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REMOVAL OF HEAVY METALS TOXICITY FROM MEDICINAL PLANTS USING NANO PARTICLES OF ACTIVATED CARBON AS ADSORBENTS

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ABSTRACT

Medicinal plants are raw material for herbal medicines. When the levels of heavy metals exceed in plants, it can induce a variety of acute and chronic effects in wide range of organism in various eco system. Medicinal herbs are easily contaminated by metals due to factors such as environment, pollution, atmosphere, soil, harvesting and handling. The control of heavy metals contents in medicinal plants represents one of the factors for the evaluation of quality. Since these plants originate from different growing areas, great difference in the uptake and concentration of heavy metals in the plant tissue can be expected. Heavy metals confined in plants finally enter the human body and may disturb the normal functions of central nervous system. Therefore a preliminary data on the various heavy metal content of the medicinal plants and removal of the heavy metal ions from contaminated medicinal plants such as *Strychnosnux-vomica* (Seeds), *Citrullus colocynthis* (Seeds), *Strychnose potatorum* (Seeds) and *Corallocarpus epigeous* (Tuber) by adsorption technique. Adsorption technique is the best, economically viable and efficient method for the treatment of heavy metal ion contaminated in medicinal plants.

Key words: Adsorption technique, Nano Particlels of activated carbon, Heavy metals and Atomic absorption spectrometry.

1.0 Introduction

The control of heavy metals contents in medicinal plants represents one of the factors for the evaluation of quality. Since these plants originate from different growing areas, great difference in the uptake and concentration of heavy metals in the plant tissue can be expected. The high heavy metal contents in some plants arise from their ability to accumulate particular metals especially lead. Heavy metals like As, Cd, Cu, Hg, Pb etc., are toxic metals cause health problems while consuming the medicinal plants contaminated with heavy metals.

Therefore a preliminary data on the various heavy metal content of the medicinal plants and various parts will be desirable before it is taken up for the preparation of the herbal medicine and suitable analytical techniques has to be made use of the determination of their elements.

2.0 Removal of heavy metals

Removal of these toxic metal ions are the best approaches to control the metal pollution problem and safeguard the environment and human health.Activated carbon is widely used as an adsorbent in industry due to its high adsorption capacity. This capacity is related to the pore structure and chemical nature of the carbon surface in connection with preparation conditions.

The present investigation deals with the studies of simultaneous removal of heavy metal ions As (III), Cd(II), Cu(II), Hg(II) and Pb(II) by adsorption on the commercially available adsorbents Nano Particals of Activated Carbon (NPAC). Results of various parameters like Initial concentration of metal ions in medicinal plants, Contact time and pH of the solution affecting the adsorption process, kinetic of adsorption are presented and discussed in this chapter.

2.1 Adsorption studies of heavy metals

The parameters which warrant the evaluation for investigating the mechanism of adsorption and deciding the efficiency of the simultaneous removal of the metal ions by adsorption on NPAC are

- Concentration of the metal ions
- Dose of the adsorbents
- Contact time between adsorbate and adsorbent
- pH of the solution
- Stirring speed.

The variations in the percentage removal of metals ion with its concentration are shown in relevant datas obtained from these experiments are given in table 1. It was observed that the percentage removal of heavy metals ions is low at higher concentrations and gradually increases the concentration of the heavy metals ions are decreases. This is due to the fact that after the formation of mono-ions layer at concentration over the adsorbent surfaces, further formation of the layer is highly hindered at higher concentration due to the interaction between metal ions present on the surface and in the solution.

When particles decreased to the nanometer range, an increasing fraction of the atoms are exposed to the surface, giving rise to excess surface energy should be prone to adsorb molecules onto their surfaces in order to decrease the total free energy.

3. Experimental

3.1 Procedures

1. Dry ashing:

Dry ashing provides good precision and rapid. Dry ashing can be used for preparation of plant samples for the determination of Cd, Cu, As, Hg and Pb. This method is relatively free from reagent contamination.

2. Wet digestion or wet oxidation.

This method is the preferred route for simultaneous oxidation of organic matter and also for dissolution of trace metal content of the sample. The main advantage of this method is that, it prevents elemental loss by volatilization, because the digestion takes place at a lower temperature

3.2. Chemicals

Analytical grade Nitric acid, hydrochloric acid and hydrogen peroxide (Merck, India) were used as received. Standard sample solutions of As, Cd, Cu, Hg and Pb, (1000 mg/ml) were obtained from Merck (Germany). All the solutions were prepared from triply distilled water. The commercially available Nano particles of activated carbon (Norit DX ULTRA) obtained from Netherlands.

3.3 Sample preparation

Known quantity of samples were transferred into a silica crucible and kept in a muffle furnace for ash at 450^{o} C for 3 hours and then 5 ml of 6M HCl was added to the crucible. Then, the crucible containing acid solution was kept on a hot plate and digested to obtain a clean solution. The final residue was dissolved in 0.1 M HNO_3 solution and made up to required volume.

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Table-1 Effect of contact time on the percentage removal of **Heavy Metals (** Cu, Cd, As, Pb & Hg) by Adsorption of Nano Particles of Activated Carbon **(NAPC), Stirring time: 10- 60 minutes** -Quantity of NPAC used: **0.1/100 (g/ml)**

Medicinal plants, part used & Initial Conc of Heavy Metals (ppm)	Time (mins)	Cu		Cd		As		Pb		Hg	
		Quantity absorbed (ppm)	% absorbed	Quantity absorbed (ppm)	% absorbed	Quantity absorbed (ppm)	% absorbed	Quantity absorbed (ppm)	% absorbed	Quantity absorbed (ppm)	% absorbed
Strychnosnux-vomica (Seeds) Cu -14.91 Cd – 0.70 As –0.126 Pb – 53.35 Hg- 0.115	10	10.55	29.24	0.40	42.86	0.081	35.71	29.34	45.00	0.075	34.73
	20	7.25	51.37	0.17	75.71	0.023	81.75	13.35	74.98	0.061	46.91
	30	3.50	76.53	0.08	88.57	0.004	96.83	8.44	84.18	0.029	74.76
	40	0.85	94.30	0.03	95.71	0.004	96.83	3.25	93.91	0.009	92.17
	50	0.15	98.99	0.03	95.71	0.004	96.83	0.55	98.97	0.003	97.39
	60	0.15	98.99	0.03	95.71	0.004	0.00	0.55	98.97	0.003	97.39
Citrullus colocynthis (Seeds) Cu –25.14 Cd – 0.79 As –0.189 Pb – 39.65 Hg – 0.028	10	17.55	30.19	0.38	51.90	0.125	33.86	28.67	27.69	0.013	53.74
	20	13.64	45.74	0.21	73.42	0.076	59.79	19.58	50.62	0.007	75.09
	30	9.40	62.61	0.10	87.34	0.028	85.19	10.94	72.41	0.003	89.32
	40	4.65	81.50	0.06	92.41	0.006	96.83	4.55	88.52	0.003	89.32
	50	1.35	94.63	0.02	97.47	0.003	0.00	1.15	97.10	0.001	96.44
	60	0.55	97.81	0.02	97.47	0.003	0.00	1.15	97.10	0.001	96.44
Strychnose potatorum (Seeds) Cu -39.74 Cd – 0.85 As –0.058 Pb – 68.59 Hg – 0.134	10	28.65	27.91	0.52	38.82	0.030	47.92	45.97	32.98	0.088	34.08
	20	20.15	49.30	0.33	61.18	0.019	67.01	32.46	52.68	0.063	52.81
	30	14.68	63.06	0.14	83.53	0.032	44.10	14.58	78.74	0.040	70.04
	40	8.55	78.49	0.08	90.59	0.007	87.85	9.65	85.93	0.011	91.76
	50	3.78	90.49	0.03	96.47	0.003	94.79	3.85	94.39	0.003	97.75
	60	1.10	97.23	0.03	96.47	0.003	94.79	0.75	98.91	0.003	97.75
Corallocarpus epigeous (Tuber) Cu -10.18 Cd – 0.42 As –0.055 Pb – 49.34 Hg – 0.061	10	6.98	31.43	0.28	33.33	0.028	48.81	30.57	38.04	0.030	50.90
	20	3.69	63.75	0.12	71.43	0.012	78.06	20.15	59.16	0.013	78.72
	30	1.15	88.70	0.05	88.10	0.003	94.52	9.86	80.02	0.007	88.54
	40	0.55	94.60	0.05	88.10	0.003	94.52	3.15	93.62	0.003	95.09
	50	0.55	94.60	0.05	88.10	0.003	94.52	1.50	96.96	0.003	95.09
	60	0.55	94.60	0.02	95.24	0.003	94.52	1.50	96.96	0.001	98.36

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Kinetic Adsorption on Nano Particles of Activated Carbon (NPAC) using Natarajan & khalaf equation

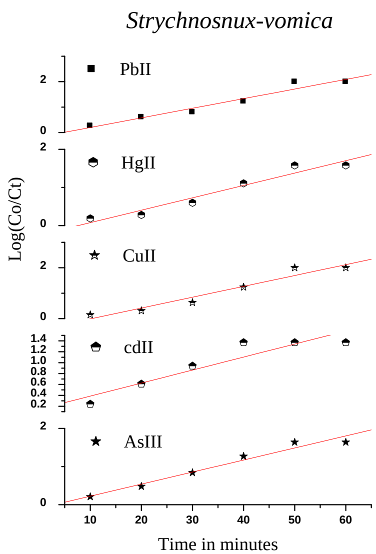


Fig: 1.1

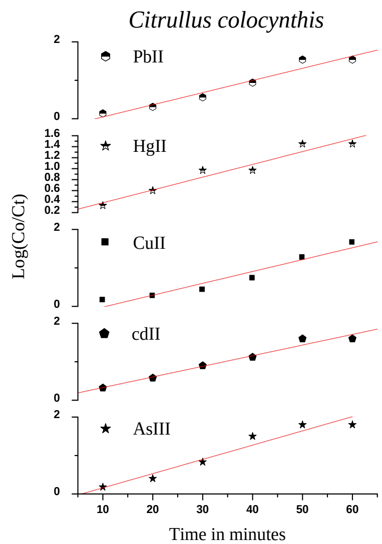


Fig: 1.2

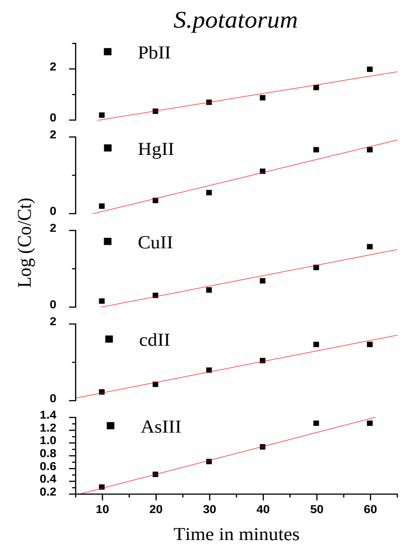


Fig: 1.3

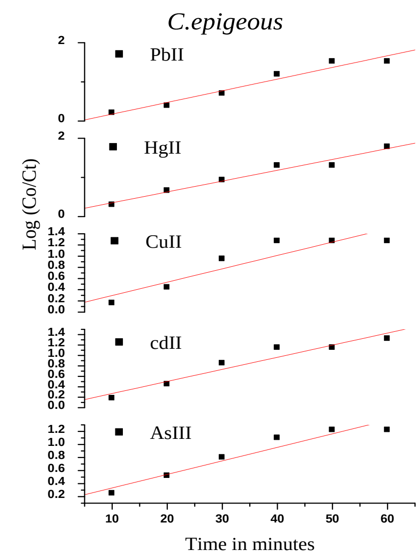


Fig: 1.4

Figure:1.1 – 1.4 Kinetic model for the removal of As^{3+}, Cd^{2+}, Co^{2+}, Hg^{2+} and Pb^{2+} ions

by Adsorption on (NPAC) using Natarajan & khalaf equation

4.0 Results and discussion

The effect of various experimental parameters like initial concentration of metal ions, contact time, dose of activated carbon and initial pH has been investigated by following batch adsorption technique at 27±1^{o}C. The percentage removal of metal ions increased with the decrease in initial concentration and increase in contact time, dose of adsorbent and initial pH. Adsorption data were modeled with, various first-order kinetic equations like Natarajan-Khalaf equations. These first order equations and model were found to be applicable. The kinetics of adsorption is found to be first order with intra-particle diffusion as the rate determining step. Removal of metal ions using activated carbons (NPAC) is found to be favorable and hence it could be used as an adsorbent for the removal of metal ions from medicinal plants.

Although far more attention has been paid towards the studies on the adsorption of metal ions, only fewer attempts has been made on the studies on the kinetics and mechanism of adsorption of metal ions on activated carbons (NPAC). The present study is therefore aimed at to study the removal of Arsenic^{3+}, Cadmium^{2+}, copper^{2+}, Mercury^{2+} and lead^{2+} ions by adsorption on activated carbons (NPAC) under various experimental conditions in order to optimise the process parameters and to apply the various, first order kinetic equations and models. The results of the present study will be very much helpful in designing the low-cost adsorbents for the removal of metal ions from the contaminated medicinal plants.

The adsorption experiments were carried out at 27±1^{o}C under batch mode under different experimental conditions in order to study the effect of initial concentration of metal ions, contact time, dose and initial pH. The results obtained are analysed and discussed.

4.1 Effect of initial concentration

The effect of initial concentration of metal ions on the extent of removal of Arsenic^{3+}, Cadmium^{2+}, copper^{2+}, Mercury^{2+} and lead^{2+} ions by adsorption on activated carbons (NPAC) was studied. The relevant data are given in Table 1.

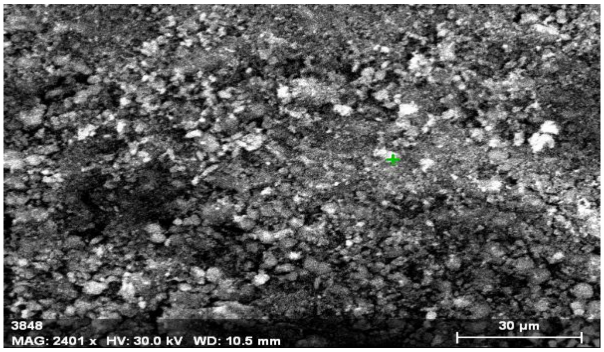
4.2 Effect of contact time

In the adsorption system, contact time plays a vital role, irrespective of the other process parameters affecting the kinetics of adsorption. The effect of contact time on the extent of removal of metal ions was studied. The extent of removal of metal ions increased with the increase in contact time and reached a constant value with the increase in contact time. The relative increase in the extent of removal of Arsenic^{3+}, Cadmium^{2+}, copper^{2+}, Mercury^{2+} and lead^{2+} ions respectively. The optimum contact time at which the maximum removal occurred is fixed as 60 minutes for nano particles of activated carbon for removal of As^{3+}, Cd^{2+}, Cu^{2+}, Hg^{2+} and Pb^{2+} ions.

The amount of heavy metals adsorbed at this point is the maximum under the particular operating conditions. The time variation curve is smooth and continuous and this indicates the formation of monolayer coverage on the outer interface of the adsorbent. Contact time required for the maximum removal of heavy metals by activated carbons (NPAC).

4.3 Scanning Electron Microscopy (SEM).

Activated carbon is widely used as an adsorbent due to its high adsorption capacity, high surface area, micro porous structure and high degree of surface respectively. The wide usefulness of carbon is a result of its specific surface area, high chemical and mechanical stability. The chemical nature and pore structure usually determine the sorption activity.



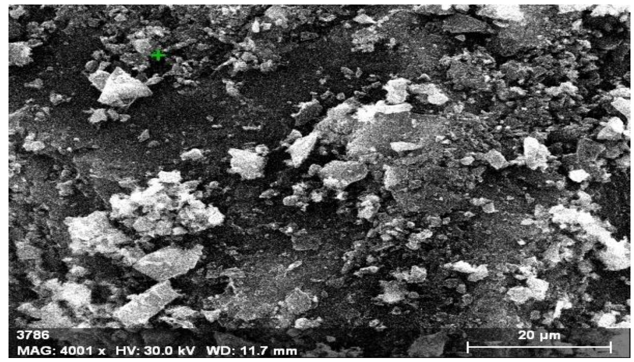
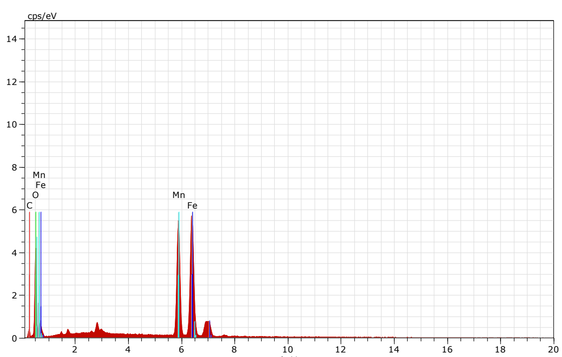


Figure-4.1 . Figure-4.2

Figure-4.1 &4.2 SEM image of Nano particles of activated carbon before and after adsorption

4.4 X-ray diffraction studies.

Activated carbon is widely used as an adsorbent due to its high adsorption capacity, high surface area, micro porous structure and high degree of surface respectively. An x-ray energy dispersion analysis (EDX) of the NPAC before and after treated with medicinal plants contains heavy metals and the composition of the heavy metals adsorbed are shown in the figure 4.3&4.4 . The pattern shows that, the presences of heavy metals are identified



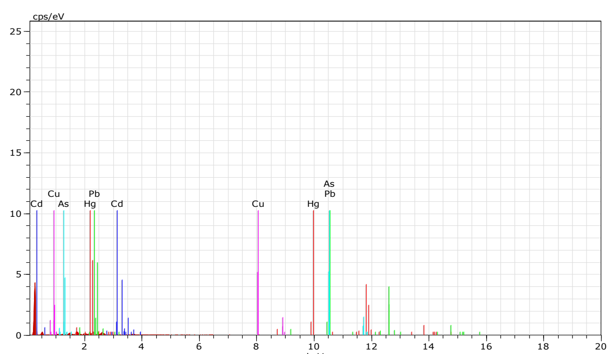


Figure-4.3 Figure-4.4

Figure-4.3 &4.4 SEM image of Nano particles of activated carbon before and after adsorption

4.5 XRD Patterns

The granular and nano particles of activated carbon samples were characterized by the XRD technique to determine their crystal phases and purity. Figure 4.5 shows the diffraction patterns of granulated activated carbon used in the study and shows two distinctive diffraction peaks corresponding to (002), (100) lattice plans, which could be indexed. No impurity peaks were detected by XRD. Figure -4.5 shows four distinctive diffraction peaks corresponding to (002), (100), (102) and (103)

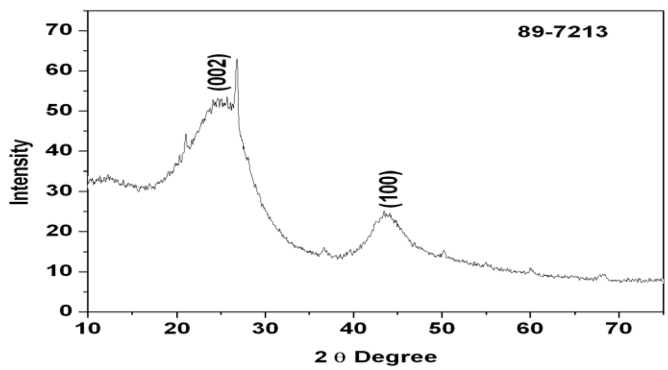


Figure -4.5

Figure -4.5 XRD Pattern of nano particles of Granulated activated carbon

5.0 Conclusion

The purpose of the current study was to standardize various indigenous medicinal plant for heavy metals contamination and to make awareness among the public regarding its safer use and collection areas, containing high level of heave metals and their adverse health effects.

The some of the plant materials and various parts have beyond the permissible limits of heavy metals and this study concluded that the plant grown on polluted area has high risk of having the heavy metals concentration above the permissible limit for each of them as compared to the unpolluted areas. The current findings indicate that the medicinal plant used for various types of ailments must be checked for heavy metals contamination in order to make it safe for human consumption. The current study will also provide useful reference data for the standardization of medicinal plant materials.

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