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Comparison of Al/Fe/Zr Performance on Coagulation

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SUMMARY

This work addresses an investigation on and comparison of the coagulation performances of aluminium, iron and zirconium coagulants on real wastewater collected in Norway. This work was carried out from January 2014 to May 2014. Coagulation is an essential part of drinking water treatment as well as wastewater treatment. Aluminium and iron coagulants have been in use for many years as part of wastewater treatment. In recent years however, research has been focused on other new coagulants such as zirconium coagulants. This is because regulatory requirements for wastewater treatment are going to become stricter in the future throughout the world. At the same time, wastewater is becoming more difficult to treat due to the presence of new manmade compounds. Thus the continued use of traditional aluminium and iron coagulants may not be able to meet the stricter standards required.

This thesis presents an overview of the coagulation performances of aluminium, iron and zirconium coagulants on various ratios of mixed greywater and blackwater collected from the wastewater collection point at the Norwegian University of Life Sciences in Norway. Analysis and comparison was done using jar tests with two initial wastewater pH values of 6.0 and 7.5. The removal efficiencies of removing orthophosphates, total suspended solids and turbidity were analysed and compared. It was found that zirconium coagulants had the best treatment efficiencies and the least dosages in all types of wastewaters analysed. The overall implications of the results obtained in this study on the future use of new coagulants such as zirconium coagulants are presented and discussed.

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1: INTRODUCTION

In recent years, the coagulation process of wastewater treatment has received considerable attention because of the declining wastewater quality and the need to increase treatment efficiencies due to stricter regulatory requirements worldwide. Coagulation is of particular interest due to its importance in removing organic matter, pathogens, phosphorus and even other inorganic matter such as fluorides and arsenic (Beddow, 2010). Man is always in the process of creating new pharmaceutical products and household chemicals which consist of new and complex compounds that end up in the wastewater. Aluminium and iron coagulants were commonly used throughout the past and are still being used at the present and they certainly work well for meeting the treatment efficiency requirements of today. However, whether they would continue working well into the future remains a question. Coagulants with an increased positive charge such as Zr^{4+}, compared to Al^{3+} and Fe^{3+}, may be a future alternative for increasing treatment efficiencies (Jarvis, et al., 2012).

1.1: Coagulation Theories

The very main aim of coagulation is to reduce particles in raw wastewater by causing them to come together and form larger agglomerations which settle easily due to gravity. Particles in wastewater usually tend to be negatively charged (Cooke, Lesson 9: Colloids and Coagulation, 2014). Due to this, they tend to repel each other and remain suspended in the wastewater without gravitational settling. To enhance settling, and therefore easy removal of particles, the charges on the particles have to be neutralized. There are four widely known theories of coagulation, namely, double layer compression, charge neutralisation, bridging and particle entrapment (Ravina & Moramarco, 1993).

1.1.1: Double Layer Compression

The presence of an electrolyte such as sodium chloride can compress a particular particle's charge influence, or commonly known as the double layer. This thus results in the lowering of the repulsive forces between the particles and the particles can then come together to agglomerate and settle gravitationally. However, it is better if the wastewater already contains these electrolytes in sufficient quantities rather than having to add them, which would lead to extra costs (Ravina & Moramarco, 1993).

1.1.2: Charge Neutralisation

Addition of coagulants such as aluminium salts, iron salts and zirconium salts can induce charge neutralisation effects. The positively charged Al^{3+}, Fe^{3+} and Zr^{4+} can be adsorbed onto the larger negatively charged particles present in wastewater. This again reduces the repulsive forces between the particles and allow them to come together to agglomerate and settle gravitationally. However, overdosing of coagulants can cause the negatively charged particles to become positively charged and again cause repulsive forces that prevent agglomeration. The important difference between charge neutralization and double layer compression is that in the latter, the ions from the salt are assumed not to adsorb to the negatively charged particles (Ravina & Moramarco, 1993).

1.1.3: Bridging

A very large coagulant molecule, such as that of organic polymers, can have large complex structures that effectively capture particles. These coagulants have molecules that are very much larger than the particles usually present in wastewater. This is referred to as bridging. However, bridging works best when used after charge neutralisation so that larger agglomerations can be captured and thus settle gravitationally (Ravina & Moramarco, 1993).

1.1.4: Particle Entrapment

Particle entrapment is similar to bridging, except that it is achieved by adding large doses of inorganic coagulants such as aluminium, iron and zirconium coagulants which precipitate as hydrous metal oxides, which settle gravitationally. The particles become trapped in the hydrous metal oxides as it settles (Ravina & Moramarco, 1993).

1.2: Factors Affecting Coagulation

Various factors can influence the coagulation performance, namely, pH, salt content, alkalinity, turbidity, temperature, mixing effects and coagulant effects (Cooke, Coagulation, 2014).

1.2.1: pH, Salt Content and Alkalinity

The levels of pH, the salt content and alkalinity in wastewater all affect the dosage of coagulant required. Generally, a low pH, high salt content and high alkalinity ensures that positively charged ions are available in the wastewater to aid coagulation. This means that the coagulant dosage can be as low as possible. However, the pH shouldn't be too low, nor the salt content too high, nor the alkalinity too high as this creates a situation of too much positively charged particles that fail to agglomerate and settle effectively (Cooke, Coagulation, 2014).

1.2.2: Turbidity

Turbidity is an indication of how much particles are present in the wastewater. A higher turbidity means higher particle content. Turbidity is measured as the amount of light scattered when light passes through the sample water. A higher number of particles would scatter a higher amount of light, indicating a higher turbidity value. Wastewater turbidity is usually sufficiently large enough to aid coagulation. However, a higher turbidity generally requires more coagulant dosing (Cooke, Coagulation, 2014).

1.2.3: Temperature

Generally as wastewater temperature is lower, the coagulant dosing has to be higher. This is because reactions occur at a slower rate as temperature decreases. A higher water temperature ensures lower coagulant dosage, but too high temperatures, beyond the ambient room temperature requires heating of the wastewater and is thus not practical due to the large energy consumptions and costs involved (Cooke, Coagulation, 2014).

1.2.4: Mixing Effects

Mixing is the process that ensures that the added coagulant can mix evenly and uniformly in the wastewater for efficient coagulation. Usually in coagulation, addition of the coagulants is followed by a short rapid mixing followed by a relatively long slow mixing. If mixing is not done sufficiently, a higher dosing may be required to bring about sufficient treatment efficiency (Cooke, Coagulation, 2014).

1.2.5: Coagulant Effects

The type and dosing of coagulant may be different for different types of wastewaters, as well as different conditions such as temperature and pH. Jar tests are necessary to conclude the optimum dosage of a particular coagulant for a particular wastewater type and condition (Cooke, Coagulation, 2014).

1.3: Aluminium Coagulants

The aluminium coagulants include aluminium sulphate, aluminium chloride and sodium aluminate, basically any compounds that provide the presence of Al^{3+} in the wastewater. Aluminium can form various multi-charged complexes, or metal hydrolysis species, depending on the pH and other conditions of the wastewater. Figure 1 below illustrates this. It is this property of aluminium that makes it a great coagulant (Beddow, 2010).

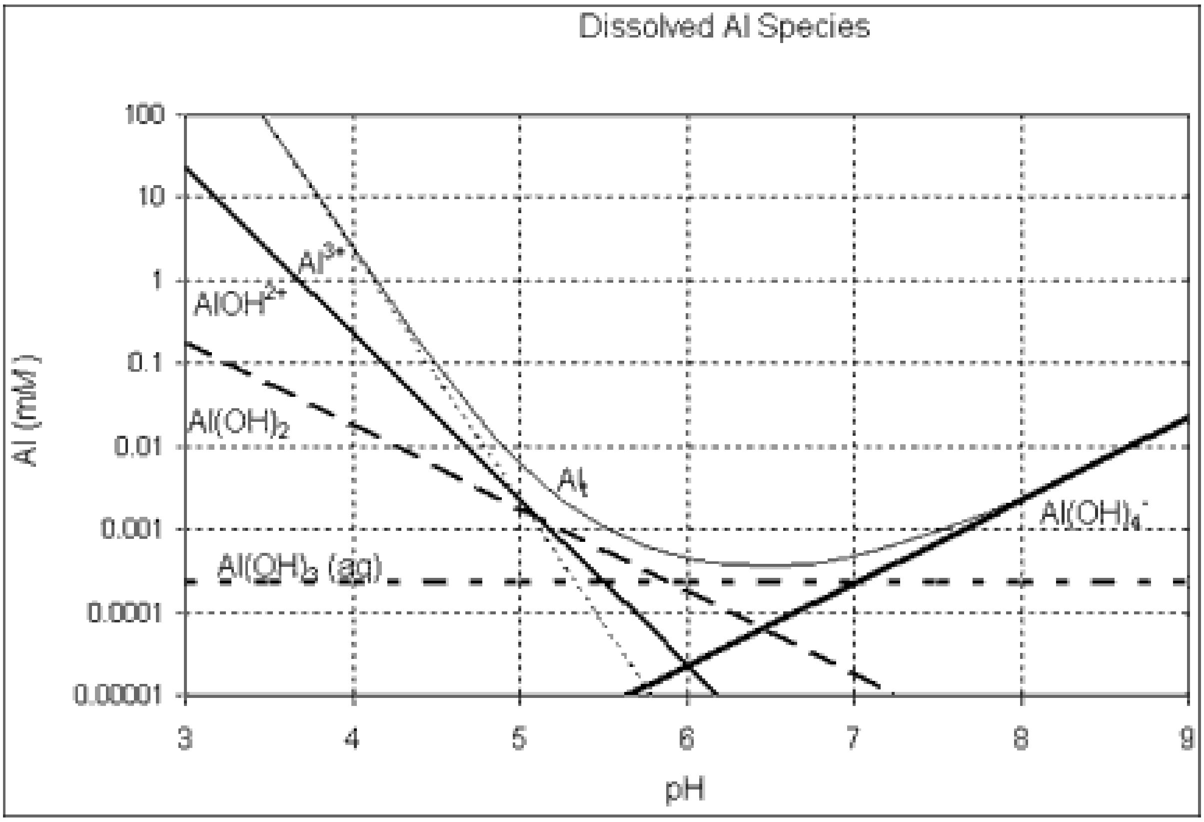

Figure 1: Hydrolysis Species of Aluminium (Environment Canada, 2013)

1.4: Iron Coagulants

The iron coagulants include ferric sulphate, ferrous sulphate, ferric chloride and ferric chloride sulphate. Just like aluminium, iron can form various metal hydrolysis species, depending on the pH and other conditions of the wastewater. Figure 2 below illustrates this. It is this property of iron that makes it a great coagulant (Beddow, 2010).

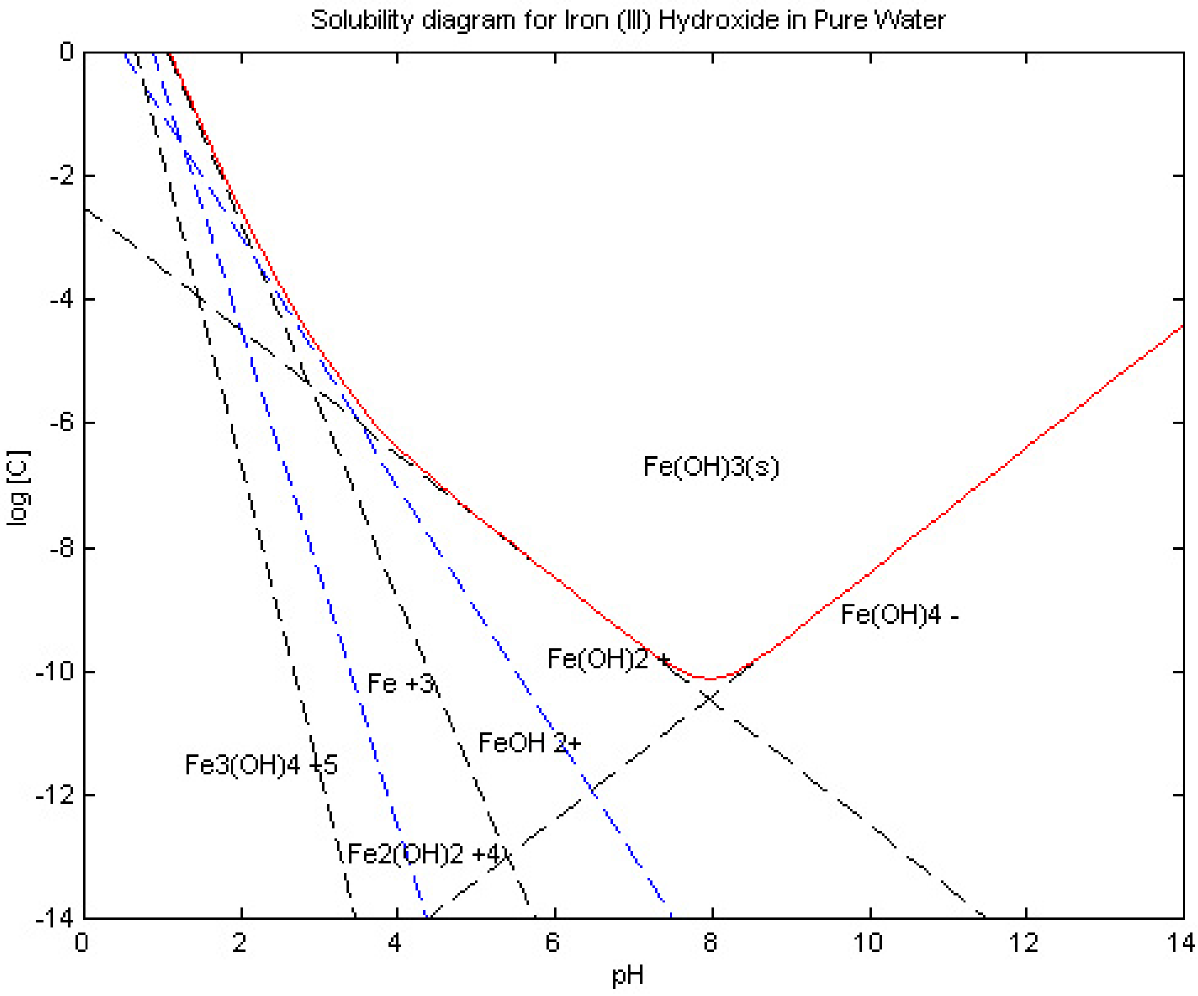

Figure 2: Hydrolysis Species of Iron (Northwestern University, 2014)

1.5: Zirconium Coagulants

Zirconium coagulants have recently been the focus of research due to its normal oxidation state of +4, which is higher than that of aluminium and iron, and also its ability to form metal hydrolysis species up to a charge of 8+ (Jarvis, et al., 2012). This would mean a lower dose for better treatment efficiency as compared to both iron and aluminium coagulants.

1.6: Advantages and Disadvantages of the Various Coagulants

Coagulants do have certain possibilities and limitations over each other, depending upon the conditions of the wastewater. Figure 3 below shows common advantages and disadvantages of the various common coagulants in use today. For example, aluminium sulphate is limited over a small pH range, but produces relatively less sludge while iron (III) chloride works over a larger pH range.

Name	*Advantages*	*Disadvantages*
Aluminum Sulfate (Alum) $Al2(SO_4)_3.18H_2O$	Easy to handle and apply; most commonly used; produces less sludge than lime; most effective between pH 6.5 and 7.5	Adds dissolved solids (salts) to water; effective over a limited pH range.
Sodium Aluminate $Na_2Al_2O_4$	Effective in hard waters; small dosages usually needed	Often used with alum; high cost; ineffective in soft waters
Polyaluminum Chloride (PAC) $Al_{13}(OH)_{20}(SO_4)_2.Cl_{15}$	In some applications, floc formed is more dense and faster settling than alum	Not commonly used; little full scale data compared to other aluminum derivatives
Ferric Sulfate $Fe_2(SO_4)_3$	Effective between pH 4–6 and 8.8–9.2	Adds dissolved solids (salts) to water; usually need to add alkalinity
Ferric Chloride $FeCl_3.6H_2O$	Effective between pH 4 and 11	Adds dissolved solids (salts) to water; consumes twice as much alkalinity as alum
Ferrous Sulfate (Copperas) $FeSO_4.7H_2O$	Not as pH sensitive as lime	Adds dissolved solids (salts) to water; usually need to add alkalinity
Lime $Ca(OH)_2$	Commonly used; very effective; may not add salts to effluent	Very pH dependent; produces large quantities of sludge; overdose can result in poor effluent quality

Figure 3: Advantages and Disadvantages of Common Coagulants (Toprak, 2006)

1.7: Wastewater Characteristics

Wastewater is the water that is discarded after being used by humans. It can contain a very wide variety of substances such as carbohydrates, lignin, fats, soaps, synthetic detergents, proteins, as well as various pharmaceutical and household chemicals, human faecal matter and urine. They may also contain toxic heavy metals such as arsenic, cadmium, chromium, copper, lead, mercury and zinc. They also contain pathogens that may affect human health if exposed (Food and Agriculture Organisation of the United Nations, 2014).

Greywater refers to wastewater generated from the kitchen, washing machines, wash basins and showers. It is all the wastewater generated that excludes wastewater from toilet flush. Blackwater refers to the toilet flush wastewater containing faecal matter and urine.

1.8: The Need to Remove Turbidity

Turbidity gives a direct indication of the amount of particles present in the wastewater, and generally, the higher the turbidity, the higher is the content of particles and the darker the colour of the wastewater. For example, blackwater has a much higher turbidity than greywater. Thus, turbidity is an indicator of water quality as clean water has an extremely low turbidity. Removing turbidity is essentially the same as removing most of the particles that have contaminated the water.

1.9: The Need to Remove Total Suspended Solids

Total suspended solids refer to the solids that are large enough to be removed from the wastewater by filtration through a specified filter pore size. It is similar to turbidity except that turbidity includes particles that are small enough to pass through the filter pores, or what is otherwise known as total dissolved solids. It is important to remove the total suspended solids before the wastewater reaches the filtration process downstream in a wastewater treatment plant. This prevents unwanted filter clogging as the filtration process is important in removing total dissolved solids, thereby further decreasing the turbidity of the wastewater.

1.10: The Need to Remove Orthophosphates

Orthophosphate, PO_4^{3-}, is the soluble form of phosphorus and is a nutrient that is easily taken up by aquatic plants such as algae. In the case where orthophosphates are discharged in large quantities into wastewater recipients, which usually are rivers or lakes, it can cause an increased growth of aquatic plant life which is detrimental to other life by taking up too much oxygen in the water (General Chemical, 2014). This phenomenon is commonly referred to as eutrophication. Thus removal of orthophosphates from the wastewater before discharge is very important to prevent large scale ecological unbalances in the recipient.

1.11: Previous Similar Studies

A very recent study on comparison of coagulation performance of a zirconium-glycine complex coagulant with traditional coagulants such as aluminium sulphate and polyaluminium chloride was published. Charge neutralization was the dominant mechanism involved. It was found that zirconium achieved a higher removal of turbidity than the other two coagulants (Zhang, Wu, Wu, & Hu, 2014).

In another study, it was again found that zirconium performed the best when compared to iron and aluminium coagulants, removing a larger amount of dissolved organic carbon in comparison to the iron coagulant (Jarvis, et al., 2012).

1.12: Objectives of Present Research Work

The present work on coagulation performances was carried out at the water laboratories of the Department of Mathematical Sciences and Technology at the Norwegian University of Life Sciences, with the following objectives:

- To understand the coagulation performances of aluminium, iron and zirconium coagulants on various ratios of mixture of greywater and blackwater, at the initial pH values of 6.0 and 7.5
- To identify the optimum coagulant dosage of all three coagulants on the various ratios of mixture of greywater and blackwater, at the initial pH values of 6.0 and 7.5, in relation to turbidity removal, total suspended solids removal and orthophosphates removal

2: EXPERIMENTAL METHODS

2.1: Collection and Preparation of Wastewater

Wastewater is separated into greywater and blackwater at the wastewater collection point at the Norwegian University of Life Sciences. In this work, it was decided to work with three ratios of mixture of blackwater and greywater, as shown in Table 1 below. Wastewater was collected manually using a 20 litre plastic container.

Table 1: Ratios of Blackwater and Greywater Used

Blackwater	Greywater
100%	0%
50%	50%
0%	100%

It was also decided to work with two initial pH values of 6.0 and 7.5 for each of the mixture ratios. The pH of each of the mixture ratios was measured using a pH meter. The temperature of the wastewater was also measured. Table 2 shows the average pH values and temperatures that were found. The temperatures and the pH values were about the same for all ratios of wastewater mixtures.

Table 2: Measured pH Values of Ratios of Blackwater and Greywater Used

Blackwater	Greywater	Average pH	Average Temperature (^oC)
100%	0%	9.2	20.6
50%	50%	9.3	20.3
0%	100%	9.3	20.5

2.2: Jar Test Setup

Six beakers were used. Each beaker was exactly identical to one another and had a capacity to hold one litre of wastewater. Six individual mixers were used, one for each beaker. The six mixers were connected to a central digital unit which can simulate rapid mixing, slow mixing and sedimentation. The following mixing conditions were used throughout all the jar tests carried out: Rapid mixing for 60 seconds at 400 revolutions per minute, slow mixing for 10 minutes at 30 revolutions per minute and sedimentation for 10 minutes.

2.3: Coagulants, pH Adjustments and Dosing

Three coagulants were used in this work, namely, aluminium sulphate, iron (III) sulphate and zirconium salt. The aluminium coagulant had 27% aluminium, iron coagulant had 43% iron and the zirconium coagulant was prepared to have 28% zirconium. In each of the jar tests, all six beakers were filled with a particular wastewater with a particular initial pH of either 6.0 or 7.5. Since the measured pH values of the wastewaters were always higher than either 6.0 or 7.5, there was a need to reduce the pH before starting the dosing. This was achieved by gradually adding small amounts of hydrochloric acid into each jar containing one litre of wastewater using a syringe and measuring the pH changes continuously using the pH meter while using the mixer to stir the wastewater. Once all the wastewaters in the six beakers were at the same pH value of either 6.0 or 7.5, the mixers were stopped and the volume was readjusted to one litre of wastewater in each beaker.

Next, six different doses in regularly increasing order, of a particular coagulant was added to each beaker using a micropipette and the rapid mixing was started immediately. pH of the wastewater in each beaker was measured at the start of slow mixing. After 10 minutes of sedimentation, a syringe was used to draw out samples of the clarified wastewater from each beaker for further analysis, namely for turbidity measurements, total suspended solids measurements and orthophosphate measurements. The entire process was repeated with new dosage values if the optimum coagulant dosage was not determined. Once the optimum dosage was found, it was repeated to ensure that it was fairly reproducible.

2.4: Measurement of Turbidity

The raw wastewater was poured into the tube designed to work with the turbidity meter. After ensuring the tube wall is clean and no air bubbles are present within the filled tube, it was placed in the turbidity meter and the value was read off directly with the units of Formazin

Nephelometric Units (FNU). This was repeated for all the clarified wastewater samples. Thus the difference in turbidity before and after coagulation can be found.

2.5: Measurement of Total Suspended Solids

First, the weight of a new, dry glass fibre filter was measured. A vacuum filtration unit was prepared and the filter placed in position. It was then switched on, and 5 ml of the raw wastewater was filtered. After filtration, the filter was removed and placed in an aluminium tray for drying in the oven for 2 hours at around 100 ^{o}C. After 2 hours of drying, the filter was weighed again. The difference in weight would be the weight of the total suspended solids in 5 ml of the raw wastewater. This can be easily converted to the units of mg/l. This was repeated for all the clarified wastewater samples. Thus the difference in total suspended solids before and after coagulation can be found.

2.6: Measurement of Orthophosphates

The ascorbic acid method is used in measuring the orthophosphate concentration. Orthophosphate in the raw wastewater reacts with ammonium molybdate and potassium antimonyl tartrate in the presence of ascorbic acid to form a blue solution. The intensity of the blue colour is directly proportional to the orthophosphate concentration. Ascorbic acid was prepared by dissolving 60 g in 1 litre of water. Ammonium molybdate was prepared by dissolving 8 g in 1 litre of water and 0.2 g of potassium antimonyl tartrate was added to this to form an ammonium molybdate – potassium antimonyl tartrate mixture (United States Environmental Protection Agency, 2014). First, a calibration curve is prepared using known concentrations of orthophosphates. 10 ml of a sample of known concentration of orthophosphates was diluted to 50 ml. 4 ml of the ammonium molybdate – potassium antimonyl tartrate mixture was added and 2 ml of ascorbic acid was added. After mixing well and waiting for 10 minutes, the blue colour was slowly formed and reached its peak. Using a spectrophotometer, absorbance of the sample was measured at a wavelength of 880 nm for 10 minutes. This is repeated for other known concentrations in order to prepare the calibration curve, and is then repeated for the raw wastewater and all the clarified wastewater samples. The absorbance of raw wastewater and clarified wastewater samples without using the ascorbic acid method was also measured as a blank to subtract any background absorbance due to the natural colour of the wastewater. Thus the difference in orthophosphate concentration before and after coagulation can be found.

3: RESULTS AND DISCUSSION

An investigation on the coagulation performances of aluminium, iron and zirconium coagulants was carried out at the water laboratories of the Department of Mathematical Sciences and Technology at the Norwegian University of Life Sciences on certain days over a period of about four months from January 2014 to May 2014. The results obtained from this study are presented and discussed in this chapter.

Jar tests are certainly a sort of trial and error method in identifying the optimum coagulation dosage for a particular wastewater at a particular pH. If the first set of jar tests cannot identify the optimum pH, then the jar tests need to be repeated with a slight shift in dosage values. Nevertheless, it is the most widely accepted method used throughout the world for studying coagulation on the bench-scale. This is important information for this study as jar tests were the central method used in this work.

3.1: Result of the Calibration Curve Preparation

Table 3 below shows the data obtained from measurements of absorbance at 880 nm of known concentrations of orthophosphates. Figure 4 below shows the calibration curve with a linear trend line that sufficiently agrees that the absorbance is directly proportional to the orthophosphate concentration.

Table 3: Data for Calibration Curve

Orthophosphate Concentration (mg P/l)	Absorbance at 880 nm
0.25	0.15
0.50	0.18
0.75	0.20
1.00	0.25
2.00	0.40
4.00	0.80

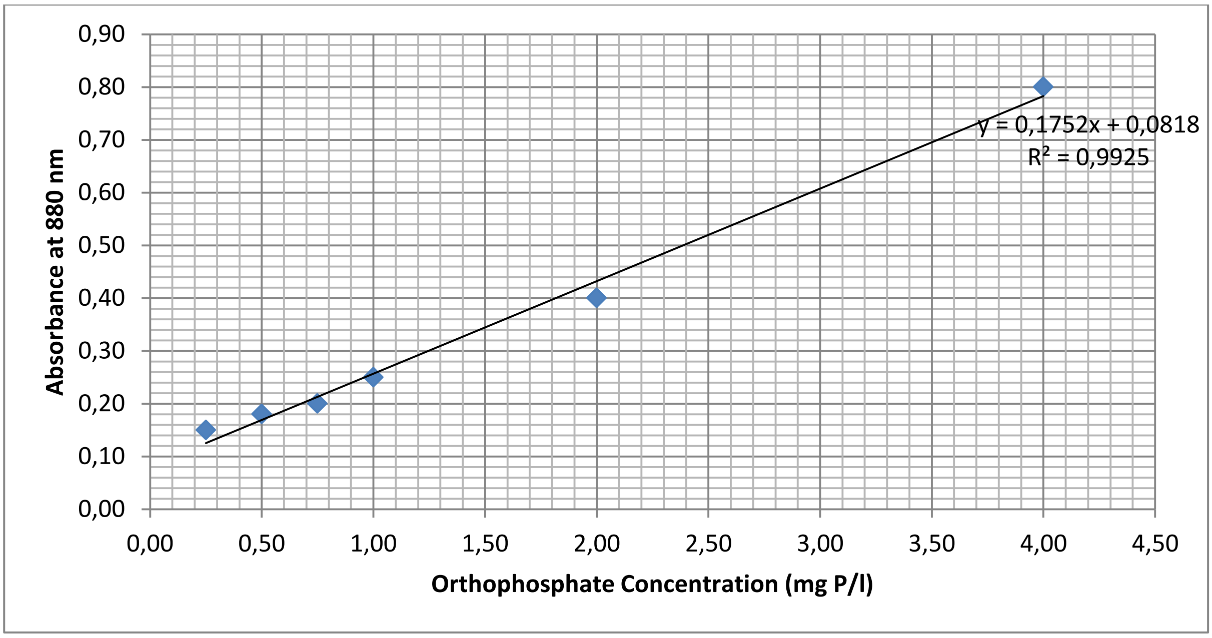

Figure 4: Calibration Curve

3.2: Results of 100% Blackwater at pH 6.0

Table 4: Data for Aluminium Coagulant

Aluminium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.95	2.28	2.60	2.93	3.25	3.58
pH	6.0	6.5	6.5	6.4	6.3	7.0	6.6
Turbidity (FNU)	1320	982	1116	812	930	598	706
Total Suspended Solids (mg/l)	2437	1940	2040	1940	1740	1600	1600
Total Orthophosphates (mg P/l)	11.1	5.3	5.1	5.2	4.3	4.4	4.5

Table 5: Data for Iron Coagulant

Iron Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.25	1.50	1.75	2.00	2.25	2.50
pH	6.0	8.9	8.7	9.0	9.2	9.1	9.3
Turbidity (FNU)	1320	1238	1234	930	1054	722	850
Total Suspended Solids (mg/l)	2437	2230	2333	2244	2015	1844	1866
Total Orthophosphates (mg P/l)	11.1	5.9	6.1	6.0	5.0	5.2	5.2

Table 6: Data for Zirconium Coagulant

Zirconium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		0.60	0.70	0.80	0.90	1.00	1.10
pH	6.0	6.2	6.1	6.2	6.1	6.5	6.3
Turbidity (FNU)	1320	860	982	760	840	488	604
Total Suspended Solids (mg/l)	2437	1654	1860	1754	1675	1456	1433
Total Orthophosphates (mg P/l)	11.1	4.8	4.9	5.0	4.0	4.0	4.2

3.3: Results of 100% Blackwater at pH 7.5

Table 7: Data for Aluminium Coagulant

Aluminium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.95	2.28	2.60	2.93	3.25	3.58
pH	7.5	8.0	8.0	7.9	7.9	8.5	8.1
Turbidity (FNU)	1300	890	940	802	890	550	680
Total Suspended Solids (mg/l)	2398	1850	1950	1888	1648	1457	1563
Total Orthophosphates (mg P/l)	10.9	5.0	4.8	5.0	4.1	4.1	4.2

Table 8: Data for Iron Coagulant

Iron Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.25	1.50	1.75	2.00	2.25	2.50
pH	7.5	9.0	8.8	8.9	9.1	9.1	9.1
Turbidity (FNU)	1300	1222	1244	920	1044	732	860
Total Suspended Solids (mg/l)	2398	2134	2256	2156	1980	1765	1799
Total Orthophosphates (mg P/l)	10.9	5.8	6.0	6.1	4.9	5.1	5.3

Table 9: Data for Zirconium Coagulant

Zirconium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		0.60	0.70	0.80	0.90	1.00	1.10
pH	7.5	7.7	7.8	7.7	7.9	7.8	7.9
Turbidity (FNU)	1300	880	1000	820	860	500	650
Total Suspended Solids (mg/l)	2398	1754	1789	1675	1476	1189	1257
Total Orthophosphates (mg P/l)	10.9	4.5	4.5	4.4	3.7	3.6	3.8

3.4: Results of 50% Blackwater and 50% Greywater at pH 6.0

Table 10: Data for Aluminium Coagulant

Aluminium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.95	2.28	2.60	2.93	3.25	3.58
pH	6.0	6.4	6.5	6.5	6.4	6.9	6.5
Turbidity (FNU)	920	560	760	412	414	230	330
Total Suspended Solids (mg/l)	1780	1459	1567	1456	1348	1135	1187
Total Orthophosphates (mg P/l)	10.1	5.0	4.9	5.0	4.0	4.1	4.3

Table 11: Data for Iron Coagulant

Iron Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.25	1.50	1.75	2.00	2.25	2.50
pH	6.0	8.7	8.8	8.9	9.0	9.1	9.1
Turbidity (FNU)	920	600	820	500	490	280	374
Total Suspended Solids (mg/l)	1780	1560	1610	1568	1478	1234	1302
Total Orthophosphates (mg P/l)	10.1	5.5	5.4	5.5	4.6	4.4	4.9

Table 12: Data for Zirconium Coagulant

Zirconium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		0.60	0.70	0.80	0.90	1.00	1.10
pH	6.0	6.1	6.2	6.2	6.3	6.3	6.4
Turbidity (FNU)	920	450	650	324	354	198	274
Total Suspended Solids (mg/l)	1780	1325	1437	1329	1129	996	1023
Total Orthophosphates (mg P/l)	10.1	4.4	4.6	4.6	3.5	3.4	3.8

3.5: Results of 50% Blackwater and 50% Greywater at pH 7.5

Table 13: Data for Aluminium Coagulant

Aluminium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.95	2.28	2.60	2.93	3.25	3.58
pH	7.5	8.0	7.9	8.1	7.8	8.3	8.2
Turbidity (FNU)	940	530	560	412	450	190	240
Total Suspended Solids (mg/l)	1830	1765	1568	1453	1265	1023	1123
Total Orthophosphates (mg P/l)	10.2	4.0	3.7	4.1	3.3	3.1	3.3

Table 14: Data for Iron Coagulant

Iron Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.25	1.50	1.75	2.00	2.25	2.50
pH	7.5	9.1	8.9	8.8	9.0	8.9	8.9
Turbidity (FNU)	940	850	864	810	850	650	726
Total Suspended Solids (mg/l)	1830	1801	1689	1674	1649	1479	1502
Total Orthophosphates (mg P/l)	10.2	5.1	5.3	5.4	4.2	4.2	5.3

Table 15: Data for Zirconium Coagulant

Zirconium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		0.60	0.70	0.80	0.90	1.00	1.10
pH	7.5	7.8	7.8	7.9	7.8	7.7	7.7
Turbidity (FNU)	940	430	420	350	320	130	190
Total Suspended Solids (mg/l)	1830	1689	1387	1298	1123	890	1089
Total Orthophosphates (mg P/l)	10.2	3.6	3.4	3.5	3.2	2.9	3.0

3.6: Results of 100% Greywater at pH 6.0

Table 16: Data for Aluminium Coagulant

Aluminium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.95	2.28	2.60	2.93	3.25	3.58
pH	6.0	6.3	6.4	6.3	6.2	6.6	6.3
Turbidity (FNU)	630	290	410	198	180	190	210
Total Suspended Solids (mg/l)	1675	1328	1438	1321	1236	1012	1045
Total Orthophosphates (mg P/l)	9.2	4.0	3.9	4.1	3.1	3.1	3.4

Table 17: Data for Iron Coagulant

Iron Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.25	1.50	1.75	2.00	2.25	2.50
pH	6.0	8.7	8.7	8.8	8.9	9.0	8.9
Turbidity (FNU)	630	350	540	302	290	280	304
Total Suspended Solids (mg/l)	1675	1456	1540	1458	1345	1199	1276
Total Orthophosphates (mg P/l)	9.2	4.5	4.4	4.6	3.7	3.6	3.9

Table 18: Data for Zirconium Coagulant

Zirconium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		0.60	0.70	0.80	0.90	1.00	1.10
pH	6.0	6.2	6.3	6.3	6.2	6.4	6.3
Turbidity (FNU)	630	230	350	130	142	142	156
Total Suspended Solids (mg/l)	1675	1130	1278	1123	1034	820	845
Total Orthophosphates (mg P/l)	9.2	3.4	3.2	3.5	2.6	2.5	3.0

3.7: Results of 100% Greywater at pH 7.5

Table 19: Data for Aluminium Coagulant

Aluminium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.95	2.28	2.60	2.93	3.25	3.58
pH	7.5	7.9	7.8	8.2	7.9	8.2	8.1
Turbidity (FNU)	620	230	190	120	112	98	120
Total Suspended Solids (mg/l)	1689	1566	1345	1233	1003	850	960
Total Orthophosphates (mg P/l)	9.3	3.4	3.1	3.4	2.8	2.7	3.2

Table 20: Data for Iron Coagulant

Iron Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		1.25	1.50	1.75	2.00	2.25	2.50
pH	7.5	9.0	9.0	8.9	9.1	9.0	8.8
Turbidity (FNU)	620	550	560	530	540	420	590
Total Suspended Solids (mg/l)	1689	1603	1450	1458	1430	1298	1378
Total Orthophosphates (mg P/l)	9.3	4.2	4.3	4.4	3.3	3.2	4.1

Table 21: Data for Zirconium Coagulant

Zirconium Coagulant	Raw	1	2	3	4	5	6
Dose (mmol/l)		0.60	0.70	0.80	0.90	1.00	1.10
pH	7.5	7.8	7.9	8.0	8.0	7.7	7.8
Turbidity (FNU)	620	190	160	110	98	76	100
Total Suspended Solids (mg/l)	1689	1356	1259	1190	960	760	850
Total Orthophosphates (mg P/l)	9.3	2.5	2.6	2.4	2.3	1.9	2.1

3.8: Removal of Turbidity for 100% Blackwater at pH 6.0

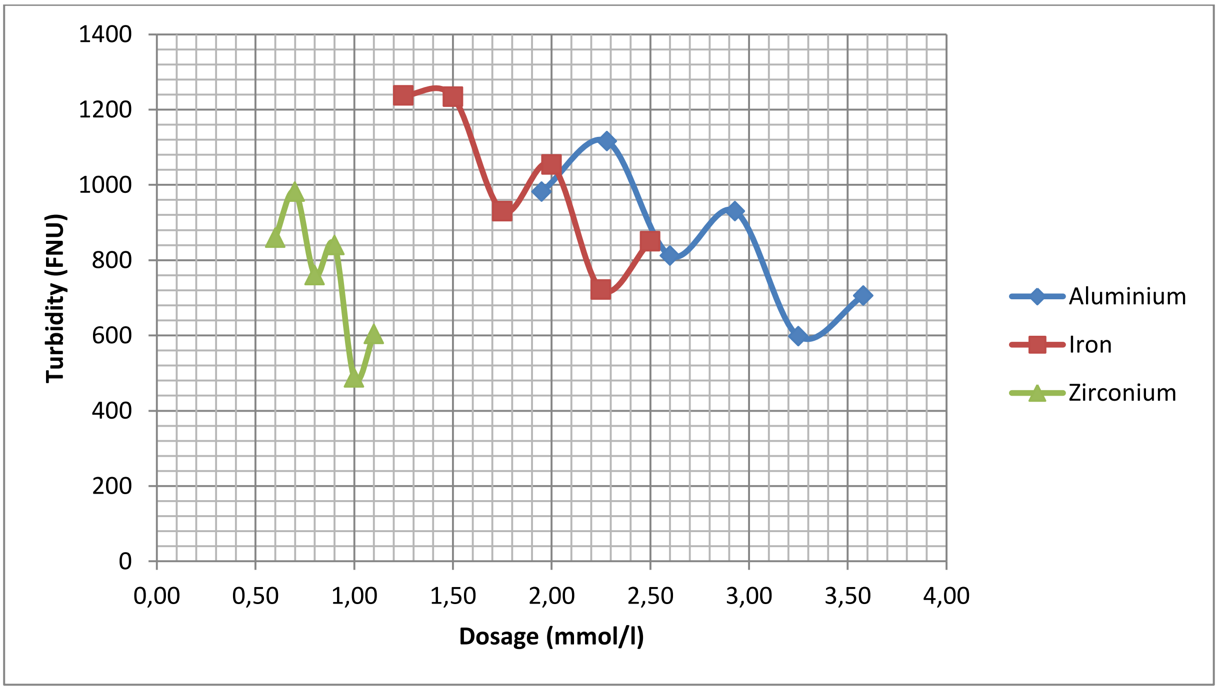

Figure 5: Graph of Turbidity for 100% Blackwater at pH 6.0

As can be seen in Figure 5 above, removal of turbidity is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher.

3.9: Removal of Turbidity for 100% Blackwater at pH 7.5

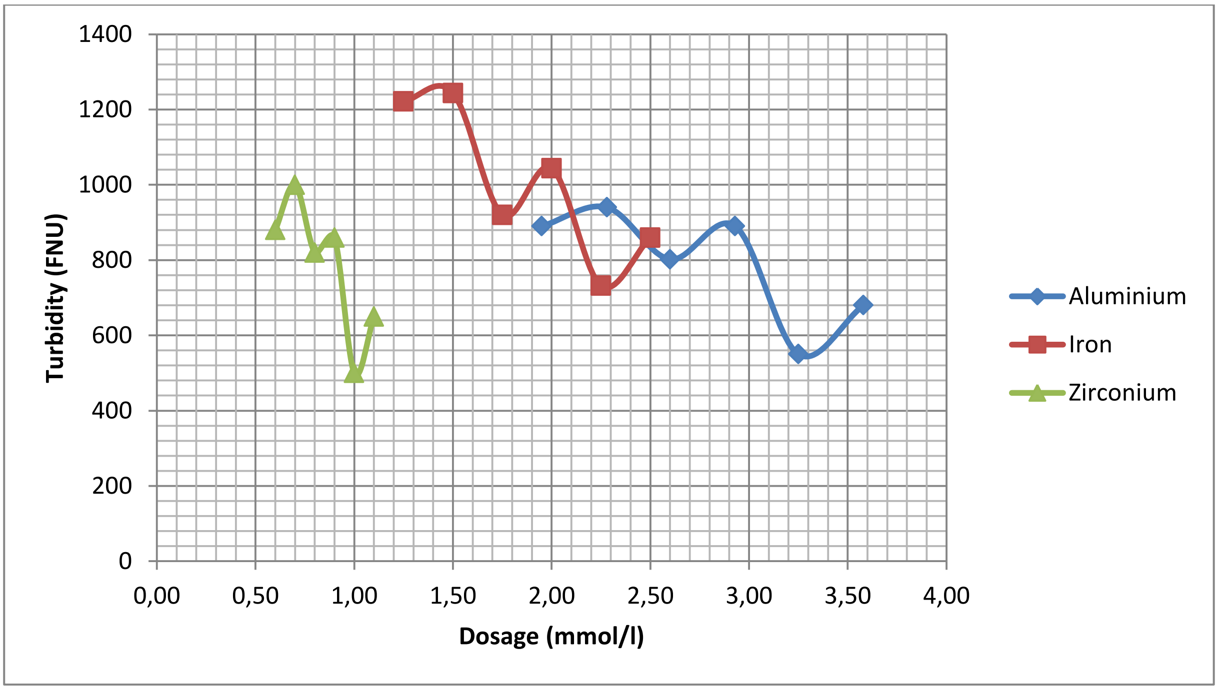

Figure 6: Graph of Turbidity for 100% Blackwater at pH 7.5

As can be seen in Figure 6 above, removal of turbidity is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher. As removal efficiencies are concerned, it is similar to Figure 5, when the water was at pH 6.0.

3.10: Removal of Turbidity for 50% Blackwater and 50% Greywater at pH 6.0

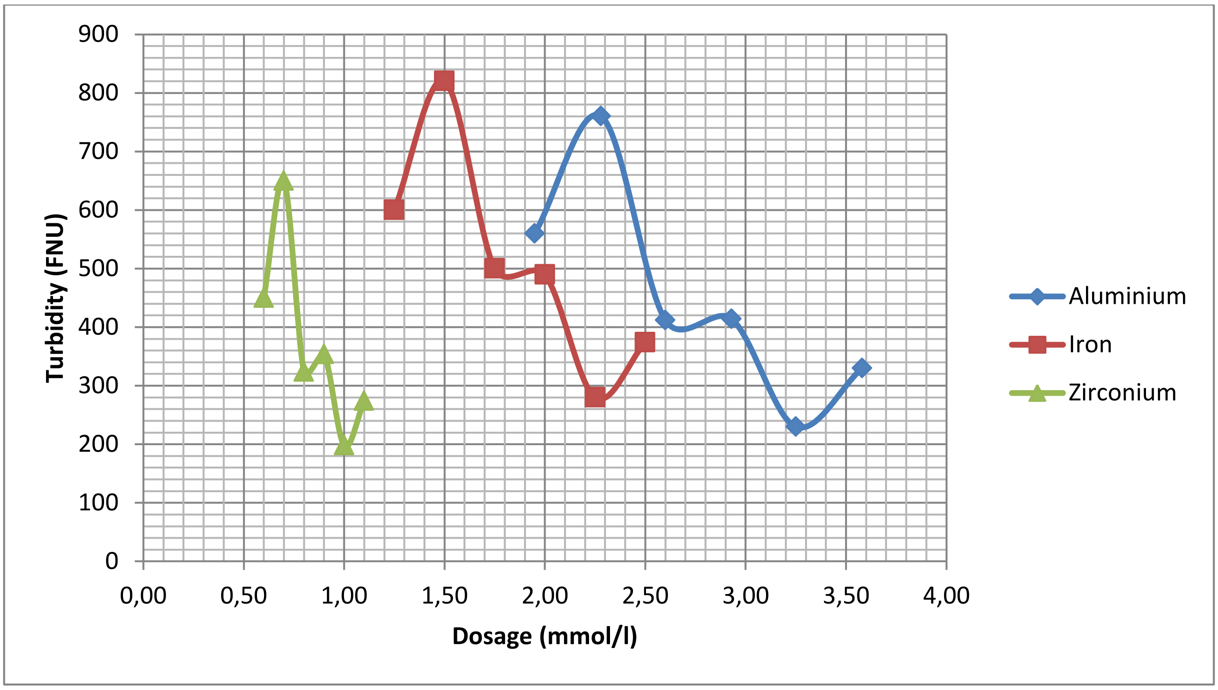

Figure 7: Graph of Turbidity for 50% Blackwater and 50% Greywater at pH 6.0

As can be seen in Figure 7 above, again removal of turbidity is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. The turbidity removal efficiencies are better than that for 100% blackwater at both pH 6.0 and 7.5.

3.11: Removal of Turbidity for 50% Blackwater and 50% Greywater at pH 7.5

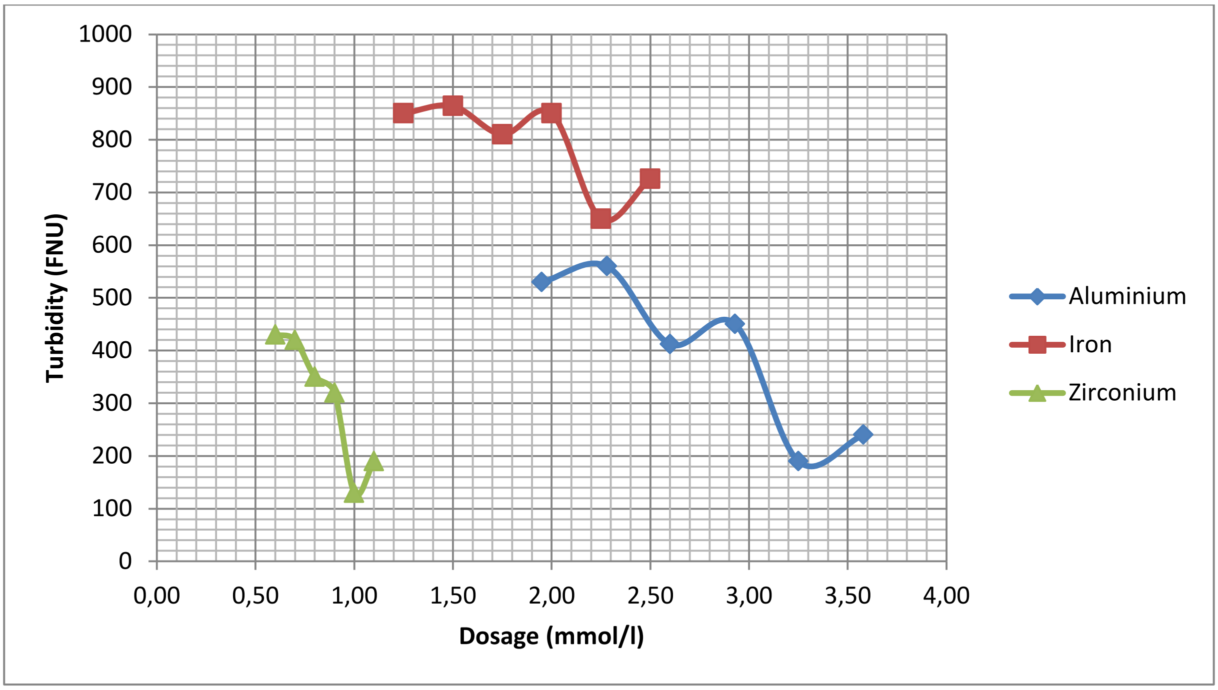

Figure 8: Graph of Turbidity for 50% Blackwater and 50% Greywater at pH 7.5

As can be seen in Figure 8 above, removal of turbidity is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. The turbidity removal efficiencies are better than that for 100% blackwater at both pH 6.0 and 7.5. The efficiencies are also slightly better at this pH as compared to pH 6.0 for the same mixture ratio. However, iron performed poorly in this test.

3.12: Removal of Turbidity for 100% Greywater at pH 6.0

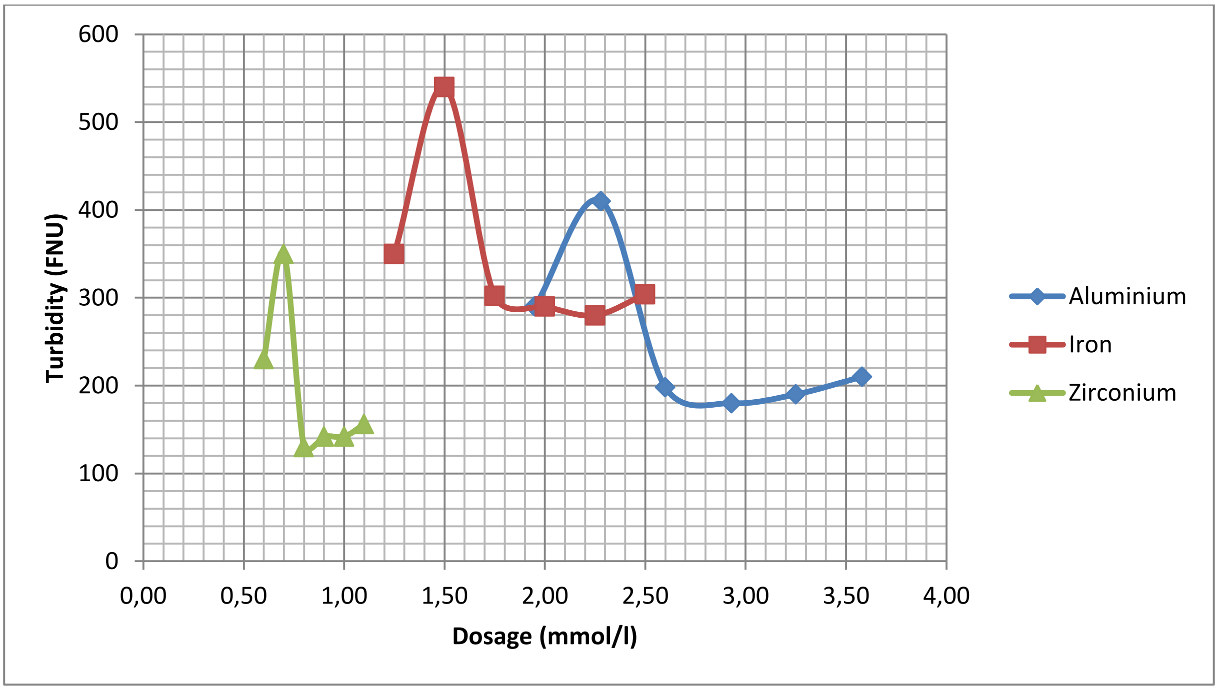

Figure 9: Graph of Turbidity for 100% Greywater at pH 6.0

As can be seen in Figure 9 above, removal of turbidity is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher.

3.13: Removal of Turbidity for 100% Greywater at pH 7.5

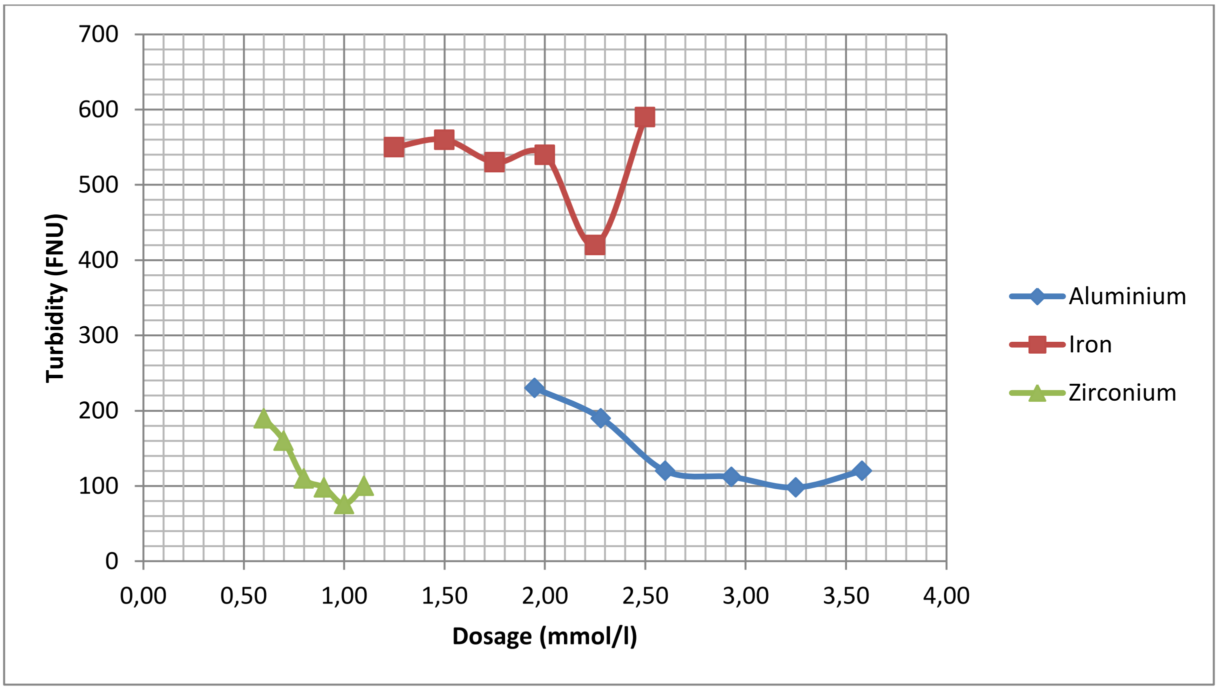

Figure 10: Graph of Turbidity for 100% Greywater at pH 7.5

As can be seen in Figure 10 above, removal of turbidity is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. The turbidity removal efficiencies are better than that for both 100% blackwater and 50% Blackwater-50% Greywater mixture. However, the removal efficiency of iron coagulant is poor as compared to the other two.

3.14: Removal of Total Suspended Solids for 100% Blackwater at pH 6.0

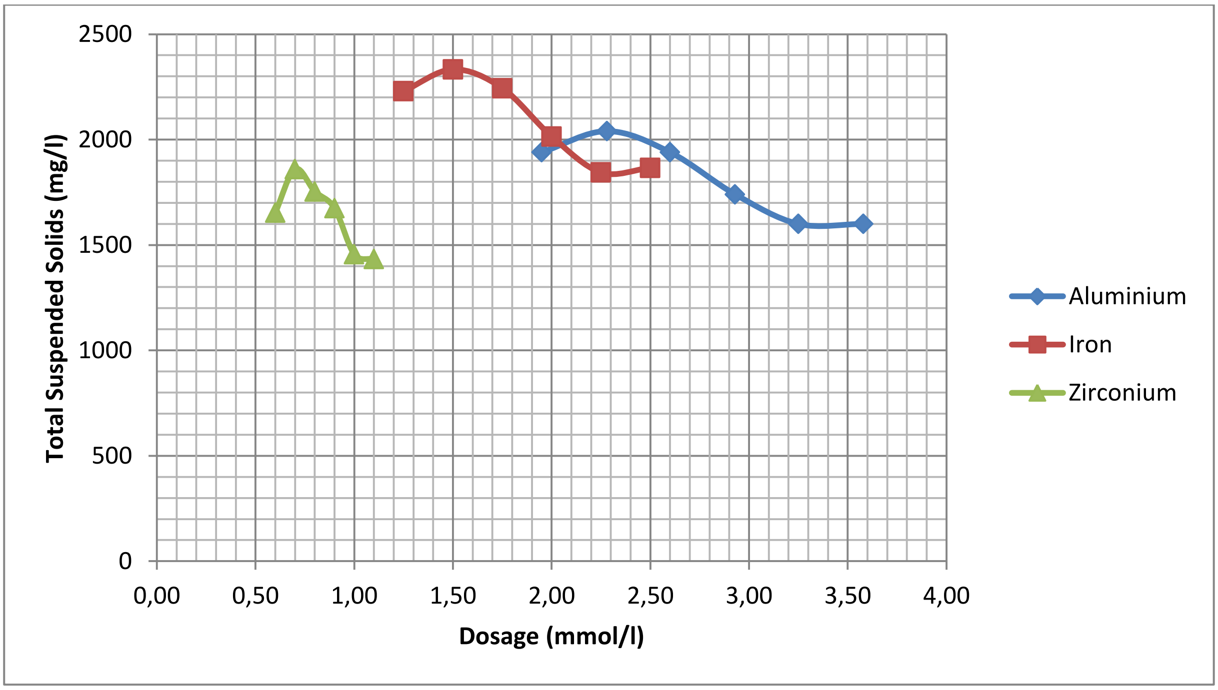

Figure 11: Graph of Total Suspended Solids for 100% Blackwater at pH 6.0

As can be seen in Figure 11 above, removal of total suspended solids is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher.

3.15: Removal of Total Suspended Solids for 100% Blackwater at pH 7.5

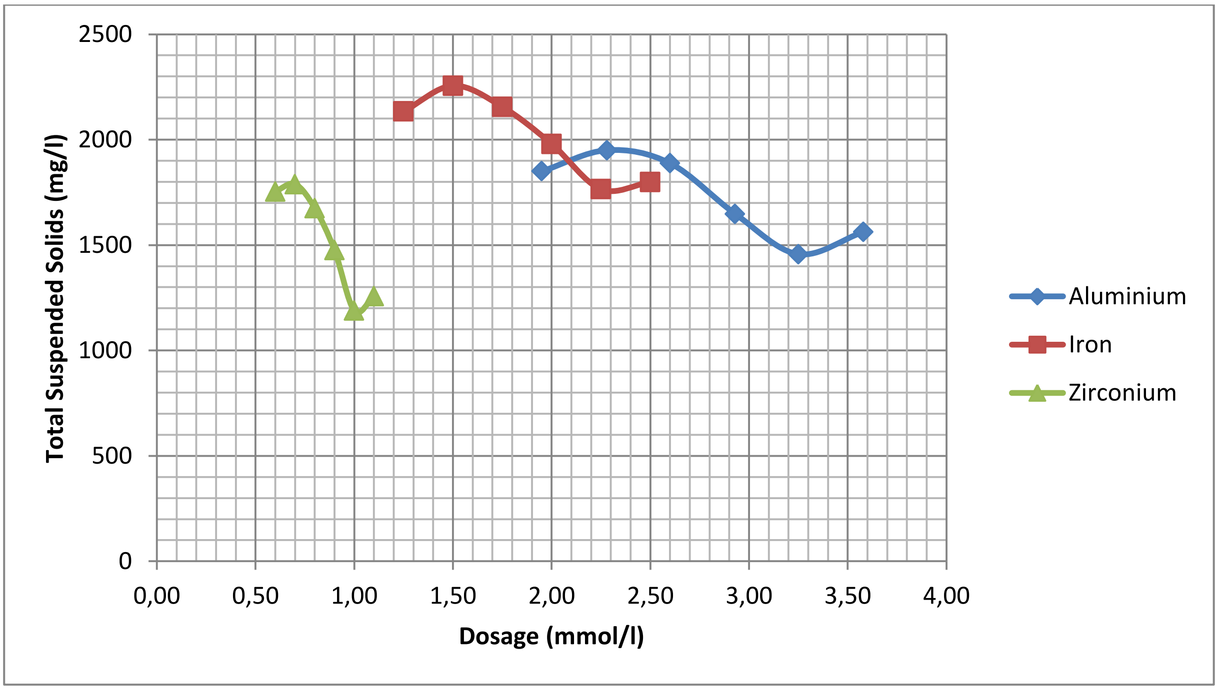

Figure 12: Graph of Total Suspended Solids for 100% Blackwater at pH 7.5

As can be seen in Figure 12 above, removal of total suspended solids is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher. Figure 12 is similar to Figure 11, which means that the removal efficiencies are somewhat similar for both pH values of 6.0 and 7.5.

3.16: Removal of Total Suspended Solids for 50% Blackwater and 50% Greywater at pH 6.0

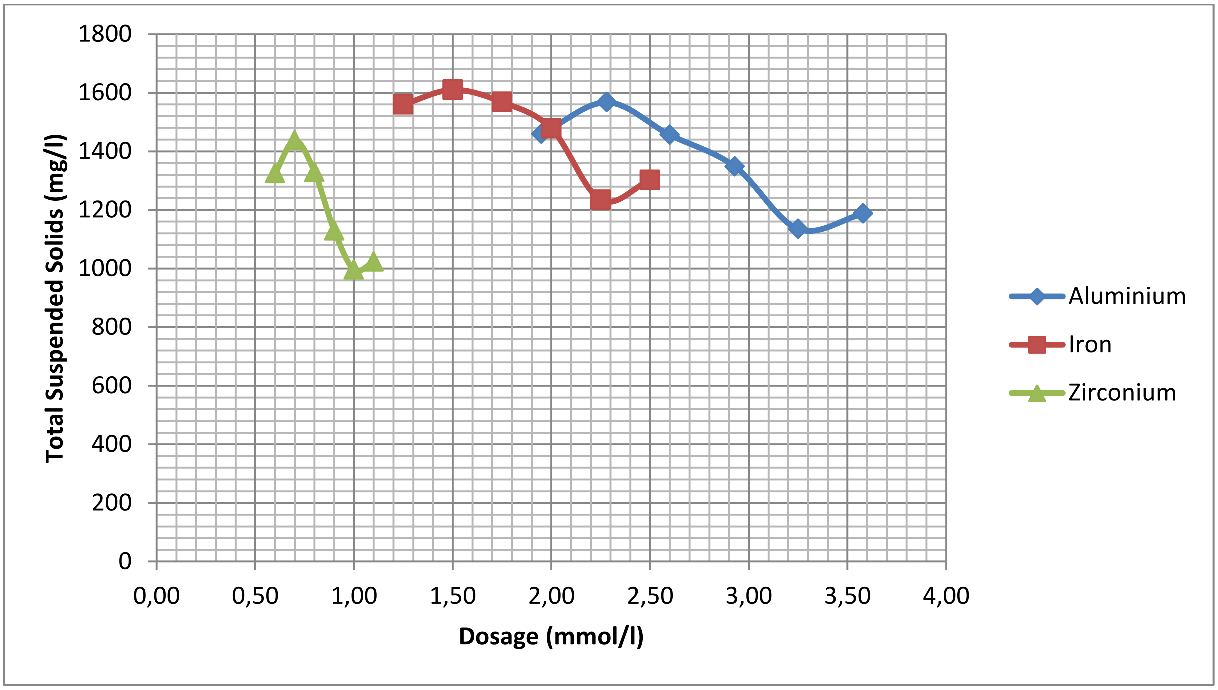

Figure 13: Graph of Total Suspended Solids for 50% Blackwater and 50% Greywater at pH 6.0

As can be seen in Figure 13 above, removal of total suspended solids is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. The removal efficiencies are slightly better here as compared to Figure 12.

3.17: Removal of Total Suspended Solids for 50% Blackwater and 50% Greywater at pH 7.5

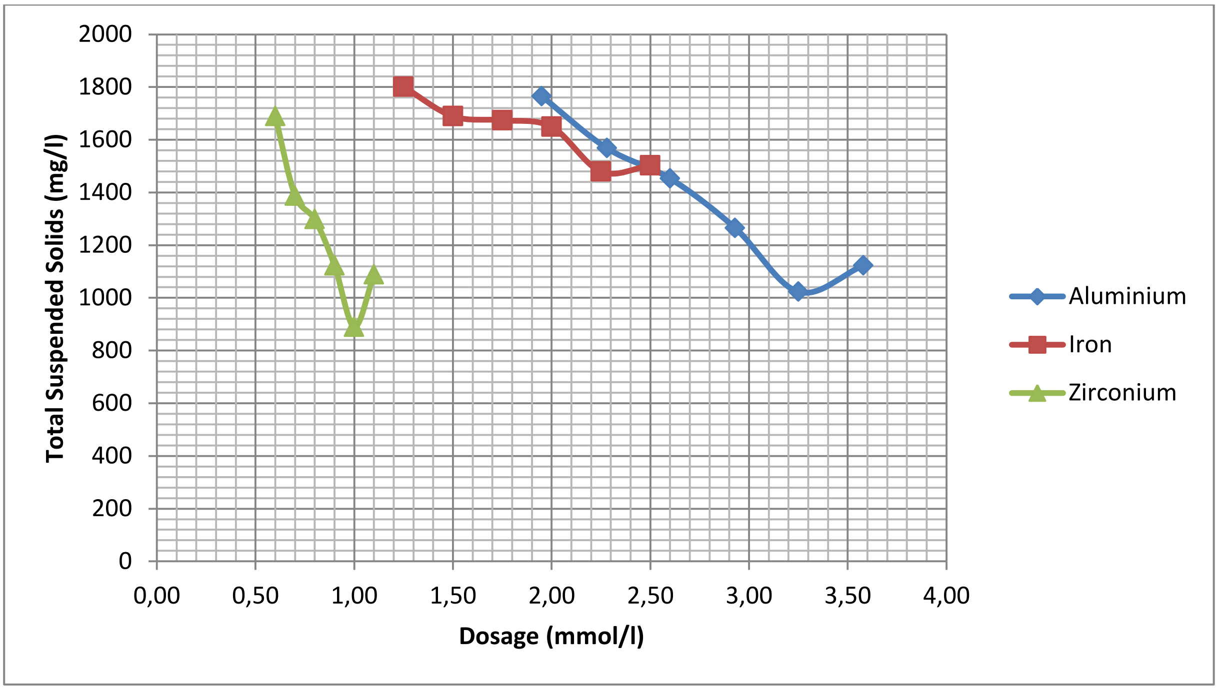

Figure 14: Graph of Total Suspended Solids for 50% Blackwater and 50% Greywater at pH 7.5

As can be seen in Figure 14 above, removal of total suspended solids is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher. The removal efficiencies are slightly better here as compared to Figure 13.

3.18: Removal of Total Suspended Solids for 100% Greywater at pH 6.0

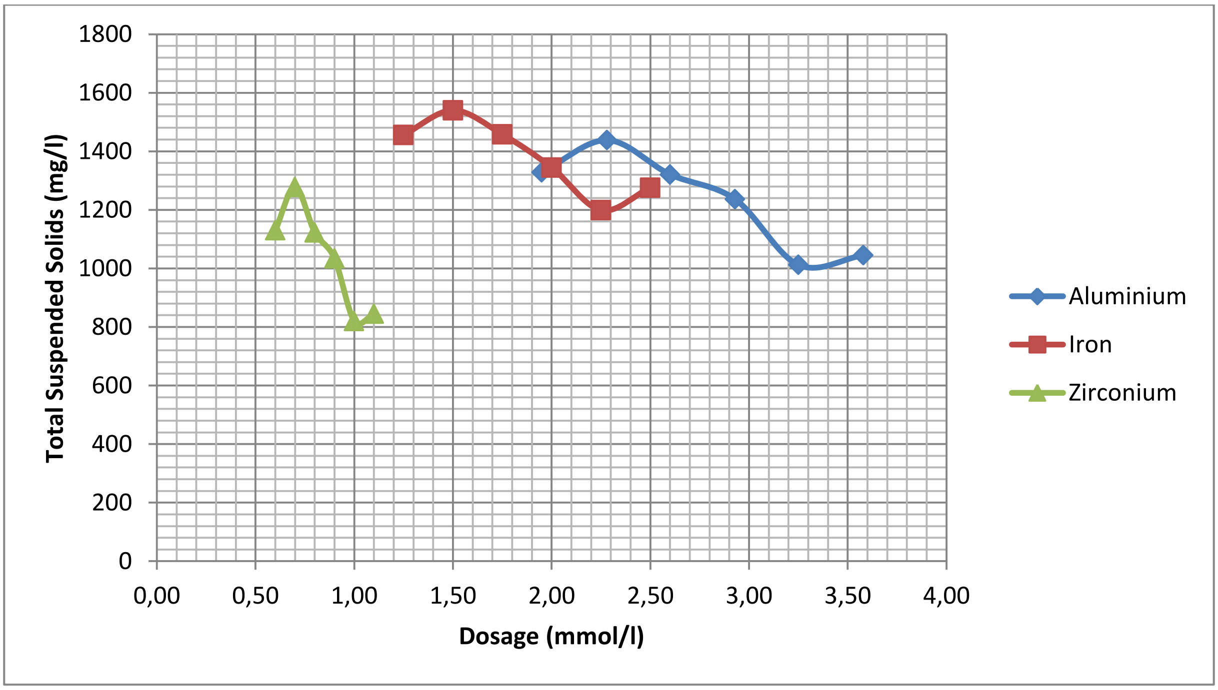

Figure 15: Graph of Total Suspended Solids for 100% Greywater at pH 6.0

As can be seen in Figure 15 above, removal of total suspended solids is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher.

3.19: Removal of Total Suspended Solids for 100% Greywater at pH 7.5

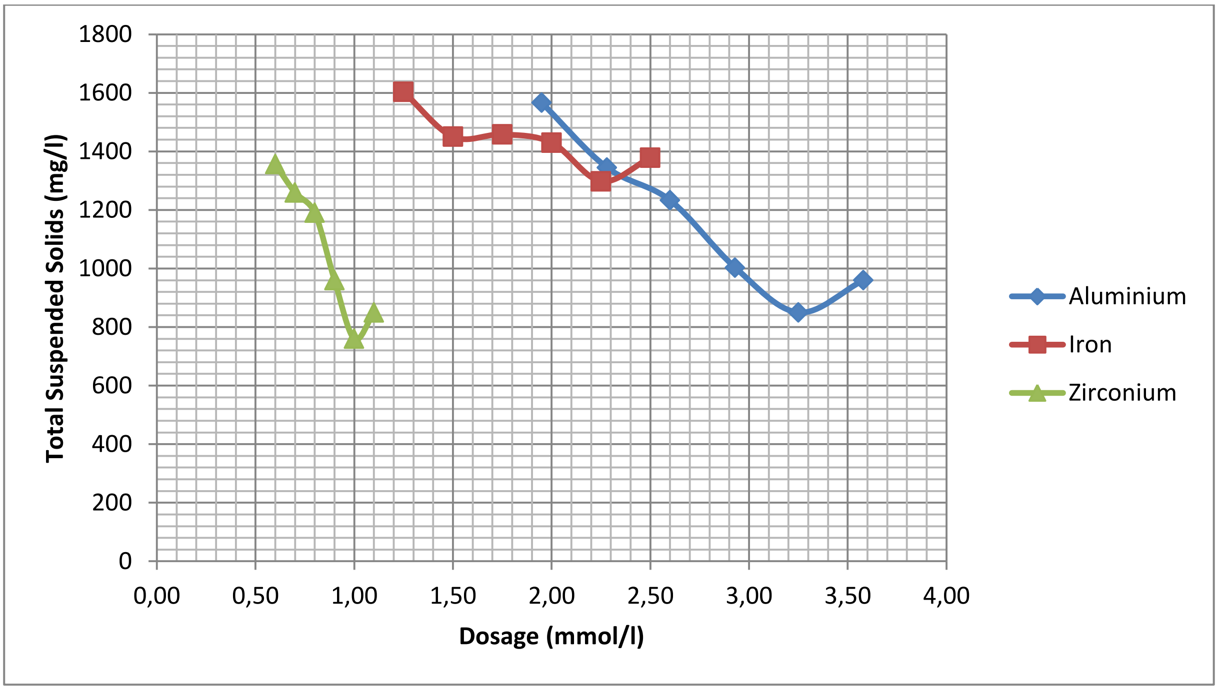

Figure 16: Graph of Total Suspended Solids for 100% Greywater at pH 7.5

As can be seen in Figure 16 above, removal of total suspended solids is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. The removal efficiencies are slightly better here as compared to that in Figure 15.

3.20: Removal of Orthophosphates for 100% Blackwater at pH 6.0

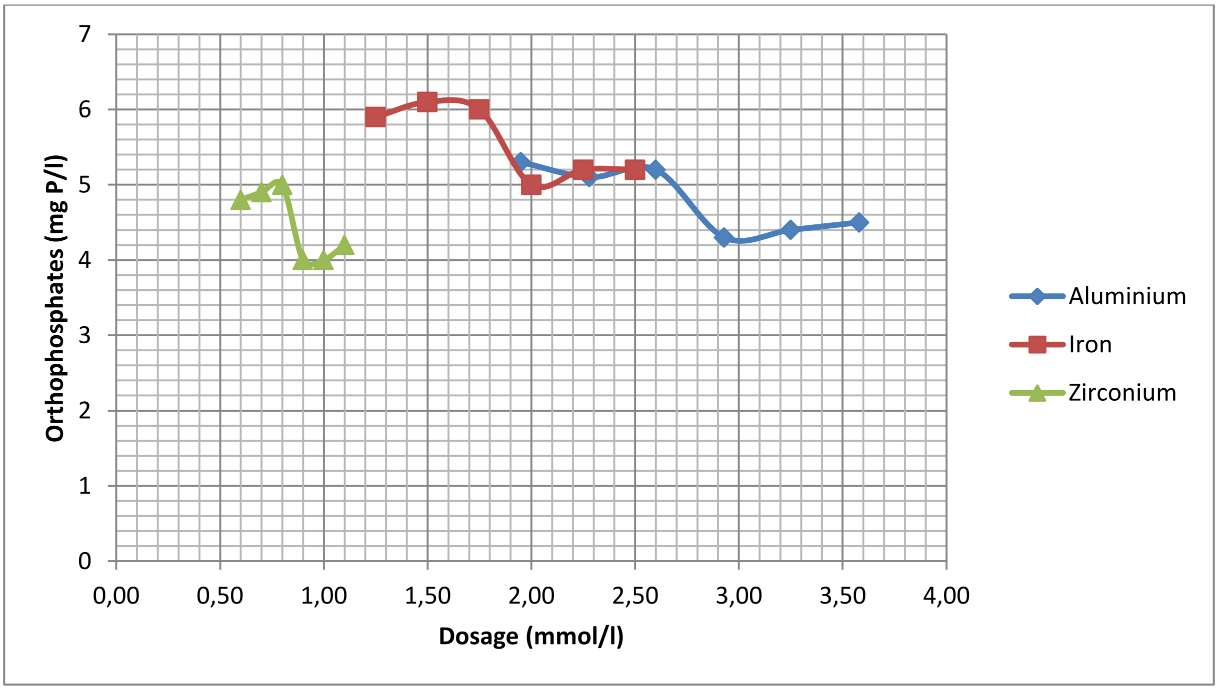

Figure 17: Graph of Orthophosphates for 100% Blackwater at pH 6.0

As can be seen in Figure 17 above, removal of orthophosphates is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher.

3.21: Removal of Orthophosphates for 100% Blackwater at pH 7.5

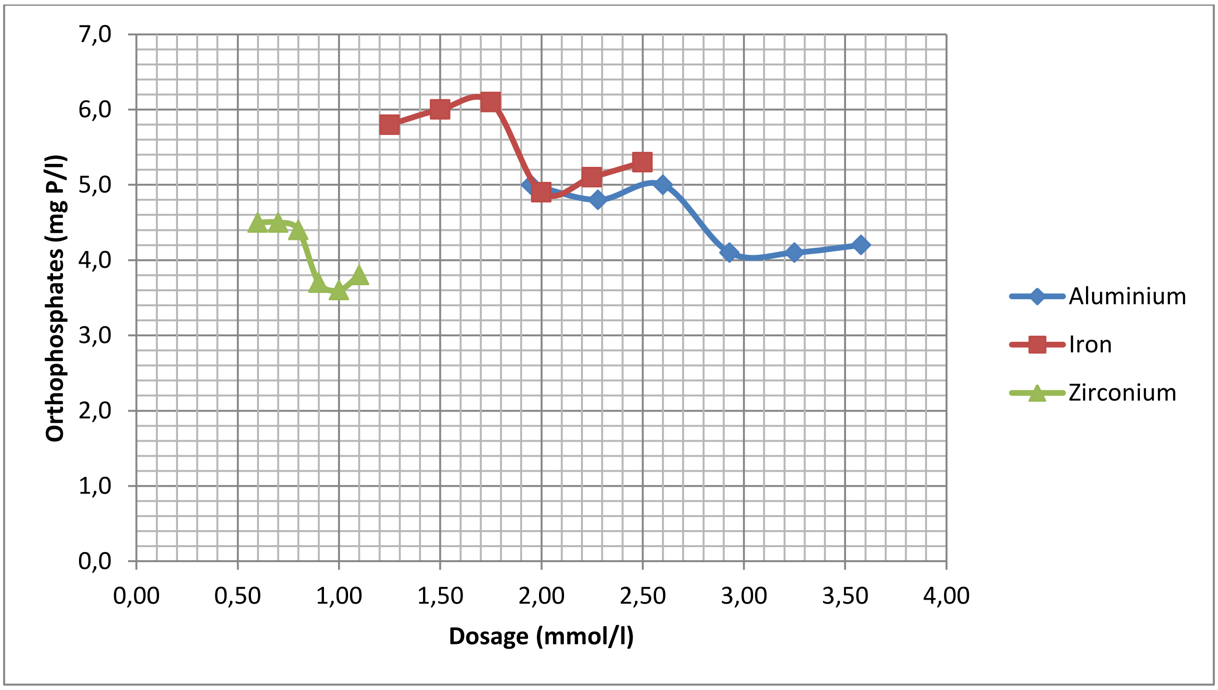

Figure 18: Graph of Orthophosphates for 100% Blackwater at pH 7.5

As can be seen in Figure 18 above, removal of orthophosphates is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher. The removal efficiencies are also slightly better here as compared to Figure 17.

3.22: Removal of Orthophosphates for 50% Blackwater and 50% Greywater at pH 6.0

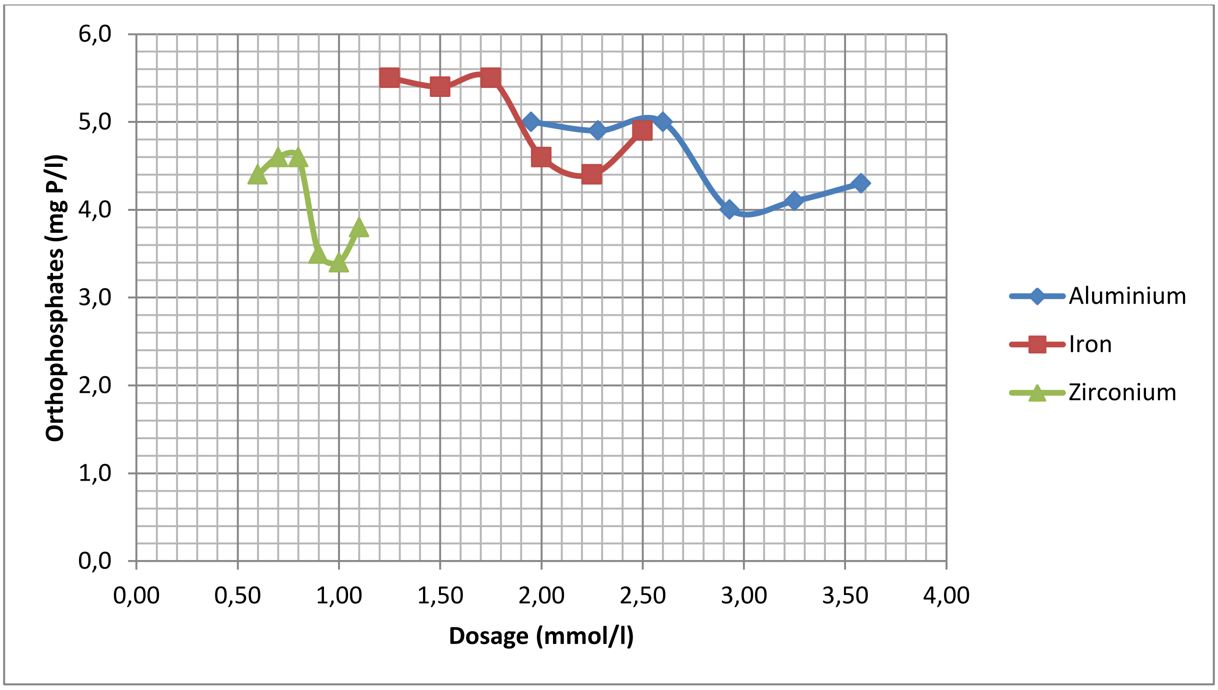

Figure 19: Graph of Orthophosphates for 50% Blackwater and 50% Greywater at pH 6.0

As can be seen in Figure 19 above, removal of orthophosphates is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher.

3.23: Removal of Orthophosphates for 50% Blackwater and 50% Greywater at pH 7.5

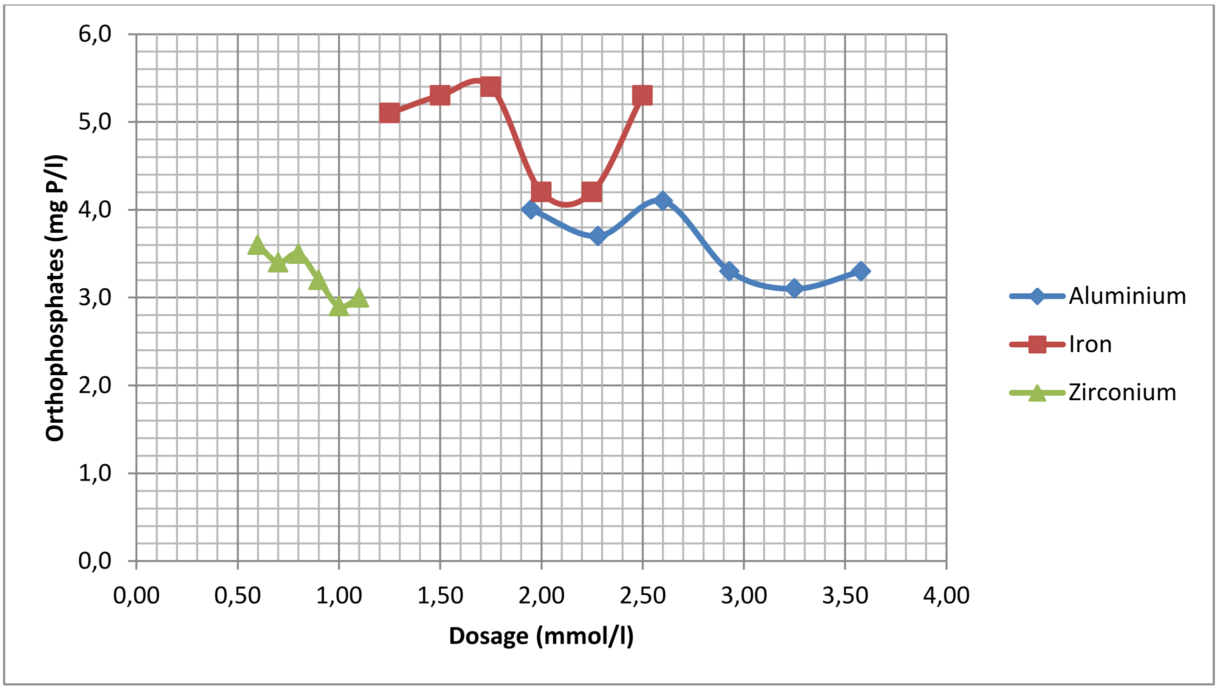

Figure 20: Graph of Orthophosphates for 50% Blackwater and 50% Greywater at pH 7.5

As can be seen in Figure 20 above, removal of orthophosphates is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. The treatment efficiencies in this case are also slightly better than that for 50% Blackwater and 50% Greywater at pH 6.0.

3.24: Removal of Orthophosphates for 100% Greywater at pH 6.0

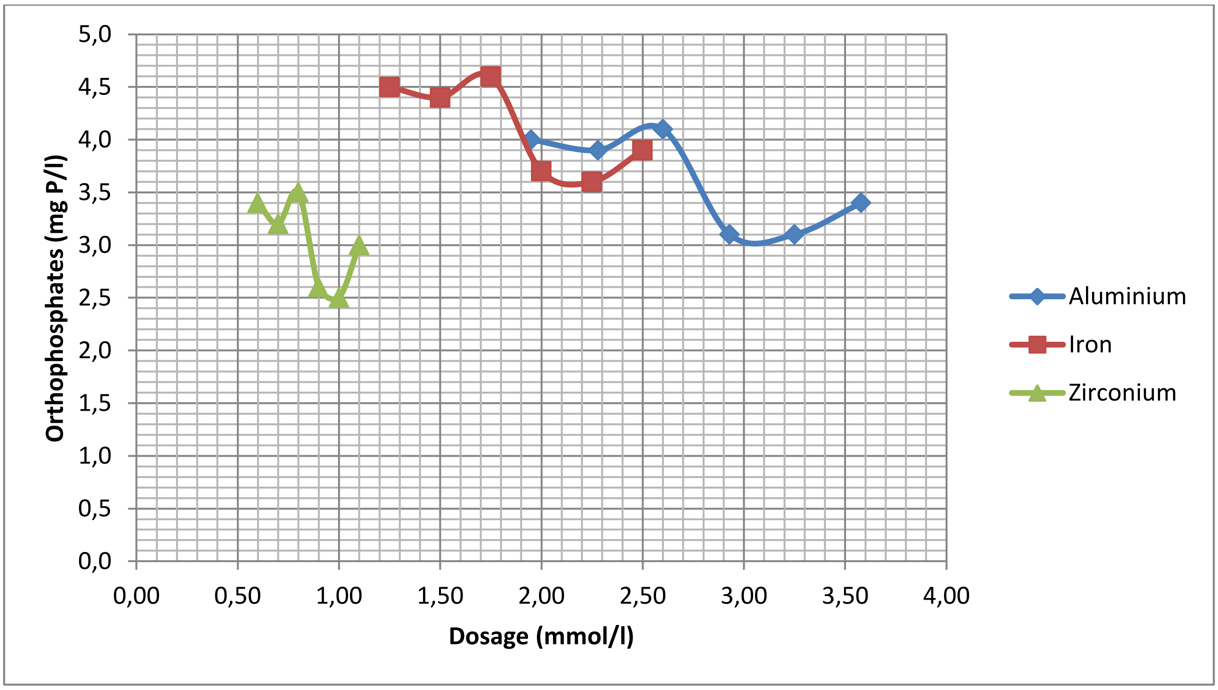

Figure 21: Graph of Orthophosphates for 100% Greywater at pH 6.0

As can be seen in Figure 21 above, removal of orthophosphates is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. Although aluminium performs better than iron in this case, the dosage needed, however is higher.

3.25: Removal of Orthophosphates for 100% Greywater at pH 7.5

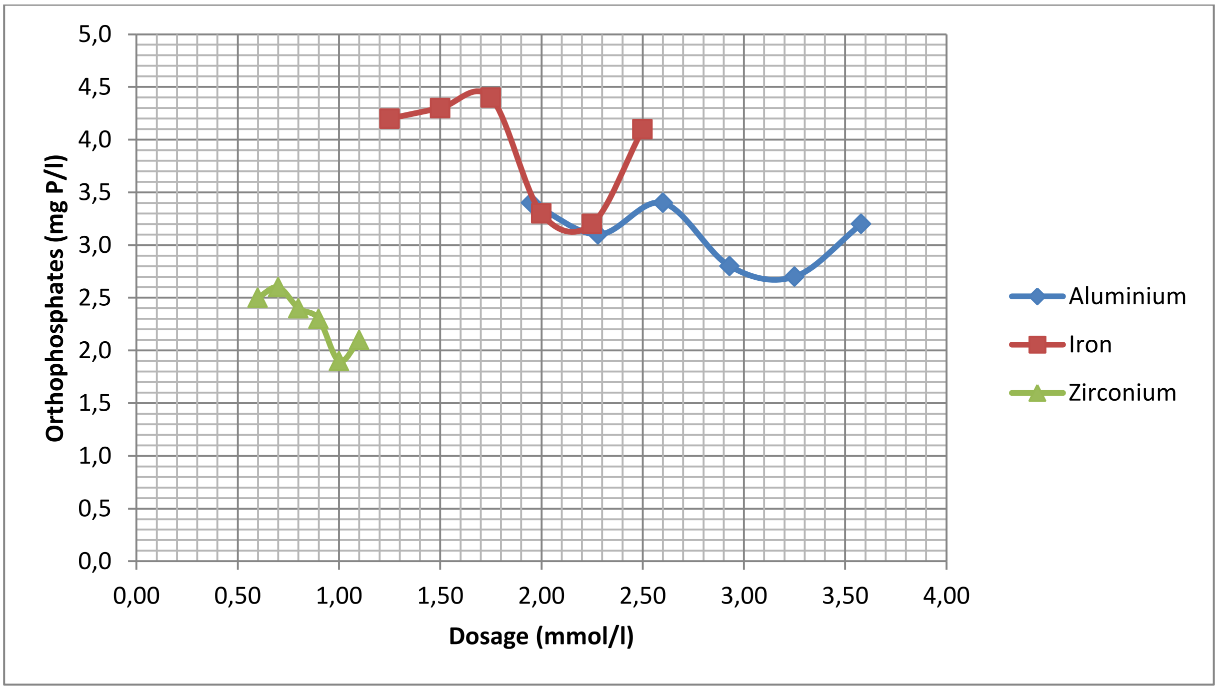

Figure 22: Graph of Orthophosphates for 100% Greywater at pH 7.5

As can be seen in Figure 22 above, removal of orthophosphates is best for zirconium, followed by aluminium and then by iron. Also dosage of zirconium is the lowest, followed by iron and then by aluminium. The treatment efficiencies in this case are better than that in Figure 21.

3.26: Discussion of Results of Present Work

The removal efficiencies in terms of turbidity removals for the aluminium coagulant ranged from 55% to 84%. For the iron coagulant, it ranged from 31% to 70%. For the zirconium coagulant, it ranged from 62% to 88%.

The removal efficiencies of turbidity in this work do not agree with the usual removal efficiencies expected from coagulation, which is at least 83% (Baghvand, Zand, Mehrdadi, & Karbassi, 2010).

The removal efficiencies in terms of total suspended solids removal for the aluminium coagulant ranged from 34% to 50%. For the iron coagulant, it ranged from 19% to 31%. For the zirconium coagulant, it ranged from 40% to 55%.

The removal efficiencies of total suspended solids in this work do not agree with the usual removal efficiencies expected from coagulation, which is at least 90% (Guida, Mattei, Rocca, Melluso, & Meric, 2006).

The removal efficiencies in terms of orthophosphates removal for the aluminium coagulant ranged from 59% to 71%. For the iron coagulant, it ranged from 53% to 66%. For the zirconium coagulant, it ranged from 64% to 80%.

The removal efficiencies of orthophosphates in this work do not agree with the usual removal efficiencies expected from coagulation, which is at least 90% (Ebeling, Ogden, Sibrell, & Rishel, 2004).

These deviations in results from expected values may be due to experimental errors. One possible error may be that clarified wastewater was collected for analysis after 10 minutes of sedimentation. A longer sedimentation time would have given better results. Another possible error may be due to the effective pH of iron, which is between 4.0 to 6.0 and 8.8 to 9.2, while the pH used in this study were 6.0 and 7.5 (Toprak, 2006). Iron performed the worst among all the three coagulants. Also the pH values were found to be increased when measured at the start of slow mixing. According to theory, pH has to decrease, because of the release of H^+ upon adding the coagulants. This deviation in pH readings may be due to errors in calibration of the pH meter.

Nevertheless, the main objective of this research work was to compare the coagulation performance of aluminium, iron and zirconium. As expected, zirconium performed better than the other two on all types of wastewaters analysed.

3.27: Implications of the Present Work and Suggestions for Future Work

From this work, it was clearly understood that zirconium coagulants work much better than traditional aluminium and iron coagulants, both by better treatment efficiencies and at smaller dosages. In all the jar test cases, it is evident that zirconium was the best amongst all three coagulants that were investigated. The investigations were indeed useful in assessing the potential usage of zirconium based coagulants in the near future as wastewater discharge requirements become stricter.

Going further, more research could be done at various pH values, as this study only focused on the pH values of 6.0 and 7.5. Furthermore, factors such as the effects of wastewater temperatures and changing the mixing conditions were not investigated in this work and are definitely points of interest for future research work, as they can affect the treatment efficiencies and dosage values.

4: CONCLUSION

The coagulation performances of aluminium, iron and zirconium coagulants were analysed on three different ratios of mixture of blackwater and greywater and at two pH values of 6.0 and 7.5. Blackwater and greywater were collected and mixed and used in jar tests to investigate the optimum dosage values of all three coagulants. Factors such as temperature of wastewater and mixing conditions were kept constant or nearly the same.

The results of this work have shown that the zirconium coagulant works generally better than both aluminium and iron coagulants, both by treatment efficiencies and by dosages.

The investigations were indeed useful in assessing the potential usage of zirconium based coagulants in the near future as wastewater discharge requirements become stricter.

Future research could focus on various pH values, factors such as the effects of wastewater temperatures and changing the mixing conditions.

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