

Experimental Study of the Thermoregulating Properties of Nonwovens Treated with Microencapsulated PCM

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Abstract

This paper reports a study on the thermoregulation properties of PCM nonwovens. Microencapsulated n-alkanes (n-octadecane and n-eicosane) dispersed in a polymer binder (acrylic-butadiene copolymer) were applied to needled and hydroentangled nonwovens by the pad-mangle or screen printing method. The surface morphology and cross section of PCM nonwovens were observed by means of scanning electron microscopy (SEM). The thermal storage/release properties of the nonwoven samples treated with microPCMs were analysed by DSC, and the thermal resistance of the nonwovens under steady state conditions was determined by means of a sweating guarded hotplate instrument. The transient thermal performance of the nonwoven samples containing microPCMs was examined using novel apparatus with a dynamic heat source. The temperature regulating factor (TRF), defined by Hittle, was determined for selected cycle times of heat flux changes (t), the results of which are shown in diagrams presenting the relation $TRF = f(t)$. The results obtained show that the main factor determining the TRF value is the amount of latent heat in a unit area of nonwoven fabric. Thermoregulating properties of the printed nonwoven sample with microPCMs are identified as being dependent on the position of the microPCMs layer. This work shows the possibility of achieving a significant thermoregulation effect even with moderate amounts of microPCMs incorporated at a proper location in the nonwoven system.

Key words: phase change material, temperature regulating factor, intelligent textiles, thermoregulation properties.

Introduction

A phase change is the process of going from one physical state to another. Phase change materials (PCMs) are those that can absorb, store and release large amounts of energy, in the form of latent heat, over a narrowly defined phase change range, during which the material changes state. PCMs use chemical bonds to store and release heat. When the melting temperature is reached during the heating process, a phase change from solid to liquid occurs, in which the PCM absorbs a large amount of latent heat from the surrounding environment. Energy is absorbed by the material and is used to break down the bonding responsible for the solid structure. This heat is then stored in the PCM and subsequently released in a cooling process starting at the PCM's crystallisation temperature. The latent heat is released to the surroundings when the material cools down. During the entire phase change process, the temperature of the PCM as well as the surrounding substrate remains constant. When the phase change is complete, continued heating/cooling results in a further temperature increase/decrease.

Well-known PCMs are linear chain hydrocarbons known as paraffin waxes (or n-alkanes), hydrated salts, polyethylene glycols (PEGs), fatty acids and a mixture or eutectics of organic and non-organic compounds. Currently, crystalline alkyl

hydrocarbons are used exclusively for textile applications, due to their large latent heat, good thermal and chemical stability, low vapour pressure and self nucleating behaviour [1].

Since 1987, PCMs have also been used as a core material in microcapsule production. Microencapsulated phase change materials (referred to as microPCMs) can be incorporated into textile structures to produce fabrics of enhanced thermal properties. Several methods of microPCMs incorporation into a fibrous structure have been developed. In present applications of PCM technology in the textile industry, for garments and home furnishing products, microencapsulated PCMs are incorporated into acrylic fibers [2] or polyurethane foams [3] or are embedded into a coating compound and topically applied to a fabric [4]. All common coating processes, such as knife over roll, knife over air, screen-printing, gravure printing, and dip coating may be adapted to apply the PCM microcapsules dispersed throughout a polymer binder to a fabric. The conventional pad – mangle systems of applying PCM microcapsules to fabrics are also suitable.

Fabrics thus treated are called “PCM treated fabrics” or “PCM fabrics”. A PCM fabric can act as a transient thermal barrier against cold or hot environments. When a PCM fabric is subjected to a hot environment, the PCM will absorb this

transient heat during the phase change, which will prevent the temperature of the fabric from rising at the melting point of the PCM. This phase change produces a temporary cooling effect in the clothing layers. In a similar manner, when a PCM fabric is subjected to a cold environment, the PCM releases the heat stored, and a temporary warming effect occurs in the clothing layers. This heat exchange produces a buffering effect in clothing layers, minimising changes in skin temperature. The active thermal insulation effect of PCM results in a substantial improvement in the garment's thermo-physiological wearing comfort.

A number of experimental conditions should be optimised before the methods mentioned are developed to produce fabrics with the structure and properties desired to meet practical applications. Most works in this field are in patent literature [2 - 6] and only a few papers in open literature [7 - 16] report the formulation of PCM microcapsules, the finishing of fabrics and the evaluation of their characteristics, including their thermal properties and durability.

Most research has concerned the effects of the thermal properties of PCM materials on wearing comfort. McCullough and Shim [11] measured the effect of layers of PCM clothing materials on reducing the heat loss or gain using PU foam containing 60% microPCMs. The results

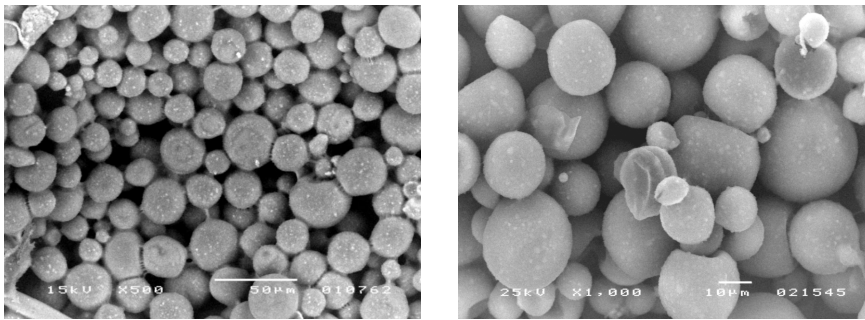


Figure 1. SEM photographs of *n*-alkanes microcapsules; a - TY83 (core material: *n*-octadecane), b - TY95 (core material: *n*-eicosane).

Table 1. Characteristics of the microcapsules.

Symbol	Core material	Density of microcapsules, g/cm ³	Solid-liquid change		Liquid-solid change	
			Temperature T _m , °C	Enthalpy ΔH _m , J/g	Temperature T _c , °C	Enthalpy ΔH _c , J/g
TY83	<i>n</i> -octadecane	0.88	30.637	208.765	21.232	208.416
TY95	<i>n</i> -eicosane	0.90	38.292	182.473	30.340	183.229

Table 2 Characteristics of basic nonwovens; ^a determined according EN 31092, under steady state conditions.

Nonwoven code	Description	Area weight, g/m ²	Thickness, mm	Air permeability (100 Pa), mm/s	Thermal resistance ^a R _t , m ² K/W
41/0	hydroentangled, 100% PET	97	0.78	1114	0.013
42/0	hydroentangled, 100% VIS	179	0.83	241	0.013
43/0	hydroentangled, 100% PET	192	1.02	373	0.017
44/0	hydroentangled, 100% VIS	107	0.68	578	0.020
45/0	needled, 100% PET	264	2.64	1057	0.051

Table 3. Characteristics of the nonwovens treated with microPCMs.

Nonwoven code	Area weight, g/m ²	Percentage of microPCMs (by weight), %	Thickness, mm	Air permeability (100 Pa), mm/s
Padded				
NP41/95/1	133	24.3	0.78	548
NP41/95/2	182	26.1	0.83	310
NP42/95	272	24.1	0.86	57
NP43/95	255	19.9	1.15	202
NP44/95	168	26.7	0.69	135
NP45/83/1	337	14.6	3.17	783
NP45/83/2	376	23.0	3.19	675
NP45/83/3	383	26.5	3.19	660
Printed				
D41/83	189	32.4	0.90	285
D42/95	237	19.5	0.96	35

Table 4. Thermal properties of padded nonwovens; ^a determined according EN 31092, under steady state conditions.

nonwoven code	Thermal resistance ^a R _t , m ² K/W	Latent heat accumulated in 1 m ² of nonwoven, kJ/m ²	Temperature regulating factor at cycle time = 900 s, TRF ₉₀₀
NP41/95/1	0.012	6.01	0.782
NP41/95/2	0.012	12.00	0.514
NP42/95	0.009	11.90	0.663
NP43/95	0.010	9.27	0.736
NP44/95	0.017	8.17	0.733
NP45/83/1	0.045	10.28	0.505
NP45/83/2	0.043	18.04	0.408
NP45/83/3	0.043	21.22	0.379

indicated that the heating and cooling effects lasted approximately 15 minutes, and the heat released depended on the number of PCM layers, their orientation to the body and the amount of body surface area covered by PCM garments. Kim and Cho [12] carried out wear trials of PCM garments and found that changes in the mean skin and microclimate temperature with PCM garments were less than those with non-PCM garments. Ghali et al. [13] analysed the effect of microPCMs on the thermal performance of fabric during periodic ventilation. Their results indicated that microPCMs in fabric cause a temporary heating effect when subjected to a sudden environmental change. Ying et al. [14] found that the rate of temperature increase of a garment with a higher PCM add-on level was lower than that of a garment with less PCM. Yoo et al. [15] studied the effect of PCMs concentration on the temperature of the air layers of a garment when the environmental temperature changes. They stated that in the case of a multilayered garment system, the effect of PCM varied according to the layer, heat gain and heat loss in the outermost layer and had to be taken into account.

The objective of this study was to assess and compare the thermal properties of nonwoven fabrics containing micro-PCMs incorporated into the pad-mangle and screen printing process.

To assess the transient thermal properties of the nonwoven, the temperature regulating factor (TRF) was determined in accordance with the ASTM standard test method [17]. The TRF was defined by Hittle and André [18] as the quotient of the amplitude of the temperature variation and the amplitude of the heat flux variation divided by the value of the steady state thermal resistance of the fabric:

$$TRF = \frac{(T_{max} - T_{min})}{(q_{max} - q_{min}) R}$$

The TRF is a dimensionless number varying in a range (0, 1). The TRF shows how well a fabric containing microPCMs moderates the hot plate temperature. A TRF value of 1 means the fabric has no heat capacitance and poor temperature regulation. A TRF equal to zero means that fabric has an infinite heat capacitance and that a body in contact with it will remain at a constant temperature. It is obvious that all fabrics fall somewhere between these extreme values.

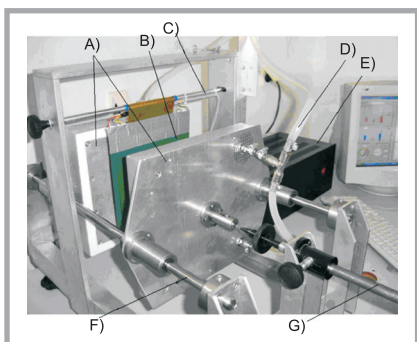


Figure 2. General view of the apparatus to determine temperature regulating factor (TRF); A) cold plates, B) hot plate, C) sample holder, D) cooling water supply to cool plates, E) thermostat, F) guide bars, G) cold plates pressure adjustment.

The TRF is a function of the frequency of the sinusoidal variation of the heat flux in the hot plate. The temperature regulation increases with an increasing frequency. Hence, the TRF increases with an increasing cycle time of the sinusoidal variation, rising exponentially from 0 to 1.

Experimental

Materials

MicroPCMs denoted as the TY83 (core material: n-octadecane) and TY95 (core material: n-icosane) from the Frisby Technologies were used. The average diameter of microcapsules was 15–60 μm . The core of the microcapsules constituted 60–85% of the particle volume, and the

polymer shell was $\sim 1 \mu\text{m}$ thick. **Table 1** shows characteristics of the microcapsules. Photographs of the microcapsules, taken with SEM (JSM 820, Jeol), are presented in **Figure 1**.

Preparing the nonwoven samples

Microcapsules containing PCM were applied to four nonwovens. **Table 2** shows characteristics of these nonwovens.

MicroPCMs were dispersed in an aqueous solution of surfactant, dispersant, an antifoaming agent and a thickener mixture, followed by dispersion in a polymer binder (acrylic-butadiene copolymer). This composition was used to treat the hydroentangled nonwovens and needed nonwovens. The composition was applied to the samples of nonwovens according to the pad-mangle technique or screen-printing technique. Then the samples were dried at 80 $^{\circ}\text{C}$ and next cured of the binder at 110 $^{\circ}\text{C}/5$ minutes.

Table 3 shows basic properties of the nonwovens tested.

Testing methods

Photomicrographs of the nonwoven surface and cross-section were taken with a scanning electron microscope (JSM 820, Jeol) to examine the nonwoven structures visually.

The thermal, storage/release properties of the nonwoven samples treated with

microPCMs were analysed by DSC (Pyris DSC6, Perkin-Elmer). The Melting T_m and crystallisation T_c temperatures, as well as the heat of melting ΔH_m upon heating and heat of crystallisation ΔH_c upon cooling of the specimens were measured in one heating and cooling cycle, with the DSC run at a heating and cooling rate of 5 $^{\circ}\text{C}/\text{min}$. The temperature range of the cycle was 0–50 $^{\circ}\text{C}$. By calculating the peak area of the DSC heating curve, the heat of melting ΔH_m was obtained. The heat of melting ΔH_m in J/g was multiplied by the nonwoven area weight in g/m^2 to give the latent heat accumulated in 1 m^2 of the nonwoven sample.

The thermal resistance of the nonwovens under steady state conditions was determined by means of a sweating guarded hotplate instrument in accordance with PN EN 31092 [19], the results of which are given in **Table 2** (untreated nonwovens) and **Table 4** (padded nonwovens).

Determination of the temperature regulating factor (TRF) of apparel fabrics was done by means of an instrument with a dynamic heat source, described in an earlier paper [20]. A general view of the instrument is presented in **Figure 2**.

This instrument simulates the following arrangement: skin – apparel – environment. The fabric sample is sandwiched between a hot plate and two cold plates,

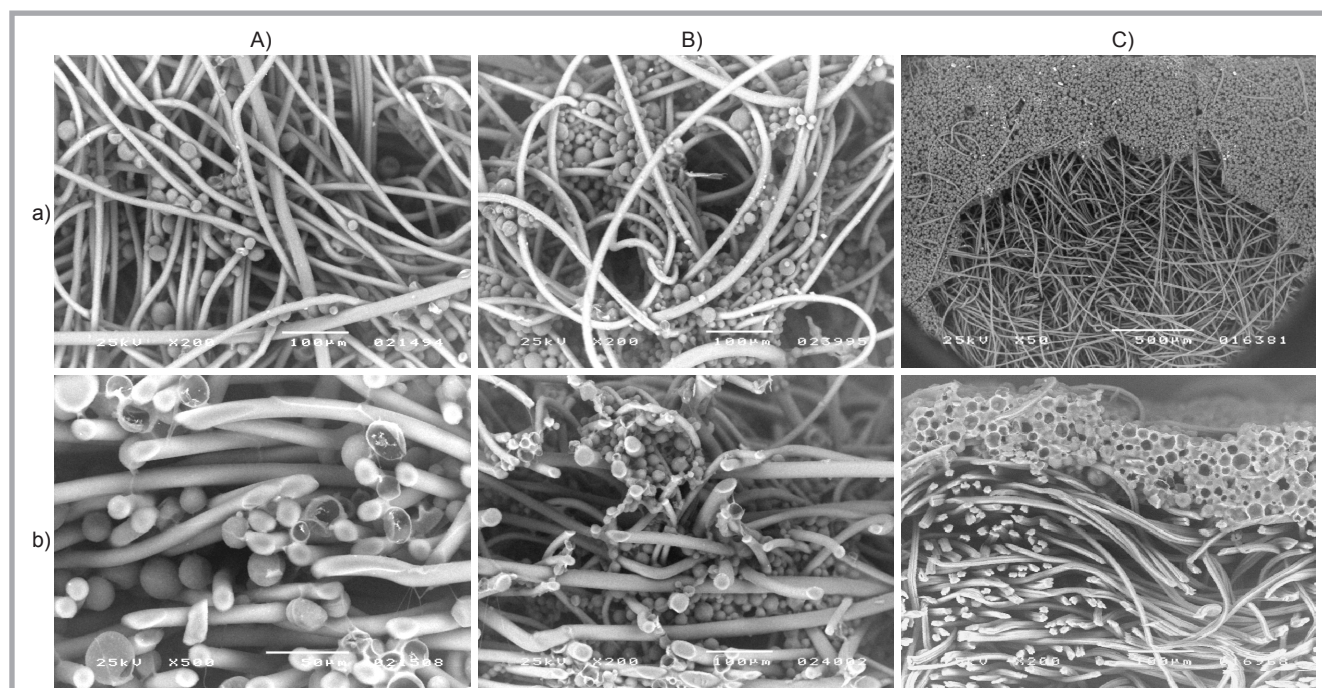


Figure 3. Microphotographs of hydroentangled nonwoven with microPCMs incorporated by pad-mangle technique (A - sample NP42/95), B - sample NP45/83/2) and by screen printing technique (C - sample D42/83); a - surface of nonwoven; b - cross-section of nonwoven.

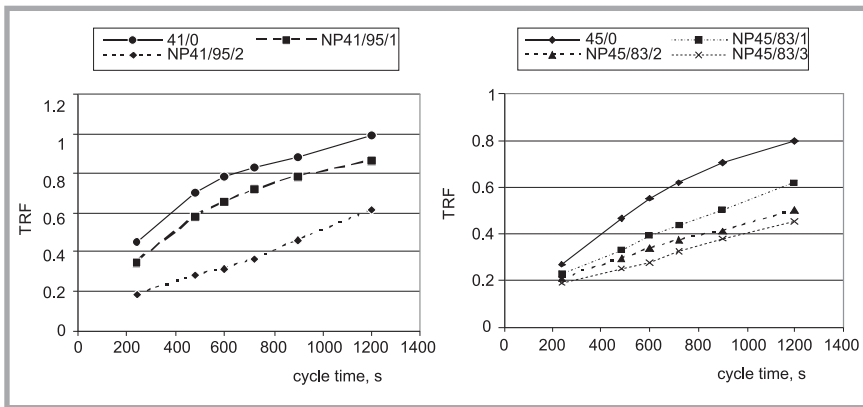


Figure 4. TRF as function of cycle time for padded nonwovens; 83 denotes microcapsules containing octadecane, 95 denotes microcapsules containing eicosane; NP41/95/1 – 24.3 % PCM; NP41/95/2 – 36.1% PCM; 41/0 – 0 % PCM, NP45/83/1 – 14.6% PCM; NP45/83/2 – 23.0% PCM; NP45/83/3 – 26.5% PCM; 45/0 – 0 % PCM; a - Hydroentangled nonwovens, b - Needled nonwovens.

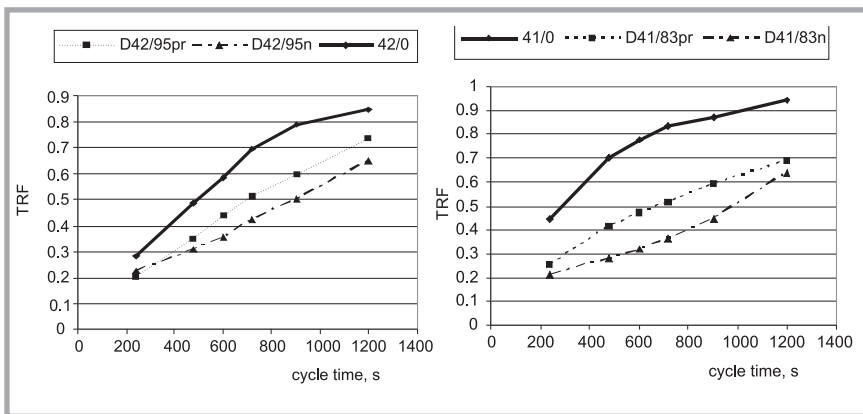


Figure 5. TRF as function of cycle time for printed nonwovens. pr – printed side in contact with hot plate; n – non printed side in contact with hot plate.

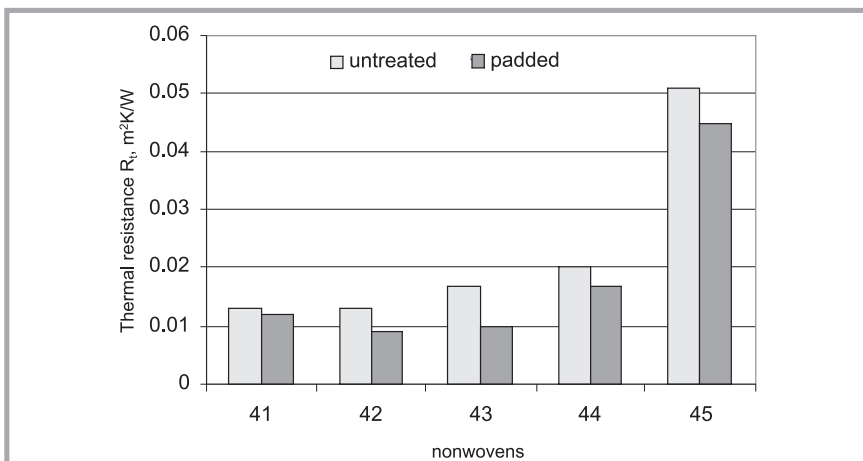


Figure 6. The effect of the applying of micro-PCMs by pad-dry-cure method on the nonwoven thermal resistance R_1 (based on the result from Table 2 and 4).

one on either side of the hot plate. These cold plates at a constant temperature simulate the environment outside the apparel. The sinusoidally varying heat input to the hot plate simulates human activity. To measure the steady state thermal resistance of the fabric, the controlled heat flux is constant, and the test proceeds un-

til a steady state is reached. To assess the temperature regulating ability, the heat flux is varied sinusoidally with time, and the temperature regulating factor (TRF) is determined.

While determining the TRF value, the following measurement parameters were

taken: a medium heat flux of 80 W/m^2 and an amplitude of the heat flux change of 30 W/m^2 . The temperature of the cold plate was selected in such a range that the temperature changes of the hot plate might fluctuate within the phase change temperature range of the microPCMs applied. The TRF was determined for 6 selected frequencies of heat flux changes (t): 240, 480, 600, 720, 900 and 1200 seconds. Three nonwoven samples were subjected to the test. The results of TRF measurements are shown in diagrams presenting the relationship $\text{TRF} = f(t)$.

Results and discussion

Figures 3 (see page 89) present microphotographs of the nonwoven surface and cross-section. Studying these photomicrographs, it was noted that in the samples of nonwovens prepared by the pad-mangle method (Figures 3.a & 3.b, see page 89), microPCMs fill up the interstices between fibers with the binder. MicroPCMs occur in the entire cross section of the nonwoven tested.

In the samples of nonwoven prepared by the screen-printing technique (Figure 3.c, see page 89), microPCMs occur on one side only, forming a very thin layer of $0.10 - 0.14 \text{ mm}$. The microcapsules – binder layer covers most of the nonwoven surface.

Differences in the nonwoven structure and microPCMs distribution observed can be the reason that nonwovens of similar or near values of latent heat per unit area vary in their thermoregulating properties.

Curves illustrating the relation $\text{TRF} = f(t)$ for padded nonwovens are shown in Figure 4, while Figure 5 shows curves for printed nonwovens. The same diagrams also show, for the sake of comparison, the $\text{TRF} = f(t)$ of reference nonwovens i.e. not containing micro-PCMs.

Analysing these diagrams, we can notice that all nonwovens with microPCMs exhibit a lower TRF value in the whole range of frequencies of heat flux changes when compared to the reference nonwovens. This is due to the increase in nonwoven thermal capacitance resulting from the incorporation of microPCMs. The results in Table 3 indicate that the nonwoven samples tested differ in the microPCMs mass contained in one square meter of the nonwoven, which means they differ in thermal capacitance.

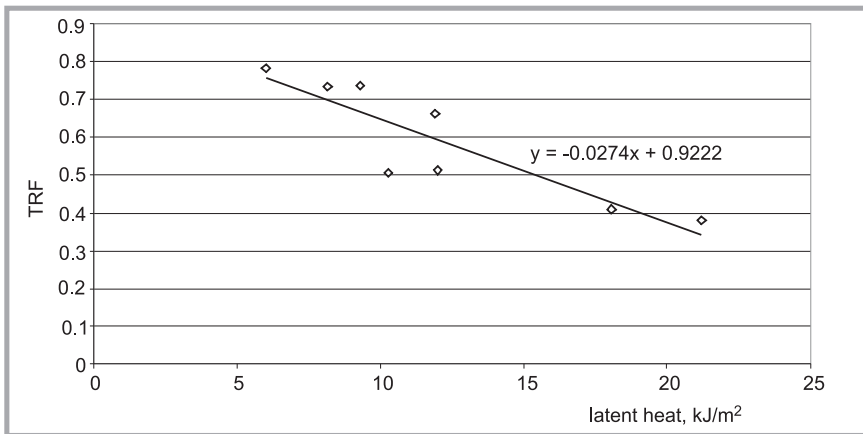


Figure 7. TRF as a function of latent heat accumulated in 1 m² of padded nonwoven.

Results of measurements of nonwoven thermal properties (i.e. thermal resistance under steady state conditions, latent heat accumulated in 1 m² of nonwoven, and TRF determined at a cycle time = 900 s) are listed in **Table 4**.

Analysing the results in **Tables 2** and **4**, it should be noted that there is a reduction in the thermal resistance of nonwovens resulting from the incorporation of microPCMs into the nonwoven structure. In the case of nonwoven without PCMs, there is a greater amount of trapped air, which has a very low thermal conductivity and is therefore a good insulator. Due to applying microcapsules to the nonwoven structure, which are dispersed through the polymer binder, the proportion of air is reduced, leading to a lowering of thermal resistance under steady state conditions (**Figure 6**).

In order to analyse the relation between the amount of latent heat and the TRF, data concerning nonwoven samples treated by the pad-mangle method are presented in **Figure 7**. It can be observed that with an increase in nonwoven heat accumulation, there occurs an improvement in thermoregulating properties defined by the lower TRF value.

Analysing the values of results obtained for nonwoven samples with microPCMs incorporated by the screen printing technique (**Figure 4**), it should be noted that the positioning of the surface covered with the microPCMs layer towards the hot plate has a significant influence on the TRF value. When a printed surface comes into contact with the hot plate, TRF values are higher than in the case where a non-printed surface touches the hot plate.

These results are in agreement with work by Lamb and Dufy-Morris [21]. In their study of the heat transfer process through fabric systems containing phase change material, they found that latent heat effect occurred only when the PCM is located below the outer surface of the fabric system, but not too close to the heat source. In the extreme case where the PCM is located on the outer surface of the fabric, the effect would be so short as to be imperceptible. If the PCM is located too close to the heat source, it will never cool down to its transition temperature, and no latent heat generation will occur. Similar results were obtained by Rossi and Bolli [22]. They studied the use of PCM to improve the thermal protection performance of fire fighter protective clothing. Their measurements of the heat transfer index during radiant and convective heat exposure showed that the effect of PCM was more pronounced when the PCM layer was placed on inner rather than outer layers of the clothing system.

Summary

The results obtained indicate the following:

- the main factor determining the TRF value is the amount of latent heat per area unit of the nonwoven fabric
- the second important factor is the way of microPCMs distribution in the fibrous substrate
- the position of the microPCMs layer was proved to be of great importance for the thermoregulating properties of the printed nonwoven samples.

This work has shown the possibility of achieving a significant thermoregulation effect even with moderate amounts of microPCMs incorporated at the proper

location in a nonwoven system. TRF results for clothing fabric assemblies can provide useful information for apparel designers and can be applied to optimise the microclimate of clothing.

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