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Kinetic and equilibrium studies on biosorption of reactive orange 107 dye from aqueous solution by native and treated fungus Alternaria Raphani

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ABSTRACT

Batch mode experiments were carried out for the removal of Reactive Orange 107 from aqueous solution using native and dead mycelia pellets/biomass of Alternaria raphani. The effect of process parameters like contact time, dosage of adsorbent, adsorbate concentration and pH on adsorption was investigated. Higher the dye concentrations lower the adsorption. Increase in biomass dosage increased the adsorption. Experimental data were analyzed by the Langmuir and freundlich isotherms. Adsorption capacity (Q_o) of autoclave biomass was 28.74mg/g, which was higher than native biomass studied. The second-order kinetic model by Ho and Mckay described well the experimental data. Acidic pH was favorable for the adsorption of RO 107. Studies on pH effect and desorption show that chemisorptions and physisorption play a major role in the adsorption process. Among the biomass studied, autoclaved biomass showed a better adsorption capacity. Utilization of autoclaved biomass is much safer as it does not pose any threat to environment. These results suggest that this fungus can be used in biotreatment process as biosorbent for reactive dyes.

Keywords: Reactive orange 107, biosorption, *Alternaria raphani*, isotherms, kinetics.

INTRODUCTION

Reactive dyes are widely used in many industrial uses due to their bright color, excellent colorfastness and ease of application [1]. Reactive dyes are typically azo-based chromophores combined with different reactive groups. They differ from all other dye classes in that they bind to the textile fibre, such as cotton, through covalent bonds; and thus are highly recalcitrant to

conventional wastewater treatment processes [2]. Textile dye effluents contain reactive dye in a concentration range of 5-1500mg/L. Therefore, the treatment of dye-contaminated effluents is currently a primary environmental concern [3]. Conventional treatment technologies such as, activated sludge process, chemical coagulation, carbon adsorption, chemical oxidation, photo decomposition, electro-chemical treatment, reverse osmosis, hydrogen peroxide catalysis etc are difficult, ineffective (or) economical disadvantage [4, 5].

Biosorption has been defined as the property of certain biomolecules (or types of biomass) to bind and concentrate selected ions/other molecules from aqueous solutions. Thus biosorption using microbial biomass especially, fungal biomass as potential sorbent for removal of dyes from industrial wastewater has gained considerable attention. Recently removal of reactive red 2 dye using activated carbon was reported [6]. Several reports on decolorisation of synthetic dyes and dye effluents by fungi [7, 8 and 9] have been reported by using various fungi. To minimize the use and generation of hazardous chemicals, while increasing the treatment efficiencies of the wastewater generated, we studied the use of fungal biomass as a cheap resource. In this study, the live and heat-treated biomass of *Alternaria raphani* were used for the biosorption of RO 107 in batch mode studies. The effects of experimental conditions such as initial dye concentration, temperature and pH are investigated to obtain information on dye removal properties of the fungal biomass.

EXPERIMENTAL SECTION

Preparation of inoculum and adsorbate

The fungus was isolated from dye industry effluent canals in the textile city, Tirupur, Tamilnadu. The isolated fungal culture was grown in Czapek-Dox agar medium for 5 days at 27±2°C in petriplates. Czapek-Dox agar medium contained potassium chloride (0.5g), potassium dihydrogen phosphate (1.0g), sodium nitrate (2.0g), ferrous sulphate (0.01g), magnesium sulphate (0.5g), yeast extract (1.0g), dextrose (30.0g), agar (20.0g), streptomycin (0.3g) and distilled water (1000mL). The plates were then flooded with sterile distilled water, brushed with sterile camel hairbrush smoothly without disturbing the mycelial growth and filtered through sterile filter. The concentration of the filtrate was adjusted to 10^6 spores/mL with sterile distilled water and used as inoculums for further studies. For the preparation of adsorbent, 1mL (10^6 spores) of fungal spore suspension was inoculated into 100mL of sterile Czapek-Dox broth (composition is similar to Czapek-Dox agar medium devoid of agar alone) in a 250mL Erlenmeyer flask and incubated at 27±2°C for 5 days in an orbital shaker at 125 rpm. After the incubation period, the mycelium developed as pellets was separated by filtration through Watman No.1 filter paper and washed with generous amounts of deionized water until free from the media components. The washed, live mycelia pellets were used as biosorbent as such [10]. The mycelia pellets were also subjected to pretreatment to enhance the efficacy of dye adsorption. The pretreatment employed were: Autoclaving: the biomass was subjected to autoclaving for 30 minutes at 121°C at 18psi. The mycelial pellets/ biomass after each pretreatment was washed with generous amount of deionized water (pH-6.8) until the pH of the wash solution was close to that of deionized water (pH 6.8-7.2).

Preparation and estimation of adsorbate

Reactive orange 107 used as a model reactive dye in the present work were procured from dye industry in Mumbai. RO 107 is based on vinyl sulphone reactive group, which have negative charges in aqueous solutions. They have moderate reactivity and are used for printing and dyeing. The general characteristics of RO 107 include, chemical formula=$C_{16}H_{18}N_4O_{10}S_3Na$; λ_{max}=415 nm. Standard solutions containing 10 to 50 mg/L of the dye were prepared by diluting a stock solution of 1000mg/L of RO 107 dye studied. The analysis of RO 107 was estimated spectrophotometrically by monitoring the absorbance at 415nm using UV-VIS spectrophotometer (Shimazdu, UV-1601, Japan).

Reactive Orange 107

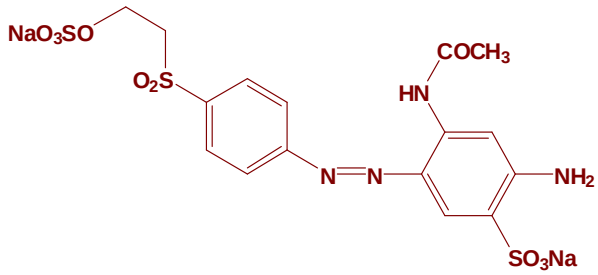


Figure 1. Structure of Reactive Orange 107 dye

Batch biosorption studies

Adsorption experiments were carried out by agitating 1g of asorbent with 50 mL of various adsorbate solution of concentration ranging from 10 to 50 mL and pH 7, 150 rpm and 30±2ºC in a thermostated rotary shaker. The flasks were withdrawn at predetermined time intervals. The adsorbate and adsorbent were separated by centrifugation at 3000rpm for 20 min. The remaining adsorbate concentration in the supernatant was determined from which the amount of dye adsorbed by the adsorbent (q, mg/g) was calculated. Control experiments were carried without adsorbent to estimate the adsorbate removal due to adsorption onto the walls of the flasks. It was observed that adsorption onto the container walls were negligible. A study was carried out with different dosages of adsorbent (0.5-3.0g/50mL) for the equilibrium time to determine the effect of adsorbent dose on RO107 removal. The effect of pH on the adsorption process was studied by varying the pH of 50mL of 50mg/L adsorbate solution in the range of 2 to 9 using dilute HCl and NaOH solutions, while keeping the other experimental parameters at the values described earlier.

Desorption experiments

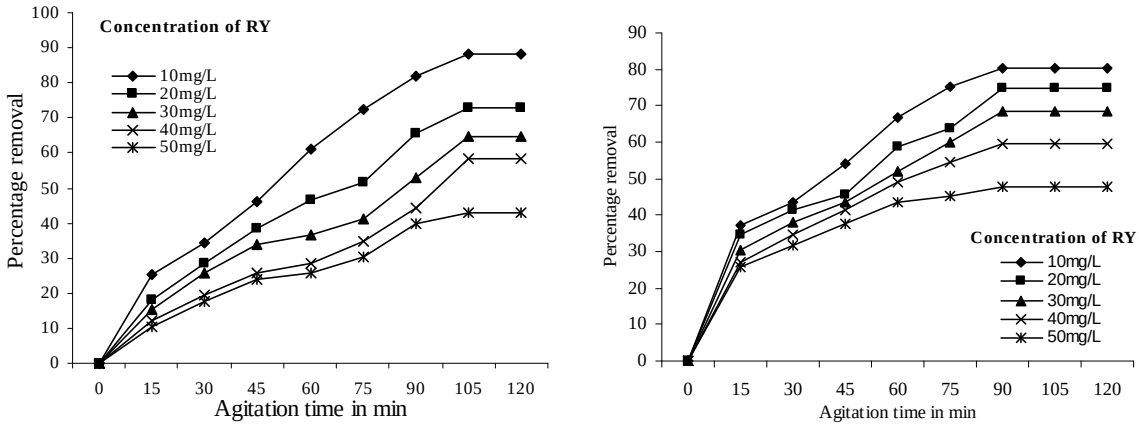
Desorption studies were carried out with adsorbate-laden adsorbent obtained from a batch process, in which the adsorbate solutions (30mg/L of RY and RB) were treated for the optimium contact time. The mycelium was washed gently with distilled water to remove unadsorbed dye. Several such samples were prepared. The spent adsorbent was then agitated at 200rpm for optimum contact time with 50 mL of distilled water, and adjusted to different pH values in the range of 2 to 10. The desorbed dye was estimated spectrophotometrically as mentioned earlier. Similarly desorption studies were carried out separately in 50 mL of 0.1-0.6N NaOH solutions. The flasks were agitated at 200 rpm until equllibrium was reached. All experiments were carried out in duplicate and the mean values are presented. The error obtained was ±2%.

RESULTS AND DISCUSSION

Effects of contact time and concentration of dye on adsorption

The effect of initial dye concentration on the rate of adsorption onto native, heat-treated fungal biomass was studied. The experiments were carried out at fixed adsorbent dose (1 g), in neutral pH with 50mL of different initial concentrations of RO107 (10-50mg/L) for different time intervals at 30°C. Autoclaved biomass showed the highest removal of RO 107 dye, where the removal was 8.04, 14.92, 20.49, 23.84 and 23.95mg/L for 10, 20, 30, 40 and 50 mg/L RO 107 solution. In live biomass the removal was 8.81, 14.54, 19.38, 20.36 and 21.45 mg/L for 10, 20, 30, 40 and 50mg/L RO 107 solution. The equilibrium time of RO 107 adsorption slightly varied with adsorbents and adsorbate concentration (Fig.2) which was 105 min and 90 min for live and autoclaved biomass respectively this may be due to affinity of adsorbates to different adsorbents.

Figure 2. Effect of Agitation time and initial dye concentration for Reactive orange 107 biosorption onto (a) live biomass and (b) autoclaved biomass of *Alternaria raphani*.



The amount of dye adsorbed (mg/g) increased with increase in contact time and reached equilibrium. Thus, it is clear that the removal of dyes depends on the concentration of the dye. The initial rapid phase may be due to availability of more number of adsorption/vacant sites, as a result there exists an increased concentration gradient between adsorbate in solution and adsorbate in the adsorbent [11].

Proposed Binding Mechanism:

The main functional groups of protonated *A.raphani* biomass are compromised of carboxyl (B-COO^-), phosphonate (B-HPO_4^-) and amine (B-NH_3^+) sites. Of these functional groups, the positively charged amine groups of the biomass are mainly responsible for reactive dye biosorption, as reactive dyes are negatively charged in aqueous solution. These negatively charged dye anions will exhibit electrostatic attraction toward the positively charged amine groups of the biomass surface. This is the reason the maximum biosorption occurred at acidic pH, which coincides with the suggestion of [12, 13] that the protonated amine could act as a binding site for the binding between the adsorbent and anionic dyes. The reduction in the reactive dye uptake with increasing pH was possible due to the electrostatic repulsion between the negative sulfonate groups (dye-SO_3^-) of the reactive dyes and negative carboxyl and

phosphonate groups on the biomass [14]. Cells when to death (or) autoclaving can suffer rupture and denaturation of cell wall that can allow free access of adsorbates to cell wall binding sites [15, 16, 17 and 18]. This could be the reason for enhanced dye adsorption by autoclaved mycelium as observed in the present study. Fu and Viraraghavan [16] also recorded an enhanced removal of congo red by autoclaved *A.niger* biomass in comparison with live biomass.

Adsorption kinetics

To study the adsorption kinetics, two kinetic models were used, which included Lagergren [19] and pseudo-second order models. In order to obtain the rate constants and equllibrium dye uptake, the straight-line plots of log were made at different initial dye concentrations. If the intercept did not equal to the experimental equilibrium dye uptake then the reaction was not likely to be first order even if plot had high correlation coefficient with the experimental data [20]. The rate constants, predicted equilibrium uptakes and the corresponding correlation coefficients for all concentration tested are summarized in Table 1 and 2.

For lagergren plot, correlation coefficients were from 0.8085 to 0.9726, but the calculated Q_e was not equal to experimental Q_e, suggesting the insufficiency of the model to fit the kinetic data for the initial concentrations examined. The reasons for these differences in the Q_e values was that there was a time lag, possibly due to a boundary layer/ external resistance controlling at the beginning of the adsorption process [21]. In most cases, the lagergren model does not fit the kinetic data well for the whole range of contact time, and generally underestimate the Q_e values (Ho and McKay, 1998).

Table 1. Kinetic parameters for the reactive orange 107 dye biosorption with live biomass of *Alternaria raphani* at different initial dye concentrations

Initial concentration of dye (mg/L)	$(Q_e)_{exp}$ (mg/g)	Lagergren model			Pseudo-second-order model		
		K_1(L/min)	Q_e(mg/g)	R^2	K_2(g/mg min)	Q_e(mg/g)	R^2
10	8.81	0.030	6.8	0.9199	5.7	8.92	0.9082
20	14.54	0.024	10.8	0.8682	2.7	15.94	0.9081
30	19.38	0.017	12.0	0.8962	2.1	21.46	0.8887
40	20.36	0.014	18.4	0.9206	2.3	22.50	0.8271
50	21.45	0.026	19.1	0.8085	2.4	24.92	0.8724

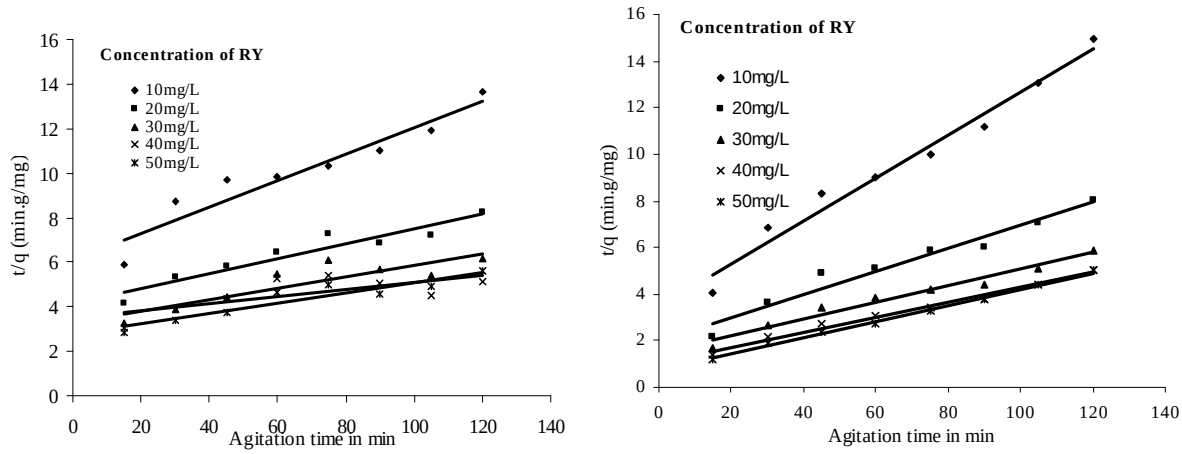
The pseudo-second order model is based on the sorption capacity on the solid phase. Contrary to other well-established models, it predicts the behavior over the whole range of studies and it is in agreement with the chemisorptions mechanism being the rate controlling step [21]. This was consistent with the better results obtained with the pseudo-second order model (Table 1 and 2) [22, 23]. Correlation coefficients were always higher than 0.95, and the lowest correlation coefficients in this case was better than the first order model correlation coefficients (Fig 3 (a)

and (b)). The values of predicted equilibrium sorption capacities showed reasonably good agreement with the experimental equilibrium uptake value.

Table 2. Kinetic parameters for the reactive orange 107 dye biosorption with autoclaved biomass of *Alternaria raphani* at different initial dye concentrations

Initial concentration of dye (mg/L)	$(Q_e)_{exp}$ (mg/g)	Lagergren model			Pseudo-second-order model		
		K_1(L/min)	Q_e(mg/g)	R^2	K_2(g/mg min)	Q_e(mg/g)	R^2
10	8.04	0.035	9.36	0.9721	2.5	10.79	0.9736
20	14.92	0.022	12.58	0.9381	1.2	20.20	0.9752
30	20.49	0.025	18.79	0.9298	8.7	23.93	0.9658
40	23.84	0.030	20.65	0.9533	9.5	26.25	0.9859
50	23.95	0.037	22.29	0.9726	1.7	27.65	0.9952

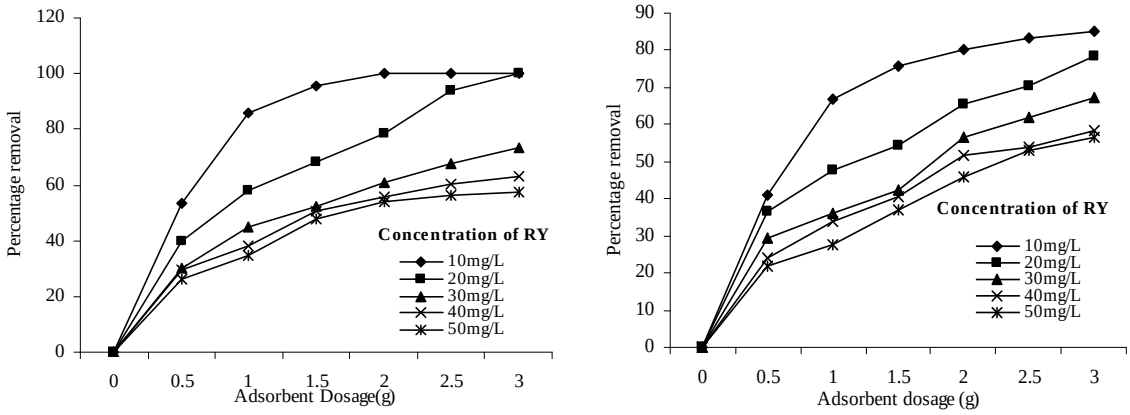
Figure 3. Pseudo second order plots for Reactive orange 107 biosorption onto (a) live biomass of and (b) autoclaved biomass of *Alternaria raphani*



Effect of carbon dosage on RO 107 removal

The removal of dye increased with increasing carbon dosage (0.5 to 3.0 g of biomass) respectively. Thus, 3 g was considered as optimum dosage for further studies. Increase in adsorbent dosage increased the percentage removal of dye which is due to the increase in adsorbent surface area of the biosorbent (Figure 4).

Figure 4. Effect of carbon dosage on Reactive orange 107 biosorption onto (a) live biomass and (b) autoclaved biomass of *Alternaria raphani*



Adsorption Isotherms

The data obtained from equilibrium studies were analyzed according to Langmuir and freundlich adsorption isotherms. The langmuir and freundlich equations are commonly used to describe adsorption isotherms at a constant temperature for water and waste water treatment applications. The Langmuir model is valid for monolayer sorption onto a surface with a finite number of identical sites. The distribution of dyes between the solid-solution interface equations has been described by the Langmuir equation [24]. The well known expression of the Langmuir model is given by equation no (1).

$$q_e = Q_o b C_e / (1+bC_e) \longrightarrow 1$$

where q_e (mg/g) and C_e (mg/g) are the amounts of adsorbed dye per unit weight of adsorbent and unadsorbed dye concentration in solution at equation respectively. Q_o is the maximum amout of the dye bound per unit weight of adsorbent to form a complete monolayer on the surface at high C_e,and b is a constant related to the affinity of the binding sites (L/mg).

The empirical Freundlich equation based on sorption into a heterogenous surface is given as

$$q_e = K_F C_e^{1/n} \longrightarrow 2$$

where K_f and n are the freundlich constants for the system, which are indicators of adsorption capacity and intensity respectively [25]. The values of the freundlich and Langmuir parameters were obtained respectively from the linear correlation between the values of C_e/q_e and C_e and log q_e and log C_e. The adsorption isotherm parameters along with the correlation coefficients are presented in Table 3.

The linear relationships were evidenced by the R^2 values (for the Langmuir model, 0.9868 and 0.9929 for live and autoclaved biomass, respectively for the fruendlich model, 0.9505 and 0.7394 for live and autoclaved biomass respectively).

Table 3. Langmuir and Freundlich model constants

	Langmiur model					Freundlich model		
Alternaria raphani	Q_o(mg/g	b, L/mg	R^2	C_0, mg/L	R_L	K_f, L/mg	n	R^2
				10	0.186			
				20	0.102			
Live biomass	23.81	0.438	0.9868	30	0.071	0.0503	1.084	0.9505
				40	0054			
				50	0.044			
				10	0.307			
				20	0.181			
Autoclaved	28.74	0.226	0.9929	30	0.129	1.038	1.038	0.7394
biomass				40	0.099			
				50	0.081			

Table 4. Comparison of Q_o values for various adsorbents

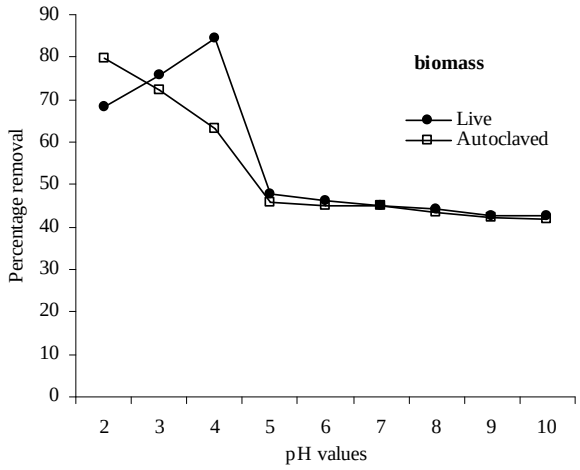
Dyes	Adsorbent	Q_o(mg/g)	Reference
Reactive orange 107	Live biomass of *Alternaria raphani*	23.81	This work
	Autoclaved biomass of *Alternaria raphani*	28.74	This work
Reactive Yellow 176	Biomass fly ash	3.65	[26]
	Modified zeolite	88.5	[27]
	Modified clay (sepiolite)	169.1	[27]
Reactive Yellow 2	Activated carbon (300–500m)	209.4	[28]
	Activated sludge	119.4	[29]
	Aeromonas sp	124.3	[30]
	Escherichia coli	52.4	[30]
	Pseudomonas luteola	102.6	[30]
Reactive Yellow 64	Calcined alunite	236	[31]
Reactive Yellow 64	Alunite	5	[31]
Sunset Yellow	Powdered peanut hull	13.99	[32]
Reactive yellow208	Hydrotalcite	47.8	[33]
Remazol golden yellow	*Kluveromyces marxianus*	33	[34]
Reactive orange 16	*Rhizopus arrhizus*	190	[1]

These indicates the applicability of the two adsorption isotherm and the monolayer coverage on the adsorbent surface. The Q_o from Langmuir isotherm indicates that the adsorption capacity of RO 107 was greater for the autoclaved biomass (28.74 mg/g) than the live biomass (23.81mg/g) of *Alternaria raphani*. The observed R_L values indicate favorable adsorption of the RO 107 on live and autoclaved biomass of *Alternaria raphani* ($0<R_L<1$).

Effect of pH on RO 107 removal

Biosorption of dye decreased with increasing pH. Lower adsorption of both dyes at alkaline pH was probably due to the presence of excess of hydroxyl ions competing with the dye anions for the adsorption sites. A maximum removal of 84.6%, 79.9% of RO 107 dye by live and autoclaved biomass was observed in values of pH 4 and 3 respectively (Figure 5). Fungal cell wall is predominantly known to contain amine, carboxyl and phosphonate groups [15]. Generally the carboxyl and phosphonate groups can't bind to reactive (direct dyes which possess negative charges in the aqueous medium, whereas positively charged amine groups are believed to be the binding sites for anionic/reactive dyes [35].

Figure 5. Effect of pH on Reactive orange 107 biosorption onto (a) live biomass and (b) autoclaved biomass of *Alternaria raphani*.

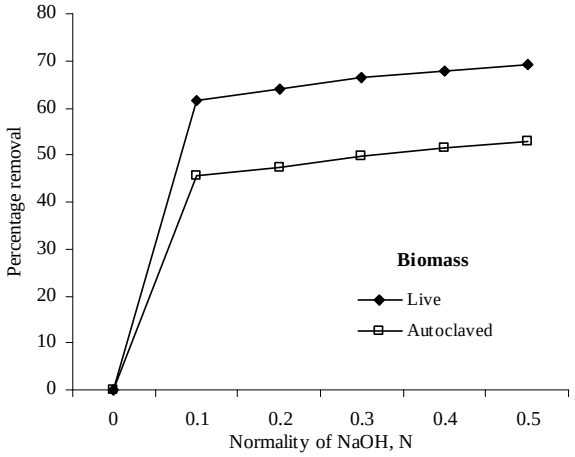


At a pH of higher acidity, that functional groups are protonated and to maintain neutrality in an aqueous environment, negative counter ions are adsorbed. These ions are mobile, and are exchanged by ions from the dyes at appropriate pH. This result shows a tendency towards great adsorption for anionic dyes in the acidic pH range [36].

Desorption studies:

Regeneration of the adsorbent may take the treatment more economical. Attempts were made to degenerate color from the mycelial pellets using various strengths of NaOH (0.1-0.6N). The percent desorption increased with increasing NaOH concentration in the aqueous medium (Fig.6) and attained a maximum desorption at 0.6N NaOH solution (Figure 6). The effect of percentage desorption was inversely correlated to pH effect, indicating that ion exchange was probably the major mode of adsorption process. Similar results were observed for the adsorption of Congo red by coir pith carbon [37].

Figure 6. Desorption studies for Reactive orange 107 biosorption onto (a) live biomass and (b) autoclaved biomass of *Alternaria raphani*.



CONCLUSION

The present study shows that autoclaved mycelial pellets/biomass of *Alternaria raphani* is an effective adsorbent for the removal of RO 107 from aqueous solutions compared to live biomass. Adsorption of RO 107 dye on to live and autoclaved biomass of *Alternaria raphani*, followed Langmuir and Freundlich isotherms, showed a good fit of the data. The adsorption capacity, (Q_o) from the Langmuir isotherm was found to be maximum for autoclaved adsorbent with 28.74mg/g. The suitability of first and second order kinetic models for the sorption of both dyes was also discussed. It was decided that the adsorption kinetics of both adsorbents obeyed the second order adsorption model. Adsorption was found to be pH dependent. Desorption tests has revealed the possibility of both chemisorptions and physisorption mechanism involved in the removal of RO 107. The whole process is more economical and eco-friendly because of the possibility of reusing the adsorbent (fungal biomass), adsorbate (dyes) and the solvent/desorption medium (NaOH). These results suggest that *Alternaria raphani* biomass can be used in biotreatment process as biosorbent for reactive dyes.

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