THE EFFICIENCY OF GUM-SILICA CMPOSITE FOR REMOVING OF WASTEWATER TURBIDITY

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ABSTRACT

A novel mixed natural coagulant has been developed to remove sewage pollutants and heavy metals from Qanat- al- Jayesh by using low cost adsorbent natural materials. In these materials, significant interaction contains Arabic gum mixed with extracted silica from rice husk ash (natural coagulants) by the Batch device approach, using two variables, pH values ranging from 5-8 and contact times between 0.25-5 hrs. All wastewater samples were collected after treatment by adsorbents and examined for determination of residual heavy metal concentrations: Pb, Ni, Zn and Cu by atomic absorption spectroscopy (AAS), turbidity, pH, total dissolved salts (TDS), electrical conductivity (EC) and total salinity (TS). The results obtained indicate The coagulation process' highest level of effectiveness was 95.2% for gum-silica composite with the weight 8 gm for reducing turbidity. In comparison, The coagulation process's least effective efficiency was 80.6%, with the weight 4 gm of gum-silica composite. On the other pH values, Turbidity, TDS, TS, and EC were reduced in the waste water sample after being treated by gum-silica composite under standard water values. This mixture can be used to remove heavy water pollutants during treatment.

Keyword: Arabic gum, Rice husk, Heavy metals, AAS, TDS.

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كفاءة مركب صمغ - سيليكا في إزالة عكورة مياه الصرف الصحي وبعض الملوثات الأخرى

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المستخلص

تم تطوير خليط من مختثر طبيعي جديد لإزالة ملوثات مياه الصرف الصحي والمعادن الثقيلة من مياه قناة الجيش في مدينة بغداد باستعمال مواد ممتزة طبيعية منخفضة التكلفة كمركب. يحتوي هذا المركب على ارتباط معنوي نشط بين الصمغ العربي مع السيليكا المستخلصة من رماد قشر الأرز وباستعمال نظام الدُفعات والأعمدة ، وباستخدام متغيرين (معلمين) ، قيم الأس الهيدروجيني التي تراوحت بين 5-8 ووقت التلامس الذي كان متفاوتًا من 0.25-5 ساعات. تم معالجة عينات المياه العادمة المجمعة بوساطة مركب الممتزات و تم فحصها لتحديد تراكيز المعادن الثقيلة المتبقية: Pb و Ni و Zn و Cu بوساطة مطيافية الامتصاص الذري (AAS). بالإضافة إلى العكورة ، ودرجة الحموضة ، وإجمالي الأملاح الذائبة (TDS) ، والتوصيلية الكهربائية (EC) ، والملوحة الكلية (TS). أظهرت النتائج أن أفضل كفاءة لعملية التخثر كانت 95.2٪ لمركب صمغ السيليكا بوزن 8 غرام لتقليل العكورة. في المقابل ، كانت أقل كفاءة لعملية التخثر 80.6٪ بوزن 4 غرام من مركب صمغ السيليكا. من ناحية أخرى ، تم تقليل قيم الأس الهيدروجيني ، العكورة ، والمواد الصلبة الذائبة ،TDS و TS ، و EC في عينة مياه الصرف بعد معالجتها بمركب الصمغ - سيليكا تحت قيم المياه القياسية. يمكن استخدام هذا الخليط لإزالة ملوثات الماء الثقيل أثناء المعالجة.

الكلمات المفتاحية: الصمغ العربي، قشر الأرز، المعادن الثقيلة، تقنية الامتصاص الذري، مجموع المواد الصلبة الذائبة.

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INTRODUCTION

In several low-income nations, a significant quantity of natural water is used for agricultural, industrial and other operations. Substant amounts of untreated waste water are disposed of directly from natural water supplies (5,6,40,41). Sometimes wastewater also contains heavy metals that are a potential concern for the environment and human safety (2). Heavy metals have been broadly released into the environment and particularly into the water due to rapid industrialization, which has caused incredible worldwide concern. Cadmium, Chromium, Copper, Lead, Mercury, Nickel, and Zinc are frequently distinguished in mechanical wastewater, which came from metal plating, mining exercises, refining, battery make, tanneries, petrol refining, paint make, pesticides, color production, printing and photographic businesses, and so on (33). Technologies for waste water management may be physical, chemical, or biological (16). A Physicochemical therapy such as; adsorption, coagulation, electrocoagulation, ion exchange, precipitation, and reverse osmosis. One of the most critical phases in water and wastewater treatment plants is the process of coagulation-flocculation, where plankton, colloidal particles and contaminants are precipitated and extracted at this point. With the assistance of coagulants, the charge of the colloidal particles is destabilized in this process (typically Al or Fe salts) this results in the development of flocculation due to the collision of destabilized particles and their aggregation, which is gradually removed from the liquid process (38). In the flocculation process, two main forms of coagulants are used, chemical or natural product. (animal or plant origin). Natural coagulants injected into the water to remove the causes of colloids balancing and stringing up in the water. The suspended solids begin to agglomerate after the coagulant is injected into the water, enlarge further in order to isolate and segregate these suspended solids from the water suspension (24), and is very effective for the treatment of waste water. From literature studies, some researchers have presented that the adsorption of less expensive natural coagulants are supplant exorbitant waste water treatment techniques such as: electrodialysis, electroflotation, chemical precipitation of compounds, ion-exchange, reverse osmosis, solvent extraction, and membrane separation attracts the interest of researchers (30). Lately, the requirement for the protected and economical methods for the expulsion of heavy metals from contaminated waters has required concentrating minimal expense agrarian waste, such as rice husk (10,11,21,23,36). Rice husk ash may have significant biosorption capacity for bioremoval heavy metal ions from industrial wastewater (7,19,26,35) , arabic gum (13,15,17) and so on, the removal of metals from wastewater have been researched by various experts (30). Our paper's goal is to determine the effectiveness of plant products in the treatment of waste water that uses the Batch system approach and has high turbidity and heavy metal concentrations, with determine the appropriate plant waste for treatment based on the shifting of a number of parameters. This study used four various samples plant products that prepared in a lab to act as natural coagulants, using silica-modified with arabic gum prepared naturally from rice husk ash locally available, less costly and more efficient materials. A composite estimation of some parameters such as: Turbidity, TDS, TS, and EC of waste water Qanat al-Jaish (Army canal water).

MATERIALS AND METHODS

Preparation of wastewater: Two liters of wastewater samples were collected from Qanat al-Jaish in eastern of Baghdad city, around 50 cm beneath the canal's surface water. Laboratory tests for waste water of many factories' pollutants were carried out before treatment to know the physical and chemical properties, which are shown in Table.1.

Table1. Initial properties of wastewater sample before treatment compared with standard values (28).

The Parameter	The Unit	Initial Values	Standard Values
pH	-	7.72	6-9.5
Turbidity	UNT	50	5
TDS	PPM	2033	933
EC	S/cmμ	4066	1400
Salinity	%	200	0.8
Pb	PPM	10	0.1
Ni	PPM	36	0.2
Zn	PPM	13	2
Cu	PPM	7	0.2

Preparation of arabic gum solution

Arabic gum (Acacia gum) bought from local Iraqi markets was crushed by a domestic blender until it became a powder. Dissolve 1 g of this powder in 1 liter of distilled water and stir well for 5 minutes. It is then filtered through a muslin cloth to produce a 0.1% concentration of the solution.

Collection and washing of rice husk samples

The rice husk samples were gathered from a local rice mill factory. Wash the rice husks well with deionized water with continuous shaking for two hours, then separate the water. This process was repeated four times until all visible excess material was removed from the rice husk. Then the rice husks are placed on a large watch glass in a heated oven at 125 ° C for 18 hr. Then, the dried rice husks were sieved using a 200 μm sieve to remove the finer fines. The larger particles of cleaned rice husks were stored in an airtight polypropylene container to avoid moisture absorption from the atmosphere. The RHA obtained in the experiment was used without any pretreatment.

Ash preparation from rice husk.

The rice husk is burned in an electric furnace at under atmospheric condition, at 700 ° C for 3 hr. 150 gm of rice husk ash was obtained from 200g of raw rice husk. In this experiment, the final ash from burning whole raw rice husk particles was 75% white ash particles Figure (1). Hence, the combustion time determined from this experiment was based on the best estimate, whereby no further change in the color of the ash product observed. These results it was better than those by Shinohara et.al(37), that was 14-35% of rice these differences depended on the varies of rice. Rice husk was utilized to create silica gel and powders(39).

Figure. 1. White ash particles (39).

Figure. 2. Powder of silica extract

Silica extraction

A total weight 1500 g of rice husk ash was added to 5L of basified distilled water concentrated of sodium hydroxide solution 2M. The mixture was then heated by hot plate stirrer at 100 °C for 1hr. Then separated through a 41μm filter paper and washed with 100 ml of deionized water to remove the remaining carbon. The filter and wash material was left to cool at room temperature. Drops of sulfuric acid were added to the solution until pH 7 was reached and left for 48 h. to allow silica gel formation. The resulting silica gel was separated from the soluble salt solution by vacuum filtration and washed with de-ionized water. The silica gel was then dried at 150 °C for 48 hr, and grinded into a white powder. The basic extraction method was chosen for the production of silica in the remaining experiments (34). Silica production by the basic extraction method showed higher amount of silica content reached to 87% as illustrated in Figure(2), and Table.2. Other research has repeatedly shown that rice husk has a significant silica concentration. such as (87.7%) (8), (90%) (27).

Table 2. Ratio of ash and silica production from rice husk

wt. of rice Husk	wt. of ash	Ash husk ratio	wt. of silica	Product ratio of silica husk	Product ratio of silica ash
200 gm	150 gm	75%	130 gm	65%	87%

Coagulant preparation: The coagulant was prepared by adding 10 g of Arabic gum with 100 ml of distilled water in a beaker and then placed on a magnetic stirrer at 20 °C until the Arabic gum completely dissolved, then adding 25 g of silica prepared in the previous step with 120 mL of ethanol in the beaker and mix them well, then add 25 mL of NH_4OH to the mixture and mix using a magnetic stirrer for 14 hours, then leave the mixture to dry in the incubator to form a solid consistency, then the mixture is finally burned in a convection oven at 600 °C.

Coagulation experimental studies

Two flasks for coagulation containing 200 mL of waste water per flask were prepared with pH 7.72. Different amounts of coagulant 4 g and 8 g were added separately to each flask and left for 60 minutes. 10 mL were taken from each flask and placed in eight tubes and shaken for 2 hrs in order to coagulate the contaminants. Then the flasks were then left for 30 minutes to allow the water to separate. Turbidity, TDS, TS, and EC values were measured after coagulation of treated wastewater samples.

Experimental studies of heavy metals removal: The study was conducted on the absorption and removal of heavy metals from waste water in a column of a translucent glass with a 30 cm height and a 2 cm inner diameter. The 8 g of Arabic gum -silica compound adsorbents were confined to the column by a 1 µm fine Teflon (PTFE) filter at the bottom of the column and a glass layer above the fluidized bed reactor (FBR) equivalent to a distance of 21 cm for the adsorption solution as shown in Figure (3).

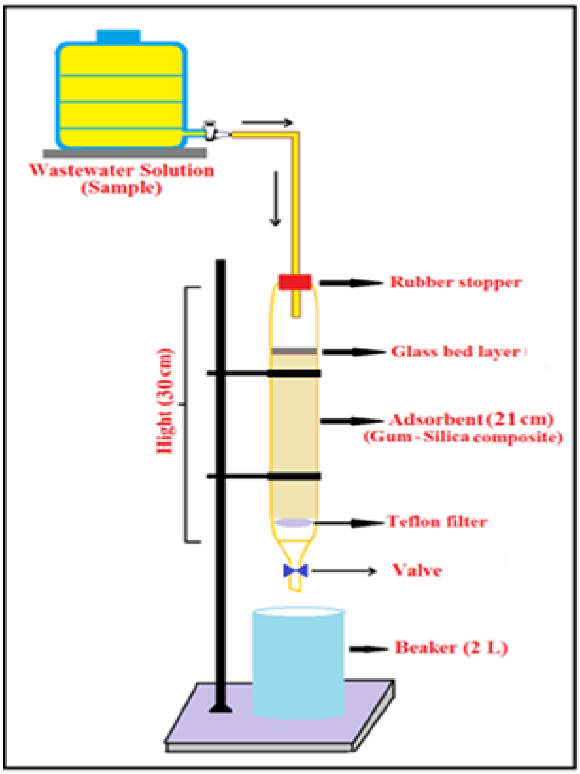

Figure. 3. Schematic of experimental column Fluidize bed reactor (FBR)

Then waste water is passed over a compound of arabic gum and silica filled inside the column to determine and estimate the efficiency of removing the absorbent material for wastewater treatment according to Box-Wilson design by studying the effect of two variables, the contact time that ranged between 0.25-5 hr. (3). All wastewater samples were collected after being treated by adsorbents and examined to determine the concentrations of residual heavy metals Lead, Nickel, Zinc and Copper by atomic absorption spectroscopy in the laboratory.

Calculations

The following equation was used to calculate the coagulation efficiency (18).

Efficiency of Coagulation (EC)%= $(C_0-C_e)/(C_0)$ X100……………(1)

"Equation (1) is coagulation efficiency" where:

C_0 = the initial concentration

C_e = the final concentration

A Langmuir model describes monolayer adsorption over distinct proximal adsorption sites. It shows that the adsorbent does not move in the plane of the surfaces and assumes that the uniform energies of monolayer adsorption on the outer layer of the absorbent material (9). The Langmuir equation is often written as a linear model to allow easy verification of whether the data conforms to the Langmuir model (1), which is expressed as:

$$\frac{Ce}{Qe} = \frac{1}{ab} + \frac{Ce}{b} \quad \text{……………(2)}$$

"Equation (2) is Langmuir model" where **C_e** is the concentration of heavy metal from wastewater (mg/L) at equilibrium, **Q_e**, maximum adsorption capacity, is the amount of heavy metal absorbed from wastewater at equilibrium (mg/g), **a** and **b** are the adsorption constant of Langmuir (L/mg) at 25 ^oC, concerning the adsorption energy. The Freundlich isotherm portrays adsorption measures that happen on heterogeneous surfaces and active sites with various energies dependent on multilayer adsorption and the equilibrium (9,12). The linear type of the Freundlich isotherm model is addressed as follows (14,22):

$$\log Qe = \frac{1}{n}\log Ce + \log K \quad \text{……………(3)}$$

"Equation (3) is Freundlich isotherm" where **K** is the constant of Freundlich isotherm, **n** is the intensity of adsorption, **C_e** is the concentration at the equilibrium of adsorbate (mg/L), **Q_e** is the heavy metals adsorbed amount at equilibrium (mg/g).

Statistical study

All data were studied and a statistical program, Anova test at (probability≤ 0.05) is used to analyze data statistically.

RESULTS AND DISCUSSION

Table 3. show the concentrations of heavy metals in wastewater after treatment by Arabic gum – silica composite.

Table 3. Concentrations of heavy metals in wastewater after treatment by Arabic gum – silica composite

Exp. No.	The variables		Concentration of heavy metals (ppm)			
	p H	C.T. (hr)	Pb (10)	Ni (36)	Zn (13)	Cu (7)
1	5.439	0.946	1.7	8	3.5	2.5
2	5.439	4.304	1.7	7.5	3.0	2.1
3	7.561	4.304	3.2	3.4	1.6	4.0
4	7.561	0.946	3.2	3.8	1.6	3.8
5	5	2.625	1.2	10.0	3.8	2.7
6	8	2.625	4.0	2.7	1.1	4.5
7	6.5	0.25	2.4	4.0	2.6	3.0
8	6.5	5	2.7	4.8	2.8	3.8
9	6.5	2.625	2.2	3.5	2.0	3.3
10	6.5	2.625	2.2	3.5	2.0	3.3
11	6.5	2.625	2.2	3.5	2.0	3.3
12	6.5	2.625	2.2	3.5	2.0	3.3
13	6.5	2.625	2.2	3.5	2.0	3.3

Effect of setting time on silica gum coagulant compound turbidity percentage

The time is one of the effective factors in the turbidity process. The degree of turbidity in UNT unit was studied against time for samples of polluted water and treated with gum and silica compound as two groups **A** and **B**, with weight 4 and 8 gm, respectively. The turbidity was measured at 30, 60, 90, and 120 minutes for each group and a comparison was made between them. The results showed in Figure (4, 5), and that the silica gum and the coagulant compound showed the best coagulation efficiency for the experimental physical and chemical parameters of both **A** and **B** groups. The turbidity ranged from 2.4 - 9.7 at 30 min.

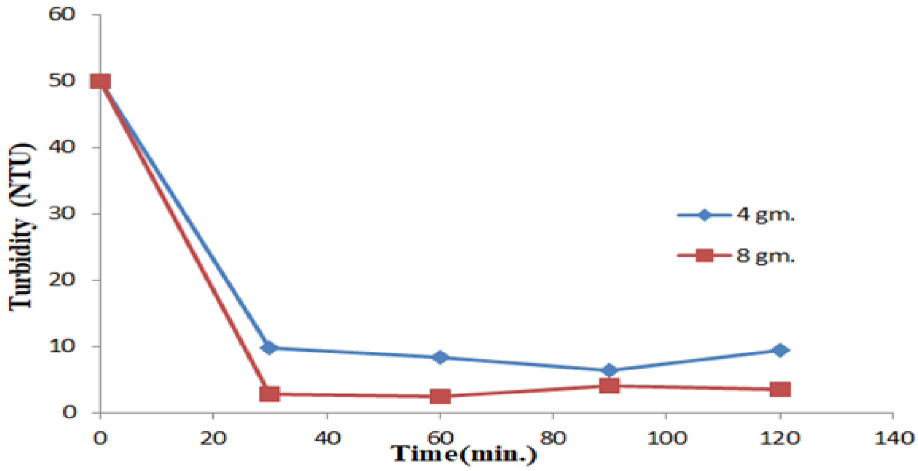

Figure. 4. Turbidity vs. Time

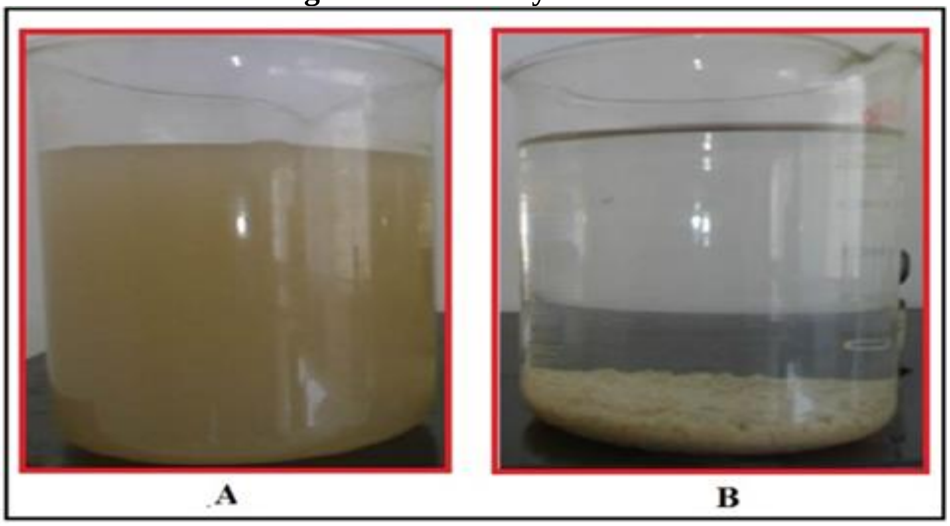

Figure.5. Reducing of turbidity of waste water sample by gum- Silica composite

Effect of setting time on silica gum coagulant compound total dissolved solids (TDS): It is found that the TDS concentration decreased considerably with the addition of silica gum coagulant compound. However, the TDS concentration was reduced immensely with 4 gm and 8 gm of gum-silica composite is 371.0 g/L and 371.0 g/L at a retention time of 90 and 60 minutes pH 7.02, respectively. The sudden decrease indicated that silica gum coagulant compound particles take suspended for a period of time after it eventually settled out. Figure (6) shows the time in minutes of TDS removal plots of the TDS removal using silica gum coagulant compound with a function of retention time, employing higher dosages, between 4-8 gm of gum-silica composite.

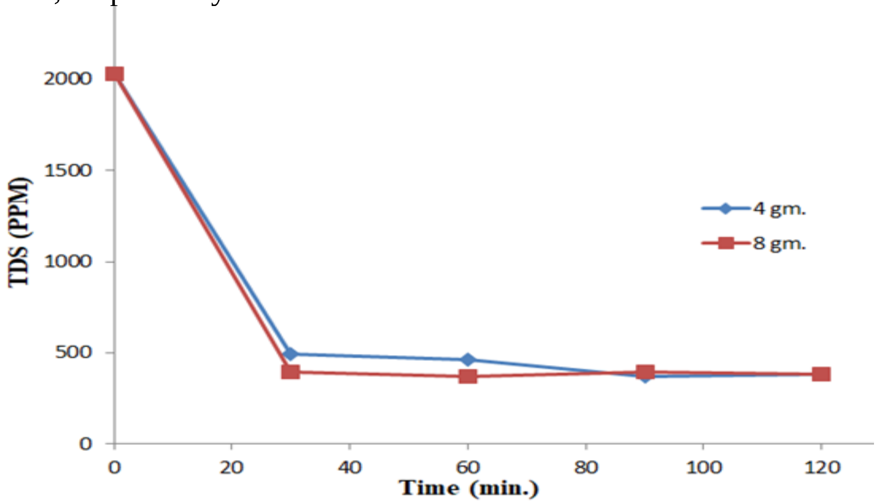

Figure. 6. TDS vs. Time

Effect of setting time on silica gum coagulant compound electrical conductivity, EC: The effect of changing the initial electrical conductivity on the efficiency of removing opacity from wastewater was studied. Maximum efficiency was observed by about 0.82% and 95.2, at 90 and 60 minutes, respectively, for 4 gm and 8 gm of gum-silica composite. The results showed that the removal efficiency increased significantly with the increase in electrical conductivity. In our paper, the impact of changing the underlying electrical conductivity was inspected for wastewater, as indicated by different rules, while keeping different factors consistent Figure (7). There is an immediate connection between the measure of coagulation delivered by the electrodes and the electrical conductivity coefficient. The decreasing electrical conductivity operates as energy progresses between the cathodes (resistance expansion) Figure (8), resulting in more coagulants and reducing the process's current costs.

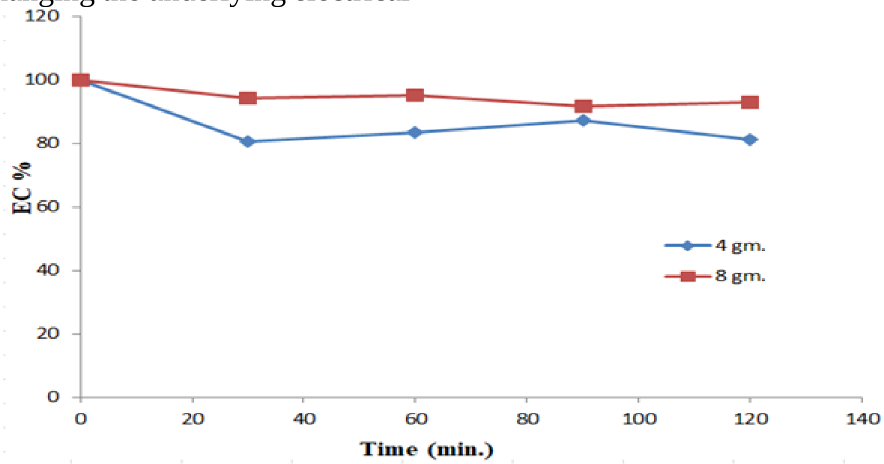

Figure. 7. EC % vs. Time

Table 4. Effectiveness of an Arabic gum-silica composite for heavy metal removal

Exp. No.	The variables		Removal Efficiency (ER %)			
	pH	C.T. (hr)	Pb	Ni	Zn	Cu
1	5.439	0.946	83	77.7	73	64.28
2	5.439	4.304	83	79	76.9	70
3	7.561	4.304	68	90.5	87.7	42.8
4	7.561	0.946	68	89.4	87.7	45.7
5	5	2.625	88	72.2	70.7	61.4
6	8	2.625	60	92.5	91.5	35.7
7	6.5	0.25	76	88,8	80	57.1
8	6.5	5	73	86,6	78.5	45.7
9	6.5	2.625	78	90.3	84.6	52.8
10	6.5	2.625	78	90.3	84.6	52.8
11	6.5	2.625	78	90.3	84.6	52.8
12	6.5	2.625	78	90.3	84.6	52.8
13	6.5	2.625	78	90.3	84.6	52.8

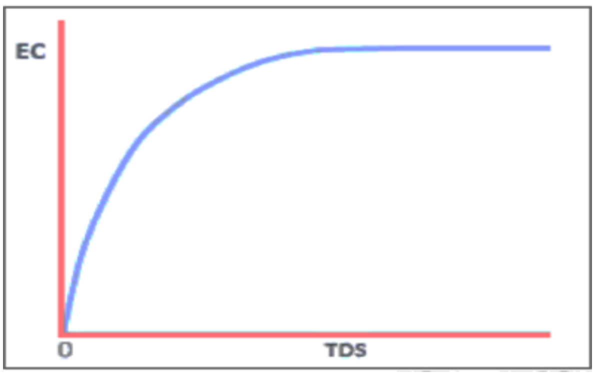

Figure. 8. The relationship between TDS and EC values (25).

Effect of removal ratio on the contact time: The effect of changes in reaction time on the rate of removal was studied. Figure (9) and Table.4. show the efficiency of removing Pb, Ni, Zn and Cu from wastewater. Based on the results, elements up to 0.946 hrs., were removed linearly to about 83, 77.7, 73.1 and 64.3% for Lead, Nickel, Zinc and Copper, respectively. This percentage was about 78, 90.3, 84.6, and 52.9% at 2.625 hours, respectively.

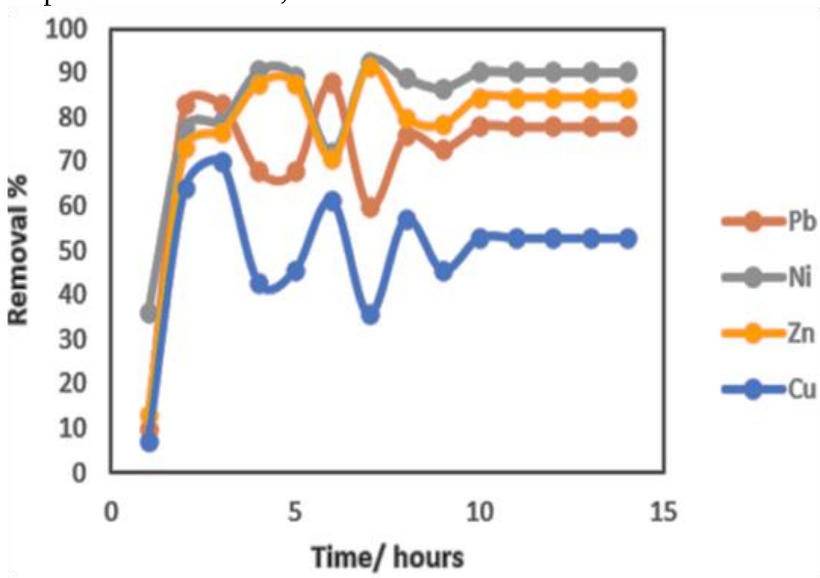

Figure. 9. Effect of reaction times on removal efficiency of heavy metals in wastewater after treatment by Arabic gum – silica composite

Figure (10) shows Removal effeicincy (RE %) & No. of Samples.

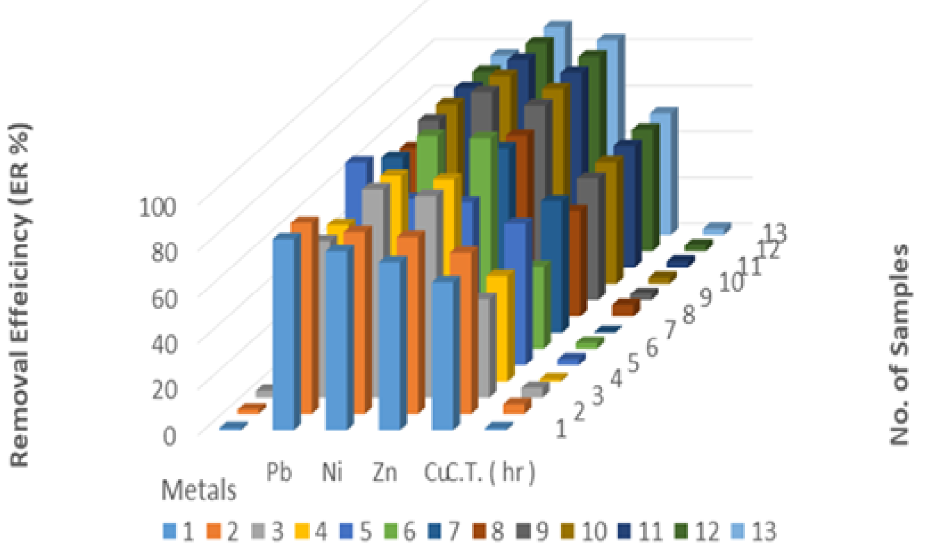

Figure. 10. Removal efficiency (RE %) & No. of Samples

Isotherm of adsorption on arabic gum– silica composite: It appears that the adsorption of arabic gum - silica compound is a great option for removing nickel to a higher degree than the rest of the contaminants, followed by Zinc, Lead and then Copper the reason may be related to the ability to exchange ions of the same charge in aqueous solutions increases with the smaller the size of the hydrated ion. It is known that there is an inverse relationship between the ion exchange capacities of the adsorbent with the size of the ions. That is, the smaller the size of the ion, the greater the ion exchange capacity. Please correct the reason in this section, as shown in Figure (11).

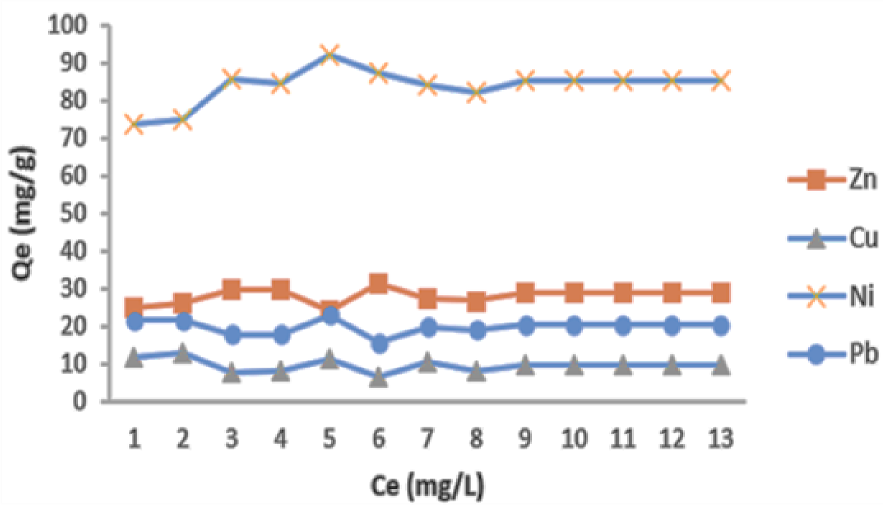

Figure. 11. Adsorption isotherm for Ni, Zn, Pb, and Cu ions in wastewater removal by arabic gum – silica composite

Figure (12 &13) summarize the results. The isothermal constants were calculated from the slope and intersection of these figures and they are shown in Table (5 & 6).

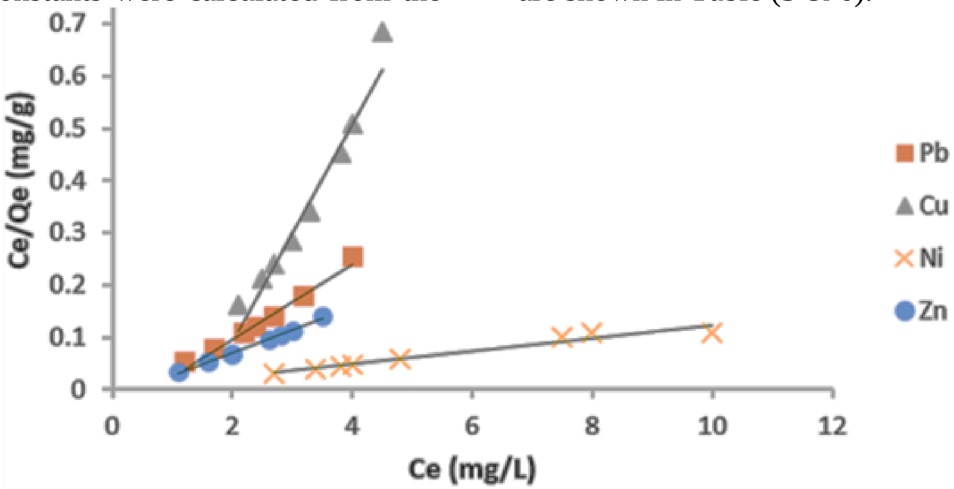

Figure. 12. Langmuir adsorption isotherm for the adsorption of Ni, Zn, Pb, and Cu ions in wastewater removal by arabic gum – silica composite at 25˚C

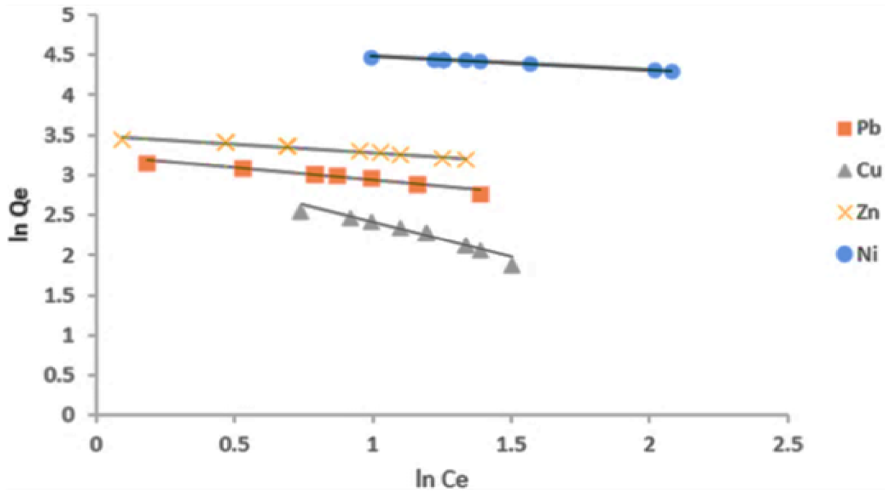

Figure. 13. Freundlish adsorption isotherm for the Adsorption of Ni, Zn, Pb, and Cu ions in wastewater removal by arabic gum – silica composite at 25˚C

Table 5. Langmir adsorption isotherm vales on arabic gum – silica composite at 25˚C

Heavy metals	Langmuir Isotherm Constants b (L/mg)	a (mg/g)	Correlation of determination (r^2)
Ni	-0.1226	-0.0015	0.9488
Zn	-58.1395	-2.5523	0.9952
Pb	-21.3675	-1.5299	0.9849
Cu	-3.0469	-0.3282	0.9502

Table 6. Frendlish adsorption isotherm vales on arabic gum – silica composite at 25˚C

Heavy metals	Frendlish Isotherm Constants n	k	Correlation of determination (r^2)
Ni	-5.9808	12.6527	0.9792
Zn	-4.7103	9.5091	0.9494
Pb	-3.1959	8.8312	0.9335
Cu	-1.1740	8.8621	0.9458

The r^2 correlation of determination values obtained was higher in the Langmuir isotherms than those in the Frendelich isotherms for arabic gum – silica composite at 25˚C, indicating that the adsorption process is good represented in the Langmuir equation. As per the r^2 for every parameter in Table (5), the Langmuir model fitted the exploratory information best by straight investigation, while the Freundlich was less fitted. These outcomes are in acceptable concurrence with the outcomes demonstrated by nonlinear analysis. This study showed the applicability of gum arabic - silica compound as a sustainable adsorbent that can be used in two stages in removing suspended pollutants and heavy metals Ni, Pb, Zn and Cu from wastewater. The results of the batch experiments indicated that the adsorption process was affected by the adsorbents dose, contact time, gum arabic concentration and silica compound with the constant temperature. Isotherm studies have shown that Langmuir can describe adsorption, a Freundlich isotherm with maximum adsorption capacities of - 0.0015, -2.5523, -1.5299, and -0.3282 mg/g for nickel, zinc, lead, and copper, respectively. As determined by a one-way test (Anova) with a F and p value at the P ± 0.05 probability level as shown in the following Table (7).

Table 7. ANOVA statistical tests for eliminating of heavy metals from wastewater

Type of Heavy Metal	Number of Subjects	Mean	Standard Deviation	F- Value	P- Value
Pb	9	2.47777	0.88991	7.365	0.001
Ni	9	5.3	2.55		
Zn	9	2.44444	0.92481		
Cu	9	3.3	0.78740		

At Level of probability (P) ≤ 0.05

The current study has been approved with a study of Majeed et al. (29). That found that rice husk showed a high adsorption efficiency, 99.02% of Ni occurred at 0.15 h. At the same time, lower removal efficiency of 94% was obtained for zinc at 2.83 hours. Majeed et al. (4) found that rice husk ash had the best lead removal efficiency from 77.75% - 71%, according to the adsorbents and the type of heavy metals in this study, and Majeed et al. (20) found that arabic gum a high adsorption efficiency, for removal were 80% for lead, 68.75% cobalt, and 90.7% cadmium, at pH 5-6.5 and 0.25 - 4.30 h. While another study, used another natural products such as; Nisreen et al. (31) used charred pistachio shell as very high in adsorption of heavy metals (Cd, Pb) for removal 98% from aqueous solutions, Rashid et al. (32) used *Eichhorniacrassipes* (water hyacinth) root and shoot powder, and ash case with chitosan were used as a removal (98%) for Pb, (98.2%) Cu, , and (96.8%) Cd within 24 h. from waste water.

CONCLUSIONS

To summarize, the physicochemical properties of the waste water sample showed high levels of pollution that are mean many harmful materials are released to the environment by industrial factories. The usage of plant wastes and plant parts as natural coagulants is to treat waste water. The removal process by the coagulant in the first stage was a high percentage with 8 gm of gum-silica composite. Thus, it was known from the turbidity, conductivity measure- ments, and the total dissolved salts of the two solutions are followed by the absorption of heavy elements Nickel, Zinc Lead, and Copper with the same coagulant. This mixture can be used to remove heavy water pollutants during treatment. Chemical modification for plant wastes and parts is necessary to develop and increase the ability of natural coagulants for waste water treatment.

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