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PRELIMINARY STUDY ON POTENTIAL OF SEAWEEDS IN DECOLORIZATION EFFICACY OF SYNTHETIC DYES EFFLUENT

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ABSTRACT: Synthetic dyes effluent comprised of color which cause damage to aquatic organisms and public human health if discharged into water bodies without prior treatment, also affects the aesthetic merit. This paper investigated the potential decolorization of industrial dye effluents by environmental species of various seaweeds collected from Beyt-dwarka, near Okha coast of Gujarat, India. Seaweeds *Ulva lactuca, Sargassum wightii, Sargassum johnstonii, Caulerpa racemosa, Acanthophora spp, U. reticulata, U. clathrata Roth, Padina pavonica* were inoculated with dyes effluents to determine its possibility for decolorization. Screening of most potential macroalgal species was studied by UV- Spectrophotometer. Among the seaweed species tested the most resulting potential seaweed were noticed as *Ulva lactuca*, *U. clathrata Roth* and *U. reticulata* & *Caulerpa racemosa* indicates 85.33 %, 81 % , 83 % & 83 % decolorization respectively for lower concentration of effluent after treatment for 16 days. Effluent characteristics in terms of physico-chemical parameters and heavy metal concentration of resulting effluent after seaweed treatment was studied indicates higher concentration of each parameter & highest level of heavy metal cupper (Cu) was detected in E_4 & E_6 as 16.865 mg l^{-1} & 16.048 mg l^{-1} respectively. The present study indicates tha seaweed can be used as biosorbent or as biodecolorising agent as it is cheaper & ecofriendly method.
Key words: Seaweed, Decolorization, Dye Effluents, Okha Coast, Physico-chemical analysis.

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INTRODUCTION

Approximately above 10,000 different types of dyes and pigments were used in the textile and printing industries. Industrial use of synthetic dyes includes complex compounds such as azo, triphenylmethane, and heterocyclic/polymeric structures [7, 33]. The studies based on physico chemical characteristics of waste water and statistical analysis has been reported [12, 13, 15, 18, 23, 24, 25, 27, 28, 30]. They are characterized as non-biodegradable and persistence in nature; it creates severe public health effects not only that it affects serious environmental problems [10]. There are numerous treatment technologies available to treat wastewater by physical and chemical processes such as flocculation, coagulation, ultra-filtration, ion-exchange process, reverse osmosis despite of their existence bio treatment process can be attractive solution because of their low coast, eco friendly and viable method [22].

Algae play an important role in wastewater treatment systems as they are universally acknowledged for that [11, 26]. Algae are classified in two types as microalgae and macro algae. Microalgae can be easily cultured and cheapest material for water treatment; the literature shows lot of research has been carried out on waste water treatment by freshwater algae such as *Chlorella vulgaris* and *Scenedesmus dimorphus* is highly efficient for ammonia and phosphorous removal during biotreatment of secondary effluents from agro industrial wastewater of a dairy industry and pig farming [16].

The role of microalgae to treat waste water was studied, *Chlorella vulgaris* shows the best removal capacity of nitrate and COD while *Scenedesmus quadricauda* shows BOD and phosphate reduction [4]. The dye Amido Black and textile dye effluent is chosen for this investigation shows that it decolorizes the textile effluent efficiently in short period of time. This can be used for the bioremediation of dye effluents [2]. Microalgal species such as *Chlorella vulgaris* LS120 & *Scenedesmus obliqus* LS121 effectively reduces the COD, BOD, TDS of tannery effluents of leather processing industry [34]. Bioaccumulation of Cadmium in Blue Green Algae *Spirulina* (Arthrospira) *Indica* was observed [5]. Bioaccumulation is the effective method for removal of heavy metal ions from wastewaters, the use of algal to extract, sequester and or detoxify heavy metals and other pollutants. Filamentous alga of *Pithophora* sp. used for the removal of cadmium, chromium and lead from industrial wastewater [6]. Biodegradation and biosorption capacity of some potential Cyanobacterial species: *Oscillatoria* sp., *Nodularia* sp., *Nostoc* sp., *Synechococcus* sp. and *Cyanothece* sp. dominated the effluents and mixed cultures showed varying sensitivity. Contaminants were removed by all the species either as individuals or in mixtures [8]. Biodegradation of tryphenylmethane dye malachite green by *Cosmarium sp.* was investigated, which shows the ability of the algal species in removing the dye [21]. Potential of seaweeds to treat waste water contaminated with heavy metals have been studied by various invigilators [9, 10, 31]. There is large abundance of seaweeds found in the shoreline and coastal areas at many parts of the oceans worldwide. They have predominant absorptive performance. They have large numbers of functional groups having strong resemblance towards dissolved cationic metals though also having comparatively higher concentrations of these same metals in the biomass. The macroalgae can be used as biosorbent material as well as biochar formation to study decolorization effieciency.

This research paper aims to determine the potential of different seaweeds in the decolorization of dyes containing industrial effluents collected from dyes manufacturing industrial sites of Gujarat, India. Present investigation focuses on screening of most potential seaweeds and resulting effluents adjacent to seaweed. It also focuses to measure the pollutant level in terms of physicochemical characterization and heavy metal content of untreated dye effluents from various dye industries from Gujarat and its co-relation was studied.

MATERIAL AND METHODOLOGY:

Collection of Effluents:

After survey of Dyes manufacturing industrial units of Gujarat, India the untreated effluent were selected for study. The effluent samples were collected from dyes manufacturing industrial units of Vadodara, Ahmedabad, Ankleshwar, Bharuch and Anand. Three effluents E1, E2, E3 were collected from industrial unit at Padra village, Vadodara District comprised of mixture of reactive red dyes, E4 effluent from Vatva GIDC, Ahmedabad District comprised of reactive yellow dyes effluent, E5 effluent from Ankleshwar GIDC, Bharuch District comprised of ink (basic) dye effluent, E6 and E7 effluents were collected from industrial unit at Kalamsar village, Khambhat taluka, and District, they were comprised of mixture of reactive black dyes, E8 effluent collected from Palsana industrial unit, Surat District comprised of disperse dyes effluent. They are collected from the various points inside industrial units such as reactor washings, filter press washings, reverse osmosis system, various glass distilleries washings and miscellaneous washings etc. These samples were untreated samples as they have not been through activated sludge systems.

Physico-chemical characterization of dye effluents:

The parameters such as pH, TDS, TSS, colour, ammoniacal nitrogen, COD, chloride, sulphate and phosphate were carried out by using APHA[3] standard method. The COD was determined by the dichromate digestion method. The chloride and ammoniacal nitrogen was estimated by titration method and colour, phosphate and sulphate were determined by using uv-visible spectrophotometer (Model-1800, Make- Shimadzu). Standard reagents and chemicals were utilized for the physico-chemical analysis. The results obtained were evaluated in accordance with the norms prescribed by Ministry of Environment and Forest of India for waste water [20]. Heavy metals from resulting effluents selected after treatment with seaweeds were analyzed by ICP-OES. (Model- Optima 3300 RL, Make- Perkin Elmer) which was evaluated with standards of Environment rules of India, 1986 [36].

Correlation studies

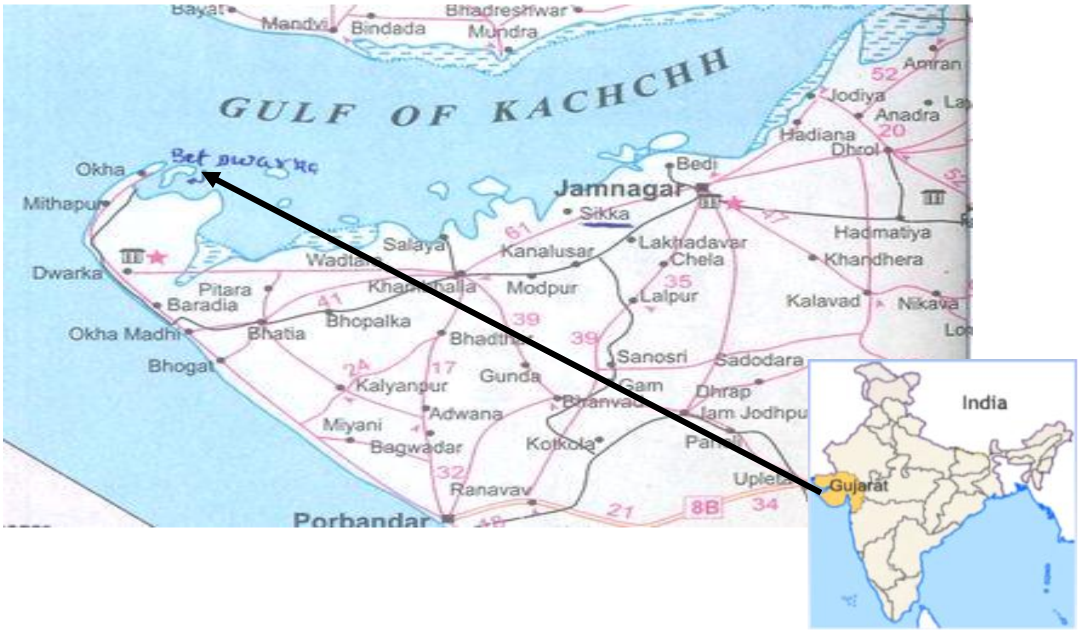
In the present paper correlation was used for declining the range of uncertainty. Correlation coefficient (r) was calculated by using the equation [29]:

r= $\sum xy \;/\; \sqrt{\sum x^2} \times \sqrt{\sum y^2}$

Seaweed collection and its identification:

The seaweed species *Ulva lactuca*, *Sargassum wightii*, *Sargassum johnstonii*, *Caulerpa racemosa*, *Acanthophora spp*, *U. reticulata*, *U. clathrata Roth* were collected from Beyt-dwarka near Okha coast of Gujarat, India (Longitude - 68°20´ E to 70°40´ E Latitude - 22°15´ N to 23°40´ N). They were collected and washed with seawater on the spot to remove debris, sand particles and surface salt and impurities. It was again washed in laboratory with tap water. Excessive water was removed by blotting paper and stored in refrigerator at freezing temperature. It was identified with the help of fisheries department, Junagadh Agriculture University, Okha, Gujarat by method described by M. Umamaheswara Rao, (1987) and from book of Study of seaweed diversity along the island of gulf of Kachchh, Gujarat (Gujarat Ecology Comission) [17].

Area of Seaweed Collection



Spectrophotometer Analysis

The Scanning was performed to determine maximum absorbance (λ max) wavelength of diluted (2:100) untreated dye effluents under 200 to 700 nm by UV-vis spectrophotometer (Make- Shimadzu, Model- 1800) [14].These effluents studied for decolorization (λ max) maximum absorbance at 535 nm, 605.5 nm, 742.5 nm, 420 nm, 602.5 nm, 742 nm,738 nm and 470 nm for E_1, E_2, E_3, E_4, E_5, E_6, E_7 and E_8 respectively. The Optical Density was measured for individual effluent (Figure.1 & Figure.2).

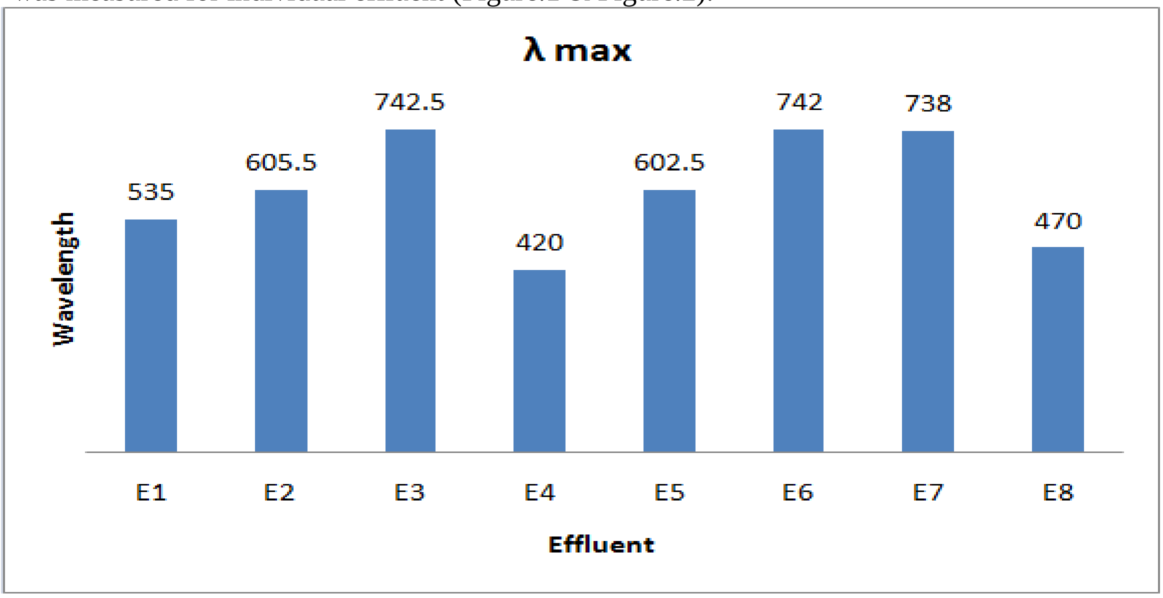


Figure-1: Spectrometer analysis for the determination of λ max

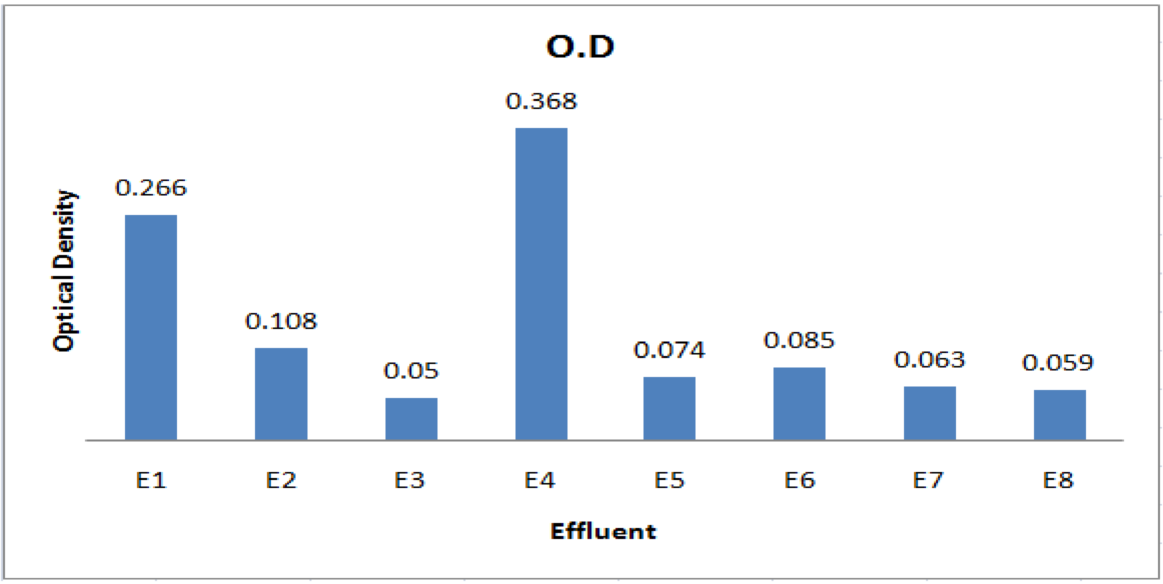


Figure-2: Spectrometer analysis for determination of OD Value

Screening of Seaweed and Effluent

The collected seaweed species *Ulva lactuca, Sargassum wightii, Sargassum johnstonii, Caulerpa racemosa, Acanthophora spp, U. reticulata, U. clathrata Roth, Padina pavonica* were entitled as A, B, C, D, E, F, G and H respectively. The experiment was carried out by inoculating 0.2 % w/v of seaweed in 50 ml of effluent samples entitled as E1, E2, E3, E4, E5, E6, E7 and E8. The experiments were carried out upto 24 days time period. The potential decolorization property of seaweed was determined by measuring optical density at every 8 days interval.

Determination of Decolorization Efficiency

This experiment was carried out for 24 days, the resultant optical density was measured at every 8 days interval with UV-Visible spectrophotometer (Make- Shimadzu, Model- 1800). Percentage reduction was calculated as below:

$$(\%) Decolorization = \frac{Initial\ absorbance - Final\ absorbance}{Initial\ absorbance} \times 100$$

RESULTS AND DISCUSSION

Characteristics of untreated dyes effluent

The untreated dyes effluents were highly colored and odour therefore it required significant treatment before disposal to main stream. The physico-chemical analysis of effluent samples namely E1 to E8 were represented in Table-1. The parameters studied are pH, TDS, TSS, colour, ammoniacal nitrogen, COD, chloride, sulphate and phosphate, these all are above the permissible limits given by MOEF Standards of India and required to be treated before discharge to Common Effluent Treatrment Plant (CETP).

pH

The colored samples with pungent smell having different pH are in the permissible limits. The effluent sample E4 is having acidic pH 3.76 which is higher than the permissible limit given by Ministry of environment and forest of India. i.e. 5.5- 9.0. The extreme pH of wastewater is generally not acceptable, as lower pH cause problems to survival of aquatic life. It obstructs with the optimum operation of wastewater treatment facilities. Water with high or low pH is not suitable for irrigation. The toxicity of heavy metals also gets enhanced at particular pH reported [25, 27].

TDS and TSS

The concentration of TDS and TSS are found high in all the samples. Highest value of TDS is found 25,080 mg/lin E3 and TSS is 2500 mg/l in E1 which exceed MOEF standard value i.e. 2100 mg/l and 100 mg/l respectively. The effluents with high TDS value may cause salinity problem if released to irrigation water [15]. Various values reported in previous work as TDS and TSS were 8445.6 mg/l and 544.95 mg/l respectively [27] & TSS ranging between 32-140 mg/l and TDS ranging between 8-20 mg/l [28].

Ammoniacal nitrogen

Ammoniacal nitrogen found higher in E2 as 760.66 mg l^{-1} where as in E1, E4 and E5 found lower values which is 29.86 mg/l, 20.53 mg/l and 13.06 mg/l respectively, which is in the range of MOEF [20] i.e. 50 mg/l. Presence of high amount of ammoniacal nitrogen in effluent is very toxic to marine biota.

COD

High value of COD cause depletion in dissolved oxygen in water which affects aquatic life difficult to survive. The standard value given by MOEF is 250 mg/l. Concentration of COD was found high in all the effluent samples. In Table-1 the maximum concentration of COD was found in E7 which is 72459 mg/l. Patale et al., 2012. [27] reported 8445.6 mg/l of COD also higher than MOEF standard & Patel et al., 2015. [28] reported 32-58 mg/l of COD which lower than permissible limits.

Color

Color is very important factor for aquatic life for food production from [18] Presence of high amount of color in water causes depletion in dissolved oxygen and biological oxygen demand which affects aquatic life for respiration and photosynthesis. In table-1 value of color is found higher in all the effluent samples. Standard value of MOEF for color is 400 Co.Pt. unit. Highest value is found in E7 which is 651450 Co.Pt. unit Which is higher than MOEF standard.

Chloride and Sulphate

High concentration of chlorides and sulphates may due to use chlorine compounds, such as hypochloric acid, chlorine gas, hydrochloric acid, and sulphate compounds such as aluminium sulphate, sulphuric acid, sodium sulphate, etc. are used as raw materials in various processes [25]. In table-1 the highest value of chloride and sulphate is 100190 mg/l and 101685 mg/l in E1 and E2 respectively which exceed MOEF value that is 1000 mg/l for both parameters.

Phosphate

The highest value of phosphate is found 1102 mg/l in E3.Standard for phosphate has not been given by any national or international authority. 0-20 mg/L of phosphates are permissible for irrigation. Over 75% of the samples are having maximum concentration levels; they are unfit for irrigation without proper treatments [25].

Correlation studies

Correlation coefficient (r) was calculated by taking the values of two factors at a time shown in Table-1. From the Table-2 it has been observed that the water pH found to show positive correlations with TSS and Phosphate it means variation of pH with TSS and Phosphate is in forward direction i.e. if one is raises other will also be raises. pH has negative correlation with TDS, ammoniacal nitrogen, COD, color, chloride, and sulphate i.e. if one is increases other will decrease. TDS has positive correlation with TSS, ammoniacal nitrogen, COD, color, chloride, sulphate and phosphate. TSS shows positive correlation with chloride whereas negative correlation with ammoniacal nitrogen, COD, color, chloride, sulphate, phosphate. ammoniacal nitrogen shows positive correlation with phosphate whereas COD, color, chloride, sulphate, phosphate. COD shows positive correlation with color, sulphate and phosphate. Color has positive correlation with chloride, sulphate and phosphate. chloride has positive correlation with sulphate whereas negative correlation with phosphate. There is the negative correlation in between sulphate and phosphate.

Heavy Metal Concentration

In this study the concentration of heavy metals such as Cr, Ni, Zn, Pb and Cd in the effluents were found below the permissible limits given for marine coastal areas as per the General Standards of The Environment Protection Rules of India, 1986 (Schedule-4) [36] as indicated in Table-3 which indicates the levels of pb found in all the effluent samples was below detection limit. The concentration of copper (Cu) in E1 is found below detection limit but found most highest in effluents E4 & E6 as 37.533 mg l^{-1} & 16.865 mg l^{-1} respectively which exceeds the limit as shown in Table-3. It has been reported that the concentration of heavy metals increases directs to bioaccumulation of metals in flora and fauna if the uptake rate of heavy metals by the organisms is more than the excretion stage [13].

Screening of seaweed from the decolorization study

The screening of macroalgae for decolorization of dyes effluent represented in graph-9 to graph-11, the result indicates that decolorization efficiency were found satisfactory in *Ulva lactuca* (A), *Caulerpa species* (D), *U. reticulate* (F), *U. clathrata Roth* (G). The *Ulva lactuca* (A) species gives good decolorization results with E1, E2 and E3 effluents. The *Caulerpa species* (D), *U. reticulate* (F) and *U. clathrata Roth* (G) species have good (65-85 %) potential for E1 and E4 and satisfactory (45-65 %) for E2 samples. All these four species gives better decolorizations for E1 effluent. The value slightly increases for 16 days, after that it becomes steady up to 24 days as shown in Figure-3 to Figure-9.

According to recent studies revealed that marine algae shows highest results 55% compare to fresh water algae 43% [32]. Previous study shows that color removal increases with increasing time period (Saraswathi and Balakumar, 2009) [35] they reported maximum time is ideal way to decrease color from dyes effluent. The present result has proved that the significant time period is 8 days then slowly increased with 16 days with no major differences and then remain steady upto 24 days. The present study shows that decolorization of above four species, *Ulva lactuca* , *Caulerpa species* , *U. reticulate* and *U. clathrata Roth* were relatively higher as compared to other research workers (Kumar et al., 2014) [32] dealt with marine species (*lsocrysis galbana*; 55.75 %).

Table-1: Physico-Chemical Analysis of dyes effluent

Parameters	E1	E2	E3	E4	E5	E6	E7	E8	MOEF Standard
pH	6.88	7.74	7.52	3.76	8.75	5.77	7.70	1.41	**5.5-9.0**
TDS (mg l^{-1})	17,180	3,62,580	25,080	17,580	9500	10740	271380	129900	**2100**
TSS (mg l^{-1})	2500	340	260	220	400	380	5040	500	**100**
Ammoniacal Nitrogen (mg l^{-1})	29.866	760.66	59.733	20.533	13.066	59.733	265.066	300	**50**
COD (mg l^{-1})	4776.64	48576	5181.44	6395.84	4533	4493	72459	8036	**250**
Colour (Co.Pt.)	160785	109065	53678	559260	3382	49483	651450	30708	**400**
Chloride (mg l^{-1})	100190	107768	8419	21048	1566	7577	55567	46652	**1000**
Sulphate (mg l^{-1})	12790	101685	6451	65210	1044	7868	64275	28600	**1000**
Phosphate (mg l^{-1})	367	328	1105	164	15.55	112	266	60.454	-

(Table-1 shows the physico chemical analysis of the dyes effluents E1, E2, E3, E4, E5, E6, E7 and E8 collected from various industries MOEF: Ministry of Environment & Forest, TDS: Total dissolved solid, TSS: Total suspended solids, COD: Chemical oxygen demand, Co.Pt: Cobalt Platinum Unit of Color parameter)

Table-2: Correlation matrix for untreated dyes effluent parameters

Analysis	pH	TDS	TSS	Ammoniacal N	COD	Color	Chloride	Sulphate	Phosphate
pH	1								
TDS	-0.11447	1							
TSS	0.14844	0.04079	1						
Ammoniacal Nitrogen	-0.02883	0.37903	-0.17347	1					
COD	-0.74287	0.47182	-0.28199	-0.25257403	1				
Color	-0.8571	0.22231	-0.28199	-0.38764487	0.92786	1			
Chloride	-0.05799	0.17094	0.9706	-0.18780321	-0.0512	0.0061	1		
Sulphate	-0.85566	0.27961	-0.22293	-0.31228657	0.96773	0.0061	0.0061	1	
Phosphate	0.21272	0.91034	-0.02119	0.588047394	0.10784	0.01994	-0.11153	-0.11153	1

(Table-2 shows correlation matrixes for untreated dyes effluents parameters)

Table-3: Heavy metal analysis of resulting effluent after seaweed treatment

Effluent	Cr (mg l^{-1})	Ni (mg l^{-1})	Cu (mg l^{-1})	Zn (mg l^{-1})	Pb (mg l^{-1})	Cd (mg l^{-1})
E_1	0.6573	0.174	BDL	BDL	BDL	0.0076
E_4	0.5179	0.2054	16.865	4.377	BDL	1.4648
E_6	0.1376	0.1754	16.048	1.4702	BDL	BDL
EPA Rules, India. 1986 (General Standard)	1.0	5.0	3.0	15	2.0	2.0

(Table-3 shows the heavy metal analysis of resulting effluents E1, E4 and E6 after seaweed treatment measured in mg l-1 unit, BDL: Below detection limit, EPA: The environment protection rules of India,1986 [schedule-4])

Screening of Seaweed and Effluent

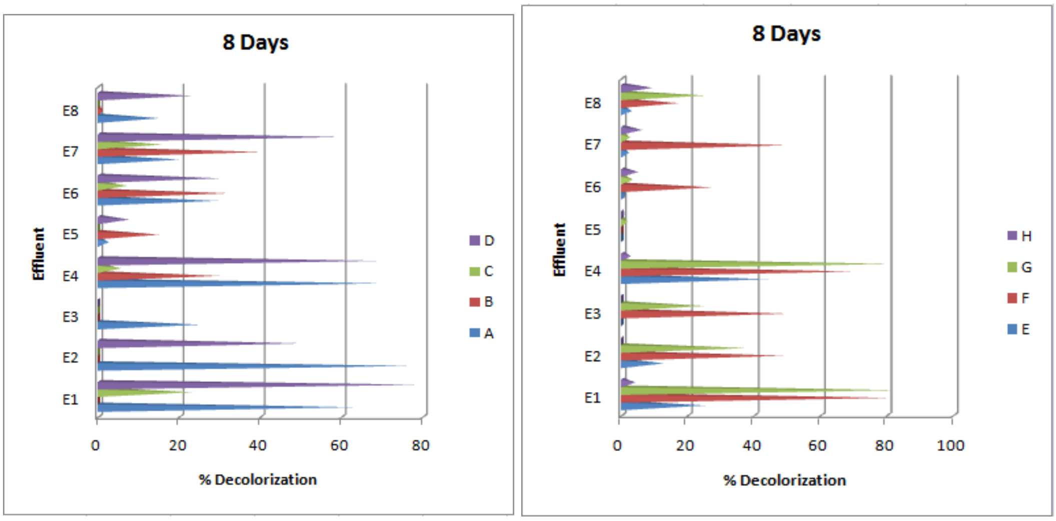


Figure 3: Percentage Decolorization in 8 Days

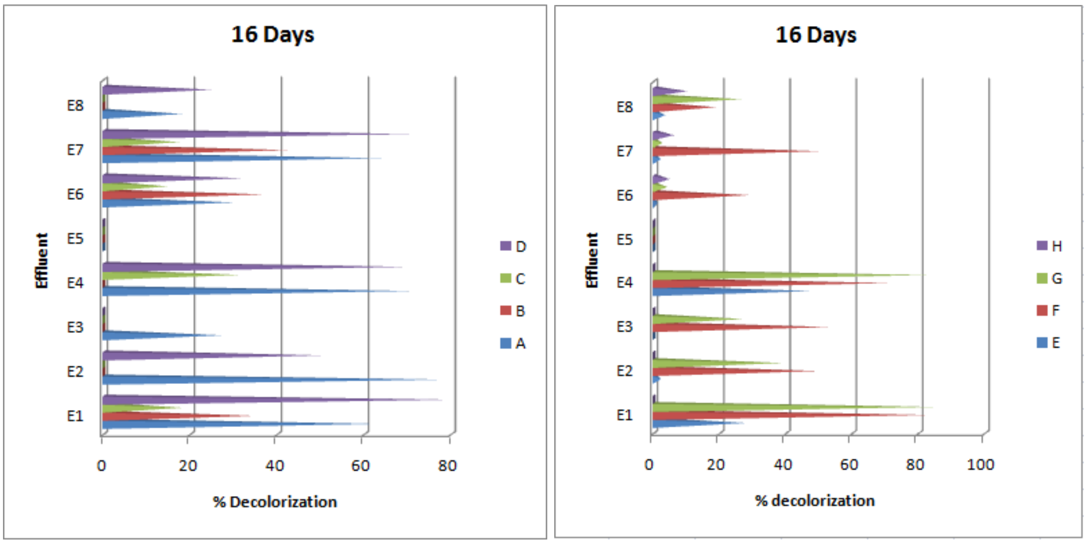


Figure 4: Percentage Decolorization in 16 Days

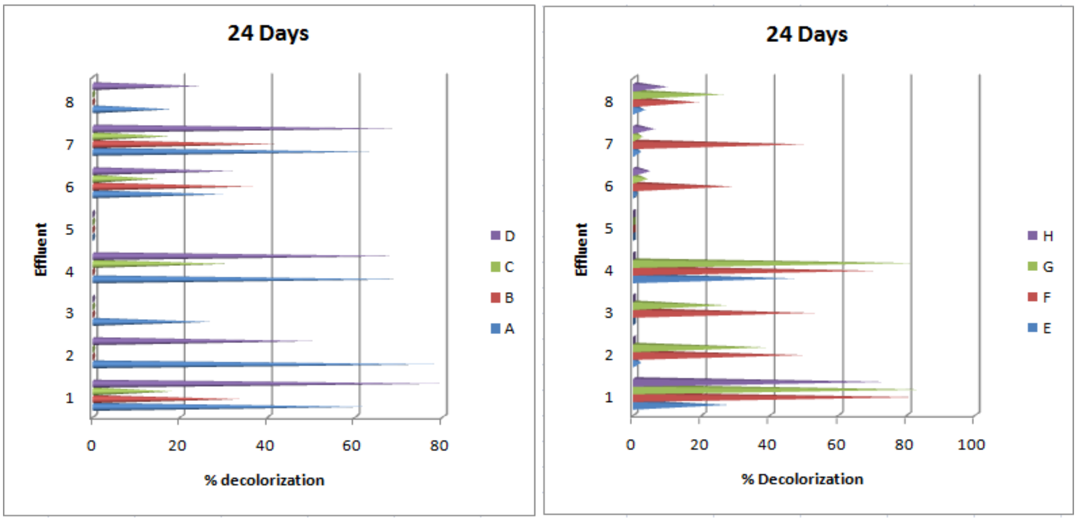


Figure 5: Percentage Decolorization in 24 Days

Effluent treatment with selected seaweed

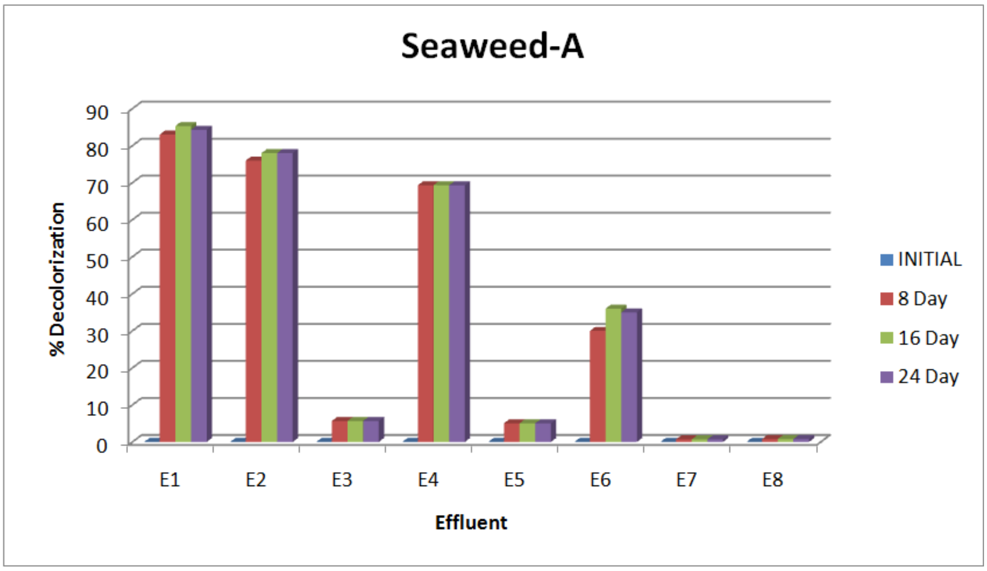


Figure 6: Percentage Decolorization observed in Seaweed-A

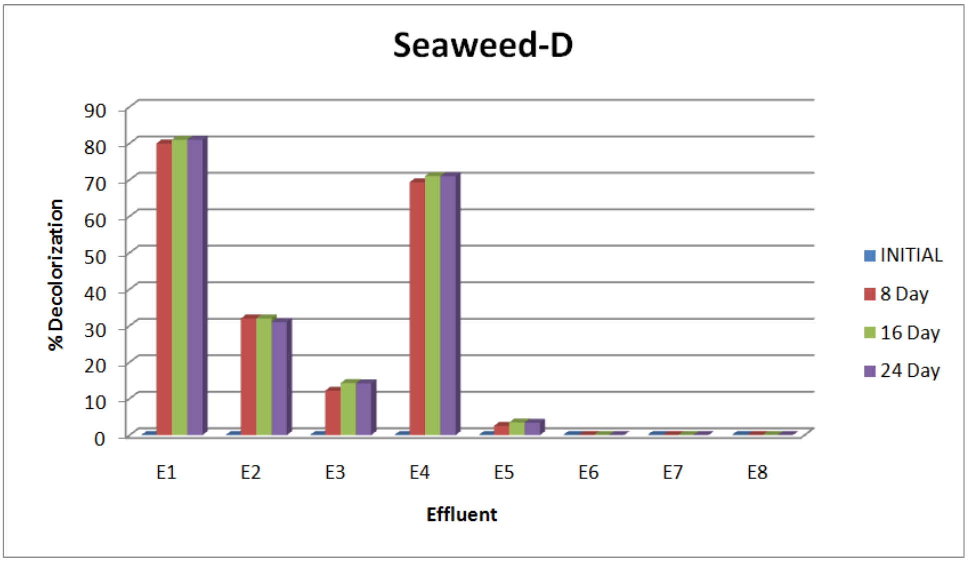


Figure 7: Percentage Decolorization observed in Seaweed-D

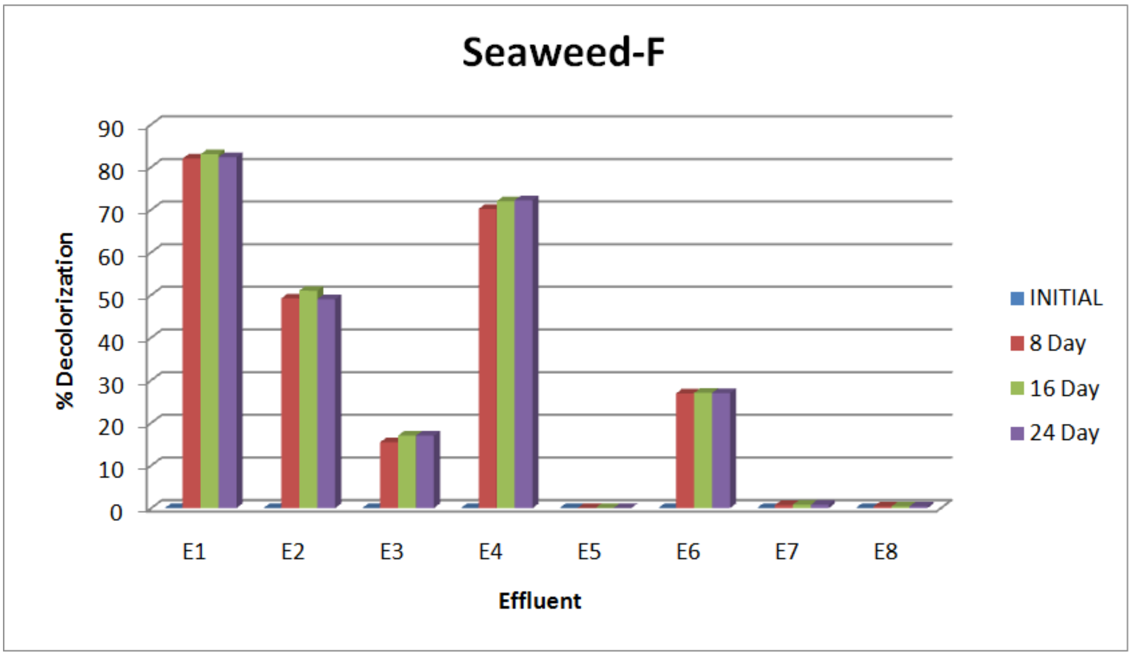


Figure 8: Percentage Decolorization observed in Seaweed-F

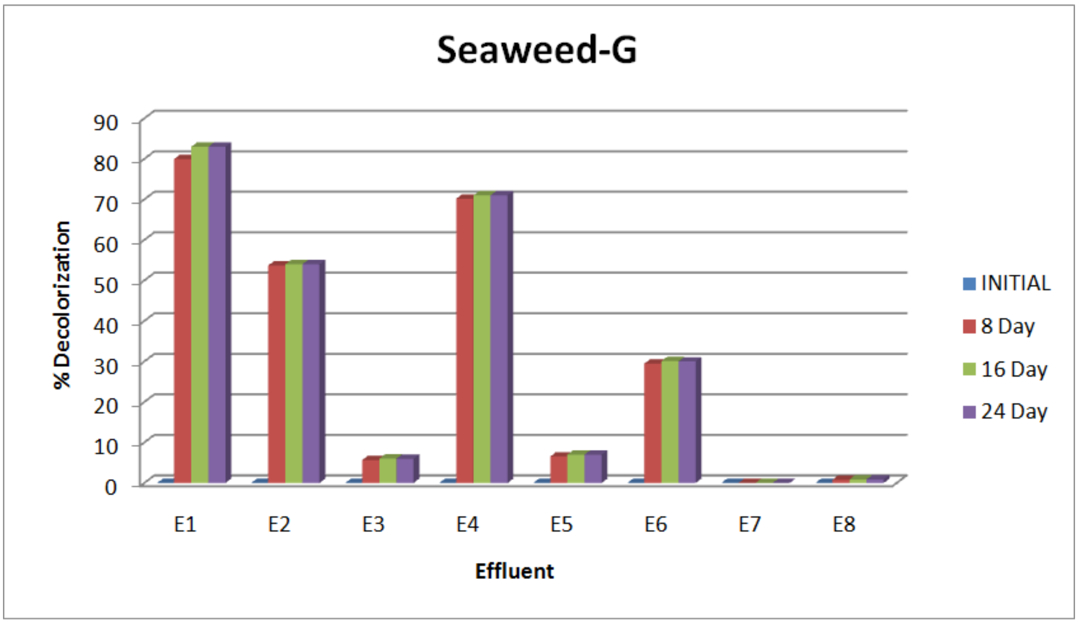


Figure 9: Percentage Decolorization observed in Seaweed-G

CONCLUSION

There are many treatment technologies available as physical and chemical treatment for waste water effluent but biological method is environment friendly and cheap method. In this research work the macroalgae shows potential result for decolorization of dyes effluent especially *Ulva lactuca* , *Caulerpa species* , *U. reticulate* and *U. clathrata Roth species* shows better efficiency which indicates it can be used as potential biomaterial to decolorize the dyes effluent. Further optimization of tolerance concentration of effluent, pH is required for commercialization.

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