



Biodegradation of triphenylmethane dye malachite green by a newly isolated fungus strain

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ABSTRACT

Malachite green (MG) is a synthetic cationic dye widely used in the textile industry as a coloring agent. It is reported to be a potential carcinogenic and mutagenic agent for the living organisms in nature. Hence, it is essential to treat and release the industrial effluents containing MG to avoid serious and irreversible effects to both the natural environment and living organisms. An effective method for mitigating the effects of toxic industrial dye is by utilizing microorganism to degrade it to non-hazardous compounds. *Lasiodiplodia* sp is a novel fungus species isolated from biota containing a high concentration of xenobiotics. It exhibits the ability to degrade malachite green in a wide range of temperatures (20 °C - 45 °C) and pH (3–12). The maximal degradation of MG was found to be 96.9% given the dye concentration of 50 mg/L, pH of 7 and temperature of 30 °C. The maximum rate of dye degradation was determined using Hanes–Wolf plot and found to be 123.5 mg-MG/g-cell/h. FTIR analysis endorses the conversion of MG into aromatic amides, carboxylic acids, alkenes and alkane suggesting that decolorization is occurred through oxidation of dyes by the action of laccases. X-ray diffraction analysis indicated that degradation was established by the process of bio adsorption and mineralization of dye in fungi mycelium. Chemical stability test on fungi mycelium indicated that the newly isolated species confer stability under adverse reaction conditions.

1. Introduction

Over 280,000 tonnes of coloring dyes are released from the industrial effluents into the aqueous environment annually, causing damages to both flora and fauna (Sen., 2016). These effluents comprising toxic dyes are to be treated suitably to mitigate the potential damages caused to the environments. This presses for a process to treat dyes in an economically feasible and environmentally friendly manner. Several methods are available and one among them is the biodegradation of dyes using microorganism. Several studies have already demonstrated biodegradation of MG using *Penicillium simplicissimum* (Chen and Yien Ting, 2015a), *Dietzia maris* (Bera et al., 2016), *Ochrobactrum pseudo-grignonense* (Chaturvedi and Verma, 2015), *Pseudomonas putida* (Kobayashi et al., 2017) and *Debaryomyces nepalensis* (Moe et al., 2015). Apart from the biodegradation process, Photocatalyst based degradation of dyes is an emerging area of research and many studies have been reported till date (Dashtian et al., 2018; Mosleh et al., 2016a, 2016b, 2016c, 2017, 2018; Mousavinia et al., 2016; Saravanan et al., 2013, 2015, 2016). *T. asperellum* was used to degrade MG (Shanmugam et al.,

2017) and reported 97.18% of dye removal compared to the control. It was primarily due to the laccase mediated enzymatic degradation and resulted in the production of less-toxic metabolites as indicated by FTIR analysis. The degradation was established by converting toxic MG to benzaldehyde via Michler's ketone pathway. Another study reported biodegradation of MG utilizing *Aspergillus flavus* as the potential organism by synthesizing laccases and manganese peroxidase (Barapatre et al., 2017). *A. flavus* degraded MG completely within 8 days, making them a successful species for biodegradation. Laccase produced from *Trichoderma harzianum* was immobilized on sol-gel and the degradation process resulted in 100% decolorization of MG. Moreover, the products produced as the result of degradation was non-toxic in nature. In another study, A rotating packed bed type reactor had been used for degradation of malachite green using HKUST-1 metal-organic framework, SBA-15 and visible light (Mosleh et al., 2016d). Chemically synthesized adsorbents are used to treat industrial effluents containing toxic dyes by the process of adsorption (Asfaram et al., 2015; Ghaedi et al., 2015; Gupta et al., 2011, 2014; Huang et al., 2017; Saleh and Gupta, 2011, 2012).

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In the current research, a novel fungal strain was isolated from biota containing a high amount of xenobiotics. It was subjected as a catalyst to degrade toxic industrial malachite green dye, resulting in the conversion to non-toxic compounds. Metabolites produced during the biodegradation process were assessed qualitatively and the mechanism of degradation is analytically represented. Degradation kinetics and various physicochemical conditions suitable for optimal degradation were determined through experiments.

2. Materials and methodology

2.1. Chemicals and reagents

Malachite green and Ethyl acetate were purchased from Himedia Laboratories Pvt. Ltd. Hydrochloric acid, Ethanol and Sodium hydroxide was purchased from Sigma-Aldrich Pvt. Ltd. MEB and MSM media were obtained from Himedia Laboratories Pvt. Ltd.

2.2. Microorganism

The fungal strain was isolated using serial dilution method from a wood-decaying mushroom sample collected from the Western Ghats, India. The fungal stock has been maintained in Potato Dextrose Agar slant and stored at 4 °C (Oduro-Mensah et al., 2018).

2.3. Culture media

Liquid mineral salt media ($\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ - 0.1 g/L, K_2HPO_4 -0.1 g/L, NH_4NO_3 -1 g/L, KCl - 0.1 g/L) with 25 ml of trace element solution (MgSO_4 -15.4 mg/L, FeCl_3 - 40 mg/L, $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ -6.3 mg/L, Traces of $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$, $\text{NH}_4 \cdot \text{MO}_7 \cdot \text{O}_{24} \cdot 4\text{H}_2\text{O}$) was prepared. The media was optimized to Malt extract 20 g/L, Dextrose 20 g/L and Peptone 6 g/L by trial and error method for decolorization and biomass growth.

2.4. 18S rRNA sequence analysis

The isolated strain was sent to Rajiv Gandhi Centre for Biotechnology (RGCB) to analyze the 18S rRNA sequence for identification of the isolated microorganism (Pandey et al., 2018). The obtained sequence was submitted to the National Centre for Biotechnology Information (NCBI) and an accession number (MG661769) was obtained. The gene sequence was analyzed using the Basic Local Alignment Search Tool (BLAST) of NCBI and a phylogenetic tree was constructed using Molecular Evolutionary Genetics Analysis Version 7.0 (Mega 7.0) as shown in Fig. 1.

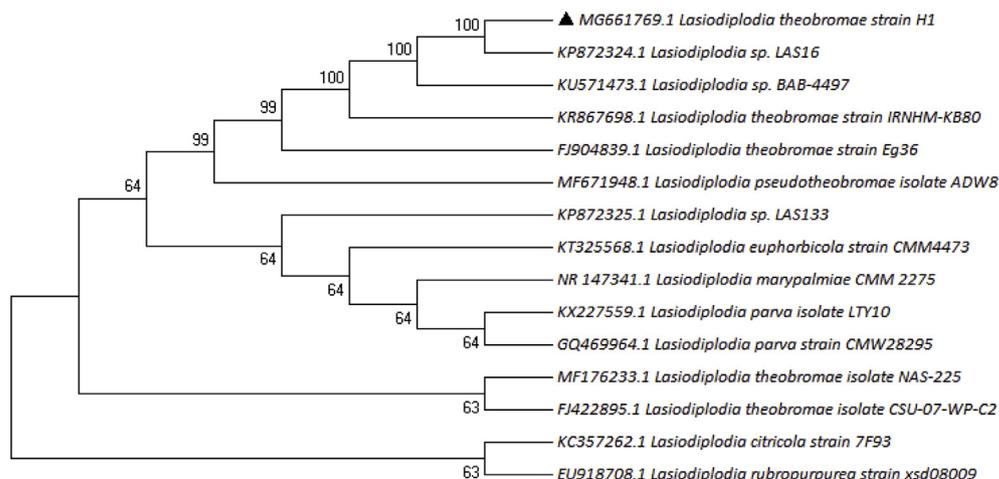


Fig. 1. Evolutionary position of isolated fungi based on sequence. The phylogenetic tree constructed in MEGA7 showing the taxonomic position of isolated fungi among the members of the genus *Lasiodiplodia*.

2.5. Degradation experiments

The newly isolated fungus was acclimatized by culturing in MEB broth containing 2.5 $\mu\text{g}/\text{mL}$ of MG. The absorbance of the supernatant was determined at regular intervals to find the amount of dye degraded. The physicochemical parameters such as pH (3–12), Temperature (25 °C - 45 °C) and concentration (50 mg/L – 500 mg/L) that influence the degradation was optimized and carried out in triplicates along with a control (Wanyonyi et al., 2017).

2.6. Analytical procedures

2.6.1. FT-IR

FTIR analysis was performed to examine the degradation of MG and determine the toxicity of degraded products through functional group modifications. The liquid-liquid extraction was performed using ethyl acetate as a mid-polar solvent on degraded metabolites. Potassium bromide was kept in a hot air oven for 30 min at 105 °C to remove the moisture present in the sample. It was further powdered and made in the ratio of 5:95 (w/w) for analysis. The data was recorded in the mid-IR range of 400–4000 cm^{-1} , for a total of 16 scans.

2.6.2. XRD

Macromolecular structure and stability of *Lasiodiplodia* sp. were determined using X-ray diffraction pattern to assess its potential to degrade the dye and interpolate the mechanism of degradation. For XRD analysis, both the fungi mycelium from MG degraded broth with initial MG concentration of 10 mg/ml was taken out, rinsed in distilled water, dried. Mortar and pestle were used to ground the dried fungal mycelium into a finely powdered form. They were packed in a zip lock cover and placed in desiccators to avoid moisture. The control contained powdered fungal mycelium cultivated in the absence of MG. Mycelium was subjected to XRD at an interval of 2θ (10–75) with the rate of $3^\circ/\text{min}$ at λ of 1.5406 Å.

2.7. Chemical stability test

The chemical stability of fungal mycelium was determined based on the amount of dye released after stability treatment. Mycelium was separated from degradation broth, washed and suspended in distilled and autoclaved at 121 °C at 15 psi. 1M HCl, 1M NaOH and 70% ethanol were used to treat the dried fungal mycelia for 1hr and optical density was measured (618 nm) using a UV-visible spectrophotometer.

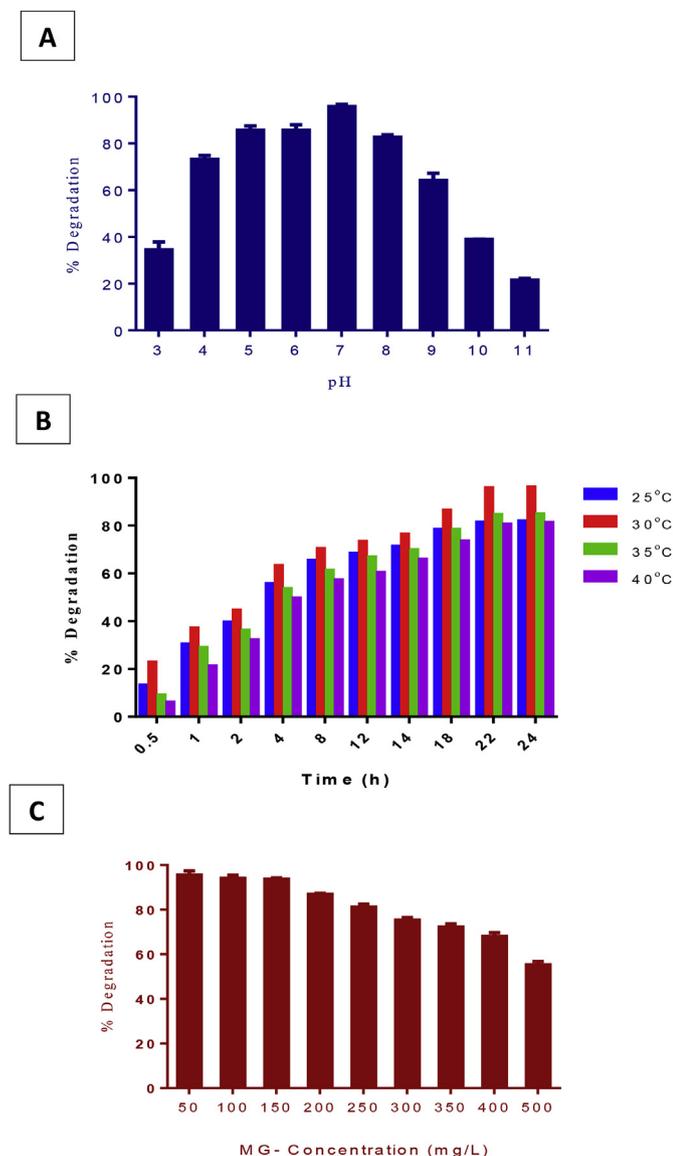


Fig. 2. (A) Effect of pH in degradation of Malachite green by specific fungal strains (B) Effect of Temperature in malachite green degradation by specific fungal strains (C) Effect of initial dye concentration percentage degradation. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

3. Results and discussion

3.1. Effect of pH

The effect of pH in biodegradation of MG was assessed between pH 3–12 to determine an optimum value for maximum degradation (Fig. 2A). The temperature and dye concentration was kept constant at 30 °C and 50 mg/L respectively. Results show that as the pH of broth increases from 3 to 7, dye degradation percentage increased and started to decrease from pH 8. This result implies that pH 7 was found to be an optimum value for MG degradation with maximum degradation percentage of 96.5% attained by the fungi as shown in Figure 2A. It also shows that *Lasiodiplodia* sp. has the potential to degrade malachite green slightly in strong acidic pH range making it a more suitable candidate for malachite green degradation and plays a vital role in bioremediation of various xenobiotics.

3.2. Effect of Temperature

Importance of temperature in degradation was studied in a temperature range of 25 °C–45 °C by keeping other parameters such as pH (7) and the dye concentration (50 mg/L) constant (Fig. 2B). Initially, the rate of degradation increased with increase in temperature (20 °C–30 °C) with maximum degradation obtained at 30 °C and decreased gradually after 30 °C. This result proves that *Lasiodiplodia* sp is a suitable candidate for bioremediation of MG at a wide range of temperatures.

3.3. Effect of dye concentration

The relationship between dye concentration and the rate of degradation was determined by varying the dye concentration ranging from 50 to 500 mg/L in MEB broth at pH 7 and incubated at 30 °C (Fig. 2C). The fungi culture was able to degrade the dye with 96.9 and 89.18 (% of degradation) for 50 mg/L and 500 mg/L respectively in 24 h (Fig. 2C). It was found that both dye degradation and fungal growth rate decreases gradually with an increase in dye concentration. The experiment was carried out under 24 h dark conditions and the dye degradation % remained the same. This shows that the fungus was able to degrade dyes equally well in both light and dark conditions (Nath et al., 2019).

3.4. Kinetics

The dye decolorization by the fungal species is an extracellular enzymatic process whose kinetics can be calculated using Hanes–Woolf Equation (Equation (1)).

$$\frac{[S]}{V} = \frac{[S]}{V_m} + \frac{K_m}{V_m} \quad (1)$$

A comparison was made between model predictions and experimental data as shown in Fig. 1. ($R^2 = 0.98$), which implies that the model describes the trend of experimental data satisfactorily well. The usage of free cells is restricted due to its instability in extreme physiochemical factors (Voberkova et al., 2018). The value of K_{dye} and V_{dye} max for *Lasiodiplodia* sp was found to be 157.94 mg/L and 20.7 mg/L/h (K_{dye} is the measure of the enzyme-substrate complex where a higher K_{dye} indicates a weak binding). The efficiency of degradation was compared with other studies (Table 1). From the analysis, the *Lasiodiplodia* species exhibit better dye degradation rate.

3.5. Fourier-transform infrared spectroscopy (FT-IR)

FTIR analysis was performed (Spectrum 100, Perkin Elmer, USA) on a sample containing MG and MEB as shown in Fig. 3. Analysis showed intense peaks at 714 cm^{-1} and 814 cm^{-1} for 1,2,3 trisubstituted aromatic and 1,3,5 trisubstituted aromatic compounds, 946 cm^{-1} for N–OH oxime, 1459 cm^{-1} due to C–C stretch in aromatic, 1585 cm^{-1} due to C=C stretch, 1522 cm^{-1} due to N–O and asymmetric stretching of CH_2 at 2981 cm^{-1} as shown in Fig. 3A. Intense peaks at 714 cm^{-1} and 814 cm^{-1} show the presence of undegraded aromatic compounds. Other peaks correspond to the presence of complex undegraded compounds. FTIR analysis of metabolites obtained by degradation of MG by *Lasiodiplodia* sp. showed a peak at 807 cm^{-1} for meta-substituted aromatic compounds for C–H bending, peak at 1032 cm^{-1} for amines (N–C), the peak for 1261 cm^{-1} due to CH_3 stretch, an intense narrow peak at 1399 cm^{-1} for C=O stretch, 2922 cm^{-1} alkenes (C–H) and 3115 cm^{-1} for carboxylic acid due to dimer OH (Fig. 3B). These results clearly indicate that the metabolites obtained by degradation of MG show a decrease in presence of hazardous components compared to control. The analysis also shows that MG is converted into aromatic amines, carboxylic acids and to alkanes. This conversion might be the possible mechanism of MG degradation by

Table 1

Comparison of malachite green dye biodegradation/biosorption using various microorganism and biosorbent reported in the literature with the present work.

Author	Microorganism/Biosorbent	Process parameters	% degradation/% of dye removal
Pandey et al. (2018)	<i>Lenzites elegans</i> WDP2	Initial dye conc: 100 mg/L pH: 7.0 ± 0.2; Temp: 30 °C ± 2 °C; Time: 12 days	21.27%
Rangabhashiyam et al. (2018)	<i>Carica papaya</i> wood	Batch: 50 ml; Biomass: 0.06 g Initial dye conc: 10–50 mg/L Temp: 50; pH: 10 RPM:120; Time:140 min	95.38%
Vyavahare et al. (2018)	Sugarcane bagasse biochar	Batch: 10 ml Biomass: 0.5 g Initial dye conc: 3000 mg/L Temp: 60 °C; pH: 7.5; Time: 51.89 min	99.99%
Gan et al. (2018)	<i>Burkholderia cepacia</i> immobilized on eucalyptus leaves	Batch: 50 ml Biomass: 0.05 g Initial dye conc: 60 mg/L Temp: 30 °C; pH: 6; RPM: 150 Time: 360 min	94.8%
Lallawmsanga Leo et al. (2018)	<i>Pleurotus pulmonarius</i> BPSM10 laccase	Batch: 50 ml Enzyme: 10 ml Initial dye conc: 100 mg/L Time: 1 h	68.6%
Zhang et al. (2018)	Laccase-biotitania biocatalysts	Batch: 10 ml Adsorbent: 20 mg Initial dye conc: 20 mg/L pH:4; Temp: 70 °C; Time: 6 h	90%
Li et al. (2018)	<i>Burkholderia cepacia</i> immobilized on graphene oxide	Batch: 50 ml Adsorbent dose: 0.11 g/L Initial dye conc: 114.5 mg/L Temp: 31.5 °C pH: 5.85; RPM: 150; Time: 96 h	99.3%
Cardoso et al. (2018)	<i>Panus strigellus</i> laccase	Batch: 4 ml Initial dye conc: 1 mg/L Enzyme: 3.6 ml Temp: 28 ± 1 Time: 72 h	68%
Wanyonyi et al. (2017)	<i>Bacillus cereus</i> KM201428 enzymes	Batch: 50 ml Enzyme conc: 10 ml of crude protease (For Biofilm growth) Initial dye conc: 9.3 mg/L Temp: 40 °C; RPM: 150; Time: 12 h	88.81%
Shanmugam et al. (2017)	<i>Trichoderma asperellum</i> laccase	Batch: 5 ml Enzyme conc: 1.50 U/mL Initial dye conc: 122.66 mg/L pH: 6.75; Temp: 25 °C; Time: 98.58 min	97.18%
Hussain et al. (2017)	<i>Cunninghamella elegans</i>	Batch: 50 ml Biomass: 5 ml (For Biofilm growth) Initial dye conc: 29 mg/L Temp: 28 RPM: 150; Time: 24 h	95%
Banerjee et al. (2016)	<i>Avena sativa</i>	Batch: 100 ml Initial dye conc: 25 mg/L particle size: 150 µm Adsorbent dosage: 0.25 g pH: 8.0; Temp: 40 °C RPM: 200; Time: 80 min	97.6%
Bera et al. (2016)	<i>Dietzia maris</i> NIT-D	Initial dye conc: 15 mg/L pH: 7; Inoculum 2% (v/v) Nitrogen source: 0.5% (w/v) Temp: 37 °C RPM: 180; Time: 4 h	72.05%
Chen and Yien Ting (2015a)	<i>Penicillium simplicissimum</i>	Batch: 100 ml Biomass: 2.0 ± 0.1 g Initial dye conc: 100 mg/L Temp: 30 ± 2 °C RPM: 150; Time: 14 days	54%
Chen and Yien Ting (2015b)	<i>Coriolopsis</i> sp	Batch: 100 ml Biomass: 2.0 ± 0.1 g Initial dye conc: 100 mg/L Temp: 30 ± 2 °C RPM: 150; Time: 9 days	52%
Dahri et al. (2015)	<i>Casuarina equisetifolia</i> needle	Batch: 20 ml Adsorbent dose: 0.04 g Initial dye conc: 100 mg/L Temp: 25 °C pH: 3.2; RPM: 250; Time: 1 h	88%
Jasińska et al. (2015)	<i>Myrothecium roridum</i>	Batch: 20 ml Inoculum: 2 ml Initial dye conc: 10 mg/L Temp: 28 °C pH: 6; RPM: 150; Time: 24 h	93%

(continued on next page)

Table 1 (continued)

Author	Microorganism/Biosorbent	Process parameters	% degradation/% of dye removal
Bhagavathi Pushpa et al. (2015)	EM based compost	Batch: 100 ml Initial dye conc: 1000 mg/L Adsorbent dosage: 4 g/L pH: 8; Temp: 30 °C; RPM: 150	95.7%
Present Study	<i>Lasiodiplodia</i> sp	Initial dye conc: 50 mg/L pH: 7; Temp: 30 °C; Time: 24 h	96.9%

Lasiodiplodia sp. Formation of amines, amide, and carboxylic acids are due to C=O stretch and N–H stretch. Presence of dimer OH confirms the degradation of MG into less toxic metabolites (Kuhar et al., 2015; Zhang et al., 2018).

3.6. XRD

The XRD data for Malachite green decolorization (before and after treatment) is shown in Fig. 4. It shows that before dye treatment *Lasiodiplodia* sp. gave peaks at 43.6615, 49.1501 and 72.7104. The peaks shifted from its original position when treated with MG to 21.3904,

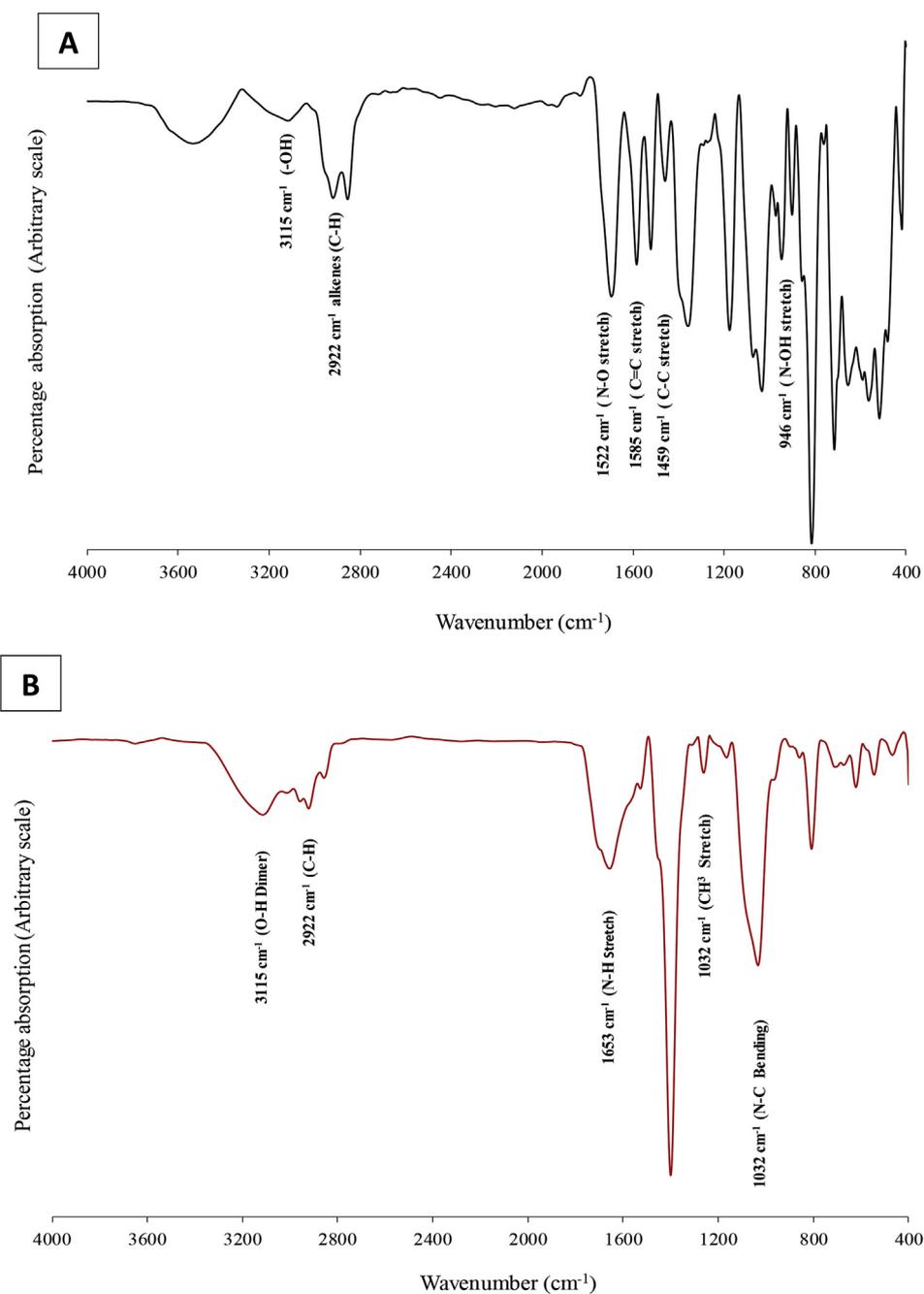


Fig. 3. (A) FTIR Spectral analysis for MG + MEB metabolites (B) FTIR Spectral reading for MG + *Lasiodiplodia* sp. + MEB metabolites.

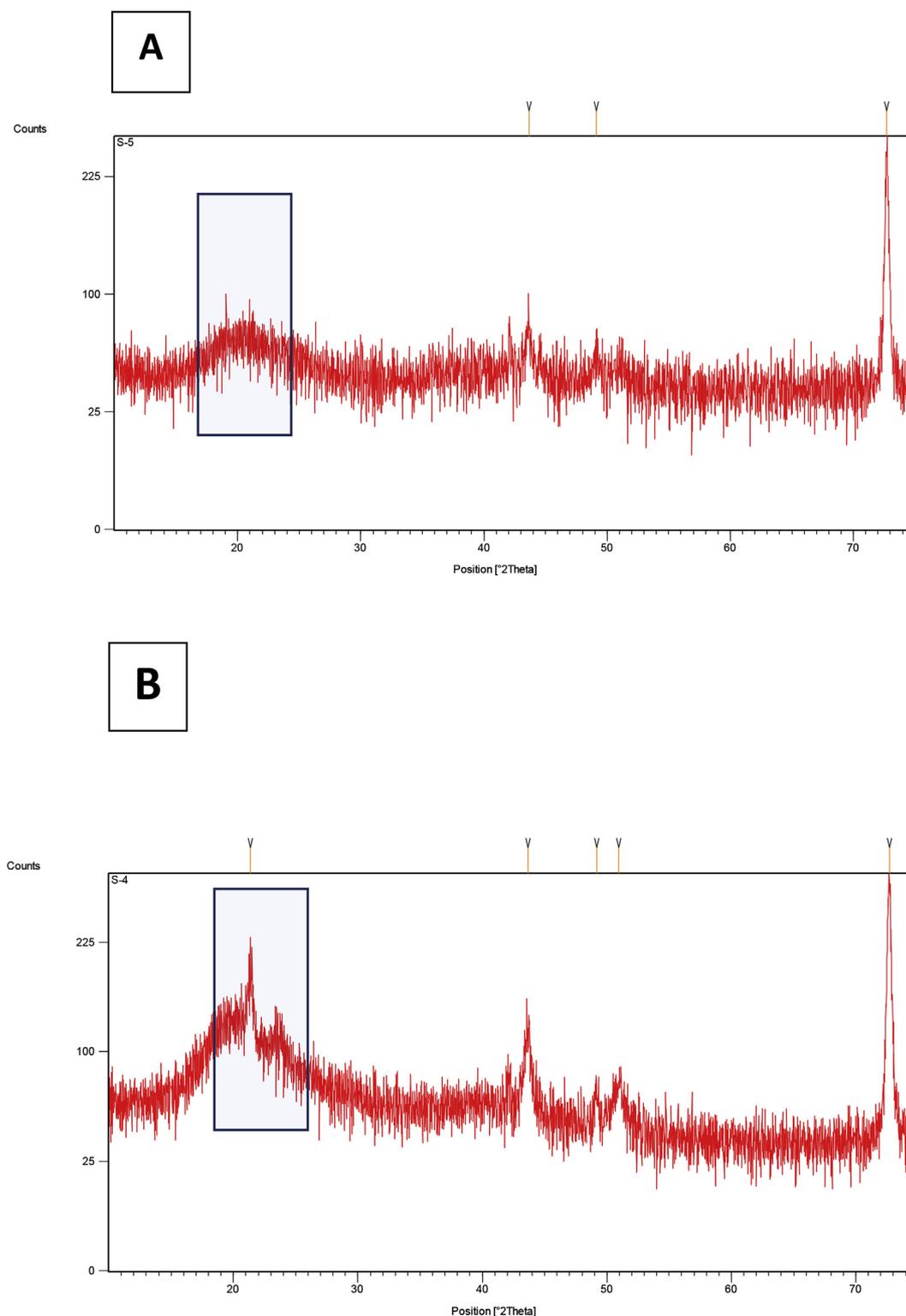


Fig. 4. XRD analysis of fungi before dye treatment (A) and after dye treatment (B).

43.6800, 49.1716, 50.9624 and 72.6849. These new peaks imply the presence of structural variation in the mycelium of *Lasiodiplodia* sp. attributed by adsorption of malachite green. The possible mechanism of dye degradation is due to adsorption of dye in mycelium body followed by mineralization of dye by metabolites of fungi (Fig. 4). The degradation of dye by *Lasiodiplodia* sp. might be due to the release of extracellular metabolites.

3.7. Chemical stability of absorbed dye

The fungi mycelium did not release any dyes when treated with chemicals. This observation revealed that the species has a greater chemical stability when subjected to adverse environmental condition and hence has the potential to act as an excellent candidate to bioremediate MG.

4. Conclusion

The study clearly shows that the novel fungi *Lasiodiplodia* sp. has the potential to degrade and mineralize MG dyes. The physicochemical parameters that influence the MG degradation by fungi were represented and found to be more tolerant to grow and degrade the dye in a wide range of pH and temperatures. The chemical stability test revealed that the fungus was found to be stable when treated with chemicals. This implies that these fungal isolates can withstand adverse environmental conditions to remediate the contaminants from the environment. Throughout the study, the initial inoculum biomass was made constant for all the varying concentrations of dyes and the percent of dye degradation was not found to be significantly varying with concentration. This leads to an interpretation that dye decolorization may be independent of the initial inoculum biomass. In addition, its degradation rate decreased gradually as the concentration of the dye increased in broth. FTIR analysis on MG degraded metabolites depicted the formation of aromatic amines, carboxylic acids, alkanes, alkenes, which shows the formation of non-toxic metabolites during degradation of MG, such as conversion of toxic aromatic amines into carboxylic acid. This justifies the involvement of laccases in the degradation process and conversion of carboxylic acids into alkanes and alkenes may be due to the lipase enzyme secreted by fungi. XRD analysis reveals structural variation in the mycelium after MG treatment, suggesting that bio-adsorption and mineralization of dye might be the possible way of dye treatment. This study reveals that *Lasiodiplodia* sp could probably be a pollution control biological candidate for proper environmental management.

Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.cbab.2019.01.030>.

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