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Fungal Biosorption: An Innovative Treatment Method for the Removal of Textile Dyes

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Abstract

Basic fuchsin is a potential toxic, irritant and carcinogenic dye. Conventional dye effluent treatment strategies can cause extreme ecological problems. Therefore, the present study was focused on the sorption of Basic fuchsin dye by dead Aspergillus niger biomass at different pH (pH 6.0, pH 6.5, pH 7.0 and pH 7.5) from aqueous dye solution. Maximum dye removal was observed at pH 7.0 (80.42%). The equilibrium data were analyzed by employing Langmuir and Freundlich isotherm equations and the results showed that the equilibrium data were better described by Freundlich isotherm model, suggested that the sorption on a heterogeneous surface. The results therefore indicated that dead biomass of Aspergillus niger could be used as natural biosorbent to remove dyes from aqueous effluents.

Keywords: *industrial effluents, basic fuchsin dye, biosorption, Langmuir and Freudlich isotherms*

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INTRODUCTION

Industrial effluents contribute colossally to water disintegration and their treatment is the subject of discussion and regulation in many countries [1]. Dyes are the vital class of synthetic organic compounds used in various industries such as manufacture of pulp and paper, leather tanning or textile dying and in manufacture of dyestuffs. Ghaly *et al*. [2] pointed out that the textile industry is one of the significant industries in the world and it assumes a noteworthy part in the economy of numerous nations. The worldwide textile dyes market was estimated to reach USD 4.7 billion in 2015 and is projected to reach USD 6.4 billion by 2019 and USD 8.75 billion by 2023 [3–5].

About 10,000 different dyes are arranged globally and approximately 8×10^5 tons of synthetic dyes is devoured in textile industries in the whole world [6]. Approximately 10–15% of dyes applied in the dyeing process are released with wastewater causing natural contamination [7]. Wastewaters from the textile industry contain a lot of colors and chemical compounds containing trace metals such as Cr, As, Cu and Zn which are capable of harming the environment and human health [2].

Different physical (color irradiation, ozonation and ultrafiltration) and chemical (coagulation–flocculation, precipitation and advanced oxidation) methods are available for the treatment of wastewater but these methods have a lot of limitations such as high chemical and operating cost, high sludge and toxic gases generation [8–10].

Subsequently, the treatment of dye wastewaters and finding dye removal efficient methods has been challenging problems among ecological advances.

Biosorption has been contemplated since 1980s for evacuating dyes, heavy metals and other organic pollutants by various microorganisms from wastewater [11]. The fundamental points of interest of biosorption are high selectivity, cost viability and good removal performance [12]. Various bacteria, fungi, yeast and algae have been tried for their biosorption abilities under different experimental conditions [13–16]. Among these microorganisms, fungal biomass can be produced cheaply and obtained as a waste from various industrial fermentation processes [17]. In the cell wall of fungi, distinctive groups are present which are

responsible for binding dye molecules. These groups include amino, carboxyl and thiol [18].

Compared with live fungal cells, dead fungal biomass possesses various advantages such as dead biomass can be easily stored and used for long periods, while live biomass require nutrients and are subject to several physiological restrictions, strict operational and maintenance conditions [19]. Some examples of using dead fungal biomass for biosorption include *Aspergillus niger* [20, 21], *Rhizopus arrhizus* [22], *Neurospora crassa* [23] and *Aspergillus wentii* [24].

In the present study, the effects of different contact time and different pH on the biosorption of basic fuchsin dye by dead *A.niger* biomass was examined and Freundlich and Langmuir isotherm models were used for the evaluation of biosorption equilibrium data.

MATERIALS AND METHODS

Isolation of Fungal Strain

A. niger was isolated from dye infested soil collected from Partapur Industrial Area, Meerut, India and identified on the basis of morphology and cultural characteristics following Gilman [25], Ellis [26] and Nagamani *et al*. [27]. *A.niger* was maintained on Potato Dextrose Agar (PDA) plates for further use.

Preparation of Biosorbent (Fungal Biomass)

A liquid growth medium was prepared, composed of 3 g Malt extract, 10 g Glucose, 3 g Yeast extract and 5 g Peptone in one liter of distilled water. After autoclaving and cooling, the medium containing flasks were inoculated with *A.niger* fungal spores and then placed in a rotary platform shaker for 10 days at room temperature at rotation speed of 150 rpm.

After 10 days of incubation, the obtained biomass was autoclaved at 121 ^{o}C /15 min, then filtered and well washed with distilled water and finally dried at 55–60 ^{o}C in an oven for 12–16 h to prepare the dead biomass. The dried biomass was crushed using a mortar and pestle and used as biosorbent for biosorption of basic fuchsin dye.

Preparation of the Adsorbate

A stock solution of 500 ppm of basic fuchsin dye was prepared. From the stock solution, various concentrations of working dye solutions (50, 100, 200 and 400 ppm) were prepared.

Biosorption of Basic Fuchsin Dye

Effects of different pH (i.e. 6.0, 6.5, 7.0 and 7.5) on the biosorption of basic fuchsin dye were determined at different concentrations. Biosorption experiments were performed in 250 ml Erlenmeyer flasks by using 40 mg biomass of *A.niger* for 100 ml solutions of 50 ppm, 100 ppm, 200 ppm and 400 ppm concentrations of basic fuchsin dye in triplicates at different pH and a repetition of dye solutions without the fungal biomass was used as control.

Samples were placed on rotary shaker at constant speed of 150 rpm at room temperature and after 20 min, samples were sieved and the supernatants were analyzed to determine the concentrations of remained dye in solution using visible spectrophotometer at 550 nm wavelength. The amounts of basic fuchsin dye adsorbed onto biomass were calculated using the following equation:

$$Qe = (C_i\text{-}Cf)V/W$$

Where, C_i was initial dye concentration, C_f was final concentration of dye in solution, W is the weight of biomass and V is the volume of dye solution [28].

Characterization of Fungal Biomass

The changes in the cell wall of fungi and surface functional groups, after biosorption of dye at different pH, were determined by Fourier Transform Infrared Spectroscopy **(**FTIR). Two mg of fungal biomass powder was mixed with 98 mg of dry powdered potassium bromide (KBr) and finally grounded. The material was used for preparing pellets by applying pressure of 10,000−15,000 psi. IR spectra were recorded on FTIR spectrophotometer at high resolution ($\leq$ 0.001 cm^{-1}). Bonding efficiencies of different functional groups were expressed as Very Strong (VS), Strong (S), Moderate (M), Weak (W) and Very Weak (VW).

RESULTS AND DISCUSSION

Effect of pH

The effect of pH on the biosorption of basic fuchsin dye by dead *A.niger* was studied at pH 6.0, pH 6.5, pH 7.0 and pH 7.5 for all dye concentrations. The results showed that when increasing the pH of the dye solution from 6.0–7.0, the sorption of dye gradually increases with increase in pH. Further increase in pH from 7.0 to 7.5, decrease in the sorption of dye was observed. At pH 7.0 maximum biosorption percentages (80.42%) and specific uptake (1608.4 mg/g) of basic fuchsin dye were obtained (Fig. 1).

The increase of the removal percentage of methylene blue (MB) with increasing pH was studied by Kanamadi *et al*. [29] and Acemioglu *et al*. [30]. Kabbout and Taha [31] observed that the biosorbed amount increases up to 65% with pH from 4 to 6 and it is similar and around 90% for pH from 7 to 13 and suggested the pH 7 for further experiments. Nanthakumar *et al*. [21] also reported that the sorption of Reactive Blue 140 by *A.niger* HM11 was maximum at pH 6.0 and 60 ppm dye concentration.

Bisorption Isotherms

The equilibrium data were acquired at pH 7.0 and described by Langmuir and Freundlich isotherm models. Langmuir isotherm assumes that the adsorption of a single adsorbate onto a series of equivalent sites (homogenous sites) on the surface of the solid (adsorbent). Each site of adsorbent can hold at most one molecule of adsorbate (mono-layer) and there are no interactions between adsorbate molecules on adjacent sites. The Langmuir equation is expressed by the following relation [32]:

$$1/q_e = 1/K_a q_m C_e + 1/q_e$$

where, q_e is the amount of dye adsorbed at equilibrium time (mg/g), C_e is the equilibrium concentration of dye in solution (mg/l), q_m is the maximum adsorption capacity (mg/g) and K_a is the isotherm constants for Langmuir (L mg^{-1}).

The Freundlich model is an empirical equation based on sorption on a heterogeneous surface or surface supporting sites of varied affinities. The Freundlich adsorption isotherm model is represented as follows [33]:

$$\ln q_e = \ln K_f + 1/n\ (\ln C_e)$$

where, q_e is the amount of dyeicion adsorbed at equilibrium time (mg/g), C_e is the equilibrium concentration of dye in solution (mg/l), K_f is the capacity of the adsorbent and n is the intensity of adsorption constant for Freundlich. The plot of $\ln q_e$ *versus* $\ln C_e$ is employed to determine the K_f and n from intercept and slope, respectively. Generally, the value of the linear regression correlation coefficient R^2 gives an indication as to which model can be chosen to give the best-fit.

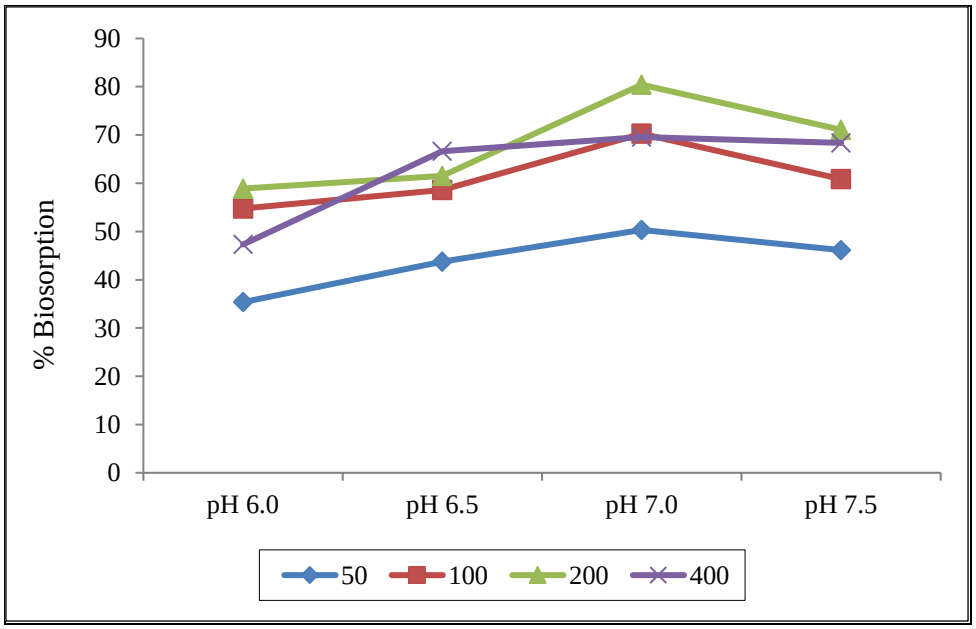

Fig. 1: *Percentage Biosorption of Basic Fuchsin Dye by Dead Biomass of A.niger at pH 7.0.*

In the present study, at pH 7.0 the R^2 values for Freundlich and Langmuir isotherms were calculated to be 0.725 and 0.657, respectively which indicated that the biosorption of basic fuchsin dye by dead *A. niger* biomass could be explained better by Freundlich as compared to Langmuir isotherm equilibrium (Figs. 2,3 and Table 1).

Sadaf and Bhatti [34] investigated the biosorption of Foron turquoise SBLN using mixed biomass of white rot fungi and found that the equilibrium data were better described by Freundlich isotherm model as compared to Langmuir equation while Dharajiya *et al*. [35] observed that the R^2 values for Langmuir and Freundlich isotherms were calculated to be 0.993 and 0.971, respectively, which indicated that the biosorption of Acid Black 52 on autoclaved biomass of *A.fumigatus* A23 dye could be explained better by Langmuir as compared to Freundlich isotherm equilibrium. Souza *et al*. [36] studied the biosorption of reactive red 120 dye (red-120) onto fungal biomass (FB) of wild *Ganoderma stipitatum* basidiocarps and they found that the biosorption equilibrium data were best fitted by Langmuir isotherm model presenting monolayer biosorption.

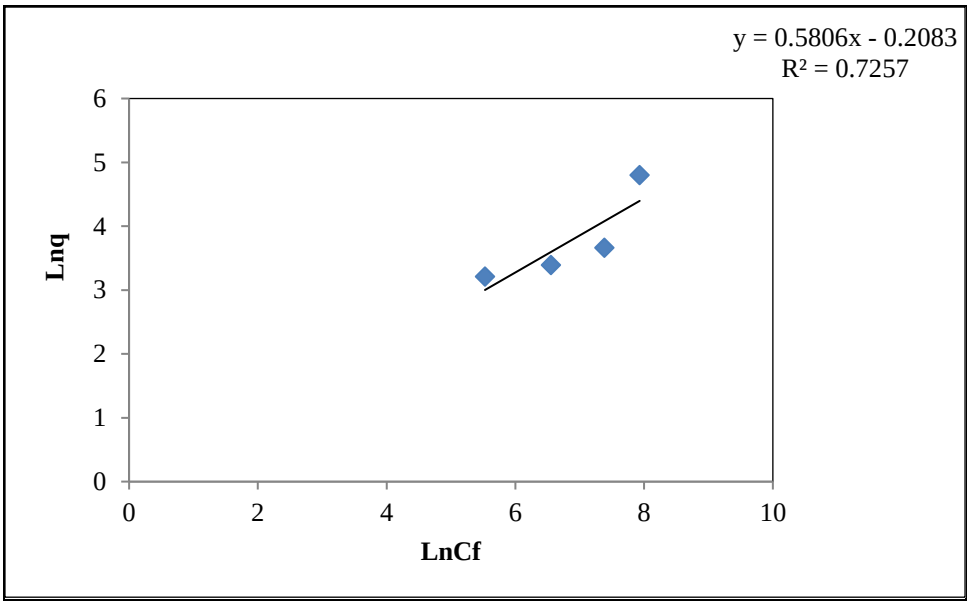

Fig. 2: *Freundlich Model for Sorption of Basic Fuchsin Dye by Dead Biomass of A.niger at pH 7.0.*

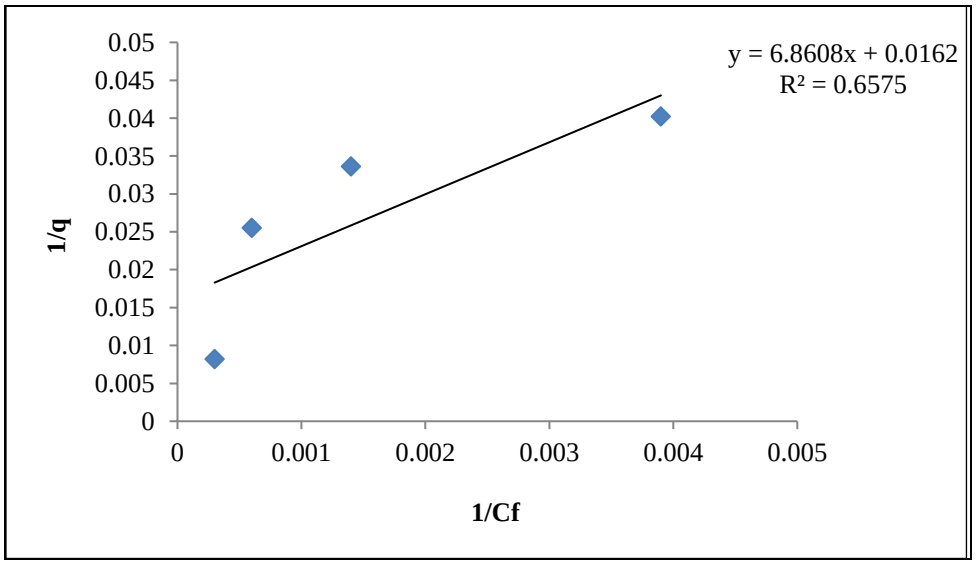

Fig. 3: *Langmuir Model for Sorption of Basic Fuchsin Dye by Dead Biomass of A.niger at pH 7.0.*

Table 1: *Isothermic Values of Basic Fuchsin Dye Sorption by Dead Biomass of A.niger.*

Freundlich isotherm				
	n	**l/n**	**Kf**	**R^2**
pH 7.0	1.724	0.580	1.231	0.725
Langmuir isotherm				
	a	b	1/ab	R^2
pH 7.0	6.860	0.0003	500	0.657

FTIR Spectra of Unloaded and Dye Loaded Fungal Biomass

Bayramoglu and Arica [18] reported that heat treatment increased the hydrophilicity of the fungal biomass by removing hydrophobic entities on the cell surfaces of the fungal biomass. Aksu [37] concluded that autoclaving also causes the disruption of the fungal structure which leads to increased porosity. Reduction in hydrophobicity can cause reduction in the functional groups which are responsible for hydrophobic nature of the biomass. Autoclaving of the fungal biomass probably caked the biomass implying that the physical strength of hyphae in the fungus was weak [35]. This is likely due to increase in surface area caused by rupture during autoclaving [38, 39].

FTIR investigation related to the fungal biosorption phenomenon was carried out using basic fuchsin-loaded fungal species. This analysis had eventually confirmed the difference between functional groups in relation to biosorption of basic fuchsin dye. The unloaded dead fungal biomass showed eleven peaks at 1035.82^{-1} [Aromatic rings (S), Si—O—C (W), Si—O—Si (W), C═S (S), Sulfonic acid (VW), C—C aliphatic chains (M)], 1078.25^{-1} [Aromatic rings (S), Si—O—C (W), Si—O—Si (W), C═S (S), Sulfonic acid (VW), Sulfonamide (M), Sulfone (M), C—C aliphatic chains (M)], 1153.48^{-1} [C—C aliphatic chains (M), C═S (S), Sulfonamide (M), Sulfone (M), Si—O—C (W), Sulfonic acid (VW)], 1317.44^{-1} [Carboxylate salt (M)], 1585.55^{-1} [Nitro (M), Aliphatic azo (M), Aromatic/hetero ring (S), Amide (S)], 1654.03^{-1} [Amide (S), Ketone (M), Carboxylic acid (M), C═C (VS), C═N (VS)], 2853.81^{-1} C—CH_3 (S)], 2925.17^{-1} [C—CH_3 (S), Aromatic C—H (S), OH (W), CH_2 (S)], 2956.04^{-1} C—CH_3 (S), Aromatic C—H (S), OH (W)], 3096.85^{-1} [Aromatic C—H (S), OH (W), CH═CH (S)], 3217.41^{-1} [OH (W), Amide (M), Amine (M), Phenol (W), Alkyne (VW)] (Fig. 4). Four peaks were completely vanished from the fungal biomass after loading the dye and one peak from 1317.44^{-1} [Carboxylate salt (M)] were shifted towards higher wavelength at 1363.73^{-1} [Carboxylate salt (M), C—CH_3 (W)]. The sorption spectrum of dye loaded dead fungal biomass also showed eleven peaks with four new peaks at 662.58^{-1} [C—Br (S), C—Cl (S), C═S (S), C—C aliphatic chains (M)], 1748.55^{-1} [Aliphatic ester (M), Lactone (M), Anhydride (M), Acid chloride (M)], 2363.87^{-1} [P—H (VW)], 2515.28^{-1} [Cyclohexylamine Perchlorate (W), Trimethylamine (W), OH (W)] and one shifting in peak at 1363.73^{-1}. Biosorption of basic fuchsin dye resulted in the reduction of five strong functional groups (aromatic ring, amide, C—CH_3, aromatic C—CH, CH═CH), four moderate functional group (nitro, aliphatic azo, amide, amine), four weak functional group (O—H) and one very weak functional group (alkyene) (Fig. 2). The Full width at half maxima (FWHM) data also showed the sharping of all the similar peaks present in both type of fungal biomass (Table 2). These vanished functional groups are responsible for the dye absorption and stress developed by the loading of basic fuchsin dye is responsible for the emergence of new peaks in the biomass. The results of FTIR spectra confirmed the modification of surface functional groups of fungal biomass due to binding of basic fuchsin dye. Dharajiya *et al.* [35] studied three methods—autoclave, acid and alkali—for pretreatments of fungal biomass and the best results were obtained with the autoclaved biomass. They further concluded that dead and dried biomass can be stored for long periods at room temperature with little risk of putrefaction. The microbial surface carries different types of functional groups such as amino, carboxylate, phosphate and hydroxyl which are responsible for the binding of hazardous chemicals and materials from industrial effluents [40]. In the present study, aromatic ring, amide, C—CH_3, aromatic C—CH and CH═CH were identified as most important moieties involved in the binding process of dyes. The amorphous homopolysaccharides and heteropolysaccharides, often in association with proteins, play the role

of cementing substances [35]. Hence, better biosorption in case of heat-treated and biomass may be related to both physical changes and changes in the functional groups. Therefore, more basic fuchsin dye molecules can enter and bind to the enlarged pores in autoclaved biomass (Fig. 5).

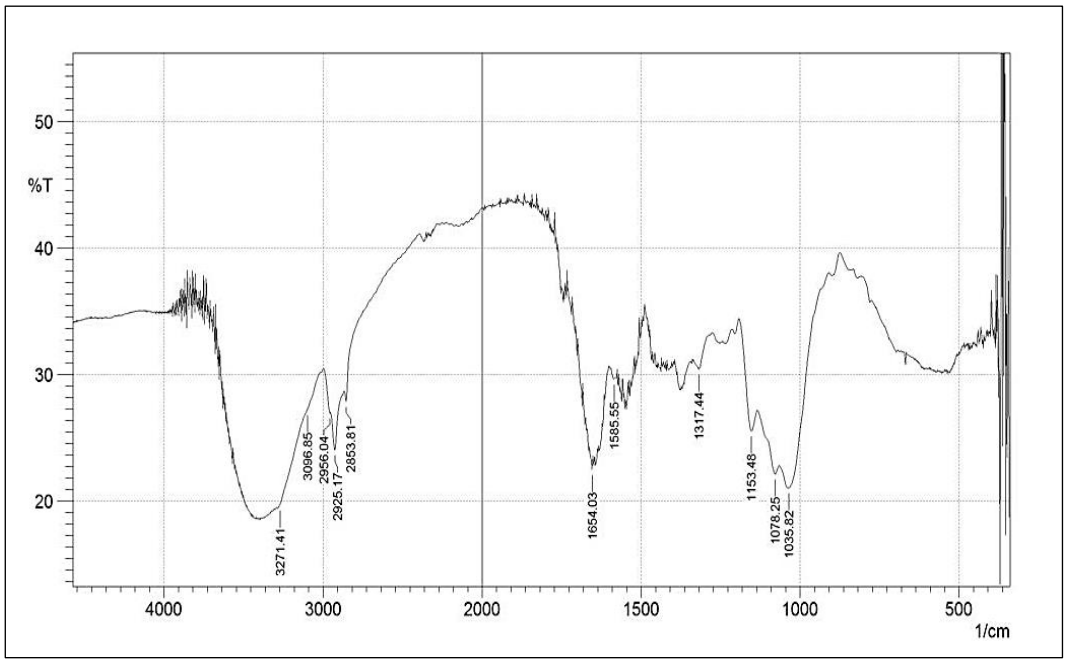

Fig. 4: *FTIR Spectra of Dead A.niger Biomass (Unloaded).*

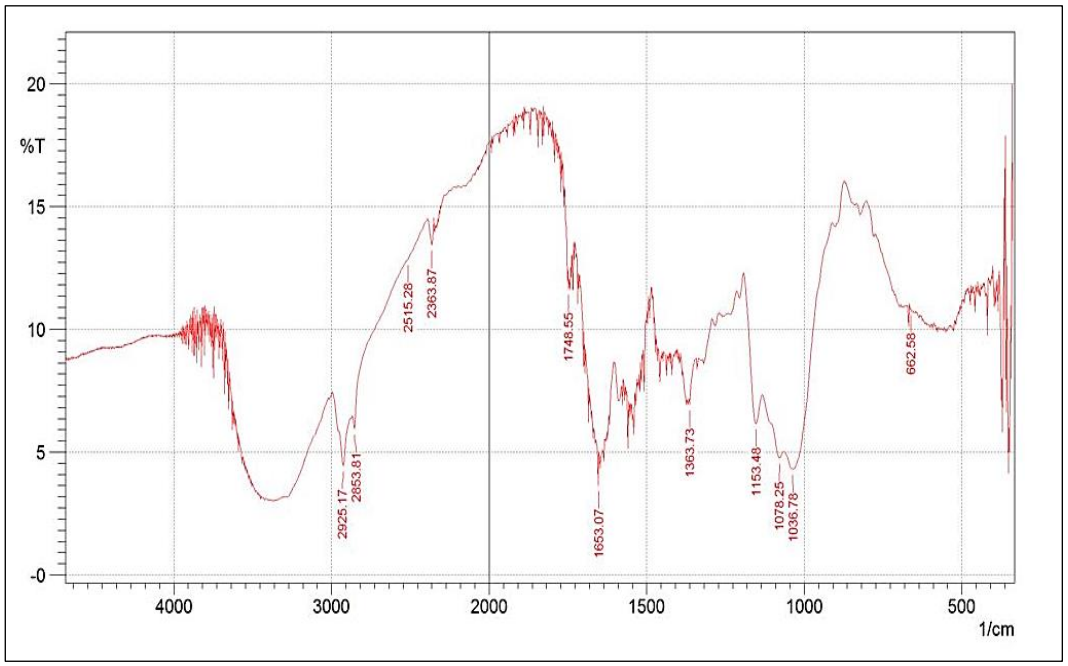

Fig. 5: *FTIR Spectra of Dead A.niger Biomass (Loaded).*

Table 2: *Full Width at Half Maxima (FWHM) of Unloaded and Dye Loaded A.niger Biomass.*

Unloaded biomass	3271.41	3096.85	2956.04	2925.17	2853.81	1654.03	1585.55	1317.44	1153.48	1078.25	1035.82
FWHM	2.4	2	2.1	2.6	2.8	3.3	2.7	2.9	2.6	2.3	3
Loaded biomass	2925.17	2853.81	2515.28	2363.87	1748.55	1653.07	1363.73	1153.48	1078.25	1036.78	662.58
FWHM	2.5	2.3	2	2	1.5	2.9	3	2.4	2	3.1	1.6

CONCLUSION

In the present study, the ability of dead *A.niger* biomass as a biosorbent to remove basic fuchsin dye from aqueous solutions was investigated. Experimental parameter such as different pH of the dye solution was found to affect the biosorption efficiency of dead *A.niger* biomass. The adsorption data present the best fit to the Freundlich model, suggesting that adsorption occurs by the formation of multilayer. This study showed that dead *A.niger* fungal biomass could be used as an efficient and inexpensive biosorbent for dye effluents treatment.

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