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IMPROVEMENTS IN DYEING OF NATURAL FIBERS WITH ECO-FRIENDLY SAFFRON NATURAL DYE USING CHITOSAN

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ABSTRACT

Natural coloring matter extracted from Saffron plant (*Crocussativus*) used for dyeing silk and poly amide fibers by conventional and microwave heating methods. Dyeing conditions as dye extracted quantity, dye concentration, pH values, temperature and time of dyeing were studied. The silk and poly amide fibers were pretreated with chitosan of different concentrations. Chitosan pre-treatment were carried out instead of mordant. Color strength (K/S) values was measured for dyed silk and poly amide fibers. The effect of the antimicrobial activity towards some kinds of bacteria and fungi were also assessed and the results obtained exhibits high inhibition percent. The results obtained showed that fastness properties such as light, washing, rubbing and perspiration of treated dyed fibers are higher than the untreated. The results of dye extraction and color strength (K/S) of silk and poly amide fibers exhibited that microwave heating method is more effective than conventional method. Dyeing by microwave heating method saving time and energy as shown from the results, it is economical method and eco-friendly, it is also produce a higher dye uptake as compared to conventional method. The results show that, the treated fibers exhibited higher results than the untreated. Natural dyes were used as substitute of synthetic dyes because it is non-polluting, and involve inexpensive equipment and small-scale operations.

KEYWORDS: Chitosan, Natural dye, Dyeing, Textile, Antimicrobial activity.

INTRODUCTION

Saffron is a perennial plant with the botanical name of *Crocus Sativus Linn* belongs to family *Iridaceae*. This plant is the red stigma of the flower is harvested in the morning when the flowers are open. The collected stigma is then dried and converted into powder. Approximately 1 kg of flower is required to produce 12g of dried saffron spice. Saffron, Carthamus tinctorius, L., is an annual plant, which is cultivated mainly for oil which is extracted from alkenes (fruits), used in food industry because of high concentration of saturated fatty acids and especially of linoleic acid (vitamin F). substance, groups which produce the color and 13 hydroxyl groups (-OH) which play the role of auxochromes intensify the color. There are only two or three places in the world where Saffron (*Crocus sativus*) grows. Kashmir is one of these places. Saffron (*Crocussativus*) plants very small and its flower is the only part which is seen above the ground. The blooming time of this flower is autumn. Saffron has a special sweet smell and it is widely used as natural dye in dyeing, cosmetic industry and cooking (Becht et al., 2006; Teli et al., 2000). It is commonly known as Crocus, it consists of dried stigmas and upper parts of styles of plant *Crocus sativus* Linn. The image of the saffron, saffron powder and the chemical structure of the coloring component are shown in Figure 1.

Recently, the textile industry must go towards developing of new technologies to minimize the energy and water consumption. The use of microwave in textile industry is one way for this purpose (Ali and.EL-Mohamedy,2011).The advantages of microwaves microwave heating technique, is the use much less liquid, they can save dyes and leave no waste of dye comparing to conventional methods. Also the less power consumption leading to production of desired shades, and fast dyeing (Hebeish et al., 2012).

Natural dyes help in protecting the environment and reduce pollution by industries (Singh, 2000). Recently, the ability of using natural dyes in dyeing textile as UV-protection and anti-microbial has been investigated. Some natural dyes require the use of mordents, such as aluminum, iron, chromium, copper and tin, to increase color fastness to light and washing. In this study we use chitosan instead of mordents. Textile materials are subjected to attack by microbes because they provide the basic requirements for growth of microbes (Ali and El-

Mohamedy, 2011; Gao, 2008). For example (keratin) and silk as natural fibers is made of protein, which provide moisture, oxygen, nutrients and temperature for bacterial growth and multiplication. This results in odors, infection, product d Avadi, eterioration allergic responses, consequently causes related diseases (Thiry, 2009; Avadi et al., 2004).

In this study we use high-quality and ecofriendly natural dye extracted from plant, and treated the fibers with natural product, this improve our environment and produce smart fibers by economic methods. The antimicrobial activity was then measured for wool and silk fibers dyed with saffron dye using different species of microbes as *Aspergillus niger*, *Pseudomonas aeruginosa* and *Staphylococcus aureus*. It is important to promote non-polluting natural dyes, which involve inexpensive equipment and small-scale operations.

A B C

Figure 1: Saffron Plant (A), Saffron Powder (B) and Structures of the Chemical Constituents of Saffron.

2 MATERIAL AND METHODS

2.1. Material

Silk and polyamide fibers were supplied by El Mahalla company-Egypt.

2.1.2. Chemicals

Chitosan high molecular weight (210,000), Poly (D-glucoseamine), was referred to as chitosan III , Chitosan medium molecular weight (100,000), Poly (D-glucoseamine), was referred to as chitosan II , Chitosan low molecular weight (1800), Poly (D-glucoseamine), was referred to as chitosan I, were purchased from ROTH, Germany.

2.1.3. Pretreatment of silk and polyamide fibers with chitosan

Chitosan (low, medium and high molecular weights) solutions were freshly prepared by dissolving1.0 g/l) in distilled water containing acetic acid (4g/l). The silk fibers were immersed in these solutions at a 50:1 liquor ratio for 30 min, and then thoroughly washed, and air dried at room temperature.

2.1.4. Dyeing silk fibers

In a dye bath containing different amounts of saffron dye with liquor ratio 30:1, silk fibers were dyed using conventional heating at different pH values (3-9) for different durations (30-60 min) and at different temperatures(50-90ºC).The silk fibers was dyed by microwave heating at pH 4 for 4 sec with a liquor ratio 1:100, . The dyed samples were rinsed by warm water and then cold water, washed in a bath containing 5g/l non-ionic detergent at 50ºC for 30 minutes, then rinsed and dried at room temperature.

2.1.5. Dyeing polyamide fibers

In a dye bath containing (10 g/l) of saffron dye with a liquor ratio 1:100, the polyamide fibers untreated and pretreated with chitosan low, medium and high molecular weight were dyed by microwave for 5 min.

2.1.6. Measurements of Color strength (K/S value)

An Ultra Scan PRO spectrophotometer was used to measure the reflectance of the samples and hence, the K/S was measured spectrophotometrically at wave length 500 nm. The K/S of untreated and pretreated wool fabrics with chitosan and tannic acid was evaluated.

2.1.7. Fastness properties

The dyed samples were washed-off using 2 g/l nonionic detergent at 80°C for 30 minutes, and tested according to ISO standard methods. The specific tests were ISO 105-X12 (1987), ISO 105-C02 (1989), ISO 105-E04 (1989), and ISO 105-B02 (1989), corresponding to color fastness to rubbing, washing, perspiration and light, respectively. The color changes of the samples were assessed against an accurate Gray scale.

2.1.8. Antimicrobial activities

The antimicrobial activity of silk and polyamide fibers pretreated with chitosan and dyed with saffron natural dye were measured according to the AATCC 100-1999 test method against Gram-positive bacteria *Staphylococcus aureus* and Gram-negative bacteria *Escherichia coli* as well as against the fungus *Aspergillus niger*. The antimicrobial organisms. The percentage reduction is calculated using the following activity is expressed in % reduction of the surviving.

Reduction % (CFU/ml) = A-B/A X100.

Where A are the surviving cells forming unities (CFU/ml) for the untreated fibers (control) , and B the surviving cells forming unities (CFU/ml) of the treated fibers (AATCC 100.1999) .

3. RESULTS AND DISCUSSION

3.1. Effect of chitosan pretreatment at different conc .on silk fibers and dyeing with saffron using microwave

Silk fibers pretreated with chitosan and dyed with saffron natural dye using microwave and conventional methods and the results indicated that chitosan III gave higher K/S values compared to I&2 as shown in Figure 2&3. We noticed also that microwave method exhibited higher results than conventional method.

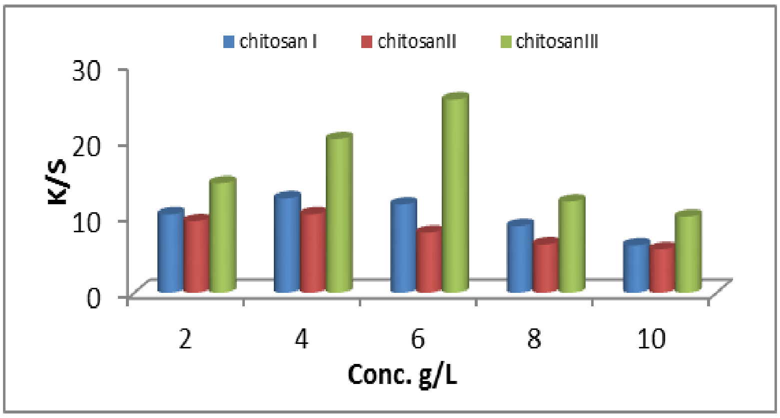

Figure 2: Effect of chitosan pretreatment at different conc. on dyeing silk fibers using microwave.

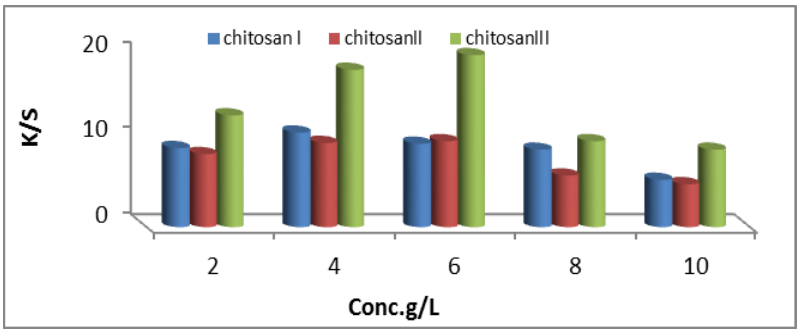

Figure 3: Effect of chitosan pretreatment at different conc. on dyeing silk fibers using conventional method.

3.2. Effect of chitosan pretreatment on dyeing poly amide fibers using microwave

Table 1 shows the colorimetric data (L*, a* and b*) of different fibers dyed with different extracted dye concentration. From data listed in table we can be concluded that increasing of the extracted dye concentration, accompanied by decreasing of L* values and thus color of samples got darker. As the dye concentration increase, a* and b* values increased in the positive direction. The color of dyed polyamide fibers turned to more reddish yellow color and became darker with increasing the dye concentration from 0.5% to 2%.

Table 1: Color data of dyed polyamide fibers pretreated with Chitosan at different conc.

Conc. %	K/S	L*	a*	b*	C*	H	∆ E
0.5	10.32	59.20	19.92	9.09	21.90	24.53	42.93
1.0	6.52	51.11	31.52	14.58	34.73	24.82	52.58
1.5	15.82	50.3	35.7	15.8	35.74	25.7	55.78
2.0	16.34	42.5	36.7	16.8	37.73	26.5	57.58
Untreated	7.61	53.90	35.05	20.94	40.83	30.85	30.12

3.3. Effect of dye amount

Chitosan bind to the fiber by chemical bonds either to the terminal $-NH_2$ of silk fiber or -COOH groups of the polypeptide chain or to the functional groups present in the side chains of the amino acids (Pascual and Julia, 2001; Dutta and Ravikumar, 2002) Figure 4 showed that the treated fibers exhibit higher values of K/S than the untreated samples (Ali and E.L. Mohamedy, 2010) .Natural dyes are substantive or adjective. Substantive dyes are absorbed and fixed by chemical bonds inside the fibers without chemical treatment. However, most natural dyes are adjective and need the use of mordents for absorption and fixing on fibers (Shin, 2010) and also changes the color produced by the dye.

Figure 4reveals that as the dye amount increase the K/S also increases when using both type of heating with much higher K/S value at all dye amount in microwave case as expected. This may be attributed to the effect of microwave heating mechanism by ionic conduction, which cause resistance heating which cause acceleration of the ions inside the dye solution, it results in collision of dye molecules with the molecules of the fiber. The results also showed that the K/S of treated silk fibers is much higher than untreated one. The results also show that microwave method exhibits higher K/S values than conventional method, Figure 5 shows that the highest K/S value at 90^{o}C and the treated is higher than the treated samples. Figure 6 shows that the highest K/S value at pH 5 for both microwave and conventional methods, it is also noticed that microwave method exhibited higher K/S values than conventional method.

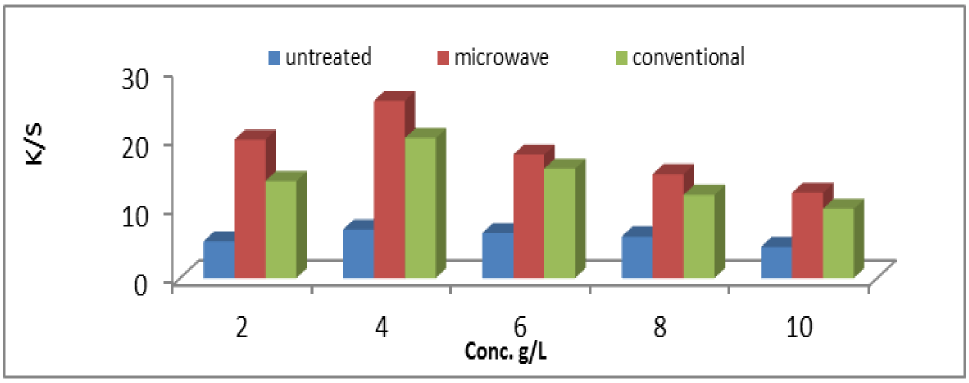

Figure 4: Effect of extracted dye concentration on color strength of treated and untreated dyed silk fibers using microwave and conventional methods.

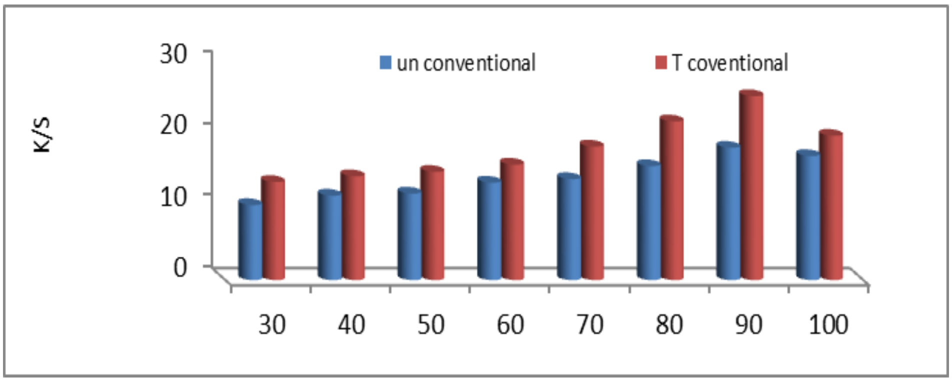

Figure 5: Effect of temperature on the color strength for treated and untreated dyed fibers. Nu: untreated, T: Treated.

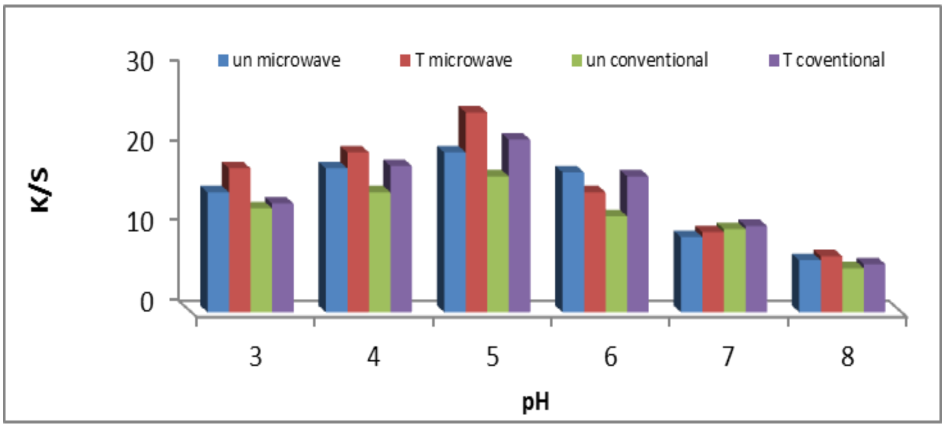

Figure 6: Effect of pH on the color strength for treated and untreated dyed fibers Nu: untreated, T: Treated.

3.4. Effect of time on the color strength for treated and untreated dyed fibers by conventional method.

Figures 7& 8 show the effect of time of dyeing silk fibers using conventional and microwave methods. The results showed that the highest values of the color strength at 50 min. for conventional method, while in microwave method the highest value of K/S obtained at 5 min.

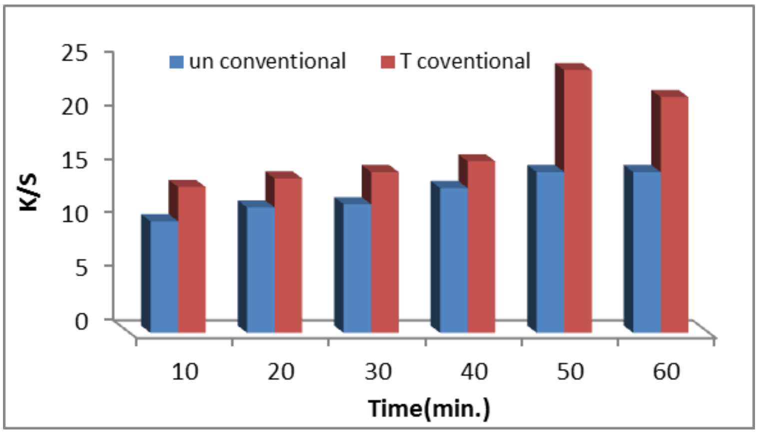

Figure7: Effect of time on the color strength for treated and untreated dyed fibers using conventional method. un: untreated, T: Treated.

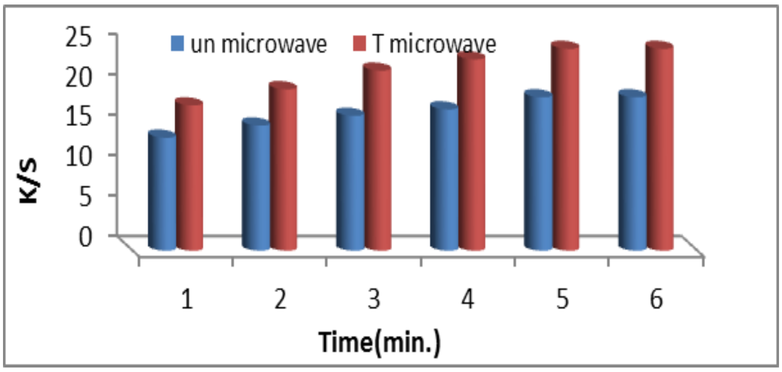

Figure 8: Effect of time on the color strength for treated and untreated dyed fibers using microwave method . un: untreated, T: Treated

3.5. Antimicrobial Activity

Two isolates of pathogenic bacteria i.e., *Staphylococcus aureus* and *Escherichia coli* as well as one isolate of pathogenic fungi *Aspergillusniger* were used to evaluate the antimicrobial activity of silk and polyamide fibers under the combined treatments effect of chitosan and saffron natural dye.

Results in tables 2 and 3 clearly show that the antimicrobial activity of silk and polyamide fibers depends on the concentration and molecular weight of chitosan. As the highest records of reduction in survived tested microbes (antimicrobial activities) were obtained at high concentrations 2 and 2.5g/L of chitosan with high molecular weight (HMW). Antimicrobial activity, varies by varying the strains of bacteria and fungi, the bacterial reduction rate increases as the molecular weight and concentration of chitosan increases. The degree of these increase depend on the kind of bacteria and fungi (Ali et al., 2015; El–Mohamedy and Aboelfetoh ,2014 a,b).Antimicrobial activities of both silk and polyamide fibers treated with natural dye and chitosan treatments were more effectiveness against *Staphylococcus aureus* G+ *than Escherichia coli* G- as well as pathogenic fungi *Aspergillus niger* .There is a slight difference in antibacterial activity between silk fibers and poly amide fibers they have the decreasing percentages of antibacterial activity. Many investigators reported that many natural dyes combined with chitosan treatment have antibacterial activity against pathogenic bacteria and fungi Antibacterial activity of prepared sample fabrics against Gram-positive (*S aureus*) is greater than antibacterial activity against Gram-negative (*E coli*), most probably due to the fact that Gram-positive bacteria cell walls consist of a single layer, whereas Gram-negative cell wall is a multilayered structure bounded by an outer cell membrane (Shtayeh et al., 1998).

Table 2: Antimicrobial activity of silk fibers pretreated with different concentrations of Chitosan with different types.

Microbes	Chitosan type	Chitosan concentration g/L				
		0	1.0	1.5	2	2.5
Staphylococcus aureus	Chitosan I	19.4	26.0	44.4	52.0	58.8
	Chitosan II	22.2	32.4	46.8	56.2	61.4
	Chitosan III	23.6	36.8	52.2	60.4	66..0
Escherichia Coli	Chitosan I	16.8	21.0	33.2	44.4	50.0
	Chitosan II	18.2	24.2	38.0	50.0	54.2
	Chitosan III	22.0	28.2	46.8	55.4	60.2
Aspergillus niger	Chitosan I	14.8	16.8	28.0	38.2	40.2
	Chitosan II	16.0	18.2	36.0	41.4	44.4
	Chitosan III	18.2	21.0	40.8	48.0	50.0

Chitosan I = Chitosan low molecular weight. Chitosan II = Chitosan medium molecular weight.
Chitosan III= Chitosan e high molecular weight.

Table 3: Antimicrobial activity of dyed poly amide fibers pretreated with different concentrations of Chitosan of different molecular weight.

Microbes	Chitosan type	Chitosan concentration g/L				
		0	1.0	1.5	2.0	2.5
Staphylococcus Aureus	Chitosan I	21.2	30.0	52.4	60.0	66.8
	Chitosan II	24.2	34.3	55.8	66.2	74.4
	Chitosan III	26.2	40.2	62.2	72.4	80.0
Escherichia Coli	Chitosan I	18.4	24.4	42.2	55.4	60.0
	Chitosan II	20.2	32.8	52.0	61.0	68.2
	Chitosan III	24.0	38.2	56.8	66.4	73.2
Aspergillus niger	Chitosan I	18.4	27.0	34.0	50.2	52.2
	Chitosan II	19.0	32.2	40.0	53.4	55.4
	Chitosan III	20.2	35.0	44.8	58.0	60.0

Chitosan I = Chitosan low molecular weight. Chitosan II = Chitosan medium molecular weight.
Chitosan III= Chitosan e high molecular weight

***3*.6. The fastness properties of the investigated dye on the silk and poly amide fibers pretreated with different concentrations of chitosan high molecular weight**.

Tables 4 & 5 showed that the fastness to washing, perspiration, rubbing and light for treated silk and polyamide fibers with chitosan high molecular weight is higher than the untreated one. It was also noticed from the results that the fastness properties enhanced at higher concentration of chitosan.

Table 4: The fastness properties of the investigated dye on the silk fibers pretreated with different concentrations of Chitosan high molecular weight.

Chitosan Conc. g/L	Perspiration		Washing		Rubbing				Light
					Acidic		Alkaline		
	S. c	Alt.	Dry	Wet	S. c	Alt	S.c	Alt	
0	3	3	4	4	4	4	4	4	5
1	4	4	4	4	4	4	4	4-5	6
2	4	4	4	4	5	5	4-5	4-5	6
3	4	5	4-5	5	4-5	5	4-5	5	6
4	4-5	5	4-5	5	4-5	5	4-5	5	7
5	5	5	5	5	5	5	5	5	7

Alt: Alteration, Sc: Staining cotton

Table 5: The fastness properties of the investigated dye on the polyamide fibers pretreated with different concentrations of Chitosan high molecular weight.

Chitosan Conc .g/L	Perspiration		Washing		Rubbing				Light
	S. c	Alt.	Dry	Wet	Acidic		Alkaline		
					S. c	Alt	S.c	Alt	
0	4	4.5	4	4	4	4.5	4	4	5
1	4	4	4	4	4	4	4	4-5	6
2	4-5	4-5	4-5	4-5	5	5	4-5	4-5	6
3	4	5	4-5	5	4-5	5	4	5	6-7
4	5	5	5	5	4-5	5	4-5	5	7
5	5	5	5	5	5	5	5	5	7

Alt: Alteration, Sc: Staining cotton

4. CONCLUSION

Chitosan low, medium and high molecular weight pretreatment of silk and poly amide fibers leads to high dye uptake. Color strength (K/S) values exhibited high values due to the treatment. The antimicrobial activity towards some species of bacteria and fungi were also measured and the results obtained exhibits high reduction percent for the treated fibers than the untreated. The fastness properties including light, washing, and perspiration of dyed fibers were assessed and gave high results. The dye extraction and color strength (K/S) of silk and poly amide fibers indicated that microwave heating is more effective than conventional method. Microwave is economical method, it save time and energy. It is also eco-friendly and produce a higher dye uptake as compared to conventional techniques.Natural dyes are non-polluting, which involve inexpensive equipment and small-scale operations.

5. ACKNOWLEDGMENTS

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