nicomel et al., 2015). The selection of an As removal technology for a particular area depends on a number of factors, including the degree of oxidation of As and the pH of the groundwater (Rahidul, 2023). As removal technologies includes precipitation, membrane separation, adsorption, ion exchange, and biotechnology (Chowdhury et al., 2019; Feroze Ahmed, 2001). The advantages and disadvantages of each of these technologies, especially with regard to cost and efficacy, dictate which treatment is used (Rahidul, 2023). These methods require a lot of energy and/or chemicals, which makes the processes of treatment expensive. Additionally, they are difficult to operate and maintain, necessitating regular part replacement, costly after-market services, and specialized people who may not be available locally. As a result, in disadvantaged populations, these technologies are not long-term viable (Annaduzzaman et al., 2021). These methods have been presented in **Table 4**.

Arsenic can be removed by precipitation methods as ferric arsenate, calcium arsenate, or arsenic sulfide. Ferric salts have been shown to be more effective than aluminum salts for precipitation (Mohan & Pittman, 2007). Using chemical precipitation to achieve As concentrations below 10 µg/L is often challenging. Additionally, removing As(III) during precipitation is notably less effective compared to removing As(V) anions. To address this, pre-oxidation is necessary to convert As(III) to As(V) ions in water (Sarwar et al., 2015; Sawana et al., 2017).

Table 4: Advantages and disdvantages of As removal methods (Rahidul, 2023)**.**

Method	Advantages	Disadvantages
Ion Exchange	Clearly defines the medium and its capacity. Less susceptible to fluctuations in water pH. Utilizes specific ion resins for targeted ion removal.	Expensive medium and operational costs. Requires sophisticated maintenance and operation. Challenges with managing sludge from resin regeneration.
Adsorption	Well-established technology with commercially available materials. High efficiency and simplicity in operation. Cost-effective with no additional chemicals or byproducts.	Adsorption beds have a limited lifespan and need regular replenishment. Generates solid waste that may be hazardous. Maintenance of the adsorbent material can be labor-intensive.
Chemical Precipitation	Simple to handle with common, inexpensive chemicals. Relatively low initial capital investment. Effective for a wide range of contaminants.	Produces toxic sludge that requires proper disposal. May necessitate pre-oxidation processes which can create harmful disinfection byproducts. The process can be inefficient for certain contaminants.
Membrane Technique	No hazardous solid waste is generated. Effective at removing bacteria and various pollutants. Eliminates the need for additional chemicals.	High maintenance and operational costs. Potential for hazardous wastewater. Often requires pre-treatment of the water to prevent fouling and extend membrane life.
Electrocoagulation	Provides an alternative to chemical precipitation. No chemicals are required. Cost-effective and relatively easy to maintain. Innovative approach with growing potential.	Requires a robust system foundation. Not yet widely commercially available. Successful implementation depends on effective design and operational management.
Phytoremediation	Environmentally friendly and sustainable. No chemicals are needed, making it a natural remediation method. Can be aesthetically pleasing and offer additional ecological benefits.	Requires a strong system foundation for implementation. Not widely available for purchase or adoption. The effectiveness can be variable depending on plant species and environmental conditions.

Ion exchange technology, which uses anion exchange resins to remove As from water, is considered to be a viable option (Laatikainen et al., 2016). The uncharged As(III) species in water cannot be effectively removed with this method; it is only effective for As(V). In addition, it is typically expensive to design high-tech water filtration systems and ion exchange resin. The process of adsorbent regeneration also generates a sludge disposal issue.

As removal from water has been reported more frequently in recent years using membrane techniques, such as reverse osmosis and nanofiltration (Bahmani et al., 2017).

These methods have several advantages such as high removal efficiency, simplicity of use, and low production of polluting sludge (Li et al., 2017). The initial investment and operating costs are high, and significant pressure is needed to push contaminated water through membranes. Additionally, issues such as concentrate discharge, membrane fouling, and flux reduction are often unavoidable in the membrane process (Kapepula & Luis, 2024; Osman et al., 2024). As and other pollutants can be removed by electro-dialysis, although a significant amount of insoluble coagulants are also deposited on the cathode (Song et al., 2017).

The adsorption process is one of the numerous methods for removing As from water that are now accessible. Due to its low cost, high efficiency, and simplicity of usage, it is seen to be one of the most promising methods (Chowdhury, 2017). Iron-based adsorbents have been considerably researched and demonstrated good efficacy in removing for As compounds from water (Hao et al., 2018). Some commercial adsorbents, such as granular ferric hydroxide (GFH) and ZVI, have been produced on a large scale (Pintor et al., 2018). Despite their demonstrated high efficacy in removing As, the majority of described adsorbents rarely find use in the field due to the interfering ions that are present in the water (Nguyen et al., 2023). It has been determined that phosphate can actively compete with As for adsorption sites, lowering arsenic adsorption capability (Lalley et al., 2016). Organic matter, such as humic and fulvic acid, can potentially have negative effects on the adsorption of As by altering the adsorption equilibrium (Fakour et al., 2015). The removal efficiencies that are often reported are summarized in **Table 5** .

Table 5: Efficiency of conventional arsenic removal techniques (Duarte et al., 2009).

Technology	Chemical reagent	Ideal conditions	% Removal As(III)	% Removal As(V)
Adsorption	Activated carbon or activated alumina	pH 5.5 - 6	30 - 60	> 95
Adsorption	Iron hydroxide	pH near 8	30 - 60	> 95
Coagulation – filtration precipitation	Ferric Chloride	pH 6 – 8	< 30	90 - 94
Coagulation – filtration precipitation	Sulphates (Al, Cu, NH_3)	pH 6 – 6.5	< 30	80 - 90
Ion exchange	Anionic resins	$SO_4^{2-} < 20$ mg/L	< 30	> 95
Membrane filtration		Presence of dissolved As	60 – 90	> 95

2.4 Arsenic removal using ZVI

ZVI has received a lot of attention for its ability to remove organic and inorganic pollutants from water (Biterna et al., 2010). The use of ZVI for pollutant removal from water is due to its low cost, non-toxicity, rapid response, and great pollutant removal capabilities (Nasseh et al., 2016). Furthermore, iron particles from industrial operations can be employed as ZVI in pollution treatment. Iron nanoparticles have been proven in studies to be effective at removing pollutants such chlorine, heavy metals, organic, aromatic nitro compounds, poly bromine diphenyl ethers, pesticides, nitrates, and pigments (Hu, Ndé-Tchoupé, et al., 2019). ZVI has been thoroughly investigated as a material to remove aqueous As, and the underlying mechanisms have received little attention (Zhao et al., 2021). The removal of aqueous As species using ZVI happens as ZVI materials corrode in water (Kanel et al., 2005; Melitas et al., 2002; Rashid et al., 2020). The removal of As ions from solution is accomplished through adsorption, co-precipitation, and surface complexation with Fe(II) and Fe(III) oxides, hydroxides, and oxyhydroxides generated by ZVI (Liang et al., 2022). The possible mechanisms are presented in **Figure 5.**

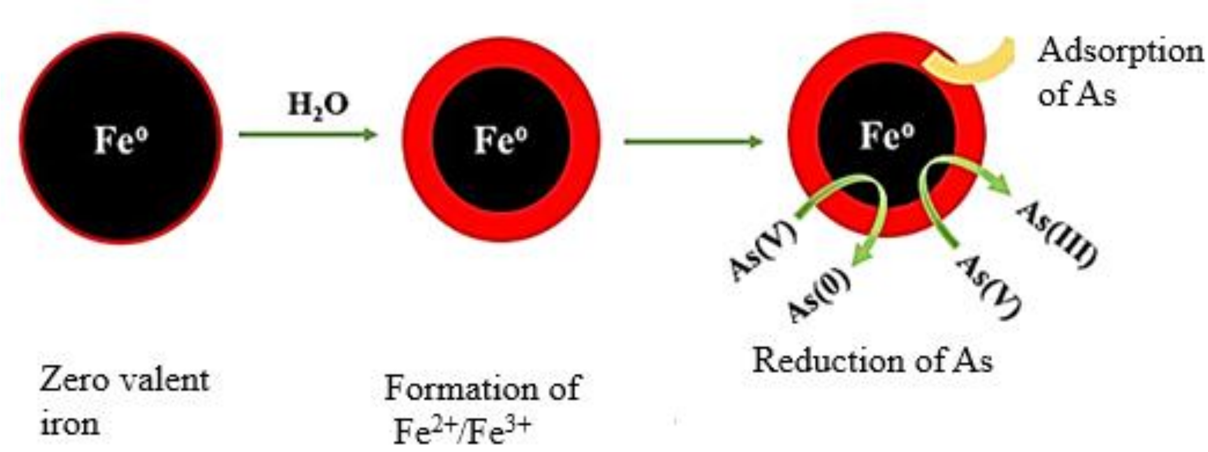


Figure 5: Schematic of the mechanisms of arsenic removal by zero valent iron (Rashid et al., 2020).

The oxidation rate at the ZVI-water interface is also known to influence the nature of the oxides and oxyhydroxides generated on the iron surface, as well as the final corrosion products. Lepidocrocite and magnetite are some of these corrosion products, with green rust and bernalite acting as intermediaries (Caloa et al., 2012). The effectiveness of As removal with ZVI is influenced by a variety of variables, including the type of ZVI material used, the physicochemical properties of the water, the initial concentration of arsenic, and the speciation of arsenic (Biterna et al., 2010).

2.5 Design sand filters with ZVI

Filter systems that use ZVI as a corrosion source for treating water are likely to clog, which negatively impact the effectiveness of the system's ability to remove pollutants (Lawrinenko et al., 2023; Weber et al., 2013). Clogging mechanisms include fouling material adsorption, bio-corrosion, cake formation, and iron volumetric expansion (Naseri et al., 2017). The volumetric expansion of iron is responsible for the majority of filter clogging, particularly in water with a pH greater than 4 (Plessl et al., 2023). Adding a chemically inert substance, such as sand, can extend filter life by increasing void space between particles and giving adequate capacity for corrosive iron expansion (Hu et al., 2020; Hu, Gwenzi, et al., 2019; Noubactep, 2015).

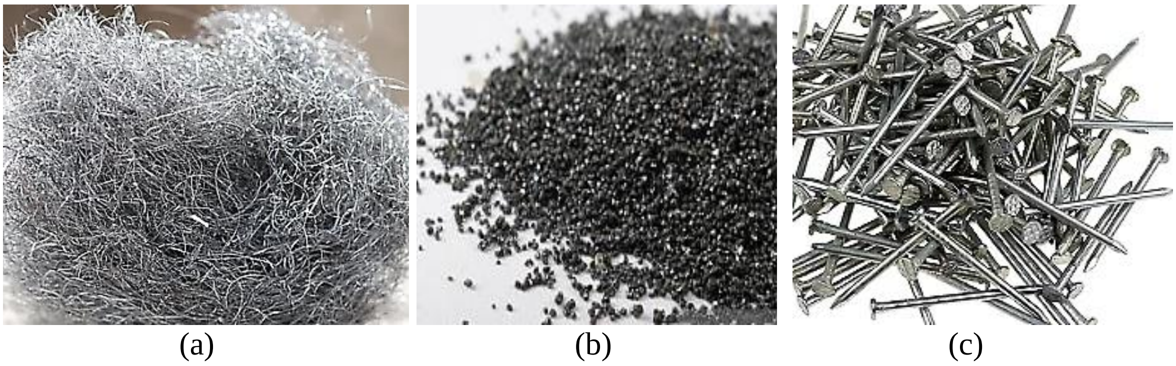
Iron (hydr)oxides are produced in the reactive zone of a ZVI/sand filter for further pollutant removal in the filter (Plessl et al., 2023). A filter that uses 100% ZVI without an inert components is not effective, even though the ZVI material is an essential component of an efficient filter. Because iron corrosion is an expanding process, a filter made entirely of ZVI material would soon lose its porosity and become unsuitable for filtering water (Btatkeu-K et al., 2014; Noubactep, 2018; Tao et al., 2022). The volumetric percentage of ZVI should not exceed 51%, or the filter would lose permeability before all of the ZVI can be exhausted, resulting in material waste (Makota et al., 2017; Noubactep, 2010; Noubactep et al., 2012; Xiao et al., 2022). The selection of the suitable ZVI form with respect to availability, reactivity, performance, cost, and environmental consideration is an important component in As remediation using ZVI (Plessl et al., 2023). The performance of several forms of ZVI has been independently investigated (Weber et al., 2013). Despite their excellent reported performance, comparison of their performance in similar experimental conditions has received little attention (Lawrinenko et al., 2023; Plessl et al., 2023; Weber et al., 2013).

CHAPTER THREE

MATERIALS AND METHODS

3.1 Zero valent iron materials

Iron wool, iron fillings, and iron nails (**Figure 6**) 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.



(a) (b) (c)

Figure 6: Forms of ZVI used in experiments (a) Iron wool, (b) Iron filings and (c) Iron nails

3.2 Materials characterization

The elemental composition of the materials was determined by X-ray fluorescence using a hand-held XRF analyzers (Hitachi X-MET8000 range). XRF spectroscopy is a non-destructive, quick, and precise technology for identifying and detecting material composition. It requires no or very little sample preparation and is suitable with solid, liquid, and powdered materials (de la Guardia & Armenta, 2011). The types of XRF spectrometers can range from small, portable instruments to large, tabletop apparatuses (Ardebili et al., 2018). XRF works by directing an X-ray beam onto the surface of the material. Atoms either scatter these X-rays or absorb them and cause an atom to release one of its inner electrons. As a result, an unstable condition develops, which is resolved by moving electrons from outer to inner shells. This results in the emission of secondary or fluorescence light with distinctive wavelengths that are used to identify the element (Goodman et al., 2015). The picture of the used handheld XRF analyzers is shown in **Figure 7**.

Figure 7: X-MET8000 hand-held analyzer

This instrument was calibrated using the appropriate alloy measuring standards. The detection limit for lighter elements (Mg, Al, Si, K, etc.) ranged between 100 and 1000 parts per million (ppm), whereas the detection limit for heavier elements (Rb, Sr, Zr, Pb, etc.) rises to 5 ppm (Simsek, 2020). Measurements and data collecting were carried out in accordance with the instrument's user manual.

3.3 Design of column filter

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 µg/L As (III). As illustrated in **Figure 8**, 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) were 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 used in experiments was collected from the Nduruma River near the bridge on Mandela Road. 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.

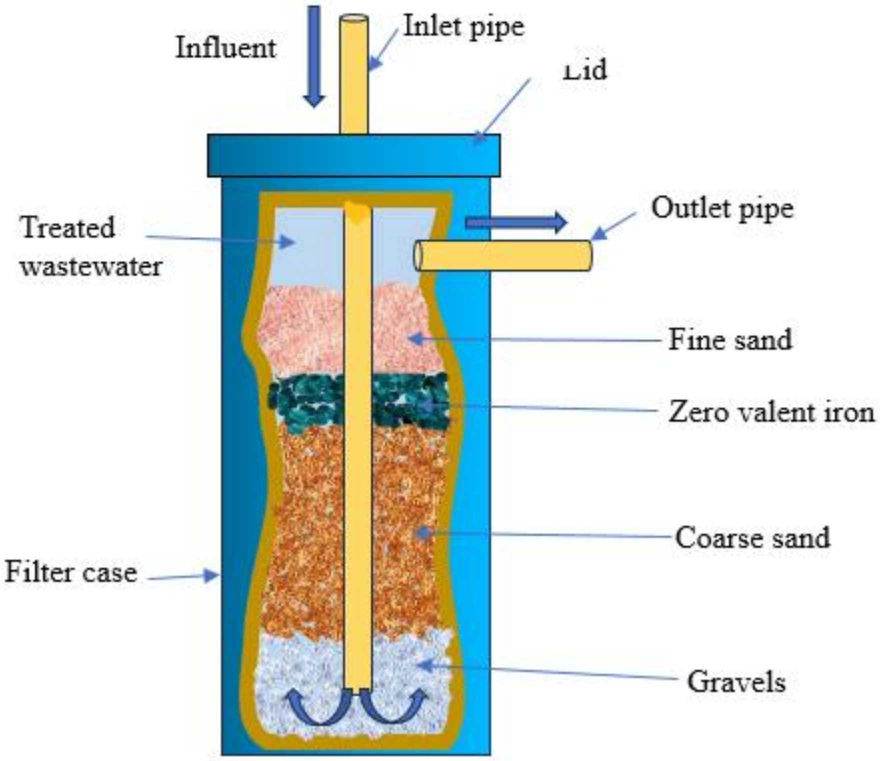


Figure 8. Schematic diagram of the column filters with layers of substrates modified (Ndé-Tchoupé et al., 2022; Ngai et al., 2006).

The plastic piping was used to create a downward flow of synthetic groundwater to the base of the filter from the storage tank situated at its top. This water is forced upward through the filter media and out of the top delivery pipe by the water in the storage tank. The filter traps and removes the suspended particles and As in three filter layers with gravel, ZVI, fine and coarse sand. A bed of gravel is placed at the bottom of the filter to allow any water that enters to distribute equally across the base.

3.4 Chemicals and reagents

Plasticware used (to avoid metal contaminations) was cleaned in hydrochloric acid (20%, v/v) followed by nitric acid (20%, v/v) and rinsed with distilled water. In this study, analytical grade chemicals were used without further purification. The mass of arsenic trioxide (As_2O_3) required to prepare 5 L of a 1000 mg/L As (III) solution was calculated. The concentration of As (III) was converted from mg/L to moles/L using its molar mass. The mole ratio between As (III) and As_2O_3 was determined, and the required mass of As_2O_3 was found by multiplying the moles needed by its molar mass. The final result showed that 6.6 g of As_2O_3 were needed for the preparation. Subsequently, this stock solution underwent a dilution process to achieve a series of solutions with concentrations of 1000 µg/L. In an effort to optimize the conditions for the removal of As, the pH of these diluted solutions was carefully adjusted to a neutral pH of 7. This adjustment was accomplished using 0.1 M sodium hydroxide (NaOH) and 0.1 M hydrochloric acid (HCl) (Azhdarpoor et al., 2015; Biterna et al., 2010; Farrell et al., 2001). These solutions were then used in experiments. All glassware and plastic bottles used in the preparation and storage of the As(III) solutions were meticulously cleansed to ensure the absence of contaminants. This was achieved by soaking the equipment in a 10% nitric acid (HNO_3) solution. Following the acid soak, the glassware and plastic bottles were thoroughly rinsed three times with distilled water to eliminate any residual HNO_3.

3.5 Batch experiments

Batch experiments were designed to compare the performance of different forms of ZVI in removing As (III) from a synthetic solution. In these experiments, the synthetic solution runs up the gravel bed and emerges at the top of the fine sand, where it is collected at the exit tap as shown in **Figure 9**. Various doses of ZVI, specifically 200 g, 300 g, 400 g, 600 g, and 800 g, were tested to determine their impact on As removal. Additionally, the experiments evaluated the influence of different contact times, ranging from 6 to 72 hours. To mimic environmental conditions as closely as possible, the experiments were conducted at room temperature which was ranging from 23 to 26 ^{0}C. Each experimental condition was replicated three times to ensure the reliability and reproducibility of the results. The average of these triplicate runs was used for further calculations and analysis. This systematic approach allowed for a comprehensive understanding of the optimal ZVI dosage and contact time needed for effective As removal.

3.6 Analysis of As in samples

Samples (100 mL) collected directly from the effluent outlet of the filter column were analyzed for arsenic (III) using WFX-210 Atomic Absorption Spectrophotometer (AAS) at Arusha Technical College's water laboratory. All preparatory steps, including acid digestion and dilution, were executed within a controlled environment of a clean laminar flow bench, equipped with a comprehensive exhaust system to minimize contamination risks. These measures are critical to maintaining the integrity of the samples and the accuracy of results. The AAS was calibrated using a linear calibration curve that passed through zero, ensuring precision across the expected range of As concentrations in the samples. The calibration process yielded correlation coefficients above 0.9900, indicating strong linearity and reliability of the instrument's measurements.

3.7 Data analysis and interpretation

MS Excel sheet was utilized for analysis of data sets, ensuring accurate interpretation through graphical representations and numerical summaries, facilitating comprehensive understanding of the data's trends and patterns. The efficiency of arsenic removal was calculated using equation 1

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

Where;

R: removal efficiency (%)

C_{in}: the influent concentration of arsenic in the solution (µg/L)

C_{eff}: the effluent concentration of arsenic in the solution (µg/L)

CHAPTER FOUR
RESULTS AND DISCUSSION

4.1 Results

4.1.1 Characteristics of the the used materials

The elemental composition analysis of sand and gravel focused primarily on oxides of iron (Fe), aluminum (Al), calcium (Ca), and silica (Si), which significantly influence their properties. Elemental analysis revealed distinct compositions among the samples: coarse sand exhibited a predominant silicon content (Si) of 43%, followed by fine sand with 38%, and gravel showing 18%. Beyond these primary elements (Fe, Al, Ca and Si), the remaining composition included significant proportions of feldspar, olivine, and various clay minerals, contributing to the overall mineralogical diversity of the samples as shown in **Table 6**.

Table 6: Elemental composition of sand and gravels used in experiments

	Percentage elemental composition				
	Si	Ca	Al	Fe	Others
Fine sand	38.0	2.1	0.9	0.4	58.6
Gravel	18.0	8.0	6.0	13.0	55.0
Coarse sand	42.0	0.5	0.9	0.3	56.2

These additional minerals can influence the physical and chemical behavior of the materials, affecting parameters such as porosity, permeability, and chemical reactivity in various environmental and industrial applications. These results have been summarized in **Figure 9**.

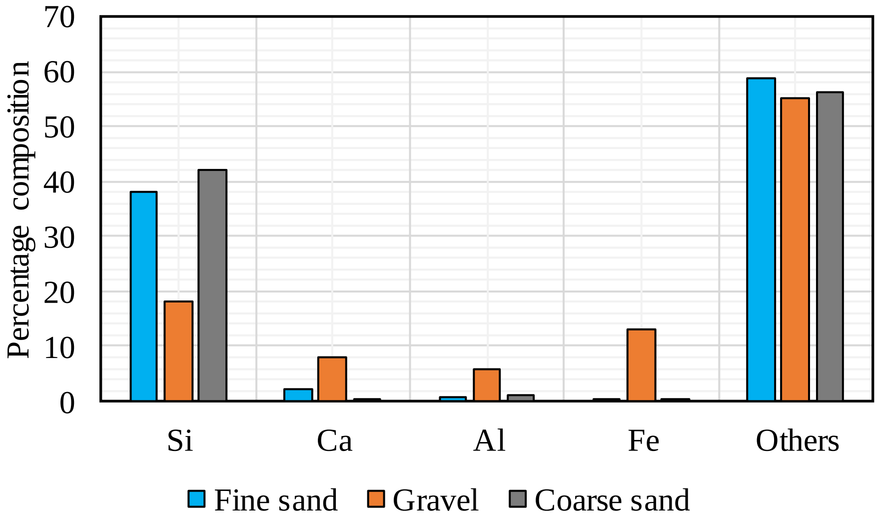


Figure 9: Elemental composition of gravels and sand used in experiments

The **Table 7** presents the elemental composition (%) of different forms of ZVI such as iron nail, iron wool, and iron filling used in experiments. It was found that the iron nail contained significant amounts of Fe (60.3%), with notable proportions of Si (2.9%), Ca (1.5%), and Al (0.9%). Other constituents contributed 34.5% to the composition. Iron wool showed higher Fe content (67.3%) compared to the iron nail, with lower proportions of Si (1.7%), Ca (0.3%), and Al (0.4%). Other constituents making up 30.4% of its composition. Iron filling had the highest Fe content (71.7%) among the materials analyzed, with minimal Si (0.3%) and no recorded Ca or Al. Other constituents contributed 28.0% to its composition.

Table 7: Elemental composition of Iron nail, Iron wool, and Iron filings used in experiments

	Percentage elemental composition				
	Si	Ca	Al	Fe	Others
Iron nail	2.9	1.5	0.9	60.3	34.5
Iron wool	1.7	0.3	0.4	67.3	30.4
Iron filing	0.3	-	-	71.7	28.0

These results have been summarized in **Figure 10**.

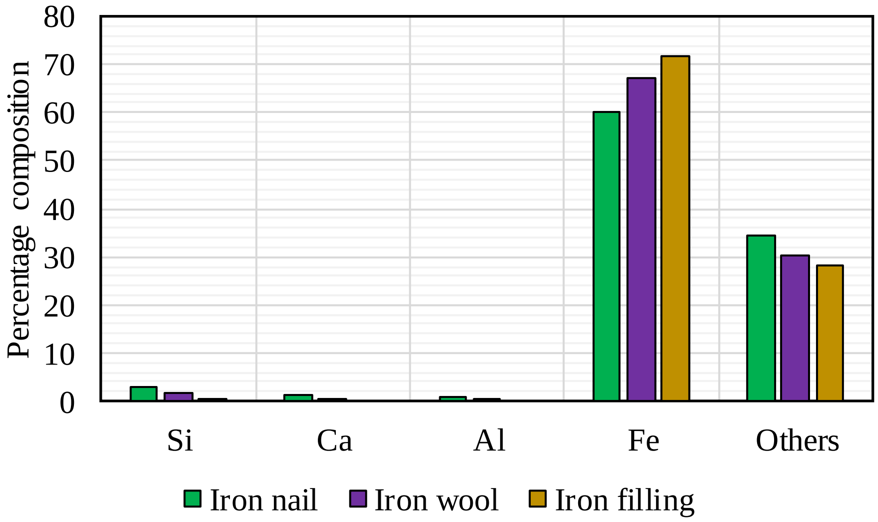


Figure 10: Elemental composition of iron nails, iron wool and iron filings used in experiments

4.1.2 Removal of As

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 µg/L. **Tables 8**, summarizes the effluent arsenic concentration (C_{eff}) and the removal efficiencies (% Removal) in all the columns.

Table 8: The removal of As using iron wool at different dose and retention time

Dose of ZVI (g)	Time (h)	C_{eff} Trial 1 (µg/L)	C_{eff} Trial 2 (µg/L)	C_{eff} Trial 3 (µg/L)	C_{eff} Average (µg/L)	SD (µg/L)	% Removal
200	6	137.50	150.80	152.55	146.95	14.47	85.3
200	12	72.80	72.10	72.72	72.54	4.09	92.7
200	24	42.70	45.10	46.12	44.64	3.13	95.5
200	48	24.60	23.50	32.84	26.98	5.96	97.3
200	72	25.30	26.00	26.79	26.03	8.34	97.4
300	6	120.10	118.00	119.05	119.05	6.99	88.1
300	12	62.90	59.90	61.34	61.38	1.52	93.9
300	24	44.80	40.70	42.81	42.77	4.70	95.7
300	48	26.50	25.70	23.13	25.11	4.16	97.5
300	72	17.30	18.30	14.62	16.74	2.50	98.3
400	6	111.00	103.50	109.17	107.89	4.72	89.2
400	12	55.30	58.20	65.03	59.51	5.57	94.0
400	24	33.48	33.48	33.48	33.48	3.01	96.7
400	48	16.20	15.50	15.76	15.82	1.35	98.4
400	72	8.30	7.10	4.10	6.50	2.31	99.4
600	6	101.30	99.00	117.76	106.02	8.34	89.4
600	12	60.60	56.20	56.21	57.67	10.07	94.2
600	24	27.50	24.60	31.60	27.90	4.61	97.2
600	48	9.90	11.40	12.18	11.16	1.33	98.9
600	72	6.30	3.90	5.94	5.38	1.53	99.5
800	6	75.20	70.30	69.36	71.62	3.68	92.8
800	12	44.90	49.40	50.75	48.35	4.02	95.2
800	24	15.50	15.20	19.52	16.74	2.09	98.3
800	48	6.60	8.20	7.55	7.45	1.11	99.3
800	72	5.10	4.40	4.48	4.66	0.70	99.5

Table 9: The removal of As using Iron filings at different dose and retention time

Dose of ZVI (g)	Time (h)	C_{eff} Trial 1 (µg/L)	C_{eff} Trial 2 (µg/L)	C_{eff} Trial 3 (µg/L)	C_{eff} Average (µg/L)	SD (µg/L)	% Removal
200	6	182.40	172.80	163.71	172.97	9.80	82.7
200	12	127.10	128.60	118.94	124.88	3.97	87.5
200	24	82.60	79.90	78.94	80.48	2.52	92.0
200	48	38.20	37.20	35.57	36.99	3.29	96.3
200	72	22.50	23.30	23.56	23.12	1.27	97.7
300	6	142.20	141.90	146.04	143.38	1.99	85.7
300	12	98.70	99.60	104.16	100.82	2.50	89.9
300	24	67.00	68.40	61.64	65.68	2.84	93.4
300	48	29.40	31.60	30.59	30.53	1.80	96.9
300	72	14.30	18.60	17.05	16.65	2.89	98.3
400	6	116.80	112.20	115.07	114.69	2.67	88.5
400	12	76.30	74.40	74.09	74.93	1.44	92.5
400	24	59.20	57.80	55.08	57.36	2.06	94.3
400	48	30.30	25.10	27.85	27.75	3.40	97.2
400	72	14.70	12.80	14.14	13.88	1.62	98.6
600	6	85.50	84.80	85.03	85.11	0.83	91.5
600	12	50.80	55.20	52.19	52.73	3.58	94.7
600	24	29.80	30.70	28.33	29.61	1.36	97.0
600	48	19.20	20.30	18.79	19.43	1.78	98.1
600	72	13.90	13.40	11.58	12.96	2.02	98.7
800	6	65.90	64.10	67.04	65.68	1.97	93.4
800	12	36.50	37.80	39.49	37.93	1.68	96.2
800	24	29.10	30.10	29.63	29.61	0.51	97.0
800	48	18.50	17.00	17.21	17.57	1.03	98.2
800	72	3.30	3.90	3.87	3.69	0.40	99.6

Table 10: The removal of As using iron nails at different dose and retention time

Dose of ZVI (g)	**Time (h)**	**C_{eff} Trial 1 (μg/L)**	**C_{eff} Trial 2 (μg/L)**	**C_{eff} Trial 3 (μg/L)**	**C_{eff} Average (μg/L)**	**SD (μg/L)**	**% Removal**
200	6	160.40	157.60	158.40	158.80	4.16	84.1
200	12	136.89	135.90	134.40	135.73	2.16	86.4
200	24	97.77	92.80	94.70	95.09	3.42	90.5
200	48	69.86	62.00	62.00	64.62	4.31	93.5
200	72	56.04	57.80	63.40	59.08	4.54	94.1
300	6	140.44	143.00	143.10	142.18	1.93	85.8
300	12	133.58	131.00	134.30	132.96	2.40	86.7
300	24	74.37	74.80	75.20	74.79	0.71	92.5
300	48	54.42	52.20	54.00	53.54	1.48	94.6
300	72	50.15	50.30	46.40	48.95	2.84	95.1
400	6	116.01	112.80	111.90	113.57	3.04	88.6
400	12	108.92	107.80	110.10	108.94	4.53	89.1
400	24	65.16	63.70	65.00	64.62	0.95	93.5
400	48	47.79	55.90	54.20	52.63	3.32	94.7
400	72	44.97	47.40	48.90	47.09	1.85	95.3
600	6	110.33	111.90	112.90	111.71	6.31	88.8
600	12	91.06	93.90	92.00	92.32	2.39	90.8
600	24	61.42	59.70	61.70	60.94	2.45	93.9
600	48	29.68	29.40	29.60	29.56	1.92	97.0
600	72	27.37	27.30	25.70	26.79	1.34	97.3
800	6	62.23	64.90	64.00	63.71	2.55	93.6
800	12	58.91	59.70	55.90	58.17	2.32	94.2
800	24	47.17	47.30	46.80	47.09	1.87	95.3
800	48	25.77	28.00	26.60	26.79	1.44	97.3
800	72	26.16	22.50	23.40	24.02	1.58	97.6

4.1.2.1 Effect of contact time on As removal efficiency

The contact time between the adsorbate and adsorbent is a crucial factor influencing the efficiency of the adsorption process. **Figures 11, 12, 13, 14** and **15** illustrate the time-dependent removal of As using varying fixed doses of ZVI. The results clearly indicate that the rate of As removal increases consistently with longer contact times. Initially, there is a rapid removal phase due to the abundance of available adsorption sites on the ZVI. As contact time progresses, the rate of removal slows down, approaching a plateau, indicating that the adsorption sites are becoming saturated. This trend highlights the importance of optimizing contact time to maximize the removal efficiency of As. Longer contact times allow for more thorough interaction between arsenic and ZVI, leading to higher removal rates. However, it is essential to balance this with practical considerations such as operational efficiency and cost. The findings underscore the significance of contact time in the design and optimization of adsorption systems for effective As removal using ZVI.

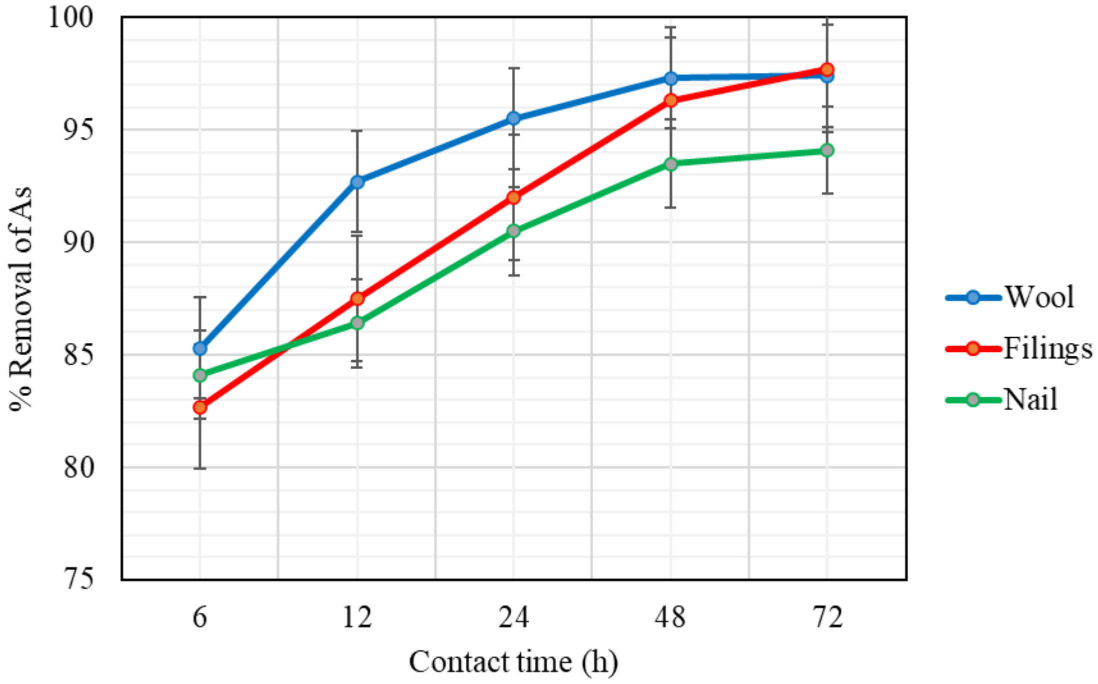


Figure 11: Effect of contact time on removal efficiency of As (200mg of ZVI)

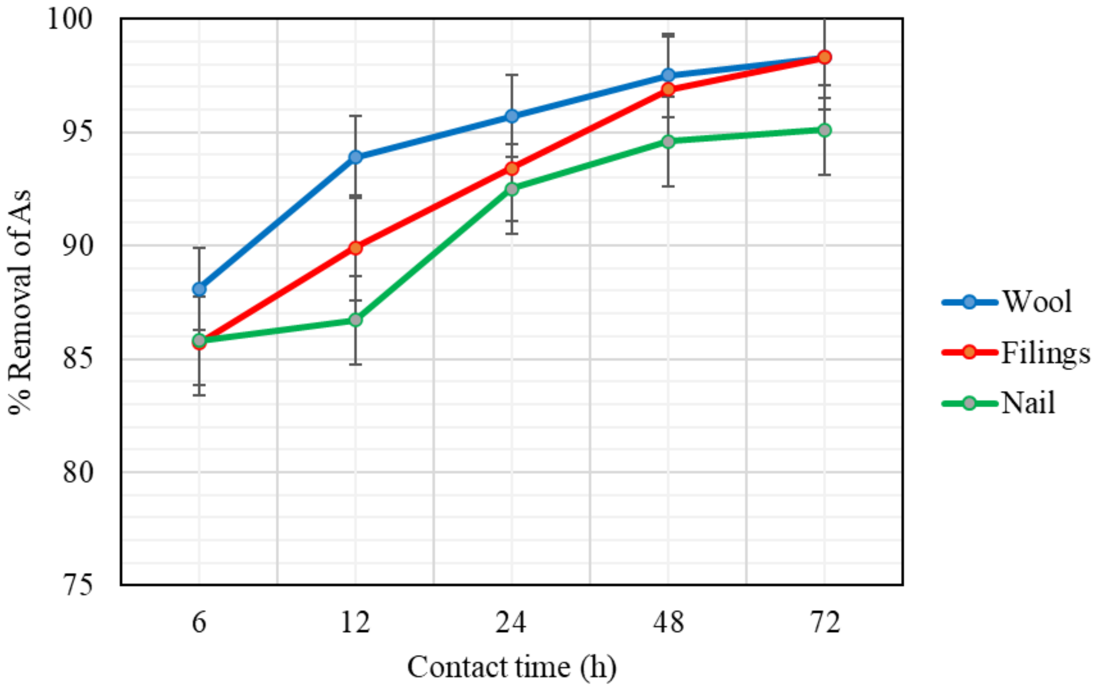


Figure 12: Effect of contact time on removal efficiency of As (300mg of ZVI)

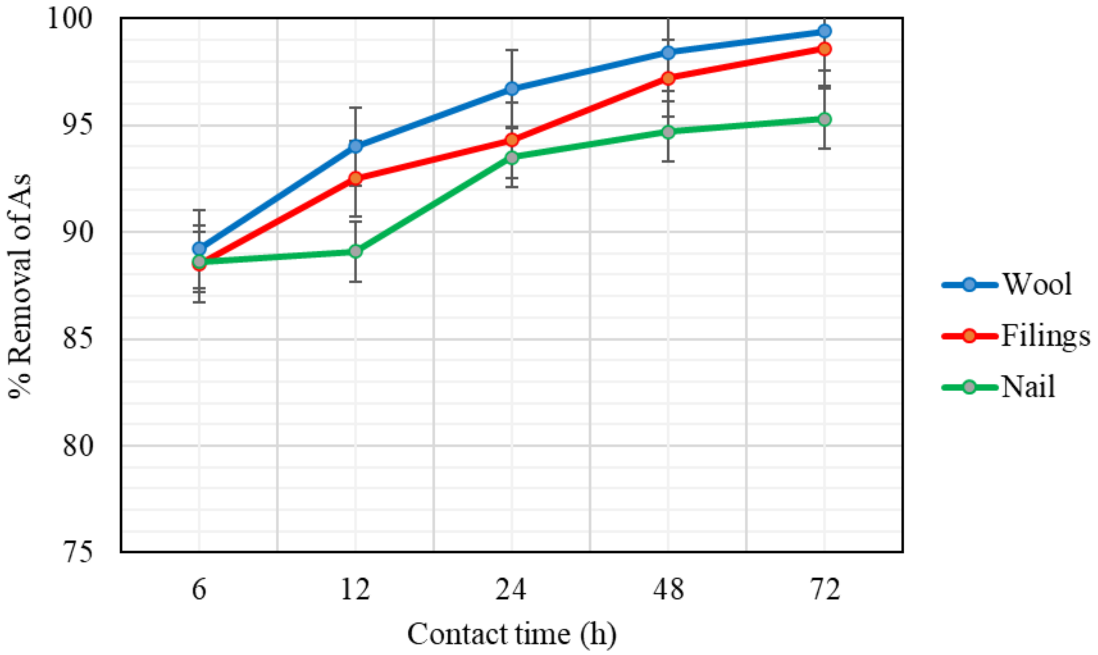


Figure 13: Effect of contact time on removal efficiency of As (400mg of ZVI)

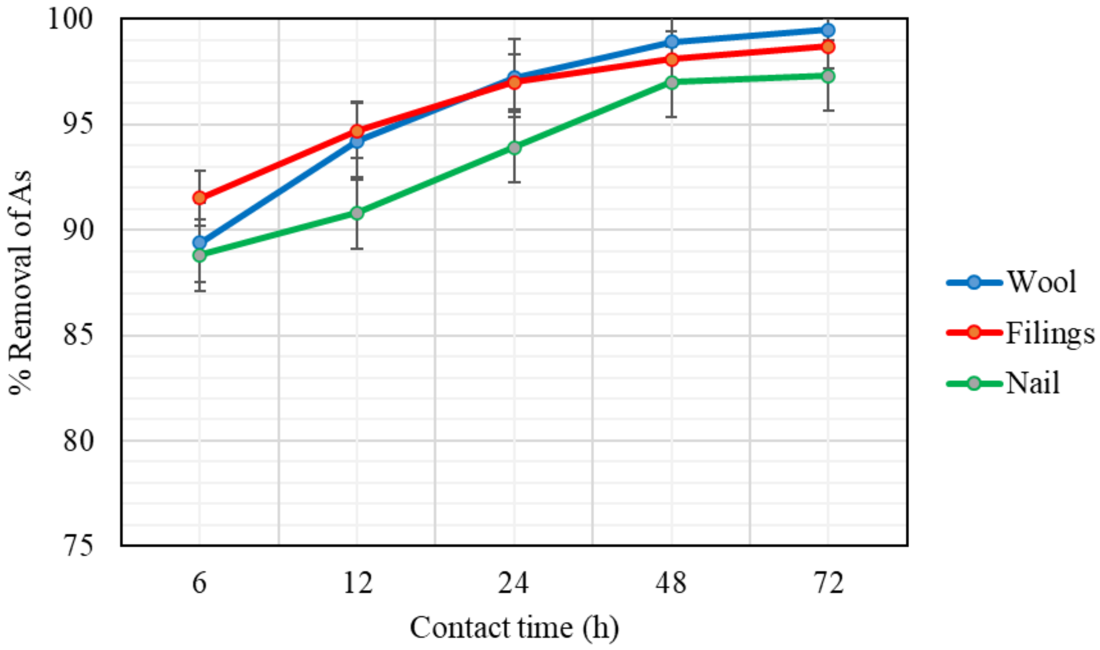


Figure 14: Effect of contact time on removal efficiency of As (600mg of ZVI)

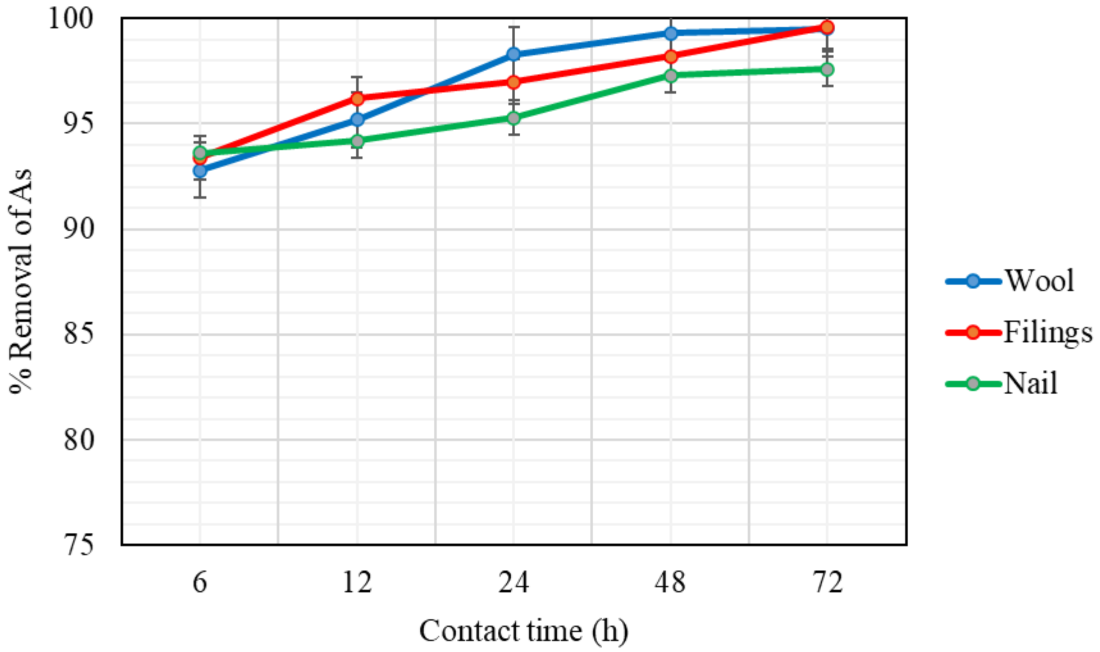


Figure 15: Effect of contact time on removal efficiency of As (800mg of ZVI)

4.1.2.2 Effect of ZVI dose on arsenic removal efficiency

The effectiveness of As removal is notably influenced by the dosage of ZVI used as an adsorbent. Optimizing this dosage is essential for developing cost-effective methods for reducing As concentrations in water. To evaluate this, the removal efficiency of arsenic was assessed using a 1000 µg/L As (III) solution across varying ZVI doses. The results consistently showed the increase in ZVI dosage and the enhancement of As removal efficiency. Specifically, the study examined the effect of ZVI dose on As removal at different fixed contact times such as 6h, 12h, 24h, 48h and 72h. As depicted in **Figures 16, 17, 18, 19** and **20**, As removal efficiency improved with higher ZVI doses for both contact times. The data indicated that longer contact times further augmented the removal efficiency, suggesting that both the amount of adsorbent and the duration of exposure are critical factors in optimizing the process. These findings underscore the importance of determining the optimal ZVI dosage to achieve effective and economical As remediation in practical applications.

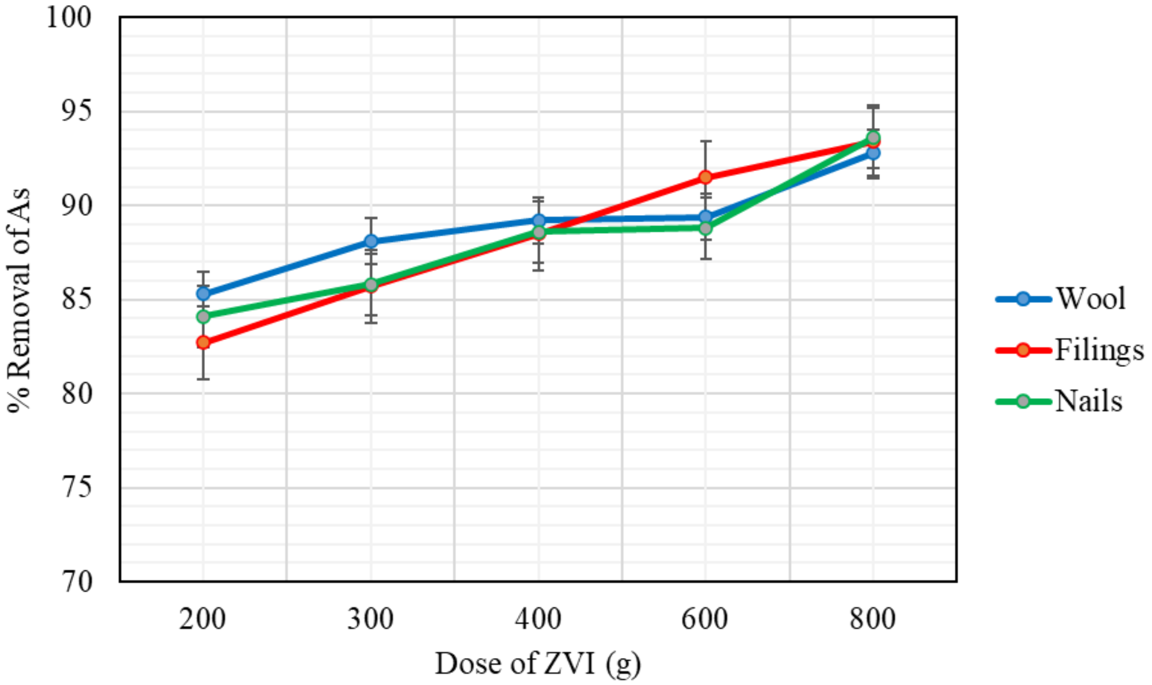


Figure 16: Effect of adsorbent dose on removal of As in 6 h

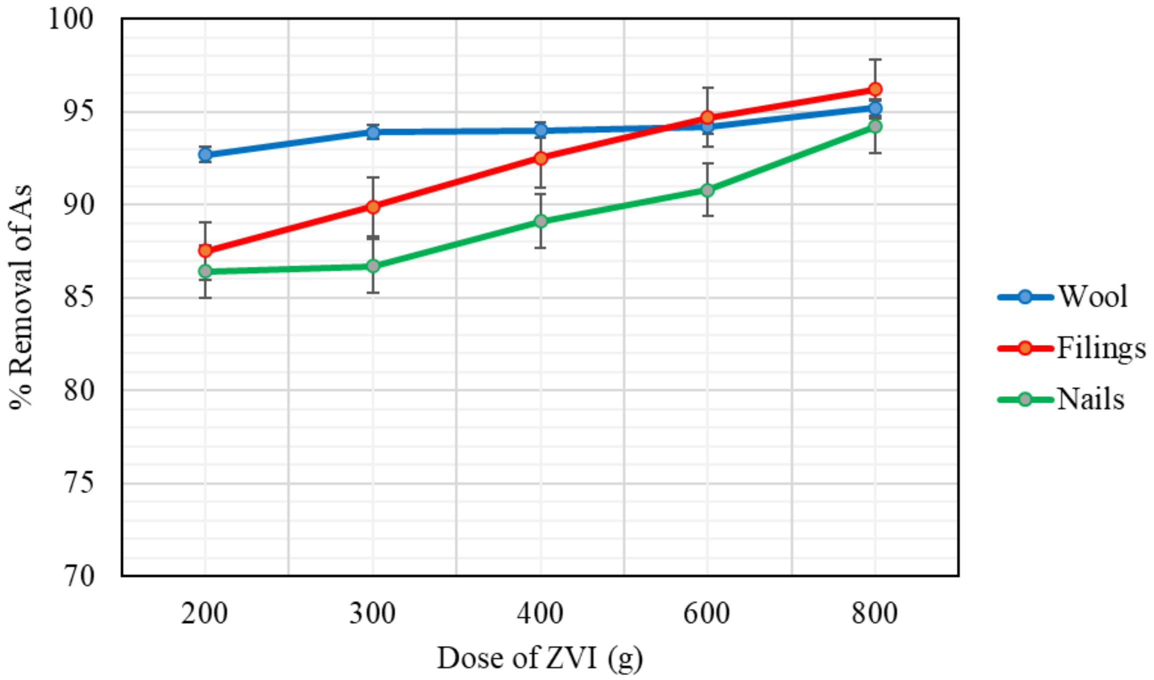


Figure 17: Effect of adsorbent dose on removal of As in 12 h

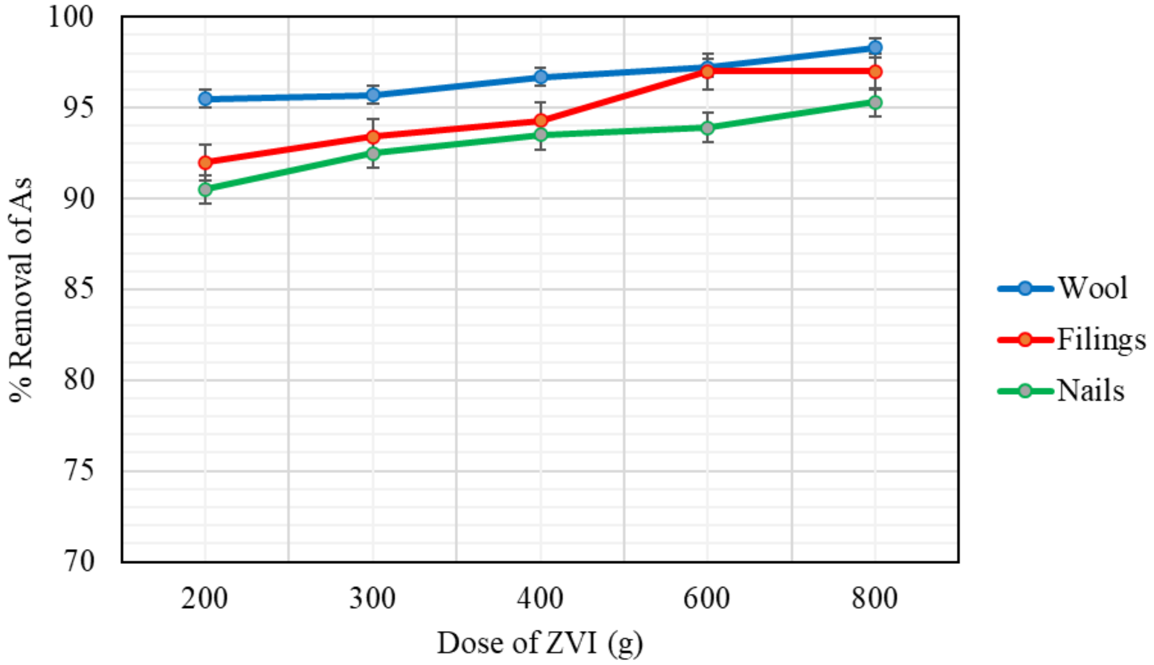


Figure 18: Effect of adsorbent dose on removal of As in 24 h

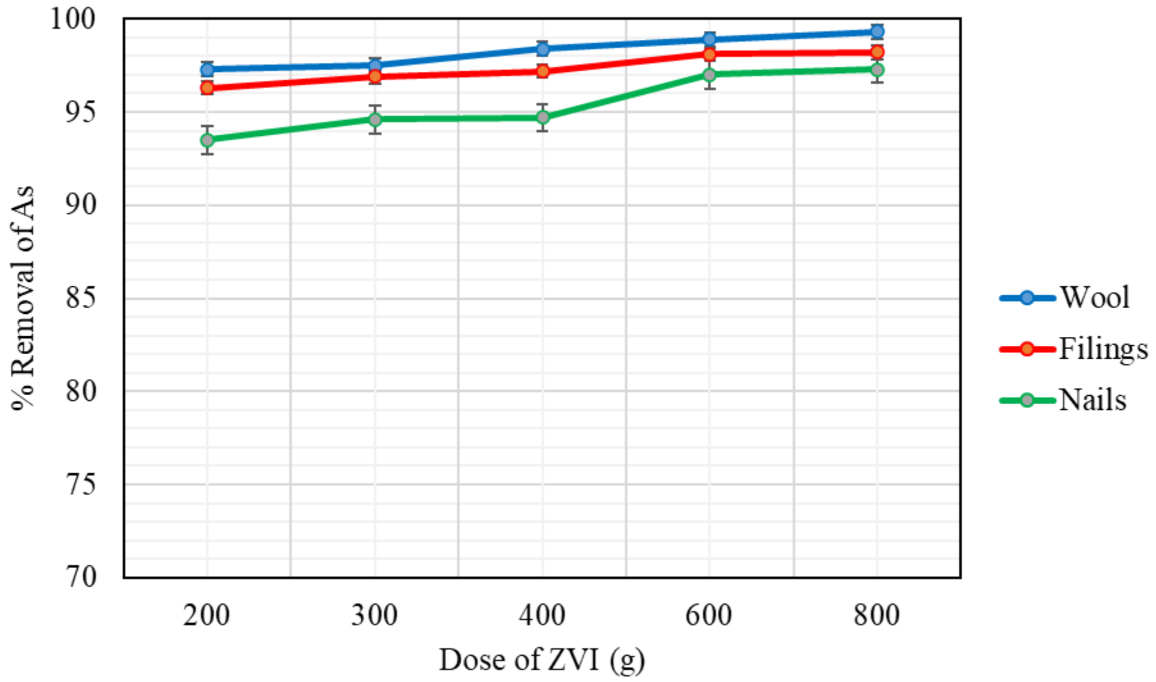


Figure 19: Effect of adsorbent dose on removal of As in 48 h

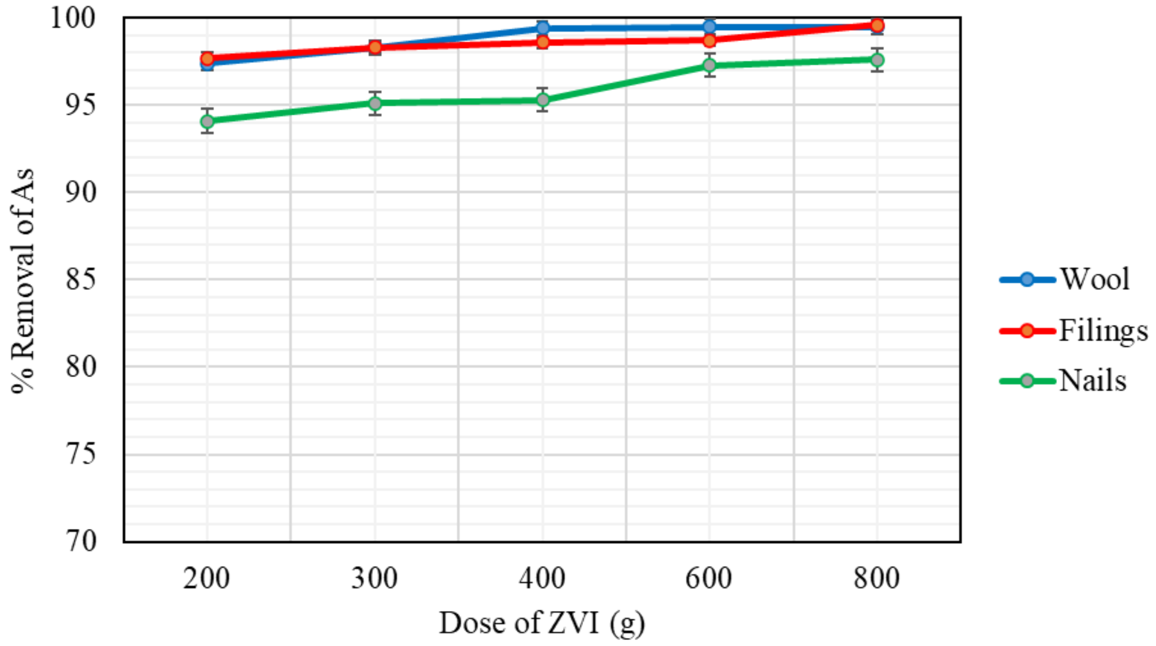


Figure 20: Effect of adsorbent dose on removal of As in 72 h

4.2 Discussion

4.2.1 Effect of contact time on arsenic removal efficiency

The use of ZVI for the remediation of As-contaminated water has gained significant attention due to its effectiveness and cost-efficiency (Fu et al., 2014). The contact time between the adsorbent and the contaminants has a major impact on the efficiency of use of ZVI to remove As (Casentini et al., 2016). Numerous studies have shown that increasing the contact time significantly improves the efficiency of As removal (Carneiro et al., 2022; Khanzada et al., 2023). The results from this study shows that the removal efficiency of As using ZVI increases significantly during the initial contact period, particularly within the first 48 h. This rapid phase can be attributed to the abundance of reactive sites on the ZVI surface (Guo et al., 2016; Morgada et al., 2009; Ullah et al., 2020). When ZVI is added to an aqueous solution, it oxidizes and corrodes (Kowalski & Søgaard, 2014). This produces a range of reactive sites that are very affinities for As species, including iron hydroxides and oxides (Guo et al., 2016; Manning et al., 2002). The fast formation of these reactive sites promotes rapid adsorption of As, which results in a significant decrease in As concentration during the initial phases of interaction (Farooqi et al., 2021; Liu et al., 2018).

Various mechanisms, including adsorption, co-precipitation, and redox reactions, are involved in adsorption of As onto ZVI (S. Wang et al., 2017). Initially, As in its predominant forms, As (V) and As (III), adsorbs onto the surface of ZVI (Xi et al., 2019; Yadav et al., 2016). ZVI undergoes oxidation to ferrous iron (Fe^{2+}), which subsequently forms iron hydroxides ($Fe(OH)_2$) and ferric hydroxides ($Fe(OH)_3$) upon further oxidation (Carabante Martinez, 2012; Ramos et al., 2009; Refait et al., 2003; Stefánsson, 2007). These iron hydroxides serve as adsorbents, binding As through complexation and electrostatic interactions (Bang & Meng, 2004; Siddiqui & Chaudhry, 2017). Furthermore, ZVI can reduce As(V) to As(III), which is then immobilized on the iron oxides (Formentini et al., 2024; F. Sun et al., 2011).

After the first 48 h, the rate of arsenic removal decreases, but steadily increases. The reason for this prolonged removal phase might be attributed to the gradual development of corrosion products on the ZVI surface, which consistently supply fresh adsorption sites (Caloa et al., 2012; Singh et al., 2021). The process of corrosion causes porous structures to form on the iron surface, boosting the material's surface area and adsorption capacity (Valencia Ramos, 2011). Furthermore, the formation of secondary iron minerals as goethite ($FeO(OH)$) and magnetite (Fe_3O_4) increases the removal efficiency by providing more arsenic species with adsorption sites (Wang et al., 2022).

Prolonged contact time increases the interaction between arsenic and the ZVI surface, which in turn promotes the diffusion of As ions into the corrosion products' porous matrix (Manning et al., 2002; Meng et al., 2024; Ramos et al., 2009; F. Sun et al., 2011). This longer contact time increases the possibility that As will be trapped in the iron oxides, which improves removal efficiency overall (Meng et al., 2024; Yu et al., 2015). Furthermore, the dynamic character of the corrosion process guarantees a steady supply of reactive sites, preserving ZVI's adsorption capability for a prolonged period of time (Lawrinenko et al., 2023; Rangsivek & Jekel, 2005; Siciliano & Limonti, 2018). In order to effectively design water treatment systems utilizing ZVI, it is imperative to comprehend the impact of contact time on the effectiveness of As removal.

4.2.2 Effect of dose of ZVI on arsenic removal efficiency

The dose of ZVI has a crucial impact on the effectiveness of As removal from waster (Bang et al., 2005; Lien & Wilkin, 2005; Qiao et al., 2021). The efficiency of removing As increases with increasing ZVI dose, and this enhancement is caused by a number of inter linked variables (Feroze Ahmed, 2001; Plessl et al., 2023). One of the primary factors for increased As removal with increasing ZVI dose is the proportional increase in the number of surface-active sites available for adsorption (Kheskwani & Ahammed, 2023; Liao et al., 2021). The adsorption process is essential to efficiently remove As from contaminated water (Cheng et al., 2021; Zhu et al., 2009). Higher ZVI dosages increase the surface area accessible for As to attach themselves to, improving the removal efficiency (Pintor et al., 2018; Xu et al., 2022; Zhu et al., 2020). Because of the larger surface area, there are more active sites for As to be absorbed, which improves the efficiency of cleansing up the contaminated water (Kheskwani & Ahammed, 2023; Zhu et al., 2009).

The oxidation and reduction activities that are essential for the removal of As are also increases in the presence of more ZVI particles in the system (Kheskwani & Ahammed, 2023; Ramos et al., 2009). ZVI functions as a potent reducing agent, capable of transforming As from its highly mobile and deadly form, As (V), to a less mobile and harmful form, As (III) (Caloa et al., 2012; Kheskwani & Ahammed, 2023). Because As (III) is more readily adsorbed onto ZVI particles, this transition is essential (Formentini et al., 2024). The overall capacity for these redox processes increases with increasing ZVI dose, which increases the rate of As removal (Pezeshki et al., 2023; Plessl et al., 2023).

The presence of free electrons (e-) is a noteworthy additional factor that enhances removal efficiency with increasing ZVI dose (Lawrinenko et al., 2023; Rashadul & Chowdhury, 2015). During redox processes, ZVI, a metallic iron in its zero oxidation state, quickly contributes electrons (Guo et al., 2016). The reduction of arsenic (V) to arsenic (III) and the entire adsorption process are facilitated by the presence of free electrons, which improves the As removal efficiency (Lawrinenko et al., 2023; Wang et al., 2022).

The adsorption kinetics also play a crucial role in the efficiency of As removal (Kanel et al., 2005; Zhu et al., 2009). As can adsorb more readily at vacant sites when ZVI dose is increased (Kapepula & Luis, 2024; Zhu et al., 2020). This results in an increased initial adsorption rate, which might be crucial in situations when quick removal of As is required (Rashadul & Chowdhury, 2015). The presence of a large number of vacant sites guarantees that As molecules come into contact with these sites more often, improving the removal efficiency overall (Kapepula & Luis, 2024; Khanzada et al., 2023).

From a practical and economical point of view, it is essential to optimize the dosage of ZVI (Ma et al., 2014; Zhu et al., 2009). While increasing the dosage increases the efficiency of As removal, it is important to take into account the possibility for reduced earnings and the associated costs (Hao et al., 2018). An excessive ZVI dose could lead to operational constraints and increased costs without commensurate increases in removal efficiency (Rahidul Hassan, 2023). Although these variables highlight the advantages of larger ZVI doses, it is essential to take into account the practical and economic implications in order to achieve the best possible balance for efficient and sustainable As removal. The advantages of a larger ZVI dose must therefore be balanced with the related costs and practical limitations.

4.2.3 Effect of type of zero valent iron

The type and properties of the ZVI employed determine its efficacy in eliminating As from water (Kheskwani & Ahammed, 2023; Liang et al., 2022; Lien & Wilkin, 2005). In this study three forms of ZVI such as iron wool, iron filings, and iron nails were compared for removal of As from synthetic solution in a sand filter. The results 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 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 As 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 instance, it has been proposed that 40 ^{0}C is the optimal temperature for H/Fe adsorption (Wu et al., 2021). Regarding pH, some studies suggests a pH of 7 as optimal for removing As (Azhdarpoor et al., 2015; Biterna et al., 2010; Farrell et al., 2001).

CHAPTER FIVE

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

In this study, the performance of three forms of ZVI, such as iron wool, iron fillings, and iron nails, were compared in terms of removing As from synthetic aqueous solution. The results showed that despite of the good performance of all forms of ZVI, iron wool performed better in removing As than iron filings and iron nails. This observation shows the potential of ZVI incorporation in filters for remediation of As contaminated groundwater. Furthermore, it was observed that. Increasing the ZVI dose increased the adsorption capacity the removal efficiency of As. The early adsorption response was quick and was observed in all columns, but later reaction involved slow absorption, producing a clear two-phase adsorption characteristic. The findings show that As can effectively be removed from water in the first 48h. These findings demonstrate that locally available materials can be used as ZVI for As remediation in As contaminated groundwater. 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 As from aqueous solution.

5.2 Recommendation

Based on the results of this study, the following recommendations are presented. It is recommended to conduct long-term studies to evaluate the sustained performance of iron wool, iron filings, and iron nails in real-world conditions. This includes assessing the impact of varying water chemistry and flow rates on the adsorption efficiency. Studies should implement pilot-scale testing of sand filters containing ZVI in areas affected by As contamination. This will help in validating the laboratory findings and determining the practical feasibility and scalability of the remediation method. Future studies should explore the development and use of composite materials that combine ZVI with other adsorbents or catalysts to enhance the removal efficiency of As. This could lead to the creation of more effective and robust remediation solutions. In addition, future study should perform a detailed cost-benefit analysis to compare the economic feasibility of using different forms of ZVI for As remediation.

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