

# Article Title Arial Size 20 Points not include abbreviation as possible

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## A B S T R A C T

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 as well as it requires relatively a long residence time. Magnetic flocculation is one of the used techniques for increase 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 (Fe<sub>3</sub>O<sub>4</sub>), 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:** Magnetic flocculation, oil refineries wastewater, nickel, cobalt, Iron oxide.

## عنوان البحث نوع الخط ايريل حجم 20 نقطة مع تجنب المختصرات

هاشم علي اسماعيل قسم الهندسة المدنية/ كلية هندسة / جامعة تكريت وسعد حامد محمد قسم الهندسة الكيميائية/كلية الهندسة / جامعة الموصل

### الخلاصة

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

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## 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 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. Slusarczyk 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  $\mu\text{m}$  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

64 aggregation (MSA) of silica nanoparticles was studied by Ref. [12]. Influences of pH, salts addition,  
65 and type of magnetite seeding particles on the turbidity removal efficiency were examined. The  
66 turbidity of CMP treated wastewater is reduced from 110.0 NTU to 7.0 NTU at pH of 6. The results  
67 showed that the residual turbidity decreases with the increase of magnetic field intensity. When the  
68 magnetic field intensity is higher than 0.08 Tesla, the residual turbidity is about 1.0 NTU. High  
69 turbidity reduction during the storm period by magnetic aggregation and separation was obtained by  
70 Ref. [13]. High turbidity raw water was prepared by mixing a sludge sample that are taken from  
71 Shiemen reservoir's tail water pond with deionized water. It was found that at magnetic field  
72 strength of 0.1 Tesla, the magnetic aggregation effects were not significant but at magnetic field  
73 strength of 0.15 Tesla, significant effects on the magnetic aggregation were observed. When the  
74 magnetic field strength raised to 0.2 Tesla, the effect on magnetic aggregation was stable. The  
75 results also showed that with increasing the magnetite dosage from 2880 mg/L to 3360 mg/L, the  
76 final turbidity is reduced from 130 NTU to 20 NTU, while raising the magnetite dosage from 3360  
77 mg/L to 4800 mg/L, the final turbidity is decreased from 20 NTU to 18 NTU. It was found that the  
78 turbidity removal efficiency at pH of 8.0 was superior than that at pH of 6. Akbar et al. [14] proved  
79 that turbidity removal is affected by pH, coagulant dosage, as well as initial turbidity. They found  
80 that the highest turbidity removal fall within 82-99.4% for initial turbidity of 10-1000NTU at pH of  
81 5-7 and coagulant dose of 10-20mg/L. Ching and Zhen [15] conducted a study on magnetic seeding  
82 aggregation of high turbid source water as a pretreatment process using magnetite nanoparticles.  
83 The effect of pH on turbidity removal efficiency was studied over pH range of 5.0-9.0 and magnetic  
84 field strength of 0.0 Tesla to 0.1 Tesla. It was found that the final turbidity is decreased with the  
85 increase of the magnetite dosages. They got a turbidity of 774, 240, 56,19 and 10 NTU when using  
86 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,  
87 and 8.0 give the residual turbidities of 80, 234, 36 and 128 NTU, respectively. Mann [16] treated  
88 North Saskatchewan River water with different concentration of combination magnetite  
89 nanoparticles, aluminum sulfate and polyacrylamide. Turbidity test reported that, 300 mg/L  
90 magnetite nanoparticles has the highest removal efficiency of 98%. It was found that the required  
91 time for removing the turbidity using magnetite was 10 minutes, while by using aluminum sulfate  
92 and polyacrylamide combination, it was 30 minutes. Basma and Hussein [17] found that turbidity  
93 removal depends mainly on the coagulant dose, pH, and settling time. They found that the turbidity  
94 could be reduced from 92 to 2.1NTU at pH of 6, coagulant dose of 80 mg/L, and 120 minutes  
95 settling time. The feasibility of turbidity removal using a high gradient superconducting magnetic  
96 separation was studied by Ref. [18]. The process variables are, polyaluminum chloride (PAC) and  
97 magnetic seeds dosages. The initial turbidity of wastewater was 110 NTU, and the applied magnetic  
98 field intensity was 5.0 Tesla. A study regarding the use of a flocculated magnetic separation

99 technology for treating Iraqi oilfield co-produced water for injection purpose was accomplished by  
100 Al-Rubaie et al. [19]. Results revealed that effluent water with low suspended solids and oil content  
101 can be obtained by applying a flocculation magnetic separation. It was also found that the required  
102 time for settling, several times less than that of the conventional methods. Treating of the emulsified  
103 oil wastewaters using a modified Fe<sub>3</sub>O<sub>4</sub> magnetic nanoparticles MNPs, was made by Ref. [20]. A  
104 chitosan grafted magnetic nanoparticles Fe<sub>3</sub>O<sub>4</sub> @APFS MNPs was used. They found a good  
105 demulsification effect via electrostatic attraction. It was also found that the demulsification  
106 performance could be further more enhanced upon Chitosan grafting especially under alkaline  
107 condition.

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

113

## 114 **2. EXPERIMENTAL PROGRAM**

### 115 **2.1. Apparatus and Procedures**

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

### 118 **2.2. Experimental Procedure**

119 The experimental procedures are listed below:

- 120 1. Beakers of 1000 ml are filled with 500 ml of wastewater after measuring its initial turbidity and  
121 adjusting the pH to the required value using 1.0 N HCl or 1.0 N NaOH.
- 122 2. The required magnetic powder dose was mixed with the wastewater at mixing speed of 250 rpm  
123 for 1.0 minute.
- 124 3. The required alum dose was added with rapid mixing of 200 rpm for 1.0 minutes, followed by a  
125 slow mixing of 30 rpm for 10 minutes. Then, the mixers are turned off and the magnets are  
126 attached to the beaker bottom from the outside for 5.0 minutes.
- 127 4. Pipette water sample from the supernatant to measure the final turbidity.

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

### 130 **2.3. Experimental Sets**

131 Three sets of experiments were examined. In the first set, all experiments were conducted using  
 132 a wastewater sample of initial turbidity 47.97 NTU, initial pH of 7.49, and temperature of 19.7  
 133 °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),  
 134 and three levels of magnetic material dose (160, 200, 240mg/L) for each one of iron oxide  
 135 nanoparticles (Fe<sub>3</sub>O<sub>4</sub>), nickel (Ni), and cobalt (Co) are performed. The second set was performed in  
 136 order to test the possibility of reducing the dose of magnetic materials. In this set, wastewater  
 137 samples which had an initial turbidity of 49 NTU, initial pH=7.60, and temperature equal to 23°C is  
 138 used. In the third set the effect of initial turbidity (49, 61, 90, and 122 NTU) on the turbidity  
 139 removal efficiency were tested after determining the best alum dose at pH of 7.

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

143  
 144 **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	$e_s$			
First	1	47.97	7.49	112 1	19.7	60,80,100,120, 140	5.5, 6.5, 7.5	160, 200, 240
Second	2	49.00	7.60	118 6	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	118 6	23.0	120	7	Fe <sub>3</sub> O <sub>4</sub> , 120, Nickel, 80, Cobalt,100
Third	3	60.00	7.60	-	23.0	120	7	Fe <sub>3</sub> O <sub>4</sub> , 120, Nickel, 80, Cobalt,100
Third	4	90.00	7.60	-	23.0	120	7	Fe <sub>3</sub> O <sub>4</sub> , 120, Nickel, 80, Cobalt,100
Third	5	122.0 0	7.60	-	23.0	120	7	Fe <sub>3</sub> O <sub>4</sub> , 120, Nickel, 80, Cobalt,100

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### 146 3. RESULTS AND DESCUSSION

#### 147 3.1. Results of First Set

148 These results are listed in [Tables 2-4](#) and samples of these results are shown graphically.  
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**Table 2** Experimental results of the first set for Iron Oxide.

<b>Turbidity Removal Efficiency (%)</b>												
<b>pH</b>												
<b>5.5</b>				<b>6.5</b>				<b>7.5</b>				
<b>Iron Oxide Dose (mg/L)</b>												
	<b>0.0</b>	<b>60</b>	<b>200</b>	<b>240</b>	<b>0.0</b>	<b>160</b>	<b>200</b>	<b>240</b>	<b>0.0</b>	<b>160</b>	<b>200</b>	<b>240</b>
60	72.13	74.88	65.42	66.56	92.31	96.32	95.89	98.01	91.00	97.23	98.22	92.07
80	77.58	78.06	68.45	74.25	94.26	96.56	94.15	97.89	92.71	97.66	93.29	96.89
100	75.11	77.17	65.03	72.88	96.12	95.40	93.10	94.04	93.67	96.66	95.74	96.78
120	73.35	76.86	69.46	73.16	93.13	96.04	96.50	93.90	95.08	97.08	95.91	96.77
140	72.89	79.67	67.50	76.19	91.07	97.01	94.36	91.53	94.39	96.32	95.31	97.25

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**Table 3.** Experimental results of the first set for Nickel.

<b>Turbidity Removal Efficiency (%)</b>												
<b>pH</b>												
<b>5.5</b>				<b>6.5</b>				<b>7.5</b>				
<b>Nickel Dose (mg/L)</b>												
	<b>0.0</b>	<b>160</b>	<b>200</b>	<b>240</b>	<b>0.0</b>	<b>160</b>	<b>200</b>	<b>240</b>	<b>0.0</b>	<b>160</b>	<b>200</b>	<b>240</b>
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

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**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

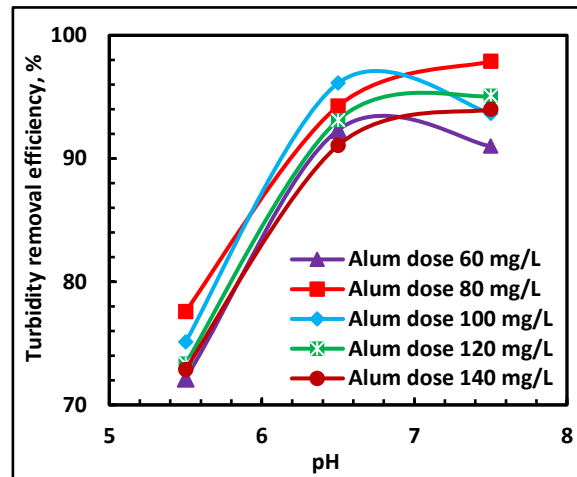
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165 **Figs. 1** and **2** represent the effect of pH on turbidity removal efficiency using alum alone and  
166 alum with 160 mg/L of iron oxide respectively. These Figures show that low turbidity removal  
167 efficiencies are obtained at pH=5.5, while a high turbidity removal efficiencies are gained at  
168 pH=6.5 and pH=7.5. These pH values which give the highest turbidity removal are within the range  
169 of operating region for alum precipitation which is from 5.0 to 7.0 with minimum solubility  
170 occurring at pH equal to 6.0 [4]. Similar trend was obtained by [14, 17, 21]. Lo et al (2007)  
171 reported that the surface of the magnetite particles is positively charged at pH=6.0. Hence, at Fe<sub>3</sub>O<sub>4</sub>  
172 doses equal to 160 mg/L and 200 mg/L the net charge of the wastewater will be positive so, a steric  
173 repulsion in the solution is occur, so high residual turbidity will remain, but at Fe<sub>3</sub>O<sub>4</sub> dose =240  
174 mg/L the weighting effect predominates and overcomes the electrostatic repulsion forces. For  
175 aluminum-based coagulants, the best coagulation performance is generally observed at pH values  
176 that are as close as possible to the pH of minimum solubility of the coagulant [22].  
177 The optimum pH value depends on the treated water properties, coagulant type, and coagulant  
178 concentration [23]. Similar trend was obtained for all magnetic powder, as it is clear from **Tables 2 -**  
179 **4** which indicate that the higher removal efficiency for all magnetic powders was obtained at pH 6.5  
180 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  
181 removal of 98.45% while the highest removal for 240 mg/L iron oxide (97.89%) was obtained at pH  
182 of 6.5 and 80mg/L alum and the highest removal for 240 mg/L of Cobalt was (97.22%) obtained at  
183 pH of 6.5 and 100 mg/L alum. Effect of pH on turbidity removal efficiency at different alum dose  
184 can be shown in Fig.2.

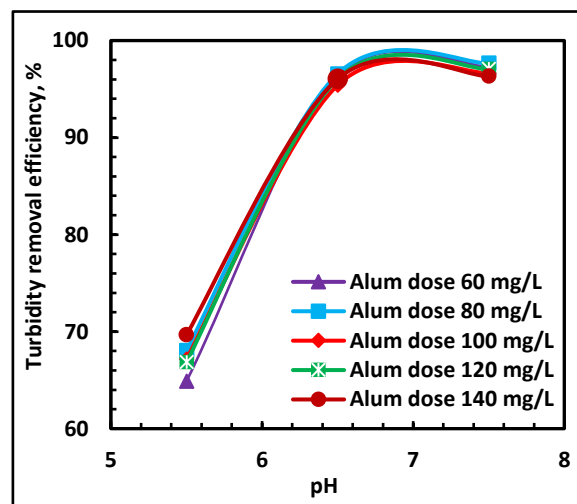
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**Fig. 1.** Effect of pH on turbidity removal efficiency at differ



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**Fig. 2.** Effect of pH on turbidity removal efficiency at different alum dose, Fe<sub>3</sub>O<sub>4</sub> dose 160 mg/L.

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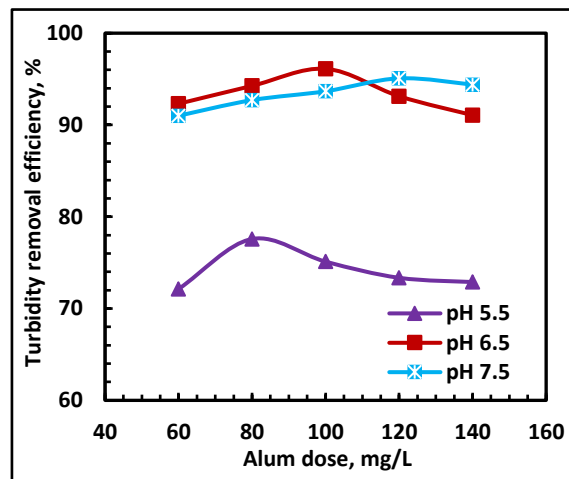
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Fig. 3 describes the influence of alum dose on turbidity removal efficiency at different pH values by applying alum only. It is clear that the removal efficiency increases with the increase of alum dose up to a certain limit then it drops. These results are in well agreement with that of [17, 24] who reported that colloidal particles are negatively charged and upon addition of aluminum sulfate, Al<sup>3+</sup> 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 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]. The highest removal (96.12%) is obtained at 100 mg/L alum dose

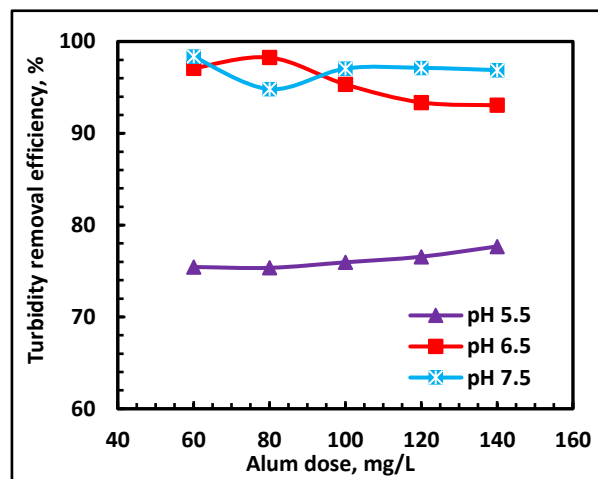
203 at pH of 6.5. The best removal (77.58%) at pH of 5.5 is obtained for alum dose of 80 mg/L, while  
 204 at pH of 7.5 and alum dose of 120 mg/L the highest removal is 95.08%. These results show that the  
 205 relationship between pH and alum dose is proportional. This may attribute to the alkalinity of the  
 206 treated water. Metal coagulants are acidic, therefore, coagulant addition consumes alkalinity. In the  
 207 case of  
 208 pH =5.5 low dose of alum is required to get good results, since a high dose of alum will consume all  
 209 the available alkalinity, lowering the pH to too low values for efficient treatment. When pH=7.5,  
 210 high dose of alum is required to depress the pH (reduce the alkalinity) to a favorable values for  
 211 coagulation. At pH=6.5, an optimum alum dose and a best removal efficiency are obtained. This  
 212 value is within the operating region range for alum precipitation which is from 5.0-7.0 with  
 213 minimum solubility occurring at pH equal to 6.0 [4].



214  
 215 **Fig. 3.** Effect of alum dose on turbidity removal efficiency at different pH.

216 **Fig. 4** describes the influence of alum dose on turbidity removal efficiency at different pH  
 217 values with the presence of 240 mg/L of Nickel. Inspection of this Figure and **Tables 2–4** indicate  
 218 that the general trend is nearly constant and the effect of alum dose with the presence of magnetic  
 219 powder is little. Turbidity removal efficiency is increased slightly with the increase of alum dose at  
 220 pH of 5.5 while it decreased slightly with the increase of alum dose at pH of 6.5 and 7.5. Moreover,  
 221 the highest turbidity removal for alum alone or alum with any of the three magnetic materials is  
 222 obtained at pH of 6.5 and 7.5 which are close to each other and the lowest removal was obtained at  
 223 pH of 5.5. As mentioned previously, at low pH higher alum dose is required to get good results,  
 224 since a high dose of alum will consume all the available alkalinity, lowering the pH to too low  
 225 values for efficient treatment. When pH=7.5, high dose of alum is required to depress the pH  
 226 (reduce the alkalinity) to a favorable values for coagulation [4]. However, at pH of 6.5 and 7.5,  
 227 there is a slight decrease of removal efficiency with the increase of alum dose. This is for two

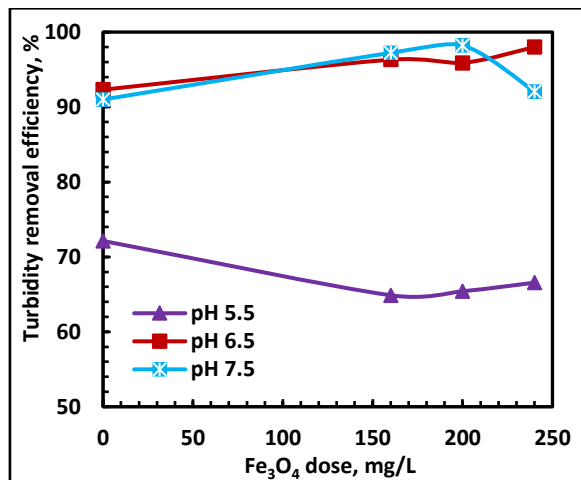
228 reasons; the first is excess alum dose may exceeds the saturation limit or produce excess aluminum  
 229 hydroxide and thus will be a source of turbidity so the removal efficiency will be decreased [4] and  
 230 the second is the fact that high magnetic powder dosage does not mean better efficiency, it becomes  
 231 a source of turbidity that is extremely difficult to be removed without externally applied magnetic  
 232 field. While at low dosage of magnetic powder, the effectiveness of magnetic aggregation will be  
 233 poor [13]. This Figure show that the main effect is for pH and magnetic powder. Akbar et. al.  
 234 (2010) stated that turbidity removal was relatively stable at all selected dosages greater than 10  
 235 mg/L when pH was kept constant, whereas turbidity removal seemed to be more influenced by pH  
 236 variation than coagulant dosage. It is also clear that alum dose can be reduced from 140 and 100  
 237 mg/L when it is used alone at pH 7.5 and 6.5 respectively to as low as 60 mg/L when magnetic  
 238 powder is added. This can reduce the excess cost of this process and satisfy one of the purposes of  
 239 this work.



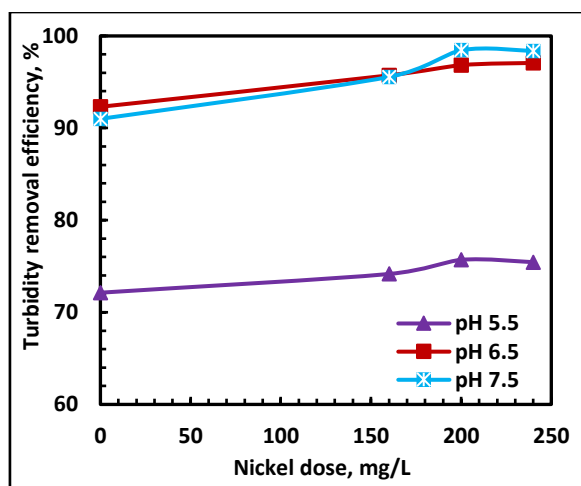
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 241 **Fig. 4.** Effect of alum dose on turbidity removal efficiency at different pH, Nickel dose 240 mg/L

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 243 Three levels of magnetic powder dose were used namely; 160, 200, and 240 mg/L. Figs. 5–7  
 244 represent samples of the results for iron oxide, Nickel, and Cobalt that gives the highest removal  
 245 efficiency respectively. A careful inspection of these figures and Tables 2–4 clarify that turbidity  
 246 removal is increased with the increase of the magnetic powder dose up to a certain limit then it  
 247 drops slightly. These results are in agreement with that of [25]. The lowest removal takes place at  
 248 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  
 249 7.5 are close together for all magnetic powder except Cobalt. Also, it could be found that the  
 250 magnetic powder value that gives the highest removal depends on both pH and alum dose. The  
 251 optimum performance for turbidity removal depends on pH, treated water properties, coagulant  
 252 type, and coagulant concentration [23]. Appropriate magnetic powder dosage is crucial, high dosage  
 253 does not mean better efficiency, it becomes a source of turbidity that is extremely difficult to be

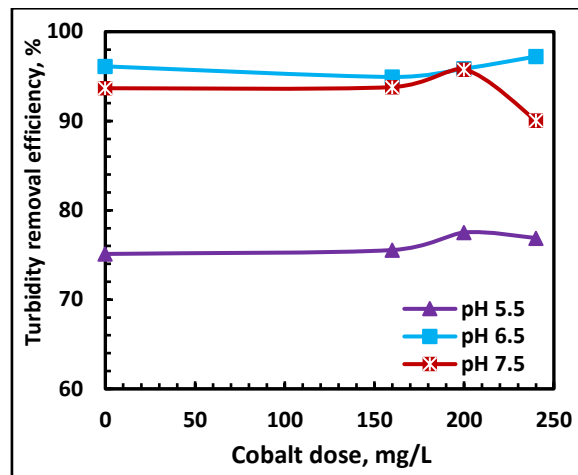
254 removed without externally applied magnetic field, in addition to high amounts of sludge formation.  
255 While at low dosage of magnetic powder, the effectiveness of magnetic aggregation will be poor  
256 [13]. The highest turbidity removal of 98.45% is obtained when using 200mg/L of Nickel with 60  
257 mg/L alum at pH of 7.5.  
258



259  
260 **Fig. 5.** Effect of Fe<sub>3</sub>O<sub>4</sub> dose on turbidity removal efficiency at different pH, Alum dose 60 mg/L.



261  
262 **Fig. 6.** Effect of Nickel dose on turbidity removal efficiency at different pH, Alum dose 60 mg/L.  
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266 **Fig. 7.** Effect of Cobalt dose on turbidity removal efficiency at different pH, Alum dose 100 mg/L.

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268 **3.2. Second Set Results**

269 Since the best pH is 6.5 and 7.5 according to the results of the first set, thus it was decided to  
 270 take the average value (7) to determine the best alum dose at this average value of pH. It was found  
 271 that 120 mg/L of alum gives the highest removal efficiency (Fig. 8). This alum dose is used to find  
 272 the effect of pH on removal efficiency and it is found that the pH range 6.5-7.5 gives the highest  
 273 removal. However at pH of 6.5, the highest removal is obtained (97.87%)(Fig. 9).

274 This result is in agreement with that of [26]. Then, in a trial to test the possibility of reducing the  
 275 magnetic powder dose, it is decided to use a range of 40 to 120 mg/L for each of the three magnetic  
 276 powders. The results were graphed on Fig. 10. It is clear that at low doses of magnetic powders the  
 277 removal efficiencies are low and it increases with the magnetic powder dose increase. At low  
 278 dosage of magnetic powder, the effectiveness of magnetic aggregation will be poor [13]. It could  
 279 be seen that the best Fe<sub>3</sub>O<sub>4</sub> magnetic powder dose is 120 mg/L, while for nickel and cobalt they are  
 280 80 mg/L and 100 mg/L respectively. The optimum performance depends on pH, treated water  
 281 properties, coagulant type, and coagulant concentration [23]. It could be concluded that nickel  
 282 magnetic powder exhibits an excellent performance, where its optimum dose is low in comparison  
 283 with iron oxide and cobalt and it gives a removal efficiency reaches to 98.57%. Effect of pH on  
 284 turbidity removal, Alum dose = 120 mg/L can be shown in Fig.9.

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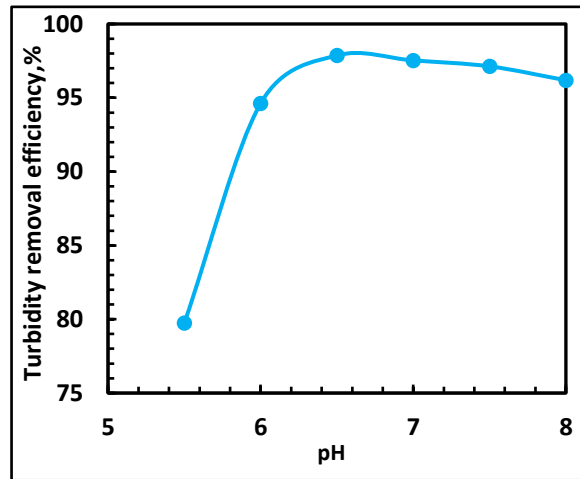
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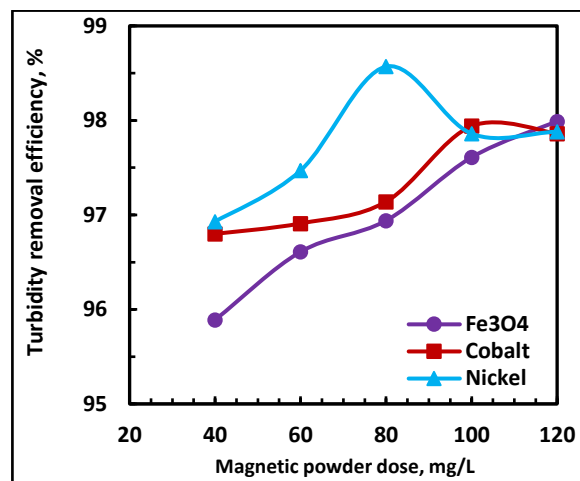
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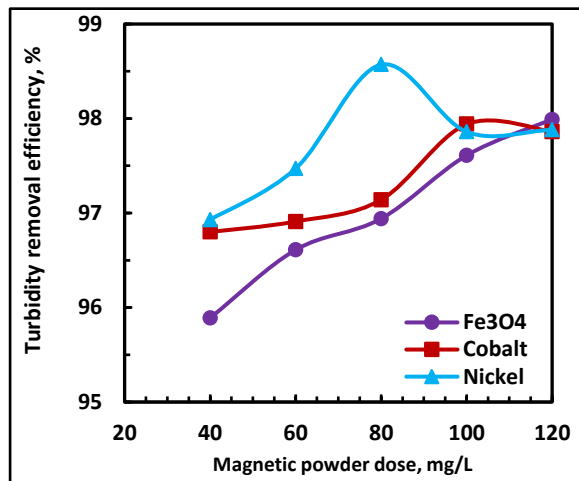


**Fig. 8.** Effect of alum dose on turbidity removal, pH=7.



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

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**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 Fig. 11.

It is clear that for alum alone, and alum with Fe<sub>3</sub>O<sub>4</sub>, 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 Fe<sub>3</sub>O<sub>4</sub> 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 experimental 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 Fe<sub>3</sub>O<sub>4</sub> (160 mg/L) at pH

352 of 7.5 is 96.66% after 5 minutes settling while with employing alum only the turbidity removal  
353 efficiencies are 67.91% and 93.67% at settling periods of 5.0 minute and 30 minute respectively.  
354 This finding can give an increase of the treated volumes by 6 folds which is an essential matter for  
355 the field units.

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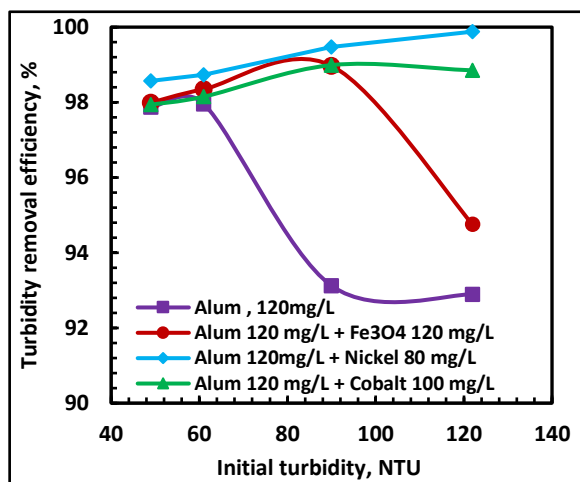
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364 **Fig.11.** Effect of initial turbidity on turbidity removal at optimum conditions.

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366 Finally, from the present work results, it could be concluded that, there is no specific value for  
367 the independent variables that gives the best results. Therefore, these values should be determined  
368 for each case depending on the experimental laboratory results. It is also revealed that the nickel  
369 magnetic powder with alum give the best results by a comparison with  $Fe_3O_4$  and cobalt magnetic  
370 powders.

#### 371 4. CONCLUSIONS

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

373 An enhancing in the turbidity removal efficiency is achieved by utilizing magnetic flocculation  
374 technique.

375 The required period for settling is very short (five minutes) in comparison with conventional method.

376 There is a potential for applying the same operating conditions for various initial turbidities, but in  
377 reasonable limits.

378 It can be concluded that nickel magnetic powder has a superior performance in comparison with iron  
379 oxide and cobalt magnetic powders.



380 The maximum turbidity removal efficiency is 99.88% when applying magnetic flocculation technology,  
381 while with applying conventional flocculation the maximum turbidity removal efficiency is 92.89 % at  
382 the same conditions.

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## 385 **NOMENCLATURE**

386 IN THIS PLACE OR AT THE BEGINNING BEFORE THE INTRODUCTION

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