(REFEREED RESEARCH)

NATURAL DYEING PROPERTIES OF WOOL FABRICS BY POMEGRANATE (*Punica granatum*) PEEL

NAR (*Punica granatum*) KABUĞUNUN YÜN KUMAŞI DOĞAL BOYAMA ÖZELLİĞİ

Mustafa TUTAK[1], Gülcan ACAR[1], Onur AKMAN[2]

[1] *Erciyes University, Department of Textile Engineering, Kayseri, Turkey*
[2] *Yünsa Textile Company, Research & Development Center, Tekirdağ, Turkey*

Received: 18.09.2013 Accepted: 24. 02.2014

ABSTRACT

Pomegranate peel natural dye solution was prepared and used to dye wool fabrics. The effects of mordant type, concentration, and chemical post-treatment on color yield and fastness properties were investigated. The naturally dyed fabrics had between 6.63-23.05 (K/S) color strength, and the color fastness results of the dyed fabrics were moderate/high level. The selected dyed fiber sample surfaces were observed using a scanning electron microscope. The SEM images of the dyed fabrics demonstrated a smooth surface profile. The experimental study showed that waste pomegranate peel extract was suitable for the natural dyeing of wool fabrics.

Key Words: Natural dye, Wool, Dyeing, Fastness, Punica granatum.

ÖZET

Nar kabuğu boya çözeltisi hazırlanarak yünlü kumaş boyamasında kullanılmıştır. Renk ve haslıklar üzerine mordan tipi, konsantrasyonu, kimyasal ard işlemin etkisi incelenmiştir. Boyalı kumaşlar 6.63-23.05 arasında renk verimine, orta/yüksek seviyede renk haslıklarına sahiptir. Seçilen lif örnek yüzeyleri electron mikroskobu ile gözlemlenmiştir. Boyalı kumaşların SEM fotoğraflarındaki lifler düzgün yüzeye sahiptir. Deneysel çalışma atık nar kabuğu ekstraktının yün kumaşı doğal olarak boyanması için uygun olduğunu göstermiştir.

Anahtar Kelimeler: Doğal boya, Yün, Boyama, Haslık, Punica granatum.

Corresponding Author: Mustafa Tutak, mtutak@erciyes.edu.tr, Tel: +90 352 207 66 66

1. INTRODUCTION

Natural dyes have been widely used in the coloring of various materials and natural fibers including wool, silk, cotton and leather since ancient times (1). Natural dyes are non-allergenic, and non-toxic to the human body and the environment in comparison to synthetic dyes; hence the usage of natural dyes in textile dyeing is increasing day by day (2-5). The dyeing of textile natural fibers with plant extracts is generally carried out by using different parts of the plant such as the peel, flowers, leaves and seed. The peel is preferred in dyeing because of its high percentage of coloring agent but removal of the peel from the plant can threaten the life of the plant. Hence the waste parts of pomegranate peel extract can be used as a natural dye for wool textile fabrics. Pomegranate (*Punica granatum*), which belongs to the family *Punicaceae*, is one of the most frequently consumed fruits in many regions across the World. Pomegranate fruit and juice exhibit medical properties such as antioxidant activity and antibacterial properties, and are beneficial to those with diabetes, heart disease, and cancer. Only the inside part of the pomegranate fruit is used for juice while the peel is waste. In this study, the unused parts of the pomegranate peel were used as a natural dyeing agent for wool fabric coloration. The chemical structure of the dye in pomegranate peel is shown Figure 1(6).

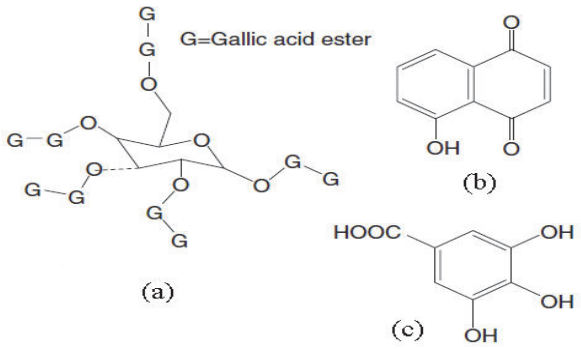

Figure 1. The chemical structure of the main components of pomegranate peel extract, (a)tannic acid, (b)juglone, and (c)gallic acid.

According to the literature survey, there are various studies on pomegranate fruit and peel. Al-Rawahi *et al*. studied the thermal characteristics of the water soluble extract of pomegranate peel. Their study demonstrated that there is a relationship among the freezing curve, glass transition line, and ultimate maximal-freeze-concentration conditions (7). Gaula investigated the ultrasound-assisted extraction of pomegranate seeds, according to seed particle size, extraction temperature, solvent/solid ratio, amplitude level, and pulse duration/pulse interval ratio (8). Dkhil studied

methanolic extract of *Punica granatum* peel *in vivo* for its pharmacological, antioxidant and anti-coccidial properties and *in vitro* for its anthelmintic activity. The study revealed that pomegranate as a natural product has protective effects against *E. papillata*-induced coccidiosis as well as possessing anthelmintic activity (9). Devatkal *et al.* demonstrated the antibacterial activity of pomegranate peel against bacteria isolated from poultry meat. Their results showed the potential application of pomegranate peel extract as an antibacterial agent against *P. stutzeri* (10). Altunkaya *et al.* investigated the antioxidant capacity of pomegranate peel powder, a by-product of the pomegranate juice industry rich in polyphenols, for use in bread production, due to its potential health effects (11). Saad *et al.* studied the phenolic extractives of pomegranate peels; their analysis shows fluctuating rates of total polyphenols, and condensed and hydrolysable tannins between the different cultivars (12). Nuamsetti *et al*. investigated the *in vitro* antibacterial activities of different extracts of pomegranate fruit peels and arils (with seeds) by the agar-well diffusion and broth dilution methods against different bacteria (13). Young *et al*. investigated the antibacterial functionality of natural colorant extracts, five kinds of natural dying aqueous solutions were obtained by extraction from peony, pomegranate, clove, coptis chinensis and gallnut (14). Eun *et al*. studied dyeing and deodorizing properties of natural colorant extracts that were obtained by extraction from gardenia, coffee sludge, and pomegranate. Their experimental results show that dyeing, color fastness, and deodorizing properties were significantly dependent on the concentration of extracts, the structure of colorant, and the kind of fabrics (15). There are limited studies related to the dyeing properties of pomegranate peel dye. Therefore, in the present study, its usage in the dyeing of wool fabric was investigated in terms of utilization of pomegranate fruit peel waste (16).

2. MATERIAL AND METHOD

2.1. Fabric

The wool fabric was supplied ready for dyeing by Yünsa Textile Company (Turkey). The characteristics of the fabric were as follows: woven twill 1/1, 155 g/m^2. The warp and weft yarns density of these fabric were 32 and 35 threads per cm, respectively.

2.2. Pomegranate peel dye extraction and wool fabric mordanting

Waste pomegranate fruit peel was used as a natural dye source in the present study; this was supplied from AKDEM herbal firm (Yalova/Turkey). The raw materials were kept in boiled distilled water (the rate of dry natural dye source to distilled water was 1:20) for the purpose of extraction. The obtained colored solutions were filtered and used in the dyeing process.

The metal salt iron sulfate was used as mordant (M1); (M0) denoted no-mordant application. The dyeing procedure of the textile fabrics using a pre-mordanting technique is shown in Figure 2.

2.3. Dyeing and post-treatment

All of the dyeing processes were carried out in a laboratory dyeing machine (Termal, Turkey) at a liquor ratio of 1/20 according to the exhaustion technique. The dyeing and post-treatment diagram is shown in Figure 2. The dyeing process was started at 25 ^{o}C. The fabric, mordant and natural dye were added respectively, and the temperature was then raised to 90 ^{0}C. Dyeing continued at this temperature for a further 45 min. After dyeing, the selected dyed fabrics were treated with tannic acid at different concentrations and then further washing of the dyed fabric was carried out in cold, boiled, boiled with non-ionic detergents and cold rinsing. It was reported in the literature (17-18) that color fastness could be developed with the use of tannic acid. In this study, it was aimed to obtain high color fastnesses by the use of tannic acid in different concentrations.

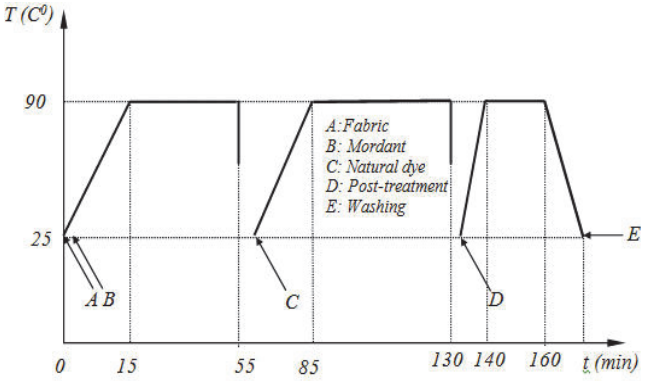

Figure 2. Mordanting, natural dyeing, and post-treatment diagram

2.4. Determination of color performance

The spectral reflectance measurements of the naturally dyed fabrics were determined using a Konica Minolta 3600d spectrophotometer. Color strength was expressed as K/S values of the dyed samples. The light fastness of the naturally dyed fabrics was determined according to the TSE 1008 EN ISO 105-B02 standard. Irradiation of the dyed fabrics was carried out using a Xenotest 150S+ light fastness test instrument (Atlas). The washing and rubbing fastnesses of the naturally dyed samples were determined according to TS EN ISO 105-C06, and TS 717 EN ISO 105-X12 standards, respectively. The dyed samples were conditioned at 25 ^{o} C and a relative humidity of 60 % for 4 h before testing.

2.5. SEM analysis

A scanning electron microscope (SEM) ZEISS EVO LS10 was used to characterize the surface morphology of the wool fibers after the dyeing process. The fiber sample surfaces were analyzed at Erciyes University Nanotechnology Research Center.

3. RESULTS AND DISCUSSION

3.1. Color and fastness properties

Table 1 shows the color properties of the selected naturally dyed fabrics in which there was no-mordant application depending on the dyeing conditions.

The selected dyeing results of the no-mordant wool fabric are given in Table 1. Light brown colors were obtained with direct application (no-mordant) of the colored solution to the wool fabric. The K/S color strength values increased as the pomegranate peel amount increased. The D2 and D3 K/S values are higher than D1. When the fastness results were examined, it was found that light, rubbing, and washing

fastness had the same level in general. Tannic acid post-treatment had no positive/negative effect on no-mordant dyeing according to the color strength and fastness levels.

Table 1. Color properties of dyed wool fabrics with no-mordant (M0) application

Sample number	Mordant and concentration	Pom. peel amountl	Post treatment type and concentration	Obtained color	K/S color strength	CIE color coordinates		
						L^*	a^*	b^*
D1	M0	50	-		10.01	63.7	2.01	29.11
D2	M0	100	3% tannic acid		16.74	59.8	5.92	33.14
D3	M0	100	5% tannic acid		15.74	59.37	5.95	32.13

Table 2. Color properties of dyed wool fabrics subjected to iron sulfate-mordant (M1) application

Sample number	Mordant and concentration	Pom. peel amount (%)	Post-treatment type and concentration	Obtained color	K/S color strength	CIE color coordinates		
						L^*	a^*	b^*
D4	M1 2%	5	-		6,63	42.48	2.97	8.79
D5	M1 5%	10	-		10,13	35.91	2.69	7.5
D6	M1 5%	100	Tannic acid 3%		21,04	24.01	3.55	4.43
D7	M1 10%	50	-		17,95	27.95	2.93	5.25
D8	M1 10%	100	-		23,05	19.54	1.15	1.55
D9	M1 20%	100	-		18,24	26.42	2.26	4.11
D10	M1 20%	100	Tannic acid 3%		20,11	25.22	2.27	4.36
D11	M1 30%	100	-		22,60	20.88	0.89	2.59
D12	M1 30%	100	Tannic acid 3%		21,84	22.97	1.29	4.01

Table 3. Light, rubbing, and washing fastness levels of naturally dyed fabrics

Sample number	Light fastness	Rubbing fastness		Washing fastness	
		dry	wet	staining	changing
D1	3/4	5	4/5	5	5
D2	4	5	4/5	5	5
D3	4	5	4/5	5	5
D4	5	5	4/5	5	4/5
D5	5	5	4/5	5	4/5
D6	6	4	3/4	5	4
D7	6	4/5	4	5	4
D8	5	4	3/4	4/5	5
D9	5	4/5	3/4	5	5
D10	5	5	4/5	5	4/5
D11	5/6	5	4	5	5
D12	6	4/5	3/4	5	5

3.2. Naturally dyed wool fiber surface analysis

The SEM images (Figure 3, a-d) show no distinguishable physical modification of the dyed fiber. The surface of the dyed wool fiber is smooth and undamaged. There are mineral deposits on the surface of the wool fibers (3b, 3c, 3d) depending on the usage of the M1 mordant but there are no mineral deposits on the wool fiber surface (4a) using the no-mordant application.

4. CONCLUSION

The waste parts of pomegranate peel were used as a natural dyeing agent for wool fabric natural coloration. For this purpose, iron sulfate mordant was used at different concentrations and tannic acid post-treatment was conducted. Pomegranate peel natural dye solutions were extracted and applied to 100% wool fabrics. The natural dyeing performances of these fabrics were determined considering color strength (K/S), SEM analysis and color fastness properties. The color strenght of the natural dye on the wool fabric was found to be highly dependent of the pomegranate peel amount (%) and mordant concentration. Un-mordanted dyed samples had good properties of rubbing and washing fastness. Mordant iron sulfate was found to give good light fastness (rating 5-6). The 5000X magnification SEM image of the treated with iron sulfate mordant showed formation mineral deposits on the surface of the dyed wool fiber. According to the experimental results waste pomegranate peel extract dye is appropriate for wool fabric natural dyeing in term of sufficient color and fastness properties.

ACKNOWLEDGEMENT

The authors are grateful to the Ministry of Science, Industry and Technology for financial support: project number “00770.STZ-2011-1” and title “Developing of natural dyed fabric friendly with nature and human health”.

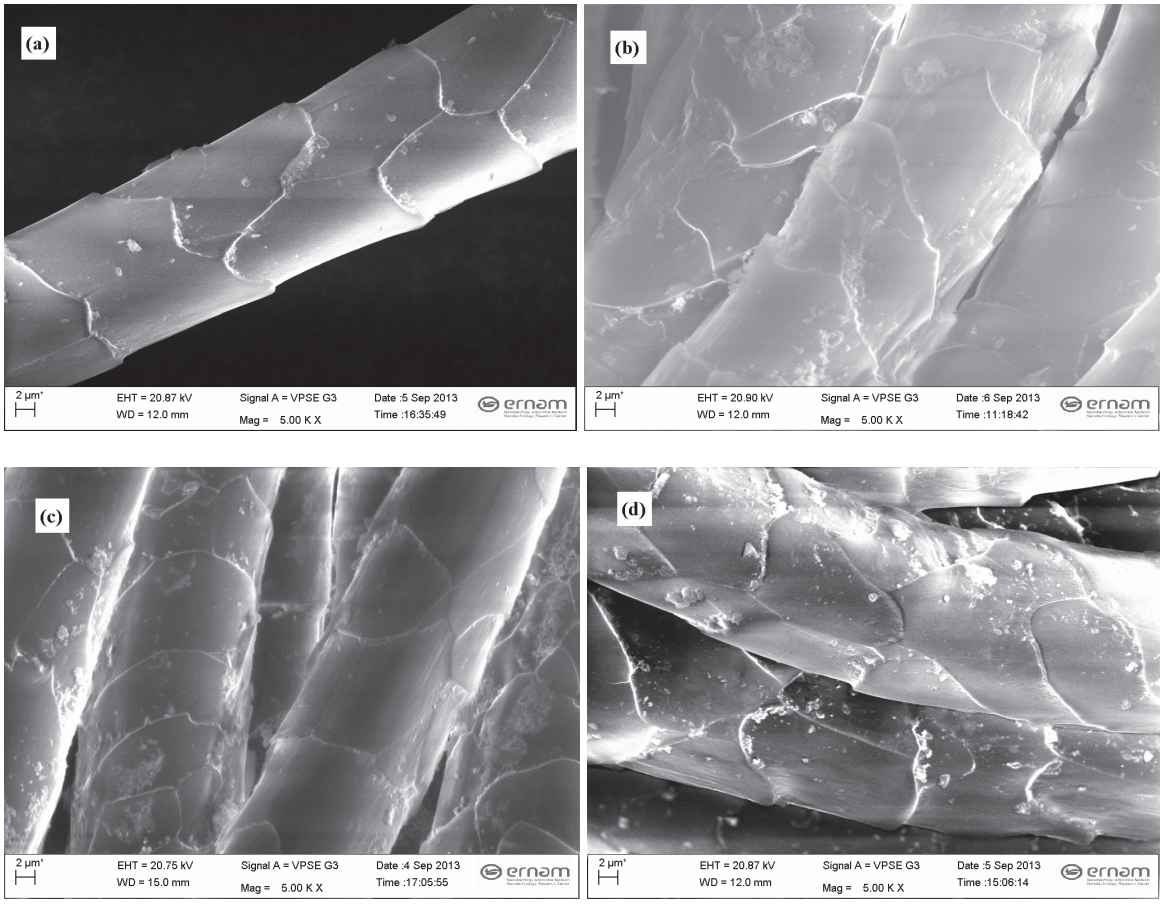

Figure 3. Selected SEM images at 5000X magnification of naturally dyed fibers (a:non-mordant D2, b:5% mordant D5, c:10 % mordant D8, d: 20% mordant, D10)

REFERENCES

1. Bechtold, T., Mussak, R., 2009, "Historical Aspect", *Handbook of Natural Colorants*, Wiley, Belgium, pp:3-17.
2. Hou, X.L., Chen, X.Z., Cheng, Y.X., Xu, H.L., Chen, L.F., Yang, Y.Q., 2013, "Dyeing and UV-protection properties of water extracts from orange peel", *Journal of Cleaner Production*, 52, pp:410-419.
3. Shahid, M., Shahid-ul-Islam, M., F., 2013, "Recent advancements in natural dye applications: a review", *Journal of Cleaner Production*, 53, pp: 310-331.
4. Boga, C., Delpivo, C., Ballarin, B., Morigi, M., Galli, S., Micheletti, G., Tozzi, S., 2013, "Investigation on the dyeing power of some organic natural compounds for a green approach to hair dyeing", *Dyes and Pigments*, 97, pp:9-18.
5. Tutak, M., Korkmaz, N.E., 2012, "Environmentally Friendly Natural Dyeing of Organic Cotton", *Journal of Natural Fibers*, 9, pp:51-59.
6. Ghaheh, F.S., Nateri, A.S., Mortazavi, S.M., Abedi, D., Mokhtari, J., 2012, "The effect of mordant salts on antibacterial activity of wool fabric dyed with pomegranate and walnut shell extracts", *Coloration Technology*, 128, pp:473-47.
7. Al-Rawahi, A., Rahman, M.S., Waly, M., Guillemin, G.J., 2013, "Thermal characteristics of a water soluble extract obtained from pomegranate skin: Developing a state diagram for determining stability", *Industrial Crops and Products*, 48, pp: 198-204.
8. Goula, A.M., 2013, "Ultrasound-assisted extraction of pomegranate seed oil - Kinetic modeling", *Journal of Food Engineering*, 117, pp:492-498.
9. Dkhil, M.A., 2013, "Anti-coccidial, anthelmintic and antioxidant activities of pomegranate (Punica granatum) peel extract", *Parasitology Research*, 112, pp:2639-2646.
10. Devatkal, S.K., Jaiswal, P., Jha, S.N., Bharadwaj, R., Viswas, K.N., 2013, "Antibacterial activity of aqueous extract of pomegranate peel against Pseudomonas stutzeri isolated from poultry meat", *Journal of Food Science and Technology-Mysore*, 50, pp:555-560.
11. Altunkaya, A., Hedegaard, R.V., Brimer, L., Gokmen, V., Skibsted, L.H., 2013, "Antioxidant capacity versus chemical safety of wheat bread enriched with pomegranate peel powder", *Food & Function*, 4, pp:722-727.
12. Saad, H., Charrier-El, B.F., Pizzi, A., Rode, K., Charrier, B., Ayed, N., 2012, "Characterization of pomegranate peels tannin extractives", *Industrial Crops and Products*, 40, pp:239-246.
13. Nuamsetti, T., Dechayuenyong, P., Tantipaibulvut, S., 2012, "Antibacterial activity of pomegranate fruit peels and arils", *Scienceasia*, 38, pp:319-322.
14. Young, H.L., Eun, K.H., Han, D.K., 2009, "Colorimetric Assay and Antibacterial Activity of Cotton, Silk, and Wool Fabrics Dyed with Peony, Pomegranate, Clove, Coptis chinenis and Gallnut Extracts", *Materials*, 2, pp: 10-21.
15. Eun, K.H., Young, H.L., Han, D.K., 2008, "Dyeing, fastness, and deodorizing properties of cotton, silk, and wool fabrics dyed with gardenia, coffee sludge, Cassia tora. L., and pomegranate extracts", *Fibres and Polymers*, 9, pp:334-340.
16. Acar, G., 2013, "Developing of natural dyed fabric friendly with nature and human health", M.Sc. Dissertation, Erciyes University, Kayseri/Turkey.
17. Burkinshaw, S.M., Kumar, N., 2008, "A tannic acid/ferrous sulfate aftertreatment for dyed nylon 6,6", *Dyes and Pigments*, 79, pp: 48-53.
18. Sheikh, M.R.K, Farouqui, A.N., Yahya, R., Hassan, A., 2011, "Effect of Acid Modification on Dyeing Properties of Rajshahi Silk Fabric with Different Dye Classes", *Fibres and Polymers*, 12, pp:642-647.
19. Guesmi, A., Ben Hamadi, N., Ladhari, N., Sakli, F., 2012, "Dyeing properties and colour fastness of wool dyed with indicaxanthin natural dye", *Industrial Crops and Products*, 37, pp:493-499.
20. Lee, Y.H., Kim, H.D., 2004, "Dyeing properties and colour fastness of cotton and silk fabrics dyed with Cassia tora L. extract", *Fibers and Polymers*, 5, pp:303-308.