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Investigation into the Dyeing of Soybean Fibers with Natural Dyes and their Antimicrobial Properties

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ABSTRACT

The dyeing behavior of annually renewable soybean protein fibers with Madder, Weld and Walnut seed husk, natural dyes, were investigated and their possible antimicrobial effects were evaluated. The results obtained from the dyeing of the Madder on the soybean fabric were compared with those on a wool fabric. It was shown that the amount of the amino acid content of the fiber has a great effect on the final color depth. The unusual change in the color of the Madder on the soybean fabric upon washing in an alkaline washing solution is also addressed. Also, a good antimicrobial activity against the gram-positive bacterium for the Walnut husk and Madder dyes were revealed while the effect of Weld dye was practically negligible. However, these natural dyes on the soybean fabric did not show any significant antibacterial properties against gram-negative bacterium. Prog. Color Colorants Coat. 7(2014), 95-104. © Institute for Color Science and Technology.

1. Introduction

Recently, the manufacture of fiber from soybean protein has received significant attention. Soybean protein fiber, SPF, is a natural-based synthetic fiber produced from annually renewable resources, combines ecological advantages with excellent performance in textiles. This regenerated protein fiber have no theoretical limit in fineness with competitive price as compared with natural protein fibers such as wool, silk and petroleum based fibers. In addition, the protein content of soybean is much higher than other comparable sources like peanuts and corn [1-4]. Also it is now possible to enhance the technical performance of SPF by molecular genetic-engineering techniques, thus broadening the fibers'commercial scope to include new and potentially specific applications [5-6].

SPF has good physical and aesthetic properties including higher breaking strength than wool, silk and cotton; outstanding crease resistance; easy wash and fast dry properties; having luster comparable to that of silk; excellent drape; fabric of yarn with high count with fine and clear appearance; outstanding comfort with optic effect; soft, smooth and light handle similar to that of blended fabrics from silk and cashmere; the same moisture absorption and higher moisture transmission as

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that of cotton which make SPF good relaxed and sanity[7-8]. However, a number of technical deficiencies are still noticeable while it is crucial to establish the processing and associated performance of the SPF materials.

Dyeing properties of SPF with acid and reactive dyes have been reported [9], however, there is little or no published information regarding dyeing this fiber with natural dyes. Many natural colorants have good light and abrasion fastness, and if they were dyed properly, they could be considered environmental friendly dyes, which have also anti-microbial properties [10-11]. These dyes also reported to have good sun protection properties [12]. This paper addresses a solution for SPF dyeing deficiencies, which are poor coloration performance and color fastness to washing, using some specific natural dyes possessing antibacterial property.

2. Experimental

2.1. Materials

The chemicals used in this study were non-ionic detergent from Dyestar, anhydride sodium carbonate, aluminum sulphate, and formic acid, all in analytic grade, supplied by Merck. The Madder and the Walnut seed husk dyes were prepared from Yazd province in Iran, and the Weld natural dye was obtained from Khorasan province in Iran. The Interlock knitted fabric, obtained from Swicofil [13], had been woven from 30 Tex soybean yarn.

The samples were dyed in an Ahiba Polymat dyeing machine from Datacolor. The fabric preparation was included scouring and bleaching. The fabric was firstly scoured to remove any possible impurities which can adversely affect the dyeing behaviuor by 2 ml/l non-ionic detergent and 1 g/l sodium carbonate with L:R of 30:1 at 50-55˚C for 15 min. Then samples rinsed and air dried without any tension. After fabric scouring, sample bleaching was done by an ozone treatment. Then, the bleached samples were mordanted using the method showed in Figure 1 in a bath containing 5% aluminum sulphate in pH=5, adjusted by formic acid. Each mordanted sample was dyed in a bath containing different concentrations of the dyes with the dyeing curve illustrated in Figure 2.

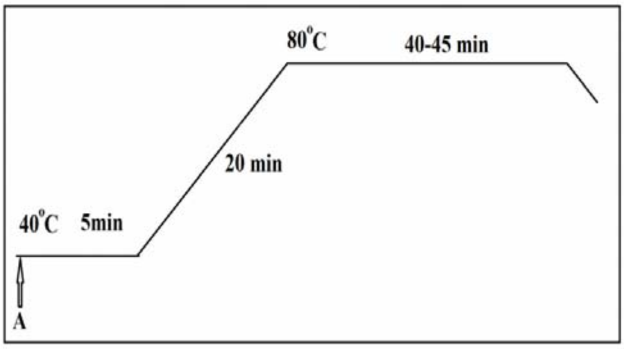

Figure 1: The method of mordanting the samples[14], A: $AlSO_4$ and formic acid added.

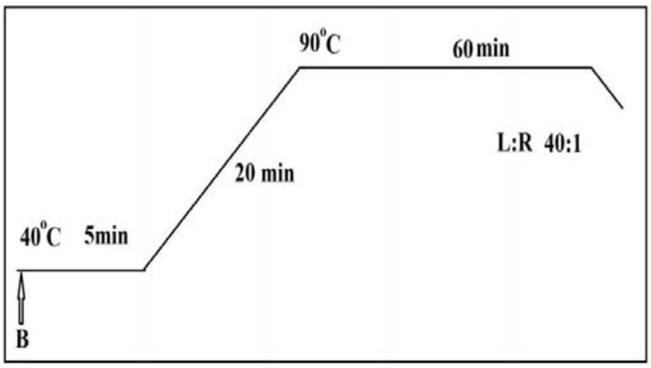

Figure 2: The dyeing curve of the soybean fabric with the natural dyes [14], B: dye added.

The wash fastness of the dyed samples was tested using the ISO105C02 standard method and the reflectance of the samples was measured by a 0/d spectrophotometer. The L*a*b* of the samples were calculated using the 10 degree standard observer and D65 standard illuminant condition. The color difference used in the research was calculated from the CIELAB color difference formula.

The method used for measuring the antibacterial properties of the dyed fabrics was based on the AATCC Test Method 100-1999, Antibacterial Finishes on Textile Materials. To begin the antimicrobial test, a bacteria solution containing constant concentration of two bacteria, the gram-positive bacterium, Staphylococcus aureus (A. aureus), AATCC 1337, and gram-negative bacterium of Pseudomonas aeruginosa (P. aeruginosa), AATCC 1074, was prepared by measuring the light absorption of the solution using a transmission spectrophotometer in 580 nm which should be in the range of 0.2-0.3. 1 ml of bacteria solution was applied evenly on sterilized cotton fabric samples cut in circular shape with 48 mm diameter (almost 1 g). The fabric samples containing the bacteria were then incubated at 35°C for 24 hours. 100 ml of sterilized water was then added to the fabric samples and 1 ml of the solution was transferred to a nutrient agar growth medium. Growth media were then incubated at 35°C for 24 hours and finally the number of bacterial colony grew on nutrient agar medium was counted using a microscope. The bacteriostatic ratio (%) was calculated using the equation (1):

$$R\ (\%)=(A-B)/A\times 100 \qquad (1)$$

where R is the reduction rate, A is the number of bacterial colonies from untreated fabrics and B is the numbers of bacterial colonies from dyed fabrics.

3. Results and discussion

Tables 1, 2 and 3 show the L*a*b* and the color difference between the appropriate pair samples of the soybean fabric dyed with the madder, the weld and the walnut seed husk, respectively. Figures 3, 4 and 5 also illustrate the corresponding reflectance of the soybean fabric dyed with the madder, the weld and the walnut seed husk, respectively, in the applied concentrations.

Considering the results in Figure 3(a) rise in the reflectance values in the range of 600-700 nm can be observed indicating a red dominant hue for the dyed fabrics. Regarding the color difference between the R23 and R24 coded dyed fabrics and by considering the economical and environmental aspects of dyeing processes, 40% was preferred as the maximum concentration of the madder to dye the soybean fabric.

Table 1: The L*a*b* and the corresponding ΔE^*_{ab} of the soybean fabric dyed with the madder.

Code	Substrate	Concentration (%) o.w.f	L*	a*	b*	ΔE^*_{ab}
R20	Soybean	10	73.0	20.6	14.0	4.83
R21	Soybean	20	68.5	21.4	15.4	
						3.25
R22	Soybean	30	65.9	19.6	16.3	
						1.92
R23	Soybean	40	65.2	21.2	17.0	
						1.97
R24	Soybean	60	63.7	22.5	17.2	
	Soybean	Reference	94.2	1.1	7.5	--
	Wool	20	41.76	37.07	31.72	--
	Wool	30	38.27	37.47	30.84	--

Table 2: The L*a*b* and the corresponding ΔE^*_{ab} of the soybean fabric dyed with the weld.

Code	Substrate	Concentration (%) o.w.f	L*	a*	b*	ΔE^*_{ab}
S20	Soybean	5	85.799	-0.009	39.151	4.13
S21	Soybean	10	82.418	-0.29	41.509	6.03
S22	Soybean	20	82.049	2.259	46.966	0.86
S23	Soybean	30	81.244	2.052	46.74	
	Soybean	Reference	94.2	1.1	7.5	--

Table 3: The L*a*b* and the corresponding ΔE^*_{ab} of the soybean fabric dyed with the walnut seed husk.

Code	Substrate	Concentration (%)o.w.f	L*	a*	b*	ΔE^*_{ab}
G20	Soybean	5	78.778	4.281	7.856	3.88
G21	Sybean	10	74.996	4.675	8.607	3.36
G22	Soybean	20	71.785	4.681	9.599	1.69
G23	Soybean	30	70.244	4.958	10.233	1.04
G24	Soybean	40	69.724	5.378	11.029	
	Soybean	Reference	94.2	1.1	7.5	--

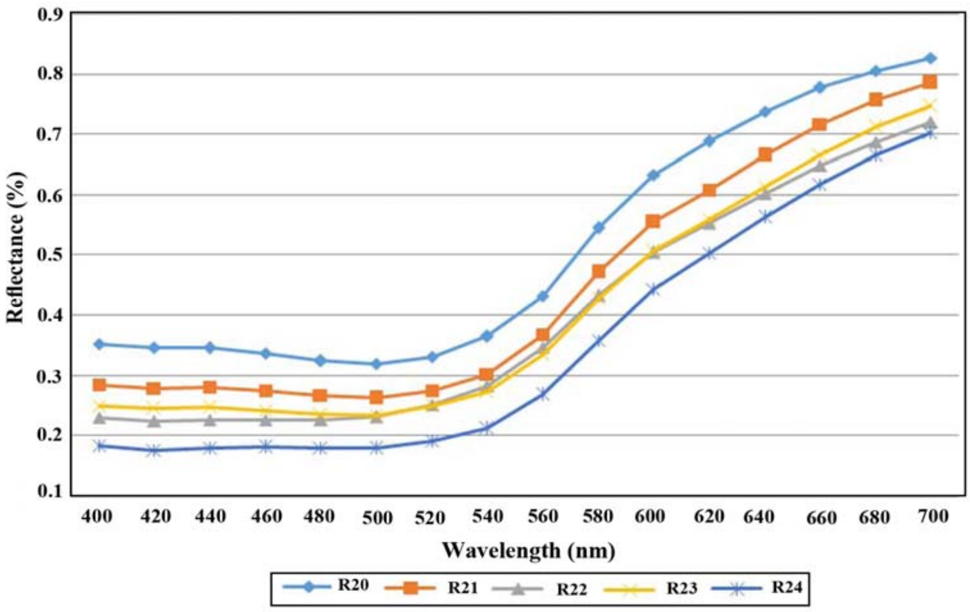

Figure 3: The reflectance of the soybean fabrics dyed with the madder in different concentrations.

Also, the results in Table 1 demonstrate that the maximum concentration of the Madder on the Soybean fabric (40%) gives less color depth than for the 20% of the dye on the wool fabric, which is due to less amino acid content (23-55%) in soybean fibers in comparison to wool fibers [9].

Concerning the results in Tables 2 and 3 and figures 4 and 5, for the same reasons explained for the madder, the maximum concentration of the weld and the walnut seed husk dyes on the soybean fabrics was considered 20% and 30%, respectively.

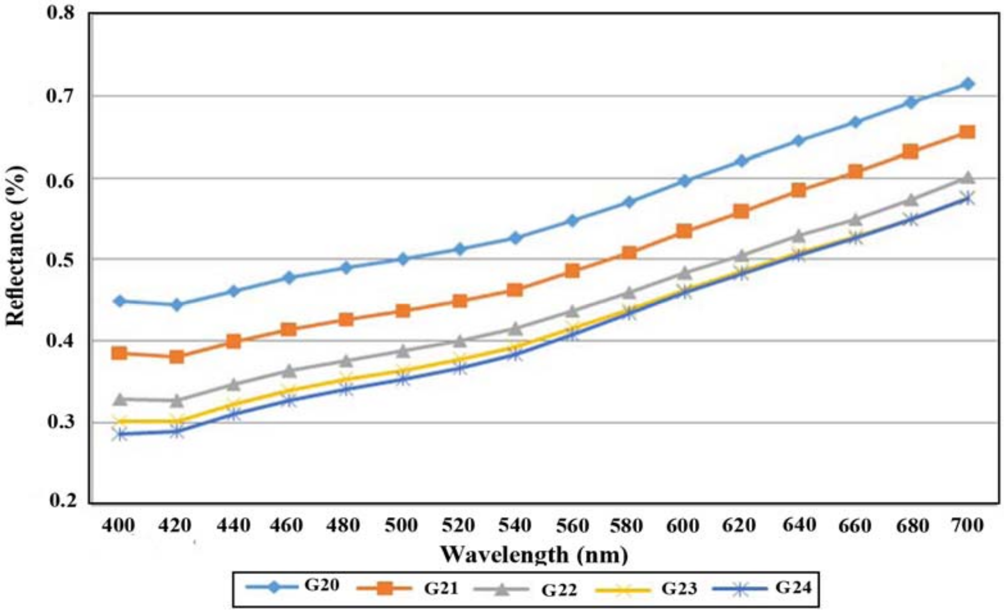

Figure 4: The reflectance of the soybean fabrics dyed with the weld in different concentrations.

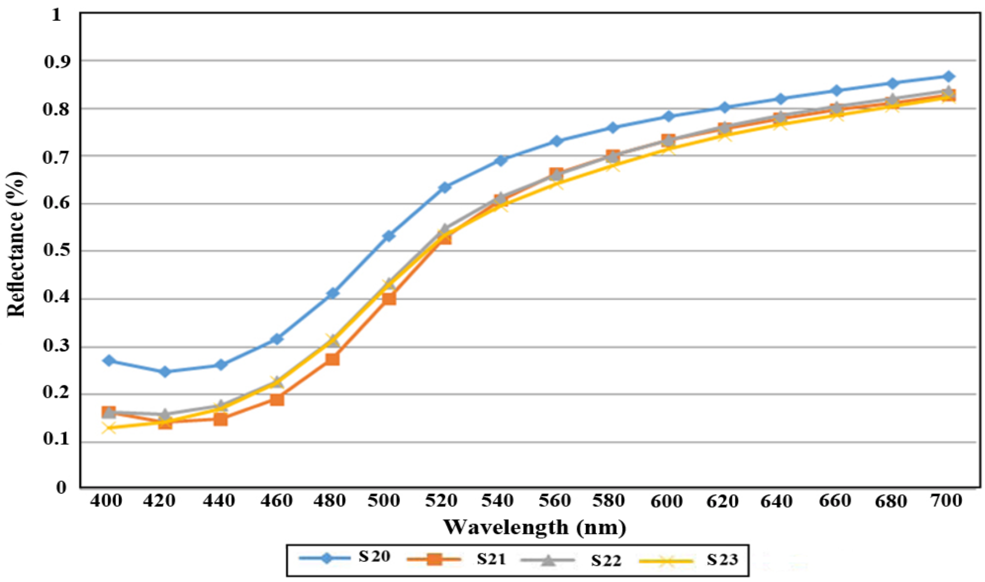

Figure 5: The reflectance of the soybean fabrics dyed with the walnut seed husk in different concentrations.

Tables 4, 5 and 6 illustrate the color fastness results for the samples dyed with the walnut seed husk, the weld an the madder, respectively.The Walnut seed husk dyed on the soybean fabric showed a good to very good wash fastness, 4-5 to 5 in the Gray Scale rating, and the fabric dyed with the Weld had a moderately good wash fastness, 3 to 3-4 in the Gray Scale rating. It is noteworthy that the soybean samples dyed with the Madder showed a color change from a dark red to a dull pink after wash fastness treatment using the ISO105C02 standard test method. This change made the wash fastness assessment of these samples difficult. However, according to the results obtained for the color fastness of these samples shown in Table 6, these samples showed good color fastness, 4 to 4-5 in the Gray Scale rating, and it should be considered that the treated samples in this table are offtone.

Considering the results reported by Montazer *et al* [14] about the same phenomenon happened in the dyeing of wool with the Madder, it seems that washing in an alkaline solution could be the reason for the color change after color fastness test done on the samples dyed with the madder.

Table 4: The color fastness of the soybean fabric dyed with the walnut seed husk.

No.	Code	Concentration (%) o.w.f	Color fastness
1	G20	5%	5
2	G21	10%	5
3	G22	20%	4-5
4	G23	30%	4-5
5	G24	40%	4-5

Table 5: The color fastness of the soybean fabric dyed with the weld.

No.	Code	Concentration (%)o.w.f	Color fastness
1	S20	5%	3-4
2	S21	10%	3-4
3	S22	20%	3
4	S23	30%	3

Table 6: The color fastness of the soybean fabric dyed with the madder.

No.	Code	Concentration (%)o.w.f	Color fastness
1	R20	10%	4-5
2	R21	20%	4-5
3	R22	30%	4
4	R23	40%	4
5	R24	60%	4

According to Montazer *et. al.,* the reason for this change in the hue of the fabric is that the alkali used in the washing solution can attack the hydroxyl groups in the position 1 and 2 of Alizarin molecule, as shown in Figure 6, and make the oxygen to have a negative charge. However, if it is assumed that all the Alizarin molecules are bounded by the mordant, their suggestion may not be correct. One possible explanation for this color change is due to the formation of quinonoid system after the treatment of the Alizarin with alkali.

Montazer *et. al.* also claimed [14] that the reaction responsible for the alteration in the color of the Madder on the wool fabric is not reversible after acid treatment. It is believed that this claim may not be optimal as the initial experiments done in the present research showed that the original color of the soybean fabric dyed with the Madder before washing can be recovered using an acid treatment. This finding is a matter of further study.

3.1. Antimicrobial activity of the natural dyes

Table 7 illustrates the results of the antibacterial properties of the natural dyes on the soybean fabrics against the gram-positive bacterium, *Staphylococcus aureus*, and the gram-negative bacterium, *Pseudomonas aeroginosa*.

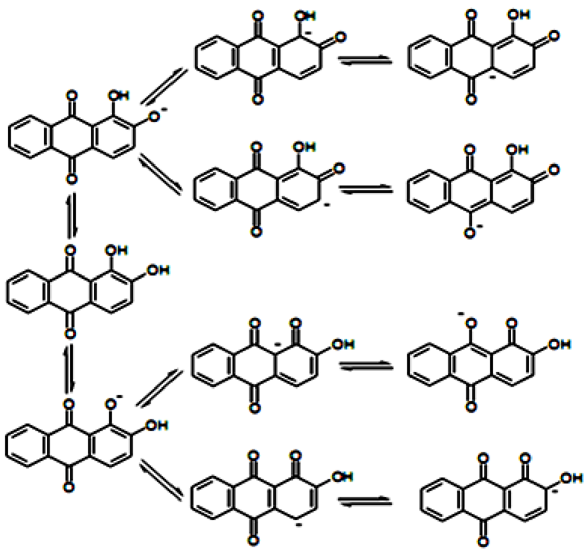

Figure 6: Different resonant structures of Alizarin [14].

Table7: Antimicrobial activity of the dyed soybean fabric with the natural dyes against Pseudomonas aeroginosa bacterium.

Code	Concentration of dye % (o.w.f)	Dye	Reduction in CFU% *Staphylococcus aureus*	Reduction in CFU% *Pseudomonas aeruginosa*
R20	10	Madder	90	0
R21	20	Madder	99	0
R22	30	Madder	100	0
S20	5	Weld	18	0
S21	10	Weld	21	0
S22	20	Weld	24	0
G20	5	Walnut seed husk	94	0
G21	10	Walnut seed husk	95	0
G22	20	Walnut seed husk	100	0
Reference	--	--	0	0

The results of antimicrobial tests indicate the direct relationship between antimicrobial properties and natural dyes concentration. The results verify the optimum dye concentration of 20-30% for the Madder and10-20% (owf) of the Walnut seed husk for gram-positive bacterium (S. aureus) while the Weld antibacterial properties is not at acceptable level with maximum 20% .

Tannins (as shown in Figure 7) and flavinoids are the most important antimicrobial phenolic components in natural dyes. It is claimed that the antibacterial mechanism of these molecules is based on making complex with the cell walls, breaking the membrane and preventing the enzyme activity.

The antimicrobial property of the Madder has been the subject of many researches and it is mentioned that due to having anthraquinone derivatives in its molecules, Alizarin is responsible for this good antibacterial properties [15]. It is mentioned [17] that anthraquinone derivates have antimicrobial property. Therefore, it seems that because of the similar anthraquinoied structure to Alizarin, Purporin is likely to have the comparable antimicrobial activity as Alizarin. The antimicrobial activity of the molecules with similar structure to Alizarin has also already been reported in a research by Wasif *et. al.*[18] on the Aleovera extract. They showed that Aleovera has good antimicrobial activity due to having Polysaccharide acemannan and Barbaloin molecules. Figure 8 shows that Barbaloin is an anthraquinone derivate and has a similar structure to Alizarin.

Another molecule based on anthraquinone and with phenol group is carminic acid showed in Figure 9. This molecule can be found in cochineal dye and it is believed that the good antibacterial property of the dye is because of carminic acid as one of its components [20].

The assumption that the Weld with a chemical structure similar to flavinoied may show moderately good antimicrobial property was declined in this research according to the results of Table 7. One reason for this observation is likely to be due to the formation of a complex between the natural dye and the mordant. This reduction in the antimicrobial activity of natural dyes due to the aforementioned reason is also observed for other dyes on different substrates [21].

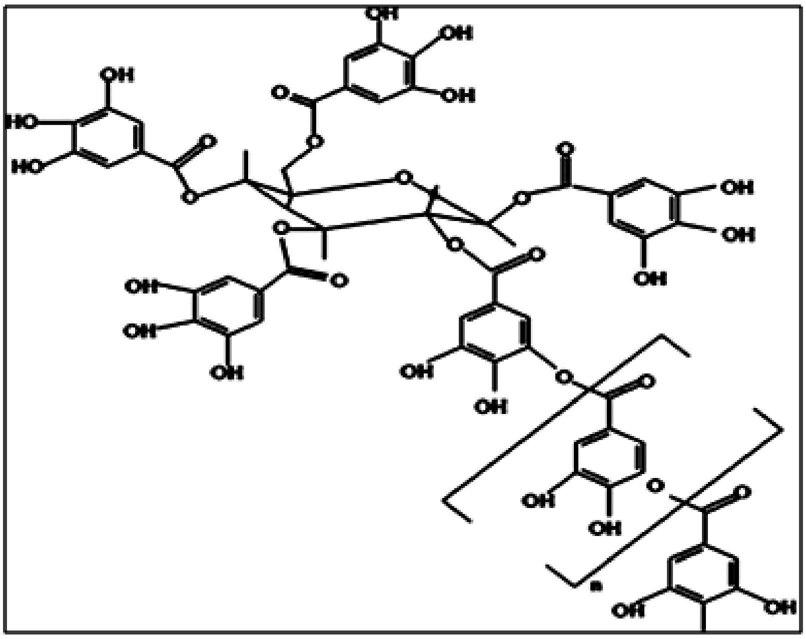

Figure 7: Chemical structure of tannin [10]

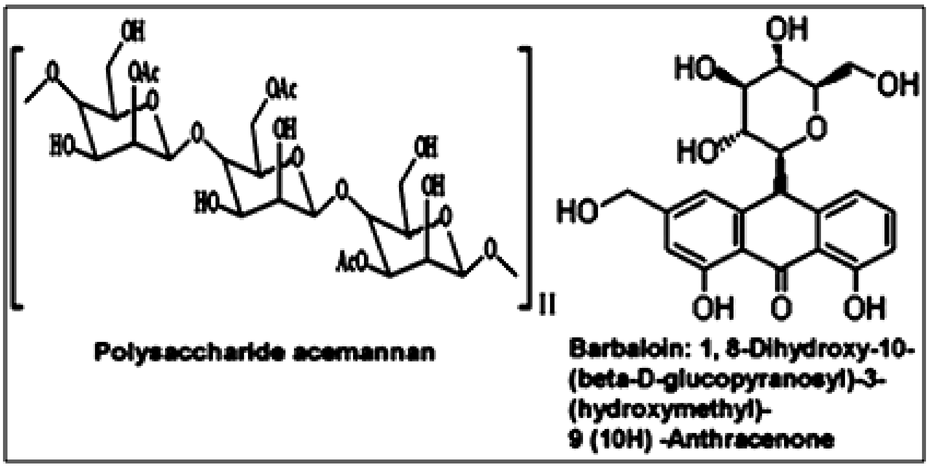

Figure 8: Components in Aloevera responsible for its antimicrobial property [19].

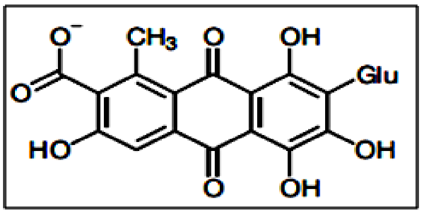

Figure 9: Chemical structure of carminic acid [14].

It is evident from the results in Table 7 that none of the natural dyes has an antimicrobial activity against *Pseudomonas aeruginosa* bacterium, which is responsible for the burn scars infection. This phenomenon has been reported for other natural dyes against gram-negative bacteria. The differences between gram-positive and gram-negative bacteria essentially rest in the structure of their respective cell walls. Gram-negative bacterium has an outer layer, lipopolysaccharide, preventing antibacterial agent penetration through the cell wall; hence, higher concentrations of these dyes are needed to destroy gram-negative bacterium. This is because of the fact that the outer layer of gram-negative bacterium, lipopolysaccharide, prevents the aggregation of antibacterial agent on the membrane cell of the bacterium [21]. Accordingly, more research is needed to find out the effect of natural dyes on different bacteria to clarify their interaction.

4. Conclusion

The dyeing behavior of the soybean fabric using Madder, Weld and Walnut seed husk natural dyes were investigated. As soybean fibers are considered environmental friendly fibers, it is more desirable to dye them with natural dyes which are of environmental friendly reagent. The results indicate that a good dye yield with acceptable fastness properties could be obtained, while the added antibacterial properties to the dyeing process pave the way for more research and introduction of facile method for multifunctional treatment that helps replacement of harmful synthetic fibers with soybean natural fibers. Considering the properties of soybean fibers stated in the introduction section together with the antimicrobial property of many natural dyes and the fact that these dyes have less harmful effect on the skin than synthetic dyes, these fibers can be used in some important applications, such as children clothing. The antibacterial properties of the dyed soybean fabric with these three natural dyes did not show a promising results and their antimicrobial activity needs to be intensified. To further explore the antibacterial properties of the natural dyes on soybean fibers, more investigations are needed.

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