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By I.M. Muhammad, S. Abdulsalam, A. Abdulkarim & A.A. Bello

*Abubakar Tafawa Balewa University, Nigeria*

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## I. INTRODUCTION

Water supply is a basic need required for living creatures and human being specifically. Developing countries and third world countries are facing potable water supply problems because of inadequate financial resources. The cost of water treatment is increasing and the quality of river water is not stable due to suspended and colloidal particle load caused by land development and high storm runoff during the rainy seasons. During the rainy seasons the turbidity level increases and the need for water treatment chemicals increase as well, which leads to high cost of treatment which the water treatment companies cannot sustain. As a result, the drinking water that reaches the consumer is not properly treated (Muyibi *et al.*, 2009). Therefore, it is of great importance to find a natural alternative for water coagulant to treat the turbidity. In this world the amount of resources available to living creatures are limited. Safe drinking water is essential to the health and welfare of a community, and water from all sources must have some form of purification before consumption (Arnoldsson *et al.*, 2008).

*Author* <sup>a</sup> <sup>σ</sup> <sup>p</sup> <sup>Ω</sup>: Department of Chemical Engineering, Abubakar Tafawa Balewa University, Faculty of Engineering & Engineering Technology, Abubakar Tafawa Balewa University, Bauchi, Nigeria.  
e-mail: idrismisau@gmail.com

Drinking water treatment involves a number of combined processes based on the quality of the water source such as turbidity, amount of microbial load present in water and the others include cost and availability of chemicals in achieving desired level of treatment (Muyibi *et al.*, 2009). Conventional methods used for purification of water include coagulation, sedimentation, filtration, aeration and also chemical treatment.

In drinking water treatment, the coagulation process is used to destabilize suspended particles and to react with organic materials in the raw water. Proper coagulation is essential for good filtration performance and for disinfection by product (DBP). Common coagulants are aluminium sulphate, ferric chloride, poly-aluminium chlorides and synthetic polymers. The use of coagulants such as alum is one of the commonest methods employed and it reduces the repulsive force between particulate matter, encouraging particle collision and floc formation (Moramudaii and Fernando, 2001).

Recent studies have indicated a number of serious drawbacks linked to the use of aluminium salts such as Alzheimer's disease associated with high aluminium residuals in treated water, excessive sludge production during water treatment and considerable changes in water chemistry due to reactions with the OH<sup>-</sup> and alkalinity of water. In addition, the use of alum salts is inappropriate in some developing countries because of the high costs of imported chemicals and low availability of chemical coagulants (Adejumo *et al.*, 2013). In addition, monomers of some synthetic organic polymers such as acrylamide have neurotoxicity and strong carcinogenic properties and because of this, there has been considerable interest in the development of natural coagulants which are safe for human health and biodegradable (Ghebremichael, 2004).

A number of studies have pointed out that the introduction of natural coagulants as a substitute for metal salts may ease the problems associated with chemical coagulants. Using natural coagulants instead of aluminium salts might give advantages, such as lower costs of water production, less sludge production and ready availability of reagents. There are also some disadvantages such as increased concentration of nutrients and chemical oxygen demand (COD) in the treated water due to the organic nature of this type of coagulants (Daniyan *et al.*, 2011).

Among plant materials that have been tested over the years, the seeds from *Moringa oleifera* have been shown to be one of the most effective primary coagulant in water treatment or purification. *Moringa oleifera* is the best natural coagulant discovered so far that can replace aluminium sulphate (alum), which is used widely for water treatment around the world (Ali *et al.*, 2010). Recently, however, there has been a resurgence of interest in natural coagulants for water treatment in developing countries. For this purpose the greatest degree of attention has been focused on the seed of *Moringa oleifera* from Sudan, Nirmali seed in India, mesquite bean and in Venezuela, red bean and common bean, sweet corn and so on. These natural coagulants can be used alone or as a substitution for chemical coagulants and flocculants. They can be used for reducing turbidity and microorganisms in water, for water softening and for dewatering sludge (Mirjana *et al.*, 2010).

Large populations in rural and semi-urban areas of Africa have no access to clean drinking water. Waterborne diseases, though a global health threat, is a feature of developing countries whose populace are compelled to use turbid and contaminated water for domestic purposes. The removal of colloidal and suspended particles present in water would be extremely beneficial as it would assuage the majority of problems associated with turbidity. Conventionally, removal of the colloids in water could be achieved by coagulation, using certain chemical coagulants like certified alum. For many developing countries, this treatment process is not feasible because of the high costs involved and the difficulty in assessing chemical coagulants including alum. Moreover, recent studies have pointed out the health threats arising from the consumption of residual aluminium present in water, such as Alzheimer's diseases and neurodegenerative illness.

This paper aimed at investigating the potential of water melon seed as a coagulant for water treatment. This material was selected because the watermelon seed has high protein content and some authors have considered that the active coagulant agents in plant extract are proteins. The objective of this study is to determine the potential of watermelon seed as a natural coagulant, and investigate the coagulation characteristics of its crude protein extracts.

## II. MATERIALS AND METHODS

### a) Materials

Materials used in this work included watermelon seed cake (coagulant), N-hexane, Gubi Dam raw water

in Bauchi-Nigeria, Distilled water, Soxhlet extractor, Digital pH meter, Electronic weighing balance, thermometer, drying oven, electric hot plate, flocculator, beakers, pipette, turbidimeter, conductivity meter, and stop watch (timer). All reagents used are of analytical grade.

### b) Methods

#### i. Water melon seed (coagulant) preparation

Fresh seeds of watermelon (*Citrullus lanatus*) of the cucurbitaceae family were obtained from the local market (*Muda Lawal*) in Bauchi, Bauchi State, Nigeria. The fruits were sliced open using a clean stainless steel laboratory knife. The seeds were washed severally with water, sun-dried for a week, sorted to remove bad ones, shelled and ground with a high speed laboratory electric blender, packed in an air tight container. 150g of the crushed seeds were then packed in a thimble and placed in a soxhlet extraction apparatus. 500ml of the n-Hexane was used to extract oil from the crushed seed in the column. The apparatus was left running for about 6hours and stopped when the extraction was complete. The cake was then washed with distilled water to remove residual n-Hexane, dried in an oven till constant weight and then sieved. The finer particles were then used as the coagulant.

#### ii. Sample water collection

The raw water sample was collected from Gubi dam, located in Bauchi state, North-East of Nigeria. The water was collected from the side of river by immersing a plastic container until it was full. The cap was inserted while it was still underway. The water was then treated using the prepared coagulant.

#### iii. Water quality tests

##### a. Turbidity

Turbidity of the water sample was measured before and after treatment using a turbidimeter in accordance with the international method of water quality measurement and the results recorded.

##### b. Total Solid

Sample of the raw water was taken in 100ml beaker. A clean and dry crucible was weighted empty and the sample was then poured into it and reweighed. The respective weights were recorded and the crucible together with the sample water were then placed on a hot plate at 104oC to evaporate the water. When all the water evaporated, the crucible was allowed to cool down and reweighed together with the residue. The total solid present was then calculated using the equation:

$$TS = 100(A - B)/200ml \quad \text{Where:}$$

$$A = \text{weight of (crucible + water) - weight of crucible empty}$$

$$B = \text{weight of (crucible + residue)}$$

*c. Total Suspended Solid (TSS)*

Sample of the raw water was 100ml in a sample bottle. The weight of a dry filter paper was taken empty and the sample water was then filtered and the residue dried at 35-40oC in an oven. The new weight of the filter paper plus residue is then taken. The difference in the weight of the filter paper empty and with residue after drying was calculated and divided by total volume of sample.

*d. Total Dissolved Solid (TDS)*

This was obtained by taking the difference between TSS and TS or two-thirds of the conductivity using the conductivity meter.

*e. pH*

The pH of the samples was taken using an electronic pH meter.

*f. Colour*

Colour of the water sample was carried out before and after treatment using a turbidity meter.

*g. Jar test*

The jar test apparatus was used to carry out coagulation and flocculation on the water samples. Six 1litre beakers were used to study the effect of coagulant dosage on coagulation, the effect of pH on coagulation and the effect of stirring time and speed on coagulation. The following parameters were then measured on the filtrate after the coagulation was completed; turbidity, colour, flocs weight, TDS and conductivity. Six different weights of the coagulant were placed in each beaker, the first having 0.1g, and the remaining five varying from 0.1-0.6g at 0.1g interval in order to determine the optimum dosage. The raw water sample was then added to make up the 250ml mark and the jars were then placed in the jar test kit and the stirrers lowered into each. The stirring speed was set at 150rpm for rapid mixing for 2 minutes and 80rpm 8minutes for slow mixing. After this was completed the samples were allowed to settle and the flocs filtered using a filter paper and the parameters listed above were measured on the filtrate. From the results obtained the dosage with the best results in colour and turbidity removal was taken as the optimum.

The procedure above was used again, however a dose of 0.1g was maintained in all six beakers. The pH was varied from 6.0-8.5 by the addition of few drops of 1M NaOH into the beakers to make it alkaline. A few drops of 1M H2SO4 solution was also added in the first beaker to make it slightly acidic at 6.0. The same speed and stirring time was used as above and the parameters listed above were measured after the coagulation-flocculation and filtration process. The pH at which the best turbidity and colour removal were observed at was taken to be the optimum pH for coagulation.

Effect of coagulant dosage was also studied. The optimum dosage of 0.1g was used in all the beakers. The stirring speed was then varied ranging

from 50rpm-300rpm at 50rpm interval. After the coagulation-flocculation process was completed for each, the samples were then filtered and the filtrate was used to test for the parameters. The same was done to determine the optimum stirring time, using the optimum speed to determine the best stirring time. The stirring time was varied at 2 mins, 5 mins, 8 mins, 10 mins and 15 mins for each beaker. After the coagulation-flocculation process was completed, the samples were then filtered and the filtrate was used for the tests (1-6).

III. RESULTS AND DISCUSSIONS

The physicochemical properties of the raw water sample used in this study are presented in Table 1. From Table 1, the turbidity value of raw water was with the range of 50-150 NTU which is classified as medium turbidity water (Doerr, 2005).

Table 1 : Initial raw water properties

Parameter	Initial Result	WHO Standard
Temperature (°C)	26.7	25-30
pH	6.82	6.5-8.5
Conductivity (µS/cm)	347	1400 Max.
Total dissolved solids (mg/L)	171	933 Max.
Turbidity (NTU)	63.5	5 Max.
Colour (TCU)	330	15 Max.

From Table 1, it can be seen that the turbidity and the colour are above the WHO's recommended value for good quality drinking water. Hence the need for treatment. However, all other components are within the accepted value and safe without treatment.

*a) Effect of dosage on coagulation*

Table 2 shows the results of the effect of coagulant dosage on coagulation. The dosages were varied from 0.1g/L - 0.6g/L for each sample treated. The settling time of 15 minutes was used and the samples filtered as longer time periods were observed for complete settling to take place.

Table 2 : Effect of coagulant dosage results

S/No	Dosage (g/L)	Temperature °C)	pH	Conductivity (µS/cm)	TDS (mg/L)	Turbidity (NTU)	Colour (TCU)
1	0.1	26.2	6.34	342	170	7.59	50
2	0.2	26.5	6.44	356	178	8.39	55
3	0.3	26.5	6.52	322	167	10.69	80
4	0.4	26.4	6.49	334	165	11.68	90
5	0.5	26.4	6.40	373	186	12.71	90
6	0.6	26.3	6.36	385	193	14.98	120

At varying coagulant dosages, the effect on constituent parameters is shown above in Table 2. At varying dosage no significant changes were observed on pH, temperature, conductivity and TDS for the water sample treated with watermelon seed cake as coagulant, however, there was a notable decrease in the turbidity of the water sample after treatment. The observation on pH and conductivity made in this present study were in accordance with previous studies on coagulation and flocculation ability of some seeds (Ndabigengesere et al., 1995). The greatest decrease was seen at the dose of 0.1g/L of raw water which reduced the turbidity from 63.5 to 7.58. This value is still above the WHO recommended level of 5NTU however according to Arnoldsson et al.,(2008), the optimal dosage for a specific water is defined as the dosage which gives the lowest turbidity in the treated water therefore the optimum dosage is 0.1g/L. At this dosage the efficiency of the coagulant in removing colour was also highest, reducing the colour from 330 to 50 TCU. This value is still large in comparison with the WHO recommended standard of less than 15TCU or PCU.

According to the findings of Ordonez et al. (2010) and Alo et al. (2012), it indicated that with increase of coagulant the conductivity increases however, this is finding is in agreement with their own as seen in Table 2 above.

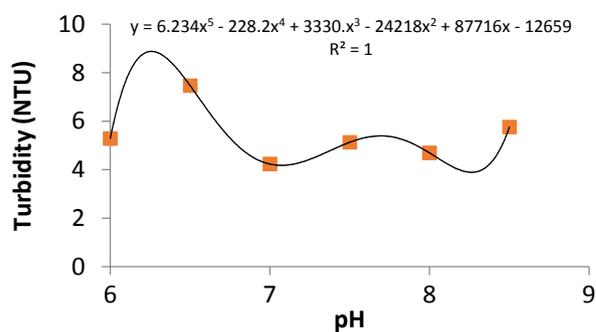
*b) Effect of pH on coagulation*

Table 3 shows the results obtained when the pH of the raw water sample was varied to study the effect of pH on coagulation.

*Table 3* : Effect of pH on coagulation results at dosage of 0.1g/L of watermelon seed cake

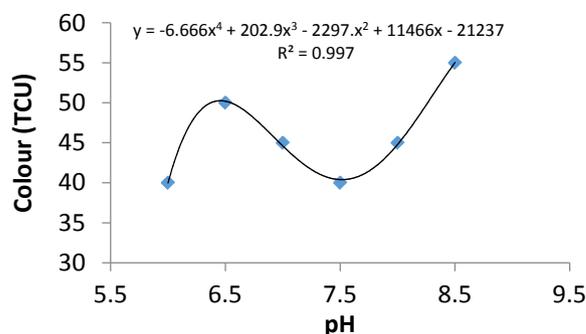
S/No	pH	Temperature (°C)	Conductivity (µS/cm)	TDS (mg/L)	Turbidity (NTU)	Colour (TCU)
1	6.0	24.4	1726	863	5.28	40
2	6.5	24.7	1588	795	7.47	50
3	7.0	24.8	1660	831	4.23	45
4	7.5	24.6	351	177	5.13	40
5	8.0	24.7	406	203	4.69	45
6	8.5	24.6	413	206	5.76	55

In the coagulation–flocculation process, pH is very important since the coagulation occurs within a specific pH range for the coagulant (Othman et al., 2008). In this study a small range of pH, between 6.0 for slightly acidic medium and 8.5 for slightly alkaline medium was selected. Figure 1 shows the pattern followed by the effect of pH on water turbidity using watermelon seed coagulant.



*Figure 1* : plot of turbidity against pH showing the effect of pH on turbidity removal

It can be deduced from Fig. 1 that, variation of the water pH resulted in a 5 degree polynomial curve with R2 value of 1. pH of between 7-7.5 provides better response in turbidity.



*Figure 3* : Plot of colour against pH, showing the effect of pH on colour removal

According to Seyrig and Shan (2007), the probable reason why different pH provides different colour is that, the colour-producing substances in water behave inconsistently. pH adjustment may cause a change in the ionization of the colour molecule with corresponding effects on bond lengths and configurations and thus light absorption.

*c) Effect of stirring time on coagulation at constant dosage*

Table 4 shows the results obtained when the effect of stirring time on coagulation was studied by varying the stirring time at a constant coagulant dosage.

*Table 4* : Effect of stirring time on coagulation at coagulant dosage of 0.1g/L

S/No	Time (min)	pH	Temperature (°C)	Conductivity (µS/cm)	TDS (mg/L)	Turbidity (NTU)	Colour (TCU)
1	2.0	7.6	23.7	421	211	4.62	65
2	5.0	7.55	23.7	424	212	4.89	55
3	8.0	7.59	23.7	418	209	3.77	40
4	10.0	7.53	23.7	423	211	4.08	50
5	12.0	7.47	23.7	411	206	4.17	45
6	15.0	7.26	23.7	462	230	5.57	70

It can be seen from Table 4 that, the effect of stirring time on coagulation and as with the effect of dosage, the results obtained showed no significant changes in pH or temperature. The temperature remained constant, over a range of stirring time. Stirring time of 2-15min was used to show this. The TDS values obtained were still below 300 mg/L which are excellent, it has its highest value at the highest stirring time. Also at stirring time of 8min a turbidity value below the WHO recommended value was obtained, best colour removal of 40 TCU.

d) *Effect of mixing speed on turbidity and colour*

From Table 5, the effect of mixing speed on coagulation was observed to only have a moderate effect on the coagulation process. This is in accordance with the findings of Othman et al., (2008).

Table 5 : Effect of mixing speed on coagulation at dosage of 0.1g

S/No	Speed (rpm)	pH	Temperature (°C)	Conductivity (µS/cm)	TDS (mg/L)	Turbidity (NTU)	Colour (TCU)
1	50	7.19	26.4	387	194	5.27	55
2	100	7.32	26.2	403	201	5.23	50
3	150	7.42	26.1	404	202	5.57	60
4	200	7.45	26.3	418	209	6.17	60
5	250	7.51	26.3	368	184	7.90	75
6	300	7.58	26.4	433	216	6.01	60

In determining the effect of stirring speed on a range of 50-300rpm at an interval of 50 rpm was chosen. Slight changes were observed for the pH and temperature values, however the pH after coagulation remained more neutral than acidic or alkaline. There was no much difference in the temperature change from the original to that after coagulation; therefore there is no marked effect on temperature. The TDS and conductivity values show a steady increase in their trend however, at speed of 250rpm there was a sharp decline in turbidity with the trend following a polynomial law (Fig. 4).

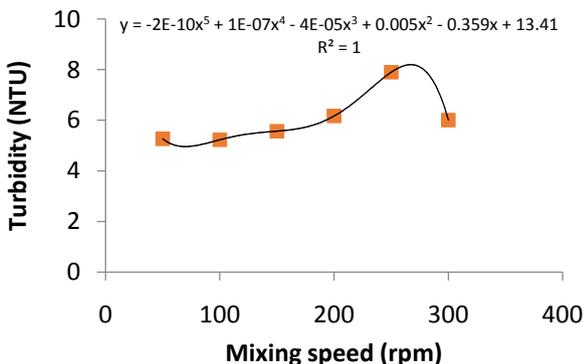


Figure 4 : Effect of mixing speed on turbidity removal

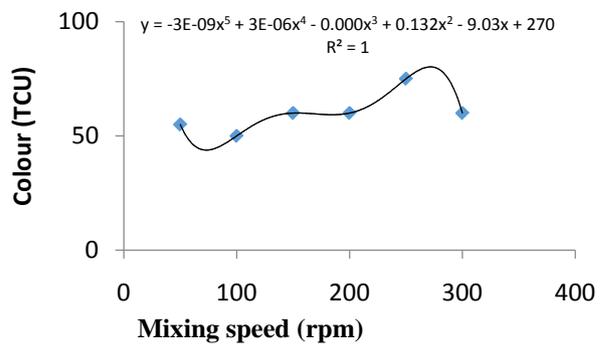


Figure 5 : Effect of mixing speed on colour removal

Also at 250rpm the turbidity and colour removal is least and best at stirring time of 100rpm with lowest turbidity value of 5.23 and colour value of 50TCU. No significant changes were recorded at speeds of 150 and 200rpm for both colour and turbidity. It can therefore mean that the lower mixing speed may improve the removal of turbidity due to reduced shearing of the flocs during initial formation which is in agreement with the findings of Ebeling et al (2004).

e) *Effect of the combination of coagulant and coagulant aid on water treatment*

Table 6 shows the results obtained after coagulation when alum was used as a coagulant aid and watermelon seed cake being used as the primary coagulant.

Table 6 : Effect of coagulant + coagulant aid on water treatment

S/No	Coagulant + Alum (Wt.%)	pH	Temperature (°C)	Conductivity (µS/cm)	TDS (mg/L)	Turbidity (NTU)	Colour (TCU)
1	100% C	7.26	25.5	419	211	6.79	20
2	80%C+20%A	6.50	25.4	472	236	0.89	15
3	60%C+40%A	6.09	25.6	478	238	0.47	5
4	40%C+60%A	4.63	25.5	564	284	1.04	5
5	20%C+80%A	4.46	25.6	646	322	1.04	0
6	100%A	4.21	25.6	873	437	0.95	5

Where:

A: Aluminium sulphate

C: Natural coagulant

The various coagulant combinations in Table 6 was used to treat the raw water sample (Gubi Dam raw water).

Results shown in Table 6, indicated significant changes in pH. With an initial raw water pH of 6.82, the sample with 100% watermelon cake, the pH recorded

was 7.26 which is neutral and well within the range of WHO standards. However steady increase in alum concentration showed a steady decline from neutral to acidic Fig. 6). This is in consonance with the results of Adejumo, et al., (2013) that showed that the pH of the water treated with natural coagulants (MO seed powder) was within the recommended WHO standards. At alum concentrations of only 40% the pH value had a significant shift towards acidic region and well within the recommended WHO standards. At concentrations of 60% and above, the pH was observed to become very acidic. This is also in consonance with the findings of Nwaiwu and Bello (2011) which showed that at alum of only 20% significant drop in pH was observed and at 80% the treated water was already acidic.

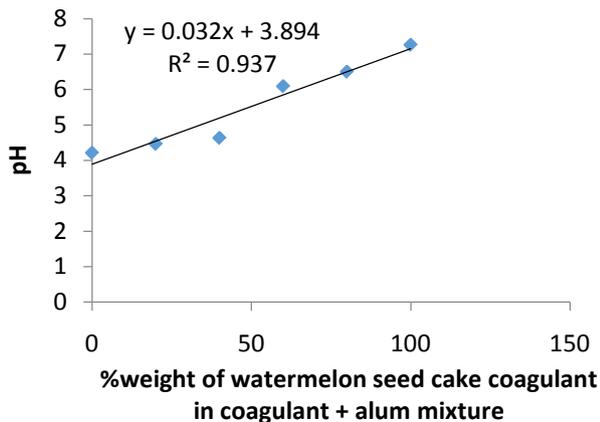


Figure 6 : Graph showing the effect of coagulant combination on pH of water

When the quantity of watermelon seed cake was more than the quantity of alum in any water sample treated, the pH reduction was within the WHO approved range of 6.0 to 8.5 but the reverse was the case when alum was either in equal quantity or greater amount than Watermelon seed cake in combination. In practical terms, this indicates that when using Watermelon seed cake alongside alum in water purification, the optimum combination should not necessitate further chemical addition for pH correction. This means that the quantity of alum in the combination should not exceed the quantity of Watermelon seed cake. The quantity of alum should be based on and should be a percentage of the Watermelon seed cake dosage suited for the water in question. Therefore a concentration of 20% alum is acceptable.

#### IV. CONCLUSION

From the study on the effects of pH, dosage, stirring speed and time, an optimum pH of 7.0, optimum dosage of 0.1g/L, optimum speed at 100rpm, and stirring time of 8 minutes were obtained respectively. Also, when watermelon seed cake was used in combination with alum higher colour and turbidity removal were observed, going as high as 100%

clarification of colour. However the recommended ratio for the combined coagulant dose was 80% watermelon seed powder and 20% alum as best water treatment was obtained. This therefore establishes that watermelon seed powder as a natural coagulant can be more efficient when used with 20% alum as a coagulant aid. From the results obtained, watermelon seed has been found to be a potential natural coagulant for surface water treatment. Medium turbid water from Gubi dam was used as case study and a good efficiency of about 89% was obtained.

#### V. RECOMMENDATION

Based on the results obtained, the following recommendations were provided:

It is recommended that further research should be carried out on pilot scale water treatment using watermelon seed as coagulant and alum as coagulant aid.

It is also recommended that more research on highly turbid water as using the water melon seed powder as coagulant.

It is recommended that more natural sources should be investigated for potential coagulation abilities.

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