

Eco-friendly synthesis of Zinc nano particles for dyes decolorization using catalpa bignonioides broth

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Abstract

Eco-friendly techniques are better ways for better society in the nearby future. As the Industrial revolution in India has broadened along with industrial waste disposal problem, it is now time to concentrate on alternate methods to reduce the industrial waste disposal problem. Present experimentation is about the treatment of synthetic alizarin dye removal using green technology. *Catalpa Bignonioides* plant was considered as promising plant for preparation of zinc nano particles broth. The effect of operating parameters like contact time, pH of the solution, concentration, amount of nanoparticles suspension and temperature. Characterization techniques involved comprises of XRD and FTIR. The maximum removal of alizarin red dye was obtained at a contact time of 30 hrs. The optimum pH obtained was at 5. The XRD presented various compounds matching with their corresponding 2θ and d -values. The entire process of alizarin red dye removal was dependent on temperature.

Keywords: Dyes, pH, Concentration, FTIR, XRD, Temperature.

1. Introduction

Economy of any nation depends on its Industrial advancement. Present scenario of Indian economy is better than many advanced countries but simultaneously industrial waste disposal is also developed [01]. Effluents from various industries like textiles, rubber, paper, plastics, cosmetics, etc are a big threat to humans, aquatic life and plants [02–08]. Though several physical or chemical processes are available like chemical adsorption, reverse osmosis, ion exchange, coordination, complexation, chelation, microprecipitation etc, but sludge remains and cost effectiveness are again adversing the problem [09–13]. In order to reduce this problem, economically viable processes must be developed [14]. The present experimentation consists of a simple green technology using nano particles for alizarin red dye removal from waste waters.

2. Materials and Methods

The present experimentation is carried out in batch process, for removal of dyes (Eosin Yellow–EY, Indigo Caramine–IC, Cresol Red–CR, Alizarine red–AR) from aqueous solutions by using *Catalpa Bignonioides* leaves with zinc oxide nano particles.

Analytical grade chemicals were used for experimentation and need no further purification. Double distilled water is used to prepare all stock and synthetic

solutions. From a stock solution containing 1000 mg of dyes in 1.0 litre, the synthetic solutions of dyes were made. By addition of 0.1 M HCl and 0.1 M NaOH solutions the pH of dyes solutions were adjusted to the desired value.



Fig.1 Dyes

2.1 Preparation of *Catalpa Bignonioides* broth

In this process 10 gm of fresh and cleaned leaves of CB are taken in a magnetic stirrer and to this 110 ml of distilled water is added and it is heated at 60°C for 10 min. After that the solution is filtered in 250 ml conical flask using whatmann's filter paper and it is kept aside for further process. The broth obtained is in pale yellow colour.

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Fig.2 Catalpa Bignonioides Leaves

2.2 Preparation of Nano Particles

In this process 10 ml of broth solution is taken and to that 5gms of zinc nitrate hexahydrate is added in a 250 ml conical flask and is kept in an orbital shaker for 24 Hrs in order to obtain nano particles. The nano particles formation is noticed when the pale yellow color is changed to dark brown color. This solution is used for various dyes degradation process of different concentrations and different dosages.



Fig. 3 Nano particles solutions for Catalpa Bignonioides

2.3 Characterization Studies

Characteristic studies were carried by XRD and FTIR. The prepared broth was centrifuged at 10,000 rpm for 2 hrs and obtained a thick paste which was dried in hot air over

and further send to advanced analytical laboratory for FTIR and XRD study.

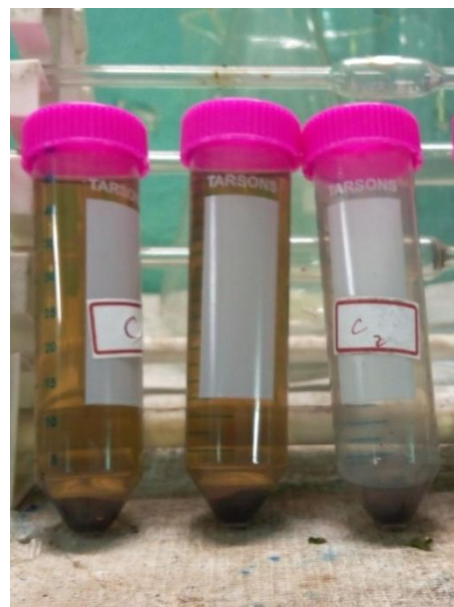


Fig. 4 Centrifuge samples for drying and Characterization analysis (FTIR & XRD)

3. Results and Discussion

3.1 Characterization

3.1.1 Fourier transform infrared spectroscopy (FTIR) Spectrum

The FTIR spectrum of zinc oxide nanoparticles is shown in Figure 5 with the broth catalpa bignonioides [15, 16]. It is an important method to determine the formation of metal nanoparticles in colloidal solution. FTIR spectrum was examined to identify the possible biomolecules responsible for capping and efficient stabilization of the biosynthesized Ag nanoparticles, which can help in further functionalization with other molecules for various applications in future.

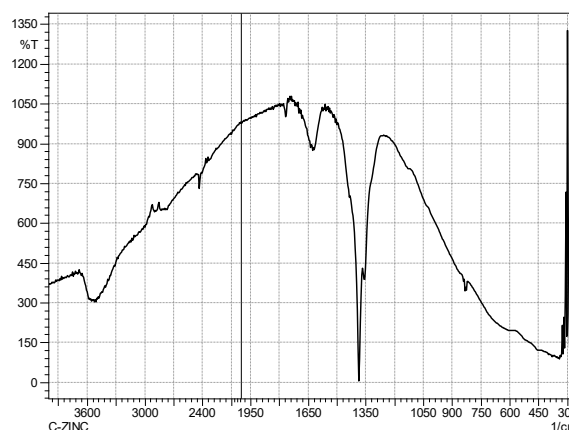


Fig.5 FTIR Spectrum of catalpa bignonioides zinc oxide nano particles

3.1.2 X-Ray Diffraction

Fig. 6 shows the XRD for the zinc oxide nanoparticles using catalpa bignonioides broth. The peak at 2θ values of 0.5345, 0.4685, 0.9285, 0.4852 and 0.8135 confirms the presence of compounds like H₃NO₅Zn, Fe_{1.04}Hli, Cr, Ag₂CSl₃ and Ge_{0.16}. Their corresponding and representing d-values are 8.2149, 2.9308, 2.0303, 2.3129 and 2.1825 respectively [17, 18].

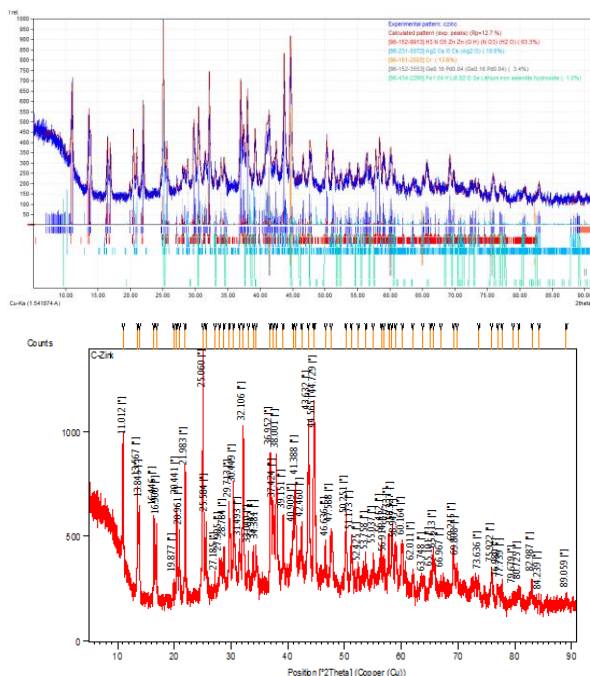


Fig. 6 XRD pattern of Catalpa Bignonioides zinc oxide nano particles

3.2 Equilibrium studies

3.2.1 Effect of contact time

The Decolourization of Dyes (AR, IC, CR, EY) was studied as a function of agitation time at Room temperature. 20 ml of 20 mg/L Dye solution was taken with 5 ml of Catalpa Bignonioides broth zinc nano solution (cb-zn-nps) at different time intervals ranging from 1 min to 72 Hrs.

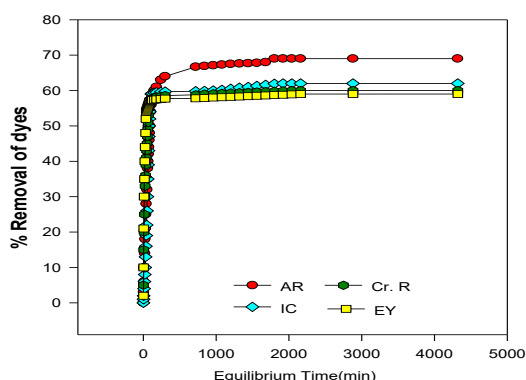


Fig.7 Effect of contact time

At the start, the ions adsorbed and occupied selectively the active sites on the cb-zn-nps solution [19, 20]. As the initial contact time increased the available free sites on the nano particles were filled and it continued till the rate of sorption gradually exhausted and reached plateau. The result obtained is shown in figure 7. As a result of the experiment, the highest % Removed for the Dyes (AR, IC, CR, EY) was 69%, 62%, 60%, 59% at the time of 30 Hrs, 32 Hrs, 34 Hrs, 36Hrs

3.2.2 Effect of pH

The pH parameter has been identified as one of the most important parameter that is effective on Dye decolourization. In order to find the effect of pH on Dye Decolourization using the cb-zn-nps, experiments have been carried out at various initial pH values and results are given in figure 8. The removal was increased from 38 % to 69% as pH was increased from 2 to 8, The pH is varied for every dye used with the broth solution catalpa bignonioides whereas further increase in pH had a negative effect. The maximum % removal was found to be 69 % at pH 5 for AR Dye. The maximum % removal was found to be 66% at pH 6 for IC dye. The maximum % removal was found to be 65% at pH 4 for CR dye. The maximum % removal was found to be 64% at pH 7 for EY dye. Therefore, the remaining all experiments were carried out at this pH value [21, 22].

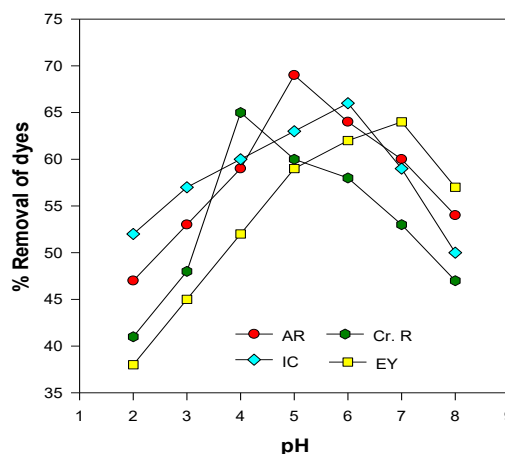


Fig. 8 Effect of pH

3.2.3 Effect of concentration

The percentage Removal of dyes at various initial concentrations is depicted in Fig. 9. At concentration of Dye solution (20 mg/L), maximum %removal is obtained and is different for every Dye using Broth catalpa bignonioides and on further increase in concentration (200 mg/L), %removal has been decreased. The capacity of % removal is increased up to a concentration of 20 mg/L. The interaction between catalpa bignonioides broth and the dyes were high initially and later subsided. The maximum removal of AR Dye is 73%. The maximum

removal of IC dye is 71%. The maximum removal of CR dye is 70%. The maximum removal of EY is 72 % [23, 24].

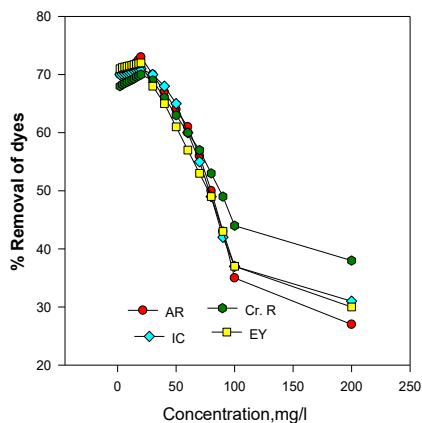


Fig.9 Effect of concentration

3.2.4 Effect of dosage

The variation of % removal of Dyes (AR, IC, CR, EY) was studied using different dosages of the broth catalpa bignonioides. Results from the fig 10 showed that % removal of Dyes (AR, IC, CR, EY) increased and uptake decreased with increase in dosage. The maximum % removal is attained at 10 ml and was almost constant at higher dosages. Therefore, the optimum dosage was selected as 10 ml for further experiments. The maximum % removal of AR dye is 80%. The maximum % removal of IC dye is 78%. The maximum % removal of CR dye is 77%. The maximum % removal of EY dye is 76% [25, 26].

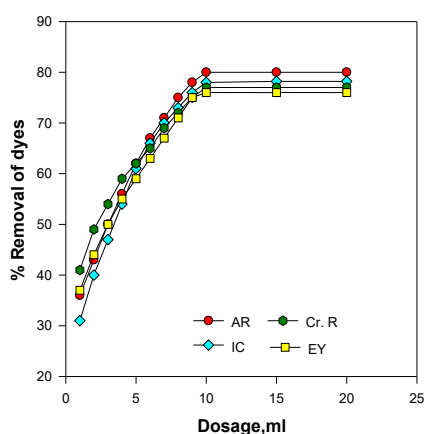


Fig.10 Effect of Dosage

3.2.5 Effect of temperature

The dependence of temperature on the % removal of dyes is investigated at different temperatures as given in fig. 11. Results from fig 11 showed that %removal of Dyes (AR, IC, CR, EY) increased from 78% to 83% with increase in temperature from 283 K to 323 K. This indicates that the % removal is an endothermic process. The increase in removal with temperature may be the results of either

increase in the number of available active surface area available for interaction on the Dyes. The maximum % removal of AR dye is 83 %, The maximum % removal of IC dye is 81%, the maximum % removal of CR dye is 79 %, The maximum % removal of EY is 80 % [27, 28].

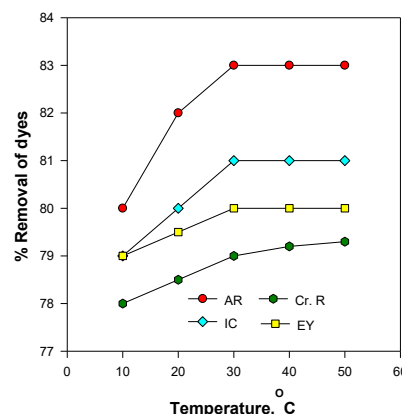


Fig.11 Effect of Temperature

Conclusions

The analysis of the experimental data results in the following conclusions:

- 1) The maximum dye decolourization of AR dye onto Catalpa Bignonioides leaves with Zinc oxide-nano solution observed when the processing parameters are set as: $t=30$ Hrs, $pH=5$, $w=10$ ml, $Co=20$ mg/L and $T=303$ K is 83%.
- 2) The maximum dye decolourization of IC dye onto Catalpa Bignonioides leaves with Zinc oxide -nano solution observed when the processing parameters are set as: $t=32$ Hrs, $pH=6$, $w=10$ ml, $Co=20$ mg/L and $T=303$ K is 81%.
- 3) The maximum dye decolourization of CR dye onto Catalpa Bignonioides leaves with Zinc oxide -nano solution observed when the processing parameters are set as: $t=34$ Hrs, $pH=4$, $w=10$ ml, $Co=20$ mg/L and $T=303$ K is 79%.
- 4) The maximum dye decolourization of EY dye onto Catalpa Bignonioides leaves with Zinc oxide -nano solution observed when the processing parameters are set as: $t=36$ Hrs, $pH=7$, $w=10$ ml, $Co=20$ mg/L and $T=303$ K is 80%.

With the above conclusions the authors confirm that the above mentioned leaves are capable of removing dyes.

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References

- [1]. Kayode Adesina Adegoke, Olugbenga Solomon Bello, "Dye sequestration using agricultural wastes as adsorbents", *Water Resources and Industry*, 12 (2015) 8–24.
- [2]. P. Nigam, G. Armour, I.M. Banat, D. Singh, R. Marchant, Physical removal of textile dyes from effluents and solid-state fermentation of dye-adsorbed agricultural residues, *Biores. Technol.* 72 (2000) 219–226.
- [3]. V.J.P. Poots, J.J. McKay, The removal of acid dye from effluent using natural adsorbents-peat, *Water Res.* 10 (1976) 1061–1066.
- [4]. G. McKay, Waste color removal from textile effluents, *Am. Dyes Rep.* 68 (1979) 29–36.
- [5]. G. Mishra, M. Tripathy, A critical review of the treatment for decolorization of textile effluent, *Colourage* 40 (1993) 35–38.
- [6]. N. Willmott, J. Guthrie, G. Nelson, The biotechnology approach to colour removal from textile effluent, *J. Soc. Dyers Colour.* 114 (1998) 38–41.
- [7]. T. Robinson, G. McMullan, R. Marchant, P. Nigam, Remediation of dyes in textile effluent: a critical review on current treatment technologies with a proposed alternative, *Biores. Technol.* 77 (2001) 247–255.
- [8]. C. Moran, M.E. Hall, R.C. Howell, Effects of sewage treatment on textile effluent, *J. Soc. Dyers Colour.* 113 (1997) 272–274.
- [9]. Chattopadhyaya G, Macdonald DG, Bakhshi NN, Mohammadzadeh JSS, Dalai AK. Adsorptive removal of sulfur dioxide by saskatchewan lignite and its derivatives. *Fuel* 2006;85:1803–10.
- [10]. Bashkova S, Bagreev A, Locke DC, Bandosz TJ. Adsorption of SO₂ on sewage sludge-derived materials. *Environ Sci Technol* 2001;35:3263–9.
- [11]. Xu L, Guo J, Jin F, Zeng H. Removal of SO₂ from O₂-containing flue gas by activated carbon fiber (ACF) impregnated with NH₃. *Chemosphere* 2006;62: 823–6.
- [12]. Mangun CL, Debarr JA, Economy J. Adsorption of sulfur dioxide on ammoniatreated activated carbon fibers. *Carbon* 2001;39:1689–96.
- [13]. Gaur V, Asthana R, Verma N. Removal of SO₂ by activated carbon fibers in the presence of O₂ and H₂O. *Carbon* 2006;44:46–60.
- [14]. J.W. Lee, S.-P. Choi, R. Thiruvengatchari, W.-G. Shim, H. Moon, Evaluation of the performance of adsorption and coagulation processes for the maximum removal of reactive dyes, *J. Dyes Pigment.* 69 (2006) 196–203.
- [15]. Zhao, Yixin, Xiaofeng Qiu, and Clemens Burda. "The effects of sintering on the photocatalytic activity of N-doped TiO₂ nanoparticles." *Chemistry of Materials* 20.8 (2008): 2629-2636.
- [16]. Sadighi, A., and M. A. Faramarzi. "Congo red decolorization by immobilized laccase through chitosan nanoparticles on the glass beads." *Journal of the Taiwan Institute of Chemical Engineers* 44.2 (2013): 156-162.
- [17]. Anandan, S., et al. "Effect of loaded silver nanoparticles on TiO₂ for photocatalytic degradation of Acid Red 88." *Solar Energy Materials and Solar Cells* 92.8 (2008): 929-937.
- [18]. Feng, Jiyun, et al. "Degradation of azo-dye orange II by a photoassisted Fenton reaction using a novel composite of iron oxide and silicate nanoparticles as a catalyst." *Industrial & engineering chemistry research* 42.10 (2003): 2058-2066.
- [19]. Salehi, Raziye, et al. "Novel biocompatible composite (chitosan–zinc oxide nanoparticle): preparation, characterization and dye adsorption properties." *Colloids and Surfaces B: Biointerfaces* 80.1 (2010): 86-93.
- [20]. A. Ozcan, A.S. Ozcan, "Adsorption of Acid Red 57 from aqueous solutions onto surfactant-modified sepiolite", *J. Hazard. Mater.*, 125 (2005), pp. 252–259
- [21]. Fan, Jing, et al. "Rapid decolorization of azo dye methyl orange in aqueous solution by nanoscale zerovalent iron particles." *Journal of Hazardous Materials* 166.2 (2009): 904-910.
- [22]. Pouretedal, Hamid Reza, et al. "Nanoparticles of zinc sulfide doped with manganese, nickel and copper as nanophotocatalyst in the degradation of organic dyes." *Journal of Hazardous Materials* 162.2 (2009): 674-681.
- [23]. Anandan, S., et al. "Effect of loaded silver nanoparticles on TiO₂ for photocatalytic degradation of Acid Red 88." *Solar Energy Materials and Solar Cells* 92.8 (2008): 929-937.
- [24]. Shu, Hung-Yee, et al. "Using resin supported nano zero-valent iron particles for decoloration of Acid Blue 113 azo dye solution." *Journal of Hazardous Materials* 184.1 (2010): 499-505.
- [25]. He, Yang, et al. "The comparative study on the rapid decolorization of azo, anthraquinone and triphenylmethane dyes by zero-valent iron." *Chemical Engineering Journal* 179 (2012): 8-18.
- [26]. Li, Fangfei, et al. "Photodegradation of an azo dye using immobilized nanoparticles of TiO₂ supported by natural porous mineral." *Journal of Hazardous Materials* 152.3 (2008): 1037-1044.
- [27]. Njagi, Eric C., et al. "Biosynthesis of iron and silver nanoparticles at room temperature using aqueous sorghum bran extracts." *Langmuir* 27.1 (2010): 264-271.
- [28]. Shahwan, Talal, et al. "Green synthesis of iron nanoparticles and their application as a Fenton-like catalyst for the degradation of aqueous cationic and anionic dyes." *Chemical Engineering Journal* 172.1 (2011): 258-266.