EFFICIENCY OF LOW COST ADSORBENTS FOR REMOVAL LEAD, COPPER AND CHROMIUM FROM WASTEWATER

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ABSTRACT

The adsorption efficiency of water hyacinth, eggshell powder and kyanite for the removal of lead, copper and chromium were studied. Kyanite as a commercial mineral has been utilized as an adsorbent, eggshell powder and water hyacinth which have many functional groups capable of binding to the metal cations. The research is a batch scale experiment using different amounts of adsorbents in solution with five different concentrations (20, 40, 60, 80, and 100 mg/L) of each metal and in mixed combination.

Two main things comes out by the above study, first, removal adsorption depends on the amount of adsorbent.The adsorption efficiency of Cu was increased from 80.5% to 95.2% ; 75.3% to 91.1% and 90.2% to 100% for Cu and 78.5% to 91.6% ; 72.9% to 90.1% and 88.7% to 97.6% for Cr. Also, about 90.2% to 97.2% , 87.5% to 95.1% and 92.6% to 100% Pb removal achieved by using 2 to 5 g adsorbent for solution having concentration of 20 and 40 mg/L of Pb for each water hyacinth, eggshell powder and kyanite in the same solution (20 mg/L). Second, it was also found for the mixture of these metals a decrease about 1.78% , 8.02% and 2.16% for lead, whereas Cu and Cr adsorption decreased about 2.74 % , 6.25% and 2.44% ; 12.11%, 7.41 % and 4.63 % respectively, to mixed metal solution, which clearly indicated that the presence of more metals in the solution will decrease the adsorption efficiency and it also found increasing the adsorbent dose, the removal efficiency of adsorbents increases, while adsorption efficiency decreases with increasing the various concentration.

Keywords: Water hyacinth, Eggshell powder, Kyanite, adsorption, lead, copper, chromium, batch experiment.

INTRODUSTION

The awareness of increasing water pollution implies studies concerning water treatment. Removal of heavy metals from industrial wastewater is of primary importance. The use of natural materials for heavy metals removal is becoming a concern in all countries. Natural materials that are available in large quantities or certain waste from agricultural operations may have potential to be used as low cost adsorbents, as they represent unused resources, widely available and are environmentally friendly *(Carvalho et al., 2003).*

Heavy metals are dangerous environmental pollutants due to their toxicity and strong tendency to concentrate in environment and in food chains*(Bulut and Baysal,2006 andSari et al., 2007)*. The source of environmental pollution with heavy metals is mainly industry, i.e. metallurgical, electroplating, metal finishing industries, tanneries, chemical manufacturing, mine drainage and battery manufacturing *(Matheickal and Yu, 1997)*.Considerable research had been carried out over the last decade on the protection against plant and animal life degradation. Several big cities contribute to increase this problem as they are sources of industrial effluents. In order to reduce the environmental pollution, a number of studies have been considered to minimize the problem caused by the commonly employed treatment of metal bearing effluents (*Valdman and Leite, 2000*and *Carvalho et al, 2003).*

Removal of metals from wastewater achieved principally by the application of several processes such as adsorption*(Fahim et al., 2006)*, sedimentation*(Song et al., 2004)*, electrochemical processes*(Tiravanti et al., 1997)*, ion exchange, cementation*(Filibeli et al., 2000)*, coagulation/flocculation*(Song et al., 2000)*, filtration and membrane processes, chemical precipitation and solvent extraction. Adsorption is the one of the important procedure for the removal of heavy metals from the environment because of strong affinity and high loading capacity. As copper, zinc and lead and its compounds used in brass industries, the continued intake of copper and lead by human leads to severe diseases like mucosal irritation, depression and most dangerous lung cancer. The adsorption efficiency of eggshell powder for the removal of Cu and Pb was studied. The research is a batch scale experiment using different amounts of adsorbents in solution with five different concentrations of both metals and in mixed combination. About 92% to 100% Cu removal achieved by using 0.5 to 1.5 g adsorbent for solution having concentration of 5 and 10 mg/l of Cu *(Agarwal. 2013)*.

In nature chromium exists in two most stable oxidation states, i.e., trivalent and hexavalent forms in aqueous system. Although, at trace level, the trivalent form is considered as an essential nutrient *(Alloway, 1995 and Rojas et al, 2005)*whereas hexavalent form of chromium is toxic, carcinogenic and mutagenic in nature *(Norseth, 1991; USDHHS, 1991; Cieslak-Golonka, 1995; Myers et al, 2000; Zhitkovich et al, 2002 and Dupont and Guillon, 2003)*. *Kowalski (1994) r*evealed that the hexavalent form is about 500 times more toxic than trivalent form. Furthermore, Cr(VI) is highly mobile in soil and aquatic system, and also is a strong oxidant capable of being adsorbed by skin *(Singh and Singh 2002).* Considering its toxicity and carcinogenic nature, the maximum levels permitted for trivalent chromium in wastewater is 5 mg/L and for hexavalent chromium as 0.05 mg/L. The adsorption process is being widely used by various researchers for the removal of heavy metals from waste streams and activated carbon has been frequently used as an adsorbent. Despite its extensive use in the water and wastewater treatment industries, active carbon remains an expensive material. In recent years, the need for safe and economical methods for the elimination of heavy metals from contaminated water has necessitated research interest towards the production of low cost alternatives to commercially available activated carbon. Therefore there is an urgent need that all possible sources of agro-based inexpensive adsorbents should be explored and their feasibility for the removal of heavy metals should be studied in detail *(Renge et al., 2012).*The removal of heavy metal ions using low-cost abundantly available adsorbent; agricultural wastes such as tea waste and coffee *(Orhan and Buyukgungor, 1993)*, hazelnut straw *(Cimino et al., 2000),* peanut hull *(Johnson et al., 2002),* sawdusts, pinus bark *(Vazquez et al., 1994)* and different bark samples, coconut husk (Babarinde, 2002), ponkan mandarin peels *(Pavan et al., 2006),* modified cellulosic materials *(Acemioglu and Alma, 2001)*, corn cobs *(Hawrhorne-Costs et al., 1995),* apple wastes *(Maranon and Sastre, 1991)*, wool fibers *(Balkose and Baltacioglu, 1992)*, tea leaves*(Tee and Khan 1988),* banana and orange peels *(Annadurai et al., 2002),* sugarcane bagasse *(Khan et al., 2001),* papaya wood *(Saeed et al., 2005)*, maize leaf *(Baharinde et al., 2006)*, leaf powder *(Hanafiah et al., 2007),* grape stalk wastes *(Villaescusa et al., 2004)* and different agricultural by-products were used and investigated. *(Marshall and Champangne, 1995 and Pehlivan et al., 2006).*

Kyanite is a triclinic crystalline mineral like other aluminosilicates and closely approximates Al_2SiO_3. Kyanite, when calcined, is used in refractory products and it is the most important of the aluminosilicates. The metal needs to be removed from industrial effluents before discharge into the environment to minimize any impact on plant, animal and human beings. In the present study, adsorption potential of low cost adsorbent (water hyacinth, eggshell powder and kyanite) towards Pb , Cu and Cr have been examined.

MATERIALS AND METHODS

Adsorbents Processing

(A)Kyanite

Kyanite was ground and washed thoroughly with tap water to clean the adhering dust. It was dried in an oven at 100 - 105 ^{0}C for 24 hrs. After drying, the adsorbent used at 100 mesh size. The chemical composition of kyanite has been reported and is shown in Table(I).

Table (I):Chemical composition of Kyanite ; eggshell and water hyacinth

Kyanite		Eggshell		Water hyacinth	
Elements	Unit	Elements	Unit %	Elements	Unit %
Specific gravity	3.70	$CaCO_3$	96.48	Hemicelluloses	22-35
SiO_2	30.2 %	S	2.31	Cellulose	17.8 – 31
Al_2O_3	65.35 %	$MgCO_3$	0.40	Lignin	7 - 26.36
Fe_2O_3	3.19 %	P (calcium phosphate)	0.50	Magnesium	0.17
TiO_2	Trace	Al	-	K_2O	28.7
CaO	Trace	K	-	Na_2O	1.8
MgO	Trace	Sr	0.07	Cl	21.0

(B) Eggshell Powder

The eggshells used in the experiment were collected free of charge from different hotels located in Cairo city, Egypt. Eggshell is largely-crystalline calcium carbonate. The calcium comes partly from the hen's bones and when necessary the hen can mobilize 10% of her bone for the purpose. Eggshells were washed with tap water several times then air-dried and incubated in hot air oven at 40 ^{0}C for 30 minutes (because protein component in eggshell can denature at high temperature(> 40^{0}C). Consequently, eggshells were ground to a powder in a grinder, and sieved to obtain between 60-100 mesh size particles *(Arunlertaree et al., 2007).*The chemical composition of eggshell has been reported according to Arunlertaree et al.,(2007) and is shown in Table (I)

(C) Water hyacinth

The water hyacinth plant is considered one of the natural and cheap materials.Water hyacinth grown on the surface of soft water in Nile River. The plantsused in this trial were collected from clean area far from those which may be industrially polluted (El-Madey area), then washed with tap water and choppedby using a small clean cutter about 5 -10 cm lengths. The chopped plants placed already on plastic sheet to minimize contamination. The plants shuffled upside down and mixed well every day to dry in sun to become acceptable, then dried at 70^{0}C for 72 hrs. The dried plants were grinded and sieved to 100 meshsizes particles.The chemical composition of water hyacinth has been reported according to Gunnarsson and Petersen (2007) and is shown in Table (I)

Adsorbate Solution

Synthetic wastewater used in this study was prepared to match the real wastewater samples used in the adsorption studies.Individual and mixed metal ion solutions of chromium, copper and lead were prepared from Merck-analytical grade stock standards of concentration 1000 mg/L. The synthetic waste water solutions were then prepared by diluting the stock standard of each metal. The pH valueof wastewater was controlled at 5 by using 1M HCl and / or NaOH. The final concentration of metal ions in the wastewater were analyzed by Inductively Coupled Plasma (ICP-OES) Perkin-Elmer Optima 7300.

Batch Adsorption Studies

Individual and mixed solutions of Pb, Cu and Cr with different concentrations 20, 40, 60, 80, and 100 mg/L were prepared.The experiment was performed using three different amounts of adsorbents 2, 3, and 5 g in single solution. 2g adsorbent was placed in a conical flask in which 200- ml of solution with known concentration of Pb was added andthe mixture was shaken in shaker. After 24 hours contact time and final concentration of metal ion was determined in filtrate by Inductively Coupled Plasma. All the experiments were carried out in triple and mean concentration was calculated by averaging them. The procedure was repeated by varying adsorbents doses and concentrations of Pb, Cr and Cu solution, both individual and mixed solution. Based on residual concentrations, the adsorption efficiencies of kyanite; water hyacinth and eggshell powder are calculated and summarized in Tables (1 to 6).The water hyacinth plant and Eggshell powder can be possible recycled by washes with 0.5 N NaOH solution then with distilled water. Excess alkali was neutralized with 0.1 N HCL solution and again washed with distilled water several times, to get rid of metals; this process will be useful for the removal of residual of metals from adsorbent after treatment to be used for several times *(Naganaik et al., 2002)*.

RESULTS AND DISCUSSION

The analysis indicates that the adsorption efficiencies of water hyacinth, eggshell powder and kyanite are high for Pb (Tables1, 2 and 3). Table (1) shows the adsorption efficiencies for various concentrations of Pb with 2g for various adsorbents. It is clear thatkyanite; water hyacinth and eggshell powderare good adsorbents for removal of Pb from wastewater. The adsorption rate is dependent on adsorbent amount and initial concentration of metal in synthetic solution. 90.2%, 87.5% and 92.6% removal of Pb from 20 mg/l solution were possible by applying 2g of various adsorbent whereas the similar amount of adsorbents were not enough to treat 100 mg/L Pb solution to above 70.7% , 67.4% and 72.4% . However, by increasing the amount of adsorbents to 5g it was possible to increase the efficiency of adsorption to about 80.6% , 73.9% and 86.8% for the same solution [100 mg/l Pb]. It shows that we would have better treatment by using different excess adsorbents.

Table(1): Different adsorptionmaterials efficiencies for lead ion at various concentrations and doses

NO.	Types of materials	Doses (g)	Initial concentration of Pb (mg/L)				
			20	40	60	80	100
			Adsorption Efficiency (%)				
1	Water hyacinth	2	90.2	88.3	80.7	75.2	70.7
2		3	92.0	90.0	87.6	80.3	76.0
3		5	97.2	97.2	90.1	85.4	80.6
4	Eggshell powder	2	87.5	87.2	78.4	71.1	67.4
5		3	90.7	88.2	80.4	75.6	70.1
6		5	95.1	95.2	87.7	78.4	73.9
7	Kyanite	2	92.6	90.2	88.0	80.0	72.4
8		3	98.7	97.6	92.4	88.7	79.9
9		5	100	100	95.4	95.1	86.8

Table(2):Different adsorption materials efficiencies for copper ion at various concentrations and doses

NO.	Types of materials	Doses (g)	Initial concentration of Cu (mg/L)				
			20	40	60	80	100
			Adsorption Efficiency (%)				
1	Water hyacinth	2	80.5	78	75	68.5	60.5
2		3	90.1	81.2	79.0	80.5	68.0
3		5	95.2	95.3	82.3	81.3	76.4

4	Eggshell powder	2	75.3	70.6	67.7	65.4	60.3
5		3	88.4	85.4	80.6	78.4	68.1
6		5	91.1	91.6	85.4	82.3	70.7
7	Kyanite	2	90.2	88.3	80.7	76.2	71.7
8		3	97.3	95.6	90.4	83.7	79.0
9		5	100	100	95.4	91.4	80.9

Table(3): Different adsorption materialsefficiencies for chromium ion at various concentrations anddoses

NO.	Types of materials	Doses (g)	Initial concentration of Cr (mg/L)				
			20	40	60	80	100
			Adsorption Efficiency (%)				
1	Water hyacinth	2	78.5	72.1	68.6	65.7	55.2
2		3	87.7	80.5	74.6	70.1	62.5
3		5	91.6	91.8	88.7	80.3	78.1
4	Eggshell powder	2	72.9	70.3	65.2	61.4	52.9
5		3	82.7	78.3	70.9	67.6	55.3
6		5	90.1	90.0	80.4	72.5	65.2
7	Kyanite	2	88.7	82.4	71.4	70.0	65.4
8		3	90.3	86.7	75.2	74.1	66.1
9		5	97.6	97.7	90.2	88.2	80.7

The adsorption efficiency and percentage adsorption of Pb, Cu and Cr at different doses of water hyacinth, eggshell powder and kyanite are shown in figs.(1, 2 and 3). The degree (%) of adsorption increases as the adsorbent dose is increased. It may be concluded that by increasing the adsorbent dose, the removal of adsorption increases, while adsorption efficiency decreases with increasing the concentration. The decrease in adsorption efficiency may be to the fact that some adsorption sites may remain unsaturated during the adsorption process whereas the number of sites available for adsorption site increases by increasing the adsorbent doses. These results are in agreement with those obtained by *Sharma and Forster(1994)*. The general removal percentage sequence is in the order:

Kyanite > Water hyacinth > Eggshell powder

Also, it can be concluded that maximum adsorptions of Pb, Cu and Cr occurs at 20 mg/L initial concentration and the minimum adsorptions are observed at 100 mg/L. It shows that we would have better treatment by using excess water hyacinth; eggshell powder and kyanite.Tables (2 and 3) indicates that adsorption efficiency is dependent on the type of metal too, as for Cu and Cr we have 80.5% and 78.5%, respectively removal in same condition [2g adsorbent in solutions 20 mg/L].Tables (4, 5 and 6) represent the results of adsorption experiments conducted on the mixture of metal solutions as mentioned before, the maximum and minimum removal efficiency in the first stage experiments with 2g of adsorbents with water hyacinth, eggshell powder and kyanite were 90.2%, 87.5% and 92.6% for Pb; 80.5%, 75.3% and 90.2% for Cu and 78.5%, 72.9% and 88.7% for Cr respectively. However, for the mixture of these metals decreases of 1.78 %, 8.02% and2.16% were found for Pb whereas Cu adsorption have decreased with about 2.74%,6.25% and 2.44%. Chromium adsorption decreased also with about12.11%, 7.41% and4.63% in the same order. The efficiency of Pb, Cu and Cr adsorption by various amounts of water hyacinth, eggshell powder and kyanite are shown in figs.(1 and2) for individual solutions and for mixed solutions of Pb, Cu and Cr. Also, effect of different adsorbents on adsorption of 20 mg/L for Pb, Cu and Cr are shown in Tables (4, 5 and 6)and (fig.3).

Table (4): Different adsorption materials efficiencies for studies heavy metals in mixed solution as affected by using 2 g absorbents

NO.	Types of materials	Metal Solution	Initial concentration (mg/L)				
			20	40	60	80	100
			Adsorption Efficiency (%)				
1	Water hyacinth	Pb	88.6	80.2	75.3	70.4	68.2
2		Cu	78.3	75.2	71.4	65.6	58.2
3		Cr	69.0	65.3	63.9	61.1	52.3
4	Eggshell powder	Pb	80.3	78.0	72.7	70.2	68.2
5		Cu	70.6	70.4	68.5	62.6	55.4
6		Cr	67.5	62.6	61.4	58.5	50.6
7	Kyanite	Pb	90.7	89.3	85.6	75.6	70.7
8		Cu	88.0	85.3	78.3	75.3	70.6
9		Cr	84.6	80.8	68.7	65.0	63.7

Table (5): Different adsorptionmaterials efficiencies for studies heavy metals in mixed solution as affected by using 3 g absorbents

NO.	Types of material	Metal Solution	Initial concentration (mg/L)				
			20	40	60	80	100
			Adsorption Efficiency (%)				
1	Water hyacinth	Pb	95.6	90.4	88.6	80.1	74.9
2		Cu	90.9	87.3	85.2	78.0	70.5
3		Cr	81.7	77.3	72.4	70.0	67.6
4	Eggshell powder	Pb	90.7	88.2	85.2	80.1	72.7
5		Cu	88.2	80.6	75.3	75.4	70.0
6		Cr	73.2	70.1	67.6	65.2	65.0
7	Kyanite	Pb	100.0	95.7	90.2	85.7	80.6
8		Cu	93.6	90.4	88.2	85.4	75.2
9		Cr	89.2	87.1	80.1	76.8	70.4

Table (6): Different adsorption materials efficiencies for studies heavy metals in mixed solution as affected by using 5 g absorbents

NO.	Types of materials	Metal Solution	Initial concentration (mg/L)				
			20	40	60	80	100

			Adsorption Efficiency (%)				
1	Water hyacinth	Pb	97.2	95.3	91.0	82.4	80.3
2		Cu	93.4	89.3	87.6	81.7	78.2
3		Cr	85.0	80.1	78.2	73.9	70.1
4	Eggshell powder	Pb	90.3	88.0	85.4	80.6	75.4
5		Cu	89.0	80.3	78.0	73.2	68.2
6		Cr	77.8	75.2	70.3	67.8	67.3
7	Kyanite	Pb	100.0	97.2	95.1	90.2	88.6
8		Cu	95.6	92.7	90.3	88.4	86.4
9		Cr	93.2	88.6	85.2	80.6	75.4

CONCLUTIONS

The above results showed that water hyacinth, eggshell powder and kyanite like the most other natural adsorbents can be used in the treatment process of heavy metals and the treatment efficiency may be as high as 100 % by precise choosing of adsorbent amount. It was also observed that the concentration of heavy metals has an important effect on the results of this treatment. Kyanite, water hyacinth and eggshell powder are waste materials and conveniently used for the treatment of industrial wastewater containing Pb, Cu and Cr metals. The most important finding of the study showed that in the mixture of metal ions the % adsorption is decreased than the individual metal. It clearly indicated that heavy metals need to be removed from the wastewater before the discharge into the rivers. The study also showed that the presence of one more metal will decrease the adsorption efficiency of adsorbents.

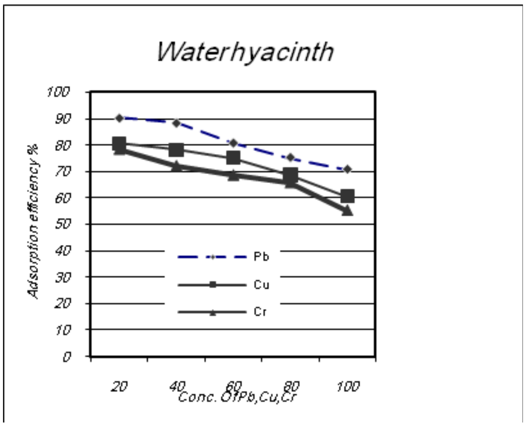


Fig.(1) : Adsorption of Lead, Copper and Chromium in mixed solution by using 2 g

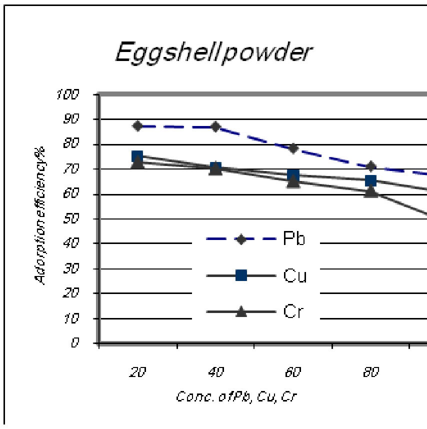


Fig.(2) : Adsorption percentage of Le
Copper and Chromium by using 2 g

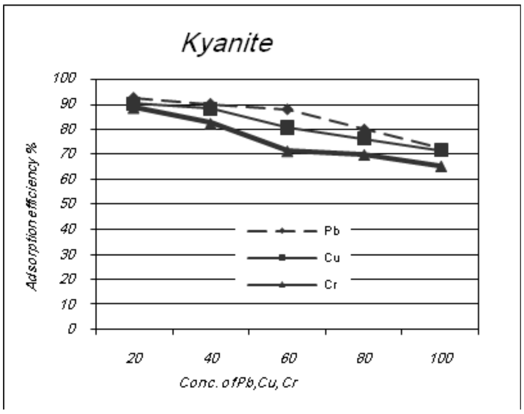


Fig. (3) : Adsorption percentage of Lead, Copper and Chromium by using Kyanite

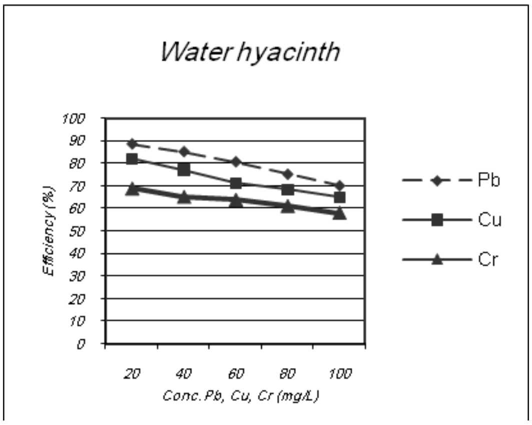


Fig. (4) : Adsorption percentage of Lead Copper and Chromium in mixed solution by Using 2 g

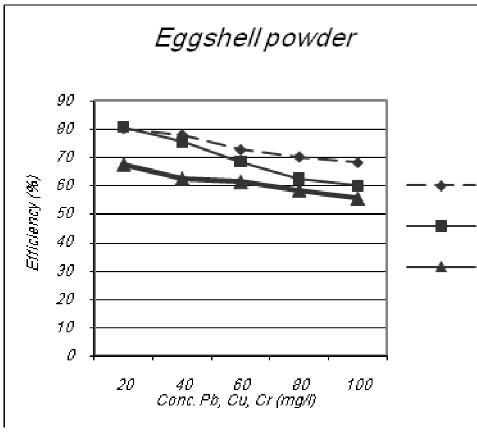


Fig. (5) : Adsorption percentage of Le
Copper and Chromium in mixed sol
by using 2 g

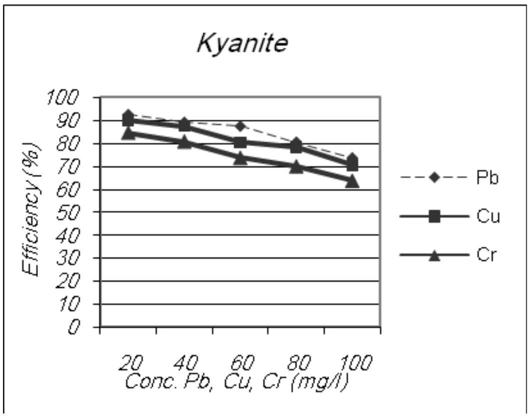
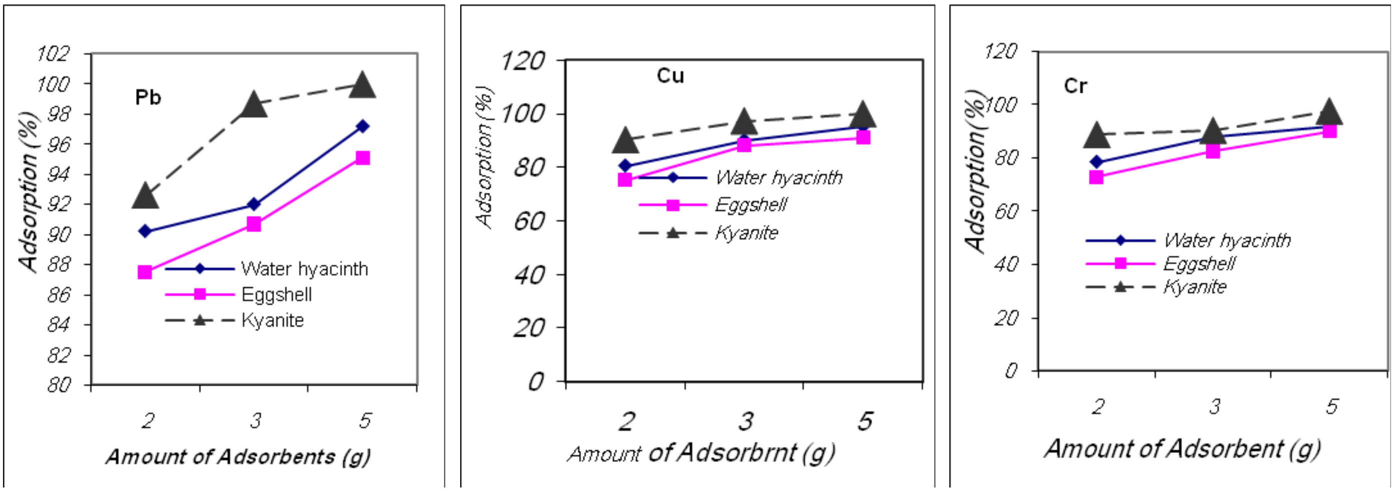


Fig. (6) : Adsorption percentage of Lead Copper and Chromium in mixed solution by using 2 g



Figs. (7, 8 and 9): Effect of adsorbent doses on adsorption of Pb , Cu, and Cr ions

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كفاءة المواد الممتصة قليلة التكلفة فى إزالة عناصر الرصاص، النحاس و الكروم من مياه الصرف

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تم دراسة كفاءة إمتصاص نبات ورد النيل ، مسحوق قشر البيض و الكينيت (سليكات الإلومنيوم) وذلك لإزالة عناصر الرصاص، النحاس و الكروم من مياه الصرف ، فالكينيت يعتبر كمادة معدنية تجارية ، مسحوق قشر البيض و نبات ورد النيل الذى يحتوى على عدة مجموعات فعالة لها القدرة على جذب كاتيونات العناصر من مياه الصرف . هذا البحث تم إجرائه كتجربة معملية وذلك باستخدام كميات مختلفة من المواد الممتصة فى

محاليل تحتوى على تركيزات مختلفة (٢٠ ، ٤٠ ، ٦٠ ، ٨٠ و ١٠٠ ملليجرام /لتر) كعناصر منفردة وعناصر مختلطة معا.

أوضحت الدراسة الآتى ، أولا ، أن كفاءة الإمتصاص تعتمد على كمية المادة الممتصة مثل كفاءه الامتصاص لكل من عنصرى النحاس والكروم حيث زادت من ٨٠.٥% الى ٩٥.٢ % و من ٧٥.٣ % إلى ٩١.١ % و من ٩٠.٢ % إلى ١٠٠ % للنحاس و من ٧٨.٥ % إلى ٩١.٦ % و من ٧٢.٩ % إلى ٩٠.١ %و من ٨٨.٧ % إلى ٩٧.٦ % للكروم. وأيضاحوالى من ٩٠.٢% إلى ٩٧.٢ % ، ٨٧.٥ % إلى ٩٥.١ % ثم من ٩٢.٦ إلى ١٠٠% رصاص قد تم إزالتها من مياه الصرف وذلك باستخدام أوزان من ٢ الى ٥ جرام مادة ممتصة فى محاليل تحتوى على تركيزات ٢٠ و ٤٠ ملليجرام / لتر رصاص لكل من نبات ورد النيل ، مسحوق قشر البيض و الكينيت فى محلول يحتوى على تركيز ٢٠ ملليجرام /لتر نحاس و كروم. ثانيا ، وجد أن مخلوط تلك العناصر يؤدى إلى نقص نسبة الإمتصاص عن تلك المحاليل المنفردة حوالى ١.٧٨ % : ٨.٠٢ % : ٢.١٦ % للرصاص بينما نقص إمتصاص عنصرى النحاس والكروم حوالى ٢.٧٤ % : ٦.٢٥ % : ٢.٤٤ % و ١٢.١١ % : ٧.٤١ % : ٤.٦٣ % للمحاليل المختلطة و التى تشير بوضوح إلى أن وجود أكثر من عنصر فى مياه الصرف سوف يؤدى الى نقص فى كفاءة الامتصاص وايضا وجد أن زيادة كمية المادة الممتصة تؤدى إلى زيادة الإامتصاص ، بينما تقل هذه الكفاءه مع زيادة التركيزات.