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Biodegradation of synthetic orange G dye by *Plearotus sojar-caju* with *Punica granatum* peal as natural mediator

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ABSTRACT

Potential of Laccase (Lac) for the decolorization of orange G dye in the presence of natural redox mediator was investigated. Dye degradation was confirmed by Ultraviolet–visible spectroscopy (UV–Vis), High-performance liquid chromatography (HPLC) and Fourier-transform infrared spectroscopy (FTIR) techniques. Lac showed promising efficiency for the degradation of dye and redox mediator enhanced it significantly. At the Lac activity of 156.78 IU/mL, the Orange G dye was degraded up to 61.45%. The Lac activity increased significantly in the presence of *Punica granatum* peel extracts as a source of redox mediator and reached at 279.27 IU/mL and orange G dye decolorization was increased up to 85% and at the optimum conditions of process variables, up to 97% dye degradation was achieved. The phytotoxicity was evaluated by measuring germination of *Trigonella foenum-graecum* L seed in treated and untreated dye. The seed germination was inhibited 100% in untreated dye solution and germination was recorded to be 81.03% when seeds are exposed to treated dye solution. Lac (*P. sojar-caju*) in the presence of natural redox mediator (*P. granatum* peal) proved to be efficient for the biodegradation of dye, which could possibly be used for the remediation of dyes in textile wastewater.

1. Introduction

The dyes mainly originate from textile, printing, cosmetics, paper and pharmaceuticals industries, which are discharged in to the water sheds without any treatment. Water contamination due to dyes is not only dangerous to the aquatic organisms, but also toxic (mutagenic and genotoxic) human beings (Selvam et al., 2013; Nikam et al., 2017; Britos et al., 2018; Agrawal and Verma, 2019; Arunprasath et al., 2019; Hassen and Asmare, 2019; Iqbal et al., 2019; Iwuoha and Akinseye, 2019; Oussama et al., 2019; Sutar et al., 2019). The pollutants in water bodies inhibit the sunlight penetration that is required for the survival of living organisms. Moreover, these pollutants exert carcinogenic effects on living organism (Bilal et al., 2016b; Iqbal, 2016). In dying process, un-reacted (dye-fiber low fixation) dyes are discharged in water sheds. The synthetic dyes have complex structures and are very stable and remained in the environment for longer time and for complete removal of dyes from effluents, efficient treatment are required (Bilal et al., 2016b). To date, different physical and chemical methods are in practice e.g. adsorption, flocculation, photolysis, chemical precipitation, electrochemical treatment, oxidation and reduction, membrane filtration for the removal of dyes from industrial wastewater (Husaain et al., 2015; Mary Jacintha et al., 2015; Mathubala et al., 2016a; Jafarinejad, 2017b, a; Laissaoui et al., 2017; Legrouri et al., 2017; Minas et al., 2017; Shameem et al., 2017). However, these treatment approaches have some drawbacks i.e., expensive, sludge formation during treatment and secondary pollution issues due to use of chemical (Manzoor et al., 2013; Iqbal and Bhatti, 2015; Ashar et al., 2016; Nadeem et al., 2016). On the other hand, the biodegradation is a phenomenon of transformation of organic compounds by living organisms and it is also a natural process. It has been reported that the biodegradation converts the organic dyes

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completely into harmless end products (Nouren et al., 2017a, 2019).

The white-rot type (WRF) fungi are capable of degrading recalcitrant compounds. WRF produce enzymes such as Lac, manganese peroxidases (MnP), lignin peroxidases (LiP) and versatile peroxidase (VP; EC 1.11.1.16), which are responsible for the degradation of dye and other pollutant in the medium (Palli et al., 2016; Rekik et al., 2019). The Pleurotus, a WRF belongs to a ligninolytic fungi class, which are widely cultivated as edible Pleurotus sajor-caju. Pleurotus sajor-caju mycelium can grow at higher temperature in wide range of pH and produces extracellular enzymes including Lac, LiP, MnP and VP (Lee et al., 2014). WRF showed promising efficiency for the degradation of pesticides, dyes, pharmaceutical agents and polycyclic aromatic hydrocarbons (Palli et al., 2016; Rekik et al., 2019). The natural mediators (redox) have been reported to be sportive the enhancement of degradation efficacy of WRF i.e., Direct yellow 4 dye biodegradation was promising in the presence of natural various mediators (veratryl alcohol, hydroxybenzotriazole, vanillin, p-coumaric acid, syringaldehyde, syringic acid and pyrocatechol). At optimum condition (pH 5.0, 50 °C, 24 U/mL and 0.25 mM H_2O_2 concentration, the C. limon peroxidase (POD) furnished 60% DY4 dye degradation (Nouren et al., 2017b). Also, the sweet lime based natural mediator showed excellent efficiency for the decolorization of textile effluents (Nouren et al., 2019) and similar findings have been reported by other researcher that bioremediation is promising for the decolonization dyes and textile wastewater (Bilal et al., 2016a; Bilal et al., 2016b, c).

Keeping in view the aforementioned facts, present study was designed to appraise the Orange G dye biodegaration efficiency of *P. sajor-caju* in combination with *P. granatum* peal as a natural source of redox mediator. Process variables (conditions) were optimized for maximum degradation of Orange G dye. The biodegradable was monitored by UV–Vis, FTIR and HPLC analysis. The phytotoxicity was evaluated by germinating the *T. foenum-graecum L* seed in untreaed and biotreated dye solution.

2. Material and methods

2.1. Chemicals, reagents and sample collection

All chemicals and reagent were of analytical grade and purchased form Sigma-Aldrich. The P. granatum peal peels were collected from the local market, Faisalabad, Pakistan. Rhodamine, Orange G, Malachite Green and Black B dyes were purchased from Sigma-Aldrich.

2.2. WRF culture preparation

Pure culture of WRF (local isolate, *P. sajor-caju*) was obtained from Horticulture Department, University of Agriculture Faisalabad, Faisalabad, Pakistan. Potato dextrose agar (PDA) slants was prepared for fungal growth and slants were stored at 4 °C. Similarly, *inoculum* medium was prepared by addition of about 1% sterilized glucose to the salt medium of Kirk's basal (Tien and Kirk, 1988). The medium was then autoclaved at 121 °C for about 15 min for sterilization. After cooling, inoculated with fresh spores of *P. sajor-caju* and placed in shaking incubator (speed: 120 rpm) at 30 °C for 6 days to get a standardized spore inoculum (1×10^8 spores/mL).

2.3. Dye degradation

The dyes decolorization was performed in 250 mL flask and conditions were optimized. In typical procedure, a 100 mL (100 mg/L) solution was placed in flask contains nutrient medium. The flasks autoclaved for 15 min at 121 °C, cooled down and inoculated aseptically (2.5 mL, 1×10^8 spores/mL) under laminar air flow. The flasks were incubated at 37 °C and 2 mL sample was withdrawn every 24 h and reaction was stopped by placing the flasks in boiling water bath for 10 min. Finally, samples were centrifuged at 10,000 rpm for 5 min. The absorbance (CE Cecil 7200, UK) was recorded and by comparing with untreated dye (control) the percentage dye degradation was estimated using relation shown in Eq. (1). Where, A_0 and A_s are the absorbance values of dyes before and after decolonization, respectively. The Orange G dye decolorization was maximum and in subsequent experiments, the effect of process variables and *P. granatum* peel extract was studied for Orange G dye.

$$\text{Decolorization } (\%) = \frac{A_0 - A_s}{A_0} \text{ x } 100 \quad (1)$$

2.4. Liginnolytic enzymes estimation

To study the enzymes secreted by the *P. sajor-caju* during decolorization processes, the supernatants of decolorized dyes samples were analysed for MnP, Lac and LiP activities. For the determination of MnP, LiP and Lac activities, the methods reported elsewhere were adopted (Wariishi et al., 1989), (Shin and Lee, 2000) and (Tien and Kirk, 1988), respectively. 2, 2 azinobis (3-ethylbenzthiazoline)-6 sulphonate (ABTS) was used for absorbance monitoring at 405 nm.

2.5. HPLC and FTIR analysis

The HPLC and FTIR analysis was performed to monitor the degradation process. The FTIR analysis is useful for functional group detection (MubarakAli et al., 2012, 2018; Jeyaraj et al., 2013; Qasim Nasar et al., 2019), while HPLC is helpful to separate, identify, and quantify the individual components in a mixture (Ashar et al., 2016; Nouren et al., 2017a). The degradation product was monitored by reverse-phase HPLC analysis (Saleh, 2005). Briefly, a 100 mL of dye solution was centrifuged at 10000 rpm and equal volume of ethyl acetate was added. Anhydrous Na_2SO_4 was used to dehydrate the extracts and dried in an oven at 80 °C. The dried extracts were mixed with acetone and used for HPLC analysis. For FTIR analysis, the sample was dried at 105 °C and mixed with KBr to prepare discs, which were scanned in the range of 400–4000 cm^{-1} (Almeida and Corso, 2014).

2.6. Phytotoxicity study

For phytotoxicity study, seeds of *T. foenum graecum* L were exposed to untreated and biodegraded dye and seed germination index was estimated (Nouren et al., 2017a). Twenty five seeds of *T. foenum graecum* L were placed in Petri plates with dimensions 115 × 25 mm and 15 mL of dye samples were added. The Petri plates were incubated 25 °C for four days. After four days, the germination index (GI) was calculated using relations shown in Eqs. (2)–(4). All experiments were run triplicate and data was averaged.

$$\text{Seed GI} = [\text{Seed germination } (\%) * \text{Root elongation } (\%)]/100 \quad (2)$$

$$\text{Seed germination } (\%) = [\text{Seed germinated in sample/Seed germinated in control}] * 100 \quad (3)$$

$$\text{Root elongation } (\%) = [\text{Root length in sample/root length in control}] * 100 \quad (4)$$

3. Results and discussion

3.1. Dye degradation efficiency

Initially, four dyes were degraded using *P. sojar-cajuon* and Orange G decolorization was significantly higher versus Rhodamine, Malachite Green and Black B. Orange G dye decolorized was 61.45% at the Lac concentration of 156.78 IU/mL. The Lac was the main enzyme in *P. sojar-cajuon* extracellular enzymes along with minor concentrations of LiP and MnP. Ligninolytic cultures of WRF have been reported to be highly efficient for the decolorization of different pollutants (Palli et al.,

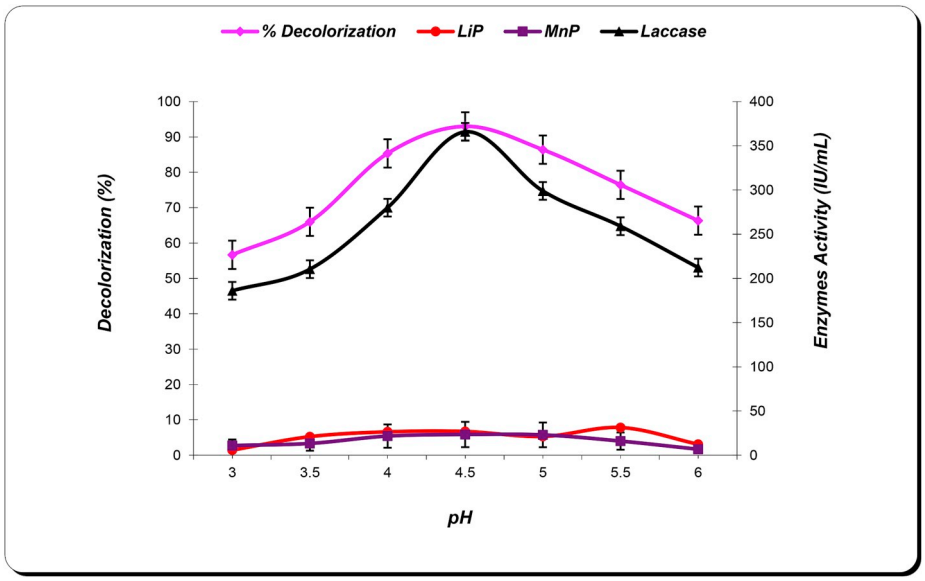

Fig. 1. Effect of pH on the removal of direct orange G and enzyme activity by *Plearouts sajor-caju.* (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

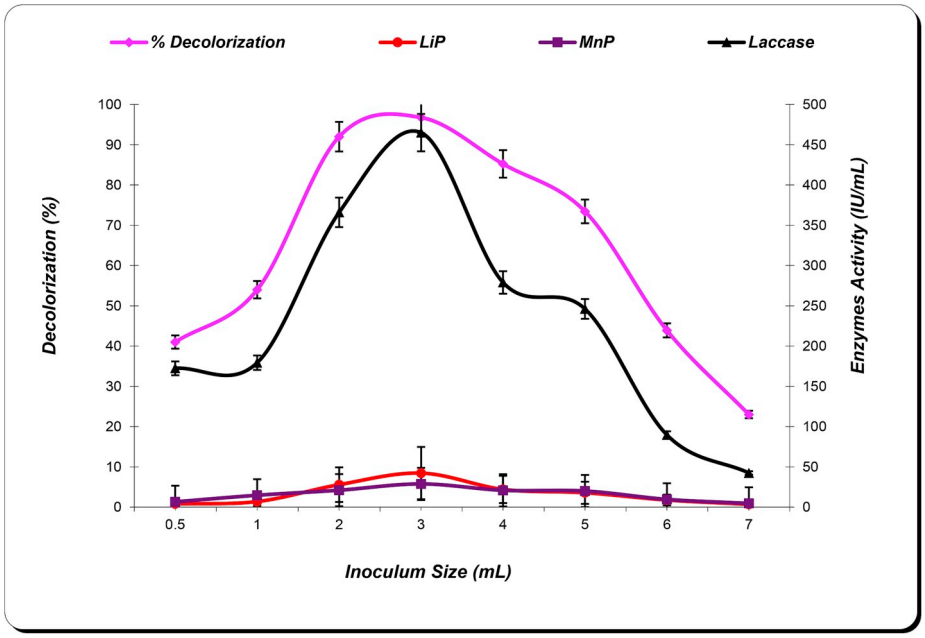

Fig. 2. Effect of inoculum size on the removal of direct orange dye and enzyme activity by *Plearouts sajor-caju.* (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

2016; Rekik et al., 2019). In present study, the different dye behavior is due to the difference in dye structure. Previous studies also documented similar findings that WRF degrade the dyes efficient and *Phellinus gilvus*, *P. sajor-caju*, *P. sanguineus* and *P. chrysosporium* decolorize the dye up to 100%, 94%, 91% and 75%, respectively (Balan and Monteiro, 2001). The effiecnt dyes degradation by fungi has been correlated with the extracellular enzymes i.e., Lac, MnP, LiP and VP (Balan and Monteiro, 2001; Palli et al., 2016; Nouren et al., 2017b, 2019; Rekik et al., 2019).

3.2. Effect of initial pH on decolorization

The dye degradation was also performed as a function of medium pH and the pH effect twas studied in the range of 3.0–6.0. The maximum Orange G dye decolorization of 92.99% was achieved at pH 4.5, followed by 86.4%, 76.45% and 66.33% dye decolorization at pH 5.0, 5.5 and 6.0, respectively (Fig. 1). The dye decolorization was increased up to 4.5 and then decreased beyond this pH values. The low dye decolorization at pH values other than 4.5 might be due to the alteration in enzyme activity since pH affect the structure of enzymes (Nouren et al., 2017a). The enzyme deactivate as a function of pH and acidic pH found more favorable for the enzymes of white rot fungus (Kariminiaae-Hamedaani et al., 2007).

3.3. Effect of inoculum size

The pH 4.5 was adjusted to evaluate the effect of inoculum size on dye decolorization and up to 96.76% decolorization of Orange G was observed using 3 mL inoculum (Lac activity 465.02 IU/mL) (Fig. 2). It

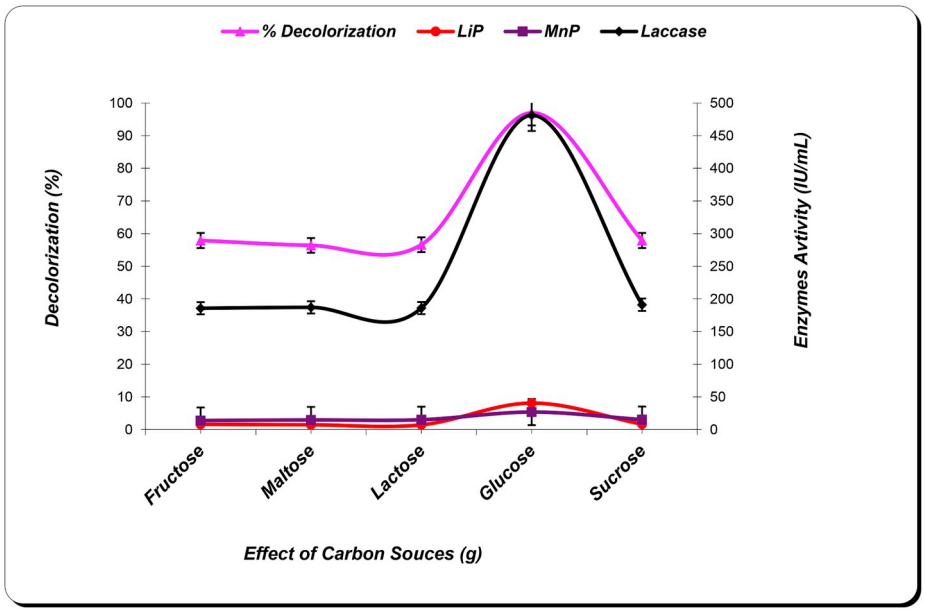

Fig. 3. Effect of carbon sources on the removal of direct orange dye and enzyme activity by *Plearouts sajor-caju.* (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

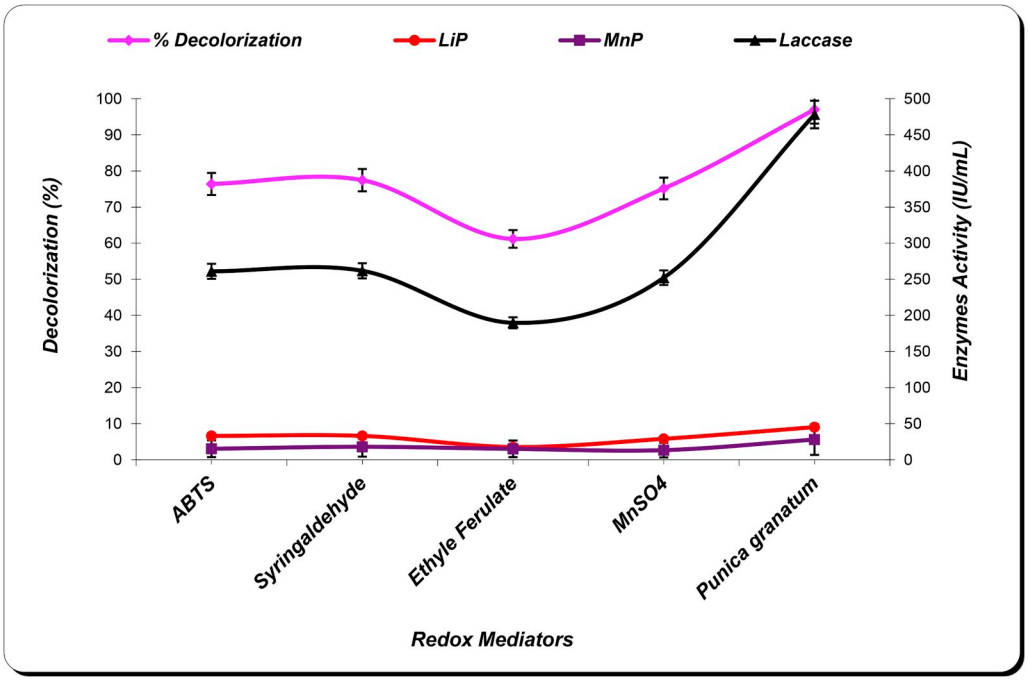

Fig. 4. Effect of Redox mediators on the removal of direct orange dye and enzyme activity by *Plearouts sajor-caju.* (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

was observed that by increasing the inoculum size, the dye degradation was also increased and maximum was observed at 3 mL and beyond this volume, the dye decolorization was decreased gradually. Previous revealed similar trend for the degradation as a function of inoculum size (Asgher et al., 2006). It has been reported that binding enzyme active site to the substrate is pH dependent (Nouren et al., 2017a).

3.4. Effect of carbon sources

The effect of carbon source on degradation was also studied i.e., glucose, fructose, maltose, sucrose and lactose. The rate of dye degradation was achieved up to 97% in the presence of glucose as a carbon source at Lac activity of 481.31 IU/mL followed by sucrose, maltose, fructose and lactose, which showed the degradation of 57.9%, 41.62%, 32.09% and 27.88% for Lac activities of 191.1, 156.89, 137.79 and 133 (IU/mL), respectively (Fig. 3). The production of enzymes depends u the growth of WRF and carbon source supplement enhanced the growth of the WRF, which indirectly, may enhance the Lac production (Nouren et al., 2017a). Previously, the WRF showed variable growth in the presence of different carbon sources. The fungus use glucose consumable glucose as carbon source efficiently, which decrease the lag phase and resultantly, the Lac activity was increased leading to

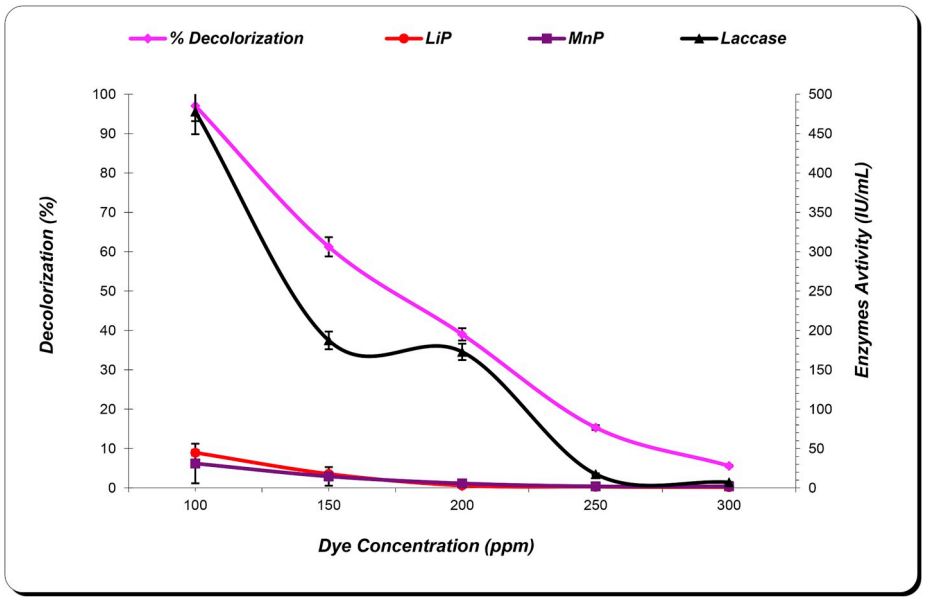

Fig. 5. Effect of dye concentration on the removal of direct orange dye and enzyme activity by *Plearouts sajor-caju.* (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

enhanced dye decolorization (Asgher et al., 2006).

3.5. Effect of redox mediators

Effect of redox mediators on dye degradation was also studied i.e., $MnSO_4$, ABTS, *P. granatum* peel extract, ethyl ferulate and syringaldehydewere and responses are shown in Fig. 4. The redox mediators showed variable effect on the dye degradation, however, *P. granatum* peel extract showed promising efficiency. Using *P. granatum* peel extract as redox mediator, and up to 97% dye degradation was achieved with Lac activity of 478.23 IU/mL followed by 77.45%, 76.4% and 31.2% degradation using syringaldehyde, ABTS and $MnSO_4$ with Lac activities of 478.23, 261.72, 261 and 137 (IU/mL), respectively. Several studies have shown that Lac production could be enhanced many folds in the presence of mediators (Sabu et al., 2005) i.e., Lac from the lignin-degrading species (*T. versicolor, P. pinisitus* and *M. thermophile*) also showed promising decolorization of dyes (Direct Red 28, indigoid Acid Blue 74 and anthraquinonic). The addition of 1-hydroxybenzotriazole as redox mediator enhanced the dye decolorization (Claus et al., 2002). In another study, the dye decolourization was also enhanced with redox mediator in combination with Lac. Violuric acid (5.7 mM) proved to be highly effective mediator for complete decolorization of dye. These findings suggest that the redox mediator along with Lac could be used for efficient degradation of dyes (Soares et al., 2001).

3.6. Effect of dye concentration

The effect of dye concentration (100–300 mg/L) was also investigated and maximum dye degradation of 97.04% was achieved at Lac activity of 477.81 IU/mL for 100 mg/L dye initial concentration. The dye degradation of 61.23%, 39%, 15.34% and 5.6% was observed for 150, 200, 250 and 300 (mg/L) dye initial concentration at Lac activities of 187.34,172.76,17.56 and 7.17 (IU/mL), respectively (Fig. 5). As the concentration of dye was increased, the dye degradation rate was decreased. This trend is in line with previous reports that dye initial concentration may exert negative impact on enzyme activity (Nouren et al., 2017a, 2019). Also (Senan and Abraham, 2004; Murugesan et al., 2007), revealed that the initial dye concentration affected the dye degradation rate negatively.

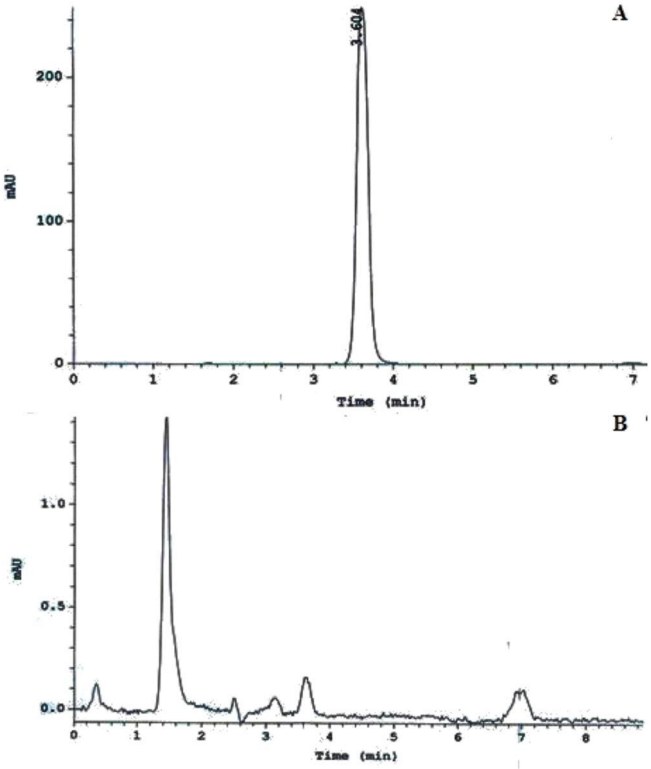

Fig. 6. (A) HPLC chromatogram of un-treated Dye (B) HPLC chromatogram of treated dye.

3.7. Dye degradation by-products

The dye biodegradation product was monitored by HPLC and FTIR analysis and response are shown in Figs. 6 and 7, respectively. The Orange G dye showed major peak at 3.604 min before treatment (Fig. 8A). The dye degradation yield different products and their peaks were appeared at retention time 0.35, 1.55, 2.5, 3.17, 3.6, and 7.0 min and major peak was observed that 1.55 min. Results revealed that the parent

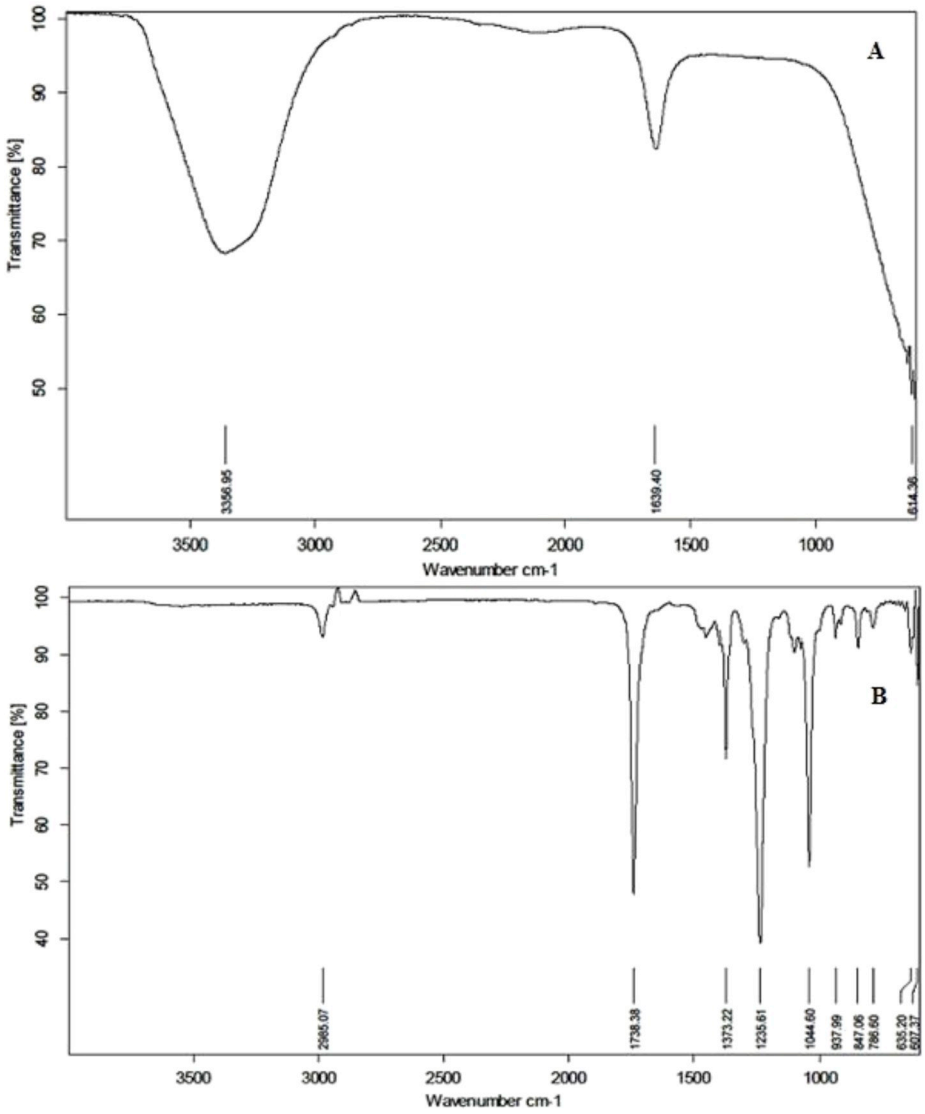

Fig. 7. (A) FTIR spectrum of un-treated dye (B) FTIR spectrum of treated dye.

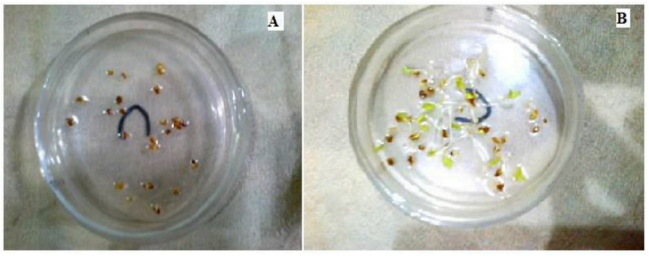

Fig. 8. (A) Germination of seed in 100 mg/L of dye (100% germination inhibition) (B) Germination of seed in 100 mg/L treated dye (81.03% germination).

dye was degraded and transformed different metabolites having low molecular weights. Further comparison of degradation was done with FTIR analysis. It was observed that the functional group in parent dye molecule was disappeared as a result of biodegradation by *P. sajor-caju* and *P. granatum* based redox mediators. A peak at 3356.95 cm^{-1} was due to–NH_2 (amine) in the dye molecule. Peak at 1639 cm^{-1} is due to–CO-NH (amide) and 614 cm^{-1} confirms the aromatic nature of dye. After decolorization the peak at 2985 cm^{-1} was due to –C-H stretching and 1738 cm^{-1} confirms the presence of –C═O (carbonyl) stretching. Weak peak at 1373 cm^{-1} indicate the –C-N stretching and strong peaks at 1235 cm^{-1} and 1044 cm^{-1} confirm –C-O stretching. The shifting, transformation, disappearance and formation of peaks indicate the degradation and generation of functional groups in by-products (Chham et al., 2018; Chidi and Kelvin, 2018; Chikwe and Onojake, 2018; Deeba et al., 2018; Gul and Hameed, 2018; Igwe and Nwamezie, 2018; Thakur et al., 2019).

3.8. Phytotoxicity

The phytotoxicity of biodegraded dye was checked by measuring germination index of *T. foenum graecum* seeds. It was observed that the untreated dye inhibited the seed germination completely (Fig. 8). Seeds exposed to treated dye (100 mg/L) solution showed 81.03% germination and 18.97% seeds were not germinated. The GI and root elongation were enhanced up to 87% when grown in treated dye solution. Phytotoxicity study clearly indicated that dye was detoxified as a results of biodegradation of dye. These observations are in line with previous studies i.e., *R. hirsuta* based Lac was employed for dye degradation and toxicity was reduced up to 80% (Abadulla et al., 2000). In another study, MnP from *G. lucidum* IBL-05 was used for the detoxification of reactive dyes and textile wastewater. The cytotoxicity (haemolytic and brine shrimp lethality) tests before and after treatment revealed that sandal reactive

dyes toxicity was reduced up to (92.29% and textile wastewater showed up to 80% less toxicity in comparison to untreated dye (Bilal and Asgher, 2015b). Similarly, MnP from IBL-05 immobilized on Ca-alginate beads showed significant toxicity reduction of after biodegradation (Bilal and Asgher, 2015a). Same author later, appraise the potential of MnP for textile effluent detoxification. *Allium cepa*, brine shrimp and heamolytic test revealed that the treated samples showed significantly low toxicity versus untreated. The immobilized MnP also revealed excellent catalytic activity for the detoxification of textile effluents (Bilal et al., 2016b). So far, the phytotoxicity study revealed that dyes can be detoxified using biodegradation and under the current scenario of environmental pollution (Iqbal, 2016; Mathubala et al., 2016b; Ogundipe and Babarinde, 2017; Silambarasu et al., 2017; Ukpaka and Wami, 2017; Yildirim and Sasmaz, 2017; Abbas et al., 2018; Abulude et al., 2018; Bomila et al., 2018; Ghezali et al., 2018; Hiwot, 2018; Ibisi and Asoluka, 2018; Mansouri et al., 2018; Palutoglu et al., 2018; Sasmaz et al., 2018), there is a need to adopt the efficient techniques for the de-pollution of water resource.

4. Conclusions

Orange G dye was biodegradation using Lac enzyme (*Plearotus sojar-caju*) in combination with natural redox mediator from *P. granatum* peel extracts. The process variables significantly affected the dye degradation. The dye degradation was reached up to 97% at optimum conditions of Lac activity 279.27 IU/mL, pH 4.5, inoculum size 3 mL, dye initial concentration 100 mg/L, glucose as carbon source and *P. granatum* peel extract. Spectroscopic techniques along with phytotoxicity analysis revealed that the Orange G dye was completely degraded into harmless end products. Results suggest that Lac (*P. sojar-caju*) in the presence of natural redox mediator is highly promising for biodegradation of dyes and could possibly be used for the remediation of dyes in textile wastewater.

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