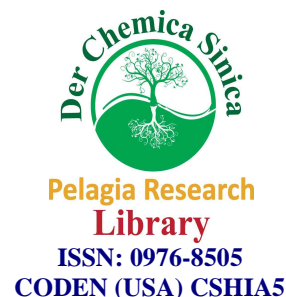




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### *Syzygium cumini* leaf powder as low cost adsorbent to remove Fe (II) from aqueous solution

Sureshkumar Halnori<sup>1</sup>, Rajesh Dandge<sup>#</sup> and Milind Ubale<sup>\*2</sup>

<sup>1</sup>Department of Chemistry Padmashri Doctor Vithalrao Vikhe Patil College of Engineering, Ahmednagar (M.S.) India.

<sup>#</sup>Department of Chemistry, Milind College of Science, Aurangabad

<sup>2</sup>Post Graduate Department of Chemistry Vasantrao Naik Mahavidyalaya Aurangabad (M.S.) India

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#### ABSTRACT

*Syzygium cumini* Leaf Powder (SCLP) was treated with nitric acid and used as a low cost, easily available and renewable biological adsorbent for the removal of Fe (II) from aqueous solution. Batch experiments were carried out for adsorption kinetics and isotherms. Operating variable studied were contact time, adsorbent dose, initial concentration of adsorbate, temperature and pH. Adsorption capacity seems to be enhanced by increasing temperature up to 40°C and adsorbent dose. Equilibrium data were well represented by the Freundlich isotherm model for all tested adsorption systems. Beside these the Thermodynamic study has showed that the Fe (II) adsorption phenomenon onto SCLP was favorable and spontaneous.

**Keywords:** Low cost material, Fe (II), adsorption.

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#### INTRODUCTION

An increase in population increased the industrialization and increase the discharge of effluents into water bodies, this discharge contain heavy metals like Chromium, Zinc, Copper, Nickel, Iron, Lead etc. Increased use of metals and chemicals in process industries has resulted in generation of large quantities of effluents that contain high level of toxic heavy metals and their presence pose environmental disposal problems due to their non-degradable and persistence nature, in addition mining, mineral processing and extractive metallurgical operations also generate toxic liquid wastes[1,2]. Heavy metals such as Nickel(II), Copper, Cadmium, Lead and Zinc have harmful effects on human physiology and other biological systems when they exceed the tolerance level[3,4].

Toxic metals can be removed by ion exchange, precipitation, reverse osmosis, evaporation, and electro dialysis etc. [5]. These methods were found unaffordable in India for removal of Fe due to operational difficulties and high treatment expenditure. Activated carbon is the most widely used adsorbent but commercially available activated carbons are very expensive [6]. Adsorption processes for removal of metals have been found cheaper and more effective than the several technologies [7]. Various low cost adsorbents have been used for the removal of heavy metals e.g. brick kiln ash, fly ash, bidi leaves, teak leaves, amla dust, wallastonite, peanut hull and neem leaves [5].

Literature survey reveals that the use of SCLP as a low cost adsorbent for removal of heavy metals from aqueous solutions has not been reported. Thus in the present study the systematic work was carried to investigate the adsorption properties of SCLP for the removal of Fe(II). The study includes adsorption as a function of time, adsorbent dose, concentration of Fe(II) solution, temperature and pH of solution. Isotherm studies were fitted for Freundlich and Langmuir adsorption isotherm.

## MATERIALS AND METHODS

### Preparation of adsorbent:

The adsorbent selected for the present study was *Syzygium cumini* which is locally available plant and was collected in Ahmednagar District of Maharashtra. The sample leaves were dried in shadow, avoiding direct sunlight on them. The dried leaves were grinded into powder and washed with distilled water and filtered. The residue left was treated with very dilute solution of nitric acid (0.1 N). It was then stirred for half an hour vigorously using mechanical stirrer at room temperature. Then it was filtered and washed with distilled water repeatedly to remove free acid. After chemical treatment the residue was dried first in air and finally in oven at 90-100°C for 8-10 hours and powdered using electric grinder. The homogeneous powder was passed through mesh for desired particle size. The adsorbent once prepared was used throughout the experimental work. The particle size of adsorbent selected for these experiments was on the basis of their settlement at the bottom of the system, so that the portion of the solution could be taken out conveniently from the supernatant liquid.

### Preparation of Fe (II) metal ion solution:

Iron was the metal ion selected for the present investigation. Fe (II) prepared by dissolving ferrous ammonium sulphate in double distilled water. The chemicals used were of analytical grade and used without further purifications. The solutions were prepared in doubly distilled water. A distilled water prepared by using first metal distillation unit and then all quick fit glass assembly in permanganatic condition, wherever necessary the prepared solutions were standardized as per literature.

### Adsorption experiments:

Each batch adsorption study was carried out by using *nitric acid treated SCLP* with iron ion solution under different conditions at maximum time 50 minutes. Iron was determined using spectrophotometer (SL-159 ELICO UV-Visible Spectrophotometer) at  $\lambda_{\text{max}}$  510 nm.

## RESULTS AND DISCUSSION

The experimental data obtained from the different batch type experiments, in the present investigation was analyzed and interpreted based on the adsorption of Fe (II) on SCLP.

### Effect of contact time:

The effect of contact time was studied by using 1 g of SCLP as adsorbent, 4 ppm of adsorbate solution, pH (2-7), room temperature (303 K) and different time (See Fig.1).

Fe (II) ions diffuse on to the surface of SCLP having active centers like pores, cracks, corners and gets adsorbed. Maximum adsorption of Fe(II) ions up to 50 minutes, beyond 50 minutes adsorption phenomenon gets retarded which may be attributed to accumulation of active centers of SCLP by Fe(II) ions and decrease in the affinity of Fe(II) for the surface of SCLP which was in good agreement with the reported work of Sayyed Hussian *et al.* [8] and therefore 50 minutes was considered as equilibrium contact time for all sets of experiment. Hence the adsorption percentage increases with increase in time and gets completed at 50 minutes.

### Effect of adsorbent dose:

Effect of adsorbent dose plays an important role in standardizing the adsorption process with quantification of adsorbate solution and the adsorbent. In our present investigation with increase in the amount of SCLP adsorbent the removal efficiency increased rapidly (Fig.2) which may be attributed to the greater availability of the exchangeable sites or surface areas at higher concentration of the adsorbent. Our findings are in good support with Hussein *et al.* [9,10].

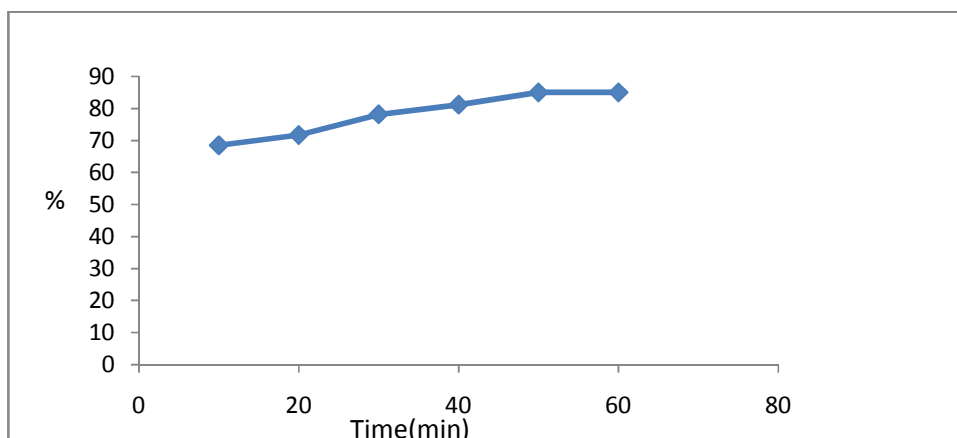


Fig.1: Effect of contact time

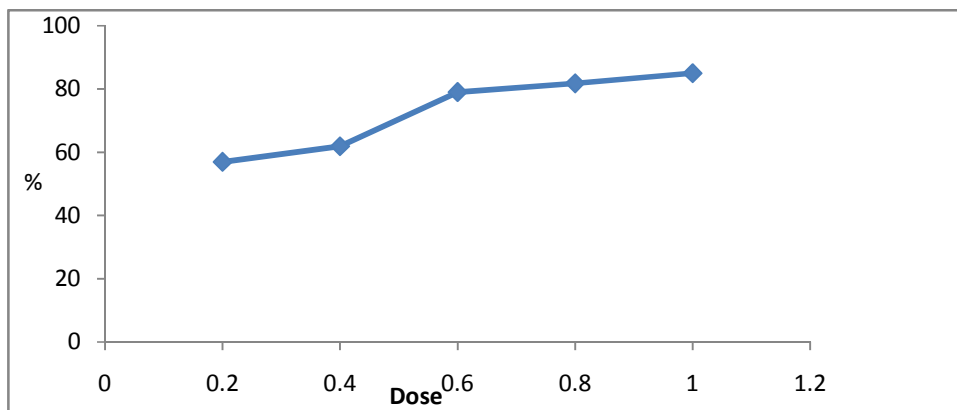


Fig.2: Effect of adsorbent dose

**Effect of initial concentration of adsorbate:**

The adsorption of iron onto the surface of SCLP was rapid initially, slows down later on and finally reached towards equilibrium indicating saturated adsorption [11]. The increased in adsorption may be attributed to increase in surface activity and due to micelle formation or the aggregation of iron ions in the concentration range studied<sup>11</sup>. The percentage removal of iron has shown significant decrease with the increase in the initial concentration of adsorbate (Fig.3). Our findings are in good agreement with the reported work of Ashoka leaf powder as adsorbent by Shelke et.al.[12].

**Effect of temperature:**

Adsorption as the effect of temperature was carried out in different temperature range (293-333 K) and remaining parameters were kept constant and found that adsorption increases with increase in temperature from 293-313 K and decreases from 323-333 (Fig.4). The increase in the extent of adsorption with the rise of temperature implies the endothermic adsorption. It has been reported that the increase in uptake with temperature is mainly due to an increase in the number of adsorption sites created by the breaking of some of the internal bonds near the edge of the crystal [13]. The decrease in adsorption with increase in temperature is due to the increase in solubility of the adsorbate with increase in temperature, or the mobility of the large ions increases with increasing temperature [14, 15]

Kinetic study was carried out and it showed second order reaction with  $R^2=0.984$ .

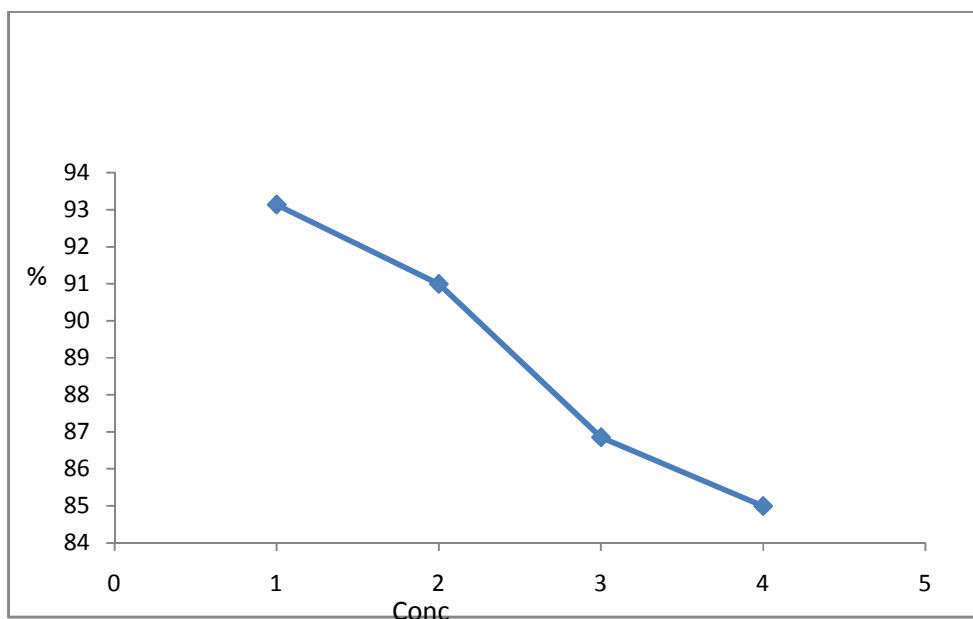


Fig.3: Effect of initial concentration of adsorbate

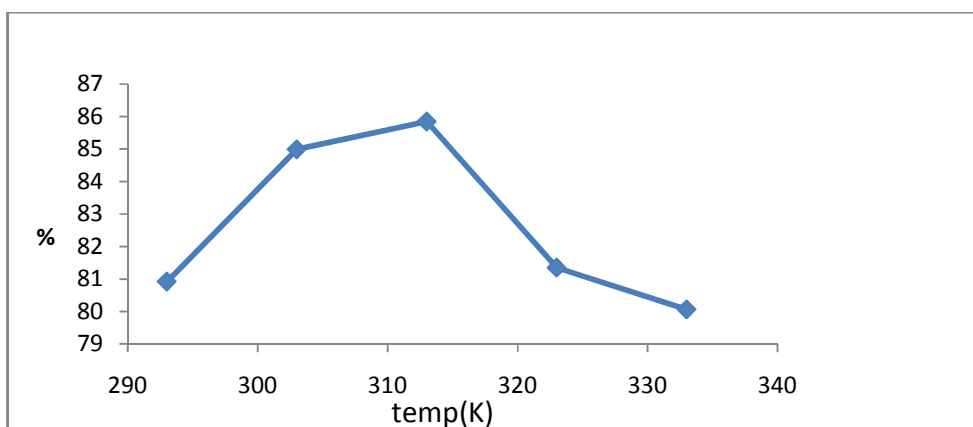


Fig.4: Effect of temperature

**Effect of pH:**

pH of solution is one of the most important parameter for adsorption of heavy metals from aqueous solution. In order to establish the effect of pH on the adsorption of iron ions, the batch equilibrium was studied at different pH values. The amount of metal adsorbed increases with increasing pH (Fig.5). Minimum adsorption at lower pH which may be attributed to presence of excess number of  $H^+$  ions onto the surface of SCLP and denies the migration of Fe (II) ions on it. With increase in pH competition from  $H^+$  ions decreases and the positive charged ions, Fe (II) take their place on the surface of SCLP. Our findings are in good agreement with reported work of Yan Liu and others [16, 17,18].

**Adsorption Isotherm:**

The adsorption isotherm is a graphical representation of amount of substance adsorbed against the residual concentration of the adsorbate concentration and adsorbent doses were analyzed using Langmuir and Freundlich isotherm in order to find the adsorption capacity of SCLP.

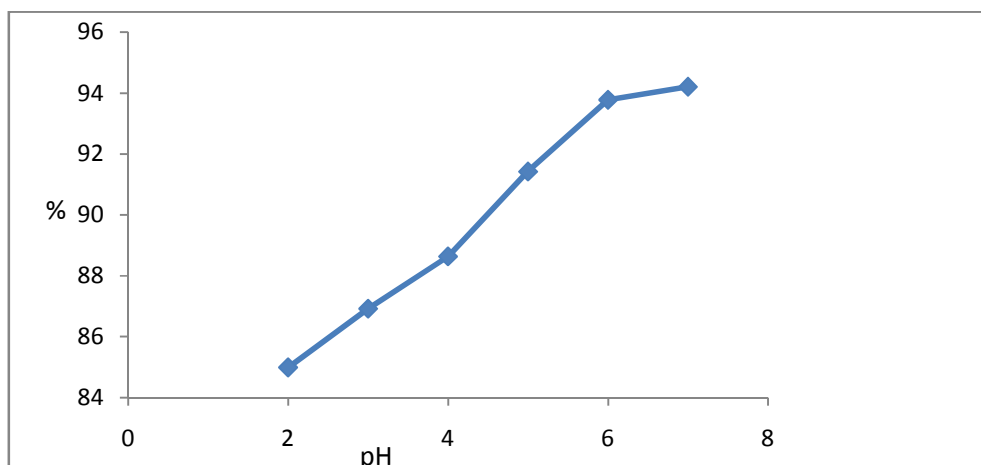


Fig.5: Effect of pH

The Freundlich isotherm was verified, the value of regression coefficient  $r^2$  found to be very close to one which indicates the good correlation exists between  $\log x/m$  and  $\log C$ . The Langmuir model represents monolayer adsorption on a set of distinct localized adsorption sites having the same adsorption energies no interaction between adsorbed molecules. The essential characteristic of Langmuir isotherm is expressed in terms of dimensionless constant or separation factor or equilibrium factor  $R_L$ , which is indicative of the nature of the isotherm and is enlisted below as,

$R_L$ Value	Type of Isotherm
$R_L > 1$	Unfavorable
$R_L = 1$	Linear
$0 < R_L < 1$	Favorable
$R_L = 0$	Irreversible

The adsorption was favorable onto the surface of SCLP as  $R_L$  value in the present study was 0.0044 which falls in the type  $0 < R_L < 1$ . Our findings are in good agreement with the findings reported by Patil and others [19, 20].

#### Thermodynamic Parameters:

Thermodynamic parameters such as free energy, enthalpy and entropy of adsorption were calculated and found that  $\Delta G$  was negative and showed spontaneous nature of adsorption process,  $\Delta H$  was negative and showed exothermic nature of adsorption process,  $\Delta S$  was positive and showed the increasing randomness at solid/liquid interface during the adsorption of iron. The values demonstrate a spontaneous and favorable adsorption process.

#### CONCLUSION

The present study showed that the nitric acid treated SCLP is an efficient low-cost adsorbent for the removal of toxic Fe (II) from aqueous solution. The adsorption of Fe (II) was found to be dependent on contact time, adsorbent dose, initial concentration of adsorbate, temperature and pH. The equilibrium adsorption data showed significant correlation to Langmuir and Freundlich adsorption isotherms and the adsorption was followed second order kinetics. Value of  $R_L$  indicates this adsorption process is favorable.

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