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Extraction of Natural Dyes from Turmeric *(Curcuma Longa)* and their Application on Polylactic Acid Fiber

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Abstract

This research presents an eco-friendly approach towards Response Surface Methodology (RSM). The current study is based on the optimization of the dyeing process of biodegradable polylactic acid (PLA) fiber using with natural dyes of turmeric extractions by using Soxhlet extraction technique. The dyeing process was carried out by using Daelim Starlet III, which is an infrared dyeing machine. Three independent variables were selected, including pH, time, and temperature. According to the standard test protocols, dyed samples were evaluated for color strength (K/S), CIELab values, fiber tenacity, and fiber elongation. ANOVA test results of the Linear Model exhibited that the model terms have a significant effect on K/S and the physical strength of fiber. Moreover, the observed values were in agreement with the predicted values and the suggested equation model was satisfactory and accurate. A temperature of 100 °C with pH 5 and time duration of 30 minutes in the dyeing process were observed to achieve optimum dyed PLA fibers, exhibiting appreciable strength and elongation. Lastly, dyed fibers exhibited satisfactory color fastness ratings.

Index Terms: Color strength, Exhaustion, Natural dyeing, Polylactic acid fiber, Turmeric

I. Introduction

In todays' world, scientists are more concerned about environmental protection. Moreover, this concern implicates the development of sustainable techniques, which is accompanied with the use of bio degradable and recyclable materials. Furthermore, polylactic acid (PLA) is a biodegradable thermoplastic aliphatic polyester, which is derived from renewable resources such as corn starch, tapioca roots, chips or starch, sugarcane and others. [1-4]. Also, its significant properties like

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biodegradability, sustainability, and recyclability have attracted researchers' attention in recent decades [3, 5-11]. However, an important area of PLA application is 'medical textiles', including fixation of fractures in bones, various delivery systems for drugs, and sutures which are further used in the process of surgery [4, 8, 11, 12]. PLA properties can be enhanced by dyeing it with certain natural dyes which are natural extracts of plants. Natural dyes are under investigation for coloration and functional finishing of textile fibers, including silk, wool, cotton and, others. [13-15]. In case of it, naturally dyed PLA exhibits many applications in the development of antimicrobial and biodegradable textiles. The current research is focused on the sustainable dyeing process of PLA by using Turmeric as natural dye. Additionally, the solubility of turmeric dyes in octanol/water system, dyeing process optimization, and evaluation of fiber properties after dyeing, such as tenacity and elongation, were investigated.

II. Materials and Methods

A. Materials

PLA fibers with denier 8 dtex, elongation 28.4%, strength 20.7 cN/tex were provided by Hebei Tianlun Textile Co. Ltd. China. However, Turmeric was used to extract natural dye. Moreover, laboratory scale acetic acid (analytical grade) for maintaining pH value of dye solution and acetone (analytical grade) for extraction of dyes were purchased from Sigma-Aldrich. Rucogal ACD (non-ionic), used as dispersing and levelling agent, was supplied by Rudolf Chemie (Dongguan) Co. Ltd. A commercially available detergent (alconox) was used for soaping off process.

B. Extraction

The natural dye from turmeric was extracted by Soxhlet extraction process by using (acetone) as a solvent. Raw turmeric powder (15 g) was placed in the soxhlet thimble followed by soxhlet extraction assembly. The extraction was carried out for 6 hours to obtain solution of yellow color. Acetone was evaporated from the extract by rotary evaporator (Buchi Rotavapor R-114). Then the extracted solution was stirred in hexane, filtered and dried by using laboratory scale spray dryer (YM-6000Y).

TABLE I
Design of Experiment (DOE)

Sample No.	pH	Time (min)	Temperature (°C)
1	4	45	90
2	6	45	90
3	4	75	90
4	6	75	90
5	4	45	110
6	6	45	110
7	4	75	110
8	6	75	110
9	3	60	100
10	7	60	100
11	5	30	100
12	5	90	100
13	5	60	80
14	5	60	120
15	5	60	100
16	5	60	100
17	5	60	100
18	5	60	100

C. Design of Experiment

The design of experiments was developed by using Minitab statistical analysis software. The data was analyzed to establish a relationship among various input variables and responses. The design of experiments, comprising various levels of input parameters has been shown in Table 1.

D. Dyeing

PLA fibers were dyed with the natural dye by using the concentration of 5.0% on the weight of fabric (o.w.f), Rucogal ACD (dispersing agent) 1 g/L, and dyebath liquor ratio of 1:40. The dyeing process was carried out by using infrared (IR) Daelim Starlet III dyeing machine, Korea. The dyeing profile has been shown in Figure 1. After optimizing the dyeing process, the samples were washed by using hot water, followed by washing with cold water, and subsequently dried in oven.

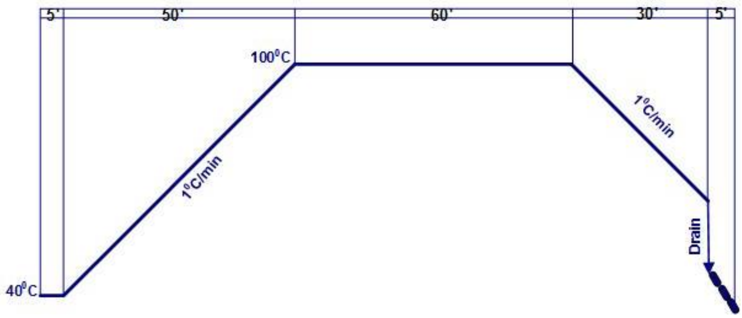

Fig. 1. Dyeing profile of PLA

E. Testing

1) Solubility:

Water solubility of turmeric dye was measured by making a solution of 0.05 g of dye in 0.05 L deionized water after continuous stirring for 24 hours. Then the solution was filtered to separate the soluble and insoluble fractions. The solid residue obtained was dried in the oven at temperature of 80 °C for 4 hours. The dried solid residue weight (W_1) and the original dye weight (W_0) were used to calculate the % dye solubility in water by using equation (1).

$$(\%) dye\ \ solubility\ \ in\ \ water\ \ = \frac{(W_0 - W_1\ )}{W_0} \times 100 \quad (1)$$

2) Hydrophobicity of Turmeric Dyes:

To determine the hydrophobicity of turmeric dyes, $\log P_{(octanol/water)}$ values were calculated by using shake-flask method. The dye concentration of 0.05 g was agitated in octanol/water system for 24 hours. The dye partitioned in octanol and water was calculated by measuring the absorbance of the dye in octanol and water separately. LogP value of dye was obtained from equation (2) by substituting the values of weight of the dye dissolved in octanol and weight of the dye dissolved in water.

$$Log\ \ P_{\left(\frac{Octanol}{Water}\right)} = Log\left(\frac{Weight\ \ of\ \ the\ \ dye\ \ dissolved\ \ in\ \ Octanol}{Weight\ \ of\ \ the\ \ dye\ \ dissolved\ \ in\ \ water}\right) \quad (2)$$

3) Color Strength Measurements:

Color strength of the dyed fibers was analyzed by measuring the *K/S* and CIELAB values by using a Macbeth Color Eye 7000 spectrophotometer. Dyed fibers were combed together to make them parallel and then attach them neatly on the cupboard. *K/S* values of the fibers were determined through Kubelka – Munk equation as given below.

$$K/S = (1-R)^2 / 2R \quad (3)$$

Where K is an Absorption Coefficient, S is Scattering Coefficient, and R is Reflectance.

4) Dye Exhaustion:

The exhaustion percentage of the dye towards PLA fibers was calculated by measuring the dyebath absorbance before and after optimizing the dyeing process. Equation (4) can be used for calculation of % exhaustion by substituting the values of A_0 and A_1.

$$Dye\ \ Exhaustion\ \ (\%) = \frac{(A_0 - A_1\ )}{A_0} \times 100 \quad (4)$$

Where, A_0 is the pre-dyeing dyebath absorbance while A_1 is post-dyeing dyebath absorbance.

5) Tensile Strength and Elongation Percentage:

PLA is a thermally sensitive fiber. Its strength can be affected by high temperature, normally which is required for the dyeing procedure of textiles. The strength of PLA fibers after dyeing procedure was tested by using FAVIMAT+ FIBER TEST. All the tests were carried out on the same fibre section.

6) Wash Fastness Test Method:

The washing fastness of dyed PLA fibers was carried out according to test method ISO 105 E01. The samples were conditioned for 5-6 hours followed by attachment with multi-fiber fabric of same size at one corner.

The samples were placed in the solution containing 4 g/L ECE detergent & 1 g/L sodium perborate in Rotawash machine, Program.: C2S Temperature: 60 °C, Time: 30 minutes, Steel ball: 25 pcs. After this, the samples were rinsed with hot water followed by cold water. Then air drying was done at a temperature not exceeding 60 °C. the staining of the multi-fiber was observed through grey scale.

III. Results and Discussion

A. Relationship Between Water Solubility and Partition of Turmeric Dyes in Octanol/ Water System

The water solubility and logP values of turmeric dye used in this study are shown in Table 2. Theoretically, logP values exhibits the degree of dye partition in octanol and water system. Dyes with higher logP values are more hydrophobic as compared to those having lower logP values. Moreover, Turmeric dye exhibits positive logP value showing that these dyes are dissolved in octanol instead of water. In textile dyeing process, this solubility behavior of dyes can be correlated to the dye ability of dyes, in terms of exhaustion (%) and color strength *(K/S)* values. However, Turmeric dye exhibits its suitability for dyeing of PLA fibers by having the poor water solubility and higher solubility in octanol as investigated by logP values. Suesat [16] reported that the dyes having solubility parameters close to the polyester (PET) would be much suitable for PLA fibers.

TABLE II

Solubility and Partition of Turmeric Dyes in Octanol/Water system

Dye	**Water solubility (% wt)**	**LogP (octanol/water)**	**Shades obtained**
Turmeric	1.80 ±0.05	1.86 ±0.5	Bright yellow

B. Effects of Dyeing Parameters on Dye Ability of PLA Fibers

The exhaustion%, *K/S* and *CIELab* values were measured after dyeing procedure as shown in Table 3. The optimization of results was carried out using 3D response surface plots by Minitab statistical software to get more conclusive results.

TABLE III

Exhaustion %, Color Strength *(K/S)*, and CIELab Values of the PLA Fibers after Dyeing with Turmeric Dye

Sample No.	**pH**	**Time**	**Temperature**	**Exhaustion**	**CIELab and Color strength (*K/S*) values**					
		Min	°C	%	*L**	*a**	*b**	*C**	*h*	*K/S*
1	4	45	90	46.24	70.83	0.76	45.46	45.47	90.96	5.37
2	6	45	90	33.16	68.19	-0.73	41.78	41.78	91.01	5.35
3	4	75	90	57.07	70.39	-0.09	48.89	48.89	90.11	6.48
4	6	75	90	53.01	72.62	-2.33	41.58	41.64	93.21	4.08
5	4	45	110	37.35	71.58	-2.02	41.08	41.13	92.81	4.02
6	6	45	110	35.00	71.83	-1.08	45.78	45.79	91.35	5.12
7	4	75	110	48.71	69.07	-0.04	44.76	44.76	90.05	5.68
8	6	75	110	50.98	71.89	-2.11	44.61	44.66	92.71	4.73
9	3	60	100	32.07	75.60	-4.60	36.84	37.13	97.12	2.88

Sample No.	**pH**	**Time**	**Temperature**	**Exhaustion**	**CIELab and Color strength (*K/S*) values**					
10	7	60	100	45.03	66.10	1.32	35.26	35.28	87.85	4.14
11	5	30	100	34.87	71.46	-0.02	47.74	47.74	90.02	5.66
12	5	90	100	48.94	71.96	0.49	43.15	43.16	89.35	4.37
13	5	60	80	38.67	72.60	-2.82	37.85	37.95	94.26	3.20
14	5	60	120	39.21	69.48	-0.76	34.25	34.26	91.27	3.44
15	5	60	100	60.15	64.83	2.93	45.90	46.00	86.35	6.45
16	5	60	100	32.11	67.52	1.73	44.26	44.30	87.76	5.88
17	5	60	100	41.46	71.28	-0.30	47.54	47.54	90.37	5.73
18	5	60	100	44.57	70.39	0.28	46.77	46.78	89.63	5.74

1) *Evaluation of Exhaustion Percentage for Turmeric Dyes:*

The exhaustion% of the dye has been investigated as a function of pH and time at constant temperature (100 °C) as shown in Figure 2. It can be observed that pH exhibits no significant effect on the exhaustion. However, the dyeing time has a significant influence on exhaustion of turmeric dyes. An increase in exhaustion was observed by increasing the dyeing time. Further, it is a common phenomenon in most cases of textile dyeing that longer the time duration of contact is between the dye and substrate, the greater the results of exhaustion. Figure 3, shows the effects of dyeing pH value and temperature range on turmeric dye exhaustion by keeping the dyeing procedure time duration constant (60 minutes). The maximum exhaustion was achieved at 100 °C and further increase in temperature caused decrease in exhaustion. In addition to it, the increase in exhaustion up to 100 °C has been attributed to the opening of fiber pores at elevated temperature. At temperature above 100 °C, it can cause the enhanced exchange of dye molecules between the fiber and dye solution which is further owing to the higher kinetic energy of dye molecules. It can compel the dye molecules to stay in solution instead of going into pores of PLA fbers. However, at lower temperature, the cohesive forces between the dye molecules are dominant because they are generating the tendency of aggregation which ultimately results in the lower exhaustion. At low temperature the dye molecules do not have the enough kinetic energy to cross the barrier and penetrate into the PLA fibers. The structure of the PLA fiber is compact, so at lower temperature the movement of molecular chains of PLA fibers is not significant. This evaluation results in low penetration among the molecular chains of fibers. But at higher temperature close to 100 °C, the movement of the molecular chains begins to accelerate to produce inter-chain spacing for the dye molecules to move into the fibers. The rate of exhaustion can be increased by using some auxiliaries but the aim of this research is to omit the addition of dyeing auxiliaries to reduce the effluent load and to develop an ecological process.

Whereas, a research group investigated a range of nine disperse dyes (concentration 2%) for PLA dyeing and found that the seven out of nine dyes exhaust below 80% [17]. They claimed that higher temperature and longer dyeing time can increase exhaustion rate. Moreover, they recommended small and linear chain dyes with no carbonyl groups for dyeing of PLA fibers to achieve higher exhaustion rates. Since, turmeric dyes are linear but the presence of carbonyl group may result in the lower exhaustion percentages. The optimization of dyeing parameters has been carried out to get enhanced exhaustion%.

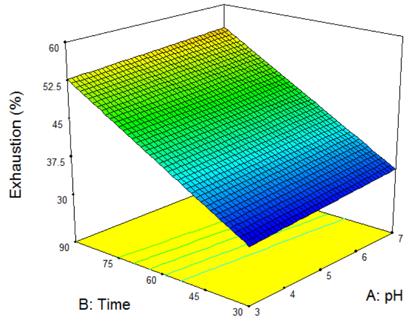

Fig. 2. Evaluation of dye exhaustion by keeping temperature constant (100 °C)

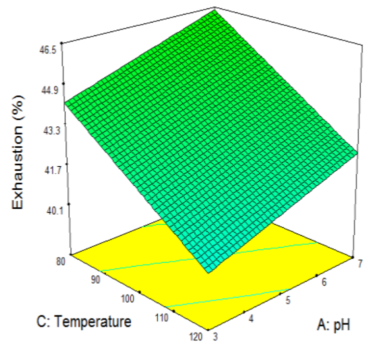

Fig. 3. Evaluation of dye exhaustion by keeping time constant (60 minutes)

2) Evaluation of Color Strength (K/S):

Figure 4, shows that the color strength value increases with an increase in pH value but after a certain threshold, it decreases with the further increase in pH value. Further, the increase in dyeing time enhances the *K/S*. A similar behavior regarding *K/S* has been exhibited by temperature as shown in Figure 5. It can be observed that with an increase in temperature the color strength value increases with the increase in pH value, then it decreases. The lower K/S at higher temperature may be attributed to the unstable structure of the natural dye (turmeric). However, PLA fiber modification by varying the proportions of the D- and L- isomers can enhance exhaustion and color strength. High D-isomer levels in PLA fiber exhibits higher exhaustion and higher color yield as compared to those having low-D isomer because of its higher amorphous, less crystalline nature [18].

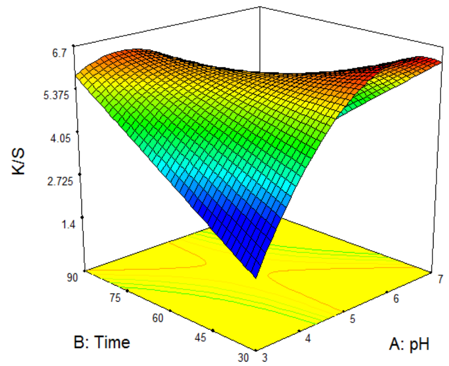

Fig. 4. Evaluation of color strength (K/S) by keeping temperature constant (100 °C)

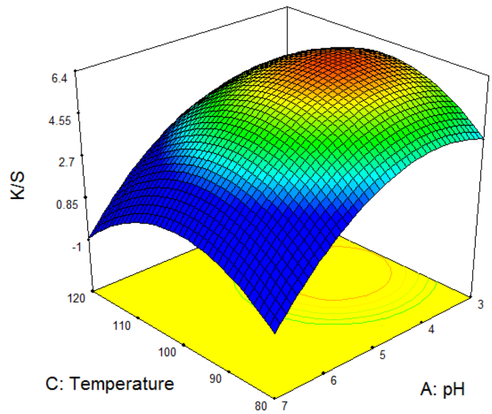

Fig. 5. Evaluation of color strength (K/S) by keeping time constant (60 min)

3) Evaluation of Lightness (L):*

Figure 6, shows the effect of dyeing time and pH value (keeping temperature constant) on lightness values of PLA fibers. It was observed that changing the dyeing time-duration does not exhibit a significant change in lightness but decrease in pH value from 7 to 3 by showing the tremendous increase in lightness values. Figure 7, shows the effect of pH and dyeing temperature (keeping dyeing time constant) on the lightness values. It was observed that changing the dyeing temperature does not exhibit a significant change in lightness. Anyhow, it is decreasing the pH value from 7 to 3, by causing to increase lightness tremendously.

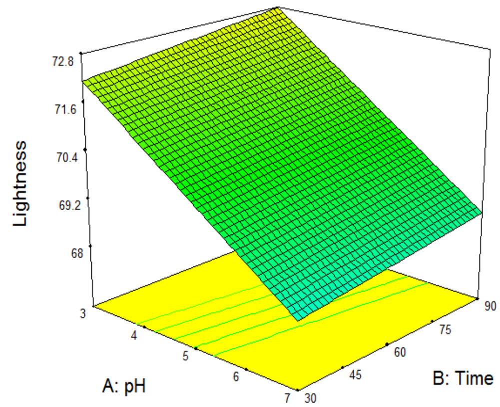

Fig. 6. Evaluation of dyed PLA lightness (L*) by keeping temperature constant (100 °C)

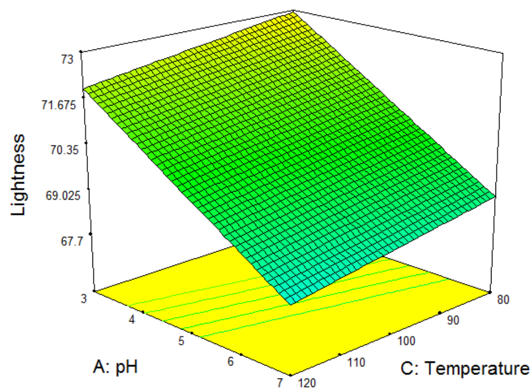

Fig. 7. Evaluation of dyed PLA lightness (L*) by keeping time constant (60 min)

4) Evaluation of Chroma (C):*

The chroma *(C*)* values of PLA fibers dyed using turmeric dyes have been measured as a function of pH and dyeing time as shown in Figure 8. It was observed that increasing pH value from 3 to 5 results in higher chroma values. While, the change in pH value from 5 to 7 results in the decrease of chroma. However, the dyeing time-duration has no significant effect on the chroma values. Figure 9, shows the effect of dyeing pH value and temperature (at constant dyeing time) on *C** values. In this case, pH value and temperature range both exhibit significant effect on chroma values. It was observed that higher temperature above 100 °C and lower pH below 5 showed a decrease in *C** values. These observations can be correlated to the effect of pH value and temperature range on the exhaustion as discussed earlier.

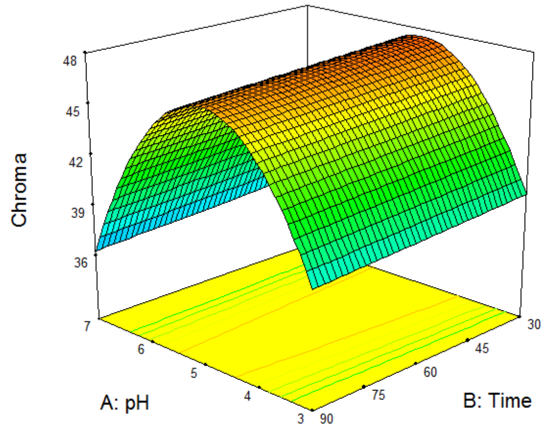

Fig. 8. Evaluation of dyed PLA chroma (C*) by keeping the temperature constant (100 °C)

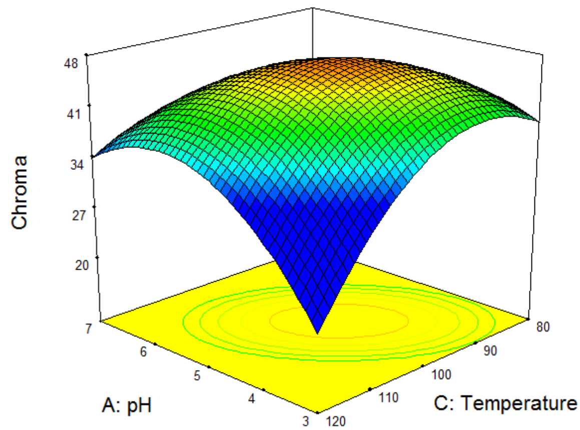

Fig. 9. Evaluation of dyed PLA chroma (C*) by keeping the time constant (60 min)

C. Effects of different dyeing conditions on strength and elongation of dyed PLA fibers

The effects of temperature, time of treatment, and pH on the tenacity and elongation of treated PLA fibers have been shown in Table 4. Graphically, it has been shown in Fig. 10 & 11.

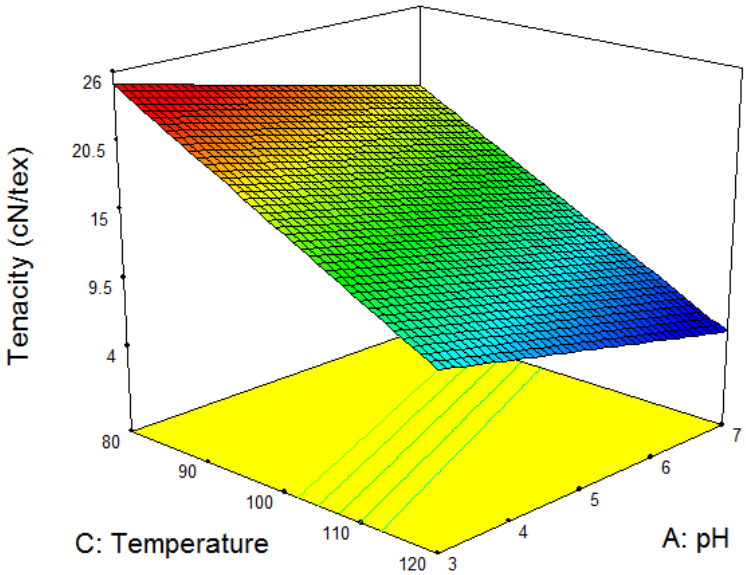

Fig. 10. Effects of dyeing temperature and pH value on the fiber tenacity (dyeing time 60 minutes)

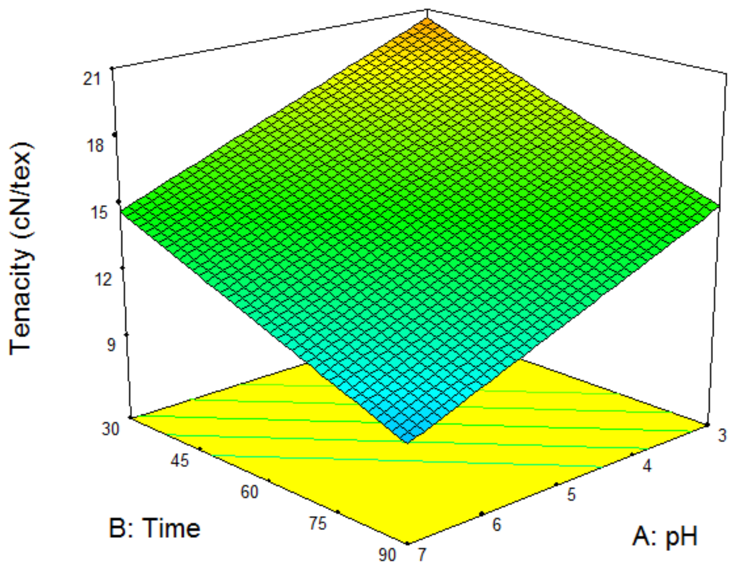

Fig. 11. Effects of dyeing time and pH value on the fiber tenacity (temperature 100 °C)

TABLE IV

Effects of pH value, dyeing time, and temperature range on fiber tenacity and elongation by using the natural dyes from turmeric

Samples	pH	Time	Temperature	Dyed PLA fibers	
		Minutes	°C	Tenacity (cN/tex)	Elongation (%)
1	4	45	90	13.7	13.67
2	6	45	90	19.41	24.83
3	4	75	90	19.73	22.09
4	6	75	90	14.22	14.47
5	4	45	110	18.09	20.40
6	6	45	110	8.21	6.19
7	4	75	110	8.81	9.04
8	6	75	110	8.66	8.95
9	3	60	100	18.19	27.11
10	7	60	100	11.33	10.97
11	5	30	100	19.44	24.57
12	5	90	100	12.61	16.02
13	5	60	80	23.47	28.37
14	5	60	120	6.26	5.32
15	5	60	100	18.23	22.44
16	5	60	100	14.21	14.08
17	5	60	100	16.43	18.3
18	5	60	100	16.29	18.25

Fig. 10, shows the change in tenacity of PLA fibers as a function of pH and dyeing temperature at fixed dyeing time. Fig. 11, reveals the change in tenacity with the change in pH and time at fixed temperature. The observations depict that the PLA fibers, dyed at higher temperature, extended dyeing time and higher pH, exhibit lower tenacity. Similarly, the milder conditions of temperature, pH and smaller dyeing time results in dyed PLA fibers with higher tenacity. The optimized dyeing temperature, pH and dyeing time are found to be 80-100 °C, 4-5, and 30-45 min, respectively. The elongation% of dyed PLA fibers has been investigated as a function of dyeing time and pH. Figure 12 shows the effect of dyeing time and pH on elongation% of PLA fibers. The higher dyeing time and higher pH rendered lower elongation% and vice versa. From Figure 13 it is obvious that higher dyeing temperature and pH exhibited lower elongation% and vice versa. Literature reported that longer dyeing time and higher dyeing temperature can cause considerable strength loss and decrease in molecular weight [19]. Dyebath pH can also affect strength and elongation of PLA fibers. A research group recommended weakly acidic dyebath for dyeing PLA with best strength and elongation [20]. It was also claimed that more acidic pH (lower than 4) or more alkaline pH (pH higher than 7) cause significant loss in fiber elongation as well as strength [21].

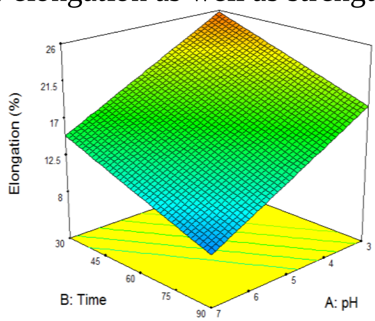

Fig. 12. Effects of dyeing, time, and pH on fiber elongation percentage (temperature 100 °C)

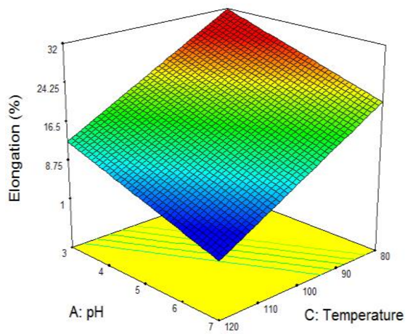

Fig. 13. Effects of dyeing, pH value, and temperature range on fiber elongation percentage (dyeing time 60 minutes)

D. Colorfastness properties of naturally dyed PLA fibers

The washing fastness properties of PLA fibers dyed along with turmeric dye were as satisfactory as the dyed samples showed fastness the rating up to 3-4. Besides, little staining was observed on polyester and cotton fibers of multifiber stripe. The washing fastness properties of PLA fibers dyed with turmeric dyes were satisfactory. Although, the colorfastness testing results were not conclusive and hence they were not added to this manuscript.

IV. Conclusion

The novel approach of dyeing PLA fiber using natural dyes extracted from turmeric has been explored along with optimization of dyeing parameters. Moreover, the obtained dye exhibited low solubility and higher hydrophobicity after exhibiting its suitability for PLA fibers. However, an increase in temperature and time duration of the dyeing process resulted in enhanced exhaustion percentages as well as better color strength values. Elevated dyeing temperature (>100 °C) and higher dyeing time duration resulted in reduced strength and lower elongation of PLA fibers. Also, pH value has significant effects on dye exhaustion and color strength. The optimized values for dyeing temperature, pH value, and dyeing time duration were found to be 100 °C, 5, and 30 minutes, respectively. Ultimately, all PLA fibers exhibit appreciable strength and elongation depending upon these parameters. Hence, the colorfastness results of dyed samples were also encouraging in showing better resistance to laundering process.

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Declaration of interest

The authors declare no potential conflict of interest.

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