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Application of an experimental design methodology for the optimization of cosmetotextile impregnation process by β -cyclodextrin based microcapsules

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Abstract

Neroline-loaded microcapsules based on β -cyclodextrin polyurethane material were fixed on cotton fabric using an impregnation technique. The process parameters were optimized so as to achieve a maximum microcapsules deposition and binding. Simultaneous evaluation and optimization of the effects of interdependent parameters was achieved using an experimental design based on response surface methodology (RSM). The selection of the mathematical model and its use for statistical analysis were done by the least squares method using the Minitab 19 software. The optimal impregnation conditions yielding a fabric mass gain of 8.56 % are as follow: 21 g·L⁻¹ for the microcapsules concentration, 31 g·L⁻¹ for the polyurethane cross-linker concentration and 0.72 g·L⁻¹ for the cationic surfactant concentration. The morphological and mechanical characterizations, and air permeability evaluations of the resulting cosmetotextile fabric show the suitability of the process in terms of applicability and end-use properties.

Keywords

Microcapsules; impregnation process; cosmetotextile; optimization; experimental design; response surface methodology.

Introduction

The modern lifestyle is currently characterized by the production of performance clothing, which is one of the most dynamic and active branches in the textile sector. This type of clothing has a very high added value which makes it possible to offer consumers more functional performance (comfort, safety, aesthetics, ...) (Jocić, 2016). Cosmetotextile products provide various cosmetic activities like insect-repelling, correction of body odors, feeling of well-being, protection, changing appearance, UV protection, anti-ageing, refreshing, moisturizing, slimming, perfuming, ... (Petrušić & Koncar, 2016; Bonaldi, 2018).

Microencapsulation is the preferred technology for the manufacture of cosmetotextiles (Jyothi et al., 2010; Mali et al., 2013). This technique involves the production of microcapsules containing active ingredients inside their core protected by a wall material. By this way, this technology enhances not only the protection of the encapsulated ingredients from hazardous environments and from interactions with other compounds in the system, but also allows sustained release and/or stimuli-triggered release of them, providing long-lasting effects and durability (Buschmann et al., 2015). Cosmetotextiles articles can be prepared using microcapsules through different processes such as incorporation into textile fibers, adsorption on fiber surface or binding microcapsules to fiber surface especially by an impregnation process.

The design of cosmetotextile is facing issues related to the many parameters that influence the functionalization process of textile fabric i.e. the temperature, the treatment duration, the choice of reagents and their concentrations, ... To find optimal conditions for the multi-parameter process, Design of Experiment (DoE) statistical methodology provides a rigorous methodology allowing the determination of the most suitable ingredients, their concentrations and the process parameters (Rezić & Kiš, 2020). Among the various multivariate DoE-based methods, the Box–Behnken Response Surface Methodology (RSM) developed by Box and Wilson (Box & Wilson, 1992) is especially used in the design of experiments for optimization purpose (Javidi et al., 2019; Karimifard & Alavi Moghaddam, 2018; Jaswir et al., 2019). The main benefit of such methodology is the reduction in the number of experimental tests necessary to evaluate the influence of several variables (Zhong & Wang, 2010). RSM has been successfully applied in optimizing different processes such as extraction (Minjares-Fuentes et al., 2014), nanocomposite and self-healing coating (Ebrahiminiya et al., 2018; Khoee et al., 2016), microencapsulation (Jaswir et al., 2019; Ning & Yue, 2019; Zhao et al., 2016; Yingngam et al., 2019), textile pretreatment (Abdelileh et al., 2019), dyeing (Haji, 2017; Ben Ticha et al., 2013), textile effluent (KweinorTetteh & Rathilal, 2021; Jadhav et al., 2015), textile functionalization (Rezić & Kiš, 2020; Salehi et al., 2018), ...

To the authors' best knowledge, there are no published studies on the application of RSM in microcapsules deposition and binding on textile fabric using impregnation process. Therefore, the aim of the present work is to show a demonstrative instance of such an optimization aiming at enhancing the functionalization of pure cotton fabric by impregnation process using neroline-loaded microcapsules based on β -cyclodextrin. The synthesis and properties of these microcapsules have been described in a previous work (Ben Abdelkader et al., 2019). The effect of the composition of the impregnation bath (concentrations of microcapsules, polyurethane cross-linking agent and cationic surfactant) on the fabric mass gain are presently studied.

Modelling and optimization of these experimental conditions are then investigated in order to improve the performances of the impregnation process. Finally, several textile properties are assessed, namely morphology, tensile strength and air permeability.

Experimental part

Materials

Microcapsules based on β -cyclodextrin were synthesized via an interfacial polycondensation method according to our previous published procedure (Ben Abdelkader et al., 2019).

For textile application, a knitted fabric (jersey, 100 % cotton) bleached and mercerized, with a mass of $138 \text{ g}\cdot\text{m}^{-2}$, was used. It was supplied by Esprit Maille (Bouhjar, Tunisia). A polyurethane cross-linking agent (Politex SW/N) was supplied by Prochimica Novarese SPA (Italy). Dodecylamino Diglycidyl Ether of Isosorbide (DoDGEDAS) was synthesized as previously reported (Ben Abdelkader et al., 2016) and used as a cationic surfactant. The application of microcapsules was carried out using a Continuous Hot Air Dryer (Mathis KTF, Switzerland). Polyurethane and surfactant molecules are shown in Figure 1.

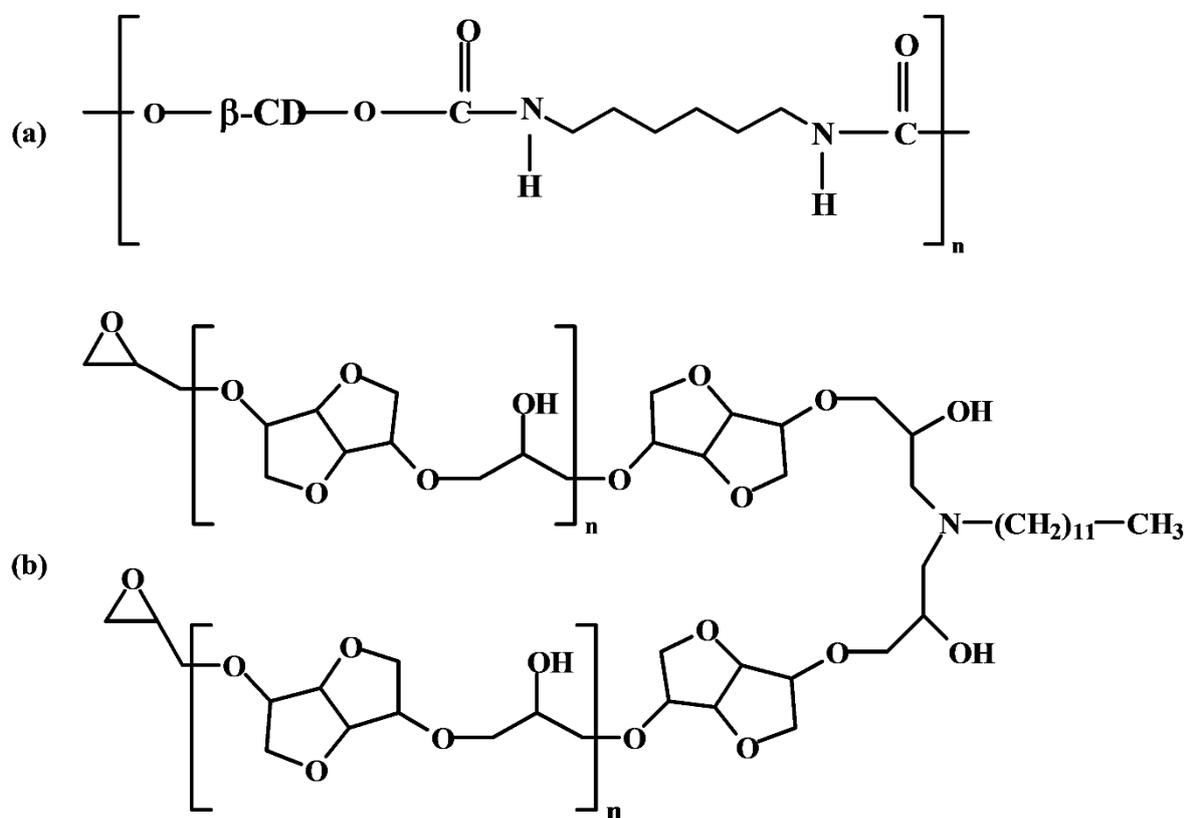


Figure 1. Molecular representation structure of: (a) the polyurethane polymer based on β -cyclodextrin forming the microcapsules shell based on β -cyclodextrin and (b) DoDGEDAS.

General methods and instrumentation

Textile application

Fragrant microcapsules were mixed with a polyurethane cross-linking solution in distilled water containing DoDGEDAS as a cationic surfactant. The cotton fabric was impregnated into this solution under ambient conditions during 20 min. Then the fabric was dried without squeezing during 6 min at $140 \text{ }^\circ\text{C}$ in air at atmospheric pressure to allow the cross-linking agent to harden the deposit.

Mass gain of the fabric

The fabric mass gain G was the ratio of the mass difference of dry fabric after and before the treatment to the initial mass of dry fabric. It was expressed as a percentage using Eq. 1 as follows:

$$G(\%) = \frac{\text{the mass difference of the dry fabric after and before impregnation}}{\text{the mass of the dry fabric before impregnation}} \times 100 \quad (1)$$

Design of experiment (DOE)

Surface Response Methodology (SRM) was used to optimize the impregnation process (Box et al., 2005; D'Agostino & Stephens, 1986; Khuri & Cornell, 1996). The experimental factors studied were the concentrations of the microcapsules (X_1), the polyurethane cross-linker (X_2) and the surfactant (X_3). The investigated experimental domain and the corresponding coded levels (-1, 0 and +1) are defined for each factor in Table 1 (Kumar et al., 2015; Singh et al., 1995; Singh et al., 2008). A Box-Behnken design was built allowing fitting a second-order quadratic model for the prediction of the fabric mass gain G (Box et al., 2005). Fifteen experiments were thus carried out including three center points (Table 2). All experiments were done in triplicate. Multiple linear regression, analysis of variance (ANOVA) and statistical analysis of data were done using the Minitab software (Version 19, State College, PA, USA).

Table 1. Experimental range and levels of independent process variables.

<i>Coded variable</i>	<i>Natural variable</i>	<i>Unit</i>	<i>Levels</i>		
			-1	0	+1
X_1	U_1 : microcapsules concentration	(g·L ⁻¹)	10	20	30
X_2	U_2 : polyurethane cross-linker concentration	(g·L ⁻¹)	20	30	40
X_3	U_3 : surfactant concentration	(g·L ⁻¹)	0.4	0.7	1

Scanning electron microscopy

The residual bath after impregnation, the shape and surface features of untreated and treated textile were observed by Scanning Electron Microscopy (SEM) after drying using a Jeol JCM 5000 (Jeol, Japan) microscope. The samples were examined at room temperature without surface metallization and operating at 5 kV and 10 kV acceleration under moderate vacuum.

Tensile strength

Tensile test of knitted fabrics was carried out based on the ISO 13934-1 standard by using a LLOYD dynamometer under standard conditions (20 ± 2 °C and 65 ± 2 % of relative humidity) after preconditioning all samples in a standardized conditioning room during 24 hours. Measurements were performed in both directions of knitting (column direction and row direction). The specimen dimensions were 200 mm in length and 50 mm in width. Distance between clamps was 50 mm and the movement speed was 10 mm·s⁻¹. Three samples were tested for each type of fabric.

Air permeability

Air permeability of textile fabrics was measured based on the EN ISO 9237 standard by using a FX3300-III instrument (TEXTTEST AG) under standard conditions (20 ± 2 °C and 65 ± 2 % of relative humidity). All samples were preconditioned in a standardized conditioning room during 24 hours. Measurements were carried out under an air pressure of 100 Pa per 100 cm² fabric surface. Tests were repeated three times and the measurements were performed by scanning the entire textile surface on both sides in order to examine the reproducibility of the results. Outcomes were rates of air flow per unit area and time expressed in “mm·s⁻¹”.

Results and discussion

Factors affecting impregnation process

Functionalization of mercerized cotton jersey fabric with neroline-loaded microcapsules based on β -cyclodextrin was studied. The variables were the main operating conditions influencing impregnation process, the microcapsules concentration, the polyurethane cross-linking agent concentration and the surfactant concentration. The effect was the mass gain of the treated textile (G). Throughout this study, each experiment was performed three times and each experimental point is an average of the obtained results. As a preliminary study, the influences of the variables were firstly investigated independently while keeping the other variable constant. Thereafter, a full optimization was performed using a multivariate statistical analysis.

Effect of the microcapsules concentration

The microcapsules concentration was varied between 10 and 40 $\text{g}\cdot\text{L}^{-1}$ for a cross-linking agent concentration of 30 $\text{g}\cdot\text{L}^{-1}$ and a surfactant concentration of 1 $\text{g}\cdot\text{L}^{-1}$. The variation of G (Figure 2) indicates that the mass gain increased with respect to the amount of introduced microcapsules up to 25 $\text{g}\cdot\text{L}^{-1}$. The mass gain declined above this concentration.

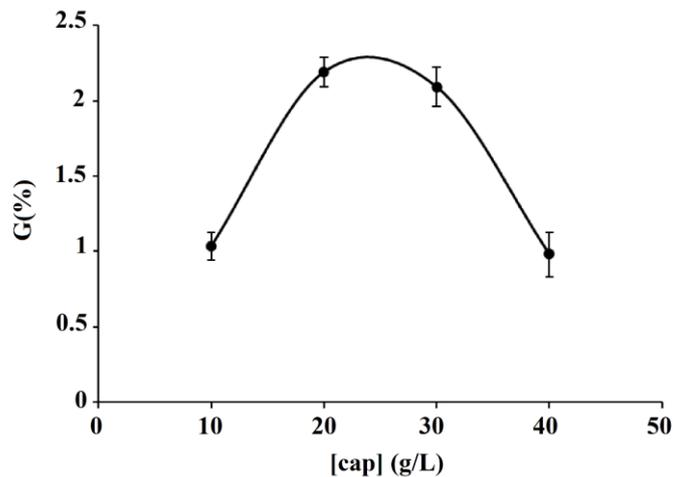


Figure 2. Effect of the microcapsules concentration on the mass gain.

To understand the origin of such loss of mass gain, residual bath initially containing the highest concentration of microcapsules (40 $\text{g}\cdot\text{L}^{-1}$) was analyzed by SEM as shown in Figure 3.

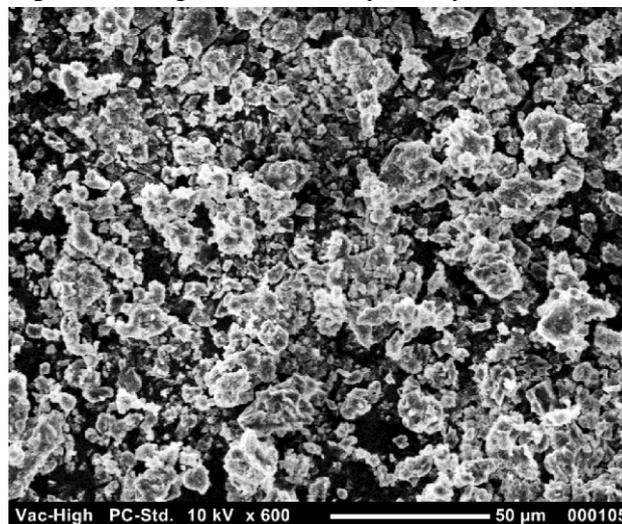


Figure 3. SEM micrograph residual bath initially containing 40 $\text{g}\cdot\text{L}^{-1}$ concentration of microcapsules.

It is clear that microcapsules are agglomerated which cause their sedimentation in the bath. The practical consequence is that there is an optimum. The concentration at optimum may depend of the concentrations of cross-linking agent and surfactant. This is farther addressed in the multivariate analysis.

Effect of the concentration of polyurethane cross-linking agent

For a microcapsules concentration of $20 \text{ g}\cdot\text{L}^{-1}$ and surfactant concentration of $1 \text{ g}\cdot\text{L}^{-1}$, the effect of the polyurethane cross-linker concentration on the mass gain percentage (G) was studied at different concentrations ($20\text{--}50 \text{ g}\cdot\text{L}^{-1}$). As shown in Figure 4, the mass gain increased by varying the concentration from 20 to $30 \text{ g}\cdot\text{L}^{-1}$, reached a maximum and decreased above $30 \text{ g}\cdot\text{L}^{-1}$. The first increase is a trivial result. The decrease above $30 \text{ g}\cdot\text{L}^{-1}$ can be explained by the fact that an excess of polyurethane resin prevents the capsules from penetrating deeply into the knit (in the inter-yarns and inter-fibers) because it forms a film coating the top surface of the fabric.

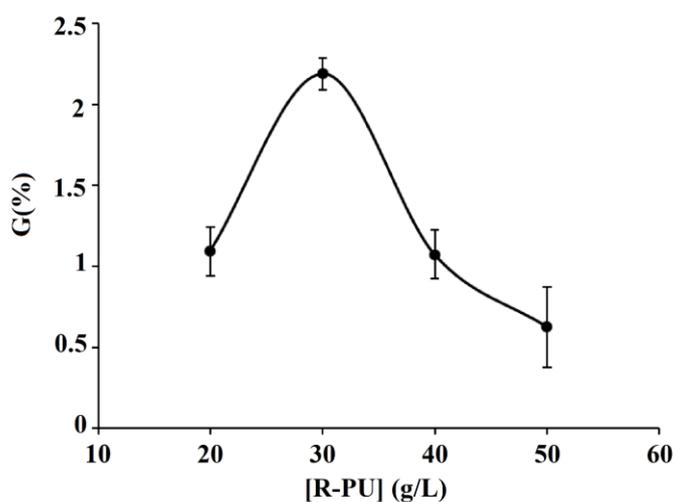


Figure 4. Effect of the polyurethane cross-linker concentration on the fabric mass gain.

Effect of the surfactant concentration

For a microcapsules concentration of $20 \text{ g}\cdot\text{L}^{-1}$ and a polyurethane cross-linker concentration of $30 \text{ g}\cdot\text{L}^{-1}$, the mass gain percentage (G) was assessed for DoDGEDAS concentrations varying between 0 and $1 \text{ g}\cdot\text{L}^{-1}$ (Figure 5). The use of a cationic surfactant contributes at improving the rate of attachment of microcapsules to the textile support. Indeed, the mass gain continuously increased with respect to increasing surfactant concentration. A larger concentration might improve again the mass gain: however, the concentration of surfactant should be kept moderate in order to avoid accumulation of it in the fabric that possibly will cause skin irritation during wearing the fabric.

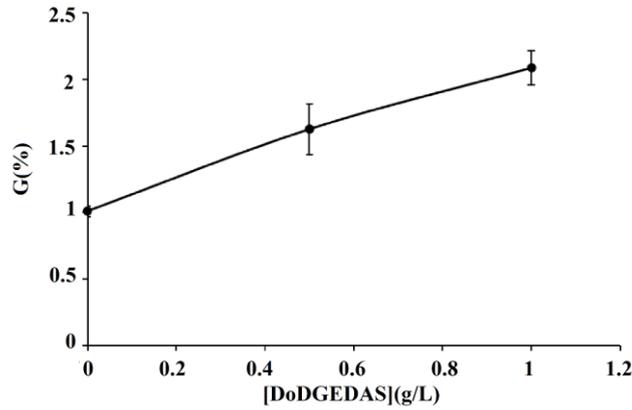


Figure 5. Effect of the surfactant concentration on the mass gain.

Optimization of the impregnation process by RSM

Model validation

Because the preliminary experiments showed that the variations of mass gain with respect to variables was not linear and there is a maximum, a quadratic model has then been postulated to explain the mass gain. For this purpose, the three experimental factors were simultaneously varied according to a Box-Behnken design. Results were investigated using the software Minitab 19 and led to the following equation:

$$G(\%) = 8.48 + 0.73 X_1 + 0.74 X_2 + 0.23 X_3 - 3.74 X_1^2 - 3.46 X_2^2 - 2.63 X_3^2 + 0.32 X_1X_3 + 0.66 X_2X_3 \quad (2)$$

where X_i are the coded factors. All the regression coefficients are significant at the 5 % level (Student t -test). For the regression equation of the gain mass percentage (G) (Eq. 2), it was found that the determination coefficient R^2 is equal to 0.977, which proved the satisfactory adequacy of the developed model. The mass gain ranges from 0.64 % up to 8.50 % depending on the applied operating conditions (Table 2). Each experiment being carried out in triplicate, the 95 % confidence interval of G has been estimated about ± 0.1 % (standard deviation ~ 0.04 %) indicating a high reproducibility of the G measurements.

Table 2. Coded actual levels of studied variables and results obtained for a surface design.

Experiment N°	Coded values			Real values			Mean response
	X_1	X_2	X_3	U_1	U_2	U_3	G (%) (n=3)
1	-1	-1	0	10	20	0.7	0.64
2	+1	-1	0	30	20	0.7	1.19
3	-1	+1	0	10	40	0.7	1.33
4	+1	+1	0	30	40	0.7	1.95
5	-1	0	-1	10	30	0.4	1.15
6	+1	0	-1	30	30	0.4	2.82
7	-1	0	+1	10	30	1.0	0.74
8	+1	0	+1	30	30	1.0	3.70
9	0	-1	-1	20	20	0.4	1.58
10	0	+1	-1	20	40	0.4	2.51
11	0	-1	+1	20	20	1.0	0.96
12	0	+1	+1	20	40	1.0	4.51
13	0	0	0	20	30	0.7	8.46
14	0	0	0	20	30	0.7	8.50
15	0	0	0	20	30	0.7	8.49

Analysis of variance was first used to assess the fitting quality of the developed model (Winer, 1971). The evaluation is based on a F -test. Results of ANOVA in Table 3 proves the high significance of the regression model for the mass gain prediction (Eq. 2) (p -value < 0.01 %).

Table 3. Analysis of variance (ANOVA).

Source	Degree of freedom	Sum of squares	Variance	F	p -value
Regression	8	117.1	14.6	33781	$< 0.01\%$
Residuals	6	2.8	0.5	-	-
Total	14	119.9	-	-	-

To finally assess the validity of the developed model, the residual errors were also determined by calculating the difference between the experimental G and the G predicted by the model (Eq. 2). These residuals errors < 0.8 % indicated that the mass gain G is satisfactorily explained by the regression model.

Surface response

Once validated, the model was used to visualize the surface responses as a bi-dimensional plot of two factors (X_1 and X_2), while keeping the third one (X_3) constant. Figure 6 shows the contour plot of the response surface for G at surfactant concentration of 0.4, 0.7 and 1 $\text{g}\cdot\text{L}^{-1}$.

According to Figure 6, maximum mass gain appears at the center of the three plots and reaches a maximal value of 5.63, 8.55 and 6.30 % for surfactant concentration of 0.4, 0.7 and 1 $\text{g}\cdot\text{L}^{-1}$, respectively. The highest mass gain is obtained when all the concentrations of microcapsules, cross-linking agent and surfactant are fixed at the center of the investigated experimental domain. The shape of the contour plot being quite circular indicates similar effect of X_1 and X_2 on G .

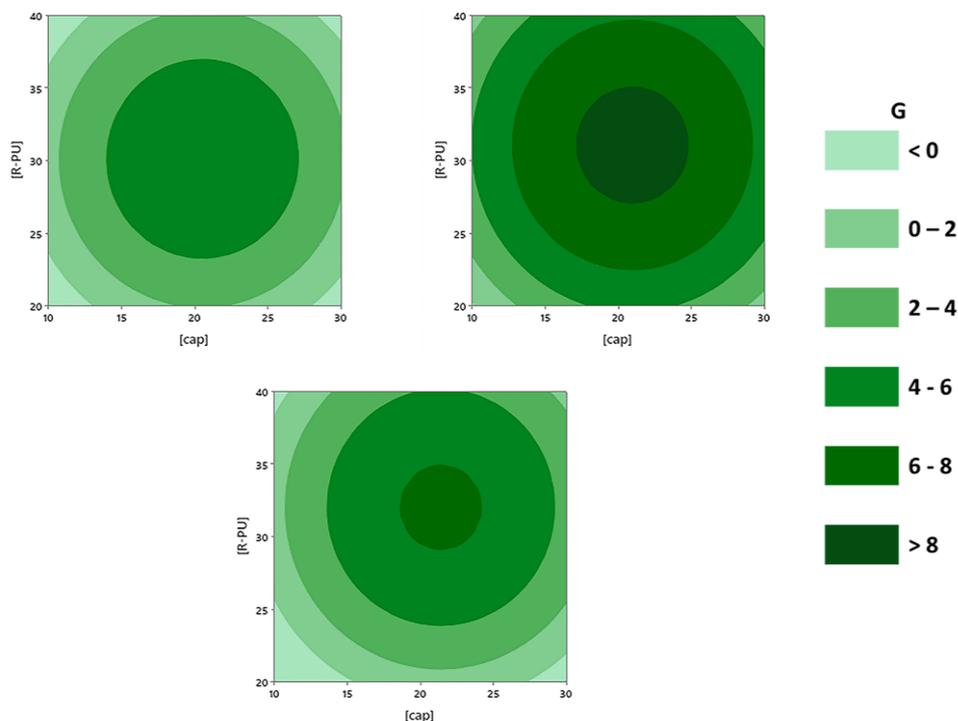


Figure 6. Contour plots showing the combined effect of microcapsules and cross-linker concentrations (b) on the mass gain (G) using different hold values of surfactant concentrations: (a) 0.4 $\text{g}\cdot\text{L}^{-1}$, (b) 0.7 $\text{g}\cdot\text{L}^{-1}$ and (c) 1 $\text{g}\cdot\text{L}^{-1}$.

Response optimization

The optimal conditions of the cotton impregnation process using fragrant microcapsules were predicted by the response optimizer tool of Minitab 19 software for maximum response. Response optimization described in Figure 7 shows that optimal experimental conditions are a microcapsules concentration of $21 \text{ g}\cdot\text{L}^{-1}$, a cross-linker concentration of $31 \text{ g}\cdot\text{L}^{-1}$ and a surfactant concentration of $0.72 \text{ g}\cdot\text{L}^{-1}$, yielding a mass gain of 8.56 %.

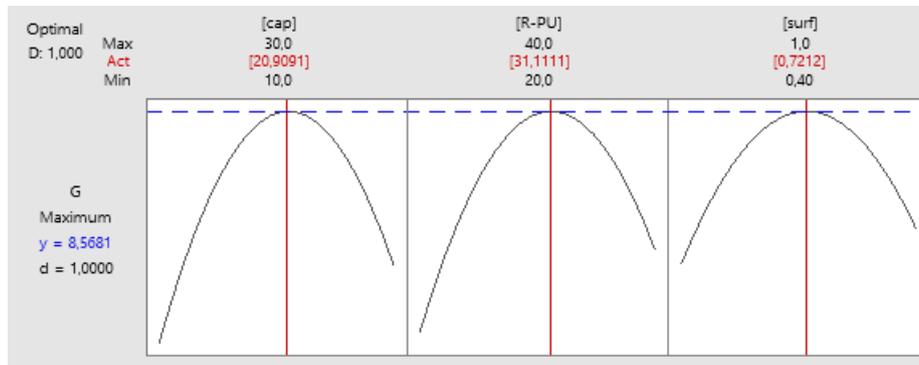


Figure 7. Response optimization of the impregnation process.

To check the validity of the model, experiments were performed to compare the experimental results with those predicted responses using the model equation. The theoretical optimal value of mass gain under these optimized conditions is 8.56 % while the experimental one is 8.61 %. The difference between these two values is well included in the experimental uncertainty thus demonstrating the good predictive performance of the developed model.

Effect of the impregnation process on textile properties

After functionalization of cotton knitted fabric by impregnation process under optimized conditions, several textile properties were studied namely morphology, tensile strength and air permeability.

Morphology

Treated textile was analyzed by SEM after being impregnated in a bath containing fragrant microcapsules, the DoDGEDAS cationic surfactant and the polyurethane cross-linking agent, under optimized conditions. Figure 8 shows SEM micrographs of the untreated cotton knitted fabric compared to the treated one.

It is clear that the β -CDHDI microcapsules are well distributed on the textile surface: between the textile yarns, the fibers and in the cotton cavity. This is due to the use of DoDGEDAS which promotes the dispersion of the microcapsules in the impregnation bath.

Indeed, fragrant microcapsules were well fixed and not altered by the heat treatment carried out (temperature of $140 \text{ }^\circ\text{C}$ for 6 min) since they kept their spherical shape intact. A higher density of microcapsules was also noticed in this case compared to previous works (Azizi et al., 2014; Ben Abdelkader et al., 2018; Azizi et al., 2019; Ben Abdelkader et al., 2013). This is the consequence of optimization. The binder is also found in an adequate quantity as no excess of it could be seen. Thus, two scenarios are possible, depending on the size of the particles. As for the small size microcapsules, they are stuck and completely covered in the polyurethane resin. The latter phenomenon promotes a longer life of the cosmetotextile.

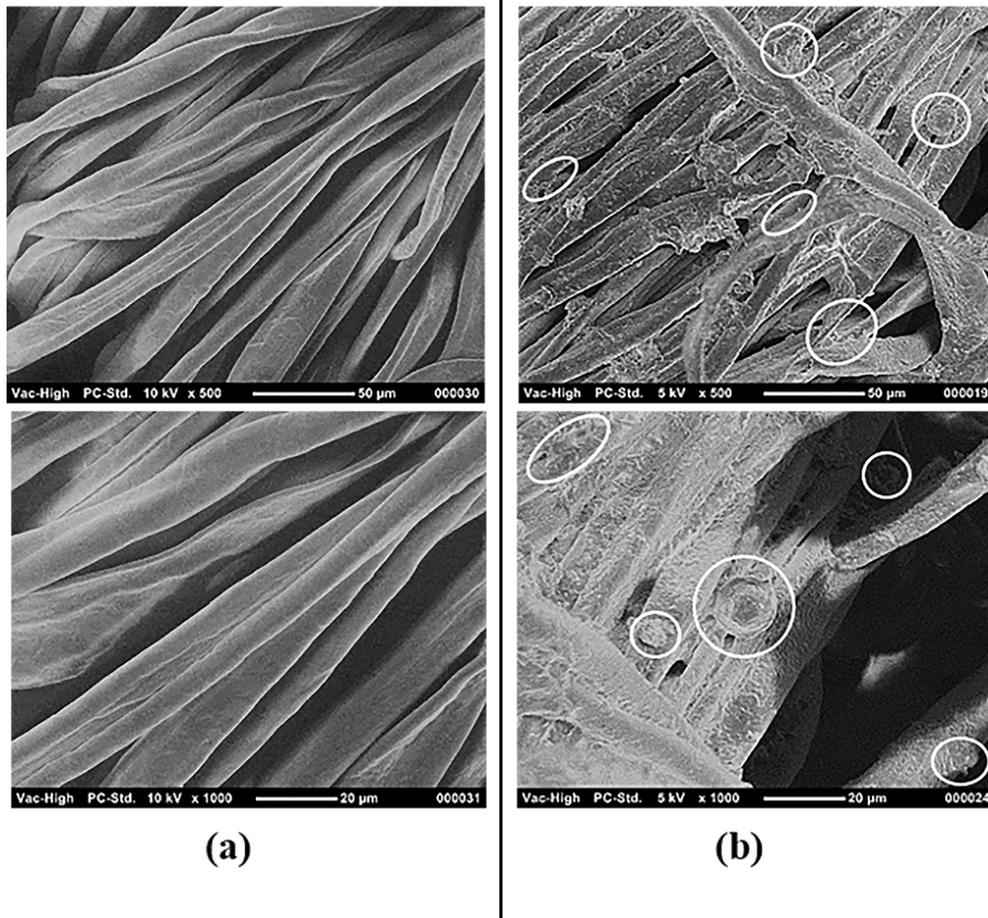


Figure 8. SEM micrographs of: (a) untreated (b) treated cotton knitted fabric.

Tensile strength

The evaluation of the effect of the optimized functionalization treatment on the tensile properties was carried out by a comparison between the tensile curves of the untreated and the treated samples in both knitting directions (row direction and column direction), in particular the maximum force, the elongation at maximum force as well as the rigidity (Figure 9).

The general tensile behaviors of the different knitted fabrics are similar in the two solicitation directions and whether before or after impregnation.

Thus, we distinguish several stages as follows (Szabo et al., 2019):

- The start of the curve corresponds to the resistance to friction between the threads and the fibers.
- The second part of the curve is linear. Significant deformation takes place under low loads. This is mainly due to a change in the configuration of the mesh which elongates in the direction of stress and tapers in the transverse direction while the yarns line up. Meshes slip and friction at the crossings takes place. The shape of the section of the yarn changes and flattens with a reorganization of the fibers under the effect of local compression. The mesh is mainly subjected to bending forces in the yarn. As the load increases, mesh legs continue to line up. The mesh lengthens in the tensile direction but no longer narrows in the transverse direction. Meshes continue their slip and the cross section of the yarn decreases until it reaches a critical mesh extension point at which the stress is transmitted to the fibers.
- The transmission of stress forces to the fibers corresponds to the third step. Thus, the fibers

are stressed in traction at the level of the legs of the meshes and in shear at the level of the crossings.

- The last step corresponds to the rupture which starts by shearing at the intersection of the adjacent meshes causing unraveling throughout the column.

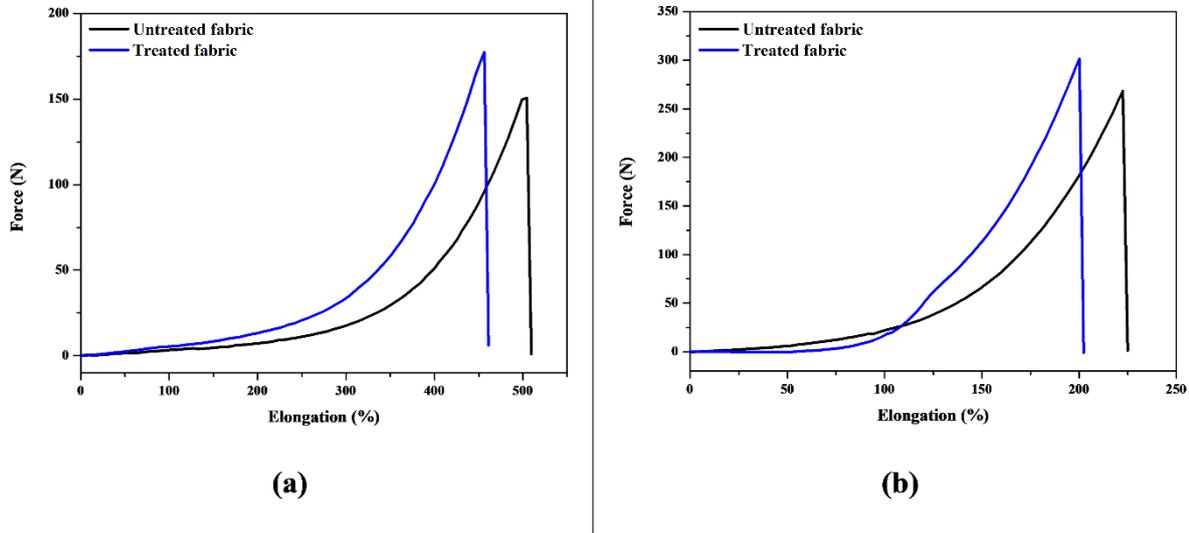


Figure 9. Tensile behavior of untreated, impregnated and treated knitted fabric in both directions: (a) row direction and (b) column direction.

A comparison was carried out between the maximum force, the elongation at maximum force as well as the rigidity of the untreated and the treated samples (Figure 10).

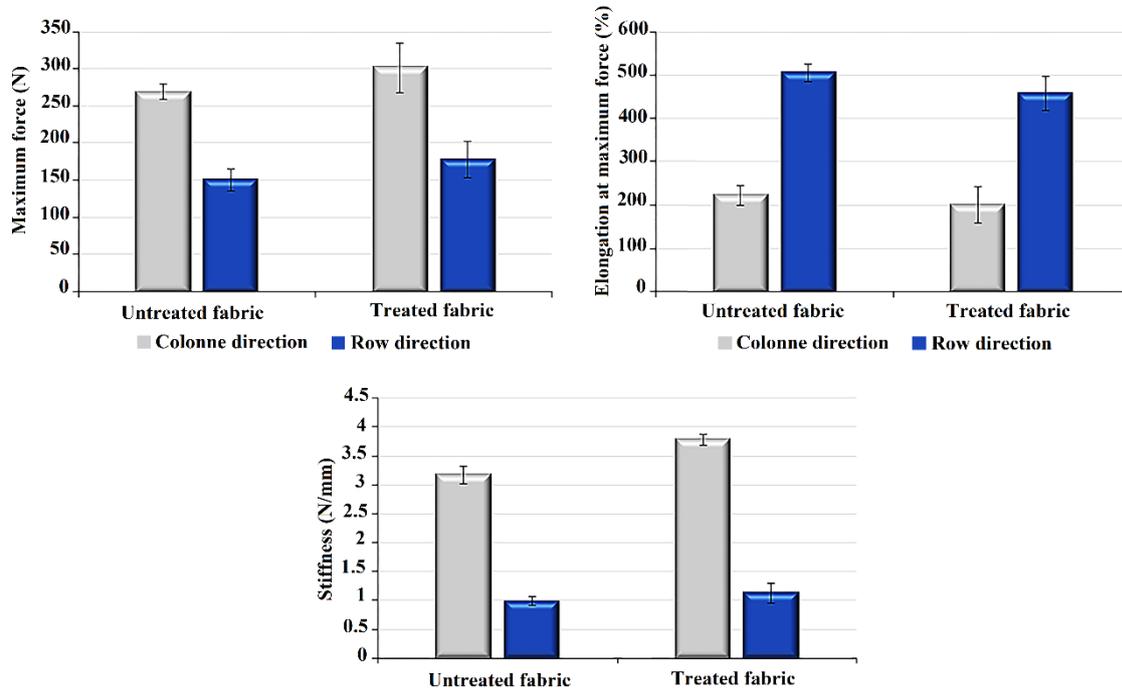


Figure 10. Variation of maximum force, elongation at maximum force and stiffness depending on the impregnation treatment in row and column directions.

The treatment of fixing the microcapsules to the cotton knitwear by the impregnation process increased the maximum force in both directions. Thus, the polyurethane resin coated the cotton

threads and fibers while partially blocking their movements. As a result, the tensile strength was improved (Potocic-Matkovic & Skenderi, 2013). Regarding the elongation at maximum force, a slight decrease in both directions of stress of the impregnated knit was noticed. This is probably due to the fact that the polyurethane resin prevented slippage between yarns and fibers during traction (Potocic-Matkovic & Skenderi, 2013). Figure 10 shows that the impregnation treatment increased the rigidity in the elastic zone. This increase was more accentuated in the column direction than in the row direction. This was probably due to the establishment of new bonds, inter-fibers and inter-yarns, of physical or covalent types by impregnation. According to these results, it appears that the studied impregnation treatment slightly modifies the mechanical properties of the textile without altering them.

Air permeability

The assessment of air permeability was carried out by measuring the speed of passage of air flowing through the fabric under a pressure differential. The evaluation of the influence of the functionalization treatment carried out on the air permeability was carried out by comparing the value of the treated textile with that of the untreated textile.

The air permeability properties of untreated and treated knitted cotton fabrics are displayed in Figure 11. Results shows that the treatment for fixing the microcapsules by the impregnation process decreased the air permeability of the functionalized textile with a percentage of 42 % for a mass gain of 8.61 %. The textile support is not yet saturated, since the porosity is not negligible and there is no excess of the impregnation bath that can form a layer right side up and on the other side of the textile substrate. In fact, microcapsules and cross-linker are distributed inside textile sample in the sites between the thread and in statements between the fibers (Romdhani et al., 2013).

According to Misra et al (2019), textile fabric become more permeable to air with decreasing GSM. That is why low GSM fabric would be more suitable for summer applications and vice versa. In addition, by the decrease of the air permeability from $992 \text{ mm}\cdot\text{s}^{-1}$ to $567 \text{ mm}\cdot\text{s}^{-1}$, there is not a fall of the wearing comfort because the textile before treatment has a very high level of openness of the structure and air permeability (Misra et al., 2020).

The uniformity of microcapsules coating was confirmed by this impregnation process as air permeability was performed at different points of the fabric. All of them presented similar air permeability behavior. The present functionalization technique seems to be efficient in not only promoting a high level of material attachment but also ensuring the coating uniformity (Ghaheh et al., 2017).

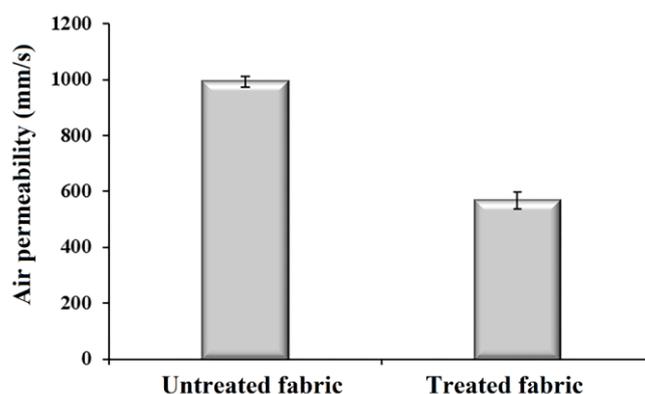


Figure 11. Air permeability of untreated and treated knitted cotton fabrics.

Conclusions

The effect of the composition of the impregnation bath (concentrations of microcapsules, cross-linker and surfactant) on the efficiency of the microcapsules deposition definitely shows sharp optima. This deserves a rigorous methodology of optimization. The use of the response surface methodology (RSM) was aiming at optimization of the fragrant functionalization of pure knitted cotton. Treated fabric was prepared using neroline-loaded microcapsules based on β -cyclodextrin by impregnation process. The concentrations of microcapsules, polyurethane cross-linker and cationic surfactant have been investigated to determine the optimal conditions. Using the developed model, the maximum possible mass gain reached 8.56 %. The optimal impregnation conditions are as follow: $21 \text{ g}\cdot\text{L}^{-1}$ for the microcapsules concentration, $31 \text{ g}\cdot\text{L}^{-1}$ for the polyurethane cross-linker concentration and $0.72 \text{ g}\cdot\text{L}^{-1}$ for the cationic surfactant concentration. Optimization using an experimental design (RMS) definitely makes sense because there is a pronounced maximum of fabric mass gain with respect to the studied variables.

A study of some properties of the cosmetotextiles produced via the optimized impregnation protocol was carried out. SEM micrographs revealed an effective adhesion between microcapsules and cotton fibers. According to tensile strength tests, it appears that the impregnation treatment slightly modified the mechanical properties of the textile without altering them. Air permeability decreased by 42 % after functionalization. Overall, the studied impregnation treatment appears adequate for the development of a wide range of other cosmetotextile articles since it allows the addition of a specific functionality to the textile support without blatantly altering its intrinsic properties.

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