Biosorption of Corafix Yellow GD3R using *Halimeda gracilis* and its Toxicological Assessments

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Abstract

The elimination of CYGD3R with *H. gracilis* was performed under *in vitro* conditions. Optimization of process variables affecting the adsorption capacity of CYGD3R including dye concentrations, biosorbent concentrations, pH, incubation time and temperature were performed to assess the well suited optimal conditions for the extreme removal of dye. By calculating the kinetics and thermodynamics, the mechanism and feasibility of adsorption process was evaluated. The elution of the dye from the biosorbent was recorded in terms of desorption percentage using different eluents. The treated dye solution was assessed to test its toxicity against green gram seedlings and selected microbes. The results revealed that maximum dye removal (88%) was recorded at pH 6 in the medium amended with 300mg/L of dye with 200mg/L of biosorbent at 25°C within 72hrs of incubation. Moreover application of *H.gracilis* for CYGD3R removal confirmed the process of biosorption strongly comply the pseudo-second order kinetic model assisted through chemisorption. The adsorption remained endothermal, feasible and spontaneous as evident from thermodynamic study. Desorption using the eluent 0.1N HNO_3 revealed 60% recovery of the dye from the biosorbent in the first cycle and a decrease was noted in further cycle. The process of dye decolorization by *H.gracilis* was confirmed by UV-Visible spectral analysis. The phytotoxicity study revealed significant growth on green gram seedlings grown with CYGD3R treated solution, thereby proving its non-hazardous nature. Absence of inhibition of zone in the CYGD3R treated solution proves its non-toxicity towards microbes. Thus, biosorption using *H. gracilis* proves to be a sustainable strategy for dye removal from waste water through ecofriendly approach.

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Introduction

Water is a vital and indispensable requisite for all living organisms because it play a noteworthy role in determining its capability for domestic, agriculture and industrial activities and also accessibility to clean water supply should be available always. Without adequate treatment millions of contaminants are released into water bodies as a results of urbanization, industrial revolution and expanding population which thereby pose water scarcity and pollution to become a major problem.[1]

One among the leading industry in the economic sector is textiles, where human not only need it for survival, but also evolved into a form of fashion. Almost 1,00,000 diverse categories of dyes are available in the market, among azo reactive dyes occupies about 30% of annual dye material production.[2] Emission of these complex organic contaminants into the ecosystem is the major cause for the decline of water and soil quality.[3] When the dye-contaminated waste water is released into aquatic bodies, they deteriorate the water quality and make it unsafe to utilize for domestic tasks and public health.[4] The occurrence of synthetic dyes in the aquatic ecosystem reduces visibility, affects the growth of fauna and flora and also lowers the photosynthetic rate. They also pose major risk to human health by causing severe impairment to brain, central nervous system, liver, kidney as well as dysfunction of reproductive systemand also cause allergic dermatoses, respiratory illness and malignant tumour.[5,6] In the current investigation, Corafix Yellow GD3R dye, was less expensive, more productive, and requires less time to color. The chemical groups in these dyes enable them to create covalent bonds with the fabrics, and give them their distinctive properties.[7] They also have toxic effect, as they decrease the water transparency, and seem to be genotoxic to aquatic plants and animals.[8] Thus, there is a pressing requisite to spot more sensible, inexpensive and green technology for dye removal before discharging into water bodies to evade destruction of aquatic ecosystem.

Numerous conventional physicochemical techniques have been recommended for the purpose of textile effluent treatment which includes ozonation, membrane filtration, activated carbon adsorption, ion exchange, flocculation, froth flotation, electro coagulation and reverse osmosis.[9] All these techniques were extremely flexible and beneficial, but because of their high cost and disposable nature, they produce secondary waste that needs to be addressed further. It has been demonstrated that removing color from textile effluent using biological such as microorganisms, algae, seaweed, agriculture wastes and plants are extincing, ecofriendly, economical and an advantageous alternative method.[10] Indeed, using seaweeds as biosorbent for removing color and other contaminants from dye contaminated waste water is a promising and an appealing environmentally friendly technique. Seaweeds can easily grow in both fresh and saline water depending on the climate, requires less financial input to cultivate and generate least amount of sludge.[11] *H. gracilis* was nominated as the candidate seaweed in the current study, because of its strong affinity to remove the dye and the presence of reactive functional compounds on its surface.[12] Literature reveals that the removal of CYGD3R using *H. gracilis* has not been renowned, and therefore the current investigation was formulated to assess its decolorization efficiency against Corafix Yellow GD3R and to evaluate its toxicity against green gram under *in vitro* conditions.

Materials and Methods
Processing of Seaweed and Adsorbate for Adsorption Study

The seaweed was procured from the Mandapam, Tamil Nadu. The acquired seaweed was cleaned in running water, air dried, powdered and stored at room temperature in an airtight container for future investigation. The stock solution was prepared by suspending 1000mg of CYGD3R dye with 1L of water and the content was left overnight. Proper dilutions were prepared for the study from the stock solution.

Zero Point of Charge Determination

Batch equilibrium approach was performed to calculate the point of zero charge (pHzpc). To a known volume of 0.1M NaCl, the biosorbent was added and the pH (2-11) was regulated with 0.1M HCl/0.1M NaOH. The solution's initial pH ($pH_{initial}$) remained documented, and it was also endlessly stirred at 150 rpm for 24 hours. The solution's pH was determined besides the results recorded as pHfinal. A plot of pH change, $\Delta pH = (pH_{initial} - pH_{final})$, against

(pH$_{initial}$), was constructed and the biosorbent pHzpc was calculated at the point where ΔpH is zero.[13]

Decolorization of Corafix Yellow GD3R through Batch Adsorption Experiments

The batch adsorption study for dye removal was performed by varying the dosage of dye and biosorbent concentrations from 100 to 500 mg/L, pH (2-11), incubation time (24-144hrs) and temperature (20-40°C) and the pH was adjusted using 1N HCl/ 1N KOH. The decolorization efficiency of *H. gracilis* against CYGD3R was assessed after each incubation period and the decolorization percentage was observed at 580nm using the formula.[14]

Decolorization (%) =Initial absorbance - Final absorbance/ Initial absorbance X 100   ...(1)

The maximum decolorization percentage was established under optimal conditions and a subsequent study was conducted there as well.

Kinetic and Thermodynamic studies

The kinetic studies was performed at varying time intervals to impart the evidence on the sorption and rate controlling mechanism with Pseudo first order (PFO) and Pseudo second order (PSO), intraparticle diffusion (ID) models respectively. The nature of biosorption at different temperature was studied using thermodynamic factors specifically free enthalpy (ΔG°), total heat (ΔH°) and entropy (ΔS°), below optimized circumstances.

Desorption study

Using the eluents such as NaOH, CH$_3$COOH, HCl and HNO$_3$, at 0.1N concentration separately the efficacy of dye desorption was performed in three consecutive cycles to observe the dye removal from the dye-loaded biosorbent at optimized conditions. The amount of dye desorbed and adsorbed denotes the efficacy of percentage desorption.[15]

UV-Visible Spectral Analysis

The spectra of CYGD3R decolorized with *H.gracilis* was analyzed through UV-visible spectrophotometer at 300-700nm (Hitachi U2800, Tokyo, Japan) and equated with untreated dye solution to determine the process of decolorization.

Phytotoxicity Study

Vigna radiata (Green gram), was selected to demonstrate the toxicity of the CYGD3R treated and untreated dye solutions. Ten healthy seeds were sown in nine pots separately and kept under room temperature. At regular intervals, the seedlings were watered with treated and untreated dye solutions as well as with tap water which served as control. Each treatment was performed in triplicates. The seedlings were removed and biometric measurements, specifically germination percentage, vigour index, shoot and root lengths were documented on seventh day.[16] Phytotoxicity index (PI) greater or less than 1, reveals the toxic or stimulatory character of the dye solution exposed to *H.gracilis* against the seedlings of *V. radiata*.[17]

Germination percentage=(seeds sprouted (number))/ (seeds sown) ×100   ...(2)

Vigour Index =Germination percentage X Seedling length   ...(3)

Phytotoxicity Index=1- (Seeds root length exposed to treated dye solution)/(Seeds root length exposed to tap water)   ...(4)

Antimicrobial Activity

Corafix Yellow GD3R decolorized with *H.gracilis* and untreated dye solution were tested against the bacterial (*Staphylococcus aureus* and *Escherichia coli*) and fungal (*Aspergillus niger* and *Rhizopus stolonifer*) cultures through agar well diffusion method. On Muller Hinton(bacteria) and Rose Bengal Chloramphenicol (fungi) agar plate's two wells were bored aseptically and added the test solutions which were kept at 37°C for 24hrs and 28oC for three days respectively. The inhibitory zone circumference (mm) served as index for the pollutant toxicity. The data were statistically determined using mean±SD.[18]

Results and Discussion
Distinctive Characters of *Halimeda gracilis*

The green macroalgae Halimeda gracilis, belongs to the phylum Chlorophyta and family Halimedaceae (Fig.1). *H. gracilis* is a surface water species which can flourish by absorbing sunlight and also found

in the subtidal zone of southeast coast of Tamil Nadu. The fresh segments *H. gracilis* was greenish, flattened, subcylindrical, ribbed, extensively calcified, trilobed, infrequently cuneate and the dry segments were pale green to white. They possess a single holdfast to secure them to the substrate and have dichotomous/trichotomous branching.[19]

Fig. 1: Dried seaweed of *H.gracilis*

Assessment of Zero Point of Charge (pHzpc) of *H. gracilis*

It is an crucial aspect in understanding the surface chemistry of the biosorbent.[20,21] The pHzpc data denotes the pH level at which the biosorbent net charge is zero. The pHzpc of *H.gracilis* was empirically determined arises at 5.4, as shown in Fig.2. This outcome implies that pH below 7 (acidic) is advantageous for anionic dyes. The research examining how pH influences dye decolorization explored further in the adsorption studies.
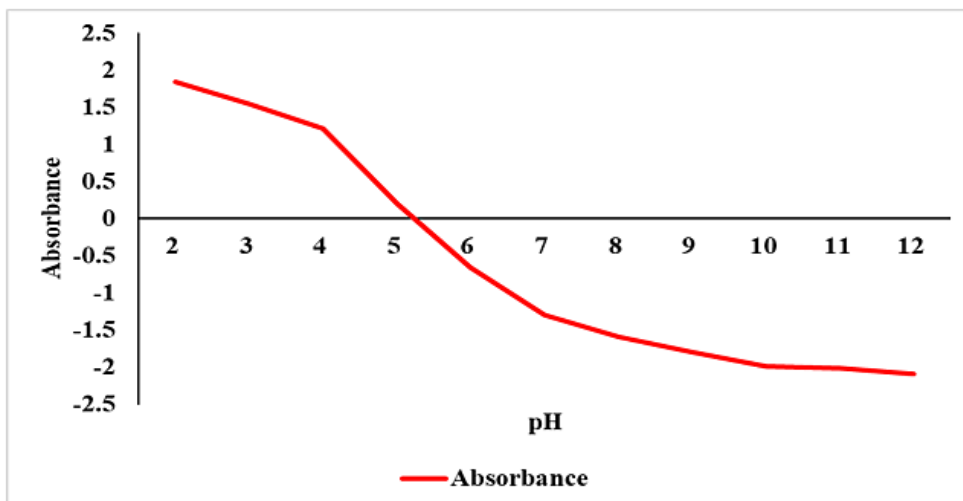

Fig. 2: pHzpc of *H.gracilis*

Batch Adsorption Studies for Corafix Yellow GD3R decolorization

The process of dye decolorization was assessed through batch mode using diverse adsorption parameters such as concentration of dye and biosorbent, incubation period, pH and temperature. A crucial aspect that influences the adsorption phenomena is the dye concentration which was differ from 100 to 500mg/L by keeping all other factors constant(Fig 3a-3e). The consequence of dye concentration on the removal of CYGD3R using *H. gracilis* was intense at 300mg/L (88%) and further rise in dye concentration reveals no substantial effect on decolorization phenomenon (60%). The adsorption sites in the biosorbent are easily occupied by the dye molecules and thereby a significant impact on adsorption capacity was recorded at varying dye concentrations. The concentration of dye offers a stronger dynamic energy, to conquer the diffusive mass transfer barrier involving the biosorbent and adsorbate. Thus at low concentrations, the dyes may interact with the binding sites to promote high removal, whereas rise in dye concentration has no effect on the adsorption phenomena after equilibrium is established because the external surface of the biosorbent gets saturated by the dye molecules. *Fucus vesiculosus* effectively removed methylene blue and rhodamine B under optimized conditions which supported the findings of the present study.[22]

The surface of the biosorbent provides binding sites for the dyes to adsorb from aqueous solution.[23] From Fig.3b, it is evident that the utmost color removal (84%) was observed in dye solution amended with 200 mg/L of biosorbent and its further increase (300-500 mg/L) did not cause any progression in biosorption. The availability of active sites in the biosorbent at low concentration allows the optimum level of dye to adsorb on its surface and thereby increases the dye removal rate.[24] Whereas, the rate of dye sorption gets declined when the concentration of biosorbent is raised, since they form aggregates at the adsorption site. Thus, the amount of available adsorption sites and the exchange of dye molecules for biosorption depend upon the concentration of biosorbent used in the adsorption process.[25] According to the results of the present investigation, *Ulva lactuca* was found to remove methylene blue and malachite green effectively at lower biosorbent concentrations.[26]

The entire adsorption process is substantially determined by the dye solution's pH, since it is vital for the protonation of the dye binding site.[27] The removal of CYGD3R using *H. gracilis* was studied at varying pH (2-11) and it evident from Fig.3c that maximum removal of CYGD3R was established at pH 6 (88%) and a further raise in the pH (7-11) declined the rate of dye absorption. With regard to zero point of charge at a low pH, the surface of *H.gracilis* was protonated (positive charge), which would leads to a powerful electrostatic attraction between the biosorbent (positive surface charge) and CYGD3R (anion) molecules which lead to increase the dye removal. As the pH increases (basic conditions), the surface charge of *H.gracilis* became negative which might have created electrostatic revulsion amidst the biosorbent and anionic CYGD3R dye molecules which assisted in decreased dye removal.[28]

Contact time is one of the vital factor for using a biosorbent successfully for practical applications and for quick adsorption.[29] To evaluate *H. gracilis* effectiveness at removing dye, the biosorption experiment was conducted over a range of time periods (24–144 hours). It was noticed that the efficacy of dye removal remained high at the early stage and maximum adsorption was found at 72hrs which subsequently falls with no discernible color removal after 72hrs (Fig.3d). Rapid adsorption at the primary stage may be due to the abundance of high number of binding positions for biosorption, but as time proceeds, the remaining binding positions become harder to occupy and takes long time to reach equilibrium because of the repulsion between the biosorbent and adsorbate.[30] At the initial stage of adsorption physical or ion exchange process may occur at the cell surface and increase the rate of dye removal whereas complexation, precipitation or saturation of binding sites might occur after a lapse of time and thereby results in decreased dye removal.[31]

The most crucial kinetic variable in the biosorption process is the temperature which has an impact on the adsorption capability of the biosorbent and the mobility of the adsorbate.[32] An increase in the temperature, increases the dye removal rate and maximum removal was recorded at 25°C (84%) using H.gracilis. Further increase in temperature (30°C -40°C) decreases the dye

removal efficiency (Fig.3e), which might be initiated by the modification in the texture of the biosorbent or material deterioration.[33] *Ulva lactuca* removed methylene blue to the greatest extent at the ideal temperature of 25°C which supported the results of the present investigation.[34] According to the temperature profile, the sorption capacity rises with temperature until it reaches a maximum and then falls. The decline in dye removal after rise in the optimal temperature may be related to denaturation, which occurs when a biosorbent is exposed to extremely high temperature which damages the active binding sites or the tendency to desorb dyes from the interface of the solution.[35] Thus at optimal conditions (Dye concentration-300mg/L, pH 6, Biosorbent concentration - 200mg/L, Temperature - 25° C,Incubation time - 72 hrs), 88% of CYGD3R has been removed by *H.gracilis.*
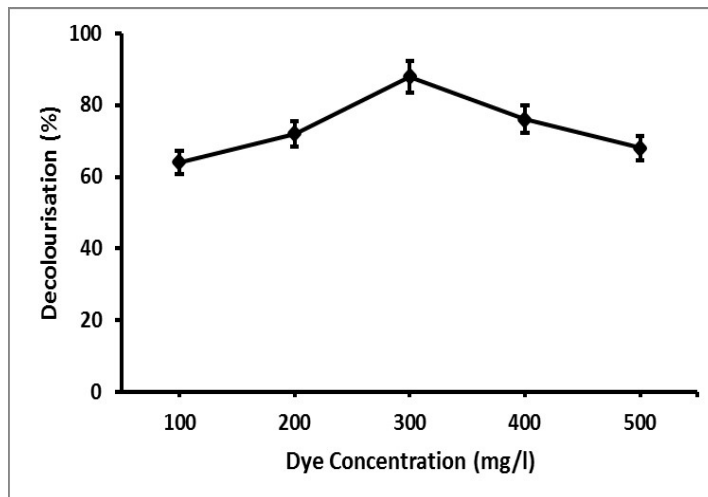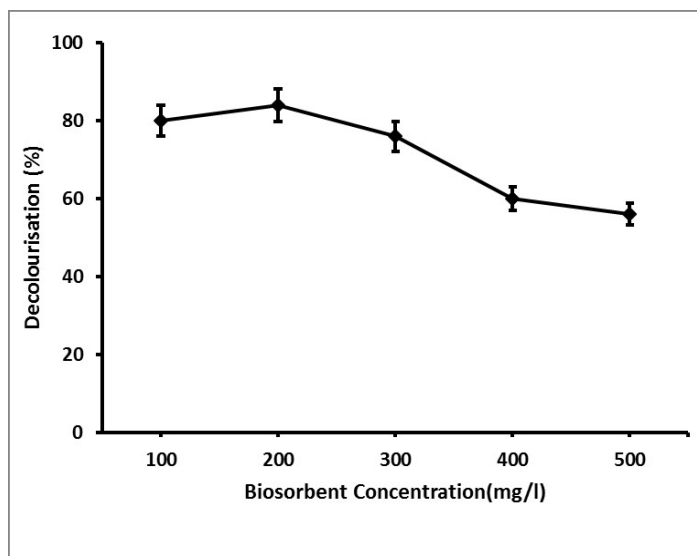

Fig. 3a: Dye concentration
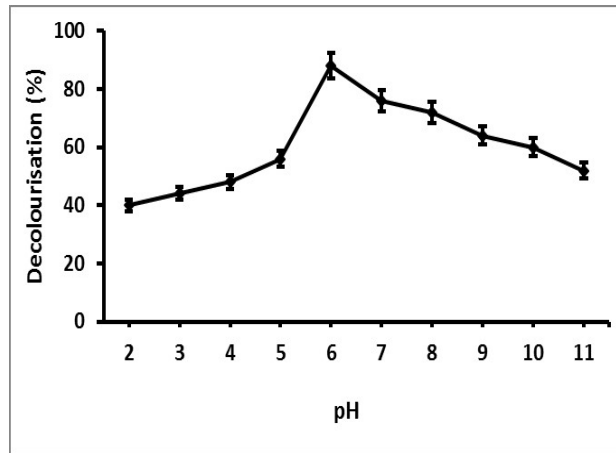

Fig. 3b: Biosorbent Concentration
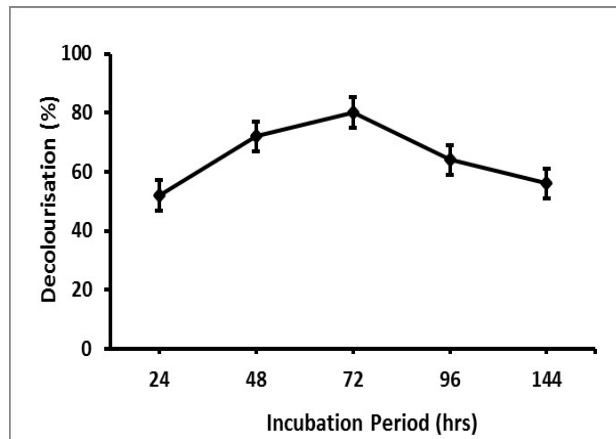
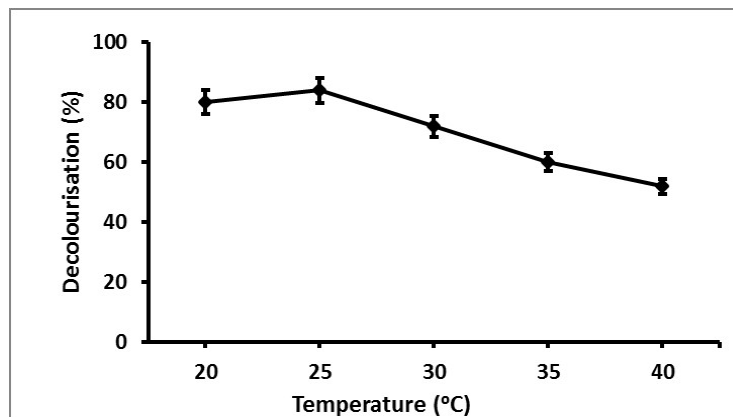**Fig. 3c: pH**

Fig. 3d: Incubation Period

Fig. 3e: Temperature

Fig. 3a-3e: Decolourization of CYGD3R by *H. gracilis* under optimized parameters

Adsorption Kinetics and Thermodynamic Studies

Adsorption kinetic models namely the PFO, PSO and ID remained assessed to demonstrate of possible physiochemical interactions involved between the biosorbent and dye molecule during adsorption process. The kinetic model's linear equations were presented below,

$$\log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \qquad ...(5)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_{e^2}} \qquad ...(6)$$

$$q = K_p\ t^{1/2} + C \qquad ..(7)$$

where the adsorption level constants are k_1 and k_2 (mgming^{-1}), q_e and q_t (mgg^{-1}) represent the quantity of dye biosorbed at the equilibrium at given time (t), q indicates sum of dye adsorbed (mgg^{-1}), $t^{1/2}$ is the square root of time t (min), K_p stands for intraparticle diffusion rate constant and the interceptis denoted as C. The datas of k_1, k_2 and qe are equated from log $(q_e - q_t)$ vs t and t/qt vs t polts separately and their findings were depicted in Table 1 and picturized in Fig.4a and 4b.The R^2 value (0.011) of PFO was less than the PSO (R2> 0.95) kinetic models which proves the later to be more suitable. This further implies that the process of adsorption occurs through chemisorption concerning the equilibrium armed forces through the exchange of electrons among the biosorbent and adsorbate.[36] The adsorption results of the present study obey pseudo-second kinetics model which agrees with the reports on the removal of yellow dye using *Padina pavonica*.[37] Fig.4c depicts that the linear form do not passes through the origin and proves the ID is not the adsorption rate controlling stage for the CYGD3R adsorption on to *H.gracilis*.

The thermodynamics factors such as free enthalpy (ΔG°), total heat (ΔH°) and entropy (ΔS°) were evaluated to study the spontaneity and the adsorption of CYGD3R on to *H.gracilis* using the following equations

$$\Delta G = \Delta H° - T\Delta S° \qquad ...(9)$$

$$\Delta G = -RT \ln K_D \qquad ...(10)$$

$$\ln K_D = \frac{\Delta S°}{R} - \frac{\Delta H°}{RT} \qquad ...(11)$$

where, K_D (thermodynamic equilibrium constan) t, R (universal gas constant -8.314 J mol^{-1} K^{-1}) and T (temperature). The ΔG° and ΔH° datas existed attained from the slope and intercept of Van't Hoff plot of experimental data on lnKDvs 1/T (Fig. 4d) and their results were depicted in Table 1. The negative datas of ΔG° show that the biosorption of CYGD3R onto *H. gracilis*is a spontaneous and feasible manner and the positive datas of ΔH° and ΔS° represents the endothermal in nature and rising randomness at adsorbate-biosorbent interface during the biosorption procedure. The rise of randomness on solid-liquid interface in the sorption system throughout the adsorption method is shown by the positive value of variation in entropy (ΔS°). Similar such endothermic, feasible and spontaneity was noticed in Direct Red 81 removal by *Argemone mexicana*.[38]
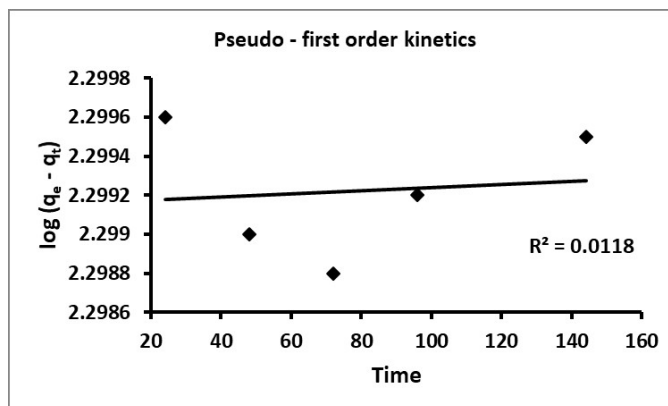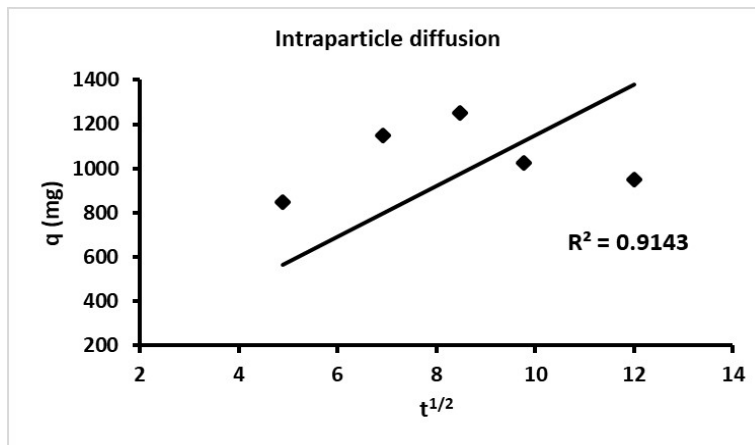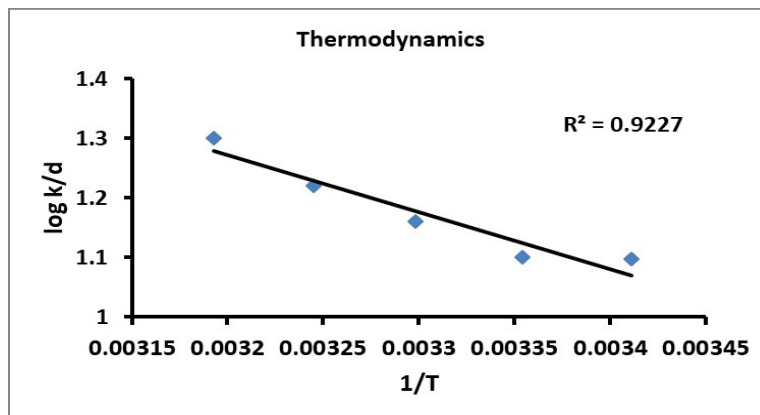

(4a)
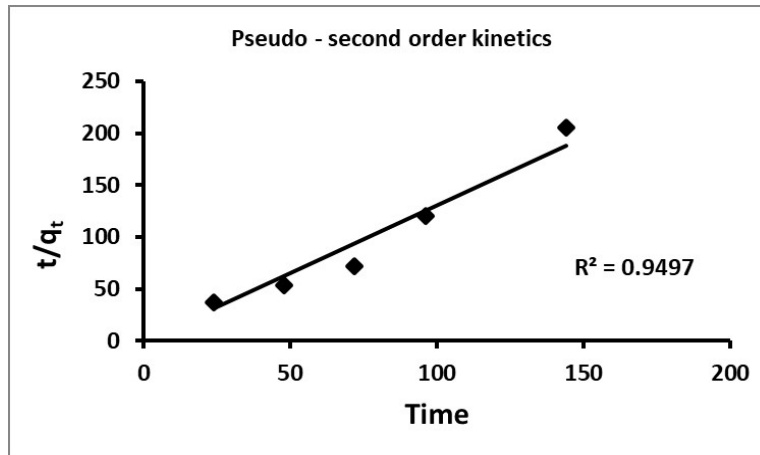
(4b)

(c)

(d)

Fig 4:PFO (4a), PSO (4b) kinetic models, ID (4c) and Van't Hoff plot (4d) for the biosorption of Corafix Yellow GD3R onto *Halimeda gracilis*.

**Table 1: Adsorption kinetics and thermodynamic parameters**

Adsorption kinetics parameters								
PFOkinetics*			PSO kinetics*			ID**		
q_e	K_1	R^2	q_e	K_2	R^2	K_p	C	R^2
2.29	7.88	0.01	-12.94	1.43	0.94	6.028	994.27	0.91

Thermodynamics					
Temperature (oC)	$\Delta G \circ$***		$\Delta H \circ$***	$\Delta S \circ$	R^2
20	-182				
25	-224		4.34	9.59	0.92
30	-285				
35	-350				
40	-432				

*(mg min g^{-1}), ** (mg g^{-1} min$^{-1/2}$), ***(kJ mol^{-1}), ****(J mol^{-1}K^{-1})

Corafix Yellow GD3R Desorption Using Different Eluents

The biosorbent needs to be recycled to make the biosorption process a profitable one. Desorption, a highly efficient chemical procedure was utilized for recycling the dye-loaded biosorbent, since it does not damage the biosorbent structure.[39] Under optimal conditions, three successive desorption cycles were employed using the selected eluents. Among the eluents employed, 0.1N HNO$_3$ seems to desorb 60% of the dye from the biosorbent, followed by others in the first cycle and thereafter a progressive decrease was detected in the second and third cycles (Fig.5). For industrial applications, the recovery of dye from the chosen biosorbent is crucial, since it must not only have great biosorptive potential, but also be easy to use economically, simple to regenerate and reuse. Therefore, the eluent, kind of biosorbent, and biosorption mechanism should be the fundamental requirement for a proper desorption process.[40] Hence the choice of desorbing solvent or eluent is crucial for optimal regeneration and is greatly dependent on the kind of biosorbent and the biosorption mechanism.[41]
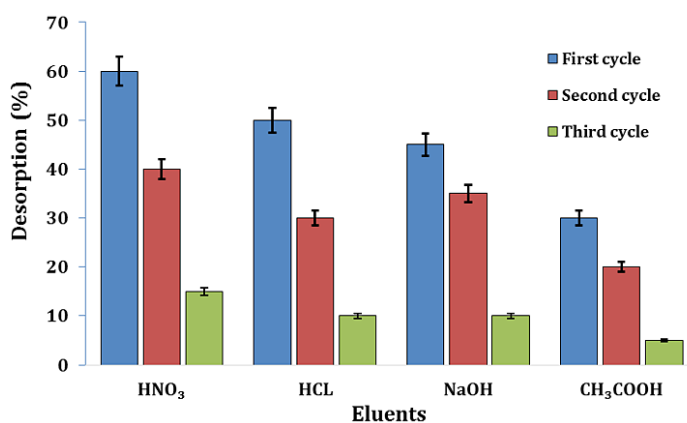
Fig. 5: Desorption of CYGD3R using different eluents

UV-Visible Spectral Analysis

The UV-visible spectrum of the CYGD3R concealed a peak at 580 nm and it was completely disappeared in the treated dye solution. The decrease in absorbance peak in the spectrum provides sturdy evidence for the decolorization of the dye (Fig.6). The diminished or disappearance of the peak may be due to the enzymatic biodegradation of dyes or by the breakdown of chromophores.[42] Similar such decrease/ disappearance in the UV-visible absorption peak was observed when methylene blue was exposed to *Arthrospira platensis* which may be either due to adsorption or biodegradation.[43]
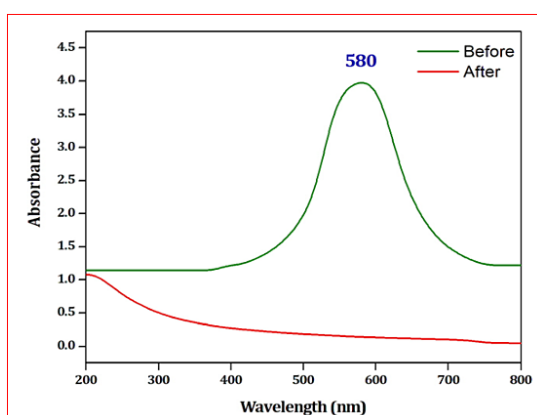

Fig. 6: UV-visible spectra of test samples

Phytotoxicity Study

When untreated textile dyeing effluent was released into the terrestrial ecosystem or utilized for agriculture, they cause harmful effects to biota. Subsequently, measuring their toxicity before and after decolorization was fraught with concern. *Vigna radiata* (Green gram) was used as the candidate plant to evaluate the toxicity of CYGD3R treated with *H.gracilis* and was compared untreated with tap water (control). The results of the biometrics revealed that the dye-treated solution enhanced the percentage of germination (93%), shoot (5.85 cm) and root (2.59 cm) length and vigour index (785) when considered with untreated dye solution (67%, 4.33cm, 1.57cm and 395) whereas the seedlings grown with the tap water (100%, 6.36cm, 3.1cm and 946) showed a positive impact. The phytotoxicity index was documented as 0.16, which is less than 1 thereby demonstrating the stimulatory effect of the treated of dye solution and thereby proves its nontoxic nature. Similar such decreased in seedling growth was observed in *Triticum* sp. seeds watered with untreated red and blue dye solution where the dyes may interact with the root and quickly penetrate into the embryonic axis membrane.[44] Also, the reduced shoot and root length of *Lemna minor* seedlings exposed to untreated solution cause harmful effects such as reduced enzyme activation, water imbalance, nutritional deficiencies, hormonal changes that affect membrane permeability and plant growth.[45]

Microbial Toxicity Studies

The results revealed that *S. aureus* and *E.coli* exhibited 6 mm and 8 mm zone of inhibition and *Aspergillus niger* and *Rhizopus stolonifer* recorded in 9mm and 11mm zone of inhibition against untreated dye solutions. In treated dye solution, there was no discernible growth inhibitory zone, indicating that the degraded dye was not harmful to the examined bacterial and fungal isolates.

Conclusion

The present study investigates the potentiality of *Halimeda gracilis* in Corafix Yellow GD3R removal under *in vitro* conditions. Corafix Yellow GD3R dye adsorption was found to be strongly reliant upon the concentration dye and biosorbent, pH, incubation time and temperature. Under optimized conditions, *H.gracilis* removed 88% of Corafix Yellow GD3R dye. The adsorption monitored pseudo second order kinetics and thermodynamic factors exhibited the adsorption of dye on top of *H. gracilis* remained spontaneous, feasible and endothermic in nature. Thus from the obtained outcomes, it can be accomplished that for the textile wastewater comprising variety of azo reactive dyes, *H. gracilis* can be used as a potent biosorbent under optimized conditions due to its great biosorption efficiency, reusability and desire to serve as the foundation for the creation of ecofriendly pollution-free environment.

Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

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1. Lebron Y.R, Moreira V.R, Santos L.V. Studies on dye biosorption enhancement by chemically modified *Fucus vesiculosus*, *Spirulina maxima* and Chlorella pyrenoidosa Algae. J. Clean. Prod.2019;240: 118197.

2. Jadhav J.P, Phugare S.S, Dhanve R.S, Jadhav S.B. Rapid biodegradation and decolorization of Direct Orange 39 (Orange TGLL) by an isolated bacterium *Pseudomonas aeruginosa* strain BCH. Biodegradation. 2010;21(3):453-63.

3. Kroumov D. Determination of the mass transfer-limiting step of dye adsorption onto commercial adsorbent by using mathematical models. *Environ. Technol.* 2014; 35: 2356-2364.

4. Robinson T, Mullar G, Marachant R. Remediation of dyes in textile dyeing industry effluents a critical review on current treatment technologies with a proposed alternative. *Environ. Technol.* 2001; 77: 247-255.

5. Salleh M.A, Mahmoud D.K, Abu N.A, Karim W.A, Idris A. Methylene blue adsorption from aqueous solution by langsat *(Lansium domesticum)* peel. *J Purity, Utility Reac and Envi.* 2012; 1: 472-495.

6. Wilkinson S. M, Mc Gechaen K. Occupational allergic contact dermatitis from reactive dyes. *Contact Derm.* 1996; 35: 376.

7. Senturk T, Camlı C, Yildiz S. Comparing the phytoremediation efficiency of three different algae for the nutrient removal of Gediz River in Manisa/Turkey. *CBU Journal of Science.* 2017; 13(3):737-743.

8. Gonçalves B.B, Giaquinto P.G, Silva D.S, Meloe C, Neto S, Lima A.S, BritoDarosci A. A, Portinho J.L, Carvalho W.F, Rocha T.L. Ecotoxicology of glyphosate-Based Herbicides on Aquatic Environment. *J. Biochem. Technol.* 2019; 55-71.

9. Wang J.L, Chen C. Biosorbents for heavy metals removal and their future- A review. *Biotechnology Advances.* 2009; 27: 195–226.

10. Nilson T, Peterson U. Successional reflectance trajectories in northern temperate forests. *Int. J. Remote Sens.*1993; 14(3):609-613.

11. Ozer A, Ozer D. Comparative study of the biosorption of Pb(II), Ni(II) and Cr(VI) ions onto Saccharomyces cerevisiae: determination of biosorption heats. *J. Hazard. Mater.* 2003; 100: 219-229.

12. Varma R, Turner A, Brown M.T. Bioaccumulation of metals by focus ceranoides in estuaries of South West England. *Mar. Pollut. Bull.* 2011; 62 : 2557–2562.

13. Hashem, A., Aniagor, C. O., Morsy, O. M., Abou-Okeil, A and Aly, A. A. Apricot seed shell: An agro-waste biosorbent for acid blue193 dye adsorption. Biomass Conversion and Biorefinery. 2022.

14. El-Sheekh M.M, Gharieb M.M, Abou-El-Souod G.W. Biodegradation of dyes by some green algae and Cyanobacteria. *Int. Biodeterior. Biodegradation.* 2009; 63: 699-704.

15. Phugare S.S, Kalyani D.C, Patil A.V, Jadhav J.P. Textile dye degradation by bacterial consortium and subsequent toxicological analysis of dye and dye metabolites using cytotoxicity, genotoxicity and oxidative stress studies. *J. Hazard. Mater.*2011: 186, 713-723.

16. Akar S.T, Akar T, Cabuk A. Decolourisation of a textile dye, reactive red 198 (RR198) by *Aspergillus parasiticus* fungal biosorbent. *Braz. J. Chem.* Eng.2009; 26: 399-405.

17. Al-Tohamy R, Sun J, Fareed M. F, Kenawy E. R, Ali S. S. 2020. Ecofriendly biodegradation of Reactive Black 5 by newly isolated *Sterigmato myceshalophilus* SSA1575, valued for textile azo dye wastewater processing and detoxification. *Scientific reports.* 2020; 10: 1-16.

18. Mekki A, Dhouib H, Sayadi S. Polyphenols dynamics and phytotoxicity in a soil amended by olive mill wastewaters. *J. Environ. Manage.* 2007; 84:134- 140.

19. Saratale R.G, Saratale G.D, Chang J.S, Govindwar S.P. Decolorization and bio-

degradation of reactive dyes and dye waste-water by a developed bacterial consortium. *Biodegradation.* 2010: 21: 999 1015.

20. Yadav M, Thakore S, Jadeja R. Removal of organic dyes using *Fucus vesiculosus* seaweed bioadsorbent an ecofriendly approach: Equilibrium, kinetics and thermo-dynamic studies. Environmental Chemistry and Ecotoxicology. 2022; 4, 67–77.

21. Berber-Villamar N. K, Netzahuatl-Muñoz A. R, Morales-Barrera L, Chávez-Camarillo G. M, Flores-Ortiz C. M, Cristiani-Urbina E. Corncob as an effective, eco-friendly, and economic biosorbent for removing the azo dye Direct Yellow 27 from aqueous solutions. PLoS ONE. 2018: 13, e0196428.

22. Xing X, Jollanda M.H, Huisman J. Competition between cyanobacteria and green algae at low versus elevated CO2: who will win, and why?.*Journal of Experimental botany*.2017: 68(14): 3815–3828.

23. Yadav M, Thakore S, Jadeja R. Removal of organic dyes using *Fucus vesiculosus* seaweed bioadsorbent an ecofriendly approach: Equilibrium, kinetics and thermodynamic studies. *J. Environ. Chem. Ecotoxicol.*2022; 4:67-77.

24. Yagub M.T, Sen T.K, Afroze S, Ang H.M. Dye and its removal from aqueous solution by adsorption: a review. *Adv. Colloid Interface Sci*. 2014 209: 172–184.

25. Azmat R. Marine green algae *Codiumyengarii* as a good biosorbent for elimination of reactive black 5 from aqueous solution. Pak. *J. Pharm Sci*. 2014; 5, 1443-1449.

26. Dharajiya D, Shah M, Bajpai B. Decolorization of Simulated Textile Effluent by *Phanerochaete chrysosporium* and *Aspergillus fumigatus* A23. Nat. Environ. Pollut. Technol.2016; 15(3).

27. Deokar R, Sabale M. Biosorption of Methylene Blue and Malachite Green from Binary Solution onto *Ulva lactuca*. *Int. J. Curr. Microbiol. Appl. Sci.*2014: 295-304.

28. Das N, Charumathi D. Remediation of synthetic dyes from wastewater using yeast-an overview. *Indian J. Biotechnol*. 2012; 11: 369-380.

29. Ramachandran P, Sundharam R, Palaniyappan J, Pudukkadu A. Potential process implicated in bioremediation of textile effluents: A review. *Adv. Appl. Sci. Res*.2013; 4(1):131-145.

30. Loong Y, Lewis W. L. 2013. The disappearing Environmental Kuznets Curve: A study of water quality in the Lower Mekong Basin (LMB). *J. Environ. Manage.*2013; 131:415-425.

31. Idris M.N, Ahmad Z. A, Ahmad M.A. Adsorption equilibrium of malachite green dye onto rubber seed coat based activated carbon. *Fudma J. Sci*. 2011; 3: 67-76.

32. Chiou M. S, Ho P. Y, Li H. Y. Adsorption of anionic dyes in acid solutions using chemically cross-linked chitosan beads. *Dyes and pigm*.2004; 60(1), 69-84.

33. Arulselvi I.P, Barathi S. Decolourization, degradation, and toxicological analysis of textile dye effluent by using novel techniques – Review. *Int. J. Sci. Res.*2015; 3:2118- 2136.

34. Nacèra Y, Aicha B. Equilibrium and kinetic modelling of methylene blue biosorption by pretreated dead *Streptomyces rimosus*: Effect of temperature. *Chem. Eng*. J.2006; 119(2-3):121-125.

35. Abdelwahab O, El Sikaily A, Khaled A, Nemr A. Removal of Methylene Blue from aqueous solution by marine green alga *Ulva lactuca*. *J.Chem. Ecol*. 2006; 22:2, 149-157.

36. Alqaragully M.B (2014) Removal of textile dyes (maxilon blue and methyl orange) by date stones activated carbon. *Int. J. Adv. Res. Chem. Sci*. 2014; 1(1): 48-59.

37. Hameed B.H. Spent tea leaves: A new non-conventional and low-cost adsorbent for removal of basic dye from aqueous solutions. *J. Hazard. Mater.*2009; 161: 753-759.

38. Fakhry E.M. Padinapavonica for the removal of dye from polluted water. AmJ Plant Sci.2013; 4, 1983-1989.

39. Khamparia S, Jaspal D. Adsorptive removal of direct red 81 dye from aqueous solution onto Argemonemexicana. *Sustainable Environment Research*. 2016;26(3): 117-123.

40. Kwak H.W, Kim M.K, Lee J.Y, Yun H, Kim M.H, Park Y.H. Preparation of bead type biosorbent from water soluble *Spirulina platensis* extracts for chromium (VI) removal, Algal Res. 2015; 7:92-99.

41. Chen S, Zhang J, Zhang C, Yue Q, Li Y, Li

E. Equilibrium and kinetic studies of methyl orange and methyl violet ad sorption on activated carbon derived from *Phragmites australis. Desalination.*2010; 252, 149-156.

42. Das C, Gupta S. De S. Treatment of dyeing effluent from tannery using membrane separation process, *Int J Environ Waste manag.*2010; 5: 354-367.

43. Bagchi M, Ray L. (2015) Adsorption behavior of reactive blue 4, a tri-azine dye on dry cells of Rhizopusoryzae in a batch system.

Chem. Speciation and Bioavailability. 2015; 27(3):112-120.

44. Mansour AT, Alprol AE, Abualnaja KM, El-Beltagi HS, Ramadan KM, Ashour M. The Using of Nanoparticles of Microalgae in Remediation of Toxic Dye from Industrial Wastewater: Kinetic and Isotherm Studies. *J Mater.* 2022; 31:15(11):3922.

45. Brahmbhatt NH, Jasrai RT. The role of algae in bioremediation of textile effluent. *Methods.* 2016; 21:28.