Comparative study of removal of phenol using *Tamarindus indica L.* seeds and *Diospyros melanoxylon Roxb.* leaves as biosorbent

H. Vignesh, M. Eajas Basha, S. Yuvaraj, D. Vinothkumar, R. Harisaranraj and N.G. Ramesh Babu

Abstract— Phenol and its derivatives are considered to be primary pollutant components in wastewater due to their high toxicity, high oxygen demand and low biodegradability. In the present scenario, it is found that the contamination of water by phenol is increasing day by day. The World Health Organization (WHO) prescribed 1 mg/l as the maximum permissible concentration of phenol in drinking water. Hence elimination of phenol from water becomes mandatory. Biological methods are the most appropriate techniques due to mineralization of toxic organic compounds and inexpensiveness. Among the biological methods, biosorption was considered as the best method because of its high removal efficiency. In this study, *Tamarindus indica L.* seeds and *Diospyros melanoxylon Roxb.* leaves are used as biosorbents and their efficiencies are compared. The maximum removal efficiency of 96.105% was obtained by *Diospyros melanoxylon Roxb.* leaves at pH-7 at time interval of 7 hrs before optimization. The optimization based on pH and time gave the highest removal efficiency of 99.06858594% which was obtained by *Tamarindus indica L.* seeds at pH-6.2 at a time interval of 31 hrs.

Index Terms— Biological methods, Biosorption, *Diospyros melanoxylon Roxb.* leaves, optimization, Phenol, *Tamarindus indica L.* seeds and World Health Organization.

1 INTRODUCTION

The contamination of water by phenol has been recognized as an issue of growing importance in recent years [1]. Phenol and its derivatives are considered to be primary pollutant components in wastewater due to their high toxicity, high oxygen demand and low biodegradability [2]. Phenol, also known as carbolic acid and phenic acid, is an organic compound with the chemical formula C_6H_5OH. The molecule consists of a phenyl group ($-C_6H_5$) bonded to a hydroxyl group (-OH). It is mildly acidic, but requires careful handling due to its propensity to cause burns. Phenol and its higher homology are aromatic molecules containing hydroxyl group attached to the benzene ring structure [3]. The origin of phenols in the environment is both anthropogenic as well as xenobiotic [4].

Phenol at concentration as low as 5 mg/l impacts typical smell upon chlorination. Although the toxicity of phenolic compounds is not as high as heavy metals or biocides, their high contamination often inhibits or even eliminates bacteriological populations in municipal biological wastewater treatment plants. They also add odour to drinking and food processing water and have mutagenic and carcinogenic effects [5] even at low concentration of 2.0 µg/l [3].

Concentration of phenol in the industrial effluents varies in the range of 10 to 17,500 mg/l. The World Health Organization (WHO) prescribed 1 mg/l as the maximum permissible concentration of phenol in drinking water [6]. Thus, elimination of phenol effectively is necessary to preserve the environment and the health of human beings [4]. Therefore, effluents containing phenolic compounds must be efficiently treated before being discharged to the surrounding environment.

Biosorption can be defined as a process in which solids of natural origin are employed for sequestration or separation of pollutants from an aqueous environment [7]. Many natural materials were used as biosorbent for phenol removal, like human hair [8] and chicken feather [9], chemically modified pistachio shells [10], peanut shell [11] and tamarind nutshell [12] etc., which are cheap viable options.

In the present work, study has been made regarding the phenol removal efficiency of activated carbon biosorbents obtained from *Tamarindus indica Roxb.* seeds and *Diospyros melanoxylon L.* (commonly known as Tendu Patta) leaves.

2 MATERIALS AND METHODS

2.1 Sample Collection

2.1.1 *Tamarindus indica* seeds

The tamarind seeds for production of biosorbents were collected from the AERI (Adhiyamaan Educational and Research Institute) campus.

2.1.2 *Diospyros melanoxylon* leaves

Diospyros melanoxylon leaves were obtained by removing the outer layer of beedis (tobacco present inside beedis were cleaned) which was bought from a local general store near

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2.2 Stock solution preparation

Stock solution was prepared by adding 1g of crystal phenol to 1 litre of distilled water to obtain 1ppm concentration. The stock solution was prepared fresh in order to carry out the further processes.

2.3 Preparation of biosorbent

2.3.1 Powdering

1. *T. indica* seeds were crushed using a roller crusher and *D. melanoxylon* leaves were cut into small pieces (2-3cm).
2. The samples were powdered using a mixer.
3. The powdered samples were sieved using a 40µm mesh to get even sized pieces.

2.3.2 Formation of activated carbon

1. Sulphuric acid was added to the powdered samples in the ratio 1:1.
2. The samples were then heated at 180°C for 3 to 5 hours.
3. 1% sodium bicarbonate was added to the samples until the effervescence ceased.

2.3.3 Drying

1. The samples were then dried at 120°C for 18 hrs.
2. The dried samples were used to carry out further processes.

2.4 Addition to the stock solution

1 g of biosorbent was added to 500 ml of stock solution and mixed well. 50 ml of the biosorbent mixed solution was taken and filtered using Wattman No. 1 filter paper so that the sample is free from the biosorbents. Estimation of amount of phenol was carried out by direct photometric method. This gives the initial phenol concentration and this procedure was carried out every 1 hr.

2.5 Estimation of phenol

Estimation of phenol was carried out by 4-aminoantipyrine method.

2.5.1 Procedure

1. 50 ml of the sample was transferred to a beaker
2. 0.3 ml of 2% 4-aminoantipyrine was added
3. 1ml of 2N Ammonium hydroxide was added and mixed thoroughly
4. 1ml of 2% Potassium ferricyanide was added and mixed again
5. The reddish colour produced by the sample in UV-spectrophotometer at 500 nm was compared

3 Results

In the present environmental status it is found that phenol is one of the most common pollutants which affects the environment by polluting water bodies, rivers, lakes etc. In this regard, it is necessary to decrease the amount of phenol in water. The present study describes the removal of phenol using *Tamarindus indica Roxb.* seeds and *Diospyros melanoxylon L.* leaves as biosorbents. The removal efficiency of microorganisms was calculated by the formula

$$\text{Removal (\%)} = \left[\frac{(\text{Intial absorbance} - \text{Final absorbance})}{\text{Initial absorbance}} \right] X\ 100$$

Results were tabulated and graph was plotted between time (hour) and removal (%).

TABLE 1: ABSORBANCE VALUE OF *TAMARINDUS INDICA* SEEDS BIOSORBENT

Time (hour)	Absorbance at 500nm	Removal (%)
0	2.192	7.19729
1	2.182	7.62066
2	2.151	8.933108
3	2.147	9.102456
4	2.141	9.356478
5	1.82	22.94666
6	1.73	26.75699
7	1.6	32.2608

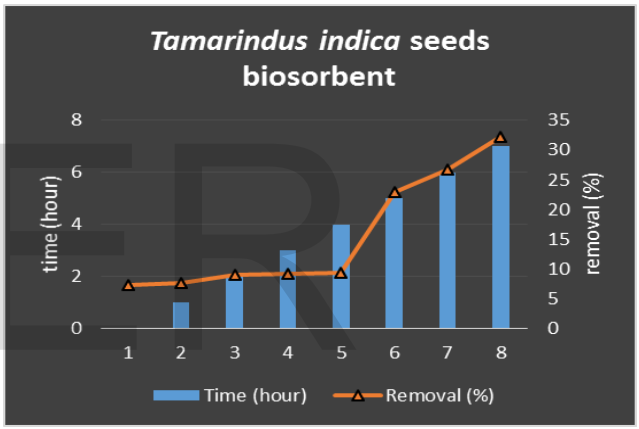

Fig. 1. Phenol removal efficiency of *Tamarindus indica* seeds biosorbent

TABLE 2: ABSORBANCE VALUE OF *DIOSPYROS MELANOXYLON* LEAVES BIOSORBENT

Time (hour)	Absorbance at 500nm	Removal (%)
0	2.145	9.18713
1	1.568	33.61558
2	1.379	41.61727
3	0.734	68.92464
4	0.438	81.45639
5	0.15	93.64945
6	0.123	94.79255
7	0.092	96.105

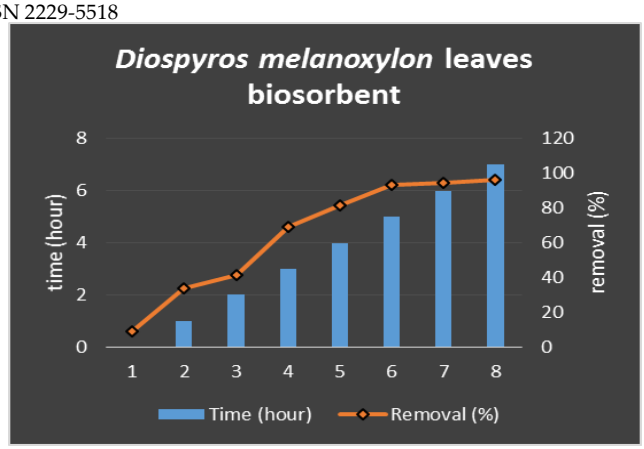

Fig. 2. Phenol removal efficiency of *Diospyros melanoxylon* leaves biosorbent

3.1 Optimization based on pH

Optimization based on pH was done by altering the pH of the solution. The efficiency of phenol removal was determined for pH 4, 6.2, 7 and 8 for both the biosorbents separately.

TABLE 3: ABSORBANCE VALUE OF *TAMARINDUS INDICA* SEEDS BIOSORBENT AT PH-4, 6.2, 7 AND 8

Time (hour)	Removal (%) at pH-4	Removal (%) at pH-6.2	Removal (%) at pH-7	Removal (%) at Ph-8
0	8.890771	7.408975	7.19729	7.281964
1	9.18713	8.933108	7.62066	7.62066
2	9.271804	9.737511	8.933108	7.959356
3	10.79594	10.66892	9.102456	8.255715
4	11.93903	12.19306	9.356478	8.594412
5	21.25318	23.7934	22.94666	9.398815
6	26.33362	29.29721	26.75699	17.01948
7	37.93911	35.64776	32.2608	22.94666

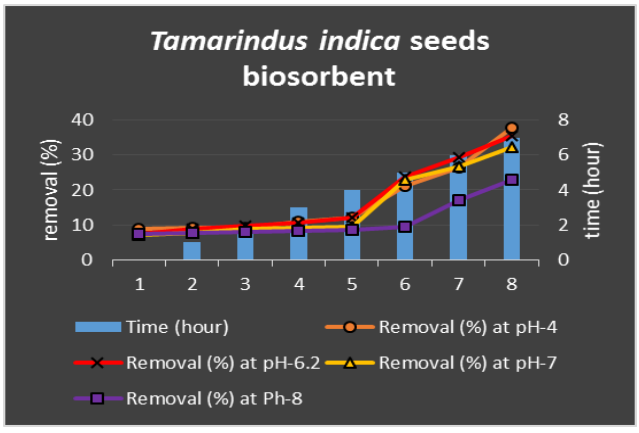

Fig. 3. Phenol removal efficiency of *Tamarindus indica* seeds biosorbent at various pH

TABLE 4: ABSORBANCE VALUE OF *DIOSPYROS MELANOXYLON* LEAVES BIOSORBENT AT PH-4, 6.2, 7 AND 8

Time (hour)	Removal (%) at pH-4	Removal (%) at pH-6.2	Removal (%) at pH-7	Removal (%) at Ph-8
0	9.18713	9.18713	9.18713	9.18713
1	11.30398	11.43099	33.61558	35.8171
2	43.98815	30.82134	41.61727	55.50381
3	55.50381	56.56224	68.92464	73.7934
4	74.76715	76.24894	81.45639	87.46825
5	92.59102	93.26842	93.64945	93.94581
6	95.25826	96.74005	94.79255	96.06266
7	98.22185	97.88315	96.105	96.69771

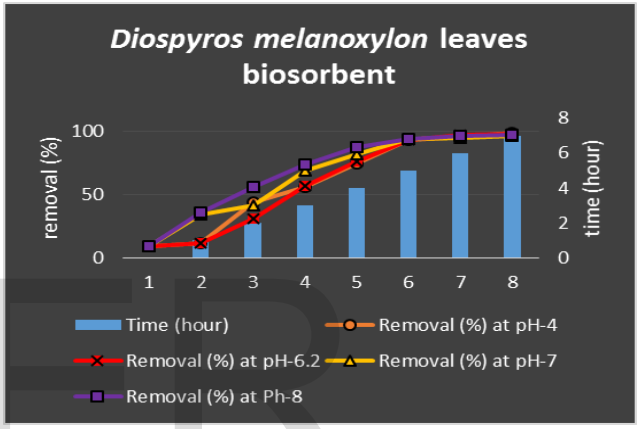

Fig. 4. Phenol removal efficiency of *Diospyros melanoxylon* leaves biosorbent at various pH

3.2 Optimization based on time

Optimization based on time has been done for biosorbents for the solutions of pH 4, 6.2, 7 and 8 at a time interval of every 1 hr starting from 24 hrs.

TABLE 5: ABSORBANCE VALUE OF *TAMARINDUS INDICA* SEEDS BIOSORBENT AT PH-4, 6.2, 7 AND 8 AT VARIOUS TIME INTERVAL

Time (hour)	Removal (%) at pH-4	Removal (%) at pH-6.2	Removal (%) at pH-7	Removal (%) at Ph-8
24	98.17951	98.47587	98.13717	97.54445
25	98.39119	98.64522	98.34886	97.92549
26	98.72989	98.72989	98.5182	98.17951
27	98.81456	98.81456	98.64522	98.30652
28	98.8569	98.89924	98.72989	98.39119
29	98.89924	98.98391	98.81456	98.47587
30	98.89924	99.02625	98.8569	98.56054
31	98.94157	99.06859	98.89924	98.60288

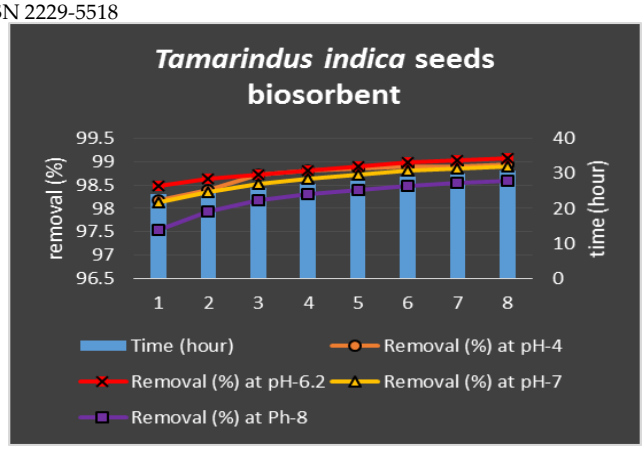

Fig. 5. Phenol removal efficiency of *Tamarindus indica* seeds biosorbent at pH-4, 6.2, 7 and 8 at various time interval

TABLE 6: ABSORBANCE VALUE OF *DIOSPYROS MELANOXYLON* LEAVES BIOSORBENT AT PH-4, 6.2, 7 AND 8 AT VARIOUS TIME INTERVAL

Time (hour)	Removal (%) at pH-4	Removal (%) at pH-6.2	Removal (%) at pH-7	Removal (%) at Ph-8
24	98.68755	98.64522	97.41744	97.88315
25	98.77223	98.64522	97.58679	98.0525
26	98.77223	98.64522	97.7138	98.13717
27	98.77223	98.68755	97.92549	98.22185
28	98.81456	98.68755	98.09483	98.26418
29	98.81456	98.68755	98.22185	98.30652
30	98.81456	98.68755	98.39119	98.34886
31	98.8569	98.72989	98.5182	98.39119

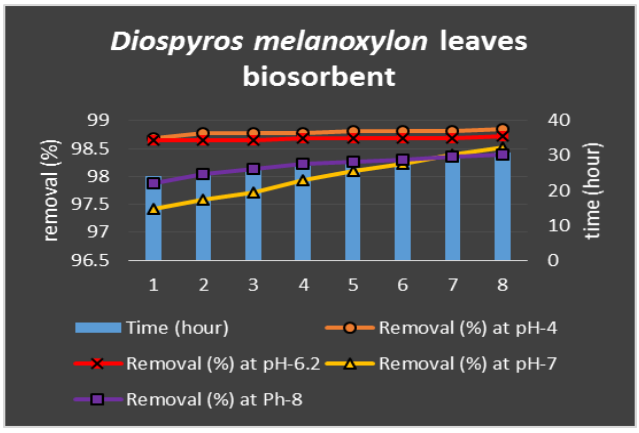

Fig. 6. Phenol removal efficiency of Diospyros melanoxylon leaves biosorbent at pH-4, 6.2, 7 and 8 at various time interval

4 CONCLUSION

The biosorbents were prepared from *T. indica* seeds and *D. melanoxylon* leaves. The production of biosorbents is found to be cost effective and has a high absorption ability of Phenol. Biosorbents removed the phenol without formation of any degraded product of phenol. Thus, phenol removal by biosorbents is an eco-friendly method and is recommended for the removal of phenol.

5 ACKNOWLEDGEMENT

The authors wish to thank Mrs. T. Pakutharivu, Head of the Department, Department of Biochemistry, M.G.R College of Arts and Science, Hosur for her constant support throughout the project.

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