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AN AGRICULTURAL WASTE AS A NOVEL COAGULANT AID TO TREAT HIGH TURBID WATER CONTAINING HUMIC ACIDS

ALTAHER H.[1,2*]
TAREK E. KHALIL[3]
ABUBEAH R.[4]

[1]*Department of Chemical Engineering Technology*
Yanbu Industrial College, Saudi Arabia
[2]*Present address: University of Nizwa, Chemical Engineering Department*
Nizwa, Sultanet of Oman
[3] *Department of Chemistry, Faculty of Science*
Alexandria University, Alexandria 21321, Egypt
[4]*Department of Chemical Engineering, Faculty of Engineering*
El-Minia University, El-Minia, Egypt

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*to whom all correspondence should be addressed:
e-mail: haltaher@hotmail.com

ABSTRACT

Production of potable water from most raw water sources usually uses the coagulation/flocculation process to remove turbidity. Alum is widely used as a coagulant. However, there is concern about its associated risk of Alzheimer disease. As a result, there has been considerable interest in the development of natural coagulants and coagulant aids in order to reduce the dose of alum. This study aimed to evaluate the use of the Corchorus Olitorius L. (COL), a leaf vegetable grown in Africa and the Middle East, as a novel coagulant aid. COL has important advantages over other coagulant aids. It is an agricultural waste that is widely produced and does not require further chemical treatment. Tests were carried out to evaluate the optimal dosages and conditions required to achieve optimum removal of both turbidity and humic acid. Based on the results of jar test, COL is an efficient coagulation aid. It has the ability to reduce both the primary coagulant dose from 600 mg l^{-1} to 300 mg l^{-1} and the residual turbidity from 5.63 to 0.26 NTU. This novel coagulant aid also reduced the total organic carbon (TOC) concentration to zero level. It also increased the rate of flocculation.

Keywords: Alum, Corchorus Olitorius L., Coagulation, Flocculation, Turbidity, Water treatment.

1. Introduction

Water from surface or ground sources is not clean enough for consumption as it contains particles of colloidal suspension. These particles cannot be separated with conventional physical methods such as filtration or settling, unless they are agglomerated through coagulation and flocculation. The colloids commonly found in wastewater are stable because of the electrical charge they carry. The charge of colloids can be positive or negative. However, most colloidal particles in wastewater have a negative charge (Farajnezhad and Gharbani, 2012). The coagulation process is effective in removing high concentration of organic pollutants, heavy metals, some anions, and in reducing color, chemical oxygen demand (COD) and total suspended solids (TSS). Aluminum salts are the most widely used coagulants. However, they may cause numerous human neurological disorders (Tassinari *et al.*, 2013). It has been reported, however, that iron salts can be more efficient and cheaper than aluminum salts but their high importation cost is seen as a serious setback for developing countries (Ugonabo *et al.*, 2012). One major disadvantage of the coagulants is the huge amounts of toxic sludge produced in the process (Zemmouri *et al.*, 2012).

There are two main types of coagulants used in water treatment; namely: primary coagulants and coagulant aids. The role of primary coagulants is to neutralize the electrical charges of particles in the water, which causes the particles to stick together, while the coagulant aids increase the floc's density, add roughness to the flocs so that they don't break up during the mixing and settling process, and reduce flocculation and settling time (Aziz *et al.*, 2007).

On the other hand; lime and bentonite, clay minerals, activated SiO_2, cationic or anionic polyelectrolyte are examples of coagulant aids (Cheng *et al.* 2008). Some studies have investigated the use of tannin as a primary coagulant and coagulant aid in water treatment (Özacar and Şengil, 2002). Chitosan has also been investigated as a coagulant or flocculent for a wide variety of suspensions (Altaher, 2012; Zemmouri *et al.*, 2012; Hu *et al.*, 2013). A new coagulant aid called Enteromorpha extract was investigated for enhancement of coagulation performance (Zhao *et al.*, 2013).

The most important benefit of using a coagulant aid is the reduction in the amount of a coagulant used in any given case; which in turn reduces sludge and chemical residues in drinking water. In an attempt to avoid the hazards of using metal salts as coagulants, more interest has been generated towards development of natural coagulants and coagulant aids (Amud and Amoo, 2007; Ugonabo *et al.*, 2012; Hu *et al.*, 2013; Yang *et al.*, 2013). However, because most natural coagulants and coagulant aids are very expensive, it has become necessary to develop non-toxic, efficient, and low cost coagulant aids (Zhao *et al.*, 2013). The natural coagulants may be of plant seed, leaves, roots, and animal extracts (Gunaratna *et al.*, 2007). Other natural substances tried as coagulant aids include Moringa Olifera, red bean, and red maize (Gunaratna *et al.*, 2007; Poumaye *et al.*, 2012). Performance of agro-based materials, Surjana seeds, Nirmali seed, and maize as coagulant aids with alum as a coagulant was investigated by Raghuwanshi *et al.*, (2002).

For any new coagulant/coagulant aid to be considered, it must be efficient, cost effective, sustainable, and environmentally safe. Corchorus Olitorius L. (COL) is a leaf vegetable grown in Africa and the Middle East. It is a vegetable native to Egypt and popular in the Mediterranean regions, Japan and Southeastern Asia. Young green leaves and shoots of this vegetable are picked and cooked. The stems and roots are considered as agricultural wastes. A through literature survey indicated that Corchorus Olitorius L. has not been used as coagulant or coagulant aid. The aim of this study is to investigate the use of COL waste as a coagulant aid to enhance turbidity and TOC removal, and accelerate the flocculation time. Operating variables that were investigated included turbid water pH, coagulant and coagulant aid dosages slow mixing time, sedimentation time, and filtration as polishing step. Coagulant aid COL was optimized on the basis of TOC removals, residual turbidity, and flocculation time.

2. Materials and methods

2.1 Materials

Stock kaolin solution was prepared by dissolving 10 g of kaolin (Acros Organics, Gee, Belgium) in 1 liter of distilled water. The solution was stirred for 24 hours for complete hydration of kaolin. The turbid water was prepared by adding part of the stock solution to distilled water to reach the required turbidity. The alum solution (Panreac Quimica SA, Spain) was prepared by dissolving 10 g of the solid salt in 1 liter of distilled water. This solution was used as a coagulant without further dilution. The coagulant aid was freshly obtained from the local market. The stems were cut into small pieces and crushed in a mortar using a pistol. The liquid extract was collected and kept in a glass container. The working solution was prepared by dissolving 1 g of this extract in 1 liter of distilled water. The humic acid solution was prepared by dissolving 1 g of the solid material (Loba Chemiw, India) in 1 liter of distilled water. A suitable volume of humic acid solution was added to reach the required humic acid concentration in turbid water.

2.2 Coagulation experiments

Coagulation/flocculation tests were conducted using a conventional jar test apparatus equipped with 6 beakers of 1000 ml volume. The turbid water samples (100 NTU) were stirred for one minute to assure the homogeneity of the turbid water. The beakers were filled with 500 ml of the turbid water for each test run. No adjustment of pH was performed unless when evaluating the effect of pH on the coagulation process. Calculated amounts of coagulant were added to each beaker to reach the required dose. Care was taken not to change the total volume of the turbid water. Rapid mixing was applied at 250 rpm for 1 minute followed by slow mixing at 50 rpm for 20 min. The flocs formed were then allowed to settle for 40 minutes. After settling period, samples were withdrawn from supernatant for analysis. All the experiments were carried out at ambient temperature of 20–22 °C. The different factors affecting the coagulation process were studied by changing one factor, at a time, while keeping the other factors constant. For instance, to study the effect of initial pH, all other factors were kept constant and the pH of turbid water was adjusted at different value using either 1N H_2SO_4 or 1N NaOH. The experimental conditions are summarized in Table 1.

Table 1. Experimental conditions of the coagulation/flocculation experiments

	Rapid mixing time,min / rate,rpm	Slow mixing time,min / rate,rpm	pH of turbid water	Dose of alum as mg l^{-1} $Al_2O_3.18H_2O$	Dose of COL, mg l^{-1}
Effect of alum as the primary coagulant dose	1/250	20/50	7.5	20-1800	-
Effect of COL as the primary coagulant dose	1/250	20/50	7.5	-	2-200
Effect of pH when using alum	1/250	20/50	1-11	600	-
Effect of pH when using alum+COL	1/250	20/50	7.5	300	3
Effect of COL as coagulant aid on removal of turbidity	1/250	20/50	7.5	20-600	2-200
Optimization of alum dose and coagulant doses for removal of humic acids	1/250	20/50	7.5	20-600	2-10
Effect of coagulant aid on the flocculation time	1/250	20/50	7.5	300	3

The filtration was conducted in a column of 60 cm height and 1 cm diameter. 10 g of sand was placed in the column and was supported with glass wool. The sand was introduced to the bed without stratification. After the sand was introduced, gentle tapping was applied on the outer surface of the column to avoid possible channeling through the sand bed. The supernatant from the coagulation/flocculation process were introduced to the column using a peristaltic pump. The effluent from the column was collected, and the residual turbidity was measured. Duplicates of the experiments were conducted, and the average was taken.

2.3 Analytical method

The TOC representing the humic acid and residual coagulant aid was determined following Hach Method 10129 of the Hach Water Analysis Handbook (Hack 1997), using DRB200 Reactor and DPR 890 colorimeter. Turbidity was measured using calibrated Nephelometer (Hack Turbidity Meter Model 2100) and pH was measured using pH 501, EuTech Instruments.

3. Results and discussion

3.1 Sand bed, turbid water, and COL characteristics

The characteristics of the sand bed used are: d10 (the grain diameter at 10% passing), d60 (the grain diameter at 60% passing), and uniformity coefficient UC (d60/d10) of the sand bed were 0.173 mm, 0.433 mm and 2.5 respectively. The turbid water characteristics are illustrated in Table 2A while the characteristics of COL are introduced in Table 2B.

Table 2A. Characteristics of turbid water

TOC (mg l^{-1})	pH	Turbidity (NTU)	EC (s m^{-1})	TDS (mg l^{-1})
50	7.5	100	0.204	102

EC: Electrical conductivity

Table 2B. Characteristics of Corchorus Olitorius L. (Ndovu and Afolayan 2008)

	Total carbohydrate, (mg g^{-1})	Crude protein, (mg g^{-1})	Crude fiber, (mg g^{-1})	Crude lipid, (mg g^{-1})	Ash, mg g^{-1}
Leaves	695.0±32.4	162±3.4	20.3±1.0	17.2±2.9	105.2±1.0
Stems	802.0±18.2	51.0±1.8	88.2±4.8	69.0±1.3	51.9±1.2

3.2 Coagulation performances of separate coagulant and coagulant aid

Effect of alum dosages on the turbidity removal efficiency is shown in figure 1A. It is apparent from the results that with increase in the alum dose from 20 to 300 mg l^{-1}; the turbidity removal efficiency decreased from 93.5 % to 88.3 %, respectively. Further increase in the alum dose to 600 mg l^{-1} caused an increase in turbidity removal to 94.4 %. After this dose the turbidity removal decreased again. Thus, the highest turbidity removal efficiency (94.4 %) equivalent to final turbidity of 5.6 NTU was obtained at an alum dose of 600 mg l^{-1} which was considered as the optimum dose of alum at these conditions. The initial increase in turbidity of the water samples after adding the coagulant may be attributed to the coagulant itself, which added turbidity to the water before starting its coagulation action. At high coagulant doses, aluminum hydroxide is produced and suspended solids causing turbidity are removed by entrapment into or sorption onto hydroxide flocs (Demirci *et al.*, 1998).

COL was tried as the primary coagulant in a separate experiment. As illustrated by figure 1B, the different doses of COL resulted in poor turbidity removal efficiency compared to alum. The highest efficiency that could be attained was 42.5 %. The lowest final turbidity was 57.5 NTU at a dose of 6 mg l^{-1} of COL. An explanation is the steric hindrance that may take place between polymer molecules at high concentration (Hu *et al.*, 2013). The poor performance of natural polymers when used as primary coagulants has been reported by other researchers (Hu *et al.*, 2013; Zhao *et al.*, 2013).

The data presented in Table 2B indicates the presence of high concentrations of both carbohydrates and lipids in the stems. The presence of these constituents increase the content of organic matter in the treated water which represents a disadvantage due to the increase in turbidity at certain situations if the dose of the primary coagulant is not well adjusted.

COL waste can be considered as a natural polymer. When used as coagulant or coagulant aid, it enables the formation of chemical bridges, through hydrogen bonds or van der Waals forces, with the suspended solids in the treated water. This will enhance the flocculation process (LIBANIUS, 2008, Theodoro *et al.*, 2013). The high carbohydrates content of COL, presenting the carboxyl and hydroxyl groups, increase the clotting ability.

It is believed that the presence of hydroxyl groups along the polymer chain provides abundant adsorption sites for the removal of suspended water (YIN, 2010).

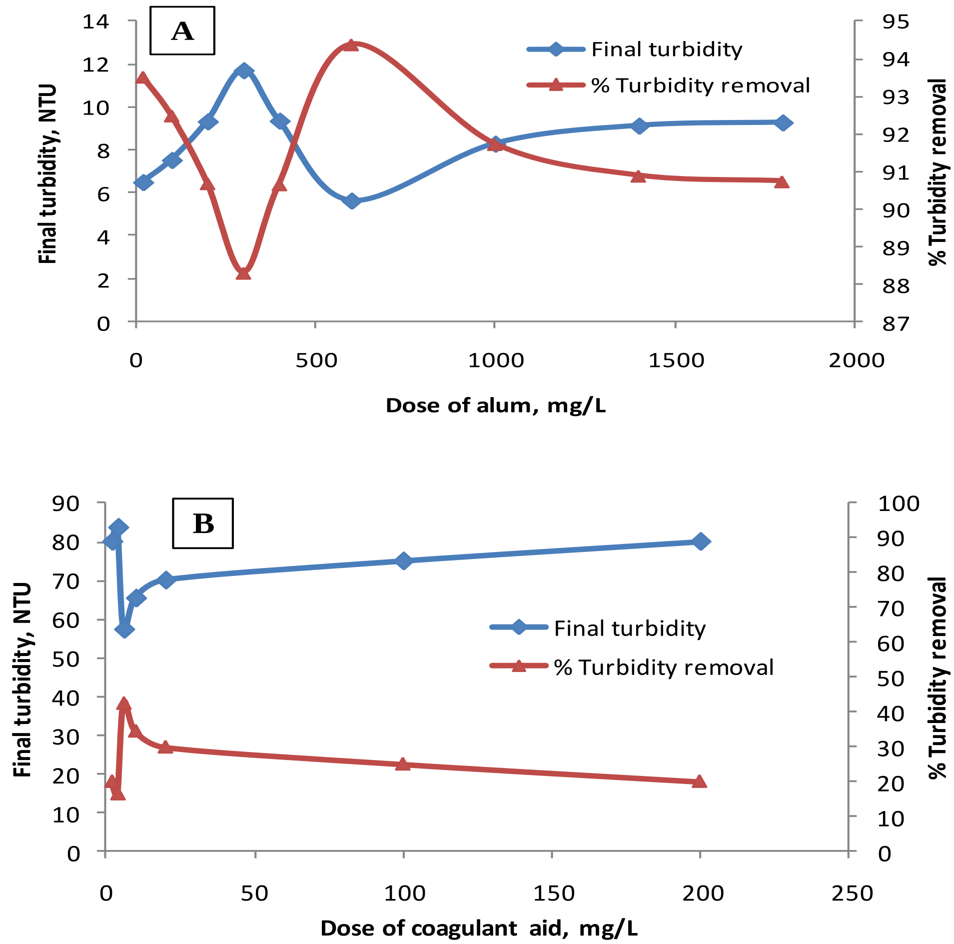

Figure 1. Effect of coagulant/coagulant aid doses on removal of turbidity; A: alum, B: COL

3.3 Effect of turbid water pH on turbidity removal

In the coagulation–flocculation process, pH is a very important factor since the coagulation occurs within a specific pH range for a particular coagulant. When alum is added to water, a series of soluble hydrolysis species are formed. The species that may be produced include $Al(OH)_3$, Al^{3+}, $Al(OH)^{2+}$, $Al(OH)^{1+}$ and $Al(OH)_4^-$. These hydrolysis species have positive or negative charges depending on the water pH. The positively charged hydrolysis species can adsorb onto the surface of colloidal particles and destabilize them. The equilibrium state between the concentrations of the above ions depends on the pH of the solution (Libecki and Dziejowski, 2008). The following equilibrium is obtained shifting to the right with increasing pH:

$$Al^{3+} \rightarrow Al(OH)^{2+} \rightarrow Al(OH)_2^+ \rightarrow Al(OH)_3 \rightarrow Al(OH)_4^-$$

The coagulation process with alum as the sole coagulant is capable of achieving significant turbidity removal, the removal is highly pH dependent (Yan *et al.*, 2008; Yang *et al.*, 2010).

As shown in figure. 2A, removal efficiency with alum decreased with the increase of pH from 1 to 3 and then increased with increasing pH reaching a considerable high level at pH 7.1. Further increase in the pH led to slightly decreased removal efficiency. At pH 11, coagulation efficiency almost reached the slightly high removal value of 97.1 % (final turbidity of 2.9 NTU). At this high pH, high precipitation of aluminum will take place leading to removal of turbidity by sweep-floc mechanism. In our tested samples, the high removal of turbidity at pH 1 may be attributed to the removal of humic acids first, followed by kaolin turbidity. The high turbidity removal at higher pH may be attributed to the association of humic acids with other suspended solids during the flocculation. On the other hand, when COL was added as a coagulant aid, illustrated by figure 2B, a slightly different behavior was observed. Increasing the pH led to a clear decrease in the removal efficiency. The removal efficiency decreased from a value of 98 % at pH 1 to 85 % at pH 7.5. It is clear that the removal efficiency when using COL as a coagulant aid has been enhanced by the effect of pH. A final turbidity of 0.214 NTU was attained at pH 11. This value is very low compared to that obtained when using alum alone and it is better than the value recommended by World Health Organization, WHO, (1 NTU).

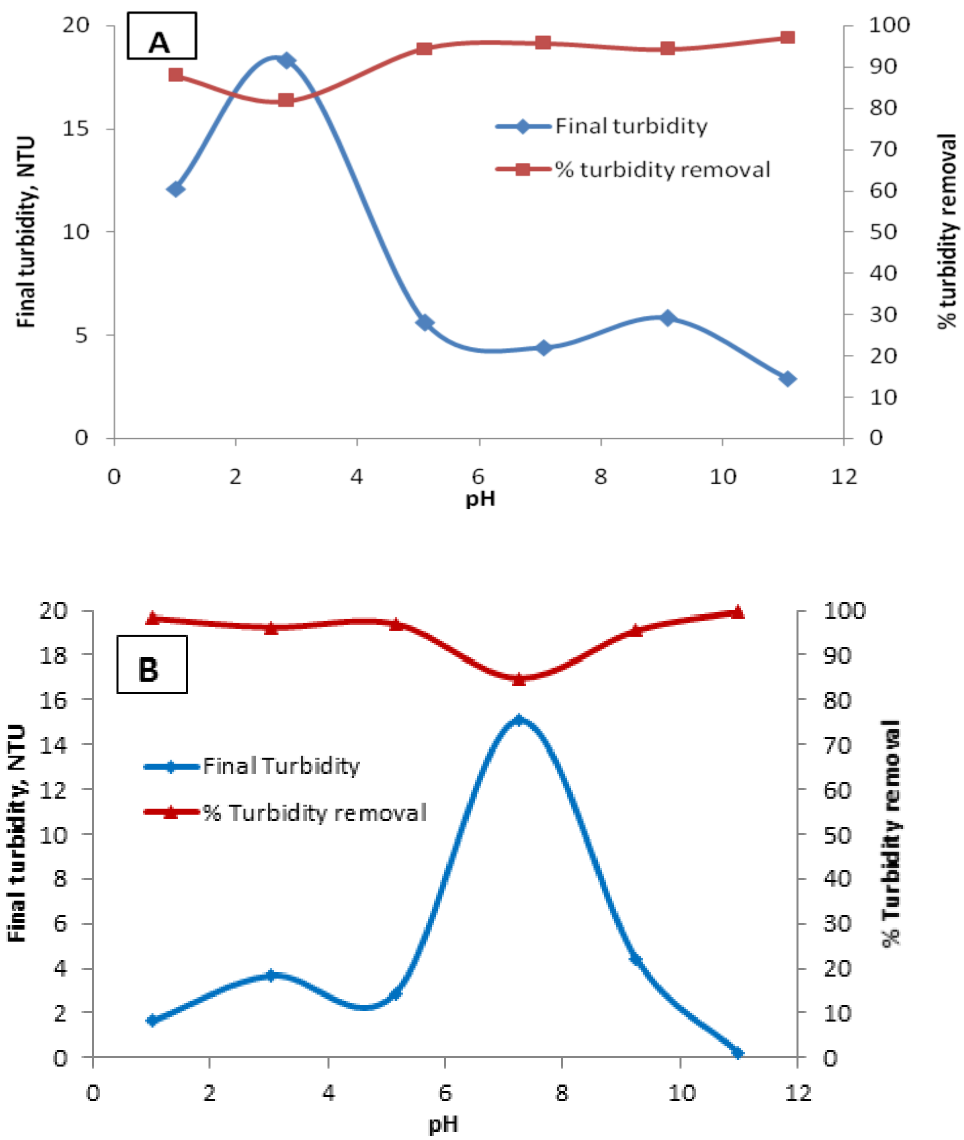

Figure 2. Effect of water pH on removal of turbidity; A: alum, B: Alum + aid. Alum dose is 300 mg l^{-1} and COL DOS IS 3 mg l^{-1}

3.4 Effect of coagulant aid on turbidity removal

Two separate experiments were performed. In the first experiment, one set of beakers containing turbid water was treated with alum alone. Another set was treated by adding the same dose of alum and different doses of COL. In the second experiment, the dose of alum was kept constant at its optimum value, and the dose of COL was varied from beaker to another. For the second set, the dose of alum was varied and the dose of COL was kept constant at its optimum value. As illustrated in figure 3, adding the coagulant aid was very influential in decreasing turbidity. The turbidity using a small dosage of the coagulant aid (3 mg l^{-1}) decreased the residual turbidity to half its value (from 5.63 NTU to 2.3 NTU). This last turbidity value wouldn't have been achieved without using the coagulant aid, no matter how optimum other conditions are.

Figure 4 represents the optimization conditions for both alum and COL. It is clear that the optimum doses of both alum and COL are 300 mg l^{-1} and 3 mg l^{-1}, respectively. The correct selection of optimum combination doses of coagulant and coagulant aid resulted in 50 % decrease in the initial primary coagulant dose. Also, the final turbidity has been enhanced to reach a value of 2.3 NTU compared to a value of 5.63 NTU when alum was used alone. As illustrated by table 2B the stem of the plant contains substantial amount of proteins. These proteins may impart positive charge to solution which bind with the negatively charged particulates causing turbidity (Tassinari *et al.*, 2013).

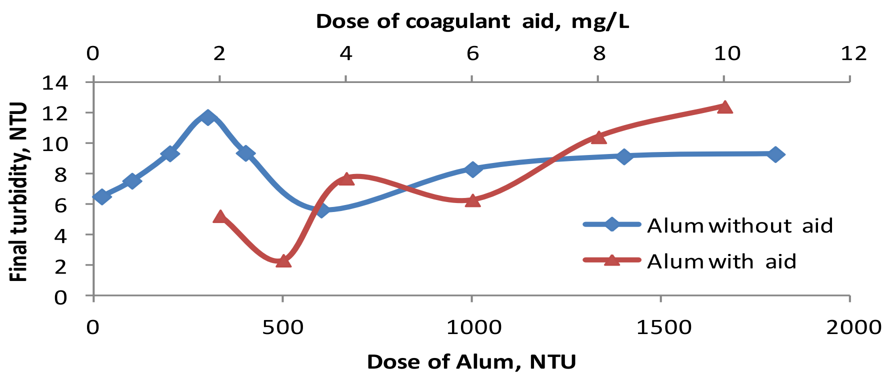

Figure 3. Effect of coagulant dose on removal of turbidity

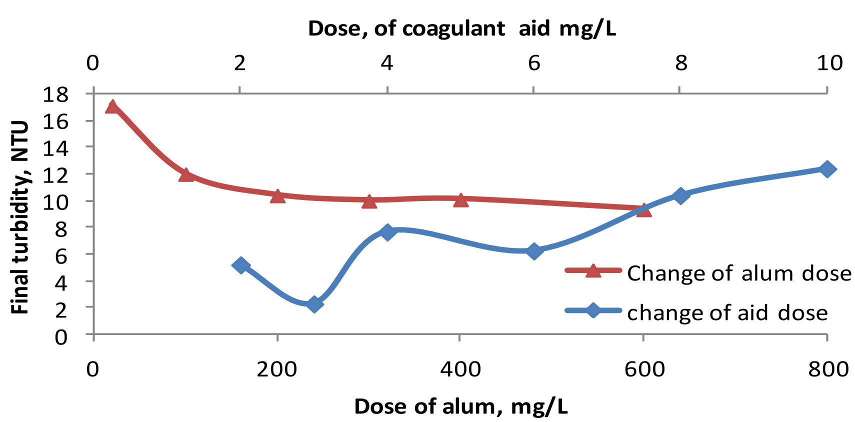

Figure 4. Optimization of alum and coagulant doses

3.5 Humic acid removal

Removal of natural organic matter (NOM) from water can take place through several mechanisms. These mechanisms are charge neutralization, entrapment and adsorption (Lanciné *et al.*, 2008). Clay minerals dispersed in natural water may adsorb humic substances. Alum will dissociate in water to produce many cations. Ion exchange between organic and clay due to presence of these cations is important in this mechanism (Duan and Gregory, 2003). Hydrogen bonding and Van der Waals forces may also play a significant role, particularly if the humic molecule has large uncharged portions and sufficient flexibility to come into contact with the surface of clay (American Water Works Association, AWWA 1979).

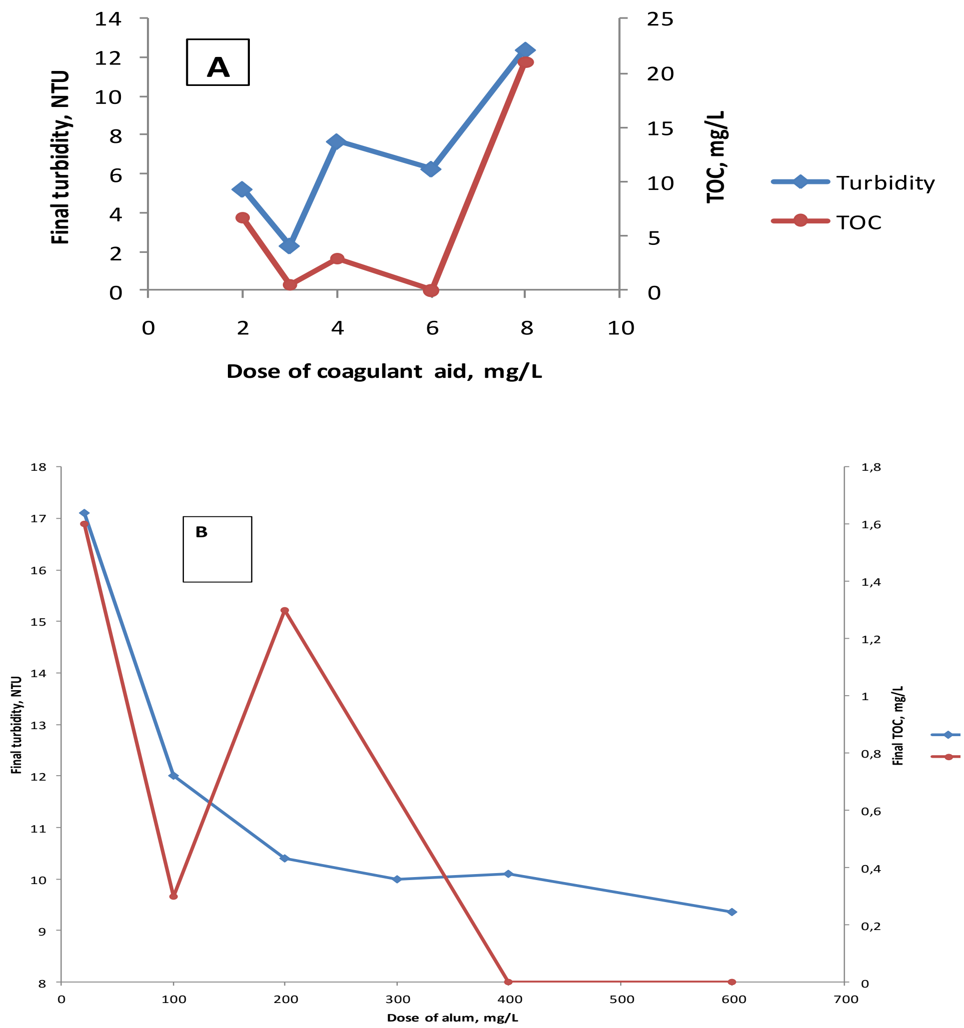

Figure 5. Optimization of alum and coagulant doses for TOC removal

To investigate the effect of the coagulant aid on the removal of humic acids, two experiments were performed. In the first one, the dose of alum was kept constant and the dose of COL was changed and in the second one, the dose of COL was kept constant and the dose of alum was changed. It is clear from figures 5A & 5B that the removal of humic acids is closely related to the removal of turbidity. That may be due to the adsorption of humic substances on the surface of kaolin and the collective flocculation of them. It is worthy notice that the TOC value representing humic acids and residual COL after flocculation reached a zero value at certain doses of the coagulant and the coagulant aid. These doses are considered as the optimum doses. The high increase in TOC in figure 5A after reaching the zero TOC value is attributed to the residual coagulant aid.

3.6 Effect of coagulant aid on flocculation and settling times

Two sets of beakers containing the turbid water were used. To the first set, the optimum dose of alum was added in each beaker, and to the second set optimum doses of both alum and coagulant aid were added. As illustrated by figure 6, clear difference can be observed at the initial mixing stages. The solutions containing the coagulant aid were destabilized very rapidly. Within 5 minutes of slow mixing followed by sedimentation, the turbidity of water treated with the coagulant aid decreased to 75 % of its initial value compared to 50 % in case of water treated with alum alone.

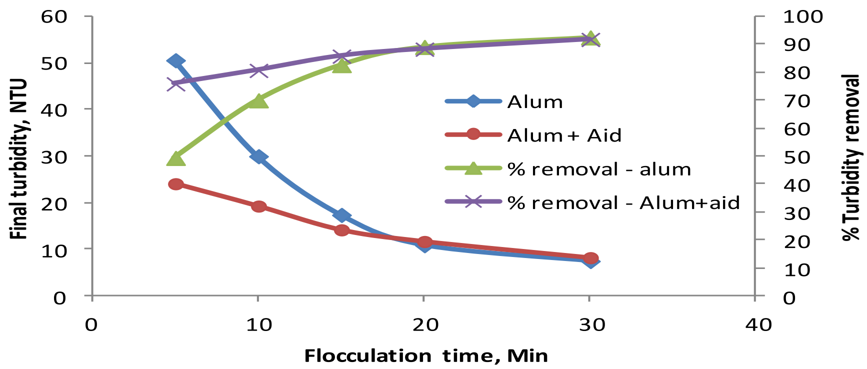

Figure 6. Effect of coagulant aid on the flocculation time

An important difference between the two systems is the size of the formed flocs. As illustrated by figure 7, in case of water treated with coagulant aid, large flocs would be observed within five minutes, though they had irregular shapes.

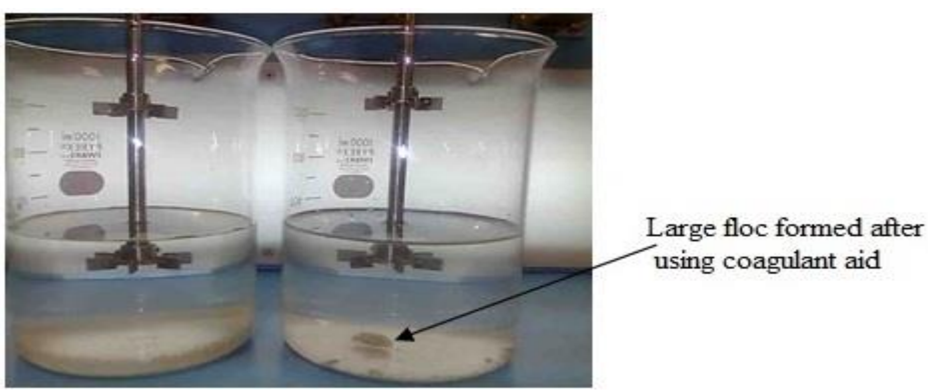

Figure 7. Effect of coagulant aid on the flocs shape and size; Left: water treated with alum alone, Right: water treated with alum + coagulant aid

The effect of coagulant aid on sedimentation time was analyzed at different sedimentation time intervals using optimum doses of alum alone and alum combined with the aid. The test was performed similar to the previous tests with one difference; the slow mixing was continued for 20 min followed by sampling treated water at different time intervals during the settling process. Figure 8 illustrates the settling rate for both systems tested. Alum shows a much faster sedimentation rate when used alone. 40 min of sedimentation can remove 92% of flocs, while alum with coagulant aid needs 70 minutes to achieve this value. These observations may be attributed to the shape of the flocs formed. The flocs formed when using alum alone are homogeneous and spherical in shape which leads to rapid rate of sedimentation, while flocs formed when using alum and coagulant aid are irregular in shape which resulted in slow rate of stirring. The flocs formed using the coagulant aid had probably lower density and gelatinous nature compared with that formed with alum. That also may reduce their settling rate. However, the water produced from the filter bed after sedimentation had better quality for systems that used the coagulant aid. The residual turbidity in such systems was 0.26 NTU compared to 0.632 NTU when using the alum alone. This can be attributed to the same reason; the irregular flocs can be captured by the filter media much better than the spherical ones. Usually, the physical characteristics of the flocs are important in determining their removal efficiency. For example, large compact flocs have a high settling rate (Wilen *et al.*, 2003), whilst large and porous flocs aid filtration due to low permeability (Bushell *et al.*, 2002).

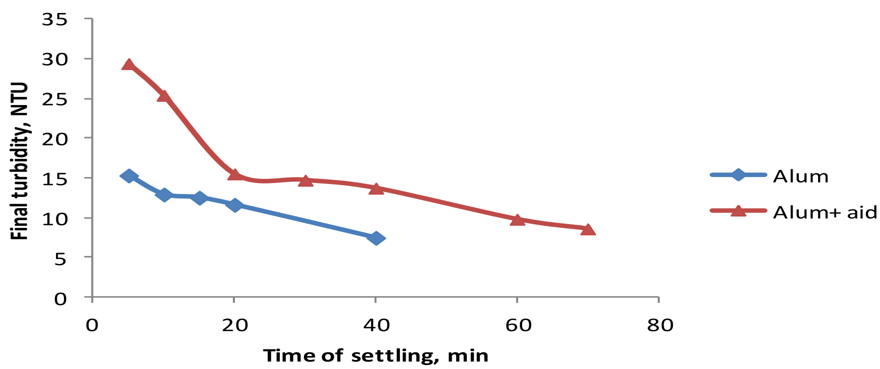

Figure 8. Effect of coagulant aid on the settling time

4. Conclusion

The experiments conducted confirm the significant effect of COL. Results showed that COL has the ability to reduce the dose of the primary coagulant (alum) and the residual turbidly to almost the half. The coagulant dose was reduced from 600 mg l^{-1} to 300 mg l^{-1} while the residual turbidity was reduced from 5.63 to 2.3 NTU when using the coagulant aid. COL was found to increase the initial rate of flocculation process. COL did not enhance the settling rate of the formed flocs. However, the water produced after filtration had much better quality compared to that produced without using the coagulant aid; 0.26 NTU and 0.632 NTU, respectively. Comparing all these results, indicates that using COL as a coagulant aid is advisable.

References

Altaher H. (2012), The use of chitosan as a coagulant in the pre-treatment of turbid sea water, *Journal of Hazardous Materials,* **233-234**, 97-102.

AWWA (1979), Committee Report, Organic removal by coagulation: a review and research.

Aziz H.A., Alias S., Assari F. and Adlan M.N. (2007), The use of alum, ferric chloride and ferrous sulfate as coagulants in removing suspended solids, colour and COD from semi-aerobic landfill leachate at controlled pH, *Waste Management Research*, **25**(6), 556-565.

Bushell G.C., Yan Y.D., Woodfield D., Raper J. and Amal R. (2002), On techniques for the measurement of the mass fractal dimension of aggregates, *Advanced Colloidal Interface Science*, **95**(1), 1-50.

Cheng W.P., Chi F.H., Li C.R. and Yu F. (2008), A study on the removal of organic substances from low-turbidity and low-alkalinity water with metal-polysilicate coagulants, *Colloids and Surfaces A: Physicochemical Engineering Aspects*, **312**, 238–244.

Demirci S., Erdogan B. and Ozcimder R. (1998), Wastewater treatment at the petroleum refinery, kirikkale, turkey using some coagulants and Turkish clays as coagulant aids, *Water Research*, **32**(11) 3495-1499.

Duan J. and Gregory J. (2003), Coagulation by hydrolyzing metal salts, *Advanced Colloid Interface*, **45,** 100-102.

Farajnezhad H. and Gharbani P. (2012), Coagulation treatment of wastewater in petroleum industry using poly aluminum chloride and ferric chloride, International Journal of Research and Review In Applied Sciences, **13**(1), 306-310.

Gunaratna K.R., Garcia B., Anderson S. and Dalhammar G. (2007), Screening and evaluation of natural coagulants for water treatment, *Water Science Technology: Water Supply*, **7**(5/6), 19 -25.

HACH Company (1997), Water Analysis Handbook. Hach Company, Colorado.

Hendricks D. (2010), Fundamentals of Water Treatment Unit Processes: Physical, Chemical, and Biological, CRC Press.

Hu C.Y., Lo S.L., Chang C.L., Chen F.L., Wu Y.D. and Ma J.L. (2013), Treatment of highly turbid water using chitosan and aluminum salts, *Separation and Purification Technology*, **104**, 322-326.

Lanciné G.D., Bamory K., Raymond L., Jean-Luc S., Christelle B. and Jean B. (2008), Coagulation-flocculation treatment of tropical surface water with alum for dissolved organic matter (DOM) removal: influence of alum Dose and pH adjustment, *Journal of International Environmental Application & Science*, **3**(4), 247-257.

Liang Z., Wang Y., Zhou Y., Liu H. and Wu Z. (2009), Variables affecting melanoidins removal from molasses wastewater by coagulation/flocculation, *Separation and Purification Technology*, **68**, 382–389.

Libanius M. (2008), Fundamentals of quality and water treatment. 2nd ed. Campinas. , Atom Ed.

Libecki B. and Dziejowski J. (2008), Optimization of humic acids coagulation with aluminum and iron(III) Salts, *Polish Journal of Environmental Studies*, **17**(3), 397-403.

Ndovu L.J. and Afolayan A.J. (2008), Nutritional analysis of the South African wild vegetable *Corchorus Olitorius L.*, *Asian Journal of Plant Sciences,* **7**(6), 615-618.

Özacar M. and Şengil I.A. (2002), The use of tannins from turkish acorns (valonia) in water treatment as a coagulant and coagulant aid, *Turkish Journal Of Engineering and Environmental Sciences,* **26**, 255-263.

Patel H. and Vashi R.T. (2012), Removal of congo red dye from its aqueous solution using natural coagulants, *Journal of Saudi Chemical Society,* **16**, 131-136.

Poumaye N., Mabingui J., Lutgen P. and Bigan M. (2012), Contribution to the clarification of surface water from the moringa oleifera: case M'poko river to Bangui, Central African Republic, *Chemical Engineering Research and Design*, **90**, 2346-2352.

Raghuwanshi P.K., Mandloi M., Sharma J., Malviya H.S. and Chaudhari S. (2002), Improving filtrate quality using agrobased materials as coagulant aid, *Water Quality Research Journal of Canada*, **37**(4), 745–756.

Tassinari B., Doherty S. and Marison I.W. (2013), Submicron capsules extracted from rapeseed as novel flocculant agents for the treatment of turbid water, *Water Research*, **47**, 4957-4965.

Theodoro J.D.P., Lenz G.F., Zara R.F. and Bergamasco R. (2013), Coagulants and Natural Polymers: Perspectives for the Treatment of Water, *Plastic and Polymer Technology,* **2**(3), 55-62.

Ugonabo V.I., Menkiti M.C. and Onukwuli O.D. (2012), Coagulation kinetics and performance evaluation of corchorus olitorus seed in pharmaceutical effluent, *International Journal of Multidisciplinary Science and Engineering*, **3**(7), 20-32.

Wilen B.M., Jin B.and Lant (2003), Impacts of structural characteristics on activated sludge floc stability, *Water Research*, **37**, 3632-3645.

Yan M., Wang D., Yu J., Ni J., Edwards M. and Qu J. (2008), Enhanced coagulation with polyaluminum chlorides: Role of pH/alkalinity and speciation, *Chemosphere*, **71**, 1665-1673.

Yang Z., Gao B., Wang Y., Zhang X. and Yue Q. (2013), Relationship between residual al species, floc operational parameters and coagulation performance during reservoir water treatment by PAC-PDMDAAC, *Separation and Purification Technology,* **102**, 147-156.

Yang Z.L., Gao B.Y., Yue Q.Y., Wang Y. (2010), Effect of pH on the coagulation performance of Al-based coagulants and residual aluminum speciation during the treatment of humic acid-kaolin synthetic water, *Journal of Hazardous Materials*, **178**(1-3), 596-603.

Yin C.Y. (2010), Emerging usage of plant-based coagulants for water and wastewater treatment, *Process Biochemistry*, **45**, 1437- 1444.

Zemmouri H., Drouiche M., Sayeh A., Lounici H., Mameri N. (2012), Coagulation flocculation test of Keddara's water dam using chitosan and sulfate aluminum, *Procedia Engineering,* **33**, 254-260.

Zhao S., Gao B., Wang Y., Yang Z. (2013), Influence of a New Coagulant Aid-Enteromorpha Extract on Coagulation Performance and Flocs Characteristics of Aluminum Sulfate Coagulant in Kaolin-humic Acid Solution Treatment, *Colloids and Surfaces: A Physicochemical Engineering Aspects*, **417**, 161-169.