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Research Article

Comparative Investigation of the Color Removal Efficiency of Different Mosses Species from Dye Solutions

Salih PAŞA[1,*], İbrahim DEMIR[2], Yasin AYTEPE[3]

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[1*]Afyon Kocatepe University, Faculty of Education, Department of Science, Afyonkarahisar, Türkiye; salihpasa@dicle.edu.tr
[2] Afyon Kocatepe University, Faculty of Arts and Science, Department of Chemistry, Afyonkarahisar, Türkiye; ibrahimdemir@aku.edu.tr
[3] Niğde Ömer Halisdemir University, Science Institute, Niğde, Türkiye; yasinaytepe@hotmail.com
*Corresponding author

Abstract: In this study, Acid Red 88 (Sodium 4-(2-hydroxy-1-naphthalenylazo) -naphthalenesulfonate) dye solution, which is used extensively in textile industry, was used. Dye removal efficiency determination studies were firstly performed by using three moss species, *Chiloscyphus polyanthus*, *Cinclidotus pachylomoides* and *Palustriella Falcata* (Hedw). In the case of comparison of color removal efficiency of *Chiloscyphus polyanthus*, *Cinclidotus Pachylomoides* and *Palustriella Falcata* (Hedw), it was seen that the species with the highest color removal efficiency with increasing dye concentration was determined for *Palustriella Falcata* (Hedw). Furthermore, the effect on color removal efficiency was determined at specific concentration, pH and under UVA (Ultraviolet-A) lamp light by using a photo reactor with these mosses species. It has also been detected that these three mosses can be used in photocatalytic studies. Thus, these three mosses species can be employed as color removing material both separately and with UVA light.

Keywords: Mosses; color removal; dye solution; wastewater; photo-reactor

Farklı Yosun Türlerinin Boya Çözeltilerinden Renk Giderme Etkinliğinin Karşılaştırmalı Olarak İncelenmesi

Özet: Bu çalışmada, tekstil endüstrisinde yaygın olarak kullanılan Asit Kırmızısı-88 (Sodyum 4-(2-hidroksi-1-naftalenylazo)-naftalensülfonat) çözeltisi, çeşitli mantarlarla boya giderimi amacıyla kullanılmıştır. Boya giderim etkinliği belirleme çalışmaları ilk olarak *Chiloscyphus Polyanthus*, *Cinclidotus Pachylomoides* ve *Palustriella Falcata* (Hedw) adlı üç karayosunu türü kullanılarak gerçekleştirilmiştir. *Chiloscyphus Polyanthus*, *Cinclidotus Pachylomoides* ve *Palustriella Falcata* (Hedw)'nın renk giderme etkinlikleri karşılaştırıldığında, artan boya konsantrasyonuyla en yüksek renk giderme etkinliğine sahip türün *Palustriella Falcata* (Hedw) için belirlendiği görülmüştür. Ayrıca bu yosun türleri ile foto-reaktör kullanılarak belirli konsantrasyon, pH ve UVA (Ultraviyole-A) lamba ışığı altında renk giderme etkinliği üzerindeki etkisi belirlenmiştir. Ayrıca bu üç karayosununun foto-katalitik çalışmalarda kullanılabileceği tespit edilmiştir. Sonuç olarak, bu üç yosun türünün hem ayrı ayrı hem de UVA ışığıyla renk giderici madde olarak kullanılabileceği tespit edilmiştir.

Anahtar Kelimeler: Yosun; renk giderimi; boya çözeltisi; atık su; foto-reaktör

1. Introduction

A rapid enhancement has been observed in industrial activities due to the hasty increase in population and the development of technology. As a result of these activities, environmental pollutants and various wastes were emerged implicitly that has become a major problem for life and nature [15; 35; 43; 45; 54; 64]. This situation has made it necessary to struggle environmental pollution more effectively. In particular, water resources have been negatively affected due to pollutants released directly and indirectly from industrial facilities to receiving environments. There are lots of industries that cause poisonous wastewater in the technological growing world. The textile industry appears to be one of the most environmentally "polluting" industries, in the case of considering the volume and composition of wastewater. The textile industry is considered to be the industry that pollutes the environment the most, considering its discharge volume and effluent content compared to other industrial sectors [70; 72].

Scientific studies and researches in recent years have focused on preventing the pollution of water resources and reducing the current water pollution load [12; 15]. Synthetic dyes are widely used in the textile industry and they contain color, high chemical oxygen demand (COD), pH, and toxic solids. Accordingly, they create important environmental problems. Industrial waste-derived organic dyes pose significant challenges, much like numerous other organic pollutants, given their elevated toxicity to aquatic life. This toxicity can lead to severe repercussions for human health, as well as for plants and aquatic ecosystems [28; 51]. Treatment of this type of wastewater with high amounts of organic pollution is difficult and problematic due to the high concentrations of complex dyestuffs and stable chemical structures of these organic compounds [48]. So, the treatment of wastewater containing dyestuff requires using advanced treatment techniques. [19; 44; 55; 73; 76]. The adsorption process is one of the most used methods that have been employed for the highest purification performance in recent years [6; 7; 13; 18; 19; 24; 30; 33; 39; 42; 48; 59]. Many researchers have focused on dye removal experiments in the last decades due to the decreased clean water resources [11; 14; 16; 32; 56; 62].

Additional measures are required to remove color, which is the main characteristic of textile dyeing industry wastewater. Because classical treatment plants can only achieve partial success in removing the color of wastewater [17]. When colored wastewater is discharged into the receiving environment, it absorbs the sun rays and prevents the light from reaching the deep part where photo-synthetic activity takes place. As a result, the primary production chain is disrupted in natural systems. Due to the presence of color in natural water bodies, the permeability of dissolved oxygen is impeded. The reduction of dissolved oxygen in water bodies seriously affects life in the aquatic environment [31].

Even at low concentrations, wastewater containing dyestuff poses an aesthetically undesirable problem when discharged into the environment [52]. Due to the complex chemical structures of synthetic organic pigments in dyes, they are resistant to bacteriological decomposition [37].

While some azoic dyes undergo anaerobic degradation, the decomposition process leads to the formation of aromatic amines, which can have highly adverse effects on the environment [10]. In our country, dyestuffs have the potential to accumulate at high rates in the environment, thanks to their resistance to breakdown due to the azo bonds they contain. Their stable structure under acidic and alkaline conditions and resistance to aerobic degradation, heat, and light make it challenging to purify dyes using conventional treatment methods. Consequently, it is known that in some cases, reactive dyestuffs exit the treatment process at high rates, up to 90%, without undergoing treatment [31].

Among diverse treatment technologies, (bio)adsorption stands out as a highly promising technique for treating dye-laden effluents, particularly when the adsorbent is affordable and easily accessible. Cost-effectiveness is a crucial factor in industrial applications, making this method particularly appealing [63]. Sorbents, especially based on biowastes have gained significance due to their comparable efficiency. Materials locally abundant in large quantities, such as natural substances, agricultural waste, or industrial byproducts, can serve as low-cost adsorbents. Various types of these economical adsorbents, including black carrot residues, wheat shell, grape stalk waste, sugarcane bagasse, maize bran, coconut and seed hull, cotton waste, apple wastes, orange residue, rice bran, and lemon peel waste, have been employed for heavy metal removal [1; 2; 3; 5; 8; 20; 26; 27; 36; 47; 60; 63; 66].

Mosses, representing approximately 23,000 species among all land plants worldwide, have found extensive application as indicators of heavy metal pollution in numerous studies. A key aspect of these investigations involves standardizing the adsorption capacities of various moss species sourced from different regions, facilitating an objective comparison of pollution levels [38]. Mosses serve as sensitive bio-indicators for detecting heavy metal contamination and offer several advantages as indicator organisms: (a) lacking an epidermis or cuticle, their cell walls readily allow penetration by metal ions; (b) devoid of organs for mineral uptake from substrates, they primarily obtain minerals through precipitation; (c) mosses reflect metal concentrations correlated with atmospheric deposition amounts [25; 65; 71].

Among various bio-indicators, mosses were among the first used to trace pollution in Europe [9; 23], particularly in industrial areas [21; 22; 34; 49; 74]. Their ability to mirror the chemical composition of the surrounding atmosphere stems from the absence of a cuticle or roots. Due to their ectohydric nature, mosses directly acquire most elements and nutrients from atmospheric deposition [13]. While several other sorbents have been tested as pollution monitors, the low production cost of mosses and their potential for reutilization, combined with their high adsorption capacity, provide moss with additional value. *Chiloscyphus polyanthus*, *Cinclidotus pachylomoides* and *Palustriella Falcata* (Hedw). were selected for the removability of dye from wastewater.

Chiloscyphus polyanthus is one of the commonest leafy liverworts to be found on rocks and other surfaces in watercourses and lakes where it usually grows at least partially submerged. It's unlikely to find it in chalk or limestone streams or in other base-rich water as it prefers water with a pH of 6.5 or less. Scapania undulata is a common associate [69].

Cinclidotus pachylomoides, also known as lattice tooth-mosses, are a genus of deciduous moss in the family *Pottiaceae*. They are aquatic mosses that live in and on streams and rivers and are widespread in the Holarctic - especially in Europe and the Mediterranean [75].

Palustriella Falcata, this large hook-moss is surprisingly variable in habit, although not habitat. It is a characteristic species of strongly basic and often stony flushes and fens. For the field bryologist, its stand-out field character (other than habitat of course) is its strongly pleated leaves. This places it in a small but select group of lookalikes and rules out others which do not have pleated leaves. Confusion is therefore only likely with *Sanionia* species, which rarely grow in wet habitats, *Hamatocaulis vernicosus*, a smaller, reddish species with more weakly pleated leaves, and the two other species of *Palustriella* [68].

In the current study, the dye removal of Acid Red 88 textile dye with three different mosses was made comparatively for the first time in the literature. In addition, it has been determined that these three mosses can be used both separately and in dye removal processes with photocatalytic oxidation.

1. Materials And Methods

1.1. Materials

Mosses used in the dye removal experiment were collected different regions. *Chiloscyphus Polyanthos* was got from Kayseri city, Soysalli village, near Buyuleyen Lake at an altitude of 1100 m with coordinates 38° 23'300"N - 35°21'580"E; *Cinclidotus Pachylomoides*, from Kayseri city, Yahyalı province near Kapuzbasi Waterfall at 750 m altitude and at 37°46'26"N-35°23'36"E coordinates; *Palustriella Falcata*, from Kayseri city, Yahyali province, near Kapuzbasi Waterfall at 750 m altitude and 37°46'26" N - 35°23'36"E coordinates.

The Acid Red 88 was obtained from Fisher Scientific (TCI, America), NaOH and HCl were obtained from Merck (KGaA, Germany) and purified accordingly [40]. UV-Visible spectra were recorded on a Shimadzu UV-160-A spectrophotometer. The pH adjustments were made with the Thermo ORION 3-STAR benchtop pH meter. Centrifugation was carried out with the Hettich Benchtop centrifuge ROTOFIX 32 A. Weighing operations were done with Shimadzu AUX320 Analytical Balance.

2.2. Methods

2.2.1. Biosorption Experiment

In this study, mosses were subjected to dye removal applications in a shaking water bath to assess their biosorption capabilities. Each experiment utilized 100 mL of dye solution with the biosorbent material, aiming to determine the optimal values for experimental parameters. Following each biosorption experiment, samples underwent centrifugation for biosorbent-solution separation, and the remaining dye concentration in the supernatant was quantified using a UV-Visible spectrophotometer [2; 3; 4; 63].

Biosorption studies were carried out at room temperature. A 250 mL capacity flask was used for this purpose. The optimum pH study was conducted to determine the pH at which the bryophytes used exhibited the best absorbance properties [5]. 100 mL dye solution at 50 ppm concentration was put into 250 mL flasks. The initial pH of each dye solution was individually adjusted to the desired values with 0.1 M HCl and 0.1 M NaOH. (Selected pH values are 3, 4, 5, 6, 7, 8, 9, and 10 for each dye). Then, 1 g of pre-prepared moss biosorbents was added to dye-containing flasks at the initial time. They were agitated at a constant speed at room temperature. [46; 57]. (Fig. 1) Samples were taken at particular time periods such 1^{st}, 2^{nd}, 3^{rd}, 4^{th}, and 5^{th} hour. After centrifugation separately, each sample was examined for dye removal activity. The dye concentration in the aqueous part was measured with a UV-Visible spectrophotometer to calculate the removal capacity. All procedures were applied in the same order for all three bryophyte types used in the experiments.

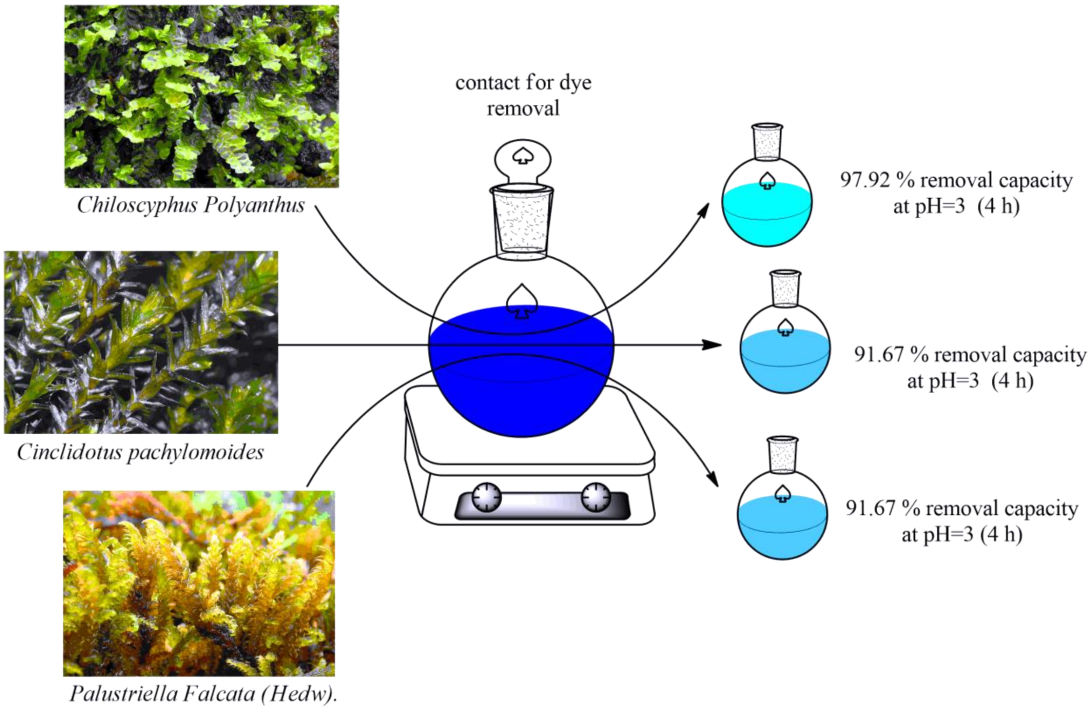

Figure 1. The illustration of biosorption experiment of dye solution by three mosses

The employed solutions at various concentrations, such as 50 ppm, 100 ppm, 200 ppm, 300 ppm, and 400 ppm were prepared from stock dye solution. The pH of the solutions was adjusted to pH=3 value, which was determined as the optimum working value. The 100 mL of each solution was taken into the flask, and it interacted with 1 g of algae in the erlenmeyer. The contact time was determined as 1, 2, 3, 4, and 5 hours in a shaker water bath at room temperature. Biosorption studies were performed for *Chiloscyphus Polyanthus*, *Cinclidotus Pachylomoides* and *Palustriella Falcata (Hedw)* at room temperature, at different dye concentrations, pH = 3, and agitation speed of 150 rpm/min.

2.2.2. Photo-reactor Process

A batch-type cylindrical Pyrex glass reactor (50 cm x 4.6 cm) with a capacity of 500 mL and a UV lamp system were used. The mixture in the reactor was provided by bottom-fed airflow. The UV lamp system used in the process is cylindrical in shape, made of flexible transparent material, with a height of 33.5 cm and a diameter of 14.3 cm. Its inner surface is covered with aluminum foil, allowing a maximum of 6 UV lamps to be placed hexagonally at equal intervals and the lamp type to be changed. Lamps with two different types of UV range were used in the lamp system. These lamps are classified as UVA (315-400 nm) and UVC (100-280 nm) according to the International Commission on Illumination.

The photo-reactor mechanism was formed by placing the lamp assembly on the shaker. The experimental setup used for photo-reactor experiments is shown in Fig. 2 below [41; 67].

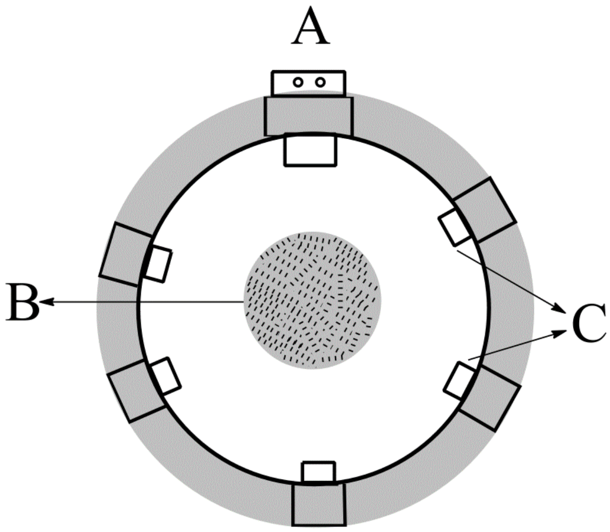

Figure 2. Schematic representation of the reactor system used by applying photoreactor and UVA light. (A: Switch On/Off, B: Erlenmeyer, C: UV A Lamps)

For photocatalytic application, 50 ppm and 100 mL of Acid Red 88 dyestuff solutions were used. It was placed in a 250 mL flask and the pH was adjusted to 3. Then, 1 gr *Chiloscyphus polyanthus*, *Cinclidotus pachylomoides* and *Palustriella Falcata (Hedw)* were added to these dye solutions. UV lamps were turned on and the reaction was started. The contact duration continued for 120 minutes. Solution samples were taken in every 20 minutes to calculate the removal capacity. They were centrifuged at 4000 rpm for 5 minutes. Then, the samples were placed in a 10 mm quartz cell in a spectrophotometer device and the absorbance was measured at 510 nm wavelength [61]. The color removal efficiencies over measured absorbance values were calculated using the equation below and the color removal graph was drawn.

$$\text{CRE } (\%) = \frac{\text{Co-Ct}}{\text{Co}} \times 100$$

CRE %: Color removal efficiency,
C_0: initial dye concentrations,
C_t: dye concentrations at t.

2. Results and Discussion

2.1. Determining the optimum pH

As can be seen from the values obtained in Table 1, the pH values determined to be pH=3 in all three bryophyte types which provided the best color removal performance and gave the most efficient absorbance results.

Table 1. *Color removal percentages of moss-dye solution at various pHs*

Plant	Time	pH 3	pH 4	pH 5	pH 6	pH 7	pH 8	pH 9	pH 10
Chiloscyphus polyanthus	1st hour	89.58	72.55	64.71	63.83	68.00	58.33	61.70	67.31
	2nd hour	93.75	74.51	66.67	65.96	68.00	64.58	61.70	67.31
	3rd hour	97.92	82.35	66.67	72.34	66.00	62.50	61.70	67.31
	4th hour	**97.92**	86.27	66.67	76.60	74.00	62.50	63.83	69.23
	5th hour	97.92	82.35	62.75	72.34	68.00	70.83	72.34	67.31
Cinclidotus Pachylomoides	1st hour	89.58	86.27	84.31	80.85	86.00	83.33	78.72	82.69
	2nd hour	89.58	86.27	86.27	80.85	86.00	85.42	78.72	82.69
	3rd hour	91.67	86.27	88.24	82.98	86.00	85.42	85.11	84.62
	4th hour	**91.67**	88.24	88.24	82.98	88.00	85.42	87.23	86.54
	5th hour	95.83	90.20	90.20	87.23	90.00	89.58	85.11	90.38
Palustriella Falcata	1st hour	85.42	72.55	62.75	53.19	66.00	56.25	53.19	59.62
	2nd hour	87.50	76.47	74.51	57.45	70.00	62.50	59.57	65.38
	3rd hour	91.67	78.43	76.47	59.57	72.00	64.58	63.83	67.31
	4th hour	**91.67**	78.43	78.43	61.70	72.00	66.67	65.96	67.31
	5th hour	93.75	88.24	86.27	76.60	84.00	79.17	76.60	80.77

3.2. Color Removal Application in Different Dye Concentrations

The dye removal efficiency of *Chiloscyphus polyanthus* was observed up to 89.58%. This result was obtained with 50 ppm dye solution at the end of the first hour. The color removal efficiency increased with the progress of application time. The highest color removal efficiency was recorded as 97.92% at the 3rd hour. In the case of examining the values in Fig. 3, the dye removal efficiency regularly increased in the first 4 hours.

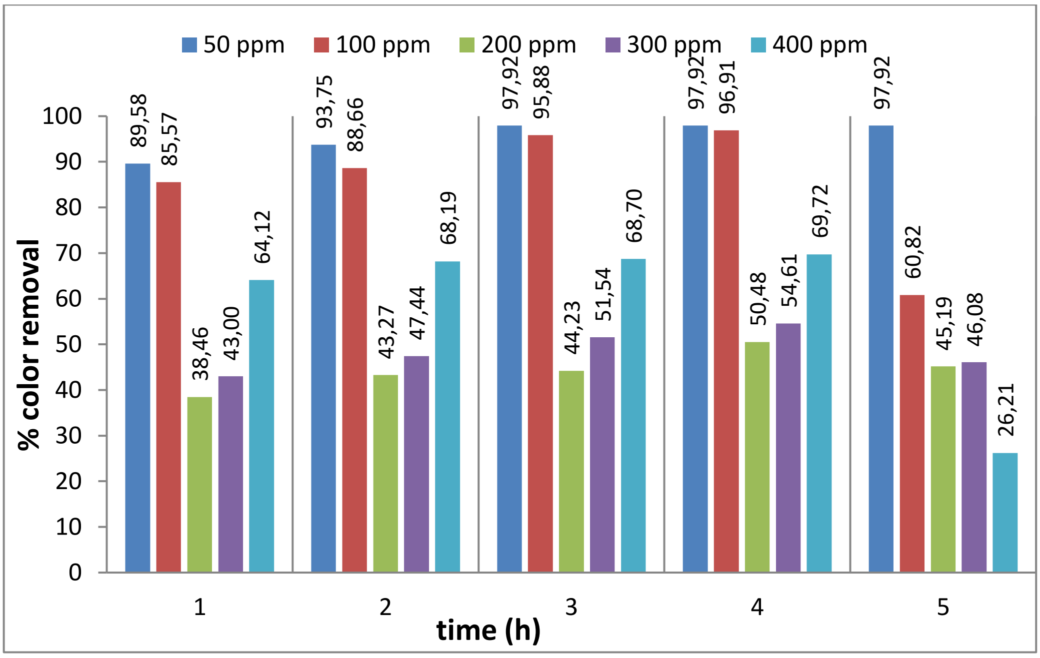

Figure 3. Color removal percentages graph of "*Chiloscyphus polyanthus*" versus dye concentration and time

In general, the *Chiloscyphus polyanthus* fungus shows increasing dye removal efficiency for Acid Red 88 with increasing concentration and contact time within the first 4 hours. However, after the 5th hour, it is thought that the dye adsorption reverses and desorption into the solution medium occurs.

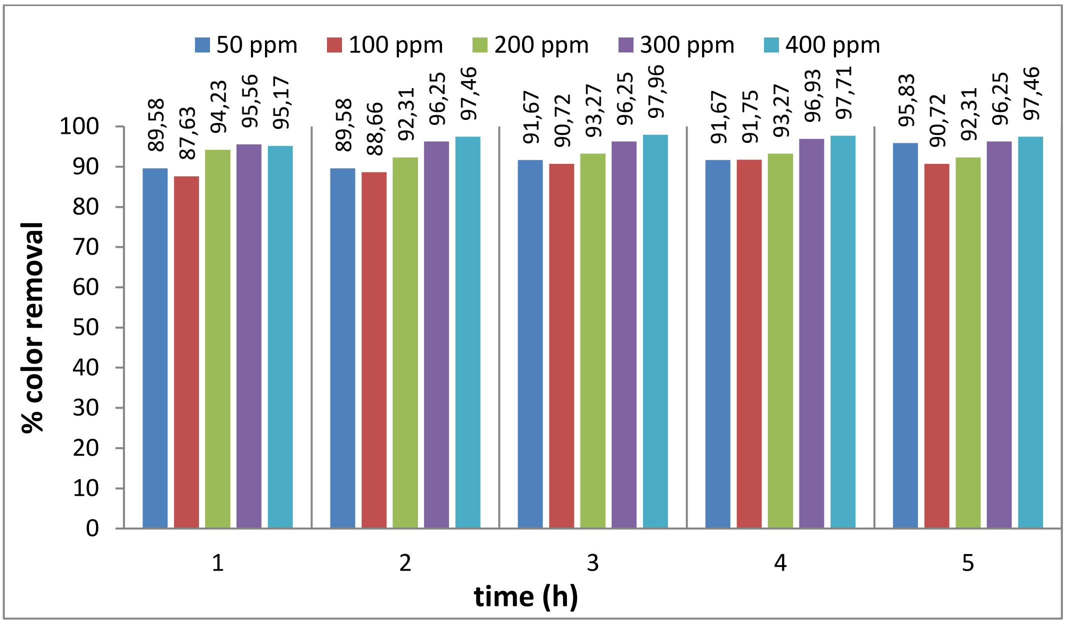

Figure 4. Color removal percentages graph of "*Cinclidotus Pachylomoides*" versus dye concentration and time

High color removal efficiency of *Cinclidotus Pachylomoides* was obtained as a result of the dye removal experiments conducted (Fig. 4). The color removal efficiency increased at low concentrations between 1st and 5th hours. The highest decolorization efficiency occurred with 50 ppm concentration at the end of the 5-hour experiment process. However, the maximum color removal was achieved in a shorter time for higher concentrations. The highest decolorization efficiency, 97.96%, was detected in 400 ppm solution after 3 hours. In addition, it is noteworthy that, generally, when the dye concentration is increased from 50 ppm to 400 ppm, the color removal efficiency is higher in the samples taken for each hour.

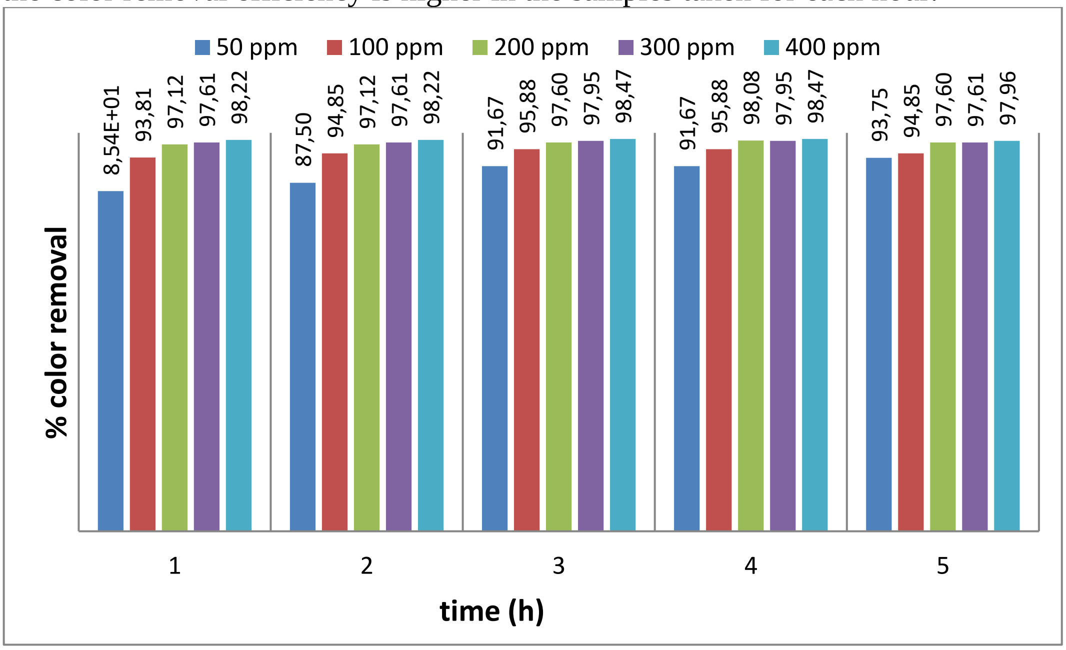

Figure 5. Color removal percentages graph of "*Palustriella Falcata*" versus dye concentration and time

Palustriella falcata (Hedw) type bryophyte has also shown significant color removal efficiency against Acid Red 88 textile dye (Fig. 5). 85.42% color removal efficiency was determined in 50 ppm solution after 1st hour. Besides, the color removal efficiency of *Palustriella falcata (Hedw)* was obtained 98.22% with 400 ppm solution at the same time interval. The highest color removal efficiency was acquired as 98.47% with 400 ppm dye solution at the end of 3rd hour.

In the case of comparison of three types of mosses, such *Chiloscyphus polyanthus*, *Cinclidotus Pachylomoides,* and *Palustriella Falcata (Hedw)*; the highest color removal efficiency during the time increasing versus concentrations was determined by employing *Palustriella Falcata (Hedw)*.

It can clearly be seen that, even in the first hour, the color removal efficiency with 400 ppm Acid Red 88 textile dye solution was found to be 98.22%. It can be evaluated that *Palustriella Falcata (Hedw)* has an effective color removable ability at higher dye concentrations in a short time. *Chiloscyphus polyanthus,* among these three mosses, achieved better color removal activity as 97.92% at 50 ppm than other moss species.

3.3. Photo-Reactor Studies

In order to determine whether *Chiloscyphus polyanthus, Cinclidotus Pachylomoides,* and *Palustriella Falcata (Hedw)* can be used with Photo reactor or not was investigated. The results of photo-reactor experiments with 50 ppm Acid Red 88 solution are given in Table 2.

Table 2. T*he color removal efficiency of "Mosses - dye solution" batches with photo-reactor*

Time	Chiloscyphus polyanthus + UV	Cinclidotus Pachylomoides + UV	Palustriella Falcata + UV
20 min.	87.50	81.25	75.00
40 min.	87.50	83.33	85.42
80 min.	93.75	87.50	85.42
100 min.	95.83	91.67	87.50
120 min.	97.92	93.75	89.58

Chiloscyphus polyanthus, Cinclidotus Pachylomoides and *Palustriella Falcata (Hedw) were employed i*n order to determine whether they could be used as water purification from dye solution or not. The results of photo-reactor experiments with 50 ppm Acid Red 88 are given in the Table 2.

Herewith, *Chiloscyphus polyanthus* demonstrated 97.92% color removal performance after 2 hours. Chiloscyphus polyanthus was found to have 93.75% color removal activity in the 2nd hour during the experiment performed without UV light alone. Photo-reactor application has increased the color removal efficiency approximately as 4.44%.

93.75% color removal efficiency with photo reactor for *Cinclidotus Pachylomoides* was obtained in the 2nd hour. It was observed that the moss alone performed 89.58% color removal activity in the 2nd hour without UV light. This shows that 4.65% increase occurred in photo-reactor investigation.

Lastly, *Palustriella falcata* showed 89.58% efficiency under UV light at the end of 2 hours. Furthermore, in the experiment conducted without UV light, it was observed that the

Palustriella falcata achieved 87.50% color removal ability after 2 hours. An increase just about 2.3% has happened in color removal efficiency.

3. Conclusion and Suggestion

The moss species such as *Chiloscyphus polyanthus, Cinclidotus Pachylomoides* and *Palustriella Falcata (Hedw)* were found low-cost biosorbent candidates for removal of Acid Red 88 dyestuff from wastewater. Therefore, they can be frequently used in the textile industry to purify water contaminants. The optimum pH at which biosorbents could get the highest efficiency was determined as 3. The increasing concentration from 50 ppm to 400 ppm of dye solution didn't significantly change the adsorption performance. It can be evaluated that selected mosses have an adequate adsorption capacity. However, increasing time had a slightly increasing effect on dye removal. But after the 5th hour, the adsorption process reversed to desorption in some cases. The highest color removal efficiency was determined as 98.47% with *Palustriella falcata* with 400 ppm dye concentration at the 3rd hour. Besides, *Chiloscyphus polyanthus* was found to be more effective in low concentrations. It was obtained 97.92% dye removal efficiency with 50 ppm at the 3rd hour.

These significant results can also be encountered from similar researches such; Nath et al. (2015) investigated the biosorption of malachite green by *Bacillus cereus*. They observed a maximum dye removal efficiency of 91% under optimal conditions: pH 5, biomass concentration of 0.5 g/L, initial dye concentration of 400 mg/L, and a contact time of 360 minutes [50]. Padmesh et al. (2008) investigated the biosorption of basic dyes (methylene blue and rhodamine B) using *Azolla filiculoides*. They found maximum biosorption capacities of 166.7 mg/g and 91.8 mg/g at 30°C, respectively. The authors concluded that the isotherm data fit best to Langmuir models, while the kinetics of adsorption followed the pseudo-second-order model most accurately [53]. Bhagavathi et al. (2016) examined the biosorption of basic dyes (methylene blue, malachite green, and basic blue 41) using effective microorganisms-based water hyacinth compost. They found that the maximum dye removal efficiencies for methylene blue, malachite green, and basic blue 41 were 98.9%, 98.4%, and 89.1%, respectively, with uptake values of 286.15 mg/g, 147.81 mg/g, and 139.29 mg/g. The optimized operational parameters for batch adsorption studies were observed to be pH (8, 8, and 7), dosage (4 g/L), and initial dye concentration (50 mg/L) [58]. Vijayaraghavan et al. (2015) explored the capability of *Gracilaria corticata* for removing crystal violet. They noted the highest sorption capacity at pH 8, with a biosorbent dosage of 5 g/L, and an initial dye concentration of 1000 mg/L [29].

It has also been studied that these three biosorbent species can also be employed in photo-reactor experiments for the purification of wastewater. The activities on color removal efficiencies were investigated with mosses by sending UVA rays to the solution, including 50 ppm dye concentration. They all enhanced the dye removal in different percentages: *Chiloscyphus polyanthus* 4.44%; *Cinclidotus Pachylomoides* 4.65%; and *Palustriella falcata* 2.30%. Thus, it can be concluded that these three mosses can remove color from wastewater solution both individually and with UVA light.

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Conflict of Interest

No potential conflict of interest was reported by the author(s).

Research and Publication Ethics Statement

The author declares that this study complies with research and publication ethics.

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