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Treatment of Al Doura Oil Refinery Wastewater Turbidity using Magnetic Flocculation

ABSTRACT

The use of conventional flocculants such as Aluminum sulphate (Alum) alone to treat the wastewater may be insufficient to get the required turbidity, suspended solids removal. It also requires relatively a long residence time. Magnetic flocculation is one of the techniques used for increasing the efficiency of the turbidity removal. In the present study, three sets of experiments are carried out in order to investigate the possibility of increasing the suspended solid removal efficiency from Al Doura oil refinery wastewater using iron oxide (Fe₃O₄), Nickel (Ni), and Cobalt (Co) ferromagnetic powders with alum. The following operating conditions namely, pH, alum dose, ferromagnetic powder dose, and initial turbidity are studied. The results revealed that an improvement in turbidity removal efficiency is satisfied, as well as, a reasonable reduction in the sedimentation period is achieved. The highest turbidity removal is 99.88% that obtained for 122NTU sample for alum dose 120 mg/L+ Nickel dose of 80mg/L and pH of 6.5.

Keywords:

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معالجة العكورة لمياه مصفى الدورة باستعمال التليبد المغناطيسي

الخلاصة

قد لا يعطي استعمال الملبدات التقليدية مثل كبريتات الالمنيوم المائية (الشب) الازالة المطلوبة للعكورة من مياه الفضلات الصناعية، هذا بالإضافة الى الحاجة الى وقت مكوث طويل نسبياً. تقنية التليبد المغناطيسي احدى التقنيات الواعدة لغرض تقليل وقت المكوث والحصول على كفاءة ازالة اعلى للعكورة. في هذا العمل تم انجاز ثلاث مجموعات من التجارب لدراسة إمكانية رفع كفاءة ازالة العكورة وتقليل وقت المعالجة لمياه فضلات مأخوذة من مصفى الدورة حيث تم استعمال الشب مع كل من الملبدات المغناطيسية: أوكسيد الحديد والنيكل والكوبلت. كما تمت دراسة تأثير العوامل التشغيلية التالية: الدالة الحامضية، جرعة الشب، جرعة المادة المغناطيسية ونوع المادة المغناطيسية على كفاءة ازالة العكورة. بينت النتائج المختبرية ان هناك زيادة ملموسة في كفاءة ازالة العكورة ونقصان كبير في وقت المكوث نتيجة لاستعمال المواد المغناطيسية مع الشب. كما بينت النتائج المختبرية ان اعلى كفاءة ازالة للعكورة قد بلغت 99.88% عند دالة حامضية مقدارها 6.5 وذلك باستعمال جرع شب مقدارها 120 ملغم/لتر إضافة الى جرعة نيكل مقدارها 80 ملغم/لتر وذلك لنموذج عكورته الابتدائية 122 وحدة عكورة.

1. INTRODUCTION

Earlier studies show that an average of 468 gallons of water were required to refine one barrel of crude oil [1]. However, recent studies show that in USA one barrel of crude oil requires 42–79.8 gallons of water to be refined, with a median of 63 gallons of water [2]. Taking into account that 18.9 million barrels per day of crude oil is

refined in USA at 2013 [3], water reuse within an industrial plant is essential [4]. Wastewaters of the oil refineries contain a large quantities of solids, salts, crude oil, aromatic and cyclic hydrocarbons, surfactants, phenols, naphthalene acids, sulfides, heavy metals, and other chemical products. In primary purification of water and industrial wastewater treatment, a widely used process is coagulation–flocculation. This process is preferable in primary treatment due to its simplicity, high efficiency and

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cost-effectiveness [5]. However, this process exhibits several disadvantages, such as the need for high amounts of chemicals for neutralizing the charges of the suspended particles, the need for pH adjustment before and after treatment, the sensitivity to temperature change, in addition to the excessive sludge production [6]. Various improvements are introduced to the coagulation–flocculation process, such as using natural or synthetic polymers as a flocculant aids to strengthen flocs, employing another technology of separation with the coagulation–flocculation process, like magnetic flocculation. Its principle is adding particles of a higher magnetic susceptibility into a conventional coagulation – flocculation process to enhance the flocculation velocity and form flocs of high density to settle quickly [7]. It combines a traditional flocculation and a magnetic separation in one process exhibiting a quick, simple, energy – efficient, and cost effective advantages [8].

Miura et al. [9] applied a ferromagnetic powder with aluminum sulfate or polyaluminum chloride in order to remove solids from the wastewater. They got a removal efficiency of 99%. It was noticed that the required time for separating flocculated suspended solids was only few seconds, while in conventional treatment, it takes about one hour. Slusarczuk and Brooks [10] added a magnetic ferric powder and polyethylene imine as a flocculant agent to treat the turbidity. It is found that ferrite powder exhibited synergism with the aqueous polyethylene imine solution. The results revealed that the sludge volume is about 80% less than the volume produced by using polyethyleneimine alone. The suspended solids removal efficiency is raised from 30% to 71 % when 1000 ppm of ferrite powder and 10 ppm of polyethyleneimine are added. Kang et.al. [11] used a magnetic ferrite powder of about 5 μm average particle size after dealing it with a solution of white alum ($\text{KAl}(\text{SO}_4)_2$), polyaluminum chloride or ferric chloride. Ferrite powder was added at stirring speed of (200 - 300 rpm) for (1.0 - 2.0 minutes). It was found that the flocculated particles settled rapidly at a rate of 5 cm/minute, whereas in conventional methods that use alum or polyaluminum chloride, a period of 2.0-4.0 hours is required for efficient settling. Magnetic seeding aggregation (MSA) of silica nanoparticles was studied by Ref. [12]. Influences of pH, salts addition, and type of magnetite seeding particles on the turbidity removal efficiency were examined. The turbidity of CMP treated wastewater is reduced from 110.0 NTU to 7.0 NTU at pH of 6. The results showed that the residual turbidity decreases with the increase of magnetic field intensity. When the magnetic field intensity is higher than 0.08 Tesla, the residual turbidity is about 1.0 NTU. High turbidity reduction during the storm period by magnetic aggregation and separation was obtained by Ref. [13]. High turbidity raw water was prepared by mixing a sludge sample that are taken from Shiemen reservoir's tail water pond with deionized water. It was found that at magnetic field strength of 0.1 Tesla, the magnetic aggregation effects were not significant but at magnetic field strength of 0.15 Tesla, significant effects on the magnetic aggregation were observed. When the magnetic field strength raised to 0.2 Tesla, the effect on magnetic aggregation was stable. The results also showed that with increasing the magnetite dosage from 2880 mg/L to 3360 mg/L, the final turbidity

is reduced from 130 NTU to 20 NTU, while raising the magnetite dosage from 3360 mg/L to 4800 mg/L, the final turbidity is decreased from 20 NTU to 18 NTU. It was found that the turbidity removal efficiency at pH of 8.0 was superior than that at pH of 6. Akbar et al. [14] proved that turbidity removal is affected by pH, coagulant dosage, as well as initial turbidity. They found that the highest turbidity removal fall within 82-99.4% for initial turbidity of 10-1000NTU at pH of 5-7 and coagulant dose of 10-20mg/L. Ching and Zhen [15] conducted a study on magnetic seeding aggregation of high turbid source water as a pretreatment process using magnetite nanoparticles. The effect of pH on turbidity removal efficiency was studied over pH range of 5.0-9.0 and magnetic field strength of 0.0 Tesla to 0.1 Tesla. It was found that the final turbidity is decreased with the increase of the magnetite dosages. They got a turbidity of 774, 240, 56,19 and 10 NTU when using 1.0, 3.0, 5.0, 7.0 and 9.0 g/L magnetic dose. Their results showed that at pH values of 5.0, 6.0, 7.0, and 8.0 give the residual turbidities of 80, 234, 36 and 128 NTU, respectively. Mann [16] treated North Saskatchewan River water with different concentration of combination magnetite nanoparticles, aluminum sulfate and polyacrylamide. Turbidity test reported that, 300 mg/L magnetite nanoparticles has the highest removal efficiency of 98%. It was found that the required time for removing the turbidity using magnetite was 10 minutes, while by using aluminum sulfate and polyacrylamide combination, it was 30 minutes. Basma and Hussein [17] found that turbidity removal depends mainly on the coagulant dose, pH, and settling time. They found that the turbidity could be reduced from 92 to 2.1NTU at pH of 6, coagulant dose of 80 mg/L, and 120 minutes settling time. The feasibility of turbidity removal using a high gradient superconducting magnetic separation was studied by Ref. [18]. The process variables are, polyaluminum chloride (PAC) and magnetic seeds dosages. The initial turbidity of wastewater was 110 NTU, and the applied magnetic field intensity was 5.0 Tesla. A study regarding the use of a flocculated magnetic separation technology for treating Iraqi oilfield co-produced water for injection purpose was accomplished by Al-Rubaie et al. [19]. Results revealed that effluent water with low suspended solids and oil content can be obtained by applying a flocculation magnetic separation. It was also found that the required time for settling, several times less than that of the conventional methods. Treating of the emulsified oil wastewaters using a modified Fe_3O_4 magnetic nanoparticles MNPs, was made by Ref. [20]. A chitosan grafted magnetic nanoparticles Fe_3O_4 @APFS MNPs was used. They found a good demulsification effect via electrostatic attraction. It was also found that the demulsification performance could be further more enhanced upon Chitosan grafting especially under alkaline condition.

In the present study, an investigation on applying magnetic flocculation to treat wastewater of Al-Doura oil refinery using iron oxide, Nickel, and Cobalt magnetic powders with alum is made. The main objectives of this study are: Increasing the removal efficiency of the suspended solids and reducing settling time and consequently treating large quantities of polluted water without a need for enlarging the treatment basin.

2. EXPERIMENTAL PROGRAM

2.1. Apparatus and Procedures

2.1.1. Jar test

The Jar test apparatus was used in this study, is pharma test PT-DT7, it was taken from Samarra'a Company for drug and medical implementations (SDI).

2.1.2. Turbiditymeter

The turbidity of water samples were detected by HANNA turbidity meter.

2.1.3. pH-meter

The pH-meter that used in the present study is Jenway 3310.

2.1.4. Electrical Balance

Precisa XB 220A electrical balance. pH and turbidity were measured three times for each sample and the average values were registered.

2.2. Experimental Procedure

The experimental procedures are listed below:

1. Beakers of 1000 ml are filled with 500 ml of wastewater after measuring its initial turbidity and adjusting the pH to the required value using 1.0 N HCl or 1.0 N NaOH.
2. The required magnetic powder dose was mixed with the wastewater at mixing speed of 250 rpm for 1.0 minute.

3. The required alum dose was added with rapid mixing of 200 rpm for 1.0 minutes, followed by a slow mixing of 30 rpm for 10 minutes. Then, the mixers are turned off and the magnets are attached to the beaker bottom from the outside for 5.0 minutes.

4. Pipette water sample from the supernatant to measure the final turbidity.

Note: When alum is used alone, the settling time is 30 minutes while 5 minutes is a settling time for all magnetic powder.

2.3. Experimental Sets

Three sets of experiments were examined. In the first set, all experiments were conducted using a wastewater sample of initial turbidity 47.97 NTU, initial pH of 7.49, and temperature of 19.7 °C. Five levels of alum dose (60, 80, 100, 120, and 140 mg/L), three levels of pH (5.5, 6.5, and 7.5), and three levels of magnetic material dose (160, 200, 240mg/L) for each one of iron oxide nanoparticles (Fe₃O₄), nickel (Ni), and cobalt (Co) are performed. The second set was performed in order to test the possibility of reducing the dose of magnetic materials. In this set, wastewater samples which had an initial turbidity of 49 NTU, initial pH=7.60, and temperature equal to 23°C is used. In the third set the effect of initial turbidity (49, 61, 90, and 122 NTU) on the turbidity removal efficiency were tested after determining the best alum dose at pH of 7.

Wastewater samples had been taken from the industrial wastewater unit of Midland Refineries Company (Al-Doura Oil Refinery), precisely before the inlet of the coagulation-flocculation unit. Table 1 includes the operating variables for these sets.

Table 1
Operating parameters values of the present work.

Scheme	Sample No.	Wastewater properties				Alum dose, mg/L	pH	Magnetic powder dose, mg/L
		Turbidity, NTU	pH	TDS, mg/L	Temperature, °C			
First	1	47.97	7.49	1121	19.7	60,80,100,120,140	5.5, 6.5, 7.5	160, 200, 240
Second	2	49.00	7.60	1186	23.0	60,80,100,120,140	5.5, 6, 6.5, 7, 7.5, 8	40, 60, 80, 100, 120
Third	2	49.00	7.60	1186	23.0	120	7	Fe ₃ O ₄ , 120, Nickel, 80, Cobalt,100
Third	3	60.00	7.60	-	23.0	120	7	Fe ₃ O ₄ , 120, Nickel, 80, Cobalt,100
Third	4	90.00	7.60	-	23.0	120	7	Fe ₃ O ₄ , 120, Nickel, 80, Cobalt,100
Third	5	122.00	7.60	-	23.0	120	7	Fe ₃ O ₄ , 120, Nickel, 80, Cobalt,100

3. RESULTS AND DESCUSSION

3.1. Results of First Set

These results are listed in Tables 2-4 and samples of these results are shown graphically.

3.1.1. Effect of pH

Figures 1 and 2 represent the effect of pH on turbidity removal efficiency using alum alone and alum with 160 mg/L of iron oxide respectively. These Figures show that

low turbidity removal efficiencies are obtained at pH=5.5, while a high turbidity removal efficiencies are gained at pH=6.5 and pH=7.5. These pH values which give the highest turbidity removal are within the range of operating region for alum precipitation which is from 5.0 to 7.0 with minimum solubility occurring at pH equal to 6.0 [4]. Similar trend was obtained by [14, 17, 21]. Lo et al (2007) reported that the surface of the magnetite particles is positively charged at pH=6.0. Hence, at Fe₃O₄ doses equal to 160 mg/L and 200 mg/L the net charge of the wastewater will be positive so, a steric repulsion in the solution is occur, so high residual turbidity will remain, but at Fe₃O₄

dose =240 mg/L the weighting effect predominates and overcomes the electrostatic repulsion forces. For aluminum-based coagulants, the best coagulation performance is generally observed at pH values that are as close as possible to the pH of minimum solubility of the coagulant [22].

The optimum pH value depends on the treated water properties, coagulant type, and coagulant concentration [23]. Similar trend was obtained for all magnetic powder, as it is clear from Tables 2 - 4 which indicate that the higher removal efficiency for all magnetic powders was obtained at pH 6.5 and 7.5. It is also clear that 200 mg/L of Nickel with 60 mg/L alum at pH of 7.5 gave the highest removal of 98.45% while the highest removal for 240 mg/L iron oxide (97.89%) was obtained at pH of 6.5 and 80mg/L alum and the highest removal for 240 mg/L of Cobalt was (97.22%) obtained at pH of 6.5 and 100 mg/L alum.

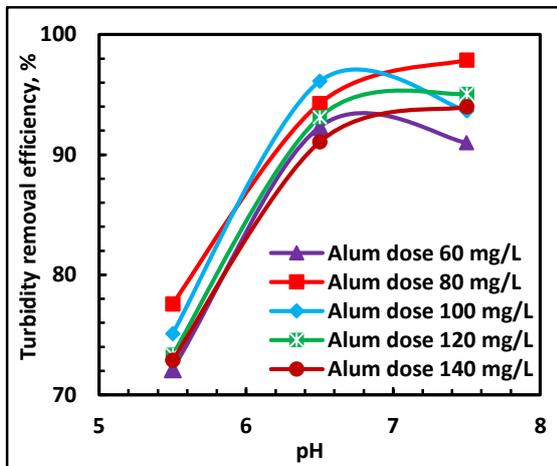


Fig. 1. Effect of pH on turbidity removal efficiency at different alum dose.

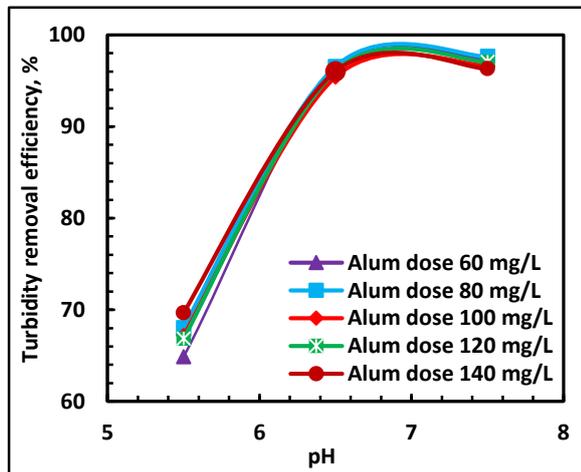


Fig. 2. Effect of pH on turbidity removal efficiency at different alum dose, Fe₃O₄ dose 160 mg/L.

3.1.2. Effect of Alum Dose

Figure 3 describes the influence of alum dose on turbidity removal efficiency at different pH values by applying alum only. It is clear that the removal efficiency increases with the increase of alum dose up to a certain limit then it drops. These results are in well agreement with that of [17, 24] who reported that colloidal particles are negatively charged and upon addition of aluminum sulfate, Al⁺³ ions are attracted to these particles. At the point of a

complete charges neutralization, the colloids begin to agglomerate due to a collisions between particles. If excess coagulant is added to the wastewater, the results are a reverse of the net charge on the colloidal particles (from negative to positive).

Particle re-stabilization by a reversal charge allowed greater amounts of smaller particles to remain in solution, thus increasing the total solids. Excess alum dose may exceed the saturation limit or produce excess aluminum hydroxide and thus will be a source of turbidity so the removal efficiency will be decreased [4]. The highest removal (96.12%) is obtained at 100 mg/L alum dose at pH of 6.5. The best removal (77.58%) at pH of 5.5 is obtained for alum dose of 80 mg/L, while at pH of 7.5 and alum dose of 120 mg/L the highest removal is 95.08%. These results show that the relationship between pH and alum dose is proportional. This may attribute to the alkalinity of the treated water. Metal coagulants are acidic, therefore, coagulant addition consumes alkalinity. In the case of pH =5.5 low dose of alum is required to get good results, since a high dose of alum will consume all the available alkalinity, lowering the pH to too low values for efficient treatment. When pH=7.5, high dose of alum is required to depress the pH (reduce the alkalinity) to a favorable values for coagulation. At pH=6.5, an optimum alum dose and a best removal efficiency are obtained. This value is within the operating region range for alum precipitation which is from 5.0-7.0 with minimum solubility occurring at pH equal to 6.0 [4].

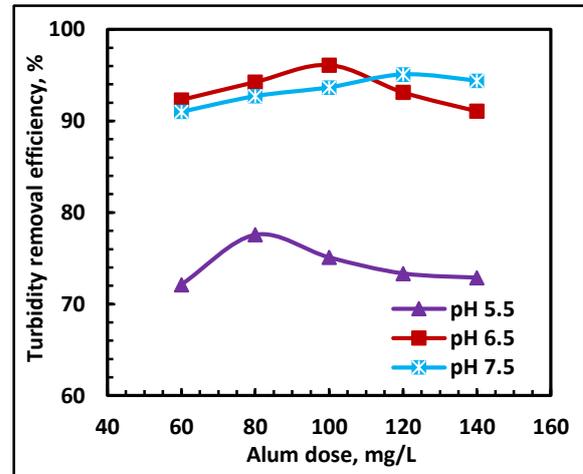


Fig. 3. Effect of alum dose on turbidity removal efficiency at different pH.

Figure 4 describes the influence of alum dose on turbidity removal efficiency at different pH values with the presence of 240 mg/L of Nickel. Inspection of this Figure and Tables 2-4 indicate that the general trend is nearly constant and the effect of alum dose with the presence of magnetic powder is little. Turbidity removal efficiency is increased slightly with the increase of alum dose at pH of 5.5 while it decreased slightly with the increase of alum dose at pH of 6.5 and 7.5. Moreover, the highest turbidity removal for alum alone or alum with any of the three magnetic materials is obtained at pH of 6.5 and 7.5 which are close to each other and the lowest removal was obtained at pH of 5.5. As mentioned previously, at low pH higher alum dose is required to get good results, since a high dose

of alum will consume all the available alkalinity, lowering the pH to too low values for efficient treatment. When pH=7.5, high dose of alum is required to depress the pH (reduce the alkalinity) to a favorable values for coagulation [4]. However, at pH of 6.5 and 7.5, there is a slight decrease of removal efficiency with the increase of alum dose. This is for two reasons; the first is excess alum dose may exceeds the saturation limit or produce excess aluminum hydroxide and thus will be a source of turbidity so the removal efficiency will be decreased [4] and the second is the fact that high magnetic powder dosage does not mean better efficiency, it becomes a source of turbidity that is extremely difficult to be removed without externally applied magnetic field. While at low dosage of magnetic powder, the effectiveness of magnetic aggregation will be poor [13]. This Figure show that the main effect is for pH and magnetic powder. Akbar et. al. (2010) stated that turbidity removal was relatively stable at all selected dosages greater than 10 mg/L when pH was kept constant, whereas turbidity removal seemed to be more influenced by pH variation than coagulant dosage. It is also clear that alum dose can be reduced from 140 and 100 mg/L when it is used alone at pH 7.5 and 6.5 respectively to as low as 60 mg/L when magnetic powder is added. This can reduce the excess cost of this process and satisfy one of the purposes of this work.

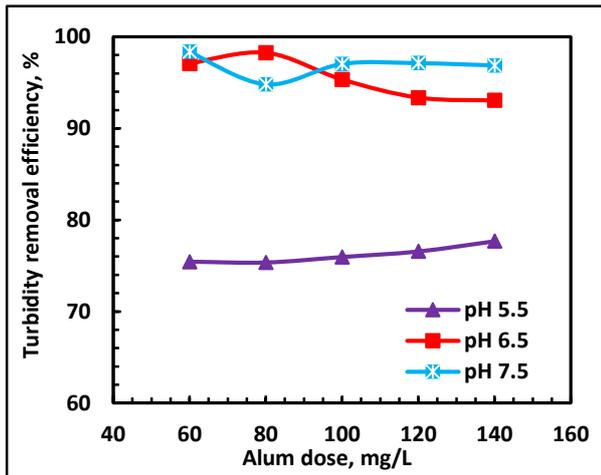


Fig. 4. Effect of alum dose on turbidity removal efficiency at different pH, Nickel dose 240 mg/L

3.1.3. Effect of Magnetic Powder Dose

Three levels of magnetic powder dose were used namely; 160, 200, and 240 mg/L. Figures 5–7 represent samples of the results for iron oxide, Nickel, and Cobalt that gives the highest removal efficiency respectively. A careful inspection of these figures and Tables 2–4 clarify that turbidity removal is increased with the increase of the magnetic powder dose up to a certain limit then it drops slightly. These results are in agreement with that of [25]. The lowest removal takes place at pH of 5.5 and the highest removal takes place at pH 6.5 or 7.5. Moreover, the removal at pH 6.5 and 7.5 are close together for all magnetic powder except Cobalt. Also, it could be found that the magnetic powder value that gives the highest removal depends on both pH and alum dose. The optimum performance for turbidity removal depends on pH, treated water properties, coagulant type, and coagulant concentration [23]. Appropriate magnetic powder dosage

is crucial, high dosage does not mean better efficiency, it becomes a source of turbidity that is extremely difficult to be removed without externally applied magnetic field, in addition to high amounts of sludge formation. While at low dosage of magnetic powder, the effectiveness of magnetic aggregation will be poor [13]. The highest turbidity removal of 98.45% is obtained when using 200mg/L of Nickel with 60 mg/L alum at pH of 7.5.

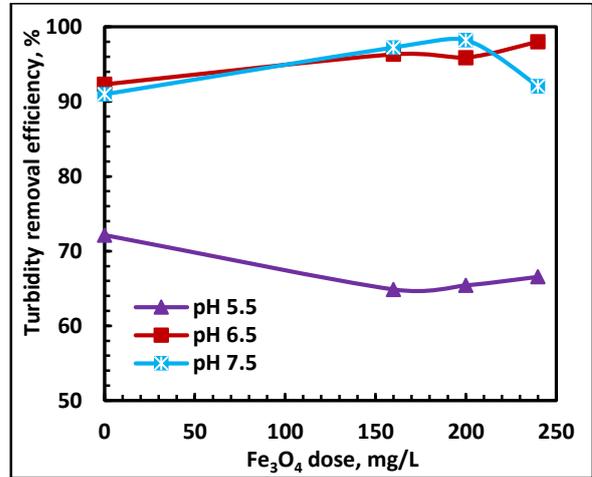


Fig. 5. Effect of Fe₃O₄ dose on turbidity removal efficiency at different pH, Alum dose 60 mg/L.

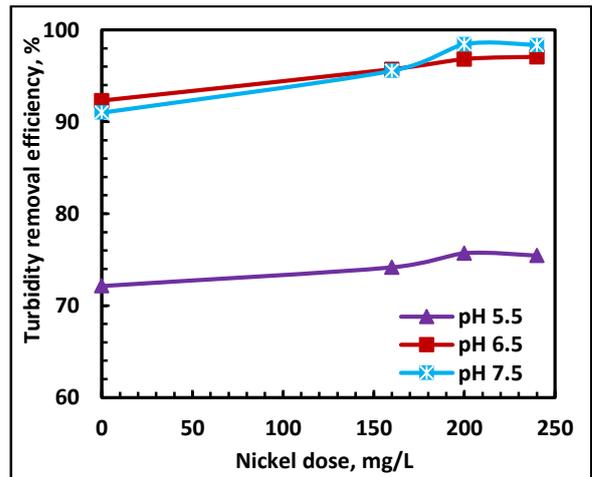


Fig. 6. Effect of Nickel dose on turbidity removal efficiency at different pH, Alum dose 60 mg/L.

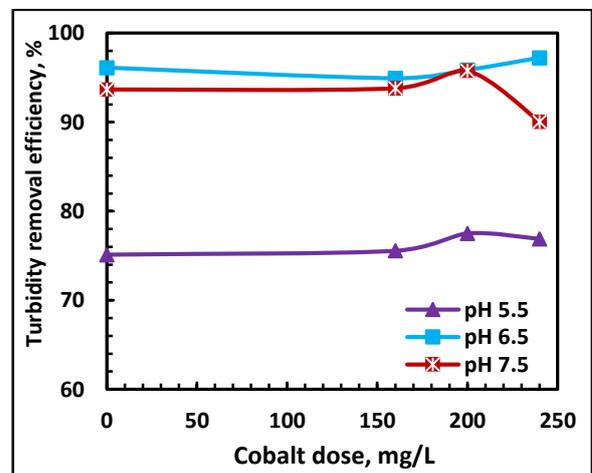


Fig. 7. Effect of Cobalt dose on turbidity removal efficiency at different pH, Alum dose 100 mg/L.

3.2. Second Set Results

Since the best pH is 6.5 and 7.5 according to the results of the first set, thus it was decided to take the average value (7) to determine the best alum dose at this average value of pH. It was found that 120 mg/L of alum gives the highest removal efficiency (Fig. 8). This alum dose is used to find the effect of pH on removal efficiency and it is found that the pH range 6.5-7.5 gives the highest removal. However at pH of 6.5, the highest removal is obtained (97.87%) (Fig. 9). This result is in agreement with that of [26]. Then, in a trial to test the possibility of reducing the magnetic powder dose, it is decided to use a range of 40 to 120 mg/L for each of the three magnetic powders. The results were graphed on Fig. 10. It is clear that at low doses of magnetic powders the removal efficiencies are low and it increases with the magnetic powder dose increase. At low dosage of magnetic powder, the effectiveness of magnetic aggregation will be poor [13]. It could be seen that the best Fe₃O₄ magnetic powder dose is 120 mg/L, while for nickel and cobalt they are 80 mg/L and 100 mg/L respectively. The optimum performance depends on pH, treated water properties, coagulant type, and coagulant concentration [23]. It could be concluded that nickel magnetic powder exhibits an excellent performance, where its optimum dose is low in comparison with iron oxide and cobalt and it gives a removal efficiency reaches to 98.57%.

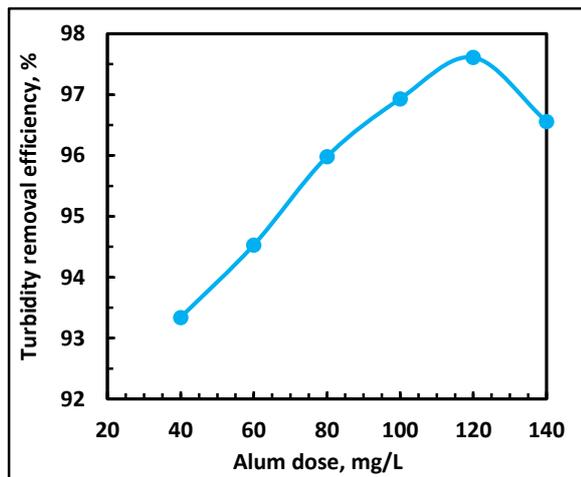


Fig. 8. Effect of alum dose on turbidity removal, pH=7.

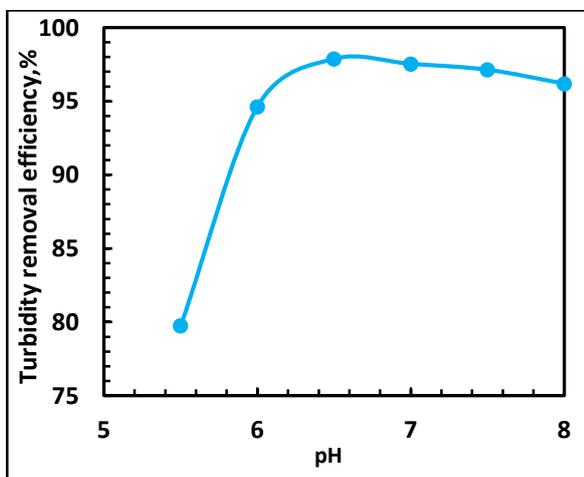


Fig. 9. Effect of pH on turbidity removal, Alum dose = 120 mg/L.

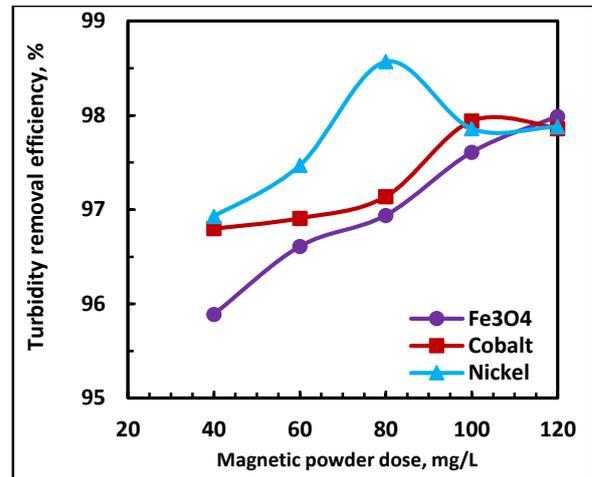


Fig. 10. Effect of magnetic powder dose on turbidity removal, alum dose = 120 mg/L & pH = 6.5.

3.3. Results of the Third Set

After finding the best conditions of alum dose, pH, and magnetic powder doses, an evaluation for using different initial turbidities with the best findings are illustrated in Figure 11.

It is clear that for alum alone, and alum with Fe₃O₄, the turbidity removal start to fall down at an initial turbidity of 61 and 90 NTU respectively, while Cobalt and Nickel still show good results at higher turbidities. This finding is in full agreement with that of [14] who stated that turbidity removal efficiency was decreased to a certain extent by increasing initial turbidity and application of higher coagulant dosage may improve turbidity removal from relatively high turbidity waters since high turbidity in addition to the dispersed Fe₃O₄ will need a lot of alum doses to neutralize their charges and overcome the mutual repulsion forces between suspended solids. They also found that there is an optimum magnetic dose for a specific initial turbidity range. However, when the raw water turbidity is altered, this optimal dosage will require an experimentally adjustment. They found that the optimum magnetic dose show a linear relationship with the initial turbidity. They stated that the coagulation process and turbidity removal was considerably effected by pH, coagulant dosage, as well as initial turbidity. Bahman (2014) reported that the increase in the turbidity removal with the increase of the initial turbidity might be attributed to other mechanisms such as sweeping flocculation rather than the neutralization of the surface charge of colloids.

For all experiment when using alum with magnetic powder, samples for the determination of removal efficiency are taken after 5 minutes while for alum only, samples are taken after 30 minutes settling. The removal efficiency when using alum (100 mg/L) and Fe₃O₄ (160 mg/L) at pH of 7.5 is 96.66% after 5 minutes settling while with employing alum only the turbidity removal efficiencies are 67.91% and 93.67% at settling periods of 5.0 minute and 30 minute respectively. This finding can give an increase of the treated volumes by 6 folds which is an essential matter for the field units.

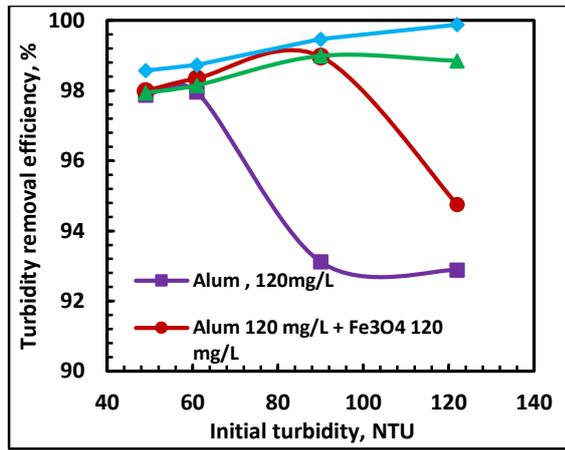


Fig.11. Effect of initial turbidity on turbidity removal at optimum conditions.

Finally, from the present work results, it could be concluded that, there is no specific value for the independent variables that gives the best results. Therefore, these values should be determined for each case depending on the experimental laboratory results. It is also revealed that the nickel magnetic powder with alum give the best

Table 2

Experimental results of the first set for Iron Oxide.

		Turbidity Removal Efficiency (%)											
		pH											
		5.5				6.5				7.5			
		Iron Oxide Dose (mg/L)											
		0.0	60	200	240	0.0	160	200	240	0.0	160	200	240
Alum dose, mg/L	60	72.13	4.88	65.42	66.56	92.31	96.32	95.89	98.01	91.00	97.23	98.22	92.07
	80	77.58	8.06	68.45	74.25	94.26	96.56	94.15	97.89	92.71	97.66	93.29	96.89
	100	75.11	7.17	65.03	72.88	96.12	95.40	93.10	94.04	93.67	96.66	95.74	96.78
	120	73.35	6.86	69.46	73.16	93.13	96.04	96.50	93.90	95.08	97.08	95.91	96.77
	140	72.89	9.67	67.50	76.19	91.07	97.01	94.36	91.53	94.39	96.32	95.31	97.25

Table 3

Experimental results of the first set for Nickel.

		Turbidity Removal Efficiency (%)											
		pH											
		5.5				6.5				7.5			
		Nickel Dose (mg/L)											
		0.0	160	200	240	0.0	160	200	240	0.0	160	200	240
Alum dose, mg/L	60	72.13	74.16	75.70	75.44	92.31	95.72	96.83	97.07	91.00	95.55	98.45	98.36
	80	77.58	74.40	75.26	75.35	94.26	96.79	95.88	98.26	92.71	97.07	97.99	94.81
	100	75.11	73.55	74.81	75.95	96.12	96.39	96.61	95.33	93.67	97.13	97.46	97.05
	120	73.35	73.59	75.58	76.56	93.13	96.86	96.01	93.36	95.08	97.10	96.74	97.13
	140	72.89	75.52	75.31	77.68	91.07	95.33	94.29	93.06	94.39	96.86	95.78	96.88

Table 4

Experimental results of the first set for Cobalt.

		Turbidity Removal Efficiency (%)											
		pH											
		5.5				6.5				7.5			
		Cobalt Dose (mg/L)											
		0.0	160	200	240	0.0	160	200	240	0.0	160	200	240
Alum dose, mg/L	60	72.13	74.89	76.73	77.73	92.31	97.10	95.37	96.55	91.00	87.44	90.53	89.21
	80	77.58	82.78	77.34	78.06	94.26	95.94	97.00	97.09	92.71	91.54	90.22	94.58
	100	75.11	75.55	77.52	76.89	96.12	94.93	95.86	97.22	93.67	93.80	95.78	90.05
	120	73.35	74.38	77.01	77.21	93.13	95.10	94.25	93.72	95.08	92.87	89.56	90.16
	140	72.89	74.81	76.26	77.72	91.07	93.76	94.60	92.42	94.39	93.32	90.66	93.66

results by a comparison with Fe₃O₄ and cobalt magnetic powders.

4. CONCLUSIONS

The main conclusions of the present study could be summarized as follows:

1. An enhancing in the turbidity removal efficiency is achieved by utilizing magnetic flocculation technique.
2. The required period for settling is very short (five minutes) in comparison with conventional method.
3. There is a potential for applying the same operating conditions for various initial turbidities, but in reasonable limits.
4. It can be concluded that nickel magnetic powder has a superior performance in comparison with iron oxide and cobalt magnetic powders.
5. The maximum turbidity removal efficiency is 99.88% when applying magnetic flocculation technology, while with applying conventional flocculation the maximum turbidity removal efficiency is 92.89 % at the same conditions.

REFERENCES

- [1] Louis EO. Water Requirements of the Petroleum Refining Industry. United States Government printing office, Washington 1963.
- [2] Pingping S, Amgad E, Michael W, Jeongwoo H, Robert JH. Estimation of U.S. refinery water consumption and allocation to refinery products. *Fuel*, Volume 221, 1 June 2018, 542-557
- [3] OPEC, Statistical Bulletin, 2014.
- [4] Metcalf and Eddy Inc. Wastewater Engineering: Treatment and Reuse, 52, McGraw Hill Series in Civil and Environmental Engineering. 4th Edition, 2003, McGraw-Hill, New York, 1819.
- [5] Mousa KM, Hadi HJ. Coagulation/flocculation process for produced water treatment. *International Journal of current Engineering and Technology*, 2016; **6**(2): 550-555.
- [6] Prakash N, Sockan V, Jayakaran P. Wastewater treatment by coagulation and flocculation. *International Journal of Engineering Science and Innovative Technology* 2014; **3**: 478-484.
- [7] Wang CR, Ren XL, Hou ZF, Ke C, Geng Q. Magnetic flocculation technology for copper and zinc ions removal from the tin smelting wastewater. *Applied Mechanics and Materials* 2013, Trans Tech Publ, 1284-1288.
- [8] Zhao Y, Liang W, Liu L, Li F, Fan Q, Sun X. Harvesting *Chlorella vulgaris* by magnetic flocculation using Fe₃O₄ coating with polyaluminium chloride and polyacrylamide. *Bioresour Technol*. 2015; **198**: 789-796.
- [9] Miura M, Matubayasi H, Iwai S. Waste water treatment method and apparatus. 1977, United States Patent No. 4,039,447.
- [10] Slusarczuk GM, Brooks RE. Ferrite flocculating system. 1980, United States Patent No. 4,193,866.
- [11] Kang YU, Taek K, Jong L, and Bom ST. Method for clarifying water by rapid flocculation and settling. 1995, Japan patent No. JP694192A.
- [12] Ching JM, Pei WC, and Li JW. Removal of nanoparticles from CMP wastewater by magnetic seeding aggregation. *Chemosphere* 2005, **63**, 1809-1813.
- [13] Lo SL, Wang YL, Hu CY. High turbidity reduction during the storm period by applied magnetic field. *Journal of Environmental Engineering and Management* 2007; **17**: 365-370.
- [14] Akbar B, Ali DZ, Nasser M, Abdolreza K. Optimizing coagulation process for low to high turbidity waters using aluminum and iron salts. *American Journal of Environmental Sciences* 2010; **6** (5): 442-448.
- [15] Ching JM, Zhen GF. Magnetic seeding aggregation of high turbid source water. *Journal of Environmental Engineering and Management* 2010; **20**: 145-150.
- [16] Mann AS. Removal of model waste-water bacteria by magnetite in water and waste-water treatment processes. M.Sc. Thesis; 2012, University of Alberta, Edmonton, Alberta.
- [17] Basma AA, Hussein BO. Evaluation of alum/lime coagulant for the removal of turbidity from al-ahdab iraqi oilfields produced water. *Journal of Engineering* 2015; **21**(7):145-153.
- [18] Zeng H, Li Y, Xu F, Jiang H, Zhang W. Feasibility of turbidity removal by high-gradient superconducting magnetic separation. *Environmental Technology* 2015; **36**:2495-2501.
- [19] Al-Rubaie MS, Dixon MA, Abbas TR. Use of flocculated magnetic separation technology to treat Iraqi oilfield co-produced water for injection purpose. *Desalination and Water Treatment* 2015; **53**: 2086-2091.
- [20] Lu T, Chen Y, Qi D, Cao Z, Zhang D, Zhao H. Treatment of emulsified oil wastewaters by using chitosan grafted magnetic nanoparticles. *Journal of Alloys and Compounds* 2017; **696**(C): 1205-1212.
- [21] Degremont. Water treatment handbook. 1979, 5th Ed; Distributed by Halsted Press N.Y.
- [22] Sahu O, Chaudhari P. Review on chemical treatment of industrial waste water. *Journal of Applied Sciences and Environmental Management* 2013; **17**: 241-257.
- [23] Farajnezhad H, Gharbani P. Coagulation treatment of wastewater in petroleum industry using polyaluminum chloride and ferric chloride. *International Journal of Research and Reviews in Applied Sciences* 2012; **13**: 306-310.
- [24] Ghaly AE, Snow A, Faber BE. Effective coagulation technology for treatment of grease filter wash water. *American Journal of Environmental Sciences* 2007; **3**(1): 19-29.
- [25] Alabdraba WM, Albayati MB, Ahmed YR, Mustafa MR. Influence of magnetic field on the efficiency of the coagulation process to remove turbidity from water. *International Review of Chemical Engineering* 2010; **5**(4):1-8.
- [26] Elleuch RS, Hammemi I, Khannous L, Nasri M, Gharsallah N. Wastewater treatment of bottle oil washing water (BOWW) by hybrid coagulation–flocculation and biological process. *Desalination and Water Treatment* 2014; **52**:1362-1369.
- [27] Bahman R. Treatment of water turbidity and bacteria by using a coagulant extracted from *Plantago ovate*. *Water Resources and Industry* 2014; **6**: 36-50.