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Removal of methylene blue from aqueous solutions with hybrid bioprocess: biosorption on modified cork powder and soluble turnip peroxidase

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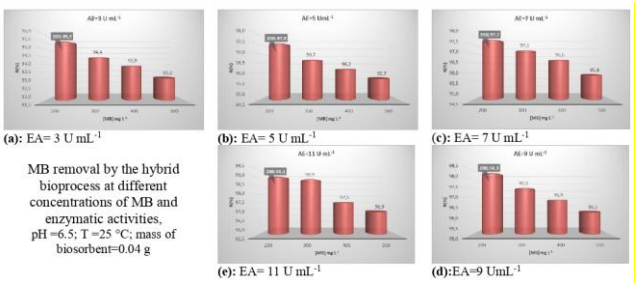
Graphical abstract



MB removal by the hybrid bioprocess at different concentrations of MB and enzymatic activities, pH =6.5; T =25 °C; mass of biosorbent=0.04 g

Abstract

Methylene blue (MB) removal from aqueous media using hybrid bioprocess: biosorption on modified cork powder (SDS-cork) and soluble turnip peroxidase (STP) was investigated. Cork powder was modified with sodium lauryl sulphate (SDS). Chemical composition of the biosorbent were characterized by infrared before and after biosorption. The hybridization was carried out under the optimal conditions of the two processes: pH=7, T=40 °C, $[H_2O_2]$=10 mM for enzyme activity(EA)=7 U mL^{-1} witch it was in excess for the EA= 5 and 3 U mL^{-1}, $[H_2O_2]$ = 15 mM for EA= 9 U mL^{-1} and $[H_2O_2]$=15 mM for EA= 11 U mL^{-1} with a treatment time that equals the sum of two optimal treatment times, and a mass of SDS-cork 0.04 g with a diameter <0.16 mm. The feasibility and performance of this hybridization was tested according to two parameters the initial concentration of MB which varied between 200 and 500 mg L^{-1} and the enzymes activities which has varied between 3 and 11 U mL^{-1}. The minimum removal percentage of MB was satisfactory according to 93% for [MB] = 500 mg/L and an EA of 3 U mL^{-1}, otherwise the maximum removal percentage was very satisfactory being in the order of 99.9% for a [MB] = 200 mg L^{-1} and EA= 11 U mL^{-1}.

Keywords: cationic methylene blue dye, wastewater treatment, modified cork powder, soluble turnip peroxidase, coupled bioprocess.

1. Introduction

The waters loaded with dyes rejected by industries have been widely studied. MB is a model dye most commonly used in cotton, wood and silk dyeing (Santoso *et al.,* 2020). It is known for being a toxic and persistent substance in the environment. This dye can cause eye burns that cause permanent injury to human and animal eyes (Santoso *et al.,* 2020; Youcef *et al.,* 2019). Inhalation may cause breathing difficulties and ingestion through the mouth produces burning sensation, nausea, vomiting, sweating and heavy cold sweat (Santoso *et al.,* 2020; Contreras *et al.,* 2019).

So, it was necessary to eliminate this pollutant by specific processes such as physical processes (adsorption, irradiation, ozonation, ... etc.) (Kong *et al.,* 2020; Li *et al.,*2019), Chemical processes (flocculation, precipitation, ion exchange, ... etc.) (Mazivila *et al.,* 2019), the membrane separation (Mazivila *et al.,* 2019; Parakala *et al.,* 2020), enzymatic (Morsi *et al.,* 2020) and biological processes (Contreras *et al.,* 2020; Katheresan *et al.,* 2020).

However, these processes still not eco-friendly or nature-friendly (e.g., membrane separation), leading to the generation of large quantities of sludge or involving poor regeneration, which makes the operation as a whole very costly (e.g., adsorption) (Zhou *et al.,* 2020). On the other hand, they are not profitable in terms of removal percentage for the high concentrations of this pollutant (e.g., enzymatic processes) (Bilal *et al.,* 2020) or the process was difficult and sensible for any error (e.g., biological process) (Contreras *et al.,* 2020).

Therefore, the idea is to study the performance and different feasibility of new hybrid bioprocess: biosorption on modified cork powder and soluble turnip peroxidase. This process is eco-friendly and nature-friendly technique. As well as, this process is fast, easy, more efficient and regenerable. The role of SDS is the fixation of MB by a chemical bond and it leads to increase the amount

adsorbed in MB (Azizi *et al.,* 2021; Que *et al.,* 2018; Irani and Jadid*,* 2015; Sahu *et al.,* 2015).

2. Materials and methods

2.1. Chemical products

4-aminoantipyrine ($C_{11}H_{13}N_3O$, 99%) purchased from Fluka Chemika; Acetone (C_3H_6O, 99.5%) purchased by prolabo chemicals; Phenol (C_6H_6O, 99%) imported by Prolabo-Chemicals; Hydrogen potassium monopotassium phosphate (136.06, 99%) imported by Biochem-Chemopharma; Hydrogen peroxide (H_2O_2, 10%) imported by SNC Boufama and Associates Z.A.C Mila 43000; Potassium hydroxide (KOH, 85%) imported by Sigma-Aldrich; Methylene blue ($C_{16}H_{18}ClN_3S$, 85%) imported by Riedel de Haen. Cork imported from the Wilaya of Jijel (North East Algeria).

2.2. Materials

FTIR Spectroscopy Tensor II (Bruker). Ohaus centrifuges, model FC5706, Kika Laborate chnik brand agitator and Hotplate Stirrer brand agitator LabTech (DAIHAN LABTECH CO.LTD); Ohaus brand analytical balance; pH meter Inlolab type with magnetic stirring and equipped with a combined glass electrode; Rotavapor (R-200) brand Buchi; Optizen 2120 UV brand UV-visible spectrophotometer, computerized PC for the storage and processing of spectra; Memmert brand oven, Magnetic stirring bars of different sizes were obtained from Cole-Parmer Canada Inc. (Montreal, QC).

2.3. Methylene blue analysis

The MB analysis was carried out by UV-visible spectroscopy with a λ_{max} = 664 nm. As such, the removal percentage of this dye (R) by the SDS-cork or the STP is calculated by the equation 1:

$$R(\%) = \frac{A(0) - A(t)}{A(0)} \times 100 \quad (1)$$

$A(0)$ and $A(t)$ are the absorbance of the untreated sample and the absorbance of the sample after a treatment time respectively.

2.4. Infrared spectroscopy analysis of cork surface

The functional groups of the virgin cork, SDS-cork and SDS-cork after biosorption of MB were detected by FTIR spectroscopy.

2.5. Extraction and purification of STP

STP was extracted from white turnip with a percentage between turnip mass and volume of distilled water of 0.5: 1. The results of the extraction filter with the gas and maintain in a temperature 4 °C. This extract, which is called crude enzyme extract (CEE), is mixed dropwise with cold acetone with stirring and ice-wrapped with the volume ratio between CEE and acetone was 1:1. The mixture acetone-CEE subsequently placed in a temperature of 4 °C for one hour in order to facilitate pellet recovery after mixing under centrifugation at 6000 rpm for 10 min. Then this pellet dissolved in different volumes of monopotassium phosphate buffer solution (pH=6.5, 0.01 M), to have different enzymes activities of STP they are subsequently preserved in the refrigerator at a temperature of 4 °C.

2.6. Enzyme activity assay

The enzyme activity assay is following the 4-aminoantipyrine (Am-NH_2) method (Nicell *et* Wright 1997), as in a 50 mL vial put: 1 mL of phenol (0.1M), 1 mL of H_2O_2 (0.1M) and 1 mL of 4-aminoantipyrine (0.01M). The rest is supplemented with a pH= 6.5 buffer solution of 0.01 M monopotassium phosphate. A volume of 4 mL of this reaction mixture (50 mL) should be placed in a test tube and added to 0.2 mL of STP. This kinetic reaction is followed by UV-vis spectroscopy after the addition of 0.2 mL of STP at λ_{max} = 517 nm in order to plot the DO= f (time) pattern. The enzyme activity was calculated by the following equation:

$$EA(UmL^1) = \frac{\Delta DO}{\Delta t} \times \frac{V_r}{V_e} \times \frac{1000}{7425} \quad (2)$$

Where EA is the enzyme activity (U mL^{-1}); Vr is the reaction volume (mL) and Ve is the volume of STP (mL).

2.7. Chemical modification of cork

Dissolve 20 g of the cork in 100 mL of SDS. This mixture should then heat to 50 °C with stirring for 4 hours and then filter. After filtration, the support is washed several times (≤10 times) with water osmosis. SDS-cork should be stored in the desiccator at a temperature of 60 °C for 24 hours before use (Azizi *et al.,* 2021). Therefore, the pH_{PZC} of SDS-cork is 5.2 (Figure 1). The overall surface charge is positive for pH solutions below this value and is negative when pHs are above pH_{PZC}. As the MB is basic, its dissolution in water causes the release of colored ions of positive charge (cations). MB contains polar groups such as hydroxyls and carboxyls. In addition, the electric charge of the biosorbant depends on the pH of the medium because of the ionization of these surface functional groups. Note that the retention of MB on a biosorbant increases with the increase of the negative charge of the surface.

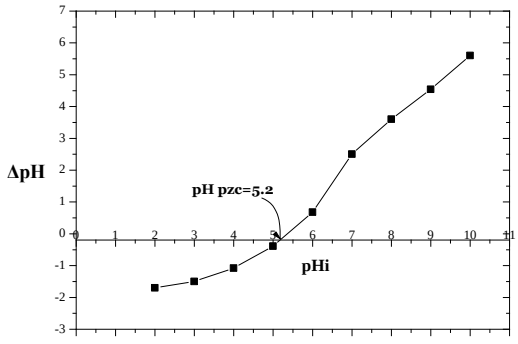


Figure 1. The zero-charge point (pH_{PZC}) plot for adsorption of MB onto SDS-cork.

2.8. Adsorption studies

The batch biosorption experiments of MB by the SDS-cork was carried out in a volume of 20 mL at stirring speed 250 rpm with predefined values for the MB concentration, pH,

temperature, treatment times and the biosorbant mass, in such a way to optimize these parameters values (Azizi *et al.,* 2021).

2.9. Oxidation studies with STP

The experiment consists of putting 1 mL of MB for a desired concentration in a reaction mixture comprising 0.2 mL of STP and 1 mL of hydrogen peroxide with 2.8 mL of well-established pH buffer solution.

2.10. Hybridization experiments of SDS-cork and STP

Hybridization experiments of two bioprocesses: biosorption onto SDS-cork and STP were carried out under optimal conditions of pH, temperature, concentration of hydrogen peroxide, enzyme activity, treatment times and mass of SDS-cork. The feasibility and performance of this coupled method was tested according to two parameters of the initial concentration of MB which varied between 200 and 500 mg L^{-1} and the enzyme activities that it has been varies between 3 and 11 U mL^{-1}.

3. Results and discussion

3.1. Cork surface analysis by infrared spectroscopy

The superimposed FTIR spectra for virgin cork, SDS-cork and SDS-cork after biosorption of MB show indifference changes in the absorption variation (Figure 2). since the adsorption of SDS on cork was carried out a physical adsorption and also the adsorption of MB on SDS-cork according to azizi et al., 2020 (Azizi *et al.,* 2021). However, we distinct peaks at 3417.98 cm^{-1} corresponding to (O–H) stretching vibration in aromatic rings, alcohols, phenols and carboxylic acids. Others peaks such as, 2924.18, 2862.48, 1627.97, 1450.52 and 1041.60 cm^{-1} presenting the organic function respectively(–CH_2), (-C-H), (C–C), (C=O stretching) and (C–O) (Liang *et al.,* 2010; Lopes *et al.,* 2001; Neto *et al.,* 1995; Prades *et al.,* 2010). Though, the FTIR spectrum of SDS-cork after biosorption of MB shows peaks at 3448.84 cm^{-1}, 2931.90 cm^{-1}, 1635.69 cm^{-1} according to (C=C stretching vibration in aromatic rings), and the peak 1049.31 cm^{-1} presenting the organic function (C–N) Liang (Liang *et al.,* 2010).

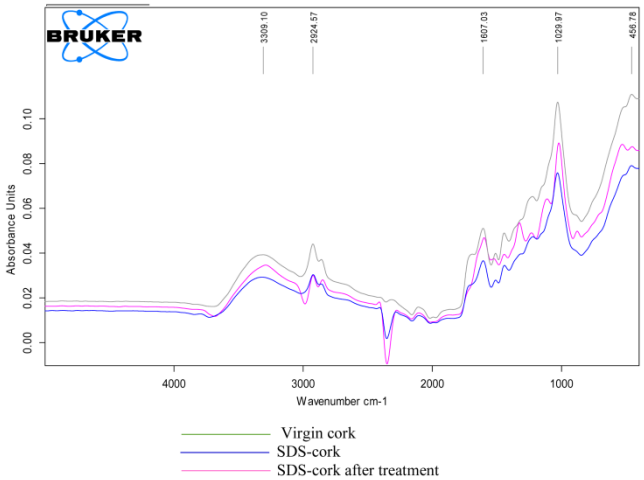


Figure 2. Cork surface analysis by infrared spectroscopy.

3.2. Elimination of MB by SDS-cork

3.2.1. Effect of pH

The study of initial pH effect on the biosorption process was done in the range of pH 2 to pH 8. The amounts of MB retained by SDS-cork from different solutions were found to be closely related to the initial pH value of the solution (Figure 3). MB biosorption is appreciable pH values between pH 4 and pH 7, with peak retention towards pH 6 corresponding to 90% of MB removing. The pH_{PZC} of SDS-cork is 5.2. So, in an acid medium the lower pH_{PZC} leads to greater adsorption capacity. A slight decrease in removal efficiency is observed with increasing pH up to pH= 9. The MB adsorption decreases from 90 to 82.2% for pH 6 to 8 respectively. Beyond pH 7, a decrease in efficiency has been observed. The same phenomenon is observed for values below pH 4.

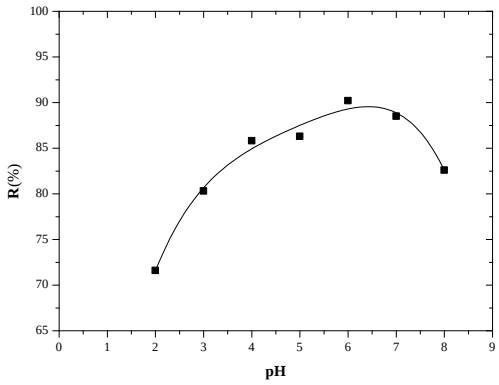


Figure 3. pH effect on MB removal by SDS-cork. Mass of SDS-cork = 0.02 g; Volume of solution=20 mL; [MB] =100 mg L^{-1},t = 120 min;T = 25 ° C.

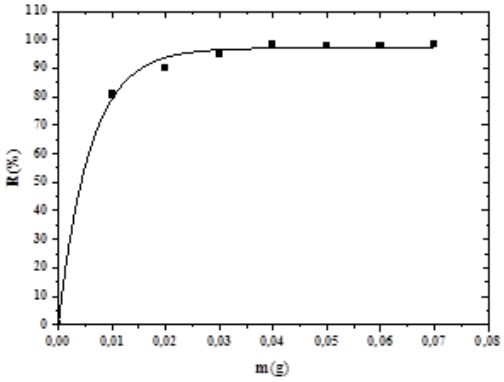


Figure 4. Mass of SDS-cork effect on MB removal by biosorption. pH=6; [MB] = 100 mg L^{-1}, t = 120 min; T = 25 °C.

3.2.2. Effect of Mass of SDS-cork

Figure 4 shows that a mass of 0.04 g of SDS-cork is capable of fixing a maximum of MB (98%). The amounts of MB stuff fixed must agree with the biosorbent doses in solution to ensure an equivalent number of adsorption sites. Beyond a certain mass, the retention rate decreases slightly, probably indicating the presence of another type of interaction between dye and SDS-cork. It may be a competition between the fibres retaining dye fractions and the free fibres of the adsorbent that attract it,

returning it to solution. It is therefore useful to work with adsorbent doses ≤ 0.04 g and to avoid an ineffective overdose. In the following work and to determine the adsorption capacity by saturating all the probable sites, we chose to work with adsorbent masses of 0.04 g.

3.2.3. Effect of contact time

Figure 5 demonstrates the temporary variation of MB removal efficiency as a function of the three initial concentrations of this dye (100, 200 and 300 mg L^{-1}). The initial rate for MB removal was inversely proportional to the initial concentration of this dye and therefore the maximum duration of treatment is properly proportional with the increase in the initial MB concentration. In such a way that the maximum R for the three concentrations 100, 200 and 300 mg L^{-1} were 81, 88 and 98% for 90, 120 and 180 minutes, respectively. The increase of loading capacity of SDS-cork with increasing initial MB concentration may be due to higher interactions between MB and SDS-cork.

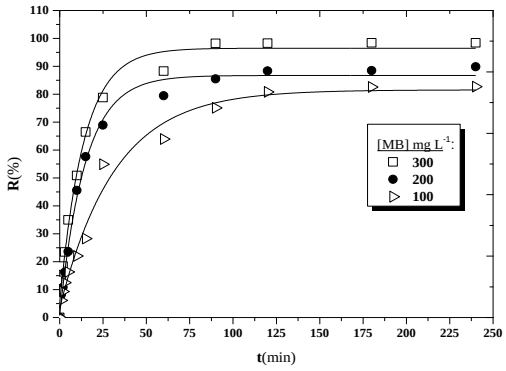


Figure 5. Effect of contact time on MB removal by biosorption onto SDS-cork. Mass of SDS-cork = 0.04 g; pH = 7; T = 25 °C.

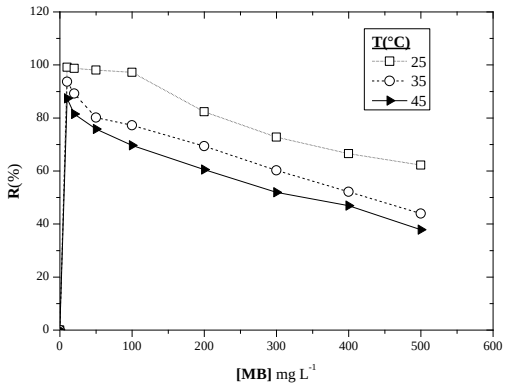


Figure 6. Effect of the temperature on MB removal by biosorption onto SDS-cork. Mass of SDS-cork = 0.04 g; pH = 7.

3.2.4. Temperature effect

Figure 6 represents the influence of temperature on the biosorption of MB onto SDS-cork. When we use different temperatures at 90 minutes of stirring, the adsorption capacity of the SDS-cork as a function of time increases between 25° and 35 °C and deprived for the temperature of 45 °C. The values obtained are R = 96% at T = 25 °C, R = 97% and at T = 35 °C and R = 93% at T = 45 °C. The experimental results obtained prove that this parameter positively affects this process by a high-energy contribution, thus making it possible to overcome the repulsive forces located at the interfaces of the liquid and solid media. Therefore, it is interesting to note that the contribution of heating plays an inhibitory role in the kinetics of retention of this dye for a temperature above 45 °C, regardless of their affinity for this support. This means that the retention process could be endothermic (ΔH> 0) and lead to physisorption under these conditions (Azizi *et al.,* 2021).

3.3. Adsorption Mechanism of MB on SDS-cork

At pH> PZC=5.2, such as the optimal pH is 6, the surface of the SDS-cork features negative charge and the functional groups like carboxyl and hydroxyl groups are free to interact with the cationic species MB^+ (Figure 7) (Que *et al.,* 2018).

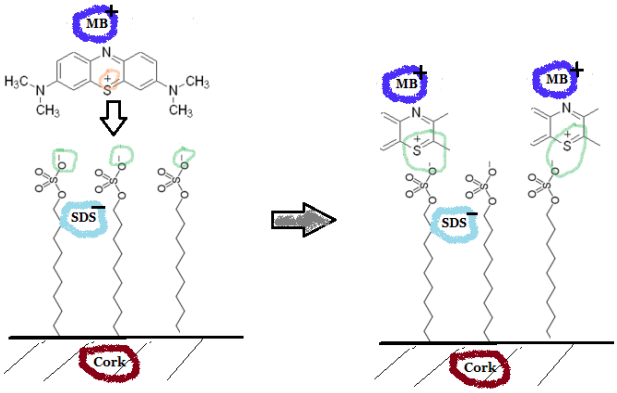


Figure 7. Adsorption mechanism of MB onto SDS-cork.

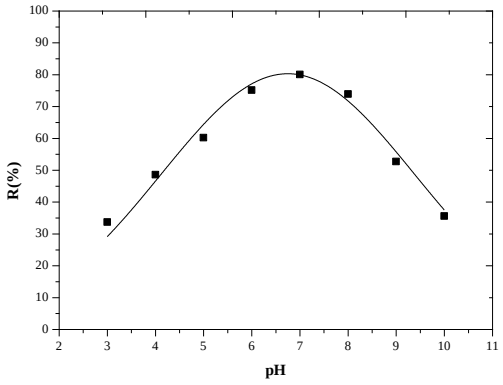


Figure 8. The pH effect on MB removal by STP. T= 25 °C; [MB]=100 mg L^{-1}; [H_2O_2]=1 mM ; EA= 7 U mL^{-1} ; t= 2 hours.

3.4. Oxidation of MB by STP

3.4.1. The effect of initial pH

The initial pH of the reaction medium has a direct impact on the removal efficiency of MB by STP. MB removal profile as a function of pH shown below was carried out in a volume of 5 mL. Therefore, the maximum MB removal according to 80% corresponding to the optimum pH = 7 (Figure 8).

3.4.2. The effect hydrogen peroxide

Batch experiences were carried out on the oxidation of a 100 mg L^{-1} of MB by STP to have the effect of the

concentration of hydrogen peroxide on the percentage removal of this dye for of the three-enzyme activities 7, 9 and 11 U mL^{-1}. The results of these tests are summarized in the Figure 9. Thereby, increasing the enzymes activities of 7 to 11 U mL^{-1} units was produced an increase in the consumption of hydrogen peroxide for the same concentration of MB. In addition, this increase in MB consumption was positively influenced on the percentage removal up to optimal values of hydrogen peroxide 10, 15, 20 mM for enzyme activities 7, 9 and 11 U mL^{-1} they percentage removal were 87.5, 95 and 99% respectively.

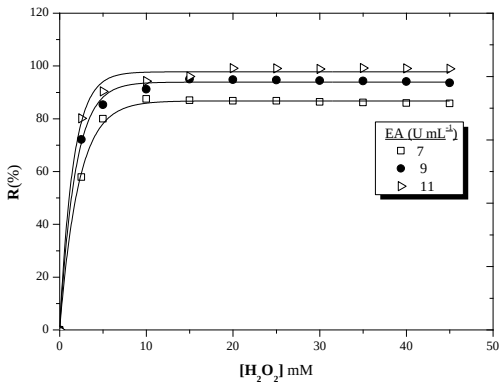


Figure 9. The effect of hydrogen peroxide concentration on MB removal by STP. pH= 7; T=25 °C; [MB]=100 mg L^{-1}; t= 2 hours.

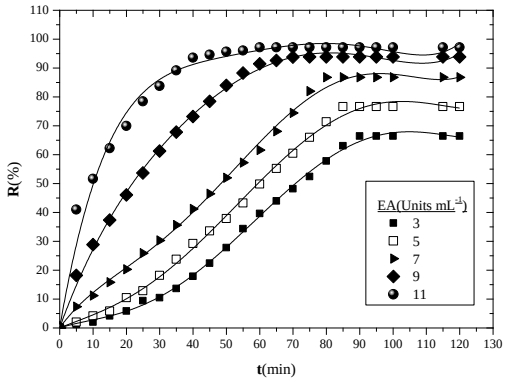


Figure 10. Oxidation kinetics of MB by different enzymes activities of STP. pH= 7; T=25 °C; [MB]=100 mg L^{-1}.

3.4.3. The effect of contact time

Figure 10 shows clearly that the increase in enzyme activity reduces the treatment time of this MB. The optimal treatment times were 90, 85, 80, 75 and 60 minutes corresponding to the enzyme activities 3, 5, 7, 9 and 11 U mL^{-1} respectively. Such as, the percentage removals were achieved 66, 76.6, 86.7, 94, and 97% for 3, 5, 7, 9 and 11 U mL^{-1} respectively.

3.5. Elimination of MB by the hybrid bioprocess: SDS-cork/STP

The minimum value of MB removal was a satisfactory in the order 93% for [MB] = 500 mg L^{-1} and enzyme activity 3 U mL^{-1} and the value of the maximum MB removal percentage was very satisfactory in the order of 99.9% for a [MB] = 200 mg L^{-1} and EA= 11 U mL^{-1} (Figure 11).

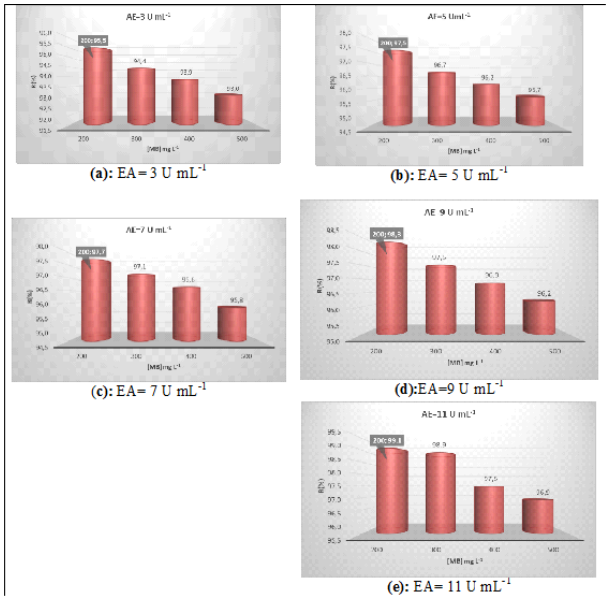


Figure 11. MB removal by the hybrid bioprocess SDS-cork/STP at different concentrations of MB and enzymes activities, pH =6.5; T =25 °C; mass of SDS-cork=0.04 g.

From the Table 1, the hybrid process (SDS-cork/STP) can treated high and low concentration of MB dye from 200 to 500 mg L^{-1} with very satisfactory performance removal which it reaches to 99.1 and 96.9% respectively according to reasonable treatment times. These percentages removals were unobtainable against the simple process: biosorption on SDS-cork and STP. Such that, the treatment of MB by the hybrid process was carried out in series according to two steps respectively (t_1+t_2). Where the first step (t_1) is carried out by SDS-cork process, such as this process is characterized by a high- performance in eliminating of high concentrations on MB (from 100 to 400 mg L^{-1}). The second step followed by STP process(t_2). The last process characterized by our short treatment time but it does not allow for treatment of high concentration on MB (≤100 mg L^{-1}).

Table 1. Performance of the three processes SDS-cork, STP and SDS-cork/STP for MB dye removal

	[MB] mg L^{-1}	t (min)	R (%)
SDS-cork	100	t_1=90	81
	200	t_1=120	88
	300	t_1=180	98
STP	100	t_2=60	97
SDS-cork/STP	200	t_1+t_2=90 + 60	99.1
	300	t_1+t_2=120 + 60	98.9
	400	t_1+t_2=180 + 60	97.5
	500	(t_1)+t_2= (180 +90) + 60	96.9

Table 2. Comparison between different performance process for MB dye removal

Process	[MB] (mg L^{-1})	t (min)	R (%)	References
SDS-cork/STP	500	330	**96.9**	This study
	200	150	**99.1**	
Photocatalytic removal using CuSCdS nanocomposite under visible irradiation in the presence of H_2O_2	10	10	99.97	Mahanthappa *et al.,* 2019
Activated carbons from vegetable sponge of cylindrical loofa	100	20	99	Cherifi *et al.,* 2020
Chemical oxidation/Biosorption	10	60	98.7	Othmani *et al.,* 2020
Photocatalysis/adsorption:(FeNiZnO/polyacrylamide nanocomposite)	8	120	97.56	Kanta *et al.,* 2014
Adsorption/Photodegradation degradation in visible light	40	120	97.54	Rong *et al.,* 2015
mesoporous titania – polyvinyl alcohol	55	8	97.1	Jaseela *et al.,* 2019
Activated sludge	500	30	96.54	Cherifi *et al.,* 2016
Adsorption/ Photocatalytic elimination under UV irradiation	16	120	90	Ahmed *et al.,* 2017

The comparison study between our hybrid process with others processes for MB removal presented on the Table 2. It indicated than, the SDS-cork/STP process can almost treated low and high MB concentration with very high removal percentage for modest treatment times.

4. Conclusion

Hybridization of two bioprocesses: biosorption on SDS-cork and STP was successful. This hybridization was carried out under optimal conditions of pH (6.5), the temperature (25 °C), $[H_2O_2]$ = 10 mM for EA = 3, 5 and 7 U mL^{-1}, $[H_2O_2]$ = 15 mM for EA = 9 U mL^{-1} and $[H_2O_2]$= 20 mM for EA = 11 U mL^{-1}, and a contact time equals the sum of two optimal treatment times for both processes and a SDS-cork mass of 0.04 g.

As such, this hybridization was tested according to two parameters, the initial concentration of MB they varied between 200 and 500 mg L^{-1} and the enzyme activities are varied between 3 and 11 U mL^{-1}. The minimum removal percentage of MB was assumed to be satisfactory on the order of 93% for [MB] = 500 mg L^{-1} and EA= 3 U mL^{-1} and the maximum removal percentage was of a very high value 99.9% satisfactory for [MB] = 200 mg L^{-1} and 11 U mL^{-1}.

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