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LOCAL RESOURCE EXPLOITATION

Optimization of the extraction of Henna (*Lawsonia inermis*) dye for cotton fabric dyeing

Optimisation de l'extraction du colorant du Henné (Lawsonia inermis) pour la teinture du tissu en coton

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ABSTRACT:

This study aimed at the optimization of the extraction process of Henna (*Lawsonia inermis*) dye for dyeing purposes. The three factors Box-Behnken design associated to the response methodology surfaces allowed to study the effect of the extraction solvent (% ethanol), the extraction time (minutes) and temperature (° C) on the extraction yield and the colourimetric characteristics (L*, a*, b*, C*, and H*). The experimental results of the responses were fitted to a quadratic polynomial model and these models were validated. To obtain the darkest as possible dyed fabric, the extract yield and C were maximized and L* minimized; and the multi-criteria analysis provided the optimum extraction conditions for 49.22 % ethanol, 61.35 °C and 18.37 minutes. The responses parameters of this optimum correspond to 24.64 % of extraction yield with the L*, a*, b*, C* and H* values of the dye respectively of 30.12, 26.64, 39.32, 47.90 and 55.87°. On the dyed fabric, the same colour parameters are respectively 51.81, -5.15, 17.24, 17.99 and 106.71°. The pH values of extract, between 3.86 and 4.30, shows that it is an acid-type dye. The models obtained can be used to define the extraction conditions corresponding to the desired colouration as long as the latter remains within the colour range of orange-red.

Keywords: Henna, Extraction, Design of experiments, Dye, Colour.

RÉSUMÉ :

Ce travail avait pour but d'optimiser le processus d'extraction du colorant du Henné (*Lawsonia inermis*) pour la teinture. Le plan de Box-Behnken à trois facteurs, associé à la méthodologie de surfaces de réponses, a permis d'étudier l'effet du solvant d'extraction (% éthanol), le temps d'extraction (minutes) et la température (° C) sur le rendement d'extraction et les caractéristiques colorimétriques (L*, a*, b*, C* et H*). Les résultats expérimentaux des réponses ont été ajustés à un modèle polynomial quadratique et ces modèles ont été validés. Pour obtenir un tissu teinté qui soit le plus foncé possible, le rendement d'extrait et C ont été maximises, et L* minimise. L'analyse multicritères a fourni les conditions d'extraction optimales pour 49,22 % d'éthanol, 61,35 °C et 18,37 minutes. Les paramètres-réponses de cet optimum correspondent à 24,64 % du rendement d'extraction avec les valeurs L*, a*, b*, C* et H* du colorant respectivement de 30,12, 26,64, 39,32, 47,90 et 55,87 °. Sur le tissu teint, les mêmes paramètres de couleur sont respectivement 51,81, -5,15, 17,24, 17,99 et 106,71 °. Les valeurs de pH de l'extrait, comprises entre 3,86 et 4,30, montrent qu'il s'agit d'un colorant de type acide. Les modèles obtenus permettent de définir les conditions d'extraction correspondant à la coloration souhaitée tant que celle-ci reste dans la gamme de couleur orange-rouge.

Mots clés : Henné, Extraction, Plan d'expérience, Colorant, Couleur.

1. INTRODUCTION

Dyes and pigments are substances which when applied to a substrate lead to selective reflection or transmission of incident daylight (Clarke and Anliker, 1980). In general, dyes consist of an assemblage of chromophore groups, auxochromes and conjugated aromatic structures (Chequer et al., 2013). Synthetic dyes have been cited as cause of dermatitis, cancer, respiratory issues, and other allergenicity (Angelini et al., 2003; Bechtold and Mussak, 2009). In addition to that, most of them are toxic and difficult to degrade (Lu et al., 2009). On the other hand, natural dyes are seen as safe since nontoxic, causing no allergy and non-harmful to the environment because they are biodegradable. Natural dyestuffs are colouring substances extracted or derived from nature, like plants, animals, minerals and microbial sources, and used for colouration of a variety of textile materials (Hofmann-De Keijzer and Heiss, 2019; Saxena and Raja, 2014) food and cosmetics. Many of these plants are known for their medicinal properties, like antibacterial activity (Mirjalili et al., 2011; Nagia and EL-Mohamedy, 2007; Prusty et al., 2010; Singh et al., 2005; Vankar and Shanker, 2009). Therefore the use of natural dye is highly promoted recently among traditional craftsmen, in the fields of textile conservation, paints and pigments production, cosmetic, and food industries (Chaudhary et al., 2010; Samanta and Konar, 2011; Qadariyah et al., 2019). Those plants, from which dyes can be extracted from some parts, are called tinctorial plants. Their extracts are commonly used to dye natural textile fibres (wool, cotton, silk) but can also be used as food or body dyes. Henna *(Lawsonia inermis*), from the *Lythraceae* family, is one of the most used tinctorial plants. The colouring molecule in Henna leaves is called Lawsone (2-hydroxy naphthoquinone) responsible for its orange-red colouration (Saxena and Raja, 2014). Extraction of particular compounds from Henna was previously done for multiple purposes, using different methods (Elaguel et al., 2019; Tan et al., 2013; Zohourian et al., 2012). Some authors worked specially to extract Henna dye for dyeing textile materials (Alam et al., 2007; Fatima et al., 2016; Qadariyah et al., 2019; Yusuf et al., 2015). Willing to defined extraction conditions that are easy to implement, inexpensive and harmless to both humans and the environment, we opted for a batch extraction with cheap and eco-friendly solvents: Ethanol and water are both. Ethanol-Water mixtures were used as extraction solvent because of the low water solubility of Henna dye (Fatima et al., 2016; Hasan et al., 2015; Saxena and Raja, 2014). Ethanolic and enzymatic extraction have shown similar extraction efficiency of Henna pigment (Sivarajasekar et al., 2018). Similarly, ethanol allow a higher colour strength compare to methanol and acetone (Hasan et al., 2015). To the best of our knowledge, batch solvent extraction, which is the easiest method to implement in a context of small investment, was not yet optimized, Therefore, the present study aimed to optimize conditions of this method for extracting natural dye from Henna leaves powder, using the experimental design approach, and observe the effects of the extraction conditions directly on a dyed cotton fabric.

2. MATERIAL AND METHODS

2.1. Material

Henna (*Lawsonia inermis*) leaves powder from Marza (at latitude 7.28 and longitude 13.58), was purchased at a market in the town of Ngaoundere-Cameroon and stored in opaque vials at 25 °C.

2.2. METHODS

2.2.1. Extraction process.

Figure 1 presents the extraction and the dyeing process. Henna powder is mixed with the extraction solvent, which is ethanol, at the ratio 1:10 (weight : volume) and stirred from time to time for few seconds in thermostaticallycontrolled water bath. The extraction parameters studied are the extraction temperature in Celsius (X_1), the percentage of ethanol (X_2), and the extraction time in minutes (X_3).

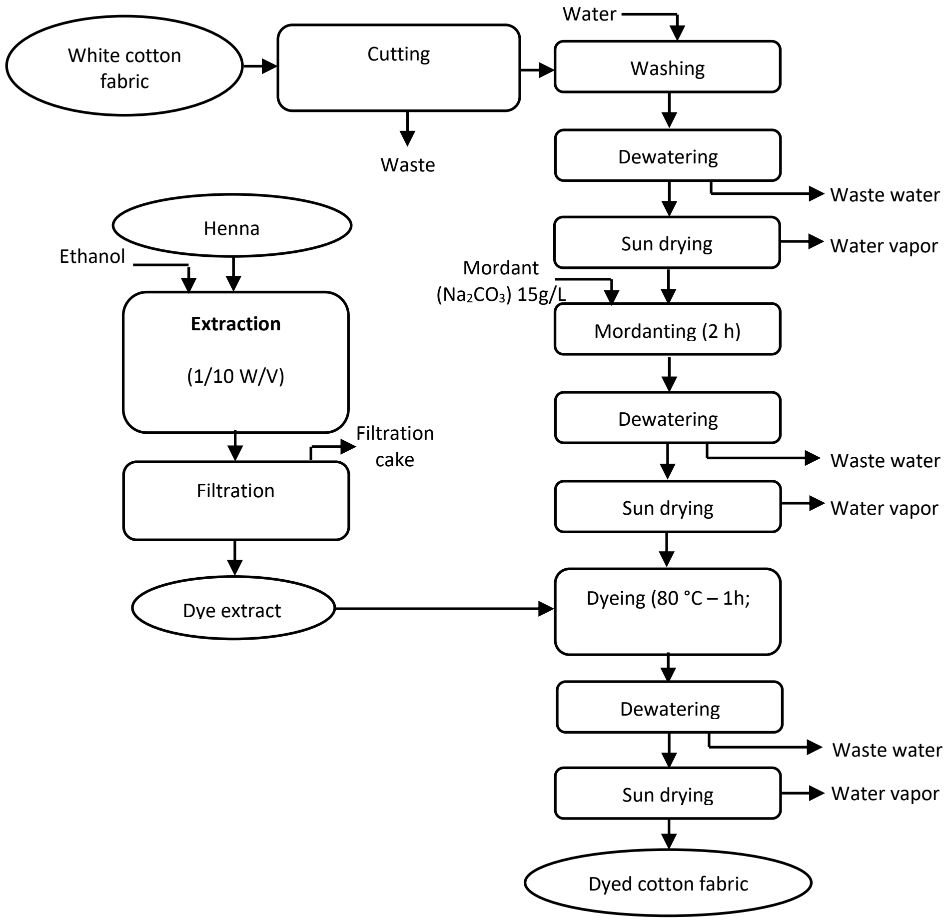

Figure 1. Dyeing process from Henna extract

The combination of these extraction conditions is defined by the Box-Behnken design (Table 1). The obtained extract is filtered and stored in opaque vials and dark place at 25 °C. Every assay has been done in triplicates.

Pieces of white cotton fabric (6 cm x 3 cm = 0.42 $\pm$ 0.03 g) have been prepared, washed with distilled water, and sun dried. The dried fabric pieces were soaked for 2 hours in a 3 % disodium carbonate solution (Na_2CO_3) used as mordant for improving dye fixation. Afterwards, they were wrung out and sun dried again. The Henna extract was heated at 80°C, and the pre-treated fabric pieces were soaked in for 1 hour. The fabrics were then wrung out and sun dried.

2.2.2. Analytical methods.

The extraction yield is evaluated by the dry soluble extract. pH of Henna extract is also evaluated with a pH-meter (Hanna instruments, HI 8033). Colour measurements of the extract and dried dyed cotton fabrics have been carried out using pictures with ImageJ software for assessing colour characteristics in terms of CIE Lab system. On each sample, the parameters L*, a* and b* of the sample are determined and the values of C* and H* are calculated according to equations 1 and 2 (Recueil des methodes internationales d'analyses, 2006). The luminance or clarity, L*, varies from black (0) to white (100). a* and b* express the difference in colour from that of a grey surface of the same clarity. They indicate the chromaticity coordinates, which is trend of the colour: a* grows from green (<0) to red (>0), and b* from blue (<0) to yellow (>0).

$$C^* = \sqrt{a^{*2} + b^{*2}} \quad (1)$$

$$H^* = arctan(a^*/b^*) \quad (2)$$

C* is the saturation of the colour. It defines the purity of a colour. It varies from bright to dull colours. H* is the hue of the colour and can be represented as the wheel of colour, with purple red at 0°, yellow at 90°, green-bluish at 180°, and blue at 270°.

2.2.3. Statistical analysis, modelling and optimization.

Box-Behnken design was commonly used to optimize extraction process. It is among the most efficient experimental designs, giving optimal conditions in a minimum number of experimental runs and the easiest to set up (Ferreira et al., 2007). The response surface methodology combined to the 3 factors Box-Behnken allows to model the responses studied in the form of a second-degree polynomial equation as presented below:

$$R = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_i \sum_j \beta_{ij} X_i X_j \quad (3)$$

R is the response; k the number of factors; X_i the factors. And β_i, β_{ii} β_{ij} are respectively the linear, quadratic factors and interactions coefficients. All data obtained were analysed with Minitab 18 software (Minitab, Ltd., Brandon Court, UK), to obtain the coefficients for the models, their significance using ANOVA and optimum conditions. The optimum was obtained by seeking the darkest colour as possible for dyed fabrics. The fitting quality was assessed through the determination coefficient of (R^2) and the adjusted determination coefficient (R^2 adj). In addition, the absolute average deviation (AAD), the Biais (B_f) and the accuracy (A_f) factors were used to validate the statistical models.

$$R_{aj}^2 = \sum_{i=1}^{n} \frac{(Y_{cal,i} - \bar{Y}_{obs})^2}{(Y_{obs,i} - \bar{Y}_{obs})^2} \quad (4)$$

$$AAD = \frac{\sum_{i=1}^{n} \frac{|Y_{obs,i} - Y_{cal,i}|}{Y_{obs,i}}}{n} \quad (5)$$

$$B_f = 10^{\left(\frac{1}{n}\sum_{i=1}^{n} \log\left(\frac{Y_{cal,i}}{Y_{obs,i}}\right)\right)} \quad (6)$$

$$A_f = 10^{\left(\frac{1}{n}\sum_{i=1}^{n} \left|\log\left(\frac{Y_{cal,i}}{Y_{obs,i}}\right)\right|\right)} \quad (7)$$

With: Y_{obs} = observed response; Y_{cal} = calculated response; "i, p" represent the rank of the experiments and "n" the total number of experiments. A valid model should have R^2 adj > 80 % (Joglekar and May, 1987), 0 < AAD < 0.3 (Baş and Boyaci, 2007) and 0.75 < B_f, A_f < 1.25 (Dalgaard and Vigel Jørgensen, 1998).

3. RESULTS AND DISCUSSION

3.1. Effect of extraction parameters on the Extraction yield

Table 1 presents all the experimental results obtained in different extraction conditions. It shows yield values between 15.73 and 25.29 %. Qadariyah et al. (2019) found a extraction yield of Henna dye in this range.

Table 1. Experimental values of responses parameters of Henna dye extract and dyed cotton fabric

X_1: temperature (30 - 70° C), X_2: solvent concentration (0 – 80 % ethanol), X_3: time (5 – 45 min) of extraction.

	Factors						**Responses**											
	Coded values			**Uncoded values**			**Henna dye extract**							**Dyed cotton fabric**				
Runs	**X_1**	**X_2**	**X_3**	**X_1**	**X_2**	**X_3**	**R_d**	**pH**	**L***	**a***	**b***	**C**	**H**	**L***	**a***	**b***	**C**	**H**
1	-1	-1	0	30	0	25	19.46	3.86	23.81	23.94	29.48	37.98	50.86	64.54	-7.40	14.32	16.12	117.11
2	0	-1	-1	50	0	5	15.73	4.20	20.62	20.55	26.36	33.45	51.99	62.44	-5.94	13.86	15.09	115.26
3	0	0	0	50	40	25	23.79	4.07	31.71	25.20	40.37	47.67	58.16	55.31	-5.66	16.92	17.84	108.3
4	1	0	1	70	40	45	25.16	4.23	33.76	25.02	41.80	49.22	58.60	51.82	-2.38	16.82	16.99	99.91
5	0	-1	1	50	0	45	17.39	4.14	23.05	24.00	30.81	39.08	51.95	61.82	-5.66	14.84	15.88	110.60
6	0	1	1	50	80	45	21.50	4.28	34.63	18.70	43.40	47.28	66.86	61.33	-5.82	17.07	18.03	106.78
7	1	-1	0	70	0	25	21.53	4.00	15.37	17.56	17.36	24.79	43.23	52.96	-5.24	10.23	11.49	115.54
8	-1	0	-1	30	40	5	22.42	4.30	35.88	37.59	45.07	58.75	50.17	62.11	-6.27	17.13	18.25	108.26
9	-1	0	1	30	40	45	25.29	4.06	30.07	26.75	38.32	46.99	54.93	62.16	-5.99	17.57	18.57	109.33
10	0	0	0	50	40	25	23.79	4.09	28.13	29.88	37.60	48.03	51.40	54.41	-5.57	17.33	18.20	108.31
11	-1	1	0	30	80	25	22.02	4.28	41.13	12.99	48.61	50.50	74.55	65.51	-7.75	17.54	19.18	115.40
12	1	0	-1	70	40	5	24.36	4.25	27.95	26.04	36.80	45.14	55.11	51.81	-4.96	16.11	16.86	106.61
13	0	0	0	50	40	25	24.71	4.25	32.38	34.09	42.52	54.50	51.28	54.01	-5.64	16.72	17.64	108.31
14	0	1	-1	50	80	5	20.61	3.99	38.36	15.64	46.73	49.35	71.59	61.90	-6.06	19.25	20.18	107.75
15	1	1	0	70	80	25	22.42	4.08	29.25	15.96	37.51	40.77	67.01	52.62	-5.27	18.73	19.46	105.92

Table 2 presents the coefficient values of validated polynomial model of extraction yield. The quadratic effect temperature (X_1^2) has a significant impact on the extraction yield. The negative coefficient of linear effect of temperature means the extraction yield reduce with the temperature increase, but not significantly. But above a certain temperature, the quadratic effect of temperature promotes the augmentation of the extraction yield. This is due to the increase in solubility and diffusion coefficients. An increasing temperature allows a decrease in the solvent viscosity which improves the solvent penetration into the plant matrix, and thus improved mass transfer (Al-Farsi and Lee, 2008; Hemwimon et al., 2007; Silva et al., 2007; Wang et al., 2008).

Active compounds from plants are organic compounds efficiently extracted from them with ethanol or methanol (Zhang and Lewis, 1997). Therefore, Henna dye was extracted with ethanol-water mixture which are usually used to extract polar molecules and is non-toxic. The ethanol concentration (X_2) and its quadratic expression (X_2^2) have a significant effect (Table 2) on the extraction yield. The positive coefficient of the direct effect of ethanol concentration implies that it will increase the extraction yield. In the same way, the negative coefficient of the quadratic component implies that, high concentration of ethanol will reduce the extraction yield. Ethanol is less polar than water which is more polar, and they can be mixed with each other in any proportion. Therefore, with the addition of ethanol to water, the polarity of the complex solvent will decrease continuously and the medium is more and more favourable to the extraction, reason why the yield increase. But, above a certain concentration, further decrease in polarity of the extraction solvent is detrimental to the extraction yield (Chandrasekhar et al., 2012; Qadariyah et al., 2019; Zhang et al., 2007).

3.2. Effect of the extraction conditions on pH of the extract

For all experimental runs, pH values of extract are ranged from 3.86 to 4.30 (Table 1). The pH of ethanol solution, the extraction solvent used, is neutral (7 ± 0.01). So, when extraction is set up, the mixture becomes acid, indicating that the Henna dye is an acid-type dye. The pH model presented on Table 2, will not be ever used for optimization because it does not fit with polynomial model because of his very low R^2 value.

3.3. Effect of the extraction conditions on the colour properties of the extract and the dyed cotton fabrics

Experimental results of colour characteristics are presented on Table 1. The luminance values (L*) are all relatively low (<50). Thus, Henna extract can be described as dark. The opposite observation can be made according to the values obtained from the dyed fabrics, since their L* values are all above 50. For both extract and dyed fabrics, the b* are all positive while a* are all positive just for the extract. The reason can be fixation of green pigments extracted from the leaves. In both cases, regardless the extraction conditions, b* values are all above a* values. With H* values between 50.86°-66.86°, all the extract presented a Hue between orange and the yellow. On the other hand, the dyed fabric presents a yellow green colouration (97.99°-117.11°). From the observation of these parameters, regardless the extraction conditions, the Henna dye extract can be qualified as dark reddish-yellow, which is a colour close to that of the Lawsone, the dyeing molecule of Henna leaves (Saxena et Raja, 2014). The fabrics end up with a lighter yellow colour, also due to the fixation of Lawsone and some green pigments. These trends could also be explained by the fact that dye in the extract is not totally fixed on the fabric pieces showing an overview of dye fixation degree.

Table 2. Coefficients values and validation parameters of the models

		Henna extract							Dyed cotton fabric				
		Extraction Yield (%)	**pH**	**L***	**a***	**b***	**C**	**H°**	**L***	**a***	**b***	**C**	**H°**
Constant		20.0113	4.1260	33.4359	33.0377	34.9759	48.3174	48.3729	75.0992	-6.4047	14.5612	16.5902	113.6320
Linear factors	**X_1**	-0.2651	0.0042	**-0.2171***	-0.0017	**0.0349***	**-0.0274***	0.0522	**-0.1941***	**-0.0025***	0.0422	**0.0156***	**-0.0574***
	X_2	**0.2770***	0.0074	**0.4435***	0.3168	**0.5355***	**0.6528***	**0.0116***	-0.2119	0.0373	**0.0636***	**0.0459***	**-0.2074***
	X_3	0.2594	-0.0221	-0.4786	-0.4127	-0.5188	-0.6581	-0.0533	-0.3416	**-0.1295***	-0.0297	0.0053	**0.4809***
Quadratic factors	**X_1^2**	**0.0035***	0.0000	-0.0008	-0.0037	-0.0041	-0.0048	-0.0007	-0.0007	0.0002	-0.0014	-0.0012	**0.0014***
	X_2^2	**-0.0026***	0.0000	-0.0019	**-0.0066***	**-0.0033***	**-0.0060***	0.0035	**0.0029***	**-0.0006***	**-0.0008***	-0.0005	**0.0029***
	X_3^2	-0.0029	0.0002	0.0037	0.0015	0.0049	0.0047	0.0035	0.0067*	**0.0016***	0.0012	0.0006	**-0.0071***
Quadratic interactions	**X_1*X_2**	-0.0005	-0.0001	-0.0011	0.0029	0.0003	0.0011	0.0000	-0.0004	0.0001	**0.0017***	**0.0015***	**-0.0025***
	X_1*X_3	-0.0013	0.0001	0.0073	0.0061	0.0073	0.0099	-0.0008	0.0000	**0.0014***	0.0002	-0.0001	**-0.0049***
	X_2*X_3	-0.0002	0.0001	-0.0019	**-0.0001***	-0.0024	-0.0024	-0.0015	0.0000	0.0000	-0.0010	-0.0009	**0.0012***
Validation parameters	**R^2**	95.80	60.54	93.50	85.88	93.89	93.66	87.13	99.09	96.45	95.50	95.87	100.00
	R^2 aj	88.25	0.00	81.79	60.47	82.88	82.24	63.97	97.46	90.05	87.39	88.44	99.99
	AADM	0.02	-	0.05	0.10	0.05	0.04	0.05	0.01	0.04	0.03	0.02	0.01
	B_f	1.00	-	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
	A_f	1.02	-	1.05	1.10	1.05	1.04	1.05	1.01	1.04	1.03	1.02	1.01
Specifications		Maximize	-	-	-	-	-	-	Minimize	-	-	Maximize	-
Optimum values	**Theorical**	24.64	-	-	-	-	-	-	51.81	-5.15	17.24	17.99	106.71
	Real	23,62	-	-	-	-	-	-	56.71	-7.16	13.12	14.94	118.63

*: significant effect ($p<0.05$)

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All models for colour characteristics of dyed fabrics were better fitted with polynomial model than Henna dye. Both of them are validated regardless the values of validation parameters presented in Table 2. The temperature (X_1) and the concentration of ethanol (X_2) have both a significant effect on the extract L*. For the dyed fabrics, temperature (X_1) and the quadratic effect of the concentration of ethanol (X_2^2). In both cases, the temperature increase tends to a lower L*, while the ethanol concentration has the opposite effect. The a* value of the extract is significantly impacted by the quadratic effect of the concentration of ethanol (X_2^2), and the interaction ethanol concentration-extraction time (X_2*X_3). Both of them tending to reduce its value, which means, making the extract greener, extracting more green pigments from the leaves. On the fabrics, the temperature (X_1), the quadratic effect of the concentration of ethanol (X_2^2), and the extraction time (X_3) have a significant negative impact on the a* value. But the quadratic effect of time (X_3^2) and the interaction temperature-time (X_1*X_3) are significant and tends to increase the yellow compounds. The temperature (X_1), the concentration of ethanol (X_2) and its quadratic (X_2^2) have a significant impact on the b* value of the extract. Both temperature and ethanol concentration tend to make the extract more yellowish, while an excess of ethanol tends to make the extract blue. On the fabric, the concentration of ethanol (X_2) and its quadratic effect (X_2^2) have a significant impact on the b* value, along with the interaction temperature-ethanol concentration (X_1*X_2). Among the three, on the excess of ethanol (X_2^2) leads to a bluer colour on the fabric. The only factor affecting significantly the Hue of the extract is the ethanol concentration (X_2), and it presents a positive effect. On the fabric, all the factors and interactions are significant. The extraction time (X_3), the quadratic effects of temperature (X_1^2) and ethanol concentration (X_2^2), and the interaction ethanol concentration-extraction time (X_2*X_3) have a positive effect on the Hue of the fabric.

With a maximum value of 58.75 %, the saturation (C*) of the extract is not very high. This can be explained by the non-specificity of the extraction solvent, which extract all soluble molecules. That observation is much more visible on the fabric. It can be explained but the low percentage of retention of the coloured molecules on the fabric. The ethanol concentration (X_2) has a significant positive effect on the saturation, while its quadratic effect (X_2^2) and the temperature (X_1) display a significant negative effect. On the fabric, all the significant effects (X_1, X_2, and X_1*X_2) display a significant and positive impact on the saturation. These can be linked to the effect of the time and the temperature which significantly and positively impact the extraction yield. The more coloured molecules are extracted, more can be fixed on the fabric.

3.3. Optimization of extraction process conditions

In this study, the aim of the optimization is to get the darkest colour as possible for dyed fabric with high extract yield. Luminance L* and saturation C* express better this aim than others parameters which are focused either on one specific colour (a*, b*) or multiple possible angle colour (H*). So models were used and specifications were to maximize the extraction yield and C* to have a saturated colour, while L* was minimized to have a colour with less clarity. Following this, the optimum extraction conditions obtained were 49.22 % of ethanol solution, 61.35 °C and 18.37 minutes. The real values of the response parameters determined by carrying out the extraction under optimal conditions are close to the theoretical values obtained from the models. This proves the repeatability of the extraction process and the validated models can indeed be used for different purposes as long as one remains within the values range of studied factors.

4. CONCLUSION

The aim of this work was to optimize the extraction of henna *(Lawsonia inermis)* dye with observation of its direct effect on a dyed fabric. The modelling allows to observe that the interactions between the studied factors are not very significant on the followed responses, in their respective studied ranges. Optimal extraction conditions (49.22 % ethanol, 61.35 °C, 18.37 minutes) can easily be implemented at this level with little resources. The dye could be extracted and used directly without storage and extraction time helps in that way. Degradation would be important if product has to be stored. Reuse of waste extract after dyeing could be explored because the dye is not totally adsorbed on fabric in one soaking. Despite extracting the coloured molecules, the extraction is not really specific. Also, the addition of a mechanical assistance as controlled-stirring, or ultrasound can improve the extraction.

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6. CONFLICT-OF-INTEREST DECLARATION

All authors have no conflicts of interest to disclose.

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