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Photocatalytic decolourization of congo red dye from aqueous solution by fungal biomass doped copper oxide nanoparticles

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ABSTRACT

*Dye-containing wastewater discharged from several industrial activities is a serious threat to the water bodies around industrial areas, because these toxic, organic or synthetic, dyes can affect plant life and human life indirectly affecting the ecosystem of the environment. The study was aimed to enhance the capacity of the bio sorption process by doping copper oxide on to fungal biomass for dye remediation. The isolated fungal strain is *Aspergillus flavus*. Copper oxide nanoparticles are synthesized by green synthetic pathway and *Coriandrum sativum* was utilized as 0.04 M Copper oxide and fungal biomass.*

Keywords— Biosorption, Dyes, Fungal Biomass, Green Synthetic Pathway

1. INTRODUCTION

The wastes of the colorant category produced from various types of industries might have injurious impacts on microbial inhabitants and may be unhealthful and sometimes even fatal to mammals. These dyes may source for eczema, irritation of skin problem, mutations and cancer. Few chemicals used to produce dye are highly toxic carcinogens or hormonal disrupters (Hunger, 2003).

Congo red is an unsafe dye which not only possesses extremely toxic Properties affecting the cells of mammals but also possesses the power to cause anaphylactic shocks in humans and is potentially carcinogenic. The dye discharged in water bodies without being treated properly disrupts the cycle of life of aquatic animals and plants by obstructing the penetration of light (Zehra et al. 2009). As per their design, dyes are relatively very steady molecules, created to fight against the ruin by light, biological, chemical and other natural modes of degradation (Emad et al., 2008). So alternate approaches come into play.

Biological synthesis of nanoparticles called the green synthesis is gaining momentum in the last decade. This method of nanoparticle synthesis is found to be very simple and cost-effective procedure which is an alternative to the chemical and physical synthesis present. For biological synthesis, plants, bacteria, fungi, algae, yeast all can be utilized (Simkiss and

Wilbur, 1989). Green synthesis is a preferred alternative of synthesis of NPs because it is safer for the biological systems, environmentally friendly and physical and chemical characteristics (Grigore et al., 2016).

2. MATERIALS AND METHODS

2.1. Green synthesis of copper nanoparticles

2.1.1. Preparation of *Coriandrum sativum* extract: *Coriandrum sativum* leaves were collected and washed with tap water followed by distilled water. Leaves were dried in hot air oven at 80°C for 4 hours and ground using mixer grinder as a fine powder. To 10 g of the ground leave powder, 200ml of distilled water was poured and boiled at 80°C for 30 min (Irvani et al. 2013). After 30 min, the extract was allowed to cool and then the plant extract was filtered using filter paper. The freshly prepared extract was used for further synthesis

2.2.2. Preparation of Copper oxide nanoparticles: To 25ml of freshly prepared leaf extract, aqueous CuSO₄ (0.1M) was added and stirred at 320 rpm for 15min at 45°C. After the 20 min the solution was filtered using 0.2 µm Whatman filter paper. Then, the solution was kept hot air oven at 100°C for 3 hours. After drying the particle, it was crushed into powder for further analysis

2.2. Fungal culture used

Fungi isolated from marine source (sedimentation soil, Thrissur beach) were used for remediation of Congo red. The microorganisms were grown in potato dextrose agar (Hi-Media) and incubated at 35°C for 5days and then stored at laboratory conditions at 4°C for further studies. Identification and confirmation of the fungal cultures were done by using – (i) Direct Microscopic Method, (ii) Culture Method and (iii) 18s sequencing.

2.3. Preparation of fungal mass doped with nanoparticles

The fungal spores were subjected to round bead formation by spore suspension method as described below:

2.3.1. Spore suspension Method using Cephadox broth:

After sub-culturing, spores can be harvested from lawn cultures of the organism on potato dextrose agar by flooding

the culture with a sterile loop and dislodging spores from the hyphae with the aid of a sterile glass spreader or loop. And thus spore is filtered and grown in cephalox broth for the production of biomass.

2.3.2. Determine the spore count: The total number of spores can be calculated using a hemacytometer. Spore count is determined by using the following equation: Spores/ml = (n) x 10⁴, where: n = the average cell count per square of the four corner squares counted.

2.3.3 Characterization of harvested biomass doped with nanoparticles: The harvested biomass is characterized by Scanning electron microscopy (SEM).

3. RESULTS

3.1. Synthesis of Copper oxide nanoparticles

Copper oxide nanoparticles was synthesized by adding Coriandrum sativum extract to copper sulphate. During the reaction conditions, the colour of the mixture changed from blue to green and then into a brown precipitate (Fig. 1). This colour change can be attributed to the reduction of the Cu⁺ ions by the plant extract as a result of excitation of Surface Plasmon vibration in metal nanoparticles (Gopinath et al. 2014).

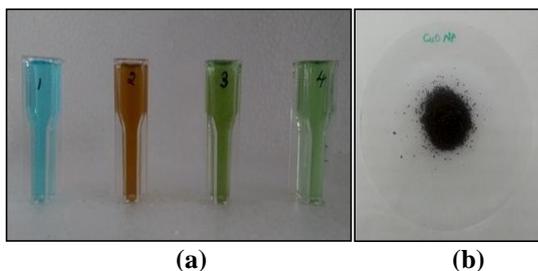


Fig. 1(a): Reduction of copper sulphate by the Coriandrum sativum leaf extract from blue to green. 1. 0.1M CuSO₄, 2. Leaf extract of coriandrum sativum, 3. 0.1 M CuSO₄ + Leaf extract of coriandrum sativum, 4. Filtrate. (b): Synthesized copper nanoparticles.

3.2. Identification of fungal strain

The fungal strain isolated from marine sediment had a morphology resembling that of Aspergillus (Fig 2a). The organism was identified as Aspergillus flavus through 18S rRNA sequencing (Fig 2b).

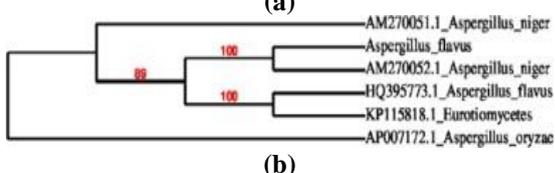


Fig. 2: (a) Fungal growth on PDA media, (b) 18S rRNA sequence map showing Aspergillus flavus

3.3. Preparation of fungal beads

After sub-culturing, spores can be harvested from lawn cultures of the organism on potato dextrose agar by flooding

the culture with a sterile loop and dislodging spores from the hyphae with the aid of a sterile glass spreader or loop. The spores are filtered and grown in cephalox broth for the production of biomass (Droce et al. 2013). Along with 1ml of spores, 0.02, 0.04, 0.06 0.08 0.1mg/100ml of copper oxide is added into cephalox broth and kept in shaker incubator for 4-5days at a temperature of 27°C with 125rpm. According to the spore count, 1ml of the suspension contained 2,080,000 spores.



Fig. 3: Fungal beads doped with copper oxide particles

3.4. Congo red removal by fungal beads

3.4.1. Comparison between the experimental conditions:

Doping of copper particles on to the fungal beads enhances effective dye removal. The experiment was carried out in three different sets under respective conditions (0.04M CuO+BM, CuO NPs alone, Biomass alone). It was found that 0.04M CuO+BM adsorbed dye up to 98%, whereas NP adsorbed 88.56% and biomass adsorbed only 76.88%. (Asses et al. 2018) reported a 97% Congo red removal with Aspergillus niger incubated at pH 5, in presence of 200 mg/L of dye during 6 days at 28°C and under 120 to 150 rpm shaking speed.

3.4.2. Congo red removal with 0.04M CuO+Biomass:

When different concentrations of dye (20ppm, 40ppm, 60ppm, 80ppm, and 100ppm) are treated with 0.04CuO+BM, it was found that the degradation of dye in 20ppm showed almost 98% of removal in 60min. As the concentration of dye increased, it was found that the degradation percentage decreased and thus a maximum concentration of dye (100 ppm) showed 40% degradation in 60min.

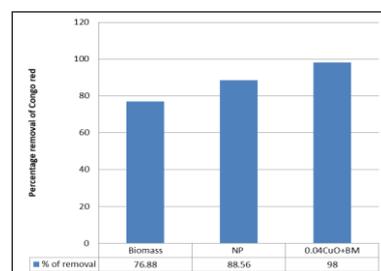


Fig. 4: Congo red removal under different experimental conditions.

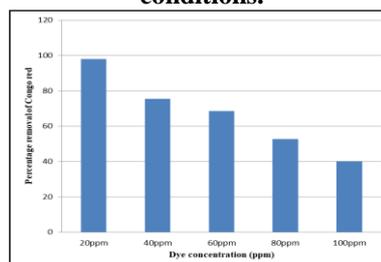


Fig. 5: Congo red removal at different initial dye concentrations.

3.4.3 Congo red dye removal with 0.04M CuO+BM under solar irradiation:

To enhance the degradation potential, the reaction was carried out under sunlight. Application of 0.04M CuO+BM showed enhanced effective degradation of Congo

red dye under solar irradiation by a process called photocatalytic activity. A highest decolorization rate of 99% was seen in this case. (Ayodhya et al. 2016) demonstrated notable photocatalytic activity of copper sulfide (CuS) nanoparticles in solar, visible and UV lights.



Fig. 6: Decolourization of Congo red solution at 0mins and 60mins

4. CONCLUSION

This *Aspergillus flavus* contains the amino groups could be the major biosorption sites while the carboxylic acid, phosphate groups and the lipid fraction may not form the binding sites, on the other hand, these fungal biomasses could act as an electrostatic attraction which binds to dye molecules. Besides, CuO NPs has been proven to be possessing excellent photocatalytic activity. Consequently, the longer the time exposure, the better the photocatalytic degradation on Congo red tested. Application of 0.04M CuO+BM showed enhanced effective degradation of Congo red dye under solar irradiation by a process called photocatalytic activity. Thus, 0.04M CuO+Biomass could be used as a biosorbent for the effective removal of Congo red from dye wastewater in terms of high biosorption capacity, available in natural and abundant with low cost.

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