

NORWEGIAN UNIVERSITY OF LIFE SCIENCES



## PREFACE

This is my 30 credits Master thesis in the Department of Mathematical Science and Technology. I am a Master's student in the Department of Plant and Environmental Science. My Masters program is Environment and Natural Resources. Specialization Sustainable Water and Sanitation, Health and Development at the Norwegian University of Life Sciences (UMB).

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Ås, March 2012

Md.Fayej Ahmad.

## Abstract:

Arsenic is widely distributed in nature in the air, water and soil. Acute and chronic arsenic exposure by drinking water has been reported in many countries, especially Argentina, Bangladesh, India, Mexico, Mongolia, Thailand and Taiwan. There are many techniques used to remove arsenic from drinking water. Among them reverse osmosis is widely used. Therefore the purpose of this study is to find the conditions favorable for removal of arsenic from drinking water by using reverse osmosis techniques. The experiments were conducted with two types of arsenic (III, V) in tap water up to arsenic concentration of 400  $\mu\text{g/l}$ . We were applied different production water flow rates and pH for removal of arsenic from drinking water. We conclude that in this experiment 99% removal efficiency was achieved for arsenic (III) and arsenic (V). Arsenic (V) is very efficiently removed by reverse osmosis and arsenic (III) removal efficiency depends on water pH. Practical processes can be developed with reverse osmosis to remove all major species of arsenic from water.

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# 1. Introduction

Water is the most essential element for life. In many parts of the world people suffer from poor drinking water quality. Especially people in developing countries are suffer because of the lack of pure drinking water. This is why people are at risk of health hazard (Nguyen, 2007). Arsenic contamination in drinking water is one of the most prominent problems in some countries of the world. Arsenic is generally distributed in the environment (air, water and soil). Arsenic distribution in nature is in both organic and inorganic form. Arsenic has a dual nature. It is a useful element for human health and, on the other hand, if the concentration is higher than the expected limit, arsenic creates problems for health. Arsenic is used for different purposes in the commercial sector. Sources of arsenic in drinking water originate largely from natural sources and create health problems in some countries in the world (Ning, 2002).

The World Health Organization (WHO) has suggested that the upper limit of arsenic in drinking water should be set to 10  $\mu\text{g/l}$  as an alternative to 50  $\mu\text{g/l}$  (Nguyen, 2007). Acute and chronic health effects by arsenic in drinking water have been reported in different countries, especially Bangladesh, India, Mexico, Argentina, Mongolia, Thailand and Taiwan. In these countries a large part of the groundwater is contaminated by arsenic, this groundwater is a source of drinking water (Ning, 2002). The concentration of arsenic in groundwater is 100 to over 2000  $\mu\text{g/l}$ . Some studies show that long term drinking of arsenic contaminated groundwater can lead to cancer of the bladder, lungs, skin, kidney, nose and liver. Long time exposure to arsenic has non cancer effects. It damages cardio-vascular, pulmonary, immunological and neurological systems. Arsenic is an environmental toxicant in drinking water (Ning, 2002).

Bangladesh is one of the most arsenic affected countries in the world, 50 million out of 140 million people are affected by arsenic by drinking water highly concentrated with arsenic from tube wells and dug wells (Geucke et al., 2008). In Bangladesh the arsenic concentration in ground water is up to 1000  $\mu\text{g/l}$ . Because of arsenicosis present and future generations in the Bangladeshi population are exposed to health risks (Geucke et al., 2008).

In the present era, technology developments allow arsenic to be removed from drinking water. Among the most commonly used technologies are oxidation, co-precipitation, adsorption onto coagulation flocks, lime treatment, sorption media, ion exchange resin and membrane technology

(Geucke et al., 2008). Membrane technology has advanced enormously for water treatment all over the world. Reverse osmosis (RO) membranes have become recognized as one of the best available technologies for cleaning inorganic hazard in water (Kang, 2000).

## 2. Chemistry and Geochemistry of arsenic:

Arsenic, atomic number 33, belongs to the VB group in the periodic table. Arsenic is a metalloid, and it occurs in various oxidation states (-III, 0, +III and +V) in nature. In natural water, arsenic is mostly found in inorganic forms as tri-valent arsenite arsenic (III) and penta valent arsenate arsenic (V) (Nguyen, 2007).

The arsenic commonly present in water is the pH dependent species of arsenic ( $H_3AsO_4$ ) and the arsenous ( $H_3AsO_3$ ) acid system. These anions have acidic characteristics. The stability and dominance of a specific species depends on the pH of the solution. Arsenate are stable in aerobic or oxidizing condition, while arsenites are stable under anaerobic or reducing condition (Figure 1)

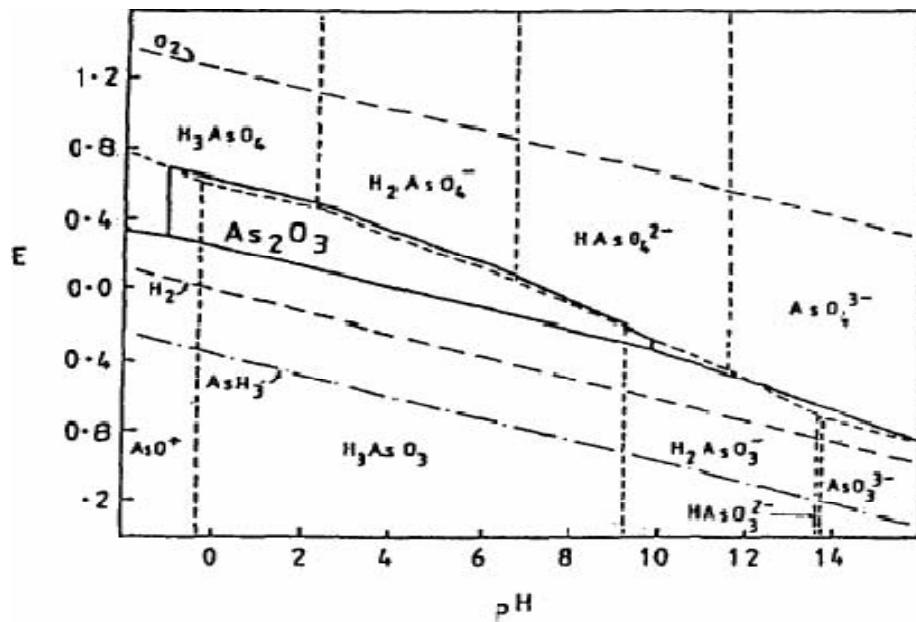


Figure 1: Potential pH diagram for the arsenic water system at unit activity of all species (Choong et al., 2007)

The concentration of arsenic in the earth's crust normally ranges from 1.5 - 5 mg/kg. Table 1 illustrates arsenic minerals in nature. Arsenopyrite, realgar, and orpiment minerals contain sulfide ores such as copper, lead, silver, and gold. The weathering process may release arsenic from sulfide ores in soil, surface water, groundwater and the atmosphere (Ning, 2002).

**Table 1. Major arsenic minerals commonly found in nature. (Gomez-Caminero et al., 2001).**

Mineral	Composition	Occurrence
Native arsenic	As	Hydrothermal veins
Proustite	Ag <sub>3</sub> AsS <sub>3</sub>	Generally one of the late Ag minerals in the sequence of primary deposition
Rammelsbergite	NiAs <sub>2</sub>	Commonly in mesothermal vein deposits
Safflorite	(Co, Fe)As <sub>2</sub>	Generally in mesothermal vein deposits
Seligmannite	PbCuAsS <sub>3</sub>	Occurs in hydrothermal veins
Smaltite	CoAs <sub>2</sub>	
Niccolite	NiAs	Vein deposits and norites
Realgar	AsS	Vein deposits, often associated with orpiment, clays and limestones, deposits from hot springs
Orpiment	As <sub>2</sub> S <sub>3</sub>	Hydrothermal veins, hot springs, volcanic sublimation product
Cobaltite	CoAsS	High-temperature deposits, metamorphic rocks
Arsenopyrite	FeAsS	The most abundant Arsenic mineral, dominantly mineral veins
Tennantite	(Cu,Fe) <sub>12</sub> As <sub>4</sub> S <sub>13</sub>	Hydrothermal veins
Enargite	Cu <sub>3</sub> AsS <sub>4</sub>	Hydrothermal veins
Arsenolite	As <sub>2</sub> O <sub>3</sub>	Secondary mineral formed by oxidation of arsenopyrite, native arsenic and other As minerals
Claudetite	As <sub>2</sub> O <sub>3</sub>	Secondary mineral formed by oxidation of realgar, arsenopyrite and other As minerals
Scorodite	FeAsO <sub>4</sub> ·2H <sub>2</sub> O	Secondary mineral
Annabergite	(Ni,Co) <sub>3</sub> (AsO <sub>4</sub> ) <sub>2</sub> ·8H <sub>2</sub> O	Secondary mineral

Hoernesite	$Mg_3(AsO_4)_2 \cdot 8H_2O$	Secondary mineral, smelter wastes
Haematolite	$(Mn,Mg)_4Al(AsO_4)$	
Conichalcite	$CaCu(AsO_4)(OH)$	Secondary mineral
Adamite	$Zn_2(OH)(AsO_4)$	Secondary mineral
Domeykite	$Cu_3As$	Found in vein and replacement deposits formed at moderate temperatures
Loellingite	$FeAs_2$	Found in mesothermal vein deposits
Pharmacosiderite	$Fe_3(AsO_4)_2(OH)_3 \cdot 5H_2O$	Oxidation product of arsenopyrite and other As minerals

The arsenic concentration in soil naturally ranges from 0.1 to 40 mg/kg, and the average concentration range from 5 to 6 mg/kg. Arsenic contaminates groundwater and surface water through processes such as erosion, dissolution and weathering (Figure 2). The geothermal process is also responsible for arsenic contamination in groundwater. Other natural sources that influence arsenic contamination include volcanic eruptions and forest fires. Volcanic activity is the principal natural source of arsenic emissions to the atmosphere, and it is predicted that 2,800 to 44,000 metric tons of arsenic emissions released due to volcanic activity annually (Ning, 2002).



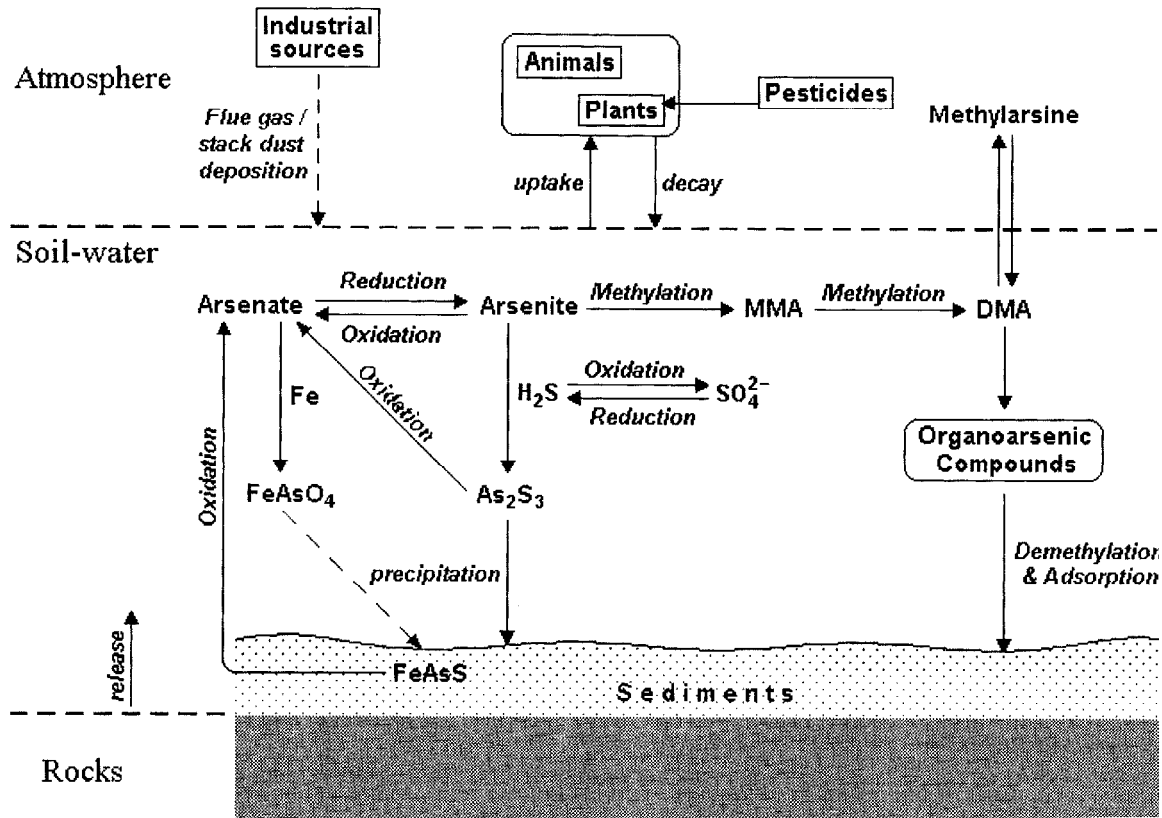


Figure 2. Arsenic cycle in the environment source (Saha, 2002).

Arsenic (V) and arsenic (III) are the most common forms of arsenic in ground and surface water. Different inorganic forms of arsenic oxides occur naturally in the environment ( $As_2O_3$ ,  $As_2O_5$ ) and sulfides ( $As_2S_3$ ,  $AsS$ ,  $HAsS_2$ ,  $HAsS_3^{3-}$ ). The inorganic forms of arsenic that are stable in oxygenated waters include arsenic acid As (V) species ( $H_3AsO_4$ ,  $H_3AsO_4^-$ ,  $H_3AsO_4^{2-}$  and  $AsO_4^{3-}$ ). In arsenic acid arsenic (III) is also stable as  $H_3AsO_3$  and  $H_2AsO_3^-$  under slightly reducing aqueous conditions. Arsenic transportation depends on different factors such as the oxidation state, pH, iron concentration, metal sulfide, sulfide concentration, oxidation and reduction potential, temperature, salinity and distribution and composition of biota (Ning, 2002). Arsenic concentration in surface water depends on some additional factors which include total suspended sediment, seasonal water flow volumes, time and day (Ning, 2002).

Sorption chemistry is also responsible for transportation of arsenic in surface water systems. If the pH and arsenic concentration in the water is high and the concentration of total suspended solids is low, the sorption process may be less important. On the other hand, pH, arsenic concentration is low and suspended sediment load is high arsenic present in suspended particle phase than dissolved phase. Arsenic is dispersed by wind and high flow scouring. "Diurnal changes as much as 21 % of arsenic concentration have been observed in river attributable to pH changes due to sunlight and photo synthesis" (Ning, 2002).

### 3. Natural distribution of arsenic:

Arsenic is the 20<sup>th</sup> richest element in the earth crust, and the 14<sup>th</sup> in sea water. Arsenic is an extraordinary crystal element comprising about five hundred thousandths of 1% (0.00005%) of the earth crust. The arsenic concentration in igneous and sedimentary rock is 2 mg/kg with different types of rocks containing different concentrations. Ranges from 0.5 - 2.5 mg/kg and higher concentrations were found in argillaceous sediments and phosphorites. Arsenic has been found to reduce marine sediment and may also co-precipitate with ironhydroxides and sulfides in sedimentary rock. Table 2 shows arsenic concentrations in some natural geochemical materials (Mandal and Suzuki, 2002).

Over 200 different minerals contain arsenic, of which 60% are arsenates, 20% sulfides and sulfosalts and another 20% are arsenides, arsenates, silicates and elemental arsenic. Arsenic concentration is categorized into three major classes: *Igneous, Sedimentary and Metamorphic rock* (Table 2).

**Table 2. Arsenic concentration in the three most common rock worldwide (Mandal and Suzuki, 2002).**

Materials	Arsenic (mg/kg)	Materials	Arsenic (mg/kg)
<i>Igneous</i>		<i>Sedimentary rocks</i>	
Acidic		Marine	
Rhyolite (extrusive)	3.2–5.4	Shale/claystone (nearshore)	4.0–25
Granite (intrusive)	0.18–15	Shale/claystone (offshore)	3.0–490
Intermediate		Carbonates	0.1–20.1
Latite, andesite, trachyte (extrusive)	0.5–5.8	Phosphorites	0.4–188

Diorite, granodiorite, syenite (intrusive)	0.09–13.4	Sandstone	0.6–9
Basalt (extrusive)	0.18–113	Non-marine Shales	3.0–12
Gabbro (intrusive)	0.06–28	Claystone	3.0–10
Ultrabasic		Recent sediments (marine)	
Peridotite, dunite, serpentinite	0.3–15.8	Lake	2.0–300
<i>Metamorphic rocks</i>		Clays	4.0–20
Quartzite	2.2–7.6	Carbonate	<1.0
Slate/phyllite	0.5–143	Stream/river	5.0–4000 (mineralized area)
Schist/gneiss	0.0–18.5	Soils	<0.1–97

**Table 3. Arsenic deposits in the world (Gomez-Caminero et al., 2001).**

Type of deposits	Arsenic mineral	Average arsenic concentration (mg/kg)	Location
Enargite-bearing copper–zinc–lead deposits	Enargite	1000 (0.1%)	United States, Argentina, Chile, Peru, Mexico, Republic of the Philippines, Spain, Yugoslavia, USSR
Arsenical pyritic copper deposits	Arsenopyrite, tennantite	40,000 (4%)	United States, Sweden, Federal Republic of Germany, Japan, Bangladesh, France, USSR
Native silver and nickel–cobalt arsenide bearing deposits	Smaltite, domeykite, safflorite, rammelsbergite, cobaltite, niccolite, loellingite, arsenopyrite	25,000 (2.5%)	Canada, Norway, Germany, Democratic Republic, Czechoslovakia
Arsenical gold deposits	Arsenopyrite, loellingite	<5000 (0.5%)	United States, Brazil, Canada, Republic of South Africa, Australia, USSR
Arsenic sulfide and arsenic sulfide gold deposits	Realgar, orpiment	2000 (0.2%)	United States, People's Republic of China
Arsenical tin deposits	Arsenopyrite	2000 (0.2%)	United States, Bolivia, Australia, Indonesia, Malaysia, Republic

Arsenical quartz, silver and lead–zinc deposits	Arsenopyrite	6000 (0.6%)	of South Africa United States, Canada, et al.
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Arsenic is found in fresh and groundwater in many countries around the world (Mandal and Suzuki, 2002). The concentration of arsenic varies in ground water across the globe (Table 4). The WHO has recommended that the maximum concentration should not exceed 10 µg/l. Sea water contains arsenic concentrations from 0.001 - 0.008 mg/l. Table 4 shows arsenic affected countries, sources of arsenic and concentrations of arsenic in water (Mandal and Suzuki, 2002).

**Table 4. Concentrations of arsenic in groundwater of the arsenic-affected countries (Mandal and Suzuki, 2002).**

Location	Arsenic source	Concentration (µg/l)
South-West Finland	Well waters; natural origin	17–980
New Jersey, USA	Well waters	1 (median) 1160 (maximum)
Western USA	Geochemical environments	48,000 (maximum)
South-west USA	Alluvial aquifers	16–62
Bangladesh	Well waters	10 -1000
Lagunera region, northern Mexico	Well waters	8–624
Shanxi, PR China	Well waters	0.03–1.41
Calcutta, India	Near pesticide production plant	<50–23,080
Fukuoka, Japan	Natural origin	0.001–0.293

In addition to geochemical factors, microbial agents can influence the oxidation state of arsenic in water. Different geochemical processes can transfer arsenic in aquatic systems. Table 5 describes arsenic concentration in different environmental media.

**Table 5: Arsenic concentration in environmental media (Ning, 2002).**

Environmental media	Arsenic concentration range
Air ng/m <sup>3</sup>	1.5-53
Rain from unpolluted ocean air µg/l	0.019
Rain from terrestrial air µg/l	0.46

Rivers $\mu\text{g/l}$	0.20-264
Lakes $\mu\text{g/l}$	0.38-1000
Ground well water $\mu\text{g/l}$	<1.0- >1000
Sea water $\mu\text{g/l}$	0.15-60
Soil $\text{mg/kg}$	0.1-1000
Stream /river sediment $\text{mg/kg}$	5.0-4000
Lake sediment $\text{mg/kg}$	2.0-300
Igneous rock $\text{mg/kg}$	0.3-113
Metamorphic Rock	0.0 -143
Sedimentary Rock	0.1-490
Biota(green algae) $\text{mg/kg}$	0.5-5.0

Arsenic is present in air, it exists in nature mainly absorbed on particulate matter. Arsenic in air is normally a mixture of arsenite and arsenate. The organic arsenic percentage in the air is very low except in areas of arsenic pesticide application (Mandal and Suzuki, 2002). Human exposure by arsenic is very low, normally the arsenic concentration in air is  $0.4\text{-}30\text{ ng/m}^3$ . The United States Environmental Protection Agency (USEPA) estimated that the average exposure in the United States (U.S) was  $6\text{ ng As/m}^3$ . Arsenic concentration depends on the particulate matter and relative vapor. In Europe, the arsenic concentration in the air is very low. Concentration depends on the region:  $0.2\text{ - }1.5\text{ ng/m}^3$  in rural areas,  $0.5\text{ - }3.0\text{ ng/m}^3$  in urban areas, and not higher than  $50\text{ ng/m}^3$  in industrial areas (Mandal and Suzuki, 2002).

In Bangladesh, groundwater contains high amounts of arsenic ( $50\text{-}2000\text{ }\mu\text{g/l}$ ) and has created a serious health hazard in human history. (Mandal and Suzuki, 2002). Figure 3 shows that arsenic comes from different sources (Aquifer, geothermal, coal, mining) and the amount of the global population at risk.

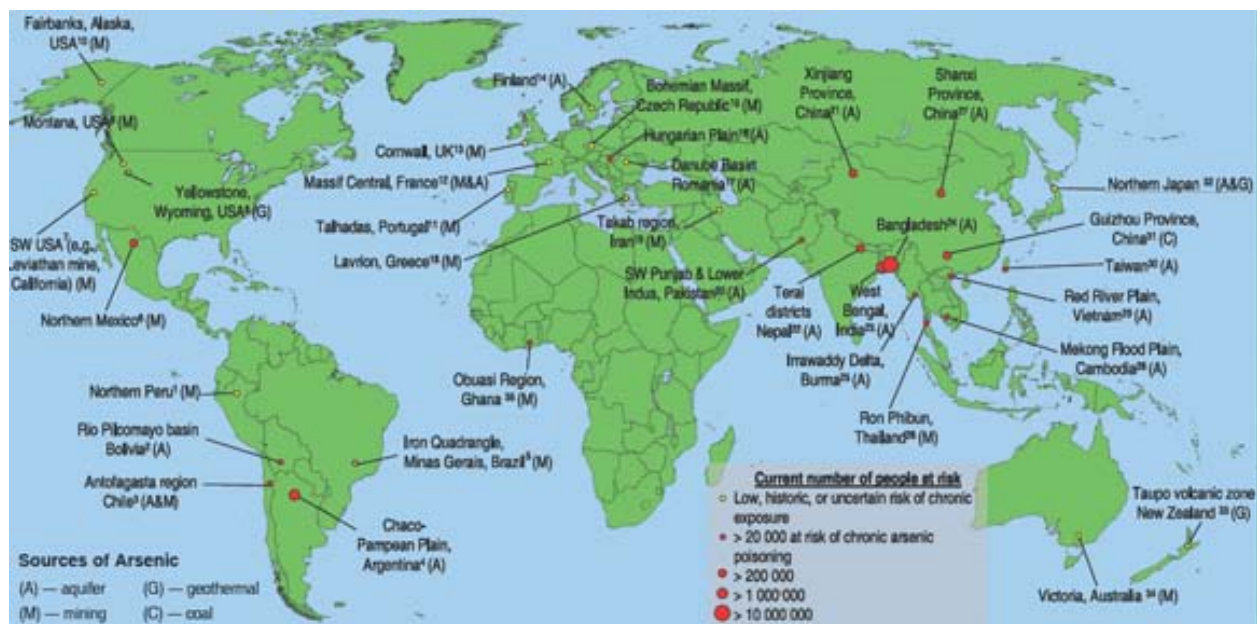


Figure 3: Sources of arsenic and people living under risk of arsenic contaminated water (Garelick and Jones, 2008).

#### 4. The effects of arsenic on human health:

Human exposed to arsenic through ingestion, inhalation, or skin absorption. High doses of arsenic may have acute toxic effects give gastrointestinal symptoms (poor appetite, vomiting, and diarrhea) and disturbed the fractions of the cardiovascular and nerve systems(Safiuddin, 2001).

The data collected by the department of public health , Non government organizations (NGOs) and private organizations have revealed that a large part of the population in Bangladesh are suffering from melanosis, leuco melanosis, keratosis, hyperkeratosis, gangrene and skin cancer (Safiuddin, 2001). Malanosis (93.5%) and caratosis (68.3%) are the most common diseases. Leuco-malanosis (39.1%) and hyper keratosis (37.6%) have been found in many cases (Safiuddin, 2001).

The extend of arsenic intoxicion depends on the type of arsenic compounds and concentration of arsenic in the human body (Safiuddin, 2001). It has been reported that 40 to 50 % of the ingested arsenic be retained in human body. The amount of arsenic contaminated water is very high in

Bangladesh, mainly in the rural areas. In Bangladesh most of the people live in rural areas, where the average water consumption is 5 liters per person/day. They also use arsenic contaminated water for cooking. Around 90 % of people have arsenic in their nail, hair, and urine above the normal level present in Table 6 (Safiuddin, 2001).

**Table 6. Arsenic level in hair, nails, skin scales and urine of among Bangladeshi rural area people (Safiuddin, 2001).**

Field Survey from August 1995 to February 2000	
Total Hair Samples Collected from 210 Arsenic Affected Villages	4386
Percentage of Samples Having Arsenic above Toxic Level	83.15%
Total Nail Samples Collected from 210 Arsenic Affected Villages	4321
Percentage of Samples Containing Arsenic above Normal Level	93.77%
Total Urine Samples Collected from 20 Arsenic Affected Villages	1084
Percentage of Samples Having Arsenic above Normal Level	95.11%
Total Samples of Skin Scales	705
Percentage of Samples Containing Arsenic above Toxic Level	97.44%
Field Survey from April 1999 to February 2000 (27 Days)	
Total Hair Samples	1054
Percentage of Hair Samples Having Arsenic above Toxic Level	89.35%
Total Nail Samples	1000
Percentage of Nail Samples Containing Arsenic above Normal Level	94%
Total Urine Samples	41
Percentage of Urine Samples Having Arsenic above Normal Level	97.50%
Total Samples of Skin Scales	115
Percentage of Samples Containing Arsenic above Toxic Level	100%

#### 4.1 Skin Cancer

A significant relationship between skin cancer and arsenic exposure has been observed (Rossman, 2004). The mechanism of action may involve effects on DNA methylation and DNA repair. It is associated with cancer of the skin and internal organs, as well as vascular disease (Rossman et al., 2004).

#### 4.2 Lung cancer

Lung cancer significantly increases with increased arsenic concentration in the body. Together with cigarette smoking arsenic exposure has a synergistic effect thus increasing the risk of lung cancer. An increased risk of lung cancer was associated with the high level of arsenic exposure via drinking water. Research revealed a decreased level of arsenic exposure also reduced lung cancer among smokers; the mortality rate of lung cancer reduces when arsenic concentration is lessened in drinking water (Hopenhayn-Rich et al., 1998).

#### 4.3 Effects on memory and intellectual function

The level of arsenic concentration in urine sample among Mexican children was inversely associated with Verbal intelligence quotient and long term memory. This study found an impact on long term memory and understanding capability of children. Children who drank water with an arsenic concentration of 50 mg/l have a lower performance of intellectual capability than children who drank water containing 5.5 mg/l of arsenic (Wasserman et al., 2004).

#### 4.4 Reproductive effects

Pregnancy complications are found due to chronic effects from ground water contamination with arsenic. A study found a positive correlation with arsenic concentration and fetal loss and premature delivery. Women who chronic exposure drank (less than 50  $\mu\text{g/l}$ ) arsenic in drinking water gave birth to children with a decrease birth weight (Chen et al., 2004).





Figure 4: Skin lesions in soles and hands due to arsenic poisoning from drinking water (Safiuddin, 2001)

Skin ailments such as those displayed in figure 4 are generally symptoms of continuous drinking of arsenic contaminated water and are symptoms of different skin diseases like hypopigmentation (white spots on the skin), hyper pigmentation (dark spot on the skin), keratosis and melanonin. More serious health problems such as skin cancer or cardiovascular and nervous affections are known to appear with a latency of 10 or more years (Zürich, 2007).

## 5. Problem and remediation methods of arsenic removal from drinking water:

Arsenic concentration in drinking water is different from place to place. In developing countries it is very expensive to remove arsenic from drinking water by central water treatment systems. People living in rural areas are mostly affected by arsenic contaminated in drinking water because of poor treatment facilities. Different technologies have been developed to remove arsenic from drinking water. Some technologies are not suitable for safe drinking water supply in small communities because of expensive have too maintenance costs.

Membrane technology is relatively new for water treatment. It is convenient for small and large scale water treatment facilities. In our experiment we use reverse osmosis system for arsenic water treatment and investigate to find out optimal conditions for better removal efficiency. Reverse osmosis systems can be prepared for small and large scale treatment unit with different

membranes. Reverse osmosis systems are very easy to operate, having low maintenance costs and can be used as a small scale treatment plant in rural areas.

## 6. Different membrane use for water treatment:

### 6.1 Ultrafiltration:

Ultrafiltration has been using for many different applications, including the field of drinking water treatment. It has used to remove pathogenic microorganisms, bacteria, viruses, colloids, turbidity, soluble macromolecules and natural organic matter. Ultrafiltration is used in lieu of coagulation/precipitation and media filtration pretreatment for reverse osmosis and nano filtration membranes (Segal and Dosoretz, 2011).

Supplying clean and, drinking water is an emerging challenge. Pollution originates from industry and commercial sources. Ultrafiltration has widely used for water treatment all over the world (Hank and Wyckoff, 2010).

Advantages of ultrafiltration (Hank and Wyckoff, 2010):

- Remove pathogen and turbidity in only one treatment step.
- Complete obstacle for bacteria, virus and parasites.
- Deactivated pathogens are fully removed in the water.
- Low operating effort: ultrafiltration systems made for small water treatment solutions are fully automated.
- Combined with activated carbon pre-filtration, it removes taste, odor, pesticides and residuals of antibiotics.
- Green technology: some ultrafiltration can be directly operated on solar power.
- No chemicals application to the water.

## 6.2 Microfiltration:

Microfiltration removes contaminants from the fluid micro porous membrane. The normal microfiltration porous range is 0.1 - 10 ( $\mu\text{m}$ ). Micro filtration is different from nanofiltration and reverse osmosis. Water passes through the membrane with low pressure to high pressure in two systems. Microfiltration is normally used for drinking water treatment. It effectively remove major pathogens and contaminants such as *Giardia lamblia* cysts, *cryptosporidium* and large bacteria. For this application the filter has to be rated for 0.2  $\mu\text{m}$  or less (Dharmappa and Hagare, 1999).

Advantages of microfiltration for water treatment (Dharmappa and Hagare, 1999).

- Bacterial decontamination with little or no chemical addition.
- Water treatment system is very compact.
- Ultra pure water is used for industrial purposes.
- Microfiltration water treatment system might be economical for small water treatment system.
- In some different cases sludge treatment can be considerably lowest amount.
- High and stable product water quality.

## 6.3 Nanofiltration:

Nanofiltration membranes can remove different valence ions and different molecular weight organic molecules from the water, it depends on membrane charge and pore size. Nanofiltration is used for purification, biochemical substance separation, waste water reclamation and water softening (Ji et al., 2011).

Nanofiltration membrane charged both positively and, negatively. Nanofiltration membranes are made from polyamids, sulfonated polysulfone and sulfonated polyphenyleneoxides. This compounds have used for negatively charged membrane. Positively charge membrane is more effective in removing multi-valent cations and remove amino acids below their isoelectric point. Positively charged Nanofiltration have been developed in recent years (Ji et al., 2011).

#### 6.4 Reverse Osmosis System:

Reverse osmosis is commonly used to reduce suspended solid particles and dissolved matter from water. Table 7 shows the common contaminants removed by reverse osmosis, including harmful pesticides (Dvorak, 2008)

**Table 7. Common contaminants removal by reverse osmosis system (Dvorak, 2008).**

Ions and metals	Arsenic,aluminum,barium,cadmium,calcium,chlorine,chromium,copper, fluoride,iron,lead,magnesium,manganese,mercury,nitrate,potassium, radium, selenium, silver, sodium, sulfate, zinc
Particles	Asbestos, protozoan cysts, cryptosporidium
Pesticides	Endrin, heptachlor. lindane, pentachlorophenol

Not all contaminants can be removed by reverse osmosis. Dissolved gases like hydrogen sulfide, will pass through the membrane. Some pesticides, solvents and volatile organic chemical compounds cannot be removed by a reverse osmosis system. Removal efficiency of reverse osmosis depends on the concentration of the contaminants and chemical properties, characteristics of the membrane and operating system of the reverse osmosis system (Dvorak, 2008).

Reverse osmosis system contain two solutions of different chemical concentrations separated by a semi permeable membrane. Water passes through the membrane from the dilute to the more concentrated solution due to a different pressure level. This pressure is called osmotic pressure and the process is called osmosis (Dvorak, 2008).

In a reverse osmosis system, pressure is applied to the concentrated side of the membrane. This force applies to the osmotic process in a reverse way. For this reason it is called the reverse osmosis system. Figure 5 shows the reverse osmosis working principal. After treatment treated water is collected in clean tank and the contaminants washed away as reject water (Dvorak, 2008).

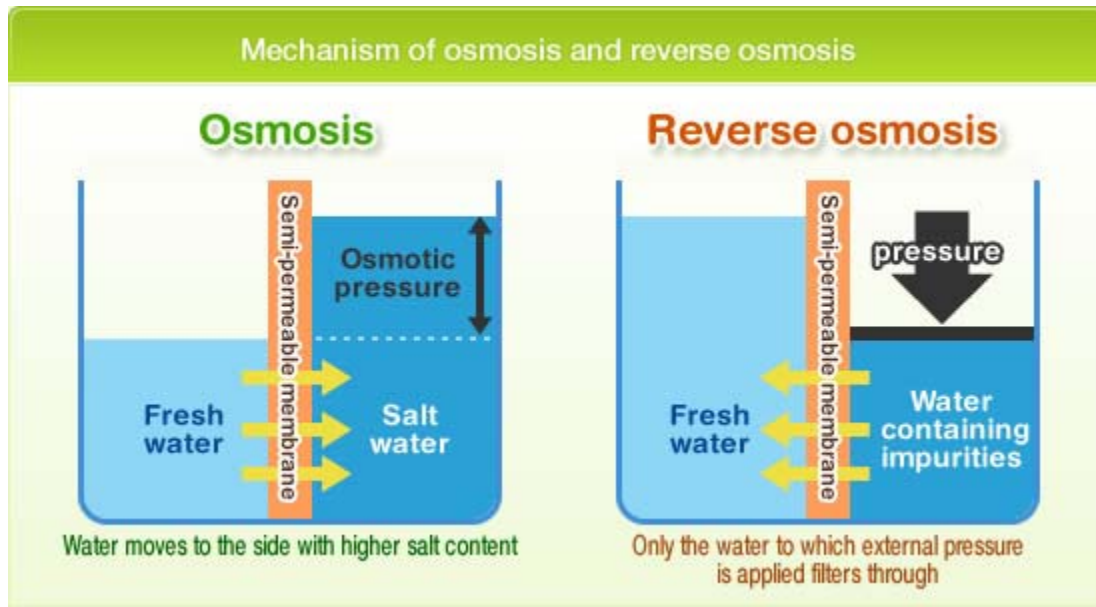


Figure 5: Osmosis and reverse osmosis main working principal (HITACHI, 2012)

When reverse osmosis is used for water treatment, a portion of water passes through the membrane. Sometimes water contains higher amounts of minerals than it did prior to entering the system. If the minerals are soluble then they will pass through the membrane to the flash water. Otherwise no soluble minerals may clog to membrane. Table 8 shows some substance that may cause clogging (Dvorak, 2008).

**Table 8: Problem that may arise during water treatment in reverse osmosis system (Fisher et al., 2008).**

Substances	Problem may arise when
Alkalinity	"If alkalinity exceeds 1000 ppm and the majority of the alkalinity is due to carbonate rather than bicarbonate, materials could precipitate"
Barium	"If barium concentrations exceed 1.5 ppm, barite (barium sulfate) precipitate may form"
Boron	"Water containing more than 50 ppm boron and an alkalinity of greater than 1000 ppm may form precipitates"
Calcium and Magnesium	"If the pH of the membrane water is greater than 10 and the combined calcium and magnesium content exceeds 60 ppm, precipitates may form"

Hardness	"Hardness is a measure of the combined total concentration of calcium, magnesium, and strontium in water. If pH exceeds 10, alkalinity exceeds 500 ppm, and hardness exceeds 150ppm, precipitates may form"
Iron and Manganese	"Combined concentrations of iron and manganese greater than 25 ppm can lead to formation of precipitates"
Silica	"The concentration of silica should be less than 100 ppm"
Sulfate	"If water has a pH greater than 10 and contains sulfate greater than 50 ppm, it may produce a precipitate"

## 7. Materials, methods and experimental design:

Arsenic (III) and arsenic (V) were mixed with Laboratory tap water and prepared arsenic contaminated water. All experiments were implemented in the department of Mathematical science and technology (IMT) and in the laboratory of Plant science (IPM) department laboratory, Norwegian University of Life Science. Malthe Winje Company, Norway installed the Reverse osmosis system. In the experiments all pipes were connected to the reverse osmosis system, production water tank and reject water collection tank. Arsenic (III) and arsenic (V) samples were prepared separately for each experiment. This experiment was divided into three steps. The first step was designed for arsenic (V). Arsenic (V) sample was prepared by  $\text{Na}_2\text{HAsO}_4 \times 7\text{H}_2\text{O}$  (Sodium Hydrogen arsenate hepta hydrate, 98%) case number -10048-95-0, formula weight-312.01 g/mole. In this experiment 500 liters arsenic (V) sample was prepared in two different concentrations 200  $\mu\text{g/l}$  and 50  $\mu\text{g/l}$  (Arsenic mixed with water), in two different pH condition pH=6 and pH=8. we calculated pressure for reverse osmosis system in production water flow rate (l/h), two production water flow rate were applied 50 l/h and 200 l/h. Arsenic sample were prepared in tank and adjusted so that the water was at pH=6 and pH=8 according to experimental design. The pH was adjusted by Nitric acid ( $\text{HNO}_3$ ) and Sodium Hydroxide (NaOH). Table 11 shows the experimental design for arsenic (V).

**Table 11. Experimental design for arsenic (V):**

Initial arsenic sample water (l)	Arsenic concentration ( $\mu\text{g/l}$ )	Flow rate in RO system (l/h)	pH	Sampling time (min)
500	50	50	6	20 40
500	50	50	8	20 40
500	50	200	6	20 40
500	50	200	8	20 40
500	200	50	6	20 40
500	200	50	8	20 40
500	200	200	6	20 40
500	200	200	8	20 40

In this experiment arsenic contaminated water was treated by the reverse osmosis system. Treated water was collected in plastic tubes for further analysis by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Sixteen samples were taken from treated water and eight samples were taken for initial concentration measurement and also reserve samples were taken from every experiment. In the beginning of the treatment when the reverse osmosis system was started this time first five minutes were taken for reverse osmosis system flushed by arsenic sample water, After cleaning, arsenic contaminated reject water was collected in tank for treatment by  $\text{K}_2\text{S}$  (Potassium sulfide).

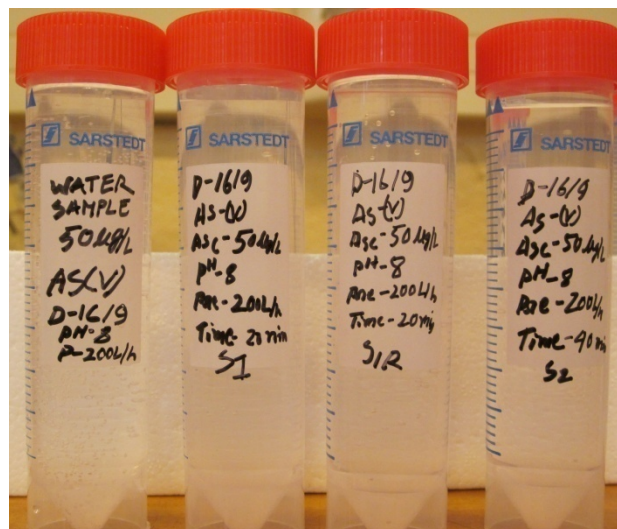


Figure 6: Picture of reverse osmosis system and Treated water collection tube for ICP-MS analysis.

The second step of the experiment was designed for arsenic (III). Arsenic (III) samples were prepared by  $\text{Na}_2\text{AsO}_2$  (Sodium arsenite solution containing arsenic trioxide, 100%) 0.05 mol/l (0.1N). In the experiment 500 liters arsenic (III) sample was prepared in two different concentrations 200  $\mu\text{g/l}$  and 50  $\mu\text{g/l}$  (Arsenic mixed with water), at two different pH conditions pH=6 and pH=8. In the reverse osmosis system two production water flow rate were applied (50 l/h and 200 l/h). Arsenic contaminated water was adjust pH=6 and pH=8 according to experimental design. The pH was maintained by Nitric acid ( $\text{HNO}_3$ ) and Sodium Hydroxide ( $\text{NaOH}$ ).



**Table 12. Experimental design for arsenic (III):**

Arsenic sample water (l)	Arsenic concentration ( $\mu\text{g/l}$ )	Flow rate in RO system (l/h)	pH	Sampling time (min)
500	50	50	6	20 40
500	50	50	8	20 40
500	50	200	6	20 40
500	50	200	8	20 40
500	200	50	6	20 40
500	200	50	8	20 40
500	200	200	6	20 40
500	200	200	8	20 40

In these experiments all steps were the same as the first step of experiment. After treating, arsenic (III) contaminated water sample was taken for ICP-MS analysis. Sixteen samples were taken from treated water and eight samples for initial concentration measurement and reserve sample taken from every experiment. Reject water was collected in a tank for treatment by  $\text{K}_2\text{S}$  (Potassium sulfide).



Figure 7: Arsenic contaminated reject water collection tank.

After finishing two steps experimented water samples were analyzed by ICP-MS, According to the results, arsenic concentration in treated water was different for Arsenic (V) and Arsenic (III). Arsenic (V) treated water concentration was less than  $1\mu\text{g/l}$ , This concentration is accepted according to the WHO limit. The reverse osmosis system showed almost 99% removal efficiency for arsenic (V) but the arsenic (III) concentration in treated water is higher than the WHO guideline, the highest removal efficiency for arsenic (III) was (80%) and not suitable for drinking water as per WHO guidelines. Therefore we decided on a new research plan for arsenic (III). In this step we prepared arsenic (III) solution like before. We prepared 500 liters of arsenic contaminated water for each experiment. In this step arsenic concentration was  $200\mu\text{g/l}$ ,  $50\mu\text{g/l}$  and  $300\mu\text{g/l}$ . we applies three pH condition  $\text{pH}=6$ ,  $\text{pH}=8$  and  $\text{pH}=10$ , Production water flow rate in the reverse osmosis system was 200 l/h previous results show that a water flow rate 200 l/h is better than 50 l/h. In this experiment arsenic (III) contaminated water passed through reverse osmosis system twice. The first time arsenic contaminated water passed through the membrane with one pH and a production water flow rate 200 l/h. After treatment pH was adjusted to initial pH conditions and two different production water flow rate 200 l/h and 50 l/h. All arsenic contaminated water was treated by the reverse osmosis system two times. We collected water in plastic tubes for ICP-MS further analysis.

**Table 13. Experimental design for arsenic (III) second step:**

Arsenic sample water (l)	Arsenic concentration (µg/l)	pH	Water flow rate in RO system (l/h)	Adjust pH after RO1	Water flow rate in RO system (l/h)	Sampling time(Min)
500	50	6	200	6	200	20
					50	40
500	50	8	200	8	200	20
					50	40
500	200	6	200	6	200	20
					50	40
500	200	8	200	8	200	20
					50	40
500	200	10	200	10	200	20
					50	40
500	300	10	200	10	200	20
					50	40

### 7.1 Description of membrane that use in experiment

Membrane company name: DWO FLIMTEC Membranes.

Membrane brand name: DOW FLIMTEC BW30-4040.

**Table 9.Product specification:**

Product	Part number	Feed spacer thickness (mil)	Permeate flow rate gpd (m <sup>3</sup> /d)	Stabilized salt rejection (%)
BW30-4040	80783	34	2400(9.1)	99.5

1. Permeate flow and salt rejection based on the following test conditions: 2,000 ppm NaCl, applied flow rate: 150 psig (10.3 bar) for LE-4040 and 225 psig (15.5 bar) for BW30-4040 and BW30-2540, 77<sup>0</sup>F (22<sup>0</sup>C) and 15% recovery.
2. Permeate flows for individual elements may vary +/- 20%.

- For the purpose of improvement, specifications may be updated periodically.

**Table 10. Operating limit of the reverse osmosis system.**

Membrane type	Polyamide Thin-Film Composite
Maximum Operating Temperature	113 <sup>0</sup> F(45 <sup>0</sup> C)
Maximum operating pressure	600 psi(41 Bar)
Maximum feed flow rate 4040 elements	16gpm(3.6m <sup>3</sup> /h)
Maximum pressure drop	15psi(I Bar)
pH range continuous operation	2-11
pH range short time cleaning	1-13
Maximum feed silt density index	SDI5
Free chlorine tolerance	<0.1ppm

## 7.2 Statistical analysis

Minitab 16 was used for statistical analysis. A generalized linear model (GLM) from Analysis of variance (ANOVA) test with 95% significant level was tested for the treatments for all the parameters. The variables involved in this test were arsenic water concentration, reverse osmosis production water flow rate, and arsenic contaminated water pH.

## 7.3 Materials use for experiment

The reverse osmosis system with connecting pipe , Sodium arsenate solution, sodium hydrogen arsenate hepta hydrate, two 500 litres capacity tanks, Nitric acid, sodium hydroxide, pH meter, Rapid chemical mixer, 16 watt electric cable. Plastic tube, ICP-MS,

## 8. Results

According to ICP-MS results for arsenic (V) concentration in treated water, we found 99% removal efficiency in several treatment applications. (Appendix A) shows the treatment efficiency in different treatment applications. Eleven treatment applications show the highest removal efficiency (99%), and the lowest removal efficiency was 95%. Arsenic concentration in treated water is acceptable according to WHO guidelines.

A significant relationship was seen between treated water arsenic (V) concentration pH and water flow rate, No significant relationship was found with initial concentration of arsenic (V).

**Table 14. Arsenic concentration with three treatment factors.**

Factor	Sample Number	Mean±SD	DF	CV	F	P
Concentration	16	125±77	1	61.97	0.54	0.475
pH	16	7 ±1	1	61.97	6.66	0.024
Water flow rate	16	125±77	1	14.75	7.99	0.015

All values are mean ± SD and significant level measure at  $p < 0.05$ .  $R^2 = 55.9\%$ ,  $n = 16$

A significant relationship was obtained with pH and treated water concentration ( $P < 0.05$ ), and also pH with water flow rate ( $P < 0.05$ ). However initial arsenic concentration and treated water concentration was not significant (Table 14).

Figure 8 shows the percentage of efficiency in two pH conditions with two different water flow rate. Sampled water at pH=8 showed a higher percentage of removal efficiency than sampled water at pH=6. Water flow rate 200 l/h is more efficient than water flow rate 50 l/h. Each sample number shows the removal efficiency of two pH and one water flow rate condition.

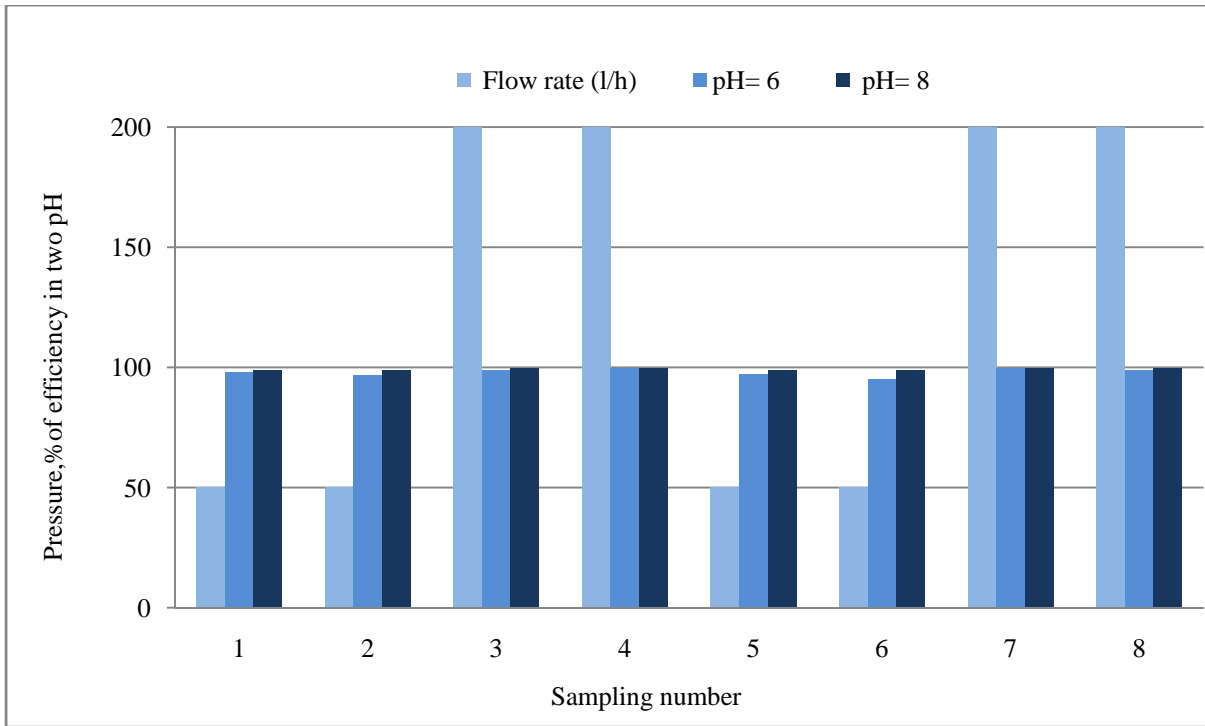


Figure 8: Arsenic (V) percentage of removal efficiency (pH=6, pH=8) at two different flow rates.

Figure 9 shows the removal efficiency in two sampled waters at two different pH. Each sample number shows the percentage of removal efficiency rate of two pH condition.

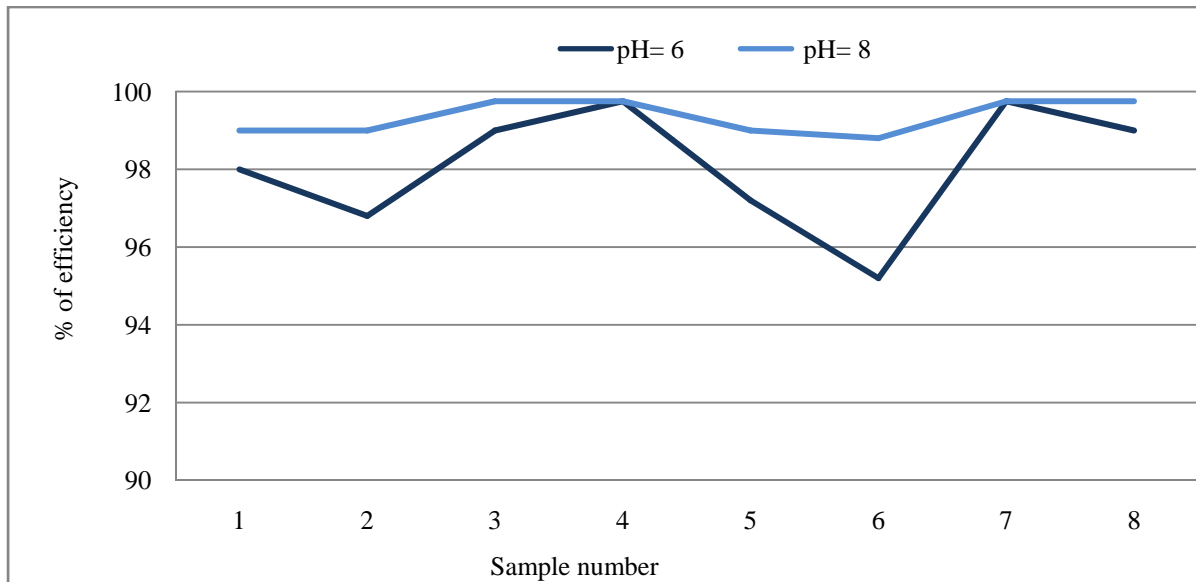


Figure 9: Arsenic (V) removal efficiency at pH=6 and pH=8 level.

In the experimental design two sets of experiments were performed for arsenic (III). Four treatment conditions had the highest removal efficiency of 80% and one had the lowest (54%) removal efficiency (Appendix B). Sixteen different treatment applications were performed.

A significant relationship was obtained between treated water arsenic (III) concentration, initial concentration and water flow rate. No significant relationship was found between pH and treated water concentration.

**Table 15. Treated water arsenic concentration tested with three treatment factors.**

Factor	Number	Mean±SD	DF	CV	F	P
Concentration	16	125±77	1	61.97	139.21	0.000
pH	16	7±1	1	61.97	1.78	0.207
Water flow rate	16	125±77	1	14.75	20.69	0.001

All values are mean ± SD and significant level measure at  $p < 0.05$ .  $R^2 = 93.09\%$ ,  $n = 16$

A significant relationship was found ( $P < 0.05$ ) with initial arsenic (III) concentration and treated water arsenic (III) concentration. A significant relationship with water flow rate and treated water concentration was also found, ( $P < 0.05$ ). No significant relationship was observed with pH and treated water arsenic concentration (Table 15).

Figure 10 shows the percentage of arsenic (III) removal efficiency in two pH conditions with two different water flow rate. Water flow rate at 200 l/h was more efficient than water flow rate at 50 l/h. Each sample number shows that removal efficiency of two different pH and one water flow rate condition.

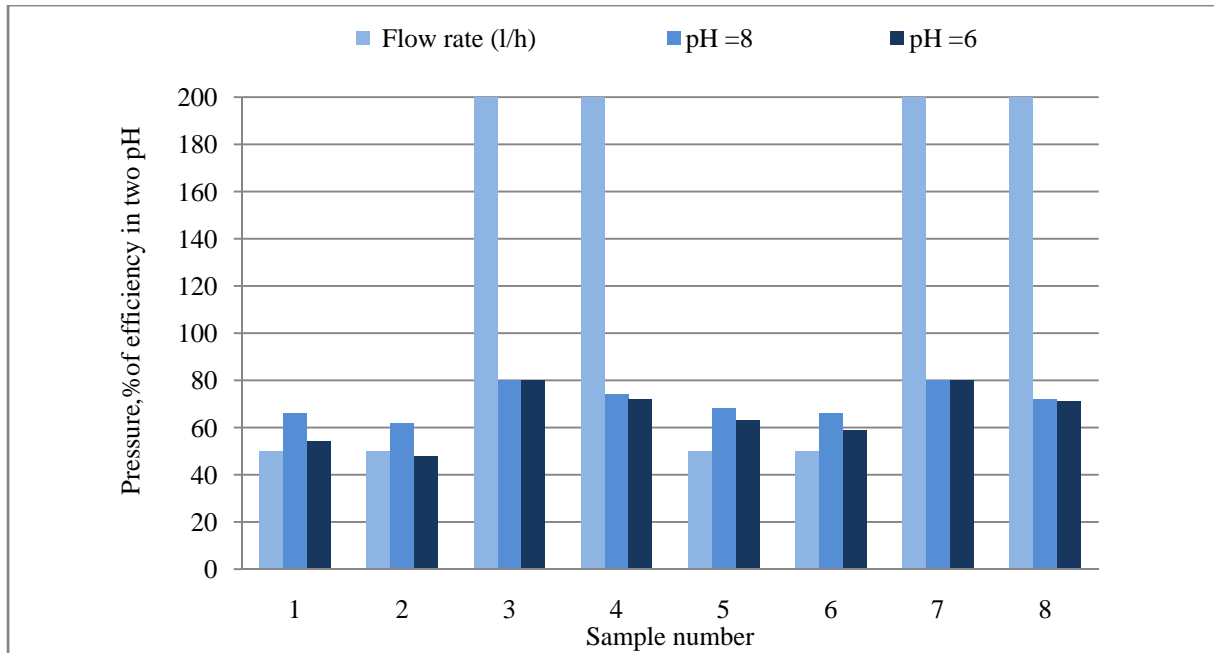


Figure 10: Removal efficiency of arsenic (III) percentage in pH=6 and pH=8, two different flow rates.

Water flow rate at 200 l/h showed more removal efficiency than water flow rate at 50 l/h, treated water arsenic (III) concentration depends on initial arsenic (III) concentration. Figure 11 shows the removal efficiency of two different water flow rate.

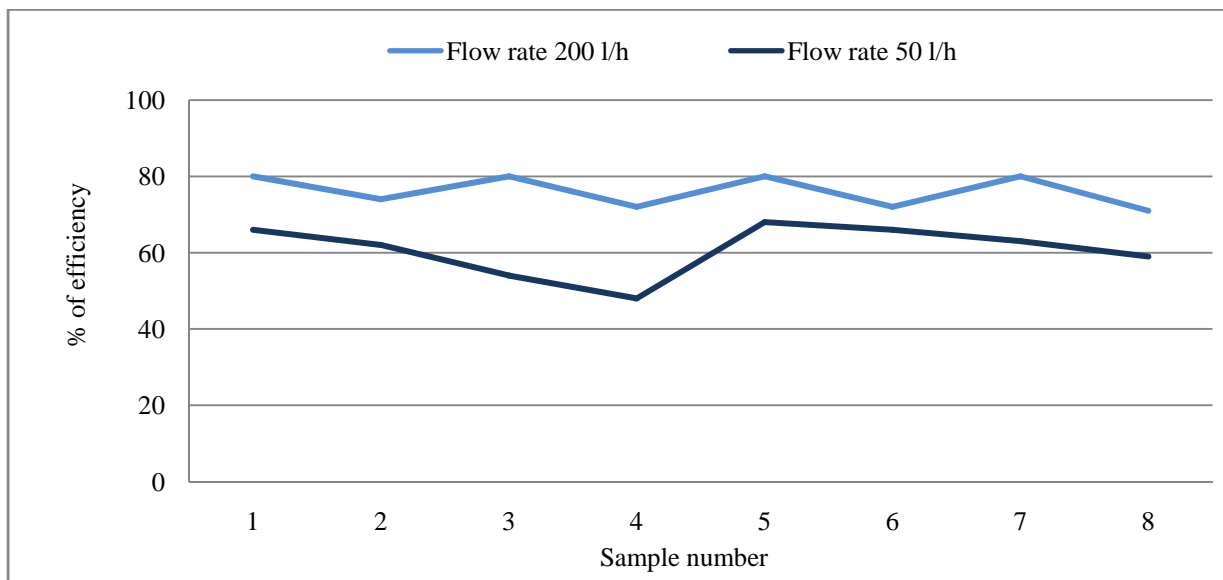


Figure 11. Arsenic (III) removal efficiency of two different water flow rates.



In the last step we performed two times treatment for arsenic (III). Highest removal efficiency was found in sample water at pH=10 condition. 99% removal efficiency was found in pH=10 level and lowest removal efficiency was found at pH=6 condition. (Appendix C)

We observed highest (99%) removal efficiency and lowest (76%) removal efficiency. In the first step flow rate was fixed for treatment.

Results showed a significant relationship of treated water arsenic (III) concentration with initial concentration and pH. There is no significant relationship between water flow rate and treated water arsenic concentration.

**Table 16. Arsenic concentration shows with three treatment factors.**

Factor	Number	Mean±SD	DF	CV	F	P
Concentration	10	140±77	1	55.33	23.55	0.005
pH	10	7.6±1.57	2	20.76	12.31	0.012
Water flow rate	10	125±79	1	63.25	5.16	0.072

All values are mean ± SD and significant level measure at P<0.05. R<sup>2</sup>=88.86%, n=10

A significant relationship was found with Initial concentration and treated water concentration (P<0.05). On the other hand there is no significant relationship with water flow rate. Figure 10 shows the percentage of arsenic (III) removal efficiency in three different pH conditions. Water at pH=10 condition had a better removal efficiency compared to the other two pH.

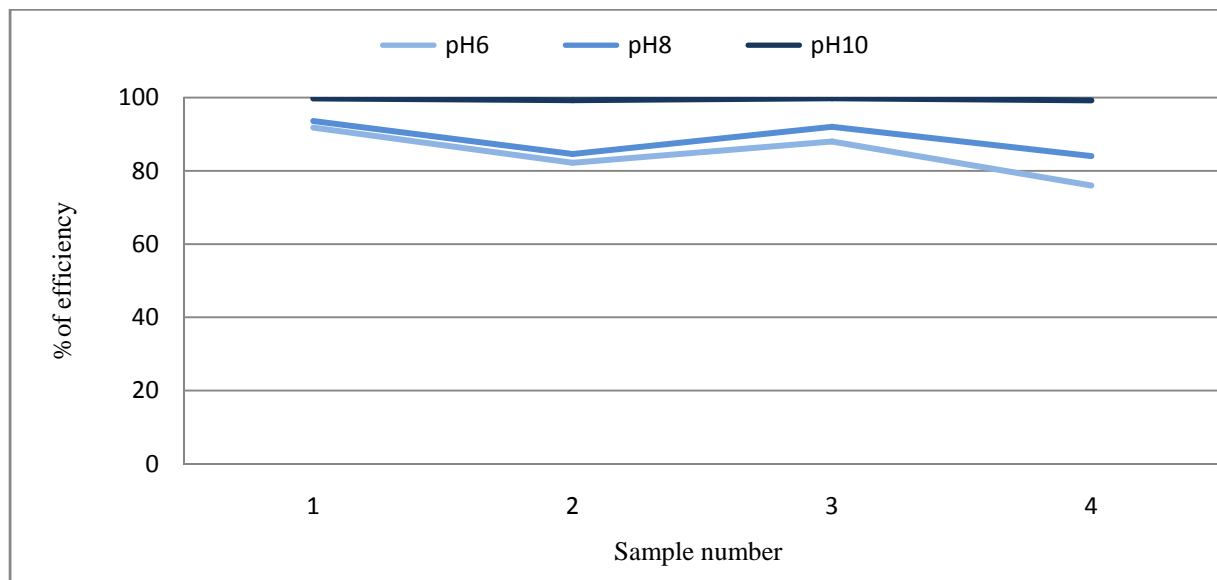


Figure 12: Arsenic (III) removal efficiency in three different pH conditions.

Figure 13 shows that removal efficiency was calculated with combined removal % of reverse osmosis 1 (RO1) and reverse osmosis 2 (RO2). While pH=10 RO1 efficiency is higher than RO2 compared to other pH condition. Each sample number represents combined arsenic (III) removal efficiency.

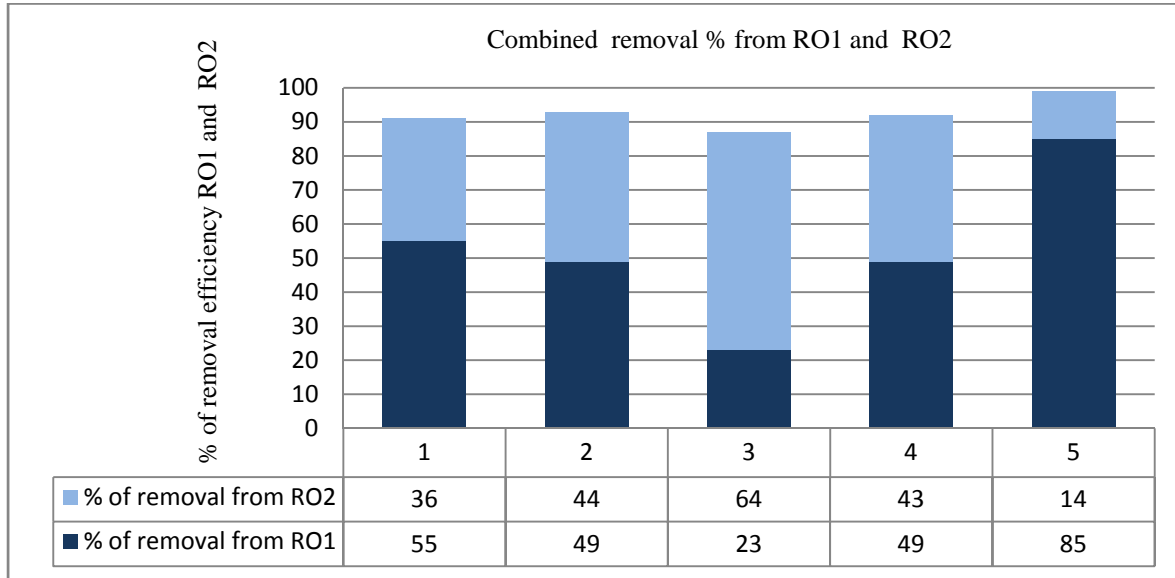


Figure13: The combined removal percentage of arsenic (III) RO1 and RO2.

## 9. Discussion

Our results suggest that the reverse osmosis system is significant to remove arsenic from drinking water. The reverse osmosis system and the membrane are very efficient for removing arsenic from drinking water. Removal efficiency level was 99% for arsenic (V) and 99% for arsenic (III) in different treatment applications.

Arsenic concentration is responsible for removal efficiency of arsenic (III) compared to arsenic (V). There are no statistically significant relationships with removal efficiency of arsenic (V). Arsenic (III) is more soluble and neutral charged in water. Arsenic (V) is negatively charged and less soluble in water. Arsenic (III) also becomes charged with higher pH condition. Arsenic (V) feed water concentration used 2000 µg/l and arsenic (III) used different concentration for treatment and pre oxidation process recommended for better removal efficiency (Geucke et al.,

2008). Organic carbon, chlorine, Monochloramine, potassium permanganate used for pre oxidation (Ning, 2002). In our experiment we got very good efficiency rate for arsenic (III) without any pre oxidation step. Further research is needed to investigate higher concentration removal efficiency.

Production of water by the reverse osmosis system depends on production water flow rate. Removal efficiency is higher for arsenic (III) and arsenic (V) at higher production water flow rate. Production water flow rate is more significant for arsenic (III) efficiency rate compared to arsenic (V).

Removal efficiency is also dependent on in arsenic contaminated water pH. The pH has significant effects on removal efficiency from water. Different pH conditions have different removal efficiencies. According to the experiments, arsenic (III) removal is more depended on pH than arsenic (V). That gradually increases pH in contaminated water and gradually increases removal efficiency. Experiments show solution pH affects the removal of arsenic (III) because most of the arsenic (III) exists in monovalent anion from pH=10, while most of the arsenic (III) exists in a neutral molecule at pH=6 and pH=8. The pH range was applied from 3 - 10 for arsenic (III) removal in two different membranes (Kang, 2000), For ES-10 membrane removal efficiency was 75% and NTR-729HF membrane removal efficiency was 43%. In our experiment arsenic (III) removal efficiency was 99% in pH=10 condition with BW30-4040.

Studies report arsenic removal ranging between 40-90% without specifying the arsenic species removed (Gomez-Caminero et al., 2001). Another pilot study reported arsenic (V) removal at 96-99% and arsenic (III) removal at 46-84%. A second pilot study (Gomez-Caminero et al., 2001) reported that arsenic (III) reduction at 73%, Bench – scale studies with reverse osmosis membrane shows arsenic (V) reduction at 88-96% and arsenic (III) reduction at only 5%.

In our experiment arsenic removal rate is more than 99% from 400 µg/l for both arsenic (III) and arsenic (V). The DOW FILMTEC (BW30-4040) membrane is efficient for arsenic removal from drinking water. This membrane has also removed other ions from drinking water. It may prove efficient at removing heavy metals from waste water and removing harmful microorganisms from drinking water. Further research is needed in this regard.

## 10. Conclusions

Arsenic rejection is significantly higher from the contaminated water. Arsenic (V) and arsenic (III) were very efficiently removed by reverse osmosis. The removal of arsenic was strongly affected by the solution pH, especially arsenic (III). Two time's arsenic contaminated water pass through the membrane and the removal efficiency rate 99%. When dealing with ground water high values of arsenic (III), a higher pH adjustment might be recommended. It was demonstrated that pH control for sample water is essential for the successful removal of arsenic compounds. Practical processes can be developed with reverse osmosis to remove all major species of arsenic from drinking water. Further studies are needed in the characterization of the Arsenic species being treated and in preparing a suitable design of the reverse osmosis process to match the demand. Reverse osmosis systems can produce safe drinking water for arsenic affected areas all over the world.

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## Appendix

Appendix A. Table shows the arsenic (V) concentration in treated water and % of removal efficiency.

Initial arsenic concentration (µg/l)	Water flow rate (l/h)	pH	Sampling time (min)	Arsenic concentration in treated water (µg/l)	% of efficiency
50	50	6	20	1	98
50	50	6	40	1.6	96
50	50	8	20	<0.5	99
50	50	8	40	<0.5	99
50	200	6	20	0.7	99
50	200	6	40	<0.5	99
50	200	8	20	<0.5	99
50	200	8	40	<0.5	99
200	50	6	20	1.4	97
200	50	6	40	2.4	95
200	50	8	20	<0.5	99
200	50	8	40	0.6	98
200	200	6	20	<0.5	99
200	200	6	40	0.6	99
200	200	8	20	<0.5	99
200	200	8	40	<0.5	99



Appendix B. Table shows the arsenic (III) concentration in treated water and % of removal efficiency.

Initial arsenic Concentration ( $\mu\text{g/l}$ )	Water flow rate (l/h)	pH	Sampling time(min)	Arsenic concentration in treated water( $\mu\text{g/l}$ )	% of removal efficiency
50	50	6	20	23	54
50	50	6	40	26	48
50	50	8	20	17	66
50	50	8	40	19	62
50	200	6	20	10	80
50	200	6	40	14	72
50	200	8	20	10	80
50	200	8	40	13	74
200	50	6	20	73	63
200	50	6	40	82	59
200	50	8	20	63	68
200	50	8	40	68	66
200	200	6	20	39	80
200	200	6	40	57	71
200	200	8	20	40	80
200	200	8	40	55	72

Appendix C: Table shows the arsenic (III) concentration in treated water and % of removal efficiency after two times treated by Reverse osmosis system.

Arsenic concentration (µg/l)	pH	Water flow rate in RO unit (l/h)	Adjust pH after RO1	Water flow rate unit (l/h)	As(III) concentration in treated water (µg/l)	% of removal efficiency
50	6	200	6	200	4.1	91
				50	8.9	82
50	8	200	8	200	3.2	93
				50	7.7	84
200	6	200	6	200	24	88
				50	48	76
200	8	200	8	200	16	92
				50	32	84
200	10	200	10	200	0.5	99
				50	1.5	99
300	10	200	10	200	0.5	99
				50	1	99