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# Congo Red Dye Color Removal Studies by Using The Biochar Of Arjuna Seeds and Ozone Treatment: Characterization and Equilibrium Studies

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***Abstract*—The present study investigates the adsorption and oxidative degradation of Congo Red dye using Arjuna seed biochar as a low-cost and eco-friendly adsorbent. Biochar was prepared through pyrolysis of Arjuna seeds under nitrogen atmosphere at 600 °C and characterized using FTIR, BET, and XRD analyses. Batch and continuous adsorption experiments were performed to evaluate the influence of operational parameters such as adsorbent dosage, temperature, pH, and initial dye concentration on dye removal efficiency. Experimental results demonstrated that dye removal efficiency increased with increase in adsorbent dosage and temperature, while acidic pH conditions favored maximum adsorption. Continuous packed bed adsorption studies showed a maximum decolorization efficiency of 98.32% for 50 ppm Congo Red dye solution. Adsorption equilibrium data were analyzed using Langmuir and Freundlich isotherm models, indicating favorable adsorption behavior of the prepared biochar. Thermodynamic analysis confirmed that the adsorption process was spontaneous and endothermic in nature. Regeneration studies revealed satisfactory reusability of the adsorbent for multiple adsorption-desorption cycles. Furthermore, ozone-assisted oxidation significantly enhanced dye degradation and reduced wastewater toxicity due to the strong oxidative nature of ozone. The combined adsorption and ozonation treatment process demonstrated efficient removal of Congo Red dye with reasonable energy consumption, suggesting its potential application for sustainable treatment of textile wastewater.**

***Keywords*— Adsorption, Arjuna seed adsorbent, Congo Red dye, Ozone oxidation, Regeneration, Textile wastewater treatment**

## I. INTRODUCTION

Rapid industrialization and extensive utilization of synthetic dyes in textile, paper, leather, rubber, cosmetic, and printing industries have generated serious environmental concerns due to the discharge of dye-containing wastewater into aquatic ecosystems [1,2]. Synthetic dyes are generally stable aromatic compounds specifically designed to resist degradation under environmental conditions, making their removal from wastewater highly challenging [3]. Among various industrial dyes, Congo Red (CR) is a benzidine-based anionic azo dye widely used because of its excellent color stability and strong binding properties [7]. However, untreated discharge of Congo Red dye into water bodies causes severe environmental pollution and may lead to toxic, mutagenic, and carcinogenic effects on living organisms [14].

Various conventional treatment techniques such as coagulation, flocculation, membrane separation, chemical precipitation, oxidation, and biological treatment have been employed for dye removal from wastewater [2,5]. Nevertheless, these methods often suffer from limitations including incomplete degradation, sludge generation, high operational cost, and low treatment efficiency [6]. Among the available treatment technologies, adsorption has emerged as one of the most effective and economical methods due to its operational simplicity, high efficiency, flexibility, and ease of regeneration [10,11].

Recently, the utilization of low-cost biochar and agricultural waste-derived adsorbents has attracted considerable attention for dye removal applications [7,10]. Biochar materials possess porous structures, high surface area, and abundant surface functional groups which enhance adsorption efficiency toward dye molecules. Arjuna seed biochar can therefore serve as an efficient adsorbent for Congo Red dye removal due to its favorable physicochemical properties. The adsorption behavior is significantly affected by several operating parameters including adsorbent dosage, pH, temperature, and initial dye concentration [12].

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In addition to adsorption, advanced oxidation processes have shown promising potential for degradation and mineralization of dye molecules [14,15]. Ozone treatment is considered one of the most powerful oxidation techniques because of its high oxidation potential and ability to convert complex dye molecules into simpler and less toxic compounds [16]. Ozonation also improves biodegradability and reduces phytotoxicity of treated wastewater [17,18]. The combined application of adsorption and ozone oxidation can therefore provide an efficient and sustainable treatment approach for textile dye wastewater remediation.

The present study investigates the adsorption and oxidative degradation of Congo Red dye using Arjuna seed biochar under batch and continuous operating conditions. The effects of various process parameters such as adsorbent dosage, temperature, pH, and dye concentration were evaluated. Langmuir and Freundlich adsorption isotherm models were analyzed to study adsorption equilibrium characteristics. Thermodynamic studies, regeneration analysis, and ozone-assisted degradation experiments were also performed to evaluate the feasibility and efficiency of the proposed treatment system.

## II. MATERIALS AND METHODS

### *A. Preparation of Adsorbent*

The seeds of arjuna tree (*Terminalia arjuna*) were collected from the campus of IIT BHU. The seeds were properly dried and then pyrolysed at 600 $^0$C temperature level in nitrogen environment. Pyrolysis produces the solid carbon content known as biochar , liquid (bio oil) and gaseous residue. Proximate analysis data shows that the adsorbent has 52.35% fixed carbon, 22.53% volatile matter, 10.54% moisture content and 14.58% ash content.

### *B. Reagents*

The major reagents used during the study were Congo Red dye ($C_{32}H_{22}N_6Na_2O_6S_2$), sodium hydroxide (NaOH), and hydrochloric acid (HCl). All stock and diluted solutions were prepared using distilled water.

### *C. Instrumentation*

1. UV-Visible Spectrophotometer: Dye concentration and absorbance measurements were carried out using UV-Visible Spectrophotometer (ELICO SL 159). The instrument operates on Beer-Lambert law relating absorbance with concentration and path length. Congo Red dye showed maximum absorbance at 500 nm and all absorbance measurements were performed at this wavelength.
2. pH Meter: The pH of dye solutions was measured using a pH meter (Labman Scientific Instruments). The instrument measures hydrogen ion concentration difference and converts the generated emf into pH values.
3. FTIR Analysis: Fourier Transform Infrared Spectroscopy (FTIR) was employed for functional group characterization of the adsorbent material. The technique enables simultaneous acquisition of high-resolution spectra over a wide spectral range.
4. XRD Analysis: X-ray diffraction (Rigaku Ultima IV 2036E202) was used to investigate the crystalline structure and surface topography of the adsorbent material based on Bragg's law.
5. BET Analysis: Surface area and porosity analysis of biochar samples were carried out using BET analyzer (Micromeritics ASAP 2020). Nitrogen adsorption-desorption technique at 77 K was employed for determination of surface area, pore volume, and pore size distribution.

### *D. Adsorption Experiments*

1. Batch Adsorption Study: Batch adsorption experiments were conducted using Congo Red dye solutions of 50, 100, 150, and 200 ppm concentrations. Different adsorbent dosages (1–2 g) were added into 100 mL Erlenmeyer flasks containing dye solutions. The pH of the solutions was adjusted within the range of 2–10 using 0.1 N NaOH and 0.1 N HCl solutions. Experiments were performed in an orbital shaker incubator maintained at 34 ± 1 °C and 120 rpm. After equilibrium, samples were centrifuged at 5000 rpm for 6 min. Dye uptake capacity was calculated using adsorption equilibrium relationship.

$$q = \frac{Ci\ Vi - Cf\ Vf}{W}$$

where q is dye uptake (mg/g), Ci and Cf are initial and final dye concentrations (mg/L), Vi and Vf are initial and final solution volumes, and W is the amount of adsorbent used.

2. Continuous Study: Continuous adsorption experiments were carried out using a packed bed column packed with biochar adsorbent. Dye solutions were introduced using a peristaltic pump at a flow rate of 100 cc/h to enhance residence time. Spargers were employed for drop-wise distribution of dye solution over the adsorbent bed. Various

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pH conditions were investigated to determine optimum operating conditions for maximum color removal.

*E. Regeneration of Adsorbent*

Regeneration of adsorbent was performed to restore adsorption capacity after saturation. The packed bed was washed thoroughly with distilled water and treated using 0.1 N NaOH and 0.1 N HCl solutions. After regeneration, the adsorbent was dried in a vacuum dryer at 50 °C for 24 h. Various regeneration techniques such as microwave, steam, vacuum, ultrasound, and oxidative regeneration are also reported in the literature.

*F. Ozone Treatment*

Ozone treatment was employed as an advanced oxidation process for enhanced degradation of Congo Red dye molecules. Ozone oxidizes complex dye molecules into smaller and less toxic compounds. Ozonation experiments were conducted in a semi-batch reactor equipped with spargers for effective gas-liquid contact. The ozone generation unit (Faraday Ozone, SL Oxygen Concentrator OXY-5L) was operated with oxygen flow rates between 2–4 L/min and ozone output levels ranging from 10–50%. The effects of ozone dosage, degradation efficiency, and energy consumption were evaluated during the treatment process.

## III. RESULTS AND DISCUSSION

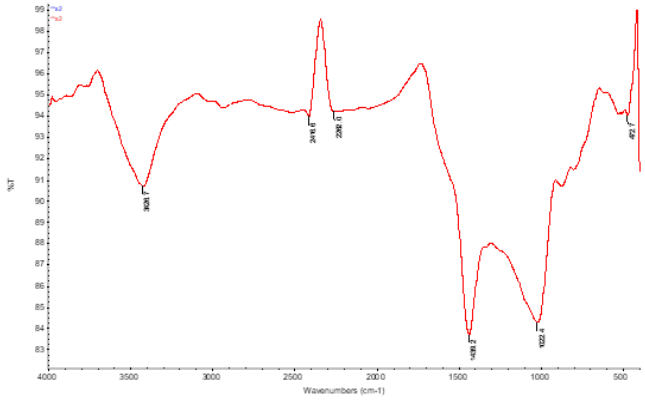
*A. Characterization*

FTIR spectra of Arjuna seed biochar before and after adsorption are presented in Fig. 1 and Fig. 2. The spectra indicate the presence of various functional groups responsible for adsorption of Congo Red dye molecules. The pristine biochar exhibited a broad absorption peak at 3426.7 $cm^{-1}$ corresponding to –OH stretching vibration. Peaks observed at 2416.6 $cm^{-1}$, 2262.0 $cm^{-1}$, 1439.2 $cm^{-1}$, and 1022.4 $cm^{-1}$ were attributed to =C–H, nitrile (C≡N), –C=C–, and C–F functional groups, respectively.

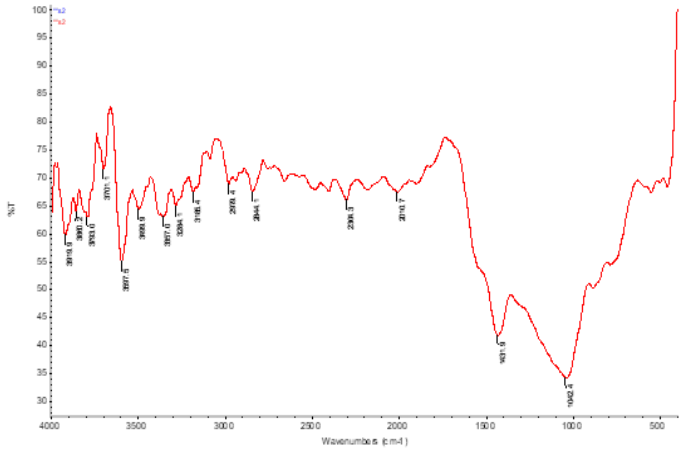
The FTIR spectra of dye-loaded biochar obtained after batch adsorption experiments showed significant peak shifting, indicating interaction between dye molecules and active adsorption sites. Peaks observed at 3697.5 $cm^{-1}$ represented –OH stretching, while peaks at 3499.9, 3357.0, and 3284.1 $cm^{-1}$ indicated amide groups and N–H stretching vibrations. Peaks at 2979.4 $cm^{-1}$ and 2844.1 $cm^{-1}$ corresponded to C–H stretching vibrations. Additional peaks at 1431.9 $cm^{-1}$ and 1042.4 $cm^{-1}$ confirmed the presence of ester and C–F functional groups.

FTIR analysis of biochar obtained after continuous adsorption experiments is shown in Fig. 3. The peak at 3410.3 $cm^{-1}$ corresponds to alcohol or phenolic –OH groups, while the peak at 2265.4 $cm^{-1}$ indicates nitrile and alkynyl stretching vibrations. Peaks observed at 1431.7 $cm^{-1}$, 1073.0 $cm^{-1}$, and 776.4 $cm^{-1}$ correspond to ester groups, C–F stretching, and aromatic C–H bonding, respectively.

BET surface area and porosity analysis revealed that the prepared biochar possessed a surface area of 770.688 $m^2/g$, pore volume of 0.19792 $cm^3/g$, and average pore diameter of 2.8733 nm. The cumulative adsorption surface area between pore diameters of 1.7–300 nm was found to be 564.9286 $m^2/g$, while cumulative desorption surface area was 565.7947 $m^2/g$. Proximate analysis indicated 52.35% fixed carbon, 22.53% volatile matter, 10.54% moisture content, and 14.58% ash content.




(a)




(b)

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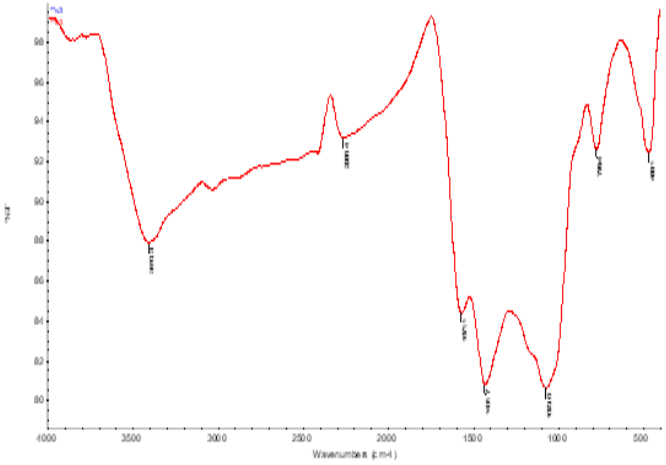


(c)

Fig.1. (a): FTIR spectra before adsorption (b): FTIR spectra after adsorption (Batch Mode Operation) and (c): FTIR spectra after adsorption (Continuous mode operation)

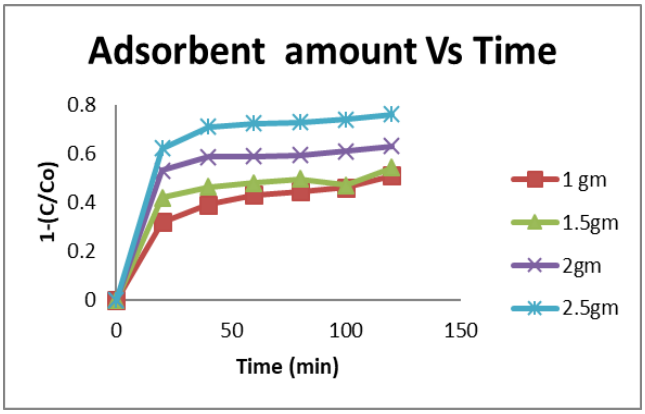
*B. Effect of Various Parameters on Congo Red Dye Removal*

1. Batch Adsorption Experiments

a) Effect of Adsorbent Dose: The effect of adsorbent dosage on dye removal efficiency was investigated using powdered biochar under batch operating conditions. The experimental results demonstrated that dye removal efficiency increased significantly with increase in adsorbent dosage due to the availability of larger active surface area and adsorption sites. Maximum color removal efficiency of 76.1% was achieved using 2.5 g adsorbent per 100 mL dye solution (Fig. 2a). Similar observations were also reported by Bharti et al., where methylene blue dye removal increased from 51.9% to 94.56% with increase in biochar dosage.

b) Effect of Temperature: The effect of temperature on Congo Red dye adsorption was investigated at 300, 310, 315, and 320 K. Dye removal efficiency increased with increasing temperature, indicating endothermic adsorption behavior. Maximum removal efficiency of 78.2% was observed at 320 K (Fig. 2b). Similar adsorption enhancement with increasing temperature was also reported by Bulut and Aydin during methylene blue adsorption using wheat shells.

c) Effect of Solution pH: The pH of dye solution significantly influenced Congo Red adsorption efficiency. As the pH increased from 4.3 to 9.1, color removal efficiency varied from 52% to 83% (Fig. 2c). The enhanced removal at lower pH conditions may be attributed to increased electrostatic attraction between positively charged adsorbent surface and negatively charged dye molecules.

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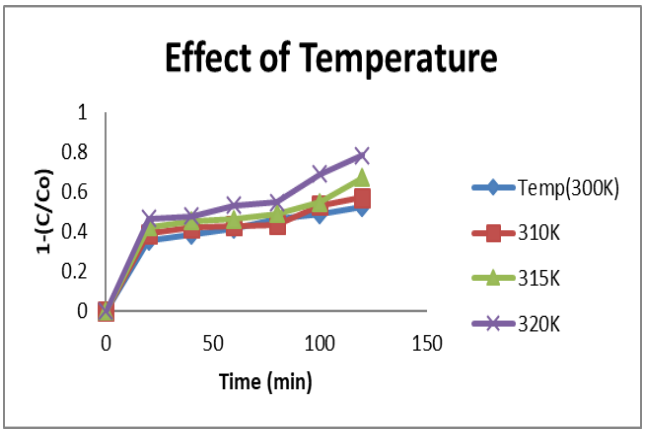

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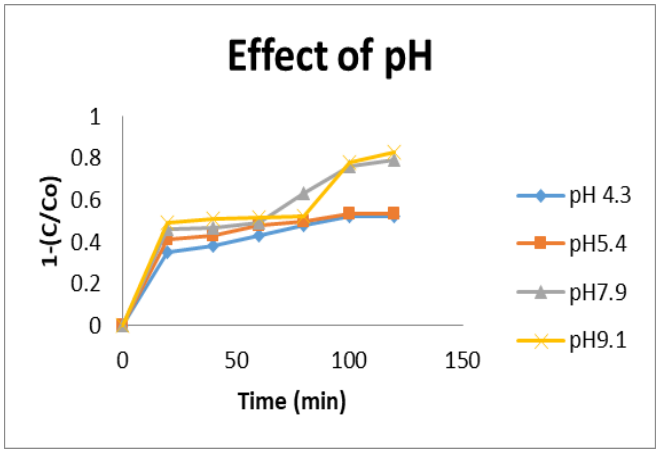



(b)

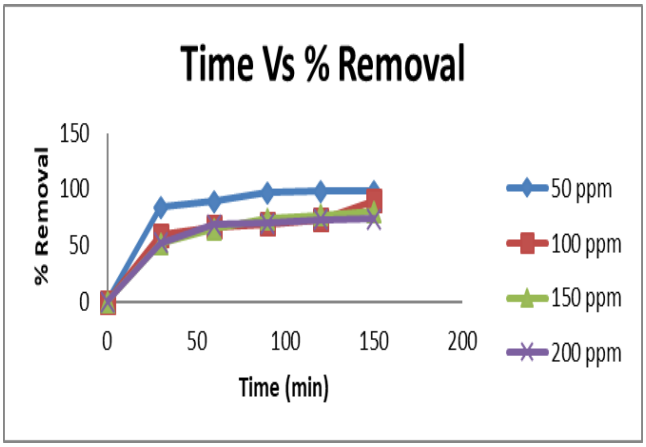
(c)

Fig 2: Effects of various operating parameters on dye color removal (a) amount (b) Temperature and (c) pH.

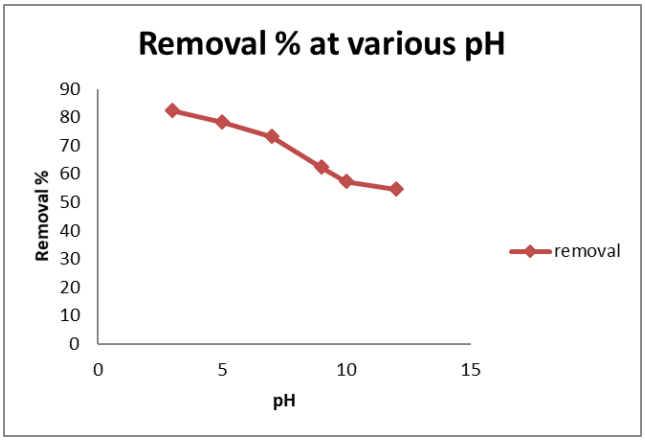
2. Continuous Adsorption Experiments

a) Effect of Initial Dye Concentration: Continuous adsorption experiments were performed in a packed bed column packed with Arjuna seed biochar. The dye solution was supplied at a flow rate of 100 cc/h using a peristaltic pump. Experimental results showed that 50 ppm dye concentration exhibited maximum decolorization efficiency of 98.32% after 150 min of operation (Fig. 3a). Higher dye concentrations showed comparatively lower adsorption efficiency due to agglomeration and micelle formation of dye molecules.

b) Effect of pH: The influence of pH on continuous dye removal efficiency indicated that adsorption efficiency decreased with increase in pH. Maximum degradation efficiency of 82.42% was observed at pH 3, while only 54.56% removal was obtained at pH 12 (Fig. 3b). At lower pH conditions, increased hydrogen ion concentration generated positively charged adsorbent surfaces which enhanced electrostatic attraction toward anionic Congo Red dye molecules.




(a)




(b)

Fig 3: Effect of (a) Initial concentration and (b) pH level on color removal.

*C. Equilibrium Isotherm Studies*

1. Langmuir Isotherm: Langmuir isotherm assumes monolayer adsorption over homogeneous adsorption sites without interaction between adsorbed molecules.

$$\frac{Ce}{qe} = \frac{1}{b\,Qm} + \frac{Ce}{Qm}$$

where $q_e$ and $Q_m$ represent equilibrium adsorption capacity and maximum monolayer adsorption capacity, respectively. The values of $Q_m$ and b obtained from curve fitting were 51.282 mg/g and 0.095965, respectively (Fig. 4a).

2. Freundlich Isotherm: Freundlich isotherm describes heterogeneous surface adsorption behavior and is represented by:

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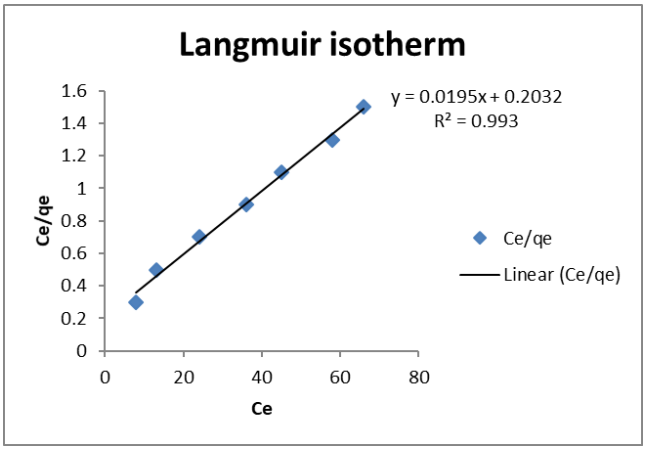
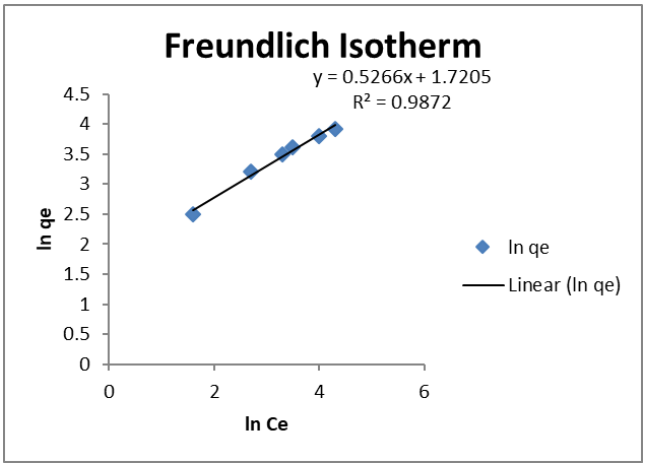
$$\log q_e = \log K_f + \frac{1}{n} \log C_e$$

where Kf and n are Freundlich constants associated with adsorption capacity and adsorption intensity, respectively. The values of Kf and n obtained from experimental data were 5.5873 and 1.8989, respectively (Fig. 4b).




(a)




(b)

Fig 4: (a) Langmuir and (b) Freundlich isotherm for CR adsorption onto AS.

*D. Adsorption Thermodynamics*

Thermodynamic analysis was performed to investigate spontaneity and feasibility of the adsorption process. Gibbs free energy change was calculated using:

$\Delta G^0 = -2.303RT \log K_d$ and $K_d = \frac{qe}{Ce}$

The thermodynamic relationship between Gibbs free energy, enthalpy, and entropy is given by:

$\Delta G^0 = \Delta H^0 - T\,\Delta S^0$

The calculated equilibrium constant $K_d$ was 4.0255, while enthalpy change ($\Delta H^0$) and entropy change ($\Delta S^0$) were 29.47 kJ/mol and 106.97 J/K, respectively, (Fig. 5) as elucidated in Table I. Negative Gibbs free energy values confirmed spontaneous adsorption behavior.

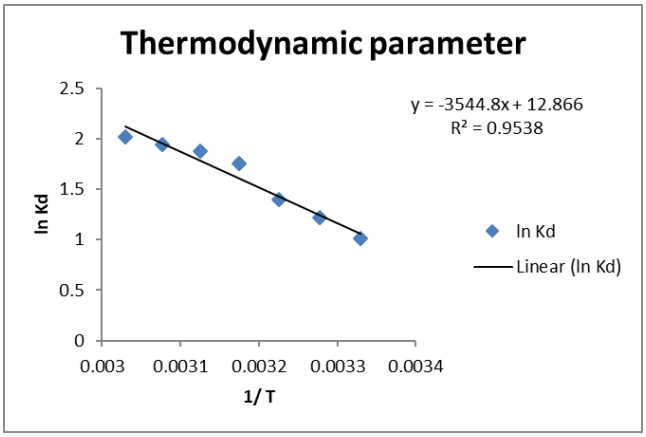



Fig 5: Thermodynamic parameter evaluation ($\Delta G^0$, $\Delta H^0$& $\Delta S^0$).

TABLE I

FREE ENERGY CHANGE ($\Delta G^0$) VALUES AT DIFFERENT TEMPERATURES

| Temperature (K) | Free Energy Change, $\Delta G^0$ (kJ/mol) |
|---|---|
| 300 | -3.47 |
| 305 | -3.53 |
| 310 | -3.59 |
| 315 | -3.65 |
| 320 | -3.70 |
| 325 | -3.76 |
| 330 | -3.82 |

*E. Regeneration of Adsorbent and Decolorization Studies*

Regeneration studies were conducted for five adsorption-desorption cycles using 0.1 N NaOH and 0.1 N HCl solutions. After each adsorption cycle, the packed bed was washed thoroughly with distilled water and regenerated chemically. The adsorption efficiency gradually decreased after repeated cycles due to saturation of adsorption sites (Fig. 6). Similar decline in adsorption efficiency after regeneration cycles was also reported by Pathania et al. using *Phoenix dactylifera* seed adsorbent.

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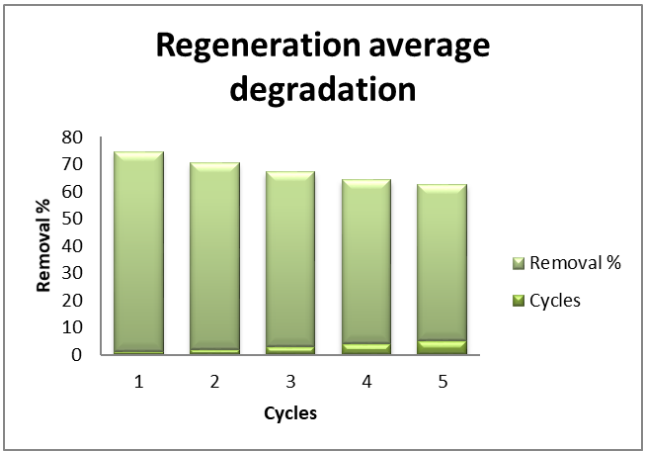


Fig 6: Removal efficiency of AS in various cycles of operation.

*F. Ozone Treatment*

Advanced oxidation treatment using ozone was employed for further enhancement of dye degradation efficiency. Ozone exhibited strong oxidative capability for degradation of azo dye molecules into simpler and less toxic compounds. The ozone oxidation process also reduced phytotoxicity and improved biodegradability of treated wastewater. Oxygen was supplied at a flow rate of 3 L/min during ozonation experiments.

Energy consumption analysis indicated that energy demand increased with increase in degradation efficiency and dye concentration. The total energy consumption required for degradation of 1 g Congo Red dye was found to be 3.5 kW. Ozone treatment exhibited higher oxidation potential compared to other oxidizing agents such as $H_2O_2$, thereby providing rapid decolorization and mineralization of dye molecules.

## IV. CONCLUSIONS

The present study demonstrated the effective removal and degradation of Congo Red dye using Arjuna seed biochar under both batch and continuous adsorption operating conditions. The prepared biochar exhibited excellent adsorption characteristics due to its high surface area, porous structure, and presence of various active functional groups confirmed through FTIR and BET analyses.

The adsorption efficiency was significantly influenced by operational parameters such as adsorbent dosage, temperature, initial dye concentration, and solution pH. Maximum dye removal efficiency was observed under acidic conditions and lower dye concentration levels. Continuous packed bed adsorption studies showed superior decolorization performance with a maximum removal efficiency of 98.32% for 50 ppm dye solution.

Langmuir and Freundlich adsorption isotherm studies confirmed favorable adsorption behavior of Congo Red dye onto Arjuna seed biochar. Thermodynamic analysis revealed that the adsorption process was spontaneous and endothermic in nature. Regeneration studies indicated that the adsorbent could be effectively reused for multiple adsorption-desorption cycles with moderate reduction in adsorption efficiency.

Advanced oxidation treatment using ozone further enhanced dye degradation and reduced the toxicity of treated wastewater. Ozonation demonstrated strong oxidative capability for mineralization of dye molecules with reasonable energy consumption. The combined adsorption and ozone treatment process can therefore be considered an efficient, economical, and environmentally sustainable approach for the treatment of textile dye-containing wastewater.

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