

## Article

# Sustainable Extraction of Colourant from Harmal Seeds (*Peganum harmala*) for Dyeing of Bio-Mordanted Wool Fabric

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**Abstract:** The recent pandemic scenario has caused demand for green products that have medicinal aspects, as well as greener approaches for global health. Natural dye from plants, particularly from harmal seeds, is an excellent alternative to carcinogenic yellow synthetic dyes. The current study has been conducted to isolate natural colorants from harmal seeds in methanolic medium through Gamma-Assisted Extraction (GAE). The dyeing variables that are necessary for shade development before and after mordanting were selected. It has been found that 6 kGy is the optimal absorbed dose for extraction of colorant from 6 g of powder to isolate the colorant in the methanolic medium through the Gamma-irradiated extraction mode (GAE). To get excellent results, 30 mL of methanolic extract containing 6 g/100 mL of Glauber salt was used for dyeing of irradiated wool at 45 °C for 65 min. For improving the color strength and acceptable rating of fastness, 9% of henna, 3% of acacia, 10% of turmeric, and 7% of pomegranate extracts as pre-bio-mordants as well as 7% of acacia, 3% of pomegranate, 9% of henna, and 10% of turmeric extracts as post-mordants have given high results compared to when chemical mordants have been used. It was concluded that Gamma-ray treatment has excellent color strength in the dyeing of bio-mordanted wool using harmal seed extracts under mild conditions, and has good fastness ratings after using chemical and bio-mordanting methods as well.

**Keywords:** bio-mordants; chemical mordants; harmal seeds; sustainability; wool fabric



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## 1. Introduction

The inclusion of advanced technologies in various fields, such as textiles, food, flavors, pharmaceutical, wood, plastic, glass, etc., has urged researchers and traders to develop methods that are not only sustainable in nature but also add value to the existence of green products [1,2]. These advanced technologies, such as ultraviolet [3], ultrasonic [4] microwave [5], Gamma, and plasma radiation [6], have provided a new horizon for traders, researchers, and consumers on account of their cost, time, and effective nature.

Of these tools, Gamma irradiation is the most powerful tool, which is gaining widespread utilization in textiles, isolation of natural products, medical textiles, anti-bacterial textiles, pharmaceuticals, and food processing [7,8]. It also has great use in the functionalization of fabrics and wastewater treatment, curing of fabrics bioremediation, and isolation of functional biological components (colorant) from plants without harming their physiological characteristics [9,10]. It also helps in tuning the surface of fabrics, which, in turn, helps to improve the uptake ability and diffusion power so that the effluent load is minimized [11–13]. Hence, due to the energy and labor-effective nature, the use of Gamma-ray treatment isolation of natural dyes and the improvement of their dyeing behavior onto natural fabric is on the rise [14]. Currently, the revival of natural dyes in different fields has

been warmly welcomed due to their sustainable and soothing nature [15]. These colorants are not only easily biodegradable but also have no effluent problem. On account of their shades and structural compatibility, these dyes are now being considered as an alternative to synthetic dyes in various applied fields, particularly in textiles, food, flavors, solar cells, pharmaceuticals, cosmetics, etc. [16,17]. Due to their beneficial role in the environment, the global community has been given awareness of their herbal and ayurvedic nature and their possible use in every field [18]. However, plant-derived natural colorants, particularly those with an ayurvedic nature, have been explored for coloring natural fabric since the last decade [19]. Thus overall, the revival of such colorants has not only helped us to save cultural heritage but also to make the globe fresher, cleaner, and healthier.

Of these plants, harmful seeds (*Peganum harmala* L.), as a source of natural colorants, are of great value. Its seeds contain alkaloids, such as harmine, harmaline, harmalol, and harmol [20–22]. Traditionally, its seeds have been used as a cure for evil eyes, skin inflammation activities, and parasitic worms [23]. The seed extracts also exhibit antimicrobial, antiprotozoal, anti-insect, anticancer, and anti-lice action [24–26]. Its extract is also considered an alternative to Turkey red dye (obtained from madders) and is used to dye carpet and wool yarns, make tattoo stains and inks, and help with the bioremediation of synthetic dyes [27].

To the author's knowledge, there are no such extensive studies that have been done to explore the dyeing behavior of harmful seeds, whereas our group is exploring the dyeing ability of harmful seeds for the first time, starting from extraction under the Gamma-ray-assisted extraction mechanism (GAE), dyeing, and mordanting conditions. The utilization of plant-based anchors (bio-mordants), such as henna, turmeric, acacia, and pomegranate [22], has made the coloring of wool using harmful seed extracts more sustainable and therapeutic [28,29]. Hence, given the excellent nature of harmful seeds and the promising role of Gamma radiation in textiles, this research work has been designed not only to extract the colorant from harmful seeds under the influence of Gamma irradiation but also to enhance the color strength and fastness properties using chemical and bio-mordants.

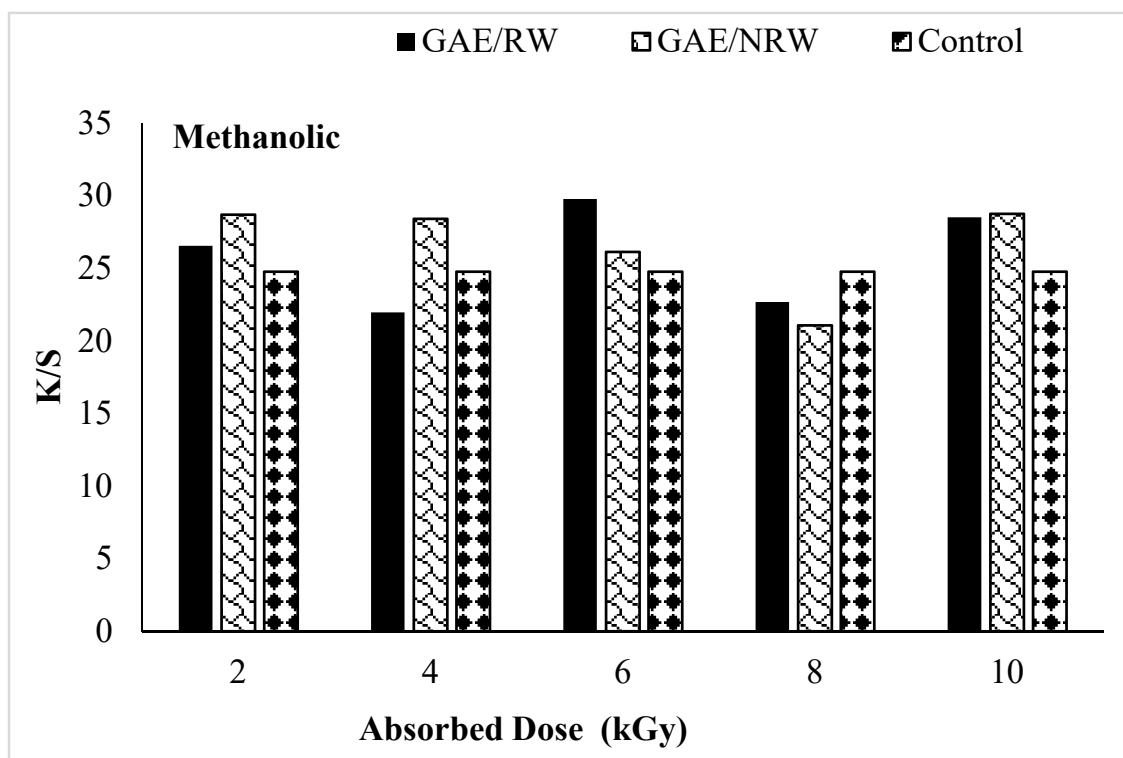
## 2. Materials and Methods

### 2.1. Material Collection

Harmful seeds (*Peganum harmala* L.) have been purchased from the local herbal market in Faisalabad. Seeds have been washed with water to remove the dirt and dried under shade. The dried seeds were ground finely and sieved to get powder of homogeneous particle size. Pakistan-made commercial chemicals such as methanol (CH<sub>3</sub>OH), hydrochloric acid (HCL), sodium hydroxide (NaOH), Glauber's salt (Na<sub>2</sub>SO<sub>4</sub>), and chemical mordants (salts of Al, Fe etc.) were purchased. For bio-mordanting, powders of turmeric rhizomes, henna leaves, acacia bark, and pomegranate rinds were purchased from the local superstore. The crude source of bio-mordants was also sieved to get powder of equal particle size. Pre-treated wool (GSM = 100 g/m<sup>2</sup>) was purchased from the Textile Market, Faisalabad, Pakistan.

### 2.2. Irradiation and Extraction Process

The absorbed dose of 2–10 kGy was given to wool fabric, and, for surface tuning, Gamma-ray-assisted extraction (GAE) was performed using a Cs-137 Gamma irradiator at N.I.A.B. (Nuclear Institute of Agriculture and Biology), Faisalabad, Pakistan. The unirradiated powder (NRP) was used for the isolation of colorant by employing 4 g with 100 mL of methanolic medium (60%) at reflux for 60 min—keeping a solid-to-liquid ratio of 1:25. After refluxing, methanolic crude mixture was filtered and the filtrate was used to dye untreated wool (NRW) at 75 °C for 65 min—keeping the fabric-to-dye-bath volume ratio of 1:25 (Khan et al., 2014; Adeel et al., 2017). This process was named control in Figure 1. In another experiment, Gamma-ray-assisted extraction (GAE) was done by proving a Gamma ray absorbed a dose of up to 10 kGy with an interval of 2 kGy, and, after extraction, the crude mixtures were filtered and used to dye irradiated (RW) and unirradiated wool fabric (NRW) at 75 °C for 65 min.



**Figure 1.** Gamma-ray-assisted methanolic extraction of colorant from harmful seeds and its application for wool dyeing.

### 2.3. Optimization of Mordanting Condition

After the optimization of the extraction condition at a particular absorbed dose (6 kGy), it was necessary to find a particular volume obtained from a particular amount of powder, and then vary the amount of powder for isolation. For this purpose, 2, 4, 6, 8, and 10 g of powder were used to isolate the colorant using 100 mL of methanolic medium upon refluxing for 60 min. In another set of experiments that utilized an optimal amount of powder (6 g), 10–70 mL of methanolic extract for 1 g of fabric was employed to keep the pH of the dye bath at 1–7. To select the optimum contact levels for the colorfast dyeing process, the contact levels (time) and heating levels (temperature) as dyeing variables from 25–85 min/°C were employed. In another set of experiments, to achieve maximum exhaustion, 1–10 g/100 mL of Glauber’s salt (GS) were employed in optimal conditions.

### 2.4. Shade Development Methods

For improvement in shade, 1–10% of the chemical mordants, such as the salt of Al, Fe, Cu, Sn and Co, and tannic acid (TA), have been utilized before (pre-) and after (post-) the dyeing process at 65 °C for 65 min, thereby keeping the mordant-to-fabric ratio of 1:25. For making the dyeing process more soothing and sustainable, 1–10% of the extracts of turmeric containing curcumin, henna containing Lawson, acacia bark containing quercetin, and pomegranate rind containing tannin have been employed before and after dyeing at 65 °C for 65 min, thereby keeping the bio-mordant –to- fabric ratio of 1:25 [22]. For the isolation of the colorants from plant-based mordants, curcumin from turmeric, Lawson from henna, quercetin from acacia, and tannin from pomegranate have been extracted using already documented methods from Adeel et al. (2021).

### 2.5. Evaluation of Characteristics of Dyed and Undyed Fabrics

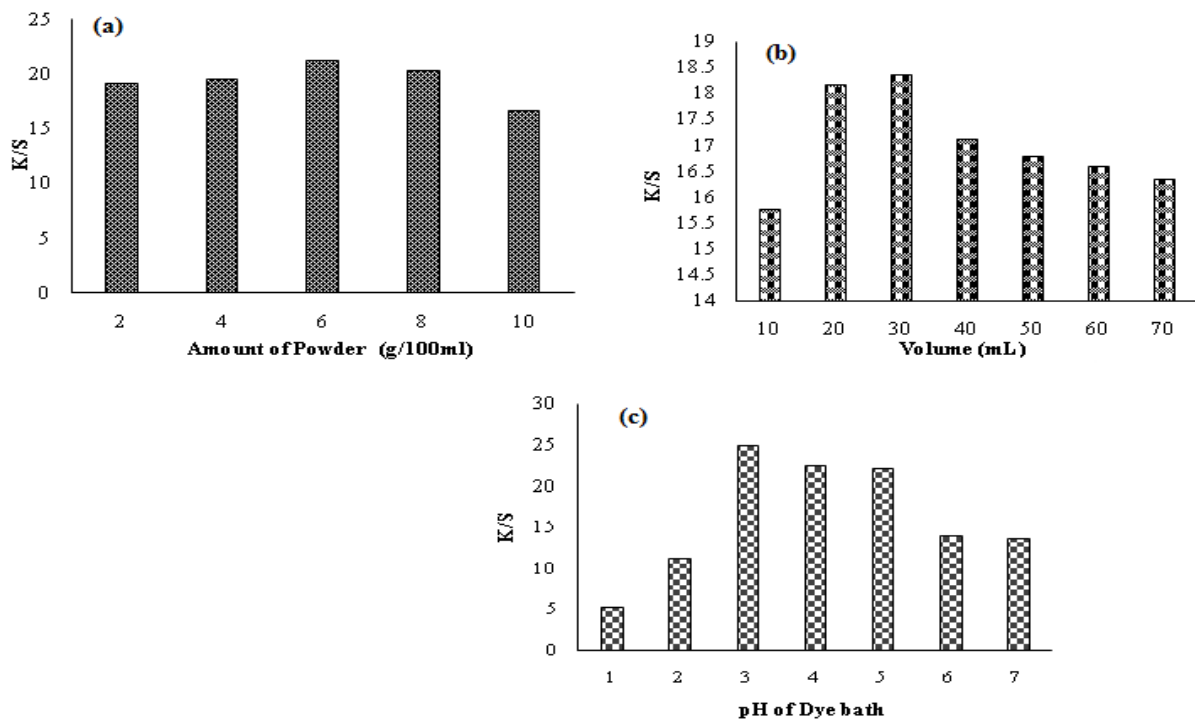
For the investigation of the changes in color strength (K/S), all the fabrics dyed before or after irradiation have been evaluated through the Kubelka–Munk equation, which was computed in a Data color SF 600 (USA) equipped with a D 6510° observer. To evaluate the

fastness, all the selected dyed, mordanted fabrics were exposed to ISO standards, such as ISO 105 CO3 for laundering (washing), ISO 105 BO5 for light, ISO 105 X12 for crocking, and ISO 105 D01 for dry cleaning (DCF) at the Q.A. & Q.C. Lab. of Noor Fatima Fabrics, Faisalabad, Pakistan.

### 3. Result and Discussion

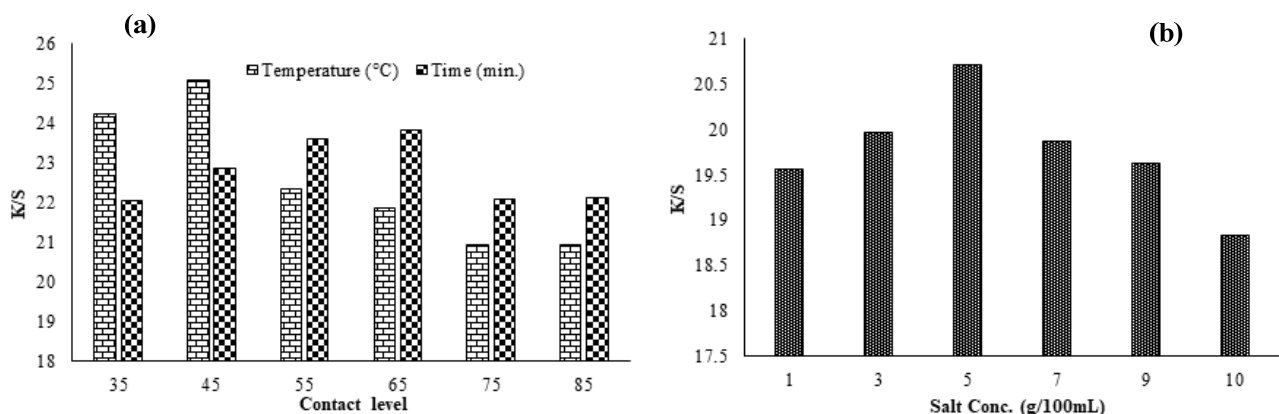
Gamma-ray treatment has given profound results in the extraction of colorant from harmful seeds in methanolic medium. The results shown in Figure 1 indicate that Gamma-ray treatment has good color depth (K/S) at low-absorbed doses (6 kGy). Depending upon the nature of the colorant (alkaloid), methanol medium has been found to be the best solvent to isolate the colorant, followed by its application onto wool fabrics at 6 kGy. Below the optimal absorbed dose, the extraction process is slow, whereas, above the optimal absorbed dose, the other phytochemicals are also isolated, which, upon coloration of the fabrics, influence the shade. Previously, it has been observed that methanol is the best solvent to extract the colorant because it has more dissipation power than aqueous, alkaline, or acidic medium [30]. The mass transfer process has occurred (solid–liquid interaction) through Gamma irradiation via rupture of the cell wall where the colorant evolved into the solvent [31]. Moreover, Gamma-ray treatment via producing free radicals modifies the isolation of functional bioactive components through the low consumption of the solvent at a reduced time. The other factor is the irradiation of wool fabrics, which shows that there is some etching occurring onto fibers—which might enhance its substantivity [32]. Hence, overall, the Gamma-ray-assisted extraction of colorant (GAE) in methanolic medium has given good color strength on irradiated wool (RW) when an absorbed dose of 6 kGy is employed.

After getting the extraction medium, the amount of powder needed is necessary to find out because of the above selected amount (>6 g), whereas the other phytochemicals are also more significantly involved in such a way that actual colorant has a low chance of absorbing onto the fabric. The results given in Figure 2a show that methanolic extract obtained from 6 g of harmful powder upon irradiation up to 6 kGy has an excellent yield on irradiated wool fabric (RW). After getting the optimal powder amount (6 g), it was also necessary to find out the volume of extract (30 mL) that was suitable for the coloration of wool fabrics. It has been observed that 30 mL of irradiated methanolic extract (RE) obtained from 6 g of powder (Figure 2b) has good color strength on irradiated wool (RW). A low volume does not impart good color depth, whereas, above the optimal volume, the clusters of molecules may get diffused into the modified wool fabric (RW) that remains on the surface. During this washing, unfixed dye is stripped off, which in turn results in low color strength. Hence, after reducing the amount of powder, the lowering of the extract volume reveals that Gamma-ray treatment has proven its cost-effective nature. As the wool contains a large number of polar groups, it has a greater ability to form a stable interaction with colorant in an acidic medium. If the pH of the extract is lowered, the anion groups of the fabric get protonated, wherein the fabrics have a significant chance of interacting with the –OH of the colorant via ionic bonding, which will give a good color yield [33]; this is because the wool keratin has amide linkages that are sensitive to the medium of the dye bath. Moreover, towards alkalinity, the –COOH of wool keratin may face a weak interaction with colorant –OH, thereby resulting in a low color strength. However, in an acidic medium, the amino group of keratins gets protonated and amide linkages are easily available for binding with the functional site [34,35]. Hence, the results displayed in Figure 2c reveal that the dyeing of irradiated wool (RW) should be done using an irradiated methanolic extract with a pH of 3 to get excellent results.



**Figure 2.** Impact of powder amount (a), volume (b), and pH of the extract (c) on the isolation of colorant from harmful seeds in a methanolic medium.

The other factors are contact variables, which play a great role in the dyeing of wool with plant-based colorants. Gamma-ray treatment has reduced not only the heating level (dyeing temperature) but also the contact time. This is because Gamma radiation is a powerful tool that has reduced the particle size of the colorant, which in turn has less time to absorb onto fabrics. Moreover, the kinetic energy of the colorant is increased when the dyeing variable is slowly increased up to 45 °C for 65 min. (Figure 3a). After these levels, the equilibrium of the dye bath is distributed, and the colorant starts stripping from the fabric to rush toward the dye bath. Hence, in order to get good coloring properties on irradiated wool (RW), irradiated methanolic extract (RE) should be used at 45 °C for 65 min.



**Figure 3.** Effect of heating levels (a) and salt concentration (b) on dyeing of irradiated wool using irradiated methanolic extract of harmful seeds.

The amount of salt to achieve maximum exhaustion needs to be optimized. This is because the natural dyeing process is dependent on the amount of salt for the interaction of the colorant with the wool via short-range attractive forces [36]. It was found that 5 g of Glauber's salt (GS) has an excellent exhaustion of the colorant to rush towards wool fabrics (Figure 3b). Above that amount (>5 g), over exhaustion has led to over aggregation, which,

in turn, upon washing, unfixed the colorant and removed it, and a low color strength was observed. The reduction in the amount of salt for exhaustion during the dyeing of wool with harmful seed extract shows that Gamma-ray treatment is a cost-effective tool. Overall, it can be seen that Gamma-ray extraction has taken less extract volume, energy, time, and salt to produce better results.

Bio-mordanting is a newly-introduced process, so called bio-dyeing, because through a special interaction, new colorfastness tints are developed; furthermore, due to their therapeutic nature, these molecules impart biological characteristics towards fabrics before and after dyeing. These bio-molecules have not only given new shades but also enhanced the color characteristics [36–38]. These functional potent isolates (as shown in Figure 4), via forming intermolecular H-bonding with colorant and functional sites of fabric, have given a high color yield with newly-developed shades [39,40]. It has been found from Table 1 that during pre-bio-mordants, 9% of Lawson from henna (structure given below), 10% of curcumin from turmeric (structure given below), 3% of quercetin from acacia (structure given below), and 7% of tannin from pomegranate (structure given below) has given a high color strength (K/S) on irradiated wool (RW) when employed before dyeing using methanolic extracts of harmful seed. A low amount of bio-mordants, when used, have given a lower yield (K/S) whereas, above the optimum amount, the aggregates formed may have difficulty diffusing into the fabric and remain unfixed on the fabric. Upon washing, the overcrowded aggregates are stripped, and a lower yield (K/S) is observed. Similarly, during post-bio-mordanting, 9% of Lawson from henna, 10% of curcumin from turmeric, 7% of quercetin from acacia, and 3% of tannin from pomegranate has given a high color yield through their application onto dyed fabric (Table 1). The tonal variation given in Table 1 shows that most fabrics that are dyed before and after bio-mordanting are darker in tone and reddish-yellow in hue, because there is a joint effect between two biomolecules, i.e., alkaloids from harmful in combination with curcumin from turmeric, quercetin from acacia, tannin from pomegranate, and Lawson from henna, which can be seen forming through additional H-bonding.

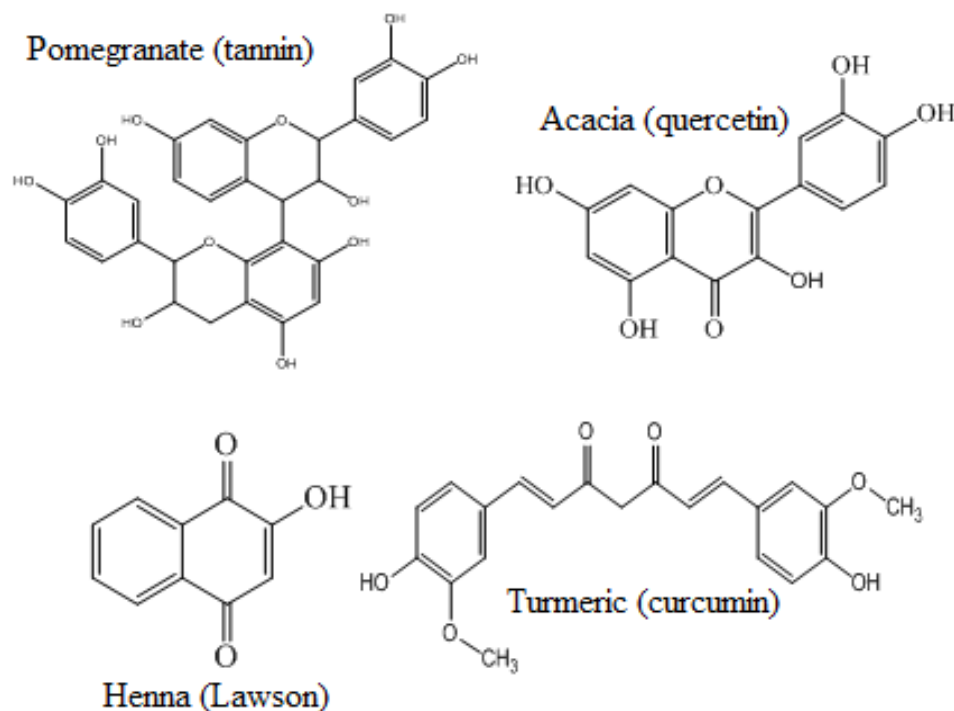


Figure 4. Functional Potent isolates of bio-mordants.

**Table 1.** Color characteristics of chemical and bio-mordanted wool fabrics dyed before, after, and during dyeing with Gamma-treated harmful seed extracts.

Mordant Concentration	K/S	L*	a*	b*	Mordant Concentration	K/S	L*	a*	b*
Al 10% (Pre)	16.341	70.80	2.83	30.04	T. A 10% (Pre)	21.281	58.45	7.66	30.69
Al 7% (Post)	7.1222	71.08	3.58	24.77	T. A 7% (Post)	8.666	54.57	8.77	22.60
Fe 10% (Pre)	19.299	59.43	6.48	30.87	Acacia 3% (Pre)	12.612	60.33	9.12	30.21
Fe 7% (Post)	7.6815	60.00	10.40	28.73	Acacia 7% (Post)	6.6678	58.62	12.19	27.35
Co 7% (Pre)	19.77	61.55	10.51	34.49	Pomegranate 7%(Pre)	19.527	61.52	7.12	30.76
Co 7% (Post)	7.2014	60.69	7.52	20.69	Pomegranate 3%(Post)	6.4293	68.68	4.78	32.29
Sn 10% (Pre)	17.439	69.86	6.50	33.30	Henna 9% (Pre)	21.578	49.70	9.46	28.65
Sn 7% (Post)	8.0339	74.57	4.29	30.94	Henna 9% (Post)	12.547	51.74	12.37	30.13
Cu 10% (Pre)	16.924	54.69	2.23	31.69	Turmeric 10% (Pre)	23.645	51.35	13.23	43.86
Cu 7% (Post)	7.6831	63.06	7.05	23.25	Turmeric 10% (Post)	23.576	59.21	16.14	64.98

The role of chemical mordants is reversed as compared to bio-mordants because during chemical treatment, the involvement of d-orbitals of the metal dye complex reduction power of metals works via coordinating the covalent bonding through metal dye complex formation [41,42]. It has been observed that 10% of tannic acid (TA) has given a high color yield, as compared to the other pre-mordants used. Similarly, 10% of Al, Fe, Cu, and Sn, and 7% of Co have a good color yield (K/S) upon dyeing of irradiated wool (RW) with irradiated methanolic extract (Table 1). This is because the metal ions have been absorbed evenly onto the wool fabric, which, upon dyeing with irradiated extract, has formed a stable metal dye complex through a coordinated covalent bond [43,44]. Similarly, during post-mordanting, 7% of TA has given a good color yield (K/S) as compared to the other metal mordants used (Table 1). Hence, 7% of Al, Cu, Co, Fe, and Sn, as post-mordants, have excellent yields. This good color strength is attributed to the even dyeing of wool after Gamma-ray treatment. The tonal variation given in Table 1 shows that most fabrics dyed before and after chemical mordanting are brighter in shade and are reddish-yellow in hue. Thus, the reduction of the chemical mordant amount used after Gamma-ray treatment reveals that this powerful tool is sustainable and cost-effective.

The rating results of colorfastness properties given in Table 2 for pre- and post-chemical mordanting, as well as bio-mordants, show that most fabrics dyed after mordanting have excellent ratings for light, washing, rubbing, and dry cleaning. This is because the conjugation system present in bio-mordants and colorants plays a greater role when the fabric is in contact via a covalent bond. The extra bonding given by bio-mordants may create more stable interactions with fabrics and colorants [45,46], which, upon exposure to agents such as heat, detergents, light, crocking, and dry-cleaning agents, resist detaching. Similarly, the rating results given in Table 2 for chemical pre- and post-mordanting reveal that the stable metal dye complex formation on fabric resists fading of the colorant [47–49]. The greater the potential efficiency of metal to form a covalent bond with an amide linkage of wool and –OH of colorant isolated from harmful seeds, the more stable the complex will be that is formed [50,51]. Hence, the overall rating results show that bio-mordants have a promising resistance to heat, light, detergent, crocking, and dye cleaning agents. The shades that were made are represented in Table 3. It can be concluded that the utilization of bio-mordants has not only made the dyeing of irradiated wool greener and more sustainable but also given improved fastness characteristics.

**Table 2.** Colourfastness rating of mordanted and dyed Gamma-treated wool fabric using harmful seed extracts.

Mordant Concentration	LF	WF		RF		DCF	PF	
		c. c	c.s	DRF	WRF		Acidic	Alkaline
Control	3/4	3	3	$\frac{3}{4}$	$\frac{3}{4}$	3/4	3	3
Al 10% (Pre)	4	4/5	4/5	4/5	4	4	4/5	4/5
Al 7% (Post)	4/5	5	5	5	5	4/5	5	5
Fe 10% (Pre)	4/5	4/5	4/5	4/5	4	4	5	5
Fe 7% (Post)	4/5	5	4/5	5	4/5	4	4/5	4/5
Co 7% (Pre)	4/5	5	4/5	4	$\frac{3}{4}$	4	4/5	4/5
Co 7% (Post)	4/5	5	5	5	4	4/5	5	5
Sn 10% (Pre)	4	4/5	4/5	4/5	4	4	4/5	4/5
Sn 7% (Post)	4/5	5	5	5	5	4	4/5	4/5
Cu 10% (Pre)	4/5	5	4/5	4/5	4	4/5	4/5	4/5
Cu 7% (Post)	4/5	5	5	4/5	4	5	5	5
T. A 10% (Pre)	4/5	4/5	4/5	4/5	4	4	4/5	4/5
T. A 7% (Post)	4/5	5	5	4/5	4	5	4/5	4/5
Acacia 3% (Pre)	4	4/5	4/5	4/5	4	4	4/5	4/5
Acacia 7% (Post)	4/5	4/5	4/5	4	4	4/5	4/5	4/5
Pomegranate 7% (Pre)	4/5	4/5	4/5	4/5	4	4/5	4/5	4/5
Pomegranate 3% (Post)	4/5	4/5	4/5	4/5	4	4/5	4/5	4/5
Henna 9% (Pre)	4/5	4/5	5	4/5	4/5	4	4/5	4/5
Henna 9% (Post)	4/5	4/5	5	4/5	4/5	4	4/5	4/5
Turmeric 10% (Pre)	4	4/5	4/5	4	$\frac{3}{4}$	4	4/5	4/5
Turmeric 10% (Post)	4	4/5	4/5	4	$\frac{3}{4}$	4	4/5	4/5

LF = Light fastness, WF = wash fastness, c.s = color stain, c.c = color change, RF = Rub fastness, DRF = dry rub fastness, WRF = wet rub fastness, DCF = dry clean fastness, PF = perspiration fastness.

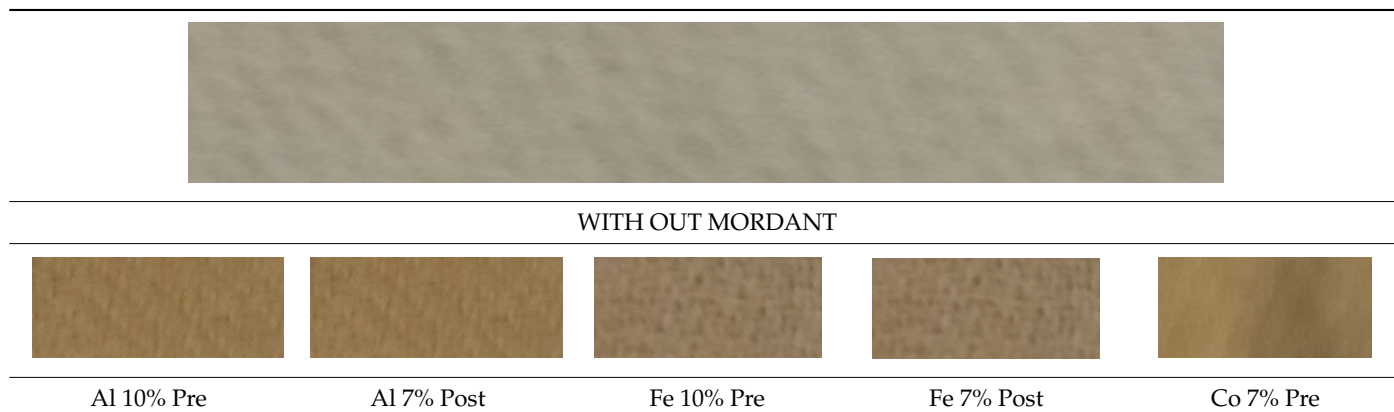

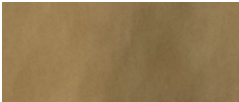
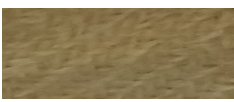



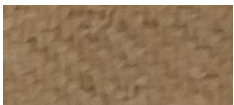
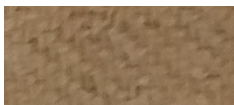
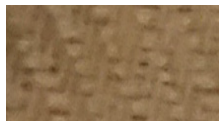


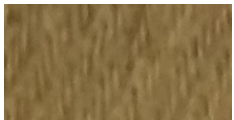




**Table 3.** Tonal variation of Gamma-treated dyed and mordanted wool fabrics using harmful seed extracts.



Table 3. Cont.

				
				
Co 7% Post	Sn 10% Pre	Sn 7% Post	Cu 10% Pre	Cu 7% Post
				
T.A 10% Pre	T.A 7% Post	Acacia 3% Pre	Acacia 7% Post	Pomegranate 7%Pre
				
Pomegranate 3% Post	Henna 9% Pre	Henna 9% Post	Turmeric 10% Pre	Turmeric 10% Post

#### 4. Conclusions

Sustainability is one of the greater demands of the global community because of global warming and global heat, and unexpected seasonal changes have been found due to carcinogenic, effluent loads from industries. Among sustainable products, natural plant pigments and colorants have taken a special place and particular attention of the people due to their excellent biological and herbal characteristics. This study is one a series of explorations of plants that have ayurvedic and biological natures and have potential to impart color onto fabric. Additionally, among modern techniques for their extraction, Gamma-ray treatment in textiles has shown promising effects not only in the exploration of new dye-yielding plants, such as harmful seeds, but also in enhancing extraction yield and color characteristics. An absorbed dose of 6 kGy was optimized to get extraction of the colorant in methanolic medium. The dyeing wool fabric for 65 min at 45 °C using 30 mL of dye bath of pH 3 using 5 g/100 mL of the Glaubar's salt as an exhausting agent was done to get colorfast shades before and after the selected amount of chemical and bio-mordants. It is concluded that this novel modern tool can be applied to explore new dye-yielding plants as a source of natural dyes for the coloration of natural fabrics, and the addition of new bio-mordants can be done to assess new shades with excellent colorant characteristics.

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