

DECOLORIZATION OF A TEXTILE DYE, REACTIVE RED 198 (RR198), BY *Aspergillus* *parasiticus* FUNGAL BIOSORBENT

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Abstract - The decolorization potential of textile dye Reactive Red 198 (RR198) by *Aspergillus parasiticus* fungal biosorbent has been investigated as a function of initial pH, contact time, biosorbent and initial dye concentration in a batch system. Maximum dye biosorption capacity 1.03×10^{-4} mol g⁻¹ was observed at pH 2.0 and 2.0 g L⁻¹ of biosorbent concentration. Biosorption equilibrium was attained within 50 min. The equilibrium data followed Langmuir, Freundlich and Dubinin-Radushkevich isotherm models at 20, 30, 40 and 50°C. An increase in the biosorption capacity of *A. parasiticus* with temperature showed that the decolorization process is endothermic. Results indicated that *Aspergillus parasiticus* was an effective candidate for textile dye RR198 removal from aqueous solutions.

Keywords: *Aspergillus parasiticus*; Biosorption; Decolorization; Isotherm; Reactive Red 198.

INTRODUCTION

As a result of many industrial activities, the discharge of large quantities of organic pollutants into the receiving waters has caused significant environmental pollution. Synthetic dyes are a group of organic pollutants that are extensively used in textile, paper, printing and dyehouses (Aksu, 2005).

The colored effluents damage the aesthetic quality of water and reduce light penetration and photosynthesis and also some of the dyes are toxic or mutagenic, carcinogenic and allergenic (Aksu and Çağatay, 2006; Kumar et al., 2006; Bakshi et al., 1999; O'Mahony et al., 2002). According to the survey of the Ecological and Toxicological Association of the Dyestuffs Manufacturing Industry (ETAD), over 90% of some 4000 dyes have LD₅₀ values greater than 2000 mg kg⁻¹ (Shore, 1996; Robinson et al., 2001). Hence decolorization of the dye-bearing effluents is of great importance.

The traditional physical or chemical decolorization methods including coagulation, flocculation, ion

exchange, irradiation, precipitation, ozonation and adsorption or a combination of these methods have been used for dye removal from wastewaters (Aksu, 2005; Fu and Viraraghavan, 2001).

However application of these methods is somewhat restricted due to some limitations such as operational costs, formation of hazardous by-products, intensive energy requirement (Padmesh et al., 2005) and limited adaptability to a wide range of effluents (Fu and Viraraghavan, 2001). The use of biological materials as sorbents for the treatment of colored effluents appeared as a potential alternative process to conventional methods (Kumar et al., 2006). Currently, numerous studies have focused on the dye biosorption and biodegradation abilities of microorganisms, including some bacteria, fungi and algae. Live and dead forms of microbial cells can be used in the decolorization process and the main mechanisms of these forms are biodegradation and biosorption, respectively (Fu and Viraraghavan, 2001).

In many treatment processes, compared with the living cells, the dead cells offer some advantages,

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which are following; dead organisms are not subject to toxicity limitations, they do not require a continuous supply of nutrients, they can be regenerated simply and reused for many cycles (Akar and Tunali, 2005; Padmesh et al., 2005), biomass can be stored until the use and their operation is easy. Also they can be obtained from industrial sources as a waste product from established fermentation processes (Fu and Viraraghavan, 2001).

Compounds such as chitin, lipids, amino acids and other cellular components present on the cell walls of microorganisms provide active binding sites for dye molecules and biosorption processes based on interactions between the dyes and binding sites in a manner of surface adsorption, ion exchange, complexation, chelation and microprecipitation (Aksu, 2001). The dye-microorganism interactions depend on different factors varying from the chemical structure of the dye being studied to the type of biosorbent used, as well as operating conditions (Kargi and Ozmişci, 2004; Šafaříková et al., 2005).

Currently research is focused on finding optimal microbial biosorbent and reaction conditions in order to develop optimal processes enabling decolorization of large volumes of waters (Šafaříková et al., 2005).

The water-soluble reactive dyes are one of the most important groups of dyes used in the textile dyeing industries due to their highly brilliant colors (Aksu, 2003). Reactive Red 198 (RR198) is also among this group of dyes and it was selected as the model reactive dye for this work.

Aspergillus parasiticus and *Aspergillus flavus* fungal strains have only limited parasitic abilities, yet they can successfully colonize and produce aflatoxin in developing host tissue in the field (Brown et al., 1999; Mayer et al., 2003). However, several studies have been reported in the literature on the biosorption potential of the dead form of *A. flavus* (Hafez, et al., 1997; Deepa et al, 2006; Akar and Tunali, 2006). The commercially available and easily cultivable fungus *A. parasiticus* was chosen as a biosorbent material because of a lack of

information on its dye sorption ability.

This research aims to investigate the removal of a textile dye RR198 from aqueous solutions by *A. parasiticus* cells as an alternative biosorbent. The effects of various operating parameters were studied including pH, biosorbent dosage and contact time. Biosorption was analyzed by the Freundlich, Langmuir and Dubinin-Radushkevich (D-R) isotherm models at 20, 30, 40 and 50°C.

MATERIALS AND METHODS

Biosorbent Preparation

The filamentous fungus, *A. parasiticus* (NRRL 502), was obtained from the culture collection of the Biology Department of Eskişehir Osmangazi University. The stock cultures were routinely maintained on agar medium at +4°C and liquid growth conditions for *A. parasiticus* were described in our previous studies (Kiran, et al., 2005).

The live *A. parasiticus* cells were separated from the growth medium by filtration after seven days of growth, spread on petri dishes and dried in an oven at 60°C overnight. They were then powdered using a mortar and pestle and sieved to select 150 µm particles for use as a biosorbent.

Dye Solutions

The dye used in this study was Reactive Red 198 (RR198) obtained from a local textile company and used without further purification. Its chemical structure is shown in Fig. 1. A stock solution (1 g L⁻¹) of RR198 was prepared by dissolving an accurate quantity of dye in bidistilled water and other concentrations varying between 100 and 300 mg L⁻¹ were obtained by dilution. Fresh dilutions were used for each biosorption experiment. The pH of the working solutions was adjusted to the required values by 0.1 M HCl or 0.1 M NaOH. All the chemicals used were analytical grade.

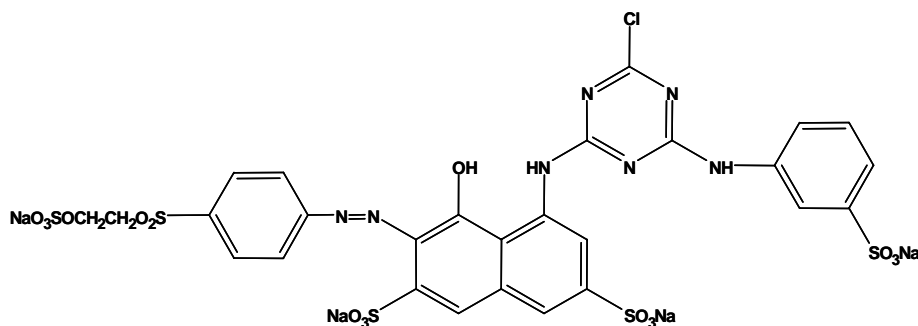


Figure 1: Chemical structure of RR198

Dye Biosorption Experiments

Dye biosorption experiments were performed on a magnetic stirrer. 0.1 g of dried biosorbent of *A. parasiticus* was put in a 100 mL beaker and 50 mL of dye solution (100 mg L^{-1}) was added to it. The mixture was stirred at 200 rpm at 20°C for 1 h to elucidate the optimum conditions of pH, biosorbent dosage and contact time.

The effect of solution pH on dye biosorption capacity of the fungal biosorbent was determined by adjusting the pH values between 1 and 10, adding freshly prepared 0.1 M HCl or 0.1 M NaOH solutions. The optimum pH value was used throughout all biosorption experiments. The effect of biosorbent concentration was investigated by using biosorbent samples ranging from 0.4 to 4.0 g L^{-1} . This was followed by the assessment of the effect of equilibrium time, varied between 10 to 90 min, on the dye biosorption capacity of the biosorbent. The initial dye concentration was varied from 100 to 300 mg L^{-1} and the isotherm appropriateness of the biosorption results was evaluated for the Langmuir, Freundlich and Dubinin-Radushkevich (D-R) isotherm models at temperatures of 20, 30, 40 and 50°C under the optimum conditions.

The biosorption yield (%) and the amount of dye sorbed by the biosorbent were calculated using the following general equations:

$$\text{Biosorption yield (\%)} = [(C_i - C_e) \cdot 100] / C_i \quad (1)$$

$$q_e = [(C_i - C_e) \cdot V] / M \quad (2)$$

where,

q_e : amount of dye biosorbed (mg g^{-1})

C_i : initial dye concentration in solution (mg L^{-1})

C_e : final dye concentration in solution (mg L^{-1})

V: volume of the medium (L)

M: amount of the biosorbent used in the reaction mixture (g).

Biosorbent was separated by centrifugation at

500 rpm for 5 min. The dye concentrations in the supernatants were determined using a spectrophotometer, (UV/Vis, Unicam UV2-100) at λ_{max} 515 nm. All the experiments were carried out in triplicate and data presented are the mean values from these independent experiments. Error bars are indicated wherever necessary. All statistical analysis was done using SPSS 9.05 for Windows.

RESULT AND DISCUSSION

Effect of pH

The pH value of the biosorption medium significantly influences the sorption properties of fungal biosorbents (Aksu and Tezer, 2000). The pH dependence of dye biosorption is related to both protonation or deprotonation of the functional groups of the biopolymers on the biosorbent surface and the ionization potential of complex dye molecules in solution (Maurya et al., 2006). Fig. 2 represents the effect of pH on the biosorption of RR198 onto dried *A. parasiticus* cells. As seen in this figure, the maximum biosorption of RR198 on *A. parasiticus* was found as 47.56 mg g^{-1} at pH 2.0. Because dye binding sites on the biosorbent were closely associated with H_3O^+ at lower pH, which cause an increase in the electrostatic attraction between positively charged biosorbent surface and the negatively charged dye molecules. The biosorption capacity of the dried cells of *A. parasiticus* decreased when the pH of the solution increased from 2.0 to 5.0. Under alkaline conditions biosorption capacity significantly decreased ($p < 0.05$). The decrease of the biosorption capacity with increase in the medium pH can be explained by the dye binding sites on the biosorbent surface becoming negatively charged, restricting the approach of the dye molecules as a result of repulsive forces.

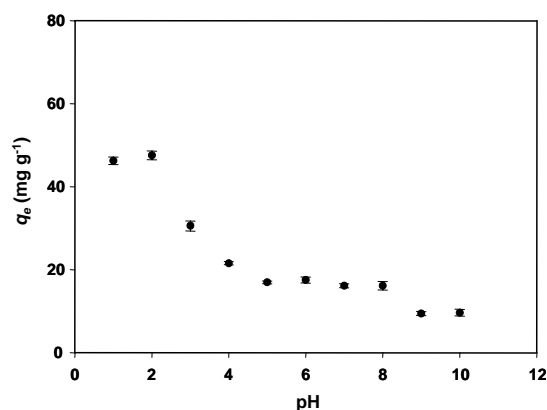


Figure 2: Effect of pH on the biosorption of RR198 by *A. parasiticus* at 20°C

Effect of Biosorbent Dosage

In order to determine the effect of the biosorbent dosage on the biosorption efficiency of RR198, the amounts of biosorbent added into the biosorption medium were varied from 0.4 to 4.0 g L⁻¹ and the results are presented in Fig 3. With increase in the biosorbent dosage, from 0.4 to 2.0 g L⁻¹, the percentage decolorization of RR198 increased from 29.40% to 98.57%, which is the maximum value obtained as the number of possible binding sites increased. Further increase up to 4.0 g L⁻¹ did not change the maximum RR198 biosorption capacity of the biosorbent and it almost stayed constant ($p > 0.05$). This is because, at higher biosorbent concentrations, there is a very fast biosorption onto the biosorbent surface that produces a lower solute concentration in the solution than when the biosorbent concentration is lower (Kumar et al., 2006).

Effect of Contact Time

Fig. 4 indicates the effect of treatment time on the biosorption of RR198 dye onto biosorbent. High biosorption rates were observed at the beginning of

the biosorption and the process reached the plateau values within ca. 50 min. After this period, no considerable change was observed in the RR198 biosorption capacity of the biosorbent ($p > 0.05$) and it was fixed as the optimum contact time.

Biosorption Isotherms

Equilibrium isotherms are plots of the quantity of sorbate retained on the biosorbent (q_e) as a function of sorbate concentration in the liquid phase (C_e). Biosorption data are commonly represented by an equilibrium isotherm. The equilibrium biosorption isotherm is of fundamental importance for the design and optimization of the biosorption system for the dye decolorization studies (Maurya et al., 2006). Therefore, the equilibrium biosorption isotherms are one of the most important data for understanding the mechanism of the biosorption (Tunali et al., 2006). Several isotherm equations are available and three important isotherm models, namely Langmuir, Freundlich and Dubinin-Radushkevich (D-R) isotherm models were chosen in this study to describe the biosorption equilibrium and the model plots at various temperatures are shown in Figs. 5, 6 and 7.

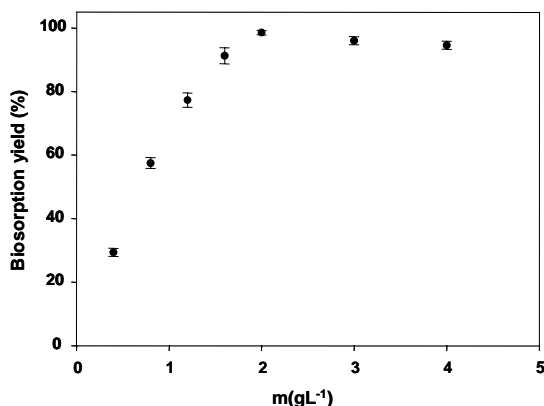


Figure 3: Effect of biosorbent dosage on the biosorption of RR198 by *A. parasiticus* at 20°C

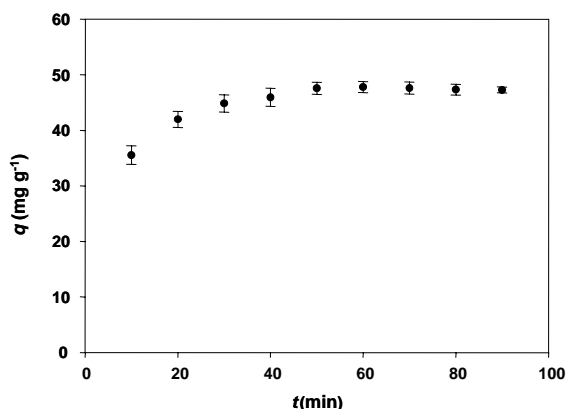


Figure 4: Effect of contact time on the biosorption of RR198 by *A. parasiticus* at 20°C

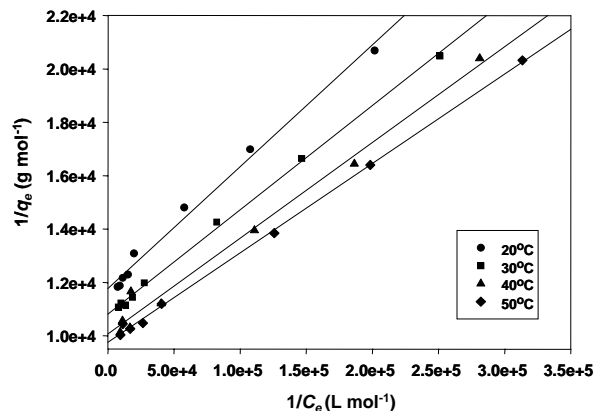


Figure 5: Langmuir plots for the biosorption of RR198 by *A. parasiticus* at various temperatures

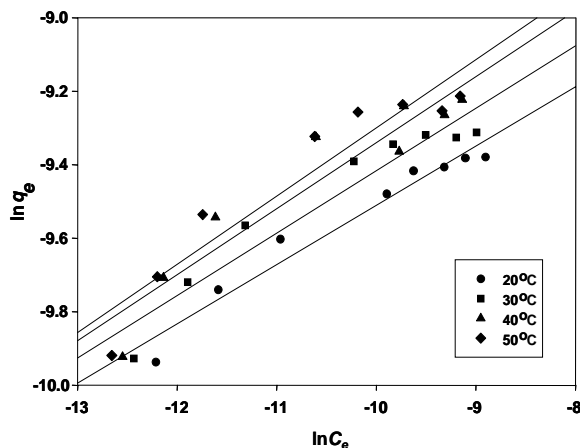


Figure 6: Freundlich plots for the biosorption of RR198 by *A. parasiticus* at various temperatures

The Langmuir isotherm is represented by the following equation (Langmuir, 1918):

$$q_e = \frac{q_{\max} K_L C_e}{1 + K_L C_e} \quad (3)$$

where q_e is the biosorbed dye concentration (mol g^{-1}), C_e is the residual dye concentration at equilibrium (mol L^{-1}), q_{\max} is the monolayer biosorption capacity of the biosorbent (mol g^{-1}), and K_L is the Langmuir biosorption constant (L mol^{-1}).

The Freundlich isotherm involves heterogeneous biosorption and the empirical Freundlich equation is (Freundlich, 1906):

$$q_e = K_F C_e^{1/n} \quad (4)$$

where K_F (L mg^{-1}) and n are Freundlich biosorption isotherm constants depend on several environmental factors.

The Dubinin-Radushkevich (D-R) isotherm is another less commonly used model. It was applied to distinguish between physical and chemical biosorption (Benhammou et al., 2005) of dye. The D-R isotherm equation (Dubinin and Radushkevich, 1947) is:

$$q_e = q_m e^{-\beta \varepsilon^2} \quad (5)$$

where q_m is the theoretical sorption capacity, and ε is the Polanyi potential; β ($\text{mol}^2 \text{J}^{-2}$), is the activity coefficient related to mean sorption energy, E (kJ

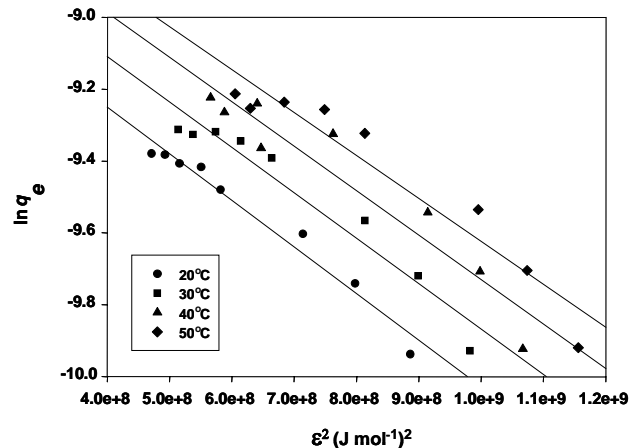


Figure 7: D-R plots for the biosorption of RR198 by *A. parasiticus* at various temperatures

mol^{-1}). This is the free energy of the transfer per mole of the biosorbate from infinity to the sorbent surface and can be calculated using the following equation (Hobson, 1969; Hasany and Chaudhary, 1996; Dubey and Gupta, 2005):

$$E = \frac{1}{(2\beta)^{1/2}} \quad (6)$$

The magnitude of this parameter is useful for information about the type of biosorption process such as chemical ion exchange or physical biosorption. It was found that E values varied from 19.634 to 20.455 kJ mol^{-1} , which are bigger than the energy range of adsorption reactions, 8-16 kJ mol^{-1} . The type of biosorption may be interpreted as chemical biosorption (Helfferich, 1962; Onyango et al., 2004).

The Langmuir, Freundlich and D-R parameters for the biosorption of RR198 dye onto *A. parasiticus* are listed in Table 1. The Langmuir isotherm model fits very well for the RR198 biosorption process based on the r^2 values in Table 1. Biosorption capacity of *A. parasiticus* increased with increasing temperature from 20 to 50°C and was found to be $1.03 \times 10^{-4} \text{ mol g}^{-1}$ at 50°C. This observation implies that the biosorption of RR198 by *A. parasiticus* biosorbent is endothermic nature. The biosorption capacity of *A. parasiticus* obtained for RR198 in this study is comparable to and moderately higher than that of many corresponding biosorbents reported in the literature (Table 2).

Table 1: Biosorption isotherm constants for the biosorption of RR198 by *A. parasiticus* at various temperatures

t (°C)	Langmuir					Freundlich			Dubinin-Radushkevich (D-R)			
	q_{\max}		K_L (L mol^{-1})	Γ_L^2	R_L	n	K_F (L mol^{-1})	Γ_F^2	q_{\max} (mol g^{-1})	β ($\text{mol}^2 \text{kJ}^{-2}$)	Γ_{D-R}^2	E (kJ mol^{-1})
	(mol g^{-1})	(mg g^{-1})										
20	8.50×10^{-5}	83.66	2.579×10^5	0.990	0.0126	6.204	3.712×10^{-4}	0.954	1.62×10^{-4}	1.297×10^{-3}	0.968	19.634
30	9.25×10^{-5}	91.04	2.766×10^5	0.998	0.0117	5.883	4.455×10^{-4}	0.924	1.83×10^{-4}	1.264×10^{-3}	0.943	19.889
40	9.94×10^{-5}	97.83	2.805×10^5	0.985	0.0116	5.561	5.305×10^{-4}	0.890	2.05×10^{-4}	1.237×10^{-3}	0.908	20.105
50	1.03×10^{-4}	101.37	2.913×10^5	0.999	0.0111	5.384	5.859×10^{-4}	0.899	2.19×10^{-4}	1.195×10^{-3}	0.921	20.455

Table 2: Biosorption results of different textile dyes from the literature for various biosorbents and operating conditions

Biosorbent material	Dye	Rating conditions					References
		pH	t (°C)	Initial concentration (mg L ⁻¹)	Biosorbent dosage (g L ⁻¹)	Biosorption capacity (mg g ⁻¹)	
<i>Rhizopus arrhizus</i>	Reactive Orange 16	2.0	-	0-500	1.0	190	O'Mahony et al.,2002
<i>Rhizopus arrhizus</i>	Reactive Red 4	2.0	-	0-500	1.0	150	O'Mahony et al.,2002
<i>Saccharomyces cerevisiae</i>	Remazol Blue	3.0	25	10-400	0.52	84.6	Aksu, 2003
<i>Saccharomyces cerevisiae</i>	Remazol Black B	3.0	25	10-400	2.87	88.5	Aksu, 2003
<i>Saccharomyces cerevisiae</i>	Remazol Red RB	3.0	25	10-400	2.60	48.8	Aksu, 2003
<i>Kluyveromyces marxianus</i>	Remazol Black B	-	-	10-500	1.0	37.0	Bustard, et al.,1998
<i>Escherichia coli</i>	Reactive Blue 5	3.0	28	200	-	89.4	Hu, 1996
<i>Escherichia coli</i>	Reactive Red 22	3.0	28	200	-	76.6	Hu, 1996
<i>Escherichia coli</i>	Reactive Violet 2	3.0	28	200	-	65.5	Hu, 1996
<i>Escherichia coli</i>	Reactive Yellow 2	3.0	28	200	-	52.4	Hu, 1996
<i>Aspergillus parasiticus</i>	Reactive Red 198	2.0	20	100-300	2.0	83.66	This study
<i>Aspergillus parasiticus</i>	Reactive Red 198	2.0	50	100-300	2.0	101.37	This study

The essential characteristics of the Langmuir isotherm can be expressed by a dimensionless constant, R_L , referred to as the equilibrium parameter. R_L is described by the following equation:

$$R_L = \frac{1}{1 + K_L C_o} \quad (7)$$

where C_o is the highest dye concentration (mol L⁻¹). The value of R_L is used for the interpretation of the sorption type (Weber and Chakravorti, 1974; Hall *et al.*, 1966). Calculated R_L values for this study are found to be between 0.0111 and 0.0126 in the temperature range of 20-50°C and these results indicate the biosorption process is favorable.

The Freundlich model constants K_F and n are indicators of biosorption capacity and biosorption intensity, respectively (Malik, 2004) and the numerical values of these constants are included in Table 1.

CONCLUSION

A. parasiticus was found to be an effective and alternative biosorbent for the removal of RR198 textile dye from aqueous solutions. The biosorption process was affected by various experimental conditions such as pH, contact time and biosorbent concentration. Maximum dye removal was observed at pH 2.0 and 2.0 g L⁻¹ of biosorbent concentration. Biosorption equilibrium was established in 50 min. Experimental data were fitted well to the Langmuir isotherm model. The maximum dye removal percentage was determined as 98.57% at 50°C. The biosorption process is endothermic.

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