Research Article

Journal of Environmental Nanotechnology

Environmental toxicity assessment on organic dyes using *Typha Angustata*

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Received: 18.01.2018 Accepted: 22.03.2018 Published: 30-06-2018
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ABSTRACT

Organic dyes are used in many industries like leathers, food, coloring, printing, cosmetics and paper making industries. The Nano level dye molecules are common water pollutants. A trace amount of dyes present in the dye effluent affect the environment. Typha Angustata Plant (sambu grass) is selected to identify the plant poisoning and non-poisoning organic dyes through the adsorption method. The adsorption of organic dyes has been performed by various conditions such as concentration, contact time, temperature and pH are optimized to find out the maximum toxic effect of organic dyes on the Typha Angustata Plant. The kinetics, thermodynamic and isotherms models are evaluated to find out the poisoning effects on Typha Angustata Plant.

Keywords: Adsorption; Environmental studies; Organic dyes; Typha angustata Plant; Toxicity.

1. INTRODUCTION

Dyes are basically natural or synthetic organic compounds that can themselves to surfaces or fabrics to provide bright and lasting color. They are applied in various industries such as leather, textile, paper, rubber, cosmetics, plastics, pharmaceuticals and food industries. Most of them are complex organic molecules. It is resistant to many things, such as the action of detergents (Chincholi *et al.* 2014; Yagub *et al.* 2014). Colored dye wastewater is regarded as a direct result of the production of the dye. It is also a consequence of its use in textile and related industries. There are more than 10,000 commercial dyes are known, with an annual production of over 7×10^5 tones per year (Sen *et al.* 2011). It is estimated that 2% of the dyes are discharged in effluent from manufacturing operation, while 10% was discharged from textile and associated industries (Allen *et al.* 2004).

Discharge of these dyes into the water streams will affect the people who may use these effluents for living purposes such as washing, bathing and drinking (Sharma and Sobti, 2000). Some dyes can cause allergy, dermatitis, skin irritation, cancer and mutations in humans (Bhatnagar and Jain, 2005). Apart from that, dyes also associated with environmental concern with their absorption and reflection of sunlight entering the water, which will inhibit the growth of bacteria, limiting it to levels insufficient to biologically degrade impurities in the water (Mckay and Al Duri, 1987). Furthermore, the dye can also affect aquatic plants as they reduce sunlight transmission through water. Therefore, the removal of such colored compounds from waste effluents becomes environmentally important because even a small amount of dyes in water can be toxic and highly visible (Chiou *et al.* 2004).

Different physical-chemical processes have been applied for the treatment of colored wastewater. These include precipitation, flocculation, electro-kinetic, coagulation, electro-flotation, ion exchange, membrane filtration, electro-chemical destruction, irradiation and ozonation. However, all these treatment methods are costly, suffering from many restrictions and cannot be utilized by small industries to treat the wide range of wastewater (Indra *et al.* 2005). Natural plant waste is widely used for the water treatment process. The agricultural waste materials such as tea waste (Uddin *et al.* 2009), wheat straw (Zhang *et al.* 2011), grass waste (Hameed, 2009), jack fruit peel, palm shell activated carbon (Foo and Hameed, 2012), coir pith carbon, sugarcane stalks, neem leaf powder, sawdust and carrot stem powder were used for the removal of methylene blue and other dyes from wastewater. Adsorption is a well-known equilibrium separation process and an effective method for water decontamination applications (Dabrowski, 2001). In terms of initial cost, flexibility and simplicity of design, ease of operation, and sensitivity to harmful contaminants, adsorption has been proven to be superior to alternative systems for water re-use. Adsorption does not cause the creation of any hazardous chemicals.

Typha angustata as an adsorbent

Typha angustata (shown in Fig. 1) is a perennial plant and is an aquatic weed and is widely grown in

waterlogged areas and is belongs to the family Typhaceae in the plant kingdom. Typha angustata is the most popular medicinal plant used for various medicinal properties and reported in many traditional literatures in India, as well as in China and Turkey.

The height is 1.5-2cm, and its leaf and stem are standing straightly. Its leaf is thick and has 5-12 mm thickness. Male and female flowers develop in dense, complex spikes on the same vertical stem. The male flowers are reduced to a pair of stamens and hairs and wither once the pollen is shed, leaving a short and bare stem portion above the female inflorescence. The stem is joint less. The dense cluster of female flowers forms a cylindrical spike some 10 to as much as 40cm and 1 to 4 cm broad. Seeds are minute about 0.2 mm long and attached to a thin hair or stalk, which effects wind dispersal.

Fig. 1: *Typha angustata* (Sambu grass).

At present work, Typha angustata raw material can be successfully employed as a low-cost alternative to the commercial adsorbents for the removal of Crystal Violet and Orange-G dye.

Table 1. Physical and Chemical properties of CV and OG.

Dye	Chemical Formula	Molecular Formula	Maximum absorption (λ_{max})	Make
Crystal Violet	$C_{25}H_{30}ClN_3$	408	591	Loba Chemie pvt ltd
Orange –G	$C_{16}H_{10}N_2Na_2O_7S_2$	452.38	480	Loba Chemie pvt ltd

2. EXPERIMENTAL

2.1 Materials and Methods

The dyes used in this study are Crystal Violet (Loba Chemie) and Orange G. The physical and chemical properties of Crystal Violet and Orange G are listed in Table 1, and the structures are given in (Fig. 2 & 3).

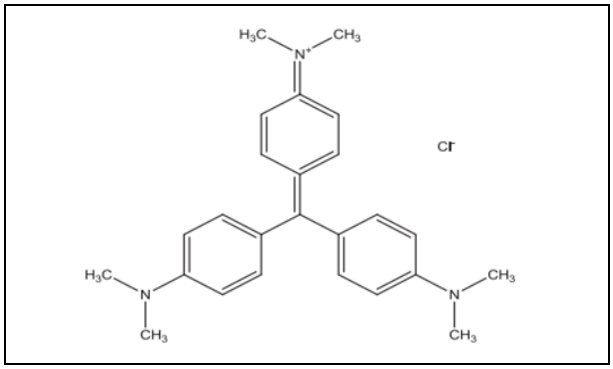

Fig. 2: Structure of Crystal Violet

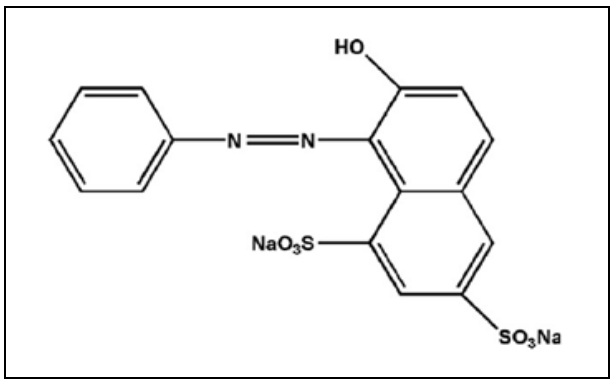

Fig. 3: Structure of Orange-G

2.2 TAP Powder Preparation

The plant *Typha angustata* is gathered without a blossom. The plants are chopped into little pieces and dried for one week in the sun. To form a powder, these little bits are finely ground. The powder is then rinsed with distilled water many times to remove surface adherent items and a water-soluble ingredient in the powder. Finally, the powder is dried in a hot air oven at 110^0C.

2.3 Adsorption Experiment

The stock solution of 1000 mg/l is prepared by dissolving 1g of CV and OG in a standard measuring flask separately. Working solutions of the necessary concentration are made by diluting the stock solution many times. A UV-Spectrophotometer is used to determine the dye concentration (Perkin Elmer model lambda-25). The dried adsorbent (0.5g) is added with 50ml of CV and OG solution in 100ml conical flask separately. The mixture is stirred on a magnetic stirrer (Remi-model-1MH), and the final concentration of the solution is measured spectrophotometrically.

The percentage removal of dye is calculated using the following equation.

$$\% \text{ removal} = (C_o - C_e)/C_o \times 100$$

The amount of dye adsorbed per g of TAP powder is calculated from the following equation.

$$Q_e = (C_o - C_e)\ V/W$$

Where,

C_o initial concentration of dye (mg/L)
C_e equilibrium concentration of dye (mg/L)
V Volume of the solution (L)
W mass of the adsorbent (g)

3. RESULT & DISCUSSION

3.1 Adsorption Studies

3.1.1 Adsorption of organic dyes on *Typha Angstata* plant

3.1.1.1 Effect of contact time

The effect of contact time for the adsorption of CV and OG are carried out at room temperature. The percentage removal of dyes at room temperature as a function of time is shown in Fig. 4. Very rapid adsorption is observed within 10 min for CV, whereas the removal of OG is 13% in 20 min. The rapid adsorption of CV is due to the interaction of cationic dye CV on the basic surface of the TAP powder. The poor adsorption of OG may be due to the less interaction between the anionic dyes on the basic cellulosic surface. The adsorption of CV is rapid. This may be due to the strong, active sites/ basic sites present in the TAP powder that may attract the cationic CV (Eren and Acar, 2006).

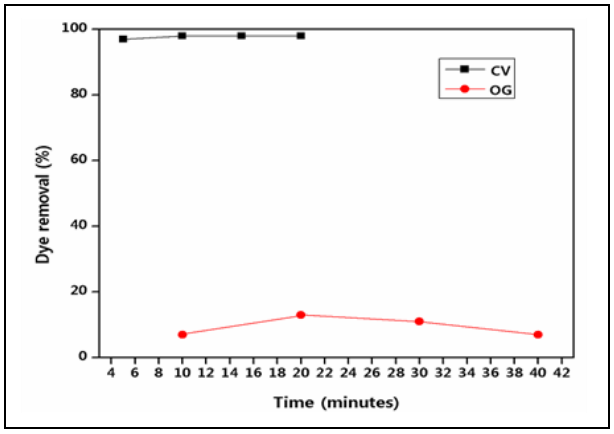

Fig. 4: Effect of contact time for the uptake of CV and OG on TAP. CV = Concentration-100mg/l, adsorbent dosage-0.5g/50ml, temperature-30 °C. OG = Concentration-100mg/l, adsorbent dosage-0.5g/50ml, temperature-30 °C.

3.1.1.2 Effect of concentration

The effect of dye concentration on the adsorption of CV and OG are shown in Fig. 5. Percentage adsorption of CV is decreased with the increase of initial dye concentration from 97 % to 49 %. Contact time for the adsorption of CV is 10 min. The decrease of adsorption with the increase of concentration may be due to the saturation of surface area and active sites available on TAP powder.

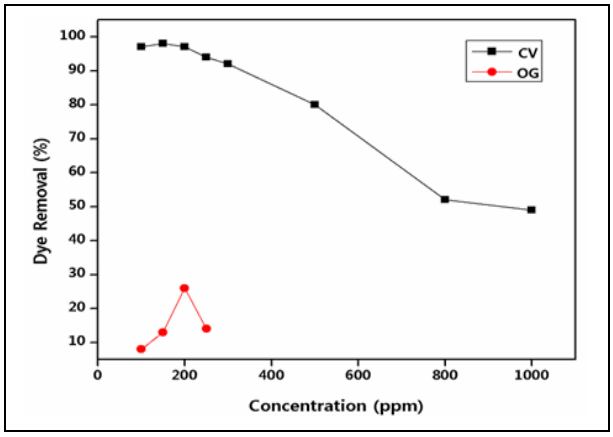

Fig. 5: Effect of concentration for the uptake of CV and OG on TAP. CV = adsorbent dosage - 0.5g/50ml, time-10 min, temperature-30 °C. OG = adsorbent dosage - 0.5g/50ml, time-20 min, temperature-30 °C.

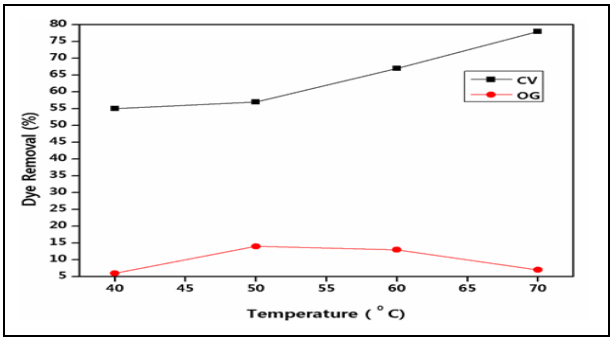

Fig. 6: Effect of temperature for the uptake of CV and OG on TAP. CV = Concentration-800mg/l, adsorbent dosage-0.5g/50ml, time-10 min. OG = Concentration-200mg/l, adsorbent dosage-0.5g/50ml, time-20 min.

Percentage adsorption increases with decreased initial dye concentration and reaches the maximum and then decreases adsorption for OG on TAP powder (Senthilkumaar *et al.* 2006).

3.1.1.3 Effect of temperature

The effects of temperature for the adsorption of dyes have been studied at 30-80°C are shown in Fig. 6. Percentage adsorption increases with the increase of temperature for CV. The reason for the increase in the adsorption of CV with an increase in temperature is due to the strong interaction between CV and the cellulosic surface of the adsorbent. When the temperature increased above 40°C, the absorption also increased, it is due to the activation of weak active sites in high temperature. Hence, adsorption increases with an increase in

temperature. But in the case of OG, the adsorption increases with increased temperature and further increased of temperature decreased to adsorption. This may be due to the repulsion of anionic OG and the basic surface of the TAP powder (Qadeer, 2007).

3.1.1.4 Effect of pH

The effect of pH of the solution on the adsorption of CV and OG is presented in Fig. 7. Experiments are performed with pH (4.0-9.0) for 50mL of dye solution and the adsorbent dose of 0.5 g. CV. Adsorption increases from 25% to 98% with an increase in pH. Further, there is no significant change at higher pH. At lower pH, the absorbent surface generates a net positive charge due to a higher concentration of H^+ ions. As a result, there is an electrostatic repulsion between cationic CV molecules and the adsorbent surface, which inhibits the CV removal. On the anionic dye, the maximum removal of the OG is decreased from 82% to 29% with increasing pH. Higher OH^-ions generate a net negative charge on the adsorbent surface. These results revealed that cationic and anionic dyes require basic and acidic medium respectively, for maximum removal (Salleh *et al.* 2011).

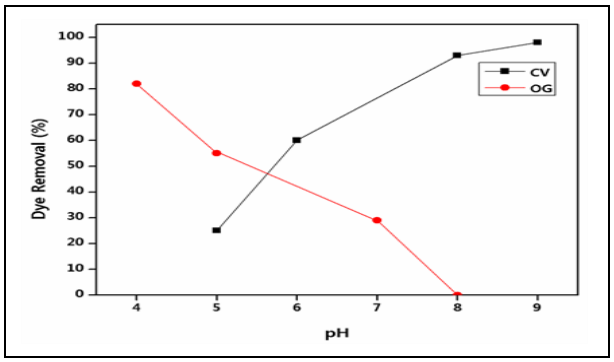

Fig. 7: Effect of pH for the uptake of CV and OG on TAP. CV = Concentration-800mg/l, adsorbent dosage-0.5g/50ml, time-10min, Temperature-30 °C. OG = Concentration-200mg/l, adsorbent dosage-0.5g/50ml, time-20min, Temperature-30 °C.

3.1.1.5 Effect of adsorbent dosage

The effect of dosage for the adsorption of CV and OG are shown in Fig. 8, and the percentage adsorption increases with increased adsorbent dosage. By increasing the adsorbent dosage, the number of active sites, as well as the surface area, also increased. For CV the adsorption increased from 52 to 76% for the adsorbent dosage of 0.5 to 1.5g. OG adsorption decreased from 26 to 3% with an increase of adsorbent dosage of 0.5 to 1.5g. In TAP, the adsorbent surface carries basic sites. Due to the presence of basic sites, the anionic dye adsorption decreases with increasing dosage (Salleh *et al.* 2011).

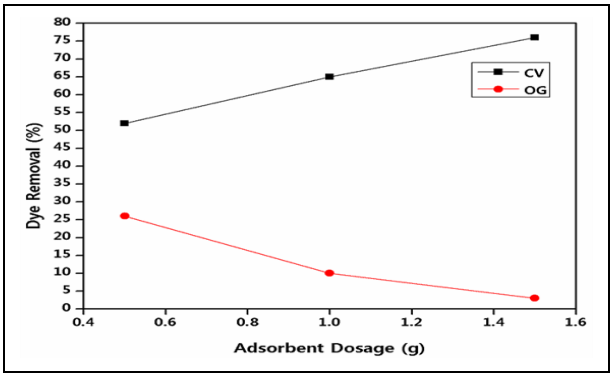

Fig. 8: Effect of adsorbent dosage for the uptake of CV and OG on TAP. CV = Concentration-800mg/l, pH-7, time-10 min. OG = Concentration-200mg/l, pH-6, time-20 min.

3.2 Adsorption Kinetics

3.2.1 Pseudo- second-order kinetics

The adsorption kinetics of CV and OG on TAP powder has been studied. The Pseudo second-order equation is represented as follows:

$$t / Q_t = 1/K_2Q_e^2 + 1/Q_et$$

Where,

K_2 is the rate constant for pseudo-second-order adsorption (g m^{-1} min^{-1}).
Q_e is the amount of dye adsorbed per unit mass of the adsorbent at equilibrium (mg g^{-1}).
Q_t is the amount of dye adsorbed per unit mass of the adsorbent at time t (mg g^{-1}).
t is time in minutes.

The plot of (t/Qt) versus t gives a straight line for the adsorption of CV and OG on TAP powder. The kinetic plot is shown in (Fig. 9 & 10). The linear regression coefficient (R^2) values are higher than 0.99, indicating that the adsorption follows the pseudo-second-order kinetics. The pseudo-second-order kinetic data for the adsorption of CV and OG on TAP powder are given in Table 2.

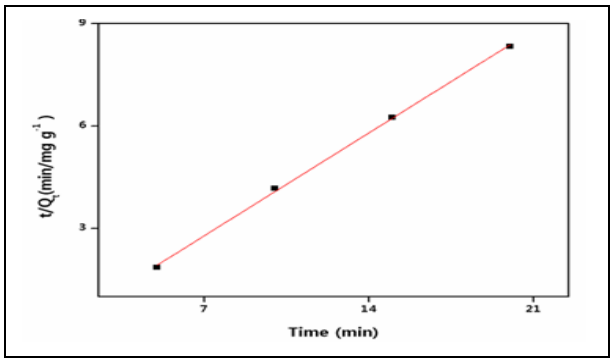

Fig. 9: Pseudo-second order kinetics plot for the adsorption of CV on TAP.

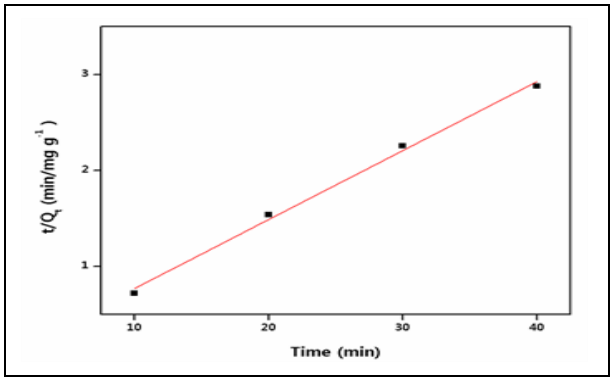

Fig. 10: Pseudo-second order kinetics plot for the adsorption of OG on TAP.

Table 2. Pseudo-second order kinetics parameters for the adsorption of CV and OG on TAP.

Adsorbent	Dye	K_2 (s^{-1})	R^2
TA	CV	0.4305	0.9989
	OG	0.0719	0.9943

3.3 Adsorption Isotherm

3.3.1 Langmuir adsorption isotherm

The adsorption capacity and interaction between adsorbent and adsorbate were described by adsorption isotherms. Langmuir and Freundlich adsorption isotherms are used to optimize the design of the adsorption system. Adsorptions of CV and OG at different concentrations are studied, and the data are given in Table 3.

A linear form of the Langmuir model may be written as

$$C_e / Q_e = (1/Q_{max}K_L) + (Ce/Q_{max})$$

Where,

Q_e is the equilibrium concentration of dyes on the adsorbent (mg g^{-1})
C_e is the equilibrium concentration of dyes in solution (mg L^{-1})
Q_{max} is the monolayer capacity of Adsorbent (mg/g)
K_L is the Langmuir adsorption constant.

A plot of C_e/Q_e Vs C_e is a straight line with slope $1/Q_{max}$ and the intercept $1/Q_{max}K_L$ (Fig. 11 & 12). The correlation coefficient (R^2) values are 0.9891 and 0.8449 for CV and OG, respectively. The correlation coefficient (R^2) value is very close to 1 for CV, indicated that the adsorption process follows the Langmuir adsorption isotherm.

Table 3. Langmuir parameters for the adsorption of CV and OG on TAP.

Adsorbent	Dye	Q_{max}(mg/g)	R^2	K_L
TA	CV	47.8927	0.9891	0.3426
	OG	0.6486	0.9674	258.86

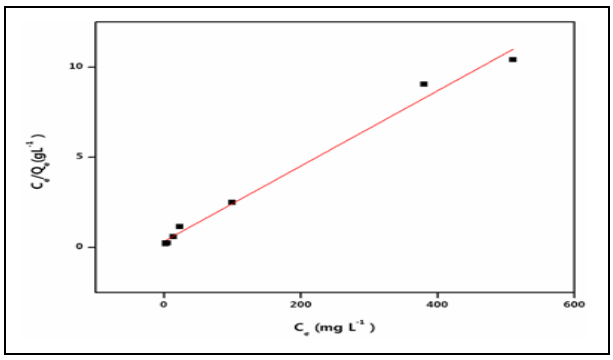

Fig. 11: Langmuir adsorption isotherms for the adsorption of OG on TAP.

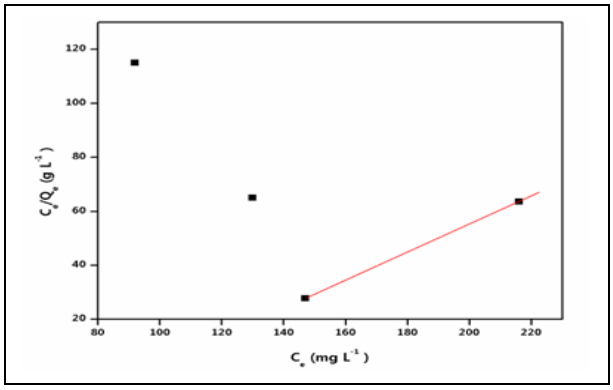

Fig. 12: Langmuir adsorption isotherms for the adsorption of CV on TAP.

3.3.2 Freundlich isotherm

The multilayer adsorption phenomena is assumed in the Freundlich isotherm model. The adsorption investigations are carried out at various concentrations at room temperature, and the adsorption results are used to validate the adsorption isotherms of CV and OG on TAP powder using the freundlich equations.

Freundlich model in linear form is represented by the following equation,

$$\ln Q_e = \ln K_F + (1/n) \ln C_e$$

Where,

K_F is the Freundlich constants
n is the number of layers

The plot of $\ln Q_e$ vs $\ln C_e$ gave a straight line (Fig. 13 & 14) with the intercept $\ln K_F$ and the slope 1/n. The R^2 values are 0.9177 and 0.8453 for CV and OG. The R^2 value is low when compared to Langmuir adsorption isotherm, and it is indicated that the adsorption is well fitted through the Langmuir adsorption isotherm. The result obtained from Langmuir and Freundlich adsorption isotherms is given in Table 4.

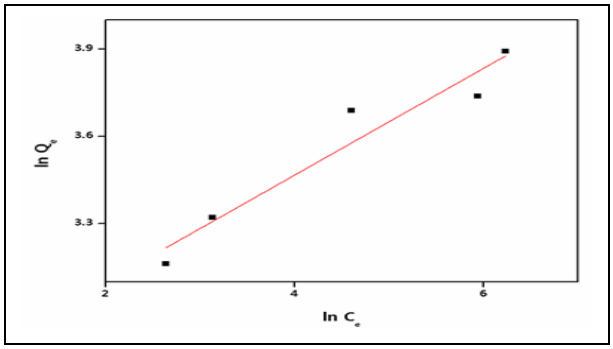

Fig. 13: Freundlich adsorption isotherms for the adsorption of CV on TAP.

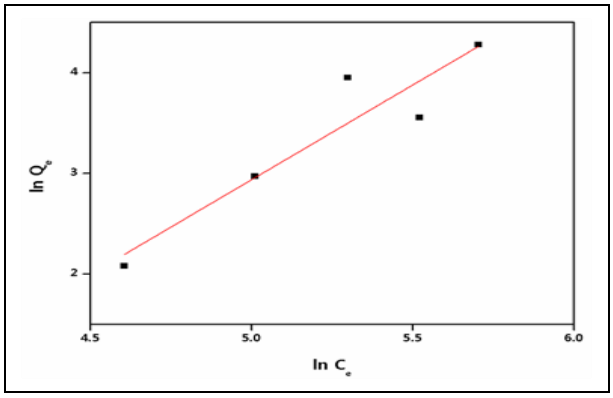

Fig. 14: Freundlich adsorption isotherms for the adsorption of OG on TAP.

Table 4. Freundlich adsorption isotherm parameters.

Adsorbent	Dye	N	R^2	K_F
Typha	CV	5.4495	0.9177	15.3567
Angustata	OG	0.5310	0.8453	0.00153

3.4 Thermodynamic Study

Thermodynamic studies were done to elaborate on the thermodynamic parameters. Experiments were carried out in the temperature ranges (30 -70 ℃), keeping all the parameters constant. Enthalpy (H^0), entropy (S^0) and change in Gibb's free energy (G^0) were calculated for both the dyes by using Van't Hoff equation.

$$K_D = q_e/C_e \quad (1)$$

$$\Delta G = -RT \ln K_D \quad (2)$$

$$\ln K_D = (\Delta S / R) - (\Delta H / RT) \quad (3)$$

From the equations eq.**2** and eq.**3**,

$$\Delta G = \Delta H - T\Delta S \quad (4)$$

Where,

K_D is the distribution coefficient of the adsorbate
q_e is the equilibrium dye concentration on adsorbent (mg g^{-1})
C_e is the equilibrium dye concentration in the solution (mg L^{-1})
R is the universal gas constant (8.314 J/mol K)
T is the temperature (K).

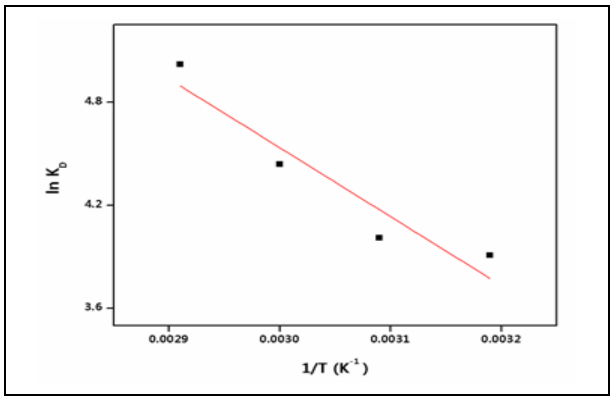

Fig. 15: Thermodynamic parameters plot for the adsorption of CV on TAP.

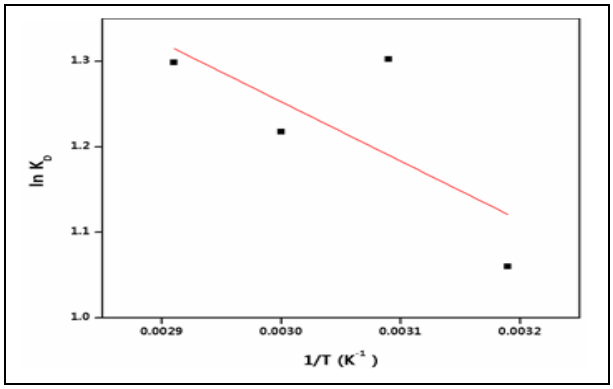

Fig. 16: Thermodynamic parameter plot for the adsorption of OG on TAP.

ΔH^0 and ΔS^0 parameters can be calculated from the slope and intercept of the plot $\ln K_D$ vs 1/T, respectively (Fig.15 & 16). Results are summarized in Table. ΔG^0 values at the temperatures of 30⁰C – 70⁰C (303 K - 333 K) are negative Table 5. This indicated that the process is a feasible adsorption process and spontaneous.

The enthalpy (ΔH^o) values are greater than 40 KJ/mol indicated that the adsorption of dye over TAP powder is chemisorption. The fact that enthalpy was positive meant that the adsorption process was endothermic. Furthermore, the positive value of $\Delta S°$ revealed that during adsorption, the degrees of freedom increased at the solid-liquid interface.

Table 5. Thermodynamic parameters for the adsorption of CV and OG on TAP.

Adsorbent	Dye	Temperature	ΔG^o(KJ/mol)	ΔH^o (KJ/mol)	ΔS^o (KJ/mol)
TA	CV	313	-9.7592	-41.309	0.1376
		323	-11.1352		
		333	-12.5112		
		343	-13.8872		
	OG	313	-2.9077	-5.7655	0.0270
		323	-3.1848		
		333	-3.4659		
		343	-3.7390		

4. CONCLUSION

The dye effluent has many organic dyes, these organic dyes polluting water, soil, plant and animal etc. The plant poisoning and soil polluting organic dyes identifications are reported in the literature. However, plant poisoning nature of organic dyes on TAP powder. In the present investigation, the adsorption of CV and OG has been studied on TAP to evaluate their toxic effect on the plant. The following experimental conditions are optimized to attain maximum adsorption of CV and OG are given below:

- The effect of contact time has been optimized to find out the equilibrium adsorption of CV and OG on TAP. Very rapid adsorption is observed within 10 min for CV, whereas the removal of OG is 13% in 20 min.
- The effect of the concentration of dye has been optimized to find out the equilibrium adsorption of CV and OG on TAP. It is observed that percentage adsorption of CV decreases with an increase of initial dye concentration from 97% to 49%, respectively. Percentage adsorption increases with the increase of initial dye concentration and reaches the maximum, and then decreases adsorption for OG on TAP powder.
- The temperature effect for the adsorption of CV and OG on TAP has been optimized to find out the percentage adsorption increases with the increase of temperature for CV. But in the case of OG, the adsorption increases with increased temperature and further increased of temperature decreases the adsorption.
- Adsorption increases from 25% to 98% with an increase in pH. Further, there is no significant change at higher pH. On the anionic dye, the maximum removal of the OG decreases from 82% to 29% with an increase in pH.
- The effect of adsorbent dosage variation shows that the adsorption increases from 52 to 76%, with an, increased of adsorbent dosage for CV. But the adsorption decreases from 26 to 3% with an increased adsorbent dosage for OG.
- The adsorption on TAP powder follows pseudo-second-order kinetics for CV and OG and obeys Langmuir adsorption isotherm only on CV.
- The thermodynamic study proved that the adsorption of CV is chemisorption, and OG is physisorption on TAP.
- Thus this adsorption study proved that CV has more affinity on TAP powder than OG. Hence it rapidly toxic to plant than OG.

FUNDING

This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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