Natural dyes in the design of textile: how to make them more competitive face to synthetic dyes

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In recent years, much attention has been focused on research to replace synthetic dyes, in a competitive manner, with natural dyes. In this work the dyeing of cotton fabric by using natural dyestuffs extracted from hibiscus, annatto, turmeric, coffee and acai berry has been studied in both magnetic stirring and ultrasonic extractions. Unlike the current literature results, the comparative values of extraction yield were found to be higher using magnetic stirring than ultrasound. Extraction medium and temperature seem to be predominant parameters on the extraction yield and dyeing performance. From the preliminary results obtained in this study, it might be suggested that, in general, it is possible to improve the performance of natural dyes on cotton fabrics when the extraction is carried out in alkaline medium at temperatures above 40 °C. An illustrative chart of the studied natural dyestuffs was assembled.

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Introduction

Nowadays being fashionable means to be worried about environmental issues and sustainable development [1]. In the current world in which sustainability and concern about the industrial residues are emerging as a concept, there is a tendency toward a cleaner industry with better use of energy replacing whenever possible synthetic chemicals for raw materials derived from renewable resources which should be used in a rational manner so that they can be preserved in nature. As pointed out by Siqueira [2], in Brazil the concern with the forests dates from the early nineteenth century. Exploration along with preservation was the policy of Portuguese Crown reported at the eleven articles of the Brazil wood regiment. It was a position of vanguard regarding the worry about deforestation, although the real intention was to safeguard the raw material for further exploitation [2].

Green Chemistry is the design, development, and implementation of chemical products and processes to reduce or eliminate the use and generation of substances hazardous to human health and the environment [3]. According to this concept, unlike regulatory requirements for pollution prevention, Green Chemistry is an innovative, non-regulatory, economically driven approach toward sustainability.

Textile processing industry is one of the major environmental polluters. In order to process a ton of textile about 230-270 tons of water is spent by generating an effluent proportional to this quantity [4]. According to these date there are two main ways to limit the environmental impact of textile processing. One is to construct sufficiently large and highly effective effluent treatment plants, and the other way is to make use of dyes and chemicals that are environment friendly.

Natural dyes appear to be the most appropriate substitute to the relatively toxic synthetic dyes. They are believed to be safe because of non-toxic, non-carcinogenic and biodegradable nature. Further, natural dyes do not cause pollution and waste water problems [5].

Natural dyes are not an innovation but a revival of a rich and prudent tradition and cannot be compared with synthetic dyes only in terms of efficiency for industrial applications. However, we need to ensure that the replacement of synthetic dyes by natural dyes will not just transform the industrial fabric into handcrafted fabric, but lead to a more environmentally friendly fabric with the same quality standards previously achieved with synthetic dyes [6].

There is an increasing demand for developing appropriate techniques for a more efficient and effective extraction of active substances from the plants and minerals in order to enable the use large scale of natural dyes [7]. From the point of view of some authors [8] for the reintroduction of natural dyes in industrial scale, the dyeing process with natural dye should be possible to be performed on the equipment available in modern textile industry without requiring large investments.

In this context, it was the aim of this work to perform a small review of new approaches for extraction of natural dyes. Some of the approaches that have been reported were tested for the extraction of natural dyes from plant raw materials easily found in Brazil.

Strategies for the extraction of natural dyes

According to Bechtold *et al*. [9], the predominance of synthetic dyes in the last decades harmed the development and adaptation of natural dyeing to the needs of modern textile industries. In their view, there is now a considerable gap separating the knowledge we have about efficient processes for the extraction of natural dyes from the demands of commercial dyeing processes. These authors concentrated their study on plant sources available in the moderate Austrian climate. The possibility for agricultural production of the plant materials, the feasibility of the extraction with water, the exclusion of chemicals or non-aqueous solvents for improvement of the dyestuff extraction among others were adopted as criteria for the selection of raw materials. The results presented in the research indicated that a general one-bath dyeing process can be established for various natural dyes and acceptable fastness properties can be achieved both on wool and on linen as substrates.

Usually it is established that the efficiency of extraction with water of vegetable dyes is low, thus affecting the production costs and restricting the use of natural dyes in comparison with synthetic dyes. With the aim of achieving the optimum conditions for colouring cotton textiles a study of the alkaline extraction of dye from henna leaves was performed [5]. Indeed the alkaline extracts were found to have much higher colour strength than that obtained in distilled water. The washing fastness, rubbing fastness and light fastness properties on cotton, when applied without any mordant, were found to be moderate to good.

Hou *et al.* [10] extracted dye from orange peel (OP) at different temperatures (room temperature, 70, 80, 90 and 100 °C) for different time periods (0.5, 1, 1.5, 2, 2.5 and 3 h) to optimise the extraction conditions. The authors found that dyeing temperature of 100 0C and dyeing time of 120 min were appropriate for direct and mordant dyeing methods. Moreover dyeing experiments showed that the colour of wool fabric dyed with water extracts from OP was deeper than that dyed with ethanol extracts.

A study for concerning the development of a technique for efficient extraction of natural dyes was conducted by Sivakumar *et al.* [7]. The authors compared the extraction of natural dye using ultrasound with the conventional process using magnetic stirring.

A magnetic stirrer is a device that employs a rotating magnetic field to cause a stir bar immersed in a liquid to spin very quickly, thus stirring it. Since glass does not affect a magnetic field appreciably (it is transparent to magnetism), magnetic stir bars work well in glass vessels.

On the other hand, when a liquid is irradiated by ultrasound, micro bubbles appear, grow and oscillate extremely quickly and even collapse violently if the acoustic pressure is high enough. The occurrence of these collapses near a solid surface will generate micro jets and shock waves which promote the extraction.

In this specific study, they used beetroot as a natural dye resource. According to their explanation, extraction of colouring matter from beetroot is a solid-liquid leaching process involving mass transfer problem. Since the colouring matter is strongly bound with plant cell membranes, extraction could be more effective with the aid of ultrasound. Significant 8% of enhancement in the yield of colorant has been achieved with ultrasound when compared to magnetic stirring process using 1:1 ethanol-water.

The aforementioned authors have continued the studies in extraction of natural dyes with the use of ultrasound and explored other natural sources of raw material [11]. Green wattle bark, Marigold flowers, Pomegranate rinds, four o'clock flowers and Cocks comb flowers were used. The results indicated that there is about 12–100% of improvement in the yield of the obtained extract due to the use of ultrasound when compared to magnetic stirring at 45 °C. As reported by the authors, they were aware that they could achieve higher extraction efficiency with organic solvents such as n-hexane. However, their objective was to develop a sustainable and effective process by using aqueous system.

According to Leitner *et al.* [8], in order to achieve a feasible strategy for dyeing with natural dyes some aspects should be considered. The following considerations were pointed out by these authors:

- The dyestuff content in the plant material is low, so large amounts of solvent are needed. Thus the extraction has to be performed by using only water.
- Huge amounts of plant material have to be handled during harvesting, storage and extraction.
- Standardisation of the dyestuff is required to minimise variability in the dyeing result due to quality differences of plant material.
- The application of natural colorants must be possible on the technical equipment available in modern textile dye house, without big additional investment.

According to the authors a strategy to overcome many of the technical barriers is to produce natural dyes in solid form by precipitation of the dyes from plant extracts. In this context they reported a model study to formulate a solid dyestuff product from aqueous extracts of dried Canadian Goldenrod plant material (Solidago canadensis).

The production of a concentrated dyestuff containing product was achieved by using simple stages including extraction, precipitation, sedimentation, filtration and drying. According to them, this process required only basic equipment and could be installed at the site of farming and harvesting.

In another investigation the precipitation of natural colorants extracted from plants by using aluminium salt instead of iron salt has been reported [12]. The formation of aluminium based dye lakes has been pointed out as more beneficial and was studied with three different plant sources, namely Canadian Onion (Allium Cepa), Canadian Goldenrod (Solidago Canadensis) and Pommegranate (Punica Granatum). As reported in this study, after dissolution of the aluminium lake in diluted acid this solution can be used for textile dyeing in procedures similar to the application of synthetic dyes.

Islam *et al.* [13] studied the effect of ammonia post-treatment on Wool yarns dyed with extract from Annatto seeds (Bixa orellana). For this purpose the woollen yarn samples dyed with annatto were treated with various ammonia solutions (1, 3, and 5% w/w) at 25 °C for 10 min. It was established that the treatment with ammonia solution results in a variety of sober and elegant shades with variation in hue and tone. They found that the highest colour depth (K/S) on woollen yarn was obtained by using 5% (w/v) ammonia concentration.

Materials and methods

Materials

The extracts were produced using parts of some plants easily found in the Brazilian flora as shown in Table 1. The photographs of these materials are shown in Figure 1 (a-f), respectively.

Other reagents used were ethanol, sodium hydroxide, sodium bicarbonate and urea from Synth-Brazil. Tannic acid was purchased from Sigma-Aldrich. All reagents were used as received without any procedure to increase the purity or remove moisture.

	Common name	Botanical name	Part used
a	Hibiscus	Hibiscus rosa-sinensis	Flowers
b	Onion	Allium cepa	Bark
c	Annatto	Bixa orellana	Seeds
d	Coffee	Coffea arabica	Dry seed powder
e	Turmeric	Curcuma longa	Root
f	Acai berry	Euterpe oleracea	Pulp and/or epicarp

Table 1: Vegetable raw materials used in the present study

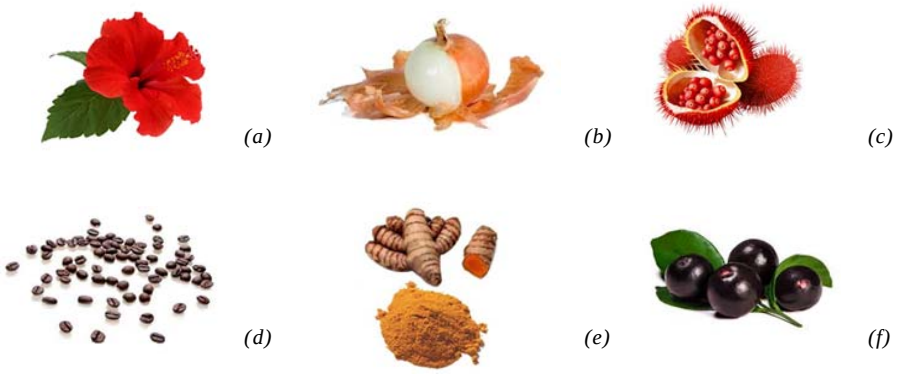

Figure 1: Natural dye yielding plants used in the present study (also see Table 1) – (a) Hibiscus, (b) Onion, (c) Annatto, (d) Coffee, (e) Turmeric and (f) Acai berry.

We understand the word harmony, as used in the context of colour and art, to broadly refer to 'good contrast'. Different to the preceding work, our aim is to define global hue contrast and on its own this does not cast any 'judgment' on the quality of contrast in an artwork. We regard the evaluation of colour harmony to be a low-level addressal of the colour in an artwork, providing detail specific to that artwork (e.g. 'this artwork employs an analogous harmony whereas that artwork employs a split complementary harmony'). Differently, our approach provides a high level and general definition of the hue organisation in an artwork wherein many artworks may be evaluated 'on a level playing field'.

Extraction using magnetic stirring (MS)

Typically 1 g of sample was taken and 40 mL of the extraction medium was added in a glass beaker. The extraction was performed with a magnetic stirrer Technal TE 0851 for 1 hour at a temperature of 40 °C.

Extraction using ultrasound (US)

The extraction using ultrasound was performed with a Branson Sonifier W 450 Digital with 50% amplitude in the pulsed mode, being on for 5 minutes and off for another 5 minutes until to complete 15 minutes of sonification.

Typically 1 g of sample was taken and 40 mL of the extraction medium was added in a glass beaker. The beaker was covered using aluminum foil to prevent loss of solvent by evaporation and an ice bath was used to avoid overheating of the extract.

The media used for extraction were water, aqueous sodium hydroxide (1 M), aqueous sodium bicarbonate (1 M) and aqueous urea solution (1 M).

At the end of the extraction process, the samples taken from both magnetic stirring and ultrasound were filtered under vacuum on filter papers pre-weighed. The filter papers with the vegetable residue were dried in an oven for 24 hours at 60 ^{0}C and weighed again to calculate the amount of residue left on the filter paper. The filtrates (aqueous extracts) were kept in clean and closed flasks and were used to dye the cotton fabric samples.

Dyeing experiments

Small samples (3 cm × 3 cm) of 100% cotton fabric washed and bleached (Horizonte Têxtil Ltda, Brazil) were immersed in a beaker containing the dyestuff extract for 20 minutes at room temperature. After this time the samples were placed in an oven for 24 hours at 60 °C for drying.

All dyeing were carried out without the presence of mordant, except in the case of the extraction of annatto dyestuff performed in water with magnetic stirrer. In this case, samples of cotton fabric were first immersed in mordant aqueous solution for 20 min at room temperature (pre-mordant method). After drying for 24 h at room temperature, samples were dyed with the same procedure described above. The mordants used were tannic acid (TA) and a resin based on itaconic acid which was synthesised in the laboratory, hereinafter named ITA.

Gravimetric analysis

The yield of the colorant extracted per gram of the plant material used was calculated using the Equation 1.

$$\%\ yield\ of\ natural\ dye = \frac{mass\ of\ extracted\ dye\ (g)}{mass\ of\ vegetable\ raw\ material\ used\ (g)} x\ 100 \quad (1)$$

The mass of extracted dye was calculated by subtracting the mass of vegetable raw material used (about 1 g) of the mass in grams of residue left on the filter paper after vacuum filtration.

The comparison between the extraction with ultrasound (US) and magnetic stirring (MS) was estimated using the Equation 2.

$$Ratio = \frac{\%\ yield\ of\ dye\ with\ ultrasound\ (US)}{\%\ yield\ of\ dye\ with\ magnetic\ stirring\ (MS)} \quad (2)$$

Colour measurements

The colour of specimens was measured by using a portable colour measurement instrument (Instrutherm, model ACR-1023). After calibrating the instrument, specimens were assayed by recording the resulting RGB and coordinates. According to this theory, the human eye perceives colour through stimulation of three visual pigments in the cones of the retina. According to this theory a colour in the RGB colour model can be described by indicating the amount of red, green and blue it contains. Each can vary between the minimum (completely dark) and maximum (quite intense). When all colours are at a minimum the result is black. If they are all at a maximum, the result is white. One of the most common representations for colours is the utilisation of the range between 0 and 255. Thus the colours can be represented as in Table 2.

	White	Blue	Red	Green	Yellow	Magenta	Cyan	Black
[RGB]	[255 255 255]	[0 0 255]	[255 0 0]	[0 255 0]	[255 255 0]	[255 0 255]	[0 255 255]	[0 0 0]

Table 2: RGB values.

Optical microscopy (OM)

Dyed fabric samples were evaluated for visual appearance using Leica DM 2500M (Reflected-light microscopy).

Wash fastness test

After the dyeing process, the fabric samples were submitted to a test for fastness wash reported elsewhere [14]. In such test the samples were stitched to “testimonies fabrics”, forming a “sandwich”, and immersed into 50 mL of a bath containing 5 g L^{-1} of detergent (sodium lauryl sulfate) and 2 g L^{-1} of commercial sodium carbonate, during 45 minutes at 60 °C. Then samples were removed without rinse and were placed in an oven for 2 hours at 60 °C for drying.

The numerical results were evaluated both for transfer of the colour (fastness) and for the colour change (degradation), as standardised scales, as shown in Tables 3 and 4 respectively.

Index	Signification
5	Negligible or does not transfer
4	Transfers slightly
3	Transfers a little
2	Transfers substantially
1	Transfers a lot

Table 3: Degree of colour transfer [14].

Index	Signification
5	Negligible or does not change
4	Changes slightly
3	Changes a little
2	Changes substantially
1	Changes a lot

Table 4: Degree of colour change [14].

Results and discussion

Influence of extraction media

The influence of the extraction media on the final result of the dyeing when magnetic stirring was used (MS), can be observed through the photographs shown in Figure 2. Table 5 shows the standard hexadecimal notation of the corresponding RGB values printed in Figure 2.

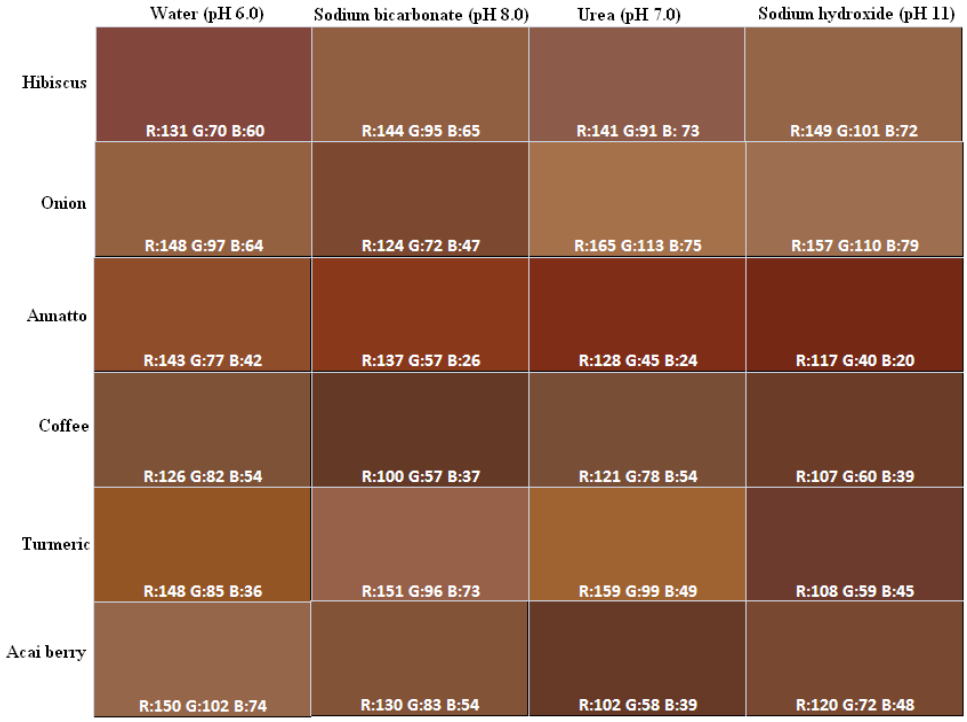

Figure 2: The influence of the extraction medium on the dyeing of cotton (MS).

	Water	Sodium bicarbonate	Urea	Sodium hydroxide
Hibiscus	#83463C	#905F41	#8D5B49	#956548
Onion	#946140	#7C482F	#A5714B	#9D6E4F
Annatto	#8F4D2A	#89391A	#802D18	#752814
Coffee	#7E5236	#643925	#794E36	#6B3C27
Turmeric	#945524	#976049	#9F6331	#6C3B2D
Acai berry	#96664A	#825336	#663A27	#784830

Table 5: Conversion of RGB values from Figure 2 to hexadecimal notations.

As can be seen in Figure 2, different extraction media give rise to different colour shades. This result can be explained by the changes occurred in the structure of the colouring matters present in the vegetable when they are subjected to different pH values.

This is the basic principle of some natural dyes which can be used as indicators of basic and acid media.

The influence of the type of stirring (extraction in water) on the final result of the dyeing can be observed through the photographs shown in Figure 3. Table 6 shows the standard hexadecimal notation of the corresponding RGB values printed in Figure 3.

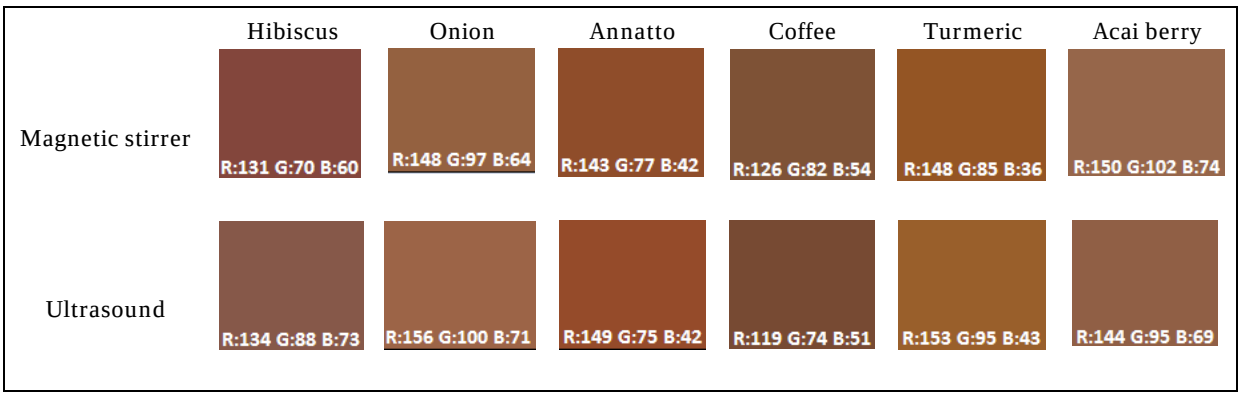

Figure 3: Photographs illustrating the influence of the type of stirring on the dyeing (extraction with water).

	Magnetic stirrer	**Ultrasound**
Hibiscus	#83463C	#865849
Onion	#946140	#9C6447
Annatto	#8F4D2A	#954B2A
Coffee	#7E5236	#774A33
Turmeric	#945524	#995F2B
Acai berry	#96664A	#905F45

Table 6: Conversion of RGB values from Figure 3 to hexadecimal notations.

Figure 3 clearly shows that the type of stirring had little or no influence on the final result of dyeing. Unlike what occurred while varying the pH of the medium, it is possible to observe that the colours obtained were the same regardless of the type of stirring. This result confirms that changes takes place in the structure of colouring matters present in the vegetable with changing pH of the medium and the type of stirring used to extract such colouring matters may only influence yield of the extract obtained.

Comparison of extraction yield

Table 7 shows the yield values for extractions carried out in various media with magnetic stirring.

According to various authors the yield extraction of natural dyes is still a challenge. Probably the high values found were related to the method used and did not correspond to the actual values. Nevertheless they were used as comparative measures between different dyes, since the conditions for determining the extraction yield were the same for all.

From the data presented in Table 7, it is not possible to make a correlation between the pH of extraction medium with the extraction yield. Probably this is due to the difference in solubility of the

various colouring matters in the different extraction media. However it should be noted that, in general, higher pH values often favor the extraction. Sodium hydroxide, as a non environmental friendly substance, was used only for investigative purposes. The aqueous urea solution is an environmental friendly option and showed good results for most of the obtained extracts. In view of this, urea could be a potential candidate for further studies related to environmental friendly media for extraction of natural dyes.

If we considered the highest values of yield in the extraction media of low environmental impact (highlighted in Table 7), the comparison of the obtained yield for the various dyes can be represented as the graph show in Figure 4.

	Water	Sodium bicarbonate	Urea	Sodium hydroxide
Hibiscus	**61**	46	52	53
Onion	34	8	**72**	23
Annatto	35	35	**47**	58
Coffee	51	53	**57**	69
Turmeric	0**	20	**34**	30
Acai berry	38	**41**	28	55

**Comparative values*
***This value was assigned to experimental error since the extract was obtained.*

Table 7: Yield values obtained for extractions carried out in various media with magnetic stirring.*

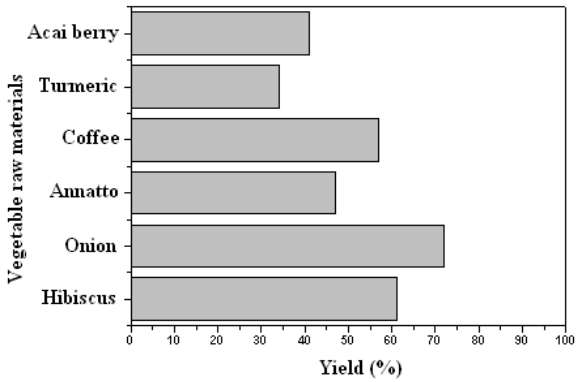

Figure 4: Maximum yield obtained with environmental friendly media.

Table 8 presents the comparative results between the water extraction carried out with magnetic stirring and ultrasound.

	Ultrasound	Magnetic stirring	Ratio US/MS
Hibiscus	52	61	0.85
Onion	31	34	0.91
Annatto	24	35	0.68
Coffee	51	51	1.00
Turmeric	54	0**	error
Acai berry	26	38	0.68

**Values without decimal places*
***This value was assigned to experimental error since the extract was obtained.*

Table 8: Values obtained for the yield of water extraction with magnetic stirring (MS) and ultrasound (US).*

As can be seen in Table 8, the type of stirring had no major influence on the yield of the extraction. Reminding that the extraction with magnetic stirring was carried out at 40 °C, it may be suggested that temperature is a parameter that has more influence on the extraction yield than the type of stirring. This result was in disagreement with those that were already reported [7, 11]. On the other hand, the stability of the colouring matters should be observed when using ultrasound technique [15]. In this way, the poor performance of the extraction with ultrasound could be attributed to some kind of degradation of the colouring matters.

Optical microscopy (OM)

Figure 5 (a-d) shows some micrographs, that is, images taken using a microscope, of cotton fabric samples dyed with annatto extract.

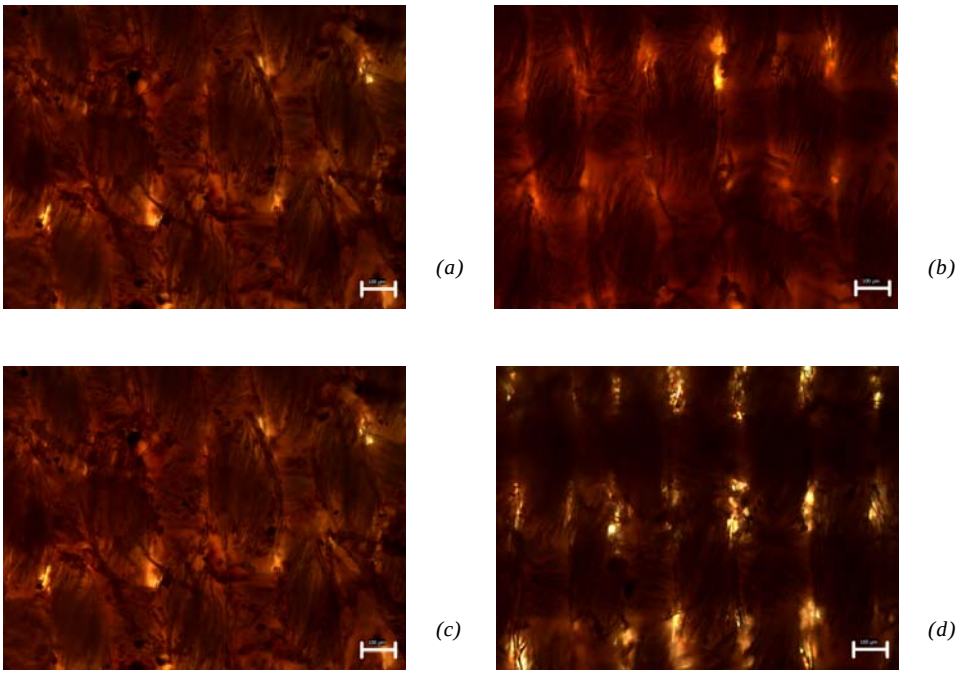

Figure 5: Optical microscopy of cotton fabric samples dyed with annatto extract: (a) water MS, (b) sodium hydroxide (MS), (c) water MS and (d) water US. Scale bar = 100 µm.

Comparing the micrographs corresponding to the extraction in water (a) with that in sodium hydroxide (b), both with magnetic stirring, it can be seen that the extraction carried out in alkaline conditions produced an extract with better covering power. This result might be related to better extraction yield in alkaline medium as showed in Table 7.

Regarding the micrographs c and d, it may be observed that the use of ultrasound was not effective to produce an extract with good covering power. This observation might be related with the results of the extraction yield presented in Table 8. Such results did not show improvement extraction yield by using ultrasound and suggests that the extraction yield is more affected by the temperature than the type of stirring.

Figure 6 shows the micrograph of the sample fabric dyed with turmeric extract obtained in water with ultrasound. As can be seen, in this case the poor covering power of the extract is not directly related to yield extraction of 54% achieved in these conditions (Table 8).

Figure 6: Optical microscopy of cotton fabric sample dyed with turmeric extract (water/ultrasound). Scale bar = 100 µm.

In view of this, it is reasonable to think that other factors in addition to the extraction yield influence the final result of the dyeing. Probably, it is not possible to directly relate the covering power with the extraction yield. One of the factors that could have influence would be the degree of interaction between the molecules of colouring matter from the vegetables with the fabric fibers.

Different dyes will certainly require different periods of fabric impregnation and different temperatures. The experiments performed in this study did not take into account these specificities.

Future investigations need to be done for the control of dyeing parameters such as extraction time, extraction temperature as well as dyeing conditions for various types of fibers.

Fastness to washing

Table 9 shows the indexes obtained in washing test. It is worth mentioning that lower indexes for colour transfer (Table 3) mean that the testimony fabrics were strongly stained. Lower indexes for colour change (Table 4) mean that dyed fabric samples faded more easily.

Annatto extract	Colour transfer	Colour change	Yield%
Water MS	2	2	35
Sodium bicarbonate MS	2	2	35
Urea MS	1	2	47
Sodium hydroxide MS	1	3	58
Water MS	2	2	35
Water US	1	2	24
Water MS TA mordant	1	4	35
Water MS ITA mordant	2	4	35

Table 9: Indexes for the washing tests according to Tables 3 and 4.

The results did not differ greatly among the vegetable sources used. For this reason, Table 7 shows only the results obtained with annatto extract. From Table 9 it could be suggested that alkaline medium is more effective for fixing the dye on cotton fabrics. Coincidently the best yield for extraction was achieved under this condition.

On the other hand, comparing the extraction carried out in magnetic stirring with that in ultrasound, it can be noticed that the dye extraction yield had no influence in fixing the dye on the fibre. In this case it is reasonable to think that the best performance in terms of fixing the dye on the fiber can be attributed to the alkaline medium and has no relationship with the extraction yield. As expected, the presence of mordant improves the dye fixing on the fibre.

An illustrative chart of the studied natural dyestuffs was assembled as shown in Figure 7. Such chart was presented in some regional textile industries to motivate them in making further studies with natural dyestuffs.

Figure 7: Colour chart of natural dyestuffs.

Conclusions

There is an increasing demand for environmentally friendly natural dyes. The re-establishment of the cultivation of dye plants in the agriculture and their use for dyeing textiles in industry is a tendency. To achieve this purpose, efficient low cost extraction methods for dye stuffs must be found and economically sustainable dyeing process will have to be developed.

In the present work, parameters for the extraction process such as influence of pH medium and type of stirring were analysed for hibiscus, onion, annatto, coffee, turmeric and acai berry and the extracts were used for dyeing cotton. Preliminary results found in this study suggest better performance when the extraction was carried out in alkaline medium at temperatures above 40 °C.

It might be concluded that it is possible to extract dyes from vegetable sources by using basic equipment. Further studies are needed to improve the conditions of the industrial extraction keeping in mind the thermal stability of natural dyes and the use of environmentally friendly mordants, among other issues.

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