Treatment of Wastewater Associated With Crude Oil in Reservoirs

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Abstract:

The produced water or wastewater resulting from the oil reservoirs in Basrah Petroleum Company / Zubair 1 Warehouse causes very high pollution in the land. The aim of this study is to find possible ways to treat such produced water and to remove turbidity through coagulation, flocculation, sedimentation and adsorption. Experimental tests of the properties for natural produced water such as turbidity, pH, TDS, TSS, and oil content found out that oil content = 137 mg/L and turbidity = 122 NTU. The produced water is treated with two types of coagulant poly aluminum chloride, alum and with two other types of adsorbent (Bentonite , Zeolite (Cation and Anion)). The coagulation, flocculation and sedimentation experiments are performed using a Jar-test device and adsorption experiments , and a magnetic stirrer device are used at different times, speeds, and concentrations.

It is found through the practical results that the best dosages of (poly aluminum chloride and alum) are (3, 35, 6.75, and 30) mg/L to obtain R% (96.3, 93, 99, and 98.9), respectively.

It is found from the experimental results of adsorption that the best results of weight adsorbent, stirrer time and stirrer speed to obtain maximum removal efficiency are (6 gram anion zeolite ,15 minute and 920 rpm) R% (98 , 97.8 and 99.2), respectively .

It is found that the best results are poor at different parameters, except when it is used (2) gram of bentonite obtained R% of turbidity weight, stirrer time and stirrer speed by bentonite and zeolite (cation and anion) were (2 gram) . Finally, it was found that removing the oil content by adsorption (97.8) is better than coagulation (96.3). And the results for removing turbidity by coagulation (99) is better than adsorption (97.4) .

Keywords: Produced water, Coagulation, Flocculation, Sedimentation, Adsorption, Poly aluminum chloride (PAC), Alum, Bentonite, Zeolite (Cation and Anion).

Symbol	Definition	Unit
C	Final Concentration	mg/l.
C_0	Initial Concentration	mg/l.
NTU	Nephelometric turbidity unit	
R%	Oil removal efficiency =	((C0 – C)/C0)*100

معالجة المياه المصاحبة للنفط الخام في الخزانات النفطية

الخلاصة:

تسبب المياه المنتجة او المياه العادمة الناتجة من الخزانات النفطية في شركة نفط البصرة / مستودع زبير 1 نسبة عالية من التلوث في الأراضي الزراعية. تهدف هذه الدراسة الى ايجاد طرق ممكنة لمعالجة هذه المياه المنتجة وازالة التعكر من خلال التخثر - التلبد - الترسيب والامتزاز. لقد أثبتت الاختبارات التجريبية لخصائص المياه المنتجة الطبيعية من الموقع مثل التعكر، ودرجة الحموضة ، و TDS، و TSS، ومحتوى الزيت.

حيث يبلغ محتوى الزيت (137 ملغم / لتر) والتعكر (122 وحدةNTU)، وتتم معالجة المياه المنتجة بنوعين من المخثرات (بولي المنيوم كلورايد والشب) مع نوعين اخرين من الممتزات هي (البنتونايت، الزيولايت (الموجبة والسالبة)). وقد أجريت تجارب التخثير- التلبيد والترسيب لـ (بولي المنيوم كلورايد و الشب) من خلال استخدام جهاز الجرة مختبريا وأجريت تجارب الامتزاز لـ (البنتونايت و الزيولايت) من خلال أستخدام جهاز محرك مغناطيسي في أوقات وسرع وتراكيز مختلفة.

تبين من النتائج التجريبية باستخدام اختبار الجرة لإزالة محتوى الزيت والتعكر ان الجرعات الافضل لـ (بولي المنيوم كلورايد والشب) هي (3، 35 ، 6.75 و 30) ملغرام/ لتر بنسبة كفاءة إزالة (96.3، 93، 99 و 98.9) على التوالي .

ايضا" تبين من النتائج التجريبية للامتزاز الذي يستخدم لأزالة المحتوى النفطي ان افضل وزن، زمن التحريك وسرعة التحريك للممتزات هي (6 غرام زيولايت سالب، 15 دقيقة و 920 دورة في الدقيقة) بنسبة كفاءة إزالة (98، 97.8 و 99.2) على التوالي.

وجد من النتائج التجريبية لعملية الامتزاز لإزالة التعكر أن أفضل النتائج رديئة في معايير مختلفة، إلا عند استخدامه (2) غرام من البنتونايت الذي تم الحصول عليه R٪ من وزن التعكر وزمن التحريك وسرعة التحريك بواسطة البنتونايت والزيولايت (الموجبة والسالبة) كانت (2 غرام).

في النهاية، توضح النتائج العملية للطريقتين المذكورتين أعلاه أن إزالة محتوى الزيت عن طريق الامتزاز (97.8) أفضل من التخثير(96.3) . ونتائج إزالة التعكر بواسطة التخثير (99) أفضل من الامتزاز (97.4).

1- Introduction:

The treatment of wastewater associated with crude oil in reservoirs is important processes, where the untreated such water may affect the quality of the oil and its price and properties …. etc. As well as, it affects such produced water on human health, the environment and soil and underground water, so this research is intended to deal with such produced water for the purpose of free of crude oil, water, materials suspended solids and other materials. Produced water is formed by oil and gas extraction with the oil industry. It produces about 14 billion barrels of water per year [1]. The water varies very greatly in quality and quantity, and in some cases water can be useful as secondary product or even a salable commodity. Produced water is one of the major and influential problems in the petroleum industry and generated in increasing volumes from both old and new wells [2]. These effluents represent 98% of the total wastewater from the petroleum industry. It also contains complex organic and inorganic substances Such as metals, salts dispersed oil and dissolved hydrocarbons and high temperature and lack of oxygen [3]. Therefore, produced water is a major and important source of pollutants, and as the environmental laws have become more stringent, the cost of treatment has become increasingly high. This fact has led to many efforts to find more efficient and less costly ways to treat this water [4].

Therefore, the main idea of this work is to use the method treatment (coagulation, flocculation, sedimentation & adsorption) to remove the oil content and the turbidity of the oil reservoirs / Basrah Oil Company. It is also to study the effect of adding (PAC, Alum, Bentonite and Zeolite (Cation &Anion)) individually and collectively, on removal efficiency of oil content and turbidity, as well as, the use of the best dose, best time and speed of the additives (poly aluminum chloride, alum, bentonite, zeolite both types) to remove the oil content and turbidity of the produced water oil in reservoirs.

Methods of Treatment:

There are many treatment processes for removing the solids and impurities from soluble wastewater. These processes can be categorized into chemical, physical or biological treatments, such as sedimentation, precipitation, evaporation, extraction, coagulation-flocculation, flotation, electrocoagulation, filtration, adsorption, bio-degradation, ion exchange, and electrochemical treatment [5, 6 and 7]. The efficient treatment of wastewater typically requires many steps, and it is often appropriate to combine more than one method [7]. Coagulation and flocculation are among the most widely used technologies for removing solids from industrial and municipal wastewater, while adsorption is the method most frequently used metals in dilute solutions [8, 9]. The grease and oil content must be treated before discharge into the city's drainage systems. The most common treatment methods for wastewater treatment are sedimentation, centrifugal separation, coagulation, flocculation, sorption, flotation, filtration ultrafiltration, and reverse osmosis. These methods can be used separately or in groups [10]. Coagulation and flocculation are simple, inexpensive, efficient and most traditional ways of treating wastewater [11, 12].

The coagulation, flocculation, sedimentation and adsorption are used in current work. Coagulation is a process in which the colloids or particles in a suspension are destabilized, leading to the formation of small groups of molecules [13, 14 and 15].

While in flocculating the resulting destabilized and aggregated particles or other colloidal and particular substances are collected into larger aggregates [13, 14].

Flocculation is the process of quiet water movement that promotes the aggregation of small floc particles (microflocs) resulting from coagulation in larger blocks suitable for removal by clarification processes. Flocculation is enhancements sedimentation or filtration treatment system performance by increasing particle size resulting in increased settling rates and filter capture rates.

Sedimentation is a treatment process in which is suspended particles, like flocs, sand and clay which are re-moved from the water. Sedimentation is the process of letting the suspended material settles by the gravity. Adsorption is very efficient, cost effective and most importantly has the ability of meeting the environmental compliance as far as the discharge standard of the oil content of the wastewater is concerned [16].

Adsorption is an increase in the concentration of certain substances in the interface of the two different phases [17].

Adsorption is used in the industrial field for the separation, purification and recovery of mixtures, chemicals, liquid and gas media [18].

2- Experimental Work:

2-1 Methods and materials:

The experimental work to remove the oil content from the produced water is considered in two stages. The first stage includes knowledge of the standard characteristics of the produced water from the Basrah Oil Company tanks. These standard properties include turbidity and oil content. All the experiment procedures are taken from the standard methods in the examination of water and produced water, was developed in 1995 (ASTM) [19]. The second stage includes methods of treatment of produced water by coagulation, flocculation, sedimentation & adsorption. The produced water treats with four types of chemical additives: poly aluminum chloride, alum, bentonite and zeolite. Two types of coagulant are used (Poly aluminum chloride and Alum) and two other types of adsorbent (Bentonite and Zeolite) are used also. The Jar test device (laboratory scale) is used to conduct coagulation, flocculation and sedimentation experiments. Adsorption experiments are performed by using a magnetic stirrer apparatus at different times, speeds and concentrations. The Jar-test device consists of a set of vertical paddles (6-paddles) respectively, so that the cups of liters of water produced appropriately under each paddle. The driving engine has a variable speed controller. The rotation speed is in the range from (0-400) rpm [20]. In the magnetic stirrer, (200) ml of produced water is placed into the beaker. Add different weights in grams of the material (such as bentonite) in beaker, and beaker puts on the magnetic stirrer apparatus. Magnetic stirrer runs at a different speed and also different times to see the removal efficiency of oil content and turbidity. The removal efficiency is calculated by the equation below

$$R\% = \frac{(C_o - C)}{C_o} * 100$$

Where Co = Initial Concentration, C = Final Concentration

Pull the top layer at high (5) cm from the sample by a pipette and then measure the oil content ratio and turbidity. The characteristics of the chemical are shown in Tables (1 to 4).

Table (1) Properties of PAC (University of Technology / Chemical Eng.)

Item	Value
Al_2O_3 %	10.2
Basicity %	64
Solubility in water g/l at 20°C	Completely Soluble
pH of 5% aqueous solution	2.5-4.5
Chloride as Cl	10.5
Specific gravity at 20°C	1.20
Physical state	Liquid
Color	*Yellowish*
Viscosity (20°C)	*< 50 m Pa. s*
Density	*1,15 –1,41 gr/cm^3*

Table (2) Properties of Alum (Basrah Oil Company)

Description	Crystalline powder
Minimum assay	99.5 %
Insoluble matter	0.005 %
Chloride (Cl)	0.001 %
Ammonium (NH_4)	0.025 %
Arsenic (As)	0.0002 %
Copper (Cu)	0.0005 %
Iron (Fe)	0.0005 %
Lead (Pb)	0.0005 %
Sodium (Na)	0.01 %

Table (3) Properties of bentonite (University of Technology / Chemical Eng.)

Characterizes	Value
Pratical size , mm	0.075
Silica (SiO_2)	54.26
Alumina (Al_2O_3)	14.87
Ferric oxide (Fe_2O_3)	4.94
Magnesium oxide (MgO)	3.8
Calcium oxide (CaO)	5.53
Sodium oxide (Na_2O)	0.98
Potassium oxide (K_2O)	0.38
Titanium oxide (TiO_2)	0.75

Table (4) Properties of Zeolite (Basrah Oil Company)

Characterize	Cation Value	Anion Value
Active Groups	$R – SO_3$	$R - N+(CH_3)_3$
Physical aspect	Dark spherical beads	Clear golden spherical beads
Particle size	0.30 – 1.20 mm	0.30 – 1.20 mm
Effective size	0.50 mm	0.50 mm
pH range	0 – 14	0 – 14
Specific gravity	1.21	1.08
Water retention	51 – 55 %	48 – 54 %

2-2 Materials:

There are four types of chemical used in treatment of produced water. Two types are used in coagulant alum and PAC. Other two types are used in adsorption process as adsorbent bentonite and zeolite (cation, anion).

3- Results and Discussion

3.1 Effect of coagulation type on the efficiency of removal of oil content and turbidity:

Figures (1 to 4) show results obtained by coagulation at the initial concentration of oil content and turbidity from produced water. These figures show that the efficiency of removal of oil content and turbidity increases with increasing coagulation doses until they reach the best dose for removal. At best dose the removal efficiency of oil content at initial concentration Co= 137 mg/L is (96.3%, 93%) for (poly aluminum chloride and alum), respectively. And at best dose the removal efficiency of turbidity at initial concentration Co= 122 NTU is (99%, 98.9%) for (poly aluminum chloride and alum), respectively.

The overdose for poly aluminum chloride and alum causes the decrease in the oil removal efficiency because most dose of poly aluminum chloride and alum causes stability again (restabilized).

These results are removed from the influence of coagulation, flocculation, which overcomes some of the forces of repulsion between the colloidal particles, and reduces the distances between them, which increases the power of attraction and van der Waals forces and makes them weak Brownian motion and then weaken the value of the existing Zeta [21]. The doses of poly-aluminum chloride and alum increases the efficiency of removing the turbidity until it reaches the best value, and after that the efficiency of de-turbidity is reduced because more doses cause the re-stabilization of colloid particles.

3.2 Effect of adsorption on the efficiency of removing oil content and turbidity:

Figures (5 to 16) show that the adsorbent at the initial concentration of oil content and turbidity in the produced water are obtained. These figures show that the efficiency of removal of oil content and turbidity increases with increasing adsorption doses until they reach the best dose of removal. The adsorbent materials (bentonite, zeolite (cation and anion)) are used, the removal efficiency of the oil content of the best weight (1, (6 and 6 gram)) is (87.6%, (97% and 98%)), respectively.

The best stirring time of adsorbent materials (bentonite, zeolite (cation and anion)) (120, (15 and 10 minute)) are (94.1%, (97.8% and 97.4%)), respectively. Also the best stirring speed of adsorbent materials (bentonite, zeolite (cation and anion)) (920, (1080 and 920 rpm)) is (99.2%, (98.8% and 98.6%)), respectively.

Overweight leads bentonite decrease in oil removal efficiency, this is due to the hydration of bentonite which causes organic compounds with average molecular weights are weakly adsorbed or not adsorbed by pure montmorillonites (Bentonite) due to their reliably large solubility in water. Soluble compounds with molecular weights, whether charged or not, are absorbed by montmorillonites because of their low solubility in water [22]. The results of the effect of stirring speed to reduce the oil content is good, where the oil content decreases with increasing stirring speed due to a decrease in the resistance to mass transfer oil from the bulk of the oily waste water to the surface of bentonite. It is scheduled to take the deployment of each particle layer of bentonite in the resistance to mass transfer. For the stirring time, as the increase in the stirring time leads to decrease the efficiency of the removal of the oil content of the inability of the withdrawal of capillary. Water absorption of bentonite occurs through diffusion and capillary suction [23]. The efficiency of removal of oil content increases with increasing (weight, stirring time and stirring speed) for zeolite, until reach the (best weight, best time and best speed) for zeolites. This is due to increase the adsorption sites available by increasing the dose of adsorbents. However, adsorption density is reduced due to unsaturated adsorption sites. Adsorption density is also reduced due to particle interactions resulting from high concentration of adsorption. The removal efficiency of the adsorbent material increases with increasing doses [24].

The adsorbent materials (bentonite, zeolite (cation and anion)), the turbidity removal efficiency of the best weight (2, (4 and 4 gram)) is (99%, (90% and 87.4%)), respectively. As well as the best stirring time of adsorbent materials (bentonite, zeolite (cation and anion)) (120, (15 and 10 minute)) is (90%, (91% and 86%)), respectively. Also the best stirring speed of adsorbent materials (bentonite, zeolite (cation and anion)) (1080, (600 and 600 rpm)) is (97.4%, (89.2% and 86.6%)), respectively. The overdose for (bentonite, zeolite (cation and anion)), reduces the efficiency of removing turbidity for the same reason.

The comparison between coagulation and adsorption for removal efficiency of the oil content, the results show that the adsorbent (bentonite and zeolite both types) better than coagulants (poly aluminum chloride and alum), as shown in Fig. 17. As well as the comparison between coagulation and adsorption of turbidity removal efficiency, the results show that the coagulants (poly aluminum chloride and alum) better than adsorbent (bentonite and zeolite both types), as shown in Fig.18. From the graphical figures it is found the results for removal oil content by adsorption (99.2%) which is showed better than coagulation (96.3%). And the results for removal turbidity by coagulation (99%) were shows better than adsorption (97.4%).

4- Conclusions:

From the pilot study for the treatment of produced water from oil reservoirs in Zubair 1 warehouse / South Oil Company some basic information, extracted from the study as following:

1. The characterization of wastewater in reservoirs is analyzed in laboratory and shows the results: oil content = 137 mg/L, pH = 6.9 and turbidity = 122 NTU.
2. For coagulation and flocculation treatment of the poly aluminum chloride and alum as coagulant. In the case, it finds a poly aluminum chloride and alum best dose to removal oil content and turbidity by jar test was to be equal to (3, 35 and 6.75, 30 mg/L), respectively. At the best dose, efficiency is the removal of oil content and turbidity (96.3%, 93% and 99%, 98.9%) from (poly aluminum chloride and alum), respectively.
3. For adsorption treatment, bentonite and zeolite (cation and anion) is as adsorbents. In the case of the adsorbent materials (bentonite, zeolite (cation and anion)), the removal efficiency of the oil content of the best weight (1, (6 and 6 gram)) is (87.6%, (97% and 98%)), respectively. Also the best stirring time of adsorbent materials (bentonite, zeolite (cation and anion)) (120, (15 and 10 minute)) is (94.1%, (97.8% and 97.4%)), respectively. The best stirring speed of adsorbent materials (bentonite, zeolite (cation and anion)) (920, (1080 and 920 rpm)) is (99.2%, (98.8% and 98.6%)), respectively.
4. In the case of the adsorbent materials (bentonite, zeolite (cation and anion)), the turbidity removal efficiency of the best weight (2, (4 and 4 gram)) is (99%, (90% and 87.4%)), respectively as well as the best stirring time of adsorbent materials (bentonite,

zeolite (cation and anion)) (120,(15 and 10 minute)) is (90% , (91% and 86%)), respectively. Also the best stirring speed of adsorbent materials (bentonite, zeolite (cation and anion)) (1080, (600 and 600 rpm)) is (97.4%, (89.2% and 86.6%)), respectively.

5. The results for removal oil content by adsorption (99.2%) show better than coagulation (96.3%) and the results for removal turbidity by coagulation (99%) shows better than adsorption (97.4%).

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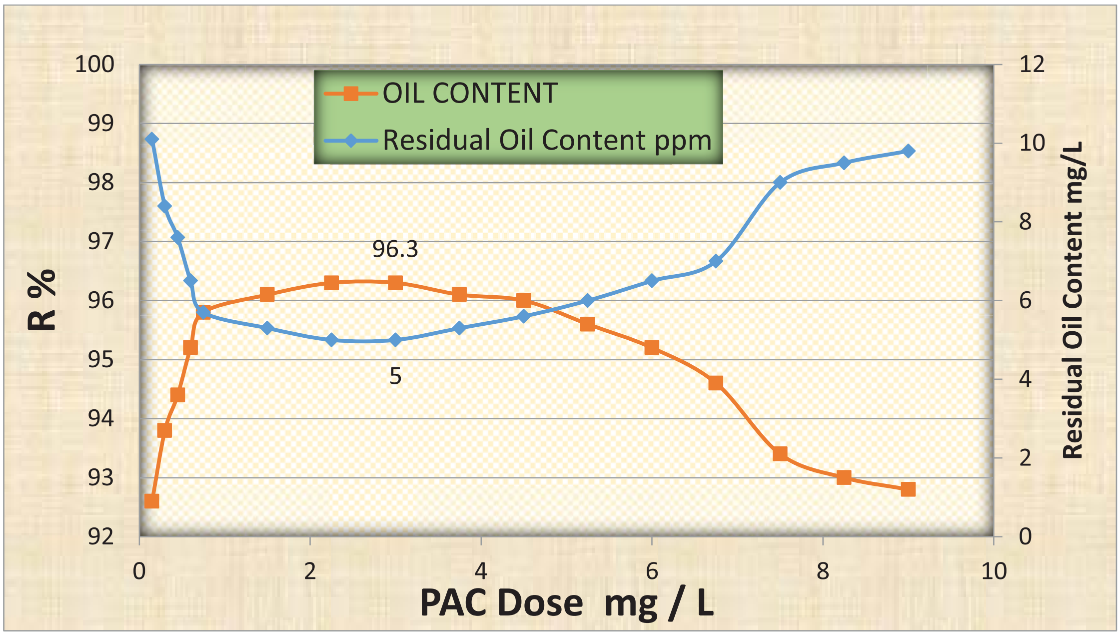

Fig. (1) Effect of PAC Dose on Oil Removal Efficiency at the best (time and speed) is (20 minute, 50 rpm). C_0 = 137 mg/L

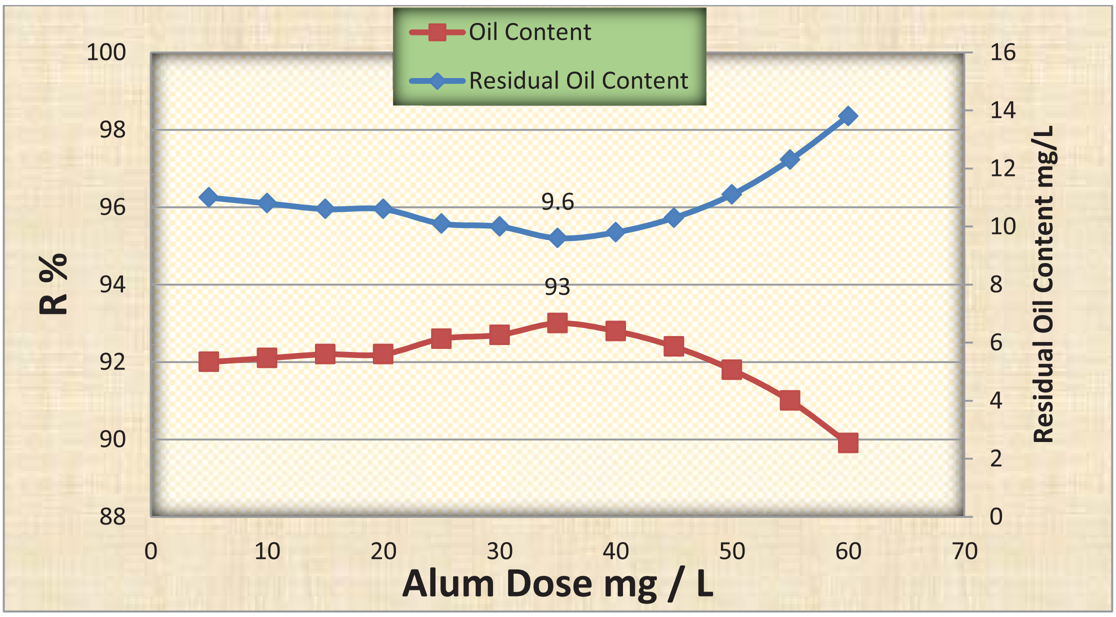

Fig. (2) Effect of Alum Dose on Oil Removal Efficiency at the best (time and speed) is (20 minute, 50 rpm). C_0 = 137 mg/L

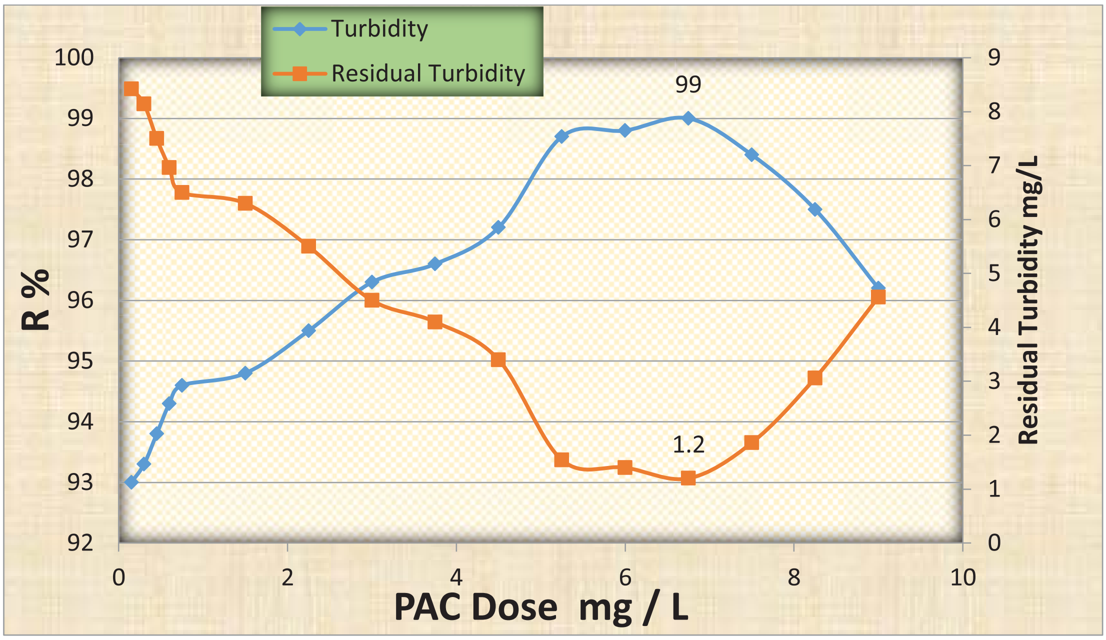

Fig. (3) Effect of PAC Dose on Turbidity Removal Efficiency at the best (time and speed) is (20 minute, 50 rpm). C_0 = 122 NTU

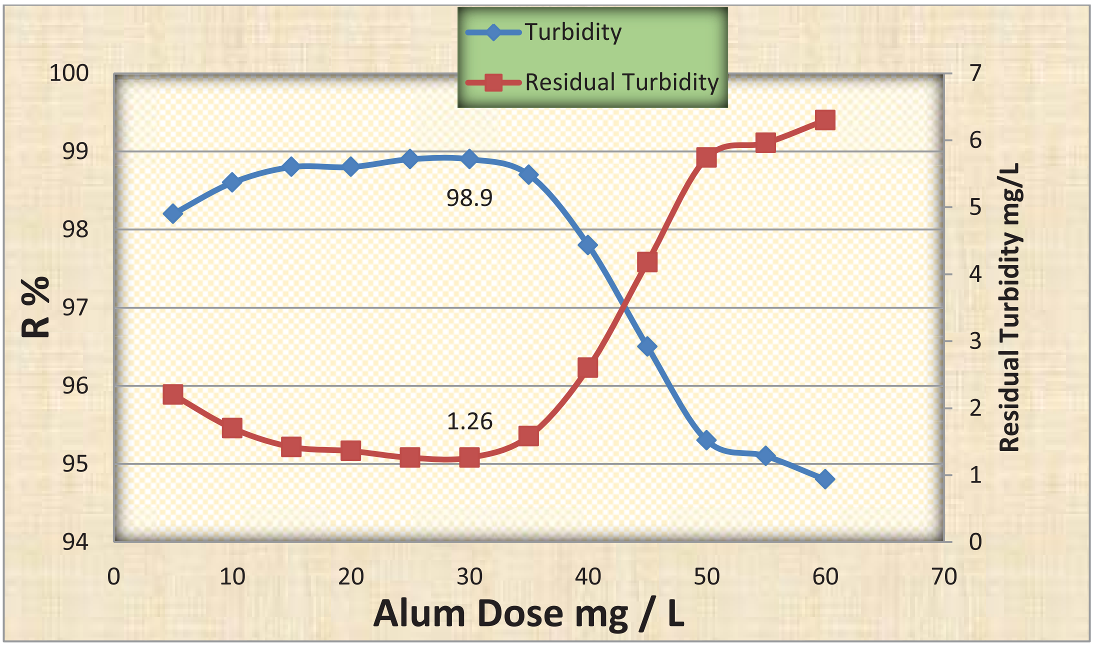

Fig. (4) Effect of Alum Dose on Turbidity Removal Efficiency at the best (time and speed) is (20 minute, 50 rpm). C_0 = 122 NTU

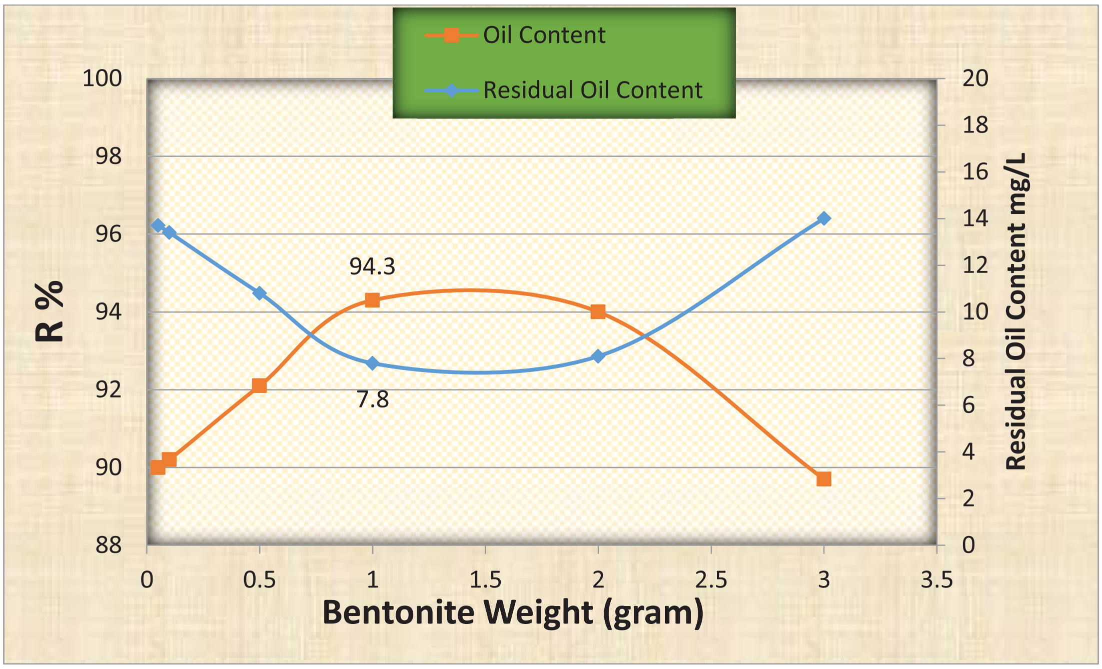

Fig. (5) Effect of Bentonite Weight on Oil Removal Efficiency for Bentonite at the stirrer time and stirrer speed is (120 minute, 300 rpm). C_0 = 137 mg/L

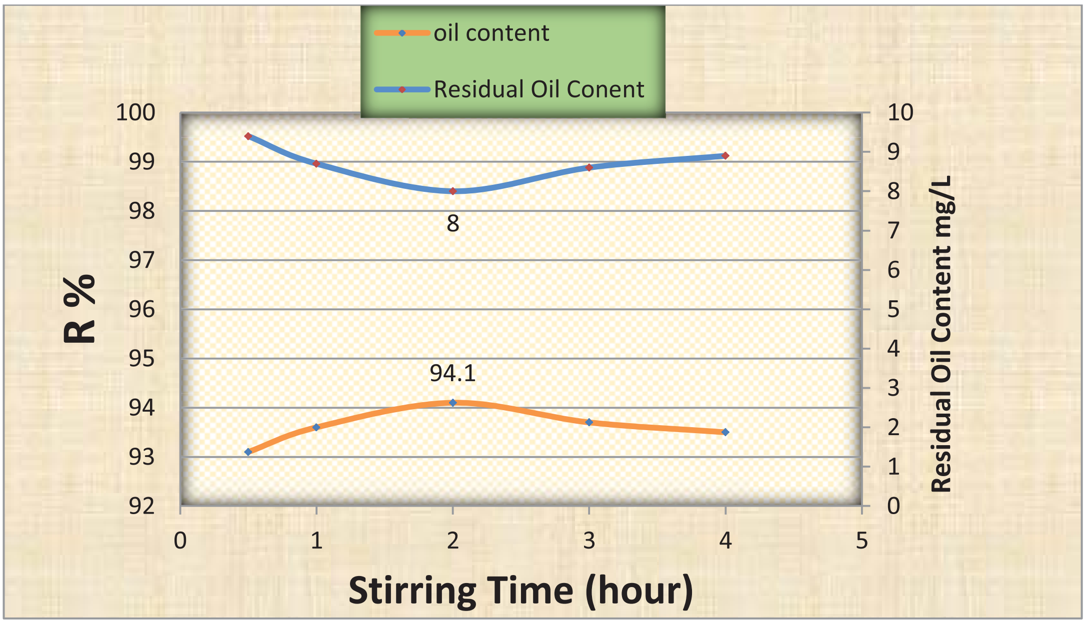

Fig. (6) Effect of Stirring Time on Oil Removal Efficiency for Bentonite at the best (weight and stirrer speed) is (1 gram, 300 rpm). C_0 = 137 mg/L

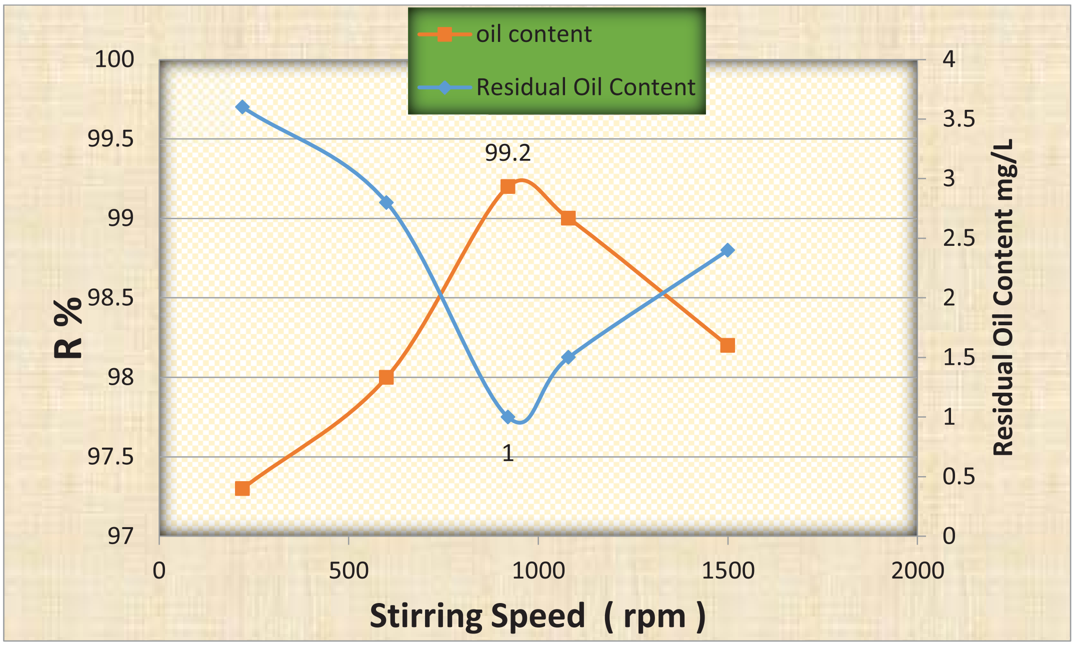

Fig. (7) Effect of Stirring Speed on Oil Removal Efficiency for Bentonite at the best (weight and stirrer time) is (1 gram, 120 minute),respectively. C_0 = 137 mg/L

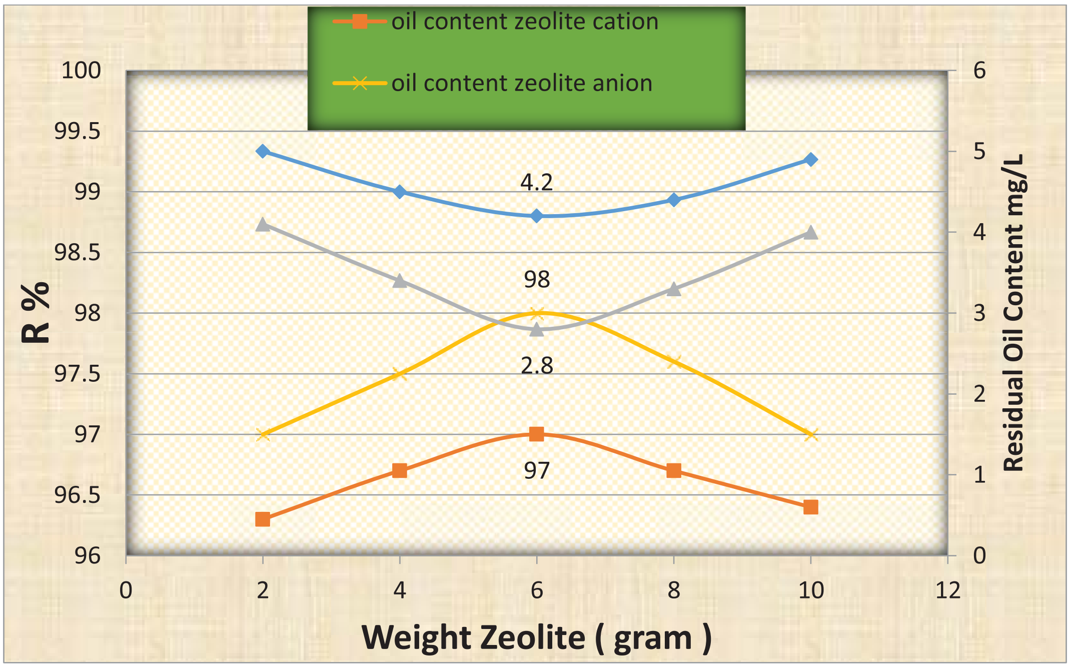

Fig. (8) Effect of Weight of Zeolite on Oil Removal Efficiency for Zeolite at the stirrer time and stirrer speed is (25 minute, 150 rpm). C_0 = 137 mg/L

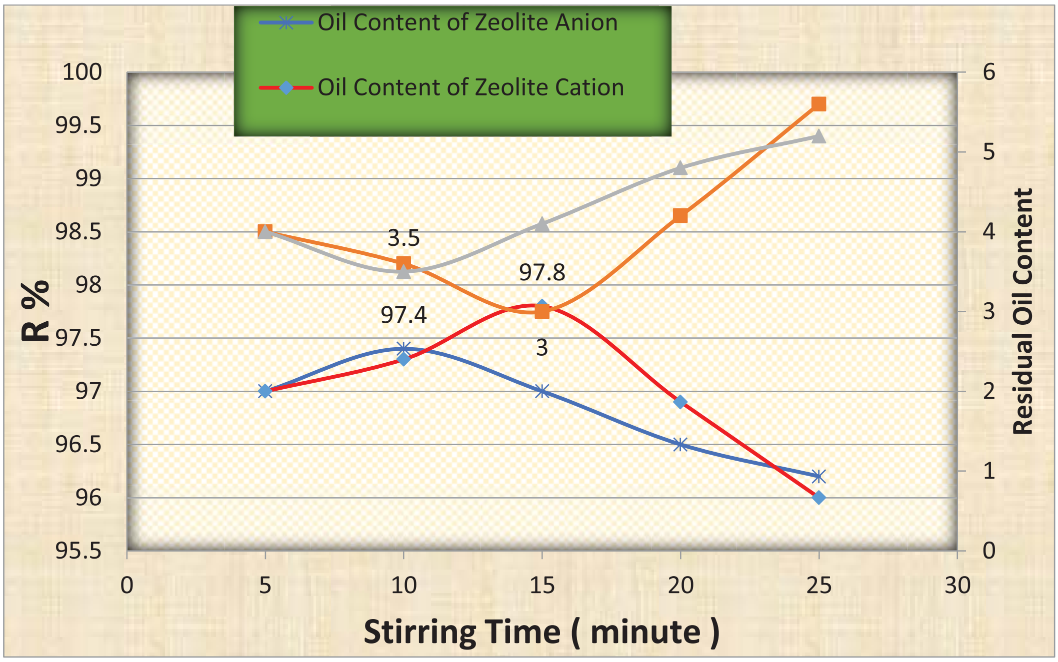

Fig. (9) Effect of Stirring Time on Oil Removal Efficiency for Zeolite at the best (weight and stirrer speed) for (cation, anion) is (6, 6 gram and 150 rpm),respectively. C_0 = 137 mg/L

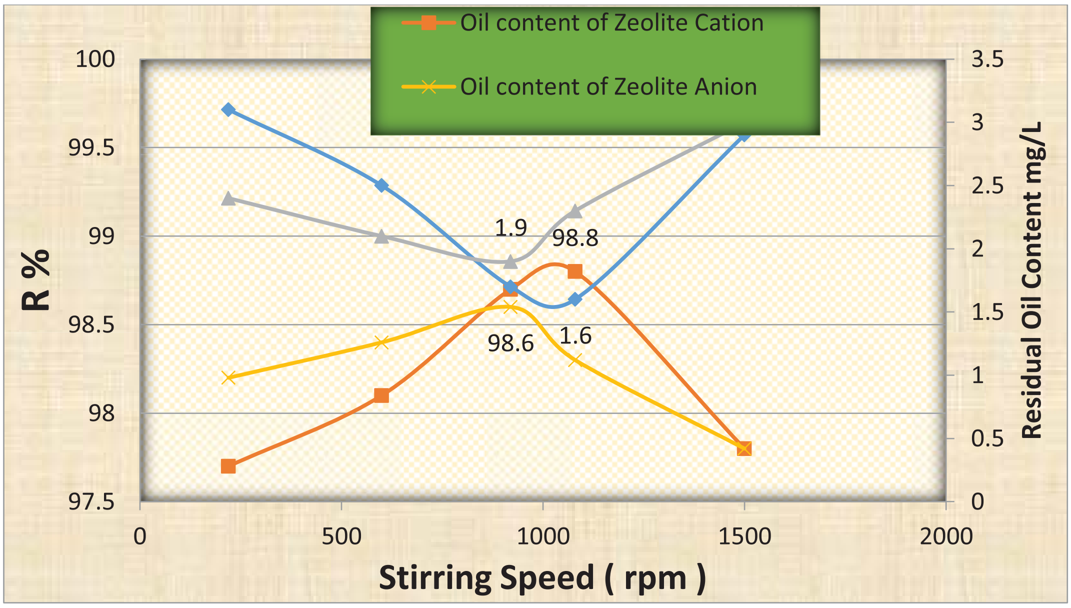

Fig. (10) Effect of Stirring Speed on Oil Removal Efficiency for Zeolite at the best (weight and stirrer time)(cation, anion) is (6, 6 gram and 15, 10 minute), respectively. C_0 = 137 mg/L

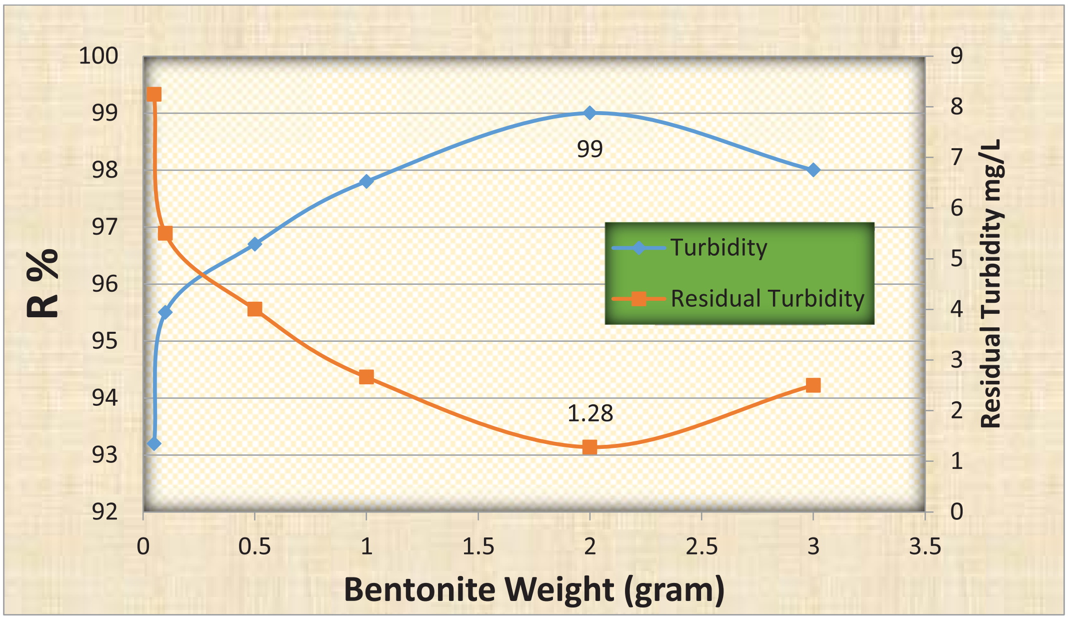

Fig. (11) Effect of Bentonite Weight on Turbidity Removal Efficiency at the stirrer time and stirrer speed is (120 minute, 300 rpm). C_0 = 122 NTU

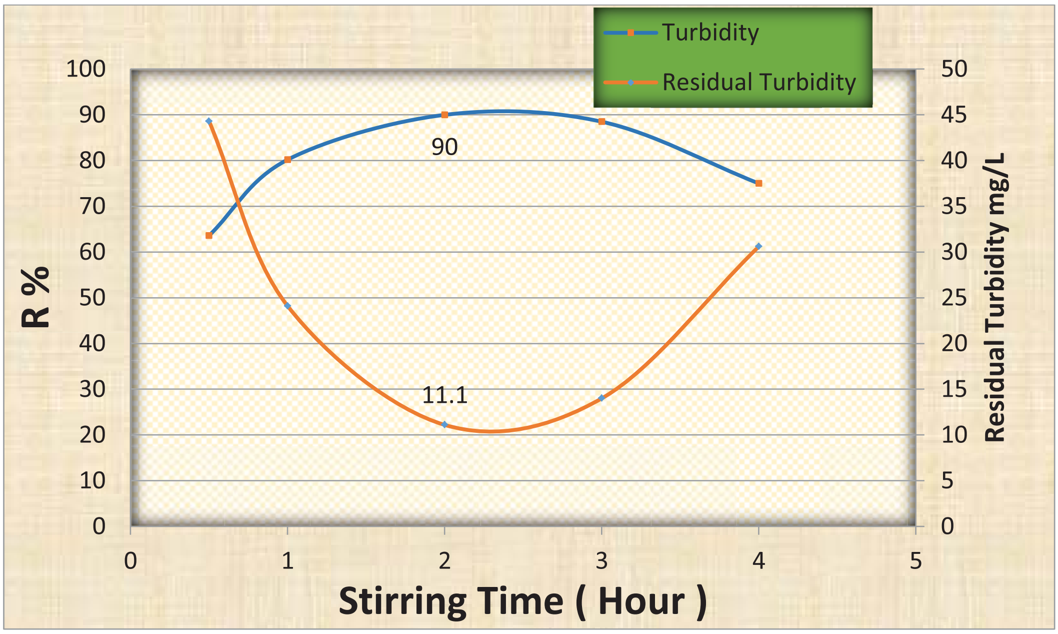

Fig. (12) Effect of Stirring Time on Turbidity Removal Efficiency for Bentonite at the best (weight and stirrer speed) is (2 gram, 300 rpm). C_0 = 122 NTU

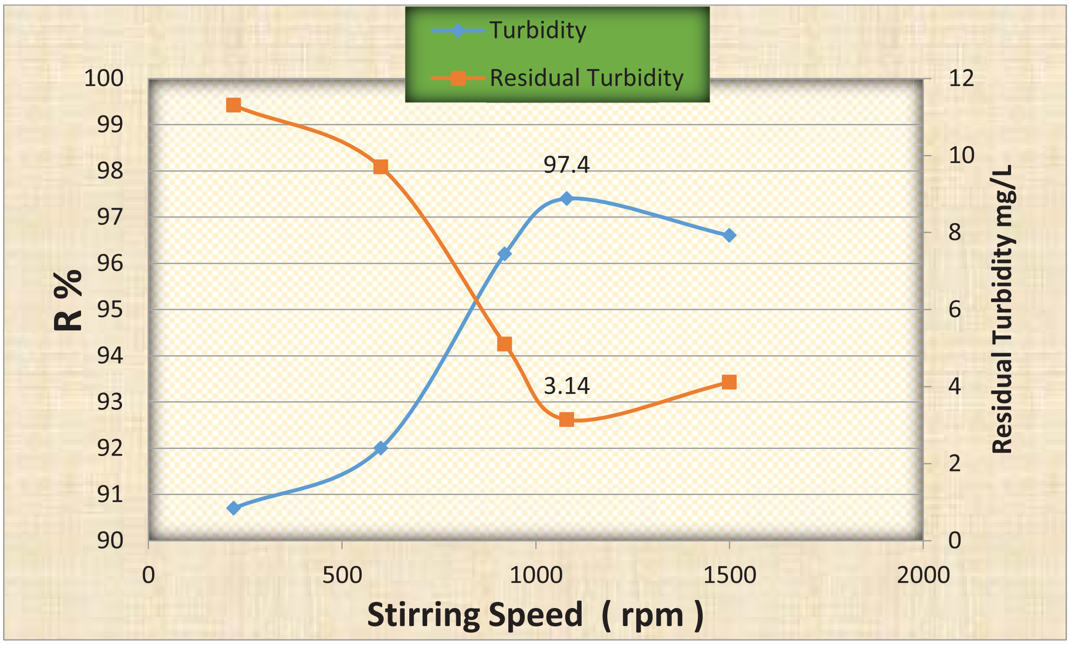

Fig. (13) Effect of Stirring Speed on Turbidity Removal Efficiency for Bentonite at the best (weight and stirrer time) is (2 g, 120 minute),respectively. C_0 = 122 NTU

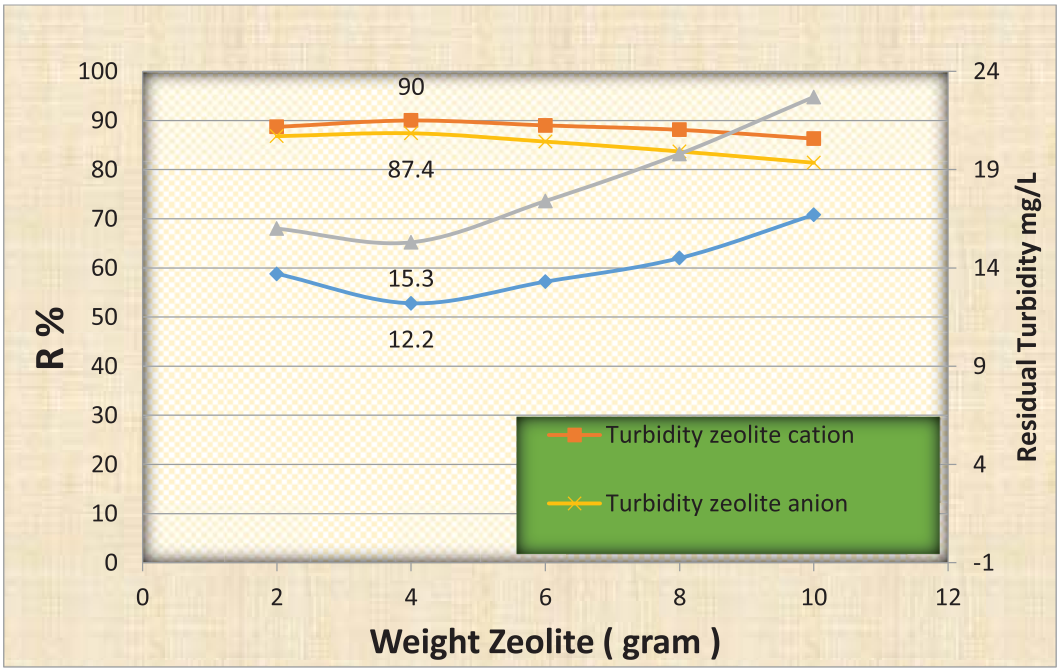

Fig. (14) Effect of Weight Zeolite on Turbidity Removal Efficiency at the stirrer time and stirrer speed is (25 minute, 150 rpm). C_0 = 122 NTU

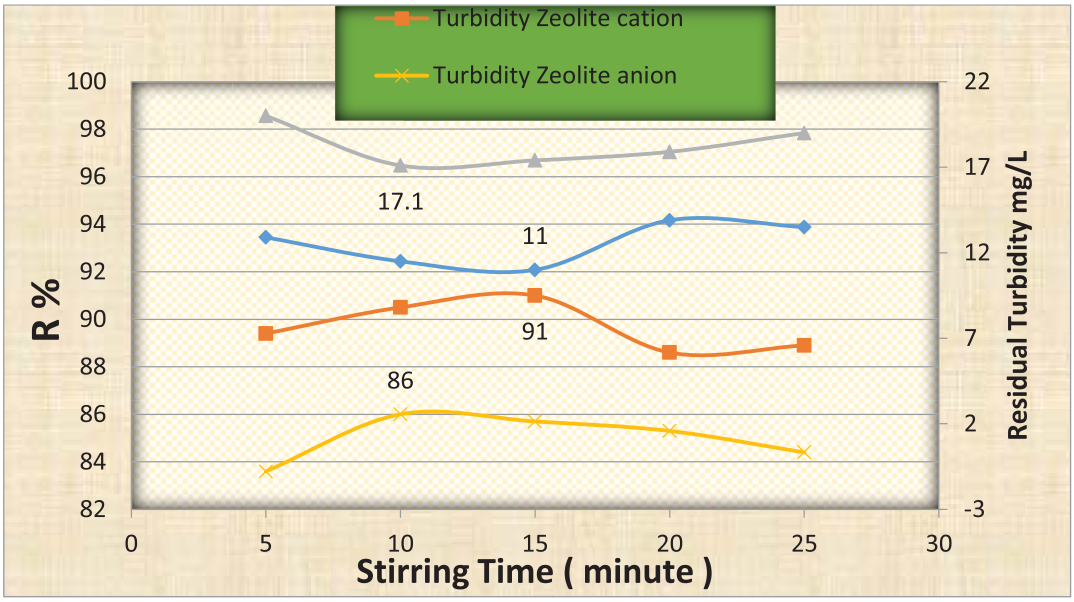

Fig. (15) Effect of Stirring Time on Turbidity Removal Efficiency for Zeolite at the best (weight and stirrer speed) for (cation and anion) is (4, 4 gram and 150 rpm),respectively. C_0 = 122 NTU

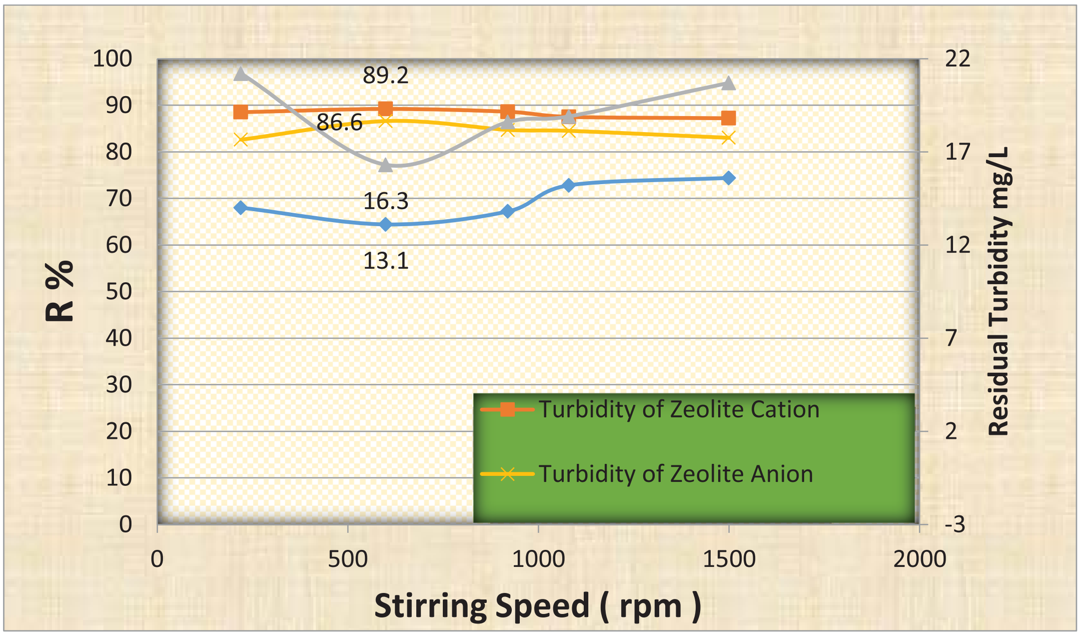

Fig. (16) Effect of Stirring Speed on Turbidity Removal Efficiency for Zeolite at the best (weight and stirrer time)(cation, anion) is (4, 4 gram and 15, 10 minute),respectively. C_0 = 122 NTU

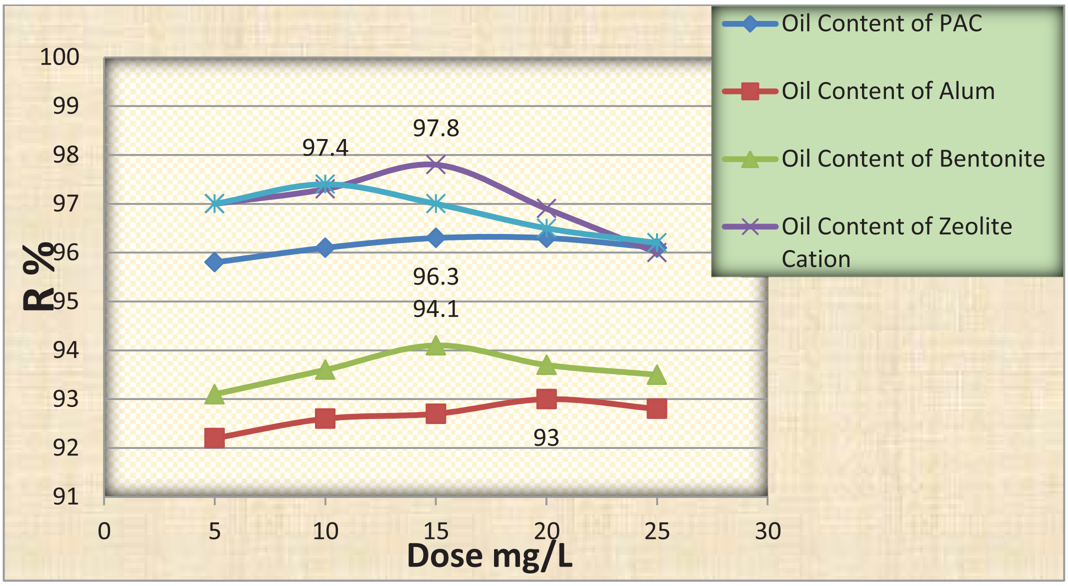

Fig. (17) The comparison between (poly aluminum chloride, alum, bentonite and zeolite both types) for removal efficiency of oil content. $C_0 = 137$ mg/L

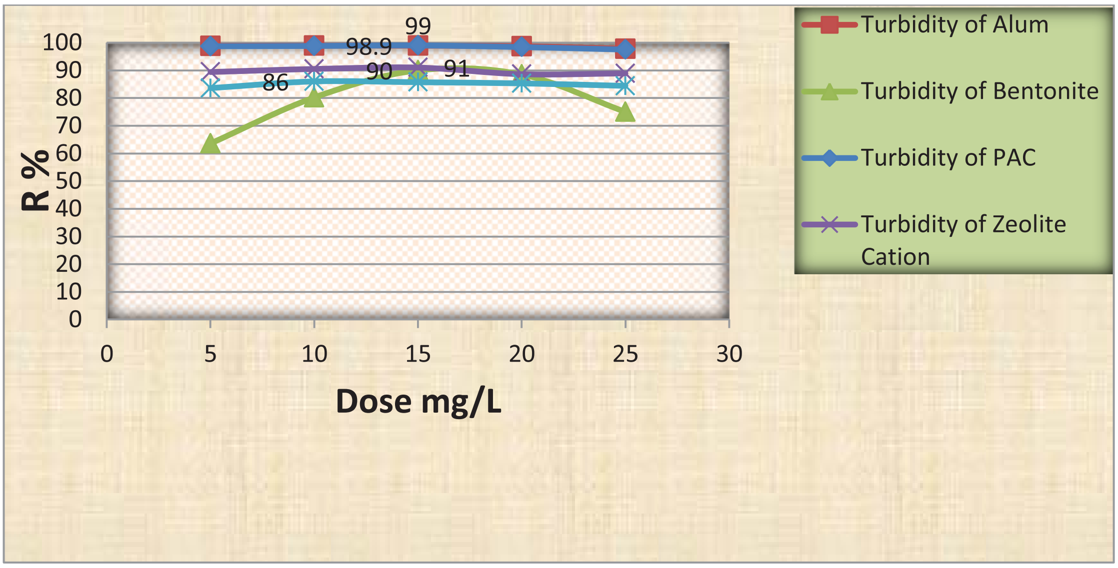

Fig. (18) The comparison between (poly aluminum chloride, alum, bentonite and zeolite both types) for removal efficiency of turbidity. $C_0 = 122$ NTU