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# Treatment of Effluents from Jewellery Industries by using Activated Carbons Prepared from *Epiphyllum Oxypetalum (Cactace)* Wastes

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## Abstract

*Epiphyllum oxypetalum is a species of cactus and one of the most commonly grown of the Epiphyllum species. It is one of the under-utilized resources available in the tropical regions of the globe and can be used as a substitute for digitalis. The Shoshone Indian tribe calls the night blooming Cereus "Pain in the heart" and used it for heart pain. Scanty work was reported on the phytochemical properties of leaf extract and no documented research work was reported on its leaf and flower for assessment of adsorption studies. Thus the present investigation was carried out to access the adsorptive studies of plant wastes. The activated carbons was prepared Epiphyllum oxypetalum waste by $H_3PO_4$ activation. The adsorption kinetics of jewellery effluents was studied by the activated carbons, which are suitably described by simple kinetic models, pseudo first order and pseudo second order equations. The optimum temperature was found to be 30 $^0$C and the adsorption data fitted well onto Freundlich, Langmuir and Tempkin adsorption isotherm models. The high surface area of activated carbons were demonstrated to be promising adsorbents for pollution control and for other applications.*

**Keywords:** *Activated carbon; Adsorption; Aurocyanide; Epiphyllum oxypetalum waste.*

## 1. INTRODUCTION

*Epiphyllum oxypetalum* is a species of cactus and one of the most cultivated species in the genus. It is a variety of night blooming Cereus. Oxypetalum (Lat.) = with acute petals, refers to the acute petals of this species. *Epiphyllum oxypetalum* was the most commonly grown of the *Epiphyllum species*, and it is known under several common names including Night-blooming Cereus, Dutchman's Pipe, Queen of the Night, Wijaya Kusuma (Indonesian), Nishagandhi in Hindi and Marathi, Table 1.

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The plant kingdom is a large reservoir of pharmacologically active molecules and large number of plant-derived medicines now commercially available. (Newman *et al.* 2007). Traditionally used medicinal plants produce a variety of compounds of known therapeutic properties. The present investigation clearly reveals the adsorption nature of this plant.

In 1894, Johnson patented the process of gold recovery using wood charcoal from a cyanide leached solution, but it was not until the early 1950's that Zadra et al. from the US Bureau of Mines developed the activated carbon based technology (Johnson, 1894; Zadra *et al.* 1952).

**Table 1. Taxonomy of *Epiphyllum Oxypetalum* Kingdom Planta**

| Kingdom | Plantae |
|---|---|
| Sub Kingdom | Tracheobionta |
| Superdivision | Spermatophyta |
| Division | Magnoliophyta |
| Class | Magnoliopsida |
| Order | Caryophyllales |
| Family | Cactaceae |
| Genus | *Epiphyllum* |
| Species | *E* oxypetalum |
| Binomial name | ***Epiphyllum oxypetalum*** |

The relative ease at which gold adsorption takes place onto activated carbon and the subsequent desorption has generated significant interest from both the scientific and industrial fields. In particular, the gold mining industry has effectively used activated carbons to extract gold (Au) from crushed ore to maximise extraction rates for many years and hence reduce processing costs. This activated carbon is then used for the adsorption of Au (I), which is in the form of $Au(CN)_2^-$ ions that are present in the cyanide leach liquors that normally contain around 10 mg/L of Au (I) (Marsden and House, 1993).

## 2. MATERIALS & METHODS

### 2.1 Plant Material *(Epiphyllum Oxypetalum)*

The plant waste of Epiphyllum oxypetalum were collected from in and around the city of Coimbatore, India in December 2012. The plant was authenticated by Tamilnadu Agricultural Univeristy Coimbatore, India. Fig 1. Chemical activation is one of the possible methods to produce activated carbons(ACs). It involves pyrolyzing the feed stock in the presence of a chemical activating agent such as $ZnCl_2$, $H_3PO_4$, $H_2SO_4$, $Na_2SO_4$, $K_2CO_3$ etc. Phosphoric acid offers some advantages as activating agent, such as ease of recovery by washing with water. The optimum temperature for phosphoric acid activation of a number of precursors of vegetable origin is 723 K (Suarez-Garcia *et al*. 2001; Molina–Sabio *et al*. 1995; Laine *et al*. 1989).

**Fig. 1: *Epiphyllum oxypetalum* flower and leaves**

### 2.2 Surface Area and Pore Size Distribution Analysis

The $N_2$ adsorption-desorption isotherms of activated carbons were measured at 77 K using a gas sorption analyzer (NOVA1000, Quanta chrome Corporation) in order to determine the surface areas and the total pore volumes Fig. 3. The surface areas were calculated using the BET equation. In addition, the t-plot method was applied to calculate the micropore volumes and external surface areas (mesoporous surface areas). The total pore volumes of the prepared carbons were estimated from the liquid volume of adsorbate ($N_2$) at a relative pressure of 0.99. All the surface areas were calculated from the nitrogen adsorption isotherms by assuming the area of a nitrogen molecule to be 0.162 $nm^2$ (Gregg *et al.* 1982).

Here we report on the porous texture characteristics of ACs prepared by $H_3PO_4$ activation of *Epiphyllum oxypetalum* wastes. These *Epiphyllum oxypetalum* wastes were impregnated with aqeous solution of phosphoric acid following a variant of the incipient wetness method, as described elsewhere. The pyrolysis treatments were carried out in a vertical tubular reactor made of quartz, using in all cases 10 g of

impregenated and dried material. The pore size distribution of activated carbon can be classified into three groups. The groups are based on the size range of pore diameters found in activated carbon: the micropores are less than 2 nm in size, the transition pores or mesopores range in size from 2 nm to 50 nm and the macropores are greater than 50 nm (Dubinin, 1960). The porosity of activated carbon is a very important property since it greatly influences the adsorption of auro cyanide. The macropores provide passage to the micropores and access to the interior of the carbon matrix. The micropores created during the activation process are responsible for the very large surface area used for Au adsorption. Typically, activated carbons have BET surface areas in the range from 500 to 1400 $m^2/g$ and the relative (%) contribution of each pore structure to the total surface area and adsorption process vary in the order: macropores (0%) < mesopores (5%) < micropores (95%) (Lartey *et al.*1999) Fig. 2 .

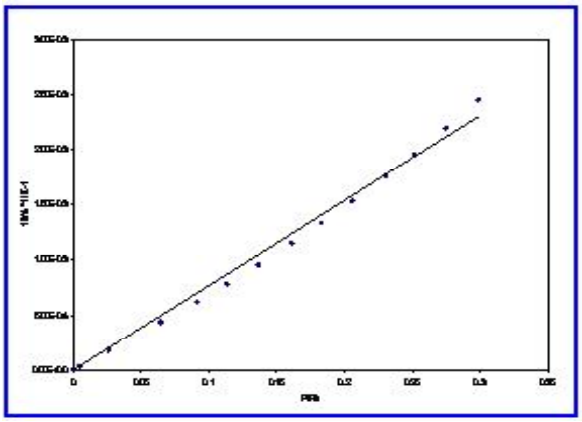

**Fig. 2: BET Plot o EOAC**

### 2.3 Scanning Electron Microscope (SEM)

The surface morphologies of activated carbons were observed with SEM, Philips SIRON with EDX facility at IISc, Bangalore.

### 2.4 Gold Adsorption Mechanism

In the case of Au adsorption on activated carbons there are three rate dependent diffusion mechanisms that have been proposed to account for the adsorption of Au based on contact time and gold loading. These mechanisms are: (i) film diffusion (mass transport), (ii) pore diffusion, and (iii) surface diffusion (Muir, 1982; Nicol *et al*. 1984a; 1984vb; Jones *et al.* 1988, 1989; Le Roux *et al*. 1991; Demopoulos and Chen, 2004; Pleysier *et al.* 2008). The initial rate of Au cyanide adsorption is rapid, with adsorption occurring at the most accessible sites in the macropores and possibly the mesopores. Under these conditions the rate is

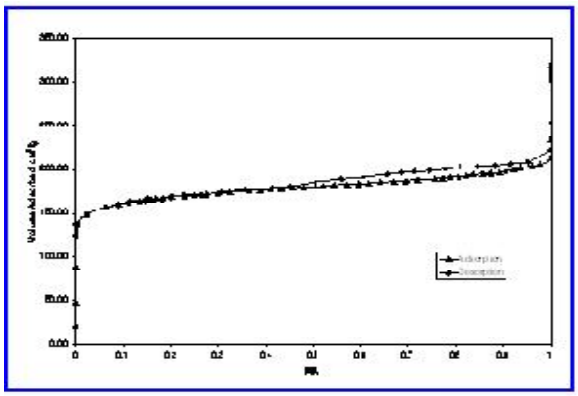

**Fig. 3: Shows adsorption – desorption isotherms of $N_2$ at 77K on ACs from *Epiphyllum oxypetalum* wastes with $H_3PO_4$**

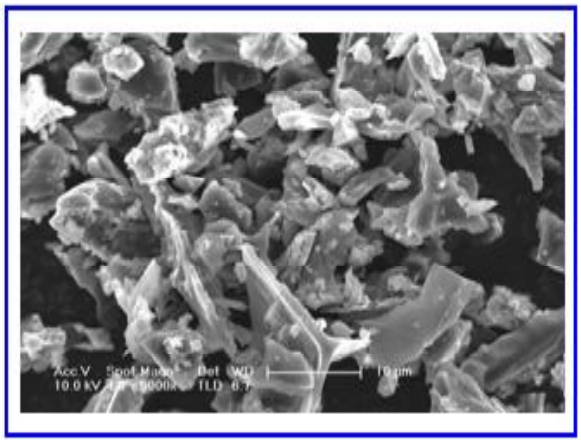

**Fig. 4: SEM image of *Epiphyllum oxypetalum* wastes with $H_3PO_4$**

controlled by the film diffusion (mass transport) of Au cyanide species onto the carbon surface. However, with time the diffusion rate decreases as equilibrium is approached. Once adsorption capacity has been achieved in the macropores and mesopores, a pseudoequilibrium is established beyond which adsorption must take place in the micropores (Marsden and House, 1993). The diffusion of gold cyanide species onto the micropores structures within the carbon structure is a much slower process, since the boundary layer or film diffusion process is complicated by the length and tortuosities of the inherent pore structures. This phenomenon has been confirmed by detailed micro-tomography studies which indicate that there is poor diffusion of gold cyanide within micropores. The findings of these studies exclude the contribution made by pore diffusion to the rate limiting mechanisms and support a surface diffusion mechanism at higher gold loadings (Pleysier *et al*. 2008).

## 3. RESULTS &DISCUSSION

### 3.1 Carbon Properties

The characteristics of activated carbon prepared from Epiphyllum oxypetalum wastes in different activation methods were given in the Tables 2 and 3. Evaluation of the characteristics of carbon (Table 2) prepared from different processes indicates that the density of carbon is in comparable range. The uniform bulk density values imply that *Epiphyllum oxypetalum* wastes has high resistance towards the action of chemicals and the pore system was also uniform, irrespective of the processing methods.

From Table 2 the moisture content was found to be high in the case of carbon obtained by $H_3PO_4$ process. This implies that extensive porosity has been introduced by this process in the carbon structure. Even though moisture content of the carbon has no effect on its adsorptive power, it dilutes the carbon which necessitates the use of additional weight of carbon during treatment process. Among the carbon prepared by various methods, the carbon obtained by carbonate process was found to contain less moisture compared to the carbon obtained by other processes. Ash content generally gives an idea about inorganic constituents associated with carbon obtained by different carbonization methods. The ash content values from Table 2 indicate that the overall ash content for all the varieties of carbon was comparatively less. This may be attributed to lower inorganic content and higher fixed carbon.

Solubility studies of carbon in acid and water were performed to evaluate the amount of impurities present in the carbon prepared by different carbonization processes. From Table 2 the data pertaining to the matter

**Table 2. EOAC waste Activated Carbon properties in various activation process**

| No | Properties | $ZnCl_2$ | $H_3PO_4$ | $H_2SO_4$ | $Na_2SO_4$ | $K_2CO_3$ |
|---|---|---|---|---|---|---|
| 1 | pH | 6.1 | 6.2 | 5.8 | 8.9 | 8.7 |
| 2 | Moisture Content, % | 15.8 | 17.6 | 13.8 | 7.1 | 6.4 |
| 3 | Ash Content, % | 10.12 | 14.2 | 8.34 | 8.76 | 10.79 |
| 4 | Bulk density, $g/cm^3$ | 0.419 | 0.419 | 0.853 | 0.526 | 0.8272 |
| 5 | Porosity, $\zeta p$ | 0.623 | 0.756 | 0.546 | 0.629 | 0.233 |
| 6 | Matter Soluble in Water, % | 1.88 | 1.18 | 1.92 | 2.8 | 2.92 |
| 7 | $pH_{zpc}$ | 3.6 | 4.6 | 3.3 | 6.6 | 7.7 |

**Table 1.3 Porosity Characteristics of EOAC Waste**

| $S_{BET}$ $m^2/g^{-1}$ | $S_{Ext}$ $m^2/g^{-1}$ | $V_{micro}$ $cm^3/g$ | $S_{micro}$ $m^2/g$ | $V_{total}$ $cm^3\ g^{-1}$ | APD Å | % micro | % meso |
|---|---|---|---|---|---|---|---|
| 621.4 | 142.46 | 0.1977 | 478.97 | 0.492 | 31.7 | 40.16 | 0.91 |

soluble in water and in acid indicate that the carbon prepared by all other processes exhibits moderate level of impurities except the carbon obtained by acid process. The high value of water soluble matter in the carbon prepared by carbonate process indicates that a large amount of carbonate salts would have been incorporated in the carbon structure. The acid leachable matter was also found to be higher in the carbon prepared by the carbonate process.

The carbon derived by acid process was acidic in nature with moderate conductivity compared to the carbon prepared by other carbonization processes, this may be due to the incorporation of acidic groups in the carbon structure. Chloride impregnated carbon was also found to exhibit moderate conductivity. This may be due to the development of exchangeable sites on the surface of the activated carbon.

The surface area of carbon prepared by various processes is found to be in the following order

$$H_3PO_4 > N_2 > ZnCl_2 > H_2SO_4 > Na_2SO_4 > K_2CO_3$$

The higher surface area of carbon prepared by $H_3PO_4$ process may be due to restricted pore shrinkage during activation. The iodine number of all carbon was found in the same order as that of surface area. This indicates that the carbon prepared by Phosphoric acid ($H_3PO_4$) process has the maximum adsorption capacity. Carbon samples with high surface area are considered to be superior for adsorption of organic substances. From Table 3 EOAC have higher BET surface area, micropore and mesopore.

From Table 2 it is clear that acid process gives more yields followed by carbonate process. When compared to other treatment processes of the acid process, $H_2SO_4$ gives high yield, due to its high charring capacity. All carbon samples except $H_3PO_4$, $H_2SO_4$ and $Na_2SO_4$ show significant nitrogen uptake at low relative pressure. This can be ascribed to the strong interaction between nitrogen molecules and the wall with closely spaced pores. Nitrogen adsorption for the $Na_2SO_4$ nd $K_2CO_3$ samples was low, since the samples have such a low degree of activation and a low pore volume.

Carbon samples from $H_3PO_4$, $H_2SO_4$ have higher BET surface area, micropore and mesopore volume. As reported by Yu and Chou (2000) the adsorption onto activated carbon surface was not only coverage at the surface of adsorbent and also filling of micropores (Yu and Chou, 2000). Mangun *et al.* (1998) also thought the total micropore volume is not important since only a small fraction of the pores is actually occupied during adsorption. Pelekani and Snoeyink (2001) also reported that the compounds are preferentially adsorbed into pores that are similar in size to the adsorbate because of greater number of contact points between adsorbent and adsorbate.

The highest surface area was obtained for activated carbon prepared using *Epiphyllum oxypetalum* wastes waste by $H_3PO_4$ process followed by activation at 800 °C under a nitrogen atmosphere (621.00 $m^2/g$). Many studies on preparation of activated carbons using $H_3PO_4$ process have been reported. Rodriguez- Reinoso *et al.* (1980) have done much work on preparation of activated carbon, including $H_3PO_4$ process.

The TG & STD curves of the activated carbon samples heated from 50 °C to 800 °C at 10 °C/minute. These curves clearly illustrate that the carbon samples begin to lose weight at about 60 °C due to the volatilization of small molecules and it started to lose weight intensively between 700 °C and 800 °C due to the pyrolysis of aromatic molecules. The weight loss and differential plot for the starting material indicates that essentially all weight loss starts at about 60 °C and corresponds to full burn off of the activated carbon. On the activation carbon samples, the weight loss starts at a higher temperature, which suggests the conversion of carbon can be taken as the oxidation barriers.

The morphological study by SEM of the above adsorbents Fig 4 reveals the highly porous nature. From the SEM results, it was found that there are holes and cave type openings on the surface of the specimen that would definitely have increased the surface that is available for the adsorption (Khattri *et al.* 1999). The amount of gold adsorption is shown in the Fig 5 with time.

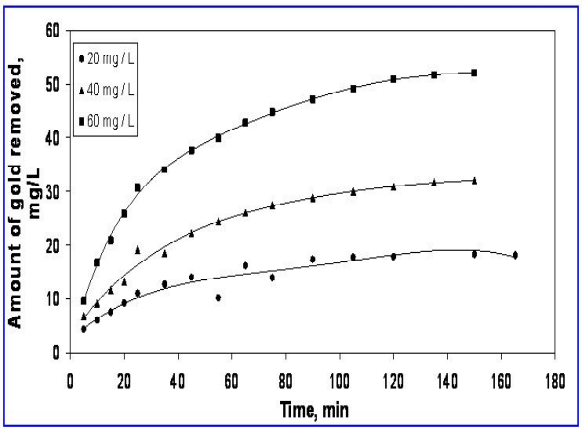


**Fig. 5: Effect of Initial concenration on amont of gold removal**

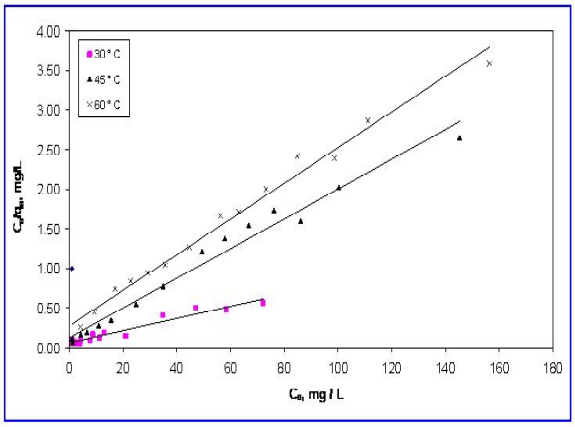


**Fig. 6: ffect of temperature variation on langmuir plot for gold adsorption**

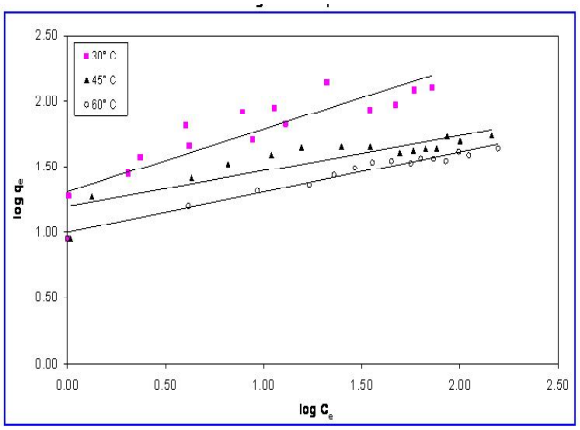


**Fig. 7: Effect of temperature variation on fruendlich plot for gold adsorption**

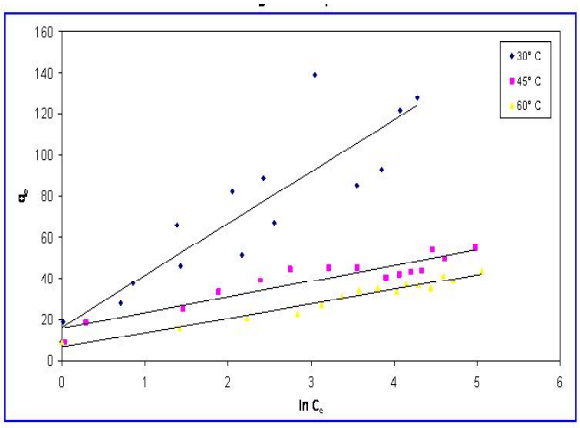


**Fig. 8: Effect of temperature variation on temkin plot for gold adsorption**

The present adsorbate –adsorbent system ia found to obey Freundlich,Langmuir and Temkin model (Fig. 6- 8)

## 4. CONCLUSION

This preliminary research project has revealed that activated *Epiphyllum oxypetalum* waste based carbons have a strong affinity for the Au (I complex) and have the potential to absorb significant levels. Further more, *Epiphyllum oxypetalum* waste based carbons could be an attractive alternative or a complementary supplement to the coconut shell based carbons that is currently being used in the Au extraction industry.

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