Adsorption Kinetics Of Chromium (VI) On Eco Friendly Adsorbents

By

A. Prithiba

A DISSERTATION SUBMITTED TO THE AVINASHILINGAM INSTITUTE FOR HOME SCIENCE AND
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Certified as Bonafide Research work

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Introduction

I INTRODUCTION

EARTH is enshielded in a marvellous mantle whose exquisite material consists of delicate interwoven threads of life. This durable filmy fabric was fashioned over several billion years and it is this factor that ensures the survival and growth of the biotic elements or living being. Any adverse effect or the disturbance on the delicate balance of the biosphere caused by man's careless or excessive exploitation of natural resources can lead to creation of unfavourable conditions for the well being of mankind. Pollution is one such process that disturbs such balance. Pollution is derived from the latin word "Pollutionem" meaning defilement.

EARTH MAKES AN INDUSTRIAL MAN. This saying although true have multiplied the hazards to which human beings are exposed. The existence of pollution in the environment as a national and world wide problem was not generally recognised until 1960's. In early days of man's existence the amount of waste was limited. It was disposed off locally and had virtually no effect upon the environment but the increase in population, urbanization and industrialization has created a lot of contaminations and disposal of waste water into natural resources. This was the origin of the problem of pollution into natural resources which developed an imbalance on the ecosystem.

Our ability to produce wastes and abuse our environment dogs every progressive step we make as if marking our own shrewdness. Our inability to foresee, much less control our waste making takes on the form of social regurgitation.

Pollution is defined as any deterioration in the physical, chemical, biological quality of the environment. Every human society, be it rural, urban, industrial or more technologically advanced disposes off certain kinds of by-products and waste products which when injected into the biosphere in great quantities affect the normal functioning of the ecosystem and have an adverse effect on plants, animals and man. These are collectively called pollutants. **(Smith 1997).**

The fruits of technological processes allowed us to overlook its more harmful consequences on the environment. Industry has generally been seen as an arch polluter on a massive scale totally insensitive to the communities - short as well as long - term interest unwilling to be open in its communications with society at large and driven almost solely by the profit motive if not greed.

The inability or unwillingness on the part of industry to articulate the facts, its changing attitudes and growing efforts to be environmentally more responsible have aggravated the adverse perception which is partially attributable to industries greatly visible than other segments of the society.

1.1. WATER POLLUTION

Pollution - the skull and cross bones of modern civilization threatens not only terrestrial vegetation but also surface water. Throughout history stability and economic welfare have been closely related to the ability of civilizations to control and manage the water surrounding them as it is the life - blood of the climate system. Water of the right quality is a necessary pre condition for nature to thrive and develop. Though water is nature's most wonderful, abundant and useful compound it is the most misused one since the time man strode the earth. Pollution of water bodies is increasing steadily due to rapid population growth, industrial proliferations, urbanizations, increasing living standards and wide spheres of human activities.

Water pollution may be defined as the addition of any organic, inorganic, biological or radiological substances to water which changes its natural qualities so that the riparian properties does not get the natural water of stream transmitted to him (or) it is a by product of rapid and unplanned industrial progress and over population.

It is mainly caused by

1. Natural process in which the decomposed vegetable animal & weathered product are brought into water resources.
2. Anthropogenic process such as industrial, agricultural, urban, domestic, radioactive, mining sources, use of pesticides & fertilizers by man.

1.2. Sources of Heavy metal pollutants:-

Both the domestic and industrial wastes contribute to the water pollution. However it is the industrial waste which is responsible for the major toxicity in the water. The effluents from a large number of industries, such as tanning, textile, mining, chemical and refineries, contain a number of toxic substances and heavy metals. These substances are discharged directly into the river water without any pre treatment. Besides biological waste, tanning industry effluent contains chemicals such as sodium sulphate, lime, sodium sulphide, caustic soda amines, chlorides acids and most predominantly Chromium salts. Chromium content in the effluent may reach as high as 9 mg/l while chlorides make it upto 4000 mg/l **Dikshit et al., 1989)** Moreover the presence of alkalinity in waste water destroys bacteria which are useful in the degradation of the organic matter, including proteins discharged during the tanning process. Refineries further deteriorate the quality of water by adding toxic metals like Chromium, Lead, Copper, Nickel, Zinc, Mercury, from the effluent **(Poddar et al., 1991)** A number of metal industries related with the electro plating and metal processing produce effluents containing copper which could reach a concentration of 0.9 mg/l while a typical wollen textile mill produces Chromium content of 100 mg/l **(Nisha et, al, 1984)** In addition to these Chromates are frequently added to cooling water for corrosion control. Some Chromium finds its way to the environment via the weathering of natural rocks. The industrial effluents are therefore the main sources of heavy metal contaminants in river water.

1.3. Toxicity of Chromium:-

Chromium is a highly active transition metal which exists is several oxidation states from divalent to hexavalent form. However the major stable aqueous species are either in the hexavalent or trivalent state. Chromium (III) is less toxic than chromium (VI). Hexavalent chromium is corrosive to flesh and is also carcinogenic. The presence of chromium (VI) in the aquatic environment at high concentration is lethal to marine species. If present in low concentration it affects fish reproduction and accumulates in fish tissue. Ingestion of Cr (VI) may cause epigastric pain, vomiting, severe diarrhoea and haemorrhage. Chromic acids and its salts have corrosive action on mucous membrane.

1.4. Problems faced in fighting pollution:-

Development without destruction is the basic need of the developing countries and this need cannot be met without strategic planning, which in turn, necessitates,

- sound natural environmental policy
- adequate legislation

- Infra structure for collecting data and implementing pollution control programmes
- acquisition of appropriate technology
- an appropriate fiscal policy

In this context the developed countries can help the developing countries by transferring the latest technology relevant to incineration, wet oxidation etc for dealing with toxic waste; providing adequate training opportunities in the developing countries and setting up demonstration plants dealing with special and toxic pollutants.

1.5. TREATMENT OF WATER POLLUTION

In recent years increasing awareness of water pollution and its far reaching effects has prompted concerted efforts towards pollution abatement. River waters contaminated by industrial wastes containing heavy metal toxic ions are hazardous to humans and animals. The most important feature that distinguishes heavy metals from toxic pollutants is their non biodegradability and they even have a tendency to accumulate in living material.Therefore the elimination of heavy metals from waste water is an important subject for public health. As the resolution of biological problems has recently gained a global significance on account of the increasingly growing pollution, the need for purification of potable and waste waters from various pollutants require the development of new methods. Since the waste contains a wide range of pollutants involving bio organics and metallic salts, it is necessary to carry out the treatment based on its composition. The control measures may be classified into three different groups - physical, chemical and biological. These methods could be used separately or in combination with each other. The coventional methods involve reduction of Cr (VI) to the trivalent state followed by alkaline precipitation of $Cr(OH)_3$, co-precipitation, reverse osmosis, solvent extraction, electro deposition, alum coagulation. However these processes are expensive and may not be an economic alternative to the industry and most of these do not show effectiveness for large scale operations. These are the chemical methods and are used when the metal concentration in the industrial effluent is very high. Biological method on the other hand may be used in the industries related with the leather treatment and food processing. For example in the case of tannery effluent it contains a large amount of biomaterial such as fleshings, hairs, skins along with other metallic chemicals used in tannery process. Under such cases a two step cleaning process is required. The biomaterial may be filtered out from the ponds containing waste water. This leaves behind effluent containing only metal salts. Metals are then removed by the physical methods which involve adsorption or ion exchange process. In this study the major interest is to explore the efficiency or removal of Chromium (VI) by using the process of adsorption.

1.6. ADSORPTION-AN OASIS IN THE DESERT OF TREATMENT METHODS

Adsorption is, by far, the most versatile and widely used technique for the removal of toxic pollutants from aqueous solution. Moreover this technique has an added advantage of having no sludge disposal problems as compared with other tecniques, for there is regeneration of the adsorbent which makes the system more economical.

Activated carbon adsorption appears to be a highly effective process for the removal of heavy metals at trace quantities. However the use of activated carbon is not suitable for developing countries because of its high cost. For that reason the use of low cost materials as a possible media for metal removal from waste water have been highlighted recently. These materials range from industrial products such as fly ash from thermal power station and waste rubber tyres **(Knocke and Hemphill, 1981)** to agricultural products such as wool, rice straw, coconut husk, peat moss **(Macchi et al 1986)**. New research shows effective adsorption of heavy metals from waste water using agricultural products and by products such as wall nut expeller meal, peanut skins, wool, rice husk, plumpit shells, peanut hults, sugar cane bagasse (Ferro-Garcia et al 1988), waste tea leaves and coffee powder.

The present study reports work on removal of Chromium (VI) by cheap carbon derived by carbonisation of waste tea leaves and rice husk with sulphuric acid.

1.7. OBJECTIVES

The objectives of this study are to

1. To determine the optimum temperature, optimum pH, optimum time, optimum dosage for the efficient removal of Cr (VI) from the effluents.
2. To determine the optimum temperature, optimum pH, optimum time, optimum dosage for the maximum recovery of Cr (VI) back from the adsorbents.
3. To interpret the results of the adsorption/desorption studies in term of Freundlich, Langmuir isotherms.
4. Tointerpret the results of the present study in terms of kinetic equations Lagergren and Weber and Morris intraparticle difussion equation.

Review of Literature

II REVIEW OF LITERATURE

All experience is an arch where through, gleams the untravelled world whose margin fades for ever & ever when we move. The advancements of today were seeded by research years ago. The development of tomorrow are the offspring of research planned today.

An acquaintance with related literature of past studies is a must for any research for formulating sound methodology, which acts as a persevearing force during the onset of research From these literature new areas of research can be inferred.

2.1. THE CHEMISTRY OF CHROMIUM

Chromium is a highly active transition metal which exists in several of oxidation states from the divalent to hexa valent form however, the major stable aqueous species are either in the hexavalent or trivalent state. The stability of the various chromium species is dependent upon the various redox and the pH conditions that is below the pH (3), temp (20-30 ° C) and the redox conditions commonly found in industrial waste waters -the predominant species are bichromate, $HCrO_4^-$, dichromate $Cr_2O_7^{2-}$ and Cr^{+3}. It is interesting to note that the divalent chromium ions Cr^{+2} may be found in extremely reducing environment.

Trivalent chromium exists predominantly in the form of Cr(III) which is a hydrolyzable cation. It is noted that for a total Cr (VI) concentration range from 10^{-4}M to 5 X 10^{-3}M , solid Chromium hydroxide may precipitate at pH values from 5.5 to 6.

2.2. REMOVAL OF CHROMIUM

Due to its toxicity it is necessary to remove chromium from the waste waters. Conventional means of treating chromium containing waste waters include the reduction of hexavalent Chromium to the trivalent state with strong reducing agents such as bisulphite or sulphur dioxide followed by precipitation of Cr (OH) at an alkaline pH and separation by gravity setting or by selective ion exchange.

Recent attention have been focussed on the high accumulation of heavy metals including chromium, in municipal sewage sludges which limits the amount of sludge that can be applied to agricultural lands, making sludge disposal increasingly difficult. It is obvious that pretreatment of plating waste water prior to municipal discharge is necessary to alleviate this problem.

2.3. METAL REMOVAL EFFICIENCY:

The removal of heavy metals during primary sedimentation is important for two reasons. For one it reduces the metal loading in the biological stage of the treatment, thus reducing possibility by impairment of the efficiency of treatment as a result of metal toxicity (**Barth et al., 1965**). Secondly, it contributes to the overall removal efficiency for the treatment work thus reducing contamination of surface water to which final effluents are discharged. Metal removal during primary sedimentation is a physical process, depending on the settling of precipitated metal or the association of metals with settlable particulate matter. Minimal revomal of dissolved metals occur during this process and the proportion of dissoved metal to total metal in the effluent increases as a result (**Oilver & Cosgrove, 1974**).

Although the principle determinants of metal removal during primary sedimentation are the efficiency of suspended solids removal and the chemical species of the metal (Soluble or insoluble) these two factors are influenced by several others. Suspended solids removal is affected by design, surface loading, flow, day, weather or storm and influent suspended solids concentration. Chemical species of metal may depend on the influent metal concentration influent chemical oxygen demand and pH of the waste waters. As a result, removal of heavy metals during primary sedimentation vary widely (**Lester, 1987**)

The fate of heavy metals in waste water treatement process was studied by**Nomura & Young (1974)** at the wahiawa sewage treatment plant in Hawaii. The step-aeration activated sludge treatment process, removed most of the heavy metals, Al, Ca, Cu, Fe, Hg, Pb & Zn that were detected at the plant in concentrations varying from a few parts per billion of Hg to a few ppm of Iron. Hexavalent Chromium was removed some what less efficiently. Results of this investigation showed that most of these metals were removed by precipitation with the sludges in primary treatment and further removal occured through biological uptake in the secondary phase of the treatment.

The removal of heavy metals during primary and secondary treatment can proceed by 2 mechanisms (**Argo and Culp, 1972**); (i) preciptation of metal hydroxides which are removed with the sludge (ii) sorption of the heavy metals. They studied the removal of heavy metals at the orange county water districts waste water reclamation pilot plant. This plant consisted of units for chemical treatment, sedimentation, ammonia stripping, recarbonation, mixed media filteration, activated carbon adsorption and chlorination. Results from the study showed that activated carbon adsorption besides time coagulation and mixed media filteration was effective in reducing the concentration of Cadmium hexavalent Chromium, Zinc and Ca. These results were of confirmation of the results obtained by **Linsted et al (1971)**.

The overall removal of total Chromium from the primary industries was 27% and from secondary was 58%. (Brown et al). This result showed that Chromium, Copper and Lead were more efficiently removed in secondary treatment processes than in primary processes. This is an added justification for the adoption of no less than secondary treatment processes in waste-water treatment.

Most of the secondary processes are found to be eiher expensive or inconsistant in operation due to many interfearing agents and recently researchers have explored the possibility of using an activated carbon adsorbent for removal of Cr (VI) which is found to be very efficient in removal of heavy metals and also cost effective. Hence the need for adsorption process.

2.4. ADSORPTION

Adsorption is the accumulation of materials at an interface liquid- solid. Adsorption is used widely in waste water treatment. Dissolved materials tend to either accumulate at interface depending on their relative strength of attraction for themselves or for the solvent.

2.4.1. Adsorption on activated carbon:-

Activated carbon adsorption appears to be a highly effective process for the removal of heavy metals at trace quantities. However the use of activated carbon is not suitable for developing countries because of its high cost. For that reason the use of low cost materials as a possible media for metal removal from waste water have been highlighted recently.

According to **Singh and Lal 1992**, the used tea leaves-a waste converted to carbonaceous meterial exhibits exceptionally high degree of Chromium (VI) adsorption and hence can be utilised for the treatment of industrial waste water effluents containing Chromium (VI) at an optimum pH - 2.

According to**Balasubramanian** 1996 goat hairs - a waste material in tanneries have been used to purify the effluent of the same tannery successfully with respect to eliminating the highly toxic Chromium ions and found to be highly technical and economical method.

Through batch experiments removal of Chromium (VI) by blast furnace flue dust generated in the steel plants have been studied, under ambient temperature and pH condition removal effected to the extent more than 95% has been observed. The adsorption process follows I order kinetics with respect to metal ion concentration (Patnaik and Das 1995).

According to Singh, D.N Saksena & N.Tiwari, 1994., the leached Acacia Arabica bark - a waste material from vegetable tanning industries converted to a sorbent material by treatment with formadehyde in acidic medium has exhibited a good sorption potential for Cr (VI) with peak value at pH 2.

Effective chromium removal from acidic electroplating waste water has been achieved by coconut shell based activated carbon (CSBAC). It is found that removal of Chromium (VI) by CSBAC is almost complete (94%) after 4 hours of contact time, whereas 82% and 88% of Chromium (VI) removal are observed after 20 minutes and one hour respectively at an optimum pH of 2.5 (Alaerts et al 1989).

Removal of Chromium from aqueous water has been investigated using adsorbents based on bagasse and coconut jute and is found to be in general most effective at low pH values (<2) (Shri Chand, Agarval and Pavankumar, 1994).

2.4.2. SOME STRIKING FEATURES OF ACTIVATED CARBON ADSORPTION PROCESS

i. The efficiency of hexavalent Chromium removal was enhanced by increasing carbon dosages and hydrogen ion concentrations. The initial stage of Cr (VI) species. The later stage of Cr (VI) removal was very slow since only adsorption occured and hydrogen ion concentration had been reduced by the reduction and the hydrolytic reactions. The magnitude of total Cr removal was decreased by the production of Cr 3, however the reduction mechanism may be eliminated and the efficiency of total Chromium removal was greatly magnified (Huang et al, 1978).

ii. Fibrous adsorbents provided much faster adsorption Kinetics compared with granular adsorbents normally used because of its small diffusion distance in the fibre. **(Motoyuki Suzuki, 1991)**.

iii. Activated carbon obtained by corbonisation with sulphuric acid alone exhibited considerable hardness and bulk density which after thermal activation was found to show high Cr (VI) removal **(Periasamy et al, 1991)**.

iv. Cr (VI) interaction with activated carbon is a complicated phenomenm, it involves reduction of the Cr (VI) into Cr (III), adsorption of Cr (VI) **(C.P. Huang & A.R. Bowers, 1978)**.

v. The process of adsorption of Chromium on activated charcoal is highly pH dependant. The amount of Chromium reduced over activated charcoal is maximum at pH 3. The percentage of reduction is high at low pH values and gradually decreases as the pH increases. This is attributed to the large number of H^+ ions present at low pH values which can neutralise the negatively charged surface or convert a neutral group to a positively charged group. The enhancement of the adsorption of Cr (VI) species is because of the decrease in the forces of repulsion between adsorbent and adsorbate. **(Narayana Nagesh & Abburi Krishanaiah)**.

vi. According to **Dhirendra B.Sigh et al, 1992,** the uptake of Cr (VI) from water by feldspar increased with time and the maximum removal was noted at a pH of 2.5 and temperature 40° C for an intial concentration of Cr (VI) 19.23 lmoldm^{-3} while low removals were noted at higher inital concentration. The removal curves were single, smooth and continuous which indicated the formation of monolayer coverage on the outer surface of the adsorbent.

vii. The adsorption of Cr (VI) from a solution of concentration 96.15 lmoldm 3 was increased from 71.68 to 80.36% with a decrease in the particle size from 200 to 125 μm at 40c. The enhanced removal of Cr (VI) with decrease of particle size of the adsorbent may be attributed to the increased surface area **(Dhirendra Singh et al, 1992).**

viii. The adsorption of Cr (VI) on tea leaves carbon increased with the decrease in pH of the solution. The optimum pH was found to be 2. The study of concentration effect revealed that adsorption capacity increased upto 5X10 3 M adsorbate concentration and then became a constant. **(D.K. Singh et al, 1992).**

ix. According to **Grover et al, 1982** when the logarithm of percentage removal of Chromium (VI) was plotted against the logarithm of the fly ash concentration the graph was found to be linear in nature. This relationship showed that as the fly ash dosage increased, the removal of Chromium (VI) also increased.

x. According to **Grover et al, 1982** an increase in Chromium (VI) concentration in the aqueous solution was found to bring about a reduction in percentage removal.

xi. According to **Grover et al, 1982**the kinetic sudies revealed that the adsorption is probably due to pore diffusion which was often rate limiting in the case of batch reactions.

xii. According to **Singh & Lal, 1993** the used tea leaves, a waste, which was converted into carbonaceous material exhibited exceptionally high degree of Chromium (VI) adsorption with a peak value at a pH of 2.

xiii. According to **D.B. Singh et al, 1992** the removal followed first order kinetics and the process was found to involve both film and pore diffusion. The diffusion process was found to be fast and the rate was relatively unirfluenced by diffusion.

xiv. According to **Srinivasan et al, 1988** initial studies with rice husk carbons proposed by pyrolysis, carbonisation in the presence of metallic chlorides, sulphates, carbonates and acids, showed that the activated carbon obtained by carbonisation with sulphuric acid alone exhibited considerable hardness bulk desity which, after activation in Co_2 atmosphere, showed high Chromium removal.

xv. According to **Srinivasan et al, 1988** at optimum conditions (Ph 2.5-3) bichromate ion was the most predominant species that gets adsorbed on the carbon because the carbon was found to high Zero point charge and hence the greater affinity for the uptake of anionic species such as bichromate rather than Cr III ion.

xvi. According to **Periaswamy et al, 1991**, the Kinetics of Cr (VI) removal by ground nut husk carbon followed a reversible first order kinetics where a single species was considered on a heterogenous surface. The overall rate constant also increased as concentration of Cr (VI) increased. Because of this, the amount of Cr (VI) getting reduced under a constant pH conditon increased due to the increase in the available carbon surface.

xvii. According to **Shri.Chand et al, 1994** thermodynamic parameters like heat of adsorption and energy of activation play an inmportant role in predicting the adsorption behaviour and both are strongly temperature dependent. It was observed that the increase in temperature leads to a linear decrease in the percent Chromium (VI) removal. The phenomena of decrease in activity with temperature rise may be explained on the basis of the overall effects by the temperature of the Cr (VI) adsorbent bond, the water adsorbent bond, the Cr (VI) -

Cr (VI) interactions and the water - Cr (VI) interactions. The water adsorbent bond is important because adsorption of Cr (VI) probably involves displacement of H_2O molecules from the adsorbent surface. Therefore it may be inferred that at higher temperature the increased adsorption could be due to weakening of either of these bonds.

xviii. **Huang and Wu (1975)** reported that maximum removal was at pH 2 using calcinated coke.

xix. **Manju et al., (1997)** inferred that adsorption is greater at lower temperatures.

2.5. RECOVERY PROCESS

i. According to **Manju et al, 1997** only 3.3% desorption took place in distilled water whereas 86.4% desorption was observed with 1m NaOH. The higher percentage desorption at higher pH may be due to the presence of large OH-ion concentration which released Cr (VI) ions from the sorbent surface to the solution. After two cycles the adsorption capacity decreased to 87.2% where as the recovery decreased to 74.6% in third cycle. The reduction in adsorption may be due to the decrease in the weight of the sorbent as a consequence of alkali treatment.

ii. According to **Srinivasan et al, 1988** the maximum desorption occured with IM solution of NaOH, the adsorbent being Rice husk carbon, with this solution the average recovery of Cr (VI) was found to be 30.6% for CAC and 22.5% for RHC. Acid regeneration of both carbons mainly resulted in the formation of Cr (III). Chromium (III) could be recovered only to an extend of 40% from both carbons when 2 M HCl was used for desorption purpose. However when regenerated with alkali followed by acid the total Chromium removal was maximum and was found to be 60% from rice husk carbon and 82% from CAC.

iii. According to **Singh & Lal, 1993** when tea leaves carbon was used, 3 M NH4OH was an important desorbing agent for Cr (VI). The average recovery of Cr (VI) was found to be 85%. The less in adsorption efficiency of the column was made up by the treatment of the same with 4 M HNO_3 This stored in ulmost to 90% adsorption effieincy of original state.

Materials and Methods

III METHODS & MATERIALS

In the present work removal of Chromium (VI) by adsorption technique has been studied. The adsorbents used were activated rice husk carbon and activated waste tea leaves carbon. The studies are compared with that of commercially activated charcoal obtained from Sarabai Chemicals Limited Baroda.

The raw materials rice husk was collected from a rice mill and the waste tea leaves were collcted from a near by bakery.

3.1. PREPARATION OF ADSORBENTS.

3.1.1 Preparation of activated rice husk carbon:-

One part by weight of raw rice husk was treated with one part by weight of concentrated suphuric acid and kept at 150 C in an oven for 24 hours. The carbonised material was washed well with water to remove the free acid and dried at 105-110°c for 24 hours. The adsorbent was gorund and sieved through a 250 mesh.

3.1.2. Preparation of activated tea leaves carbon:-

One part of weight of waste tea leaves which was washed well and dried previously, was treated with one part by weight of concentrated suphuric acid and kept at 150°c in an oven for 24 hours. The carbonised material was washed well with water to remove the free acie and dried at 105-110°c for 24 hours. The adsorbent was ground and sieved through a 250 mesh.

3.2. REAGENTS

All reagents used were of AR grade. Synthetic effluent was prepared by dissolving potassium dichromate in distilled demineralized water (141.4 mg of potassium dichromate in 1 litre of water). Complexing reagent was prepared by dissolving 250 mg of diphenyl carbazide in 50 ml of acetone. In order to assess the performance of each adsorbent and to avoid interferance by other elements in the waste water the experiments were conducted with aqueous solution of hexavalent Chromium in redistilled water.

3.3. EQUIPMENTS

pH measurements were made with Digital pen type pH meter (Henna). Digital systronics colorimeter 112 was used for spectro colorimetric work.

3.4. BATCH EXPERIMENTS

To study the adsorption capacities of these adsorbents batch experiments were carried out. Though industrial operations are not carried out batch wise these are simpler and effective in evaluating the basic parameters affecting the adsorption.

3.5. PROCEDURE

3.5.1. EFFECT OF ADSORBENT DOSAGE:-

From the synthetic effluent standard solution was prepared by pipetting out 10 ml and diluting if to 100 ml. From this 46 ml was pipetted our acconting to 230 ppm of Chromium was made upto 100 ml and were contacted with adsorbent at a pH of 2. The system was equilibrated thoroughly on a horizontal bench shaker at room temperature for 1 hour. The weight for the adsorbent was varied from 150-450 mg for all the three adsorbents. After adsorption the suspension was filtered through a whatman number 42 filter paper. The filterate was analysed colorimetrically using filter number 5 (540 nm) by complexing with diphenyl carbazide for residual Chromium (VI) concentration in the solution. From the readings measured in terms of optical density the concentration of the remaining Chromium level was found out and it was taken along y axis and plotted against the concentration of the adsorbents taken along the x axis. The curve obtained gave the effect of adsorbent dosage on the adsorption of Chromium (VI).

3.5.2. EFFECT OF pH:-

The effect of pH was studied by varying the pH from 0.5-10.5. The pH of the solution was adjusted using dilute hydrochloric acid & sodium hydroxide and measured using Digital pen type pH meter. The optimum dosage of adsorbent was found out from the curve obtained during the effect of adsorbent dosage and this adsorbent dosage is used for finding out the effect of pH. 100 ml samples containing 230 ppm of Chromium were contacted with the optimum dosage and the pH of the solution was altered and shaken for 1 hour. The solution was filtered and measured colorimetrically for the optical densities of the solution and the concentration so obtained were plotted against pH on the x-axis and a curve is got for the effect of pH on the adsorption process.

3.5.3. EFFECT OF TIME:-

The effect of time was studied by varying the shaking time. 100 ml of samples of 230 ppm Chromium containing the optimum adsorbent dosage, whose pH was adjusted to the optimum pH level, were shaken for a time interval of 15 minutes, 30 minutes, 45, 60, 75, 90 and 105 minutes. The suspension was filtered through a whatman filter paper number 42 and analysed colorimetrically. From the value the concentration was found out and plotted against time in x axis to know about the effect of time on adsorption of Chromium.

3.5.4. EFFECT OF TEMPERATURE:-

100 ml of samples containing 230 ppm of Chromium with an optimum adsorbent dosage were adjusted to optimum pH level. These samples were than shaken for 1 hour in a temperature controlled shaker (NEOLAB SHAKING BATH). The temperature was altered from 30-70°c. The effect of temperature was found out from the curve obtained from the plotting of temperature in °C along x axis versus the concentration of residual Chromium along the y axis.

3.6. DESORPTION

Recovery of the adsorbate material as well as the metal for further use is an important process in waste water treatment. For this desorption studies were carried out. Desorption was done using 2 M Na OH for CAC & RHC. 3M NH4OH was used for TLC.

3.7. KINETIC STUDIES

To interpret the kinetics of the reaction, the following equations were studied.

1. Langmuir
2. Freundlich
3. Lagergren equation and intraparticle diffusion model for first order kinetics

Results And Discussion

4. RESULTS & DISCUSSION

In the present investigation, locally available economical adsorbents namely rice husk and waste tea leaves have been used and their adsorption potential is compared with that of commercially available carbon. Prior to adsorption studies, the adsorption potential of the two adsorbents were enhanced by activation with concentrated sulphuric acid.

The aftermaths of the investigation have been tabulated and discussed below.

4.1. EFFECT OF ADSORBENT DOSAGE

4.1.1. ADSORPTION STUDIES

Batch adsorption studies were carried out at room temperature to determine the effect of variation of adsorbent dosage of the three adsorbents, on the effective removal of Chromium (VI) at a given sorbate concentration (230, ppm) pH 2 and contact time 60 minutes. In order to strike a balance between recovery and removal of Chromium (VI) a pH of 2 was maintained in the study.

The results have been tabulated in Tables 1 to 3. It is evident from the tables that adsorption potential of the three adsorbents increases with increase in the dosage from 150-240 mg.

Figure 1 is a plot of dosage of adsorbent and adsorption potentiality. It is well evident from the figure that adsorption potential of CAC is higher than the two bio adsorbents among which RHC has a higher adsorption potential than TLC.

The higher efficiency of CAC when compared to the two bioadsorbents may be attributed to the greater surface area of CAC which makes available more number of adsorption sites and hence higher Chromium (VI) removal. A maximum Chromium (VI) removal at pH 2 using calcinated coke (Huang and Wu 1975) and a higher adsorption removal by activated carbon due to its greater surface area were reported by Tanada et.al (1990).

It was observed from the study that the optimum adsorbent dosage for the maximum removal of Chromium (VI) is 450 mg.

4.1.2. DESORPTION STUDIES

Batch desorption studies were conducted with the three adsorbents at the same conditions of pH, temperature and contact time as in the case of adsorption studies. The initial concentration varied with regard to the corresponding amounts adsorbed. The results have been tabulated in Tables 4 to 6 and graphically represented in figure 2.

It is well evident from the tables that desorption which is recovery of Chromium (VI) from the adsorbent is high with bioadsorbents than the CAC.

Among the bioadsorbents RHC was found to have a comparitively higher desorption potential than TLC. In general the desorption potential of the three adsorbents varied from 69-77%. It ws noted from the study that optimum dosage for maximum Chromium recovery from the adsorbents varies from 150-200 mg.

4.2. EFFECT OF pH VARIATION ON ADSORPTION/DESORPTION OF CHROMIUM (VI) TO/FROM ADSORBENTS

The pH of the aqueous solution is an important controlling parameter in the adsorption process and thus the role of Hydrogen ion concentration was examined from solution at different pH covering a range of 0.5-10.5. The results have been depicted in Table 7.

The results indicate that the process of adsorption of Chromium on the activated adsorbents is highly pH dependent. Also it is well evident from Table 7 that adsorption is efficient at low pH as was also noticed by Periasamy et al (1991).

The variation of adsorption of Chromium with pH is graphically shown is figure 3.

From the figure it is very vivid that the adsorption of Chromium for all the three adsorbents is maximum at pH 2.5 with 300 mg of adsorbents. The percentage of adsorption is high at low pH which may be attributed to the large number of Hydrogen ions, present at low pH values, which can neutralise the negatively charged group.

A gradual decreasein adsorption was noted as the pH was increased and this may be possibly due to increased solubility of Chromium (VI) and the abundance of hydroxyl ions, therby increasing hinderance to diffusion of dichromate ions.

The enhancement of the adsorption of Chromium (VI) is because of the decrease in the force of repulsion between the adsorbent and the adsorbate. The phenomenon has also been reported by Weber and Morris for the adsorption of sulphonated alkyl bonzenes. This phenomenon was also noticed by Kannan and Vanangamudi (1991).

From the present study it has been observed that the optimum pH at which maximum removal of Chromium (VI) is at a pH of 2.5 for all the three adsorbents. However, it must be remembered that under low pH conditions all the Chromium (VI) species would be converted into dichromate ions (Huang C.P and Wu,M.H., 1975). Because of this the oxidising ability of Chromium (VI) over carbon is increased. Due to this, high reduction process on the carbon surface may be occuring under low pH conditions.

In the case of desorption studies of Chromium (VI) from the adsorbents at varying pH and initial concentration (Table 8) It was noted that the desorption potential increased as the pH was decreased and as initial concentration was decreased. Since in the present study the initial concentration also is decreasing while the pH is increased, the effect of pH alone on desorption cannot be arrived at.

The variation of desorption of Chromium with pH is graphically shown in figure 4.

The recovery of Chromium using CAC and RHC was found to be comparably same which implies that efficient removal of Chromium (VI) from the adsorbents can be achieved at lower pH and lower concentration as was noted by Desh Deepak and Ajay Kumar Gupta (1991).

4.3. TIME STUDIES

Th dependance of percentage removal of Chromium (VI) on time has also been studied. The results have been portrayed in Table 9 and graphically depicted in figure 5.

The trend of removal curve as from figure (5) is found to be linear.

The time taken for reaching the equilibrium as determined from the figure is 105 minutes at fixed initial concentration and pH2. The removal curves are single, smooth and continuous, indicating the formation of monolayer coverage on the outer surface of adsorbent (Pandey et al, 1984).

It is well evident from the tables that the efficiency of adsorption is increased the contact time. Thus on varying the contact time from 15-105 minutes, it was seen that the percentage adsorption increased from 60-99% in the case of all the three adsorbents.

CAC was found to be more efficient than bioadsorbents when the percentage removal at equilibrium time was taken for comparison.

Among the bioadsorbents RHC was found to be more efficient than TLC. The optimum time for the three asorbent for maximum removal of Chromium (VI) was found to be 105 minutes.

In the case of desorption studies at varying contact time ranging from 15-105 minutes at fixed pH and varying initial concentration it was observed that as the contact time was increased the desorption potential decreased.

This implies that maximum recovery of Chromium (VI) from the adsorbents can be achieved within the first few minutes itself.

Also recovery is found to be comparably equal between the bioadsorbents and CAC.

Thus maximum removal of Chromium from adsorbents which is a significant factor in waste water treatment plants can be achieved efficiently with economical bioadsorbents within few minutes (15-30 min). The results of desorption studies are depicted in table 10 and graphically represented in figure 6.

4.4. EFFECT OF TEMPERATURE

Temperature affects the adsorption rate by altering the molecular interactions and the solubility. The uptake of Chromium (VI) in the present study was found to increase as the temperature was raised from 30°C to40°C after which there is a gradual decline in adsorption potential as the temperature is decreased to 70°C (Table - 11).

The results are graphically portrayed in figure 7.

Although increased adsorption at higher temperature is difficult to explain, many workers have reported increased adsorption with increasing temperature (Pandey et al., 1986 and 1989).

The removal of Chromium (VI) from the synthetic effluent is found to be efficient at a optimum temperature of 40° C for all the three adsorbents. The enhanced adsorption might be due to adsorption of Chromium (VI) due to change in size of pores and enhanced rate of intra particular diffusion.

The decrease in adsorption with increase in temperature from 40° - 70°Cmay be due to weakening of adsorptive force between active sites of the adsorbents and the adsorbate species and also between the adjacent molecules of adsorbed phase.

The results obtained in the present study conform to those noticed by Mohammad Ajmal et al (1998).

Comparing the adsorption potential, of the three adsorbents with temperature variation it is seen that CAC has higher adsorption potential (95-82%) than bioadsorbents among which RHC is more efficient (93-79%) than TLC (90-76%).

4.5. KINETIC STUDIES

In the present investigation the following two approaches have been used for explaining the adsorption kinetics

- Kinetic modelling using Lagergren equation
- Weber & Morris approach for intraparticle diffusion rate constant.

4.5.1. KINETIC MODELLING USING LAGERGREN EQUATION

The rate constant of adsorption were calculated (Table 12) using the following model proeposed by Lagergren for a 1st order rate process.

$$\text{Log}\ (qe-q) = \log qe - ka/2.303\ t$$

Where,

q&qe are the amount of Chromium (VI) adsorbed at time t & at equilibrium. Ka is the rate constant for adsorption studies which is evaluated from the slope of linear plot ie, log qe-q Vs t shows the validity of Lagergren equation & suggets that removal of Chromium (VI) obeys Ist order kinetics. The rate constants Ka for the three adsorbents are given in the table below. It is seen from this table that the rate constant for CAC is greater than that of bioadsorbents among which RHC is found to be more efficient than TLC.

Adsorbents	Ka
CAC	-0.01124
RHC	-0.01167
TLC	-0.0163

In a similar manner the rate constant for the backward reaction were found using Lagergren's equation. The rate constants Kd are tabulated below for the three adsorbents. The linearity of the plot of log (qe-q) Vs t shows the applicability of Lagergren equation to the desorption studies which is hence obeying Ist order kinetics (figure 9).

Adsorbents	Kd
CAC	-0.01354
RHC	-0.01372
TLC	-0.01761

From this it is evident that the rate of desorption is higher for CAC than that of bioadsorbents. Among bioadsorbents the recovery of Chromium (VI) back from the adsorbents is found to be higher with RHC than with TLC.

4.5.2. WEBER AND MORRIS APPROACH FOR INTRAPARTICLE DIFFUSION RATE CONSTANT

A functional relationship commonly to describe the intraparticle transport is the plot of mass of solute adsorbed versus square root of contact time which is linear if the controlling mechanism for adsorption is intraparticle diffusion (Weber & Morris 1989).

The adsorption and desorption data was tested in the light of the following equation to ascertain the role of pore diffusion

$$\Delta q = k \times t^{n}$$

$$\log \Delta q = \log k + n \log t$$

Where Δq is the percentage removal

t is the mixing time

and k is a constant

The plots of Chromium (VI) adsorbed & desorbed versus square root of time were linear over a wide range of contact period for all the three adsorbents indicating that controlling step is intraparticle diffusion. A typical representation of intraparticle diffusion plot for all the three adsorbents is given in figure 10.

According to Weber and Morris a value of $n = 0.5$ implies pore diffusion to be the rate controlling step. Here the value of $n < 0.5$ indicated that the process is not purely pore diffusion controlled.

Process	Adsorbents	n
Adsorption	CAC	0.1837
	RHC	0.2020
	TLC	0.2455
Desorption	CAC	- 0.2218
	RHC	- 0.1642
	TLC	- 0.2073

4.6. ADSORPTION ISOTHERMS

Adsorption equilibrium data which expresses the relationship between mass of adsorbate adsorbed per unit weight of adsorbent and liquid phase equilibrium concentration of adsorbate are represented by adsorption isotherms and provide important design data for adsorption system.

The equilibrium data for removal of Chromium (VI) in the present investigation were analysed using Langmuir & Freundlich Isotherms.

4.6.1. LANGMUIR ADSORPTION ISOTHERMS

Langmuir Adsorption isotherm is based on the assumption that points of valency exist on the surface of the adsorbent and that each of these sites is capable of adsorbing one molecule. Thus the adsorbed layer, will be one molecule thick. Further it is assumed that all the adsorption sites have equal affinities for molecules of the adsorbant and that the presence of adsorbed molecules at once site will not affect the adsorption of molecules at an adjacent site.

The Langmuir adsorption isotherm is commonly given by

$$x/m = \frac{K_{,}' \, Ce}{1 + K_{,}Ce}$$

x = amount of Chromium (VI) adsorbed (ppm)

m = Weight of adsorbent (mg)

Ce = Concentration of Cr at equilibrium

K_1' & K_1 = Constants

on rearranging

$$\frac{1}{x/m} = \frac{1}{K_1'/K_1} + \frac{1}{K_1' \, Ce}$$

The plot of (1/x/m) versus 1/Ce is linear with slope equal to $1/K_1'$ and intercept $1/(K_1'/K_1)$.

The slope, intercept and the regression coefficients for CAC and bioadsorbents were calculated, from the linear regression for adsorption and desorption studies.

The validity of the Langmuir adsorption isotherm was verified by linear regression coefficients (Table 24).

The values are presented in tables 18,19 and 20 for the three adsorbents. The linear plots (Figure 11) shows the applicability of Langmuir model for the present system indicating the formation of monolayer coverage of adsorbate on the surface of the adsorbent.

Attempts were also made to fit the desorption data (Table 21, 22, 23) into the Langmuir isotherm. The linear plots show the applicability of the Langmuir model to the system (Figure 12).

4.6.2. FREUNDLICH ADSORPTION ISOTHERM

The equilibrium data obtained at different carbon dosage can be explained on the basis of the well known Freundlich adsorption isotherm

$$x / m = K_k Ce^{1/n}$$

Where

x/m is the amount of Cr (VI) adsorbed on unit weight of adsorbent in equilibrium with Ce, the concentration of solute in aqueous solution.

The freundlich parameters K and 1/n are indicators of adsorption capacity and intensity respectively. The values have been incorporated in Tables 25,26 and 27. The slopes, and intercepts have been evaluated from the (Figure 13). The value of K for the 3 adsorbents have been tabulated in Table 18.

Since 1/n is less than 1, it indicated favourable adsorption.

The Freundlich equation is better obeyed by the system than the Langmuir equation as it is evident from the values of regression coefficients shown in Table 28.

The values of Freundlich constant is high for CAC than the bioaadsorbents which indicates that Cr (VI) is strongly adsorbed on the adsorbent.

The ultimate adsorption capacity of the adsorbent can be calculated by substituting the required equilibrium concentration in freundlich equation.

Thus for an equilibrium concentration of 1 mg/l of Chromium (VI) 1 g of adsorbent can remove 506 micro gram Cr (VI) at 30 C with CAC adsorbents 502 micro gram with RHC and with TLC 497 micro gram.

Attempts were also made to fit the desorption data into Freundlich isotherm (Tables 29, 30 & 31). The linear plot shows the applicability of the Freundlich model to the system (Figure 14).

TABLE - 1

ADSORPTION POTENTIAL OF Cr VI WITH DOSAGE VARIATION OF CAC

CONDITION: `m' mg of CAC shaken with 100 ml of synthetic effluent containing a Cr (VI) concentration of 230ppm at room temperature 32 ± 1°C
pH - 2 Contact time - 60 minutes

S.No.	Initial Cr VI concentration Co ppm	Wt of adsorbent	Chromium (VI) adsorbed	Ce	Adsorption potential %
1.	230	150	190	40	82.6
2.	230	200	195	35	84.7
3.	230	250	210	20	91.3
4.	230	300	217	13	94.3
5.	230	350	222	8	96.5
6.	230	400	227	3	98.6
7.	230	450	228	2	99.1

TABLE - 2

ADSORPTION POTENTIAL OF Cr VI WITH DOSAGE VARIATION OF RHC

CONDITION: `m' mg of RHC shaken with 100 ml of synthetic effluent containing a Cr (VI) concentration of 230ppm at room temperature 32 ± 1°C
pH - 2 Contact time - 60 minutes

S.No.	Initial Cr VI concentration Co ppm	Wt of adsorbent	Chromium (VI) adsorbed	Ce	Adsorption potential %
1.	230	150	180	50	78
2.	230	200	190	40	82.6
3.	230	250	204	26	88.6
4.	230	300	212	18	92
5.	230	350	218	12	94
6.	230	400	225	5	97
7.	230	450	226	4	98.2

TABLE - 3

ADSORPTION POTENTIAL OF Cr VI WITH DOSAGE VARIATION OF TLC

CONDITION: `m' mg of TLC shaken with 100 ml of synthetic effluent containing a Cr (VI) concentration of 230ppm at room temperature 32 ± 1°C
pH - 2 Contact time - 60 minutes

S.No.	Initial Cr VI concentration Co ppm	Wt of adsorbent	Chromium (VI) adsorbed	Ce	Adsorption potential %
1.	230	150	175	55	76
2.	230	200	188	42	81.7
3.	230	250	197	33	85.6
4.	230	300	209	21	90.8
5.	230	350	215	15	93.4
6.	230	400	222	8	96.5
7.	230	450	224	6	97.3

TABLE - 4

DESORPTION POTENTIAL OF Cr VI WITH DOSAGE VARIATION OF CAC

CONDITION : `m' mg of CAC shaken with 50 ml of 2 M Sodium hydroxide containing varied concentration Co at room temperature 32 ± 1°C
pH - 2 Contact time - 60 minutes

S.No.	Initial Cr VI concentration Co ppm	Wt of adsorbent	Chromium (VI) desorbed	Ce	Desorption potential %
1.	190	150	148	42	77.8
2.	195	200	149	46	76.4
3.	210	250	150	60	71.4
4.	217	300	157	60	72.3
5.	222	350	160	62	72.07
6.	227	400	165	62	72.6
7.	228	450	166	62	72.8

TABLE - 5

DESORPTION POTENTIAL OF Cr VI WITH DOSAGE VARIATION OF RHC

CONDITION: 'm' mg of RHC shaken with 50 ml of 2 M Sodium hydroxide containing varied concentration Co at room temperature 32 ± 1°C
pH - 2 Contact time - 60 minutes

S.No.	Initial Cr VI concentration Co ppm	Wt of adsorbent	Chromium (VI) desorbed	Ce	Desorption potential %
1.	180	150	125	55	69.4
2.	190	200	138	52	72.6
3.	204	250	140	64	68.6
4.	212	300	143	69	67.4
5.	218	350	148	70	67.8
6.	225	400	152	73	67.5
7.	226	450	157	69	69.45

TABLE - 6

DESORPTION POTENTIAL OF Cr VI WITH DOSAGE VARIATION OF TLC

CONDITION: 'm' mg of TLC shaken with 50 ml of 3 M Ammonium hydroxide containing varied concentration Co at room temperature 32 ± 1°C
pH - 2 Contact time - 60 minutes

S.No.	Initial Cr VI concentration Co ppm	Wt of adsorbent	Chromium (VI) desorbed	Ce	Desorption potential %
1.	175	150	120	55	68.57
2.	188	200	125	63	66.48
3.	197	250	130	67	65.98
4.	209	300	135	74	64.57
5.	215	350	140	75	65.1
6.	222	400	145	77	65.3
7.	224	450	147	77	65.6

TABLE - 7

ADSORPTION POTENTIAL OF CHROMIUM VI WITH pH VARIATION

CONDITION: Weight of adsorbent 300 mg, Initial concentration of Chromium VI 230 ppm, Temperature 32 ± 1°C
Contact time 60 minutes

S.No.	pH	CAC		RHC		TLC	
		Cr ads	Adsor Poten	Cr ads	Adsor Poten	Cr ads	Adsor Poten
1.	0.5	192	83.5	189	82.2	178	77.5
2.	2.5	210	91.3	205	89.1	200	87.0
3.	4.5	190	82.6	185	80.4	167	72.6
4.	6.5	182	79.1	178	77.4	150	65.2
5.	8.5	160	69.6	155	67.4	140	60.9
6.	10.5	140	60.9	135	58.7	126	54.8

TABLE - 8

DESORPTION POTENTIAL OF CHROMIUM VI WITH VARIATION OF pH

CONDITION: Weight of adsorbent 300 mg,
Temperature 32 ± 1°C
Contact time 60 minutes

S.No.	pH	CAC		RHC		TLC	
		Cr des	Desor Poten	Cr des	Desor Poten	Cr des	Desor Poten
1.	0.5	138	71.88	129	68.25	95	53.37
2.	2.5	150	71.43	138	67.32	107	53.5
3.	4.5	147	77.37	118	63.78	88	52.69
4.	6.5	130	71.43	102	57.3	79	52.67
5.	8.5	102	63.75	89	57.44	75	53.57
6.	10.5	82	58.57	77	57.05	60	47.62

TABLE - 9

ADSORPTION POTENTIAL OF CHROMIUM VI WITH VARIATION OF CONTACT TIME

CONDITION: Weight of adsorbent 300 mg, Initial concentration of Chromium VI 230 ppm, Temperature 32 ± 1 °C pH 2

S.No.	Time in minutes	CAC		RHC		TLC	
		Cr ads	Adsor Poten	Cr ads	Adsor Poten	Cr ads	Adsor Poten
1	15	160	69.6	150	65.2	135	58.7
2	30	178	77.4	170	73.9	150	65.2
3	45	197	85.7	180	78.3	160	69.6
4	60	205	89.2	198	86.1	177	77.0
5	75	215	93.5	200	87.0	188	81.74
6	90	220	95.7	215	93.5	210	91.3
7	105	228	99.1	223	97.0	215	93.5

TABLE - 10

DESORPTION POTENTIAL OF CHROMIUM VI WITH VARIATION OF CONTACT TIME

CONDITION: Weight of adsorbent 300 mg, Initial concentration of Chromium VI 230 ppm, Temperature 32 ± 1°C pH 2

S.No.	Time in minutes	CAC		RHC		TLC	
		Cr des	Desor Poten	Cr des	Desor Poten	Cr des	Desor Poten
1	15	101	63.1	87	58.0	70	51.8
2	30	108	60.6	95	55.8	80	53.3
3	45	142	72.08	124	68.8	85	53.12
4	60	147	71.7	135	68.18	122	68.1
5	75	155	72.0	137	68.5	125	66.4
6	90	159	72.2	143	66.5	135	64.2
7	105	166	72.8	150	67.2	138	64.18

TABLE - 11

ADSORPTION POTENTIAL OF CHROMIUM VI WITH TEMPERATURE VARIATION

CONDITION: Weight of adsorbent 300 mg, Initial concentration of Chromium VI 230 ppm, Contact time 60 minutes pH 2

S.No.	Temperature in degree °C	CAC		RHC		TLC	
		Cr ads	Adsor Poten	Cr ads	Adsor Poten	Cr ads	Adsor Poten
1	30	215	93.48	210	91.3	205	89.13
2	40	220	95.65	215	93.48	208	90.43
3	50	218	94.78	208	90.43	203	88.26
4	60	210	91.3	199	86.5	195	84.78
5	70	190	82.6	182	79.13	176	76.52

TABLE - 12

KINETIC MODELLING FOR ADSORPTION USING LAGERGREN EQUATION FOR CAC

S.No.	Time in min	Initial concentration	Chromium adsorbed (q)	qe - q	Log (qe - q)
1.	15	230	160	68	1.8325
2.	30	230	178	50	1.6990
3.	45	230	197	31	1.491
4.	60	230	205	23	1.3617
5.	75	230	215	13	1.1139
6.	90	230	220	8	0.9031
7.	105	230	228	0	-

TABLE - 13

KINETIC MODELLING FOR ADSORPTION USING LAGERGREN EQUATION FOR RHC

S.No.	Time in min	Initial concentration	Chromium adsorbed (q)	qe - q	Log (qe - q)
1.	15	230	150	73	1.8633
2.	30	230	170	53	1.7242
3.	45	230	180	43	1.6335
4.	60	230	198	25	1.3979
5.	75	230	200	23	1.3612
6.	90	230	215	8	0.9031
7.	105	230	223		-

TABLE - 14

KINETIC MODELLING FOR ADSORPTION USING LAGERGREN EQUATION FOR TLC

S.No.	Time in min	Initial concentration	Chromium adsorbed (q)	qe - q	Log (qe - q)
1.	15	230	135	80	1.9031
2.	30	230	150	65	1.8129
3.	45	230	160	55	1.7404
4.	60	230	177	38	1.5798
5.	75	230	188	27	1.4314
6.	90	230	210	5	0.6990
7.	105	230	215		-

TABLE - 15

KINETIC MODELLING FOR DESORPTION USING LAGERGREN EQUATION FOR CAC

S.No.	Time in min	Initial concentration	Chromium adsorbed (q)	qe - q	Log (qe - q)
1.	15	160	101	65	1.8129
2.	30	178	108	58	1.7634
3.	45	197	142	24	1.3802
4.	60	205	147	19	1.2788
5.	75	215	155	11	1.0414
6.	90	220	159	7	0.8451
7.	105	228	166	-	-

TABLE - 16

KINETIC MODELLING FOR DESORPTION USING LAGERGREN EQUATION FOR RHC

S.No.	Time in min	Initial concentration	Chromium adsorbed (q)	qe - q	Log (qe - q)
1.	15	150	87	63	1.7993
2.	30	170	95	55	1.7404
3.	45	180	124	26	1.415
4.	60	190	135	15	1.1761
5.	75	200	137	13	1.1139
6.	90	215	143	7	0.8451
7.	105	223	150	-	-

TABLE - 17

KINETIC MODELLING FOR DESORPTION USING LAGERGREN EQUATION FOR TLC

S.No.	Time in min	Initial concentration	Chromium adsorbed (q)	qe - q	Log (qe - q)
1.	15	135	70	68	1.8325
2.	30	150	80	58	1.7634
3.	45	160	85	53	1.7243
4.	60	177	122	16	1.2788
5.	75	188	125	13	1.1139
6.	90	210	135	3	0.4771
7.	105	215	138	-	-

TABLE - 18

REGRESSION TABLE FOR LANGMUIR ADSORPTION ISOTHERM FOR CAC

S.No.	M	Co	x	Ce	x/M	1/(x/M)	1/Ce	Y′
1.	150	230	190	40	1.26	0.78	0.020	1.095
2.	200	230	195	35	0.975	1.025	0.028	1.111
3.	250	230	210	20	0.84	1.190	0.05	1.156
4.	300	230	217	13	0.72	1.382	0.076	1.209
5.	350	230	222	8	0.634	1.570	0.125	1.308
6.	400	230	227	3	0.567	1.760	0.330	1.725
7.	450	223	228	2	0.506	1.970	0.500	2.070

TABLE - 19

REGRESSION TABLE FOR LANGMUIR ADSORPTION ISOTHERM FOR RHC

S.No.	M	Co	x	Ce	x/M	1/(x/M)	1/Ce	Y′
1.	150	230	180	50	1.2	0.83	0.02	1.095
2.	200	230	190	40	0.95	1.052	0.025	1.105
3.	250	230	204	26	0.816	1.225	0.038	1.131
4.	300	230	212	18	0.706	1.416	0.055	1.166
5.	350	230	218	12	0.622	1.607	0.083	1.223
6.	400	230	225	5	0.562	1.779	0.2	1.461
7.	450	223	226	4	0.502	1.992	0.25	1.562

M - Weight of adsorbent
Co - initial concentration of Chromium (VI) in ppm
x - Chromium adsorbed
Ce - chromium equivalance
y′ - Regressed value

TABLE - 20

REGRESSION TABLE FOR LANGMUIR ADSORPTION ISOTHERM FOR TLC

S.No.	M	Co	x	Ce	x/M	1/(x/M)	1/Ce	Y′
1.	150	230	175	55	1.167	0.857	0.018	1.10
2.	200	230	188	42	0.94	1.06	0.023	1.135
3.	250	230	197	33	0.78	1.28	0.030	1.185
4.	300	230	209	21	0.69	1.449	0.047	1.310
5.	350	230	215	15	0.61	1.639	0.066	1.398
6.	400	230	222	8	0.55	1.818	0.125	1.86
7.	450	223	224	6	0.49	2.040	0.16	2.10

TABLE - 21

REGRESSION TABLE FOR LANGMUIR DESORPTION ISOTHERM FOR CAC

S.No.	M	Co	x	Ce	x/M	1/(x/M)	1/Ce	Y′
1.	150	190	148	42	0.986	1.0135	0.023	1.0315
2.	200	195	149	46	0.745	1.342	0.217	1.256
3.	250	210	150	60	0.6	1.66	0.0166	2.138
4.	300	217	157	60	0.523	1.910	0.0166	2.1389
5.	350	222	160	62	0.457	2.187	0.0161	2.225
6.	400	227	165	62	0.412	2.42	0.0161	2.225
7.	450	228	166	62	0.368	2.71	0.0161	2.225

TABLE - 22

REGRESSION TABLE FOR LANGMUIR DESORPTION ISOTHERM FOR RHC

S.No.	M	Co	x	Ce	x/M	1/(x/M)	1/Ce	Y′
1.	150	180	125	55	0.83	1.2	0.0181	1.446
2.	200	190	138	52	0.69	1.449	0.0192	1.1729
3.	250	204	140	64	0.56	1.785	0.0156	2.069
4.	300	212	143	69	0.476	2.097	0.01449	2.346
5.	350	218	148	70	0.422	2.36	0.01428	2.398
6.	400	225	152	73	0.38	2.631	0.0136	2.567
7.	450	226	157	69	0.348	2.866	0.0144	2.368

TABLE - 23

REGRESSION TABLE FOR LANGMUIR DESORPTION ISOTHERM FOR TLC

S.No.	M	Co	x	Ce	x/M	1/(x/M)	1/Ce	Y′
1.	150	175	120	55	0.8	1.25	0.0181	1.061
2.	200	188	125	63	0.625	1.6	0.0158	1.778
3.	250	197	130	67	0.52	1.923	0.0149	2.057
4.	300	209	135	74	0.45	2.22	0.0135	2.496
5.	350	215	140	75	0.4	2.5	0.0133	2.558
6.	400	222	145	77	0.36	2.77	0.0129	2.683
7.	450	224	147	77	0.326	3.06	0.0129	2.683

TABLE - 24

VALUES OF THE LANGMUIR SLOPES INTERCEPTS AND CORRELATION COEFFICIENTS DURING ADSORPTION AND DESORPTION

Adsorption			Desorption			
Adsorbent	Intercept	Slope	Correlation Coefficient	Intercept	Slope	Correlation Coefficient
CAC	1.056	2.031	0.8890	5.011	-173.03	-0.859
RHC	1.024	4.062	0.907	5.955	-249.06	-0.871
TLC	0.972	7.10	0.9310	6.709	-312.04	-0.9396

TABLE - 25

REGRESSION TABLE FOR FREUNDLICH ADSORPTION ISOTHERM FOR CAC

S.No.	M	Co	x	Ce	x/M	log(x/M)	log Ce	Y′
1.	150	230	190	40	1.26	0.1004	1.6	0.02494
2.	200	230	195	35	0.975	-0.0110	1.54	-0.0092
3.	250	230	210	20	0.84	-0.0760	1.30	-0.05393
4.	300	230	217	13	0.72	-0.1427	1.113	-0.1031
5.	350	230	222	8	0.634	-0.1979	0.90	-0.1591
6.	400	230	227	3	0.567	-0.2464	0.477	-0.2703
7.	450	230	228	2	0.506	-0.2958	0.30	-0.3168

TABLE - 26

REGRESSION TABLE FOR FREUNDLICH ADSORPTION ISOTHERM FOR RHC

S.No.	M	Co	x	Ce	x/M	log(x/M)	log Ce	Y′
1.	150	230	180	50	1.2	0.079	1.69	0.0159
2.	200	230	190	40	0.95	-0.0223	1.60	-0.0112
3.	250	230	204	26	0.816	-0.0883	1.42	-0.06553
4.	300	230	212	18	0.7	-0.1549	1.25	-0.01168
5.	350	230	218	12	0.62	-0.2076	1.07	-0.1711
6.	400	230	225	5	0.56	-0.2518	0.69	-0.2857
7.	450	230	226	4	0.5	-0.3010	0.60	-0.3128

TABLE - 27

REGRESSION TABLE FOR FREUNDLICH ADSORPTION ISOTHERM FOR TLC

S.No.	M	Co	x	Ce	x/M	log(x/M)	log Ce	Y ′
1.	150	230	175	55	1.16	0.0644	1.74	0.0117
2.	200	230	188	42	0.94	-0.0269	1.62	-0.0305
3.	250	230	197	33	0.78	-0.1079	1.52	-0.0657
4.	300	230	209	21	0.69	-0.1612	1.32	-0.1361
5.	350	230	215	15	0.61	-0.2147	1.18	-0.1853
6.	400	230	222	8	0.55	-0.2596	0.90	-0.2838
7.	450	230	224	6	0.49	-0.3098	0.78	-0.3260

TABLE - 28

THE VALUES OF K, n AND r FROM FREUNDLICH ADSORPTION ISOTHERM

Adsorbent	Adsorption			Desorption		
	K	n	r	K	n	r
CAC	0.4020	3.8	0.9528	1.35x10	0.5149	-0.9189
RHC	0.3207	3.31	0.9586	4.85x10	0.4539	-0.9027
TLC	0.2510	2.84	0.9663	1.43x10	0.4108	-0.9752

TABLE - 29

REGRESSION TABLE FOR FREUNDLICH DESORPTION ISOTHERM FOR CAC

S.No.	M	Co	x	Ce	x/M	log(x/M)	log Ce	Y′
1.	150	190	148	42	0.9867	-0.0058	1.623	-0.0192
2.	200	195	149	46	0.7450	-0.1278	1.663	-0.0968
3.	250	210	150	60	0.6000	-0.2218	1.778	-0.3200
4.	300	217	157	60	0.5233	-0.2812	1.778	-0.3202
5.	350	222	160	62	0.4572	-0.3399	1.792	-0.3474
6.	400	227	165	62	0.4125	-0.3846	1.792	-0.3474
7.	450	228	166	62	0.3689	-0.4331	1.792	-0.3474

TABLE - 30

REGRESSION TABLE FOR FREUNDLICH DESORPTION ISOTHERM FOR RHC

S.No.	M	Co	x	Ce	x/M	log(x/M)	log Ce	Y′
1.	150	180	125	55	0.833	-0.0793	1.74	-0.1478
2.	200	190	138	52	0.69	-0.1611	1.716	-0.0949
3.	250	204	140	64	0.56	-0.2510	1.806	-0.2930
4.	300	212	143	69	0.476	-0.3220	1.838	-0.3654
5.	350	218	148	70	0.422	-0.3740	1.845	-0.3790
6.	400	225	152	73	0.38	-0.4200	1.863	-0.4194
7.	450	226	157	69	0.348	-0.4580	1.838	-0.3654

TABLE - 31

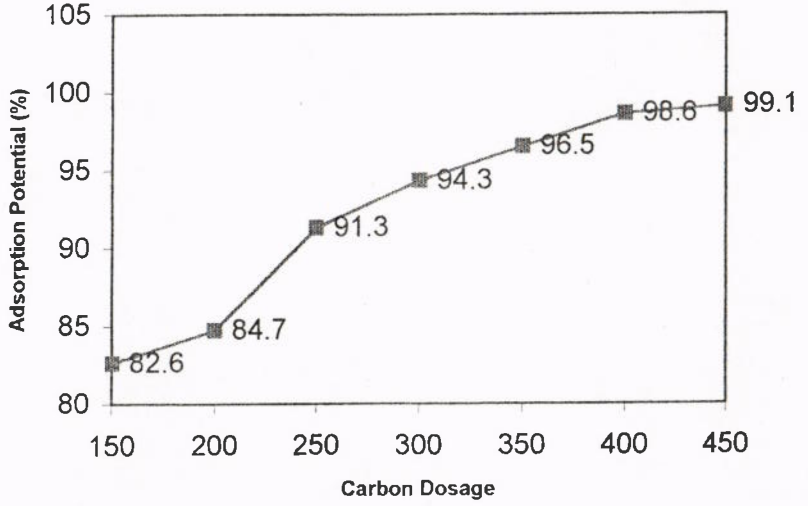
REGRESSION TABLE FOR FREUNDLICH DESORPTION ISOTHERM FOR TLC

S.No.	M	Co	x	Ce	x/M	log(x/M)	log Ce	Y '
1.	150	175	120	55	0.8	-0.0969	1.74	-0.0780
2.	200	188	125	63	0.625	-0.2041	1.799	-0.2219
3.	250	197	130	67	0.52	-0.2830	1.826	-0.2816
4.	300	209	135	74	0.45	-0.3467	1.869	-0.3920
5.	350	215	140	75	0.4	-0.3979	1.875	-0.4060
6.	400	222	145	77	0.362	-0.4412	1.886	-0.4330
7.	450	224	147	77	0.326	-0.4850	1.886	-0.4330

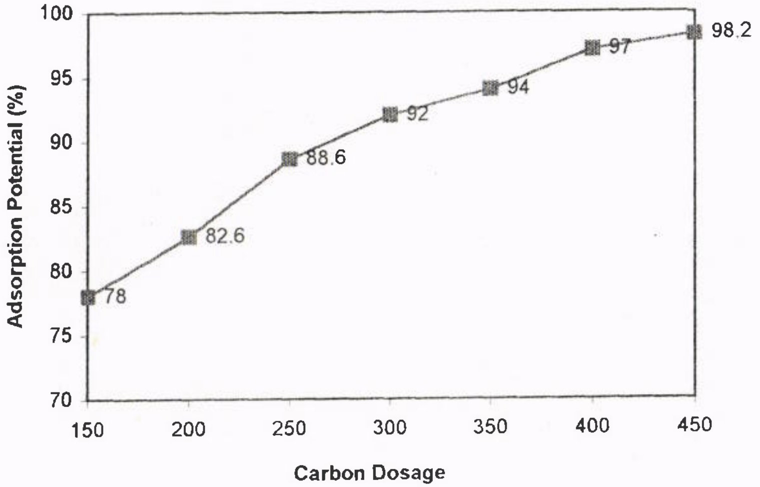
Figure - 1

Adsorption Potential of Chromium VI with Dosage Variation

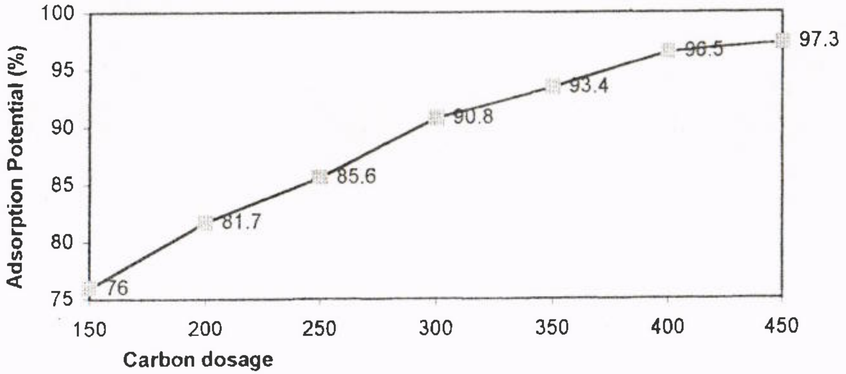
CAC



RHC



TLC



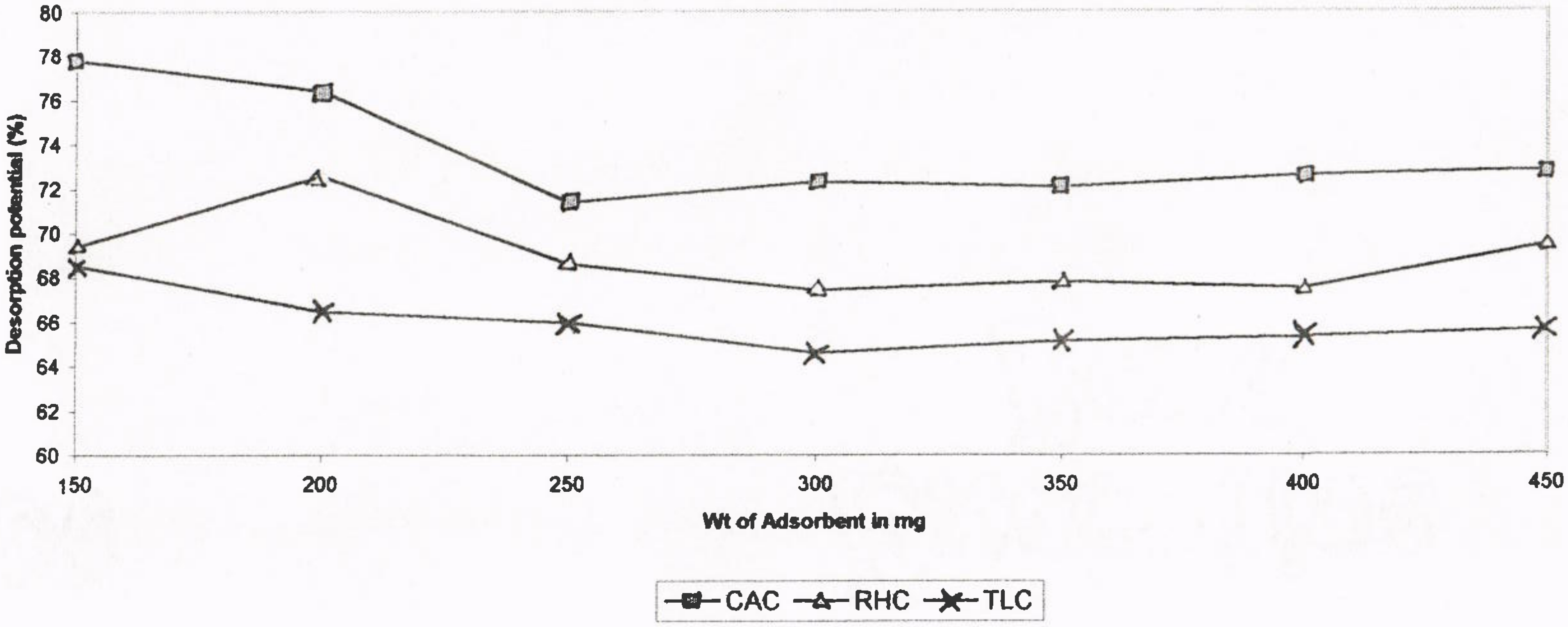
Condition - m mg of adsorbent shaken with 100 ml of synthetic effluent containing a chromium VI con. Of 230 ppm at room temprature 32 + /- 1 degree

pH - 2

Contact time - 60 minutes

Figure - 2

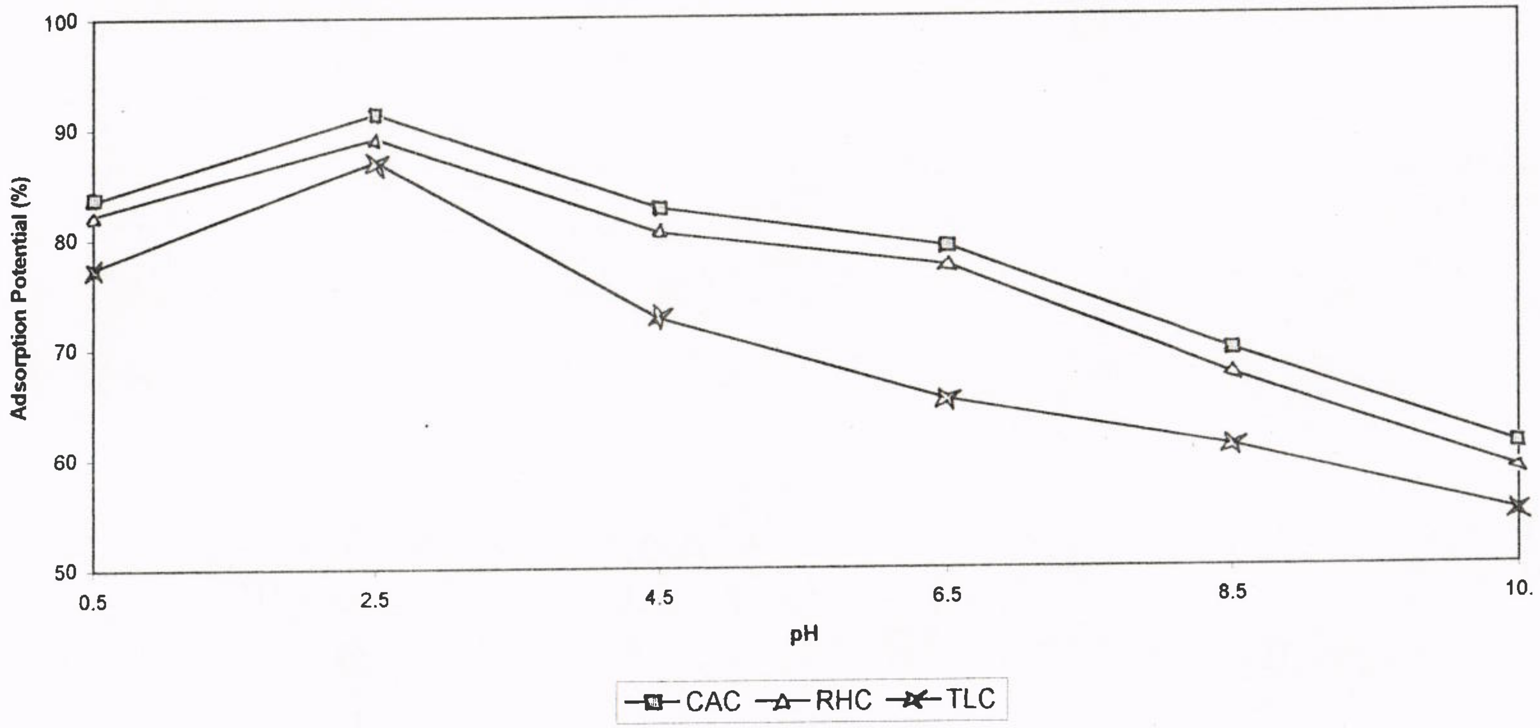
Desorption Potential of Cr VI with dosage variation of various Adsorbents



Conditions - pH - 2   Contact time - 60 min   Temperature - 32 + 0.1 C

Figure -3

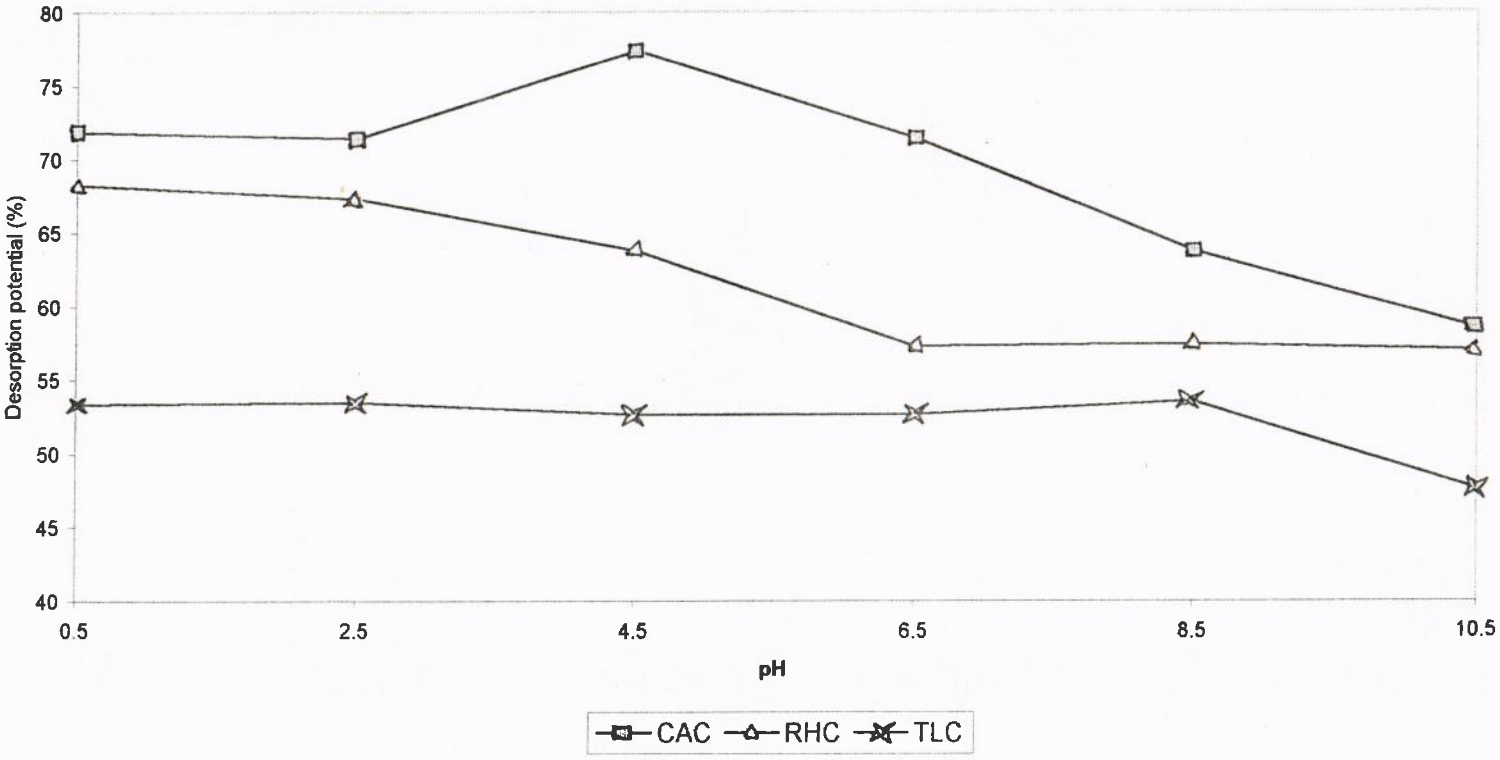
Adsorption Potential of Cr VI with pH variation



Condition - Wt of adsorbent - 300 mg   Initial Conc. Of Cr VI - 230 ppm   Temperature 32 + 0.1 C   Contact time - 60 min.

Figure - 4

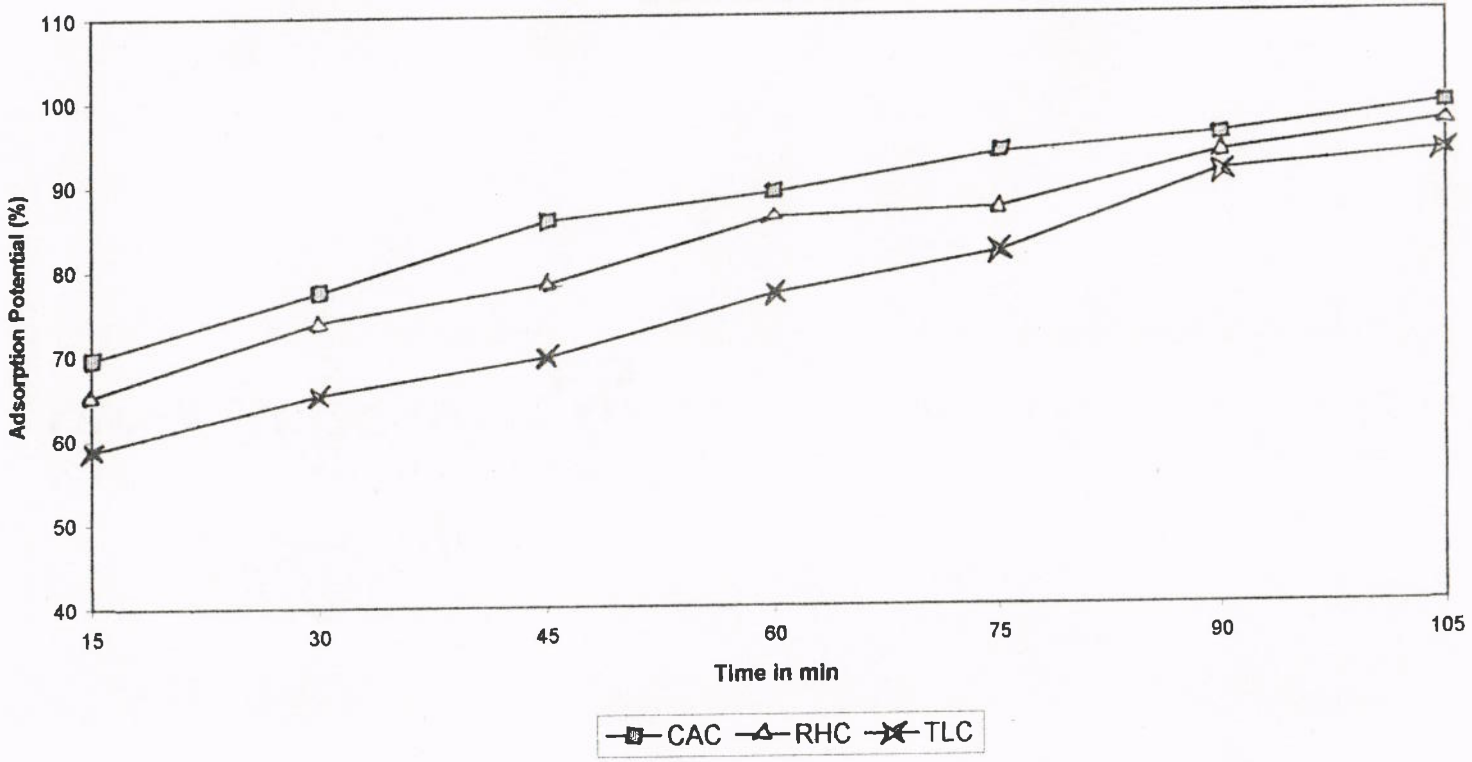
Desorption potential of Cr VI with pH variation



Condition - wt of adsorbent 300 mg Temperature - 32 + 0.1 C Contact time - 60 min

Figure - 5

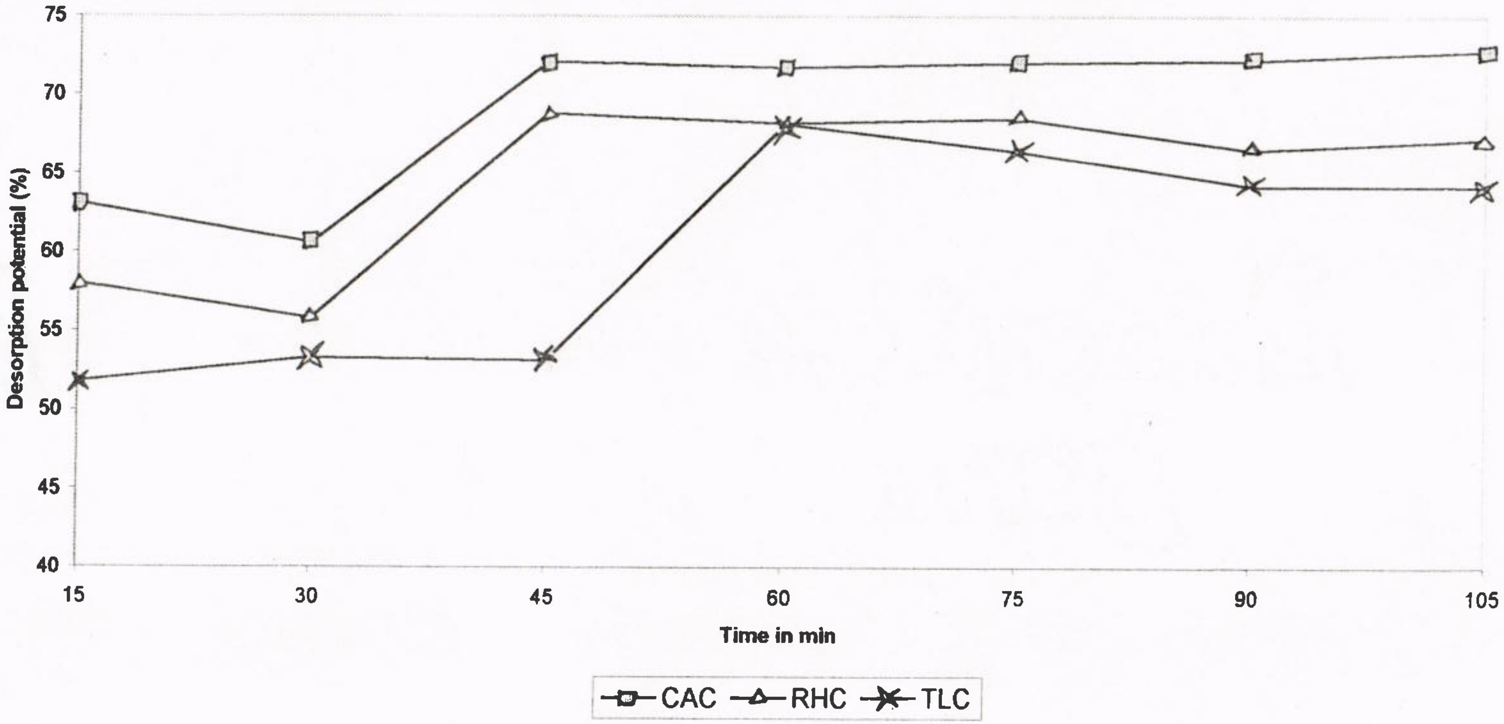
Adsorption potential of Cr VI with variation of Contact time



Condition - wt of adsorbent 300mg , Initial Concentration of Cr VI - 230 ppm , pH - 2 temperature - 32 + 0.1 C

Figure - 6

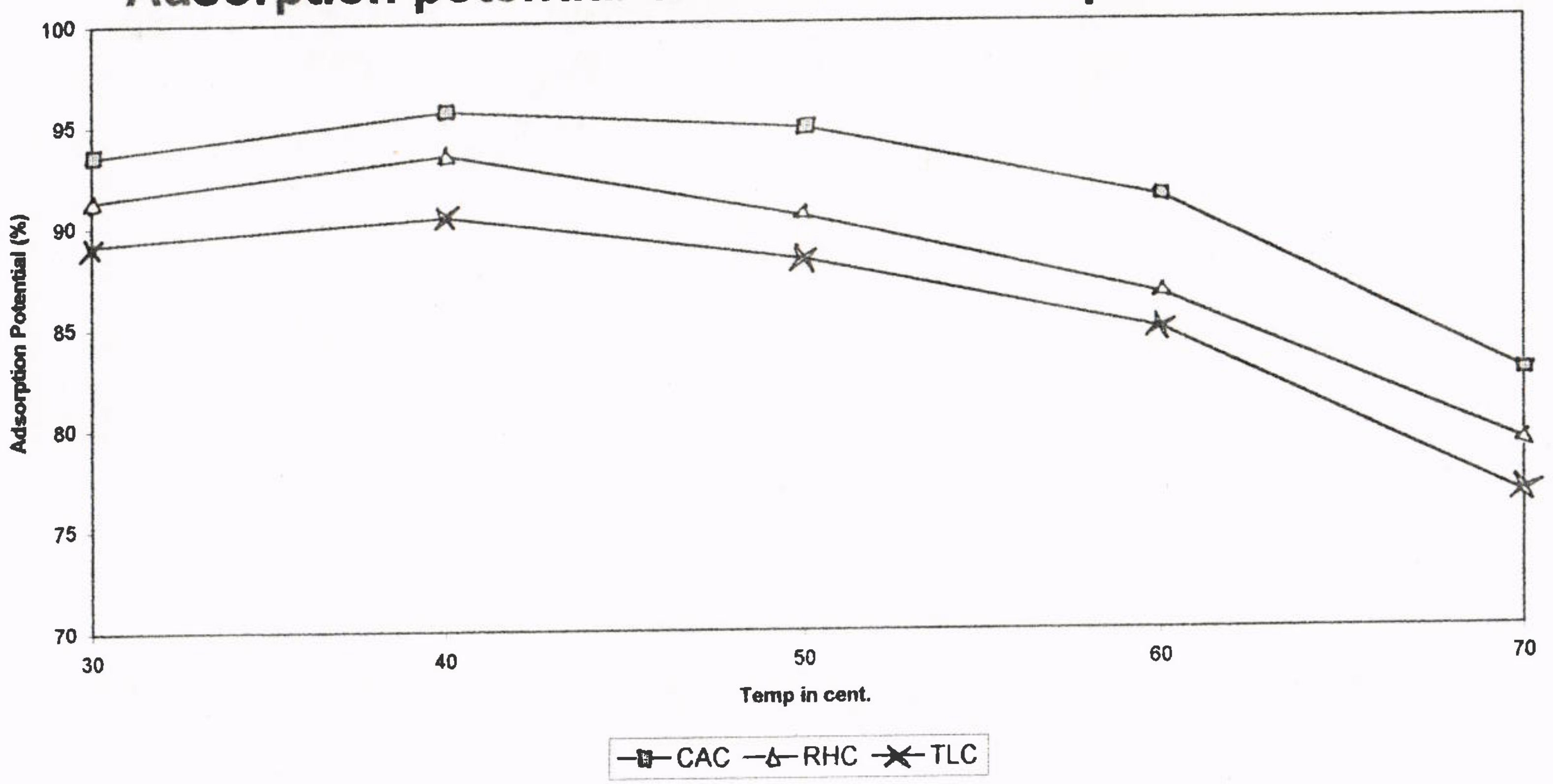
Desorption potentialof Cr VI with variation of contact time



Conditions - wt of adsorbent - 300mg pH - 2, Temp - 32 + 0.1 C

Figure - 7

Adsorption potential of Cr VI with Temperature Variation



Condition - wt of adsorbent - 300mgm Initial con. Of Cr VI - 230ppm Contact time - 60 mins pH - 2

Figure - 8

Kinetic Modelling for adsorption using Lagergren equation for various adsorbents

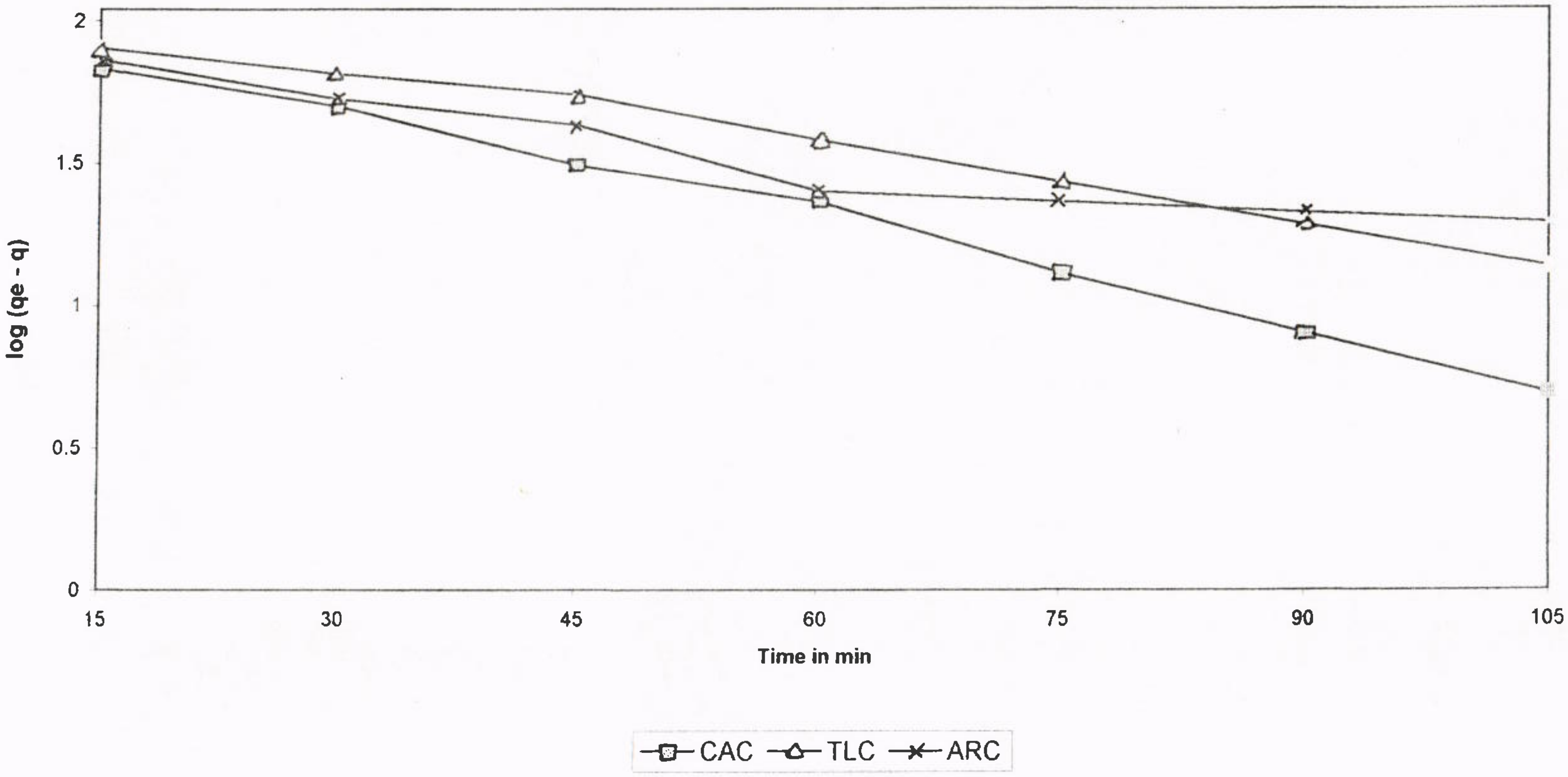


Figure - 9

Kinetic modelling for desorption using Lagergren Equation for various adsorbents

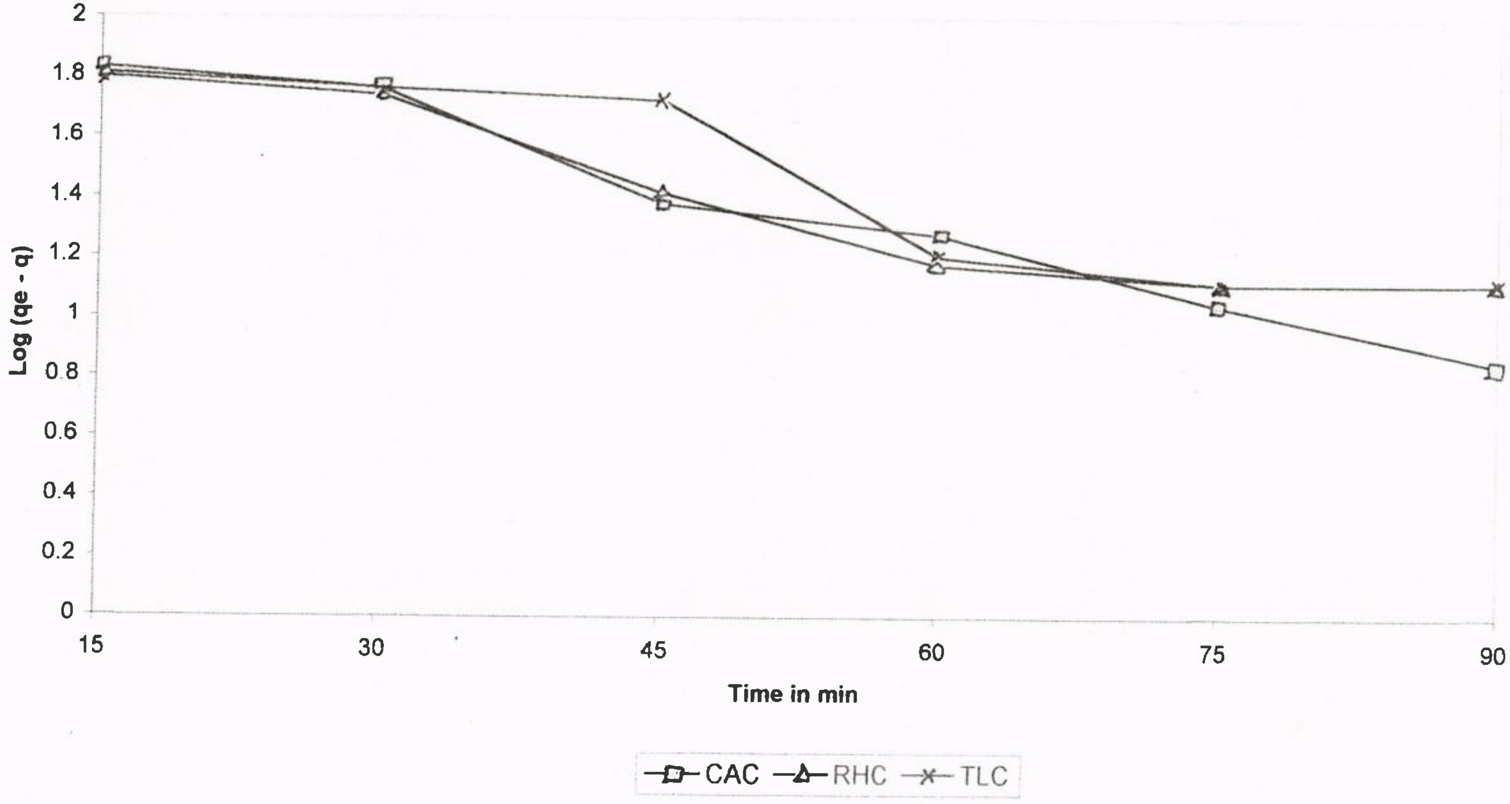


Figure - 10

A Typical representation of Intraparticle Diffusion Plot for various Adsorbents

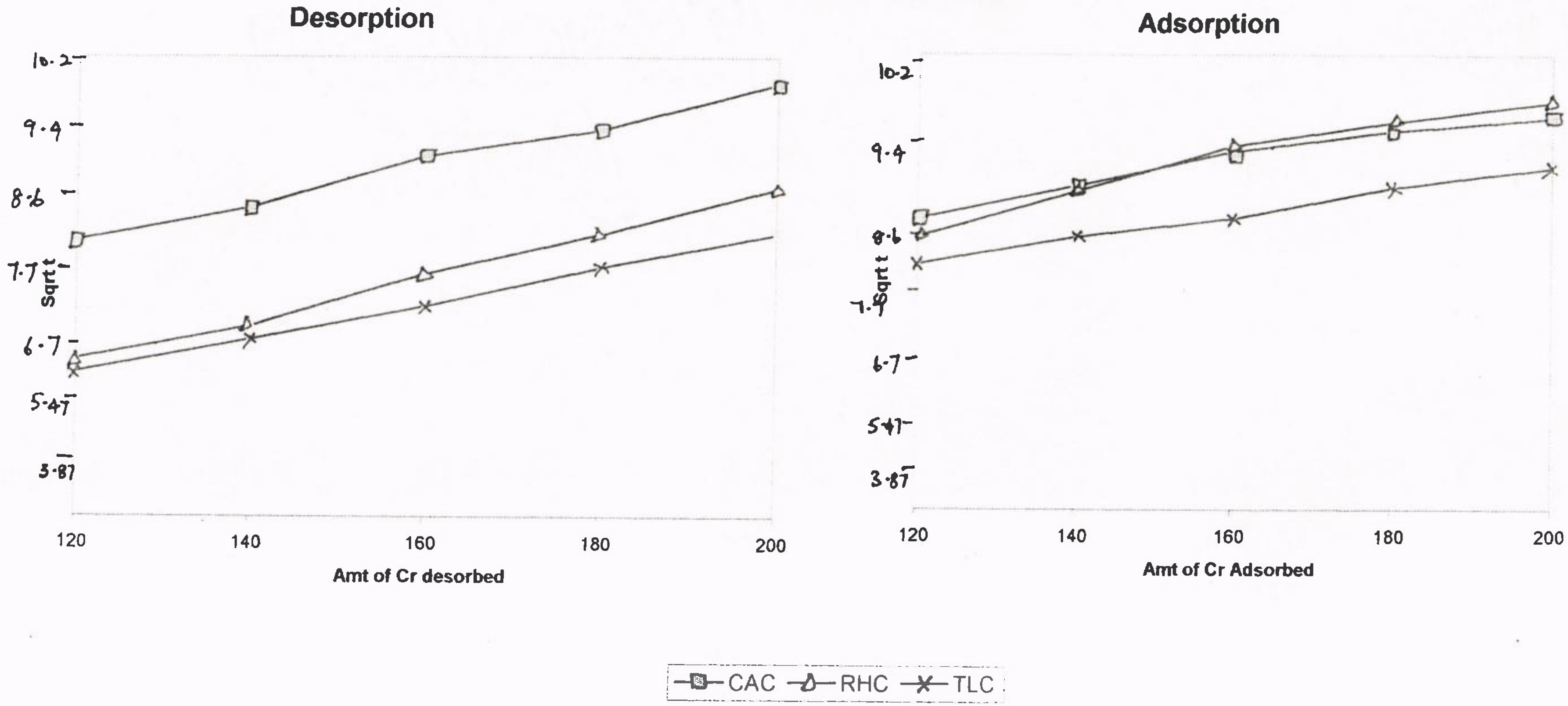


Figure - 11

A Typical representation of Langmuir Adsorption Isotherm curves for various adsorbents

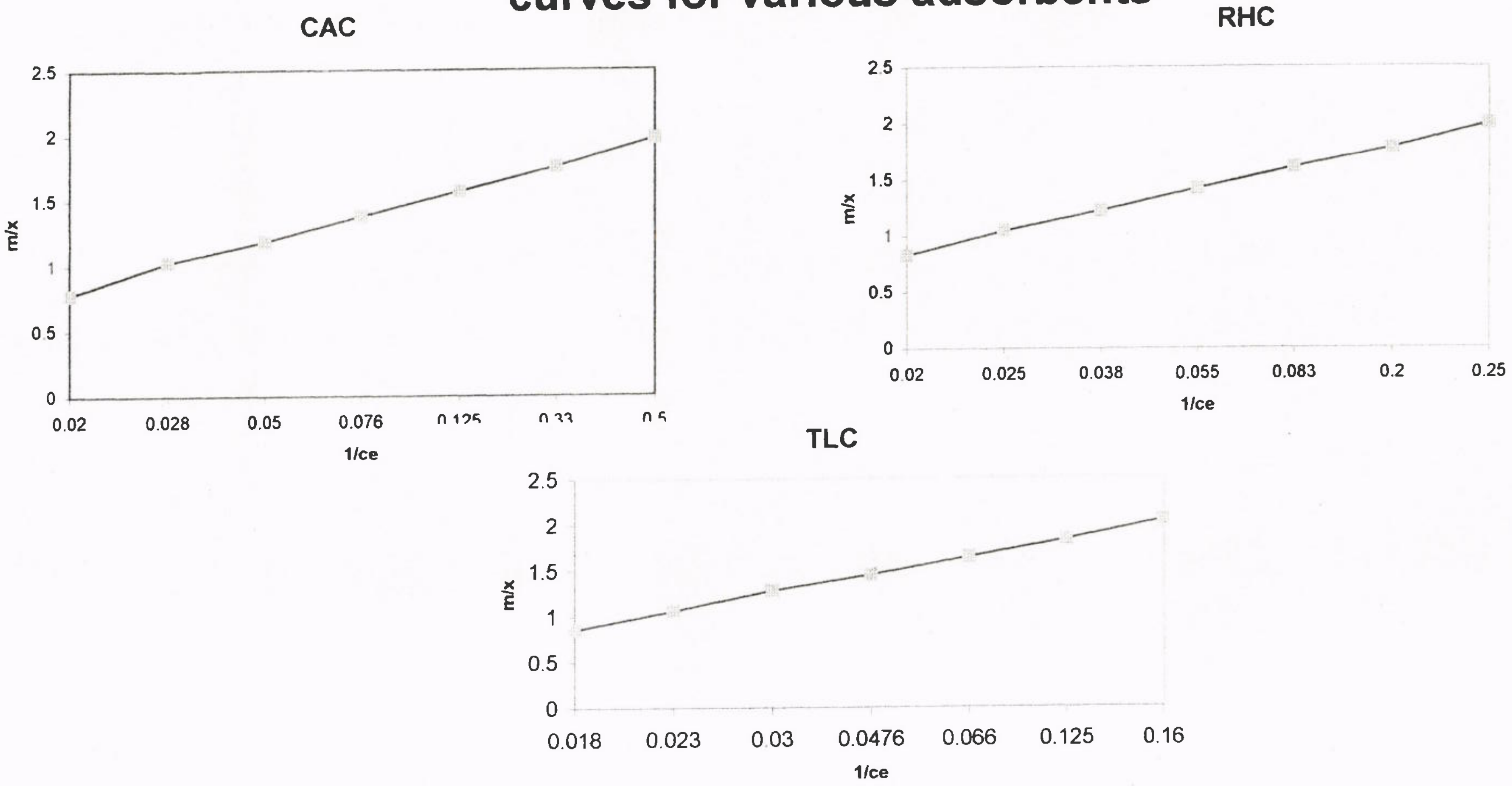


Figure - 12

A typical representation of Langmuir desorption Isotherm curves for various adsorbents

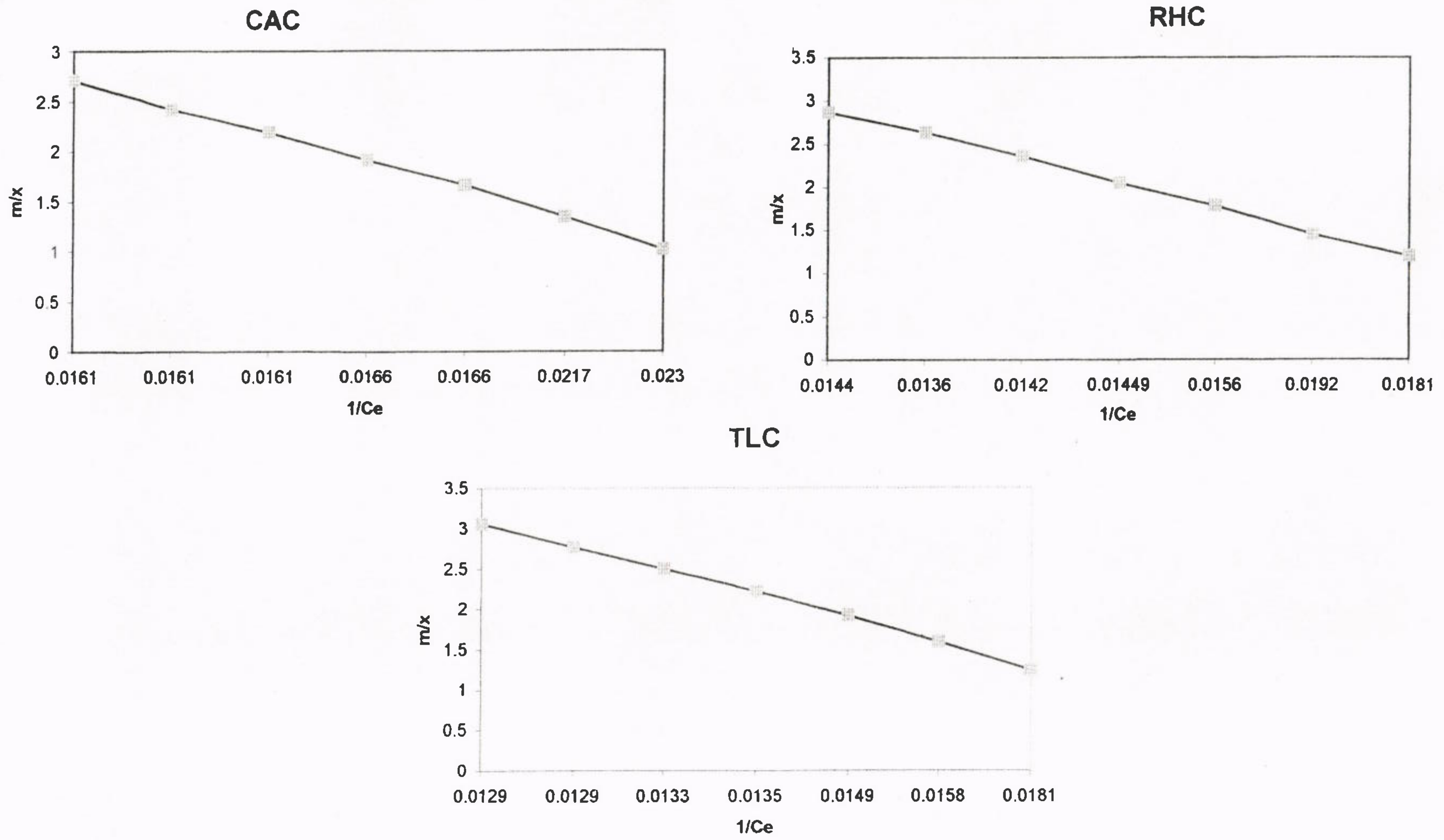
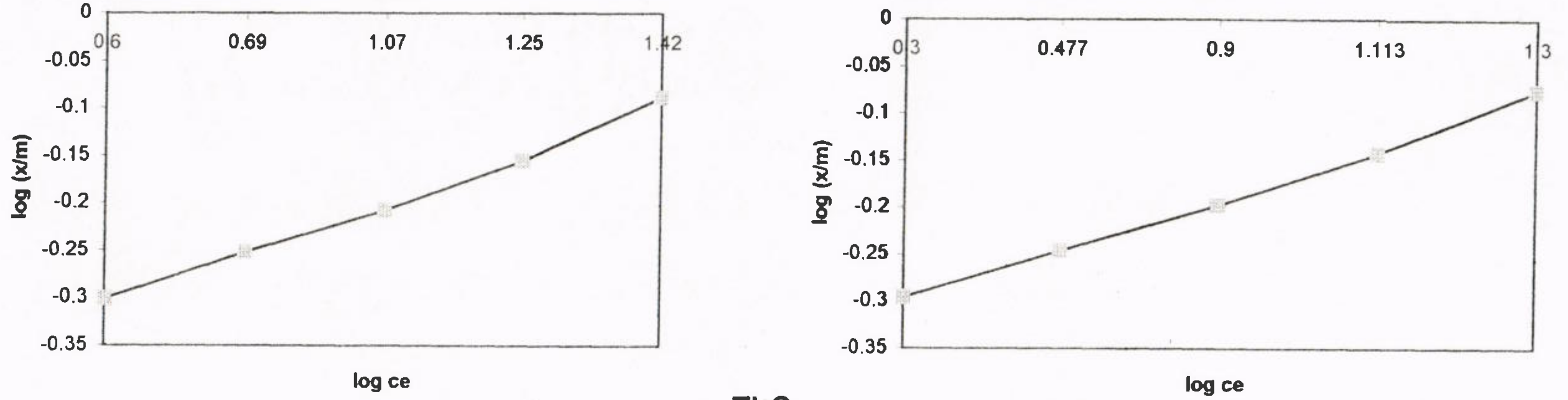


Figure - 13

A typical representation of Freundlich adsorption isotherm curves for various adsorbents


RHC

CAC

TLC

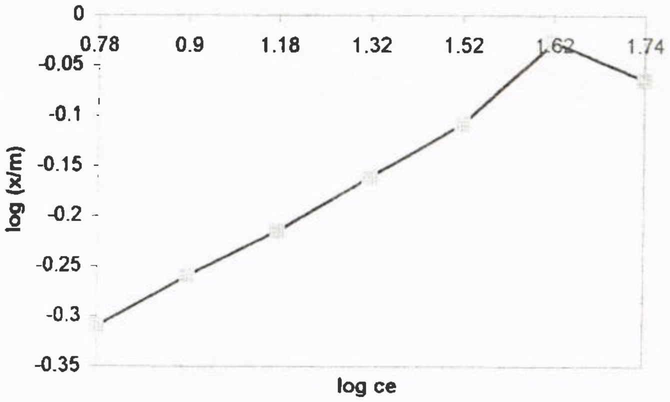
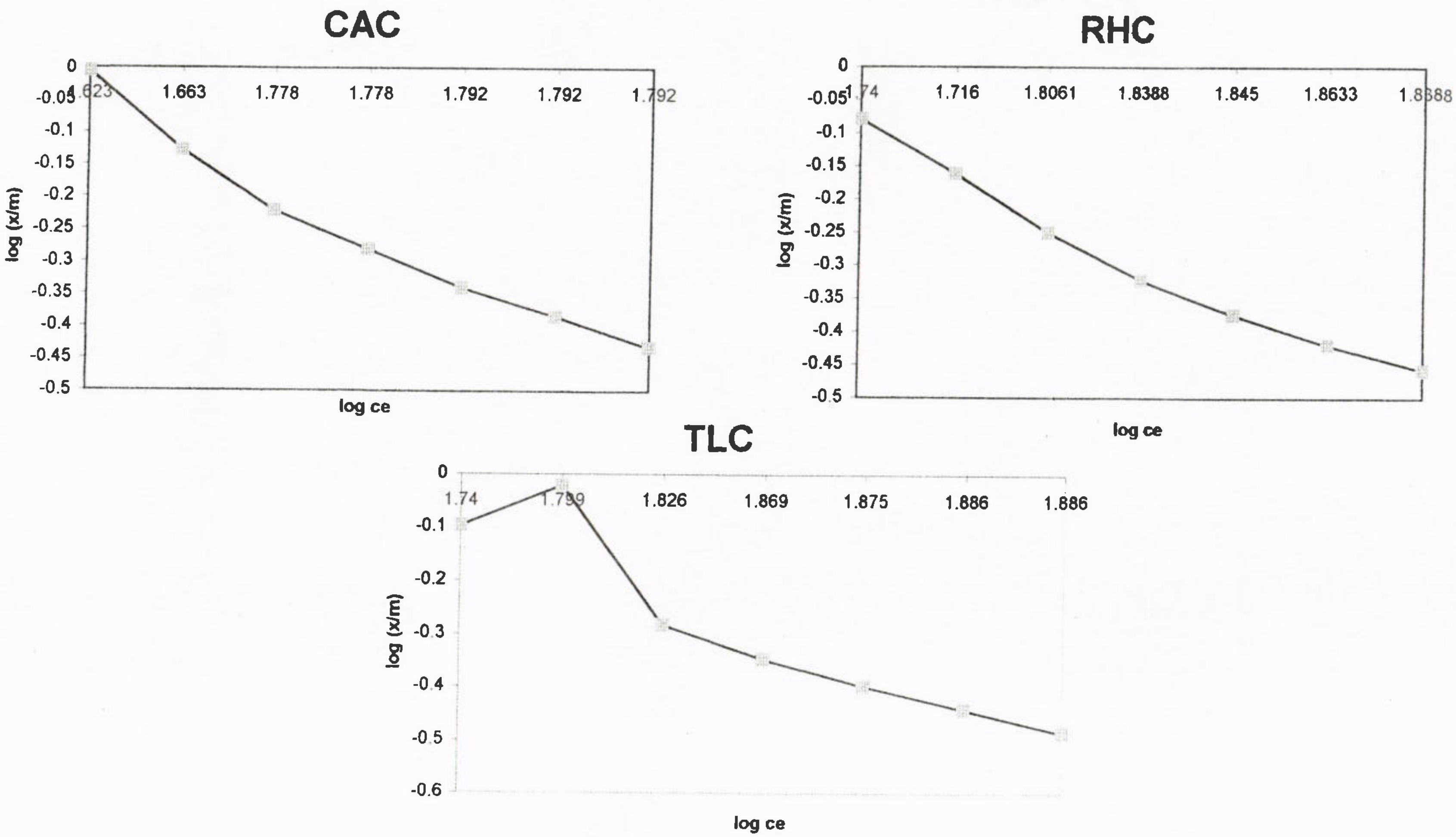


Figure - 14

A Typical Representation of Freundlich Desorption Isotherm curves for various adsorbents



Summary and Conclusion

5. SUMMARY AND CONCLUSION

The results of the present study can be summarised as follows.

- Maximum removal of Cr (VI) at room temperature (+10) can be achieved with minimum amounts of adsorbents (450 mg/100ml) at acidic pH (pH-2.5) and 105 minutes as equilibration time.

The adsorption and desorption processes are found to confirm to Ist order Kinetics.

Equilibrium data showed the applicability of Langmuir and Freundlich isotherm models.

The findings of this study reveal that RHC is a fairly good adsorbent for the removal of hexavalent Chromium from aqueous solutions.

The adsorption potential is higher than the desorption potential in the removal of Cr to and from the adsorbents respectively.

The experimental results obtained confirm the possibility of producing activated carbons with well developed pore structure, high surface area and adsorption capacity from agricultural waste by activation with concentrated sulphuric acid.

It is established that in identical conditions the adsorption characteristics of activated carbon is determined by the nature of raw material.

Removal of Cr (VI) using ecofriendly and economic adsorbents is feasible and efficient.

The study indicates that adsorption and desorprion potentials depend on adsorbent dosage pH, contact time and temperature.

It can be concluded that the bioadsorbents could be employed successfully for the removal of Cr VI from industrial effluents.

The economics of an adsorption process depends upon the regeneration and reutilisation of the adsorbent. The study shows that the spent sorbent can be regenerated using 2 M Sodium hydroxide. During regeneration a considerable amount of Chromium is obtained whihc has some economical value. The study suggests the reutilisation of the spent sorbent after making up for the loss in active adsorption sites during regeneration.

The study can be done at higher temperatures to determine the thermodynamic parameters and hence to the study of adsorption dynamics.

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Appendices

APPENDIX - 1
ESTIMATION OF CHROMIUM (COLORIMETRIC METHOD)

Principle:-

Using acidic conditions hexavalent chromium reacts with s-diphencyl carbazide to form a reddish violet coloured complex which can be determined colorimetrically.

Reagents:-

Double distilled water was used for the preparation of reagents.

a. **SYNTHETIC EFFLUENT:**

141.4 mg of potassium dichromate solution was dissolved in water and diluted to 1000 ml (1 ml=50μg of Chromium).

b. **STANDARD CHROMIUM SOLUTION:**

10 ml of sythetic effluent was diluted to 100 ml. 1 ml = 5 μg of Chromium.

c. **0.2N SULPHURIC ACID:**

8ml of concentrated Sulphuric acid was diluted to 1 litre with water.

d. **1 M SODIUM HYDROXIDE:**

40 g of Sodium hydroxide was diluted to 1 litre.

e. **3 M AMMONIUM HYDROXIDE:**

135 of Ammonium hydroxide was diluted to 1 litre.

f. **DIPHENYL CARBAZIDE SOLUTION:**

250 mg of s-diphenyl carbazide was dissolved in acetone and made up to 50 ml. Stored the solution in a brown botle.

APPENDIX - 2

PREPARATION OF CALIBRATION CURVE

To compensate for possible slight losses of Chromium during digestion or other analytical operation Chromium standards were treated as samples.

Accordingly measured volume of standard solution (1ml=5μg of Chromium) ranging from 20 ppm to 240 ppm of Chromium ie, from 4 ml to 48 ml of standard solution, were pipetted out in 100 ml. And added 2 ml of diphenyl carbazide solution to each of these solutions. After 15 minutes the violet colour developed was read at 540 nm. Using the reagent blank absorbance was corrected to zero.

The above procedure was repeated for sample also. From the corrected absorbance Chromium in the sample was determined with reference to the calibration curve.

APPENDIX - 3

ABBREVATIONS

CAC - COMMERCIALLY ACTIVATED CARBON

RHC - RICE HUSK CARBON

TLC - TEA LEAVES CARBON