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REMOVAL OF CHROMIUM (VI) FROM WATER USING BIO-ADSORBENTS DERIVED FROM LEAVES OF *Salvadora persico* AND *Caesalpinia bonduc* PLANTS

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ABSTRACT

Bio sorbents derived from leaves of *Salvadora persico and Caesalpinia bonduc plants* and their ashes are investigated for their affinity towards Cr (VI) ions from polluted water using batch mode of extractions. The influence of different physicochemical characteristics viz., pH, equilibration time, sorbent dosage, the initial concentration of Cr (VI) and presence of co-ions, are studied and optimized for maximum Cr (VI) extraction. The removal is found to be 81.0% and 86.0% respectively with leaves and their ash of *Salvadora persico plant while 95.0% and 98.0% for Caesalpinia bonduc plant adsorbents.* Some co-cations are synergistically enhancing the % removal. Monovalent anions have a marginal effect but PO_4^{3-}, SO_4^{2-} and CO_3^{2-} are interfering with the extraction of Cr (VI). The adsorption capacity is found to be 16.2 mg/g (leaves) and 22.9 mg/g (ashes) of *Salvadora persica plant*; 12.7 mg/g (leaves) and 19.6 mg/g (ash) *of Caesalpinia bonduc plant*. The procedures are successfully applied to the samples collected at various divergent sources.

Keywords: Chromium, extraction, bio-adsorbents, *Salvadora persico, Caesalpinia bonduc.*

INTRODUCTION

Intensive research interest at present is devoted throughout the Globe in developing water purification methods based on using bio-adsorbents. These methods are economical, effective and biocompatible. Our research group successfully developed methodologies for the extractions of pollutants such as Cr (VI) [1-5], Zn(II)[6], Al (III)[7-9], F^- [10-15], NO_2^- [16-18] , NH_3[19-22] , PO_4^{3-} [23-25] and Dyes.[26-30]

While we are probing the various biomaterials for their affinity towards potential pollutants, we noticed the bio-materials of *Salvadora persico* and *Caesalpinia bonduc plants* have a strong affinity towards Cr (VI), a potential pollutant. The toxicity of Chromium ions (both trivalent and hexavalent) is well known and the maximum permissible limit for Chromium ions in water is 0.05 ppm[31-33] . Salts of Chromium are extensively used in various industries viz., tanning, inks, fungicides, textiles, photography, paints etc and effluents from these sources are turning to be a potential source of Chromium pollution [31-36] . The contamination of Chromium salts in natural water is further amplified due to process like bio-amplification in water bodies.

Different bio-adsorbents are investigated for the removal of Cr (VI).[37-48] Coconut fiber[39], eucalyptus bark [41], maple sawdust [42], *Hevea brasilinesis* sawdust activated carbon[43] , waste tea leaves and rice husk [44] and Neem Sawdust,[45,46] are investigated as adsorbents for the removal of Cr(VI). The bio-adsorbents pertaining to *Justicia adhatoda*, *Cissus quadrangularis*, *Soapnut Acacia, Syzygium Cumini, Azadirachta Indica and AcaciaArabica*, are also investigated.[3, 4] Comparative studies using different bio-absorbents are also reported to literature.[1-5, 47,48]

In the present investigation, bio-adsorbents prepared from the leaves of *Salvadora persico and Caesalpinia bonduc plants* have been explored for their sorption nature towards Cr (VI) by changing the

different physicochemical equilibrium conditions. Optimum conditions for the maximum Chromium (VI) extraction are investigated. Thus, developed methods are applied to various polluted real water samples.

EXPERIMENTAL

Adsorbents

Our Preliminary investigations revealed that the leaves powders and their ashes of *Salvadora persico and Caesalpinia bonduc plants* had a strong affinity towards Cr (IV) ions.

Salvadora persico

Caesalpinia bonduc

Fig.-1: Plants having affinity to Chromium (VI)

Salvadora persica plant is a small shrub grows to a maximum height of three meters and it belongs to *Salvadoraceae* family of the plant kingdom. This plant stems are used to clean teeth and leaves are eaten in salad and are used in traditional medicine in curing asthma, rheumatism, scurvy etc. *Caesalpinia bonduc plant* belongs to Grey Nicker species of senna family of the plant kingdom. It is a climber creeps over other vegetation. It's bio-parts are used in various traditional medicines.

Salvadora persica and Caesalpinia bonduc plant leaves were cut, washed with distilled water, the sun dried and pulverized to a size less than 75µ and then activated at 105^0 C for three hours in hot-air oven Further, the dried leaves were burnt to ashes. Both the thermally activated leaves powders and their ashes were investigated in the present work.

Adsorption Method

Batch methods were adopted.[49-51] Known amounts of bio-sorbents were equilibrated for a desired time with predetermined concentrations of Cr (VI) solutions taken in stoppered 500 ml bottles in mechanical shakers. Using pH meter, initial pHs of the equilibration solutions were adjusted with dil NaOH and dil HCl. After certain agitation time, the samples were analyzed for Chromium content spectrophotometrically by using "Diphenyl Carbazide" method as described elsewhere.[52] All other studies were made using this general procedure in finding the effect of pH, sorbents concentration, time of equilibration, the initial concentration of Cr (VI), etc. The results were presented in the Fig.- 2 to 6 and Table -1 and 2.

Effect of Interfering Ions

Interference caused by fivefold excess of co-ions generally existing in water such as PO_4^{3-}, SO_4^{2-} NO_3^-, CO_3^2, Cl^-, F^-, Ca^{2+}, Mg^{2+}, Cu^{2+}, Zn^{2+}, Ni^{2+} and Fe^{2+} , on the % removal of Cr(VI) was studied. For this, known quantities of Cr (VI) are mixed with five fold excess of one of the co-ions and then the solutions were subjected to extraction procedure. The results were given in Table-1.

Applications

The methodologies developed in this work for the removal Cr (VI) were studied using samples collected from diverse sources such as effluents from tannery industry, chromate plating industry and natural lake samples (fed with known amounts of Cr ions). The results obtained were given in the Table -2.

RESULTS AND DISCUSSION

Effects of various parameters on adsorption characteristics of the bio-adsorbents developed in this work towards the extraction of Cr (VI) ions are discussed hereunder.

Influence of pH

pH conditions of equilibration system have marked effect on the removal of Cr(VI). As the pH decreases, % removal is increased (Fig.- 2).

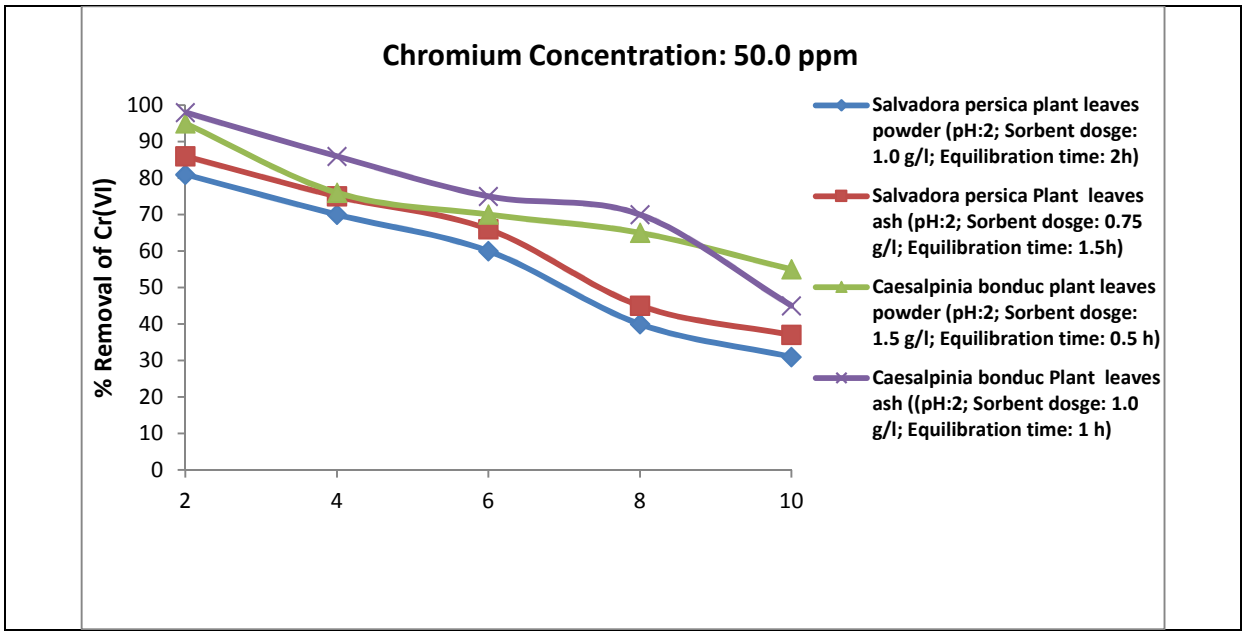

Fig.-2 : pH Vs % removal of Cr(VI) at Optimum Equilibration conditions

With *Salvadora persica* leaves powders, % remove of Cr (VI) is: 31% at pH 10; 40.0% at pH 8; 60.5% at pH 6 ; 70.0% at pH 4 and 81.0% at pH 2 at other optimum conditions of equilibration. With the *Salvadora persica leaves ash*, the extraction of Cr (VI) is: 37.0% at pH 10, 45.0% at pH 8, 66.0% at pH 6, 75.0% at pH 4 and 86.0% at pH 2. Similarly, with the *Caesalpinia bonduc leaves powder,* the removal is : 55.0 % at pH 10; 65.0% at pH 8; 70.0% at pH 6; 76.0% at pH 4 and 95.0% at pH 2 ; and with *the ashes of Caesalpinia bonduc ,* the removal is : 45.0% at pH 10; 70.0% at pH 8; 75.0% at pH 6; 86.0% at pH 4 and 98.0 % at pH 2.

These observations are on expected lines. Bio-adsorbents are endowed with functional groups such as hydroxyl, carboxyl etc., and their dissociations depend upon the pH conditions. At high pH values, dissociation occurs resulting negative charge to the surface of the adsorbent. At low pH values, dissociation is not favored and even protonation occurs, and thereby, imparting to the surface a positive charge. Cr (VI) being an anion, it is favorable 'onto' the surface of the adsorbent at low pHs and hence more removal. At high pHs, the negative charge of the surface repels the anion, Cr (VI) and hence, adsorption is less favored.

Contact time

Removal of Cr (VI) increases with equilibration time until a certain interval and after that time, the removal remains almost constant. With the adsorbents of *Salvadora persica*, the removal is found to be: 50.0% at 0.5 h , 62.0% at 1.0 h, 76.0% at 1.5 h, and 81.0% at 2.0 h or above with the leaves powders; and 61.0% at 0.5 h, 70.0% at 1.0 h and 86.0% at 1.5 h or above with the ashes of the said leaves (Fig.- 3a and 3b).

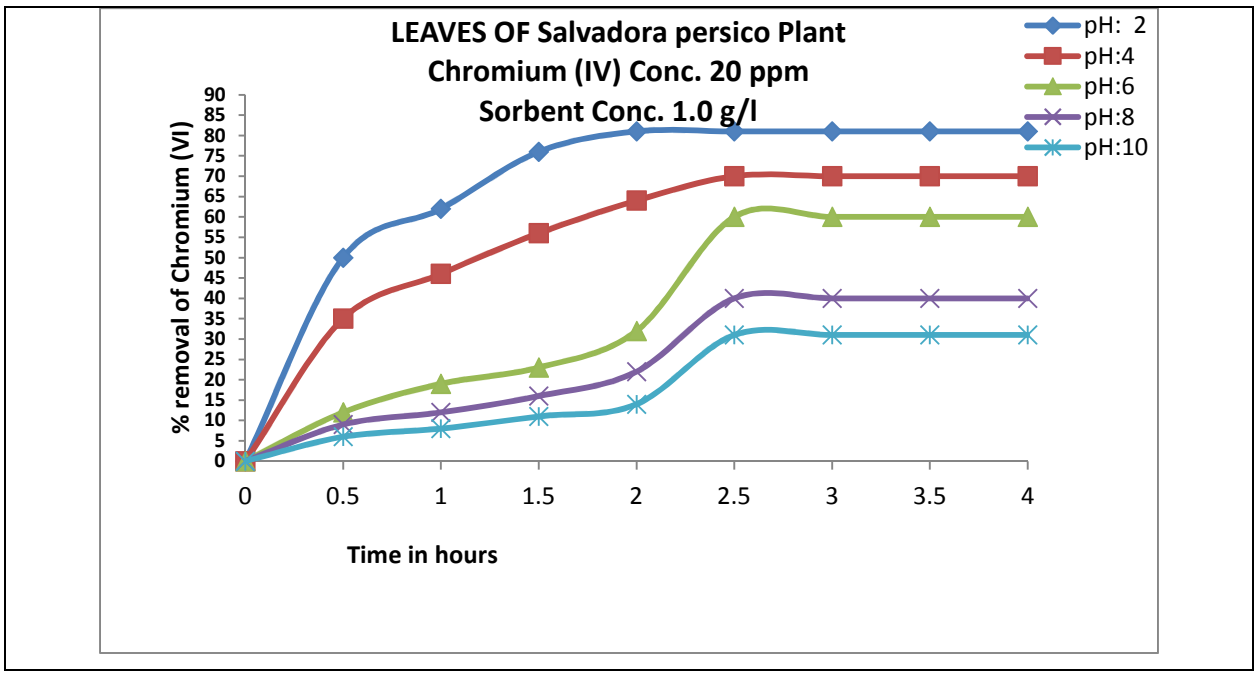

Fig.-3a: Effect of equilibration time on the extraction of Chromium (VI)

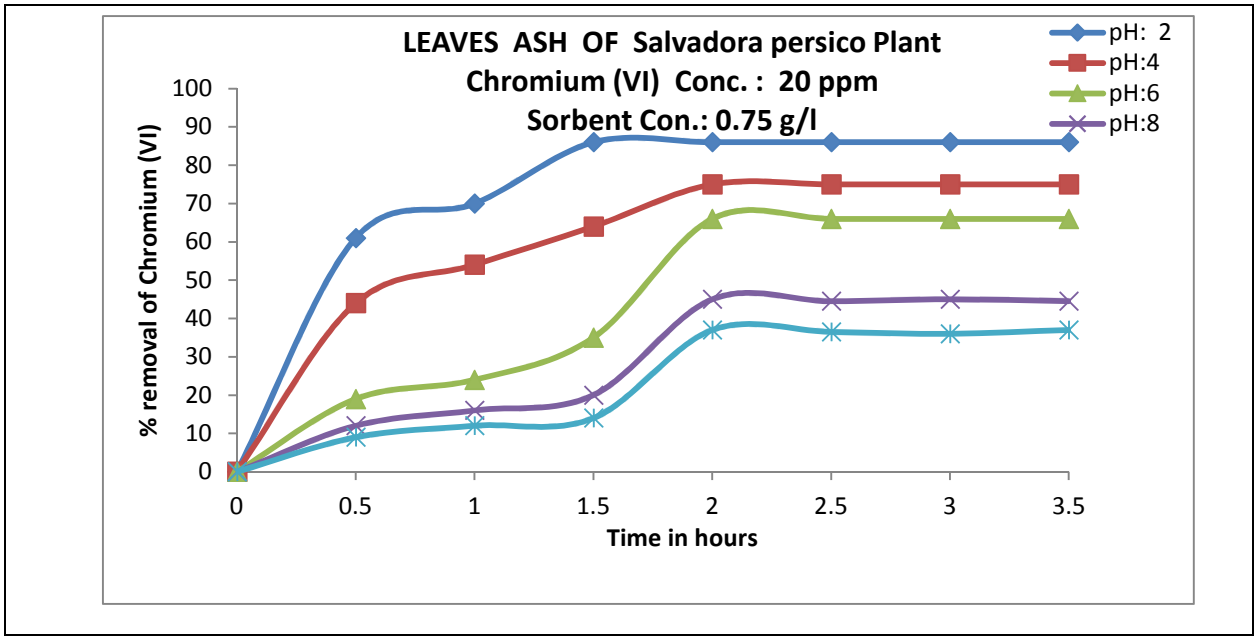

Fig.-3b: Effect of equilibration time on the extraction of Chromium (VI)

Similarly, with the leaves powder of *Caesalpinia bonduc* plant, the time dependent removal is found to be: 95.0% at 0.5 h or above and with their ashes, 61.5% at 0.5 h and 98.0% at 1.0 h or above (Fig.-4a and 4b). It may be noted that initially, the adsorption is more and it is slow down with time and it reaches a steady state after a certain time and then onwards, the removal almost remains constant. This is due to the fact that initially, more number of active sites are available for the fruitful adsorption of Cr (VI), and as the sites are progressively used up with time, the rate of adsorption is slow down.

Further, it is interesting to note that the steady state is reached quickly in the case of adsorbents *of Caesalpinia bonduc plant* than with *Salvadora persica* plant. The study state is reached after 2.0 h and 1.5 h of equilibration with the leaves and their ashes of *Salvadora persica plant* respectively while 0.5 h and 1.0 h are sufficient to reach the equilibration with the leaves and their ashes of *Caesalpinia bonduc plant* respectively (Fig.- 3 and 4).

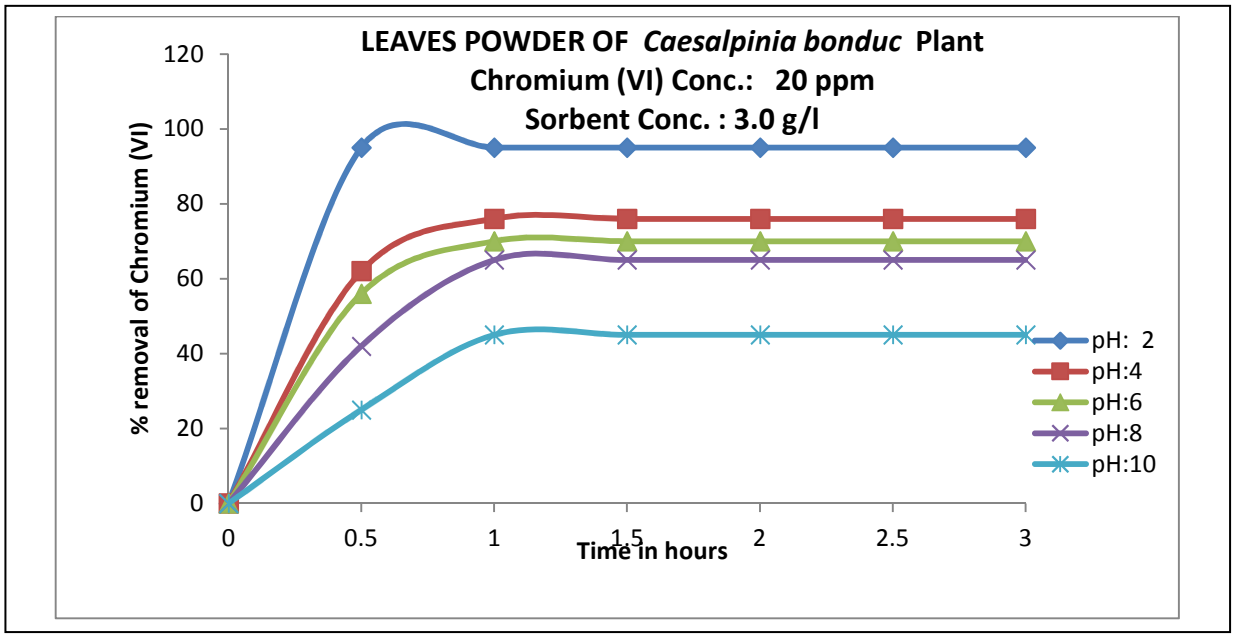

Fig.-4a : Effect of equilibration time on the extraction of Chromium (VI)

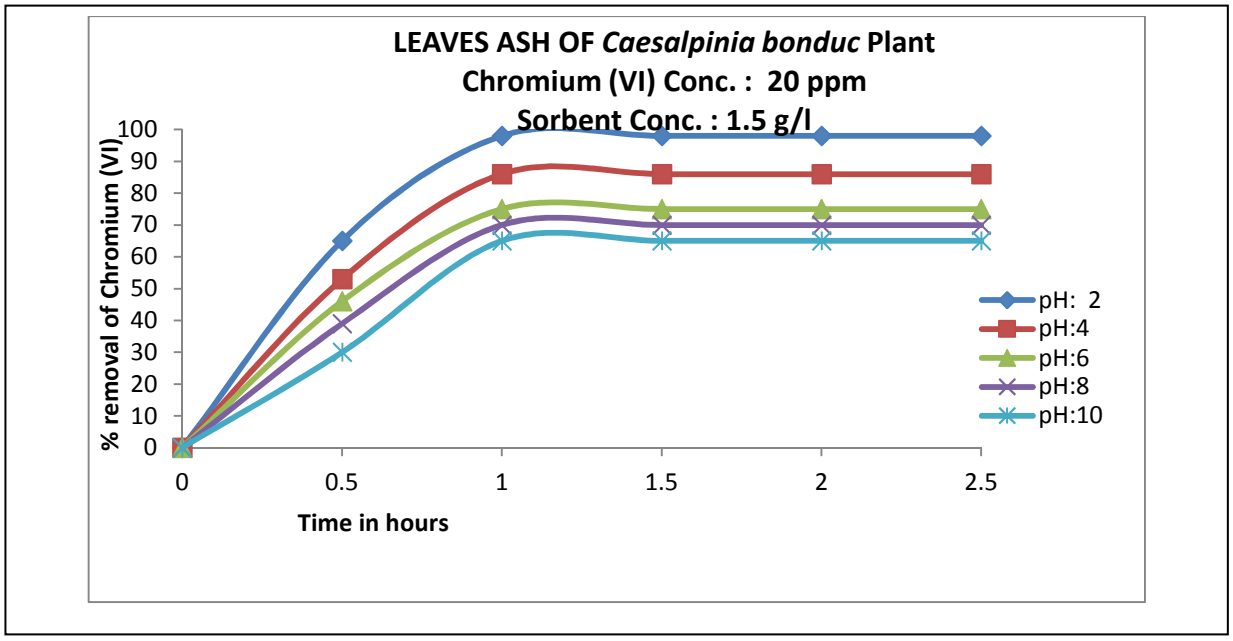

Fig.-4b: Effect of equilibration time on the extraction of Chromium (VI)

Adsorbent Concentration

The influence of dosage of adsorbent on the extraction of Cr (VI) was investigated by changing the concentrations from 0.25g/l to 2.0g/l while keeping the other extraction conditions at optimum levels (Fig.- 5).

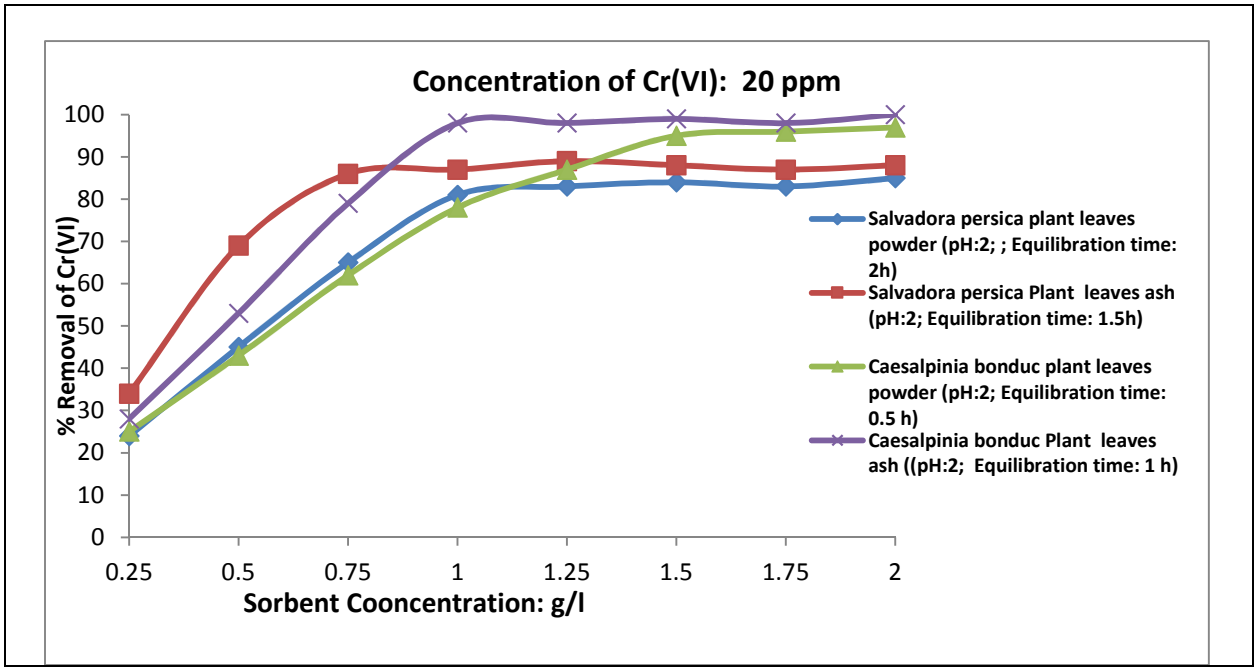

Fig.- 5: Sorbent Concentration Vs Removal of Cr(VI)

It may note from Fig.-5 that % removal is more with low adsorbent concentration and after certain dosage, there is no proportional increase of adsorption with further increase in the adsorbent dosage. For example, with the *Salvadora persica plant* leaves powder, there is a proportional increase in the removal of Cr (VI) until the dosage reaches: 1.0 g/l and after that, no significant rise is noticed (Fig.-5). With *Salvadora persica plant* leaves ash, removal is increased from 34% to 86% with the increase in dosage from 0.25 to 0.75 g/l and after that, there is no significant increase in the % removal of Cr (VI). In the case of leaves of *Caesalpinia bonduc plant,* the % removal increases from 25% to 95% as the dosage is increased from 0.25 g/l to 1.5 g/l and then onwards, there is no marked removal. With the ashes of leaves of *Caesalpinia bonduc plant,* the increase is from 28% to 98% as the sorbent dosage increases from 0.25 g/l to 1.0 g/l and after that, there is no significant removal.

The adsorption capacity is observed to be 16.2 mg/g and 22.9 mg/g for the adsorbents derived from leaves and ashes of *Salvadora persica* plant respectively. In the case of the adsorbents obtained from leaves and ashes of *Caesalpinia bonduc plant*, the sorption capacity is observed to be 12.7 mg/g and 19.6 mg/g respectively.

Initial Concentration

The initial concentration of Cr (VI) has an influence on the % removal of Cr (VI). % removal is decreased with increase in the concentration of Cr (VI) as is evident from the Fig.-6.

As the Cr (VI) concentration is varied from 5 to 80 ppm, the % removal of Cr(VI) is decreased from 90.0% to 20% for the leaves powder *of Salvadora persica plant* (pH 2; sorbent dosage: 1.0 g/l; equilibration time: 2h) and 95.0 % to 25.0% (pH 2; sorbent dosage: 0.75 g/l; equilibration time: 1.5h) for the ash of leaves of *Salvadora persica* plant. In the case of *Caesalpinia bonduc plant* adsorbents, the decrease is from 100% to 30.0% with leaves powders (pH 2; sorbent dosage: 1.5 g/l; equilibration time: 0.5 h) and 100% to 40.0% with the leaves ash (pH 2; sorbent dosage: 1.0 g/l; equilibration time: 1.0 h) as the initial concentration of Cr (VI) is varied from 5 to 80 ppm.

These observations may be accounted for the fact that at low concentrations of adsorbate, Cr(VI), the availability of active sites on the fixed amount of adsorbent are more and hence, more adsorption. But

with enhancing the adsorbate concentration, the demand for adsorption sites is more. As the adsorbent amount is fixed, the sites are also fixed and this result in a situation where in the demand for active sites of adsorption is more but only a fixed number of sites are available. This results in the decrease in % of adsorption as the concentration of adsorbate, Cr(VI) is increased.

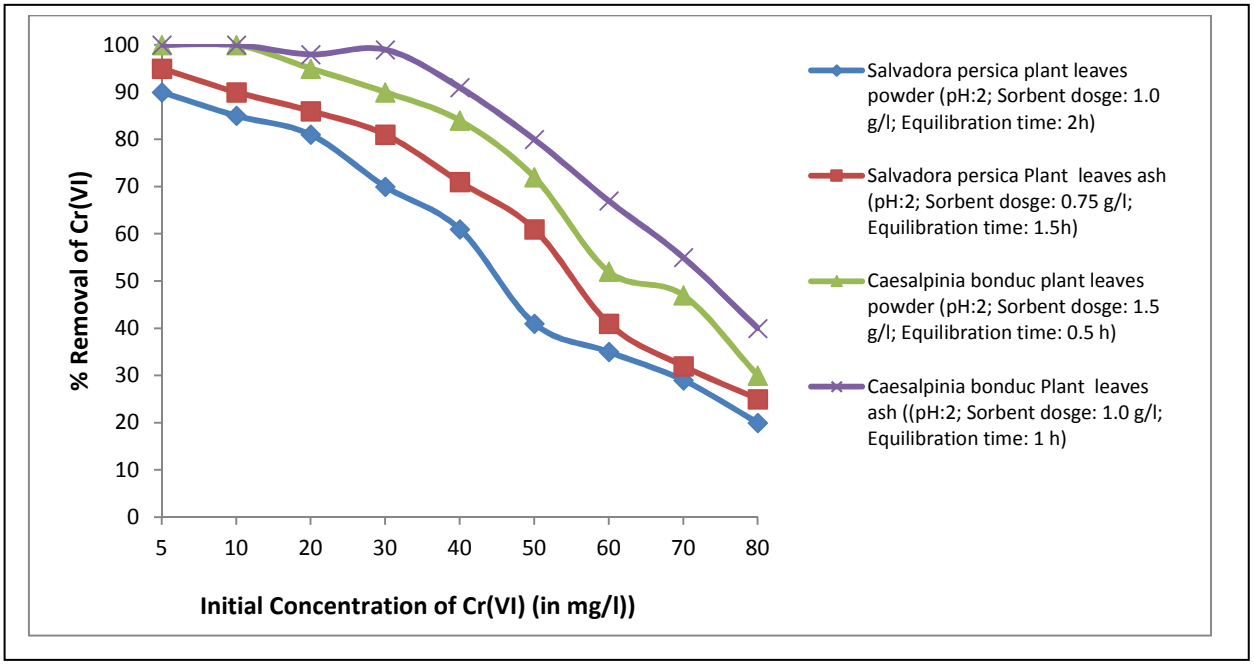

Fig.-6: Effect of Initial concentration of Cr(VI) on % removal of Cr(VI)

Influence of Co-ions

The observed data on the interferences of co-ions on the extraction of Cr (VI) are presented in Table -1. It is interesting to note that some cations markedly enhanced the % removal. For instance, with *Salvadora persico plant* leaves powder as adsorbent, the removal of Cr(VI) is 84.0% but in the presence of foreign cations, it is increased to 96.3% for Ca^{2+}, 90.0% for Cu^{2+}, 98.0% for Mg^{2+}, 83.0% for Ni^{2+}, 91.0% for Zn^{2+} and 89.0% for Fe^{2+} ions. This kind of synergetic enhancement in the % removal is phenomenal and needs further investigations. The interference of mono valent anions is marginal. But PO_4^{3-}, SO_4^{2--} *and* CO_3^{2-} are interfering in the order: $PO_4^{3-} > SO_4^{2--} > CO_3^{2-}$.

Table - 1: Influence of Co-ions on the % removal of Chromium (VI)

S. No.	Interfering ions fivefold excess	% of extraction in synthetic water samples: Cr(VI) conc. 50.0 ppm			
		Salvadora persico plant leaves		*Caesalpinia bonduc* plant leaves	
		Activated thermally (size:<75µ): pH:2;2.0 h and 1.0 g/l	Ash pH:2;1.5 h and 0.75 g/l	Activated thermally (size:<75 µ) pH:2 ; 0.5 h and 1.5 g/l	Ash pH:2;1.0 h and 1.0 g/l
	Without interfering ions	81.0%	86.0%	95.0%	98.0%
1	NO_3^-	80.0%	82.0%	93.0%	94.0%
2	Cl^-	80.5%	85.0%	93.0%	95.0%
3	F^-	80.0%	83.0%	94.0%	95.1%
4	PO_4^{3-}	62.0%	65.0%	70.3%	73.2%
5	SO_4^{2-}	64.0%	67.0%	77.0%	78.5%

6	CO_3^{2-}	79.8%	78.0%	87.0%	88.0%
7	Ca^{2+}	96.3%	92.0%	96.0%	93.0%
8	Cu^{2+}	90.0%	95.1%	97.0%	96.0%
9	Mg^{2+}	98.0%	92.0%	92.0%	93.0%
10	Ni^{2+}	83.0%	93.0%	94.0%	98.0%
11	Zn^{2+}	91.0%	92.0%	96.0%	96.0%
12	Fe^{2+}	89.0%	85.0%	93.0%	94.0%

Applications

The methodologies based on the adsorbents developed in the present work, were applied to the effluents or polluted water and the observations were noted in Table -2. It is inferred from the Table that substantial amounts of Chromium are removed.

With the leave of *Salvadora persico plant*, percentage removal is varied from 70.5 % to 77.5% while with their ashes, it is varied from 79.9% to 82.5%. In the cases of *Caesalpinia bonduc* plant, % removal varies from 86.7% to 91.1% with leaves powder and 90.0% to 93.5% with their ashes. Relatively *Caesalpinia bonduc plant* based adsorbents are more effective than *Salvadora persico* plant based adsorbents.

Table - 2: Removal of Chromium (VI) from the samples collected from divergent sources

Samples	Cr(VI) present in sample	% of Maximum extractability			
		Leaves of *Salvadora persico* plant		Leaves of *Caesalpinia bonduc* plant	
		Thermally activated powders (mesh:75 µ) pH:2;2.0 h and 1.0 g/l	Ashes pH:2;1.5 h and 0.75 g/l	Thermally activated powders (mesh:75 µ) pH:2 0.5 h and 1.5 g/l	Ashes pH:2; 1.0 hrs and 1.0 g/l
1: Tannery Industry Effluents					
1	10.5 ppm	77.0%	82.0%	90.5%	92.0%
2	21.0 ppm	75.0%	80.0%	91.0%	93.0%
3	25.0 ppm	78.0%	81.0%	89.5%	90.0%
2: Chromate Plating Industry Effluents					
1	15.0 ppm	72.0%	82.0%	90.0%	93.0%
2	22.0 ppm	73.5%	82.5%	89.5%	94.0%
3	26.8 ppm	71.5%	80.0%	90.6%	93.0%
3: Polluted Lake Water					
1	10.0 ppm	70.5%	79.9%	88.5%	93.5%
2	15.0 ppm	73.0%	81.0%	87.0%	92.0%
3	20.0 ppm	77.5%	82.5%	86.7%	90.0%

CONCLUSION

1. Bio-sorbents derived from leaves of *Salvadora persico* and *Caesalpinia bonduc* plants are investigated for their adsorption nature towards the Cr (VI) ion.
2. Various extraction conditions such as pH, sorbent dosage, equilibration time and initial concentration are optimized for maximum Cr (VI) extraction from the water. Maximum removal of 81.0% and 86.0% are observed for leaves powder and their ashes of *Salvadora persico plant* respectively at the other optimum conditions as specified in Table-1 and 2. With *Caesalpinia bonduc* plant leaves, the removal is 95.0% and with their ashes, the % removal is 97.0%

3. The adsorption capacities of the four investigated adsorbents are: 16.2 mg/g (leaves) and 22.9 mg/g (ashes) for *Salvadora persica plant* and 12.7 mg/g (leaves) and 19.6 mg/g (ash) for *Caesalpinia bonduc plant.*
4. Effects of co-ions on Cr (VI) extraction are studied. Some cations synergistically enhanced the percentage of extraction while mono valent anions marginally interfered. PO_4^{3-}, SO_4^{2-} and CO_3^{2-} have shown interference.
5. Bio-adsorbents based methodologies developed in this work are successfully applied for the removal of Chromium (VI) from various samples collected at divergent sources as is evident from Table-2.

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