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A Study on the effect of Zeta potential on coagulation process in water treatment

A thesis submitted to the Department of Physics in the
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in partial fulfillment of the requirements for the degree of Master of Science in Physics

By

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بِسْمِ اللَّهِ
الرَّحْمَٰنِ
الرَّحِيمِ

وَيَسْأَلُونَكَ عَن الرُّوح ۖ قُلِ الرُّوحُ مِنْ أَمْر رَبِّي وَمَا
أُوتِيتُم مِّنَ الْعِلْمِ إِلَّا قَلِيلًا ﴿٨٥﴾

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Maher Khleel
2013

This thesis is dedicated to
The soul of my father … Respect to
spring of tenderness. My mother…in recognition
To My wife and my children ..
My brothers and sisters…
And all those who helped me ……..

الخلاصة

تعتبر العكارة واحدة من أهم المشاكل التي تعاني منها مياه الشرب.ويتم التعامل عادة مع المياه التي تحتوي على جزيئات وعوالق وغرويات، من خلال عملية التخثير والتلبيد. والمخثرات الأكثر استخداما في عملية معالجة المياه هي كبريتات الألمنيوم (الشب) وكلوريد ألحديديك والبولي ألمنيوم التي اكتشف مؤخرا أن لها تأثيرات سلبية جانبية على صحة الإنسان إذا لم تراعى كميات النسب والأوزان المضافة إلى الماء وبمعدلات مسموح بها .

في هذا البحث تم دراسة إمكانية السيطرة على الدالة الفيزيائية جهد زيتا من خلال دراسة تأثيرها على عملية التلبيد للعوالق والغرويات الموجودة في المياه والمسببة للعكارة .وذلك للتقليل من استخدام المواد الكيماوية في عمليات معالجة المياه .

تتكون الدراسة من مرحلتين الأولى هما المقارنة بين كفاءة المخثرات الثلاثة لاختيار المخثر الأكفأ من بينها من حيث قدرته على إزالة عكارة المياه والوصول إلى المواصفات القياسية لمياه الشرب . والمرحلة الثانية تمثلت بمقارنة المخثر الذي تم اختياره في المرحلة الأولى مع المتسعات عالية الجهد (Zeta Rod) لدراسة تأثير هذه التقنية على عملية التلبيد من خلال تسليط جهد مستمر عالي(من ٥ كيلو فولت الى ٣٠ كيلو فولت) على عينات الماء العكر.

أجريت التجارب باستخدام مياه ذات عكارة مسيطر عليها من خلال إضافة الكائولين، وتم تقييم المخثرات باستخدام اختبار الجرة. وأجريت الاختبارات على ثلاث مستويات من العكارة ٥٨٠ و١٥٠و٥٠ (NTU). أظهرت النتائج تفوق البولي المنيوم كلورايد على الشب وعلى كلوريد الحديديك في إزالة العكارة، حيث تبين أن جرعة مقدارها ٧٥ ملغم /لترمن هذا المخثر قد استطاعت أن تخفض عكارة الماء من ٥٨٠ إلى ٤,٥ NTU أي بكفاءة إزالة مقدارها (٩٩,٢%) لتتوافق مواصفة المياه مع المواصفة العراقية لمياه الشرب. نفس الجرعة استطاعت أن تخفض العكارة من ١٥٠ إلى ١,١٤ NTU (٩٩,٢٤%) ومن ٥٠ إلى ١,٥١ NTU (٩٦,٩%) بنفس ظروف التشغيل والاختبار. في حين بلغت كفاءة إزالة العكارة لكل من الشب وكلوريد الحديديك لنفس الجرعة والظروف التشغيلية بمقدار (٩٨,٥%) و(٩٨,٨%) لمستوى عكارة ٥٨٠ NTU و(٩٥,٦%) و(٩٧,٤%) لمستوى عكارة ١٥٠ NTU و(٩٣,٥%) و (٨٥,٦%) لمستوى عكارة ٥٢ NTU على التوالي.

نتائج المرحلة الثانية وهي محور هذا البحث أظهرت انه بعد استخدام تقنية المتسعات عالية الجهد وتسليط قيم فولتية عالية مستمرة من ٥كيلو فولت إلى ٣٠ كيلو فولت أن مستويات عكارة المياه ازدادت عن مثيلاتها عند استخدام نفس قيم الجرع المستخدمة في المرحلة الأولى للمخثر بولي ألمنيوم كلورايد وبنفس ظروف التشغيل.مما يدل على أن هناك عملية فصل للعوالق والأطيان حدثت بسبب هذه الفولتيات المسلطة والتي أثرت بدورها على الدالة الفيزيائية لجهد زيتا أي على الشحنات المتكونة على سطوح جزيئات العوالق والأطيان مما أدى إلى زيادة مستويات العكارة وهذا يعني أن في هذه الحدود من الفولتيات أي من ٥ إلى ٣٠ كيلو فولت مستمر وباستخدام تقنية (Zeta Rod) لن يكون هناك تعزيز لعملية التلبيد وإنما سيكون هناك فصل للجزيئات الكبيرة من الأطيان والعوالق وهذه خاصية يمكن الاستفادة منها في منظومات معالجة المياه والتي تسمى بمنظومات التناضح العكسي (RO) لزيادة كفاءة المرشحات والأغشية المستخدمة .

Abstract

Turbidity is considered one of the most important problems of drinking water and it is usually treated with water containing molecules, of plankton and colloids through the process of coagulation and flocculation. And coagulants most commonly used in the water treatment process are aluminum sulfate (alum), ferric chloride,Poly aluminum chloride, which was recently discovered to have negative side effects on human health if are not taken into account the amount and weights added to water and allowed rates.

This research aims to study the possibility of controlling the physical function of " Zeta potential " through the study of its impact on the process of coagulation/flocculation of plankton and colloids in the water and causing turbidity in order to minimize the use of chemicals in water treatment processes.

The study consists of two phases The first is the comparison between the three efficiency coagulations .The most efficient of them was chosen in terms of its ability to remove the turbidity of the water and meet standard specifications for drinking water. and second phase consists comparing the coagulant that has been chosen in the first phase with the high-voltage capacitor (Zeta Rod) to study the impact of this technology on the coagulation process by applying a high DC voltage (5 kV to 30 kV) to the turbid waters samples.

Experiments were conducted by on samples of water whose turbidity was controlled from through the addition of Alkaúolin coagulants remove was evaluated using the jar test. The tests were carried out at three levels of turbidity 580 , 150, 50 NTU. The results show superiority of poly aluminum chloride over alum and ferric chloride to remove turbidity, It was found that a dose of 75 mg / l of this coagulant is able to reduce the turbidity of the water from 580 to 4.5 NTU that is, removal efficiency of (99.2%) which conforms to the specification of water of the Iraqi standard for drinking water, The same dose was able to reduce the turbidity of 150 NTU to 1.14 (99.24%) and 50 to 1.51 NTU (96.9%) in the same operating conditions and testing. While the efficiency of removal of turbidity for each of alum and ferric chloride in the same dose and operational conditions by (98.5%) and (98.8%) of the level of turbidity of 580 NTU and (95.6%) and (97.4%) of the level of turbidity of 150 NTU and (93.5%) and (85.6%) to the level of turbidity of 52 NTU, respectively.

The results of the second phase which is the focus of this research show that after the use of high voltage capacitor technology and the application of high DC voltage ranging from 5 kV to 30 kV, the levels of turbidity of the water increased over those when using the same values doses used in the first phase of the coagulant poly aluminum chloride and under the same operating conditions. which indicates that there is a separation process for plankton and clays because of these voltages applied, which in turn affects the physical function of" Zeta potential" , that is, it charges formed on the surfaces of plankton and clays particles resulting in increased turbidity levels.

This means that with these values of voltages ranging from 5 to 30 kV DC using a technique of (Zeta Rod) there will be no enhance for the process of coagulation, but there will be a separation of particles of larger of clays, plankton and this property could be used in systems of water treatment and so-called systems of reverse osmosis(RO)to increase the efficiency of filters and membranes used. ....

ABBREVIATIONS

Term	definition
Alum	Aluminum sulfate
ASTM	American Society for Testing and Materials
AWWA	American Water Works Association
AWWARF	American Water Works Association Research Foundation
CF	Chemical Flocculation
DBPs	Disinfection by-products ($\mu g\ L^{-1}$)
EC	Electro-coagulation
EF	Electro-flocculation
EPA	Environmental Protection Agency
E_r	Removal efficiency
FCWTF	Full-Scale water treatment facility
$FeCl_3$	Ferric chloride
FTU	Formazin Turbidity Units
HMM	High molar mass
HVC	High Voltage Capacitors
IVCl	Iraq Virtual Science Library
JTU	Jackson Turbidity Units
JWWA	Japan Water Works Association.
kV	Kilo volte
Mol.Wt.	Molecular weight (Daltons)
Mu	Millmicron
NF	Nano Filter Membranes
NOM	Natural organic matter ($mg\ L^{-1}$)
NTU	Nephelometric Turbidity Units
PACl	Poly aluminum chloride
PACls	Poly aluminum chloride (high-purity)
PAS	Poly aluminum sulfate
PASiC	Poly aluminum silicate chloride
pH	Potential of hydrogen(measurement of acidity of a water sample)

ppm	Parts per million(equivalent to milligram per liter)
PZC	Point of zero charge
RO	Reverse osmosis
RPM	Round per Minute
SDI	Silt density index
SUVA	Specific ultraviolet absorbance ((L.mg-1m-1)
T_{in}	The Initial Level Of Turbidity (NTU)
TOC	Total organic carbon (mg L^{-1})
T_{out}	Final Turbidity Level (NTU)
TSS	Total Suspended Solids
UM	Micrometer
UV	Ultraviolet irradiation
WRC	Water Research Commission
ζ	Zeta potential (mV)

Physical variables and constants

Symbol	definition
u	Electrophoretic mobility $m^2.V^{-1}.s^{-1}$
v	Velocity of particles $m. s^{-1}$
E	Electric field strength $V.m^{-1}$
e	Electronic charge $1.602x10^{-19}$ c
K	Boltzmann's constant $1.3806504 \times 10^{-23}$ J/K
n	Amount of substance . mol
T	Absolute temperature
N	Avogadro's number $6.02214179 \times 10^{23}$ particles/mol
I	Ionic strength
ε	Electrical permittivity of the solvent F/m
$\varepsilon_{\circ}$	Permittivity of a vacuum 8.8542×10^{-12} F/m
η	Viscosity $kg. m^{-1}. s^{-1}$
q	Charge of the particle . coulomb
δ	Thickness of the zone of influence of the charge on the particle
D	Dielectric constant of the liquid

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1:1 Introduction

Water is a very important resource for the human life. The Earth is mostly covered by water but most of it is not potable. 97% of water is salt that comes from oceans;3% is freshwater but less than 1% of this is usable from river and groundwater.[1]

With the development of industry and agriculture more and more water is becoming polluted. The rapid increase in population exacerbates water shortage, since the limited supply of fresh water cannot meet the future demand. Water availability and security is now a critical global priority, and the search for new water is more the urgent than ever.[1]

Water is the lifeblood of man, so attention to the quality of the water five thousand years ago. This interest was limited to the color of the water taste and smell. For this some treatment and operations have been used in a limited way during spaced historical periods (such as boiling, filtration and sedimentation and the use of salts such as alum) . Most of the Arab countries do not possess advanced processing units, so they depend mainly on traditional processing units . That leads to the elimination of three types of impurities: suspended solids, organic materials, microbes and other pathogens.[2]

The United Nations reports indicate that the using of contaminated and unhealthy water leads to the death children every 8 seconds as a result of water pollution.[3]

The reports indicate also that 50% of the population in developing countries suffer from water-related diseases and about (80%) of the total diseases in developing countries are due to water pollution , and more than (16%) of the world's population use contaminated water. The industrial development after the Second World War , and the increasing rate of industrial development in the last quarter of the twenteth century, in addition to the rapid population growth in that period have a significant impact on the environment in all its components , the effect of acid rain problems began , pollution of surface water and increasing demand for fresh water rates are almost in constant presence.[3]

Turbidity is one of the important characteristics to evaluate water quality and increased bacterial contamination. It is reason for increasing brownish colour, especially in winter and spring as a result of heavy rains that lead to soil and cliff erosion reaching surface water.[4]

At first it was human search for quality of water through its physical properties such as taste, smell and color only . But the developments of chemical and biological sciences and medicines, methods have made available for measuring water quality and determining its impact on human health and the living organisms.[5]

Turbidity is not only an aesthetic problem in water. But also water with a high turbidity can be very difficult or impossible to properly disinfect. As a result, the maximum allowable level of turbidity in water is 0.5 NTU, while the recommended level is about 0.1 NTU. (**NTU**, or **TU**, stands for nephelometric turbidity units, a measurement of the turbidity of water).[6]

In addition to removing turbidity from the water, coagulation and flocculation is beneficial in other ways. The process removes many bacteria which are suspended in the water and can be used to remove color from the water. Turbidity and color are more common in surface water than in groundwater. As surface water flows over the ground to streams, through streams, and then through rivers, the water picks up a large quantity of particles. as a result, while aeration is more commonly required for groundwater, treatment involving coagulation and flocculation is typical of surface water.[6]

However the overall development of science and technology since the beginning of this century and the discovery of the relationship between drinking water and some diseases prevalent, there has been rapid developments in the field of processing techniques that have added many processes which are generally designed to have water with a high degree of purity , so as to be free of turbidity and colorless, tasteless, odorless and safe from chemical and biological aspects . The treatment processes used to remove water hardness, such as facilitation or traditional treatment process include coagulation, sedimentation, filtration and injection . The process of coagulation plays an important role in the conventional treatment process of water through destabilized colloids which include organic and inorganic materials.[7]

Studies have shown that organic materials which are normally not considered harmful in themselves, but are transformed into outputs after secondary sterilization when they interact with the chlorine used to disinfect water can cause health problems.[7]

Coagulation is one of the traditional stages of treatment and it aims to remove suspended colloids (causing turbidity), such as amino acids and proteins, colloidal silica and toxic substances, , And this materials are not soluble and as do not deposit as easily as sand and clay . This colloidal solution is characterized by stability and its colloidal fractions enjoys at surface area belonging large and thus a high level of energy and are usually these molecules for larger fractions as a result of overflowing with energy level by assembling these molecules and the formation of particles, The stability of the colloidal solution is caused by the electrical charge of the colloid molecules (negative or positive) and these charges are not unusual electrical discharges , because of the presence of a double layer of negative charges and positive surrounding colloid part, where the electrolytes on the surface of the colloid molecule because of the forces of Van der waals and these ions in turn attract adverse ions because of static electrical forces of attraction and thus is formed the double electrical layer, In general there are two forces affecting the surface of the colloid .They are momentum thermodynamic and momentum of electric kinetic forces (Zeta momentum) ,and the ionic layer causes foreign particle movement toward the opposite pole reference and with the greater the number of foreign ions strength of Zeta increases and so as becomes firmer colloidal solution. In other words, to lose the stability of the solution Zeta force must be reduced and this is done by adding coagulates factors (electrolytes).Note that if the colloidal solution is positive, the negative ions are affected only and opposition is true, and generally water intended for purification contains colloids materials with a negative charge and thus coagulants are added, coagulation, such as solvents, salts of aluminum and iron with a positive charge and includes aluminum sulfate (alum), aluminum sulfate plus aluminate sodium alum black plus ferric chloride and ferric sulfate and adding electrolytes returns the charge particles to close to zero and then become easy to correlation with aluminum ions iron sediment that are formed as a result of the interactions, Figure(1-1)shows the stages of the conventional treatment.[8]

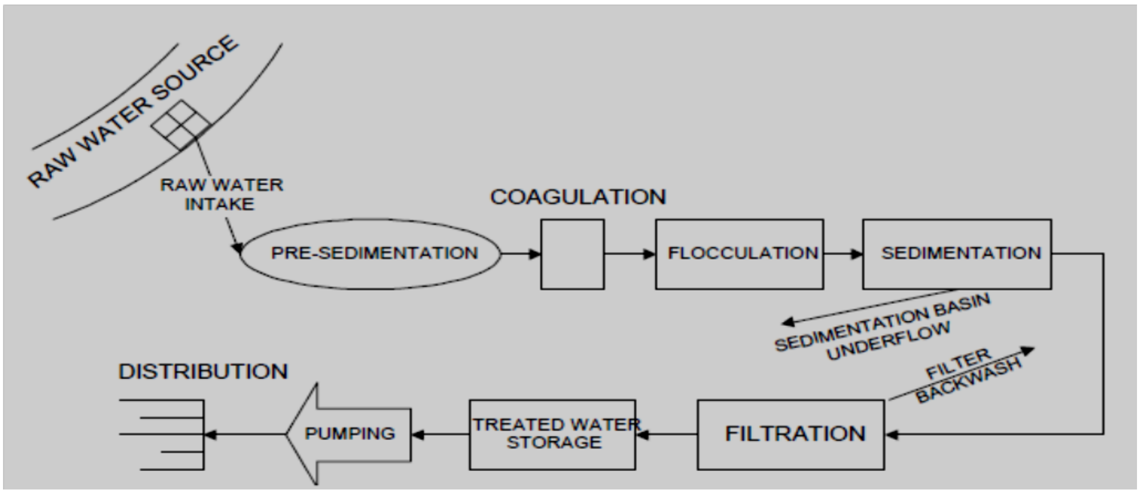


Figure (1-1) The traditional stages of treatment

Colloidal particles in nature normally carry charges on their surface, which leads to the stabilization of the suspension. By addition of some chemicals, the surface property of such colloidal particles can be changed or dissolved material can be precipitated so as to facilitate the separation of solids by gravity or filtration. Conversion of stable state dispersion to the unstable state is termed destabilization and the processes of destabilization are coagulation and flocculation.[9]

Often the terms coagulation and flocculation are used synonymously in spite of existing a subtle difference between the two. If destabilization is induced through charge neutralization by addition of inorganic chemicals, the process is called coagulation. On the other hand, the process of forming larger agglomerates of particles in suspension or of small agglomerates already formed as a result of coagulation through high molecular weight polymeric materials is called flocculation. No substantial change of surface charge is accomplished in flocculation. The agglomerates formed by coagulation are compact and loosely bound, whereas the flocs are of larger size, strongly bound and porous in case of flocculation.[9]

Oppositely charged ions in an electrolytic solution are attracted to the surface of a charged particle and can either be closely associated with the surface or distributed some way into the solution. Thus the two opposite forces, electrostatic attraction and ionic diffusion, produce a diffuse cloud of ions surrounding the particulate, which can extend up to 300 nm This co-existence of original charged surface and the neutralizing excess of counter-ions over co-ions distributed in a diffused manner is known as the electrical double layer.[10]

The double layer consists of two major regions, an inner layer (called Stern layer) where the initial layer of adsorbed ions and molecules are located at the particle surface; and the outer layer (called Gouy-Chapman layer) of oppositely charged counter-ions. The stability of colloidal suspension is greatly influenced by the potential of the Stern layer. Though this potential cannot be measured directly, it is approximated to the Zeta potential representing the electrical potential between the shear plane and the bulk solution.[11]

A charged particle dispersed in an ionic medium tends to have a concentration of opposite ions attracted towards it. For example, a negatively charged particle collects a number of positive counter-ions. As one moves further away from the particle, concentration of counterions decreases due to diffusion until ionic equilibrium is reached. A plot of the chargis contributed by these ions versus distance from the particle surface. Now, if the particles were imagined to be moving, they would tend to drag their counter ions along with them while leaving behind the ions that are further away from their surface. This would set up a plane of shear – the potential difference which is called the Zeta potential (ζ).[9]

In the water treatment technology called reverse osmosis (RO) is used as forms and different types of membranes are widely used in plants and desalination systems as water filter, These include types of nano filter membranes(NF) ,(Ultra filter) and (Micro filter) . The such systems are used for desalination for drinking or wastewater purification for re-use.[12]
Methods of desalination membranes provide nearly 5.8 million cubic meters per day of fresh water, this amount represents approximately 41% of total global production capacity for water desalination. It also represents the number of units that operate desalination techniques membranes more than 73% of the number of desalination units in the world.[13]
The membrane desalination ways use the natural properties of different types of membranes manufactured, some of the semi-permeable polymers allow the passage of water just below the salt ions dissolved under the influence of hydraulic pressure, such as the situation in reverse osmosis membranes There are other types of membrane which are impermeable to water which conduct electricity, and which allow selective passage of salt ions dissolved in water under the influence of voltage.[13]

One of the main problem of these system the occurrence of blockages in the membranes used in water purification process with the passage of time so as to a layer of sediment on the surface of these membranes. There are two ways form by which are used to remove and prevent blockages and debris from the membranes and work to raise the efficiency of the new system, they are:
1 - The first method is to remove sediment from the membranes after deposition and the remove the membranes of the system, and wash some of the chemicals (raising operational costs of the station whenever blockages occur rapidly).
2 - The second method depends on preventing sediment before reaching the membrane through the control of some of the physical properties of the membrane itself and the water that passes through the membrane, including the method of control of Zeta potential.[14]
Zeta potential to membrane surfaces can be controlled through with coating surfaces some chemicals, which in turn work on raising or lowering the value of Zeta potential, according to what is required to work with Zeta potential to water (which is usually neglected by workers in water treatment plants)and which reflects the significance of stability, in colloids is considered collided electric and a material considered that where Zeta potential will high(Whether this increase in value is positive or by negative 30 mV ±) . Evidence of high stability, which a colloid with low Zeta or with Zeta value equal zero are considered unstable and cause clotting and the formation of sediment harmful holes filters in part processing using filters(see Figure(1-2) and thus reduce the filtration efficiency and thus this technique is employed to find solutions easy and inexpensive to control the sediment in the parts used in water purification through the use of materials and devices to achieve that goal.[12]

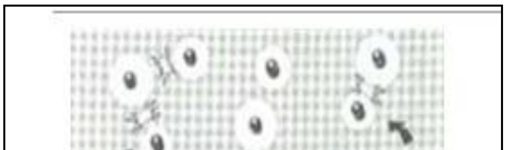


Figure (1-2): (1 - The scattered particles. Zeta potential high positively or negatively, 2- Molecules grouped . zeta potential and sessile or equal to zero)

1:2 Literature Survey

Increasing demand for water, especially with the population growth in the world leads to the need for improvement in the methods of treatment of water, especially in light of environmental pollution after the development of industry and agriculture in the world so as to prevent or minimize the negative effects resulting from water pollution, and the treatment methods that do not follow the correct measurements for addition of chemicals. Therefore there is a growing need to find alternative ways for traditional methods of water treatment .Over the past decades there have been a number of independent studies that seek to improve water treatment techniques and methods. One of these studies are :

Menachem Elimelech et.al. 1994 [12], studied the effect of changing Zeta-potential of water and adding some salt with changing concentrations on the efficiency of the reverse osmosis membranes used in the systems of water treatment, which is called (RO) to prevent the blockage of these membranes by preventing the layer on the surface of the membrane. Also they showed that Zeta-potential to reverse osmosis membranes affects the effective and strong presence of non-interacting chemicals and impurities on the surfaces of those membranes.

Zhu and Elimelech. **1997**[13] noted that there is a decrease in the rate of sediment inside the vehicle and membranes due to the addition of cellulose acetate . This decline is due to the growing electrical repulsion between colloids and the surface of the membrane. And the increase in the charge is responsible for a surface dispersion and that this increase is the result of the solution ionic forces in line with the principles of the dispersion of the movement known as electric particles and thus become less negative because of the double layer pressure and that leads to the collapse of the layer.

Romo and Pitts .1999 [15] present a study on cleaning the membranes used in water treatment systems of (RO) reverse osmosis. The process period was shortened and reduced to four times the period of time after the application of dispersion technique using high-voltage capacitors.

Shivaji S. Deshmukh et.al.2001[14]

The aim of this research is to study the impact of change in Zeta effort on wastewater treatment, as well as on some stations membranes in sewage treatment. The researchers found out that the presence of certain ions of parity bilateral such as (Ca $^{+\,+}$, Mg $^{+\,+}$) in wastewater works to increase the effect Zeta effort and more on membrane leading to increased life and improved efficiency.

Mohamed.h.El-masry, Olfet.M.Sadek, et.al. **2004** [16] achieved raw surface water purification from organic, inorganic and microbial content using the electro-coagulation method. Batch coagulation experiments were conducted using the jar-test method. A pair of aluminum electrodes (suspended in the jar) was charged with low voltage current for releasing aluminum ions in the raw water to precipitate the suspended matter. The optimum current density was 0.6325 mA cm^{-2}. The coagulation efficiency was evaluated by determining the turbidity of treated water. The efficiency reduction in raw water turbidity was 90%, leading to a change in water Zeta potential from −85 mV (before treatment) to −40 mV (after treatment), i.e. the particles tended to be destabilized and the coagulation process became predominant. Water contents of nitrates, phosphates and sulfates were considerably reduced by 77.5, 83.3 and 20.0%, respectively.

E. Ofira, , Y. Orenb , A. Adina. **2007** [17] present a study on comparison between two technologies: electro flocculation (EF) and chemical flocculation (CF), which may be of used as pretreatment in wastewater treatment. Natural water reservoir or wastewater intended for reuse, that contains high concentrations of iron particulates, often requires particle destabilization and removal. These particles can be removed by flocculation process applying ferric chloride (CF) or by Fe^{3+} ion (EF) that changes to Fe^{2+} in a short time. EF, a fairly new environmentally-friendly process has been developed with the aim of reducing, or even avoiding altogether, the amount of chemicals required during pretreatment. The behavior of iron oxide and hydroxide colloid in EF aims at solving the problem of contaminated water or industrial wastewater. This research investigated EF with iron electrodes and chemical dosing with ferric chlorides for several types and different concentrations of iron suspension. The results show a gradual rise in absolute ζ-potential and an increase in particle size with flocculation time. The results of both processes are compared in terms of the efficiency of solid/liquid separations performed by sludge sedimentation and filtration. Preliminary testing identified the process conditions, pH of the water treated, dosage of the coagulant, mixing conditions, conductivity, and current density. The target was to find the method which provides optimum coagulation and avoids excessively high energy consumption in EF.

E. Ofira, , Y. Orenb , A. Adina .**2007**[18] studied Electro flocculation (EF) of wastewater or water which reused with colloidal particles, such as kaolin and iron and with the right pH, might be optimal for aggregation during water treatment. The EF process is compared to chemical flocculation (CF) in terms of ζ-potential and particle size parameters. The results demonstrate that the particle becomes larger, except in the case of pH 4, where, after 50 min of flocculation, there is a jump in the value and the particle attains a diameter of more than 800 nm. Throughout the entire flocculation time, the ζ-potential values indicated a stronger negative charge on the particle. Basically, there was a gradual rise in absolute ζ-potential with the increase in particle size. The reason for this behavior can be explained by the solubility of kaolin with a decreasing ionic strength, when certain iron ions are released from the electrode and the charge of the particle is altered. The aggregation process, whereby a few small particles with a negatively-charged surface are rendered into a larger-sized particle, results in a larger total negative charge. It has long been recognized that ζ-potential is a very good index of the magnitude of the repulsive interaction between colloidal particles. Measurements of ζ-potential are commonly used to assess the stability of colloidal and particle size produced by EF. Thus, the coupling of the EF of kaolin and iron with the right pH might be optimal for producing particles.

A.I. Zouboulis, N.D. Tzoupanos .2009[19] in their study investigated the behavior of the new coagulant agent polyaluminium silicate chloride (PASiC).The main purpose was the examination of several possible derivatives of polyaluminium silicate chloride, based on different basicity (OH/Al molar ratio), silica content (Al/Si molar ratio) and preparation method .Moreover, a systematic study was conducted to define the optimum values of aforementioned major parameters, in order to produce an improved product, in comparison with the commonly applied polyaluminium chloride. The coagulation performance of PASiC products was evaluated for the treatment of contaminated tap water (in terms of turbidity and of NOM removal, as well as of residual Al concentrations and of zeta-potential measurements) . Additionally, the new products are compared with the laboratory of PACl, with results alum (i.e. $Al_2(SO_4)_3\cdot18H_2O$), as well as with commercially available PACl samples . Overall, the obtained results suggest that in order to produce a silica-based polyaluminium coagulant with improved coagulation properties, the basicity (OH/Al ratio) should be between 1.5 and 2.0, the silica content (Al/Si molar ratio) between 10 and 15 and should be prepared preferable with the co-polymerization technique. However, attention has to be paid to the specific application of these products.

Fengting Li , Jia-Qian Jiang, et.al . 2010 [20] provide a new method for the preparation of high-purity PACl , because it contains high aluminum oxide content (>10%), high basicity (>90%), and low insoluble substance (<0.04%), iron (<300 mg L^{-1}) and heavy metals. results show that for the whole dose range studied, high-purity PACls removed more turbidity than conventional PACl. On the other hand, when the dose was greater than 4 mg L^{-1} as Al, the high-purity PACl showed great UV_{254}-absorbance removal capacity; which was even much greater at high doses (e.g., 10 mg L^{-1}). This could be partly attributed to the charge effect where a relatively high Zeta potential in the test water was achieved by dosing high-purity PACl.

Xiao Zhan , Baoyu Gao , et.al . 2010 [21] investigated the effect of polyaluminum chloride (PAC) dosage on the coagulation performance and the floc characteristic with respect to the treatment of surface water with low specific UV absorbance(SUVA)value. The results show that natural organic matter (NOM) removal increases with the increasing dosage of PAC, while the turbidity removal decreased as the Zeta potential further increases to the positive side. At low PAC dosages, charge neutralization is the main mechanism for NOM removal. If the dosage of PAC is increased, entrapment, adsorption and complication play important role in removing(NOM.). The growth rate of the flocs was raised with the PAC dosage. Flocs formed at a PAC dosage of 3 mg/L were larger than those at high dosages (12 mg/L and 20 mg/L) and became more compact during the slow stir process. Floc breakage behavior show that flocs formed at different dosages of PAC gave different floc strength and the strength factors are in the order: 3 mg/L < 12 mg/L < 20 mg/L. On the other hand, the floc regrowth factors at different dosages of PAC were in the order: 3 mg/L > 20 mg/L > 12 mg/L. Coagulation treatment with 20 mg/L of PAC resulted in lower chlorine decay rate, but did not lower the total reacting compounds in the effluent when compared with coagulation with the PAC dosage of 12 mg/L .

Kherfan Sadeddin , Alnaif Naser, et.al.2011[22] focus on the use of electric coagulation method in water treatment . Electro-coagulation EC is a simple and efficient method for water treatment. It was not widely applied because of high capital cost compared to other treatment technologies. In recent years, many investigations have focused on the use of EC owing to the increase in environmental restrictions on effluent wastewater. The results show that feed water treated by EC is very clear and its quality exceeds the standard of feed water quality of reverse osmosis units.

Removal efficiencies of total suspended solids TSS and turbidity are more than 99% and 98%. The results presented in this paper on two water reverse units operated for 6 months show that total treatment of feed water by addition of EC reduces all fouling factors indicators such as water flow, pressure drop and silt density index SDI.

Hideto Yoshida , Takahisa Tatekawa , et.al. 2013 [23] found a new method of Zeta-potential measurement by the use of the liquid sedimentation balance method . The test samples used were spherical acrylic and silica particles, and beads mill was used to enhance particle dispersion. The new method can be used to estimate the zeta-potential of each particle size range. The absolute value of the negative Zeta-potential of small particles is greater than that of large particles. The results of measuring Zeta-potential by the new method agree with those of other conventional measurement devices.

The slope of the Zeta-potential with respect to particle diameter decreases as the waiting period following the beads mill pre-treatment process increases.

1:3 Aim Of The Work

This work aims to study the effect of physical function of Zeta potential on the coagulation process in water treatment and of the extent of its impact on the course of this process.

The work involves two phases. The first is a comparison between the effectiveness of three kinds of coagulant used in the coagulation process to remove the turbidity of the water (the more common chemicals used in drinking water treatment) which are *alum, ferric chloride, poly aluminum chloride* by using conventional jar test experiments. The second is to compare the effectiveness of the best coagulant with the use of ***Zeta Rod*** technique (High-Voltage Capacitors) and the impact of this technology on the coagulation process and turbidity levels in the water treatment in an attempt to reduce the use of chemicals to reduce the adverse impact of negative effects on human health.

Chapter Two : Theoretical Background

2 .1 Turbidity

Turbidity is a physical principal characteristic of water and is an expression of the optical property that causes light to be scattered and absorbed by particles and molecules rather than transmitted in straight lines through a water sample. It is caused by suspended matter or impurities that interfere with the clarity of the water. These impurities may include clay, silt, finely divided inorganic and organic matter, soluble colored organic compounds ,and plankton and other microscopic organisms.

Simply stated, turbidity is the measure of relative clarity of a liquid. Clarity is important when producing drinking water for human consumption and in many manufacturing uses. Once considered as a mostly aesthetic characteristic of drinking water, significant evidence exists that controlling turbidity is a competent safeguard against pathogens in drinking water.[24]

Photo (2-1) Turbidity standards of 5, 50, and 500 NTU [25]

Turbidity is the cloudiness or haziness of a fluid caused by individual particles (suspended solids) that are generally invisible to the naked eye, similar to smoke in air. The measurement of turbidity is a key test of water quality. Fluids can contain suspended solid matter consisting of particles of many different sizes. While some suspended material will be large enough and heavy enough to settle rapidly to the bottom of the container if a liquid sample is left to stand (the settable solids), very small particles will settle only very slowly or not at all if the sample is regularly agitated or the particles are colloidal. These small solid particles cause the liquid to appear turbid.[25]

2:1:1 Causes Of Turbidity Of Water

At open water, turbidity may be caused by growth of phytoplankton. Human activities that disturb land, such as construction, mining and agriculture , it can lead to high sediment levels entering water bodies during rain storms due to storm water runoff..

Certain industries such as quarrying, mining and coal recovery can generate very high levels of turbidity from colloidal rock particles. In drinking water, the higher the turbidity level, the higher the risk that people may develop gastrointestinal diseases. This is especially problematic for immuno-compromised people, because contaminants like viruses or bacteria can become attached to the suspended solid. The suspended solids interfere with water disinfection with chlorine because the particles act as shields for the virus and bacteria. Similarly, suspended solids can protect bacteria from ultraviolet(UV)sterilization of water. [26]

Typical sources of turbidity in drinking water include the following according to [24] . (see Figure (2-1)) :

- Waste discharges;
- Runoff from watersheds, especially those that are disturbed or eroding;
- Algae or aquatic weeds and products of their breakdown in water reservoirs, rivers, or lakes;
- Humic acids and other organic compounds resulting from decay of plants, leaves, etc. in water sources; and

- High iron concentrations which give waters a rust-red coloration (mainly in ground water and ground water under the direct influence of surface water).
- Air bubbles and particles from the treatment process (e.g., hydroxides, lime softening) .[24]

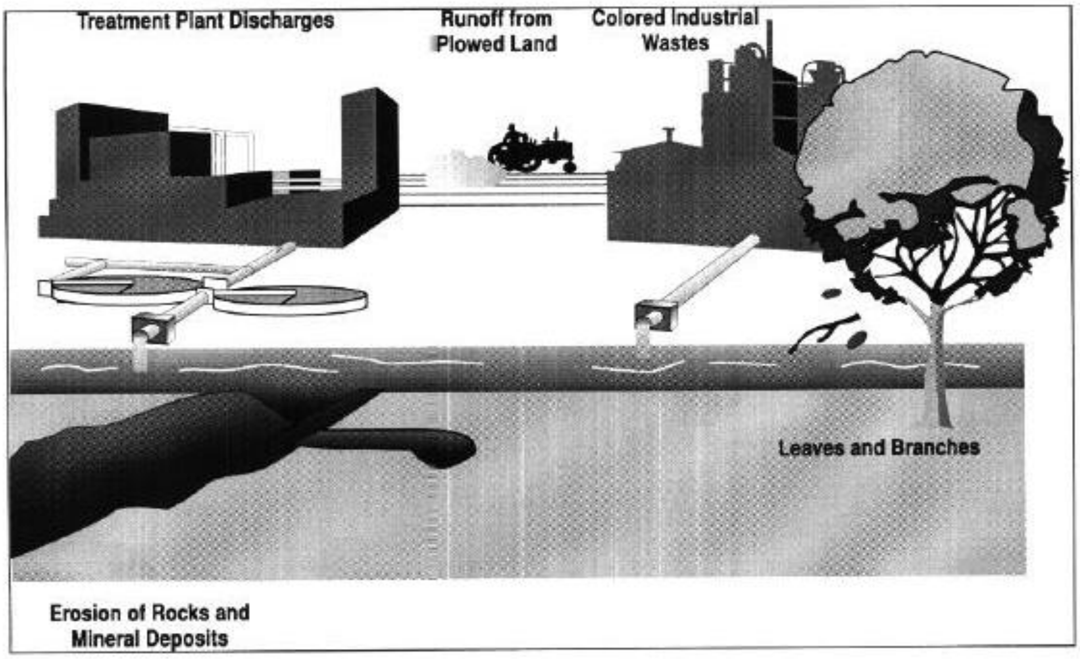


Figure (2-1). Typical sources of turbidity in drinking water

2:1:2 Turbidity Measurement Units

The first practical attempts to quantify turbidity date to 1900 when Whipple and Jackson developed a standard suspension fluid using 1,000 parts per million (ppm) of diatomaceous earth in distilled water. Dilution of this reference suspension resulted in a series of standard suspensions, which were then used to derive a ppm-silic scale for calibrating turbid meters.[24]

The standard method for determination of turbidity is based on the Jackson candle turbid meter, an application of Whipple and Jackson's ppm-silica scale. The Jackson candle turbid meter consists of a special candle and a flat-bottomed glass tube, and was calibrated by Jackson in graduations equivalent to ppm of suspended silica turbidity.A water sample is poured into the tube until the visual image of the candle flame, as viewed from the top of the tube, is diffused to a uniform glow. When the intensity of the scattered light equals that of the transmitted light, the image disappears; the depth of the sample in the tube is read against the ppm-silica scale, and turbidity was measured in Jackson turbidity units (JTU). Standards were prepared from materials found in nature, such as Fuller's earth, kaolin, and bed sediment, making consistency in formulation difficult to achieve.[24]

In 1926, Kingsbury and Clark discovered formazin, which is formulated completely of traceable raw materials and drastically improved the consistency in standards formulation. Formazin is a suitable suspension for turbidity standards when prepared accurately by weighing and dissolving 5.00 grams of hydrazine sulfate and 50.0 grams of hexamethylenetetramine in one liter of distilled water. The solution develops a white hue after standing at 25°C for 48 hours. A new unit of turbidity measurement was adopted called formazin turbidity units (FTU).[24]

Even though the consistency of formazin improved the accuracy of the Jackson Candle Turbid meter, it was still limited in its ability to measure extremely high or low turbidity. More precise measurements of very low turbidity were needed to define turbidity in samples containing fine solids. The Jackson Candle Turbid meter is impractical for this because the lowest turbidity value on this instrument is 25 JTU. The method is also cumbersome and too dependent on human judgment to determine the exact extinction point.

Indirect secondary methods were developed to estimate turbidity. Several visual extinction turbid meters were developed with improved light sources and comparison techniques, but all were still dependent on human judgment. Photoelectric detectors became popular since they are sensitive to very small changes in light intensity. These methods provided much better precision under certain conditions, but are still limited in ability to measure extremely high or low turbidities.[24]

Finally, turbidity measurement standards changed in the 1970's when the nephelometric turbid meter, or nephelometer, was developed which determines turbidity by the light scattered at an angle of 90° from the incident beam. A 90° detection angle is considered to be the least sensitive to variations in particle size. Nephelometry has been adopted by *Standard Methods* as the preferred means for measuring turbidity because of the method's sensitivity, precision, and applicability over a wide range of particle sizes and concentrations.The nephelometric method is calibrated using suspensions of formazin polymer such that a value of 40 nephelometric units (NTU) is approximately equal to 40 JTU. The preferred expression of turbidity is NTU.[24]

2:1:3 Particles Contributing To Turbidity

Particles in a raw water supply may be composed of inorganic materials, pathogens, or toxic materials. These particles may also provide sorbent sites for pesticides and other synthetic organic chemicals and heavy metals. Particles are undesirable not only for the cloudy appearance they impart to finished water, but because they also have the ability to shelter microorganisms from inactivation by disinfectants . Consequently, a principal element in supplying quality drinking water is the maximum removal of particles. To establish or optimize a particle removal process, it is important to understand the physical properties of particles.

Particles suspended in water can be categorized into three classes based on their origin:

1. Inorganic materials, such as silt or minerals;

2. Living or dead organic matter; and
3. Biotic material including algae, viruses and bacteria.

Due to the range of small sizes for common particles in water, it is common to find sizes termed in "microns" within the water industry. A micron, or micrometer, is equal to 1 x 10^{-6} meters, or 0.00004 inches. Generally, particulate contaminants to be removed from a raw water source range from the larger macro sized particles visible to the naked eye, to the ionic particles viewed only by scanning electron microscopes. [27]

2:1:3:1 Inorganic Particles

Inorganic particles in water are produced by the natural weathering of minerals, including both suspended and dissolved materials. Inorganic particles may consist of iron oxides, salts, sulfur, silts and clays such as bentonite or muscovite. Depending on the concentration of inorganic particles present in raw water sources, human health effects can vary from beneficial to toxic. Naturally occurring minerals find their way into raw water sources either naturally through the breakdown of minerals in rock, or through industrial process discharges which have contaminated a raw water source. Industrial contributors can include mining, smelting, coal burning power producers, oil and gas companies, and electroplating operations. Clays, metal hydroxides, and other particles originating from mineral sources typically vary from several nanometers to several microns in diameter, with a continuous size distribution over this range. In surface waters, the majority of these particles are within a 0.1 to 1 micron size range. as a result of their settling characteristics, particles in this size range have the ability to remain in suspension in moving water. Particles of this size range scatter visible light efficiently, due to the larger surface areas which are created as particles decrease in size. This scattering gives the water a turbid, or cloudy, appearance at very low concentrations.[28]

The large surface areas created by even a small mass concentration of the colloid particles provide abundant adsorption sites for natural and synthetic organic matter, metals, and other toxic substances. Bacteria and viruses can also attach to these particles, and there is some concern that inorganic particulate contamination has the ability to shield microorganisms from inactivation by disinfectants.[28]

Inorganic particles such as clays make up a large proportion of the particles present in natural waters. These particles may consist of iron oxides, silicates, calcites, clays, aluminum oxides, and many other minerals.[29]

Although the particles in each watershed are unique, all show similar electrochemical behavior, since their surfaces are generally covered with surface hydroxyl (OH) groups. Depending upon the pH of solution, the charge of these hydroxyl groups may be positive or negative.[30]

2:1:3:2 Organic Particles

Organic materials are compounds, natural or manmade, having a chemical structure based upon the carbon molecule. Millions of organic compounds containing carbon have been identified and named, including; hydrocarbons, wood, sugars, proteins, plastics, petroleum-based compounds, solvents, pesticides and herbicides.

Both naturally-occurring and synthetic organics are present in surface waters and typically originate from the following sources: [31]

1. The decomposition of naturally occurring organic materials in the environment;
2. Industrial, agricultural and domestic activities; and
3. Reactions occurring during the treatment and distribution of drinking water.

Organics may have adverse human health impacts, such as toxicity, or as carcinogens when ingested. In addition, naturally occurring organics, most widely referred to as natural organic matter (NOM), can give raw water a characteristic color, taste, or odor. Furthermore, organics in water can be altered by treatment processes resulting in disinfection byproducts (DBPs).[31]

In organic particles such as bacteria, under the pH conditions of most natural waters, these particles have a negative surface charge.[30]

2:1:3:3 Particles of Biotic Origin

Four categories of waterborne microorganisms exist as particles contributing to the turbidity of raw water:

- Protozoans;

  Protozoans are organisms that can exist in colonies or as single cells. Some protozoans are capable of producing spores, a small reproductive body capable of reproducing the organism under favorable conditions. In water, most spores resist adverse conditions that would readily destroy the parent organism.

  Of the tens of thousands of species of protozoa, the principal protozoan pathogens of concern in potable water are *Cryptosporidium*, *Giardia lamblia,* and *E. histolytica*. When these organisms are ingested by humans, they can cause symptoms including; stomach cramps, diarrhea, fever, vomiting, and dehydration. These parasites are typically more resistant to traditional chlorine disinfection than coliforms.[32]

- Enteric viruses;

  A virus is a parasitic, infectious microbe, composed almost entirely of protein and nucleic acids that can cause disease in humans and other living organisms. Viruses can reproduce only within living cells, and typically range from 0.004 to 0.1 micron in diameter.[28]

- Algae;

Algae are common and normal inhabitants of surface waters and are encountered in every water supply that is exposed to sunlight. [33]

Algae typically range in size from 5 to 100 microns.

Algae are not typically a threat to public health in a drinking water supply. Concerns in potable water treatment arising from the presence of algae include; the ability to create large quantities of organic matter; the production of turbidity, tastes and odors in source water, and; the physical impact on the water treatment plant processes.[28]

- Bacteria.

Bacteria are single-celled organisms that lack well-defined nuclear membranes and other specialized functional cell parts. Bacterial cells typically range from 1 to 15 microns in length. They vary in shape from simple spheres to filamentous threads.

Bacteria can exist almost anywhere on earth and in almost any medium. Some are beneficial to man while others are harmful, or even fatal. The principal bacterial pathogens of concern in water treatment are the *Salmonella, Shigella, Yersinia enterocolitica, enteropathogenic E. coli, Campylobacter jejuni, Legionella, Vibrio cholerae,* and *Mycobacterium*..[28]

It can be concluded from the above that there are three types of objects which can be found in water. from smallest to largest, these objects are chemicals in solution, colloidal solids, and suspended solids. Coagulation/flocculation will remove colloidal and suspended solids from water.

Chemicals in solution which have been completely dissolved in the water are electrically charged and can interact with the water, so they are completely stable and will never settle out of the water. Chemicals in solution are not visible, either using the naked eye or using a microscope, and are less than 1 Mu in size. (A **Mu**, or **millimicron**, is equal to 0.000000039 inches.) An example of a chemical in solution is sugar in water.[6]

Colloidal solids, also known as **nonsettleable solids**, do not dissolve in water although they are electrically charged. Still, the particles are so small that they will not settle out of the water even after several years and they cannot be removed by filtration alone. Colloidal solids range between 1 and 500 Mu in size and can be seen only with a high-powered microscope. Examples include bacteria, fine clays, and silts. Colloidal solids often cause colored water, such as the "tea color" of swamp water.[6]

Finally, **suspended**, or **settleable**, **solids** will settle out of water over time, though this may be so slow that it is impractical to merely allow the particles to settle out in a water treatment plant. The particles are more than 1,000 Mu in size and can be seen with a microscope or, sometimes, with the naked eye. Examples of suspended solids include sand and heavy silts.[6]

2:2 Treating Water To Remove Turbidity

One of the most commonly used methods for the removal of suspended solids in drinking water is the addition of coagulant and flocculation aids, such as alum, ferric chloride, and long chain polymers.[34]

Coagulation is the process of decreasing or neutralizing the electric charge on suspended particles or Zeta potential. Similar electric charges on small particles in water cause the particles to naturally repel each other and hold the small, colloidal particles apart and keep them in suspension.

The coagulation/flocculation process neutralizes or reduces the negative charge on the particles. This allows the Van der Waals force of attraction to encourage initial aggregation of colloidal and fine suspended materials to form microfloc. Flocculation is the process of bringing together the microfloc particles to form large agglomerations by physically mixing or through the binding action of flocculants, such as long chain polymers. A classical coagulation/flocculation unit process consists of three separate steps: according to [35]

1) Rapid or flash mixing: the suitable chemicals (coagulants/flocculants and if required pH adjusters) are added to the wastewater stream, which is stirred and intensively mixed at high speed.

2) Slow mixing (coagulation and flocculation): the wastewater is only moderately stirred in order to form large flocs, which are easily settled out.

3) Sedimentation: the floc formed during flocculation is allowed to settle out and is separated from the effluent stream.[35]

2:2:1 *Coagulation and Flocculation*

The chemistry of coagulation and flocculation is primarily based on electricity.

Electricity is the behavior of negative and positively charged particles due to their attraction and repulsion.

Like charges (two negatively charged particles or two positively charged particles) repel each other while opposite charges (a positively charged particle and a negatively charged particle) attract.[6]

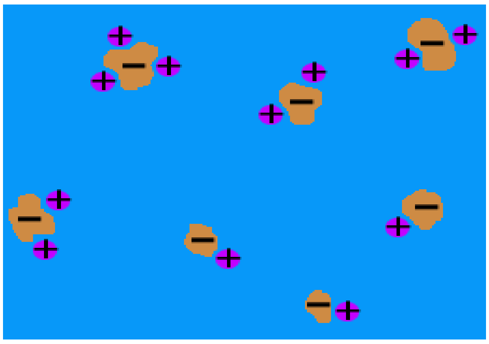
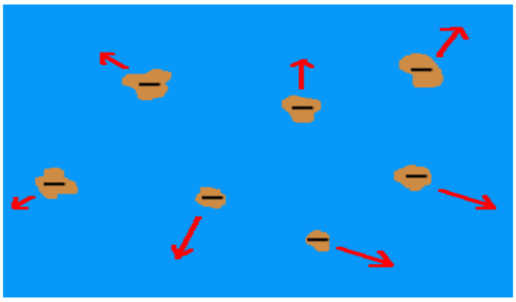


Figure (2-2). Negatively charged particles repel each other due to electricity

Most particles dissolved in water have a negative charge, so they tend to repel each other. As a result, they stay dispersed and dissolved or colloidal in the water, as shown above in Figure (2-2) .The purpose of most coagulant chemicals is to neutralize the negative charges on the turbidity particles to prevent those particles from repelling each other. The amount of coagulant which should be added to the water will depend on the **Zeta potential**, a measurement of the magnitude of electrical charge surrounding the colloidal particles. [6]

You can think of the Zeta potential as the amount of repulsive force which keeps the particles in the water. If the zeta potential is large, then more coagulants will be needed. Coagulants tend to be positively charged. Due to their positive charge, they are attracted to the negative particles in the water, as shown below in Figure(2-3).[6]



Figure(2-3). Positively charged coagulants attract to negatively charged particles due to electricity.

The combination of positive and negative charges results in a **neutral**, or lack, of charge. As a result, the particles no longer repel each other.

The next force which will affect the particles is known as Van der Waal's forces. **Van der Waal's forces** refer to the tendency of particles in nature to attract each other weakly if they have no charge.[6]

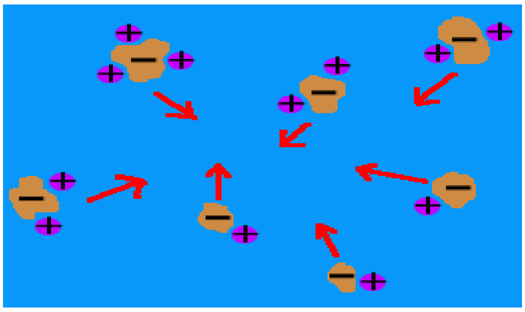


Figure (2-4). Neutrally charged particles attract due to Van der Waal's forces

Once the particles in water are not repelling each other, van der Waal's forces make the particles drift toward each other and join together into a group. When enough particles have joined together, they become floc and will settle out of the water.[6]

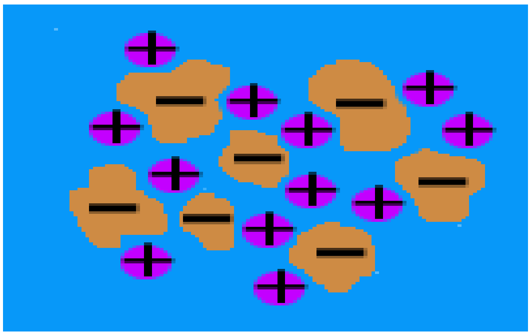


Figure (2-5). Particles and coagulants join together into floc.

Coagulation is defined as the addition of chemicals and mixing such that particles and some dissolved contaminants are aggregated into larger particles that can be removed by solids removal processes, such as clarification and filtration.[36]

Traditional coagulation has the primary purpose of removing suspended materials and organic matter with minimal sludge production in order to decrease turbidity.[37]

Coagulation is a water treatment process that adds a substance, typically a metal, to a solution. The most common coagulants include ferric sulfate ($(Fe_2(SO_4)_3.8H_2O)$, ferrous sulfate ($FeSO_4.7H_2O$), ferric chloride ($FeCl_3$), aluminum sulfate, also known as alum, ($Al_2(SO_4)_3.14H_2O$), aluminum chloride ($AlCl_3.6H_2O$), and sodium aluminate ($NaAlO_2$).[38]

Alum is most commonly used in drinking water whereas the iron compounds (both ferric and ferrous forms) are mostly used in treating waste waters. However, ferric chloride has been increasing in its application in drinking water. For alum, the stoichiometric coagulation reaction is seen in Eqn. 1

$$Al_2(SO_4)_3 \cdot 18H_2O + 3Ca(HCO_3)2 = 2Al(OH)_3(s) + 3CaSO_4 + 6CO_2(g) + 18H2O \qquad \text{(Eqn 1)}$$

For iron coagulation, the stoichiometric coagulation reaction is seen in Eqn. 2:

$$2FeCl_3 + 3Ca(HCO_3)_2 = 2Fe(OH)_3(s) + 3CaCl_2 + 6CO_2(g) \qquad \text{(Eqn. 2)}$$

In these reactions, calcium bicarbonate represents natural or added alkalinity which is needed in order to generate the precipitate as one of the products. as a result, these reactions consume alkalinity in addition to lowering the pH, thus creating water that is considered unstable if sufficient alkalinity is not present in the system.

The goal of coagulation is to change the oxidation state, and therefore change the chemical properties, to remove unwanted negatively charged colloids. Colloids typically have a diameter less than 10 µm which causes their sedimentation in water by gravity to be less than 10^{-2} cm/sec and therefore remain suspended.[39]

Coagulation masks the charge repulsion between these suspended particles so that they group together and form larger, heavier particles which are easier to remove from a solution than small particles. Typical coagulation is intended to reduce the turbidity (improve clarity) of the water. In short, coagulation is the addition of chemicals to water to destroy or reduce repulsive forces and induce particle agglomeration. Destabilization reduces the energy barrier between repulsive forces, providing conditions in which Van der Waals or other adhesive forces can dominate.[38]

In general, coagulation tends to remove more hydrophobic NOM compounds than hydrophilic NOM compounds.[40]

In natural waters, colloids predominantly carry a negative charge due to a variety of functional groups on the surface of the particle.[39]

Chemicals used in coagulation are positively charged and act as neutralizing agents when interacting with the negative charges on NOM.[41]

Coagulation causes destabilization by either reducing the energy barrier so Van der Waals forces (or other adhesive forces) can dominate or by double layer compression by reducing the surface potential as a result of changes in the surface chemistry (bridging or entrapment).[42]

Following coagulation, flocculation is the physical process of promoting particle contact to facilitate the agglomeration to larger floc which can then settle by gravity. In a typical water treatment facility(illustrated by Figure(2-6)).[43]

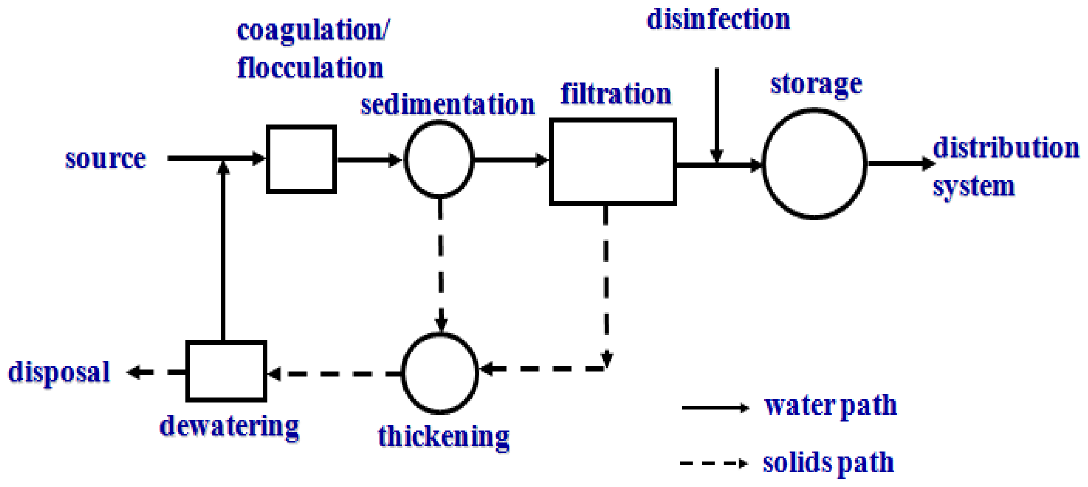


Figure (2-6): Theoretical water treatment process

these processes are accomplished using two different tanks in series. The first tank is a rapid mix tank, with velocity gradients preferred in the range of 3000 to 5000 s^{-1}, to which the coagulant dose is added so that the particles become destabilized. The second tank is the flocculation tank, which is a slow mix tank, with velocity gradients preferred in the range of 20 to 80 s^{-1} for solids and color removal coagulation or 130 to 300 s^{-1} for softening, to promote particle collision and aggregation. An illustration of coagulation, flocculation and settling can be seen in(Figure 2-7). [43]

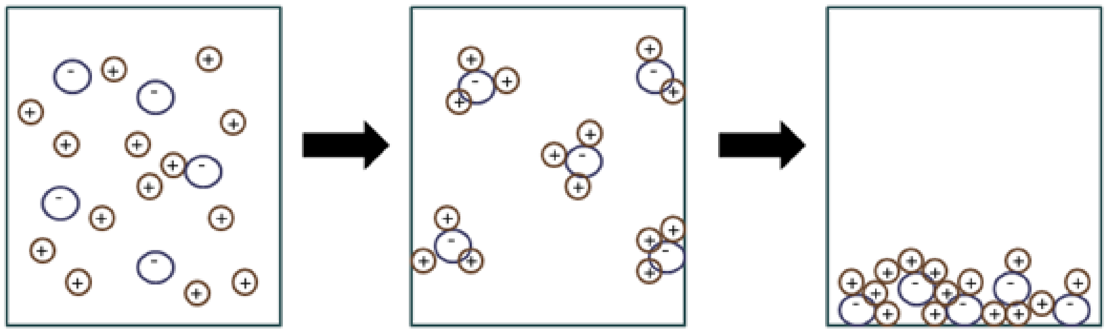


Figure (2-7): Coagulation (ion dispersion), flocculation (floc formation), and settling (flocs settle by gravity)

2:2:2 Coagulants Used In Water Treatment

A coagulant is any chemical that is used to bring about the agglomeration of colloidal particle. Inorganic or mineral coagulants, such as aluminum sulfate have been used for the coagulation of colloidal suspension in water for centuries, while in comparison, polyelectrolytes are a relatively new innovation in water treatment. Polyelectrolytes are used in water treatment both for coagulation and as coagulant/flocculant aids to strengthen flocs and improve their settleability .The choice of coagulant depends on the chemical requirements of the treatment process.[44]

Chemical coagulation of turbid water is one of the conventional techniques used for purifying raw water. The technique involves the interaction of particulate and/or colloids with a destabilizing agent. The essential purpose of coagulation is to aggregate these particles into large sizes that will settle quickly.[45]

Several inorganic salts of aluminum and iron are commercially available for chemical coagulation. Alum (aluminum sulfate) is almost exclusively used in the treatment of drinking water, while iron – as ferric chloride – is predominantly used in treating domestic and industrial waste waters . [46]

The effectiveness of coagulation to remove NOM and particles depends on several factors, including coagulant type and dosage, mixing conditions, pH, temperature, particle and NOM properties (such as size, functionality, charge and hydro phobicity), as well as the presence of divalent cations and concentrations of destabilizing anions (bicarbonate, chloride, and sulfate). Ferric chloride and ferric sulfate as well as alum are acidic, thus lowering the pH after application. Hence, low alkalinity waters may need addition of base, while high alkalinity may sometimes need to be lowered by acid addition to reach efficient pH range .However, high alkalinity waters have higher buffer capacity over low alkalinity waters against pH decrease .[47]

Coagulant chemicals come in two main types - primary coagulants and coagulant aids. **Primary coagulants** neutralize the electrical charges of particles in the water which causes the particles to clump together. **Coagulant aids** add density to slow-settling flocs and add toughness to the flocs so that they will not break up during the mixing and settling processes.Primary coagulants are always used in the coagulation/flocculation process. Coagulant aids, in contrast, are not always required and are generally used to reduce flocculation time. Chemically, coagulant chemicals are either metallic salts (such as alum) or polymers. **Polymers** are man-made organic compounds made up of a long chain of smaller molecules. Polymers can be either **cationic** (positively charged), **anionic** (negatively charged), or **nonionic** (neutrally charged.) The table below shows many of the common coagulant chemicals and lists whether they are used as primary coagulants or as coagulant aids.[6]

Table.(2-1) Coagulant chemicals, primary coagulants or as coagulant aids

Chemical Name	**Chemical Formula**	**Primary Coagulant**	**Coagulant Aid**
Aluminum sulfate (Alum)	$Al_2(SO_4)_3 \cdot 14\ H_2O$	**X**	
Ferrous sulfate	$FeSO_4 \cdot 7\ H_2O$	**X**	
Ferric sulfate	$Fe_2(SO_4)_3 \cdot 9\ H_2O$	**X**	
Ferric chloride	$FeCl_3 \cdot 6\ H_2O$	**X**	
Cationic polymer	Various	**X**	**X**
Calcium hydroxide (Lime)	$Ca(OH)_2$	**X**	**X**
Calcium oxide (Quicklime)	CaO	**X**	**X**
Sodium aluminate	$Na_2Al_2O_4$	**X**	**X**
Bentonite	Clay		**X**
Calcium carbonate	$CaCO_3$		**X**
Sodium silicate	Na_2SiO_3		**X**
Anionic polymer	Various		**X**
Nonionic polymer	Various		**X**

2:2:2:1 ***Aluminum Sulfate***

Aluminum sulfate is a compound that has the formula $Al_2(SO_4)_3$, and its needle-shaped crystals are colorless, crystalline contains in its network eighteen water molecule($Al_2(SO_4)_3.18H_2O$).
it compound degrades well in water, especially in hot water, but it is not soluble in ethanol

Aluminum sulfate loses crystal water on heating above 86 ° C, and its anhydrous form above 340 ° C (white powder). Solution is prepared from aluminumhydroxide in concentrated sulfuric acid. [48]

$$2Al(OH)_3 + 3H_2SO_4 \rightarrow Al_2(SO_4)_3 + 6H_2O$$

Principal aluminum compounds that are commercially available for suspended solids removal are dry and liquid alum. Commercial dry alum most often used in wastewater treatment is known as filler alum and has the approximate formula $Al_2(SO_4)_3$, $14H_2O$ and a molecular weight of about 600. Activated alum contains about 9% sodium silicate, which improves the coagulation in some waters. Black alum is the alum containing activated carbon. Another aluminum compound in use is sodium aluminate. It is much more alkaline compound than alum. When added alkalinity is found to be necessary for the efficient coagulation,
sodium aluminate may be substituted for all or part of the needed alum dose.[9]

photo (2-2): Aluminum sulphate

Aluminum sulfate or alum has been used for several centuries in water treatment and is probably the most well known and commonly used coagulant. The chemical is prepared by reacting bauxite or certain clays with sulfric acid. Alum when prepared from virgin chemicals does not normally contain any impurities of concern. Pure alum 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. Iron is not a problem as an impurity as it is a coagulant in its own right and tends to assist the coagulation process.

Alum is sometimes prepared from low-grade clays and from waste acid. This in some cases leads to the presence of undesirable concentrations of heavy (toxic) metals in the solution. Although the alum is usually much cheaper in such cases care should be taken when approving its use and it is probably better to confine the use of low grade alum to wastewater treatment.[44]

Alum can be bought in liquid form with a concentration of 8.3%, or in dry form with a concentration of 17%. When alum is added to water, it reacts with the water and results in positively charged ions.[6]

Alum chemistry can be described by the presence of three species: Al^{+3} , $Al(OH)^{+2}$, and $Al(OH)^{-}_{4}$ in equilibrium with an amorphous $Al(OH)_{(3am)}$ solid phase . [49][50]

the alum is least soluble at pH 6.0. This means that at pH 6.0, the maximum amount of

coagulant is converted to solid-phase floc particles. At pH values higher or lower than this pH of minimum solubility, dissolved Al levels in the treated water will increase. The solid phase formed upon precipitation, $Al(OH)_{3(am)}$, has a surface charge that is dependent on pH, due to the hydroxyl groups present. As is described for inorganic particles and NOM, the surface charge is more positive at lower pH. This has implications for the adsorption of NOM onto the floc surface, and the filterability of the floc.[50][51][52]
When alum is added to a wastewater, the following reaction takes place:

$A12(SO4)3 \cdot 18\ H2O + 3\ Ca(HCO3)2 \Leftrightarrow 3\ CaSO4 + 2\ Al(OH)3 + 6\ CO2 + 18\ H2O.$

The insoluble aluminum hydroxide, $Al(OH)_3$, is a gelatinous floc that settles slowly through the wastewater, sweeping out the suspended material. Alkalinity is required for the reaction and, if not available, must be added at the rate of 0.45 mg/L as $CaCO_3$ for every 1 mg/L alum.[53]

Table.(2-2) The features and positives and negatives of alum [47]

Coagulant type	**Features**	**Positive**	**Negative**
Alum	Trivalent aluminum ions are released into a solution from the respective salt. They are hydrolyzed and form soluble complexes possessing high positive charges. Coagulation efficiency depends on e.g. coagulant dose, mixing, pH, temperature, particle and NOM properties. During coagulation, the most effective range of pH is suggested to be about 5–6.5.	Stable, easily handled, readily soluble. Better turbidity removal than with ferric salts in many cases. Can be more effective than ferric in low doses. Higher color removal efficiency.	Relatively high coagulant residuals in the finished water in some cases.Possibility towards Alzheimer's disease. Ferric salts have been noted to be better in removing NOM than aluminum salts in many investigations. High alkalinity consumption. Sulfate and/or chloride in finished water increases corrosivity.

2:2:2:2 Ferric Chloride

Triple iron chloride chemical compound with the formula $FeCl_3$, and in the form of yellow crystals (hexagonal hydrate)**.** has property to be significantly damp and it is often in the form of hexagonal hydrate, and often has the smell of hydrogen chloride.
Ferric chloride compound degrades triple well in water, forming an acidic solution of all property .It is prepared from the interaction of chlorine with iron chloride in aqueous medium according the equation:

$2[Fe(Cl_2)(H_2O)_4]\ .2H_2O + Cl_2 \rightarrow 2[Fe[H_2O]_6]Cl_3$

The non-aqueous him matter is prepared by passing chlorine gas on iron at high temperatures.[48]

photo (2-3): Ferric chloride

Iron compounds possess pH coagulation ranges and floc characteristics similar to aluminum sulfate. The cost of iron compounds may often be less than that of alum. However, the iron compounds are generally corrosive and often present difficulties in dissolving, and their use may result in high soluble iron concentration in process effluents. The iron salts most commonly used as coagulants include ferric sulfate, ferric chloride and ferrous sulfate. These compounds often produce good coagulation when conditions are too acidic for best results with alum. Sometimes the particles are best removed under acidic conditions and iron compounds give better results.[9]

Ferric chloride, as is the case for the other iron salts, was originally a waste product from spent pickling solution. However, now that demand has increased it is on purpose made. It is readily prepared from reacting scrap iron with hydrochloric acid. Because it is highly deliquescent in its crystalline form it is almost invariably supplied as a solution. Ferric chloride is highly acidic and the solution contains free hydrochloric acid. The solution is highly corrosive to nearly all normally used metals including all grades of stainless steel and needs to be stored, pumped and conveyed in synthetic corrosion-resistant materials. The chemical is normally supplied as a solution of about 40% strength as $FeCl_3$ with an SG of about 1,4 and a pH of less than 1.[44]

When ferric chloride is added to a wastewater, the following reaction takes place:

$$2\ FeCl_3 \cdot 6\ H_2O + 3\ Ca(HCO_3)_2 \Leftrightarrow 3\ CaCl_2 + 2\ Fe(OH)_3 + 6\ CO_2 + 12\ H_2O$$

The insoluble ferric hydroxide, $Fe(OH)_3$, is also a gelatinous floc that settles in the wastewater, sweeping out the suspended material. Alkalinity is required for the reaction and, if not available, must be added at the rate of 0.55 mg/L $CaCO_3$ for every 1 mg/L ferric chloride. [53]

Table.(2-3) The features and positives and negatives of Ferric chloride[47]

Coagulant type	**Features**	**Positive**	**Negative**

Ferric chloride	Ferric salts hydrolyse similarly as aluminum salts when added to water and form different hydrolysis products. Effectiveness of coagulation depends on the same factors during alum salt coagulation. The most effective range of pH is suggested to be pH 4.5–6	Ferric salts have been noted to be better in removing NOM than aluminium salts in many investigations. Especially the removal of middle size NOM fractions is noted to be more efficient. Not so sensitive to temperature changes compared to alum..	Ferric-based coagulant with acid strength and associated optimized flocculation pH ranges can produce purified water with less buffering capacity and require greater chemical addition for stabilization and corrosion control. High alkalinity consumption. Sulfate and/or chloride in finished water increases corrosivity.

2:2:2:3 Polyaluminum Chloride (PACl)

The polyelectrolytes used in water treatment are high molecular weight, synthetic organic polymers, produced by the polymerization of one (homopolymer) or more (copolymer) types of monomer units. Since the type and number of monomer units can be varied during the manufacture of polyelectrolyte's, a wide variety of polymers can be produced. In addition to this, the polymer chains can be linear, branched or cross-linked, adding to their complexity.

The molecular weight, solubility and electronic charge can provide useful information regarding the efficacy and the toxicity of a particular polymer. Polymers can contain both negatively and positively charged sites and are usually classified according to this, cationic having an overall positive charge, anionic and overall negative charge, non-ionic being neutral and amphoteric having both positive and negative sites.

The more highly charged a polymer, the more soluble it is likely to be and therefore also the more bioavailable to aquatic organisms.

Cationic polyelectrolytes are usually referred to as primary coagulants, while non-ionic and anionic are referred to as coagulant aids or flocculants and these have relatively high molecular weights, often in the region of ten times or more that of the typical primary coagulant.[44]

In recent years, prehydrolyzed aluminum coagulants, e.g. polyaluminium chloride (PACl) and polyaluminium sulfate (PAS),have been developed and researched . PACl has been made by partially neutralizing AlCl$_3$ to different basicity ratios, and its use has been continuously spreading. Prehydrolyzing the AlCl$_3$ can enhance the amount of Al$_{13}$ (AlO$_4$Al$_{12}$(OH)$_{24}^{7+}$) in the coagulation process, which, in turn, has been noted to be the most efficient Al-species for contaminant removal . These coagulant species (Al$_{13}$ or Al$_b$) are considered to be the most efficient Al-species due to their larger size and higher positive charges. In several studies the flocs formed under various aluminium coagulant species are observed to have different properties. and also the destabilization mechanisms of different aluminium species have been investigated . The prehydrolyzed polymer coagulants have been reported to have advantages over conventional coagulants, including less temperature or pH dependence, as well as smaller alkalinity consumption . but the characteristics of the water to be treated (e.g. alkalinity, pH, and NOM content) play a major role in the choice of a proper coagulant. Consequently, prehydrolyzed coagulants have not been noticed to enhance the removal efficiency of NOM in all cases.[47]

A range of factors such as the nature of the water, the coagulation pH and the dose of coagulant together influence the range of species formed and subsequently, the treatment performance. Considerable interest and attention have been paid in last three decades to prepare pre-hydrolysed metal-ion coagulants, based on either aluminium (e.g., poly-aluminium chloride). These have been shown to perform better in some cases, in comparison with conventional coagulants such as aluminium sulfate or ferric sulfate. The superior performance of pre-polymerized coagulants is attributed to their wider working pH range, a lower sensitivity to low water temperature, lower doses required to achieve the equivalent treatment efficiency, and lower residual metal-ion concentrations.

One of the major pre-polymerized coagulants, poly-aluminum chloride (PACl) is used broadly for water and wastewater treatment.PACl contains different amounts of hydroxyl, and the molecular formula of PACl is $[Al_m(OH)_n(H_2O)_x].Cl_{3m-n}$ $(n \leq 3m)$.[54]

Polyaluminum chlorides (PACl) are synthetic polymers that serve as an inorganic aluminum-based coagulant. PACl is a potential substitute for alum because: (1) PACl tends to be more robust than alum with regard to achieving coagulation goals because their precipitates vary less under changing environmental conditions; and (2) PACl has less potential toxicity-related concerns than alum. PACl products react to form insoluble aluminum polyhydroxides which precipitate in big volumetric flocs similar to those formed with alum to absorb and precipitate suspended pollutants in the water.PACl compounds contain supplemental hydroxide (OH)- ions which cause lower pH depression and alkalinity impacts during coagulation processes. PACl products generally do not require the addition of acid or base to control the pH within the natural water range.

PACl has less toxicity potential than sulfate-containing alum. Sulfate loading from alum can be a concern because of the link of sulfur cycling in wetland environments with microbially-mediated methylation of mercury. Methyl mercury is toxic, and once formed can bioaccumulate among trophic levels.

PACl is synthetic polymers designed for coagulation based on optimum charge neutralization and bridge binding. Precipitates formed by alum and ferric salt application are amorphous hydroxides and the exact characteristics of those products and the efficiency of the chemicals used are dependent upon a number of variables such as temperature and mixing energy.[55]

Engineered polymers like PACl tend to be more efficient and robust with regard to achieving coagulation goals because their precipitates are less variable. PACl has very good performance over a broad dosing range, and inorganic/organic polymer blends appear to be the most difficult to overdose. Even so, however, more optimal dosing of PACl improves coagulant performance. Thus, more optimal dosing of PACl leads to more efficient coagulant utilization and better performance.

Compared to alum and ferric chloride, the performance of PACl with regard to P and turbidity removal is minimally affected by changes in temperature, mixing regimes, storm water quality and dose. The performance of PACl is also less affected by different rapid or slow mixing specifications.

The advantages of PACl include: (1) it reduces total dissolved P as well as alum; (2) it typically produces less flocculate than alum; (3) it has a broader range of pH over which they are effective (i.e., optimally effective for waters with pH ranging from 6 – 8, and relatively effective up to a pH of 10); and (4) it reportedly has ten to twenty times less dissolved aluminum in solution than does alum.

The disadvantages of PACl are:

(1) it is substantially more expensive than alum, and distributors are more limited; (2) it is available in a wide range of products that are not all equal in performance, which requires an understanding and assessment of their properties in the context of water treatment goals in order to select a preferred PACl coagulants; and (3) it is reported that some PACl, are less effective at removing P than alum. [55]

Table.(2-4) The features and positives and negatives to (PACl) [47]

Coagulant type	Features	Positive	Negative
Polyaluminum chlorides (PACl)	Made by partially neutralized (prehydrolyzed) aluminium chloride.Enhanced amounts of high-charged, moderate-molar-mass hydrolysis species e.g. Al_{13}.	Less temperature and pH dependent than alum salts. Smaller alkalinity consumption. Better NOM removal capacity than alum in many cases. Lower dose requirement and less sludge produced. Lower residual aluminium in treated water.	The effectiveness of coagulant is significantly affected by coagulant hydrolysis species speciation. Preformed Al species are stable and cannot be further hydrolysed during coagulation. Might not be so efficient in removing HMM and highly hydrophobic NOM..

2:2:3 <u>Advantages and Disadvantages of Coagulants</u>

The most common coagulants used in drinking water treatment are liquid alum and ferric chloride..etc.Table(2-5) lists alternative inorganic coagulants as well as their advantages and disadvantages.[56]

Table.(2-5) Advantages and disadvantages of coagulants
(US Army Corp of Engineers, 2001)

Name	Advantages	Disadvantages
Aluminum Sulfate (Alum) $Al_2(SO_4)_3.18H_2O$	Easy to handle and apply; most commonly used; produces less sludge than	Adds dissolved solids (salts) to water; effective over a limited pH range.

	lime; most effective between pH 6.5 and 7.5	
Sodium Aluminate $Na_2Al_2O_4$	Effective in hard waters; small dose-ages usually needed	Often used with alum; high cost; ineffective in soft waters
Polyaluminum Chloride (PACl) $Al_{13}(OH)_{20}(SO_4)_2.Cl_{15}$	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 needs 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 $FeSO_4.7H_2O$	Not as pH sensitive as lime	Adds dissolved solids (salts) to water; usually needs 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

Chapter Two : Theoretical Background

2:3 Electro Kinetic Properties of Particles

It is noticed from the above coagulation is a process where the repulsive potential of electrical double layers of colloids is reduced in such a way that micro-particles can be produced. These micro-particles collide with each other and form larger structures (flocs) in the flocculation process. Historically, coagulation has been employed in water treatment to decrease turbidity and color and to remove pathogens. However, optimum conditions for turbidity or color removal are not always the same as those for NOM removal. In the baseline coagulation, the coagulation conditions are optimized for turbidity removal, whereas optimized coagulation refers to dose and pH conditions is where optimized especially for organic matter reduction. Enhanced coagulation refers to conditions where more (excess) coagulant is used than would be needed for baseline coagulation, with also changes in pH, order of chemical addition or use of alternative coagulant chemicals, thus gaining more efficient TOC and thus NOM is removed.[47]

Colloidal particles comprise a large portion of the turbidity-producing substances in waters.Examples of colloidal particles include color compounds, clays, microscopic organisms and organic matter from decaying vegetation or municipal wastes. Colloidal dispersions are stable in water, as they poses a large surface area relative to their

weight. Therefore, gravitational forces alone will not remove colloids during sedimentation. Effective removal of these colloidal dispersions is greatly impacted by the electro kinetic properties on the surface of the colloids. Each colloid carries a similar electrical charge that produces a force of mutual electrostatic repulsion between adjacent particles. If the charge is high enough, the colloids will remain discrete and in suspension. The addition of coagulants or polymers reduces or eliminates this charge and colloids will begin to agglomerate and settle out of suspension or form interconnected matrices which can then be removed during filtration. This agglomeration causes the characteristics of the suspension to change by creating new particle viscosity, settling rates and effective size properties for the colloids. Colloids are classified as hydrophobic (resistant to water bonding) or hydrophilic (affinity for water bonding). Hydrophilic colloids are stable because their attraction to water molecules will overcome the slight charge characteristic they possess. This attraction makes hydrophilic colloids difficult to remove from suspension. Examples of hydrophilic colloids include soaps and detergents, soluble starches, soluble proteins and blood serum. On the other hand, hydrophobic particles are dependent on electrical charge for their stability in suspension. The bulk of inorganic and organic matter in a turbid raw water is of this type.[57]

2:3:1 Electrical Potential

Most colloidal particles in water are negatively charged as a result of differences in electrical potential between the water and the particle phases. This charge is due to an unequal distribution of ions over the particle surface and the surrounding solution. The charge on a colloidal particle can be controlled by modifying characteristics of the water which holds the particles in suspension. Modifications include changing the liquid's pH or changing the ionic species in solution. Another, more direct technique is to use surface-active agents, such as coagulants, that directly adsorb to the surface of the colloid and change its characteristics.[57]

2:3:2 Electrical Double Layer Theory

The double layer model explains the ionic environment surrounding a charged colloid and explains how the repulsive forces are set up around a colloid. Figure (2-8 a and b) illustrates the resulting colloidal state. A single negative colloid will initially attract some of the positive ions in the solution to form a firmly attached layer around the surface of the colloid, known as the Stern layer. additional positive ions are still attracted by the negative colloid, but are also repelled by the Stern layer as well as by other positively charged ions trying to get close to the negatively charged colloid. This constant attraction and repulsion results in the formation of a diffuse layer of charged ions surrounding the colloid and Stern layers. The diffused layer can be visualized as a charged atmosphere surrounding the colloid. Together, the attached positively charged ions in the Stern layer and the charged atmosphere in the diffuse layer are referred to as the double layer.[57]

The charge is a maximum at the particle surface and decreases with distance from the surface. The thickness of this layer depends on the type and concentration of ions in solution . When two similar colloidal particles with similar primary charge approach each other, their diffused layers begin to interact.The similar primary charges they

possess result in repulsive forces. The closer the particles approach, the stronger the repulsive forces. Repulsive forces which keep particles from aggregating are counteracted to some degree by an attractive force termed Van der Waals attraction. All colloidal particles possess this attractive force regardless of charge and composition. As Van der Waals forces tend to have relatively weak attractions, the force decreases rapidly with an increasing distance between particles.[57]

The balance of the two opposing forces, electrostatic repulsion and Van der Waals attraction, explains why some colloidal systems agglomerate while others do not. As particles with similar charge approach one another, the repulsive electrostatic forces increase to keep them separated. However, if they can be brought sufficiently close together to get past this energy barrier, the attractive Van der Waals force will predominate, and the particles will remain together. The random motion of colloids caused by the constant collisions with water molecules, termed Brownian Movement, will bring particles in close proximity and aggregation may occur. However, the addition of coagulant and polymers is typically used to lower the energy barriers between particles and provide efficient agglomerations for settling.[57]

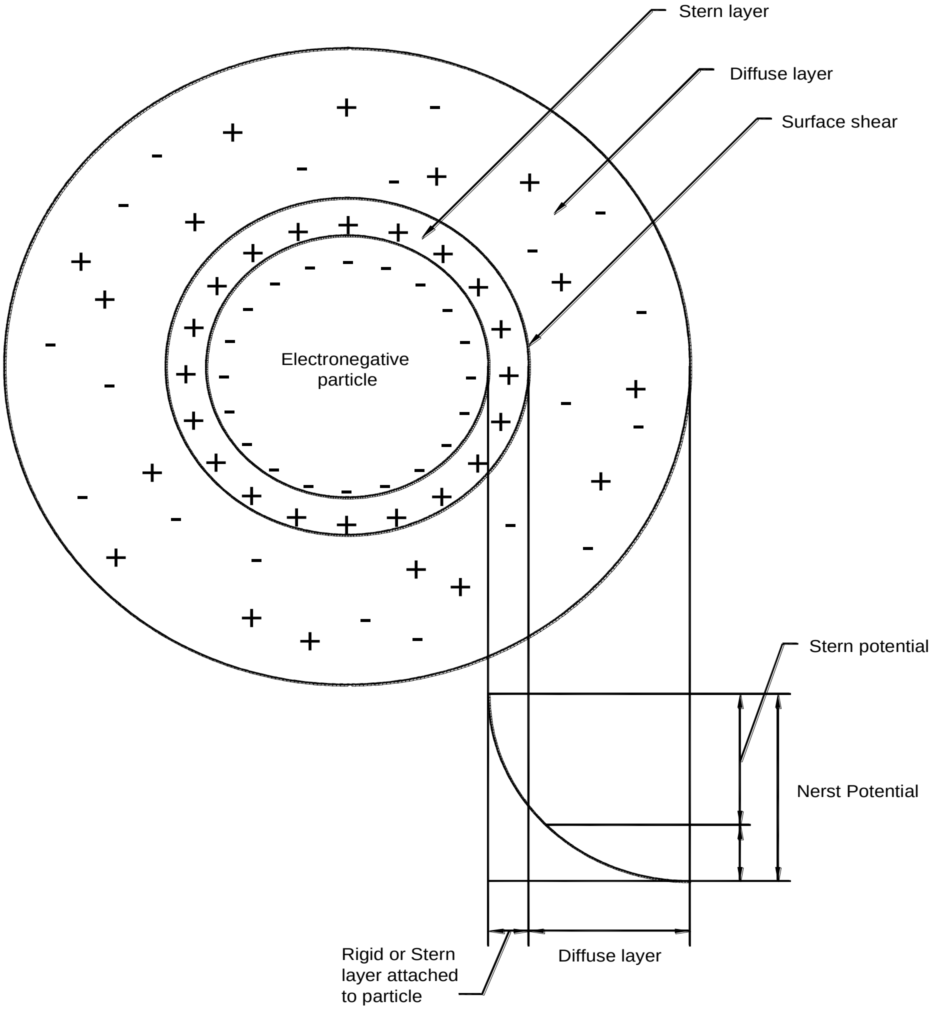


Figure (2-8 a). Double layer theory (Guoy-Stern Colloidal Model)

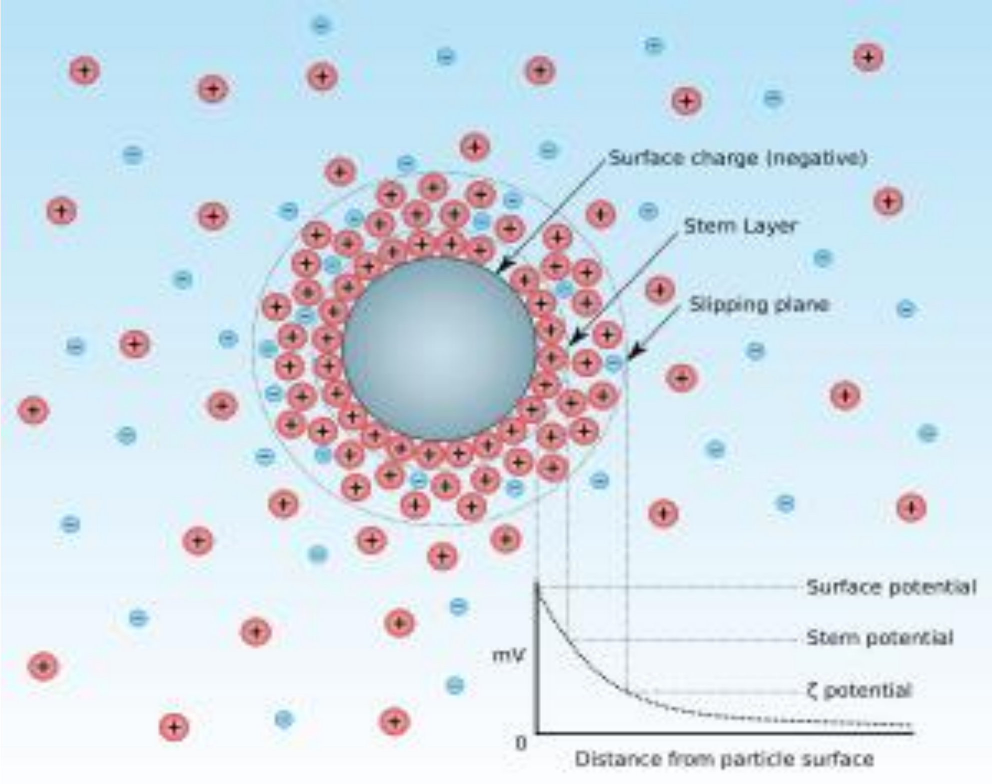


Figure (2-8 b). Double layer theory (Guoy-Stern Colloidal Model)

To explain the electrokinetic processes in the immediate vicinity of the charged surface of the particles, the potential model will be explained. As can be seen from Figure. (2-8), this model is based on a spherical solid particle with a certain base potential, also called the “Nernst potential”. Counter-ions are adsorbed directly at the phase boundary due to Coulomb or Van der Waals forces. This mono- to bimolecular layer is called the “Stern layer”. This is the followed by the “diffuse double layer”, in the region of which negative ions are also present. By addition of counter-ions into the surroundings of the negatively charged surface, the electrical potential is weakened continuously. The force of repulsion or attraction predominates, depending on the ionic strength. The situation of the resultant force is an important criterion for coagulation and/or agglomeration of the suspended solid particles in the waste water. It is possible to determine the potential relationships at the solids/liquids shear
surface by electrokinetic measurements.The result of these measurements is the "Zeta potential”. In case of flocculation surface forces have to overcome these electrical forces. That means for inert particles, the best flocculation can be achieved for a Zeta potential that is zero.[58]

One of the requirements for removing or inactivating particulate contaminants as well as biological pathogens is to enhance the coagulation capabilities in water treatment facilities. Impurities in wastewater are primarily anionic, negatively charged species. Cationic (positively charged) additives have been developed to neutralize these charged anionic contaminants. By introducing optimal quantities of cationic additives that will cause the system to reach a neutral charge, it is possible to improve the efficiency of removing contaminants via sedimentation or filtration. ζ potential measurements provide a tool to quantify the optimal concentration of the oppositely charged additives required to enhance coagulation of contaminants from a water supply.

In the case of water treatment applications, charged particles should always be considered to be stable particles and therefore undesirable. The objective is to reduce the surface charge of the contaminants by treating them with oppositely charged additives to the point where the charge of the contaminating material is close to zero or neutral. These oppositely charged additives (cationic flocculating agents in this case) will adsorb to the surface of the (anionic) particles. When the particle/ additive complexes have little or no charge, the ζ potential approaches zero (termed point of zero charge) and there will then be no repulsive forces maintaining the stability of these particles in the dispersion leading to their aggregation and sedimentation.[59]

Zeta potential measurements, used in conjunction with turbidity measurements, provide a method to (1) monitor the particle stability which varies over time and (2) identify maximum aggregation and flocculation conditions.

Since raw (input) water quality can vary significantly with time due to many factors previously discussed, these measurements provide a method to continuously assess process parameters that indicate water quality so that a quick response can be made when high raw water variability occurs. It should be noted that the case study below presents results from a full-scale water treatment facility (FCWTF) and provides a year's worth of data monitoring the Zeta potential, turbidity measurements, and alum dosage in the water treatment process.[59]

2:3:3 Zeta Potential

The Stern layer is considered to be rigidly attached to the colloid, while the diffuse layer is a dynamic layer of charged particles. The ***Nernst Potential*** is the measurement of voltage (in the order of millivolts) in the diffuse layer. The potential is a maximum at the Stern layer and drops exponentially through the diffuse layer. The ***zeta potential*** is the electrical potential representing the difference in voltage between the surface of the diffuse layer and the water. It is important to know the magnitude of the zeta potential, as it represents the strength of the repulsion between colloid particles and the distance which must be overcome to bring the particles together.

The primary charge on a colloid cannot be measured directly. However, the Zeta potential can be computed from measurements of particle movement within an electrical field (electrophoretic mobility). Therefore, the Zeta potential, ζ, is defined by the equation: [57]

$$\zeta = \frac{4\pi\delta q}{D}$$

where q = charge of the particle

δ = thickness of the zone of influence of the charge on the particle

D = dielectric constant of the liquid

Zeta potential measurements can be made using a high-quality stereoscopic microscope to observe colloidal particles inside an electrophoresis cell .
An electric field is created across the cell and charged particles move within the field. Their velocity and direction are then related to the Zeta potential. Measurements of Zeta potential can give an indication of the effectiveness of added electrolytes in lowering the energy barrier between colloids, and can direct the optimization of coagulant dose in water treatment. [60]
The destabilization of colloids is accomplished by the reduction of the zeta potential with coagulants such as alum, ferric chloride and/or cationic polymers. Once the charge is reduced or eliminated, no repulsive forces exist. Gentle agitation in a flocculation basin will cause numerous, successful colloid collisions.[57]

2:3:4 Principle Of Measuring Zeta Potential

Zeta potential was calculated using Smouluchowski equation as early as 1903. The sample to be measured is dispersed in suitable liquid phase and placed in the path of a beam of laser light. A pair of electrodes is introduced into the sample. Charged particles in the sample will move under the influence of an electric field applied across the electrodes. The direction of the motion indicates the sign of the charge on the particles:
negatively charged particles will gravitate towards the positive electrode and vice versa. The velocity of particles, per unit electric field, can be measured and is called the electrophoretic mobility (u). Thus,

$$u = v/ E \qquad m^{2}.V^{-1}.s^{-1} = m.\ s^{-1}/ V.m^{-1}$$

where v is the particle velocity and E is applied electric field strength. The equation used for converting the observed mobilities into effective electrokinetic potential depends upon the value of dimensionless quantity ‘ka’ in which ‘a’ is the radius of the particle (assumed spherical) and ‘k’ is the quantity given by

$$k = \frac{4\pi e^{2}\sum nZ^{2}}{\varepsilon\ KT}$$

where, e = electronic charge,
ε = electrical permittivity of the solvent ,
K = Boltzman constant
T = absolute temperature

From the expression given, k = 1 × 106 cm at 25^{o}C in water ($\varepsilon/\varepsilon o = 78$) containing 1 mM of 1:1 electrolyte. Values of ‘k’ at other concentration follow by simple proportion. If ‘ka’ > 200 it will be usually sufficiently accurate to use the Smoluchowski formula which in the original unrationalised form is $u = \varepsilon\zeta/4\pi\eta$, where ε is the permittivity of the suspending medium and η is the viscosity. Typical units would be to have u in micron per second under one volt per cm, in which case for water at $25^{o}C$, Zeta potential in mV would be given by: [61]

$$\zeta = 12.83\ u \quad .$$

2:3:5 Zeta Potential and Suspension Stability

Knowledge of Zeta potential can be used to predict and control the stability of colloidal suspensions or emulsions. The greater the Zeta potential, more likely the suspension is to remain in stable form.

The Zeta potential is very much dependent on the pH of the suspension. A plot of Zeta potential vs pH is called an iso-electric curve. The pH for which Zeta potential is zero, is called the ‘iso-electric point’ or ‘point of zero charge’ (PZC).[62]

It has been already pointed out that if the particle surface charge is strong, it results in significant mutual repulsion prohibiting the particles from coming closer. If the charge on these particles is reduced to zero, these repulsion forces are eliminated. Gentle mechanical agitation will then create numerous particle collisions instead of ‘near misses’. The forces of adhesion, cohesion, and mechanical interlocking by ‘polymer bridging’ will result in agglomeration.[62]

Turbidity (representing suspended particulate matters) consists of two distinct fractions: a coarse fraction (1 mm to 1 mm) and a colloid fraction (1 mm to $10A^{o}$). The coarse fraction may be successfully removed by conventional coagulation/flocculation treatment, but the colloid fraction cannot. As the floc formed during coagulation/flocculation is also of the same charge as that of the particles before treatment, a mutual repulsion exists between the floc particles and the colloids, and these repelling forces prevent the colloid from making permanent contact with the floc, regardless of agitation. Such colloids can effectively be removed by lowering the Zeta potential of both the floc and colloid to a value of approximately zero plus or minus 5 mV. This can be done by simultaneously employing the proper dosage of an inorganic coagulant coupled with an appropriate organic polyelectrolyte. The water treatment plant at Waterford, New York, is believed to be the first to demonstrate purification of water based on the principle of continuously controlled Zeta potential.[61]

2:4 Flocculation Mechanism

As discussed earlier, the charge structure surrounding the particles is called the electrical double layer, which, for convenience is divided into Stern, and Gouy-Chapman layers. The former is the initial layer of adsorbed ions and molecules located at the particle surface. The charge presented to the solution at the Stern layer naturally attracts a diffuse layer of free ions with a net different opposite charge, i.e., the Gouy-Chapman layer. For particles to make contact and aggregate, the potential at the stern layer must be overcome. Unfortunately, this potential, which is of interest in determining particle stability, cannot be measured experimentally. In order to cause the particles of a stable dispersion to flocculate, it is necessary to provide enough kinetic energy to particles to overcome the potential energy barrier. Alternatively, the barrier can be eliminated by surface-charge neutralization. This may be accomplished either by double layer compression (charge neutralization mechanism) or adsorption of flocculent onto the particle surface (bridging mechanism).[9]

2:4:1 Charge Neutralization Mechanism

Charge neutralization by double layer compression is accomplished when flocculation is effective through an increase in solution ionic strength. The expression for the Debye-Huckel length, as derived from the Gouy-Chapman model of the electric double layer, is given as

$K = (4 \times 10^{-3} e^2 NI/\varepsilon kT)^{1/2}$

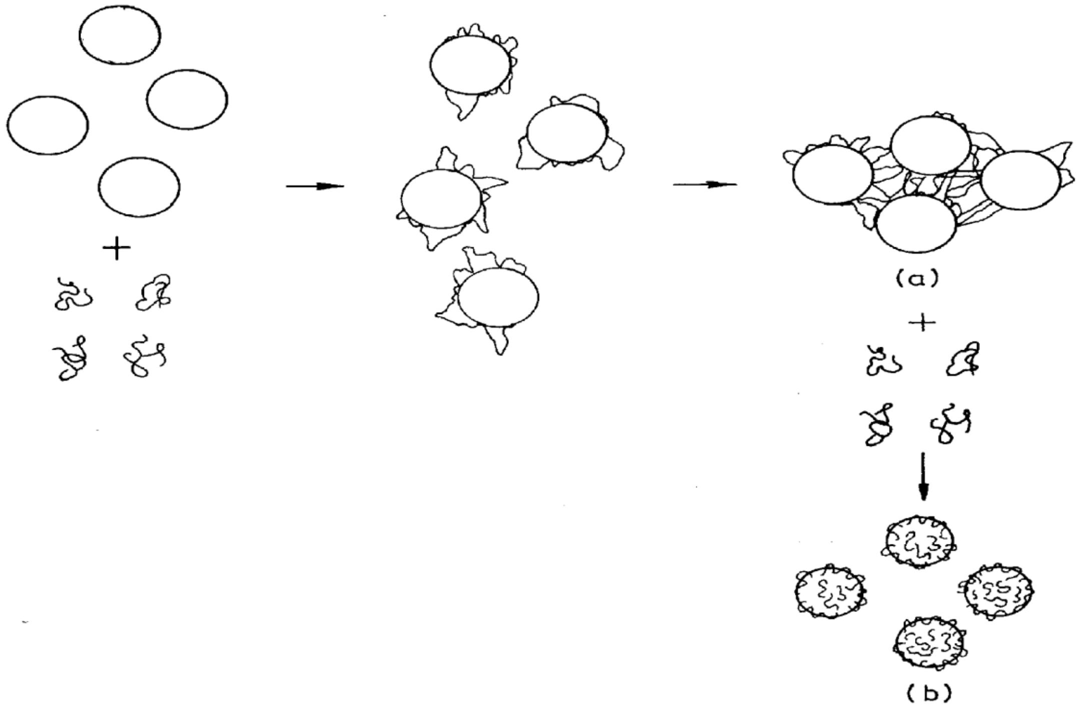
where, e = electronic charge
N = Avogadro's number
I = ionic strength
ε = electrical permittivity of the solvent
k = Boltzman constant, and
T = absolute temperature

Since the double layer thickness is approximated by $1/K$, increase in ionic strength (I) decreases the thickness of the double layer . This compression allows the approach of the colloidal particles to where short-range attractive forces predominate over electrostatic repulsive forces. In the presence of hydrolysable metal ions such as Al^{3+} and Fe^{3+} or polyelectrolyte's of opposite charge to colloid surface , the charge is neutralized by adsorption of these species onto the particle surface. An important case of this is the flocculation of negative colloidal particles with cationic polymers. In fact, in many cases, the action of cationic polymers can be explained in terms of their strong adsorption on negatively charged particles and consequent reduction of double layer repulsion, allowing aggregation to occur. It may be noted that the most effective cationic flocculants are often those of high charge density, with molecular weight playing only a minor role. [63].[64]

That charge neutralization in fact occurs is reported by comparison of Zeta-potential measurements with flocculation results, when it is found that optimum flocculation occurs at the point of total charge neutralization. For flocculation to proceed with pure charge-neutralization mechanism, it is natural to expect that Zeta-potential will be zero at the point of optimum flocculation. But in practice, the zeta potential tends to become negative at optimum flocculation with an increase in molecular weight of the polyelectrolyte. This is because increasing molecular weight of flocculent favors bridging relative to charge neutralization mechanism. Thus in any system where flocculation is affected by the addition of electrolyte or oppositely charged polyelectrolyte, it is likely that some degree of charge neutralization occurs, the extent depending upon the system characteristics. [63].[64]

2:4:2 Bridging Mechanism

Long chain polymers when added in small dosage to a suspension of colloidal particles, adsorb onto them in such a manner that an individual chain can become attached to two or more particles thus “bridging” them together (Figure.2-9a).But interestingly this phenomenon is observed up to a particular optimum polymer dosage beyond which flocculation diminishes, a process being known as steric stabilization (2-9b). The essential requirements for polymer bridging are that there should be sufficient unoccupied particle surface for attachment of polymer segments from chains attached to other particles and that the polymer bridges should be of such an extent that they span the distance over which antiparticle repulsion prevails. Thus, at lower dosages, there is insufficient polymer to form adequate bridging links between particles. With excess polymer, there is no longer enough bare particle surface available for attachment of segments and the particles become destabilized, which may involve some steric repulsion. On average, bridging flocculation gives aggregates (flocs) which are much stronger than those produced by addition of salts (i.e., by reduction in electrical repulsion)Figure(2-10). However, such stronger flocs produced by the bridging mechanism may not reform once broken at high shear rates.[63]



Figure(2-9) (a) Schematic illustration of bridging flocculation and (b) restabilization by adsorbed polymer

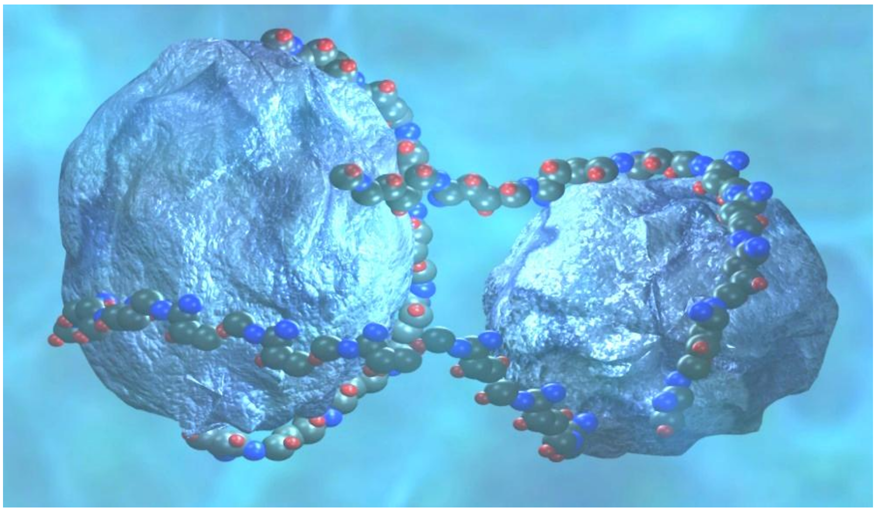


Figure (2-10) Polyelectrolyte chains on colloidal particle

Chapter Two : Theoretical Background

2:5 <u>Zeta Potential and Its Impact On The Water Treatment Applications</u>

Forms and different types of membranes are widely used in plants and desalination systems and water purification, and those kinds of reverse osmosis membranes (RO) , nano filter membranes (NF), (Ultra filter), (Micro filter). Such systems are used for desalination and filtering drinking water and purifying wastewater for re use . [12]

The desalination uses the natural properties of different types of membranes manufactured from some of the semi-permeable polymers which allow the passage of water but not the dissolved salt ions under the influence of hydraulic pressure, such as the situation in reverse osmosis membranes(RO).[13]

One of the main problems of these systems is blockages in the membranes used in water purification process with the passage of time because a layer sediments on the surface of these membranes.

One of the ways in which they can remove or prevent the formation of settles on the membrane is to control some of the physical properties of the membrane itself and the water that passes through the membrane, including the method of control by Zeta potential.[65]

The advantages of using Zeta potential in desalination systems and water filters containing the membranes and filters are the following : [66]

- remove and prevent debris found in the membranes.
- increase the life of the membranes and filters in the systems and minimize the impact of corrosion to certain chemicals.
- Zeta potential works to remove and prevent the formation of layer Albaeofflm (biological layer).
- reduce the cost of maintenance.
- improve the efficiency of the equipment.

2:6 High Voltage Capacitance Device

High Voltage Capacitance Device Zeta Rod™ .photo (2-4) (HVC) functions by inducing an alteration of the natural surface charge density of dielectric colloidal particles irrespective of the particle composition.

The installation of the HVC forms a cylindrical capacitor, much like those pictured in physics texts. Electrochemical dispersion of colloidal organic and inorganic particles, as well as microorganisms, has been practiced for many years to treat the water in small commercial and industrial water systems.[67]

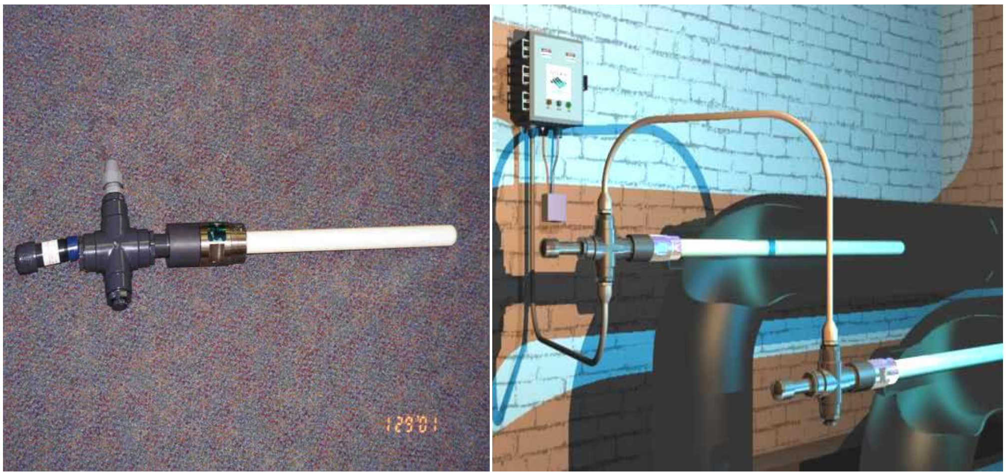


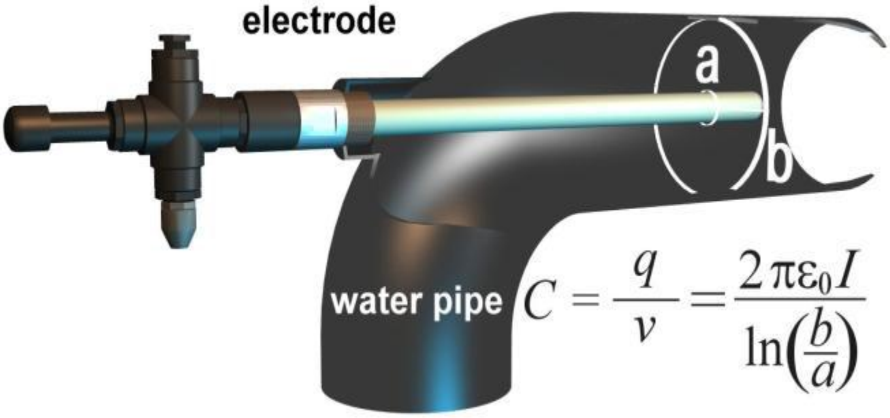
Photo (2-4) High voltage capacitance device.

The HVC has also shown it is effective as a means of preventing biofilm formation on RO membranes.[15]

The conducting surfaces are the metallic lining of the HVC and the metal of the pipes or vessels. The ceramic dielectric of the HVC establishes a static electric field within the piping system or the vessel. A direct current power supply charges the capacitor system. The field strength across the water is a function of charge voltage, system dimensions, and the dielectric constant of the water.[67]

At sufficiently elevated voltage, the field strength across the water influences the capacitive charge of the particle. The result is a sharp increase in the surface charge of all wetted surfaces .

The primary field of the HVC (potentials above about 5 kilovolts) extends for about 20 feet in either direction from the electrode surfaces. For a cylindrical capacitor, the total capacitance of the system is given by the equation Figure (2-11) :

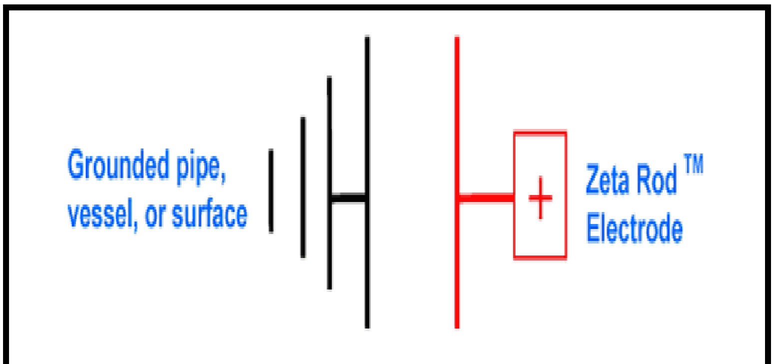


Figure(2-11) Total Capacitance - High Voltage Capacitance Device

Where
C = capacitance (units)
q = charge
v = voltage
I = length
a, b = radii of the electrodes forming the capacitor . [67]

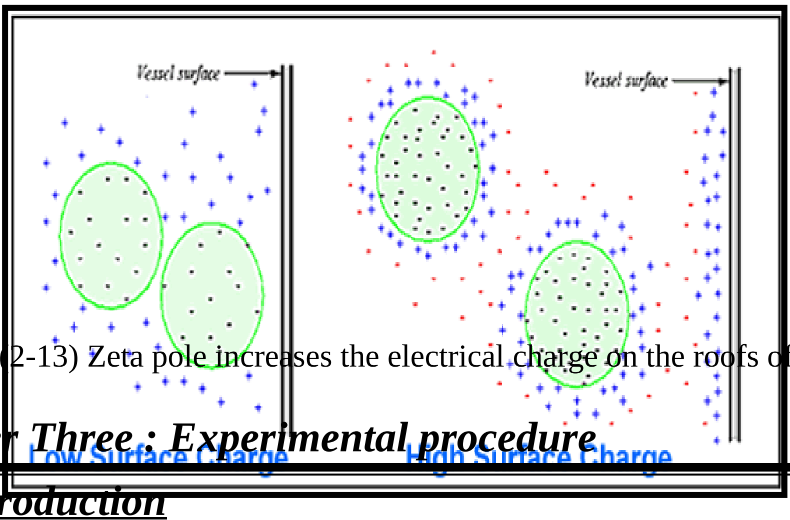
Dispersion technique uses colloidal particles in electrochemical way on an experimental level for many years for water treatment in small systems as well as commercial and industrial systems.

The basic principles of electrostatic dispersion in aqueous systems include, capacitor, double layer, surface charge, insulation properties of colloids, The ionic strength of the solution. to produce a dispersion to induce electro static, capacitor created by the introduction of cylindrical electrode insulated and locked in a metal tube or tank as shown in Figure (2-11) .The function of this system is to change the charge colloid particle surfaces above the natural state . Conductive paint inside a ceramic electrode is placed on capacitor plate. And ceramic insulating pole prevents the flow of current to the other plate . Ground, or the opposite surface the capacitor is represented by metal pipes or reservoirs. as shown in Figure(2-12). Capacitors are charged continuously to very high voltage limits (30 - 35) kV through power supply (AC) . [68]



Figure(2-12) High voltage capacitance device

It is seen from the above that Zeta potential technical can be employed for finding solutions in an easy and inexpensive way to the problem of plaque formation in the membranes used in water purification through preventing the formation of these deposits of all kinds (and by working on particle dispersion according as shown in Figure(2-13), thus increasing the efficiency and life of the membranes that are of great importance in water desalination plants. As a result of this Zeta potential can be used to monitor and control the electrical stability of the colloidal solution or emulsion.



Figure(2-13) Zeta pole increases the electrical charge on the roofs of colloidal particles and dissonance

Chapter Three : Experimental procedure

3:1 Introduction

This chapter describes all the equipment and instruments used in the test and the chemicals used and the method of preparation of solutions . The experimental part will consist of two phases , The first compares between the effectiveness of the three coagulants used in this study They are (alum, ferric chloride, poly aluminum chloride) . and gives selection of the optimal dose of coagulant efficient.

The second phase is the using of Zeta Rod technology (high voltage capacitor) and studies the behavior of the coagulation process and compares it with the effectiveness of more efficient coagulant for the coagulation process.

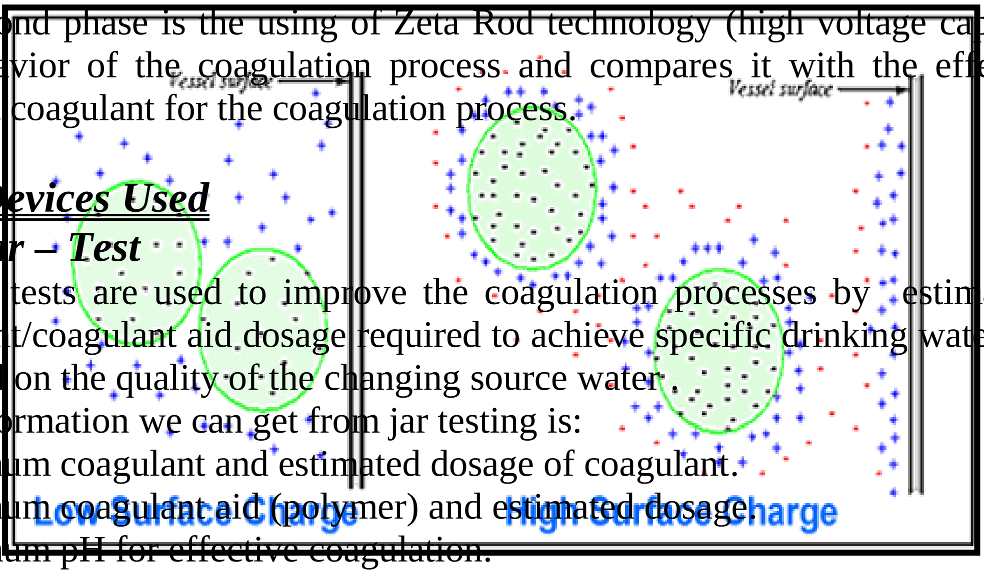
3:2 Devices Used

1 - Jar – Test

Jar tests are used to improve the coagulation processes by estimating the optimum coagulant/coagulant aid dosage required to achieve specific drinking water quality goals that is based on the quality of the changing source water .

The information we can get from jar testing is:

- Optimum coagulant and estimated dosage of coagulant.
- Optimum coagulant aid (polymer) and estimated dosage.
- Optimum pH for effective coagulation.
- Sequence of chemical application.
- Time and energy mixing ideal
- Evaluation of floc settle ability. [69]



For over 50 years, the jar test has been the standard technique used to optimize the addition of coagulants and flocculants used in the wastewater and drinking water treatment industry. The Standard Practice for Coagulation_/Flocculation Jar Test of Water, ASTM D 2035, was first approved in 1980 and reapproved in 1999. The scope of this practice ‘‘covers a general procedure for the evaluation of a treatment to reduce dissolved, suspended, colloidal, and non settleable matter from water by chemical coagulation_/flocculation, followed by gravity settling.’’ This standard is utilized to provide a special technique to systematically evaluate the variables normally encountered in the coagulation_/flocculation process. Since coagulant interactions are very complex, laboratory studies are used to determine the optimal dosage, duration, and intensity of mixing and flocculation. The coagulation_/flocculation process consists of three distinct steps. First, the coagulant is added to the effluent water and rapid and high intensity mixing is initiated. The objective is to obtain complete mixing of the coagulant with the wastewater to maximize the effectiveness of destabilization of colloidal particles and initiate coagulation. Critical parameters for this step are the time and the intensity (velocity gradient, G) or speed of mixing. The velocity gradient is a measure of the mixing energy and can be determined for a specific piece of equipment or specified in the engineering design analysis. Second, the suspension is slowly stirred to increase contact between coagulating particles and to facilitate the development of large flocs. Again, the flocculation duration and intensity are critical parameters. For example too high intensity can break up the aggregate floc. Third, mixing is terminated and the floc is allowed to settle.[35]

One of the most common pieces of bench test apparatus found in water treatment laboratories to identify potential coagulation-flocculation conditions in liquid suspensions is the jar test apparatus. The test is used to confirm the preferred chemicals and also to identify the best concentrations. The batch-test consists of using six identical jars containing the same volume and concentration of feed, which are charged simultaneously with six different doses of a potentially effective flocculant. The six jars can be stirred simultaneously at known speeds. The treated feed samples are mixed rapidly and then slowly and then allowed to settle. These three stages correspond respectively to the fundamental processes of dispersing the flocculant onto the particle surface, gently sweeping the colloidal particles together to form large flocs, and allowing the flocks to settle. At the end of the settling period, test samples are drawn from the jars and turbidity of supernatant liquid is measured. A plot of turbidity against flocculant dose gives an indication of the optimum dosage (i.e. the minimum amount required to give acceptable clarification). The criteria thus obtained from a bench jar test are the quality of resultant floc and the clarity of the supernatant liquid after settling. The design of the full-scale plant process is then done based on the bench-scale selection of chemicals and their concentrations.[9]

The jar test consists of six glass beakers which are usually cylindrical in shapes and have capacity of 1 liter and iron rotary paddle is placed inside each one. The mixing and time of movement can be controlled quickly. The beakers are placed on the illuminated base which helps observation of the floc formation and settling characteristics. as photo (3-1) below shows :

Photo (3-1) Jar test device

2- A device for measuring turbidity (Turbidirect)

As photo (3-2) below shows **:**

Photo (3-2) A device for measuring turbidity

3- Sensitive balance (precisa Swiss Manufacturing)

(Mix 220 gm—Min 0.0001 gm)

As shown in photo (3-3) below:

Photo (3-3) Sensitive balance

4- pH and temperature measuring device shown in photo (3- 4) below:

Photo (3-4) pH meter

5-Spectrophotometer (colorimeter **DR5000** (**HACH**)) shown in photo (3-5) below:

A device used to examine solutions concentrations by measuring the absorbance at a specific wavelength(522 nm) [71]. The mechanics of optical spectrometer is designed to measure the absorption of radiation energy where the colored material to be identified or react with the compound to produce a colored compound so that the sensitivity of the test is accurate and thus low concentrations of substance is estimated .

Photo (3-5) Spectrophotometer

6- Zeta Rod device (HVC)

Variable voltages (5 kV – 25 kV) DC shown as in the photo (3-6 a) below**:**

Fixed voltages (30 kV DC) as shown photo (3-6 b) below:

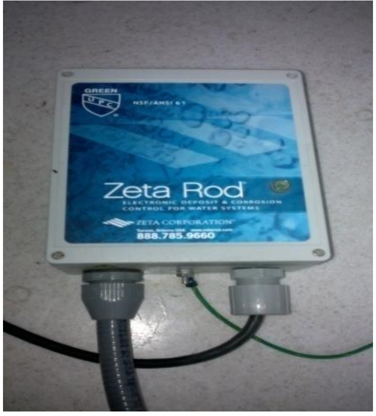


Photo (3-6 a) Photo (3-6 b)

3:3 Tools and Materials Used

- Standard volumetric glassware ***(500ml,250ml)***
- Pipettes **.**

Photo (3-7) Tools used

- Aluminum Sulfate **$Al_2(SO_4).16H_2O$** Assy 98% .
  (BDH Chemical Ltd. Pool England)
- Ferric Chloride **$FeCl_3$** Assy 98% .
  (HIMEDIA . India)
- Poly aluminum Chloride (PACl) $Al_2Cl_{12}(OH)_{24}$.
  (HIMEDIA . India)
- Kaolin powder .
- Distilled water .
- Steel cylindrical container of 1 liter.

3:4 Procedure

A) The First Phase

It is to select optimum coagulant and estimate dosage of coagulant.

1 - Chemical Preparation

Stock solutions of the coagulants and flocculants were used to improve the ease of handling and measuring, and ensure good mixing in the jars and the Treatment chemicals used in full-scale operations, Stock Solution cannot be used for jar test purposes at their delivered strength because of the difficulty of measuring the volumes needed for various dosages Therefore a diluted solution of known strength must be generated for jar test purposes . This solution is known as a STOCK SOLUTION.

Chemical Dosing of Jar Test Beakers: ……….[69]

Amount of Stock Chemical (mL) Solution =

$$\frac{\text{Dosage Desired (mg/L) X Beaker Volume (L) X 1000 (mL/L)}}{\text{Stock Chemical Concentration (mg/L)}}$$

Example Calculation of a required dosage of 20 mg/L or 20 ppm

$$\text{Amount of Stock Chemical solution (mL)} = \frac{20\ (\text{mg/L}) \times 1\ \text{L} \times 1000\ (\text{mL/L})}{10{,}000\ \text{mg/L}}$$

$$= 2\ \text{ml}$$

This means that to get a dose of 20 ppm withdrawn 2 ml of Stock solution. (*In water treatment chemical dosage is expressed as parts per million (ppm) or mg/L*)

1- 2.5 g of alum is dissolved in250 ml of distilled water to obtain the Stock solution. Each 1 ml of it is equal to 10 ppm and the same process is repeated for ferric chloride.
2- 0.5 g of (PACl) is dissolved in500 ml of distilled water to obtain the Stock solution. Each 1 ml of it is equal to 1 ppm.
3- Samples of synthetic turbid water were prepared by adding a kaolin powder to tap water in all coagulation experiments at a rate of 1 g per 1 liter to obtain turbidity level of approx 500 NTU and so on for other required levels of water turbidity(150,50 NTU) at each step.

2 - Jar-test

Experiments were run following a modified version of the standard jar test procedure**[Standard Practice for Coagulation-Flocculation Jar Test of Water (Designation: D 2035 – 80 (Reapproved 2003)]** . [70]

And the coagulation_ flocculation tests were carried out following the standard practice for coagulation_ flocculation testing of water to evaluate the chemicals, dosages, and conditions required to achieve optimum result**.**

To do the experiment, these steps are followed**:**

● put 1 liter of raw water(which was prepared)in each beaker of six beakers of a jar-test device **(Lovibond com. German Manufacturing . Mix 300 Rpm)** Then they were placed in allocated places in the machine.

● Specific dosages of coagulant was added according to the required ratios to each beaker at predetermined dosage levels and sequence **.** ● Start the multiple stirrer operating at the “flash mix" speed of approximately 120 rpm . Flash mix is for approximately 1 min after the additions of chemicals. and the flash mix time and speed (rpm)were recorded.

● Reduce the speed as necessary to the minimum required to keep floc particles uniformly suspended throughout the “slow mix” period. Slow mix is for 20 min at a rate of 20 rpm.

● After the slow mix period, withdraw the paddles and observe settling of floc particles In most cases this time will be that required for the particles to settle to the bottom of the beaker; however, in some cases there may be interfering convection currents. If so, the recorded settling time should be
that at which the unsettled or residual particles appear to be moving equally upward and downward.

● After 15 min of settling, record the appearance of floc on the beaker bottom. Record the sample temperature. By means of a pipet, withdraw an adequate sample volume of supernatant liquid from the jar at a point one half of the depth of the sample, After that turbidity is measured by turibidmeter device **(Lovibond com. German Manufacturing . Mix 800 NTU**),pH and temperature by pH meter device**(InoLab (WTW)**.**German Manufacturing)** and the weight of aluminum remaining in each sample for alum and for poly aluminum chloride . The remaining weight of iron of the ferric chloride per sample drawn was measured using the Spectrophotometer devices**(DR5000 Hach company 2005)**.[71]

- And repeat the same process for of the three coagulants and for the three levels of water turbidity(500,150,50 NTU) and record results .

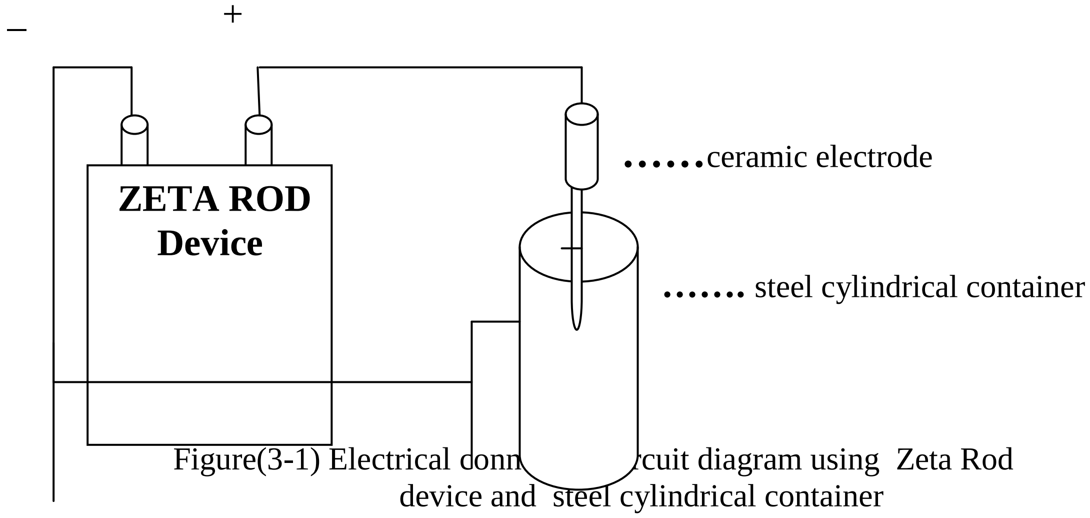
B) The Second Phase

Zeta Rod technology(high voltage capacitor)is used to study the behavior of the coagulation process and compare it with the effectiveness of more efficient coagulant in the coagulation process that has been achieved in the first phase.

At this stage, the optimum dose of coagulant was found out for the better removing turbidity by the results of the first stage. The technique is used where high-voltage capacitors use a Zeta Rod**(Zeta Corporation U.S. Manufacturing)**.

To do the second phase follow the following steps:

- Repeat the same steps used in the first stage of the preparation of concentrations. as well as the action steps in jar test but use best coagulants chosen from the results of the first phase .
- In the jar test steps here use steel cylindrical container Instead of glass beaker so as to complete the electrical circuit according to the chart shown below:



Figure(3-1) Electrical conn[illegible]cuit diagram using Zeta Rod device and steel cylindrical container

- After the completion of the sedimentation process (15 minutes) place the ceramic electrode into the steel container which contains troubled waters . The change voltages of 5 kV to 25 kV DC gradually re-process each dose, and the same steps followed for the other use of the device at fixed voltage of (30 kV DC) are per dose as the photo(3-8) below shows:

Photo(3-8) The ceramic electrode into the steel container

- Upon completion of each step after applying different voltages withdraw a sample of the water and measure the turbidity and pH and record the results against each dose of coagulant used .
- Relationship diagrams are drawn between the voltages used, and turbidity according to the results to study the behavior of the effect of voltages applied in the process of coagulation.

Chapter Four : Results and Discussions

This chapter deals with the results and discusses the impact of three coagulants used *(alum, ferric chloride and PACl)* on the troubled waters and compare their efficiency in the removal of turbidity . This chapter presents the results and discusses of the impact of the use of Zeta Rod technology on the process of coagulation and flocculation in water treatment processes.

4:1 The First Phase:

Comparison between Chemical Coagulants

Once the jar tests were completed , the final results of turbidity and pH values were measured. In addition, weight of aluminum remaining in the samples, as well as iron were measured and compared with the weights in the potions added.

This result can be seen in Table (4-1) for alum , Table (4-2)for ferric chloride and Table(4-3) for PACl and Figures(4-1)and(4-2) in the high level of turbidity(500NTU).

- (Alum) $Al_2(SO_4)_3.16H_2O$ Assay 98% Mol.Wt. 821.96

Wt.Al in 15 ppm from Alum = X

X /2Al=15mg/mol.wt. (Alum) → X /2 *26.98=15mg/821.96

X =0.98 mg

Table (4-1) Results when using alum at high turbidity levels (500 NTU)

Beaker Number	Stock Chemical to Add (ml)	Desired Dose (mg/l) or ppm	Weight of (Al)mg Before Process	Turbidity NTU	pH	Temp.ċ	Weight of (Al)mg After Process
1	0	0	0	146	7.34	15.9	0
2	1.5	15	0.98	38.9	7.31	15.9	0.02
3	3.0	30	1.96	31.1	7.20	15.9	0.19
4	4.5	45	2.94	16.2	7.10	16	0.2
5	6.0	60	3.92	10	7.00	16	0.25
6	7.5	75	4.9	8.2	6.90	16.2	0.3

- $FeCl_3$ Assay 98 % Mol.wt 162.21

Wt. Fe in 15 ppm from $FeCl_3$ = X

X /Fe = 15mg/mol.wt. ($FeCl_3$) → X /55.5=15mg/162.21

X =5.13 mg

Table (4-2) Results when using FeCl3 at high turbidity levels (500 NTU)

Beaker Number	Stock Chemical to Add (ml)	Desired Dose (mg/l) or ppm	Weight of (Fe)mg Before Process	Turbidity NTU	pH	Temp.ċ	Weight of (Fe)mg After Process
1	0	0	0	172	7.60	18.4	0
2	1.5	15	5.13	46.1	7.28	18.4	0.01
3	3.0	30	10.27	33.3	7. 03	18.4	0.2
4	4.5	45	15.39	22.1	6.81	18.4	0.33
5	6.0	60	20.5	13.1	6.65	18.5	0.36
6	7.5	75	25.65	6.8	6.47	18.4	0.44

●

PACl ($Al_{12}Cl_{12}(OH)_{24}$) Mol.Wt.1156.56

Wt.Al in 15 ppm from(PACl) = X

X /12Al=15mg/mol.wt. (PACl) → X /12 *26.98=15mg/1156.56

X =4.19 mg

Beaker Number	Stock Chemical to Add (ml)	Desired Dose (mg/l) or ppm	Weight of (Al)mg Before Process	Turbidity	ph	Temp.ċ	Weight of (Al) After Process

1	**0**	**0**	**0**	**211**	**7.65**	**17.2**	**0**
2	**15**	**15**	**4.19**	**28.6**	**7.62**	**17.2**	**0**
3	**30**	**30**	**8.39**	**13.4**	**7.63**	**17.2**	**0**
4	**45**	**45**	**12.59**	**8.43**	**7.61**	**17.3**	**0**
5	**60**	**60**	**16.79**	**6.91**	**7.60**	**17.3**	**0**
6	**75**	**75**	**20.99**	**4.55**	**7.59**	**17.4**	**0**

Table (4-3) Results when using PACl at high turbidity levels (500 NTU)

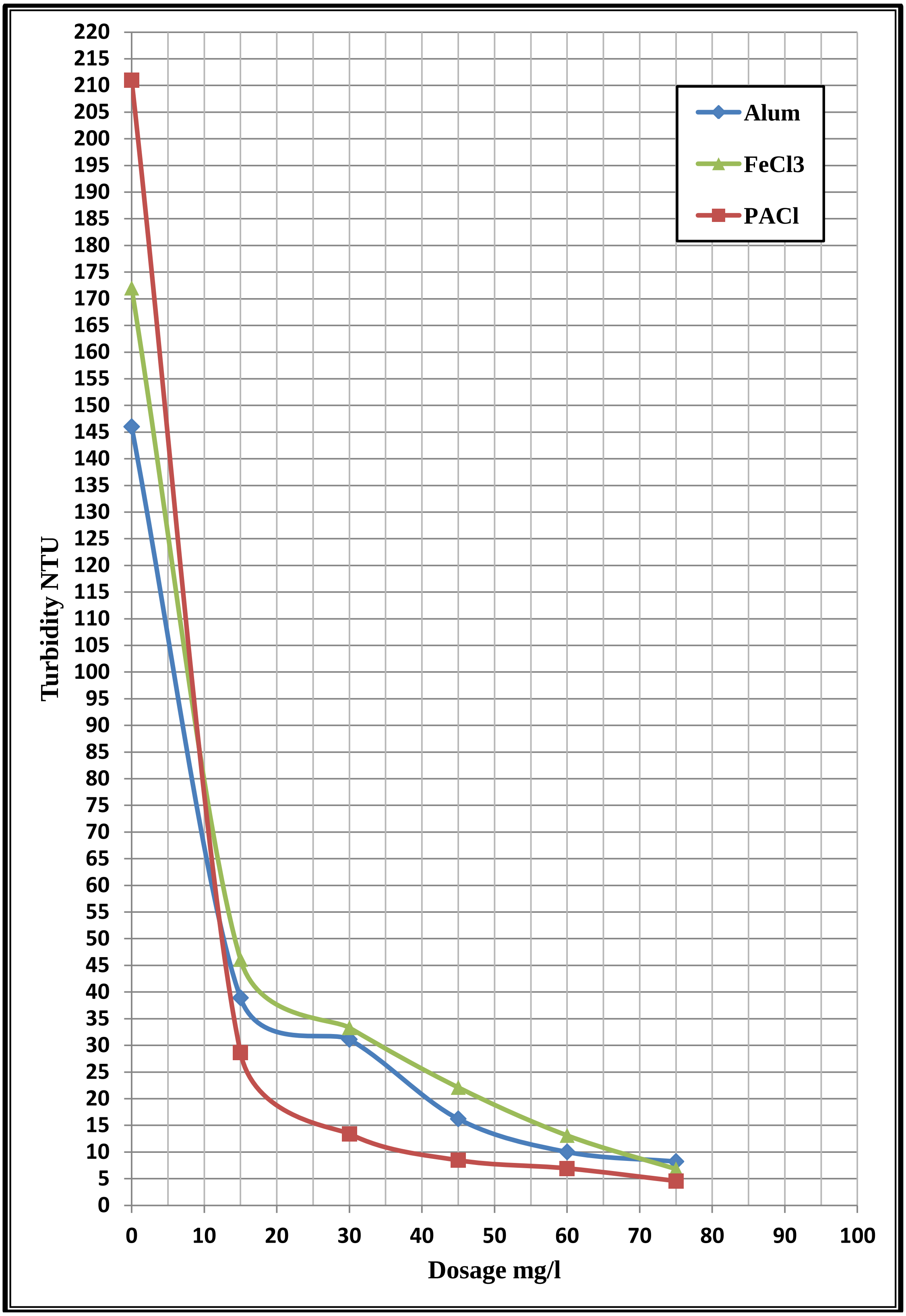


Figure (4-1) Relation between the desired doses and turbidity(500NTU)

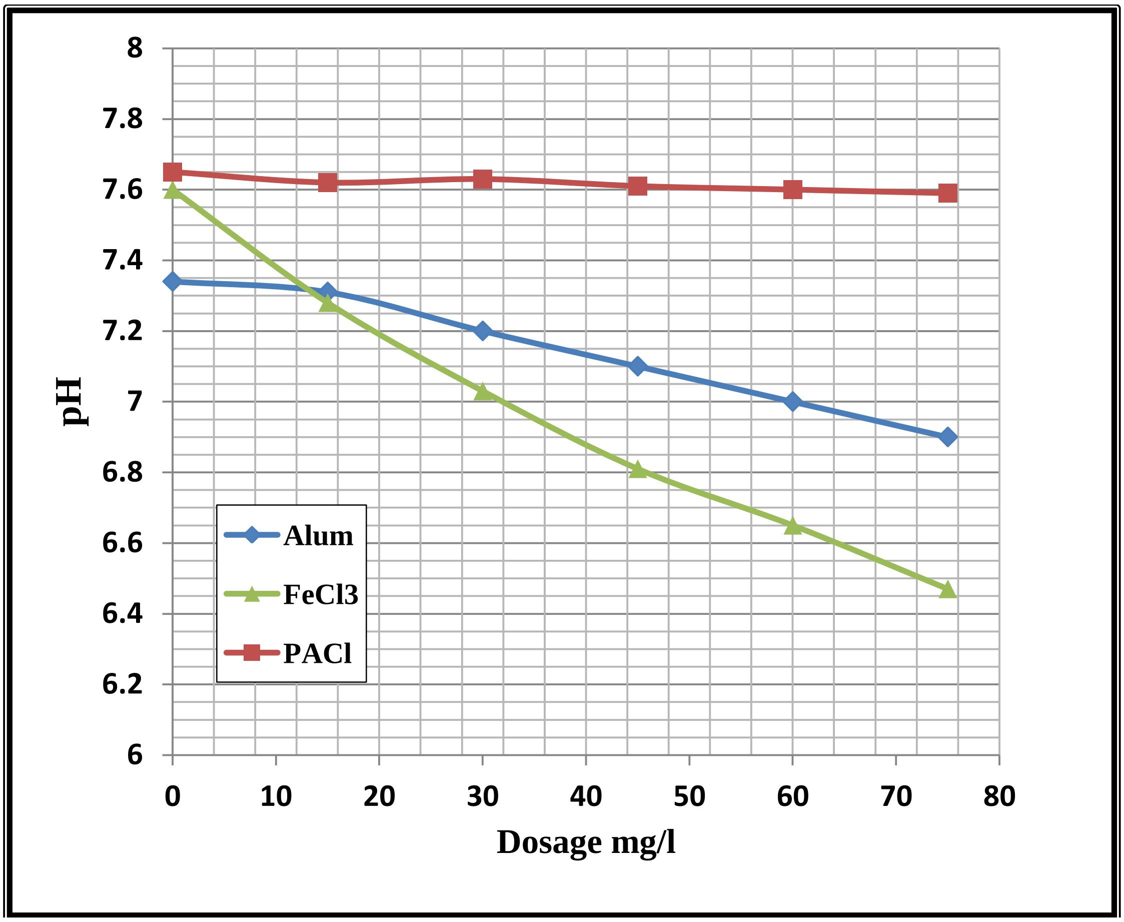


Figure (4-2) Relation between the desired doses and pH

From the tables above(4-1)(4-2)(4-3)and Figure(4-1), in case of not adding any dose of coagulant(beaker no.1) notes that turbidity has decreased to some extent as a result of steps of mixing fast and slow as suspended particles begin to gather with each other as shown photo (4-1a,b). As for the other samples note that there is a difference in the efficiency of removal of turbidity of the water.

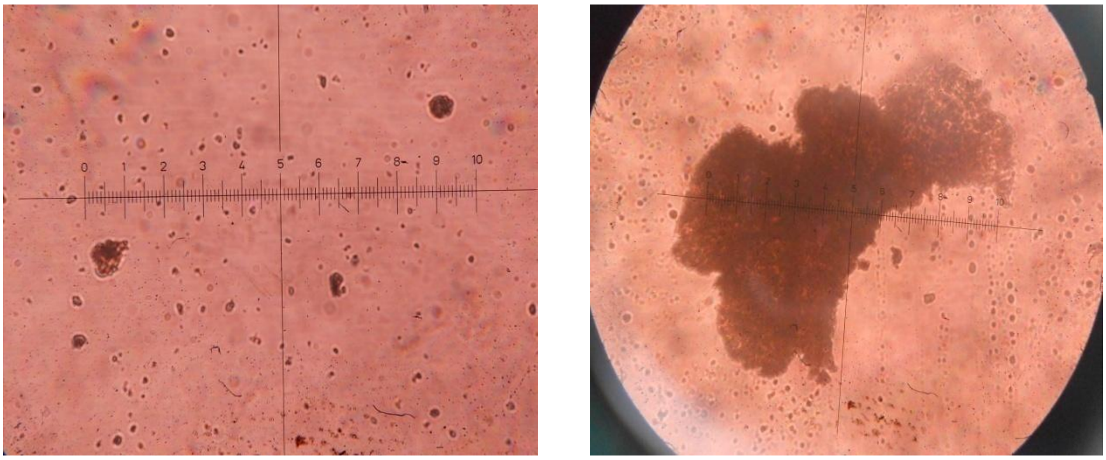


Photo (4-1) a. Kaolin particles before **b. Kaolin Particles after the**

the treatment **the treatment**

Also the tables above show the dose of 75mg/l of alums able to reduce the turbidity of the water to 8.2NTU ,that is, efficiency removal of (98.5%) and the same dose of ferric chloride reduces turbidity to 6.8NTU (98.8%) while the dose of 75mg/l of Poly aluminum chloride reduces turbidity of the water to 4.55 NTU(99.2%).

$E_r = 1- (T_{out}/ T_{in}) \times 100$
where E_r : efficiency removal
T_{out} : final turbidity level
T_{in} : the initial level of turbidity …….[3]

Also, there is a high weights of aluminum and iron remaining in water after treatment when using alum, ferric chloride(0.3mg),(044mg) respectively and these weights increase with increasing concentrations of doses.Otherwise when (PACl) does not have any residual amount of aluminum in the water.

pH values have changed significantly when using alum and ferric chloride ,the solution tends to be acidic, with more dose concentrations of additive coagulants. pH values have changed insignificantly and very little when using (PACl) as shown in Figure(4-2).

Chapter Four : Results and Discussions

The results when using the jar test at intermediate levels of turbidity (150NTU) of the water are shown in the Table (4-4) for alum , Table (4-5)for ferric chloride and Table(4-6) for PACl and Figures(4-3)and (4-4).

- (Alum) $Al_2(SO_4)_3.16H_2O$ Assay 98% Mol.Wt. 821.96

Table (4-4) Results when using alum at intermediate levels of turbidity(150 NTU)

Beaker Number	Stock Chemical to Add (ml)	Desired Dose (mg/l) or ppm	Weight of (Al)mg Before Process	Turbidity	pH	Temp.ċ	Weight of (Al)mg After Process
1	0	0	0	126	7.86	15.4	0
2	1.5	15	0.98	43.2	7.68	15.5	0.3
3	3.0	30	1.96	27.8	7.45	15.5	0.6
4	4.5	45	2.94	16	7.27	15.6	0.3
5	6.0	60	3.92	10.2	7.14	15.7	0.2
6	7.5	75	4.9	6.5	7.01	15.7	0.1

- $FeCl_3$ Assay 98 % Mol.wt 162.21

Table (4-5)Results when using FeCl3 at intermediate levels of turbidity(150NTU)

Beaker Number	Stock Chemical to Add (ml)	Desired Dose (mg/l) or ppm	Weight of (Fe)mg Before Process	Turbidity	pH	Temp.ċ	Weight of (Fe)mg After Process
1	0	0	0	128	7.95	19	0
2	1.5	15	5.13	35.4	7.7	18.8	0.24
3	3.0	30	10.27	22.6	7. 5	18.8	0.25
4	4.5	45	15.39	13.4	7.32	18.5	0.25
5	6.0	60	20.5	6.0	7.10	18.7	0.27
6	7.5	75	25.65	3.8	7.04	19	0.29

- PACl ($Al_{12}Cl_{12}(OH)_{24}$) Mol.Wt.1156.56

Table (4-6)Results when using PACl at intermediate levels of turbidity(150 NTU)

Beaker Number	Stock Chemical to Add (ml)	Desired Dose (mg/l) or ppm	Weight of (Al)mg Before Process	Turbidity	pH	Temp.ċ	Weight of (Al)mg After Process
1	0	0	0	120	7.81	16.4	0
2	15	15	4.19	13.8	7.78	16.5	0
3	30	30	8.39	6.76	7.73	16.6	0
4	45	45	12.59	3.63	7.63	16.7	0
5	60	60	16.79	2.25	7.58	16.8	0
6	75	75	20.99	1.14	7.61	17.0	0

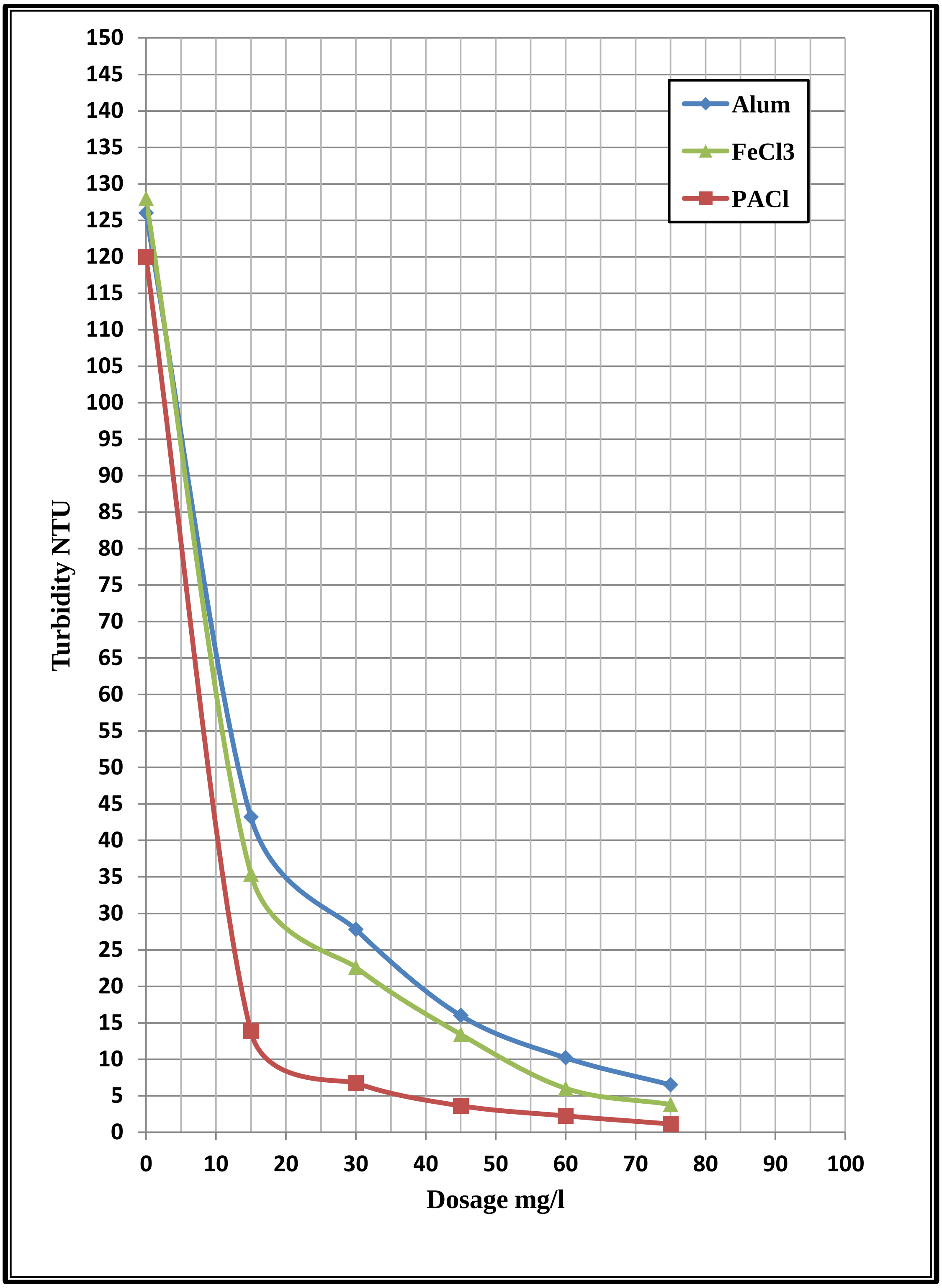


Figure (4-3) Relation between the desired doses and turbidity(150NTU)

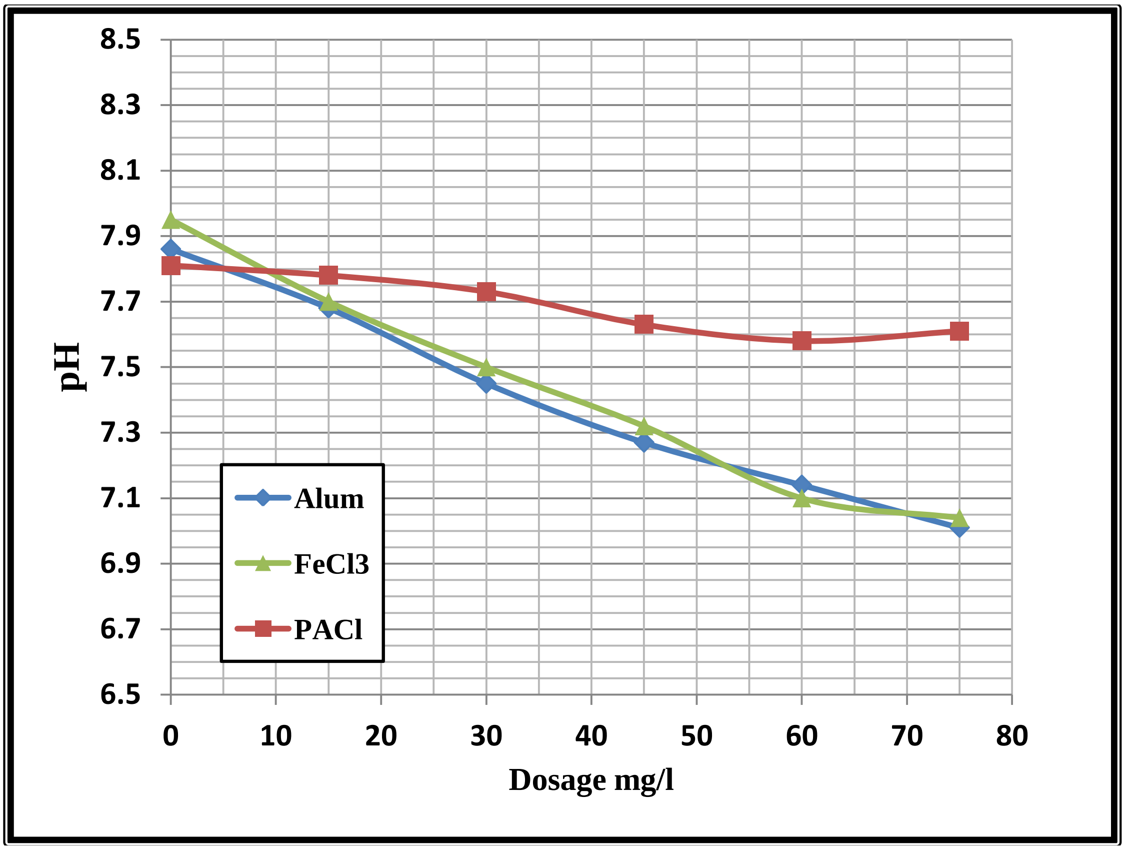


Figure (4-4) Relation between the desired doses and pH

At the levels of medium turbidity the results also show that the efficiency of coagulant poly aluminum chloride compared with that of alum and ferric chloride. From the Tables(4-4)and(4-5)(4-6) and Figure(4-3), it is seen a dose of 75mg/l reduces the turbidity of the water from 150NTU to 1.14NTU and the removal efficiency is (99.24%) While the same dose of alum and ferric chloride has reduced the turbidity of the water to 6.5NTU and 3.8NTU and efficiently remove is (95.6%),(97.4) respectively under the same operating conditions of the test. There is 0.6 mg of aluminum remaining in the water when using a dose of 30 mg/l of alum but there is no residual amount of aluminum when using any dose of PACl.

Figure (4-4)shows that there is a noticeable change in pH values when using alum and ferric chloride, while the change in pH values when using PACl is slight and unnoticeable .

In the jar tests low levels of water turbidity(50NTU) results are similar to the previous results of the high and medium levels of turbidity. This can be seen in Tablets(4-7),(4-8),(4-9) and Figure (4-5).

- (Alum) $Al_2(SO_4)_3.16H_2O$ Assay 98% Mol.Wt. 821.96

Beaker Number	Stock Chemical to Add (ml)	Desired Dose (mg/l) or ppm	Weight of (Al)mg Before Process	Turbidity NTU	pH	Temp.ċ	Weight of (Al)mg After Process
1	0	0	0	48.3	7.83	18.3	0

2	**1.5**	**15**	**0.98**	**16.5**	**7.56**	**18.3**	**0.190**
3	**3.0**	**30**	**1.96**	**10.6**	**7.38**	**18.3**	**0.113**
4	**4.5**	**45**	**2.94**	**7.09**	**7.22**	**18.3**	**0.10**
5	**6.0**	**60**	**3.92**	**4.69**	**7.10**	**18.4**	**0.12**
6	**7.5**	**75**	**4.9**	**3.21**	**6.98**	**18.4**	**0.113**

Table (4-7) Results when using alum at low levels of turbidity(50 NTU)

- $FeCl_3$ Assay 98 % Mol.wt 162.21

Table (4-8) Results when using FeCl3 at low levels of turbidity(50 NTU)

Beaker Number	Stock Chemical to Add (ml)	Desired Dose (mg/l) or ppm	Weight of (Fe)mg Before Process	Turbidity	pH	Temp.ċ	Weight of (Fe)mg After Process
1	**0**	**0**	**0**	**48.7**	**7.62**	**18.2**	**0**
2	**1.5**	**15**	**5.13**	**16.2**	**7.30**	**18.0**	**0.8**
3	**3.0**	**30**	**10.27**	**12.9**	**7. 03**	**18.1**	**1.5**
4	**4.5**	**45**	**15.39**	**10.2**	**6.85**	**18.1**	**1.6**
5	**6.0**	**60**	**20.5**	**8.72**	**6.70**	**18.0**	**1.8**
6	**7.5**	**75**	**25.65**	**7.2**	**6.55**	**18.1**	**2**

- PACl ($Al_{12}Cl_{12}(OH)_{24}$) Mol.Wt.1156.56

Beaker Number	Stock Chemical to Add (ml)	Desired Dose (mg/l) or ppm	Weight of (Al)mg Before Process	Turbidity	pH	Temp.ċ	Weight of (Al)mg After Process

1	**0**	**0**	**0**	**43.4**	**7.88**	**18.8**	**0**
2	**15**	**15**	**4.19**	**7.81**	**7.88**	**18.7**	**0.08**
3	**30**	**30**	**8.39**	**4.92**	**7.87**	**18.7**	**0.073**
4	**45**	**45**	**12.59**	**2.26**	**7.83**	**18.8**	**0.039**
5	**60**	**60**	**16.79**	**1.63**	**7.79**	**18.8**	**o.o88**
6	**75**	**75**	**20.99**	**1.51**	**7.73**	**18.8**	**0.072**

Table (4-9)Results when using PACl at low levels of turbidity(50 NTU)

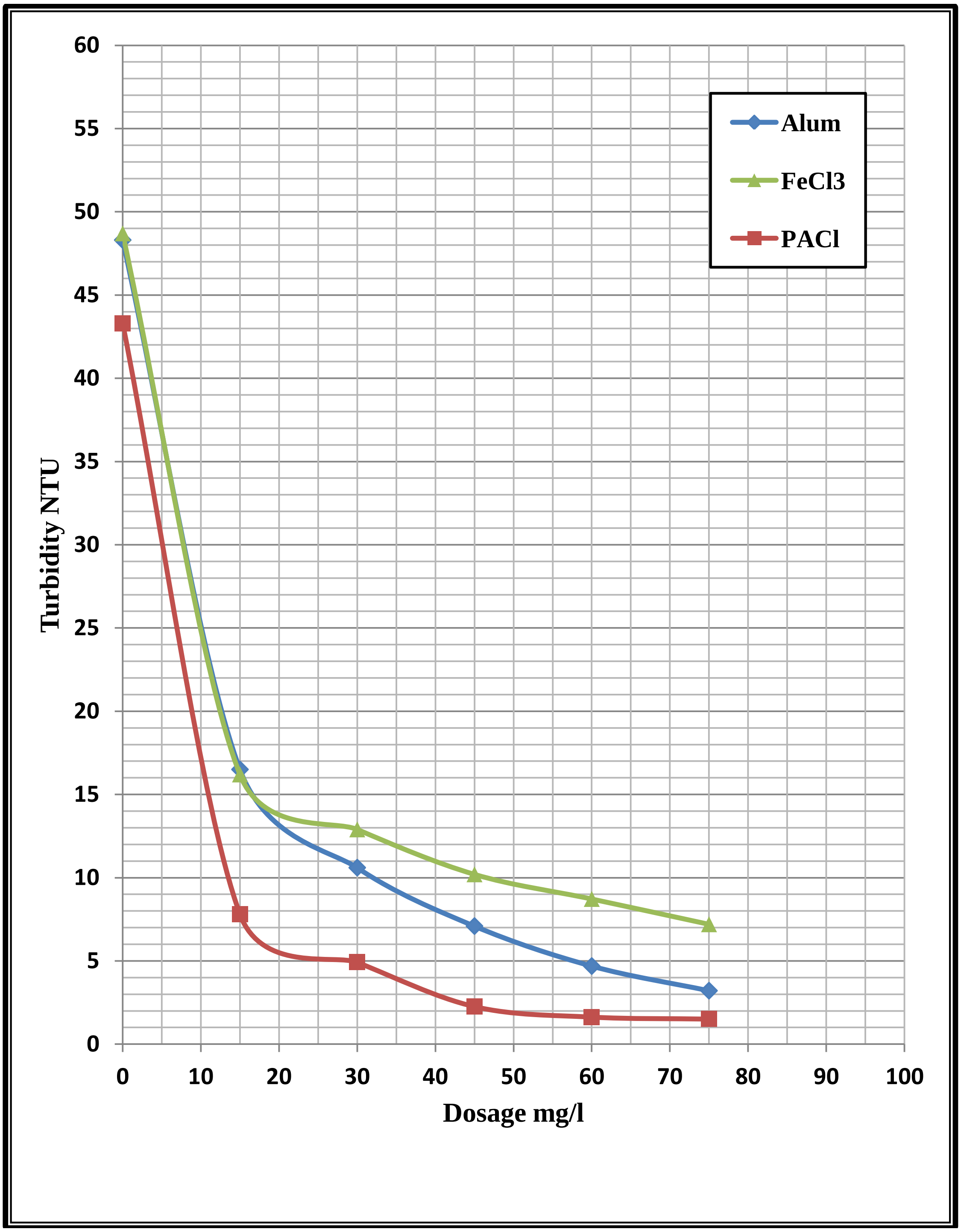


Figure (4-5) Relation between the desired doses and turbidity(50NTU)

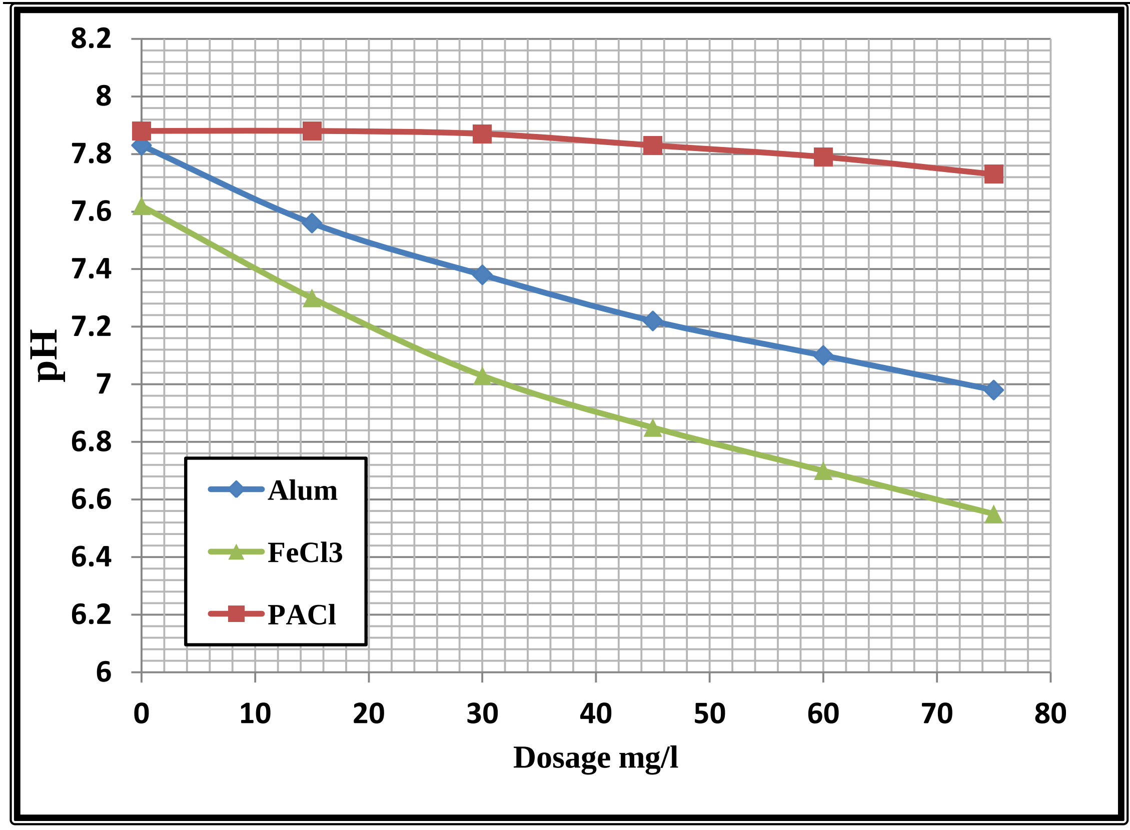


Figure (4-6) Relation between the desired doses and pH

Tables (4-7), (4-8) and (4-9) and Figure(4-5) show is coagulant PACl clearly superior to alum and ferric chloride in the removal of turbidity of the water at low levels because the water turbidity decreases from 50 to 1.51NTU using a dose of 75 mg/l of PACl (96.9%).While the turbidity decreases to 3.21NTU when using 75mg/l doses of alum(93.5%) and to 7.2NTU when using 75mg/l doses of ferric chloride (85.6%). Also, the remaining quantities of aluminum when using PACl are very small and insignificant while the remaining quantities of aluminum and iron in the water after treatment by alum and ferric chloride are great and noteworthy.

Figure (4-6) shows a very slight in pH values change at use of PACl doses while pH values when using doses of alum and ferric chloride shows a large and noticeable change. It should be noted that in all the previous results the three levels of turbidity, the temperature change when measured variables shows a very slight and intangible change.

Taking higher temperatures into consideration it is noticed chemical reactions occur more quickly and lower temperatures usually cause poor coagulation and flocculation.Therefore,at lower temperatures, coagulated water requires longer flocculation time.[69]

4:2 The Second Phase : Use of Zeta Rod Technique.

After the completion of the first phase and choosing the best coagulant from coagulants used. jar tests were made under the same previous conditions and the same operating doses for poly aluminum chloride and turbidity of the water level of (500NTU), but using a technique Zeta Rod(HVC) on recent sedimentation process (15 min). Using a variable voltage from 5kV to 30kV with each dose of PACl gives the results shown in tables and figures below:

a) Dose from the Stock Solution = 15 mg/l (PACl)

Table (4-10)Results when using 15 mg/l from PACl

voltage kV DC	Turbidity NTU	pH	Temp.c°
0	23.3	7.82	19.3
5	22.1	7.81	19.3
10	22.4	7.8	19.3
15	25.7	7.9	19.2
20	29.5	7.83	19.3
25	29.1	7.85	19.2
30	29.8	8.01	19.3

b) Dose from the Stock Solution = 30 mg/l (PACl)

Table (4-11)Results when using 30 mg/l from PACl

voltage kV DC	Turbidity NTU	pH	Temp.c°
0	12.6	7.8	19.2
5	16.1	7.79	19.2
10	18.2	7.82	19.1
15	16.9	7.81	19.1
20	17.2	7.80	19.2
25	17.1	7.82	19.0
30	18.5	7.99	19.1

c) Dose from the Stock Solution = 45 mg/l (PACl)

Table (4-12)Results when using 45 mg/l from PACl

voltage kV DC	Turbidity NTU	pH	Temp.c°
0	7.2	7.8	19.2
5	12.6	7.79	19.2

10	**11.8**	**7.80**	**19.5**
15	**11.7**	**7.82**	**19.3**
20	**12.**	**7.83**	**19.4**
25	**12.2**	**7.81**	**19.3**
30	**16**	**8.0**	**19.4**

d) Dose from the Stock Solution = 60 mg/l (PACl)

Table (4-13)Results when using 60 mg/l from PACl

voltage kV DC	Turbidity NTU	pH	Temp.c◦
0	**5.83**	**7.72**	**20.0**
5	**13.9**	**7.78**	**20.1**
10	**13.7**	**7.73**	**20.2**
15	**14.1**	**7.75**	**19.9**
20	**14.4**	**7.72**	**20.3**
25	**14.2**	**7.76**	**20.2**
30	**14.9**	**7.9**	**19.9**

e) Dose from the Stock Solution = 75 mg/l (PACl)

Table (4-14)Results when using 75 mg/l from PACl

voltage kV DC	Turbidity NTU	pH	Temp.c◦
0	**3.9**	**7.78**	**19.5**
5	**10.9**	**7.74**	**19.6**
10	**11**	**7.72**	**19.4**
15	**11.3**	**7.75**	**19.8**
20	**11.2**	**7.76**	**19.6**
25	**11.6**	**7.71**	**19.6**
30	**12.5**	**7.80**	**19.9**

f) Using a Zeta Rod variable voltages of (5 kV-30 kV) DC without using any coagulant and at jar-test.

Dose from the Stock Solution = 0

Table (4-15)Results without any dose from PACl

voltage kV DC	Turbidity NTU	pH	Temp.c◦

0	208	8.0	20.1
5	233	8.03	20.2
10	240	8.0	20.2
15	246	8.04	20.1
20	251	8.03	20.2
25	254	8.03	20.3
30	262	8.0	20.1

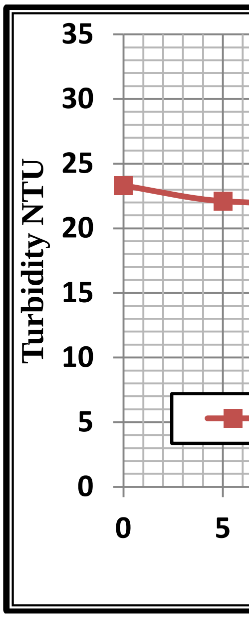


Figure (4-7) Relation between the voltages and turbidity at a dose of 15 mg/l of PACl

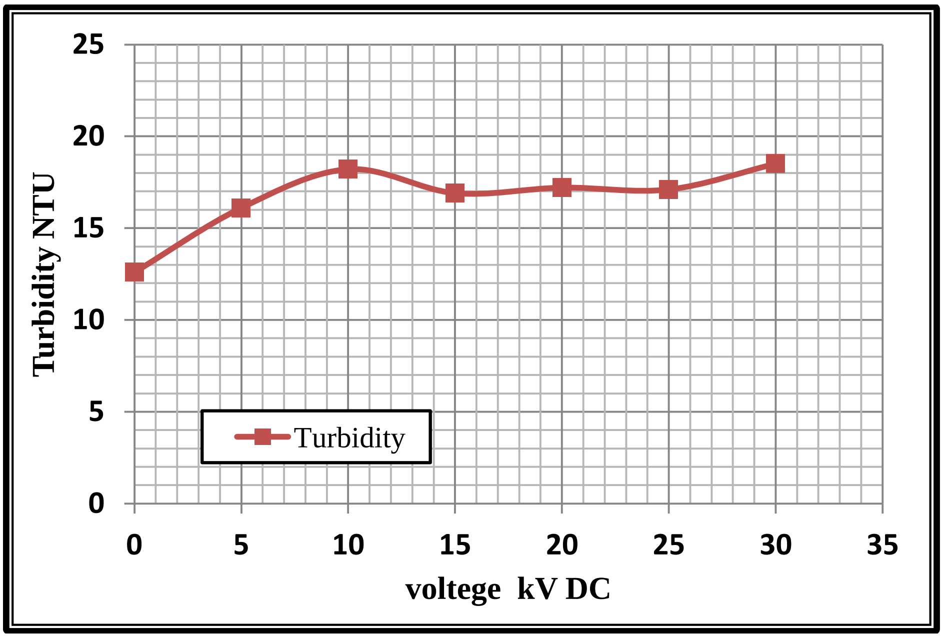


Figure (4-8) Relation between the voltages and turbidity at a dose of 30 mg/l of PACl

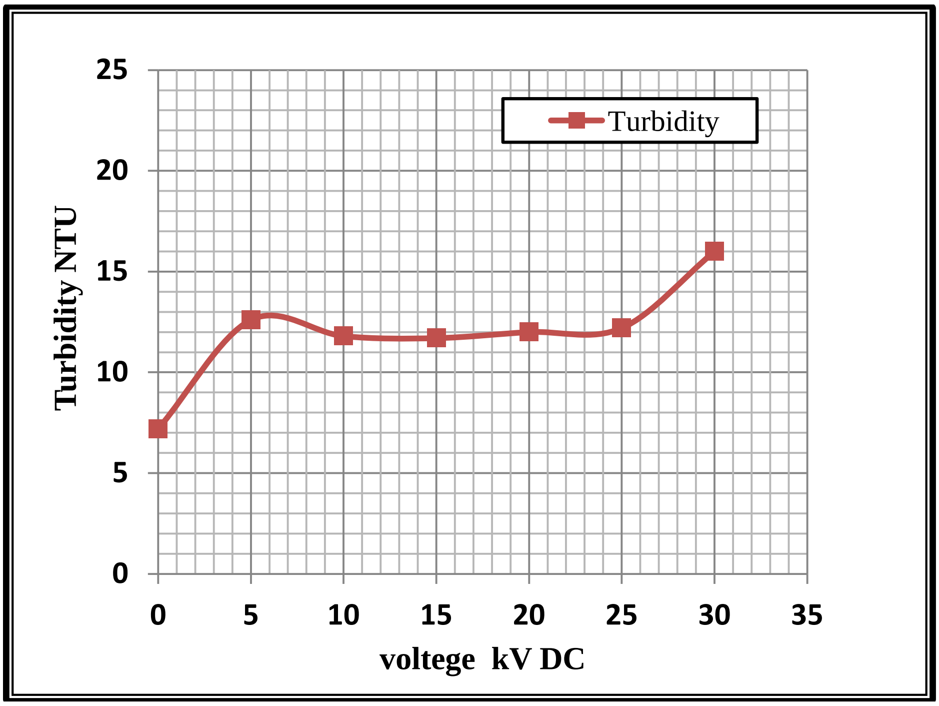


Figure (4-9) Relation between the voltages and turbidity at a dose of 45 mg/l of PACl

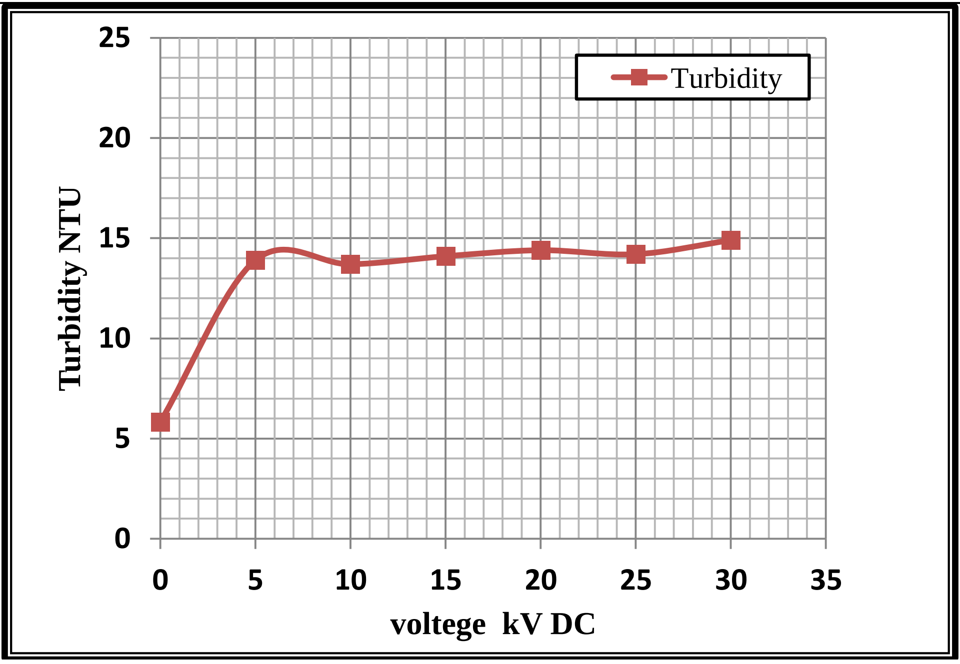


Figure (4-10) Relation between the voltages and turbidity at a dose of 60 mg/l of PACl

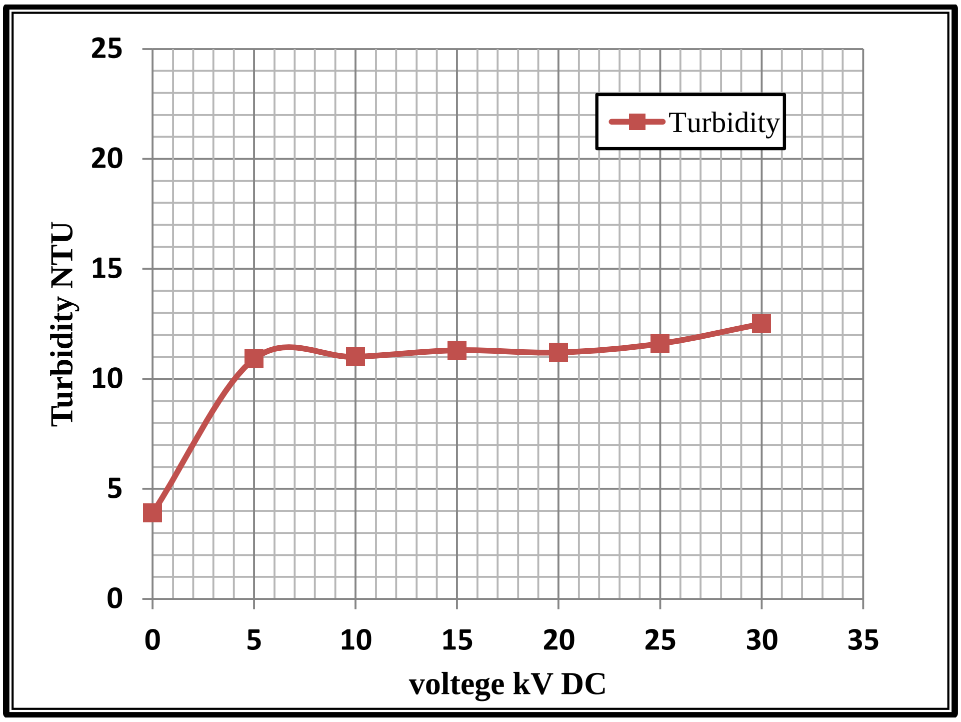


Figure (4-11) Relation between the voltages and turbidity at a dose of 75 mg/l of PACl

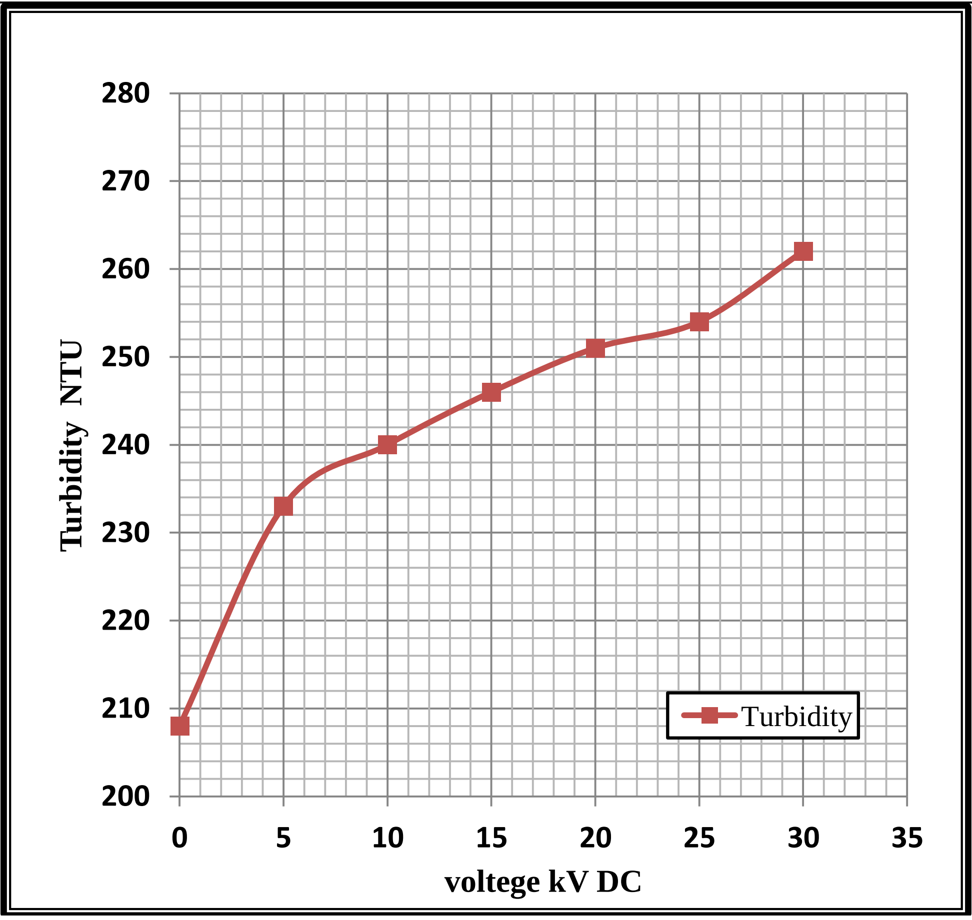


Figure (4-12) Relation between the voltages and turbidity at a dose of 0 mg/l of PACl

Results according to Tables from(4-10) to (4-15) and Figures from (4-7)to(4-11) show that the turbidity level increases to values higher than the values obtained in the first stage of each dose after using Zeta Rod technology or high-voltage capacitors to try to change the surface charge of particles.

Table (4-10) shows turbidity begins to rise from 22.1NTU to 29.8NTU after using the voltage from 5kV DC to 30kV DC, respectively. Table (4-14) shows as well increase in turbidity reach 12.5 NTU despite using a dose of 75 mg / liter of PACl. Moreover the use of Zeta Rod without adding any dose of coagulant shows that the turbidity level has increased from 208NTU to up to 262NTU when using the voltage from 5kVDC to 30kVDC gradually as shown in Table (4-15) and Figure (4-12).

Chapter five : Conclusions and Recommendations

5:1. Conclusions

From these results, it can be concluded the following :

1 - According to the first phase, it can be deduced that the coagulant of poly aluminum chloride is better then alum and ferric chloride in the process of removing the turbidity of the water. which has high turbidity levels and using a dose of 75mg/L of PACl gives turbidity level of 4.55NTU which is less than permitted levels of turbidity in the Iraqi specifications for drinking water which is 5NTU [72] . On the other hand, levels are not reached when using the same dose of alum or ferric chloride and under the same operating conditions. Moreover at the medium levels of water turbidity, the dose of 45mg/l of PACl has achieved specifications allowed and brought the turbidity level to 3.63NTU.At low levels, the dose of 30mg/l of PACl has achieved specifications allowed and brought the turbidity of the water level to 4.92NTU, while the same could not be achieved doses with alum and ferric chloride under the same operating conditions.

PACl has superiority because Polyelectrolyte's are long-chain, high-molecular-weight molecules which bear a large number of charged groups. The net charge on the molecule may be positive, negative, or neutral.The chemical groups on the polymer are thought to combine with active sites on the colloid, combining them into larger particles which will then settle by gravitational force.

This corresponds to the findings of the researchers when comparing coagulants used in water treatment.[55]

pH values do not change through the addition of PACl showing the non-disintegration of long strings of coagulant, The chains of the polymer can then attach to the neutralized particles and generate a larger, single particle which contributes to the deposition of plankton while the pH values have changed significantly with the use of alum and ferric chloride solution and the tendency to be acidic means that coagulants have disintegrated into the water.

2 - It has been noted that when using PACl in all stages ,the quantities of aluminum remaining in the water after treatment are very small and are almost unnoticeable in contrast when alum is used , the remaining quantities of aluminum are large and exceed the limits allowed in the standard specifications Iraqi for drinking water which is 0.2 mg/l [72] excess of which causes neurodegenerative diseases, such as Alzheimer’s disease.[73]This is one of the reasons for the PACl advantage over Alum.

3 - The application of high-voltage capacitors technology using a technique Zeta Rod raises the following question: Dose enhance coagulation process or is Poly aluminum chloride the best of all in the removal of turbidity.

The answer obtained from the results that show increase in turbidity after using voltages and imperceptibly.

This means that there is a process of separating molecules accumulated and deposited because of the voltages used .the use of Zeta-Rod does not reinforce the flocculation process, but the separation of the particles from each other thus increasing the turbidity of the water . This result is due to the devices available in the laboratories of the Ministry of Science and Technology which limit voltage from 5 to 30kV DC . It was found that within these voltages the process is of separating , not assembling molecules. Them can be concluded that the use of high voltage through the ceramic pole of Zeta(5-30 kV DC) will increase the negative charge of the colloidal particles in the water and therefore can through this techniques control Zeta potential for water treatment and thus control the operations making the water with the values of high Zeta potential (negative or positive). The use of high voltages will prevent pooling particles and the formation of large blocks in the water. From previous studies it is shown that this technique can be used in water treatment plants, which is called(RO)to keep efficiency of the membranes used.

Chapter five : Conclusions and Recommendations

5:2 Recommendations

1- From this dissertation, it has been noted to poly aluminum chloride coagulant is preferred because of its efficiency in the process of coagulation, it helps to accumulate relatively high weight (heavy)particles making it easier for them deposit and reduce the time needed for deposition and this feature makes it better than the rest of the coagulants used in the water treatment . Therefore, we recommend using Poly aluminum chloride as an alternative from other coagulants in water treatment processes .

2- There is a need to adjust the concentrations of substances which coagulate because not adjusting concentrations leads to operational problems in the filter units . The survival of large proportions of harmful ions of aluminum and iron salts has a negative impact on public health .

Therefore, we recommend taking into account the allowable ratios which are added to the water for the purpose of treatment.

3- For the purpose of the study to take advantage of Zeta Rod technology to enhance the coagulation process in water treatment we recommend using a higher voltage of 30 kV or less than 5 kV DC in devices of Zeta Rod .

4- We recommend of using the technique of Zeta potential in desalination systems and water filter (voltage of 5kV to 30 kV DC) because it will contribute to raising the efficiency of these filters, and will work to prevent the collection of particles with each other and form large blocks into the water to settle and close the openings in filters, because aggravation of this problem with time leads to a reduction in the efficiency of these filters.

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رسالة مقدمة إلى جامعة تكريت/ كلية التربية – قسم الفيزياء
وهي متطلب جزئي لنيل شهادة الماجستير في الفيزياء
تقدم بها

ماهر خليل إبراهيم

إشراف

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