Removal of Chromium from Leather Industry Effluents

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Abstract - *Environmental pollution with heavy metals is a serious issue worldwide posing threats to humans, animals and plants and to the stability of overall ecosystem. Chromium (Cr) is one of most hazardous heavy metals with a high carcinogenic and recalcitrant nature. Removal of Chromium ions from synthetic tannery wastewater was investigated by using different Adsorbents like Activated Carbon, Bentonite, coffee Powder and Moringa powder with various Parameters including contact time and adsorbent dosage. Activated carbon Adsorbent was the Effective and removal efficiency reached about 98% at contact time 120 minute. The result Shows that Activated Carbon, Bentonite, coffee Powder and Moringa powder can be used as good adsorbents for the removal of Cr ions from synthetic tannery wastewater.*

Keywords - *Adsorption, Chromium ions, Synthetic Tannery Effluent, Activated Carbon,Bentonite, Coffee powder and Moringa powder adsorbents.*

I. INTRODUCTION

To live a healthy life, people all around the world need access to safe water. A lot of industries, together with the fast growing population in world pose a threat to the access of safe water for the citizens. Several of the industries in the world release contaminated water to the nearby streams. One serious pollutant emitted from a leather industry is chromium (Cr), Chromium is the 7th most abundant metal on the earth, which present in the environment, mainly two oxidation states Cr3+ and Cr6+ having average concentration 100 mg/kg [1]. Chromium is normally present in rocks, soil, volcanic and combustion dust [10]. The major source of chromium in industrial waste water is electroplating, aluminum conversion, metal finishing, pigments, leather fungicides, photographic materials and wood preservatives [6].

The global production of chromium is 107 tons per year among them 60-70% is used in alloys, including stainless steel and 15% in various chemical processes [2]. The permissible limit of chromium in drinking water is 0.1mg/L and in waste water is 5 mg/L for trivalent chromium and 0.05 mg/L for hexavalent chromium[2]Leather is tanned with chromium because of hydrothermal stability and excellent physical properties. Leather uptakes only 60-70% of chromium used in tanning process and untreated waste water contain 1500-3000mg/L of chromium[6].A very little amount of chromium is essential for glucose, lipid and amino acid metabolism but long time exposure causes skin disorders, lung carcinoma, kidney damage, dermatitis ulcers and skin lesions[2]. There are several traditional methods to remove chromium from industrial waste water Such as ion-exchange, membrane separation, Electroplating bioaccumulation, precipitation and modified zeolite etc. These methods suffer from various problems like costly, generation of secondary sludge, incomplete metal removal, creation of toxic waste products and high reagent cost etc. Many reports have appeared on the development of low-cost activated carbon from renewable resources and also how to decontaminate water in an environmentally friendly manner. Agricultural and industrial waste materials have been utilized as activated carbon precursors, by a large number of researchers for the removal of chromium. Activated carbons with their large surface area, micro porous character and chemical nature of their surface have made them potential adsorbents for the removal of heavy metals from industrial waste water However, trivalent chromium contained in leather can be under various conditions in small amounts oxidized to form hexavalent chromium[30]. Heavy metal ions are wide spread pollutants of great environmental concern as they are non-degradable and thus persistent in nature that accumulates in the food chain, which with time reach detrimental levels in living systems, resulting in serious health hazards such as irritation in lungs and stomach, cancer in digestive tract, low growth rates in plants and death of animals [5].

In recent years, contamination of the environment by Cr especially hexavalent chromium has become a major area of concern. To wards this direction, several conventional waste water treatment technologies were developed and are used successfully at large scale, to reduce the hazardous compounds concentration in effluents from higher to lower levels [16].Researchers from around the world used various Adsorbents. In this study we utilize Activated Carbon, Bentonite, coffee powder and Moringa Powder adsorbents to remove chromium from liquid waste water leather industry effluent and found that activated carbon adsorbent was best at 120 min with 3gm dose of Adsorbent. This work aims at the removal of chromium from the tannery waste water by abundantly available Activated Carbon, Bentonite,Coffee Powder and Moringa Powder Adsorbents . The present study also aims to develop a suitable methodology to maximize the removal efficiency of chromium by the Adsorption technique [31].

Significance of the study

This study has great significance after it successfully completed. It will used generally in two ways for the whole environment. In one way it will use for leather industries to remove chromium from waste effluents and as a result it will save aquatic ecosystem as well as other living organisms. This will contribute in keeping the countries ecosystem safer. Similarly it will used for the industry to reduce cost of waste treatment since Activated carbon, Bentonite, Coffee powder(used) and Moringa powder are effective and cheaper than other adsorbents. In the other way it will used for the utilization of waste materials for chromium removal application rather than disposal in a landfill. Since it utilizes waste from industries as adsorbent material to clean the waste of other industry, it can create industrial zone. This will help to reduce the environmental burden and will enhance economic benefit of Activated carbon, Bentonite, Coffee powder(used) and Moringa powder. For the researchers it will provide opportunities to increase the practical knowledge. Furthermore for the University it will help to promote its mission and build up the link with the industry.

II. LITERATURE REVIEW

***Tanning and Characteristics of Tannery Waste*:** Tanning is a process by which putrefable biological material, such as leather, is converted into a stable material which is resistant to microbial attack and has enhanced to wet and dry heat. Tanning consists of a series of successive operation converting raw hide and skin into leather. During the tanning process at least about 300 kg chemicals are added per ton of hides. The uncontrolled release of tannery effluents to natural water bodies increases health risks for human beings and environmental pollution. Tannery wastewater is characterized by being strongly alkaline with a high salt content, one of which is chromium [17].

Chemistry of Chromium: Chromium is a heavy metal, symbolically represented by Cr, with atomic number of 24, and mass number of 51.9961. It belongs to the first series of transition metals. It is the 17th most abundant element in the earth's crust. Chromium occurs naturally in ultramafic igneous rocks, soil, plants, animals, and in volcanic dust and gases. It may occur in nine different forms of oxidation state ranging from Cr (-II) up to Cr (VI), but the two common valence states are trivalent Cr (III), the most stable, and hexavalent Cr (VI) which is not thermodynamically stable. Solubility of chromium (III) depends on pH; decreases dramatically at pH value greater than 4.5.Cr (III) can form stable complexes with organic legends at pH value as high as 8.5[13]. During the tanning process only Cr (III) salts are used. Nevertheless, under certain prevailing conditions, the Cr (III) can be transformed into Cr (VI). Also, the conversion of Cr (VI) to Cr (III) occurs readily by a variety of oxidizing agents in water and soil, but a few oxidants in natural water can oxidize Cr (III) to Cr (VI). Cr (VI) is known human carcinogen, where as Cr (III) is less toxic. Chromate is toxic to both plants and animals and strong oxidizing agent and corrosive in nature [11].

Chromium In Drinking Water Standard: The Safe Drinking Water Act (SDWA) requires EPA to determine the level of contaminants in drinking water at which no adverse health effects are likely to occur. These non-enforceable health goals, based on possible health risks from exposure over a lifetime, are called maximum contaminant level goals (MCLGs).EPA sets enforceable standards for drinking water contaminants based on the best available science to prevent potential health problems. In most cases, the enforceable standard is known as a maximum contaminant level (MCL), the maximum permissible level of a contaminant in water which is delivered to any user of a public water system. MCLs are set as close to the health goals as possible after considering costs, benefits, and the ability of public water systems to detect and remove contaminants using suitable treatment technologies. The national primary drinking water regulation that established the MCL for total chromium of 0.1mg/L was promulgated in 1991. The SDWA requires EPA to periodically review the national primary drinking water regulation for each contaminant and revise the regulation, if appropriate [3].

Toxicity of Chromium: Chromium has a ‘chronic’ toxic effect upon aquatic life. Cr (VI) is toxic to fish life since they rapidly penetrate cell walls. They are mainly absorbed through the gills and the effect is accumulative. Ingesting small amounts of Cr (both Cr +3 and Cr +6 forms) will generally not harm, however

ingesting above recommended levels over long periods of time can result in adverse health effects including gastrointestinal irritation, stomach ulcers, heart burning, respiratory tract infection, sever cough, fever and loss of eyesight. Lung cancer and kidney failure were the reported causes of death in many cases. Skin contact may result in systemic poisoning, damage or even severe burns, interference with the healing of cut or scrapes which, if not treated properly, may lead to ulceration and severe chronic allergies. Eye exposure may cause permanent damage. In general, Cr (VI) is more toxic, more soluble, more mobile and hence absorbed into cells more readily than Cr (III)[8]. This chromium can replace other metals in biological systems with toxic effects and its accumulation throughout the food chain leads to serious ecological and health problems. Chromium is also toxic to agronomic plants at about 5 to 100 µg/mg of available Cr in the soil. Plants absorb Cr (VI) better than Cr (III); whereas Cr (VI)
is more toxic. Plant growth studies of solution cultures with low levels of Cr have indicated that
Cr is not an essential component of plant nutrition. Although some crops are not affected by low concentration of Cr, it is toxic at high concentration or may reduce yield. Higher application of chromium to the plant soil growing media brought poor quality and poor growth of the seedlings [5].

Adsorption: Adsorption may be defined as the process of accumulation of any substance giving higher concentration of molecular species on the surface of another substance as compared to that in the bulk. When a solid surface is exposed to a gas or a liquid, molecules from the gas or the solution phase accumulate or concentrate at the surface. The phenomenon of concentration of molecules of a gas or liquid at a solid surface is called adsorption. Adsorption is a well-established and powerful technique for treating domestic and industrial effluents. In water treatment, the most widely method is adsorption onto the surface of activated carbon. Activated carbon is used for adsorption of organic substances and non-polar adsorbate and it is also usually used for waste gas (and waste water) treatment. It is the most widely used adsorbent since most of its chemical (surface groups) and physical properties (pore size distribution and surface area) can be tuned according to what is needed. Its usefulness also derives from its large micro pore (and sometimes mesopore) volume and the resulting high surface area[14].
Adsorption is a process that uses solids for removing substances from either gaseous or liquid solutions. Adsorption phenomena are operative in most natural physical, biological, and chemical systems. The process of adsorption involves separation of a substance from one phase accompanied by its accumulation or concentration at the surface of another. Adsorption capacity depends on adsorbate surface properties, adsorbate chemical properties, temperature, pH etc.There are number of adsorbents are available for the removal of chromium from water. The adsorption will take place in two ways.

Physical Adsorption: **It** is a result of intermolecular forces of attraction between molecules of the adsorbent and the adsorbate. In this case, the adsorbate merely condenses in a thin on the surface of adsorbent solid. The intermolecular attractive forces that retain the adsorbent on the surface are purely physical and are called Van der waals forces. As these forces are very weak, the adsorbate is loosely bound to the surface of the adsorbent [9].

Chemical Adsorption or Chemisorption:In this case, molecules of the adsorbate are retained on the surface of the solid by chemical bonding that means it involves chemical interaction between the adsorbent solid and the adsorbed substance. Identifiable chemical compound may not actually result but the adhesive force is much greater than that in physical adsorption. In many cases, it is irreversible. Adsorbents are classified into two types, synthetic adsorbents and natural adsorbents [9].

Synthetic adsorbents: Synthetic adsorbent are those which are prepared artificially. Such are zero-valent iron, granular ferric hydroxide, goethite, activated alumina; iron-oxide coated sand, activated carbon. These are some of the synthetic adsorbents used for removal of chromium from water which are having the arsenic removal efficiency more than 90% but the cost of these synthetic adsorbents are high and the regeneration cost is also high[9].

Natural adsorbents*:* Natural adsorbent are those which are naturally available in atmosphere. Extensive literature indicated that agricultural wastes either in natural form or modified form are highly efficient for the removal of chromium ions from aqueous solution, those are Moringaoleifera leaves, rice husk, wheat husk, banana peels, peanut peels, pea peels, tree barks of mango, java plum,neem, clay, fuller's earth, betonies, fly ash; bagasse, zeolite,sepiolite, pyrolusite, limonite, dry plants, red mud, and various other materials such as wood, peat, clay, kaolin,humic acid, human hair, hematite or feldspar, pine needles, cactus leaves, polymer materials, tea leaves, tree fern, orange juice residue, coconut coir pith, ferruginous manganese ore etc.[10].

III. MATERIALS AND METHODS:

Chemicals and Reagents

The major chemicals and reagents used to carry out batch adsorption experiment are: Dilute (0.1M) HCl and NaOH for pH adjustments and as cleaning agents; Chromium sulfate (Cr2(SO4)3 powder form to prepare stock solution; Activated carbon,Bentonite,coffee powder and Moringa powder as adsorbents and Distilled water to prepare solution.

Methodology

The adsorbents used in this study were activated carbon, Moringa seed powder, Bentonite and Coffee residue. All chemical solutions were prepared from certified lab grade chemicals. Chromium sulfate was used as the source of Cr (IV) in the synthetic wastewater. The Cr stock solution (1000 mg/l) was prepared by dissolving 2.828 g Cr2(SO4)3 in 1000 ml of deionized water, while Cr concentration of synthetic wastewater was varied from 25 to 75 mg/l by diluting the stock solution. Before being used for adsorption studies, the pH of Cr solution at different of Cr concentration was measured using a pH meter. Adjustment of pH was carried out using 0.1N dilute HCl. Agitation of the system under investigation was carried out on a magnetic stirrer (2 MLH Model), at different time intervals, different concentration and different adsorbent doses. Finally, Chromium concentration was analyzed by using Atomic Absorption spectrophotometer (model noVAA400p)[23,19].

Preparation activated carbon adsorbents

Preparation of Activated Carbon: Activated carbon purchased from Puretlitte Company with a size Of 20microns, and sieve size is 250-500 μm. This sample is used for the study of adsorption for removal of Chromium in leather industry effluent at different time intervals and different doses. All the experiments are carried out at room temperature[28].

Figure 1 Activated carbon adsorbent

Preparation Bentonite Adsorbents

Bentonite sample was collected from ArbaminchUniversity,college of natural sciences, Geology department. The bentonite sample was oven dried at 100 for 2 hours. The sample was crushed to fine powder and was passed through 25 microns sieve. This Bentonite sample was used to study the removal of chromium in leather industry effluent at different time intervals and different adsorbent doses. All experiment are carried out at room temperature [20]

Figure 2 Bentonite adsorbent

Preparation coffee powder (Used) Adsorbents

This sample was collected from college of natural sciences cafeteria in the form of wet. The sample was dried for two days at room temperature. Dried sample used as adsorbent for removal of chromium from leather industry effluent at different time intervals and different adsorbent doses. All the experiments are carried out at room temperature [29].

Figure 3 coffee powder adsorbent

Preparation Moringa powder Adsorbents

This sample was collected from around Arbaminch in the form of seed. The seed sample was sundried for four days at room temperature. Dried seed sample were grinded in fine powder form. The fine powder form of sample used as adsorbent for removal of chromium from leather industry effluent at different time intervals and different adsorbent doses. All the experiments are carried out at room temperature[25].

Figure.4 Moringa powder adsorbent

Preparation of Chromium (Violet bluegreen) stock solution

First 2.828g of powdered Chromium was weighted using electronic weight balance. The weighted Chromium sulfate was dissolved in 1000ml of distilled water to prepare 1000mg/l of a stock solution in volumetric flask. The pH of the stock solution was adjusted in the range of (6.8 to 7) using either 0.1 M HCl or 0.1 M NaOH. Then a 20ml of diluted stock solutions with different concentration of (0ppm (distilled water), 25ppm, 50ppm and 75ppm) were prepared by taking some calculated Chromium sulfate solution from the stock solution and diluting with distilled water [22].

Adsorption Experiments

Batch adsorption experiments were conducted using three levels and three factors in order to determine the optimum adsorption capacity of adsorbent, adsorption equilibrium time and acceptable initial chromium concentration. After a standard calibration curve was prepared,150ml of stock solution with different chromium concentrations (25 mg/l, 50mg /l and 75mg/l) were prepared in 250 ml Erlenmeyer flasks from the stock solution[20]. Activated carbon, bentonite, coffee powder, moringa powder samples with adsorbent dose of (1 gm, 2 gm and 3 gm) were weighted. Depending on the order of experiment from experimental design, the weighted Activated carbon, bentonite,coffee powder,moringa powder samples were dosed into the prepared chromium solutions turn by turn and mixed using magnetic stirrer for the maximum time of 2 hours. During mixing the sample was taken in time intervals of (60 min, 90 min, and 120 min) from each beaker without stopping the stirrer with the help of micropipette. This work was repeated until 27 total numbers of experiments were carried out. After the concentrations of Chromium in final were determined and the adsorption capacity (qe), was calculated as:

Qe=(C0-Ce)*V/m

Where Co and Ce are the initial and equilibrium chromium concentrations in (mg/lit),respectively, V is known volume of chromium solution in (litter), and **m** is a known mass of dry Activated carbon,bentonite,coffeepowder(used),moringa powder in (gram). Finally, the adsorbent or performance of Activated carbon,bentonite,coffeepowder, moringa powder are Compared [18,27].

IV. RESULTS AND DISCUSIONS

Studies on activated Carbon adsorbent at different concentration and different time intervals

In the following figure.5 shows that, the percent removal of chromium on adsorbent dose at 25mg/lit concentation.The results of adsorbent dose of 1gm,2gm and 3gm and time intervals 60,90 and 120 min. shows that, the time increases removal of chromium increases with increasing of adsorbent dose and time. As time time reaches at 120 min adsorbent dose 3 mg/lit shows maximum removal of chromium around 96%.If continue further no much more removal of chromium takes place. Based on the experimental results and literature shows that around 100 min to 120 min and adsorbent dose 3gm is the optimum. For adsorbent dose 2gms maximum removal chromium around 94% and for dose 1gm 93%.In the following figure.2 shows that, the

Percent removal of chromium on adsorbent dose at 50mg/lit concentation.The results of adsorbent dose of 1gm,2gm and 3gm and time intervals 60,90 and 120 min. shows that, the time increases removal of chromium increases with increasing of adsorbent dose. As time reaches at 120 min adsorbent dose 3gm shows maximum removal of chromium around 98%.If continue further no much more removal of chromium takes place. Based on the experimental results and literature shows that around 110 min to 120 min and adsorbent dose 3gm is the optimum. The same type results are shown in concentration 75mg/lit in figure 3.maximum removal of Chromium has taken place around 98.9% at 115 min.

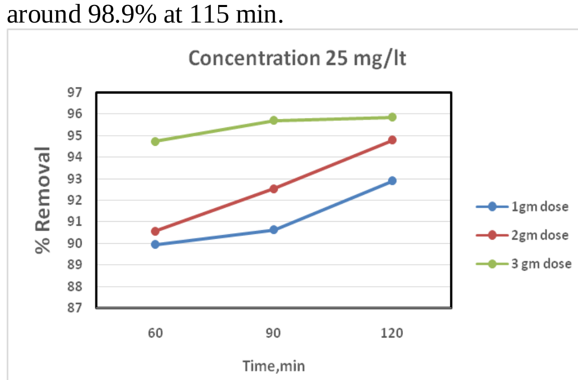

Figure.5:Removal of chromium Versus time with changes of adsorbent dose at concentration 25mg/lit

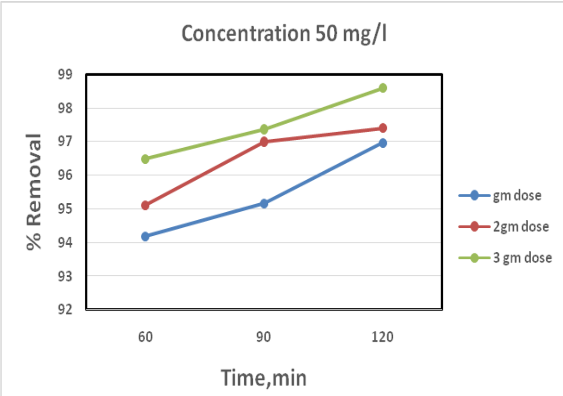

Figure: 6.Removal of chromium Versus time with changes of adsorbent dose at Concentration 50mg/lit

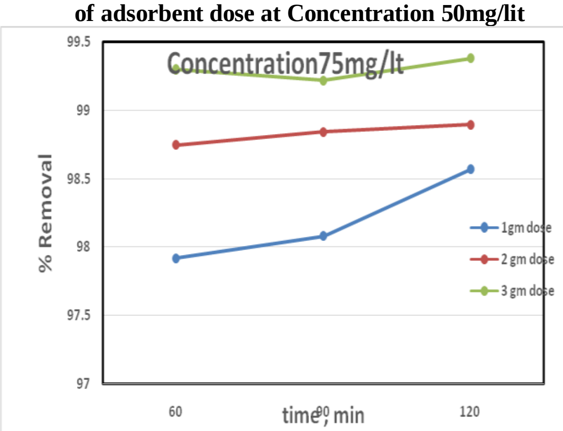

Figure: 7.Removal of chromium versus time with changes of adsorbent dose at concentration 75mg/lit

Studies on Bentonite Adsorbent at different concentrations and different time Intervals

In the figures bellow shows that, the percent removal of chromium versus time at 25mg/lit ,50mg/lit and 75mg/lit concentration, adsorbent dose of 1gm,2gm and 3gm and time intervals 60,90 and 120 min. As time increases removal of chromium increases with increasing of adsorbent dose. All results are shown in figures 8,9,10. As time reaches at 120 min adsorbent dose 3 mg/lt shows maximum removal of chromium.

For concentration 25mg/lit at adsorbent dose 1gm shows 89% removal at 120 min,where as for dose 2 and 3 gms it shows that 92 and 94% as shown in figure 8.

For concentration 50mg/lit at adsorbent dose 1gm shows 94% removal at 120 min, where as for dose 2 and 3 gms it shows that 96 and 97%as shown in figure 9.For concentration 75mg/lt at adsorbent dose 1gm shows 98% removal at 120 min,where as for dose 2 and 3 gms it shows that 98.57 and 99%as shown in figure 10.

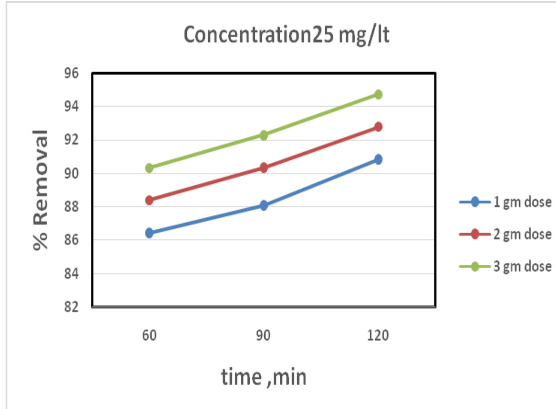

Figure.8 :Removal of chromium Versus time with changes of adsorbent dose at concentration 25mg/lit.

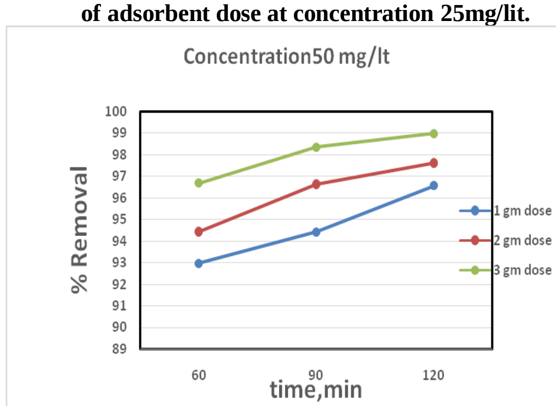

Figure.9: Removal of chromium Versus time with changes of adsorbent dose at concentration 50mg/lit.

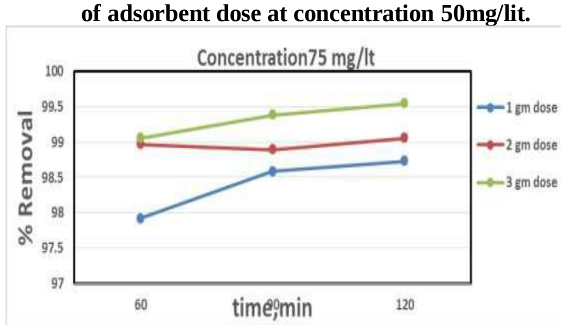

Figure.10: Removal of chromium Versus time with changes of adsorbent dose at concentration 75mg/lit.

Effect of Adsorbent on Chromium concentration 25 mg/lit at diffent time interval

The following results shows that, the removal of chromium at 25 mg/lit and adsorbent dose 1gm.The percent removal of chromium increases in increasing time. The highest removal chromium takes place for activated carbon but lowest for moringa powder, this is due the pore sizes of adsorbents. Activated carbon pore size more than other adsorbents as per studies. Pore size directly proportional to the adsorption capacity. Even at 60 min activated carbon removes more than 90%.At 120 min more 95% removes, where as other adsorbents are not removing chromium that much. The interesting point is that removal of chromium is more and more from time 1 min to 60 min, after that the capacity of adsorption slowly increasing with time. The capacity of adsorption is more by the increasing time 1 to 60 to 120 min for all adsorbent as result shown bellow the figures 13,14,15. According our result adsorption capacity of adsorbent is increased by the order of Activated carbon Bentonite Coffee powder(used) Moringa powder.

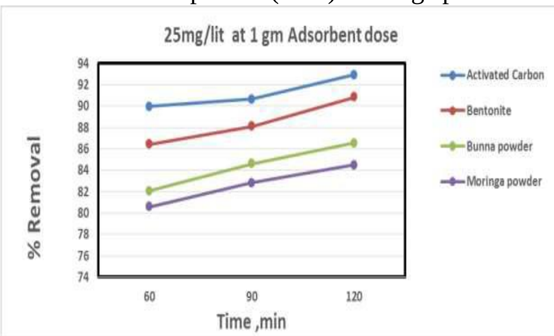

Figure:13. Removal of chromium Versus time with different adsorbents dose (1gm) at concentration 25mg/lit

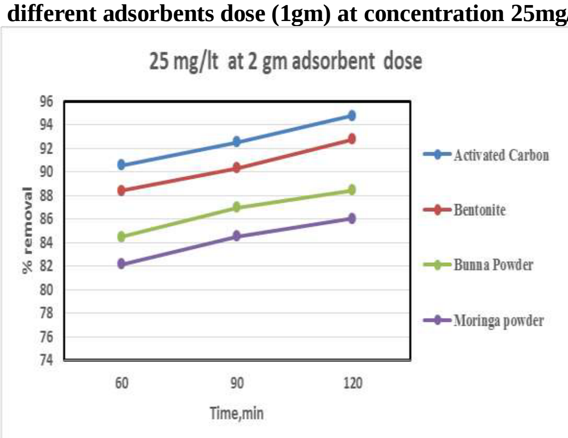

Figure: 14.Removal of chromium Versus time with different adsorbents dose (2gm) at concentration 25mg/lt

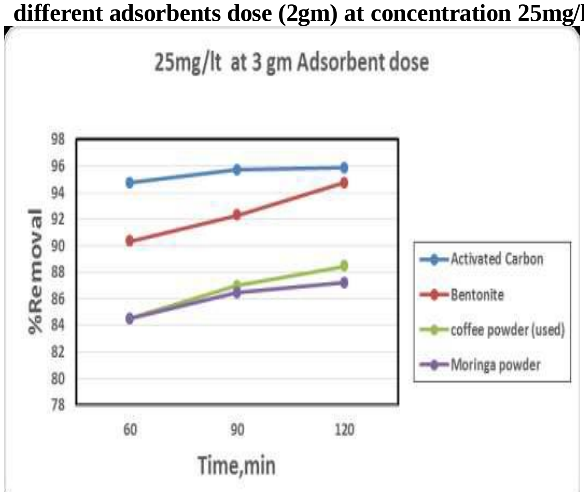

Figure:15.Removal of chromium Versus time with different adsorbents dose (3gm) at concentration 25mg/lit

***Effect of Adsorbent on Chromium concentration 50 mg/lit at diffent time interval*.**

The following results shows that, the removal of chromium at 50 mg/lit and adsorbent dose 1gm.the percent removal of chromium increases in increasing time. The highest removal chromium takes place for activated carbon but lowest for moringa powder, this is due the pore sizes of adsorbents. Activated carbon pore size more than other adsorbents as per studies. Pore size directly proportional to the adsorption capacity. Even at 60 min activated carbon removes more than 94%.At 120 min more 96.8% removes, where as other adsorbents are not removing chromium that much. The interesting point is the removal of chromium is more and more from time 1 min to 60 min.after that the capacity of adsorption slowly with increasing with time i.e., the adsorbent has removed more at initial time means at 60 minutes. The capacity of adsorption is morethe increasing time from 60 to 120 min for all adsorbent as result shown bellow the figures 16,17,18. According our result adsorption capacity of adsorbent is increased by the order of Activated carbon Bentonite coffee powder(used) moringa powder.

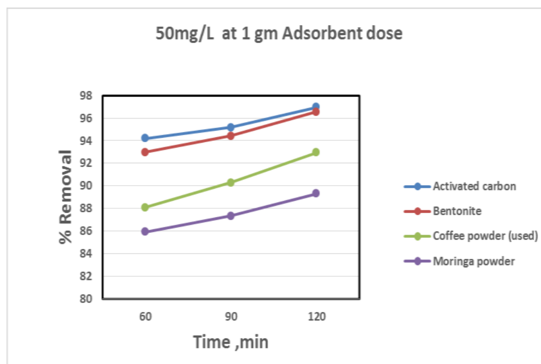

Figure:16.Removal of chromium Versus time with different adsorbents dose (1gm) at concentration 50mg/lit

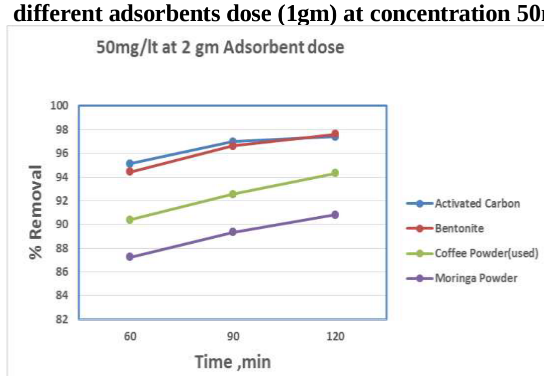

Figure:17.Removal of chromium Versus time with different adsorbents dose (2gm) at concentration 50mg/lit

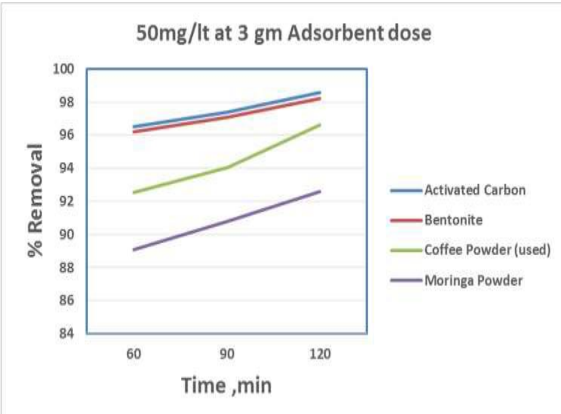

Figure: 18.Removal of chromium Versus time with different adsorbents dose (3gm) at concentration 50mg/lit

CONCLUSION

From the experimental result it can be concluded that adsorption is an effective method of lowering and removing the concentration of dissolved Chromium in the waste water effluent. It is one of the most efficient methods and has an advantage over the other methods due to its sludge free clean operation, relative lower cost and complete removal of Chromium. In this work Activated Carbon, Bentonite,Coffee powder(used) and Moringa powder was successfully employed as an adsorbents for the quantitative removal of chromium from aqueous solution. Experiments were performed at room temperature (25oC) and PH of 6.8 as a function of mixing/stirring time, initial Chromium concentration and adsorbent dosage. Based on the experimental data, removal efficiency increased with increasing initial chromium concentration, increasing adsorbent dose and mixing time. In the case of adsorbent dose, the increment Chromium removal with adsorbent dosage is due to the increase of available amount of
adsorption site. In the effect of adsorbent dose, treated activated carbon has more performance
than others to remove Chromium. As for the effect of chromium initial concentration, increasing the initial concentration enhances the contact of adsorbent surfaces with chromium. However,after 75 mg/l of chromium concentration, 3 g of adsorbent dose and 120 min of contact time a saturation point was appeared. After saturation point the removal of chromium may increase in decreasing rate. Batch studies confirm that this cost effective adsorbents can be used as a substitute for high cost adsorbent in the market nowadays such as activated carbon

RECOMMENDATIONS

The approach used in this study has given very promising results; however, there are a number of areas that need further investigation. Studies on the effects of other process parameters such as temperature, PH and particle size as well as the effect of different sources of adsorbents on adsorption performance can also be an interesting area of future study. In order to widen the applicability of the Activated carbon, bentonite, coffee powder and Moringa powder as adsorbents, it must be tested using the real Leather wastewater and the leaching experiments must be carried out to check for the regeneration capability of Adsorbents. Further pretreatment experiments for adsorbents must be carried out in order to improve its adsorption capacities by increasing its physicochemical properties such as specific surface area, pore volume and active sites. Finally further research could also be carried out to study the adsorption properties of Activated Carbon, Bentonite, Coffee powder (used) and Moringa powder in the form of pellets rather than powders.

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