JARTE
Journal of Applied Research in Technology & Engineering

1(1): 3-7, 2020
https://doi.org/10.4995/jarte.2020.13480

Received: April 4, 2020; Accepted: June 17, 2020; Published: July 31, 2020

OPTIMIZATION OF TEA EXTRACTS COMPOSITION TO DYE COTTON. TIME AND TEMPERATURE INFLUENCE

Louise Lambrecht[a1], Jaime Gisbert-Payá[b2], Eva Bou-Belda[b3], Marilés Bonet-Aracil[b4*]

[a] HO University College Ghent, Valentin Vaerwyckweg 1, 9000 Ghent, Belgium.
[a1] louise.lambrecht@student.hogent.be

[b]Universitat Politècnica de València, Plaza Ferrandiz y Carbonell s/n., E-03801, Alcoy, Spain.
[b1]jaigispa@txp.upv.es, [b2]evbobel@upv.es, [b3]maboar@txp.upv.es

Abstract:

The last few years natural dyes rose in value. Some synthetics dyes are proven to be environmentally harmful and can cause negative effects. Due to the eco awareness the natural dyes were again wildly used. Tea extracts from the *Camellia sinensis* plant were used. Tea has a large range of phytoconstituents and some can be transferred onto the cotton fabric. The optimal time and temperature to obtain as many phytoconstituents as possible had to be established. The aim of this study was to optimize the extraction process from tea so as to apply the maximum concentration of phytoconstituents onto the textile fibers and improve the cotton functionalization (Ultra violet protection for example) once it is dyed with the extract. Results demonstrate time and temperature had a great influence on the optimization of the tea extracts. We could conclude that after 2 hours the most polyphenols, hydrolysable tannins and condensed tannins are obtained and increasing the time didn't add any value. The temperature was a really important factor because the polyphenols derived around 70 °C so both the extraction temperature and dyeing treatment should be below 70 °C. Wastewater were characterized in order to determine the phytoconstituents were in the cotton fibres.

Keywords: tea extracts; cotton; Extraction; dyeing.

Cite as: Lambrecht, L., Gisbert-Payá, J., Bou-Belda, E., Bonet-Aracil, M. (2020). Optimization of tea extracts composition to dye cotton. Time and temperature influence. *J Appl Res Eng Technol & Engineering, 1*(1), 3-7. https://doi.org/10.4995/jarte.2020.13480

1. Introduction

Color is an important sensory attribute of food that often plays an important role in the market success of a product. Color is often used by the consumer as an indicator of a variety of qualities of the food product, such as flavor, safety, nutritional value and more. (Sigurdson et al., 2017). Synthetic dyes are used because of their high stability and low cost, high tinctorial strength and chemical stability. Due to the rising health concerns and eco-safety risks of synthetic dyes a non-toxic alternative had to be found. Eco-friendly natural dyes reemerged as a potential dye. Natural flora and fauna have an amazing variety of colors, this leads to endless possibilities. Natural dyes, obtained from plants, insects/animals and minerals are renewable and sustainable bioresource products with minimum environmental impact. (Kadoph, 2008)But there are still a lot of issues that are connected with natural dyes, that's why synthetic dyes are still wildly used. (Sigurdson et al., 2017). The pigment and biomolecules can be derived from plant pigments, animals, microbial organism and minerals dependent on the source the properties change. Plant pigment and dyes and animal ones can selectively absorb certain wavelengths of light and reflect other wavelengths. This property will be further discussed in the paper. Animal pigments are able to transport blood oxygen, protect predators, UV radiation, mating and more. Pigment production by various microbial organism has been used to help identify certain species. (Joshi et al., 2003).

Natural dyes can by classified in different groups according to its chemical structure such as anthraquinone (madder), apha napthoquinones (henna), flavones (weld), indigoids (indigo and tyrian purple), carotenoids (annatoo, saffron) etc. The replacement of synthetic dyes is among other things limited by the inability of natural pigments to match the color characteristics of synthetic colorants. The classification of natural derived colorants is complex because of the wide variety of innate properties of the coloring substances. (Sigurdson et al., 2017)

Any process based on the use of natural dyes will have to consider the environmentally sound processing techniques and the overall process must be competitive with regard to the use of chemicals, energy, wastes and wastewater and overall costs. (Bechtold et al., 2008) This is challenging because during extraction a lot of solvents are used and they have a big environmental impact and can be damaging for the environment. Not only the extractions have to be considered but also the coloring process itself. Sometimes mordants and auxiliary chemicals are used which can be even more pollutant than dyes. (Patil, 2019). Studies have shown

*Corresponding author: M. Bonet-Aracil, maboar@txp.upv.es

that tea extracts obtain other properties as well such as antimicrobial activity of the fabric, antibacterial, sun protection factor, pH indicator, deodorizing finishing, antifeedant finishing of textiles, UV protective clothing, antimicrobial finishing of textiles, Dye sensitized solar cells, cosmetics and pharmaceuticals (Banerjee, 2015). Thus, knowing the extraction liquor composition can be of interest to determine which are the compounds which migrate to water.

Some studies demonstrated the pertinence of conventional methods by UV-vis spectrophotometer to determine the presence of Polyphenols, tannins and condensed tannins (Naima et al., 2015). A lot of research has been done about the determination of the presence of polyphenols. The most well-known method is the Folin-Ciocalteu method from (Singleton & Rossi, 1965). Gallic acid is used as a standard to obtain a calibration curve and in this way the concentration of polyphenols can be derived. To determine the concentration of proanthocyanidin or condensed tannins the BuOH/HCl test was found. (Scalbert et al., 1989). The hydrolysable tannin content was measured by using the potassium iodate test described by (Bossu, 2006). All the test measured the asborbance of the samples using the UV-Vis spectrophotometer ZUZI 4251/50 HJD003. However, tea extracts have not been characterized yet and much less when used for dyeing.

Color when dyeing with natural dyes, concisely with tea, can vary depending on the extraction procedure. (Bonet-Aracil, et al., 2016). However, colour and other properties, can be attributed to different compounds on the extraction. This paper will examine the parameters temperature and time during the extraction procedure which influence the liquor composition from tea extracts. Tea will be reused in order to evaluate if phytoconstituents remain after one extraction, focused on the reuse of tea byproducts from food industry. The tea extracts will be used to color textile and improve the UPF property (Rungruangkitkrai, 2012). Cotton will be dyed, and wastewater will be analyzed in order to determine the presence of polyphenols, tannins and condensed tannins on the cotton fiber.

2. Experimental

2.1. Materials

Every test was conducted with deionized water. Red tea was bought in a local herbalist´s shop, it was in bulk and it was said to be Spanish Red tea from Galicia region.

To characterize the tea extraction liquor it was used: KIO_3 from R.P. NORMAPUR -VWR international S.A.S): tannic acid ($C_{76}H_{52}O_{46}$) from Sigma Aldrich); HCl 37% from PanReac AppliChen; Butanol (BuOH) 99.5% PS $CH_3(CH_2)_3OH$ from PanReac AppliChen; Na_2CO_3 from PanReac AppliChen; Folin-Ciocalteu reactive from PanReac AppliChen); Gallic acid 97% $(HO)_3C_6H_2CO_2H$ (Sigma Aldrich).

2.2. Methods

2.2.1. Extraction method

The optimal temperature and time of tea extracts have to be determined to obtain as many polyphenols and tannins as possible. The tea extracts were obtained using the infusion procedure. To prepare the red tea extracts 10 g of red tea were mixed with 200 mL of distilled water. The water was preheated to 5 °C, 70 °C and 100 °C. These temperatures were chosen because previous research stated that polyphenols derive at 60 °C (Gironi & Piemonte, 2011). The polyphenols and the tannin concentration were determined of each sample at different times. Every 30 min for 2 h a sample was taken. After two hours the same tea leaves were mixed with fresh preheated distilled water to compare the concentration of biomolecules to see if the concentrations stayed the same or decreased.

2.2.2. Polyphenols characterizations

The Folin-Ciocalteu method (Singleton & Rossi, 1965) was used to obtain the polyphenols in the red tea extracts. The Folin reagent was diluted 10 times. From this dilution 5 mL was taken and 1 mL of tea extract/water (1/50) was added. Two millilitres of Na_2CO_3 (75 g/L). The mixture was heated to 50 °C for 5 min. The absorbance of the mixture was read at λ_{max} centered at 655 nm. A calibration curve was drawn using a gallic acid solution (80 mg/L, UV-Vis spectrophotometer ZUZI 4251/50 HJD003).

The results were expressed in milligram of gallic acid equivalent (GAE) per gram of dry tea.

2.2.3. Hydrolysable Tannins characterizations

The hydrolysable tannin content was determined with potassium iodate test described by (Bossu, 2006). (2.5% v/v) was made. This solution was heated for 7 min at 30 °C, then 1 mL of red tea extract was added. The mixture was placed in a water bath and heated to 30 °C for 2 min before putting into the UV VIS spectrophotometer. First a calibration curve had to be obtained by using the tannic acid solution of 5000 mg/L. This solution was carried out by mixing 0.25 of tannic acid with $MeOH/H_2O$ (40/10). Different concentrations of tannic acid were made: 10, 50, 100, 250, 500, 1000 and 1250 mg/L. The absorbance of this mixture was measured by using UV-Vis spectrophotometer ZUZI 4251/50 HJD003 at a wavelength of 500 nm, and a curve was drawn. Results were expressed in milligram of tannic acid equivalent (TAE) per gram of dry tea.

2.2.4. Condensed tannins characterization

Condensed tannins or Proanthocyanidin content was determined with a BuOH/HCl test. (Scalbert et al., 1989). The red tea extract was diluted 50 times and added to 5 mL of an acidic ferrous solution (77 mg of $FeSO_4 \cdot 7H_2O$ in 500 mL of HCl/BuOH (2/3). The tubes were heated to 95 °C for 15 min. The absorbance was read at 530 nm (UV-Vis spectrophotometer ZUZI 4251/50 HJD003).

The results were expressed in milligram of cyanidin acid equivalent (Cya) per gram of dry tea. The condensed tannins content was calculated according to (Naima et al. 2015) using this formula:

$$\left[\frac{mg\ CyaE}{g\ bark}\right] = \frac{A \cdot V \cdot D \cdot M \cdot V_2}{l \cdot \epsilon \cdot v \cdot m}$$

Where:

A = absorbance at 530 nm; V = total reaction volume (mL); D = dilution factor; M = cyanidin molar mass (g/mol); V_2 = aqueous volume extract used to extract the red tea; L=path length (1/cm); ϵ=molar extinction coefficient 34700 L/(mol·cm); v=0.5 mL; m=dry weight mass of tea.

2.2.5. Dyeing procedure

The textile was colored using the exhaustion method. Dyed samples were prepared for color measurement, which was carried out by following a standard procedure as described in the next item. The tea extracts were used to color cotton, 40 mL of tea extract obtained at 50 °C was mixed with 160 mL of distilled water. The solution was put into a water bad at 50 °C. This was done twice, in one beaker a piece of cotton (5 g) was placed. The bath ratio was 1/40. This was colored for 45 min and every 15 min a sample of the wastewater was taken from the cotton beaker and the beaker without cotton.

2.2.6. Colour characterization

In order to objectively compare color difference measurements, the chromatic coordinates (CIE L*, a*, b*) of the CIELAB color space of the dyed samples were obtained using a MINOLTA CM-3600d reflection spectrophotometer. UV energy was included. The measurements were made with the standard observer CIE-Lab 10° and the standard illuminant D65.

On the other hand, the color difference of the samples was obtained according to the following equation:

Colour difference $(\Delta E^*) = ((\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2)^{1/2}$

Where ΔL*=L* with microcapsules–L* without microcapsules; Δa*=a* with microcapsules–a* without microcapsules; Δb*=b* with microcapsules–b* without microcapsules; “L*” describes the luminosity, “a*” measure of red-green hues, “b*” measure of blue-yellow shades. It should be noted that three measurements were made for each sample and the mean value was calculated.

3. Results

3.1. Polyphenols

Three temperature extractions (50 °C, 70 °C, 100 °C) were tested on red tea from the *Camellia sinensis* plant. Results are presented in Figure 1. The choice of the best temperature given to a good yield of polyphenols and tannins using distilled water as an extraction solvent.

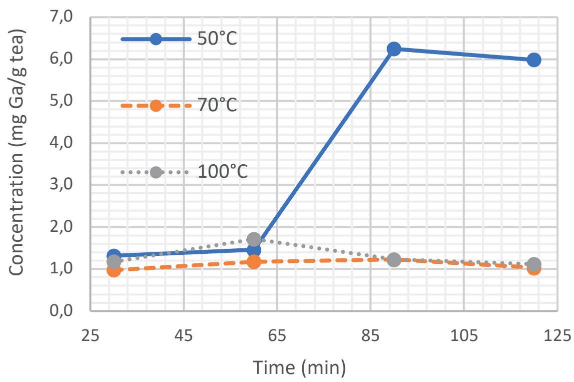

Figure 1: Polyphenol content in red tea.

As it can be observed in Figure 1, testing the behavior for 120 min, the most polyphenols were obtained at 50 °C after 90 min. This is in line with previous research stating that polyphenols derive at 70 °C and after 1 h. (Bindes et al., 2019). When the extraction is only 60 min a higher temperature can be used, despite the percentage of polyphenols extracted is considerable lower than for 90 minutes. The polyphenol content in red tea increases exponentially after 90 min and then decreases a bit. This is due to the rising to a higher temperature. Due to this tendency, reduction in polyphenols content, authors considered it has no sense to increase the period.

3.2. Hydrolysable tannins

Regarding the hydrolysable tannins, all absorbances obtained from the UV-spectroscopy, according to the methods described in methods section, were processed using the calibration curve and shown in Figure 2. Every value was expressed in mg TAE/g dry red tea. The optimal temperature to extract the hydrolysable tannins was at 100 °C, disregard the time for the evaluation however, 120 min shows a stabilization for 70 °C and 50 °C, and slightly increasing for 100 °C. It can be clearly observed a difference in comparison with polyphenols extraction. Polyphenols extraction is similar at short periods for each tested temperature, but tannins increase with temperature and time.

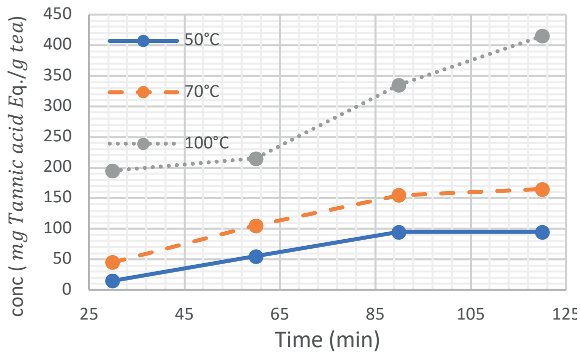

Figure 2: Hydrolysable tannins content in red tea.

3.3. Condensed tannins

Condensed tannins content was calculated using the formula and was put in Figure 3.

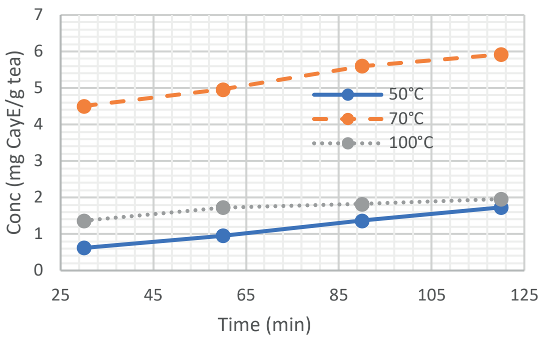

Figure 3: condensed tannin content in red tea.

The graph, similarly to hydrolysable tannins, shows an increasing tendency, but in contrast, the best temperature to obtain condensed tannins is at 70 °C, and not 100 °C as for hydrolysable. The difference in concentration between 50 °C and 100 °C is rather small, and a considerable increase can be observed at 70 °C. The best extraction time is 2 h, and apparently it tends to stabilize the concentration for this period. The concentration of mg cyanidin per grams of dry tea is equivalent to the mg of condensed tannins in red tea.

3.4. Dyeing

When fabric has been dyed (Table 1) it is clearly observed a decrease in luminosity (L*) which shifts from 92.678 to 85.207. Furthermore, it is noticeable there is a colour difference (ΔE*ab) of 12.656. Usually, ΔE*ab higher than 1, implies the colour difference can be observable by human eye. Hence, ΔE*ab > 12 implies an considerable intensity in the dyed sample. This difference is attributes not only to the L* but also to a* and b* variations. Colour coordinates allow to appreciate a movement towards more reddish due to an a* increase and towards more yellowish colour due to the b*increase. As both a* and b* values are closer to the center (0 value) we can confirm that we are not moving on the region of pure colours what would offer an orange. Thus, according to CIELab values we can objectively confirm the dyed fabric with red tea shows a light brown. This result can be observed on the K/S curve shown in Figure 4. K/S equals the strength of colour. It is observable, that there is an increase in K/S mainly from the region between 400-550 nm, this equals to a brownish colour.

Table 1: CIE L*a*b*.

	L*	a*	b*	ΔE*ab
Cotton	92.678	-0.147	3.097	
Red cotton	85.207	3.136	12.769	12.656

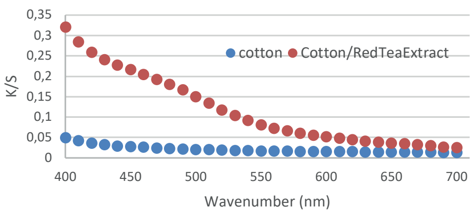

Figure 4: coloration cotton with red tea extract.

In order to show the final aspect of the dyed fabrics apart from the CIELab coordinates or colour strength (K/S). Figure 5 shows the white cotton fabric and the dyed one.

a) b)

Figure 5: Cotton dyed with red tea extract. a) cotton b) cotton + red tea;

It can be appreciated (Fig. 5a) that cotton results on a white fabric, whereas when the red tea has been used to dye cotton, the fabric (Fig. 5b) shows a brownish colour. This fits with a higher L* for the sample dyed with red tea.

3.5. Tea reuse

The use of natural dyes is increasing due to environmental concerns, as the components are biodegradable and they are not as pollutant as the synthetic ones. Tea is widely used for food industry. Tea used for other purposes apart from food, can imply side effects such as the increase of price, etc. which is not ethically correct at all. Hence, it was considered the possibility to get tea as a byproduct from food industry. The tea leaves used during the extraction were dried and used again with 200 mL of fresh distilled water. The reused tea had almost none polyphenols, condensed tannins and hydrolysable tannins left. The values that were obtained, were negligible in comparison with the high values after 2 h of extractions. Table 2 shows the values. The reference code can be explained as follows: R is red tea extract- the extraction temperature- the extraction time.

As it can be appreciated, Table 2 shows no significative results according to the concentration of phytoconstituents. Thus, tea byproduct from food industry is not likely to be used in order to functionalize textiles.

Table 2: Polyphenols and tannin content reused tea leaves.

Red tea extract	Cond. Tannins (mg CayE/g tea)	Hydrol. Tannins (mg TAE/g tea)	Polyphenols (mg GAE/g dry tea)
R-50°C-30’	0.546	5	0.946
R-50°C-60’	0.546	5	1.174
R-50°C-90’	0.546	15	1.516
R-50°C-120’	0.637	25	1.630
R-70°C-30’	0.273	35	0.689
R-70°C-60’	0.546	35	0.889
R-70°C-90’	0.637	55	0.889
R-70°C-120’	0.637	125	0.860
R-100°C-30’	0.592	115	1.174
R-100°C-60’	0.592	135	1.459
R-100°C-90’	0.637	165	1.202
R-100°C-120’	0.865	195	1.145

4. Conclusion

Results from this paper reveal that tea extracts obtain different amounts of phytocomponents depending on the extraction method. The most important factor is the temperature because polyphenols can derive if the temperature is too high. The temperature should range around 50 °C - 60 °C to obtain as many polyphenols as possible. The most condensed tannins can be obtained at 70 °C and the optimal temperature for hydrolysable tannins is at 100 °C. But this would lead to a degradation of polyphenols, so the temperature used for the extractions should be kept lower than 70 °C. The optimal time to extract phytoconstituents of red tea is 2 hours. After 2 hours most components have been extracted and further reheating doesn't add value to the process.

To sum up the optimal temperature to take the extracts is at 50 °C for 2 hours.

Furthermore, we considered reusing tea in order to consider the possibility of taking profit from the byproducts from food industry. However, results demonstrate it is not feasible to get enough phytoconstituents when the tea has been treated for two hours.

References

Banerjee, S., & Chatterjee, J. (2015). Efficient extraction strategies of tea (*Camellia Sinensis*) biomolecules. *Journal of food science and technology*, *52*(6), 3158-3168. https://doi.org/10.1007/s13197-014-1487-3

Bechtold, T., Mahmud-Ali, A, Ganglberger, E., & Geissler, S. (2008). Efficient processing of raw material defines the ecological position of natural dyes in textile production. *International Journal of Environment and Waste Management*, *2*(3), 215-232. https://doi.org/10.1504/IJEWM.2008.018244

Bindes, M.M., Cardoso, V.L., Hespanhol, M., Reis, M., & Boffito, D.C. (2019). Maximisation of the polyphenols extraction yield from green tea leaves and sequential clarification. *Journal of Food engineering*, *241*, 97-104. https://doi.org/10.1016/j.jfoodeng.2018.08.006

Bonet-Aracil, M.A., Díaz-García, P., Bou-Belda, E., Sebastiá, N., Montoro, A., & Rodrigo, R. (2016). UV protection from cotton fabrics dyed with different tea extracts. *Dyes and Pigments*, *134*, 448-452. https://doi.org/10.1016/j.dyepig.2016.07.045

Bossu, C.M., Ferreira, E.C., Chaves, F.S., Menezes, E.A., & Nogueira, A.R.A. (2006). Flow injection system for hydrolysable tannin determination. *Microchemical Journal, 84*(1-2), 88-92. https://doi.org/10.1016/j.microc.2006.04.022

Gironi, F., & Piemonte, V. (2011). Temperature and solvent effects on polyphenol extraction process from chestnut tree wood. *Chemical Engineering Research and Design*, *89*(7), 857-862. https://doi.org/10.1016/j.cherd.2010.11.003

Joshi, V., Devender, A., Bala, A., & Bhushan, S. (2003). Microbial pigment. *Indian Journal of Biotechnology*, 2(3), 362-369.

Kadolph, S. (2008). Natural dyes: a traditional craft experiencing new attention. *Delta Kappa Gamma Bulletin*, *75*(1), 14.

Naima, R., Oumam, M., Hannache, H., Sesbou, A., Charrier, B., Pizzi, A., & Charrier–El Bouhtoury, F. (2015). Comparison of the impact of different extraction methods on polyphenols yields and tannins extracted from Moroccan *Acacia mollissima* barks. *Industrial Crops and Products*, *70*, 245-252. https://doi.org/10.1016/j.indcrop.2015.03.016

Patil, D.P. (2019). Short communications Padding technique for natural dyeing. *Indian Journal of Fibre and Textile Research*, 118-121.

Rungruangkitkrai, N., & Mongkholrattanasit, R. (2012). Eco-Friendly of textiles dyeing and printing with natural dyes. In *RMUTP International Conference: Textiles & Fashion* (Vol. 3, pp. 1-17).

Scalbert, A., Monties, B., & Janin, G. (1989). Tannins in wood: comparative study on the phenolic acids identified and quantified in dry beans using HPLC as affected by different extraction and hydrolysis methods. *Journal of Agricultural and Food Chemistry, 37*(5), 1324-1329. https://doi.org/10.1021/jf00089a026

Shahid, M., & Mohammad, F. (2013). Recent advancements in natural dye applications: a review. *Journal of Cleaner Production*, *53*, 310-331. https://doi.org/10.1016/j.jclepro.2013.03.031

Sigurdson, G.T., Tang, P., & Giusti, M.M. (2017). Natural colorants: Food colorants from natural sources. *Annual review of food science and technology*, *8*, 261-280. https://doi.org/10.1146/annurev-food-030216-025923

Singleton, V.L., & Rossi, J.A. (1965). Colorimetry of total phenolics with phosphomolybdic-phosphotungstic acid reagents. *American journal of Enology and Viticulture*, *16*(3), 144-158.