Functionalization of Linen Knitted Fabric with Beeswax/Essential Oil Systems

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The aim of this work was to design a controlled-release beeswax-essential oil systems and applying them on linen knitted fabrics. The active systems were evaluated by optical microscopy and by determining the emulsifying activity index. The treated linen knitted fabrics with active systems were evaluated by SEM analysis and essential oil release was evaluated by UV analysis. The samples treated with emulsions 7 (9.3%w/v beeswax: 18.5% w/v essential oil), 8 (9.3%w/v beeswax: 27.7% w/v essential oil) and 9 (9.3%w/v beeswax: 37.0% w/v essential oil) gave the highest C_{max} value at 4 h, essential oil release depended on the beeswax: essential oil ratio.

Keywords: beeswax, linen, sage essential oil, controlled release

The last few years collaboration of experts from medical and textile sphere conducted to a multitude of innovative applications from the bio functional fabric domain [1-6]. As the healthcare industry is growing enormously, the demand for the healthcare textiles is also on the rise [7-11]. At the moment there is a rising interest towards textile materials with controlled release of biologically active principle [12-15]. Ensuring an optimal therapeutic action depends on the choice of the carrier substance which facilitates penetration by reversible alteration of the skin structure [16]. The most used substances for therapeutic excipients are polyester-based synthetic polymers (poly lactic-co-glycolic acid, poloxamer, polyvinylpyrrolidone ethylcellulose, sodium pyrrolidone carboxylate, povidone, polylactic acid, polyethylene glycol, polyvinyl alcohol), natural-origin polymers (starch, hyaluronate, human albumin, gelatin, alginic acid, collagen, chitosan) [17-19] and lipids (carnauba wax, fatty alcohol, glycerol palmitostearate, stearyl alcohol, beeswax, aluminum monostearate and glycerol monostearate) [20, 21].

Beeswax appear as very attractive due to protection of biologically active compound against chemical degradation, prolonged release of active compound, lower cytotoxicity and the relatively low costs of the excipients [22, 23]. Controlled release delivery is available for many routes of administration. One of them is topical administration of biologically active compound. For topical use the next forms of presenting the compound are used widely: liquid forms (solutions, lotions, emulsions, suspensions, tinctures, sprays, mousses), semi-solid forms (ointments, rigid foams, semi-solid liniments) and solid forms (powders, microspheres, tablets, granulates) [24, 25].

In this study it was used emulsions with possible topical applications. Beeswax was used as carrier and sage essential oil was used as biologically active compound. The prepared emulsions were evaluated by optical microscopy and by determining the emulsifying activity index. Treated linen knitted fabrics with emulsions were analysed by scanning electron microscopy (SEM) and ability to release of active compound.

Experimental part

Materials

Beeswax was procured from a private apiary in the North-East region of Romania. Sage essential oil-extract of *Salvia Officinalis L* purchased from Fares SA Romania. Tween 80 was supplied by Merck, Germany, purity 99.5%. Vegetable glycerine was purchased from SC Elemental SRL, Romania. For the purpose of the study, 100% linen knitted fabric with rib structure was selected.

Obtaining emulsions

Beeswax was melted in water bath at 63°C and appropriate amount of sage essential oil was added dropwise. The mixture was poured into solution containing Tween 80, glycerine and water, under stirring. Stirring process was maintained for 30 min.

The emulsions were obtained using the next treatment variants (table 1).

Characterization of prepared emulsions

The describing of the prepared emulsions was done through optical microscopy and through the calculation of the emulsifier activity index. The prepared emulsions were observed through a KRUSS optical microscope, and the microscopic images were transferred for computerized analysis using a Nikon, Coolpix P5100 camera. A way of assessing the thermodynamic stability of a emulsion is the determining of that emultions interfacial surface (SI), this thing being done using turbimetric measurements. The data of those measurements are at the base of the determining the emulsifier activity index (IAE) which offers in the end information about the stability of the emulsions. IAE was calculated with the following formula:

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Emulsion no.	Beeswax %(w/v)	Gliceryne %(w/v)	Water %(w/v)	Tween 80 %(w/v)	Sage essential oil %(w/v)
1	1.9	18.5	55.5	5.6	18.5
2	1.9	18.5	46.3	5.6	27.7
3	1.9	18.5	37.0	5.6	37.0
4	5.5	18.5	51.9	5.6	18.5
5	5.5	18.5	42.6	5.6	27.8
6	5.5	18.5	33.3	5.6	37.1
7	9.3	18.5	48.1	5.6	18.5
8	9.3	18.5	38.9	5.6	27.7
9	9.3	18.5	29.6	5.6	37.0
10	18.5	18.5	38.9	5.6	18.5
11	18.5	18.5	29.7	5.6	27.7
12	18.5	18.5	20.4	5.6	37.0

Table 1
TREATMENT VARIANTS

$$IAE = \frac{SI}{w} = \frac{2\tau}{C(1-\varphi)} \quad (1)$$

where:
τ - the turbidity of the emulsion;
C - the concentration of emulsifier (g/mL);
φ - the volume fraction of oil in the emulsion.

Coating of the textile with emulsions

The linen knits were immersed in emulsions prepared according to the treated variants presented in table 1, to 100% squeezing degree. The treated samples were dried out at room temperature.

Electron microscopic analysis

The morphology of the sample surface was studied by scanning electron microscopy, carried out with Analytical Scanning Electron Microscopes (VEGA Tescan LMH II), using an SE detector and a 30-kV filament (W) power supply.

Release kinetics regarding the biologically active compound

For the quantitative dosing of the immobilized sage essential oil, the treated knits were characterized by UV spectral analyses by use of CarWin 50 UV-VIS spectrophotometer. The measurements were performed in triplicate at the room temperature, by use of 2mm quartz dishes.

Results and discussions

Characterization of the prepared emulsions

The micro photos of the emulsions prepared according to the 12 treated variants (table 1) are presented in figure 1.

The stability of the emulsions is an important factor in its evaluation. Accordingly to the results presented in figure 1, the uniformity of the drops of essential oil rises proportional to the rise in the concentration of beeswax.

The rise of the concentration of beeswax involves a rise of homogeneity and density of the emulsions. Accordingly to figure 1, the samples treated with 18.5% beeswax have a more homogeneous and denser structure than those treated with 1.9% beeswax.

Research in the emulsifying activity index

Experimental results and the calculated values of the emulsifying activity index (m^2 interfacial surface per gram of emulsifier) and of the interfacial surfaces are presented in table 2 and figure 2. For the determining of the turbidity, it was determined the transmittance, using a CarWin 50 Spectrophotometer.

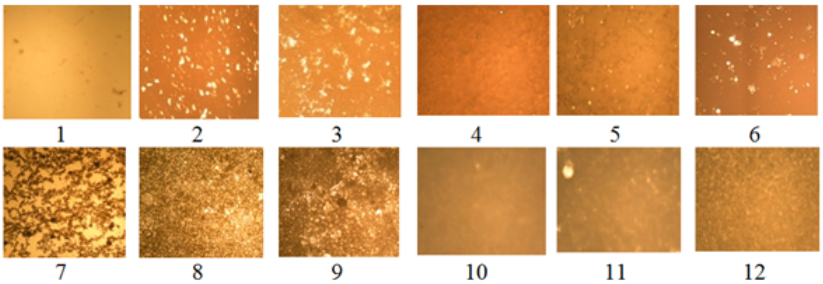


Fig. 1. Photomicrographs of the emulsions

Emulsion no.	φ	τ*	IAC
1	0.185	2.2	64.036
2	0.276	2.5	62.057
3	0.37	2.7	60.750
4	0.185	2.4	69.857
5	0.276	2.6	67.229
6	0.37	2.9	65.250
7	0.185	3.1	90.232
8	0.276	3.3	85.329
9	0.37	3.5	78.750
10	0.185	2.7	78.589
11	0.276	3.1	77.571
12	0.37	3.2	72.000

*The concentration of emulsifier: 0,056g/mL

Table 2
EXPERIMENTAL RESULTS REGARDING THE EMULSIFYING ACTIVITY INDEX (IAE)

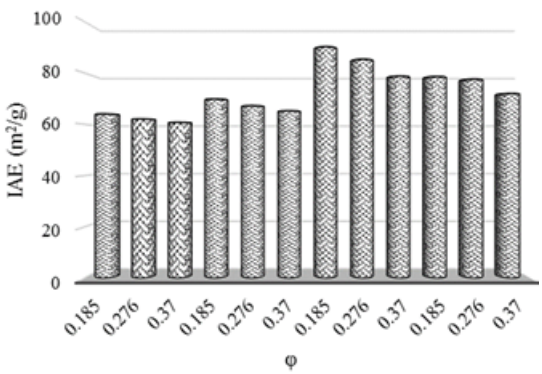
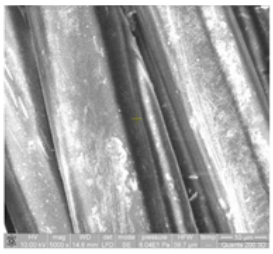
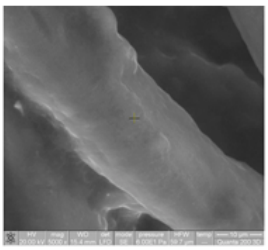


Fig. 2. IAE variation depending on φ



Untreated knitted sample



Knitted sample cover with emulsion

Fig. 3. Electronic images obtained at 5000x magnification

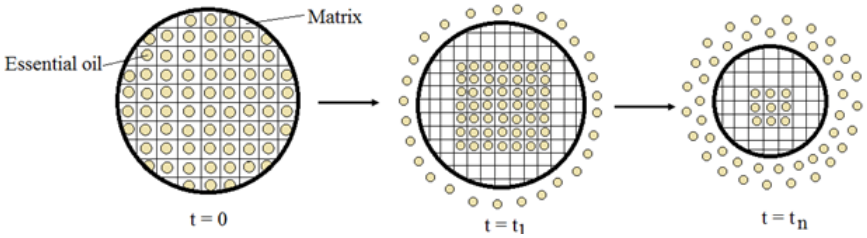


Fig. 4. Essential oil delivery system

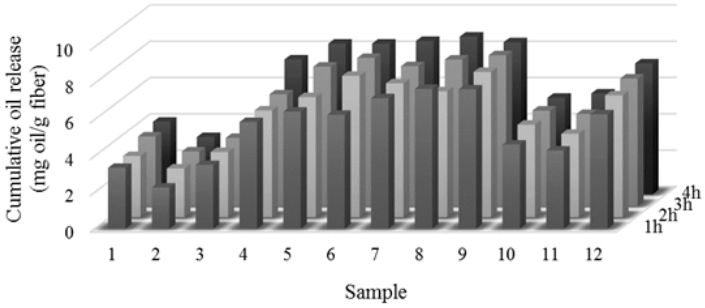


Fig. 5. Cumulative sage essential oil release

In conformity with the experimental data, the turbidity index rises proportionate to the volume fraction of oil in the emulsion; at the same time the emulsifying activity index and the interfacial surface present a minimal value in case of $\varphi = 0.185$.

The stability of the emulsions depends on the interfacial surface. The bigger IAE, the greater is the stability of the emulsion. According to the table 2 results, emulsions 7, 8 and 9 (emulsions with 9.3% beeswax) are the most stable.

Electron microscopic analysis

Because the emulsion with the biggest stability is emulsion 7 (9.3% (w/v) beeswax : 27.7% (w/v) essential oil), a SEM analysis was made for this sample. The SEM analysis of untreated and treated knitted sample with the emulsion 7 is shown in figure 3.

Release kinetic of essential oil

Beeswax is a slowly eroding matrix. Because only surface erosion is very difficult to achieve, essential oil is releasing from beeswax matrix by diffusion and erosion. After contact with physiological medium, essential oil starts to be released by diffusion in surrounding media following by beeswax erosion (fig. 4).

The essential oil release characteristics from the beeswax matrix were studied by UV Spectroscopy. The *in vitro* release was made according to the ISO 105-E04:2008 standard using a sweat solution of *p*H = 5.5, which is the human dermis specific *p*H.

1g of treated fabric was put in a sweat solution (0.05% (m/v) L-histidine monochloride monohydrate, 0.5% (m/v) sodium chloride and 0.22% (m/v) sodium dihydrogen orthophosphate dihydrate) on a liquor-to-material ratio of 30:1. The treated fabric that was put in the sweat solution was kept at 37°C. At regulate time periods, were extracted 5 mL solution, filtrated and exposed to spectrophotometry (at a maximum wavelength specific to sage essential oil used in this study: $\lambda = 236$ nm), to determine the cumulative quantity of essential oil released in time. For keeping a constant solution volume after every extraction of 5 mL of solution, were added 5 mL of sweat solution. For every solution extracted after a t time period it was determined spectrophotometrically the absorbance. The concentrations of essential oil released at the t moment, it was calculated from the calibration curve $y = 303.x$ ($R^2 = 0.999$), where y- absorbance and x- the concentration of sage essential oil.

The profile of the essential oil release from the beeswax matrix for the 12 samples is presented in figure 5.

The release of the sage essential oil from the beeswax matrix was observed for 4h. According to the release profile presented in figure 5, the biggest quantity of sage essential oil was released in samples treated with emulsions 4-9, and the smallest quantity of essential oil was released by samples treated with emulsions 1-3 and 10-12.

Conclusions

In this study, controlled release of sage essential oil was realized successfully utilizing beeswax as a matrix. The emulsions containing 9.3% beeswax shows the most stability. Sage essential oil release rates from linen knitted fabric varied according to the matrix/active compound ratio. With increasing the concentration of beeswax from 1.9% w /v to 9.3% w/v increases the amount of sage essential oil released. The samples treated with emulsions 7, 8 and 9 gave the highest amount of sage essential oil released (8.444 mg, 8.685 mg and respectively 8.376 mg sage essential oil/g linen fiber).

References

1.ALONSO, C., MARTINEZ, M.M.V., RUBIO, L., PARRA, J.L., CODERCH, L. Eur. J. Pharm. Biopharm., **84**, 2013, p. 192.
2.CEREMPEI, A., MURESAN, E.I. CIMPOESUC, N., J. Essent. Oil Res., **26**, 2014, p. 267.
3.CEREMPEI, A., GUGUIANU, E., MURESAN, E.I., HORHOGEA, C., RÎMBU, C., BORHAN, O., Fiber Polym., **16**, 2015, p. 1688.
4.CIOBANU, B.C, CADINOIU, A.N., POPA, M., DESBRIERES, J., PEPTU, C.A., Cell Chem. Technol., **48**, 2014, p. 485.
5.YANG, W.W., PIERSTORFF, E., J. Lab. Autom., **17**, 2012, p. 50.
6.POSHADRI, A., KUNA, A., J. Res. Angrau., **38**, 2010, p. 86.
7.ANCHISI, C., MELONI, M.C., MACCIONI, A.M., J. Cosmet. Sci., **57**, 2006, p. 205.
8.CEREMPEI, A., CIOBANU, L., MURESAN, E.I., MALUTAN, C. BUTNARU, R., Rom. Biotech. Lett., **15**, 2010, p. 5537.
9.PATIL, P., BHOSKAR, M., I.J.C.P.R., **6**, 2014, p. 7.
10.MORGANTI, P., PALOMBO, M., TISHCHENKO, G., YUDIN, V.E., GUARNERI, F., CARDILLO, V.E., DEL CIOTTO, P., CAREZZI, F., MORGANTI, G., FABRIZI, G., Cosmetics, **1**, 2014, p. 140.
11. DEY, N.S., MAJUMDAR, S., RAO, M.E.B., Trop. J. Pharm. Res., **7**, 2008, p. 1067.

12.CEREMPEI, A., MURESAN, E.I., SANDU, I., CHIRILA, L., SANDU, I.G., Rev. Chim. (Bucharest), **65**, no. 10, 2014, p.1154.
13.SOUZA, J.M., CALDASA, A.L., TOHIDIB, S.D., MOLINAC, J., SOUTOB, A.P., FANGUEIROB R., ZILLE, A., Rev. Bras. Farmacogn., **24**, 2014, p. 691.
14.JYOTHI, S., SEETHADEVI, A., PRABHA, K.S., MUTHUPRASANNA, P., PAVITRA, P., Int. J. Pharm. Biol., **3**, 2012, p. 509.
15.MARTEL, B., MORCELLET, M., RUFFIN, D., VINET, F., WELTROWSKI, L., J. Incl. Phenom. Macro., **44**, 2002, p. 439.
16.RAJENDRAN, A., KUMAR BASU, S., Trop. J. Pharm. Res., **8,** 2009, p. 433.
17.MANSOUR, H.M., SOHN, M.J., AL-GHANANEEM, A. DELUCA, P.P, Int. J. Mol. Sci., 2010, **11**, p. 3298.
18.JANTHARAPRAPAP, R., STAGNI, G., Int. J. Pharm., **343**, 2007, p. 26.
19.GHOSAL, K., RAJAN, R., NANDA, A., Int. J. Chem.Tech. Res., **1**, 2009, p. 1128.
20.RANJHA, N. M., KHAN, H., NASEEM, S., J. Mater. Sci., **21**, 2010, p. 1621.
21.RAMYA, K., MAHESHWARI, V., J. Textile Sci. Eng., **3**, 2013, p. 1.
22.AKTER, S., E.S.J., **10**, 2014, p. 488.
23.SHIVARAJ, A., SELVAM, R.P., MANI, T.T., SIVAKUMAR, T., Int. J. Pharm. Biomed. Res., **1**, 2010, p. 42.
24.LUPULEASA, D., BELU, I., SEVASTRE, A.S., MITU, M., Îndreptar practic pentru prepararea medicamentelor, Vol I Ediþia a II-a, Editura Medicalã Universitarã Craiova, 2009, p. 171.
25.GAUR, P.K., MISHRA, S., BAJPAI, M., J. Food Drug Anal., **22**, 2014, p. 542.

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