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THE COST ANALYSIS AND ECONOMIC FEASIBILITY OF AGRO WASTES TO ADSORB CHROMIUM (VI) FROM WASTEWATER

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ABSTRACT

In the present study biodegradable agro and horticultural waste materials have not only been used to adsorb pollutant Cr (VI) from wastewater but it will help in the agro waste management also. Using agro waste materials to purify water for reuse indicates indirect social welfare for the organization which is not directly measured. Low cost sorbent used by tanneries, electroplating industries and other metal finishing industries that release Cr (VI) in effluent wastewater joining natural streams it is sure that these industries will control this pollution at the source itself. So the Cr (VI) removal comparatively at a low cost will be the financial benefit to such type of industries. This benefit will certainly lower the overall costing of the process and products as well.

First time an attempt has been made to estimate the cost of adsorption while studying economic feasibility of various agricultural and horticultural wastes like pea (Pisum sativum) pod peels, tea (Camellia sinensis) & ginger (Zingiber officinale) mix and banana (Musa lacatan) peels to adsorb Cr (VI) from wastewater. The removal cost of 1g of Cr (VI) from wastewater using these adsorbents like; Pea Pod Peels, Tea & Ginger mix and Banana peels Waste were estimated as Rs. 9.14, Rs. 4.71, Rs. 3.11 respectively. The findings indicate that the cost of using these agro wastes as adsorbents is lesser than that of commercial activated carbon which is Rs. 142.145.The developed adsorbents were prepared from agricultural wastes which undoubtedly require proper waste management.

Adsorbents pea pod peels waste; tea & ginger waste and banana peels waste the positive enthalpy ΔH values 1.089, 1.627 and 1.494 respectively showed the endothermic sorption and the strong binding, the positive free energy change ΔG; 1.31632, 1.946 and 1.752 showed the random feasibility. Adhering, fixing adsorbate on the interface between two phases result in loss of the degree of freedom showing a *negative entropy (ΔS) for pea pod peels waste (-0.0126 and tea & ginger waste (-0.0055) whereas positive 0.0107 for banana peels waste.*

Keywords: Sustainability, Peels, Effluent, Greenhouse, Chromium Etc.

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Abbreviations: Pea pod peels waste (PPP), Used Tea and Ginger Waste (T&G), banana peels waste (BW)

PUBLIC INTEREST STATEMENT

The study shows that agricultural wastes like pea pod peels, used tea & ginger mix waste and waste of banana peels are the potential adsorbents for the removal of Cr (VI) from the effluent wastewater of tanneries and metal finishing industries. Once the chromium is removed at the source it will not at all join our natural streams of water. And this way it will protect water quality of surface as well as ground water against chromium pollution and toxicity.

RESEARCH CONTRIBUTION

The paper will contribute a lot to the society in terms of sustainability. It deals with all the three dimensional aspects of sustainability i.e. social, environmental and economic. It will also help in solid waste management as it is converting the agricultural wastes in to value added products. The advantage to the environment is that Cr (VI) removal from wastewater is achieved without damaging ecosystem. Another environmental advantage is high quality refining and reuse of refined water for different purposes. As a result of the environmental and financial benefits, various chromium releasing industries will take measures to protect the environment at the source only and make a profit at a low cost. Social aspect covers the agricultural waste management without any over burden to the society otherwise the farmers especially in developing countries like India burn their agricultural wastes and lot of carbon dioxide emission pollutes the atmosphere causing green house effect.

RESEARCH HIGHLIGHTS

1. *The adsorbents were not at all burnt to convert them in to carbon but processed for efficiency thereby saving energy in burning.*
2. *Optimization of various affecting parameters using batch set-ups.*
3. *Feasibility study of low cost agro and horticultural adsorbents for removal of chromium using Gibb's Energy Equation in Thermodynamic study.*
4. *Characterization of adsorbent surface using Scanning Electron Microscopy (SEM) analysis.*
5. *First time an innovative attempt to estimate the cost of agro and horticultural wastes used as low cost adsorbent for the removal of chromium (VI) as compared to the expansive activated carbon.*

1. INTRODUCTION

After 1970 when Environmental Protection Agency established; toxic metals degrading surface and subsurface water quality, have been a global issue and a centre of researcher's attraction. The researchers are continuously focusing on to control the emissions of heavy metals at source only to within safe industrial and drinking standards so as to achieve quality surface and subsurface water. Many projects in developing countries suffer due to environmental clearances for inadequate pollution regulations, impact on Ozone layer, air pollution and negligence of drinking water standards. The research projects of environmental concern must use the novel green technologies and the pollution control at the source only without spreading and making it uncontrollable and environmental issue as well. Many researchers have emphasized on many fold sustainable aims like; focusing on pre and post issues, abatement of heavy metals at source and their recovery for reuse, waste management and converting them in to value-added products for reuse and last but not the least controlling thereby all kind of environmental pollution with a great economy. Broadly it is an approach targeting "Six Rs" i.e. Reuse polluting agricultural and horticultural waste, Revalue converting them in to value-added wastes, Remediation of toxicity due to heavy metals, Recovery of heavy metals, Recirculation and Reuse.

Heavy metals such as chromium (Cr), lead (Pb), cadmium (Cd), nickel (Ni), mercury (Hg), copper (Cu), arsenic (As) and zinc (Zn) etc. are being widely used in various industries where from these join the environment. These metals either in form ions or compounds are water soluble and consumed by living world and being very toxic in nature their excess amount adversely affect health of all living beings. Here we are only concerned with the chromium. Though chromium (III) is very good for sugar metastasis (Katz, 1991) but chromium (VI) has been most dangerous. The industries such as electroplating, tanneries, metal finish, pigmenting and wood preservation etc. discharge chromium (VI) in their effluent wastewater which generally in developing countries like India joins natural water streams and severe contamination occurs.

In India the adsorption of chromium from industrial wastewaters using agro and horticultural wastes as adsorbent has become a popular, efficient, potential and cheaper solution which offers flexible design to produce quality wastewater within safe standards of disposal and these agro and horticultural adsorbents could also be reused but being very cheaper desorption for reuse is never ever encouraged (Sharma P.K., et al., 2013; 2015, 2016 a, 2016 b, 2017; Raji, et al., 1998; Ajmal, et al., 1995, 2000; Ayub,et.al., 1998,1999,2001,2002,2003, 2014). The potentiality of agro wastes to adsorb chromium from wastewater has been established outstandingly by a number of researchers in recent years (Sharma, P.K., et al.,2017, 2016 a, 2016 b, 2015, 2013,2010; Ayub S., et al., 2014, 2006,2002, 2001,1999, 1998, Veena Devi B., et al 2012, Baskaran P. K., et al 2010; Camino, et al., 2000; Weber, 1996; Drake, et al., 1996; Chand, et al., 1994; Siddiqui, et al., 1994; Deo, et al., 1992; Periasamyet al., 1991; Shukhla, and Sakhardane, 1991; Vaishya and Prasad, 1991; Huang, et al., 1975;). The conventional and commercial expensive activated carbon was substituted using non-conventional, cheaper and local agro wastes as adsorbents.

The effluent wastewater from tanneries containing chromium is usually converted to beneficial Cr (III) from dangerous Cr (VI) using various chemicals like sulfur dioxide, sodium bisulfate or sodium meta-bisulfite etc.

Using calcium hydroxide for pH above 8.0 less toxic hydroxide of chromium [$Cr(OH)_3$] is precipitated which is adsorbed and disposed easily as shown in following reaction.

$3\,Ca(OH)_2 + Cr_2(SO_4)_3 = 2\,Cr(OH)_3 + 3\,CaSO_4$ (Sharma, P.K. et al. 2016)

The physical adsorption mechanism comprises of surface forces imbalance. The adsorbate molecules of aqueous solution develop interface on solid adsorbent in contact through the abridgement occurring in the adsorbent capillaries and the chemical adsorption results due to adsorbate molecular layer on its base formed by residual valence forces of the base molecule. It is observed that the substances with higher molecular weight can be efficiently adsorbed from aqueous solutions and hence the adsorption process best suits to adsorb chromium from tannery effluents. A quick equilibrium interface concentration is achieved by slow diffusion. Though the adsorption varies with varying combinations of adsorbent and adsorbate along with interference of other interfering ions in complex wastewaters.

Though a lot of literature is available on low cost adsorptions but none of the researchers has estimated the cost of adsorbents so as to prove their economy. First time it is tried to analyze the adsorption cost so as to coin the economic feasibility of the so called agro and horticultural wastes used as adsorbent to remove chromium from the tannery effluents. The study comprises of estimation of cost and economical feasibility of different agro wastes like used mixture of tea and ginger (T&G), pea pod peelings (PPP) and banana waste (BW) which were used to adsorb Cr (VI) from tannery waste waster. Various parameters like pH, adsorbent dose, temperature, adsorbate concentration, contact duration, sorbent size and mixing rpm etc. were renovated during adsorption. The sorbents surfaces were described prior to batch adsorption and later; using Scanning Electron Microscopy (SEM) analysis. Thermodynamic characterization had also been made.

2. MATERIALS AND METHODS

2.1. Preparation of Sorbents and Sample

Through batch adsorption agro wastes like pea peelings (*Pisum sativum)* pod (PPP), a mixture of used ginger (*Zingiber officinale*) and tea (*Camellia sinensis***)** waste (T&G) along with banana (*Musa lacatan*) peels waste (BW) had been utilized to remove chromium (VI) from tannery wastewater.

2.1.1. Adsorbents

All the adsorbents as above were sun dried for 15 days first, then dried in oven five hours daily at 95°C for three days and then after repeated distilled water wash again dried in sun light for a week. After sun drying all the adsorbents were powdered and double dips washed by 0.1 NaOH and 0.1N H2SO4 to get rid of lignin and alkaline. Powdered sorbents after distilled water wash were dried and kept in desiccators. Before use these ready sorbents were sieved through Indian Standard Sieve for the segregation of grains.

2.1.2. Experimental

For all adsorbents a batch adsorption of chromium as per Standard Method were conducted at 25-28^o C during summer to study the effects of various parameters like sorbent dose, pH, grain size, contact time, temperature and the mixing rpm using Chromium Standard Solution for different chromium concentrations and thus optimum values of all parameters were determined so as to use them for actual tannery wastewater. Then for all optimized affecting parameters Whatman No.1 filter paper was used to filter the real suspension and the filtrates were analyzed for residual chromium by AAS (Make-Perkin Elmer, AAS, Pin AAcle 900K Model) at CED, EE section, Z.H.C.E.T., AMU, Aligarh (India).

2.2. Thermodynamic Feasibility

Using Gibb‟s free energy relation; **$\Delta G = -RT \ln K$** at equilibrium the values of thermodynamic constants indicated the adsorption process feasibility. In this relation **Δ G** represents free energy change in joule/mole, R; the universal gas constant as 8.314 and T the absolute temperature in Kelvin.

Graphs with Δ G on Y-axis and T on X-axis were drawn for all the adsorbents at the adsorption state of equilibrium temperature and then the best fit lines were mathematically simulated to the relation **$\Delta G = \Delta H^* - T.\Delta S$** [Sharma, P.K. et al; 2012 (a) & (b)]

Here, Δ H stands for enthalpy change, Δ S stands for entropy change for activation [Ayub, S. et al, 2006]. Using plots of Gibb's free energy as shown in Figure-1 PPPTP for pea pod peels, Figure-2 T&GTP for used tea and ginger waste and Figure-3 BWTP for banana peels waste as adsorbent along with their respective best fit lines for simulation and mathematical modeling to find the adsorption feasibility; ΔH, ΔG and ΔS were computed at 301 K as shown in Table-1.

All the positive enthalpy ΔH values 1.089, 1.627 and 1.494 for each adsorbent confirmed the endothermic adsorption and also indicated strong attachment; the entire positive changes in free energy ΔG values 1.31632, 1.946 and 1.752 for each adsorbent showed the random feasibility. Whenever there is attachment and fixing of adsorbate on the surface of adsorbent there occurs the degree of freedom loss indicating negative value of entropy ΔS which was really found negative for pea pod peels -0.0126 and for used tea and ginger waste as -0.0055 but this entropy value for banana waste was found to be + 0.0107 (Ayub, et al, 2006) as shown in Table-1.

2.3. Langmuir Isotherms

The Langmuir equation $q_e = \frac{(Q_o K_L C_e)}{(1+K_L C_e)}$ transformed to straight line equation as $\frac{1}{q_e} = \frac{1}{q_o} + \left(\frac{1}{q_0.K_L}\right)\left(\frac{1}{C_e}\right)$

Notations Ce stands for adsorbate content at equilibrium in mg/l, Qe stands for amount of sorbate attached on 1 gm of the sorbent at equilibrium in mg/g, Qo stands for maximum single layer covering capacity of adsorbent in mg/g which is very important parameter for estimating the cost of adsorbent here in this study. KL stands for Isotherm constant of Langmuir curve expressed in L/mg. The KL and qmax were determined by intercept and slope of the best fit line for Langmuir curve i.e. $\frac{1}{q_e}$ Vs $\frac{1}{C_e}$

Equilibrium constant RL, a dimensionless pure number is also a very important parameter to check the feasibility of the sorption. It tells the nature of adsorption i.e. if RL is greater than unity means unfavorable adsorption, linear if RL equals to unity, favorable if greater than zero and less than unity and irreversible if RL equals to zero.

RL = $\frac{1}{[1+[1+KL.Co\}]}$ Where, Co stands for adsorbate initial concentration which was 30 mg per litre, KL the dimensionless Langmuir constant of equilibrium concerned with the energy of adsorption [Dada, et al, 2012].

The Langmuir plots for pea pod peels, used tea and ginger waste and banana peels waste used as adsorbent are shown in Figure-4 PPPL, Figure-5 T&GL and Figure-6 BWL respectively. The various constants values of adsorbents are in Table 1.

2.4. Scanning Electron Microscopy

Scanning Electron Microscopy (SEM) for all the adsorbents was also done to understand the characteristics and surface textures of adsorbents both before and after use in adsorption process (Model No. JSM 6510 LV Make; JEOL Japan, at USIF, AMU, Aligarh). The samples were gold coated as found non-conducting before SEM. SEM analysis revealed sorbents showing sufficient surface with undulations for binding adsorbate.

3. RESULTS AND DISCUSSIONS

Using optimized values of adsorbent grain size, pH, sorbent dose, contact period, mixing speeds and temperature from batch experimental sets adsorption isotherms were plotted to study economic adsorption and its feasibility with cost estimation. Thermodynamic parameters so obtained clearly showed the feasibility of the adsorption.

3.1. Optimization of Affecting parameters

3.1.1. Optimum pH 2.5-3.5

Best efficiency of adsorption was observed in pH range 2.5-3.5 for all adsorbents (Sharma, P.K. et al., 2016). Similar range was obtained by various researchers who removed dihydric phenol by activated carbon (Mahesh, et al.,1999) and chromium by sphagnum moss peat (Sharma, et al., 1993).

3.1.2. Optimum Dosage 10 g/L

Banana waste dose @ 10 g/L was found suitable to adsorb almost 70 % chromium at initial concentration 30 mg/L in 1.0 hour. With the same dose of adsorbent 10 g/L used T&G removed 75 % chromium whereas PPP adsorbed 65% at optimum pH that is very near to The results by (Bansal and Sharma, 1992; Mall, 1992; Rao, et al., 2002; Ayub, S. et al., 2001, 2002; 2014).

3.1.3. Optimum Contact Time 45 Minutes

T&G and BW removed 90% chromium in first 45 minutes but PPP removed 65% only.

3.1.4. Optimum Grain Size 600 µm to 300 µm

Average grains ranging from 600 µm to 300 µm found optimum for PPP, T&G and BW.

3.1.5. Optimum Temperature 25^0C to 30^0C

The best suited temperature found 25^0C to 30^0C for all the adsorbents. used in batch set ups.

3.1.6. Optimum Mixing Speed 80 to 100 rpm

The optimum mixing speed at initial stage was observed 80 to 100 rpm neither belo nor above rpm was found suitable.

3.2. Physical Adsorption Feasibility

The value of ΔH positive showed endothermic adsorption. The value of ΔG positive showed random feasibility. Entropy (ΔS) being negative for all the adsorbents at equilibrium revealed loss of degree of freedom may be due to watering of the adsorbent surface (Baisakh, et al., 1996, Baisakh, et al., 2002)

The Langmuir adsorption isotherms for PPP, T&G and BW had been plotted as shown in Figures 1 PPPL, 2 T&GL and 3 BWL. R^2 value for Langmuir was 0.963 (PPP), 0.986 (T&G) and 0.982 (BW). Using Langmuir Isotherms for PPP, T&G and BW the maximum monolayer coverage (Q_0) were determined as 4.33 mg/g, 7.29 mg/g and 10 mg/g respectively and also the separation factors (R_L) were computed as 0.0331, 0.0343 and 0.0756 which indicated the favorable sorption. The maximum Gibb's free energy (ΔG) was found for PW as 6.0679 joule/mole showing banana peels potential low cost sorbent.

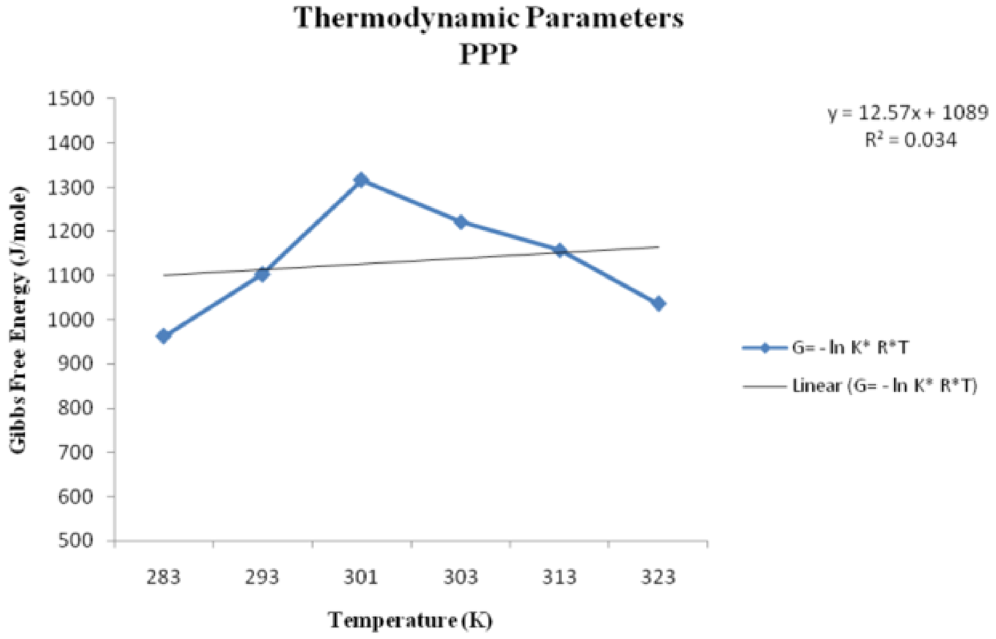

Figure 1 PPPTP: Thermodynamic Sorption Parameters for Pea pod peels Waste

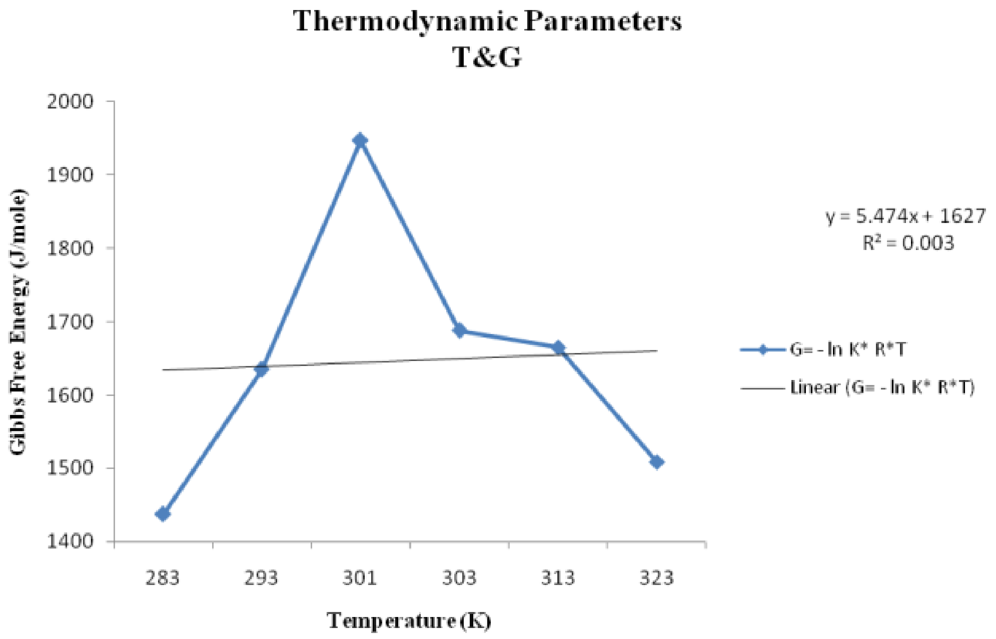

Figure 2 T&GTP: Thermodynamic Sorption Parameters for Tea & Ginger Waste

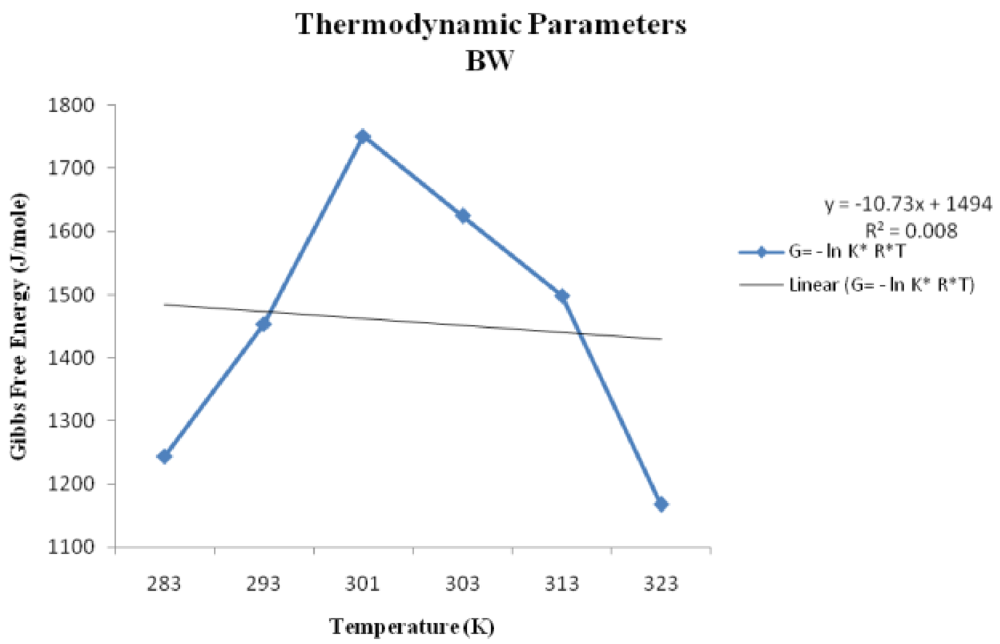

Figure 3 BWTP: Thermodynamic Sorption Parameters for Banana Waste

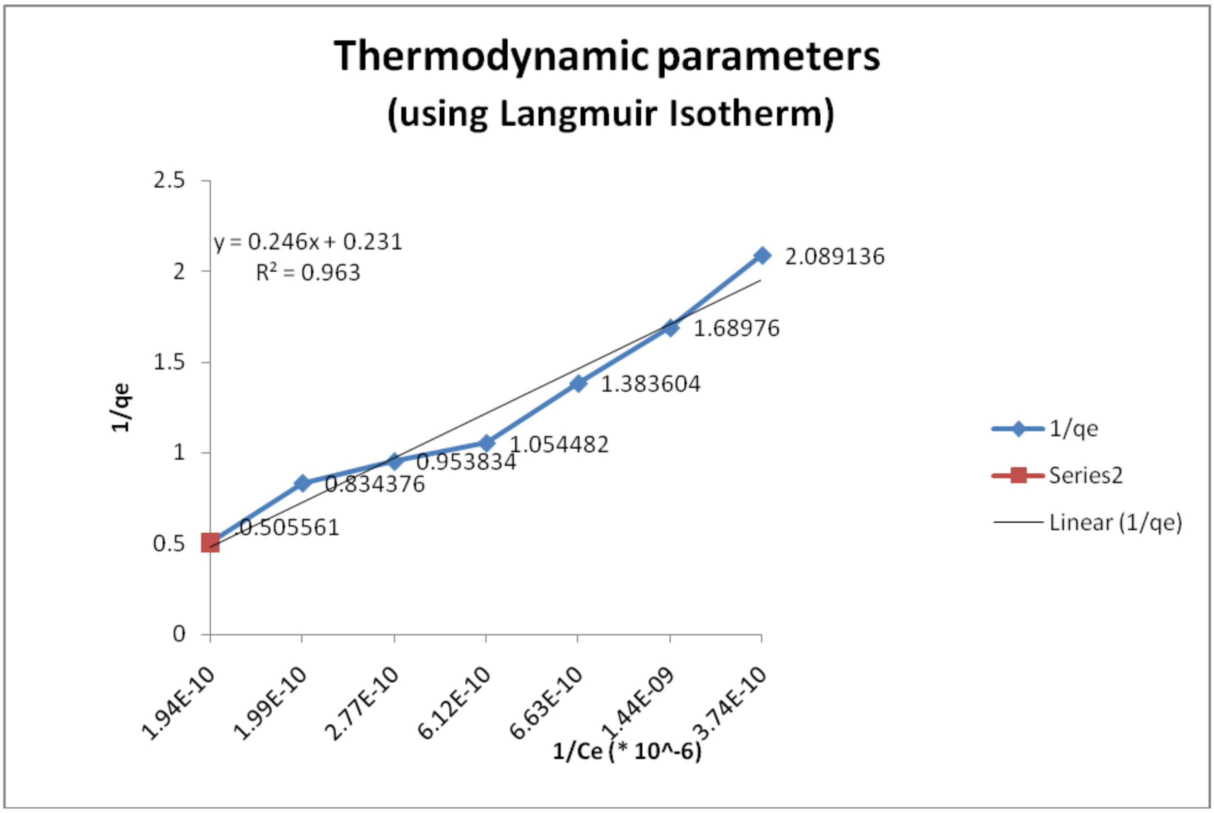

Figure 4 PPPL

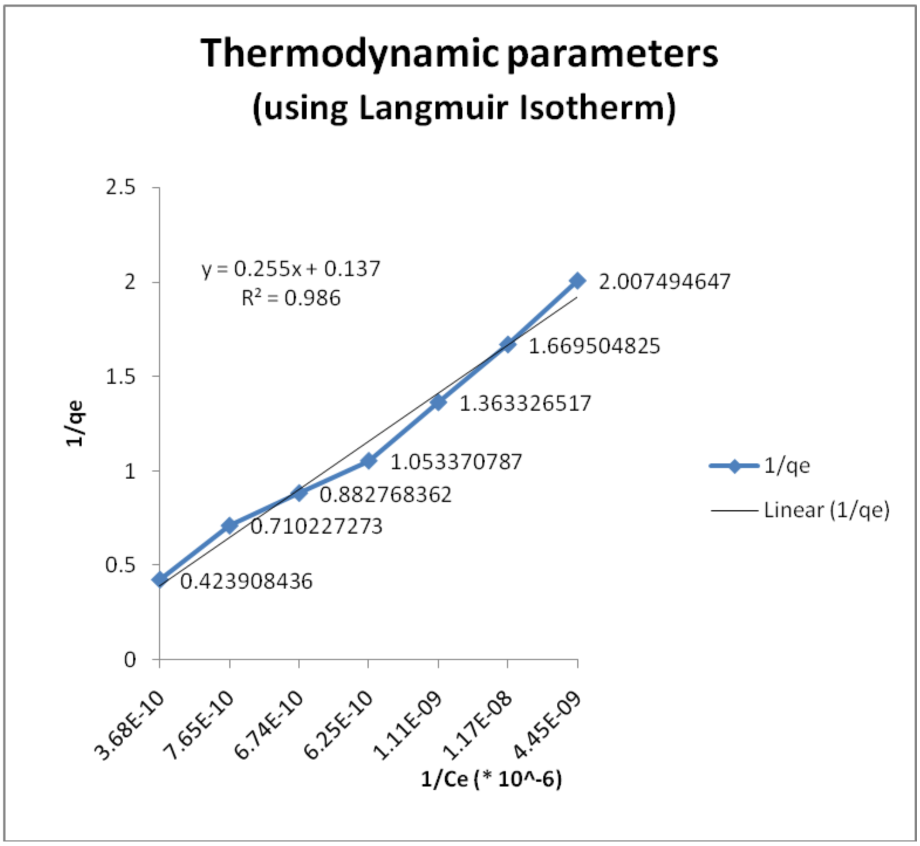

Figure 5 T&GL

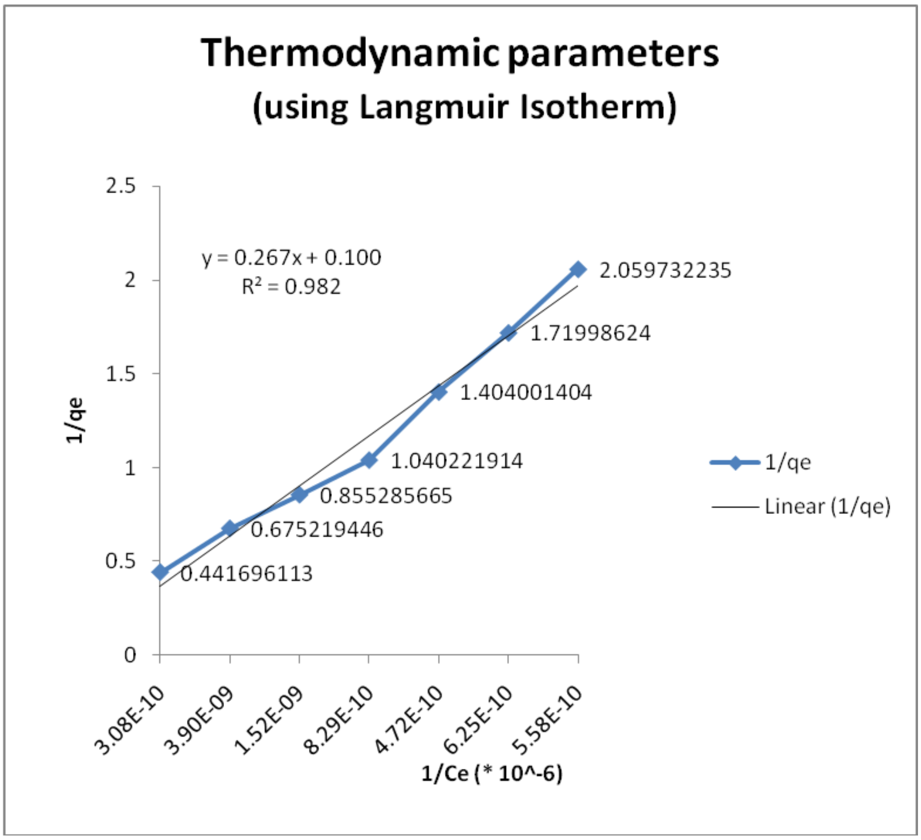

Figure 6 BWL

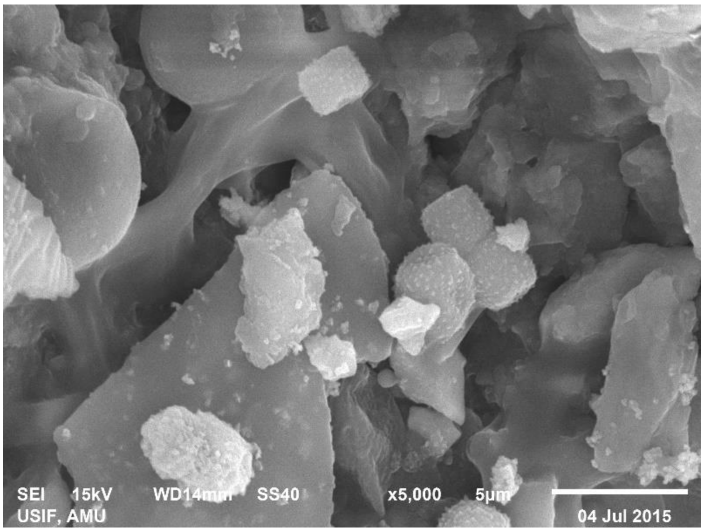

Figure 7 PPP RAW-0005.tif

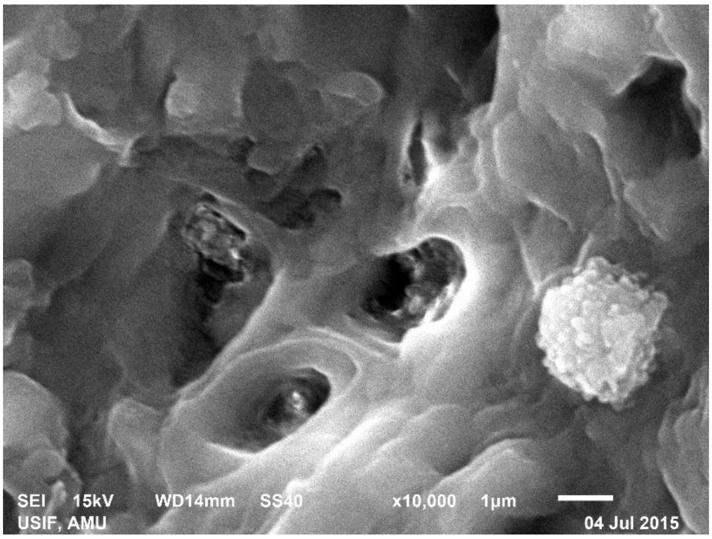

Figure 8 PPP USED-0005.tif

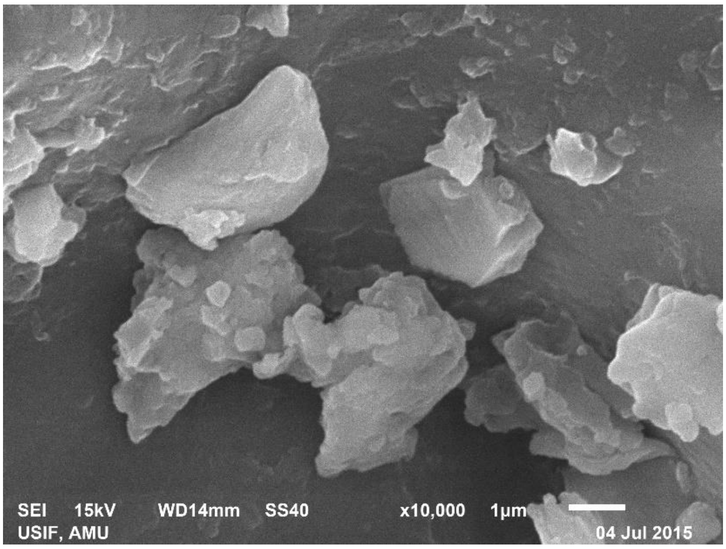

Figure 9 T&G RAW 0005

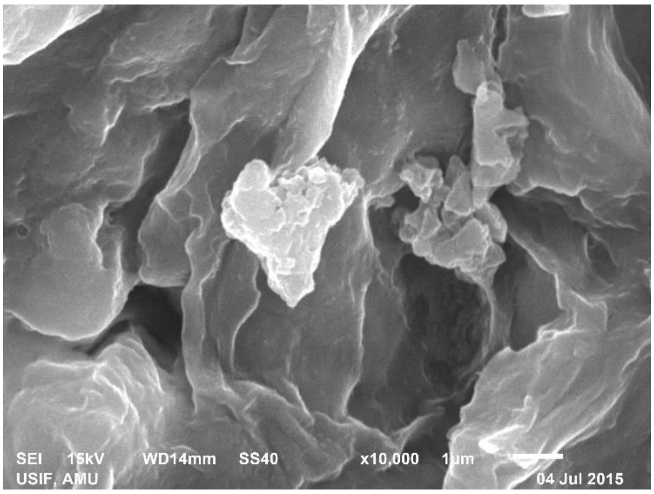

Figure-10 T&G USED-0005.tif

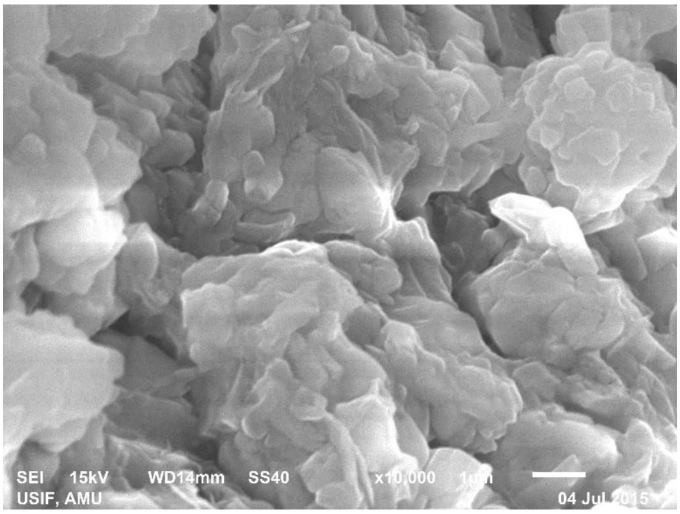

Figure 11 BW RAW-0005.tif

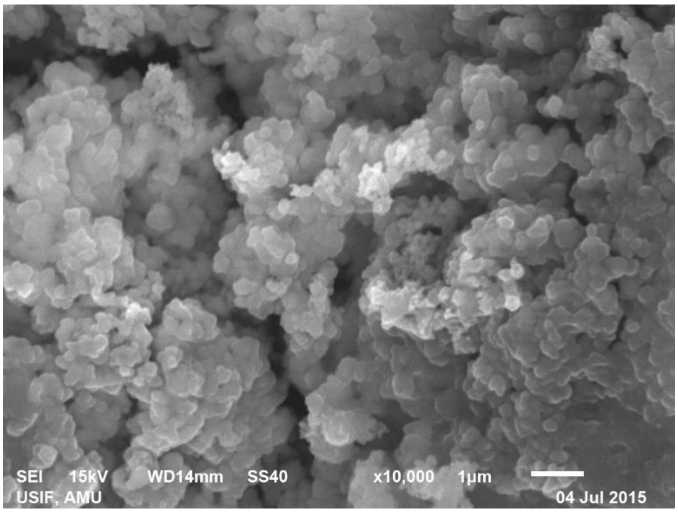

Figure-12 BW USED-0005.tif

4. COST EVALUATION

Cost evaluation is very useful tool to decide and fix the environment protection criteria provided probable environmental effects on application of physical and chemical processes are considered. Without cost evaluation of any process it becomes very difficult to select a process for the adsorption of chromium or some other heavy metal. So the cost evaluation of any process or method is very important part which was left by most of the researchers so far. The sorption process cost mainly depends on the adsorbent cost. From the ancient times activated carbon has been most used for the adsorption of chromium from wastewater and it was very expensive and unaffordable by developing countries like India; the problem couldn't be solved here it is hardly available so to meet out its day by day increasing demand as a result low cost materials become today's need which are comparable to activated carbon with respect to local environmental aspects, availability, feasibility, efficiency and economy etc. So keeping all these points in mind, the authors in this study, have attempted to evaluate the cost of adsorption using PPP, T&G and BW as adsorbents for the removal of chromium from aqeous solutions. The steps for the calculation of cost for the adsorbents preparation have been taken from the rare literature available (Gupta, 2008, Changani, 2014). The cost evaluation and economic feasibility of above agro wastes adsorbents in this research work to adsorb chromium from leather manufacturing industrial effluents has been done and at a glance shown in Table-2 and Table-3 for all the adsorbents used here.

Including all procedural steps like physical and chemical activation costs; the total cost of Adsorbents preparation as shown in Table-2. Each 1 Kg of adsorbent prepared costed as Rs. 39.56, Rs. 34.36, Rs. 31.14 respectively for activated PPP, T&G and BW.

The commonly used adsorbent activated carbon costs around Rs. 600 a kg. To remove 1 gm chromium from tannery effluents the estimated cost of sorbents; PPP, T&G and BW is ahown in Table-3. These estimated costs of the adsorbents; PPP, T&G and BW are **Rs**. 9.14, **Rs**. 4.71, Rs. 3.11 respectively. These costs are very low as compared to that of activated carbon Rs. 142.145. The developed adsorbents are prepared from agricultural wastes otherwise these agrowastes need to be managed because their disposal is also a big problem of environmental concern. So the utilization of these agro wastes to adsorb chromium from aqueous solutions helps in agro waste management. the agrowastes not only adsorb chromium but also reused and revalued giving indirect benefit to society and environment otherwise farmers usually burn them due to expensive transport. The burning of these agro wastes contributes to the green house gases emission which is of great environmental and climatic concern. Chromium is removed at a low cost which is the direct monetary benefit to the industry. This benefit lowers overall costs of the process and products as well. The advantage to the environment is that chromium is removed from tannery effluent without any disturbance to ecosystem. The quality of refined water is very high so it can be reused for various purposes. So in addition to the environmental and monetary advantages tanneries play a big role for employment and protection of environment at very low cost. This research opens the pathways of the Sustainable development of industries, society and nation. All the required aspects of sustainable development like social, environmental and economical are being achieved. It is the beauty of this research.

Table 1

At 301 K	Langmuir Constants					Thermodynamic Parameters		
Sorbent	**(Max. monolayer coverage capacity, mg/g) qmax**	**1/q0**	**(Langmuir Constant) KL**	**RL (favourable if 0< RL<1)**	**R^2 (Best Error distribution or perfect correlation)**	**ΔG (KJ/mole)**	**ΔH (KJ/mole)**	**ΔS (KJ/mole)**
PPP	4.329004	0.231	0.939	0.0331	0.955 (should be <1)	1.31632	1.089	-0.0126
T&G	7.29927	0.137	0.9059	0.0343	0.5555	1.946	1.627	-0.0055
BW	10	0.1	0.3745	0.o756	0.5831	1.752	1.494	0.0107

Table 2 Estimating and Costing of adsorbent per Kg of Agro wastes (PPP, T&G, BW).

Item	Unit Cost (Rs.)	Pea Peels	Pod Waste	Tea & Mix	Ginger Waste	Banana Waste	Peels
		Amount	Price	Amount	Price	Amount	Price
H2SO4	6.00 per L	1.5 L	9.00	1.4 L	8.40	1.2	7.20
NaOH	1.60 per L	5.0 L	8.00	4.0 L	6.40	3.0	4.80
Drying Cost	5.80 KWh	1.0 KWh 70^0, 10 h	5.80	1.0 KWh 70^0, 10 h	5.80	1.0 KWh 70^0, 10 h	5.80
Heating Cost	5.80 Kwh	1.2 Kwh 90^0, 10 h	6.96	1.2 Kwh 90^0, 10 h	6.96	1.2 Kwh 90^0, 10 h	6.96
Grinder	5.80 Kwh	0.80 Kwh, 1hr	4.64	0.4 kwh 30 min	2.32	0.4 kwh 30 min	2.32
Net Cost (Rs.)			34.40		29.88		27.08
Other Overhead Cost (Rs) (15% of Net Cost)			5.16		4.48		4.06
Total Cost (Rs)			39.56		34.36		31.14

Table 3 Unit chromium removal cost

Sorbent	Sorption Capacity (mg/g)	Sorbent Cost (Rs/kg)	Sorbent Cost for unit g chromium removal (Rs.)
PPP	4.33	39.56	9.14
T & G	7.30	34.36	4.71
BW	10	31.14	3.11

5. CONCLUSIONS

- That agro and horticultural wastes; PPP, used T&Gr and BW have been proved to be potential sorbents for the sorption of Cr (VI) from leather and electroplating industrial effluent discharges.
- The chromium (VI) removal efficiency of these agro and horticultural waste was best found between pH 2.0 to 3.0.
- The positive enthalpy (ΔH) showed endothermic sorption and strong attachment. Gibb's free energy (ΔG) positive proved non-spontaneous sorption. Entropy value ΔS negative tells poor attachment of the adsorbate on to the surface of.
- Leaching test failed as traces of chromium showed but banana peels didn't show anything like that and observed satisfactory.
- No change in BOD of leachate showed that the agro waste adsorbents are of stable nature in the adsorption.

- The agro waste adsorbents not only found cheaper and better than conventional activated carbon but also solid waste management became easier in rural areas because they simply require acid/alkali washing to enhance the adsorption efficiency.
- Scanning Electron Microscopy showed sufficient surface and irregular spaces within which chromium is adsorbed.

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