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Original article

Using Melon and Watermelon Peels for the Removal of Some Heavy Metals from Aqueous Solutions

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Abstract
In this study, the activated carbon of melon and watermelon peels was used as a solid adsorbent for the removal of ferric and copper ions from aqueous solutions through the batch equilibrium technique. The influence of contact time and initial concentration of metal ions on the adsorbed amount of metal ions was investigated. The highest adsorption capacity was found to be 3.32 and 74 mg of metal ions per gram of adsorbent at an initial concentration of 3.35 mg/L and 100 mg/L of ferric and copper ions, respectively. The melon peels were used and found to be 3.26 mg of metal ions per gram of adsorbent at an initial concentration of 3.35 mg/L, when watermelon was used, and this would be higher with a higher initial concentration. Ferric ion was removed by 86.83 - 99.55%, and copper ion by 18-88% along the whole range of initial concentrations when melon was used, and 77.55%-98.86% for ferric when watermelon was used. Isotherm studies showed that the data best fit the Langmuir isotherm model.

Key words. Removal, Heavy Metals, Melon, Watermelon, Peels.

Introduction

Heavy metal pollution in aquatic systems has become a serious threat and has a great potential to cause environmentally derived cancer because these metals are non-biodegradable and therefore persistent. Metals are mobilized and carried into the food web as a result of leaching from waste dumps, polluted soils, and water. The contamination of water with substances that hurt human beings, animals, and plants is called water pollution. Water pollution is a worldwide problem; its control has become increasingly important in recent years [1]. Conventional methods for removing metals from aqueous solutions include chemical precipitation, chemical oxidation and reduction, ion exchange filtration, electrochemical treatment, reverse osmosis, membrane technologies, and evaporation. The major disadvantage of conventional treatment technologies is the production of toxic chemical sludge, whose disposal/treatment becomes a costly affair and is not eco-friendly. Therefore, removal of toxic heavy metals to an environmentally safe level in a cost-effective and environmentally friendly manner is of great importance [2,3].

In recent years, considerable attention has been devoted to the study of the removal of heavy metal ions from solution by adsorption using agricultural materials. Natural materials that are available in large quantities or certain wastes from agricultural operations may have the potential to be used as low-cost adsorbents, and they represent unused resources, which are widely available and are environmentally friendly. Some investigations on the removal of heavy metal ions with agricultural by-products have been previously reported [4]. Biosorption is defined as the ability of biological materials to accommodate heavy metals from wastewaters through the metabolically mediated physico-chemical pathways of uptake. Algae, fungi, bacteria, parts of some higher plants, and yeasts have proven to be potential metal biosorbents. When the heavy metal concentration exceeds the tolerance level, it will show harmful effects on human physiological and other biological systems [5].

Many different approaches have been studied and developed for the effective removal of heavy metals using biosorbents such as peat [6], fly ash [7,8], microbial biomass and other agricultural by-productssuch as sugarcane bagasse [9], soya bean hulls [10], walnut hulls [11], cotton seed hulls and corn cobs [12]. It has also been observed that these biosorbents require further modifications to increase the number of active binding sites and make them readily available for sorption. Huang and Huang have stated that pre-treatment of the biomass removes the surface impurities on the biosorbents and exposes the available binding sites for metal sorption [13,14]. Copper is considered one of the most toxic metals and poses a potential threat to human health and the environment, even at low concentrations. It has been well reported that the accumulation of copper in the human body causes brain, skin, pancreas, and heart diseases [15]. The permissible limit of copper is 2.5 mg/L in water. Wastewater from various industries, such as electroplating, plastic, metal finishing, pigments, and mining, contains copper. To alleviate the problem of water pollution by copper, various methods have been used to remove copper from wastewater, such as chemical precipitation, coagulation, flotation, adsorption, ion exchange, reverse osmosis, and electrodialysis.

The production of the sludge in the precipitation method poses challenges in handling, treating, and land-filling of the solid sludge. Ion exchange usually requires a high-capital investment for the equipment as well as high operational costs. Electrolysis allows the removal of metal ions with the advantage that there is no need for additional chemicals, and also there is no sludge generation. However, it is inefficient at a low metal concentration. Membrane processes such as reverse osmosis and electrodialysis tend to suffer from the

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instability of the membranes in salty or acidic conditions and fouling by inorganic and organic substances present in wastewater [16].

Adsorption is highly effective, economical, and promising, and is widely applied for scavenging the metal-bearing wastewaters. Different conventional and non-conventional adsorbents have been used for the removal of heavy metals [17]. Hen eggshell is one of the major byproducts of the food industry. The by-product eggshell weighs approximately 10% of the total mass (60 g) of the hen egg, representing a significant waste from the egg-derived products processor because it was traditionally useless and commonly disposed of in landfills without any pretreatment [18,19].

Fruit peel waste (FPW) has the highest potential as an active adsorbent for wastewater treatment. One of the conventional treatments of FPW is through biochar production. There are a few routes to produce biochar, such as pyrolysis, slow pyrolysis, fast pyrolysis, gasification, and hydrothermal carbonization. Biochar produced at high temperatures (800°C) is efficient for the removal of copper (Cu) and lead (Pb) from wastewater. Pyrolysis is the most common method to produce biochar. It is a process where organic compounds (solid or liquid) undergo thermal decomposition in the absence of oxygen or with little oxygen. The thermal treatment breaks down the larger organic molecules into smaller ones. The production of biochar is influenced by various parameters such as temperature, heating rate, steam charge, and time [20,21]. This research aimed to investigate the use of melon and watermelon in the removal of metal ions from aqueous solutions. melon and watermelon a low-cost adsorbent, which a waste material. The study involved the examination of experimental conditions such as time and adsorbent loading on the removal of Cu (II), Fe (III), and from aqueous solutions. The Langmuir adsorption isotherms were used to investigate the adsorption process. A kinetic study was also carried out to evaluate the order of adsorption. The present study was undertaken to evaluate the efficiency of melon and watermelon fruit powder for the removal of ferric and copper ions in aqueous solution, in addition to estimating the optimum removal conditions and the suitable adsorption isotherms with their related constants.

Materials and Methods

Dried melon and watermelon were milled and ground until they passed through an 80-mesh sieve. The powdered melon and watermelon fruits were washed repeatedly with distilled water to remove dust and soluble impurities. All chemicals used are of high purity, commercially available Analar grades. A stock solution of 1000 mg/L of ferrous sulfate ($FeSO_4.7H_2O$), and copper sulfate ($CuSO_4.5H_2O$) was prepared using deionized water. From that, 3.35 mg/L of ferrous ion solutions and 100 mg/L of copper ion solutions were prepared in buffer solution. The adsorption was carried out in a batch process at (32 ±1 °C) in the presence of 100 mg of adsorbent (peel fruit powder) under constant shaking with an orbital shaker. The effect of contact time and initial concentration of metal ions was also examined by choosing different time intervals of shaking the melon and watermelon fruit powder with metal ion solutions. The sealed conical flasks were shaken at constant temperature for 15, 30, 45, 60, 90, and 120 minutes for ferrous and copper ions, respectively, where equilibrium was attained. The resultant metal ion solution concentration was determined by the spectrophotometer. The amount of metal adsorbed at equilibrium, qe mg/g, was determined using the following equation:

$$(q_e) = \frac{(ci - c_e)}{m} \times v \text{ ....} \mathbf{(1)}$$

Where: V = Volume of equilibrated solution. M =mass of adsorbent. Ci = initial concentration of metal ion. Ce = equilibrium concentration of metal ion. Percent removal (R%) was also evaluated from the following equation

$$\textbf{Removal \%} = (c_i - c_e) \ / \ (c_i) \times 100 \text{................(2)}$$

The adsorption process was carried out according to the methods described by different studies [22 -29].

Results and Discussion

The results of the effect of adsorbent doses, time, and their removal values of Ferric were shown in Table 1.

Table 1. Effect of adsorbent (Melon) doses on the concentration of Fe^{+3} ions in different time

Wt of adsorbent	C_i	TIME 15 min Ce	qe	R%	TIME 30 min Ce	Qe	R%	TIME 1:30 Ce	qe	R%	TIME 2:00 Ce	qe	R%
100	3.35	0.3856	2.96	88.84	0.114	3.23	96.59	0.0368	3.28	98.90	0.028	3.32	99.16
200	3.35	0.244	1.55	92.71	0.187	1.58	94.41	0.0385	1.65	98.85	0.015	1.66	99.55
300	3.35	0.149	1.06	95.55	0.122	1.076	96.35	0.132	1.072	96.05	0.16	1.06	95.22
400	3.35	0.456	0.723	86.38	0.358	0.748	89.31	0.062	0.822	98.14	0.1	0.812	97.01
500	3.35	0.211	0.627	93.70	0.251	0.619	92.50	0.145	0.641	95.67	0.045	0.661	98.65

The essential characteristics of the Langmuir adsorption isotherm can be expressed in terms of a dimensionless constant, the separation factor or equilibrium parameter 'RL', which is defined by,

$$\textbf{R L} = (1) \ / \ 1 + (\text{SLOP} * \text{Ci}) \text{ ....(3) [27,28].}$$

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RL >1 Unfavorable, RL=1 Linear, 0< RL <1 Favorable, RL=0 Irreversible. The results of Langmuir isotherms of Ferric ions by using melon peels as adsorbent are given in Table 2 and Figure 1.

Table 2. The values of Langmuir adsorption isotherms for Fe^{+3} ions by using 100mg of adsorbent (melon)

Time	Ce	qe	Ce /qe
15	0.3856	2.96	0.130
30	0.114	3.23	0.044
90	0.0368	3.28	0.0112
120	0.028	3.32	0.0084

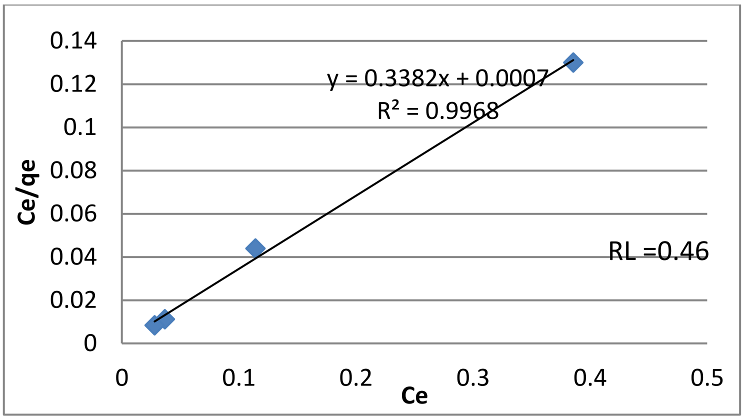

Figure 1. Langmuir adsorption isotherms for Fe^{+3} by using 100mg of adsorbent (melon).

Table 3. The values of Langmuir adsorption isotherms for Fe^{+3} ions using 200mg of adsorbent(melon)

Time	Ce	qe	Ce /qe
15	0.224	1.55	0.144
30	0.187	1.58	0.118
90	0.0385	1.65	0.023
120	0.015	1.66	0.0090

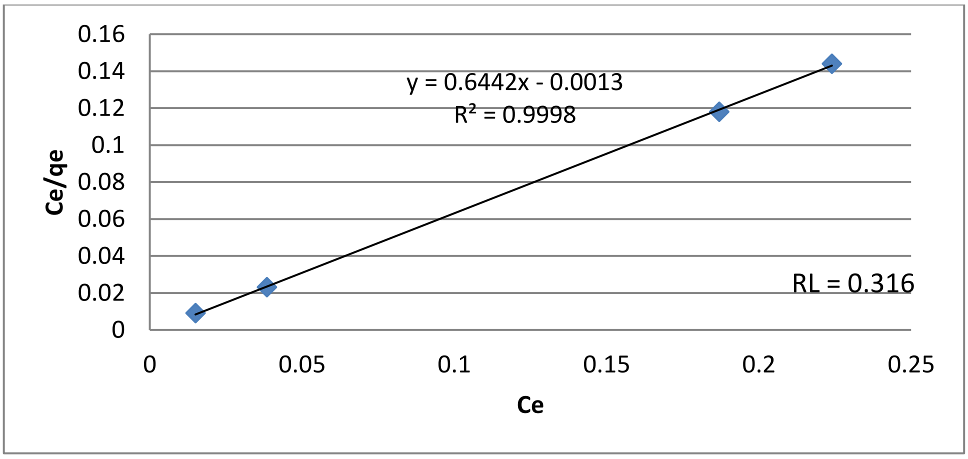

Figure 2. Langmuir adsorption on isotherms for Fe^{+3} by using 200mg of adsorbent (melon)

Table 4. The values of Langmuir adsorption isotherms for Fe^{+3} ions by using 300mg of adsorbent (melon)

Time	Ce	qe	Ce /qe
15	0.149	1.06	0.140
30	0.122	1.076	0.113
90	0.132	1.072	0.123
120	0.16	1.06	0.150

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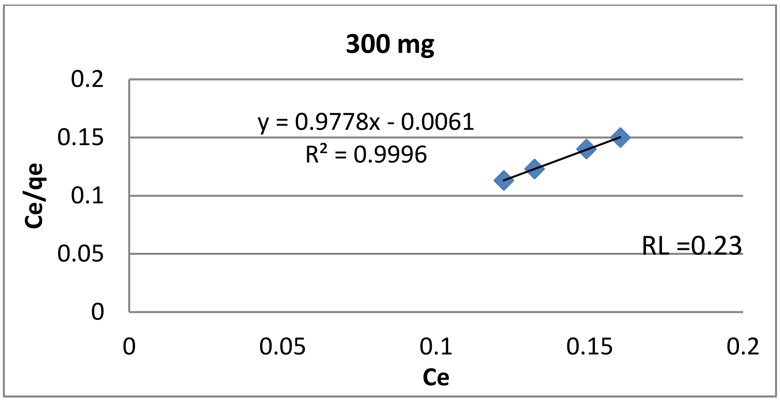

Figure 3. Langmuir adsorption on isotherms for Fe $^{+3}$ by using 300mg of adsorbent (melon)

Table 5. The values of Langmuir adsorption isotherms for Fe^{+3} ions by using 400mg of adsorbent (melon)

Time	Ce	qe	Ce /qe
15	0.456	0.723	0.630
30	0.358	0.748	0.478
90	0.062	0.822	0.0075
120	0.1	0.812	0.123

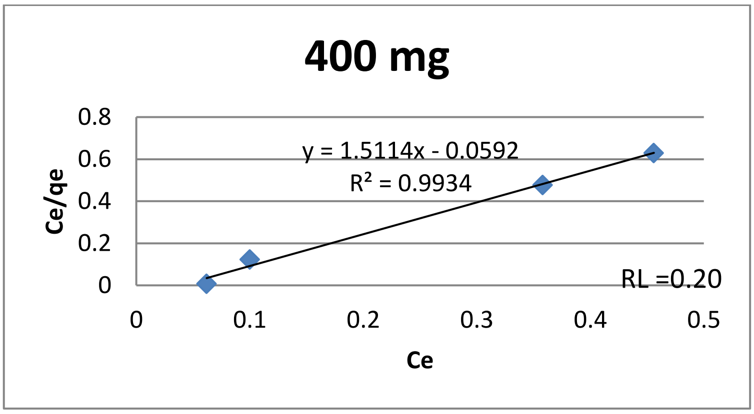

Figure 4. Langmuir adsorption on isotherms for Fe^{+3} by using 400 mg of adsorbent(melon)

Table 6. The values of Langmuir adsorption isotherms for Fe^{+3} ions by using 500mg of adsorbent (melon)

Time	Ce	qe	Ce /qe
15	0.211	0.627	0.336
30	0.251	0.619	0.405
90	0.145	0.641	0.226
120	0.045	0.661	0.068

Table 7. Effect of adsorbent doses (Melon) on the concentration of Cu^{+2} ions in different time

Wt of adsorbent	C_i	Time 15 min Ce	qe	R%	Time 45min Ce	qe	R%	2:00 Time Ce	qe	R%
100	100	47	53	53	34	66	66	26	74	74
200	100	72	28	28	50	25	50	12	44	88
300	100	77	7.66	23	53	15.66	47	33	22.33	67
400	100	64	9	36	69	7.75	31	19	20.25	81
500	100	82	3.6	18	51	9.8	49	47	10.6	53

Table 8. The values of Langmuir adsorption isotherms for Cu $^{+2}$ ions by using 100 mg of adsorbent (melon)

Time	C_e	qe	Ce /qe
15	47	53	0.886
45	34	66	0.515
120	26	74	0.351

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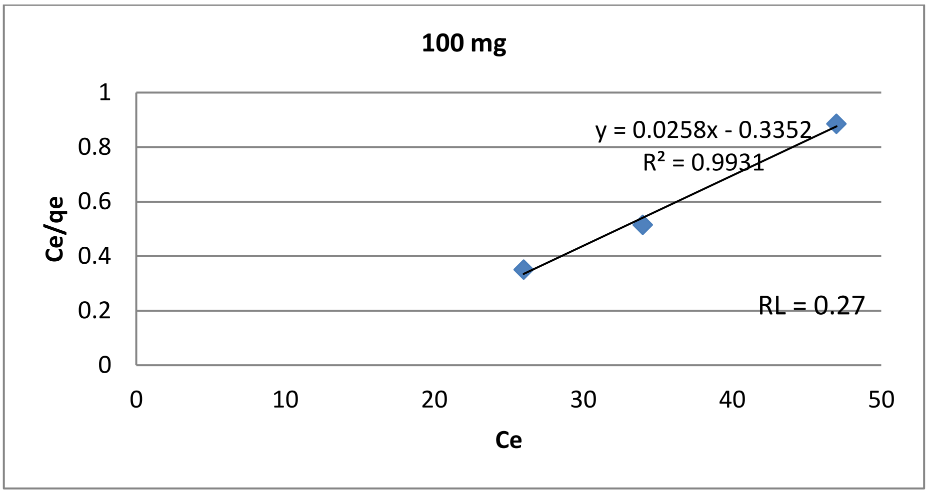

Figure 5. Langmuir adsorption on isotherms for Cu $^{+2}$ by using 100 mg of adsorbent (melon)

Table 9. The values of Langmuir adsorption isotherms for Cu $^{+2}$ ions by using 200 mg of adsorbent (melon)

Time	Ce	qe	Ce /qe
15	72	28	2.57
45	50	25	2
120	12	44	0.27

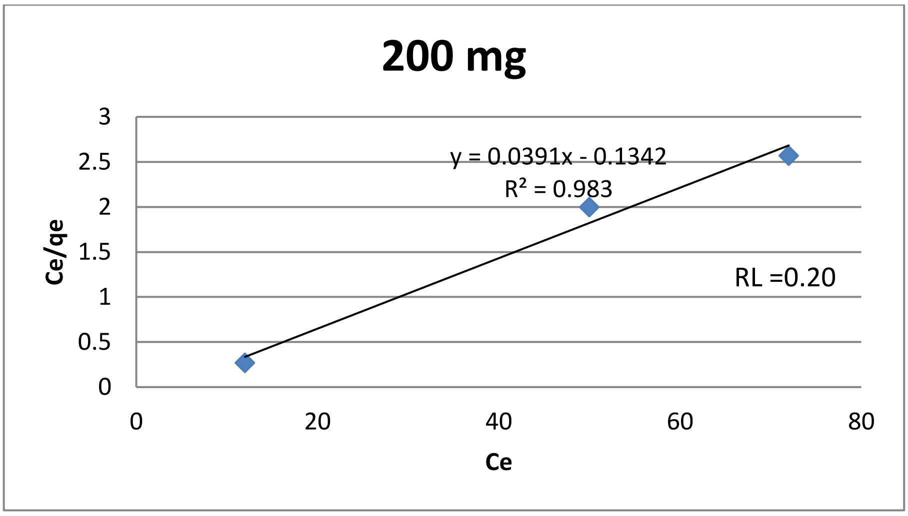

Figure 6. Langmuir adsorption on isotherms forCu^{+2}by using200 mg of adsorbent(melon)

Table 10. The values of Langmuir adsorption isotherms for Cu $^{+2}$ ions by using 300 mg of adsorbent (melon)

Time	**Ce**	**qe**	**Ce /qe**
15	77	7.66	10.05
45	53	15.66	3.38
120	33	22.33	1.47

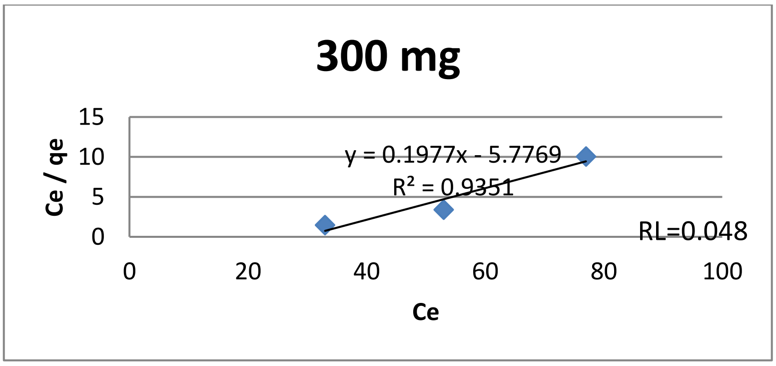

Figure 7. Langmuir adsorption on isotherms for Cu^{+2} by using 300 mg of adsorbent(melon)

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Table 11. The values of Langmuir adsorption isotherms for Cu $^{+2}$ ions by using 400 mg of adsorbent (melon)

Time	Ce	qe	Ce /qe
15	64	9	7.11
45	69	7.75	8.90
120	19	20.25	0.93

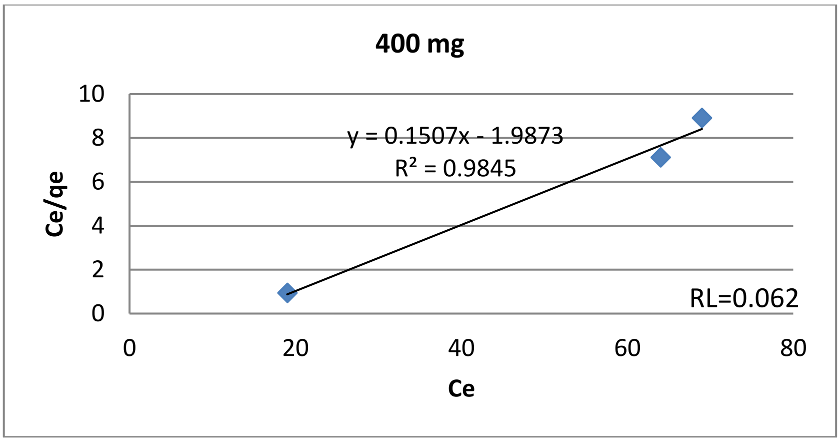

Figure 8. Langmuir adsorption on isotherms for Cu^{+2} by using 400 mg of adsorbent(melon)

Table 12. The values of Langmuir adsorption isotherms for Cu^{+2} ions by using 500 mg of adsorbent (melon)

Time	Ce	qe	Ce /qe
15	82	3.6	22.77
45	51	9.8	5.20
120	47	10.6	4.43

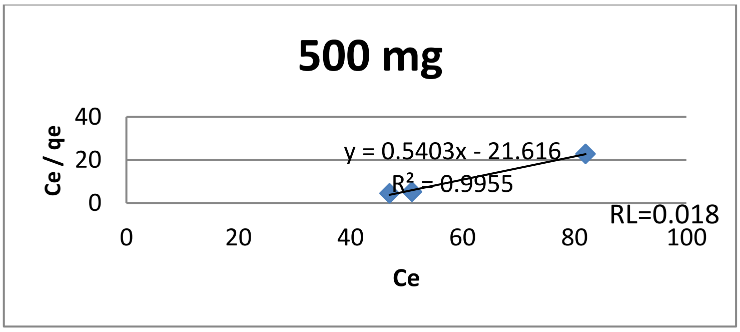

Figure 9. Langmuir adsorption on isotherms for Cu^{+2} by using 500 mg of adsorbent(melon)

Table 13. Effect of adsorbent (watermelon) doses on the concentration of Fe $^{+3}$ ions in different time

Wt	Ci	Time 15 Ce	qe	R%	Time 30 Ce	qe	R%	Time 45 Ce	qe	R%	Time 60 Ce	qe	R%	Time 01:30 Ce	qe	R%
0.1	3.35	0.459	2.89	86.29	0.13	3.22	96.11	0.078	3.27	97.67	0.085	3.26	97.46	0.083	3.26	97.52
0.2	3.35	0.414	1.468	87.64	0.32	1.55	90.44	0.217	1.56	93.52	0.179	1.58	94.65	0.038	1.65	98.86
0.3	3.35	0.6	0.916	82.08	0.112	1.07	96.65	0.352	0.999	89.49	0.09	1.086	97.31	0.159	1.06	91.17
0.4	3.35	0.725	0.656	77.55	0.348	0.75	89.61	0.36	0.747	89.25	0.187	0.790	94.41	0.135	0.803	95.97
0.5	3.35	0.467	0.576	86.06	0.29	0.61	91.34	0.273	0.615	91.85	0.125	0.645	96.26	0.097	0.650	97.10

Table 14. The values of Langmuir adsorption isotherms for Fe $^{+3}$ ions by using 100 mg of adsorbent (watermelon)

Time	Ce	qe	Ce /qe
15	0.459	2.89	0.158
30	0.13	3.22	0.040
45	0.078	3.27	0.023
60	0.085	3.26	0.026
90	0.083	3.26	0.025

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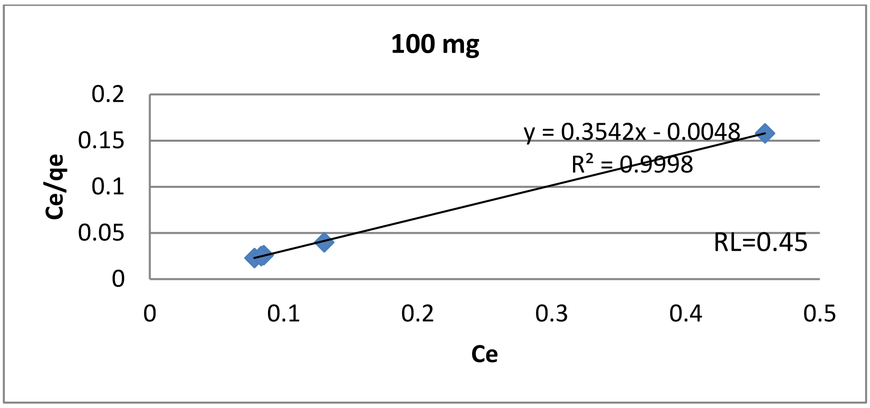

Figure10. Langmuir adsorption on isotherms for Fe $^{+3}$ by using 100 mg of adsorbent(watermelon)

Table 15. The values of Langmuir adsorption isotherms for Fe $^{+3}$ ions by using 200 mg of adsorbent (watermelon)

Time	Ce	qe	Ce /qe
15	0.414	1.46	0.28
30	0.32	1.55	0.20
45	0.217	1.56	0.13
60	0.179	1.58	0.11
90	0.038	1.65	0.023

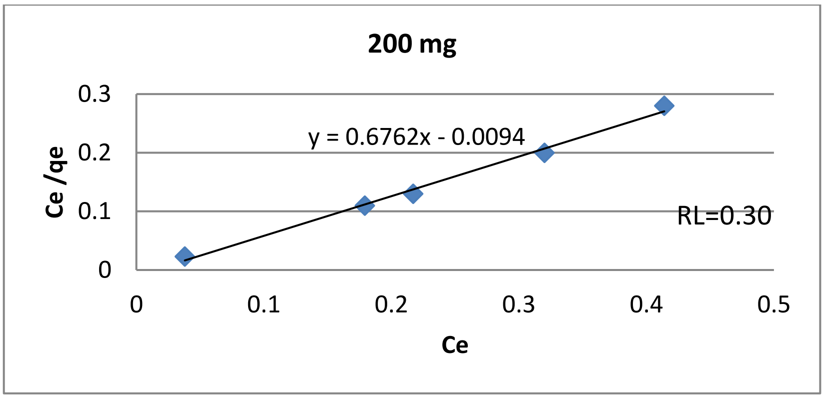

Figure 11. Langmuir adsorption on isotherms for Fe^{+3} by using 200 mg of adsorbent(watermelon)

Table 16. The values of Langmuir adsorption isotherms for Fe $^{+3}$ ions by using 300 mg of adsorbent (watermelon)

Time	Ce	qe	Ce /qe
15	0.6	0.916	0.65
30	0.112	1.07	0.10
45	0.352	0.99	0.35
60	0.09	1.086	0.08
90	0.159	1.06	0.15

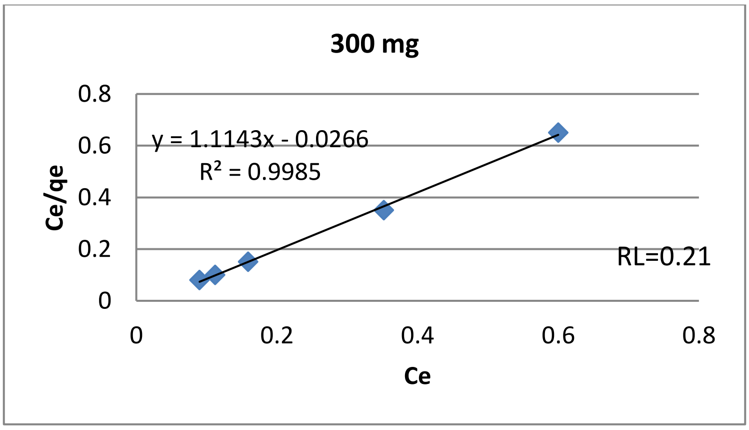

Figure 12. Langmuir adsorption isotherms for Fe^{+3} by using 300 mg of adsorbent (watermelon)

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Table 17. The values of Langmuir adsorption isotherms for Fe^{+3} ions by using 400 mg of adsorbent(watermelon)

Time	Ce	qe	Ce /qe
15	0.725	0.656	1.10
30	0.348	0.75	0.46
45	0.36	0.747	0.48
60	0.187	0.790	0.23
90	0.135	0.803	0.16

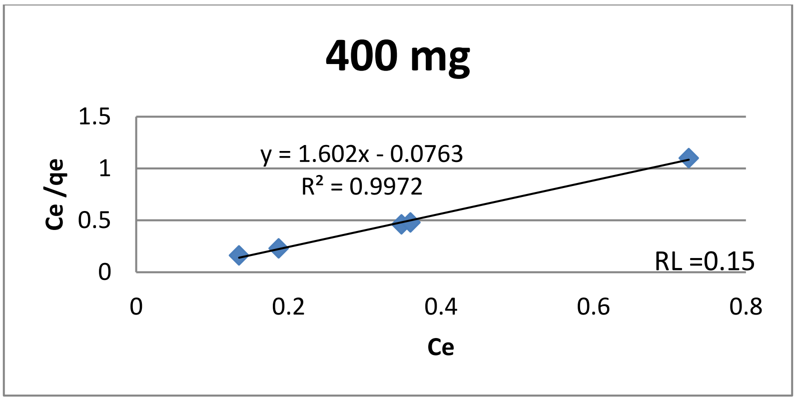

Figure 13. Langmuir adsorption on isotherms for Fe $^{+3}$ by using 400 mg of adsorbent(watermelon)

Table 18. The values of Langmuir adsorption isotherms for Fe $^{+3}$ ions by using 500 mg of adsorbent (watermelon)

Time	**Ce**	**qe**	**Ce /qe**
15	0.467	0.576	0.81
30	0.29	0.61	0.47
45	0.273	0.615	0.44
60	0.125	0.645	0.19
90	0.097	0.650	0.147

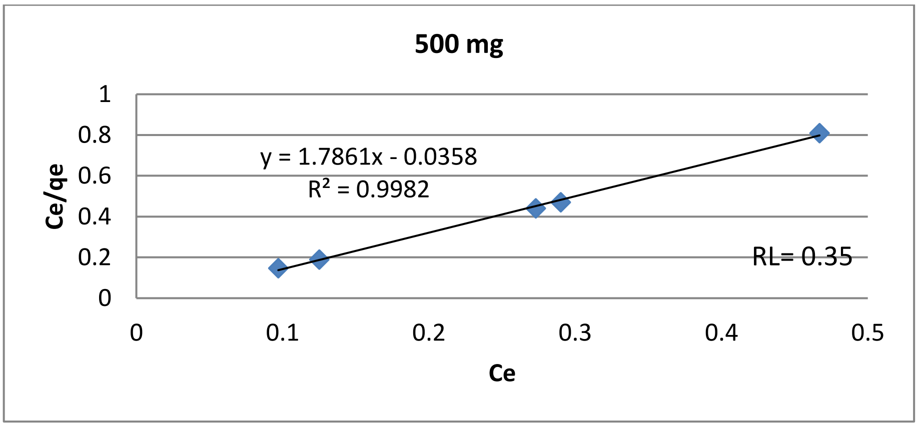

Figure 14. Langmuir adsorption isotherms for Fe $^{+3}$ by using 500 mg of adsorbent

Discussion

The carboxylate group at the active site can adsorb Fe^{+3}, Cu^{+2} ions very well. Therefore, the melon water melon fruit powder adsorbents had more metal-binding active sites and more negative surfaces, which enhanced the retention of Fe^{+3}, Cu^{+2} onto the surface and the percentage of Fe^{+3}, Cu^{+} removal. However, the adsorption mechanism of metal ions depends on various factors, such as the pH of the solution and binding characteristics. Usually, metal ion adsorption is accomplished through ion exchange, complex formation, electrostatic interaction, and precipitation [29]. The effects of contact time on the amount of metal ions on the adsorbent surface are Equilibrium have established at 15,30, 45, 60,90, and 120 min for ferric ions and copper ions. Once again, there is a big change in the amount of metal ions adsorbed with time, which indicates an ion exchange mechanism. Our findings are in harmony with those studies, which stated that the removal of pollutants from aqueous solutions depended on the values of Langmuir and Freundlich isotherms. The adsorption mechanism is attributed to the porosity size of the activated carbon, as well as the type of activation, mainly affecting the removal of compounds by activated carbon, including temperature of activation, PH values of activated carbons, presence of surface minerals, and others. The advantage of

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this study is that it used low-cost material (residual sea grasses) and did not add other materials during the activation.

Conclusion

From the experimental data of adsorption of ferric and copper ions on melon water melon fruit powder surface, the following points can be concluded. Melon and watermelon powder, an agricultural waste, could be used as a potential adsorbent for the removal of ferric and copper ions from polluted solutions. The adsorption process for both metals is the same because the adsorption of Fe(III) and Cu(II) ions is well described by the Langmuir isotherm. The adsorption of metal ions on melon and watermelon powder was dependent on the time. The percent removal of ferric and copper ions by the new adsorbent is fairly high. The amount of metal ions adsorbed increased with an increase dose of adsorbent factor.

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الملخص

في هذه الدراسة، استُخدم الكربون المنشَّط من قشور البطيخ والشمام كمادة ماصة صلبة لإزالة أيونات الحديديك والنحاس من المحاليل المائية باستخدام تقنية توازن الدفعات. ودُرِسَ تأثير زمن التلامس والتركيز الابتدائي للأيونات المعدنية على الكمية الممتصة منها. وُجِد أن أعلى سعة امتزاز بلغت 3.32 و74 ملغ من أيونات المعادن لكل غرام من المادة الماصة عند تركيز ابتدائي قدره 3.35 ملغ/لتر و100 ملغ/لتر من أيونات الحديديك والنحاس، على التوالي. أما قشور البطيخ، فقد وُجِد أنها تحتوي على 3.26 ملغ من أيونات المعادن لكل غرام من المادة الماصة عند تركيز ابتدائي قدره 3.35 ملغ/لتر، عند استخدام البطيخ، وتكون هذه القيمة أعلى مع التركيز الابتدائي الأعلى. أُزيل أيون الحديديك بنسبة تراوحت بين 86.83% و99.55%، وأيون النحاس بنسبة تراوحت بين 18% و88% على امتداد نطاق التركيزات الأولية عند استخدام البطيخ، ونسبة تراوحت بين 77.55% و98.86% للحديديك عند استخدام البطيخ. أظهرت دراسات تساوي درجة الحرارة أن البيانات تُطابق نموذج تساوي درجة الحرارة لانغموير على النحو الأمثل.

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