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OPTIMIZATION OF MIX-CHEMICAL COAGULANTS IN WATER PURIFICATION TECHNOLOGY

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Optimization of Mix-chemical Coagulants in Water Purification Technology

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ABSTRACT

Coagulation and flocculation followed by clarification are the most used and important process in the purification of turbid water containing colloidal particles. Salts of aluminum and iron are the most commonly used chemical coagulants in water treatment. The study was aimed at optimizing mix-chemical coagulants in water purification technology. An experimental comparative study where controlled factors were evaluated under various experimental set up. Jar tests were conducted to assess the efficiency of alum and ferric chloride coagulants in dual (1:1 and 3:1 alum to ferric chloride) combinations as well as separately. The highest percentage turbidity removal performance of alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination were 98.7%, 99.1%, 98.7% and 97.8%, respectively. The highest percentage COD removal performance of 71%, 58.1%, 63.6%, and 50.9% were demonstrated for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination, respectively. And the highest percentage TDS removal performance of 55.8%, 72.6%, 81.4% and 81.4% were exhibited for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination. The ferric chloride coagulant demonstrated highest (94.8%) average varying initial turbidity removal. Whereas, 1:1 alum-ferric chloride coagulant combination shows highest (80.8%) concurrent TDS, turbidity and COD average removal. The coagulation experiments indicated that coagulation process effectively removed turbidity from water using 15 to 25 mg/L dosage at 7 to 8 pH range of raw water for the applied coagulants. The selected optimal doses were more effective for medium (150 NTU – 300 NTU) initial turbidity. The study demonstrated strong positive correlation ($r = 0.93$) between coagulant dose and residual aluminum ion concentration which was statistically significant ($P < 0.05$). Also strong negative correlation ($r = -0.97$) was observed between the water pH and residual aluminum ion concentration which was similarly statistically significant ($P < 0.05$). The residual aluminum and ferric ion concentration were greatly reduced when alum and ferric chloride coagulants were used in combinations than used separately. The regular monitoring of residuals in water when using chemical coagulant should be practiced as well as control of coagulant dosage and water pH should be considered in water treatment plants.

Keywords: Optimization, Coagulant, Residuals.

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ACRONYMS

ANOVA	Analysis of Variance
AWWA	American Water Works Association
JU	Jimma University
EDTA	Ethylene Di-amine Tetra-acetate
EPA	Environmental Protection Agency
JUCVAM	Jimma University College of Agriculture and Veterinary Medicine
JWTP	Jimma Water Treatment Plant
NTU	Nephelometric Turbidity Unit
SM	Standard Method
U.S. EPA	United States Environmental Protection Agency
WTP	Water Treatment Plant
DBP	Disinfection by Product
NSF	National Sanitation Foundation

CHAPTER ONE: INTRODUCTION

1.1 Background

Rapid growth of population, urbanization and industrial as well as agricultural activities have increased water demand, particularly in recent decades (Bahvand *et al*., 2010). Particulate and dissolved impurities in water result from land erosion, pickup of minerals, and the decay of plant material with additional impurities from airborne contamination, sewage and industrial discharges, and from animal wastes. Thus, surface water sources, polluted by man and nature, are likely to contain suspended and dissolved organic (plant or animal origin) and inorganic (mineral) material, and biological forms such as bacteria, spores, cysts and plankton (EPA, 2002).

The essential requirements to the treatment of the surface water are removal of particles, removal of dissolved matter, hardness increase and neutralizing, disinfection removal of micropollutants. The most common method to remove particulate matter from surface waters is by sedimentation and/or filtration following the conditioning of the water by coagulation (Meier *et al*., 2006). In North America, both local and national governments recommend the use of optimized coagulation, flocculation and sedimentation as the best available technology to treat surface water by removing organic matter, turbidity, pathogens, and to control chlorination by-product formations (EPA, 1998).

Coagulation process date back hundreds of years and has a great importance in water treatment plants. In this process, chemicals are added which will initially cause the colloidal particles to become destabilised and clump together. When pieces of floc clump together, they may form larger, heavier flocs which settle out and are removed as sludge. In other cases flocs are removed from the water by flotation (Engelhardt, 2010). Salts of aluminum or iron are the most commonly used coagulant chemicals in water treatment because they are effective, relatively low cost, available, and easy to handle, store, and apply. They are rapidly hydrolyzed in water to give a range of products including cationic species, which can be absorbed by negatively charged particles and neutralize their charge. This is one mechanism whereby particles can be destabilized, so that flocculation can occur. Aluminum sulphate commonly called alum or sulphate of alumina is still very widely used although concern about the possible adverse effects of dissolved aluminum has recently been expressed in some quarters. The degree of aluminum absorption depends on a number of parameters such as the aluminum salt administered pH (for aluminum speciation and solubility), bioavailability and dietary factors (Niquette *et al*., 2004).

The alternative coagulants presently available for water treatment are iron salts. The substitution of alum by iron-based coagulants would certainly decrease aluminum residues in water (Douglas *et al*., 1998). This decrease would be lower when complex aluminum based coagulants are used since they do not produce as much aluminum residues as alum (Simpson *et al*., 1988). All iron-based coagulants recommended for drinking water treatment meet the National Sanitation Foundation (NSF) Standard and 60 potable water standards at normal dosages (NSF, 2002). Other aluminum salts used are PAC (polyaluminum chloride), which may have some advantages over aluminum sulphate particularly in the coagulation of `difficult' waters, polyaluminum chlorosulphate and polyaluminum silicate sulphate (EPA, 2002).

1.2 Statement of the problem

Solids are present in water in three main forms: suspended particles, colloids and dissolved molecules. Suspended particles, such as sand, vegetable matter and silts, range in size from very large particles down to particles with a typical dimension of 10 µm. Colloids are very fine particles, typically ranging from 10 nm to 10 µm (Bratby, 1980). Dissolved molecules are present as individual molecules or as ions. In general, suspended particles are simply removed by conventional physical treatment like sedimentation and filtration. Dissolved molecules cannot be removed by conventional physical treatment. Thus, the removal of colloids is the main objective and the most difficult aspect in conventional water treatment. Most suspended solids smaller than 0.1 mm found in waters carry negative electrostatic charges. Since the particles have similar negative electrical charges and electrical forces to keep the individual particles separate, the colloids stay in suspension as small particles (Shammas, 2002).

The possibility of a human health hazard arising from toxicity, infection or gastrointestinal irritation is appreciated regarding the possible sources of turbidity i. e. silt, bacteria, algae, viruses, macromolecules and material derived from organic soil matter, mineral substances and many industrial pollutants. Moreover, apart from the health point of view, turbidity is objectionable on aesthetic grounds: in other words, the potability of water less than a certain degree of clarity is considered to be doubtful by the general public. Furthermore, turbid supplies are unacceptable to many industrial consumers: for example, process water used in food and paper industries is required to have a high degree of clarity to assure adequate final product quality (Bratby, 1980).

Regardless of aluminum and iron salts are amongst widely available and most commonly used commercial coagulants for water treatment (AWWA and ASCE, 1990) considerations concerning potential ancillary costs and trade-offs associated with each particular coagulant during the evaluation of the coagulants is mandatory.

Hospitals and clinics may have problems with aluminum sulphate water. Kidney disease patients, who are exposed to high levels of aluminum in dialysis fluids and medications, develop dialysis encephalopathy, a progressive form of dementia characterized by speech and behavioral changes, tremors, convulsions, and psychosis (EPA, 2002). Orally ingested aluminum is acutely toxic to humans although this element occurs widely in foods, drinking water and many antacid preparations (Niquette *et al*., 2004).

According to the AWWA and ASCE (1990), in some applications, ferric chloride may be more effective than alum, while ferric sulphate is as effective in some waters and more economical. A case study done by Douglas *et al.* in 1998 illustrated that the use of ferric chloride, at a lower pH, was more efficient than alum for organic removal using enhanced coagulation with comparable costs. The performance of each coagulant fluctuates according to the type of water being treated (Niquette *et al*., 2004).

Alum was shown to have the higher cost relative to the ferric chloride (Yonge, 2012). The complex forms of aluminum coagulant usually cost twice as much as alum because they are derived from these salts (Tardat-Henry, 1989). The production of iron-based coagulants often depends on the availability of iron powders. Iron residues may be used as inexpensive sources. Liquid residues from metallurgical industries, containing ferrous iron in acid solution, may also be used to make iron salt solutions

(Niquette *et al.*, 2004). The substitution of aluminum-based coagulants by alternative coagulants will certainly increase the short term cost of the produced drinking water. The cost of iron-based coagulants depends on its availability and on the cost of transportation. The cost of coagulants accounts for about 5% of the price of the drinking water produced (Niquette *et al.*, 2004).

In order to remove the contaminants from drinking water various coagulation processes studies have been done. Some of these include the study of the effect of dosage and mixing conditions on the flocculation of concentrated suspensions using polymeric coagulants (Ghaly *et al.*, 2007); coagulation of synthetic water by plant seeds (Diaz *et al.*, 1999); optimizing coagulation process for low to high turbidity waters using aluminum and iron salts (Baghvand *et al.*, 2010) and coagulation of low turbidity water using bentonite (Shen, 2005). Guida *et al.* (2007) used alum as coagulant to remove COD and Total Suspended Solids (TSS) from municipal wastewater samples.

This study has been done in reference to aforementioned problems especially pertaining to residual aluminum and ferric ion. In this study the mix-coagulants, namely 1:1 and 3:1 alum-ferric chloride combinations, which are the mixture of most common types of coagulants in water treatment plants of many countries, in comparison to single alum and ferric chloride usage were investigated with the aim of determining their capabilities to reduce turbidity, total dissolved solid and chemical oxygen demand of drinking water. Their effectiveness was evaluated at different pH values and coagulant dosage to find optimal operational conditions for all coagulants.

1.3 Significance of the study

The results of the current study can be used as the baseline data for drinking water treatment facilities to consider the combination of coagulants as alternative to common single coagulant use.

This study can be used as bench mark in investigating a middle ground between aluminum and iron coagulant pitfalls by evaluating different alternatives of coagulant usage in drinking water treatment purification in order to intensify contaminant removal efficiency.

In addition it can be used as a base line data for further studies regarding coagulant optimization and residual coagulant control in water.

1.4 Research Questions

- What is the efficiency of aluminum and ferric chloride in removing turbidity, TDS and COD when used separately and in combination?
- What is the concentration of residual aluminum and iron ion in treated water.
- Under the prevailing experimental set up, what are the optimized parameters and their values?
- Does the combined form of the specified coagulants produce lower or higher result in comparison with the single coagulants used?

CHAPTER TWO: LITERATURE REVIEWS

2.1 Specific water contaminants

2.1.1 Turbidity

Turbidity may be identified as the lack of clarity of water. However, turbidity should not be confused with colour; it is very possible for a water to be dark in colour yet clear and not turbid. Suspended matter results in 'cloudiness' of the water and a very common measurement used to determine 'clarity' or 'cloudiness' of water (or any other liquid) is turbidity. Suspended matter giving rise to turbidity include silt, bacteria, algae, viruses, macromolecules and material derived from organic soil matter, mineral substances, many industrial pollutants and so on. Hence, it is self apparent that removal of turbidity from water involves the removal of a wide variety of substances. Turbidity measurements are strongly influenced by the nature, size, concentration and refractive index of particles in suspension. Therefore, as a consequence there is no direct, correlation between the amount of suspended material in a water sample and the turbidity of the sample (Bratby, 1980).

Turbidity is defined as a 'reduction of transparency caused by the presence of undissolved matter' (WRc, 1994) and provides an indication of the amount of suspended matter present in a liquid. Essentially turbidity is a measurement of the light scatter due to suspended particulates, the size and number of the particles affecting the amount of scatter. Turbidity is most commonly used in the measurement of suspended matter in water and is particularly useful for monitoring of especially potable water treatment, but also wastewater treatment processes as it encompasses a large number of different factors that can affect the treatment processes. Turbidity can easily be measured in Nephelometric turbidity unit (NTU) using a laboratory turbidity meter. Suspended particles, also often referred to as colloidal particles, are negatively charged and therefore continually repel each other. This forces the particles to travel minute distances in constantly changing direction so that they don't settle out, but remain in suspension (Leopold and Frees, 2009). Typical values for lakes and reservoirs range between 1 and 20 NTU where rivers and streams can range from under 10 to 4,000 NTU (Crittenden *et al.*, 2005)

2.1.2 Total dissolved solids

The total amount of dissolved material in water is referred to as the TDS. The unit of measure is milligrams per litre (mg/L) and it is obtained by filtering a known volume of water into a pre-dried and weighed crucible to remove all suspended material and then evaporating the filtered water to dryness (method number 2540-C, Standard Methods, 2000). The weighed residue is adjusted to mg/L value and is referred to as TDS. These dissolved materials cannot be seen and don't contribute to the clarity of the water but they can result in colour in the water and their presence can cause problems later in the water treatment process. Some surface water can contain such a high concentration of dissolved humic and fulvic acids, that they resemble the colour of black tea. In spite of this the water is quite clear and is low in turbidity. However it is obvious that such water would be unacceptable as a purified drinking water (Leopold and Freese, 2009).

2.1.3 Chemical oxygen demand (COD)

COD test is commonly used to indirectly measure the amount of organic compounds in water. It can be used to quantify the NOM and DPP precursors in drinking water treatment (Wallace Brian, 2002). Most applications of COD determine the amount of organic pollutants found in surface water (e.g lakes and rivers), making COD a useful measure of water quality. It is expressed in milligrams per liter (mg/L), which indicates the mass of oxygen consumed per liter of solution. COD is the measurement of the amount of oxygen in water consumed for chemical oxidation of pollutants. COD determines the quantity of oxygen required to oxidize the organic matter in water or waste water sample, under specific conditions of oxidizing agent, temperature, and time (Crittenden *et al*., 2005).

2.2 Coagulation and flocculation

In water treatment, coagulation is the process through which suspended, colloidal and dissolved matter are destabilized by the addition of a chemical (Crittenden et al., 2005). Once the particles have been destabilized by a coagulant and have undergone flocculation, the particles begin to settle (Edzwald, 2011). Traditional coagulants such as ferric chloride, ferric sulfate and alum hydrolyze rapidly when dispersed in water forming insoluble precipitates and compressing the electrical double layer of the suspended NOM particles. The precipitates adsorb to the particles in the water neutralizing their charge, and subsequently allow for interparticle bridging. The concentration of NOM and its composition greatly affect the chemistry of the coagulation process. Flocculation is the process by which the destabilized particles agglomerate and form flocculant particles, or "floc." Velocity gradients and particles undergoing random browning motion cause particles to collide and attach to other particles, increasing the effectiveness of removing turbidity and dissolved material (Crittenden *et al.*, 2005).

2.3 Factors influencing coagulation

In addition to mixing, many factors affect the coagulation process. Some of them are:

2.3.1 Colloid concentration

Colloidal concentration has a large impact on both the required dosage and the efficiency of the coagulation process itself (Shammas, 2002). There are two types of colloids: hydrophilic colloids and hydrophobic colloids. Hydrophobic colloids, including clay and non-hydrated metal oxides, are unstable. These colloids are easily destabilized. Hydrophilic colloids like soap are stable. When these colloids are mixed with water, they form colloidal solutions that are not easily destabilized (Bratby, 1980). The dosage of coagulants required for the destabilization of a colloidal dispersion is stoichiometrically related to the amount of colloidal particles present in solution (Stumm and O'Melia, 1968). However, for dilute colloidal systems, the rate of coagulation is very slow because of the small number of colloids in suspension and therefore, not enough contact between particles is available. Under such conditions increasing the concentration of particulate matter by the addition of a coagulant aid or recycling of settled sludge would improve the coagulation rate. Application of a large coagulant dosage to a dilute colloidal suspension would result in a greater chance of restabilizing the colloids (Shammas, 2002).

2.3.2 Coagulant dosage

The effect of aluminum and iron coagulant dosage on coagulation, as measured by the extent of removing particles causing turbidity in water, has been studied and evaluated in great detail by Stumm and O’Melia. They divided the relationship into four zones starting with the first low-dosage zone and increasing the dosage progressively to the highest dosage that is applied in zone four:

Zone 1: Not enough coagulant is present for the destabilization of the colloids.

Zone 2: Sufficient coagulant has been added to allow destabilization to take place.

Zone 3: Excess concentration of coagulant can bring about charge reversal and restabilization of particles.

Zone 4: Oversaturation with metal hydroxide precipitate entraps the colloidal particles and produces very effective sweep coagulation.

The range of coagulant dosage that triggers the start, end, or elimination of any of the above zones is dependent on colloidal particle concentration and pH value. The zeta potential represents the net charge of colloidal particles. Consequently, the higher the value of the zeta potential, the greater is the magnitude of the repulsive power between the particles and hence the more stable is the colloidal system. The magnitude of the zeta potential is determined from electrophoretic measurement of particle mobility in an electric field (Shammas, 2002).

2.3.3 Affinity of colloids for water

Hydrophilic (water-loving) colloids are very stable. Because of their hydration shell, chemicals cannot readily replace sorbed water molecules and, consequently, they are difficult to coagulate and remove from suspension. The stability of hydrophilic dispersions depends more on their “love” for water than on their electrostatic charge. It has been estimated that suspensions containing such particles require 10–20 times more coagulant than what is normally needed to destabilize hydrophobic particles (Hammer, 1986). Typical examples are the color-producing material in surface water and organic colloids present in wastewater. On the other hand, examples of hydrophobic (water-hating) particles are metal oxides that can be easily coagulated and destabilized. However, the bulk of colloidal particles in turbid water usually exhibit a mixture of hydrophobic–hydrophilic properties resulting in suspensions that are intermediate in the degree of their difficulty to coagulate (Shammas, 2002).

2.3.4 PH value

PH is a measure of H^+ and OH^- ion concentration. The presence of these ions in the potential-determining layer may cause particle charge to be more positive or less negative at pH values below the isoelectric point. At high pH values above the isoelectric point the reverse effect takes place, whereby particle charge becomes more negative or less positive. The isoelectric point is the pH value at which charge is most nearly neutralized. The isoelectric point for aluminum hydroxide is around pH 8. It varies with the ionic strength in solution but is normally in the pH range of 7 to 9 (Amirtharajah and O'Melia, 1999). The solubility of colloidal dispersions is affected radically by pH. Aluminum hydroxide is amphoteric in nature and is soluble at low and high pH. The greatest adsorption occurs in the pH range where there is minimum solubility. Amirtharajah and Mills (1982) reported that optimal coagulation with alum takes place at pH values near 5 and 7. At these points, the positively charged aluminum hydroxide neutralizes the negatively charged turbidity producing colloidal particles, resulting in zero zeta potential. However, in the pH range from 5 to 7 the colloidal particles are restabilized due to charge reversal brought about by excess adsorption of the positively charged aluminum hydroxide species. pH also plays a part in affecting the amount of aluminum residual in the water. The influence of pH on the polymer's behavior and effectiveness in coagulation is particularly important because of the interaction between pH and the charge on the electrolyte. The extent of charge change with pH is a function of the type of active group on the polymer (carboxyl, amino, etc.) and the chemistry of those groups (Shammas, 2002).

3.3.5 Anions in solution

One of the constraints in using alum and iron as coagulants is the occurrence of charge reversal and restabilization of colloids. However, this behavior can be suppressed or eliminated in the presence of high concentrations of anions such as sulfate, silicate, and phosphate (Shammas, 2002). It was found that background concentration of sulfate in excess of 10 to 14 mg/L has the ability to prevent restabilization. Coagulation with alum is brought about by various species of positively charged aluminum hydrooxides. Aluminum hydroxide possesses its lowest charge and lowest solubility at its isoelectric point that lies in the pH range of 7 to 9 (Amirtharajah and O'Melia, 1999). As a result, when the alum dosage is increased within this pH range, sweep coagulation takes place due to the formation of the aluminum hydroxide precipitate. However, at lower pH values (5–7), higher dosages of alum will tend to increase the positively charged alum species that get adsorbed on particles' interface leading to charge reversal and the restabilization of the colloidal particles. Similar concepts and conclusions are applicable to iron coagulants (Shammas, 2002).

2.3.6 Cations in solution

The presence of divalent cations, such as Ca^{2+} and Mg^{2+} in raw water is commonly considered not only to be helpful in the coagulation of negatively charged colloidal clay particles by anionic polymers but also to be necessary. Three reasons have been suggested to be behind this beneficial effect (Black *et al.*, 1965).

1. Compression of the colloidal double layer.
2. Reduction of the colloidal negative charge and minimization of repulsive potential.
3. Reduction in the range of repulsive barrier between adsorbed polymers.

2.3.7 Temperature

Coagulation by metallic salts is adversely affected by low temperature (Amirtharajah and Mills, 1982). However, the effect has been reported to be more pronounced in using alum, hence the recommendation to switch to iron salts when operating under low water temperatures (Morris and Knocke, 1984). Another alternative option is to add bentonite as a coagulation aid. The addition of the negatively charged clay particles will enable the coagulation process to proceed as a result of charge neutralization rather than by sweep coagulation (Amirtharajah and O'Melia, 1999). The increase in rate and effectiveness of coagulation at higher temperatures can be attributed to the following:

1. Increase in velocity of molecules and hence in kinetic energy.
2. Increase in rate of chemical reactions.
3. Decrease in time of floc formation.
4. Decrease in viscosity of water.
5. Alteration in the structure of the flocs resulting in larger agglomeration (Shammas, 2002).

2.4 Chemicals used in coagulation/flocculation

Numerous chemicals are used in coagulation and flocculation processes having their own advantages and disadvantages. Effectiveness, cost, reliability of supply, sludge considerations, compatibility with other treatment processes, secondary pollution, capital and operational costs for storage, feeding, and handling should be considered in selecting these chemicals. Chemicals used in coagulation / flocculation are referred to either as primary coagulants or as coagulant aids. Primary coagulants are used to cause the particles to become destabilized and begin to clump together. The purpose of coagulant aids may be to condition the water for the primary coagulant being used, to add density to slow-settling flocs or toughness so the floc will not break up in the following processes (EPA, 2002).

2.4.1 Coagulant chemicals

Salts of aluminum or iron are the most commonly used coagulant chemicals in water treatment because they are effective, relatively low cost, available, and easy to handle, store, and apply (EPA, 2002).

2.4.1.1 Aluminum sulphate

Aluminum sulphate has been used for several centuries in water treatment and is probably the most well known and commonly used coagulant. Pure aluminum sulphate is white and gives a water-white solution. However the presence of iron as an impurity is common which gives the chemical or the solution a yellow or even an orange colour. Compared to other coagulants, the use of aluminum-based coagulants is not restricted by the availability of their prime components. The cost of these coagulants generally varies with the volume produced and the distance between the production site and the water treatment plant. Aluminum salts are widely used in surface water treatment as coagulants to reduce organic matter, color, turbidity and microorganism levels (AWWA and ASCE, 1990). A large proportion of drinking water treatment plants use aluminum-based coagulants for their clarification treatment (WHO, 1998).

Aluminum sulphate is widely used in the treatment of potable water and behaves in a similar way to ferric salts in that the primary behavior is sweep flocculation following the formation of hydroxides such as $Al\ (OH)_3$. As previously mentioned, aluminum hydroxides have a limited span of pH over which they are insoluble and it is therefore important that the operating pH be carefully controlled to maintain it in the range of 5.8-8.5. Because aluminum hydroxide redissolves at high pH (i.e. pH values over 8.5), it is important that there is sufficient alkalinity in the water during flocculation to ensure complete formation and settling of the insoluble hydroxide (Leopold and Freese, 2009).

The dissolved (or residual) aluminum sulphate then passes through the filters and later reprecipitates in the reservoir or distribution system. A deep layer of aluminum hydroxide precipitate is often found at the bottom of reservoirs where aluminum sulphate is used for treatment of the water. It is therefore important to ensure good pH control throughout the process when using aluminum sulphate. The other situation in which aluminum sulphate is often the product of choice is for the removal of colour (Leopold and Freese, 2009)

Aluminum sulphate - commonly called alum or sulphate of alumina - is still very widely used although concern about the possible adverse effects of dissolved aluminum has recently been expressed in some quarters. Other aluminum salts used are PAC (polyaluminum chloride), which may have some advantages over aluminum sulphate particularly in the coagulation of `difficult’ waters, polyaluminum chlorosulphate and polyaluminum silicate sulphate (EPA, 2002).

2.4.1.2 Ferric chloride

Solutions of ferric sulphate and chloride are aggressive, corrosive acidic liquids, the chloride more so than the sulphate. They behave as coagulants in a corresponding fashion to aluminum sulphate but form ferric hydroxide floc in the presence of bicarbonate alkalinity (EPA, 2002). Ferric coagulants may have some advantages when coagulating certain types of water. First, coagulation is effective over a wider pH range, usually from pH 4 to 9. However, best performance is between pH 3.5 and 6.5 and above 8.5. Second, a strong and heavy floc is produced, which can settle rapidly. Third, ferric salts are more effective for removing color, taste, and odor-producing matter **(**Shammas, 2002).

Ferric chloride is used in both drinking water and waste water treatment although the application technology is completely different in each. In drinking water, ferric chloride can be used as a coagulant both for turbidity removal and for the removal of colour or other organic contaminants. When carrying out turbidity removal only, ferric chloride is usually used at pH values of around neutral pH where sweep coagulation tends to be the predominant reaction (Amirtharajah and O'Melia, 1990).

One of the main advantages of ferric chloride as a drinking water coagulant is the fact that it is a relatively inexpensive chemical, although this advantage is offset to some extent by the following:

- Ferric chloride consumes alkalinity and therefore lime addition is often required in order to ensure that the water remains stable
- Handling problems, since ferric chloride is highly acidic and corrosive and tends to stain all surfaces with which it comes into contact (Amirtharajah and O'Melia, 1990).

When using ferric chloride for the removal of colour and other organic contaminants, the coagulation reaction is most efficient at a pH of around 5. In fact optimal colour and organics removal is achieved using elevated dosages of ferric chloride to achieve a pH of around 5 simply through the effect of the ferric chloride itself (i.e. the reduction in alkalinity caused by ferric chloride will cause the pH to drop), or ferric chloride can be used together with an acid such as hydrochloric acid, to reduce the pH. In fact in such applications the dose selection is based on the amount of ferric chloride required to reduce the pH to around 5. Ferric chloride is generally a very cost effective option for colour and organics removal, especially at smaller plants. Using elevated inorganic coagulants in this way to remove organic contaminants is commonly referred to as 'enhanced coagulation' (Amirtharajah and O'Melia, 1990).

2.4.2 Alkaline chemicals and acids

In soft waters, where the natural alkalinity is insufficient to react with the coagulants, alkalinity must to be added either as lime or soda-ash. The alkalinity serves to neutralize the sulphuric acid which forms, together with hydroxide, when sulphates hydrolyse. If left in the water the acid would recombine with the hydroxide and revert to sulphate. Hydroxide is the desired end product as it is insoluble, floc-forming and heavier than water, and it carries the positive electric charge necessary to neutralize the negative charges of the colloidal particles. Alkaline chemicals are added also, if required, to provide the optimum pH value for coagulation to take place. In very alkaline waters, the addition of the optimum dose, for colour and turbidity removal, of a metallic salt such as alum or ferric sulphate, may not depress the pH to the optimum level for coagulation. Increasing the dose of metallic salt will further depress the pH but will result in a very undesirable increase in the soluble metallic content of the water, which will carry through the filters to supply. Dosing with a strong mineral acid, such as sulphuric acid, will depress the pH without the unwelcome increase in the soluble metallic content of the water (EPA, 2002).

2.4.3 Coagulant aids and polyelectrolytes

Coagulant aids are used to improve the settling characteristics of floc produced by aluminum or iron coagulants. The coagulant aid most used for a number of years was activated silica. It can still give the best results, mainly when used with aluminum sulphate, but a high level of technical expertise is required for its successful use (EPA, 2002).

2.5 Basic coagulant chemistry

Coagulation is a set of physical and chemical reactions, occurring between the alkalinity of the water, the impurities in it and the coagulant(s) added to the water, which results in the formation of insoluble flocs. These are agglomerations of the particulate suspended matter in the raw water, the reaction products of the added chemicals, as well as colloidal and dissolved matter from the water adsorbed by these reaction products. For a specific coagulant (such as aluminum sulphate), the pH determines which hydrolysis species (chemical compounds) predominate. Lower pH values tend to favor positively charged species, which are desirable for reacting with negatively charged colloids and particulates, forming insoluble flocs and removing impurities from the water (AWWA and ASCE, 1990).

The optimum pH for alum coagulation, which is very dependent on the water being, usually falls in the range of pH 5 to 8, approximately. The aluminum hydroxide floc is insoluble over relatively narrow bands of pH, which may vary with the source of the raw water. Therefore pH control is important in coagulation, not only in the removal of turbidity and color but also to achieve the minimum level of dissolved residual aluminum in the clarified water. Residual alkalinity in the water serves to buffer the system (prevent pH from changing) and aids in the complete precipitation of the coagulant chemicals. The amount of alkalinity in the source (raw) water is generally not a problem unless the alkalinity is very low. Alkalinity may be increased by the addition of lime, caustic soda or soda ash. Polyelectrolytes are used to improve the settling characteristics of floc produced by aluminum or iron coagulants (Shammas, 2002).

In the cases of Al^{3+} and Fe^{3+}, it is known that the primary hydration shell consists of six water molecules in octahedral co-ordination. Owing to the high charge on the metal ion, water molecules in the primary hydration shell are polarized and this can lead to a loss of one or more protons, depending on the solution pH. Effectively, this means that the water molecules in the hydration shell are progressively replaced by hydroxyl ions, giving a lower positive charge, according to the following sequence (Leopold and Freese, 2009).

$$Me^{3+} \longrightarrow Me(OH)^{2+} \longrightarrow Me(OH)_2^{+} \longrightarrow Me(OH)_3 \longrightarrow Me(OH)_4^{-}$$

This is an oversimplified scheme, since it is known that dimeric, trimeric and polynuclear hydrolysis products of Al and Fe can form. However, these can often be ignored, especially in dilute solutions, and may not greatly affect the overall metal speciation. The hydrolysis scheme above will proceed from left to right as the pH is increased, giving first the doubly- and singly-charged cationic species and then the uncharged metal hydroxide, $Me(OH)_3$. In the case of both aluminum and iron, the hydroxide of very low solubility and an amorphous precipitate can form at intermediate pH values. This is of enormous practical significance in the action of these materials as coagulants. With further increase in pH, the soluble anionic form $Me(OH)_4^-$ becomes dominant (Leopold and Freese, 2009)

2.6 Optimizing coagulation

The objective of coagulation is to condition, by chemical addition, the impurities in the water so that they will coalesce in flocculation and be removed in clarification and filtration together with the added chemicals. If the water to be treated were as constant in quality and character as groundwater from a deep well, it would theoretically be possible to calculate from analyses of the water the optimum chemical conditions and dose of coagulant required. Surface water is never as constant in quality and character as groundwater; even in a large lake, temperature changes with the season, particulate matter is blown in by wind which may also cause turbidity to increase due to disturbance of sediment. The optimum chemical conditions in terms of coagulant dose and pH for treatment of the water concerned are assessed by means of the Jar test also called the laboratory coagulation test (EPA, 2002).

2.7 Coagulants residual

2.7.1 Residual aluminum

Use of aluminum salts as coagulants in water treatment may lead to increased concentrations of aluminum in treated water. Where residual concentrations are high, aluminum may be deposited in the distribution system. Disturbance of the deposits by change in flow rate may increase aluminum levels at the tap and lead to undesirable colour and turbidity (WHO, 1996). Residual aluminum in treated waters has become a great concern. Residual aluminum comes from two sources: floc carryover and soluble aluminum (FSC Architects and Engineers, 2003).

Concentrations of aluminum at which such problems may occur are highly dependent on a number of water quality parameters and operational factors at the water treatment plant. High soluble concentrations result when the pH is less than 6.0. In order to minimize the amount of soluble aluminum the pH should be maintained in the range of 6.0 to 6.8. Floc carryover can be reduced through the use of coagulant aids, sedimentation and filtration (FSC Architects and Engineers, 2003). It is important to undertake routine unit process analysis when aluminum sulphate is added to the treatment process to ensure that chemicals such as aluminum are not carried over in the treated water. Though the Ethiopia does not have any regulations on aluminum in drinking water, many other jurisdictions do including those listed in Table 1.

Table 1: - Aluminum guidelines in drinking water

S. No.	Jurisdiction	Aluminum Limit
1	Guidelines for Canadian Drinking Water Quality	100 µg/l
2	USEPA (1991)	50 to 200 µg/l
3	European Economic Community	50 µg/l to 200 µg/l
4	World Health Organization	200 µg/l
5	AWWA Recommended Operating Level	200 µg/l
6	California Code of Regulations	1000 µg/l

As a best practice, residual aluminum concentrations should be maintained below 0.1 mg/L whenever possible. All conventional treatment plants should be able to approach this value. If aluminum residuals exceed 0.3 mg/L, plant operations should be assessed (FSC Architects and Engineers, 2003).

2.7.2 Residual iron

The iron element has no any direct adverse health effects for humans. It is found in multivitamins; however, iron in normal drinking water has no nutrient value. For the water to contain beneficial amounts, the taste of the water would be rather unpleasant. Clothes laundered in water containing iron or manganese above certain concentrations often come out stained. They can also lead to stains on plumbing fixtures such as sinks and toilets. The biggest problem, however, is that they promote the growth of a group of organisms called autotrophic. These bacteria use non-carbon sources such as iron and manganese for their food. They form thick slime layers inside pipes and storage tanks. These slime layers can cause their own problems when they become loose and create dirty water and customer complaints. But the slime layers also consume chlorine and can harbor pathogenic organisms. The guidelines state an aesthetic objective of 0.3 mg/L for iron, and 0.05 mg/L for manganese (Manual, FSC Architects and Engineers, 2003).

2.8 Jar tests

The best approach for determining the treat ability of a water source and determining the optimum parameters (more effective coagulant, required dose rates, pH, flocculation times, most effective coagulant aids) is by use of a Jar tester. The normal procedure when conducting a Jar test is to initially find the best performing coagulant and dose rate and then to determine the optimum pH and temperature for the chosen coagulant and dose rate. Performance is usually judged on turbidity and then on color removal (EPA, 2002).

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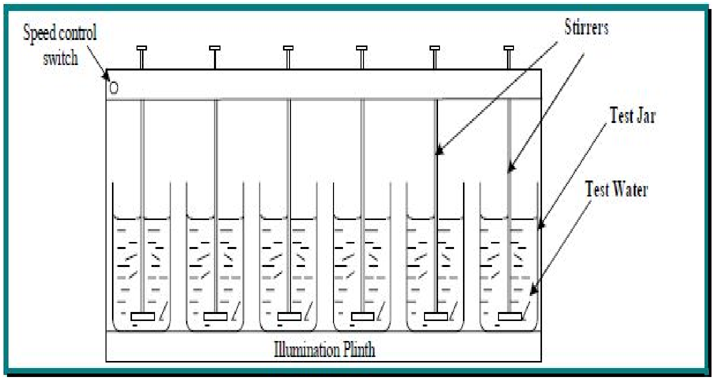

Figure 1: - Jar test apparatus.

CHAPTER THREE: OBJECTIVES

3.1 General objective

The main objective of this study is to optimize mix-chemical coagulants in water purification technology.

3.2 Specific objectives

- To investigate the removal performance of aluminum sulphate and ferric chloride for turbidity, TDS and COD under separate experiment.
- To determine residual iron and aluminum concentration in treated water.
- To search optimized parameters of mix chemical coagulants in turbidity removal.
- To evaluate the best performing coagulant or/and coagulant combination to remove turbidity, TDS and COD.

CHAPTER FOUR: METHODS AND MATERIALS

4.1 Study area and period

The study was conducted in Jimma University School of Civil and Environmental Engineering laboratory and Department of Environmental health science and technology; Environmental biology laboratory from July 11, 2016 – September 9, 2016.

4.2 Study design

An experimental comparative study where controlled factors were evaluated under various experimental set up. Two test sets were designed where one set was employed as a control group and the other one was experimental group.

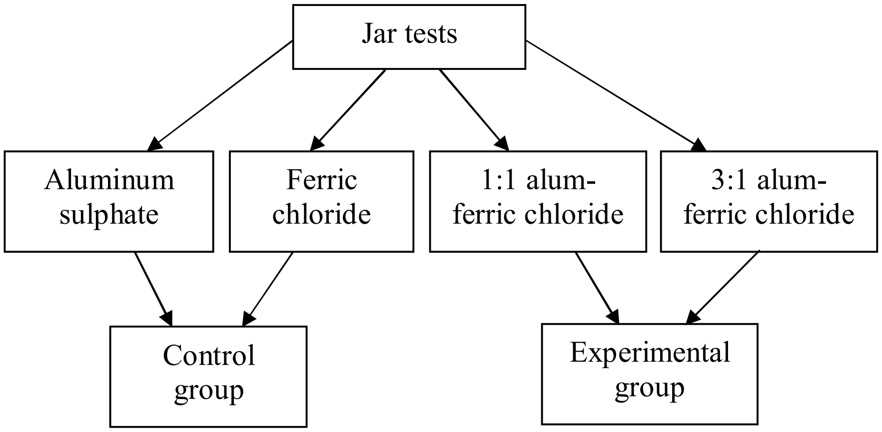

Figure 2: - Principal Study design.

4.2.1 Sample preparation and sampling technique

For reagent preparation, deionized water was used throughout the study for the sake of quality assurance. It was transported from JUCVAM Horticulture and Plant Science department. The deionized water was stored by Jerrycan which was thoroughly washed by 0.5N HCl and rinsed three times by deionized water before collection.

In this study, synthetic test water and real surface water was used as a sample. Four sample of synthetic test water was prepared under different levels of initial turbidity that was within the range of 30-500 NTU, which is a common characteristic range of the influent of water treatment plants. The samples were defined as follows: 30, 150, 300 and 500. These levels were obtained by adding different samples a certain volume of artificial water that was prepared with different proportions of water and clay passed through the sieve no.200 in order to introduce suspended solids and organic matter (Fazeli *et al.*, 2014).

Raw real water sample was collected from the influent of coagulation process unit of Jimma water treatment plant on different three days to get the required varying initial turbidity.

4.2.2 Coagulant stock preparation

Aluminum sulfate, $Al_2(SO_4)_3.18H_2O$ and ferric chloride, $FeCl_3$, was used as a coagulant. Stock solutions were prepared by dissolving 10.0 grams of alum and/ or ferric chloride in to 1,000 mL deionized water, in which 1 mL applied on a sample of 1000 mL represents a concentration of 10 mg /L when added to 1,000 mL of water to be tested (Satterfield, 2005). The 1:1 and 3:1 alum-ferric chloride coagulant combination were formed by mixing the standard stock solution of alum and ferric chloride to make the required coagulant dosage in mix-coagulation experimental study.

4.2.3 Experimental procedures

Jar test was conducted to assess the efficiency of chemical coagulants in Jimma University School of Civil and Environmental Engineering laboratory unit and Department of Environmental health to determine the effectiveness of alum, ferric chloride and alum-ferric chloride coagulant combinations in treating synthetic test water and real surface water.

Each jar was filled with 1000 mL of sample measured with a graduated cylinder. The coagulant dose destined for each jar was carefully measured into 1000 mL beakers. The stirrer speed was set on 200 rpm for 1 minute. After 1 minute, the mixing speed was reduced and was set on 20 rpm for 15 minutes. After this time period, the stirrer was turned off and flock allowed settling for 30 minutes (EPA, 2002). Samples were then withdrawn from the where located 20 mm below the water level for analysis (Baghvand, 2010). The turbidity, residual pH and temperature were measured immediately after the end of settling time of Jar test. Summary of experimental characteristics for Jar test experiments and coagulant physical property were presented in Table 2.

Table 2: - Experimental characteristics for Jar test experiments

Characteristics	Description
Coagulants	Aluminum sulfate and ferric chloride
Coagulant dose range	5-45 (mg/L)
pH range	4-9
Initial turbidity	30-500 (NTU)
Rapid mixing	60 sec at 200 (rpm)
Slow mixing	15 min at 20 (rpm)
Settling	30 (min)

4.2.4 Experimental set up

The effectiveness of the two coagulants (alum and ferric chloride) for the turbidity, TDS and COD was determined according to the aforementioned procedure. Both coagulants were applied separately as well as in 1:1 and 3:1 alum to ferric chloride ratio combinations. The dosage of coagulants applied for each experiment started at 5 mg/L, with an increase of 10 mg/L between each sample. After preparing the samples and stock solutions, Jar tests were carried out for each level of initial turbidity defined in sample preparation, where a certain amount of coagulant was added to each beaker.

Optimization of pH and coagulant dose in the coagulation process

A known volume of prepared aluminum sulphate or ferric chloride solution was added to jars containing 1000 mL of raw water at different pH values adjusted with 0.5N H_2SO_4 and 1N NaOH. Then, it was mixed at a speed of 200 rpm for 1 minute followed by 20 rpm for 15 minute and 30 minute settling time. To optimize the pH of the coagulation process, Jar tests were conducted over the pH range of 4 - 9 and constant coagulant dosages of 15 mg/L. Similarly, to investigate the optimum coagulant dose, the pH value of the raw water was maintained at an optimum pH as determined above and the coagulant dosages ranged from 5 to 45 mg/L (Davis, 2010).

Optimized pH and coagulant dosage suitability test to varying initial turbidity

A known volume of prepared aluminum sulphate or ferric chloride solution was added to the jars with 1000 mL of raw water. To investigate suitable initial turbidity of the optimized pH and coagulant dosage, the pH value of the raw water and coagulant dosage was maintained at an optimum as determined above where as the raw water initial turbidity is ranged from 30 – 500 NTU.

Determined optimal conditions verification using real water

The real samples were taken on three different days using jerry can from Jimma water treatment plant coagulation process unit influent/aeration process unit effluent. Temperature, pH, conductivity and turbidity were measured onsite during sampling. The Jar tests were done immediately after the arrival of the sample in order to prevent the degradation of sample. One liter sample volume were preserved in refrigerator below $4^{o}C$ for every water sample collected and further water samples were analyzed alkalinity, hardness, COD, TDS, sulfate, phosphate, chloride, calcium and magnesium after three days of the collection. The supernatants of the jars which demonstrated optimal conditions in turbidity removal were stored in plastic bottles and preserved in refrigerator below $4^{o}C$ for further analysis of residual aluminum and iron.

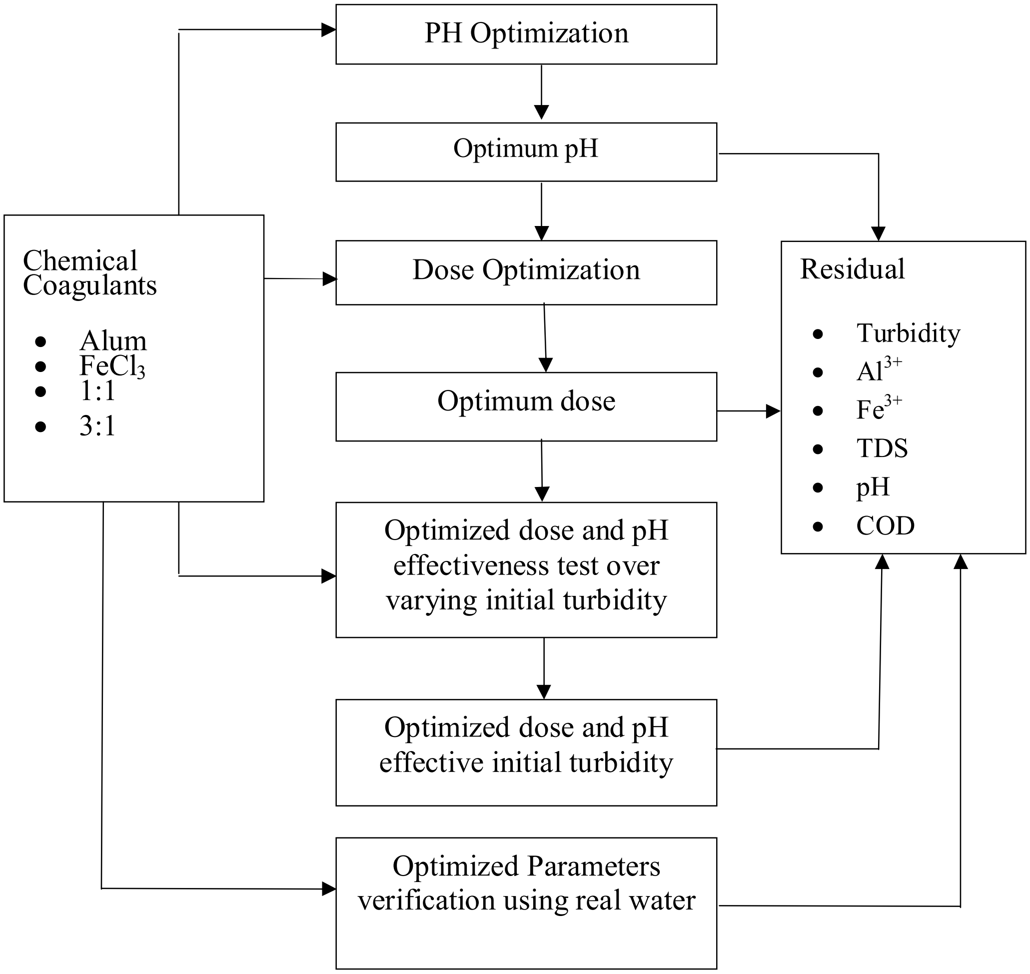

Figure 3:- Conceptual framework of the experimental setup.

The study variables in turbidity, TDS and COD removal include concentration of coagulant (5, 15, 25, 35, 45 mg/L), pH (4, 5, 6, 7, 8, 9) and initial turbidity (500, 300, 150, 30 NTU). After finishing the Jar tests with each of the samples, the reference parameters for coagulation and flocculation were measured again (pH, turbidity and temperature for both real and synthetic water, COD and TDS for real water) in order to determine the coagulant dosage that caused the highest percentage of turbidity removal, in other words the concentration of coagulant that showed the lowest level of turbidity in a sample of each of the 30 Jar tests. Residual turbidity was used as the indicator of performance. The optimal pH and dose for turbidity removal with both aluminum sulfate and ferric chloride were attained by the Jar test experiments. Only the water samples with optimal coagulant dose were analyzed for residual aluminum ion and ferric ion. The water sample water stored by 600mL plastic bottle which was thoroughly washed and rinsed by deionized water three times to reduce contamination. Selected samples were stored in the laboratory refrigerator below $4^{o}C$ and pH 2 by adding HCl acid for further analysis such as residual aluminum and ferric ion which was tested after two weeks of its production.

4.3 Study variables

4.3.1 Dependent variable

- ✓ Coagulants' removal efficiency

4.3.2 Independent variables

- ✓ Coagulant dose
- ✓ Coagulant type
- ✓ Initial turbidity
- ✓ pH
- ✓ TDS
- ✓ COD

4.4 Data collection techniques

Data relevant to the study objectives i.e turbidity, TDS and COD removal was collected. Collection of data was carried out by the principal investigator and laboratory assistant. All the data were recorded on the prepared format.

4.5 Data processing and analysis

Data was compiled and analyzed using Microsoft excels 10 – ANOVA and Minitab 16 – Correlation Coefficient and presented using tables and graphs. All result finding from experimental study was compared with other finding.

4.6 Ethical considerations

Ethical approval and clearance was obtained from School of Civil and Environmental Engineering Student Research Project (SRP).

4.7 Data quality assurance

Extensive quality control measures were implemented throughout this study. Triplicate measurements were taken to assess the consistency of the precision of the analytical instrumentation. Duplicates were done for each Jar test run as well as for each analysis. Quality control measures for laboratory data collection were performed according to the Standard Methods for the Examination of Water and Wastewater (Eaton et al., 2005). Controls and blanks were used to assure the quality of data produced throughout this study. Coagulants were stored in a cool dry, well ventilated cabinet as specified in the material safety data sheets (Yonge, 2012). Samples were stored in the laboratory refrigerator below $4^{o}C$ by adding HCl for metal analysis such as residual aluminum and ferric ion which was tested after two weeks of its production.

4.8 Limitations

During this study we have encountered a lot of challenges for instance lack of access to enough distilled water and well equipped laboratory unit even though they were solved later. However, the time and budget constraint limited further detail studies.

CHAPTER 5: RESULTS

5.1 Experimental studies using synthetic water

5.1.1 Turbidity removal efficiency at different pH and coagulant type

Aluminum sulphate turbidity removal efficiency in the pH range of 5-8 was greater than 95%, while at pH 4 and 9 it was below 90%. The maximum turbidity removal performance of ferric chloride was at pH 5 (96.4%) and pH 8 (98.1%). The 1:1 alum-ferric chloride combination has highest (97.5%) turbidity removal efficiency at pH 8 followed by 95.1% removal efficiency which was observed at pH 6. While, 3:1 alum-ferric chloride coagulant combination highest turbidity removal efficiencies were 97.37% at pH 7 and 96.7% at pH 8. In general, the optimum pH for alum and 3:1 alum-ferric chloride combination was 7 at concentration of 25 mg/L. And for ferric chloride and 1:1 alum-ferric chloride combination it was 8 at concentration of 15 mg/L. Figure 4 gives percentage turbidity removal efficiency at constant dose of 15 mg/L and initial turbidity of 150 NTU for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combinations as a function of solution pH, respectively.

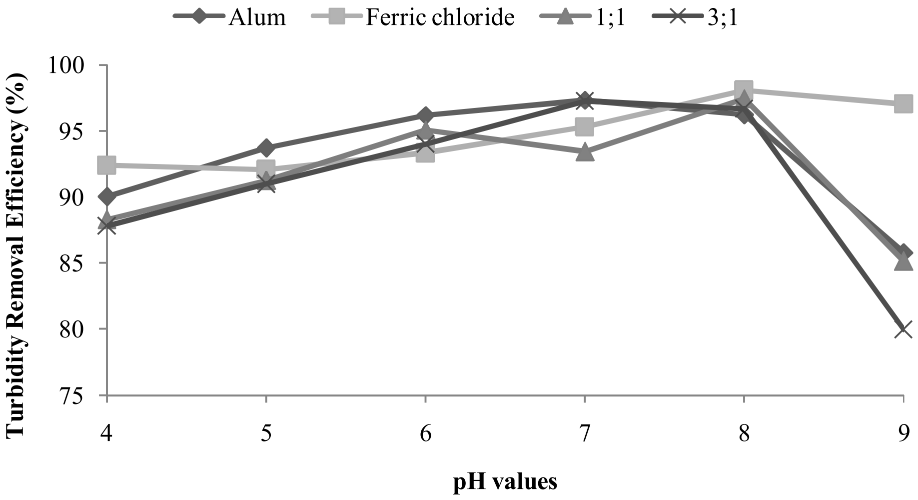

Figure 4:- Percentage turbidity removal efficiency at constant dose and initial turbidity as a function of solution pH.

5.1.2 Turbidity removal as a function of coagulant dose and type

The maximum turbidity removal efficiency was 98.2% at dosage of 15 mg/L, 96.7% at dosage of 25 mg/L, 97.5% at dosage of 25 mg/L and 96.7% at dosage of 25 mg/L for ferric chloride, aluminum sulphate, 1:1 and 3:1 alum-ferric chloride combination, respectively. Ferric chloride turbidity removal efficiency was greater than 94% in 5-45 mg/L coagulant dose range. However, relatively lower turbidity removal efficiency (<87%) was observed for alum, 1:1 and 3:1 alum-ferric chloride combinations at a dosage of 5 mg/L and 45 mg/L. Therefore, the optimum coagulant dose for alum and 3:1 alum-ferric chloride combination was 25 mg/L whereas the optimum coagulant dose for ferric chloride and 1:1 alum-ferric chloride combination was lower than alum, 15 mg/L. The results are presented in Figure 5.

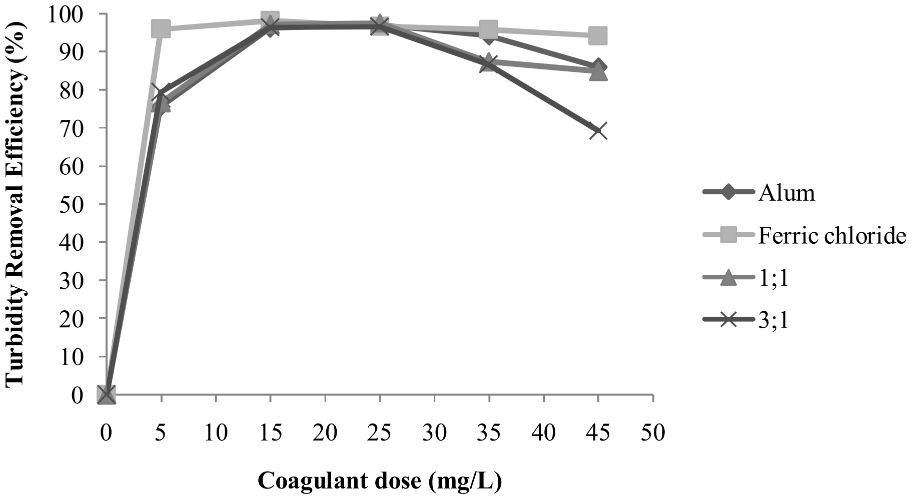

Figure 5:- Percentage turbidity removal as a function of coagulant dose at constant initial turbidity of 150NTU and optimum pH.

5.1.3 Coagulant dose effect on treated water pH

The solution pH is an important factor in determining the physical and chemical properties of the sample and it can be affected by the coagulants (Shammas, 2002). Figure 6 shows that an increase in the coagulant dose is associated with a decrease in the solution pH. The initial pH of solution at which ferric chloride and 1:1 alum-ferric chloride combination dose effect on final solution pH studied were 8. The initial pH of solution at which alum and 3:1 alum-ferric chloride combination dose effect on final solution pH studied were 7. As coagulants were added at a concentration of 5-45 mg/L the final pH value of the solution decreased in proportional to the dose increment rate.

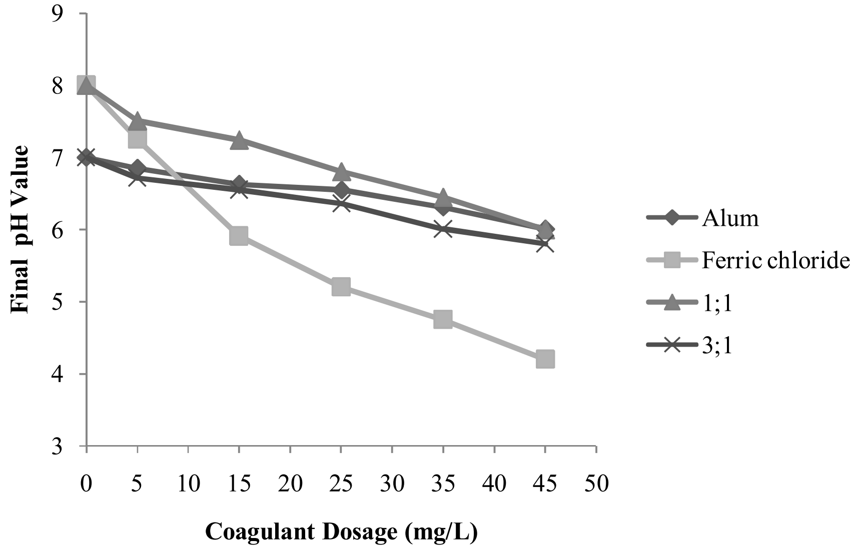

Figure 6: - Effect of coagulant dose on treated water pH.

5.1.4 Effect of initial concentration of turbidity on coagulation process

The optimum dosages and pH of coagulants used in this study were tested for their effectiveness in varying initial turbidity. Accordingly, 25 mg/L alum coagulant concentration was effective over 150-300 NTU which has an efficiency of greater than ninety seven percent. Ferric chloride coagulant dose of 15 mg/L was effective through 30-500 initial turbidity at efficiency of greater than eighty six percent. The highest removal efficiency of ferric chloride was 99.06% at coagulant dose of 15 mg/L at initial turbidity of 300 NTU. At lowest (30 and 150 NTU) initial turbidities 1:1 alum-ferric chloride coagulant combinations was more effective than the rest three coagulants having efficiency of 87% and 98%, respectively. The optimized doses of alum and 3:1 alum-ferric chloride coagulants combination were overdosing for synthetic water having initial turbidity of 30 NTU which require much less concentration to destabilize colloidal. Figure 7 gives percentage turbidity removal at optimal dose 25 mg/L, 15 mg/L, 15 mg/L, and 25 mg/L and optimal pH of 7, 8, 8 and 7 for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combinations as a function of initial turbidity, respectively.

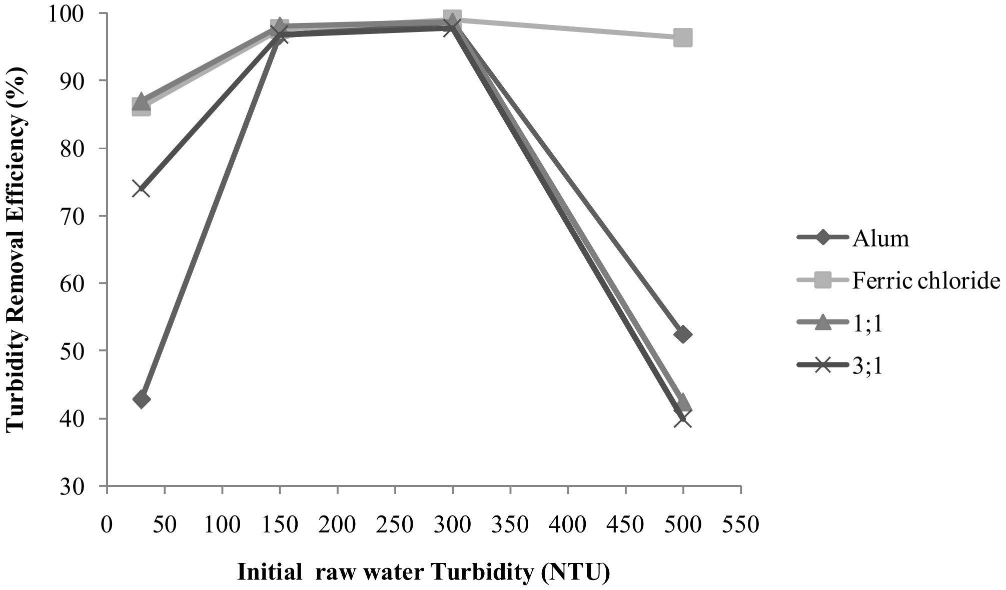

Figure 7: - Percentage turbidity removal at optimal dose and pH as a function of initial turbidity.

5.2 Experimental studies using real water

5.2.1 Raw water characteristics

A 25 L raw water sample collected from the influent of coagulation process unit of Jimma Water treatment plant was delivered to Jimma University Technology Institute Environmental Engineering Laboratory on different three days to get the required varying initial turbidity. Parameters such as pH, temperature and conductivity of the raw water were measured onsite. The collected samples were preserved below $4^{o}C$ physicochemical analysis.

The raw water collected on date 26/08/16 has initial turbidity of 86.2 NTU, TDS of 450 mg/L and COD of 102 mg/L. Also it has low alkalinity (38 mg/L), very low hardness (30 mg/L) and low conductivity (77µs/m). The concentration of cations and anions such as calcium, magnesium, sulphate, phosphate and chloride were 6.4 mg/L, 3.4 mg/L, 0.23 mg/L, 0.02 mg/L and 6 mg/L, respectively. The raw water collected on date 31/08/16 has initial turbidity of 173.6 NTU, TDS of 565mg/L and COD of 149mg/L. Also it has low alkalinity (22 mg/L), very low hardness (24 mg/L) and low conductivity (71.8 µs/m). The concentration of cations and anions such as calcium, magnesium, sulphate, phosphate and chloride were 4.8 mg/L, 2.88 mg/L, 0.26 mg/L, 0.009 mg/L and 4 mg/L, respectively. The raw water collected on date 03/09/16 has initial turbidity of 388 NTU, TDS of 465 mg/L and COD of 146 mg/L. Also it has low alkalinity (32 mg/L), very low hardness (16 mg/L) and low conductivity (77 µs/m). The concentration of cations and anions such as calcium, magnesium, sulphate, phosphate and chloride were 4.8 mg/L, 0.96 mg/L, 0.33 mg/L, 1.1 mg/L and 4.0 mg/L respectively. The results were presented in Table 3.

Table 3: - Raw water quality characteristics

S. No	Parameter		Sample taken on		
			26/08/16	31/08/16	03/09/16
1	Turbidity(NTU)		86.2	388.2	173.6
2	Alkalinity (mg/L)		48	34	44
3	Hardness (mg/L)	Calcium	16	12	12
		Total	30	24	16
4	Conductivity (μs/m)		77	71.8	78.6
5	PH		7.6	7.7	7.3
6	Temperature (^{o}C)		19.5	19.2	19.3
7	Chloride (mg/L)		6.0	4.0	4.0
8	Phosphate (Po$_4^{-3}$ –p (mg/L)		0.02	0.009	1.1
9	Sulphate (mg/L)		0.23	0.26	0.33
10	Calcium (mg/L)		6.4	4.8	4.8
11	Magnesium (mg/L)		3.36	2.88	0.96
12	TDS (mg/L)		450	565	465
13	COD (mg/L)		102	149	146

5.2.2 Optimized coagulant dose verification with real water

Optimized doses of coagulants and coagulant combinations by synthetic water should be verified with real which can contain much interference. The verification of optimum coagulant doses were performed by setting three Jar tests for three samples with initial turbidities of 86.2 NTU (low), 173.6 NTU (medium) and 388 NTU (high) which were collected on three different dates. These tests were duplicated for each level of initial turbidity and the optimal dose values obtained for synthetic water was used as a dose. The Jar test was conducted at optimal dose of 25 mg/L for alum and

3:1 alum-ferric chloride combination and 15 mg/L for ferric chloride and 1:1 alum-ferric chloride combination. The initial turbidities of real water were classified as low (86 NTU), medium (176.3 NTU) and high (388.2 NTU) and the characteristics were aforementioned in Table 3.

For real water having low initial turbidity, the turbidity removal efficiency of alum, ferric chloride, 1:1 and 3:1 alum- ferric chloride combinations were 94.1%, 95.4%, 95.2% and 94.4%, respectively. And for real water having medium initial, the turbidity removal efficiency of alum, ferric chloride, 1:1 and 3:1 alum- ferric chloride combinations were 97.4%, 97.6%, 97.3% and 97.4%, respectively. Similarly, for real water having high initial turbidity, the turbidity removal efficiency of alum, ferric chloride, 1:1 and 3:1 alum- ferric chloride combinations were 97.7%, 96.9%, 95.6% and 96.6%, respectively. Figure 8 gives percentage turbidity removal in low (86.2NTU), medium (173.6NTU) and high (388.2NTU) initial turbidity at optimal dose of 25 mg/L, 15 mg/L, 15 mg/L and 25 mg/L for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combinations, respectively.

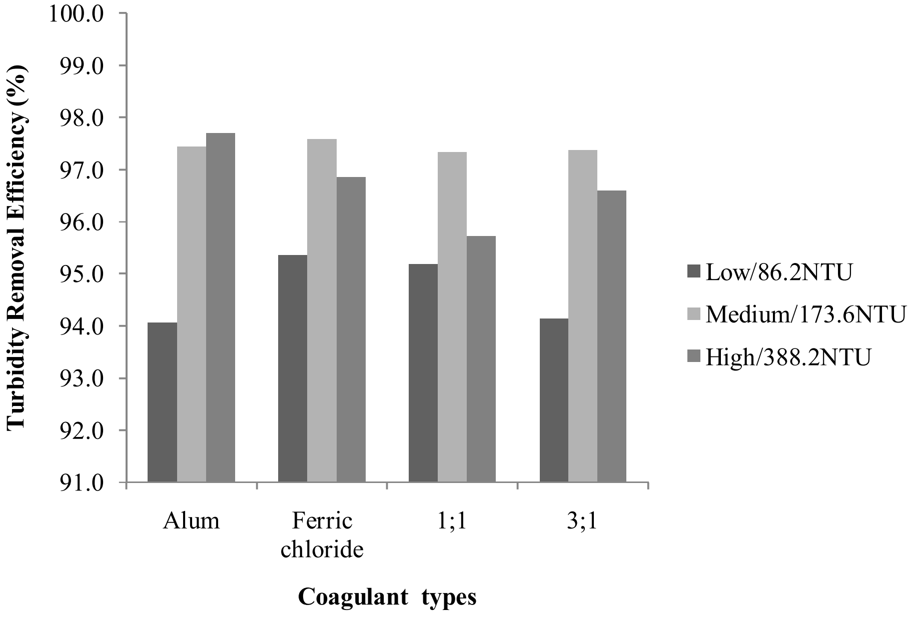

Figure 8: - Percentage low to high initial turbidity removal at optimal dose of the coagulants.

5.2.3 Concurrent removal of contaminants

The coagulation process removal of turbidity, TDS and COD from real water using aluminum sulfate, ferric chloride, 1:1 and 3:1 alum-ferric chloride combinations were presented in Figure 9. As presented in Figure 13, the turbidity and TDS removal efficiency for 1:1 alum-ferric chloride combination was higher than the rest coagulants used in this study. However, aluminum sulphate shows a highest COD percentage removal efficiency (71%). All four coagulants applied in this study show almost similar efficiency at removing turbidity. The TDS removal efficiency of alum was relatively lower (55.8%) than other coagulants. The 1:1 and 3:1 alum-ferric chloride combination coagulants have better TDS removal efficiency (81.42%) than either alum (55.7) or ferric chloride (72.566%). The COD removal efficiency of alum, ferric chloride and 1:1 alum-ferric chloride combination were in a range of 60-70%. While, 3:1 alum-ferric chloride combination shows lower removal of COD (40.94%). Figure 9 gives percentage turbidity, TDS and COD removal in medium (173.6 NTU) initial turbidity at optimal dose of 25 mg/L, 15 mg/L, 15 mg/L and 25 mg/L for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combinations, respectively.

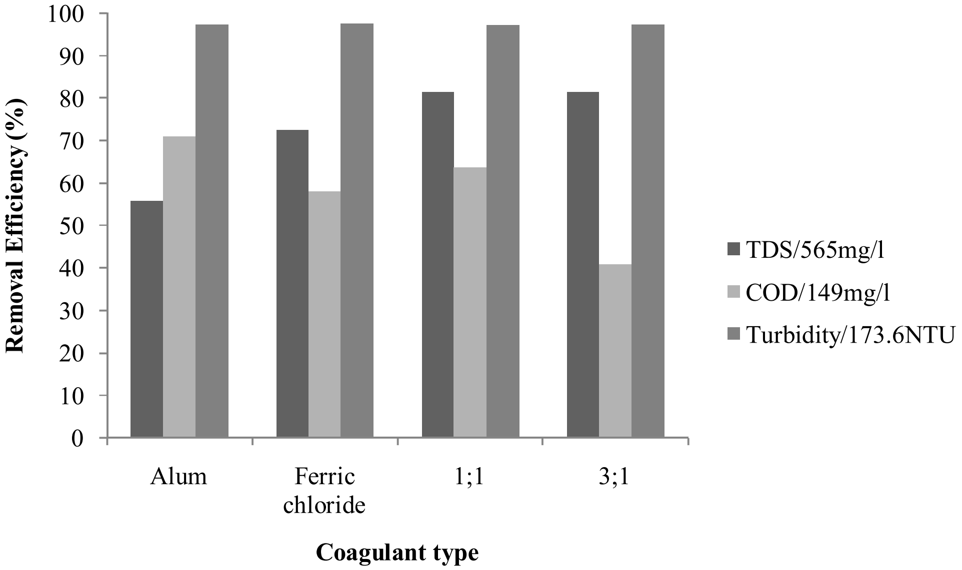

Figure 9: - Percentage turbidity, TDS and COD removal in medium initial turbidity at optimal dose.

5.3 Residuals

5.3.1 Residual ferric ion, Fe^{3+} and aluminum ion, Al^{3+} in treated water

The residual aluminum and ferric ion concentration in treated water were 0.1 mg/L and 0.02 mg/L up on alum coagulant usage at optimal conditions (dose 25 mg/L and pH 7), 0.01 mg/L and 0.4 mg/L up on ferric chloride usage at optimal conditions (dose 15 mg/L and pH 8), 0.06 mg/L and 0.32 mg/L at optimal conditions (dose 15 mg/L and pH 8) when 1:1 alum-ferric chloride combination was used and when 3:1 alum-ferric chloride combination was used 0.09 mg/L and 0.11 mg/L at optimal conditions (dose 25 mg/L and pH 7), respectively as presented in Figure 10.

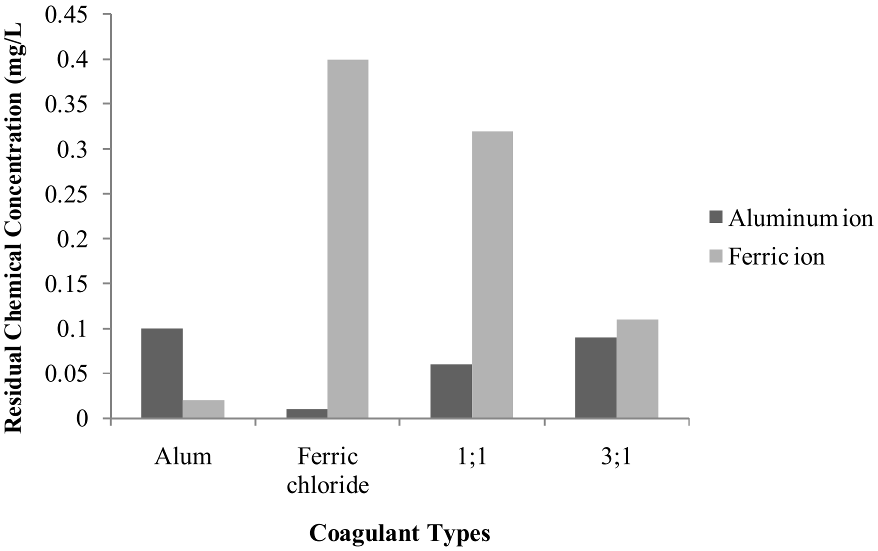

Figure 10: - Residual ferric ion, Fe^{3+} and aluminum ion, Al^{3+} in treated water.

5.3.2 Dose optimization effect on residual aluminum ion (Al^{3+})

In this study as the dosage of chemical coagulant increase the residuals observed increasing. The residual Al^{3+} ion concentration is directly proportional to the dosage of coagulant. However, at optimum alum dose (25 mg/L) the residual Al^{3+} is relatively low. Up on the use of optimal alum coagulant dosage 25 mg/L to treat water having a turbidity of 150 NTU the aluminum ion concentration at solution pH of 6.5 was 0.1 mg/L. The results are presented in Figure 11 as below.

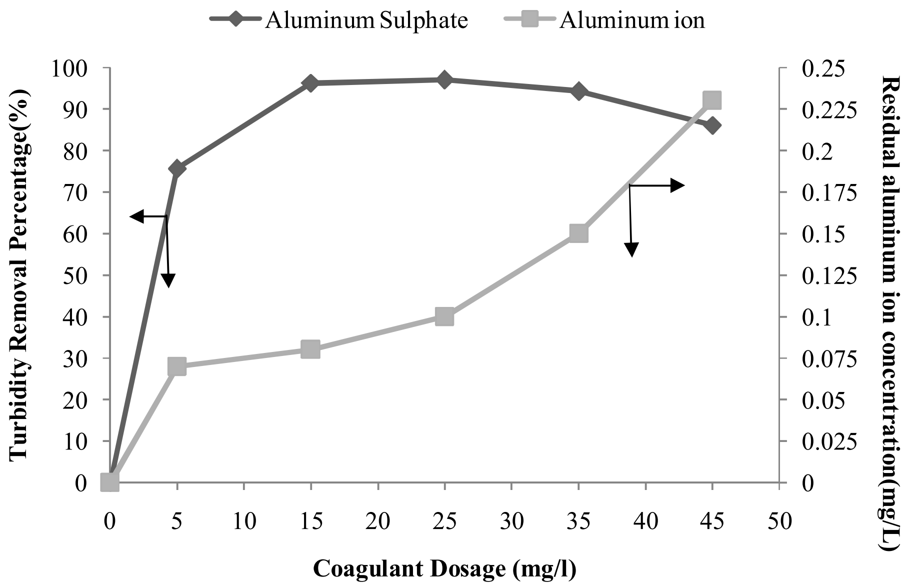

Figure 11: - Dose optimization effect on residual aluminum ion (Al^{3+}) concentration.

5.3.3 Optimal coagulant conditions residuals versus standard

At optimal conditions the residual turbidities were below the guideline of WHO for alum, ferric chloride and 1:1 alum-ferric chloride which was 3.8, 2.8 and 3.9, respectively. However, when 3:1 alum-ferric chloride was used the residual turbidity was above the guideline. The residual aluminum was 0.1 mg/L up on the application of alum at optimal conditions which is within the maximum permissible limit of WHO. The 1:1 and 3:1 alum – ferric chloride coagulant combination contribute 0.06 mg/L and 0.09 mg/L residual aluminum, respectively; which is lower than single alum contribution. The residual iron was 0.4 mg/L up on the application of ferric chloride at optimal conditions which above the maximum permissible limit of WHO. The 1:1 and 3:1 alum – ferric chloride coagulant combination contribute 0.32 mg/L and 0.11 mg/L residual iron, respectively; which is lower than single ferric chloride contribution but above the guideline. The residual TDS concentration on the application all four coagulants were below the guideline of WHO. The results are presented in Table 4.

Table 4: - Residual turbidity, TDS, pH, aluminum and iron ion concentration versus standard at optimal conditions

Residuals in mg/L	Coagulant type				WHO Guideline
	Alum	Ferric chloride	1:1	3:1	
Turbidity	3.82	2.8	3.99	6.7	5 mg/L
Aluminum	0.1	0.01	0.06	0.09	0.2 mg/L
Iron	0.02	0.4	0.32	0.11	0.3 mg/L
TDS	250	155	105	105	500 mg/L
pH	6.52	5.97	6.5	6.28	6.5-8.5

CHAPTER 6: DISCUSSION

Coagulation is used to condition, by chemical addition, the impurities in the water so that they will coalesce in flocculation and be removed in clarification and filtration together with the added chemicals. The optimum chemical conditions in terms of coagulant dose and pH for treatment of the water concerned are assessed by means of the Jar test also called the laboratory coagulation test or the sedimentation Jar test (EPA, 2002).

The best performance of alum was observed at pH 7 over the selected turbidity range but its performance decreased to some extent at pH values of 4 and 9. The coagulation efficiency of alum at pH 6 and 8 was almost close to that of pH 7. The highest turbidity removal was attained at pH 7 when 25 mg/L alum concentration was used except for initial turbidity of 30 NTU and 500 NTU. At optimal dose and pH, turbidity removal efficiency of alum was 42.8%, 96.7%, 98.7% and 52.5% for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU, respectively. Overdosing was observed for low initial turbidity (30 NTU) when the optimal dose (25 mg/L) which was gained at initial turbidity of 150 NTU was used. Turbidity removal efficiency was slightly decreased by increasing alum concentration from 35 to 45 mg/L which may be related to charge reversal and destabilization of colloidal particles due to overdosing as suggested by Stumm and O'Melia (1968). The 1:1 alum-ferric chloride combination shows highest (94.82%) average turbidity removal and lowest standard deviation of 5.58 than the rest three coagulants (Annex III, Table 21). There was statistically significant difference ($P<0.05$) between means of coagulants removal efficiency at varying initial turbidity.

Application of higher alum dose range may improve turbidity removal from relatively high initial turbidity water. Unfortunately, aluminum sulphate coagulant may increase aluminum ion concentration in treated water. Recent research has shown that aluminum from drinking water can be absorbed to some extent in both animals and humans so aluminum in drinking water considered a hazard (WHO, 1996). This is because the aluminum in water following water treatment processes seems to be in a largely "free" (i.e. unbound) form which is a prevalent bioavailable source ingested by humans. In this study as the dosage of chemical coagulant increase the residuals observed increasing. The residual Al^{3+} ion concentration is directly proportional to the dosage of coagulant. However, at optimum alum dose (25 mg/L) the residual Al^{3+} is relatively low. Up on the use of alum as a coagulant to treat water having a turbidity of 150 NTU the aluminum ion concentration at pH 6.5 is 0.1 mg/L. Optimum aluminum sulphate coagulant dose (25 mg/L) contribution to residual aluminum ion concentration is 0.1 mg/L which is below drinking water quality standard set by WHO and USEPA (0.2 mg/L). Based on correlation coefficient analysis done for this study, the coagulant dose and residual aluminum ion concentration were strongly positively correlated (r= 0.94) which was statistically significant (Annex III, Table 22).

According to the finding of this study, the turbidity removal efficiency was varied by pH, alum dose and initial turbidity of water. The obtained result was in accordance with Baghvand *et al*. (2010) and Volk *et al*. (2000) which suggested that the pH of coagulation and dose of coagulant were influential parameter affect turbidity removed. The Baghvand *et al*., (2010) study indicated that the initial turbidity of water was also affecting the turbidity removal efficiency. Under dosing was observed when 5 mg/L alum was used. The added coagulant dose was not much enough to form floc which can be categorized under zone 1 type of coagulation as stated by Shammas 2002.

The best performance of ferric chloride was observed at pH 8 and 15 mg/L dosage. The optimal ferric chloride coagulant dose 15 mg/L was used for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU. But, this dose causes under dosing for raw water having initial turbidity of 500 NTU and overdosing for raw water having initial turbidity of 30 NTU. In this study, turbidity removal efficiency of ferric chloride was relatively stable at all dosages which were in the range of 94% -99%.

At optimal pH and dose, turbidity removal efficiency of ferric chloride coagulant combination was 86.17%, 97.63%, 99.06% and 96.35% for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU, respectively. The highest (99.06%) removal efficiency of ferric chloride was shown at dose of 15 mg/L which is at initial turbidity of 300 NTU. The optimized dose of ferric chloride coagulant was overdosing for synthetic water having initial turbidity of 30 NTU. Therefore, optimal dosage of 15 mg/L ferric chloride can be selected over the applied range of turbidity except for initial turbidity of 30 NTU which require much less concentration to destabilize colloidal. Based on ANOVA done, overall initial turbidity range (30-500 NTU) ferric chloride coagulants has much higher average turbidity removal efficiency (94.8%) and lower standard deviation (5.6) than the rest three coagulants used in this study which have less than 81.6% removal efficiency and greater than 26.6 standard deviation. The difference between means of coagulants removal efficiency was statistically significant ($P<0.05$).

The best performance of 1:1 alum-ferric chloride combination was observed at pH 8 over selected range of turbidity but its performance decreases at pH 4 and 9 which is below 90%. The coagulation efficiency of 1:1 alum-ferric chloride at pH 6 and 8 was almost close to each other. The highest turbidity removal was attained at pH 8 when 15 mg/L alum-ferric chloride 1:1 coagulant combination was used. Overdosing was observed for low initial turbidity (30 NTU) when the optimal dose (15 mg/L) gained at initial turbidity of 150 NTU was used. Turbidity removal efficiency was decreased by increasing coagulant concentration from 25 to 45 mg/L. At optimal pH and dose, turbidity removal efficiency of 1:1 alum-ferric chloride coagulant combination was 87.03%, 98.03%, 98.67% and 42.5% for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU, respectively. For 1:1 alum-ferric chloride coagulants combination the highest turbidity removal efficiency, 98.03% at standard deviation of 0.042 and initial turbidity of 300 NTU was observed. On the contrary, the lowest efficiency (42.5%) was shown at standard deviation of 0.42 and initial turbidity of 500 NTU which is due to under dosing of the optimal dose determined.

The best performance of 3:1 alum ferric chloride combination was observed at pH 7 and dosage of 25 mg/L which was similar optimal pH and dose for alum. The optimal coagulant dose 25 mg/L was used for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU. But this dose shows under dosing for initial turbidity of 500 NTU where as overdosing for initial turbidity of 30 NTU. Up on the 3:1 alum ferric chloride coagulant combination optimal dose and pH usage the residual aluminum ion was found decreased.

Optimized doses of coagulants and coagulant combinations by synthetic water should be verified with real which can contain much interference. To verify the effectiveness of the selected optimal dosages, the Jar tests were conducted at 25 mg/L optimal dose of alum and 3:1 alum-ferric chloride combination and at 15 mg/L optimal dose of ferric chloride and 1:1 alum-ferric chloride combination. The turbidity removal efficiency of alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination were 97.4%, 97.6%, 97.3% and 97.4% for real water having medium (173.6 NTU) initial turbidity, respectively. Whereas relatively lower percentage removal of turbidity was observed for real water having low, 86.2 NTU and high, 388.2 NTU initial turbidity.

The colloidal concentration and pH of the solution, cations and anions in solution, alkalinity of water are among many factors that influence the coagulation process (Shammas, 2002). Based on this fact the optimum conditions set previously using synthetic water were confirmed with real water.

The presence of divalent cations such as Ca^{2+} and Mg^{2+} in raw water is helpful in the coagulation of negatively charged colloidal particle (Black *et al*., 1965). Based on this fact the calcium (Ca^{2+}) and magnesium (Mg^{2+}) concentration in the real water sample taken was low so that their effect in helping the compression of the colloidal double layer was not significant. The presence of anions like sulphate and phosphate will eliminate the occurrence of charge reversal. The occurrence of charge reversal and restablization is one of the constraints in using alum and iron as a coagulant. The concentration of sulphate anion that can suppress the restablization is 10-14 mg/L as stated by EPA 2002. However, in this study the sulphate anion concentration in the real water was in the range of 0.12-0.33 mg/L. Hence, we may conclude that the contribution of sulphate in suppressing the restablization was insignificant.

The pH of low and medium turbidity real water was 7.6 and 7.7, respectively. It is not much far from the optimal pH gained during the study (pH 7 for alum and pH 8 for ferric chloride). As a result the interference of pH of the real water sample collected on the removal of turbidity was low. But, the pH of high turbidity real water sample was 7.3 which to some extent affected the effectiveness of ferric chloride in contaminant removal where as the optimal pH was 8.

The colloidal concentration has considerable impact on both the required dosage and efficiency of the coagulation process. The dosage of coagulants required for the destabilization of colloidal dispersion is stoichiometrically related to the amount of colloidal particles present in solution (Shammas, 2002). Based on this fact the overall efficiency of all coagulants tested in this study become lower for low initial turbidity raw water sample. Additionally, the alkalinity of the raw real water sample taken observed was low so that it negatively affects the efficiency of chemical coagulants since chemical coagulants are more effective in moderately alkaline water.

All four coagulants applied in this study show similar efficiency which ranges from 97.3% to 97.6% at removing turbidity from real water having medium initial turbidity. The total dissolved solid (TDS) removal efficiency of alum (55.8%) was found relatively lower than other coagulants applied in this study. The 1:1 and 3:1 alum-ferric chloride combination coagulants have better TDS removal efficiency (81.42%) than either alum (55.7%) or ferric chloride (72.57%). The COD removal efficiency of alum, ferric chloride and 1:1 alum-ferric chloride combination were in a range of 60 - 70%. While, 3:1 alum-ferric chloride combination shows lower removal of COD (40.94%).

Based on ANOVA done to proof that there is difference between means of coagulants turbidity, TDS and COD removal efficiency the highest COD average removal was about 71% at standard deviation of 1.08 which is achieved by aluminum sulphate followed by 64% at standard deviation of 0.67 which was attained by 1:1 alum-ferric chloride combination. There was statistically significant difference between coagulants mean removal efficiency ($P<0.05$). Total dissolved solid average removal of 81.4% was achieved by 1:1 and 3:1 alum-ferric chloride combinations which show better performance than alum (55.75%) and ferric chloride (75.6%). For all four coagulants studied the average turbidity removal was high (97.3% - 97.6%) up on treatment of the real water having medium (173.6 NTU) initial turbidity. In general 1:1 alum-ferric chloride combination shows the highest (80.8%) concurrent average removal percentage of turbidity, TDS and COD at standard deviation of 15.1 which is relatively lower. The total average removal of turbidity was highest (97.4%) for all types of coagulants than TDS (72.8%) and COD (60.9%) removal. When we evaluate the effectiveness of coagulants in terms of initial turbidity the highest average turbidity removal efficiency of all four coagulants were 97.3% for initial turbidity of 150 NTU at standard deviation of 0.55 and 98.56% for initial turbidity of 300 NTU at standard deviation of 0.52 (Annex III, Table 20). There was statistically significant difference between coagulants mean removal efficiency ($P<0.05$).

According to the statistical tests analysis ferric chloride coagulant demonstrated highest (94.8%) average varying initial turbidity removal. Whereas, 1:1 alum-ferric chloride coagulant combination shows highest (80.8%) concurrent TDS, turbidity and COD average removal. It can be concluded that in the analysis table of four coagulants ferric chloride and 1:1 alum-ferric chloride combination are the most suitable material to perform the coagulation process. This is because of their lower standard deviation and higher average removal efficiency than the other coagulants in concurrent removal of contaminants (Annex III, Table 21). Even though ferric chloride performed better turbidity removal than 1:1 alum-ferric chloride combination in wider initial turbidity range, it was observed that it cause red color water at low and high initial turbidity treatment. According to the optimal rate of alum, ferric chloride and 1:1 alum-ferric chloride combination which is 15 mg/L, 25 mg/L and 15 mg/L, respectively. 1:1 alum-ferric chloride combination was economically suggested for doing chemical treatment. In this statistical test, alpha was 0.05. The optimal pH to use ferric chloride was 8 and for aluminum sulfate it was 7. Turbidity removal efficiency was sufficient to meet national drinking water limits of WHO (5 NTU) at optimum alum, ferric chloride and 1:1 alum-ferric chloride combination dose for waters with initial turbidity of 150-300 NTU.

The beneficial effects of the use of aluminum as a coagulant in water treatment are recognized. Taking this into account and considering the potential health concerns (i.e. neurotoxicity) of aluminum, a practicable level is derived based on optimization of the coagulation process in drinking-water plants using aluminum-based coagulants, to minimize aluminum levels in treated water. Aluminum ion concentration in drinking water above 0.3 mg/L poses a health hazard to human being (WHO, 1996). World Health Organization suggested a number of approaches for minimizing residual aluminum concentrations in treated water. These include use of optimum pH in the coagulation process, avoiding excessive aluminum dosage, good mixing at the point of application of the coagulant, optimum paddle speeds for flocculation, and efficient filtration of the aluminum floc (Letterman and Driscoll, 1988; WHO, 1996). However, this study considered the use of aluminum sulphate in combination with ferric chloride in different ratio as another alternative solution. The combinations of aluminum sulphate and ferric chloride in 1:1 and 3:1 ratios provide similar removal efficiency at dosage of 15 mg/L and 25 mg/L, respectively, which is proposed in order to bargain disadvantages of individual coagulants. Up on usage of aluminum and iron salts as a coagulant it is important to determine the residual aluminum and ferric ion concentration into treated water.

During the use of optimized pH and dose the residual aluminum ion concentration were 0.1 mg/L, 0.06 mg/L and 0.09 mg/L for alum, 1:1 and 3:1 alum-ferric chloride coagulants, respectively. While, the ferric ion concentration were 0.4 mg/L, 0.32 mg/L and 0.11 mg/L for ferric chloride, 1:1 and 3:1 alum-ferric chloride coagulants, respectively. The aluminum ion residual concentration was higher when alum was used alone than when alum-ferric chloride 1:1 and 3:1 combinations was used. According to this study the use of 1:1 alum-ferric chloride at a dosage of 15 mg/L reduce the residual aluminum ion by an average of 40%. Also for iron salts the residual ferric ion in treated water greatly decreases as we use it in combination with aluminum sulphate.

According to correlation coefficient analysis, the coagulant dose and aluminum sulphate turbidity removal efficiency were weakly positively correlated ($r = 0.332$) which is statistically insignificant ($P > 0.05$). Similarly, aluminum sulphate turbidity removal efficiency and residual aluminum ion concentration correlation was week but positively correlated ($P = 0.0135$) which is statistically insignificant ($P > 0.05$). Whereas the coagulant dose and residual aluminum ion concentration were strongly positively correlated ($r = 0.94$) which is statistically significant ($P < 0.05$) (Annex III, Table 22). The correlation coefficient of dose and final pH of the water was $r = -0.98$ which is strong negative correlation and statistically significant ($P < 0.05$). In the similar manner, the correlation coefficient of water pH and residual aluminum ion concentration was $r = -0.97$ which also shows strong negative correlation and statistically significant ($P < 0.05$) (Annex III, Table 22).

The 1:1 alum-ferric chloride was found as an alternative to reduce the residual aluminum and ferric ion concentration in treated water based on the finding of the study regardless of ferric chloride lowest aluminum ion contribution (Table 4).

CHAPTER 7: CONCLUSIONS AND RECOMMENDATIONS

7.1 Conclusion

In this study the Jar test experiments were performed on low to high turbidity waters to find optimal conditions at which better turbidity removal performance of alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride coagulants. The results of the data obtained from over 30 Jar tests conducted throughout the study have been presented using graphical representations shown in Figure 8 through Figure 15 and tables (Table 3 and 4). The synthetic water was used to determine the optimum pH and coagulant concentration ranges required to achieve the maximum turbidity removal percentage and it was later verified with real water. Residual concentration of aluminum and iron ions determination were performed for each coagulant and their combinations using the optimum conditions which were established from the completed Jar tests.

Results indicated that turbidity removal efficiency was varied by pH, alum dose and initial turbidity of water. The coagulation experiments using aluminum sulfate, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination indicated that coagulation process effectively removes turbidity from water using 15-25 mg/L dose of the coagulants. The result of this study shows that ferric chloride was more effective in turbidity removal compared with alum, 1:1 and 3:1 alum-ferric chloride combinations. However, alum attained higher percentage removal in COD. The alum-ferric chloride 1:1 and 3:1 combinations were more effective in the removal of TDS. The optimum pH range for turbidity removal was found 7, 8, 8 and 7 for alum, ferric chloride 1:1 and 3:1 alum-ferric chloride combinations, respectively. The turbidity removal efficiency was within 42.8% – 98.7%, 86.2% – 99.1%, 42.5% – 98.7% and 39.9% – 97.8% for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination over 30 to 500 NTU range of initial turbidity, respectively.

Generally results demonstrated that turbidity removal efficiency was higher for ferric chloride compared to aluminum sulfate, 1:1 and 3:1 alum-ferric chloride combinations at optimum conditions. Also it has narrow removal efficiency range which indicates its consistency. Turbidity removal efficiency was sufficient to meet national drinking water limits of WHO (5 NTU) at optimum alum, ferric chloride, 1:1 alum-ferric chloride combination dose for waters with initial turbidity of 150-300

NTU. The coagulation process and turbidity removal was considerably affected by pH, coagulant dosage, as well as initial turbidity of water for both alum and ferric chloride.

The Jar tests experiment done to verify the optimal doses using real water sample indicated the following results. The highest percentage turbidity removal of alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination were 98.7%, 99.1%, 98.7% and 97.8%, respectively. The highest percentage removal of COD, TDS and turbidity were 71%, 55.8% and 97.4% for alum; 58.1%, 72.6%, and 97.6% for ferric chloride; 63.6%, 81.4% and 97.3% for 1:1 alum-ferric chloride; 50.9%, 81.4% and 97.4% for 3:1 alum-ferric chloride coagulants. The ferric chloride coagulant demonstrated highest (94.8%) average varying initial turbidity removal. Whereas, 1:1 alum-ferric chloride coagulant combination shows highest (80.8%) concurrent TDS, turbidity and COD average removal. In the analysis of four coagulants, 1:1 alum-ferric chloride was found the most suitable coagulant to perform the coagulation process because it has lower standard deviation and higher average efficiency than the other coagulants in concurrent removal of contaminants. And also its residual aluminum and ferric ion contribution to treated water was fair. According to the optimal rate of alum, ferric chloride and 1:1 alum-ferric chloride combination which is 15 mg/L, 25 mg/L and 15 mg/L, respectively, 1:1 alum-ferric chloride combination was economically suggested for doing chemical treatment.

The study revealed that the optimum pH was 7 for alum and 3:1 alum-ferric chloride combination and 8 for ferric chloride and 1:1 alum-ferric chloride combination. Whereas, optimum doses of alum, ferric chloride, 1:1 and 3:1 ferric chloride combinations were 25 mg/L, 15 mg/L, 15 mg/L and 25 mg/L, respectively. The selected optimal doses were more effective for medium (150 NTU-300 NTU) than low (30 NTU) and high (500 NTU) initial raw water turbidity. However, ferric chloride was more effective over wider raw water initial turbidity range as well as pH.

The result of the study shows an increase of aluminum sulphate concentration above an optimum dose greatly increase the residual aluminum ion concentration. The study demonstrated that the residual aluminum and ferric ion concentration when alum-ferric chloride coagulant combinations used were greatly smaller than when they are used separately to treat water having the same initial turbidity. The study exhibited that the aluminum ion and ferric ion concentration was significantly reduced when coagulants used in combination than used separately. Residual aluminum and ferric ion concentration was increasing as coagulant dosages increase when pH and initial turbidity of water was kept constant, whereas residual aluminum and ferric ion concentration is seemed to be more influenced by final pH of water followed by coagulant dose applied. Therefore, pH control is important in coagulation, not only in the removal of contaminants but also to achieve the minimum level of dissolved residual aluminum and iron in the water. The optimization of coagulant dose and pH of raw water were vital in order to produce better drinking water in water treatment plant.

7.2 Recommendations

Based on the finding of this study the following recommendation has been forwarded:

- The optimized 1:1 alum-ferric chloride combination can be used as an alternative coagulant in order to reduce the individual coagulant residual aluminum ion and ferric ion concentration.
- The regular monitoring of residuals in treated water when using chemical coagulant should be practiced and routine control of coagulant dosage should be considered in water treatment plants.
- The influence of rapid mixing parameters, time and intensity of mixing, as well as slow mixing parameters and temperature on turbidity removal by other mix ratio of alum- ferric chloride combinations should be further investigated.
- Currently, the use of mix-chemical coagulant is not at a stage where it can be implemented at full scale. Application of coagulant combinations to meet allowable limits of contaminant should be further studied.

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ANNEXS

Annex I: - Jar test procedure

The basic procedure in carrying out coagulation tests to study the effects of dose of coagulant chemicals is:

i. A bulk raw water sample is obtained and mixed well;
ii. 1 litre samples are measured into the reaction vessels;
iii. Rapid stirring (200 rev / min) is applied for 1 minute;
iv. Coagulant solution to give the desired range coagulant of doses is added to all vessels simultaneously;
v. Rapid stirring is maintained for 1 minute;
vi. Slow stirring (20 rev / min) is applied for 15 minutes, estimating flocculation time and final floc size using the Floc Comparator
vii. Settling is allowed for 30 minutes;
viii. Supernatant samples are decanted and colour, turbidity and pH determined.

The best result will be the dose that has produced the most rapidly-settling floc in the shortest time. If the test has been carried out with a difference of say 10 mg/L of coagulant between each of the vessels, the test can be repeated, centered on the result just obtained and with differences of 2 or 3 mg/L of coagulant between vessels. The best floc will not necessarily be the largest, since large flocs often settle slowly and are easily broken. The best flocculation is shown by coarse dense granular particles which, while settling down, show streaks of clear (not dull or smoky) water between individual particles (EPA, 2002).

Annex II: - Laboratory and analytical result

Table 5: - Optimization of pH for alum

Experiment I - Optimization of pH for Alum														
Parameter	**Final concentration @ dose 15 and Initial Turbidity of 150 NTU**													
	A=4		**B=5**		**C=6**		**D=7**		**E=8**		**F=9**		**CONTROL**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual turbidity	16.12	13.8	10.5	8.3	6.0	5.5	4.3	3.8	6.1	5.2	23.5	19.1	135.2	138.3
pH	3.9	3.8	4.5	4.51	5.7	5.8	6.7	6.4	7.7	7.6	8.1	7.9	7.01	6.85
Temperature	20.1	19.9	19.8	20.1	19.7	19.9	20.1	19.9	19.6	19.8	20.1	19.9	19.01	19.8

Table 6: - Optimization of pH for ferric chloride

Experiment II - Optimization of pH for Ferric chloride														
Parameter	**Final conc. @ dose 15 and Initial Turbidity of 150 NTU**													
	A=4		**B=5**		**C=6**		**D=7**		**E=8**		**F=9**		**CONTROL**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual turbidity	10.7	12.1	5.1	5.7	10.6	9.4	7.93	6.1	3.0	2.8	5.4	3.5	141	136
pH	3.7	3.66	4.16	4.07	4.3	4.2	5.1	4.6	5.4	5.7	5.8	6.2	6.54	6.5
Temperature	22.3	21.8	21.0	21.7	22.1	21.8	22.03	21.1	21.9	21.5	19.7	19.8	21.9	21.3

Table 7: - Optimization of pH for 1:1 alum-ferric chloride combination

Experiment III - Optimization of pH for Alum (50%) + Ferric Chloride (50%)														
Parameter	**Final conc. @ dose 15 and Initial Turbidity of 150 NTU**													
	A=4		**B=5**		**C=6**		**D=7**		**E=8**		**F=9**		**CONTROL**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual turbidity	17.7	17.5	14.3	11.9	6.9	7.8	4.79	5.1	3.4	4.2	31.8	28.3	130.4	133.2
pH	3.8	3.79	4.4	4.5	4.9	4.8	6.0	6.1	6.5	6.5	8.0	8.1	6.6	6.8
Temperature	19.5	20.1	19.7	19.5	19.3	19.5	20.0	19.6	18.9	19.4	20.1	20.1	19.5	20.2

Table 8: - Optimization of pH for 3:1 alum-ferric chloride combination

Experiment IV - Optimization of pH for Alum (75%) + Ferric Chloride (25%)														
Parameter	**Final conc. @ dose 15 and Initial Turbidity of 150 NTU**													
	A=4		**A=5**		**B=6**		**C=7**		**D=8**		**E=9**		**CONTROL**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual turbidity	17.9	18.7	12.1	14.9	8.4	9.6	4.6	3.6	4.1	5.9	26.7	17.9	134.0	129.3
pH	3.8	3.81	4.5	4.6	5.1	5.3	62	6.3	6.6	6.5	6.78	6.8	6.91	6.9
Temperature	19.8	19.9	19.9	20.0	19.4	19.3	19.8	20.0	19.9	19.9	20.01	20.0	20.2	21.1

Table 9: - Optimization of dose for alum

Experiment V - Optimization of dose for Alum												
Parameter	**Final conc. @ Ph 7 and Initial Turbidity of 150 NTU**											
	A=5		**B=15**		**C=25**		**D=35**		**E=45**		**CONTROL**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual turbidity	38.9	34.3	6.5	4.9	4.8	4.1	7.04	10.1	23.2	18.6	136.1	135.4
pH	7.6	7.7	7.1	7.2	6.5	6.6	6.02	5.8	5.3	5.4	6.81	6.9
Temp	19.5	20.1	19.7	20.0	20.01	19.5	20.1	19.6	20.6	20.01	19.9	20.0
Al^{3+}	0.072	0.068	0.093	0.067	0.08	0.12	0.15	0.151	0.248	0.212	0.01	0.021

Table 10: - Optimization of dose for ferric chloride

Experiment VI - Optimization of dose for Ferric chloride												
Parameters	**Final conc. @ Ph 8 and Initial Turbidity of 150 NTU**											
	A=5		**B=15**		**C=25**		**D=35**		**E=45**		**CONTROL**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual turbidity	6.1	6.2	2.4	2.9	5.3	4.6	6.1	6.4	8.79	8.7	141.9	135.2
pH	6.9	7.6	5.8	6.02	4.9	5.1	4.5	5.0	4.2	4.8	6.82	6.9
Temp	20.1	20.2	20.0	20.0	20.3	20.3	20.1	20.1	20.2	20.0	20.9	20.8

Table 11: - Optimization of dose for 1:1 alum-ferric chloride combination

Experiment VII - Optimization of dose for Alum (50%) + Ferric chloride (50%)											
Final conc. @ Ph 8 and Initial Turbidity of 150 NTU											
A=5		B=15		C=25		D=35		E=45		CONTROL	
1	2	1	2	1	2	1	2	1	2	1	2
36.7	33.3	3.6	4.8	13.6	14.01	18.3	19.5	25.2	19.9	134	135
5.9	6.1	6.5	6.4	6.7	6.9	7.2	7.3	7.5	7.5	6.9	6.8
19.9	20.1	19.8	20.1	20.6	21.0	19.9	19.8	20.0	20.0	20.1	19.7

Table 12: - Optimization of dose for 3:1 alum-ferric chloride combination

Experiment VIII- Optimization of dose for Alum (75%) + Ferric chloride (25%)												
Parameters	Final conc. @ Ph 7 and Initial Turbidity of 150 NTU											
	A=5		B=15		C=25		D=35		E=45		CONTROL	
	1	2	1	2	1	2	1	2	1	2	1	2
Residual turbidity	28.1	33.7	5.1	5.5	4.1	6.2	21.9	18.2	49.9	42.3	132.6	139.9
pH	6.5	6.53	6.2	6.3	6.1	6.0	5.7	5.76	6.71	6.79	6.78	6.81
Temp.	19.8	19.4	20.0	19.9	19.8	19.7	20.0	20.2	19.3	19.2	19.9	19.8

Table 13: - Optimization for varying turbidity, alum

Experiment IX - Optimization for varying turbidity, Alum								
Parameters	Final conc. @ Ph 7 and dose of 25 mg/L							
	A=30		B=150		C=300		D=500	
	1	2	1	2	1	2	1	2
Residual turbidity	18.1	16.2	5.1	4.8	4.03	3.61	243	232
pH	6.61	6.53	6.6	6.51	6.53	6.50	6.61	6.63
Temp	19.8	20.01	20.8	20.1	19.8	19.9	19.7	20.0
Fe^{3+}	-	-	-	-	0.023	0.017	-	-
Al^{3+}	-	-	-	-	0.087	0.12	-	-

Table 14: - Optimization for varying turbidity, ferric chloride

Experiment X - Optimization for varying turbidity, Ferric chloride								
Parameters	Final conc. @ Ph 8 and dose of 15 mg/L							
	A=30		B=150		C=300		D=500	
	1	2	1	2	1	2	1	2
Residual turbidity	4.9	3.4	3.6	3.5	2.6	3.03	6.9	9.6
pH	5.8	5.9	5.79	6.0	6.01	5.92	5.89	5.91
Temp	20.01	20.0	19.8	20.0	19.9	20.1	19.7	19.9
Fe^{3+}	-	-	-	-	0.43	0.451	-	-
Al^{3+}	-	-	-	-	0.0102	0.0098	-	-

Table 15: - Optimization for varying turbidity, 1:1 alum-ferric chloride combination

Experiment XI - Optimization for varying turbidity, Alum (50%) + Ferric chloride (50%)								
Parameters	**Final conc. @ Ph 8 and dose of 15 mg/L**							
	A=30		**B=150**		**C=300**		**D=500**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual turbidity	3.79	3.99	3.10	2.8	3.9	4.09	289	286.1
pH	6.5	6.48	6.5	6.4	6.51	6.50	6.52	6.49
Temp	19.9	20.1	20.0	20.01	19.8	19.9	19.8	19.9
Fe^{3+}	-	-	-	-	0.319	0.322	-	-
Al^{3+}	-	-	-	-	0.059	0.061	-	-

Table 16: - Optimization for varying turbidity, 3:1 alum-ferric chloride combination

Experiment XII - Optimization for varying turbidity, Alum (75%) + Ferric chloride (25%)								
Parameters	**Final conc. @ Ph 7 and dose of 25 mg/L**							
	A=30		**B=150**		**C=300**		**D=500**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual turbidity	7.5	8.1	5.1	4.5	6.45	7.0	307	294.2
pH	6.2	6.18	6.21	6.29	6.3	6.25	6.3	6.28
Temp	20.1	19.8	19.9	20.0	20.0	19.7	19.9	18.8
Fe^{3+}	-	-	-	-	0.188	0.0314	-	-
Al^{3+}	-	-	-	-	0.089	0.093	-	-

Table 17: - Optimal doses verification with low initial turbidity real water

Experiment XIII – Real water with LOW (86.2 NTU) initial turbidity clarification										
Parameter	**Coagulant type**									
	A=Alum; 5 mg/L		**B=Ferric chloride; 15 mg/L**		**C=Dual, 1:1 ; 15 mg/L**		**D=Dual, 3:1; 25 mg/L**		**CONTROL**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual Turbidity	5.21	5.01	4.2	3.8	4.1	4.2	5.07	5.03	86.1	85.4
pH	7.0	7.01	6.55	6.58	6.68	6.68	6.78	6.73	7.55	7.5
Temperature	20.1	20.2	20.4	20.5	20.4	20.3	20.1	20.5	20.2	20.3

Table 18: - Optimal doses verification with medium initial turbidity real water

Experiment IXV – Real water with MEDIUM (173.6 NTU) initial turbidity clarification										
Parameters	**Coagulant type**									
	A=Alum; 25 mg/L		**B=Ferric chloride;15 mg/L**		**C=Dual, 1:1 15 mg/L**		**D=Dual, 3:1; 25 mg/L**		**CONTROL**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual Turbidity	4.24	4.63	4.41	3.96	4.2	5.01	4.6	4.5	167.1	164.9
pH	7.0	7.01	6.58	6.61	6.71	6.7	6.81	6.85	7.68	7.62
Temperature	20.2	20.2	20.1	20.2	20.5	20.3	20.2	20.2	20.4	20.5
TDS	235	265	145	165	115	95	105	105	564	565
COD	45.4	41.2	61.49	63.33	53.09	55.31	87.03	88.07	148	145

Table 19: - Optimal doses verification with high initial turbidity real water

Experiment XV – Real water with HIGH (388.2 NTU) initial turbidity clarification										
Parameters	**Coagulant type**									
	A=Alum 25 mg/L		**B=Ferric chloride 15 mg/L**		**C=Dual, 1:1 15 mg/L**		**D=Dual, 3:1 25 mg/L**		**CONTROL**	
	1	**2**	**1**	**2**	**1**	**2**	**1**	**2**	**1**	**2**
Residual Turbidity	8.2	9.6	11.2	15.5	17.0	16.2	11.1	13.3	384.7	379.3
pH	7.01	7.2	6.61	6.67	6.79	6.7	6.8	6.92	7.38	7.36
Temperature	20.1	20.2	20.2	20.19	20.18	20.12	20.2	20.1	19.9	20.0

Annex III: - Statistical analysis results

Table 20: - Optimum initial turbidity ANOVA

Anova: Two-Factor With Replication					
SUMMARY	Alum	Ferric chloride	1;1	3;1	Total
30					
Count	2	2	2	2	8
Sum	85.7	172.4	174.1	148	580.2
Average	42.85	86.2	87.05	74	72.53
Variance	19.85	12.5	0.25	2	370.87
150					
Count	2	2	2	2	8
Sum	193.4	195.3	196.06	193.6	778.36
Average	96.7	97.65	98.03	96.8	97.29
Variance	0.02	0.005	0.02	0.08	0.38
300					
Count	2	2	2	2	8
Sum	197.46	198.12	197.34	195.52	788.44
Average	98.73	99.06	98.67	97.76	98.56
Variance	0.01	0.01	0.00	0.02	0.27
500					
Count	2	2	2	2	8
Sum	105	192.72	85	79.8	462.52
Average	52.5	**96.36**	42.5	39.9	57.82
Variance	2.42	0.13	0.18	3.38	592.15
Total					
Count	8	8	8	8	
Sum	581.56	758.54	652.5	616.92	
Average	72.69	94.82	81.56	77.12	
Variance	732.51	31.14	605.73	631.75	

ANOVA						
Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Sample	9455.01	3	3151.67	1233.89	0.000	3.24
Columns	2192.86	3	730.95	286.17	0.000	3.24
Interaction	4511.99	9	501.33	196.27	0.000	2.54
Within	40.87	16	2.55			
Total	16200.72	31				

Table 21: - Concurrent removal of contaminants ANOVA

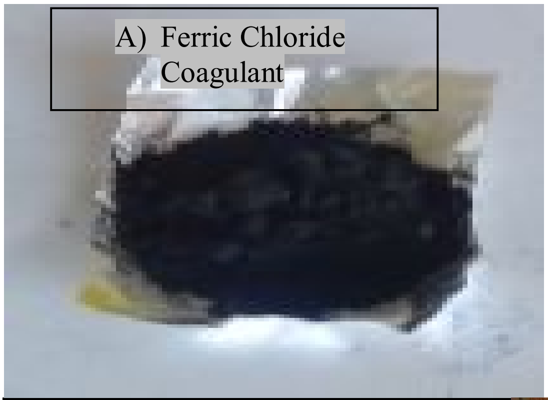
Anova: Two-Factor With Replication					
SUMMARY	Alum	Ferric chloride	1;1	3;1	Total
COD/149 mg/L					
Count	2	2	2	2	8
Sum	141.88	116.24	127.25	101.88	487.25
Average	70.94	58.12	63.62	50.94	60.90
Variance	1.16	7.14	0.45	39.7832	68.40
TDS/565 mg/L					
Count	2	2	2	2	8
Sum	111.50	145.13	162.83	162.83	582.29
Average	55.75	72.57	81.42	81.41	72.78
Variance	11.73	1.87	0.03	53.15	135.01
Turbidity/173.6 NTU					
Count	2	2	2	2	8
Sum	194.9	195.18	194.69	194.76	779.53
Average	97.45	97.59	97.35	97.38	97.44
Variance	3.125	0.32	2.67	0.9522	1.02
Total					
Count	6	6	6	6	
Sum	448.28	456.55	484.77	459.47	
Average	74.71	76.09	80.79	76.58	
Variance	359.49	320.89	228.28	464.15	

ANOVA						
Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Sample	5556.77	2	2778.38	272.46	0.00	3.89
Columns	123.73	3	41.24	4.04	0.03	3.49
Interaction	1184.88	6	197.48	19.37	0.00	2.99
Within	122.36	12	10.19			
Total	6987.75	23				

Table 22: - Relations of parameters

Correlation Coefficient Matrix				
	Dose	Alum removal efficiency	Aluminum ion	Final pH
Dose	1			
Alum removal efficiency	0.33 P = 0.59	1		
Aluminum ion	0.937 P = 0.019	0.013 P = 0.983	1	
Final pH	-0.983 P = 0.003	-0.240 P = 0.698	-0.974 P = 0.005	1

Annex IV: - Pictures of laboratory work


A) Ferric Chloride Coagulant


B) Stock solution preparation


C) Stock solution preparation

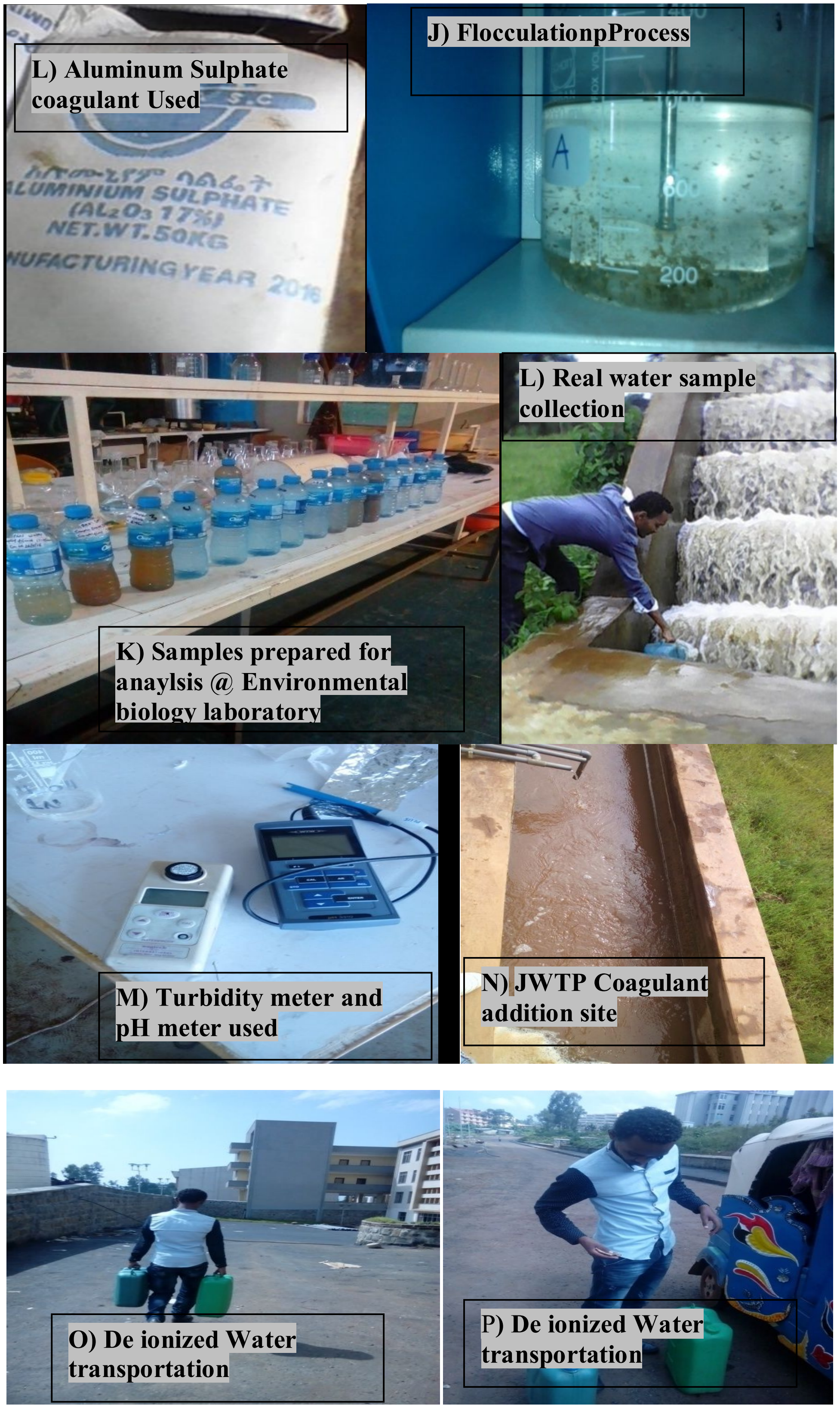

D) Prepared Stock Solutions


E)De ionized water transportation


F) Clay collection


G) Required turbidity preparation


H) Passing prepared sample to beaker



L) Aluminum Sulphate coagulant Used

J) FlocculationpProcess

K) Samples prepared for anaylsis @ Environmental biology laboratory

L) Real water sample collection

M) Turbidity meter and pH meter used

N) JWTP Coagulant addition site

O) De ionized Water transportation

P) De ionized Water transportation

Figure 12: - Laboratory work testimonies (A-P).