Journal of science faculty at Sirte University
JSFSU
2021
Peer-review Periodical Journal

جامعة سرت
Sirte University
1989

ISSN INTERNATIONAL STANDARD SERIAL NUMBER

eISSN: 2789-858X

SCIENTIFIC JOURNAL FOR THE FACULTY OF SCIENCE – SIRTE UNIVERSITY

SJFSSJ

DOI: 10.37375/issn.2789-858X - Indexed by Crossref, USA

VOLUME 3 ISSUE 2 OCTOBER 2023

Bi-annual, Peer- Reviewed, Indexed, and Open Accessed e-Journal

Legal Deposit Number@NationaL Library (Benghazi): 990/2021

1.02/2022

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Scientific Journal for the Faculty of Science-Sirte University Vol. 3, No. 2 (2023) 1-8

Scientific Journal for the Faculty of Science-Sirte University

Journal home page: http://journal.su.edu.ly/index.php/JSFSU/index

DOI: 10.37375/issn.2789-858X

Treatment of Turbid Water Collected from Oil Mill Wastewater (OMWW) by Using Low Content of Gum Arabic Polymer (GA)

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DOI: https://doi.org/10.37375/sjfssu.v3i2.1307

ARTICLE INFO:

Received: 09 April 2023

Accepted: 02 August 2023

Published: 26 October 2023

Keywords: Gum Arabic, Turbidity, Cellulose, Kinetics, Oil mill wastewater.

A B S T R A C T

Using synthetic polymers in clearyfing of turbid wastewater is very expensive method, in addition to their monomers toxicity produced from the polymerisation reaaction. Those drawbacks encouraged several scientist who intresed in an eco-friendly sustainable materials to discover an upright replacement to clarify turbid wastewater. The present study introduces Gum Arabic (GA) as a health-friendly natural polymer flocculant, which could be applied in pre-filtration process of oil mill wastewaters (OMWW) treatment. GA-cellulose as a model work and GA-OMWW interaction was investigated experimentally by turbidimetric technique and mathematically by GraphPad Prism® software, since plateau followed by one phase decay was good fit at adding 1250 and 12500 mg of polymer, while one phase decay was selected as a proper fit for 12.5 and 125 mg. Results supposed that the biopolymer improves markedly the sedimentation performance of suspended materials. Digital micrographs agreed with the turbidity results.

1 Introduction

Acacia Gum or Gum Arabic (GA) is an exudate produced by the *Leguminosae* tree Acacia Senegal (Fig. 1). It is a complex, branching heteropolysaccharide that can be neutral or slightly acidic and is made up of 1, 3-linked *β*-D-galactopyranosyl units (Fig. 2). This natural macromolecule also contains arabinose, L-rhamnose, and D-glucuronic acid as components (Patel & Goyal, 2015; Shirwaikar et al., 2008). The branches of GA chain are composed of two to five 1, 3-linked *β*-D-galactopyranosyl units, which linkage the main chain by 1, 6-linkages. Some sources report that GA is a mixture of polysaccharides and glycoproteins (Patel & Goyal, 2015). Water dissolves the light-orange or pale-white GA fragments. These plants are found from the west coast of Africa to the Indian peninsula. The majority of GA comes from the arid plains of Sudan, Chad, Nigeria, Senegal, and Ethiopia. Sudan is the greatest exporter, accounting for up to 80% of commerce, and Nigeria is the second largest. The dried saps are extracted as translucent masses, cleaned of foreign matter, kibbled or powdered, and sold to Western countries. GA got its name from Arabian traders who brought it to Europe and helped it become popular (Patel & Goyal, 2015).

Figure (1): Tree with gum Arabic exudates (Musa et al., 2018)

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GA has been used for 5000 years ago (Nasir et al., 2008; Nasir et al., 2012; Patel & Goyal, 2015; Shirwaikar et al., 2008). GA has been used to treat chronic renal disease in Middle Eastern nations (Nasir et al., 2008; Nasir et al., 2012; Patel & Goyal, 2015; Shirwaikar et al., 2008). GA has found widespread use in the food industry due to its edibility, crucial water solubility, generally recognized as safe (GRAS) status, absence of aftertaste, and other favorable properties (Patel & Goyal, 2015).

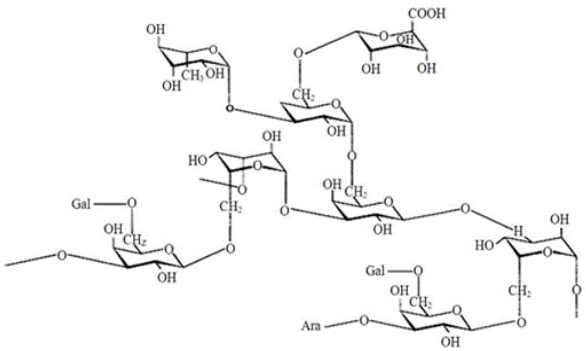


Figure (2): Structure of Gum Arabic.

GA is used in food compositions such as ice creams, jellies, candies, soft drinks, beverages, syrups, and chewing gums due to its emulsifying, stabilizing, thickening, and binding properties. Because of its film-forming characteristics, it is perfect for confectionery coatings and glazes (Patel & Goyal, 2015). Its ability to extend the shelf-life of flavors makes it appealing as a food ingredient (Nasir et al., 2012). The European Union has approved GA for food applications (Shirwaikar et al., 2008). Codex Alimentarius, an international collection of standards, codes of practice, and guidelines, has also endorsed it (Nasir et al., 2008). Its coatings pills and lozenges in pharmaceuticals and natural treatments. It is also utilized in the manufacture of creams and lotions in cosmeceuticals. It is an essential element in traditional lithography, printing, and water color paints due to its great binding property (Patel & Goyal, 2015). GA has also found application in the textile industry due to its potential to improve yarn tensile strength (Nasir et al., 2012). From this perspective, it appears useful to examine the evolution of GA technology over the last decade and the trajectory of future breakthroughs (Shirwaikar et al., 2008).

The quality of GA is determined by factors such as color, odor, moisture and ash content, viscosity, pH, specific rotation, tannins, and metal concentration (Katayama et al., 2006; Nasir et al., 2008; Patel & Goyal, 2015). Ca, Na, K, P, and traces of Pb, Co, Cu, Zn, Ni, Cd, Cr, and Mn are the most common minerals detected. Only when the quality of GA meets international requirements is shipped. As a result, constituent proportions are an important criterion in quality regulation. According to Nasir et al. (Shirwaikar et al., 2008) GA solutions have notably high concentrations of Ca^{2+}, Mg^{2+}, and K^{+}.

The Mediterranean countries that produce olive oil, such as Spain, Italy, Greece, and Tunisia, are confronted with the problem of removing the agro-industrial residues that result from this production, namely, olive-oil mill wastewaters (OMWWs), which account for approximately 1-1.6 m^3 per ton of olive fruits processed. The worldwide estimated volume of OMWW produced during the olive oil extraction process is 7-30 million m^3 per year, with Mediterranean areas producing 30 hm^3. This affluent has a high saline charge and is acidic, with a brown aqueous liquid with a hazy appearance. Its heavy organic load of sugars, tannins, polyalcohols, pectins, lipids, and primarily phenolic chemicals renders it practically unbiodegradable by ordinary biological processes (Hachicha et al., 2023). Olive oil mill wastewater (OMWW) has recently expanded dramatically due to the rapidly expanding demand for olive oil and current oil extraction processes that require large amounts of water. (Benitez et al., 1997; Lourenço et al., 2017).

The composition of the generated effluent varies according to climate and milling procedures (Tsigkou et al., 2022). OMWW has properties such as a high concentration of particles due to washing operations, a deep dark hue, an acidic pH, and a pungent odor (Tsigkou et al., 2022). When these wastes are disposed of in the environment, they produce major difficulties such as water coloring and pollution, changes in soil quality, plant growth suppression, and odor nuisance (Marques, 2001). Furthermore, direct runoff on fields reduces dissolved oxygen levels, hurting aquatic biodiversity (Marques et al., 1998). As a result, OMWW must be treated before disposal, and different treatment technologies must be combined to create an acceptable and effective means of dealing with the produced wastewater (Hamdi & Ellouz, 1992).

There are numerous strategies for treating OMWW; biological treatment, co-digestion, aerobic and anaerobic digestion are some examples (Hamdi & Ellouz, 1992). Nonetheless, the OMWW contains a high concentration of fats, lipids, and phenols, which can inhibit the growth of microbes (Cereti et al., 2004). Co-digestion is the co-treatment of one wastewater with another, which has the advantage of supplying the required pH or nutrient level for further treatment (Borja et al., 1995; Borja et al., 1995). The aerobic treatment stage can lower toxicity by reducing phenols (Borja et al., 1995; Borja et al., 1995).

This work aims to use gum Arabic (GA) as a health-friendly polymer that could be introduced as a good replacement instead of synthetic polymers in clarification of turbid water collected oil mill waste (OMWW) using low content of Gum Arabic polymer.

2 Materials and Methods

All materials in the present study have been used without further purifications unless was mentioned.

2.1 Materials

In this work GA from soluble pure, as shown in Fig. 1 was obtained from a local store. Cellulose (Sigma). Double distilled water (H_2O) (Research laboratory). Sodium Azide (NaN_3) (Sigma). Propanol (C_3H_8O) (Sigma). Olive oil mill waste (Local oil mill) in Benghazi city (Libya).

2.2 Synthetic Turbid Water: Model Work

Synthetic turbid water was prepared by dispersion of 10 grams of cellulose as a model suspended material, in 100 mL of water, immediately turbidity was recorded as a function of time, the same protocol was repeated but by adding 10, 100, and 1000 mg of GA respectively. Turbidity was measured by a conventional turbidity meter (2020 Nephelometer), after calibration with distilled water.

2.3 Kinetic Experiment

12.5, 125, 1250, and 12500 mg of GA polymer were added to 250 mL of OMWW water in separated beakers, then the turbidity was measured immediately versus time in minutes scale. For kinetic study data were fitted to an exponential function via Graph Pad Prism Software®. The final turbidity was also measured after filtration after each addition of GA polymer.

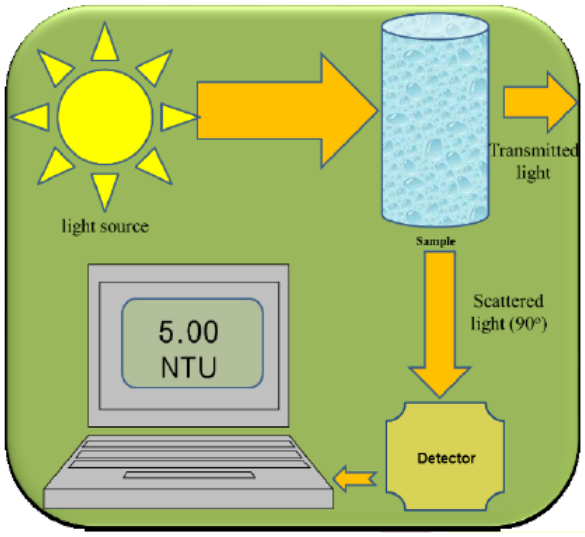


Figure (3): Turbidity measurements by 2020 Nephelometer.

2.4 Statistical Analysis

Results were expressed as means ± standard division of the mean (n = 3).

3 Results

The interaction of a polymer with suspended materials is accompanied by changes in the turbidity of the mixture solution (Eltaboni et al., 2015). Since the adhesion of this macromolecule onto solids led to settling down of the polymer chains, as a result, the suspension concentration decreases in the bulk solution, thus the turbidity of the supernatant solution will be decreased (Eltaboni et al., 2015). A versatile technique like a turbidity meter, which monitors polymeric solution transmittance, should be useful for investigating macromolecule-solids sedimentation.

3.1 Model Study of Gum Arabic-Cellulose Interaction

In order to prove the ability of gum Arabic (GA) polymer for acting as a good natural sedimentation agent, a model study was designed by monitoring the decay in the turbidity values as a function of GA-cellulose interaction time at different amounts of polymer, as shown in Fig 4. As an overall trend, the addition of polymer to the cellulose suspension exhibited a marked decay in turbidity over a stated period of time (2 hours), after this time no significant change was observed. It's worthy to note that the turbidity decay at 1000 mg of GA addition is lower than that of 10 and 100 mg of added GA, since at which turbidity decreased from about 60 NTU to 20 NTU.

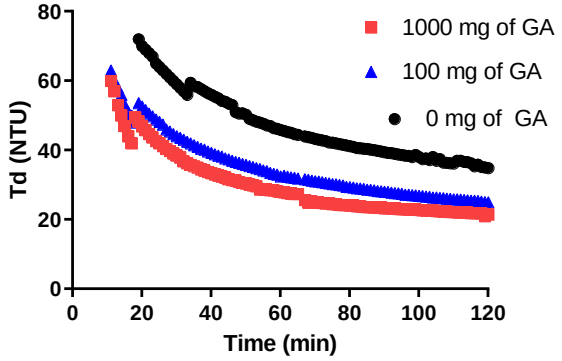


Figure (4): Turbidity decay of 10 wt% of cellulose suspension in different Gum Arabic additions (10, 100, and 1000 mg) at pH 7 and 25 °C.

3.2 Effect of Gum Arabic Content on the OMW Water Turbidity

Despite the OMWW being a turbid suspension, its turbidity varied over time from approximately 100 NTU to 65 NTU, as depicted in Fig 5. The final decayed

turbidity was still high compared with the turbidity of distilled water which is less than 5 NTU as stated by Ministry of Health standard (Azman et al., 2016).

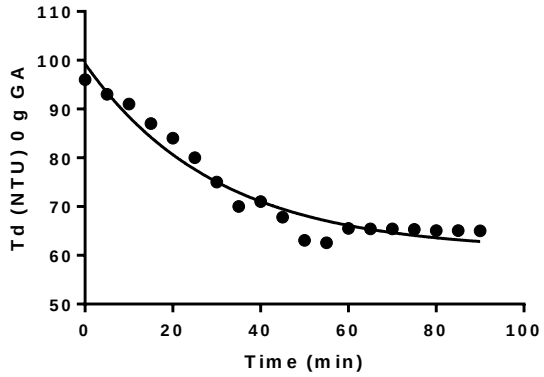


Figure (5): Turbidity of OMWW suspension as a function of time (without adding GA).

The mathematical fitting by Prism software for the experimental turbidity results in Fig 5 was carried out via using one phase model (equation 1), the generated lifetime of sedimentation for OMWW suspension and its related parameters are displayed in Table 1.

$$Td = (Td_0 - Plateau)*\exp^{(-K*t)} + Plateau \quad (1)$$

Where Td_0 is the turbidity (Td) value when t (time) is zero. It is expressed in the same units as Td (NTU), Plateau is the Td value at infinite times, expressed in the same units as Td. K is the rate constant, expressed in the reciprocal of the time in minutes. τ is the time constant, expressed in the same units as the t. It is computed as the reciprocal of K. Half-life ($t_{0.5}$) is in the time at which the half amount of OMWW is stilled down on the bottom of the container. It is computed as ln (2)/K. All the fitted data is statistically acceptable with a good regression coefficient $R^2 = 0.9507$.

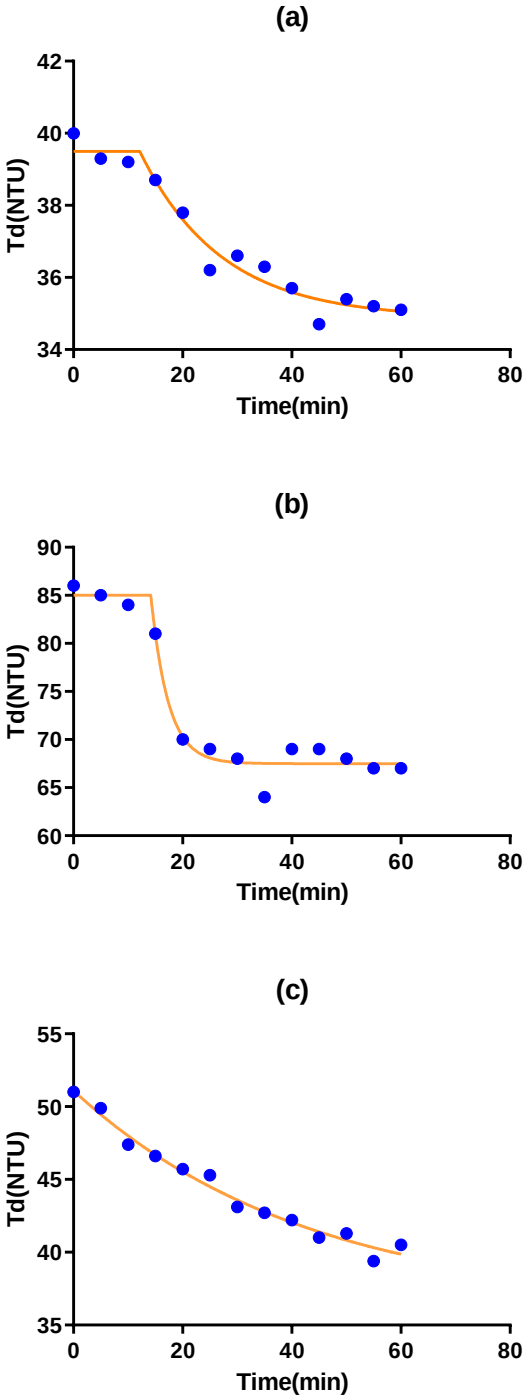
Table (1): OMWW suspension lifetime and its parameters.

One phase decay parameter	Value
K (min^{-1})	0.03329
$t_{0.5}$ (min)	20.82
τ (min)	30.04
R^2	0.9507

3.3 Kinetic study of gum Arabic interaction with OMW water

As proved by the model study, the effectiveness of GA polymer as a natural coagulant could make this naturally occurring material a credible alternative to current synthetic polymers. The kinetic study of GA-OMWW interaction was carried out by monitoring the decrease in the turbidity values as a function of time in minutes units, as shown in Fig 6 (a, b, c, and d), the dots represent the real data and the solid line decay represent the mathematical fitting. Fig 6 (a) and (b) plateau followed one phase least squares fit was found suitable to simulate the experimental results (equation 2).

$$Td = IF(t<t_0, Td_0, Plateau+(Td_0-Plateau)*\exp^{(-K*(t-t0))}) \quad (2)$$



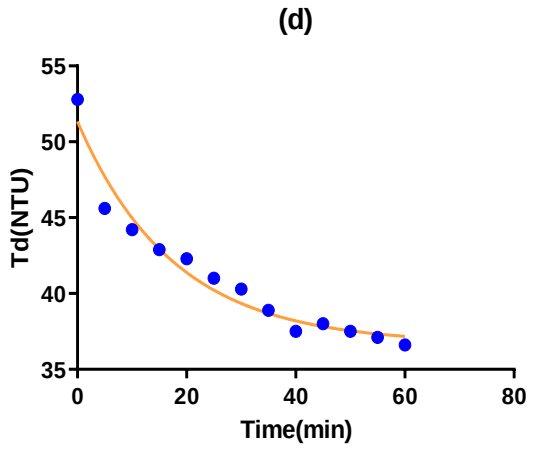


Figure (6): Kinetic decay of OMWW-GA interaction at 25 ºC an pH 7, adding 12.5 mg (a), 125 mg (b), 1250 mg (c), and 12500 mg of GA polymer.

Unlike the Fig 6 (a and b), in Fig 6 (c and d) the mathmatical fitting for the exprimental data was done by using one phase least sequares fit (equation 1), the produced lifetime of that interaction and its related parameters are dipicted in Table 2.

Table 1. Kinetic data for OMWW-GA interaction.

Kinetic parameter	12.5 mg of GA	125 mg of GA	1250 mg of GA	12500 mg of GA
K (min^{-1})	0.06675	0.3124	0.02342	0.05688
$t_{0.5}$ (min)	10.38	2.219	29.60	12.19
τ (min)	14.98	3.201	42.70	17.58
R^2	0.9599	0.9726	0.9806	0.9578

As shown in Table 2, the rate constant of GA-OMWW interaction had its maximum value when 125 mg of the polymer was added, while the lower value of K was recorded during adding 1250 mg of GA. In contrast, the half and lifetimes of interaction reached the maxima at the addition of 1250 mg of polymer. But only 3 minutes were enough to get an interaction between the suspended materials and the polymer.

Fig 7 shows the turbidity dependence on added polymer, which was measured after filtration to remove the precipitated GA-OMWW. As an overall trend, increasing the amount of added polymer was accompanied by a slight increase in the turbidity of the bulk solution.

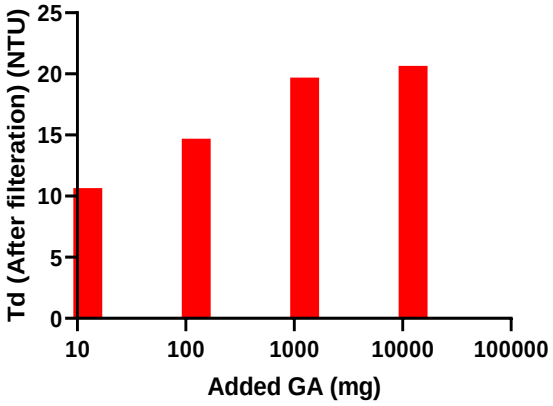


Figure (7): Turbidity of bulk solution as a function of GA amount after filteration (at natural pH and room temperature)

3.4 Digital Micrographs of OMW Water Samples

As it can be observed from the turbidity results, when the GA polymer was added to the cloudy OMWW suspension a clear water was formed. To support that results digital micrographs were taken for OMWW treated with GA as shown in Figure 8.

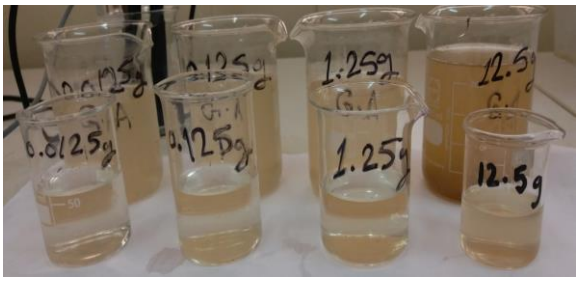


Figure (8): Clreafication of OMWW by adding different amount of GA polymer.

4 Discussion

Fig 4 shows the kinetic study of GA interaction with a cellulose suspension, as a model work, the experiment was determined by monitoring the variation in turbidity values as a function of time in minutes scale and was done at room temperature in natural pH medium. As it can be noted that without adding GA polymer the value Td decreased up to 40 (NTU) but by adding GA the value decreased to about 20 (NTU), this could be because of a quick interaction (occurred in 30 minutes, Table 1) occurred between GA chain and cellulose molecules, as proposed in Fig 9, the result of this binding let the suspended cellulose to settle down in the bottom of the aqueous solution, as a result, a clear solution (decrease in turbidity) is noted. This study proved that model-suspended cellulose was a good candidate to examine the

ability of GA to eliminate any suspensions that could exist in the OMWW sample. The suggested mechanism is in good agreement with our previous studies which used the fluorescence technique to monitor silica binding with polymers similar in structure to GA like polyacrylic acid, alginate, and lipopolysaccharides (Sparks et al., 2015; Eltaboni et al., 2020; Eltaboni et al., 2022).

Table 1 displays lifetime (τ) and its parameters for OMWW suspension. The lifetime was determined using the turbidity technique. A one phase decay model (Equation 1) was used to fit the kinetic data, hence the stability of the suspension lifetime was simulated. The results in Table 1 show that the first order model is a good fit for experimental data as its corresponding R^2 value is considerably high (0.9507). The turbidity values of suspension were found to be time dependent. It was found that the turbidity decays until reaching a plateau with the lifetime reached to 30 minutes, combined with a rate constant equals 3.3 x 10^{-2} min^{-1}.

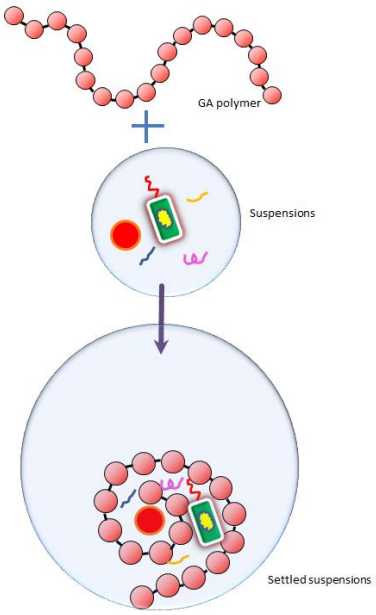
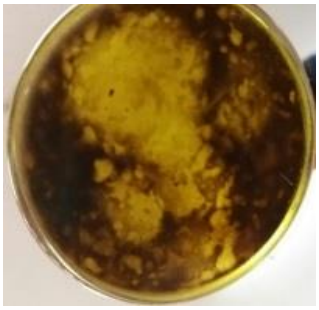


Figure (9): Proposed scheme shown cloudy water suspended with different materials turned clear when GA was added.

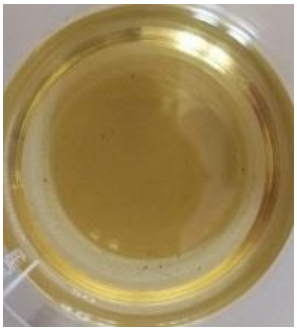
When OMWW samples were treated with 12.5, 125, 1250 and 12500 mg of GA (Fig 6 a-d), it was interestingly found that at low content of gum Arabic the polymer-OMWW particles remain suspended in solution for a couple of minutes and then stilled down on the bottom as a result of both particle interaction. This suggestion was confirmed by plateau following one phase model with a good regression coefficient (Table 2). On the contrary, at adding 1250 and 12500 mg of GA a direct interaction happened between polymer and OMWW, which was confirmed by one phase decay. However, increasing the amount of polymer could let some of it undissolved in solution and this was accompanied by an increase in the turbidity values (Fig 7).

The proposed mechanism in Fig 9 was established by Fig 10, in which the variation in clarification of OMWW suspension upon adding polymer proposed that a kind of interaction occurred between suspended materials and GA.

(a)



(b)



(c)

Figure (10): Initial OMWW (a) and OMWW after treatment with GA flocculant(b), and filtrate (c), in optimized conditions of pH and concentration.

5 Conclusion

The experimental results showed that the highest efficiency for GA as water clearer was increased by increasing its content to 12500 mg per 250 mL of treated

OMWW samples, before filtration. Possibly, the increase in the amount of settling suspension upon adding polymer causes a decrease in the turbidity. Since the stronger interaction between the GA-OMWW is more favorable. Digital micrographs agreed with the turbidity results, these proposed that health friendly polymer (GA) could be a promising material to use in OMW treatment, but further examination should be carried out to get pure water.

Acknowledgements

Authors would like to thank the chemistry department and faculty of science at the University of Benghazi for their support.

Conflict of Interest: The authors declare that there are no conflicts of interest.

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