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Sustainable Isolation and Application of Plant Extract-Based Natural Dye for Bio-Dyeing of Silk Fabric

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Abstract: Green products such as plant pigments are gaining fame globally due to their excellent ayurvedic and biological characteristics. In this study, microwave (M.W.) has been employed to isolate colourant from black pepper (*Piper nigrum* L.), and bio-mordants have been included to obtain colourfast shades. Central Composite design (CCD) was formulated under response surface methodology (RSM) to optimize dyeing variables statistically. The results obtained after a series of experiments show that using 35 mL of irradiated acidic extract (RE = 6 min) of (pH4) in the presence of 1 g/100 mL of table salt as an exhausting agent at 65 °C for 55 min has demonstrated excellent colour depth (K/S) onto microwave-treated silk fabric (RS = 6 min). By applying bio-mordants, it has been found that walnut extract (1.5%), pinenut hull extract (1%), and orange peel extract (1.5%) are there before dyeing. In contrast, walnut extract (1.5%), pinenut hull extract (1.5%), and orange peel extracts (1.5%) after dyeing have shown colourfast shades of high strength. Comparatively, salts of Al^{+3} (1.5%), salts of Fe^{+2} (1.5%), and T.A. (tannic acid = 1.5%) before dyeing, while salts of Al^{+3} (0.5%) and salts of Fe^{+2} (2%) and T.A. (Tannic acid = 1%) after dyeing, have given good results. It has been observed that the use of M.W. radiation has not only improved the yield of dyes extracted from leaves when applied to silk fabric but also the inclusion of the optimum amount of bio and chemical mordants have resulted in moderate to good and excellent colour fastness ratings.

Keywords: bio-mordanting; microwave radiation; *Piper nigrum* L. silk; sustainability; walnut shell

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1. Introduction

Natural dyes are bio-colours recovered and extracted from natural sources, and their stability is solvent [1,2]. The demand for natural dyes has been increasing in the current moment due to rising environmental concerns [3,4]. Many chemicals, such as synthetic dyes, have mutagenic and carcinogenic components used daily in the synthesis formulation and finishing processes [5]. Moreover, synthetic dyes' effluents are hazardous for water bodies and agri-land as these effluents imbalance the pH, COD, and BOD levels of water [6]. Consequently, environmental organizations raise awareness that eco-labelled and ayurvedic-based products should be used to improve lifestyle [7]. Therefore, green products such as the demand for natural dyes can be used as an alternative to synthetic colourants.

On the other hand, numerous studies have also shown that natural dyes are nontoxic and sustainable and have no disposal issues. Their effluents are nontoxic, biodegradable, and could be reused as a bio-fertilizer to improve crop growth [8]. Secondary metabolites of the plant extracts are used as tonic and herbal system that transfers their biological activities when employed on matrices such as silk, wool, and cotton [9,10]. The most vital factor is that the cultural heritage with a broad spectrum of colours is now being revived as state-of-the-art for the colouration of different substrates [11,12]. Besides the advantages of natural dyes, these have limitations, such as poor colour yield, low fastness rating, reproducibility of shades, etc. [13]. To overcome these limitations, mordants such as salts of Fe^{2+}, Al^{3+}, Cr^{1+}, and Cu^{1+} are used, but their applications also create environmental hazards [14]. Nowadays, plant-based functional isolates may be alternatives to toxic mordants have been included, which not only have excellent herbal and biological characteristics, but also have added value to the bio-colouration of fabric dyeing [15] to form a variety of firm and fast shades.

Extracting natural dyes from plant sources is a significant step in natural dyeing. Previously, various conventional methods, such as heating, stirring, soxhlet, etc., were used to extract natural colourants from plants [16]. These methods have a low yield by consuming a lot of solvent volume, time, labour, and energy, or may destroy the colouring behaviour of the actual molecules [17]. However, scientists have now found modern extraction methods induced by enzymes, radiations, supercritical phenomena, etc., to obtain maximum colour yield. They are also modifying the fabric surface to appraise its uptake behaviour. Microwaves have unique characteristics among these modern extraction methods [18,19]. Such acts uniformly heat, penetrate the material, enhance the effective yield of the extract by a solid-liquid mechanism, and ultimately enhance the colour strength [20]. These microwaves transfer energy into solvent molecules by which, when they collide with the plant cell walls, the high yield of colourants by this fast mass transfer kinetics is observed [21,22]. These M.W. rays also tune the fabric surface by penetrating its void, causing scaling on its surface. As a result of this surface modification, the fibers' sorption behaviour is enhanced, and upon dyeing high yield in terms of K/S is achieved. To overcome fastness-related issues, plant-based functional colouring matter has been used as bio-mordants despite toxic mordants [23]. These plant-based metabolites not only have excellent herbal properties but also develop new colourfast shades.

Black pepper (*Piper nigrum* L.), the king of spices, belongs to the family of Piperaceae [24–26]. Its leaves contain phytochemicals such as alkaloids, flavonoids, anthraquinone, phenolic compounds, etc., in their specific parts, such as flowers, leaves, bark, fruits, seeds, and roots [27,28]. The main phytochemicals in leaves possess antioxidant, antibacterial, antifungal, anti-carcinogens, anti-diabetic, anti-inflammatory, and insecticide properties [24,29–31]. In the traditional medicine system, black pepper is used to treat several diseases, such as liver, stomach, digestive, aphrodisiac, etc., and respiratory tract diseases such as asthma, cough, bronchitis, analgesic for muscular pains, obesity, worms, piles, congestion, paralytic, sinusitis, arthritic disorder, fever, snakebite, cholera, and diarrhea [27,32,33]. Here, in our studies, we have used extracts for bio-dyeing of proteinous fabric such as silk. Silk is an animal protein obtained from cocoons, where the main silk fibre part is fibroin with amido units (-N.H., -C=O) as the main functional site.

The novelty of this study is that, until now, no one has explored the colouring behaviour of black pepper leaves as a natural dye source. In exploring its colouring behaviour, no one has included M.W. as a sustainable isolation tool and plant-based extracts as bio-anchors to obtain excellent quality colourfast shades.

In this study, our research group has selected a suitable medium and irradiation time to obtain excellent colourant yield in terms of k/s onto fabric, and optimised dyeing variables using central composite design (CCD) under response surface methodology (RSM) as a statistical approach as a function of K/S value. Using selected irradiation extraction and dyeing conditions, shades have been developed after applying selected sustainable chemical and plant-based extracts bio-anchors. For future studies, the kinetics and thermodynamics parameters, as well as the exhaustion of dye (%) before and after treatment and other physical analysis of fabrics such as silk and cotton nylon, will be made.

2. Materials and Methods

2.1. Collection of Materials

To make crude powder, dried leaves of black pepper (*Piper nigrum* L.) from the Herbal Store (H.S.) in Faisalabad were pulverized coarsely. The textile market provided ready-to-dye silk fabric (GSM = 60 g/m^2). Commercial available chemicals such as Conc. HCl, Formic acid, salt of Al, Fe, and Tannic acid (T.A.) chemicals were used for the extraction, colouring, shade formation, and evaluation processes.

2.2. Dye Extraction and Irradiation Processes

To extract the natural dyes, the aqueous dye extract was obtained by mixing 4 g of powder with 100 mL of water on boiling for 45 min. Similarly, the acidic extract was prepared by boiling 4 g of black pepper leaves powder with 100 mL of water (1% HCl *v/v* %) for 45 min. Then, after obtaining the colour in aqueous and acidic media, the extracts and silk fabrics were irradiated using an M.W. source for 2–10 min. These M.W.-treated and untreated black pepper extracts have been used to dye M.W.-treated and untreated fabric by maintaining an extract-to-fabric ratio of 25:1 at 75 °C for 45 min.

2.3. Dyeing Conditions Optimization

Response Surface Methodology (RSM) as a satisfied model was employed for following dyeing variables: pH = (3–7), salt= (0.5–2.5 g/100 mL), dyeing temperature = (55–95 °C), and dyeing time= (25–65 min). Thirty-two trials narrated in Table S1 were conducted as per central composite design given, and the results were recorded and put under analysis to see the significance of the results.

2.4. Mordanting

To create colourfast new shades at optimal conditions, pre-and post-mordanting using electrolytes of chemical mordants Al and Fe have been utilized at 80 °C. Furthermore, plant phenolics isolated from walnut shells (*Juglans regia*) and pine nut shells (*Pinus pinea* L.), and orange peel (*Citrus sinensis*) as a source of bio-mordant have also been used to compare fast colour shades by these two types of chemical For this purpose, 35 mL from 0.5–2.5 g/100 mL of each mordant extract (bio-mordant) or solution (chemical) has been employed at 65 °C for 55 min, keeping an M:L of 1:35. The process of shade development was adapted according to Adeel et al. (2022b) and Yameen et al. (2022).

2.5. Evaluation of Fabrics

The colour strength and shade variable (L^*, a^*, b^*) have been assessed in Colori-spectrophotometer CS-410 (China), having an illuminant of D65 with a 10° observing angle. The rating for Colourfastness properties of selected dyed mordanted fabrics and the un-mordanted dyed fabric has been evaluated using ISO standard methods of light, rubbing, and washing fastness at the Department of Applied Chemistry, Government College University Faisalabad Pakistan.

3. Results and Discussion

The problem of poor extraction yield in natural dyeing has tried to be overcome by local methods, but these processes need a lot of solvent time and labour with low colourant yield [14,18]. Now energy- and time-efficient methods such as irradiation tools such as M.W. rays are under use to obtain a high yield. In this study, M.W. rays have been selected to enhance silk fabric's sorption behaviour and break the clusters of functional molecules into small molecules. These rays tune the fabric surface and enhance uptake behaviour [10,15]. For extraction, these rays enable small molecules, after rupturing the plant wall, to interact with solvent molecules rapidly. This transfer mechanism through effective solid-liquid interaction enhances the rate of colour transfer into a selected solvent, which upon Application furnishes high yield (K/S) onto fabric [16,17]. Using water-based extract, the results given in Figure 1a reveal that, after treatment of extract and fabric for

6 min, silk dyeing has given a good colour yield (K/S = 6.7901). The shade obtained is darker (L^* = 55.44) and reddish yellow in tone (a^* = 5.73; b^* = 23.55). Before treatment, the colour strength was very low (K/S =2.9164) and the shade was brighter (L^* = 65.33), less red but more yellow in tone (a^* = 4.49; b^* = 21.05) (Table S1). Using an acidic extract, the colour depth (K/S) was increased. The results shown in Figure 1b show that silk dyeing produced excellent yield (K/S = 13.599) when both acidic extract and fabric were treated for up to 6 min and utilized. The shade obtained was darker and much more reddish yellow in hue (L^* = 54.31; a^* = 8.80; b^* = 28.36). Before treatment, silk dyeing gave a better yield (K/S = 8.5685) than fabric dyed with water-based extract. The shade was darker and more reddish yellow in hue (L^* = 56.80; a^* = 8.60; b^* = 26.09). Statistical analysis (2-Way Anova) given in Table 1 reveal that irradiation of both fabric and aqueous extract should be conducted as the results are significant (p = 0.070) in terms of sample codes i-e RE/RF, NRE/NRF/ NRE/RF etc. Similarly for acidic extract, the irradiation of both fabric and extract has given highly significant results (p = 0.002). This good colour depth could be explained by the action of M.W. rays, which transfer their energy through ionic conduction and the polar nature of the solvent [34–36]. These solvent molecules make levelled and uniform energy transfer to rupture the plant boundary upon collision [19,21,22]. Hence, upon rupture of the plant cell wall, the molecules have interacted with solvent, which are used for dyeing of the M.W.-treated surface tunned fabric, and show maximum yield. Therefore, it can be recommended that dyeing of M.W. treated silk should be conducted with M.W.-treated acidic extract of black Pepper leaves powder (BPLP) extract.

Table 1. Statistical analysis of variance (Two-way Anova) for the optimization of aqueous and acidic extract.

Analysis of Variance					
Aqueous					
Source	**DF**	**Adj SS**	**Adj MS**	**F-Value**	***p*-Value**
time	1	0.5293	0.5293	0.40	0.534
sample	1	4.7923	4.7923	3.62	0.070
Error	22	29.1490	1.3250	———	———
Total	24	34.4706	———	———	———
Model Summary					
S	R-sq	R-sq (adj)	sq(pred)	———	———
1.15107	15.44%	7.75%	0.00%	———	———
Acidic					
time	1	16.37	16.366	2.45	0.131
sample	1	79.42	79.420	11.91	0.002
Error	22	146.67	6.667		
Total	24	242.46	———	———	———
Model Summary					
S	R-sq	R-sq(adj)	R-sq(pred)	———	———
2.58201	39.51%	34.01%	19.91%	———	———

After sustainable treatment and extraction medium for encapsulation of colourant from black pepper leaves powder, the dyeing factor has been optimised. For this purpose, a set of 32 experiments have been employed by varying five factors, i.e., extract volume, pH, salt as an exhausting agent, dyeing time, and dyeing temperature as contact levels (Table S3). The results in Table S3 show that 35 mL of irradiated extract with 1 g/100 mL of salt solution as per o.w.f. have produced excellent yield when used for 55 min at 65 °C. The low factors of hand volume and salt show that M.W. rays are cost–time effective. After irradiation of fabric and extract, the modification has enabled efficient yield under mild conditions. Table S3 shows that the model is fit (p = 0.01) and linear (p = 0.016), and the role of extract volume with salt as exhausting agent and time as contact level, i.e., for 55 min, is significant. In one way, only the extract nature plays a role, but in a two-way interaction, salt with time and volume, and volume with time and salt, is contributing to obtaining a high yield. Hence, three-factor, i.e., extract volume (35 mL), salt as exhausting agent (1 g/100 mL) and contact time (55 min) have been reduced after the application of M.W. rays, and it shows that the use of M.W. rays is a volume-, cost-, and time-effective tool for silk dyeing using black pepper leaves extract. Hence, it is revealed that dyeing should be performed at 65 °C for 55 min. Using 35 mL of pepper leaves extract at pH 4 obtains a good yield.

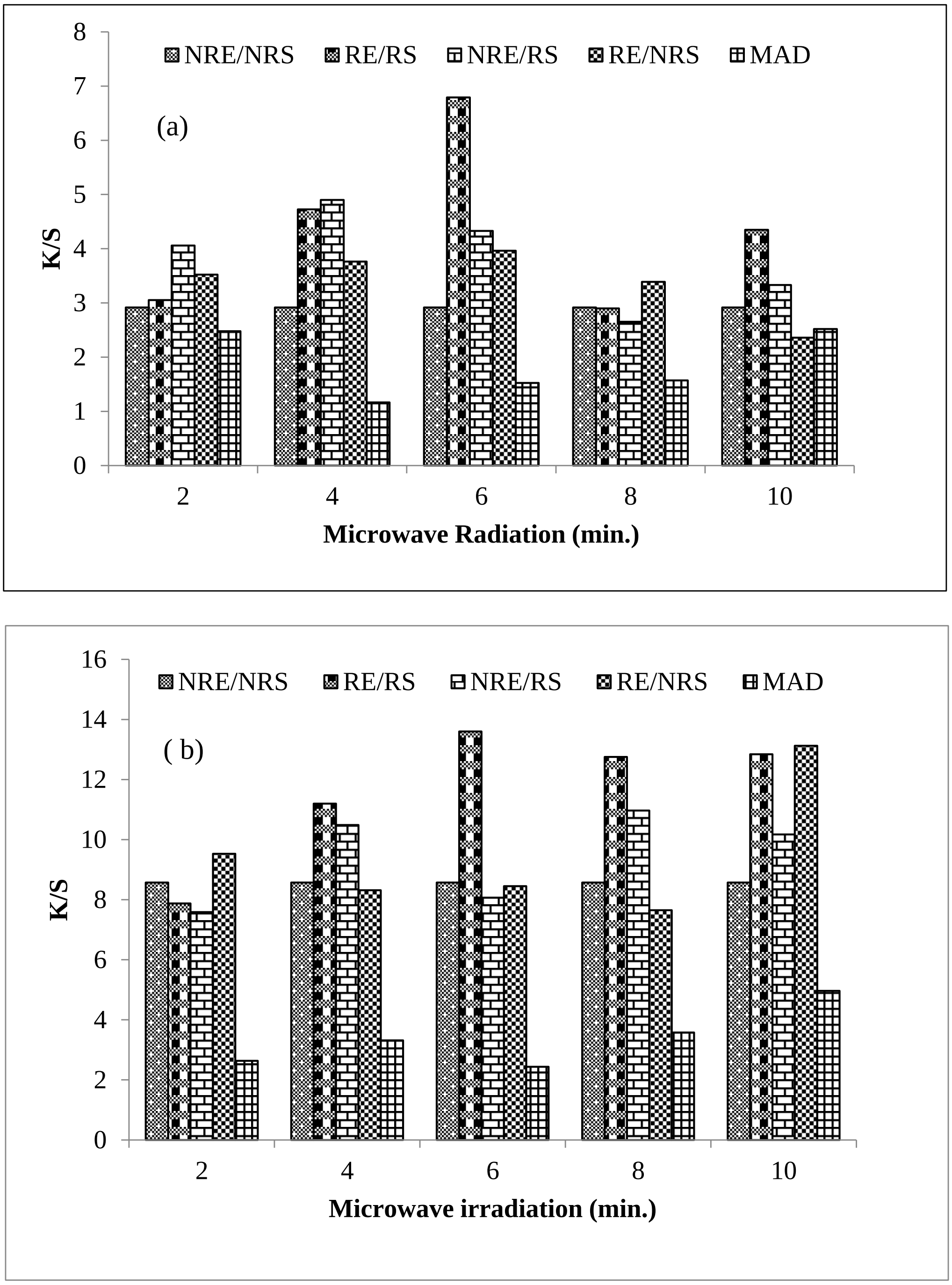

Figure 1. Utilization of microwave-irradiated and un-irradiated aqueous (**a**) and acid extract (**b**) of black pepper for dyeing irradiated and un-irradiated silk textiles.

Bio-mordanting results are given in Figure 2a, which showed that 35 mL of extract from 1.5 g/100 mL of walnut shell powder, orange peel powder, and 1.0 g/100 mL of pinenut shell powder were given high yield onto fabric before dyeing. However, after dyeing 35 mL of extract from 1.5 g/100 mL of pinenut, walnut, and orange peel, it developed high strength onto silk fabric. Overall, 1.5 g/100 mL of orange peel before dyeing and 1.5 g/100 mL of the walnut shell after dyeing have given excellent results. The colour variables shown in Table 2 reveal that most of the shades are dark reddish yellow in hue, but before dyeing (pre-bio-mordanting), walnut extract (1.5 g/mL) has given a darker shade of reddish yellow hue (L^* = 57.66; a^* = 8.11; b^* = 25.84). Similarly, orange peels (1.5 g/100 mL) have also given a darker reddish yellow hue (L^* = 59.05; a^* = 5.89; b^* = 28.40). However, 1.0 g/100 mL of pine nut extract has developed a bright or shade of reddish yellow tone (L^* = 63.76; a^* = 6.71; b^* = 22.42). Similarly, Figure 2b displayed that, after dyeing, 1.5 g/100 mL of walnut showed a dark but more reddish-yellow tone (L^* = 57.57; a^* = 10.25; b^* = 22.56), while 1.5 g/100 mL of pine nut hull extract has also shown a bright reddish yellow hue (L^* = 63.98; a^* = 5.45; b^* = 22.38). Similarly, 2 g/100 mL of orange peel has given a dark reddish-yellow hue (L^* = 61.18; a^* = 5.91; b^* = 23.11). This good strength might be due to the interaction of -C=O, -O.H. from walnut, -O.H. from catechin of pinenut shells, and -O.H. of flavonoids from the orange peel with -C=O, -N.H. of fabric, and -O.H. of flavonoids from black pepper extract (BPE). Here, the additional interaction, i.e., H-bonding, is developed, which allows colourant molecules to bind with bio-mordant the fabric [37–39]. Before and after the dyeing, this application gives new tints of high strength and good fastness ratings. Overall, bio-mordanting with orange peel extract and, after dyeing, bio-mordanting with walnut extract should be performed to obtain colourfast shades (Figure 3a,b).

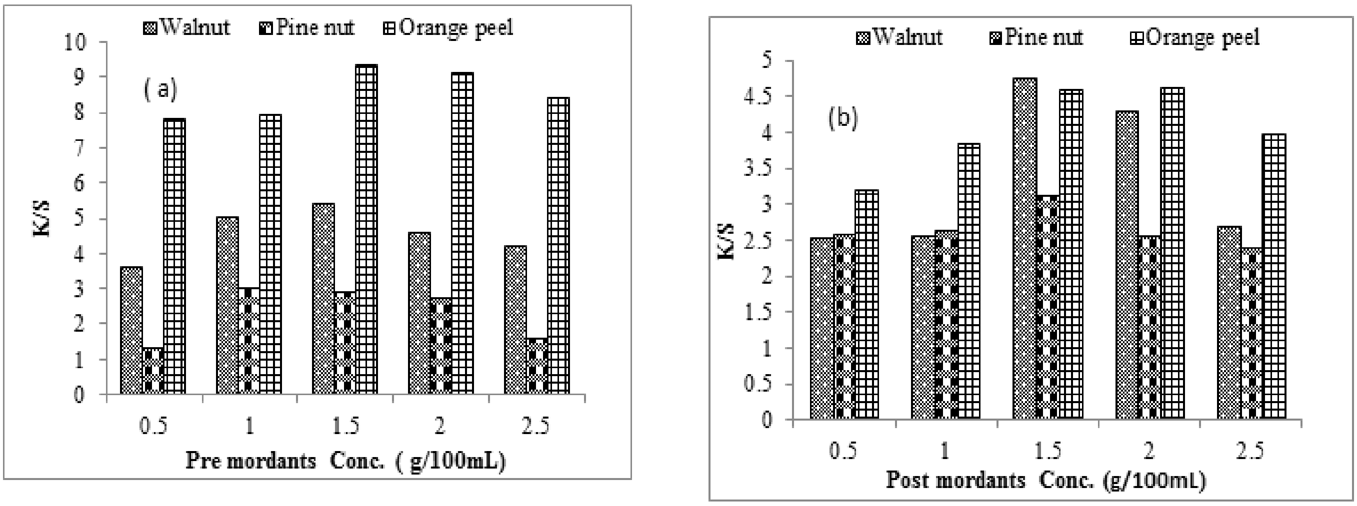

Figure 2. (**a**) Pre-bio-mordant of silk with black pepper extracts at optimized conditions. (**b**) Post-bio mordants of silk with black pepper extracts under selected conditions.

In comparison, chemical mordanting has also been performed. Many electrolytes are present that can act as additive [34,35], but owing to sustainability, green processing, and eco-friendly formations [36,40]. The salts of Al^{3+} and Fe^{2+} and tannic acid have been used. Before dyeing, 35 mL from 1.5 g/100 mL of Al^{3+} salt and Fe^{2+} salt and 35 mL from 1.5 g/100 mL tannic acid have produced a high yield (Figure 2a). After dyeing, 35 mL from 0.5 g/100 mL of aluminum salt, 2 g/100 mL of iron salt, and 1 g/100 mL of tannic acid (T.A.) have given a high yield. Hence, before dyeing, 35 mL of 1.5 g/100 mL of iron salt and 35 mL from 1 g/100 mL of tannic acid furnished high strength. Comparatively, among chemical mordants, iron salt and bio-mordants' orange peel extract have developed a high yield on silk fabric when employed at given conditions. The rating of colourfastness reveals that using the selected chemical and bio-mordants, light fastness has been improved, and washing and crocking have been enhanced from poor to good. Here, not only have the natures of the fabric, dyes, and metal used played their role in creating coordinate covalent bonds to form a complex, but also conjugation in bio-mordant molecules, flavonoid O.H., and amido linkage via additional H bonding have developed new colourfast gamutes [39–43]. The proposed bio-mordant and dye metal dye interaction have been shown in Figure 4a,b. Hence, the applications of mordant solution were reduced before and after dyeing, which shows that M.W. rays have also reduced the mordant amount after modification of the extraction and dyeing process.

Table 2. Colorfastness ratings of chemical mordant and bio-mordanted silk fabric dyed using an acid-solubilized extract of black pepper.

Mordant Name	Mordant Conc. %	*L**	*a**	*b**	Light Fastness	Washing Fastness	Rubbing Fastness	
							Dry	Wet
Pre-Mordanting								
No Mordant	0.00	54.31	8.80	28.36	4	3/4	4	3/4
Al^{3+}	1.5	69.55	5.46	23.21	5	4/5	4/5	4/5
Fe^{2+}	1.5	41.25	6.96	17.99	5	4/5	4/5	4/5
T.A.	1.5	46.96	11.98	18.28	5	4/5	4/5	4/5
Walnut	1.5	57.66	8.11	25.84	5	4/5	4/5	4/5
Pine-nut	1	63.76	6.71	22.42	5	4/5	4/5	4/5
Orange Peel	1.5	59.05	5.89	28.40	5	4/5	4/5	4/5
Post-Mordanting								
Al^{3+}	0.5	74.45	2.91	16.29	5	4/5	4/5	4/5
Fe^{2+}	2	45.08	2.06	11.72	5	4/5	4/5	4/5
T.A.	1	49.82	9.84	20.33	5	4/5	4/5	4/5
Walnut	1.5	57.57	10.25	22.56	5	4/5	4/5	4/5
Pine-nut	1.5	63.98	5.45	22.38	5	4/5	4/5	4/5
Orange Peel	1.5	61.18	5.91	23.11	5	4/5	4/5	4/5

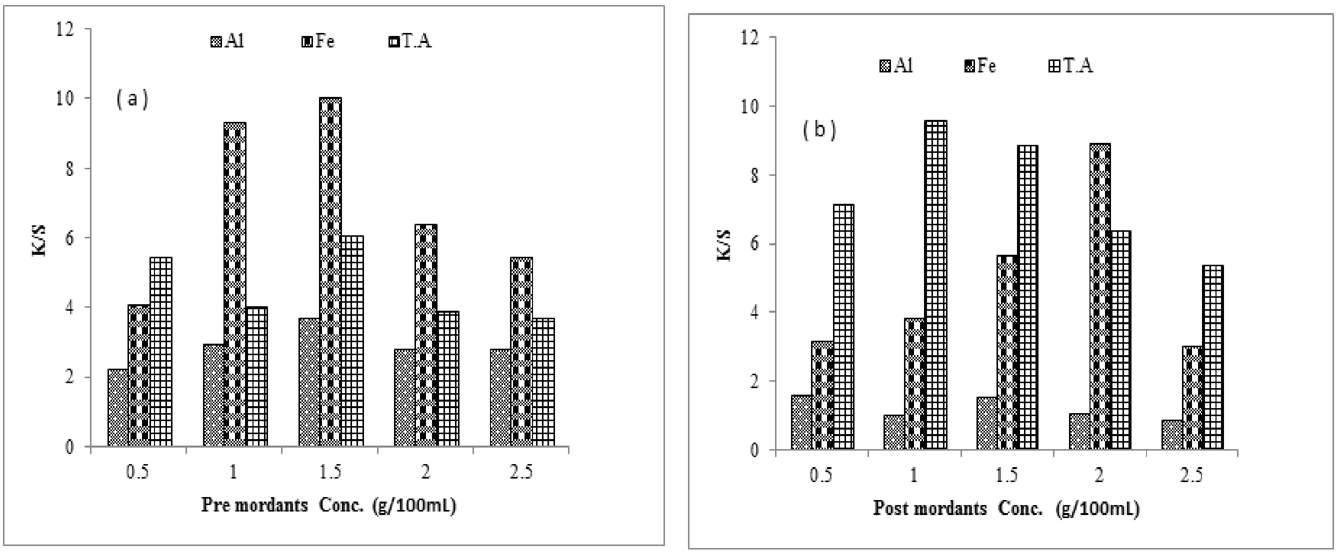

Figure 3. Pre-chemical (**a**) and post-chemical mordanting (**b**) of silk with black pepper extracts under selected conditions.

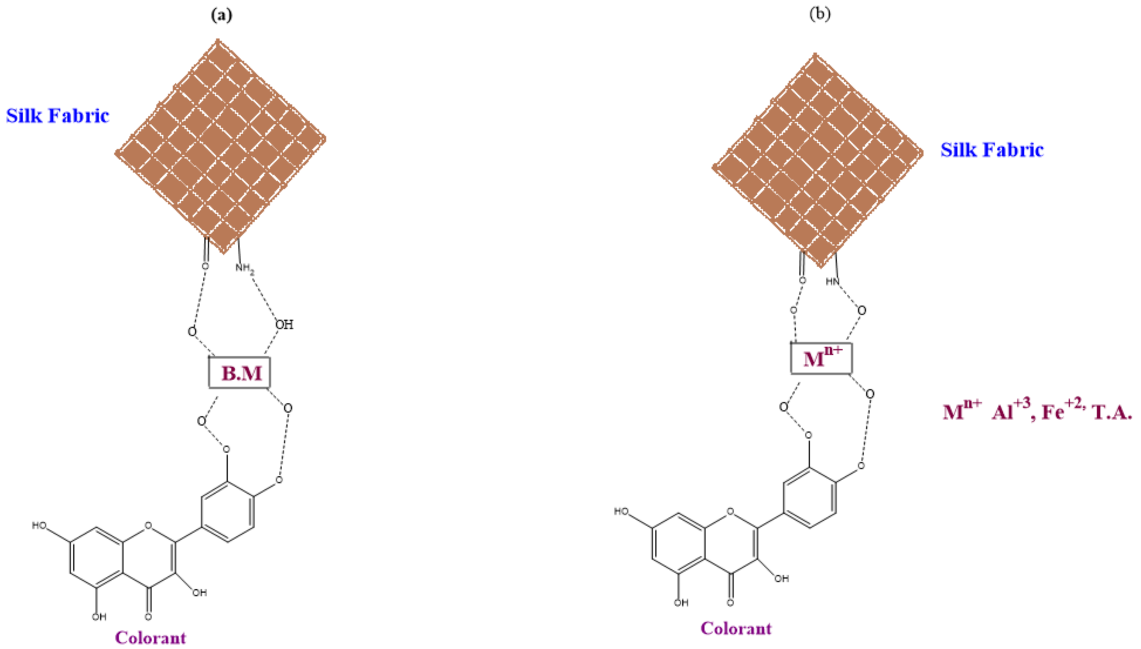

Figure 4. Proposed interaction of bio-mordant (**a**) and chemical anchor (**b**) with dye and silk fabric.

4. Conclusions

Sustainable products such as natural dyes are gaining worldwide fame because of their excellent ayurvedic nature. Considering these advantages of natural dyes, the present study has been conducted to see the effect of black pepper (*Piper nigrum* L.) as a source of natural colourant for dyeing silk fabric. The results show the irradiation of silk fabric (RS = 6 min) for dyeing with irradiated acidic extract of black pepper (RE = 6 min) and filtrate containing 1% of table salt at 65 °C for 55 min as an exhausting agent. Then, sustainable mordanting has given acceptable shade strength and fastness ratings compared to chemical mordanting. Statistical optimisation of the dyeing variable by performance formulated by Central Composite Design (CCD) under response surface methodology shows that M.W. treatment has reduced not only time and temperature but also the extract volume and salt amount. It is concluded that the M.W. treatment has excellent sustainable efficacy in isolating colourant from black pepper powder for silk dyeing. In contrast, adding new bio-mordants has made the process more sustainable and eco-friendly.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/coatings13010112/s1, Table S1. Color coordinates of bio-dyed silk fabric at selected conditions; Table S2. Trials for experiments under central composite design through response surface methodology; Table S3. Response Surface Regression: K/S Versus pH, Volume, Time, Temperature & Salt using acidic extract.

Author Contributions: A.T. is an M.Phil student who has conducted experiments with A.A. S.A. is the major supervisor, whereas F.-u.-R. has provided scientific and technical guidance for the smooth running of experiments for results. T.A. has analysed the data statistically. M.A.Q. and M.H. helped to write the manuscripts as per the results achieved. All authors have read and agreed to the published version of the manuscript.

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