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Removal of Arsenic from Water by Electrolysis

by

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The undersigned certify that they have read, and recommended to the institute of engineering for acceptance, a project report entitled “Removal of Arsenic in water by Electrolysis”, submitted my MrPurushottam Sharma in partial fulfilment of the requirement for the degree of Master of Science in Environmental Engineering.

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ABSTRACT

Arsenic is a toxic element which occurs in water due to mineral dissolution. Arsenic occurs in water mainly in the form of arsenite [As(III)] and arsenate [As(V)] . Maximum allowable limit for arsenic in drinking water is 0.01 mg/L as per WHO guidelines. In Nepal the most severe outbreak of arsenic poisoning have been associated with ground water in the Terai Region, including Bangladesh and West Bengal (Eastern India) where an estimated total of 120 million people are at risk.

The purpose of this study was to investigate the effects of the operating parameters, such as pH, treatment time, and inter-electrode spacing on arsenic removal from drinking water in the electrolysis process.

The removal of arsenic increased with decreasing inter-electrode spacing. Better removal of arsenic was observed with increasing current passing time. With increase in pH, the removal of arsenic increases.

Using the inter electrode spacing 2cm, pH 7 and current passing time 20 minutes with the DC power source of 2A/12V with iron and aluminum electrodes, the concentration of arsenic in natural water samples after electrolysis was reduced below the WHO standard for drinking water.

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List of abbreviation

NASC	National Arsenic Steering Committee
NSSC	National Sanitary Steering Committee
WHO	World Health Organization
NRC	National Research Council
DC	Direct Current
DWIDP	Department of Water Induced Disaster prevention
ENPHO	Environment and Public Health Organization
µg/L	micrograms per liter
ppm	Parts Per Million

CHAPTER 1

1.0 INTRODUCTION

1.1 Background

Arsenic is metalloid found in rocks, soil, natural water, organisms. Being a transitional reactive element, it forms chemical and organic complexes together with other metals including iron, carbon, sulphur and oxygen. Arsenic is commonly used in glass making and termite control, as a pesticide and herbicide. (NASC, 2011)

Arsenic (atomic number 33) is ubiquitous and ranks 20th in natural abundance, comprising about 0.00005% of the earth's crust, 14th in the sea water, and 12th in the human body. It's concentration in most rocks ranges from 0.5 to 2.5 mg/kg. It is a silver-grey brittle crystalline solid with atomic weight 74.9, specific gravity 5.73, melting point 817 °C (at 28atm) and sublimation point at 613°C. (Pendias et al., 2000 and Mandal et al., 2004). Existence of arsenic in chemistry and nature is shown in Figure 1.1

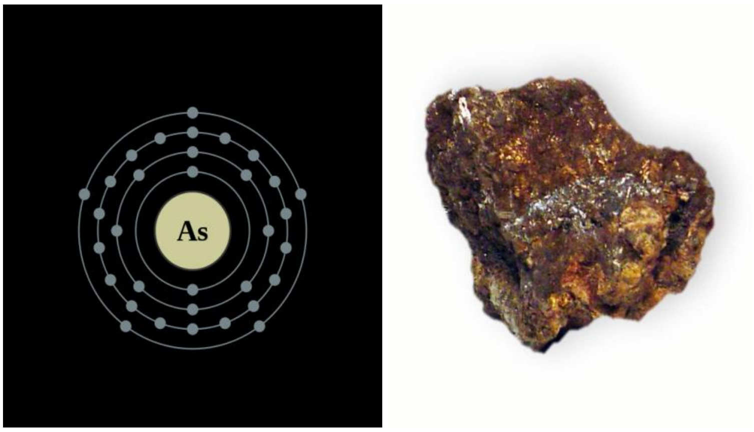

Figure1.1: Arsenic atom and arsenic ore

Arsenic is mobilized by natural weathering reactions, biological activity, geochemical reactions volcanic emissions and other anthropogenic activities (Mackenzie et al., 1979). Most environmental arsenic problems are the result of mobilization under natural conditions. However, mining activities, combustion of fossil fuels, use of arsenic pesticides, herbicides, and crop desiccants and use of arsenic additives to livestock feed create additional impacts. Arsenic exists in the - 3, 0, +3 and +5 oxidation states (Smedley et al., 2002). Arsenic (III) is a hard acid and preferentially complexes with oxides and nitrogen. Conversely, arsenic (V) behaves like a soft acid, forming complexes with sulfides. Inorganic forms of arsenic most often exist in water supplies (Bodek et al., 1998).

Occurrence of arsenic in water:

Arsenic is widely distributed throughout the earth's crust. Arsenic is introduced into water through the dissolution of minerals and ores, and concentrations in groundwater in some areas are elevated as a result of erosion from local rocks. Dominant natural arsenic bearing rocks include realgar (AsS), orpimand (As_2S_3), lollingite ($FeAs_2$), and arsenopyrite (FeAsS). Industrial effluents also contribute arsenic to water in some areas. Arsenic is also used commercially e.g. in alloying agents and wood preservatives.

Background arsenic concentrations in natural water are low, but elevated arsenic concentrations are common in groundwater as a result of natural conditions or anthropogenic impacts. Natural oxidation and/or reduction reactions involving arsenic bearing rocks under favorable temperature and pH conditions may mobilize the arsenic and increase arsenic concentrations in groundwater. Human activities that could increase arsenic concentrations in ground waters and surface waters include: oil and coal burning power plants, waste incineration, cement works, disinfectants, household waste disposal, glassware production, electronics industries, ore production and processing, metal treatment, galvanizing, ammunition factories, dyes and colours, wood preservatives, pesticides, pyrotechnics, drying agents for cotton, oil and solvent recycling and pharmaceutical works.

In organic arsenic can occur in the environment in several forms but in natural waters, and thus in drinking water. It is mostly found as trivalent arsenite [As (III)] or penta-valent arsenate [As(V)]. Organic arsenic species, abundant in seafood, are very much less harmful to health, and are readily eliminated by the body. Arsenic contaminated drinking water poses the greatest threat to public health (Lee et al., 2002).

Environmental levels and standards:

Concentrations in water are usually < 10 μg/l, although higher concentrations can occur near natural mineral deposits or anthropogenic sources. Arsenic levels in groundwater average about 1–2 μg/l, except in areas with volcanic rock and sulfide mineral deposits where arsenic levels can range up to 3400 μg/l.

In Nepal, arsenic (As) contamination is a major issue of current drinking water supply systems using groundwater and has recently been one of the major environmental health management issues especially in the plain region, i.e., in the Terai districts, where the population density is very high. The Terai inhabitant still use hand tube and dug wells (with hand held pumps that are bored at shallow to medium depth) for their daily water requirements, including drinking water. The National Sanitation Steering Committee (NSSC, 2010), with the help of many other organizations, has completed arsenic blanket test in 25 districts of Nepal by analyzing 737,009 groundwater samples. Several organizations, including academic institutions, made an effort to determine the levels of arsenic concentrations in groundwater and their consequences in Nepal. The results of the analyses on 25,058 samples tested in 20 districts, published in the status report of arsenic in Nepal (2003), demonstrated that the 23% of the samples were containing 10–50 μg/L of As, and the 8% of the samples were containing more than 50 μg/L of As. Recent status of over 737,009 samples tested, the 7.9% and 2.3% were contaminated by 10–50 μg/L and >50 μg/L, respectively of As. The areas that are mostly affected by this influx of arsenic are at Terai region of Nepal hence, the findings of this thesis will be appropriate for that area. Percentage of arsenic contaminated samples in various districts of Nepal is shown in the table in Appendix A.8.

The International Standards for drinking water was established 0.20 mg/L as an allowable concentration for arsenic by WHO in 1958. In 1963 the standard was re-evaluated and reduced to 0.05 mg/L. In 1984, this was maintained as WHO's "Guideline Value"; and many countries have kept this as the national standard or as an interim target. According to the WHO guidelines for drinking water quality:

- Inorganic arsenic is a documented human carcinogen.
- 0.01mg/L was established as a provisional guideline value for arsenic.
- Based on health criteria, the guideline value for arsenic in drinking water would be less than 0.01mg/L.

- The guideline value is restricted by measurement limitations, and 0.01 mg/L is the realistic limit to measurement, this is termed a provisional guideline value.

Interim Nepal guidelines and policies for arsenic in drinking water:

The interim Nepal guidelines and policies for arsenic in drinking water were adopted on first June 2001 by NASC. The policy provides guiding principles for all government and non-government agencies when formulating, designing and implementing asrsenic programs. The policy has established permissible arsenic concentration values of 50 ppb for drinking water. The policy focuses on immediate attention to be given by stakeholder agencies in identifying the existing “arsenic hot spots” and carrying out more testing on hot spots to better understand the extent and magnitude of arsenic contamination. The policy also highlights health care issues as well as communication with communities. (NASC, 2011)

1.2 Rationale of the study

Arsenic has effects on widely different organ systems in the body. It has produced serious effects in humans after both oral and inhalation exposure. A peculiarity of arsenic carcinogenicity is that the information mainly comes from experience with exposed humans.

Ingestion of high doses of arsenic may lead to acute symptoms within 30–60 min, but the effects may be delayed when the arsenic is taken with food. Acute gastrointestinal syndrome is the most common presentation of acute arsenic poisoning. This syndrome starts with a metallic or garlic-like taste associated with dry mouth, burning lips and dysphagia. Violent vomiting may ensue and may eventually lead to haematemesis. Gastrointestinal symptoms, which are caused by paralysis of the capillary control in the intestinal tract, may lead to a decrease in blood volume, lowered blood pressure and electrolyte imbalance. Thus, after the initial gastrointestinal problems, multi-organ failure may occur, including renal failure, respiratory failure, failure of vital cardiovascular and brain functions, and death. Survivors of the acute toxicity often develop bone marrow suppression (anaemia and leukopenia), haemolysis, hepatomegaly, melanosis and polyneuropathy resulting from damage to the peripheral nervous system. Polyneuropathy is usually more severe in the sensory nerves, but may also affect the motor neurons (National Research Council (NRC), 1999).

Fatal arsenic poisonings have been described after oral exposure to estimated doses of 2 g, 8 g and 21 g, and cases with non-fatal outcome (usually after treatment) have been reported after oral doses of 1–4 g up to 8–16 g arsenic. Incidents of continuous or repeated oral exposure to arsenic over a short period of time have been described. When they drank water containing 108 mg As/liter for 1 week, 2 out of 9 exposed persons died, 4 developed encephalopathy and 8 gastrointestinal symptoms. No deaths, but symptoms mainly from the gastrointestinal tract and skin, were observed among 220 patients studied among 447 who had been exposed to arsenic in soy sauce at a level of 100 mg/litre for 2–3 weeks; the estimated daily dose of arsenic was 3 mg. A case of lung cancer associated with exposure to arsenical dust was brought to the notice of the British Factory Department, and some further cases were detected in the early 1940's. These reports were followed by an investigation of the matter, and a remarkably elevated relative cancer mortality rate from lung and skin cancer was observed in a sheep-dip factory manufacturing sodium arsenite. Chronic skin effects of arsenic, including pigmentation changes, hyperkeratosis and skin cancer, from medicinal use but also from drinking water, were reported as early as the 19th century. A large number of case series on arsenical skin cancer after exposure via drinking water were published from Argentina, Chile, Mexico and Taiwan in the early 1900s. An endemic peripheral vascular disease (PVD), known as black foot disease (BFD), leading to progressive gangrene of the legs, has been known in Taiwan since 1920 (Acharyya et al.,1999). Figure1.2 shows the several harmful effects of drinking arsenic contaminated water (Zhuo,2004).

(Arsenic lesions on hands, missing finger, spotted, kertosis on palm, arsenic lesions on head)

Figure 1.2 Harmful effects of arsenic (Zhuo, 2004)

Unfortunately, there is no known cure for arsenic poisoning and therefore providing arsenic free drinking water is the only way to diminish the adverse health effects of arsenic. And through this thesis optimum condition for the removal of arsenic by electrolysis method will be proposed.

1.3Objectives of the study

Purpose of this study was to conduct experimental investigation of arsenic removal using the electrolysis. The main objectives of this study are:

- To investigate the effectiveness of batch electrolysis process for arsenic removal.
- To study the effect of following parameters in arsenic removal
  - pH
  - Current passing time
  - Initial concentration
  - Inter electrode spacing

1.4 Scope and limitations of the study

The study will focus on electrolysis process in removing Arsenic from ground water. However, there are some limitations in this study as listed below:

- Temperature effect is not considered during the study period.
- The study is based on batch reactor only.
- The study outcome might be limited to the knowledge and adopted methodology.
- Due to resource constraints the analysis of the arsenic in water is done by digital arsenator only.

1.5 Organization of report

This report is organized into five chapters as

Chapter I deal with introduction, rationale of the study, objectives of the study, and limitation of the study.

Chapter II describes the theories, which is related with the study. It contains relevant information and data available in past research, papers, journals etc.

Chapter III describes the methodology adopted for this research.

Chapter IV includes the data observed and measurements during lab tests and their analysis and presentation terms as results and discussions.

Chapter V includes conclusions and recommendations.

CHAPTER 2

2.0 LITERATURE REVIEW

2.1 General review of past experience

Several investigations have been worked on the removal of arsenic by electro-coagulation. These are reported below:

Mollah et al. (2000) studied electro-coagulation is an evolving technology that is being effectively applied today for wastewater treatment, the paucity of scientific understanding of the complex chemical and physical processes involved is limiting future design and hindering progress. Chemistry and physical processes involved into perspective are fully explained. EC process involves three successive stages: (a) formation of coagulants by electrolytic oxidation of the ‘sacrificial electrode’; (b) destabilization of the contaminants, particulate suspension, and breaking of emulsions; (c) aggregation of the destabilized phases to form flocs.

Gregor (2001) studied the effect of forms and concentrations of arsenic on its removal through aluminum-based coagulation treatment processes were tracked for three drinking water treatment plants. This has provided direct evidence of where and how arsenic is removed. In general, soluble As(V) is converted to particulate As(V) by adsorption during rapid mixing, and is removed along with naturally occurring particulate arsenic predominantly by clarification. Soluble As(III) tracks through the treatment processes and is converted to soluble As(V) during final chlorination. The ability of a water treatment process to achieve the maximum acceptable concentration for arsenic in drinking water is dependent on the concentration of As(III) in the source water.

Kumar et al. (2004) studied (EC) as a treatment technology for arsenite [As (III)] and arsenate [As(V)] removal from water. Laboratory scale experiments were conducted with three electrode materials namely, iron, aluminum and titanium to assess their efficiency. Arsenic removal obtained was highest with iron electrodes. EC was able to bring down aqueous phase arsenic concentration to less than 10 µg/l with iron electrodes. Current density was varied from 0.65 to 1.53 mA/cm^2 and it was observed that higher current density achieved rapid arsenic removal. Effect of pH on arsenic removal was not significant in the pH range 6–8. Comparative evaluation of As (III) and As (V) removal by chemical coagulation (with ferric chloride) and electro-coagulation has been done. The comparison revealed that EC has better removal efficiency for As (III), whereas As (V) removal by both processes was nearly same. The removal mechanism of As (III) by EC seems to be oxidation of As (III) to As (V) and subsequent removal by adsorption/complexation with metal hydroxides generated in the process.

Srivastava et al. (2008) investigated the removal of arsenic (As) and chromium (Cr) from aqueous solution by electro coagulation using iron electrodes to evaluate the influence of various experimental parameters on the removal of metal ions. The parameters were initial pH, electrolysis time, initial concentration, electrode gap, stirring rate and current density. Optimum value of current density for As and Cr removal were found to be 75 and 50 A/m^2, respectively. Removal efficiency increased with decrease in the initial concentration and electrode gap.

Balasubramanian et al. (2009) studied removal of arsenic from aqueous solution. Removal was carried out using electro-coagulation. Experiments were conducted using mild steel sacrificial anode covering wide range in operating conditions to assess the removal efficiency. The maximum arsenic removal efficiency was reported as 94% under optimum conditions. Further the experimental data were tested with different adsorption isotherm model to describe the electro-coagulation process.

Lakshmanan et al., (2009) Electrolysis is an emerging water and wastewater treatment technology that involves electrolytic oxidation of an anode material and in situ generation of coagulant (Kumar et al., 2004; Lakshmanan et al., 2009). When a current is applied between two electrodes, metal ions such as Fe^{2+} and Al^{3+} that can contribute to coagulant formation are released by anode oxidation. The Fe^{2+}can subsequently be oxidized in solution to produce an Fe(III) hydroxide or oxy-hydroxide.

Electrolysis is an alternative to using chemical coagulants for arsenic removal; thus it can be beneficial for communities with better access to electricity than to chemicals. So far, several studies have reported arsenic removal from water and wastewater by electrolysis (Balasubramanian and Madhavan, 2001; Kumar et al., 2004; Parga et al., 2005). During such processes, arsenic removal by electrolysis involved metal oxide formation followed by arsenic removal (Balasubramanian and Madhavan, 2001). In addition, electrolysis may also control oxidation-reduction reactions; species such as As(III) may be oxidized on the anode and other species may be reduced on the cathode.

2.2 Processes for arsenic removal

The general categories of remediation processes for arsenic removal are discussed below:

2.2.1 Oxidation

Of the two predominant forms of arsenic in water, arsenate and arsenite, most treatment processes are effective at removing arsenate, but not arsenite, since arsenite is typically non-charged below pH 9.2. Therefore, treatment for the removal of arsenic often includes an oxidation step to convert arsenite to arsenate.

Oxidation can be simply the addition of oxygen to a compound, or more generally, any reaction involving the loss of electrons from an atom. Aeration, the supplying of air, oxidizes arsenic, converting arsenite to arsenate, and the iron that co-occurs. This is precipitated as $FeAsO_4$. Arsenic can also be oxidized by a number of other chemicals including chlorine, hypochlorite, ozone, permanganate, hydrogen peroxid and Fenton’s reagent (H_2O_2/Fe^{2+}). Photochemical oxidization proceeds from the reaction of radiant energy and a chemical system. Oxidation alone does not remove arsenic from solution but must be combined with an arsenic removal process (Borho et al., 1996).

2.2.2 Filtration

Conventional filtration is the separation of solid particles from water by passing the solution through a medium. Particles are removed during filtration as a result of any one or combination of mechanisms: mechanical straining, sedimentation, flocculation, adsorption and/or biological metabolism. The filter medium may be of various materials, for example, sand, anthracite coal, activated carbon, cloth, paper, that retains the solid on its surface and allows the water to pass through. Common particulates removed by filtration include silt, clay, colloidal and precipitated natural organic matter, naturally-occurring iron and manganese precipitates, precipitates from metal salt or polymer coagulation, microorganisms. Filters may be classified in various ways, according to the type of granular medium used, by the hydraulic system (e.g. gravity, up-flow, etc.), rate of filtration, and/or by the location of particle accumulation (Clifford et al., 1999)

2.2.3 Absorption

Adsorption is the accumulation of materials at an interface, the liquid/solid boundary layer. It is a mass transfer process where a substance is transferred from the liquid phase to the surface of a solid and becomes bound by chemical or physical forces. Adsorption can take place on suspended particles, as part of the process of coagulation/co-precipitation, or on fixed media. Since adsorption is a surface phenomenon, the greater the surface area of the medium, the greater it's capacity to accumulate material. Each adsorbent medium has different associated properties, performances and costs. Arsenic is adsorbed onto the surface of various granular, activated, clay and celluosic adsorbents, including the following:

Oxides (e.g. hydrated ferric oxide, titanium oxide, silicium oxide).

Iron oxide-coated or MnO_2- coated sand

Bauxite, hematite, feldspar

Clay minerals (e.g. kaolinite, bentonite, Bijoypur clay)

Synthetic anion exchange resins.

Chitin and chitosan

Bone char.

Cellulose materials (sawdust, newspaper pulp) (Nenov et al., 1986)

2.2.4 Ion Exchange

Ion exchange is the reversible interchange of ions between the solid and the liquid phase where there is no permanent change in the structure of the solid. Developed for large-scale applications, ion exchange is probably not appropriate for small hand-pumped wells, but could potentially be used on a small scale.

Synthetic ion exchange resins are based on a cross-linked polymer matrix, typically composed of polystyrene cross-linked with vinyl benzene. Charged functional groups are attached to the matrix through covalent bonding and fall into the following four groups:

Strongly acidic

Weakly acidic

Strongly basic

Weakly basic

Various strong base anion exchange resins are commercially available that can effectively remove arsenic from water, producing effluents with less than 1 µg/L arsenic. Arsenic, being uncharged, is not removed, unless an oxidation step to convert arsenite to arsenate is included in the process.

Conventional sulfate-selective resins are particularly suited for arsenate removal. Nitrate- selective resins also remove arsenic, but arsenic breakthrough occurs earlier. Ion exchangers are typically down-flow, packed bed columns with ion exchange resin beads pre-saturated with an exchangeable ion. Source water is passed through the packed bed until the appearance of the unwanted contaminant in the effluent. At this stage, the ion exchange media is reactivated with a regenerant solution and rinsed with water in preparation for another treatment cycle. Both the red-ox potential and pH are important factors with regard to arsenic removal by ion exchange (Benefield et al., 1990).

2.2.5 Coagulation/ Co-precipitation

Coagulation encompasses all reactions, mechanisms and results in the overall process of particle growth (floc formation) and particle aggregation within water being treated, including in situ coagulant formation, chemical particle destabilization and physical inter-particle contacts. Coagulation involves the removal of colloidal (0.001 - 100 microns) and settleable (>100 microns) particles. However the term also commonly refers to the removal of dissolved ions (< 0.001 microns), which is actually precipitation. Chemical precipitation is the process by which dissolved ions in solution form an insoluble solid via a chemical reaction. For example, naturally occurring dissolved iron in groundwater, when exposed to oxygen, forms a precipitate. Co-precipitation occurs when an inorganic contaminant forms an insoluble complex with the coagulant. Both the valence of the inorganic contaminant and the pH of the solution are important removal by co-precipitation.

Coagulation converts soluble arsenic into insoluble reaction products, allowing separation by sedimentation and/or filtration. Factors affecting arsenic removal by coagulation/precipitation include coagulant type and dose, mixing time and speed, pH, arsenic oxidation state and concentration, presence of inorganic solutes.

Three mechanisms are mainly involved in arsenic removal:

Precipitation: The formation of insoluble compounds Al (AsO_4) or Fe (AsO_4).

Co-precipitation: Incorporation of soluble arsenic species into the metal hydroxide floc.

Adsorption: The electrostatic binding of soluble arsenic to the external surfaces of the insoluble metal hydroxides.

Direct precipitation plays the least important role in arsenic removal however; co-precipitation and adsorption are both active arsenic removal mechanisms (Novikova et al., 1982).

2.2.6 Membrane/Reverse osmosis

Membrane separation uses semi-permeable membranes that are selectively permeable to water and certain solutes to separate impurities from water. Membranes are able to remove many different kinds of dissolved solids, including arsenic, from water. However, they are usually expensive and therefore are typically considered in applications such as desalination, brackish water conversion and for removal of specific ions, such as arsenic, that are difficult to remove by other means. There are many different membrane alternatives including microfiltration, reverse osmosis, electro-dialysis, ultra filtration and nano-filtration. Membrane process treatment performance is dependent on the quality of the feed water and the desired quality of the product water. Generally the more contaminated the feed water and the higher the desired product water quality, the greater the likelihood of membrane fouling caused by particulate matter, scaling and bio fouling (Kang et al., 2000).

2.2.7 Biological

Biological treatment transforms, stabilizes and/or removes arsenic by means of microorganisms. Microorganisms, primarily certain specific bacteria, accomplish this by oxidation/reduction, mineralization, detoxification or methylation. Critical factors include energy and carbon source, aerobic-anoxic or anaerobic conditions, temperature, and pH (Mohan et al., 2007).

2.2.8 Solar distillation

Solar distillation uses the sun's energy to evaporate water, which then recondences. The process of evaporation and re-condensation separates all chemicals, including arsenic, from the water. In Bangladesh, where solar energy is plentiful, this approach may be especially suited for application in crisis areas, and, if cost-effective approaches can be developed, in rural areas (Susan Murcott, 1999).

2.3 Electrolysis

Electrolysis (EC) is an electrochemical method of treating polluted water whereby sacrificial anodes corrode to release active coagulant precursors (usually aluminum or iron cations) into solution. Accompanying electrolytic reactions evolve gas (usually as hydrogen bubbles) at the cathode. Electrolysis has a long history as a water treatment technology having been employed to remove a wide range of pollutants (Mameri et al., (1998).

2.3.1 Concept and definition of electrolysis

Electrolysis is the passage of a direct electric current through an ionic substance that is either molten or dissolved in a suitable solvent, resulting in chemical reactions at the electrodes and separation of materials. The main components required to achieve electrolysis are:

- An electrolyte: a substance containing free ions which are the carriers of electric current in the electrolyte. If the ions are not mobile, as in a solid salt then electrolysis cannot occur.
- A direct current (DC) supply: provides the energy necessary to create or discharge the ions in the electrolyte. Electric current is carried by electrons in the external circuit.
- Two electrodes : an electrical conductor which provides the physical interface between the electrical circuit providing the energy and the electrolyte.

In its simplest form, an electrolysis reactor may be made up of an electrolytic cell with one anode and one cathode. When connected to an external power source, the anode material will electrochemically corrode due to oxidation, while the cathode will be subjected to passivation. But, this arrangement is not suitable for wastewater treatment, because for a workable rate of metal dissolution, the use of electrodes with large surface area is required.

2.3.2 Arrangement of electrodes in electrolysis

A simple arrangement of an EC cell with anodes and cathodes is shown in Figure 3.1. D.C source of 12v/2 Ampere is connected between the two electrodes. The conductive metal plates are commonly known as ‘sacrificial electrodes’. The sacrificial electrodes may be made up of the same or of different materials (Pretorius et al.,). In the setup, iron was used as anode and aluminum was used as cathode.

2.3.3 Theory of electrolysis

It is generally accepted that the EC process involves three successive stages: (a) formation of coagulants by electrolytic oxidation of the ‘sacrificial electrode’; (b) destabilization of the contaminants, particulate suspension; (c) aggregation of the destabilized phases to form flocs/complexes.

2.3.4 Arsenic removal mechanism

The main electrode reactions are as shown in following equations described by (H.K. Hansen et al., 2007);

Anodic reactions

(a) Single step oxidation of Fe into ferric ion

$Fe = Fe^{3+} + 3e^-$

(b) A two step process where iron is firstly oxidized to ferrous ion which, depending on anode potential, then oxidizes to ferric ion

$Fe = Fe^{2+} + 2e^-$

$Fe^{2+} = Fe^{3+} + e^-$

Cathodic reactions

$2H_2O + 2e^- = H_2(g) + 2OH^-$ (in alkaline solution)

$2H_3O^+ + 2e^- = H_2(g) + 2H_2O$ (in acid solution)

Typically at the cathode the solution becomes alkaline with time. The applied current forces OH^- ion migration to the anode, so the pH near the anode is higher than in the bulk solution, thus favoring ferric hydroxide formation:

$Fe^{3+} + 3OH^- \rightarrow Fe(OH)_3(s)$

Arsenate Co-precipitation with or adsorbs to $Fe(OH)_3(s)$

$Fe(OH)_3(s) + AsO4^{3-(}aq) = [Fe(OH)_3*AsO4^{3-}](s)$

Thus the arsenic from the groundwater is removed.

CHAPTER 3

3.0 Methodology

Direct current electro-coagulation technique was used for arsenic removal from drinking water. Electrolysis was conducted in a batch setup to investigate the effect of pH, current density and inter-electrode spacing on arsenic removal.

3.1 Experimental setup

The electrolysis reactor (beaker) having one liter of capacity was used. Iron plate (12 cm × 2 cm) with a thickness of 2 mm and the aluminum plate of (12 cm × 2 cm) with a thickness of 1 mm were used as the electrodes for the experiments. A regulated direct current supply of 2 Ampere and 12 Volts was supplied using a DC adapter was used for the experiments. The experimental setup is shown in Figure 3.1.

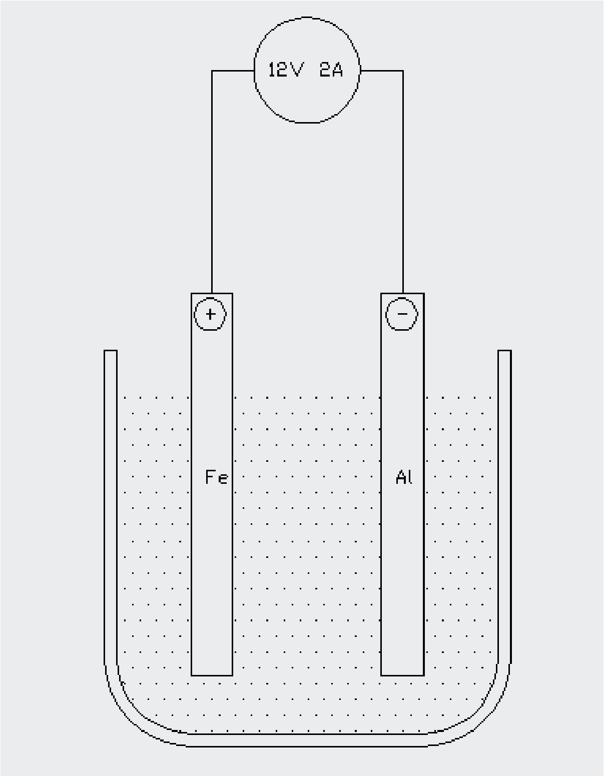

Figure 3.1: Laboratory setup for electrolysis

3.2 Preparation of stock solution

The chemicals used were analytical reagent grade. The arsenate solution was prepared from the Sodium arsenate, $Na_2HAsO_4.7H_2O$. The stock solution was prepared by dissolving 0.416 gm of sodium arsenate in 100 ml distilled water [1ml = 1mg As

(V)]. The intermediate arsenic solutions were freshly prepared before each experiment. The arsenic sample solution was prepared by dissolving appropriate amount of intermediate solution in tap water.

3.3 Experimental procedure

The complete experimental procedure is as follows:

1. The sample solution was prepared from the stock solution as required.
2. The pH of the solution was adjusted by adding either dilute HCl or NaOH as per the requirement.
3. Before each experiment, the electrodes were abraded with sand paper to remove scale and then cleaned with successive rinses of water and 1N H_2SO_4
4. The inter-electrode spacing was fixed as per the requirement.
5. DC power supply was used to pass 2A current at 12V.
6. The samples were taken for residual arsenic analysis.

3.4 Analysis of concentration of arsenic

The analysis of initial arsenic content and residual arsenic content in water sample was carried out using WAGTECK Digital Arsenator (figure 3.2)

Figure 3.2: WAGTECK digital arsenator

The arsenator system is the lowest cost, most portable and accurate method for determining arsenic in safe limits (WHO guide-lines 10 ppb/µgl).Carefully designed to be simple and safe to use either in the lab or in the field, the complete system comes with sufficient reagents and consumables for over 400 tests.

Advantages of WAGTECK digital arsenator

- Low cost digital arsenic testing device
- Fully portable, designed especially for field use
- Immediate results in the field in less than 20 minutes
- Simple, safe and easy to operate
- Gives accurate test results between the critical range of 2µgl (ppb) to 100µgl (ppb)
- Designed in conjunction with Prof. Walter Kosmus and laboratory tested by Imperial College London
- Field tested in conjunction with UNICEF/ WHO/WAT/SAN monitoring programs
- Environmentally friendly

Digital Arsenator kit comprises the following components:

- Portable Digital Arsenator® System
- Tri-Filter Arsenic gas trap
- Arsenic collection filters
- Arsine gas removal filters
- Arsenic collection filter holders (Black)
- Arsine gas removal filter holders (Red)
- Hydrogen Sulphide removal filters
- Reagent A1 sulphamic acid (dry powder sachet)
- Tablet A2 sodium borohydride
- Colour comparison chart (<10 to 500ppb)
- Dilution tube
- Spare battery
- Waste disposal bags

- Gloves and tweezers
- Rugged carry case with Instruction manual

Procedure for analysis of concentration of arsenic

- In testing flask 50 ml of sample was taken.
- Arsenic collection filter paper strip was fixed in holders (Black) and Arsine Gas removal filter paper strip was fix in holders (red).
- Reagent A1 (dry powder sachet) and Table A2 was mixed in sample and the flask was closed immediately.
- After waiting 20 minutes, arsenic collection filter holder (black) was inserted in arsenator to get the concentration of arsenic.

CHAPTER 4

4.0 RESULT AND DISCUSSION

The experiment results of arsenic removal from drinking water by electrolysis are presented in this chapter. The treatment process has to ensure that arsenic concentration in the supernatant is within the permissible limit as prescribed by WHO. Therefore, the concentration of arsenic in treated water is more important than the percentage of arsenic removed.

4.1 Effect of initial concentration

The initial concentration of arsenic in water affects the arsenic removal during its treatment. The high initial concentration results in the higher effluent making the water unsuitable for drinking purpose. It is thus required that the initial concentration is limited to a certain label so that the water after treatment is suitable for drinking purpose as per the drinking water quality standards. The effect of the initial concentration of the arsenic in its removal by electrolysis has been studied. The initial concentration of the arsenic varied from 9 to 96 µg/L. The electrolysis was carried out at current of 2A/12 volt, inter-electrode spacing of 4 cm and current passing time of 20 minutes. The arsenic concentration in water was measured at the end of 20 minutes. The results of the analysis are shown in Figure 4.1 and Figure 4.2.

Figure 4.1 shows the final concentration of arsenic at various initial concentration of arsenic. The result showed that the final concentration of arsenic increases as the initial concentration of arsenic increased. The final arsenic concentration of 2 µg/L arsenic was observed at initial concentration of 9 µg/L. The final of arsenic increased to 33 µg/L when initial concentration was increased to 96 µg/L. The arsenic removal, however, was found to be increased as its initial concentration increased. The arsenic removal was found to be 7 µg/L at initial concentration of 9 µg/L which increased 63 µg/L at initial concentration of 96 µg/L.

Figure 4.2 shows the percentage of arsenic removal at various initial concentration of arsenic. The result showed that the percentage of the arsenic removal decreases as the initial concentration increases. The arsenic removal was observed 78 % at the initial arsenic concentration of 9 µg/L and the arsenic removal was found to be decreased to 66% at initial arsenic concentration of 96 µg/L. As the initial concentration of arsenic increased, it required more amount of iron oxides to remove arsenic and this in turn required higher amount of dissolved oxygen content to oxidize As^{3+} to As^{5+}. The amount of dissolved oxygen available in water limits the oxidation of arsenic and its removal at higher initial concentration. Thus the arsenic removal percentage was found lower at higher initial concentration.

The experiment to study the effects of initial concentration on removal of arsenic was carried out at current 2A/ 12V and at an inter-electrode spacing of 4cm and current passing time of 20 minutes. The pretreated sample was found to have different arsenic content. The treated water sample was analyzed after the experimental run.

Percentage of arsenic removed was less when the solutions had higher initial arsenic concentrations as compared to lower initial concentrations. When the initial arsenic concentrations were higher, more iron oxides were needed to decrease the dissolved arsenic concentrations. This was in agreement with the result obtained which is shown in the figure-4.1and figure-4.2.

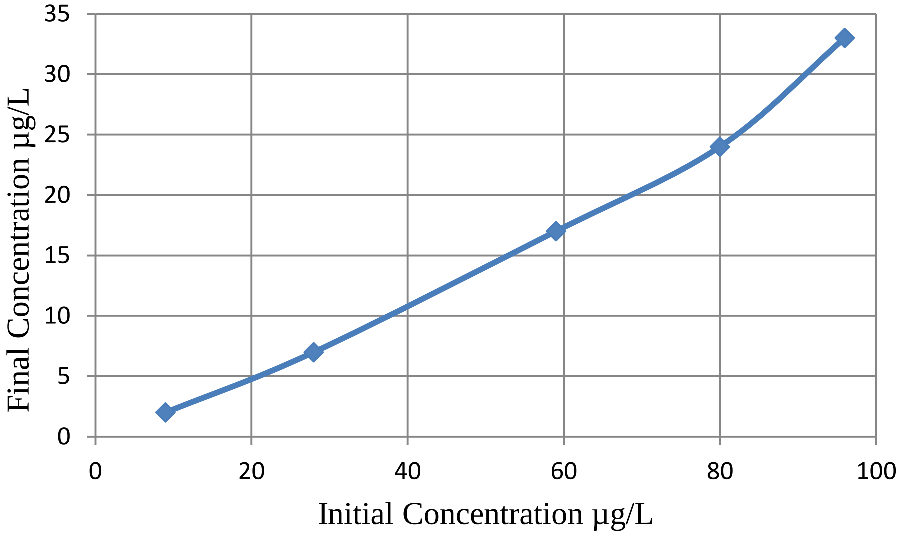

Figure4.1 Effect of Initial concentration on Arsenic removal

(Current 2A/12 V, Spacing 4 cm, current passing time 20 minutes)

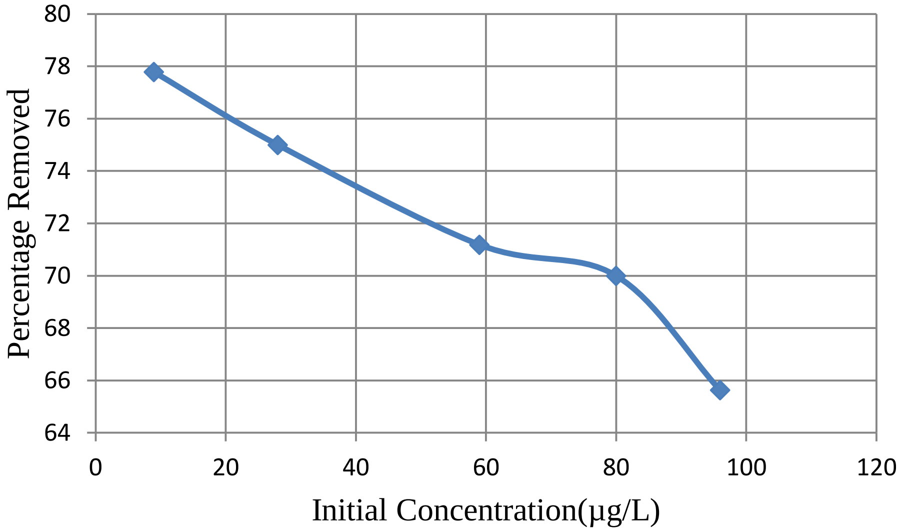

Figure4.2: Effect of Initial concentration on Arsenic removal efficiency
(Current 2A/12 V, Spacing 4 cm, current passing time 20 minutes)

4.2 Effect of current passing time

The experiment to study the effects of current passing time on removal of arsenic was carried out at current 2A/ 12V and at an inter-electrode spacing of 4cm. The experiment was performed at pH of 7. The pre-treated sample was found to have different arsenic content. The treated water sample was analyzed after electrolysis and the results are given in figure 4.3 and figure 4.4.

The result obtained showed most of the arsenic removal occurred within 10 minute of the electrolysis time as shown in figure 4.3. After the initial time period, the removal rate decreased gradually for rest of the processing time. Arsenic ions are more abundant at the beginning of the EC process, and the generated iron hydroxides due to corrosion of the anode at that time will form complexes with arsenic and therefore rapid removal of arsenic was observed. However , as the experiment proceeds the aqueous phase arsenic concentration goes on decreasing and simultaneously hydrous ferric oxides concentration increases, thereby abundance of hydrous ferric oxides occur at the end of the process

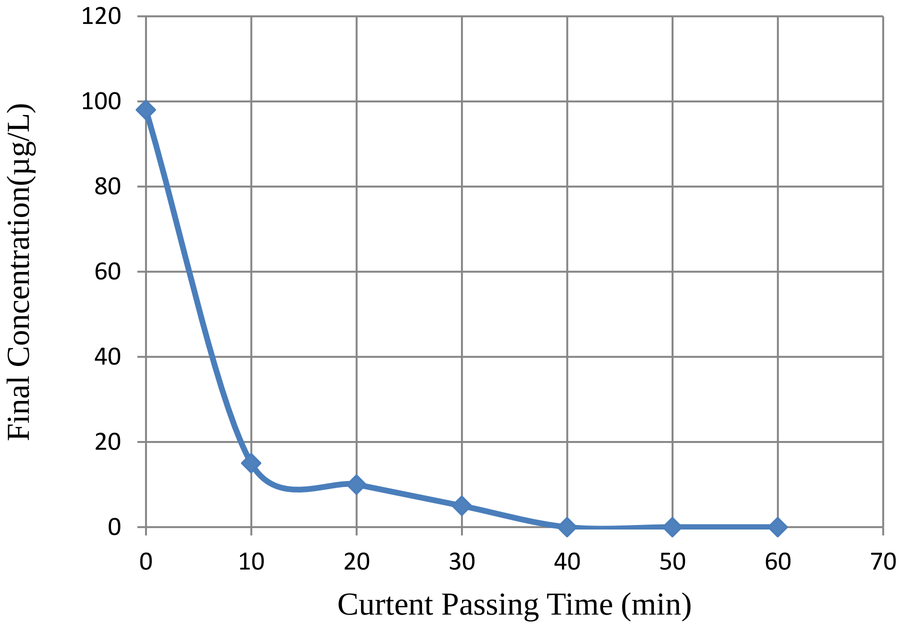

Figure4.3: Effect of current passing time on arsenic removal
(Current 2A/12 V, Spacing 4 cm, pH 7)

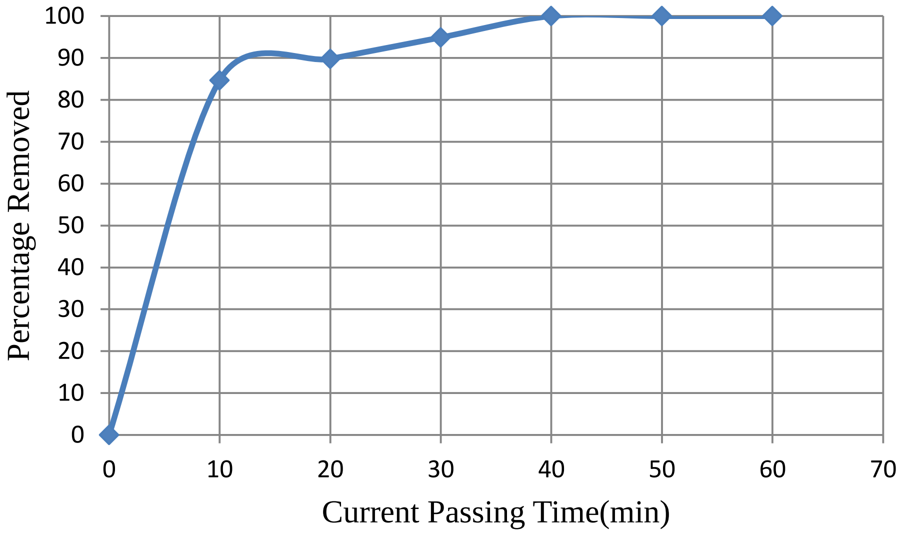

Figure 4.4: Effect of current passing time on removal efficiency

(Current 2A/12 V, Spacing 4 cm, pH 7)

4.3 Effect of inter electrode space

To study the effects of inter electrode space on removal of arsenic, the electrolysis was carried out at current 2A/ 12V and pH 7 for 20 minutes. The inter electrode spacing was varied to study the effect on residual arsenic content in the treated sample. The results obtained are reported in Figure 4.5 and Figure 4.6.

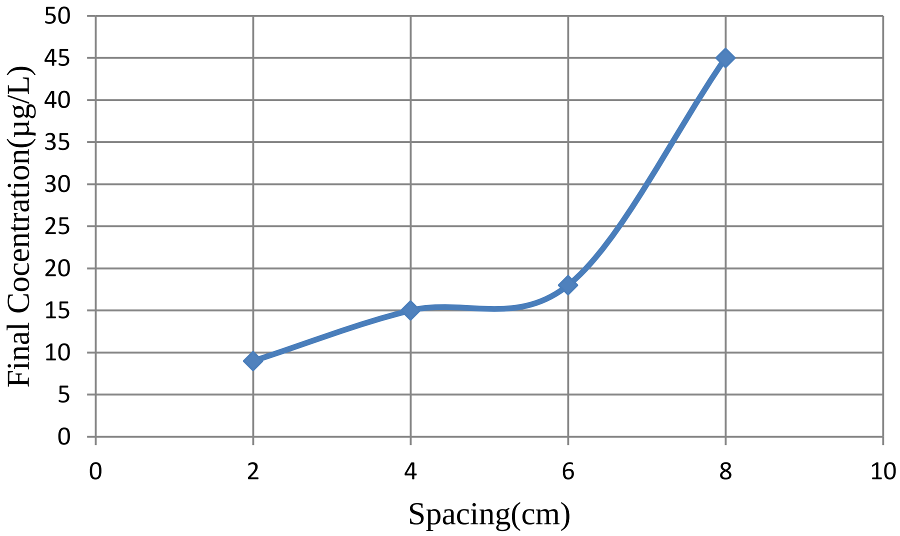

Figure 4.5: Effect of inter-electrode spacing on arsenic removal

(Current 2A/12 V, pH 7, current passing time 20 minutes)

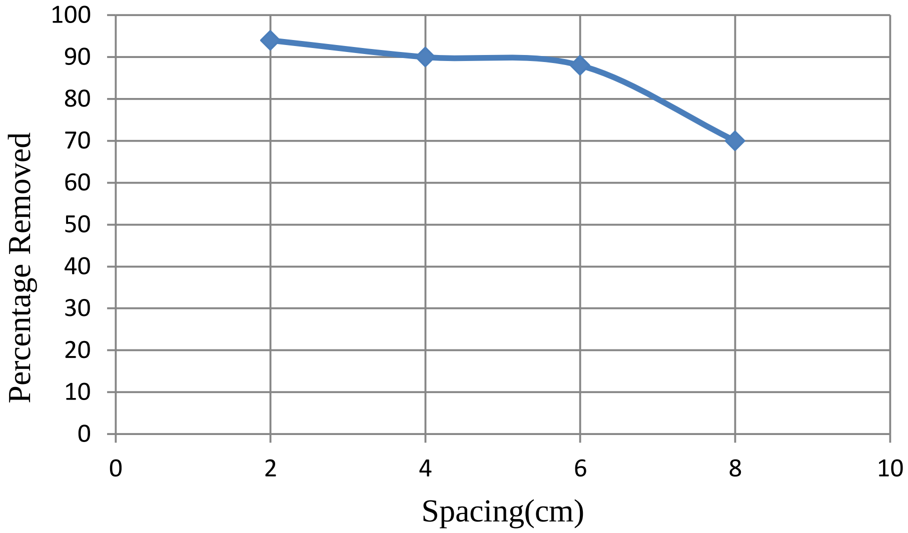

Figure 4.6: Effect of inter-electrode spacing on arsenic removal efficiency

(Current 2A/12 V, pH 7, current passing time 20 minutes)

With short inter-electrode distances the current density becomes too high and can cause short circuiting. There is no significant difference in the results for lowering the electrode spacing beyond 2 cm as the hydrogen gas evolved at the cathode increases the electrolytic resistance for very small inter-electrode spacing. This observation is in accordance with Paul (1996).

4.4Effect of pH on removal

The result has been discussed at current 2A/ 12V and at inter-electrode spacing 4cm. The pretreated sample was found to have arsenic content of 100 ppb. The treated water sample was analyzed after the experimental run. The results are given in Figure 4.7 and Figure 4.8.

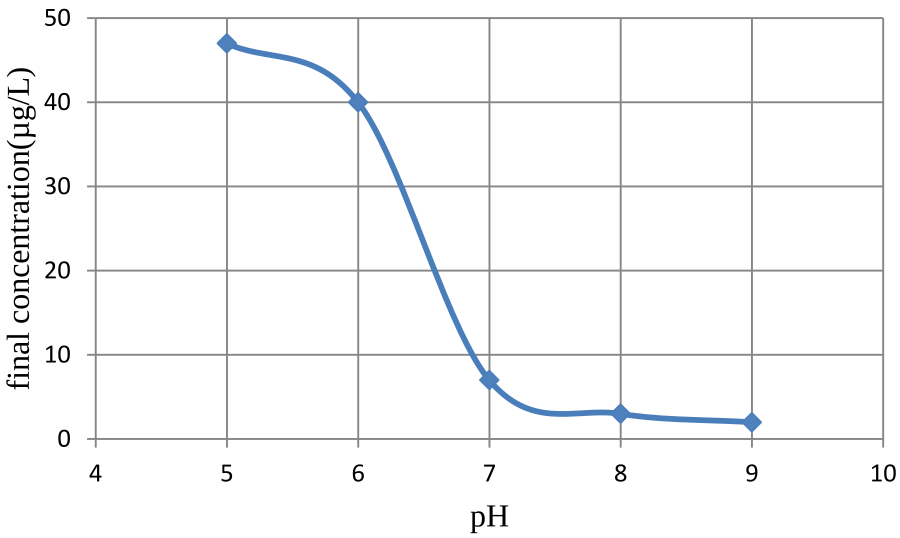

Figure 4.7: Effect of pH on arsenic removal

(Current 2A/12 V, spacing 4cm, current passing time 20 minutes)

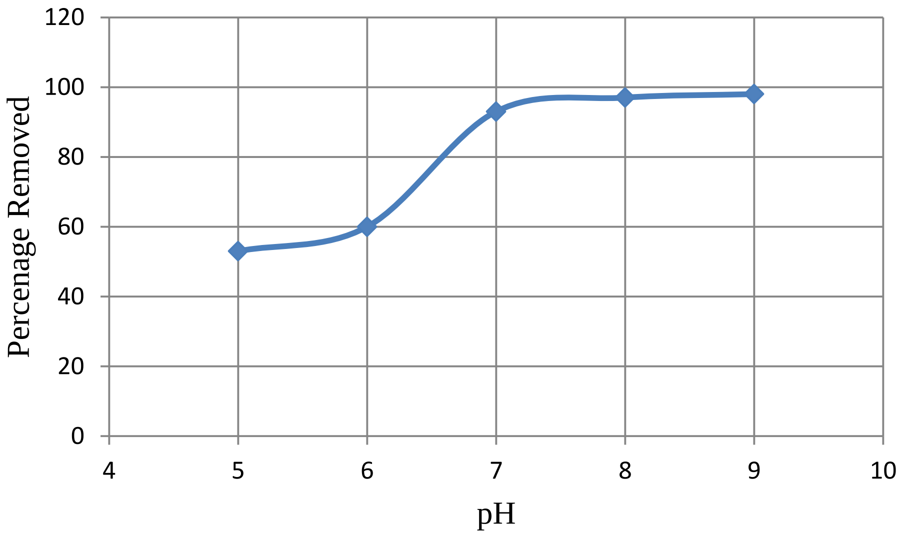

Figure 4.8: Effect of pH on arsenic removal efficiency

(Current 2A/12 V, spacing 4cm, current passing time 20 minutes)

The results show that with increasing pH the residual arsenic content in the water sample decreases. At low pH the solution protons were reduced at the cathode to hydrogen gas and the same proportion of hydroxide ions cannot be produced. Hence, at higher pH, the residual arsenic concentration was found to be lower. Kumar et al. (2004) in their work have also made the similar observation. The further experimentations were performed at pH 7 as the drinking water is near to this range and increasing the pH for the removal process results in increase of cost as the water after treatment needs to be neutralized in order to be drinkable.

4.5 Combined analysis

The combined effect of different initial concentration, varying current passing time and pH at 2cm inter electrode spacing was carried out and the result obtained are shown on figure 4.9, figure 4.10, figure 4.11, figure 4.12.

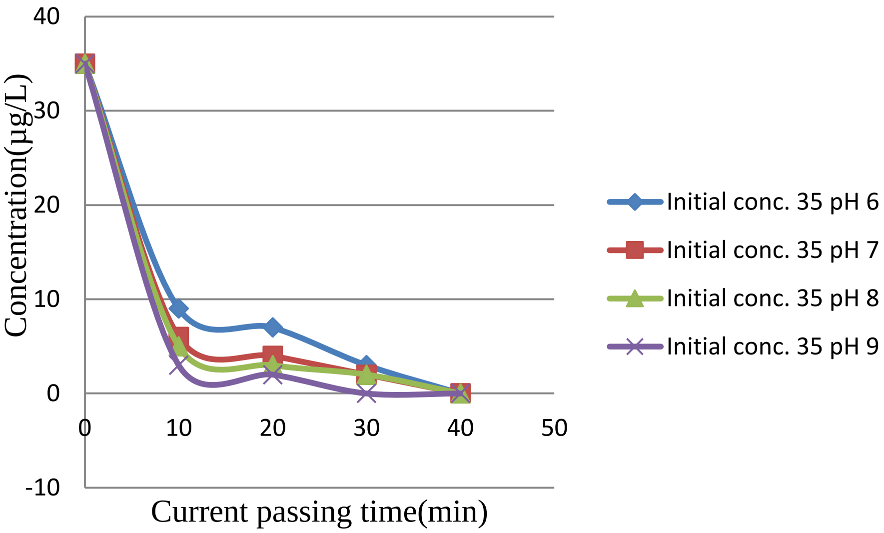

Figure 4.9 Concentration vs time for varying pH at initial concentration 35 (µg/L)

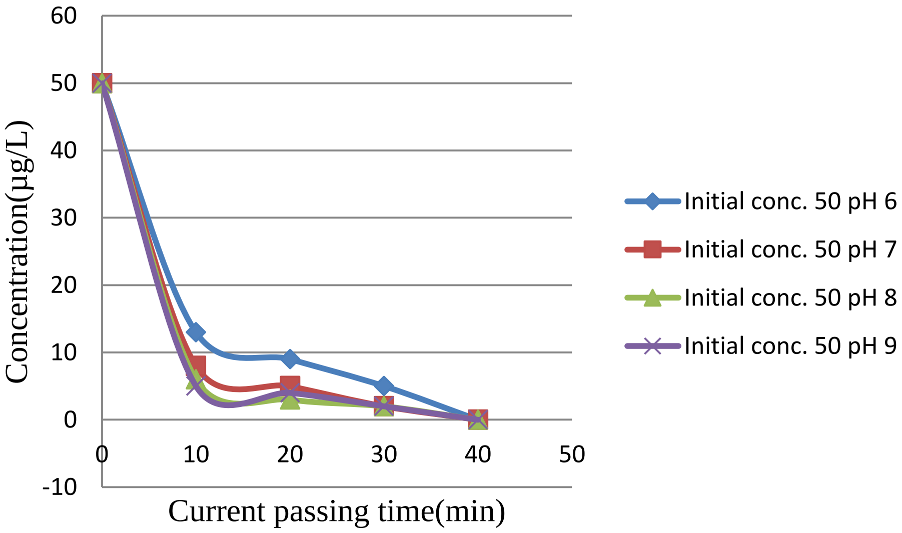

Figure 4.10 Concentration vs time for varying pH at initial concentration 50 (µg/L)

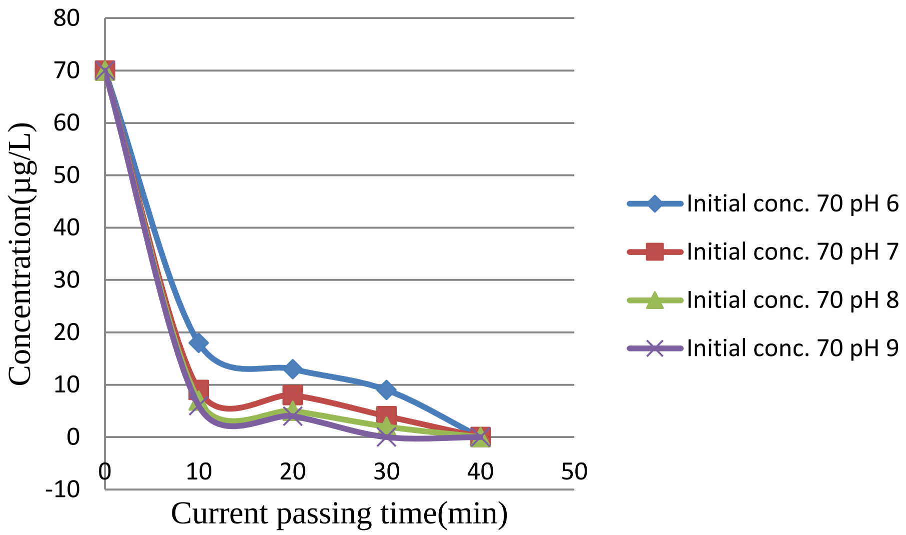

Figure 4.11 Concentration vs time for varying pH at initial concentration 70 (µg/L)

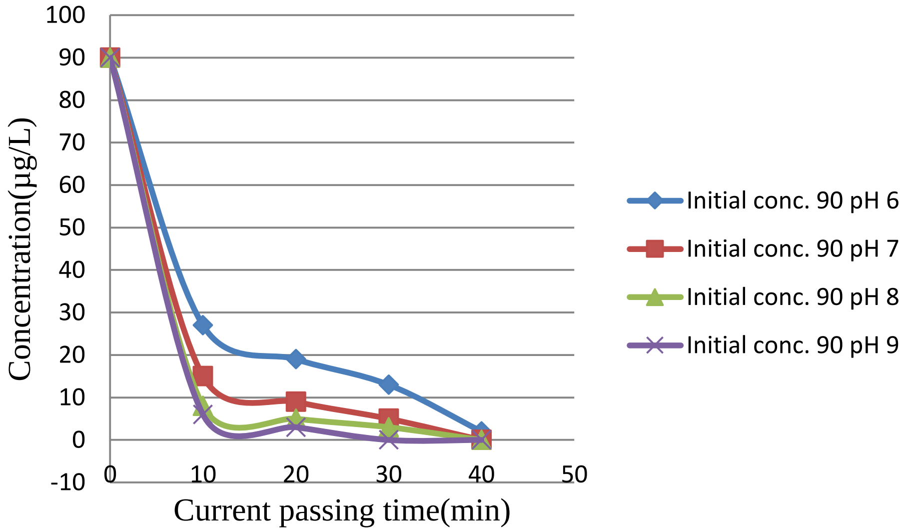

Figure 4.12 Concentration vs time for varying pH at initial concentration 90 (µg/L)

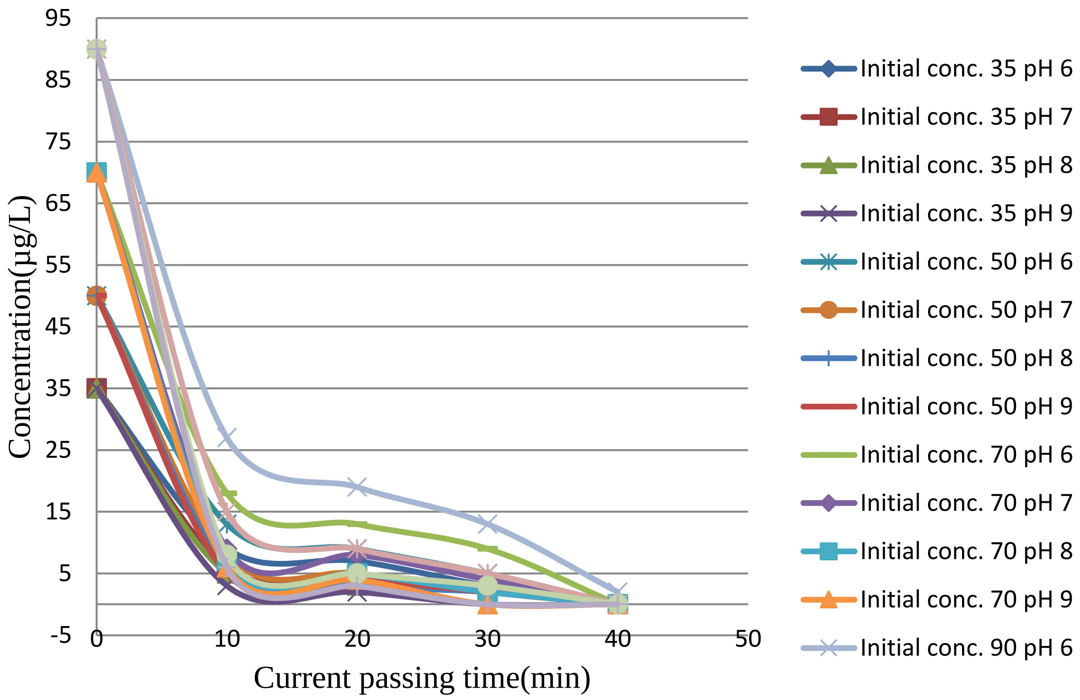

Figure 4.13 Concentration vs time for varying pH and initial concentration

The result obtained from the combined analysis of the parameters stated above reaffirms the findings obtained in the individual analysis of the parameters.

Thus, it can be inferred that for the removal of arsenic below the WHO standard for drinking water using the electrolysis method, inter electrode spacing of 2cm, current passing time 20 minutes and pH 7 can be used. These parameters are verified using natural water samples found in and around Kathmandu valley for effectiveness in removing arsenic.

4.6 ANALYSIS OF NATURAL WATER

Using electrolysis time of 20 minutes at pH 7 and inter electrode spacing 2cm, various natural water samples were analyzed and the results obtained are shown below. The location of the natural water sources with dissolved arsenic was obtained with reference to data provided by ENPHO (Environment and Public Health Organization).

Table 4.1 Analysis of natural water samples

S N	Source	Initial concentration (µgm/L)	Final concentration (µgm/L)
1	DWID	65	8
2	Dasharath Rangashala	55	6

3	Blue Bird Mall	45	4
4	Prashuti Griha	50	5

(Time 20 min, inter electrode spacing 2cm and pH 7)

CHAPTER 5

5.0 CONCLUSIONS

5.1 Based on the present work the following conclusions can be drawn:

- With increase in pH the removal efficiency of arsenic by the electrolysis increased.
- Arsenic removal improves with decreasing inter-electrode spacing.
- Final concentration of arsenic was found to be decreasing with increasing current passing time.
- The final arsenic concentration of sample after electrolysis was found to be lower at lower initial concentration.

- It was possible to bring the residual arsenic content in the treated natural water within WHO standard for drinking water (less than 10 ppb) in 20 minutes of electrolysis time with inter electrode spacing 2cm and pH 7.

5.2 Recommendation

- The arsenic removal from drinking water by electrolysis may be studied in continuous mode for continuous treatment plant.
- The analysis can be further optimized by using more precise equipments for detection of arsenic concentration in water such as atomic absorption spectrometer.
- Financial analysis of similar methods also needs to be carried out in order to study the feasibility.

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APPENDICES

A.1 Effect of initial concentration on removal of arsenic

Table A.1 Effect of initial arsenic concentration

SN	Initial conc. (µg/L)	Final conc. (µg/L)	Percentage Removed
1	9.00	2.00	78

2	28.00	7.00	75
3	59.00	17.00	71
4	80.00	24.00	70
5	96.00	33.00	66

(Current 2A/12 V, Spacing 4 cm, current passing time 20 minutes)

A.2 Effect of current passing time on removal of arsenic

Table A.2 Effect of current passing time on arsenic concentration

SN	Initial conc. (µg/L)	Current passing time (min)	Final conc. (µg/L)	Percentage Removed
1	98.00	0	98.00	0
2	98.00	10	15.00	85
3	98.00	20	10.00	90
4	98.00	30	5.00	95
5	98.00	40	<2	100
6	98.00	50	<2	100
7	98.00	60	<2	100

(Current 2A/12 V, Spacing 4 cm, pH 7)

A.3 Effect of inter-electrode spacing on arsenic removal

Table A.3 Effect of inter electrode spacing on arsenic removal

SN	Initial Conc. (µg/L)	Spacing (cm)	Final conc. (µg/L)	Percentage Removed
1	150.00	2	9.00	94
2	150.00	4	15.00	90

3	150.00	6	18.00	88
4	150.00	8	45.00	70

(Current 2A/12 V, pH 7, current passing time 20 minutes)

A.4 Effect of pH on removal of arsenic removal

Table A.4 Effect of pH on arsenic removal

SN	Initial Conc. (µg/L)	pH	Final conc. (µg/L)	Percentage Removed
1	100	5	47	53
2	100	6	40	60
3	100	7	7	93
4	100	8	3	97
5	100	9	2	98

(Current 2A/12 V, spacing 4cm, current passing time 20 minutes)

A.5 Combined analysis of pH and current passing time on various initial concentration on removal of arsenic

Table A.5 Combined analysis

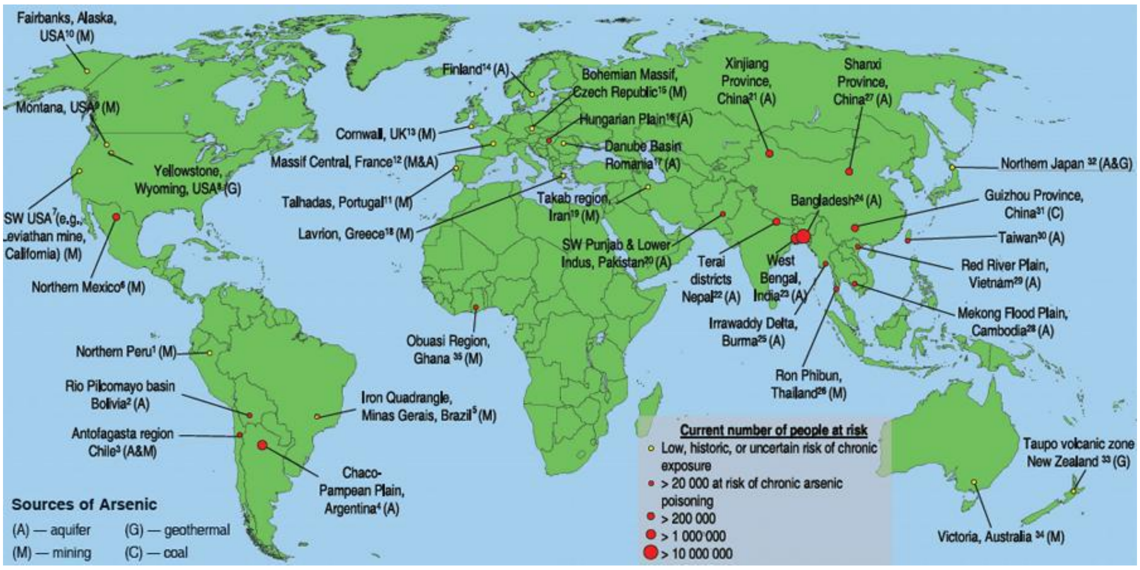
Initial concentration (µgm/L)	Final concentration (µgm/L)	pH	Current passing time (min)	Percentage removed
35	9	6	10	74

	7		20	80
	3		30	91
	0		40	100
	6	7	10	83
	4		20	89
	2		30	94
	0		40	100
	5	8	10	86
	3		20	91
	2		30	94
	0		40	100
	3	9	10	91
	2		20	94
	0		30	100
	0		40	100
50	13	6	10	74
	9		20	82
	5		30	90
	0		40	100
	8	7	10	84
	5		20	90
	2		30	96
	0		40	100
	6	8	10	88
	3		20	94
	2		30	96
	0		40	100
	5	9	10	90
	4		20	92
	2		30	96
	0		40	100

Initial concentration (µgm/L)	Final concentration (µgm/L)	pH	Current passing time (min)	Percentage removed
70	18	6	10	74
	13		20	81
	9		30	87
	0		40	100
	9	7	10	87
	8		20	89

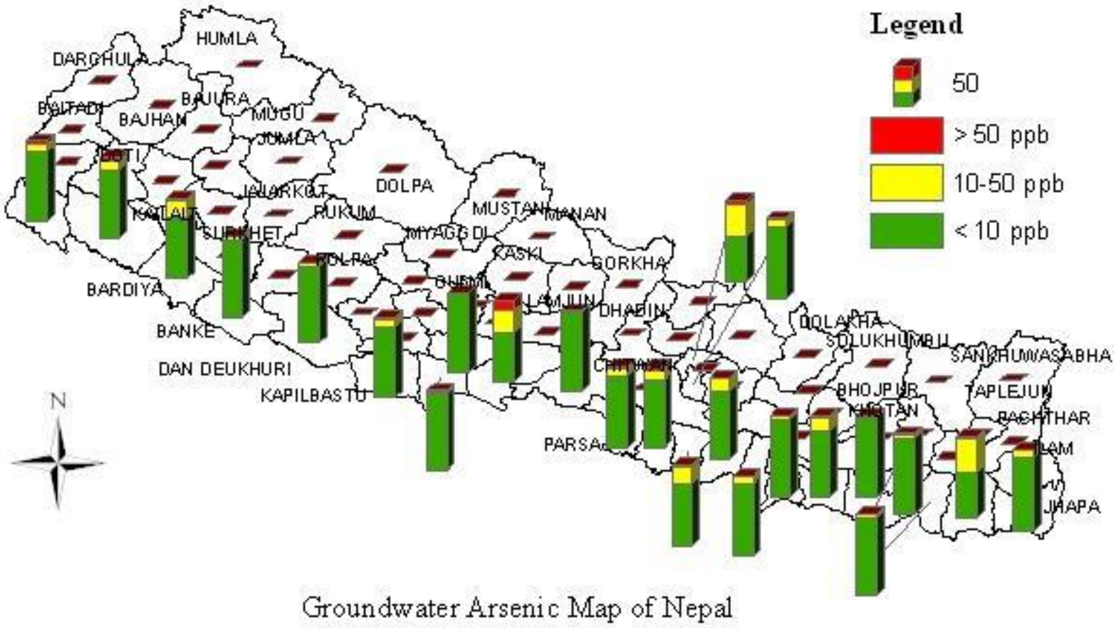
	4		30	94
	0		40	100
	7	8	10	90
	5		20	93
	2		30	97
	0		40	100
	6	9	10	91
	4		20	94
	0		30	100
	0		40	100
90	27	6	10	70
	19		20	79
	13		30	86
	2		40	98
	15	7	10	83
	9		20	90
	5		30	94
	0		40	100
	8	8	10	91
	5		20	94
	3		30	97
	0		40	100
	6	9	10	93
	3		20	97
	0		30	100
	0		40	100

(Inter electrode spacing 2cm for all analysis)



A.6 Worldwide distribution of arsenic contaminated regions, showing source of arsenic and numbers of people at risk of chronic exposure

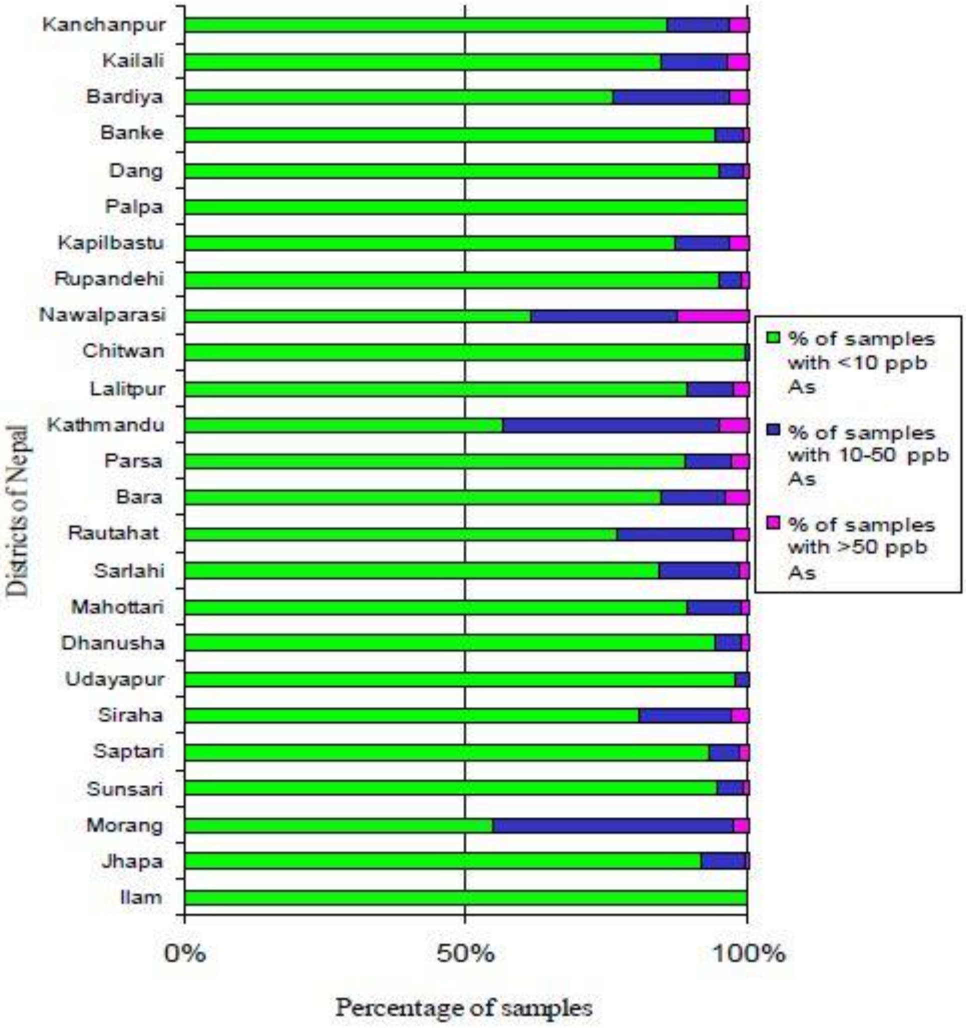
(Source: NSSC, 2010)



A.7 Groundwater arsenic map of Nepal showing proportion of arsenic contaminated samples found in various districts of Nepal.

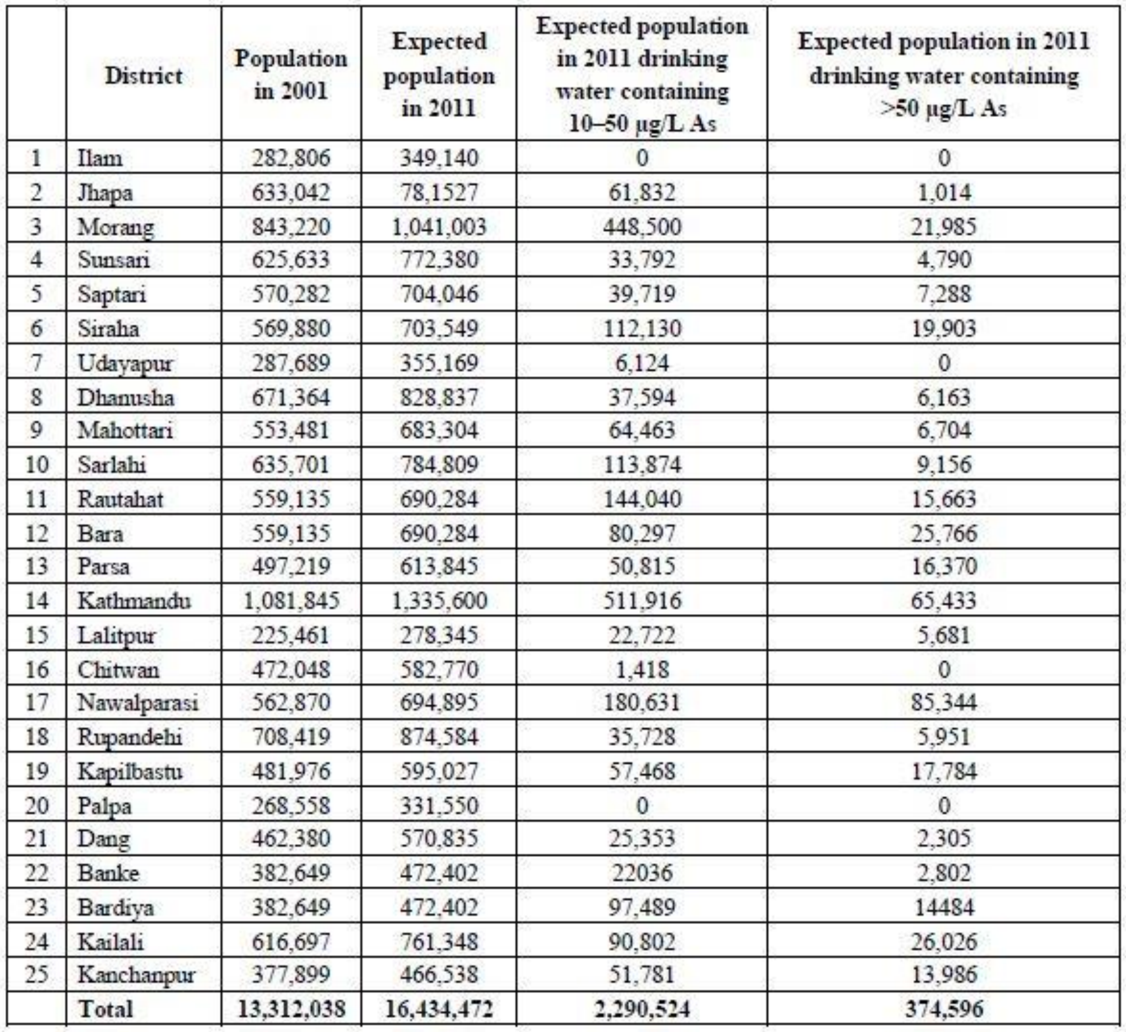
(Source: NSSC, 2010)

A.8 Percentage of arsenic contaminated samples in various districts of Nepal



(Source: NSSC, 2010)

A.9 Table of district wise expected number of population drinking arsenic contaminated water containing 10–50 µg/L As and >50 µg/L As.



	District	Population in 2001	Expected population in 2011	Expected population in 2011 drinking water containing 10–50 µg/L As	Expected population in 2011 drinking water containing >50 µg/L As
1	Ilam	282,806	349,140	0	0
2	Jhapa	633,042	78,1527	61,832	1,014
3	Morang	843,220	1,041,003	448,500	21,985
4	Sunsari	625,633	772,380	33,792	4,790
5	Saptari	570,282	704,046	39,719	7,288
6	Siraha	569,880	703,549	112,130	19,903
7	Udayapur	287,689	355,169	6,124	0
8	Dhanusha	671,364	828,837	37,594	6,163
9	Mahottari	553,481	683,304	64,463	6,704
10	Sarlahi	635,701	784,809	113,874	9,156
11	Rautahat	559,135	690,284	144,040	15,663
12	Bara	559,135	690,284	80,297	25,766
13	Parsa	497,219	613,845	50,815	16,370
14	Kathmandu	1,081,845	1,335,600	511,916	65,433
15	Lalitpur	225,461	278,345	22,722	5,681
16	Chitwan	472,048	582,770	1,418	0
17	Nawalparasi	562,870	694,895	180,631	85,344
18	Rupandehi	708,419	874,584	35,728	5,951
19	Kapilbastu	481,976	595,027	57,468	17,784
20	Palpa	268,558	331,550	0	0
21	Dang	462,380	570,835	25,353	2,305
22	Banke	382,649	472,402	22036	2,802
23	Bardiya	382,649	472,402	97,489	14484
24	Kailali	616,697	761,348	90,802	26,026
25	Kanchanpur	377,899	466,538	51,781	13,986
	Total	**13,312,038**	**16,434,472**	**2,290,524**	**374,596**

(Source: NSSC, 2010)

Photographs

Photo 1 Electrolysis and sampling for analysis