

Optimal Coagulation/Flocculation Process For Water Treatment Plants Located On Damietta Branch Of River Nile, Egypt

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ABSTRACT: A coagulation-flocculation process, using three different phases of aluminum derivatives, namely; Alum $Al_2(SO_4)_3 \cdot 18H_2O$, Poly-aluminum chloride (PAC) $[Al_2(OH)_xCl_{6-x}]_n$; Potassium aluminum sulfate (PAS) $KAl(SO_4)_2 \cdot 12H_2O$ have been studied, respectively. Optimum operating conditions have been established to achieve the optimal parameters and to achieve the maximum removal of pollutants in the constructed water treatment plants. River Nile, Egypt is the main source for drinking water and major other activities. Nile water contains suspended solids and colloidal particles that are normally treated by coagulation-flocculation followed by clarification. Alum, PAC and PAS have been studied to establish the optimum suspension removal, which are the most common types of coagulants in water treatment plants in Egypt as well in many other countries. Coagulants were investigated with the aim of determining their capabilities to reduce turbidity and other contaminants in drinking water to the required drinking standards. In Egypt; conventional drinking water treatment plants are including pre-chlorination, coagulation/flocculation, filtration and disinfection which didn't improve the utilization of coagulation that led to high consumption of coagulant and coagulant aid, pH adjustment, high residual aluminum, high cost treatment and mass production of sludge. The effect of seasonal variation, including temperature, pH fluctuation, alkalinity, pre-chlorination, and coagulant's doses relative to Zeta potential have been studied to find out the optimal operating conditions.

Keywords : Coagulation-flocculation; Colloidal materials; Turbidity, Alum, Ferric chloride, Poly-aluminum chloride

Objective: The trend of urbanization in Egypt is exerting convince on civic authorities to provide basic requirement such as safe drinking water, and sanitation. The primary objective is to ensuring good drinking-water supply practice with minimizing any contamination of water source. The removal of contamination was through treatment processes and prevention of contamination during storage, distribution and handling of the treated drinking water.

1 INTRODUCTION

The most effective means of consistently ensuring the safety of a drinking water supply is through the use of a comprehensive risk assessment and risk management approach that encompasses all steps in water supply from catchment to consumer. Optimization of conventional drinking water treatment plant means "to attain the most efficient or effective use" of water treatment plant regarding some principles, there are: achievement of consistently high quality finished water on a continuous basis and the importance to focus on overall plant performance, instead of focusing too much on individual processes [1]. The River Nile with its two branches is the main water source in Egypt. It was recently exposed to draught, contamination and pollution sources that affect its water quality as a result of uncontrolled disposal of industrial and agricultural wastewater. This is in addition to rapid population growth and bad habits for water use. Consequently, the conventional water treatment technology has to be developed to face this water demand and requirements. Conventional Zifta water treatment plant (ZWTP) under investigation was proposed to be upgraded, while Figure (1) is representing an Aerial photo. The ZWTP is including pre-chlorination, coagulation/flocculation, filtration and finally disinfection. Figure (2) shows a schematic diagram of the treatment process along the existing treatment plant. Coagulation/ flocculation process as a conventional is of great importance in solid liquid separation practice. Its role is to remove Turbidity due to the presence suspended solids, colloidal matters, fine particles, as well as sand and silt from the raw water. This followed by sedimentation and filtration steps, while the dissolved molecules can't be removed by conventional treatment [2].



Figure 1 Aerial Diagram of Zifta Drinking Water Treatment Plant (ZWTP)

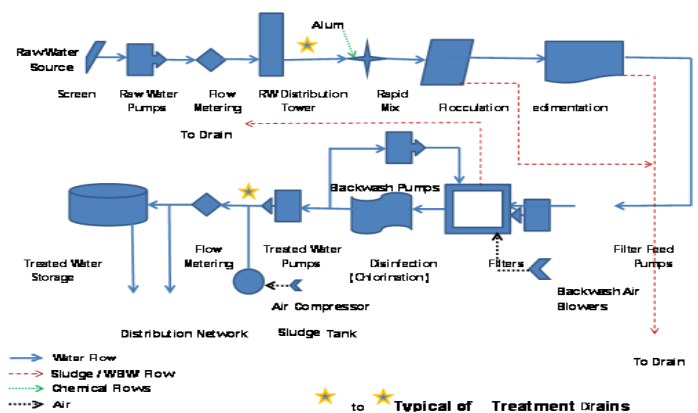


Figure 2 Schematic diagram of water treatment process at ZWTP

Also, Giani, 2011 showed that a coagulation/flocculation process, based on aluminum sulfate, has been studied by using Response Surface Methodology (RSM), in order to establish the optimum parameters to achieve a maximum suspensions removal: Al dosage, initial turbidity and water pH. The RSM is an useful tool, which allows a treatment plant operator, to determine easily, after a small number of trials, the optimum conditions to achieve a maximum suspensions removal efficiency [3]. Ferric chloride and alum, which are the most common types of coagulants in water treatment plants, were investigated to reduce turbidity of drinking water. Turbidity removal efficiency was higher for ferric chloride compared to aluminum sulfate at their optimum conditions. Results demonstrated that coagulation process can assure turbidity removal from low to medium turbidity waters effectively, using relatively low levels of aluminum sulfate and ferric chloride (10-20 mg/l). Turbidity removal efficiency still remained high when the initial turbidities of water were increased [4]. Koohestanian et al, 2008 found that on using Alum and ferric chloride as a coagulant followed by anionic polymer as coagulant aid, the process that changed the particles from nano-scale to micro-scale and larger by a physico-chemical process. The influence of pH, temperature, coagulant and coagulant aid dosages on the coagulation process was studied and conditions were optimized corresponding to the best removal of organic matters, viruses, colloids, bacteria, color and decrease in turbidity. Ferric chloride produced better results than alum. Higher dosages did not significantly increase pollutant removal and were not economical [5]. Moreover Baroniya et al, 2012, showed that conventional treatment is provided having a sequence of alum addition, coagulation, flocculation, sedimentation, filtration and disinfection by chlorination. Water treatment plants are playing an important role in purifying and supplying the pure water to the people. The operation and maintenance needs to be updated for the current requirements of people and to match up with some other plants at national and international level [6]. For a very long time, aluminum sulfate was a well known coagulant, being used in water and wastewater treatment as a preferred reagent. Usage of aluminum sulfate is based on several advantages, as low cost, high efficiency at low doses, low toxicity and high availability [7]. It was also agree with the EPA Guidance Manual Turbidity Provisions which stated that the coagulation/flocculation purpose is to remove colloidal suspensions, as inorganic form clays and silts, or organic compounds, which could be a good support for pathogens development, and presents a great threat to drinking water aspect [8]. In general; the efficiency and mechanism of coagulation-flocculation process is depend on several factors, the most relevant being initial turbidity, pH, coagulant, dosage and type, polymer, system hydrodynamics in coagulation and flocculation stages, temperature, and alkalinity [9]. According to Water Safety Plan Guide [2014]; Conventional Coagulation/Flocculation/ Sedimentation Version 1, Reliable information about water quality is essential for the proper management of a water supply. Knowledgeable and skilled staffs are also essential for minimizing the public health risks associated with water supplies [10].

2. Experimental Section; Materials and Utilities:

2.1. Materials: The coagulants used were:

- Liquid Aluminum sulfate Octadeca-hydrate 50% $[Al_2(SO_4)_3 \cdot 18H_2O]$;

- Poly aluminum chloride (PAC) $[Al_2(OH)xCl_{6-x}]_n$; and
 - Potassium aluminum sulfate (PAS) $AlK(SO_4)_2 \cdot 12H_2O$,
 - Nalclear 8173 PULV (polymer) as a coagulant aid;
- Tables 1 and 2 show chemical structures & specifications.

Table 1 Chemical structures and specifications

Specification	Unit	Alum *	PAC	PAS
Appearance	= =	Clear	yellow	white
Insol matters	%	≤ 0.02	≤ 0.2	NI
Density, 20 °C	g/ml	1.33	1.18	1.757
pH-value	= =	3.10	3.00 min	3.3
Al ₂ O ₃ content	%	8.13	8.9	NI
Molecular Wt	g	666	NI	474.4
Molecular Formula	= =	Al ₂ (SO ₄) ₃ · 18H ₂ O	[Al ₂ (OH) _x Cl _{6-x}] _n	KAl(SO ₄) ₂ · 12H ₂ O
Iron	%	0.10	0.35 max	NI
Nitrogen, N ₂	%	0.003	0.01 max	NI
Arsenic, As	mg /kg	8	0.2	NI
Cd	mg /kg	20	0.02	NI
Cr	mg /kg	400	0.05	NI
Hg	mg /kg	9	0.001	NI
Pb	mg /kg	300	0.01	NI
Ni	mg /kg	300	NI	NI
Sb	mg /kg	N.D.	NI	NI
Se	mg /kg	0.12	NI	NI

*Alum: Egyptian Co, NI: not identified, N.D: not detected

Table 2 Anionic polymer coagulant aid specifications

Chemical Name	Formula	Mol Wt
Hydrolyzed Polyacrylamide	$[CH_2-CH-CO-NH_2]_{n-m}$ ---- $[CH_2-CH-COO]_n$	10 ⁴ - 10 ⁷

The following instruments were used as:

2.1.1. Turbidity NTU has been determined by Jenway 6405 UV-VIS spectrophotometer, at two wavelengths: 420 and 500 nm, with NTU scale. It was used for measuring Residual Turbidity and expressed in Nephelometric Units (NTU), HANNA pH-meter to measure pH- values, and Zeta potential (mV) with electrode of temperature Compensation.

2.1.2. Conductivity meter Jenway 720 to measure conductivity (µs/cm), total dissolved solids in mg/l and temperature.

2.1.3. Digital chlorine analyzer to measure concentration of chlorine solution, as free residual chlorine and total. It is well mentioned that all instruments and glassware have been calibrated according to the National Institute of Standards (NIS), and all experiments have been carried out on the raw Nile water of Damietta branch around the four seasons of the year. Both 5% hydrated lime and 1.0 N H₂SO₄ were used to adjust the pH of raw water to the desired value. Liquid Alum was supplied by the National Company for Alum production; Egypt, while caustic soda, lime, and other chemicals were supplied from ZWTP stock chemicals. PAC and Polymer were purchased from the local market.

2.2. Experimental setup: Raw water samples were freshly collected from Zifta WTP with each run. Figure (3) shows sampling Inlet as well as outlet locations, respectively. Characterization of water quality was carried out in the laboratory parallel to the routine analysis of the plant schedule. All analyses were carried out in accordance with the American

Standard Methods; APHA, 2012 [22].



Figure 3 Inlet and outlet of ZWTP

2.3. Jar Test Experiment: Coagulation tests have been performed according to Jar-test method, using a standard Flocculator *Stuart Scientific*, equipped with two mixing posts. This step was done by flash mixing step (200 rpm) for 2 minutes, while flocculation step was done for 10 minutes, at (28-30) rpm, followed by a settling time of 30 minutes. Tests have been performed in 1 liter cylindrical bakets, at room temperature (20±1) °C, using ZWTP raw inlet water. Coagulants, beside initial pH's, turbidity, temperature, pre-chlorine, and polymer dosages used in all experiments were the dependent variables in the coagulation process. Supernatant samples have been siphoned to determine turbidity NTU of the treated samples in each run. Figure (4) shows its front side.



Figure 4 Standard jar test apparatus

It is used to detect the optimum operating condition and to evaluate the removal efficiency of each experiment during the applied runs Figure 3. Aluminum sulfate (50%), Potassium aluminum sulfate, and Polyaluminum chloride 10.0%, solutions were used as separate coagulants as indicated in Table 3. Freshly prepared 3.0% cationic polymer; Table 3-1 was added prior each runs to enhance flocks formation and to reduce solids separation time in each run. Turbidity; NTU has been determined by Jenway Model 6405 UV-VIS spectrophotometer, at two wavelengths: 420 and 500 nm, with NTU scale. It was used to measure the residual NTU, and to detect the efficiency of each run. Electric Conductivity meter Jenway 720 was used to measure conductivity in $\mu\text{s}/\text{cm}$, while total dissolved solids was measured in mg/l and temperature in degrees centigrade. These parameters were used to detect the values of Z-potential.

3. Results and Discussion

3.1. Characterization of raw wastewater:

Table 3 shows the main characteristics of the raw water at the entrance of ZWTP, it represents the variations in quality during individual months with minimum, maximum and average records.

Table 3 Analysis of raw water quality characteristics; based on monthly bases.

Parameter/ Date	Ave	Min	Max
Turbidity(NTU)	5.82	2.84	10.96
pH	8.12	7.91	8.36
Zeta pot.(mV)	-82	-87	-78
Temp, °C	23.7	17.0	28.14
TDS (mg/l)	207	183	251
EC, $\mu\text{s cm}^{-1}$	392	345	475
Alkalinity(mg/l)	145	134	164
T. Hardn(mg/l)	129	112	144
Ca hardness(mg/l)	85	78	96
Mg hardness(mg/l)	44	33	53
Sulfate (mg/l)	16	10	30
Chlorides (mg/l)	26	17	37
Al (mg/l)	0.02	0.00	0.03
Fe(mg/l)	0.062	0.017	0.1
Mn (mg/l)	0.05	0.02	0.07
NH ₄ (mg/l)	0.18	0.16	0.21
NO ₃ (mg/l)	3.2	2.7	3.9
NO ₂ (mg/l)	0.02	0.02	0.02

* All monthly experiments were carried out for three runs each

This water characteristic represents the water criteria along one year which gives features through the twelve months from January to December, 2013 with averages, minimum and maximum values; respectively. On the other hand, Table 4 represents the water quality during the main four seasons. Results showed that the variations in the quality were affected by ambient temperature and the noticeable variation was a result of oxygen production and carbon dioxide absorption during photosynthesis of the water hyacinth and other climatic variation in the Damietta Nile branch.

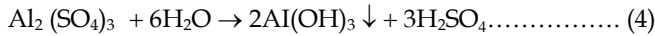
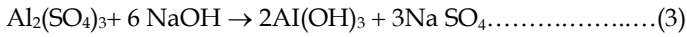
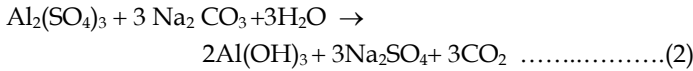
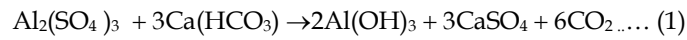
Table 4 Analysis of raw water as an average based on seasonal variation*

Parameter	Spring	Summer	Autumn	Winter
Turbid.,NTU	8.67	7.46	3.69	3.47
pH - Value	8.15	8.00	8.17	8.14
Zeta pot.,mV	-83	-78	-83	-69
Temp, °C	25.37	26.89	23.29	19.1
TDS, mg/l	186	194	235	217
EC, $\mu\text{s}/\text{cm}$	350	365	444	409
Alka.,mg/l	141	138	151	151
T. hard,mg/l	126	115	136	138
Ca hard,mg/l	82	80	91	87
Mg hard,mg/l	44	36	45	50
Sulfate.mg/l	11	11	15	26
Cl,mg/l	22	19	30	32
Al,mg/l	0.025	-	-	0.018
Fe,mg/l	0.07	-	-	0.06
Mn.mg/l	0.07	-	-	0.04
NH ₄ ,mg/l	0.18	-	-	0.19
NO ₃ ,mg/l	3.0	3.5	2.8	3.3
NO ₂ ,mg/l	0.04	0.05	-	0.02

* All experiments were carried out for three runs.

Detection of the optimum coagulation/flocculation operating conditions:

The optimum operating conditions of the treatment will be carried out according to coagulation/ flocculation process. The coagulation reactions were followed the equations:



The following results will give and discuss the effect of adding different coagulants for removing the turbidity from raw water in presence and in absence of pre-chlorination cases to give the most economic and visual treatment alternatives.

3.2. Detection of the optimum coagulant dose in the absence of pre-chlorination through Zeta-potential for the raw water.

3.2.1. Detection of preliminary alum dose as a function of Z-potential (mV), in the absence of pre-chlorination. Figure (5) represents alum dose versus the value of Zeta potential, and

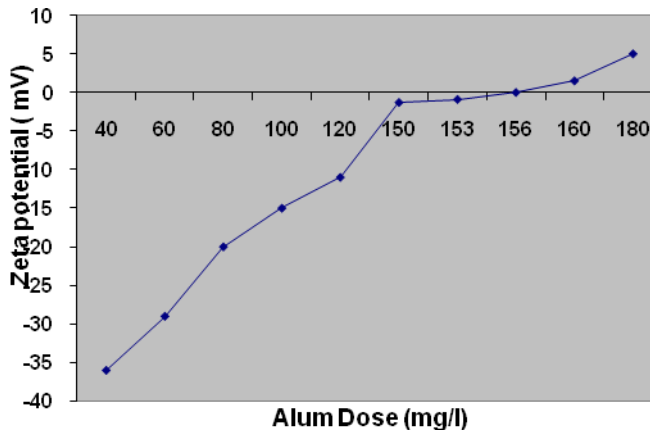


Figure 5 Detection of the Alum dose

Table 5 shows the detection of optimum alum dose on the removal of turbidity as a function of Z- potential.

Table 5 Detection of optimum Alum dose/ turbidity removal*

Alum*	160	156	153	150	120	100	80	60	40
Z-Pot	+1.5	0.0	-0.9	-1.3	-11	-15	-20	-29	-36
NTU	0.37	0.5	0.48	0.43	0.48	0.54	0.8	1.1	1.35
pH	6.66	6.7	6.74	6.80	7.00	7.08	7.2	7.7	7.52
Temp	15.3	16.3	16.2	16.4	15.3	15.5	15.3	17.4	17.1

* Alum: in mg/l; Zeta pot. mV; temp. °C, and the experiments were replicates of three runs.

The obtained results showed the effect of alum dose on Zeta potential (-73 mV); at Turbidity (3.5 NTU), pH (8.26) value, and in absence of pre-chlorination dose. Figure (6) represents this variation.

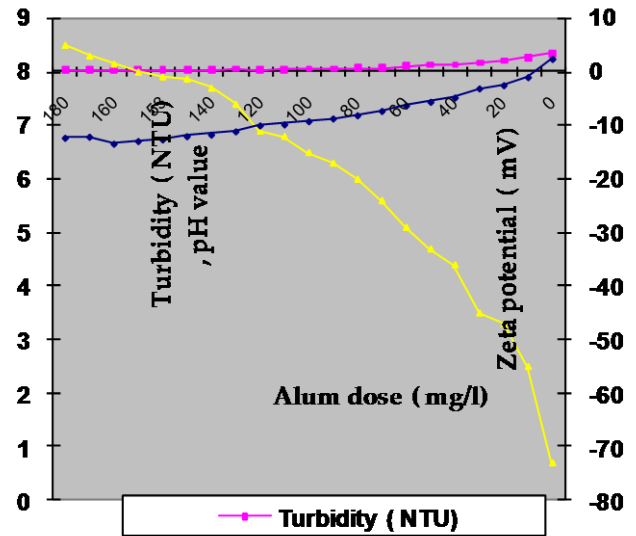


Figure 6 Detection of the optimum dose of Alum

3.2.3. Use of PAC as a dual coagulant and coagulant aid:

The optimum dose of poly-aluminum chloride played an effective role for the removal of suspended solids as well as other pollutants at its optimal operating dose. The obtained results showed that the optimum dose of PAC at its optimal operating conditions that fulfill the maximum removal of turbidity at zero pre-chlorination; 5.1 NTU; -93 mV Zeta potential; and pH (8.31). This value is representative in Figure (7).

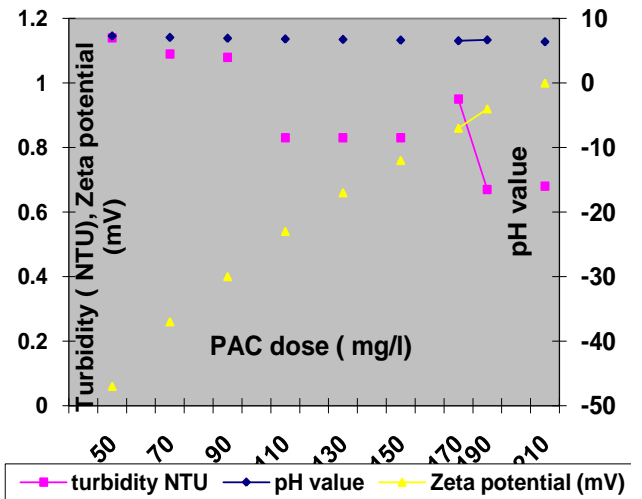


Figure 7 Detection of the optimal dose of PAC

3.2.4. Effect of PAC dose on Zeta potential (mV) at zero chlorine dose:

To study the effect of addition of different doses from PAC to the raw water river to detect the optimal dose that fulfill the maximum removal of pollutants from the raw river water, the experimental investigation has been carried out. The obtained results are shown in Figure (8), and Table 6 show the detection of PAC dose on the removal of turbidity as a function of Z- potential.

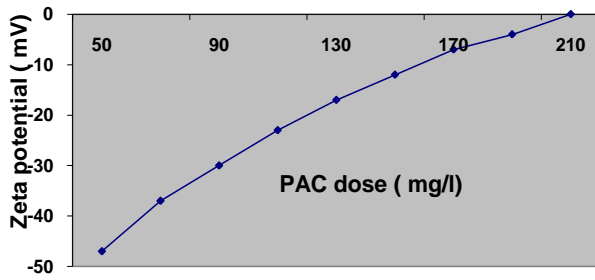


Figure 8 Detection of optimal Zeta-potential on using PAC

Table 6 Detection of PAC dose on the removal of NTU as a function of Z-potential

PAC*	10	20	30	40	50	60	70	80	90
Z-Pot	-75	-64	-58	-51	-46	-40	-36	-33	-29
NTU	2.36	1.54	1.46	1.05	1.08	0.83	0.80	0.93	0.73
pH	7.93	7.72	7.58	7.43	7.34	7.22	7.14	7.07	6.99
Temp	24.4	24.3	24.3	24.5	24.4	24.7	24.3	25.0	24.8

* PAC, in mg/l, Z- pote in mV, temp in °C, and the experiments were replicates of three successful runs.

3.2.7. Effect of PAS dose on NTU removal: To study the effect of potassium aluminum sulfate dose in mg/l on the removal of turbidity (2.50 NTU), at pH-value of 6.76, and -62 mV Zeta potential at zero mg chlorine dose; Figure 9 shows the detected required dose of the coagulant at the optimum operating conditions.

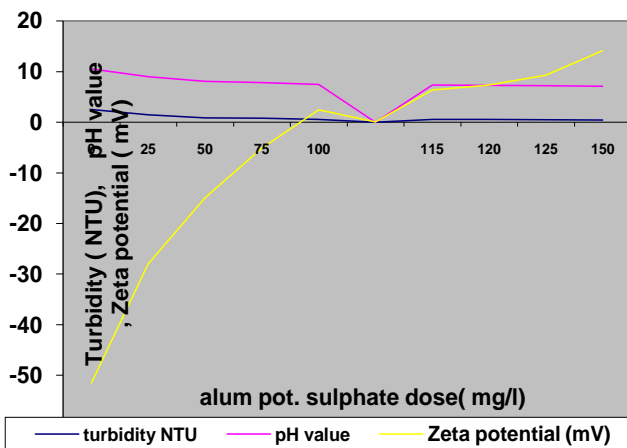


Figure 9 Detection of optimal dose of PAS

3.2.8. Effect of PAS dose on Zeta-potential: The effect of potassium aluminum sulfate dose in mg/l on the stability of Zeta potential has been studied. Results showed that the removal of turbidity (2.50 NTU), at pH-value of 6.76, and Zeta potential of -62 mV at zero pre-chlorination. Figure 10 demonstrates the detected required dose of the coagulant to fulfill the optimum Z-potential at the optimum operating conditions. The results obtained and represented in Figure 10 give the optimum dose relative to Z-potential.

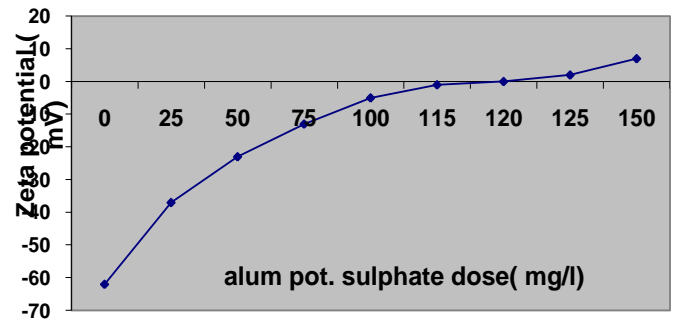


Figure10 Detection of Zeta-potential on using PAS

3.3. Detection of the optimum coagulant dose in the presence of pre-chlorination through Zeta-potential for raw water.

3.3.1. Detection of alum dose on the removal of turbidity as a function of Z-potential: The obtained results recorded in Table 7 represent the detection of alum dose on the removal of NTU as a function of Z-potential in presence of pre-chlorination. The effect of alum dose on the removal of turbidity at optimum Zeta potential in mV, and pH value as a second step after addition of 8.5 mg/l as pre-chlorination dose was studied. Figure 11 illustrates the results related to this item.

Table 7 Detection of alum dose on the removal of NTU as a function of Z-potential in presence of pre-chlorination

Alum*	10	20	30	40	50	60	70	80	90
Z-Pot	-51	-45	-42	-38	-34	-31	-27	-24	-20
NTU	1.43	0.62	0.47	0.31	0.27	0.34	0.20	0.16	0.15
pH	7.46	7.43	7.28	7.19	7.11	7.06	6.96	6.89	6.83
Temp	25.1	25.2	25.4	25.3	25.3	25.0	24.1	24.3	24.4
Alum*	10	20	30	40	50	60	70	80	90
Z-Pot	-51	-45	-42	-38	-34	-31	-27	-24	-20
NTU	1.43	0.62	0.47	0.31	0.27	0.34	0.20	0.16	0.15
pH	7.46	7.43	7.28	7.19	7.11	7.06	6.96	6.89	6.83
Temp	25.1	25.2	25.4	25.3	25.3	25.0	24.1	24.3	24.4

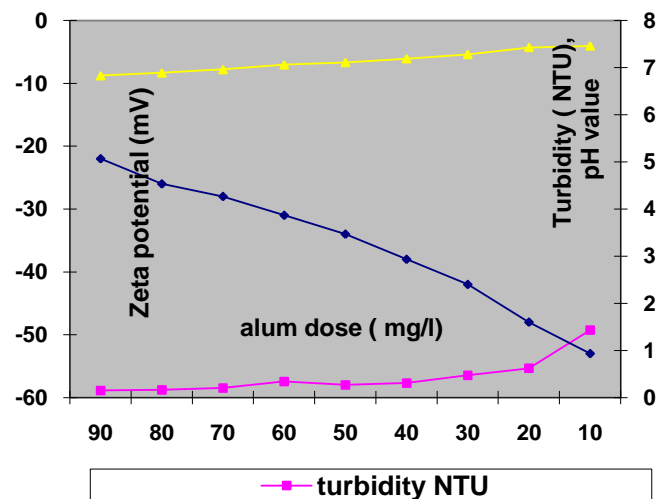


Figure 11 Detection of optimal Z-potential on using PAS

3.3.2. Detection of alum dose as a relation to Z-potential for the removal of turbidity in presence of pre-chlorination: A treatment investigation has been carried out to detect the effect of alum dose versus Zeta potential in mV; on the removal of turbidity (NTU), at in presence of 8.5 mg/l as pre-chlorination dose. Figure 12 shows the results obtained to this item.

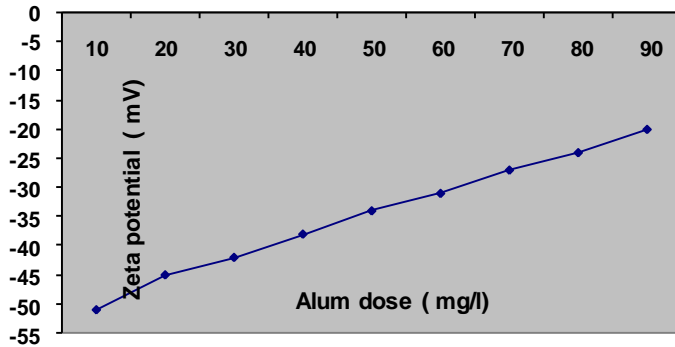


Figure 12 Detection of Alum dose on Z-potential at pre-chlorination

3.3.3. Detection of PAC dose on the removal of turbidity as a function of Z-potential:

Table 6 shows the detection of PAC doses in mg/l, as a function of Zeta potential in mV, and at the temperature in °C.

3.3.4. Effect of PAC dose on Zeta potential (mV) at chlorine dose (2.5 mg/l):

Figure 13 shows the detection of PAC dose on Zeta-potential

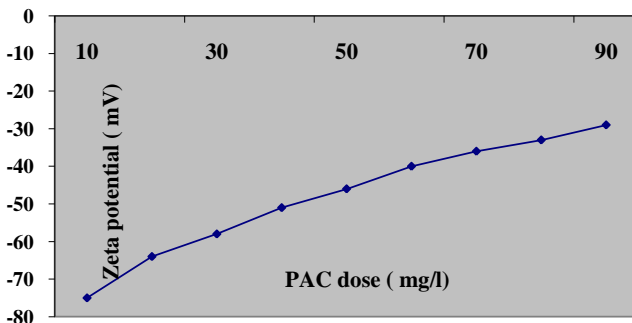


Figure 13 Detection of PAC dose on Zeta-potential

3.3.5. Effect of PAC dose (mg/l) on turbidity removal (NTU), Zeta potential (mV), pH value at chlorine dose (2.5 mg/l):

Figure 14 shows the detection of optimal PAC dose on Zeta-potential.

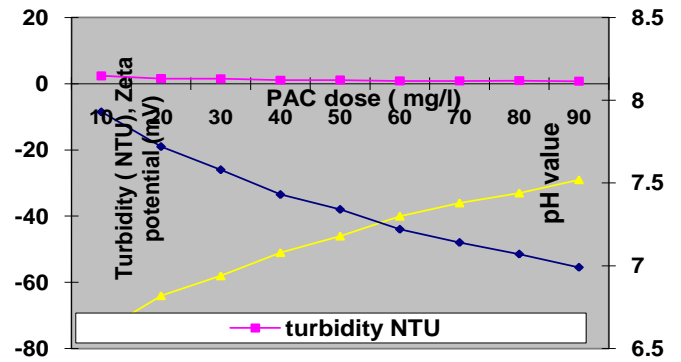


Figure 14 Detection of optimal PAC dose on Zeta-potential

4.1. Comparative study between experimental analysis of Alum efficiencies in presence and in absence of pre-chlorination

Table 8 Comparative study between experimental analysis of Alum efficiencies in presence and in absence of pre-chlorination

Alum mg/l	In presence of Cl ₂ (4.6 mg/l)			
	T °C	pH	Z- pot.	NTU
70	17.8	6.98	-10	0.82
80	17.5	7.26	-7	0.70
90	17.6	6.91	-4	0.54
100	17.7	6.86	-3.5	0.50
110	17.7	6.82	-2.0	0.45
120	17.8	6.77	0.0	0.42

Alum mg/l	In absence of Cl ₂			
	T °C	pH	Z-pot.	NTU
70	15.2	7.27	-24	0.83
80	15.3	7.19	-20	0.79
90	15.3	7.13	-17	0.61
100	15.5	7.08	-15	0.54
110	15.5	7.02	-12	0.5
120	15.3	7.00	-11	0.48

4.2. Physico-chemical analysis of ZWTP inlet and outlet samples compared to laboratory's experimental results at the optimum operating conditions: ZWTP at total chlorine dose (8 mg/l) with pre-chlorination dose (5.5 mg/l), alum dose (45 mg/l), Experimental analysis at pre-chlorine dose (2.5 mg/l), alum dose (30 mg/l), TRC: total residual chlorine (mg/l).

Table 9 Physico-chemical analysis of ZWTP inlet and outlet samples compared to laboratory's experimental results at the optimum operating conditions *

Parameter	Experimental analysis		ZWTP *	
	Treated	Raw water	Plant out	Plant In
Temp	26.5	25.37	16.4	16.5
pH	7.54	8.15	7.16	7.93
NTU	1.37	8.67	0.27	5.7
NTU; % R	84	==	95.3	= =
Alkalinity	124	141	140	158
EC	374	350	461	451
TDS	198	186	244	239
Chlorides	32	28	40	37
T. Hard	131	126	140	136
Ca Hard	86	82	92	90
Mg Hard	45	44	48	46
TRC	0.61	-	2.75	==

Obtained Results showed that: Conventional running treatment method was developed to achieve upgraded optimal coagulation that reduces: NTU; disinfection dose by-products (DBPs); heavy metal levels. On the contrary; it increases filtration time duration and to reduce running cost of the treatment. Although the theoretical value of Zeta potential is proposed to be Zero at the neutralized point, but in the actual experimental levels, the reaction doesn't fulfill the Zero value. Results showed that the potential of raw water ranged between 45 and 35 mV, that will achieve the highest removal levels of colloidal matters; wide range of pH (8.5–7.8) of raw water that is very limited. This has no remarkable effect on coagulation process. Moreover, results showed that the reaction temperatures were widely affected the consumed alum doses due to the big variation of temperatures from 14 to 30°C during winter to summer, passing by autumn and spring seasons, respectively. The 33% alum doses were fluctuated between 30 to 45 mg/l, this is due to the highly wide range of temperature from 14 to 28-30°C; that decrease alum dose. In fact, this paper represents a real trial for saving chemicals, reduces chlorinated bi-products by reducing the pre-chlorination to the minimum. Some modifications may be needed on the treatment sequence.

Conclusion and Recommendations:

- Due to the low levels of algal growth with noticeable DBPs reduction during summer and spring seasons, the required pre-chlorination decreased from 1.5 to 0.5 mg/l, which led to corresponding variation of Z-potential of the raw water from 45 to 35 mV, and achieved a high removal of colloidal matters.
- Although the sludge produced from alum was higher than PAC; but, alum was highly effective than PAC.
- The optimum operating conditions for the chosen factors that fulfilling the final turbidity 0.3 NTU, were pH range 7.8 - 8.5, and coagulant dose 30- 45 mg/l, respectively.

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