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Toxicity and environmental impacts approached in the dyeing of polyamide, polyester and cotton knits

Jorge Marcos Rosa[a,b,*], Vanessa S.G. Garcia[a], Nathalia F. Boiani[a], Camila G. Melo[a], Maria C.C. Pereira[a], Sueli I. Borrely[a]

[a] Instituto de Pesquisas Energéticas e Nucleares, São Paulo, Brazil
[b] Faculdade de Tecnologia SENAI Antoine Skaf, São Paulo, Brazil

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ABSTRACT

Three colouration process was analyzed, in which consumption of water, the spent of electrical and thermal energy, emission of carbon dioxide besides effluent toxicity of dyeing of polyamide, cotton and polyester knits were approached. the dyeing of polyamide knit presented lowest consumption of electrical energy, the dyeing of polyester knit presented the lowest consumption of thermal energy and emission of co_2 molecules into atmosphere, and the effluent of cotton dyeing presented lowest acute toxicity (CE_{50}) to *Daphnia similis*.

1. Introduction

The textile activity in Brazil represents approximately U$ 45 billion, equivalent to 7% of the total production value of the Brazilian manufacturing industry. The jobs generated by the textile chain reached 1.5 million in 2017 roughly 18.7% of the total industrial production [1]. Despite of its importance for developing countries, the manufacturing of textiles is a complex issue in relation to the environment, water usage, and resultant pollution for receiving water systems. The textile processing consists of many operations, such as scouring, desizing, washing, bleaching, dyeing and printing. Therefore, real industrial textile wastewater has an extremely complex matrix containing many chemical ingredients in addition to dyestuff [2,3]. Among various complex constituents present in textile wastewaters, the dyestuff can be considered as the most peremptory source of contamination followed by the surfactants. The direct discharge of the coloured textile effluent into the freshwater bodies adversely affects the aesthetic aspect, water transparency and dissolved oxygen content [4]. The final results of such type of discharges are the damages to aquatic biota.

Several methods and processes for textile wastewater treatment have been studied such as membrane system [5–8], ultrafiltration [9], adsorption [10–12], hybrid process [13,14], electrochemically advanced oxidation [15], biological processes [16,17], degradation of dyestuff from soil by ornamental plants [18] and utilization of textile effluent as the nutrient to *Chlorella variabilis* [19].

Looking for a cleaner production, novel fabrics and fibers were obtained as man-made Lyocell fibers [20], the ionic liquid application in dyeing [21,22] new dyestuffs were obtained from renewable sources [23], catalyst immobilization and development for the application of enzymes for textile processing [24,25], cleaner colouration with reactive dyestuff using a pad-batch-steam process [26] and disinfection of textiles using low temperature plasma [27]. Technologies, ideas, and actions for a better environment into the textile sector have to achieve pollution discharge reduction mainly for liquid effluents forcing/and technology development also due to the demand of water and tightening environmental legislation on the effluents generated by textile industry and water scarcity in different areas of the world [28].

Monitoring toxic charge of effluents is a mandatory action nowadays for the protection of water resources. As an example, three types of reactive dyestuffs were assessed for toxicity by Baumer et al. [29]. The authors applied an oxidative system including enzymatic systems as an alternative for textile effluents treatment. However, during their studies the oxidative processes were able to degrade organic products but toxicity was not reduced (for acute and chronic effects - *D. magna* and *V. fischeri.*), they also obtained good results for the decolourisation. Croce et al. [30] investigated the aquatic toxicity of 42 commercial dyestuffs using the application of in silico tools. Ecological bioassays acute and long-term exposure with *Daphnia magna* and *Raphidocelis subcapitata* were applied in order to evaluate the potential impact of these products. The authors concluded that only 9 formulations showed toxicity lower than 100 mg L^{-1} for *D. magna* while 30 dyestuffs were toxic for *R. subcapitata*.

Toxicity evaluation of textile dyeing effluent and its possible relationship with chemical oxygen demand was studied by Liang et al.

* Corresponding author.
E-mail address: jotarosa@hotmail.com (J.M. Rosa).

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[31], testing the acute toxicity against *Vibrio fischeri* and *Desmodesmus subspicatus*. The authors identified a positively correlation with COD in different textile dyeing effluents, with R^2 values higher than 0.84.

So, the objective of this present paper was the analysis of several factors related to ecological costs: water consumption, the emission of carbon dioxide, the consumption of electrical and thermal energy during dyeing processing of cotton (CO), polyamide (PA) and polyester (PET) fibers that have been intensively studied mainly because of the microplastic introduced into effluents [32,33].

Furthermore, nowadays in Brazil there is a research group which intend to suggest an insertion of a cloth tag, containing information about water and energy consumption, as well as a toxicity of the effluent that was generated by the dyeing process of this cloth. Based on those facts, the paper also was suggesting a methodology in order to contribute with it.

2. Materials and methods

All fibers were dyed in a specific colour, Pantone 19-1619, selected from SENAI Fashion Design Book of Tendency Autumn/Winter 2019. The acute toxicity of the real effluents obtained from the dyeings was tested with *Daphnia similis*. These fibers, PA, CO and PET, were chosen for this evaluation once they are the most used fibers in the Brazilian clothing making [1].

2.1. Materials

2.1.1. Auxiliaries

Acetic acid 98%, sulfuric acid 98%, ammonium sulphate 98%, sodium hydroxide 98%, sodium metasilicate, sodium carbonate 98%, hydrogen peroxide 50% and sodium chloride 98%, supplied by Labsynth; sequestering, nonionic detergent and catalase enzyme supplied by Golden Technology; leveling agent, dispersant and fixating agent supplied by Archroma. All auxiliaries were used with no previous purification.

2.1.2. Dyestuffs

Acid Orange 67 (AO67), Acid Red 299 (AR299), Acid Blue 113 (AB113), Disperse Yellow 235 (DY235), Disperse Red 73 (DR73) and Disperse Blue 165 (DB165) supplied by Archroma, Reactive Yellow 145 (RY145), Reactive Red 239 (RR239) and Reactive Blue 222 (RB222) supplied by Golden Technology. All dyestuffs were used with no previous purification.

2.1.3. Substrates

The dyeing have been done using knits produced in an Orizio circular monofronture, John/C model, 3.0 feeders per inch, diameter of 30 inch, 28 gauge, 30 RPM, being 30/1 Ne carded yarn and gramature of 142 g m^{-2} to CO knit; 200/72 dTex multifilament yarn and gramature of 140 g m^{-2} to PET knit; 200/96 dTex multifilament yarn and gramature of 140 g m^{-2} to PA knit, all of them with 0.90 m of width.

2.2. Methods

2.2.1. Dyeing

The dyeing processes were carried out using a 10:1 liquor ratio. The formulations with the amount of chemicals used in the dyeing with acid (PA), disperse (PET) and reactive (CO) dyestuffs and its auxiliaries, as described in Table 1. All steps, pre-treatment, dyeing and after-treatment, were conducted according to literature about dyeing of CO with reactive dyestuffs [34–41], dyeing of PA with acids dyestuffs [35–40,42–44] and dyeing of PET with disperse dyestuffs [35–40,45–48], as well as according both chemicals and dyestuffs suppliers' recommendations.

The process of PA dyeing was shown as Fig. 1, while CO dyeing process was represented in Fig. 2 and PET dyeing process in Fig. 3. The figures showed the time of textile processing, which were used to determinate the electrical energy consumption, the sequence of chemicals addition and the heating temperature, in order to calculate the necessary amount of thermal energy and the consequent amount of CO_2 released into the atmosphere.

Table 1
Amount of chemicals used in the dyeing according to fiber type.

Chemicals			PA	CO	PET
a	Sequestering	(g L^{-1})	1.00	1.00	1.00
b	Nonionic detergent		1.00	1.00	1.00
c	Sodium carbonate		0.50	5.00	0.50
d	Sodium hydroxide		–	1.40	1.00
e	Sodium metasilicate		–	0.50	–
f	Sodium chloride		–	50.00	–
g	Fixation agent		2.00	–	–
h	Ammonium sulphate		1.00	–	–
i	Dispersant		–	1.00	1.00
j	Catalase enzyme		–	0.50	–
k	Levelling agent		1.00	–	–
l	Sulfuric acid	(mL L^{-1})	–	0.14	–
m	Hydrogen peroxide		–	2.00	–
n	Acetic acid		0.30	0.50	0.50
o	RY145	(% / owm*)	–	0.91	–
p	RR239		–	1.22	–
q	RB222		–	0.76	–
r	AO67		0.66	–	–
s	AR229		0.34	–	–
t	AB113		0.22	–	–
u	DY235		–	–	0.82
v	DR73		–	–	0.78
x	DB165		–	–	0.49

* = on weight of material.

The reflectance (R%) and the differences of CIELab system (ΔL^*, Δa^* and Δb^*) from all dyeing were determined by spectrophotometry under illuminant D_{65}, 10° (Konica Minolta CM-3600d) in the wavelength of maximum reflection of the colour.

For the measurement of colour, standard values are used worldwide, for example as determined by an organization called *Commission Internationale de l'Eclairage* (CIE). The values used by CIE are called L^*, a^* and b^* and the colour measurement method is called CIELab [49].

The system can be plotted in a colours space with three axis (Fig. 4), where L^* axis represents the difference between white ($+L^*$) and black ($-L^*$), a* axis represents the difference between green ($-a^*$) and red ($+a^*$) and b* axis represents the difference between yellow ($+b^*$) and blue ($-b^*$).

The colours measurements were determined by spectrophotometry under illuminant D_{65}, 10° (Konica Minolta CM-3600d). The values of Euclidean distance (ΔE^* - also known by colour difference) in the colours space was calculated using the Eq. (1) given below [41].

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

2.2.2. Ecological costs

In order to determine the consumption of electrical energy for dyeing, the parameters of Jet HT Riviera Eco Metalwork were used, considering the capacity of 50 kg and installed potency of 7.4 kW. The theoretical consumption for each kilogram of processed substrate was determined by the time of the process, in minutes, applying Eq. (2) [50].

$$Q_E = \frac{t \times I_P \times 6.00 \times 10^4}{E_C} \quad (2)$$

$Q_E = J\ kg^{-1}$; t = *process time in min*; I_P = *installed potency*; E_C = *equipment capacity*

For the amount of required thermal energy for each kilogram (substrate), the Eq. (3) was applied.

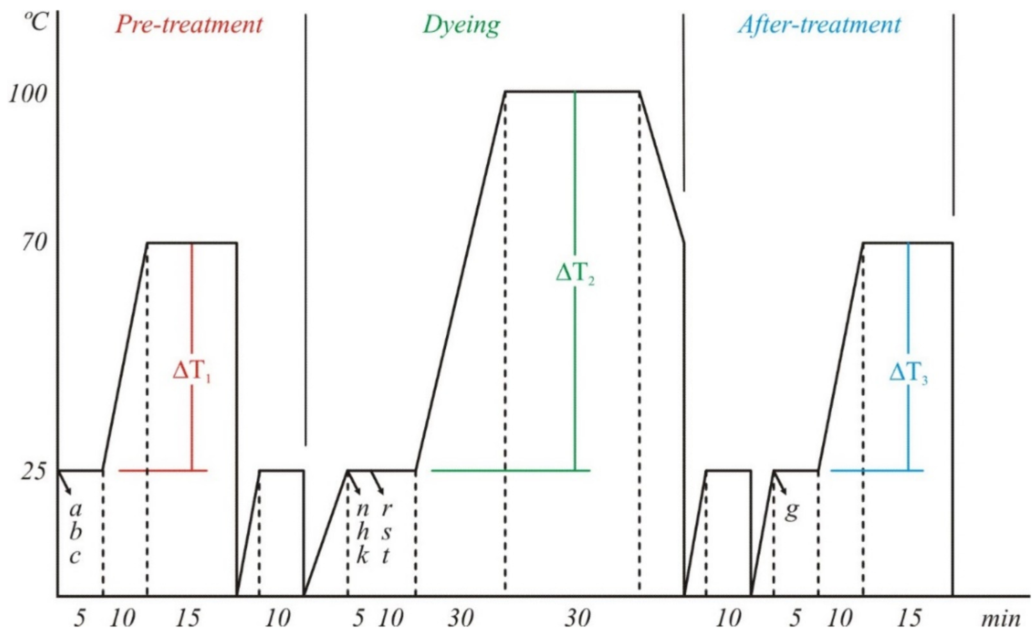

Fig. 1. Dyeing procedure of PA with acid dyestuffs.

$$Q_T = (\Delta T_1 + \Delta T_2 + \Delta T_n) \times Cp_{H_2O} \times m_{H_2O} \times 10^{-3} \quad (3)$$

$Q_T = J\ kg^{-1}$; T in Kelvin; Cp in $J\ kg^{-1}K^{-1}$, and m in grams, adopting specific mass of water = 1.0 g cm^{-3} and liquor ratio equal to 10 liters per kilogram of fiber.

According to the supplier of fuel gas, (Comgas Company of São Paulo), the fuel gas is a gas mixture containing 89.0% of methane, 6.0% of ethane, and 1.8% of propane. Based on the inferior calorific power (ICP) standards laid down by ASTM D 3588-98 as being $3.70 \times 10^7\ J\ m^{-3}$ for methane, $7.00 \times 10^7\ J\ m^{-3}$ for ethane, and $9.23 \times 10^7\ J\ m^{-3}$ for propane. An ICP of $4.02 \times 10^7\ J\ m^{-3}$ was calculated for the gas mixture [50]. In order to calculate the volume of gas needed for heating the amount of water for dyeing a kilogram of substrate, using an Etna GHV-2000 steam generator with production capacity of 5.56×10^{-1} kg s^{-1}, maximum allowable operating pressure (MAOP) equal to 1.0 MPa, and operating with 85% of efficiency as a parameter, the Eq. (4) was used.

$$V_1 = \frac{Q_T}{4.02 \times 10^7 \times E_{SG}} \quad (4)$$

V_1 is in $m^3 kg^{-1}$, Q_T in $J\ kg^{-1}$ and E_{SG} is the efficiency of the steam generator

To calculate the mass of CO_2 emissions during the supply of heat energy, assuming that the gas is ideal and it is in normal conditions of pressure and temperature, the Eq. (5) was used.

$$mCO_2 = \frac{P \times V_1 \times 4.40 \times 10^{-2} \times 2.66}{R \times T} \quad (5)$$

mCO_2 in kg, P = 101.3 kPa, V_1 in m^3, R = 8314 m^3 kPa $mol^{-1} K^{-1}$, T = 273.15 K

The consumption of water (C_W), adopting liquor ratio equal to 10 liters per kilogram of fiber, was determined by Eq. (6).

$$C_W = LR_1 + LR_2 + LR_3 + LR_n \quad (6)$$

C_W in $L\ kg^{-1}$, LR_1, LR_2, LR_3 and LR_n = liquor ratio used in each process step

2.2.3. Toxicity measurements

Ecotoxicity assays were performed with daphnids, using a serial dilution of the effluents and twenty organisms were exposed to each effluent concentration. The effective lethal concentration was the endpoint (EC_{50}), meaning the concentration at which 50% of the

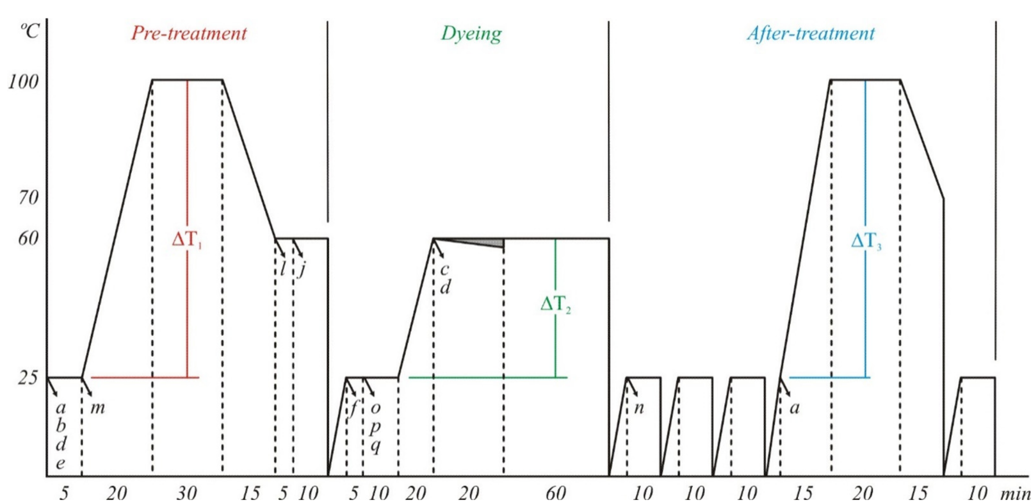

Fig. 2. Dyeing procedure of CO with reactive dyestuffs.

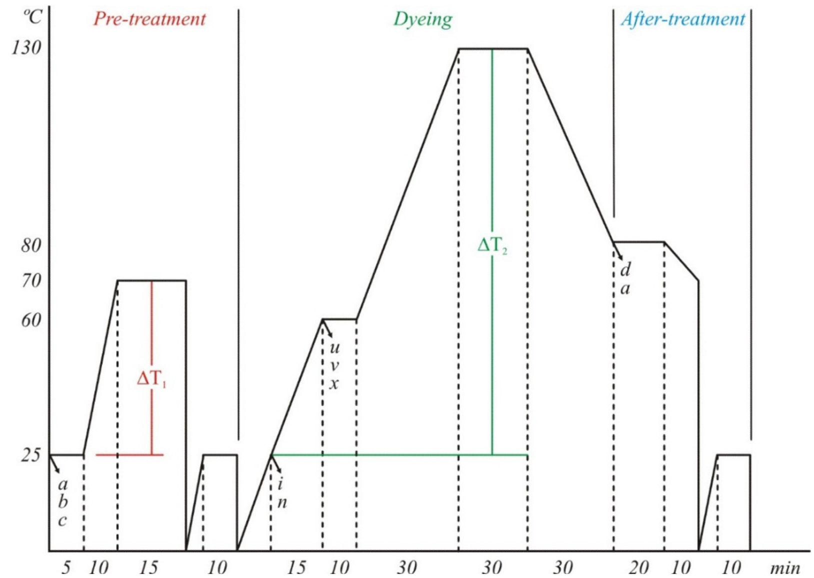

Fig. 3. Dyeing procedure of PET with disperse dyestuffs.

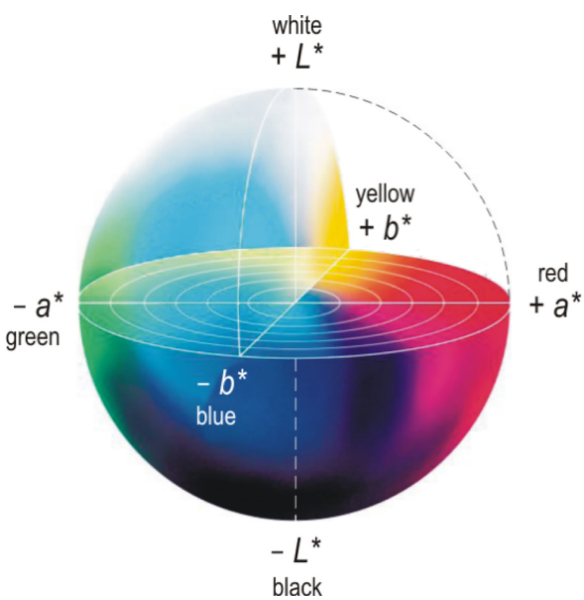

Fig. 4. Colours space in CIELab system [49].

individuals were damaged after a specified length of exposure: 48 h for *D. similis* water fleas, which means immobilization.

All the methods carried out during toxicity assays were based on standard laboratory conditions, following ABNT Brazilian Methods [51], since the raring of *D. similis*, until the exposition to the textile samples. Trimmed Sperman Karber method was applied as statistical analysis after the exposure.

Acute toxicity tests with this organism are very used in environmental reports of water quality and required in monitoring by environmental monitoring Brazilians agencies. It has been used in many scientific studies, such Leite et al. [52] and Vacchi et al. [53,54] that assessed the ecotoxicology of Disperse Red 1 dyestuff in their studies; Rocha et al. [55] whose performed the ecotoxicological risk assessment of Acid Black 210 dyestuff through testing its toxicity; Meireles et al. [56], that assessed the ecotoxicology of Disperse Red 73 dyestuff or even by Luna et al. [57], in which authors evaluated the ecotoxicity of five dyestuff to freshwater organisms before and during their photo-Fenton degradation.

3. Results

3.1. Dyeing

The results of dyeing processes are described in Table 2 in terms of the values of reflectance (R%), partial differences (ΔL^*, Δa^*, and Δb^*) and the colour difference (ΔE^*), being the Euclidean Distance in CIELab colour space calculated between the samples colour and the standard colour (Pantone 19–1619).

The colour difference between PA and CO was $\Delta E^* = 1.76$, between PA and PET was $\Delta E^* = 1.06$ and between CO and PET was $\Delta E^* = 1.50$. All values of ΔE^* between dyeing did not exceed 2 points. These data are acceptable when compared with the standards used in the Brazilian

Table 2
Results of dyeings according CIELab system.

Pantone 19-1619 at 690 nm		PA	CO	PET	Partial deviation			Total deviation (ΔE^*)		
					PA/CO	PA/PET	CO/PET	PA/CO	PA/PET	CO/PET
Data	%R	13.72	13.36	14.24	0.36	−0.52	−0.88	0.36	−0.52	−0.88
	L*	44.57	43.89	45.01	0.68	−0.44	−1.12	1.76	1.06	1.50
	a*	14.92	14.34	14.12	0.58	0.80	0.22			
	b*	−13.56	−12.04	−13.02	−1.52	−0.54	0.98			

OBS: %R = reflectance.
L* = lightness axis values.
a* = green-red axis values.
b* = yellow-blue axis values, CIELab system.

Table 3
Ecological costs of dyeing.

Assessed variables per kg		PA	CO	PET
Consumption of water (L)		50	70	40
Energy	Electrical (Q_E)	1.38×10^6 J	2.58×10^6 J	1.64×10^6 J
	Thermal (Q_T)	6.89×10^3 J	7.73×10^3 J	6.27×10^3 J
Fuel consumption (V_1)		1.46×10^{-4} m^3	1.63×10^{-4} m^3	1.33×10^{-4} m^3
Mass of CO_2 (mCO_2)		568×10^{-5} kg	6.37×10^{-5} kg	5.16×10^{-5} kg

industry of clothing [58].

3.2. Ecological costs

The values of consumption of water, electrical energy, and thermal energy and CO_2 emission were presented in Table 3.

The dyeing of PET spent 30 liters less than cotton and 10 liters less than polyamide. Even though, the amount of 40 liters per kilogram is still high, due to the scarcity of clean water around the world [59,60].

Higher amount of electrical and thermal energy was required for dyeing CO in comparison to the others: with a difference of 1.20×10^6 J kg^{-1} and 1.46×10^3 J kg^{-1}, respectively. Obviously, the amount of thermal energy spent in a higher volume of fuel gas, that was 1.77×10^{-5} m^3 higher than PA and 309×10^{-5} J kg^{-1} higher than PES. This accounted for CO_2 emission as 7.77×10^{20} molecules for dyeing of PA, 8.71×10^{20} molecules for dyeing of CO_2 and 7.06×10^{20} molecules for the dyeing of PET. The consumption of fuel gas has generated an emission of high number of CO_2 molecules, being 7.77×10^{20} molecules for dyeing of PA, 8.71×10^{20} molecules for dyeing of CO_2 and 7.06×10^{20} molecules for the dyeing of PET.

In the dyeing of cotton, other class of dyestuff could be used in order to minimize the consumption of energy and water, as demonstrated by Rosa et al. [50]. However, the acute toxicity of the effluent was not assessed by authors.

3.3. Toxicity

The acute toxicity values obtained for PA, CO and PET dyeing effluents were presented at Table 4.

Despite having lower values of electrical energy consumption, the effluent of PA dyeing was relatively of presented higher values of acute toxicity than CO or PET samples. Nowadays, toxicity in textiles, or presented in effluents of dyeings, have been monitored and researched, being one of the principal concern about environmental [31,32,54,55,61,62].

Besides, the growing demand of dyestuff in the textile industry makes it one of the main sources of water pollution problems and, in the last decade, removal of azo dyestuff from wastewater has attracted considerable attention because of the damage it causes to aquatic organisms, and short-term public health damage [55,56].

Considering Brazilian textile industry issues related to safety use of the products many actions have been proposed in order to avoid chemicals residues into the finished clothes, that represent harmful effects to human health. In terms of environmental issues and textile wastewater Brazilian Environmental Council (CONAMA) is in charge of regulations [63].

Table 4
Average acute toxicity of effluents.

Test	CE_{50} (%)		
	PA	CO	PES
1	0.52	4.71	1.83
2	0.85	4.35	1.75
3	0.46	4.67	1.56
media	0.61	4.58	1.71

Table 5
Ecological information about knit piece.

T-shirt	Consumption of			Molecules of CO_2 into atmosphere	Toxicity of effluent (before treatment)
	Water (L)	Energy (J)			
		Electrical	Thermal		
PA	12.5	3.45×10^5	$1{,}72 \times 10^3$	$1.94 \cdot 10^{20}$	High
CO	17.5	6.45×10^5	1.93×10^3	$2.18 \cdot 10^{20}$	High
PET	10.0	4.10×10^5	1.57×10^3	$1.77 \cdot 10^{20}$	High

All the fabrics covered in this study have yield equal to 3.90 m kg^{-1}, making possible a production of four t-shirts, size G (Brazilian measurement). Therefore, each t-shirts dyed in Pantone 19–1619 colour, assessed in this paper, could have a tag with information such as described in Table 5.

4. Conclusions

CONAMA, edited two recommendations for controlling toxicity before discharging industrial effluents, and also controlling organic / inorganic matter into the rivers and water reservoirs [64]. This regulation states that “any effluent from any pollution source is obligated treated before being discharged and only if the quality parameters of receiving system is guaranteed”. Besides, Article 18 states that the effluent cannot cause toxic effects to the to the living organisms in the receptor system or rivers, in order to protect aquatic life.

Concerned about the toxicity of textiles at the global level, the Brazilian Association of Textile Industry (ABIT) formally requested to Brazilian Association of Technical Standard (ABNT) to create a research group in order to study and control the possible chemical residues into the clothes [65].

Furthermore, faced with worldwide concern, a group called Sustainable Fashion Laboratory was formed in 2017, with many Brazilians companies and research institutions. The main objective of this group is creating a tag containing information about toxicity, water and energy consumption, as well as the emission of CO_2 in the atmosphere.

Int this paper, in terms of water consumption, PET was the fiber that presented the lowest value, 40 L kg^{-1}.

Regarding electric energy consumption, PA required the lowest value, being 1.20×10^6 J kg^{-1} less than CO and 2.66×10^5 J kg^{-1} less than PET.

Thermal energy data revealed PET fiber as the less expensive 1.77×10^5 less than PA and 3.09×10^5 less than CO. In this case, thermal energy impacted directly in the emission of CO_2 molecules into the atmosphere. PET emitted 7.10×10^{19} molecules less than PA and 1.65×10^{20} less than CO.

Even though spending more water and energy, the effluent of CO dyeing was relatively less toxic than the others. However, all the effluents were very toxic to *D. similis* exposed for 48 h.

A suggestion of tag is demonstrated in the Fig. 5, in which was used the data from Table 5.

The total energy (electrical + thermal) spent is equivalent to a

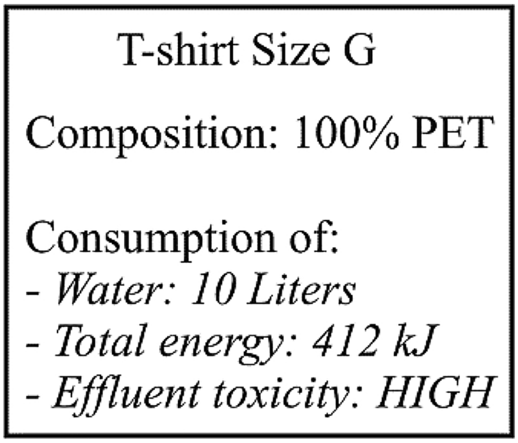

Fig. 5. Tag for 100% PET T-shirt, size G.

shower of approximately 1.5 min, using a 5500 W of potency equipment.

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