

## Evaluation of Phase Change Materials for Personal Cooling Applications

Teunissen, Lennart; Janssen, Emiel; Schootstra, Joost; Plaude, Linda; Jansen, Kaspar

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# Clothing and Textiles Research Journal

## Evaluation of phase change materials for personal cooling applications

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Keyword:	phase change material, PCM, cooling garment, cooling power, hotplate
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Abstract:	<p>Eleven phase change materials (PCMs) for cooling humans in heat-stressed conditions were evaluated on their cooling characteristics. Effects of packaging material and segmentation were also investigated. Sample packs with a different type PCM (water- and oil-based PCMs, cooling gels, inorganic salts) or different packaging (aluminum, TPU, TPU+neoprene) were investigated on a hotplate. Cooling capacity, duration and power were determined. Secondly, a PCM pack with hexagon compartments was compared to an unsegmented version with similar content.</p> <p>Cooling power decreased whereas cooling duration increased with increasing melting temperature. The water-based PCMs showed a &gt;2x higher cooling power than other PCMs, but relatively short-lived. The flexible gels and salts did not demonstrate a phase change plateau in cooling power, compromising their cooling potential. Using a TPU or aluminum packaging was indifferent. Adding neoprene considerably extended cooling duration, while decreasing power. Segmentation has practical benefits, but substantially lowered contact area and therefore cooling power.</p>

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## **Evaluation of Phase Change Materials for Personal Cooling Applications**

Working and exercising in thermally challenging conditions have a negative impact on health, comfort and performance (Flouris et al., 2018; Galloway & Maughan, 1997; Gonzalez-Alonso et al., 1999; Lucas et al., 2014; Maughan et al., 2012). To attenuate these adverse effects, multiple types of personal cooling garment are available. One of the main technologies in this area is the use of phase change material (PCM). PCM uses its latent heat capacity of melting for cooling; the material changes phase by heat extraction from an object or medium with a temperature above the PCM's melting point. The functionality can be restored by solidifying the PCM again at a temperature below its melting point. The usability of PCM in cooling garments, without the need for power supply and bulky equipment, offers practical benefits over motorized liquid or air cooling systems. Its application areas are therefore widespread, from industrial work to medical settings and outdoor sports.

As the cooling mechanism of a PCM relies on conduction, the method is particularly suited when evaporative cooling is limited (Zhao et al., 2012). Low evaporation rates occur when someone wears personal protective clothing that reduces breathability. This may result in severe heat strain, e.g., on professionals fighting forest fires or infectious diseases (Petruzzello et al., 2009). Low evaporation rates are also induced by climatic conditions with a high relative humidity, imposing heat strain on outdoor workers and athletes in warm humid areas. The summer Olympics in Tokyo were expected to be challenging in that respect (Gerrett et al., 2019) and turned out to be.

Multiple studies showed that cooling the skin by PCM improves thermal strain, thermal comfort and performance (Bongers et al., 2015; Gao et al., 2012; House et al., 2013; Itani et al., 2018; Reinertsen et al., 2008; Ruddock et al., 2017). Even in the absence of a reduced rectal temperature, the increased temperature gradient from core to skin

26 and/or improved thermal perception provide these benefits (Kay et al., 1999; Ruddock et  
27 al., 2017; Tyler et al., 2011). In general, the phase change energy in a package ( $E_{pc}$ ) is  
28 proportional to the mass ( $m$ ) and given by  $E_{pc} = m \times L_{pc}$ , in which  $L_{pc}$  is the latent heat of  
29 the material. The balance between cooling duration ( $t_{cool}$ ) and intensity ( $P_{cool}$ ), given by  
30  $E_{pc} = P_{cool} \times t_{cool}$ , is determined by physical and environmental factors. Physical factors  
31 comprise both the contact area with the skin and the heat resistance between PCM and  
32 skin. Environmental factors comprise the external insulation of the PCM and the ambient  
33 temperature (Lu et al., 2015; Udayraj et al., 2019; Wan et al., 2018).

34 Different types of work or sport require different demands regarding cooling intensity,  
35 duration and flexibility. Gao et al. (Gao et al., 2010, 2011), investigating three types of  
36 PCM, found that the PCM cooling effect was correlated to PCM melting point, PCM mass  
37 and PCM contact area. Hamdan et al. (Hamdan et al., 2016) recommended, from their  
38 modelling studies, to use a higher melting point and mass for prolonged duration, while a  
39 lower melting point would provide a faster cooling effect. Despite these general guidelines  
40 and some quantitative estimations by manufacturers, empirical and comparative data on  
41 the cooling potential of different types of PCM are scarce, as well as on the influence of  
42 packaging. To be able to design cooling solutions in a more systematic way, an in-depth  
43 study on the cooling capacity of a range of PCMs and packaging configurations was  
44 performed.

45 Classical PCMs provide cooling by changing phase from a solid to a liquid state.  
46 While this process is going on, their temperature will remain at their phase change  
47 temperature. They usually contain water, paraffin or (biobased) oil as a main component  
48 and are available in a wide range of melting temperatures. The most well-known example  
49 is pure water, changing phase at  $0^{\circ}\text{C}$ . However, such a low melting temperature may lead  
50 to vasoconstriction and discomfort when applied directly on the skin. PCM with a higher

51 melting point reduces those risks and requires less refrigeration power to solidify  
52 (Bendkowska et al., 2010), although at the cost of a weaker and slower cooling effect.

53 A disadvantage of these classical PCMs might be the fact that they become completely  
54 rigid in a solid state. To improve freedom of movement, cooling clothing with macro-  
55 encapsulated PCM granules or micro-encapsulated PCM-coatings have been developed  
56 (Mokhtari Yazdi & Sheikhzadeh, 2014). However, as cooling capacity is determined by  
57 the PCM-mass, granules or coatings (usually containing only several grams of PCM) are  
58 unlikely to have a measurable physiological effect. To attain a relevant amount and  
59 duration of cooling in hot conditions, larger PCM-compartments or packs are required  
60 (Mokhtari Yazdi & Sheikhzadeh, 2014), typically 200-400 g each. To still increase the  
61 flexibility of these larger PCM units, the use of a cooling gel can be considered. Cooling  
62 gel usually consists of a mixture of common PCM and a compound with a very low  
63 melting point. In the right proportions, the material remains flexible in the freezer, while  
64 keeping sufficient cooling power due to crystallization of the PCM. Extra additives can be  
65 used to create a gel-like structure. This might be desirable for more comfort, better  
66 handling, prevention of sagging of the material, and prevention of fluid loss in case of  
67 leakage. However, as these additives hinder PCM crystallization and conductivity within  
68 the pack, it will compromise the phase change effect.

69 Next to the classical PCMs changing from solid to liquid, hydrated inorganic salts are  
70 able to store and release latent heat as well. The most known example in this category is  
71 Glauber's Salt ( $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$ ). When the salt is cooled to a temperature lower than the  
72 saturation point, a supersaturated solution with crystal growth appears (Mondal, 2008).  
73 Compared to PCMs like paraffin waxes, hydrated salts have a larger energy storage  
74 density and a higher thermal conductivity (Farid et al., 2004). In addition, they are less  
75 flammable and remain more or less flexible in a cooled state. However, they experience

76 problems with supercooling, phase segregation, corrosiveness and instability on thermal  
77 cycling, limiting their application (Chandel & Agarwal, 2017; Safari et al., 2017). Further,  
78 the typical phase change temperature of hydrated salts ( $\sim 32^{\circ}\text{C}$ ) is too high for personal  
79 cooling. It can be lowered to  $18^{\circ}\text{C}$  by using additives, but remains in a higher range of  
80 melting points than the classical PCMs.

81 The primary aim of this study was to evaluate a selection of classical PCMs (water-  
82 and oil-based), cooling gels and inorganic salts on their cooling characteristics. For that  
83 purpose, several commercially available and custom made PCMs were selected on their  
84 proposed suitability for human cooling in highly heat-stressed conditions. Sample packs  
85 with the different PCMs were tested on a hotplate in a climate controlled environment.

86 Next to the PCM itself, packaging material may also impact cooling characteristics.  
87 Therefore, our second aim was to study 1) the difference between thermoplastic  
88 polyurethane (TPU) and aluminum packaging and 2) the effect of an insulating neoprene  
89 layer for skin protection. By affecting conductivity, it was hypothesized that aluminum  
90 would increase cooling power and decrease duration ( $H_1$ ), while an additional neoprene  
91 layer would decrease cooling power and increase duration ( $H_2$ ).

92 A final point of interest was the packaging pattern. Applications with a large cooling  
93 surface (e.g., a PCM pullover) will generally consist of different compartments to ensure  
94 an even PCM distribution and to improve freedom of movement. However, this  
95 segmentation will diminish contact area and may consequently affect cooling power.  
96 Recently, the authors developed a new PCM packaging design with interconnected  
97 hexagon-shaped compartments. The hexagon shape features three axes around which a  
98 sample can bend. As a result, it improves the flexibility to adapt to body curves and  
99 movements, increasing wearing comfort in work and exercise. The interconnections  
100 between segments enable a 'waterbed mechanism' to reduce the risk of puncturing a

101 compartment by high impact force. The third aim of this study was to investigate the  
102 effect of this packaging design on contact area and cooling characteristics. For that  
103 purpose, a cool pack with hexagon compartments was compared to an unsegmented cool  
104 pack with similar PCM-content. We hypothesized that the hexagon pattern would reduce  
105 cooling power in proportion to contact area ( $H_3$ ).

## 106 **Materials and Methods**

### 107 **Study Design**

108 Cooling power, cooling duration and cooling capacity of eleven PCM cool packs were  
109 determined on a custom-made hotplate. The PCMs were suited for human body cooling in  
110 highly heat-stressed conditions, but varied in composition, melting temperature and/or  
111 packaging material. In a second evaluation, cooling characteristics of a cool pack with  
112 hexagon segmentation was compared to a similarly sized unsegmented cool pack. Both  
113 packs were filled with the same PCM type and mass.

### 114 **PCM Samples**

115 Eleven PCM samples, varying in composition and/or packaging were included in the  
116 study (Table 1).

117 *[Place Table 1 about here]*

118 **Water.** Water has a very large heat capacity, with a latent heat of 334 J/g. However,  
119 because of its low phase change temperature and subsequent high cooling intensity,  
120 vasoconstriction and even frostbite might occur when applied for too long on bare skin.  
121 Therefore, we both tested water in just a standard thin film TPU packaging (denoted as  
122 H<sub>2</sub>O) and in TPU-packaging added with an insulating neoprene layer on top at the contact  
123 side (denoted as H<sub>2</sub>O-neo). A 0.8 mm thick neoprene foam rubber sheet was used (Bardy  
124 et al., 2005), supplied with an adhesive layer to ensure optimal connection between the  
125 rubber and the PCM's packaging. Although it will somewhat reduce the cooling power,

126 closed cell neoprene rubber contains the appropriate material properties for this purpose; it  
127 does not soak up water, is flexible, durable and it insulates very well (thermal conductivity  
128 of 0.05 W/mK).

129 **Ethanol.** Because of its low melting point (-114°C) and non-toxic nature, ethanol is a  
130 suitable compound to create a flexible water-based PCM. Pilot experimentation revealed  
131 that a mixture with 17% ethanol provided the right flexibility after freezing, while still  
132 enabling sufficient crystallization for appropriate cooling characteristics. The melting  
133 point of this mixture amounts -9°C. In addition to this basic mixture (denoted as EtOH), a  
134 more practically applicable version with a gel-like structure (denoted as EtOHgel) was  
135 created adding carboxymethylcellulose (CML). Because of the low melting point, ethanol-  
136 water mixtures also pose risks when directly applied on bare skin. Therefore, a third  
137 sample was evaluated, in which the ethanol-gel pack was covered with a neoprene layer,  
138 similar to the water sample (denoted as EtOHgel-neo).

139 **Inuteq-PAC.** Inuteq-PAC is a commercially available PCM marketed by Inuteq BV  
140 (The Netherlands) and produced by CrodaTherm™ (UK), consisting of bio-based oil and  
141 available in four different melting temperatures (6.5, 15, 21, 29°C). The PCM is  
142 completely rigid in a solid state. We only included the lowest two melting temperatures  
143 (Inu6.5 and Inu15) in the study because of their higher relevance for application during  
144 exercise in the heat. Manufacturer specifications indicate a latent heat of fusion of 184 J/g  
145 for Inu6.5 and 177 J/g for Inu15. In addition, Inuteq-PAC gel (Inu15gel) was evaluated. It  
146 is a commercially available variation of Inu15, consisting of 30% Inuteq-PAC 15°C and  
147 70% unspecified additives. Below its melting point, the PCM remains a flexible gel.

148 **IZI Flexible PCM.** IZI Flexible PCM is a commercially available PCM (IZI Body  
149 Cooling, The Netherlands), consisting of a sodium sulfate decahydrate ( $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$ )  
150 and additives. It is available in three different crystallization temperatures (18, 24 and



151 32°C) and remains relatively flexible in a frozen state. In view of the aimed application,  
152 we only included the 18 and 24°C variants in this study. Manufacturer specifications  
153 assign this PCM a latent heat of fusion of 228 J/g. The manufacturer packages the PCM in  
154 a specific highly conductive aluminum foil (0.15 mm) that is said to improve the PCM's  
155 performance. In order to evaluate the effect of different packaging with similar PCM  
156 content, we compared both the original aluminum (Izi24-alu) and a customized TPU  
157 packaging (Izi24-TPU).

158 **Sample preparation.** For the first evaluation, all PCMs were packaged in square  
159 sample packs of equal size and weight. As the aluminum packaging of IZI Flexible PCM  
160 could not be reproduced by the researchers, the size (empty) and weight of all other test  
161 samples was adjusted accordingly (116 x 140 mm inner bottom surface of the empty pack,  
162 250 g PCM). These other samples were packaged in 0.1 mm thick TPU foil and sealed  
163 using a laser cutter (Figure 1a). As the samples of H<sub>2</sub>O and EtOH exceeded the maximum  
164 cooling power of the hotplate, two smaller packs of half the size and mass (81 x 98 mm  
165 inner bottom surface of the empty pack, 125 g) were produced, providing a similar contact  
166 pressure on the plate. These small packs were used to check the full power profile and the  
167 extent of the missing data during the standard pack measurement.

168 For the second evaluation regarding packaging pattern, two equally sized hexagon  
169 shaped samples of 0.1 mm TPU were filled with 150 g EtOHgel. The first sample was  
170 divided in small communicating hexagon segments with a rib length of 3 cm (honeycomb  
171 structure). In this way, the pack maintained optimal flexibility in a frozen state. The  
172 second sample was unsegmented, resulting in a pack with less thickness (Figure 1b and  
173 1c).

174 *[Figure 1 about here]*

## 175 **Measurements**

176 To determine the PCMs' cooling characteristics, a custom-made guarded hotplate was  
177 used (Figure 1d). This hotplate aims to keep the aluminum plate at the top of the device at  
178 the set temperature, adjusting its power output accordingly. When placing a cool pack on  
179 top of the aluminum plate, the required power equals the heat extraction of the cool pack.  
180 This enables a controlled comparison of the cooling power of the different PCM packs  
181 over time. The measuring area of 150x150 mm is surrounded by a 25 mm wide edge with  
182 separate temperature control to minimize lateral heat losses (guarded hotplate principle).  
183 Maximal cooling power was 71.7 W. Sample rate was 1 Hz and the data were filtered by  
184 an exponentially weighted moving average filter with a weighting factor of 0.1. An  
185 exponentially weighted moving average filter is used for smoothing data series readings. It  
186 allows to specify the weight of the last reading versus the previous filtered value, by  
187 setting the alpha parameter:  $Y_{n\_filtered} = \alpha * Y_n + (1 - \alpha) * Y_{n-1\_filtered}$ . By setting the alpha  
188 parameter at 0.1, the result will be approximately the average of the last 10 readings. The  
189 hotplate was validated by determining the thermal conductivity of EPS and Perspex plates  
190 of various thickness, resulting in realistic thermal conductivity coefficients of 0.03 and  
191 0.15 W/mK, respectively.

192 The contact area of the squared cooling packs with the hotplate was estimated by  
193 making a contact print of the frozen and melted samples on paper. If applicable, the solid  
194 and fluid state measurement were averaged. For determining the contact area of the  
195 hexagon packs, the packs have been placed between two flat pieces of plexiglass. In order  
196 to see where the sample pack touched the plexiglass, a small amount of water was  
197 distributed over the surface of the top plexiglass plate, causing the contact areas to show a  
198 clearer outline. Pictures of the sample packs in between the plexiglass have been made in  
199 top view (53 mm lens), to minimize the effect of distortion. Top view pictures of the  
200 sample packs in between the plexiglass have been made with a 53 mm lens to minimize

201 ~~the effect of distortion due to perspective.~~ The outlines of the surface areas have been  
202 traced in photoshop (darker areas) and the pixels have been scaled to millimeters to be  
203 able to measure the contact surface in SI units.

## 204 **Procedures**

205 PCM packs were frozen in a refrigerator at  $-21^{\circ}\text{C}$  for at least six hours. During  
206 freezing, samples were compressed between two Perspex plates with a 1 kg weight on top,  
207 in order to create a flat surface for optimal initial contact with the hotplate.

208 The hotplate was placed in a bench-top climatic chamber (Espec SH-661, Espec corp,  
209 Japan) to create a temperature- and humidity-controlled environment. The climate  
210 chamber contained a fan continuously blowing at  $\sim 1.5$  m/s. To shield the samples from  
211 airflow and create a stable environment around the samples, a styropor foam cover with a  
212 thickness of 10 mm was placed on top of the samples on the hotplate. During the first 10  
213 minutes (during start-up), the contact resistance increases due to the formation of a thin  
214 melt layer, which evens out the surface roughness that is inherent to a frozen packet. The  
215 temperature of the hotplate was set at  $35^{\circ}\text{C}$ , in line with previous studies (Mairiaux et al.,  
216 1987) and mimicking skin temperatures in hot conditions. The climatic chamber was also  
217 set at  $35^{\circ}\text{C}$ , preventing any heat loss from the hotplate to the surrounding air. Humidity  
218 was controlled at 65%.

219 The test was stopped when the power withdrawal of the sample consistently fell below  
220 5 W. At that stage, the PCMs had completely melted and heated up to above  $25^{\circ}\text{C}$ ,  
221 asymptotically rising towards ambient temperature. The power threshold was based on the  
222 period that PCM cooling would outperform natural evaporative cooling (sweating). Pilot  
223 measurements in a similar set-up indicated that evaporation of water provided about 5 W  
224 cooling power. All samples were tested two times and data were averaged over these  
225 trials.

## 226 **Data Analysis**

227 Data from the hotplate was aggregated with LabVIEW and analyzed using MATLAB  
228 (version R2018b). Power data were averaged per minute from the start of the cooling  
229 period. This start was determined by the first instance that cooling power exceeded the  
230 threshold value of 5 W for three samples (3 sec) in a row. Cooling duration was defined as  
231 the number of minutes during which average cooling power remained above the 5W  
232 threshold. The cooling capacity and average cooling power were calculated by  
233 respectively integrating and averaging power measurements over the total cooling  
234 duration. The mean power was divided by the contact area, normalizing values to W/m<sup>2</sup>.  
235 When the PCM mass deviated from the standard 250 g, the cooling capacity and duration  
236 was normalized to 250 g. Values were averaged over the two trials.

## 237 **Results**

238 The eleven squared test samples had a mean PCM mass of  $246 \pm 7$  g. The mean  
239 bottom surface area of the flat empty packs was  $161 \pm 4$  cm<sup>2</sup>, while the contact area of the  
240 filled packs with the hotplate amounted on average  $111 \pm 9$  cm<sup>2</sup>. The Izi18-alu sample  
241 deviated most ( $131$  cm<sup>2</sup>); without this outlier, mean contact area was  $109 \pm 6$  cm<sup>2</sup>.

242 The two half-sized packs had a mean PCM-mass of  $123.5 \pm 1.5$  g. The mean bottom  
243 surface area of the empty half-sized packs was  $78$  cm<sup>2</sup>, while the mean contact area with  
244 the hotplate was  $43.5$  cm<sup>2</sup>. ~~So~~ Therefore, although the mass and surface area of the empty  
245 packs was nearly 50% of the standard packs, filling reduced the contact area to about 40%  
246 of the standard packs. This will have had a slight effect on cooling rate and contact  
247 pressure. Nevertheless, as the total cooling capacity is determined by the PCM-mass, the  
248 smaller packs were still assumed to have 50% of the cooling capacity of the standard  
249 packs.

## 250 **Power Profiles**

251 Figures 2 and 3 show the cooling power profiles ( $> 5$  W) of the different samples over  
252 time, for clarity grouped into different categories. PCM power profiles typically start at a  
253 relatively high cooling power, associated with the energy needed to raise the temperature  
254 of the contact layers to the melting point. During the subsequent stage the PCM melts,  
255 resulting in a constant power plateau. After complete melting of the PCM the cooling  
256 power drops rapidly. The total cooling capacity of a sample is given by the area below the  
257 cooling power curve.

258 *[Place Figure 2 about here]*

259 In Figure 2, the four classical PCMs ( $H_2O$ , EtOH, Inu6.5 and Inu15) have been  
260 depicted. As the peak cooling power of the  $H_2O$ - and EtOH-samples exceeded the  
261 maximum power of the hotplate, their possible plateau cannot be seen. Therefore, the  
262 additional measurements with half of the PCM-mass have been added to the figure,  
263 demonstrating that the typical plateau phase is indeed present in these PCMs. The Inu-  
264 samples also showed the typical phase change power plateaus. The small fluctuations  
265 during their melting process are likely due to the liquification of the PCM, which causes  
266 flow and enlarges the contact area. In all of the depicted samples, the cooling power  
267 quickly decreased when all material had melted. The higher melting temperatures and  
268 different latent heats of the Inu-oils led to an approximately three times lower cooling  
269 power than the  $H_2O$ - and EtOH-samples. However, the cooling time was much longer.

270 Figure 3a provides an overview of the different cooling gels and salts. Two inflexible  
271 (ungelled) counterparts have been added to the figure for comparison. It is clearly visible  
272 that the phase change plateau characterizing the classical PCMs is absent for the gels and  
273 salts (EtOHgel, Inu15gel, Izi18-alu and Izi24-alu). Instead, they show an instant cooling  
274 peak followed by a gradual decrease. Nevertheless, also among the flexible PCMs, the

275 lowest melting point (EtOHgel) results in the highest cooling power. Further, the Izi18 salt  
276 shows a higher peak power than Izi24 and Inu15gel for the first 5 minutes.

277 The use of aluminum and TPU foil as packaging material for Izi24 flexible PCM  
278 barely showed any difference. The cooling power patterns of Izi24\_TPU en IZI24\_alu never  
279 deviated more than 1.7 W and have therefore not been depicted both. It shows that the packaging  
280 materials had a negligible effect on the cooling performance and both samples are considered  
281 identical.

282 Figure 3b illustrates the effect of adding a layer of neoprene to the sample. The  
283 corresponding samples without neoprene layer have been added to the graph for  
284 comparison. The neoprene layer considerably lowered initial cooling power of the H<sub>2</sub>O-  
285 sample, but increased the cooling duration by using the PCM's latent heat capacity at a  
286 lower rate. The H<sub>2</sub>O-neo sample reached the 5W threshold at about 108 min (not shown in  
287 the graph), a factor three longer than the H<sub>2</sub>O package without insulation. However, the  
288 mean cooling power dropped by a factor 4. For EtOHgel, these factors were much more  
289 limited (1.3 and 1.5 respectively). *[Place Figure 3 about here]*

### 290 **Cooling Duration, Average Cooling Power and Cooling Capacity**

291 Figure 4 shows the cooling capacity, cooling duration and mean cooling power of all  
292 PCM samples, for the interval above the 5 W cooling threshold. All results are averages of  
293 two tests; error bars indicate the individual values. As the upper part of the H<sub>2</sub>O- and  
294 EtOH-profiles was missing, their capacities (123 and 87 kJ respectively) were  
295 underestimated. The additional measurement with a half-sized pack resulted in cooling  
296 capacities of 66 and 51 kJ respectively. Therefore, it can be estimated that the real  
297 capacity of the standard sized packs is approximately 131 and 103 kJ, implying an initial  
298 underestimation of about 6 and 16%. Applying these corrected capacities to the cooling  
299 duration of the original pack, leads to a 7% higher mean power for H<sub>2</sub>O (5820 vs. 5458

300 W/m<sup>2</sup>) and an 18% higher mean power for EtOH (5547 vs. 4694 W/m<sup>2</sup>). Figure 4 has  
301 been adjusted accordingly. Cooling duration of the small packs was nearly similar to the  
302 standard packs (2 min deviation), as can be expected at a roughly comparable mass to  
303 surface ratio.

304 *[Place Figure 4 about here]*

### 305 **Packaging Pattern**

306 Figure 5 illustrates the different cooling power profiles of the segmented and  
307 unsegmented configurations of the EtOHgel samples. Both packs contained 150 g PCM,  
308 but the segmented EtOHgel-hex pack (Figure 1c) had a contact area of 57 cm<sup>2</sup> compared  
309 to 126 cm<sup>2</sup> for the unsegmented EtOHgel-nohex pack (Figure 1c). So the use of hexagon  
310 compartments led to a reduction in contact area of 55%. The mean cooling power  
311 decreased nearly proportional by 56% (30.3 W for EtOHgel-nohex vs. 13.3 W for  
312 EtOHgel-hex), while the cooling duration increased by 60% (25 min vs. 40 min  
313 respectively). Cooling capacity above the 5 W cooling threshold also substantially  
314 diminished in the segmented sample (30%, from 45.4 to 32.0 kJ).

315 *[Figure 5 about here]*

## 316 **Discussion**

### 317 **PCM Characteristics**

318 The primary aim of this study was to evaluate 11 cool packs with classical PCMs,  
319 cooling gels and inorganic salts on their cooling characteristics. The power profile of the  
320 water-based PCMs (H<sub>2</sub>O and EtOH) showed a stable phase change interval at a high  
321 cooling power. Although the peak power could not exactly be determined, it is clear that  
322 both peak power and mean power are considerably higher than the oil and salt-based  
323 PCMs. The trade-off is a 25-60% shorter cooling duration than these latter PCMs. Despite  
324 the shorter cooling duration, the total cooling capacity (power\*duration) of the water-

325 based PCMs is still distinctly higher than the other samples. The addition of 17% ethanol  
326 to water (EtOH) lowers the melting point of the PCM to  $-9^{\circ}\text{C}$  and keeps the PCM in a  
327 non-rigid state. However, the increased flexibility goes at the cost of some cooling  
328 potential, as both power and duration decrease. This is likely due to incomplete  
329 crystallization of the water part.

330 The power profile of the Inuteq-oils (Inu6.5 and Inu15) also showed a relatively stable  
331 phase change interval, but at a substantially lower level. Although they maintained this  
332 stable level for a longer time, cooling capacity and mean cooling power were 27-52%  
333 lower than the water-based PCMs. The cooling capacities of Inu6.5 and Inu15 are  
334 comparable, but the lower melting point of Inu6.5 leads to a 25% shorter depletion time at  
335 a higher power level. These results are in line with previous studies indicating that a PCM  
336 with lower melting temperature should be used for fast intense cooling, whereas a PCM  
337 with higher melting temperature is more beneficial when a prolonged cooling duration is  
338 required (Hamdan et al., 2016; Zhao et al., 2012).

339 The cooling gels and inorganic salts demonstrated a clearly different power profile  
340 from the above-mentioned PCMs. Instead of a 20-60 minute cooling plateau, the gel and  
341 salt containing specimens showed a first intense cooling peak, followed by an immediate  
342 and steady decrease. The lack of a cooling plateau suggests the absence of a crystal-  
343 melting phase. Therefore, it could be hypothesized that the additives for structuring or  
344 stabilizing the material obstruct crystallization of the PCM and suppress the phase change.  
345 Nevertheless, comparison of the experimental heat capacity with calculations on the  
346 substance specifications suggests that the phase change cannot have been completely  
347 absent. An alternative explanation for the lack of a phase change plateau could be the  
348 reduced circulation within the viscous gels and salts. It was observed that at the end of the  
349 test, the upper part of these sample (particularly EtOHgel) was still substantially colder



350 than the lower part, which was in contact with the hotplate. Therefore, there does not seem  
351 to be a stable melting phase, but rather a simultaneous warming of the lower PCM layers  
352 and melting of the higher layers. During practical use in cooling garment, however, body  
353 movement may reduce this effect by stimulating the mixing of the PCM fluid.

354 The different cooling behavior of the gels and salts compromised their cooling  
355 potential, which is illustrated by comparing Inu15gel and EtOHgel to their pure  
356 counterparts Inu15 and EtOH. Inu15gel is a convenient smooth gel, but loses nearly half  
357 of its cooling capacity compared to Inu15. The loss in cooling potential seems  
358 predominantly caused by the large percentage of thickening agent and consequently lower  
359 percentage of active PCM in the substance. In EtOHgel, the thickening agent forms a  
360 much smaller fraction of the total mass and the cooling capacity decrement is limited to  
361 24%. Cooling duration of the EtOHgel is even longer than the fluid EtOH due to its more  
362 viscous structure, as explained above.

363 Both the inorganic salts (Izi18 and Izi24) and Inu15gel reached a higher peak power  
364 than the Inu-oils (Inu6.5 and Inu15), but due to their gradual decrease after this peak, they  
365 end up with a lower mean power. Both cooling power and capacity are lower than the  
366 other PCMs, the former making sense in view of their higher melting temperatures.  
367 Cooling duration of the salts is longer than Inu15gel and comparable to Inu15 and  
368 EtOHgel. Notably, the 6°C difference in melting temperature between Izi18 and Izi24  
369 only results in a higher cooling power during the first five minutes.

### 370 **Packaging Material**

371 The second aim of this study was to investigate the effect of two specific adaptations  
372 in packaging material. First, the standard aluminum packaging of the IZI Flexible PCM  
373 cool packs was compared to the TPU packaging which enclosed the other samples.  
374 Results indicate that an aluminum packaging does not have a clear advantage in cooling

375 potential to TPU, so we have to reject hypothesis  $H_1$ . However, the aluminum laminate  
376 packaging may have other advantages which were not investigated here. For example,  
377 aluminum laminate is known to effectively block water loss by evaporation, which may  
378 benefit the stability of the cooling packages. Moreover, its high conductivity may provide  
379 an optimized homogenization of the surface temperature, preventing spots with too strong  
380 cooling. Finally, it might be beneficial for the cooling perception on the skin.

381 Adding an insulative layer of neoprene does not only protect the skin from frostbite or  
382 worse, but also has a considerable impact on the heat conductivity of the pack and as a  
383 result on the cooling power profile. Especially in the  $H_2O$ -neo pack, the mean power was  
384 about four times lower than without neoprene. In addition, the cooling duration increased  
385 by a factor three. Effects on the EtOH-sample were more modest (mean power reduced by  
386 nearly one third, duration increased by a quarter), but still substantial, thus confirming our  
387 hypothesis  $H_2$ . This means that the addition of a (thin) insulation layer provides a simple  
388 means for fine-tuning cooling power and duration of a given cooling pack.

### 389 **Packaging Design**

390 When cooling larger areas, dividing cooling packs in (interconnected) segments is  
391 often required to create more flexibility and less obstruction in movements. The design  
392 with hexagon-shaped segments of 3 cm rib length (EtOHgel-hex) could fulfill these  
393 requirements very well but was seen to reduce the contact area by 55%. This led to a  
394 reduction in mean cooling power of 56%, confirming our hypothesis  $H_3$  that cooling  
395 power is proportional to the contact area. As PCM mass of the two samples was equal, the  
396 mass-to-contact-area ratio of EtOHgel-hex increased by 55%. This roughly fits with its  
397 60% longer cooling duration and confirms that mass (per surface area) determines cooling  
398 duration. These relationships of contact area and mass with cooling intensity and duration  
399 have been previously reported (Gao et al., 2011; Hamdan et al., 2016).

400 Cooling capacity would be expected to be comparable in view of the equal PCM-mass,  
401 but appeared to be lower in EtOHgel-hex. This is likely due to the previously mentioned  
402 problem of reduced circulation in the EtOHgel. The higher mass-to-contact-area ratio in  
403 EtOHgel-hex increases this issue and could have reduced the effective use of the cooling  
404 capacity. The power profile in Figure 5 suggests that the segmented sample has more  
405 cooling capacity left below 5 W than the unsegmented sample. However, these low  
406 cooling powers are only relevant in conditions without the need for aggressive cooling and  
407 without the opportunity for evaporative cooling.

### 408 **Practical Implications**

409 Multiple factors need to be considered in choosing the right PCM for cooling garments:

410 **Cooling intensity.** This study shows that the cooling power of a PCM primarily  
411 depends on its melting temperature and contact area. So if cooling intensity is the main  
412 criterium, a PCM with low melting temperature applied over a large contact area is  
413 recommended. The studied materials with the highest cooling power were water and a  
414 17% ethanol-water mixture, providing more than twice the cooling power of the oil- and  
415 salt-based materials. However, a higher cooling power is often accompanied by a shorter  
416 cooling duration and may pose a risk for skin damage on direct contact. Consequently,  
417 these PCMs are particularly suited for applications that require intense cooling for a  
418 limited time on top of a clothing layer. In addition, PCMs with low melting temperatures  
419 are more difficult to keep frozen inside a cool box, so its use requires access to appropriate  
420 cooling facilities.

421 **Cooling duration.** The best way to increase the cooling duration is by increasing the  
422 PCM mass per contact area. If it is also required to minimize the PCM mass, cooling  
423 duration can be extended considerably by attaching an insulative (neoprene) layer to the  
424 cooling pack. Although such a layer reduces the cooling power, it may provide an

425 additional benefit in protecting the skin. Therefore, this option is particularly applicable  
426 for long-term cooling with moderate to low intensity, directly on the skin. Similar effects  
427 on cooling duration and power can be expected when a clothing layer separates cool pack  
428 and skin, or when a thicker TPU-layer is used. Finally, cooling duration can also be  
429 increased by adding a thickening agent, making the substance more viscous. The  
430 suppression of crystallization makes this solution less effective in terms of cooling power,  
431 but provides more convenience in terms of flexibility.

432 **Flexibility.** When comfort and freedom of movement are important, large packages  
433 with PCMs being rigid in a frozen state should not be used. There are three ways to  
434 prevent this. First, a substance with very low melting point (like ethanol) may be added to  
435 keep the PCM in a fluid state. Additives may be necessary to create a stable and  
436 convenient consistency. Using additives that do not replace a substantial amount of the  
437 active PCM mass reduces the loss of cooling potential. Secondly, inorganic salts may be  
438 used, although they lack a stable cooling power plateau and seem to have a somewhat  
439 lower cooling potential than other PCMs. Thirdly, flexibility can be improved by  
440 segmentation of the packaging. However, this study showed that the subsequent decrease  
441 in contact area leads to a reduction in cooling power. So depending on the requirements  
442 and intended use of a cooling application, a trade-off should be made between the pros  
443 and cons of a certain segment size. The use of a flexible PCM may allow somewhat  
444 larger segments without losing flexibility. Using interconnected segments, e.g., to spread  
445 and attenuate impact forces, requires small segments in any case.

#### 446 **Limitations and Recommendations**

447 Although PCM samples were of approximately similar size, slight variations in contact  
448 area were apparent within trials and between samples. Variations within trials occurred as  
449 a result of the melting process, levelling the surface roughness of the frozen pack during

450 the first 10 min by a thin melt layer. By averaging measured contact areas in solid and  
451 fluid states (if applicable), a best guess has been made. Variations between samples were  
452 due to differences in material properties like viscosity, phase change behavior, density,  
453 etc. As a result, the contact area of the EtOHgel samples was slightly smaller than  
454 average, whereas the contact area of the Izi-alu samples was somewhat larger. Mean  
455 power values have been normalized and thus corrected for these differences. Nevertheless,  
456 small inaccuracies in contact area estimation cannot be excluded, although its effect on the  
457 main results is considered limited. Further, some condensation was inevitable during the  
458 measurement. This water could have affected the conductivity of the plate and PCM. Its  
459 evaporation could have had some additional impact.

460 The use of a hotplate at fixed temperature does not mimic the complex  
461 thermoregulatory responses of the human skin, like vasomotion. Application of a cool  
462 pack on human skin can cause vasoconstriction and conductive cooling of the skin,  
463 reducing the temperature difference and heat transfer. In addition, the use on a curved  
464 body, possibly with clothing layers in-between, will compromise tight contact. These  
465 factors will slow down the melting process compared to the hotplate. On the other hand,  
466 using PCM cooling garment will stimulate kneading and mixing of the PCM, increasing  
467 the melting process and heat flux towards the skin. Absolute cooling duration and mean  
468 power will therefore differ from a real-life situation, and validation of the findings of this  
469 study through human subject testing is recommended. Nevertheless, the hotplate is an  
470 excellent research tool for straightforward comparison of PCMs on basic cooling  
471 characteristics and for establishing the cooling capacity without the need of subjects or  
472 manikins. Further, knowing a PCM's cooling capacity from the hotplate, mean cooling  
473 power in practice can be estimated by additionally measuring its cooling duration in  
474 practice. This could be achieved by monitoring the PCM's temperature using a thermistor.

475 A neoprene layer can provide skin protection when using a PCM with low melting  
476 temperature directly on the skin. Next to providing skin protection, the application of a  
477 neoprene layer has been shown to attenuate cooling power and extend its depletion time  
478 considerably. The use of alternative insulation materials may enable to tune cooling power  
479 and cooling duration to a desired level, while keeping its skin protective function. In  
480 addition, applying neoprene to both sides of the pack could reduce experimental heat loss  
481 and extend depletion time even further (Udayraj et al., 2019). By applying neoprene only  
482 to the contact side in the current study, environmental heat loss was in fact  
483 underestimated. On the other hand, in practice, applying an additive layer on just one side  
484 of the package provides the flexibility to either or not use it: the side with neoprene  
485 directly on the skin or the side without neoprene on top of a clothing layer.

486 PCM cooling packs are non-breathable and therefore block sweat evaporation,  
487 possibly cancelling the net cooling effect. Therefore, ~~their use is, therefore,~~ most  
488 effective in conditions that substantially reduce evaporation, like protective clothing.  
489 When evaporation is still possible, PCM cooling garments are more useful in hot and  
490 humid environments than in hot and dry conditions (Cleary et al., 2014; Zhao et al., 2012).  
491 In addition, breathability may be increased by the creation of small openings in the PCM  
492 garment, though at the cost of some contact area.

### 493 **Conclusions**

494 In general, a lower melting point of a PCM is related to a higher cooling power and  
495 shorter cooling duration. Water-based PCMs show in this respect a very high cooling  
496 power, ranging up to over 5500 W/m<sup>2</sup>, for a relatively short period of about half an hour.  
497 Gels and salts do not demonstrate a phase change plateau in cooling power which  
498 compromises their cooling potential. This is probably due to incomplete crystallization  
499 and hindered fluid circulation in the cool pack. Further, the cooling power of a certain

500 PCM is proportional to its contact area; the cooling capacity and duration is related to the  
501 PCM mass per contact area. Adding an insulative layer to a cool pack considerably  
502 extends the cooling duration but decreases the average cooling power. Using a TPU  
503 instead of an aluminum foil to pack the PCM does not affect the cooling characteristics.  
504 Segmentation of cooling packs in small compartments has practical benefits but  
505 substantially lowers the contact area and therefore reduces cooling power.

506 The findings of this study can assist in making more considered choices for testing and  
507 designing specific sports, medical and occupational PCM applications, in order to  
508 alleviate heat stress more effectively. That is, it demonstrates the impact of PCM choice  
509 on the cooling intensity/duration ratio, including the opportunity to fine-tune this ratio  
510 using insulative packaging. Further, it increases understanding of the trade-off between  
511 flexibility and cooling requirements when using flexible PCM and/or segmentation,  
512 highlighting the magnitude of both the crystallizing mass and the contact area as important  
513 parameters. Human subject testing is suggested to validate the presented findings and to  
514 further specify design guidelines for garment applications.

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Table 1. PCM samples included in the hotplate test. PCM = phase change material; T<sub>melt</sub> = melting temperature; TPU = thermoplastic polyurethane.

PCM	Composition	T <sub>melt</sub> (°C)	Consistency when frozen	Packaging	Short name	Manufacturer
<b>Water</b>	H <sub>2</sub> O	0	Solid	TPU	H <sub>2</sub> O	Custom made
<b>Water</b>	H <sub>2</sub> O	0	Solid	TPU Neoprene	H <sub>2</sub> O-neo	Custom made
<b>Ethanol</b>	C <sub>2</sub> H <sub>5</sub> OH 17% H <sub>2</sub> O 83%	-9	Flexible	TPU	EtOH	Custom made
<b>Ethanol gel</b>	C <sub>2</sub> H <sub>5</sub> OH 17% H <sub>2</sub> O 83% + CML	-9	Flexible	TPU	EtOHgel	Custom made
<b>Ethanol gel</b>	C <sub>2</sub> H <sub>5</sub> OH 17% H <sub>2</sub> O 83% + CML	-9	Flexible	TPU Neoprene	EtOHgel-neo	Custom made
<b>Inuteq-PAC 6.5</b>	Bio-based oil	6.5	Solid	TPU	Inu6.5	Inuteq
<b>Inuteq-PAC 15</b>	Bio-based oil	15	Solid	TPU	Inu15	Inuteq
<b>Inuteq-PAC gel</b>	Bio-based oil 30% Additives 70%	15	Flexible	TPU	Inu15gel	Inuteq
<b>IZI Flexible PCM 18</b>	Na <sub>2</sub> SO <sub>4</sub> .10H <sub>2</sub> O	18	Flexible	Aluminum	Izi18-alu	IZI BodyCooling
<b>IZI Flexible PCM 24</b>	Na <sub>2</sub> SO <sub>4</sub> .10H <sub>2</sub> O	24	Flexible	Aluminum	Izi24-alu	IZI BodyCooling
<b>IZI Flexible PCM 24</b>	Na <sub>2</sub> SO <sub>4</sub> .10H <sub>2</sub> O	24	Flexible	TPU	Izi24-TPU	Customized (from Izi24-alu)

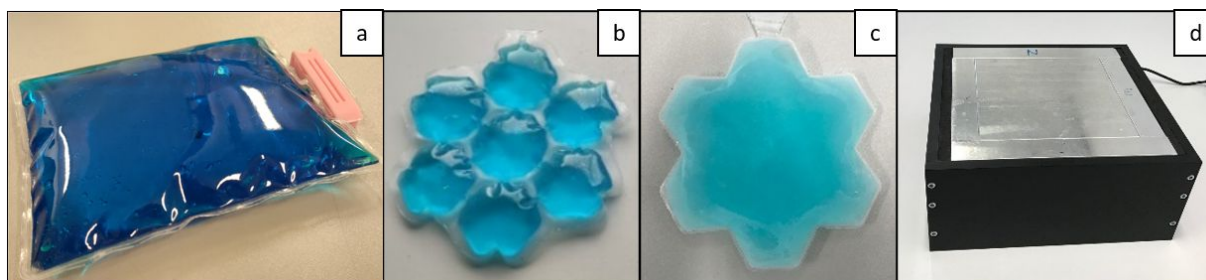


Figure 1. Test material and equipment: a) square sample pack, b) segmented hexagon pack, c) unsegmented hexagon pack and d) custom-made guarded hotplate.

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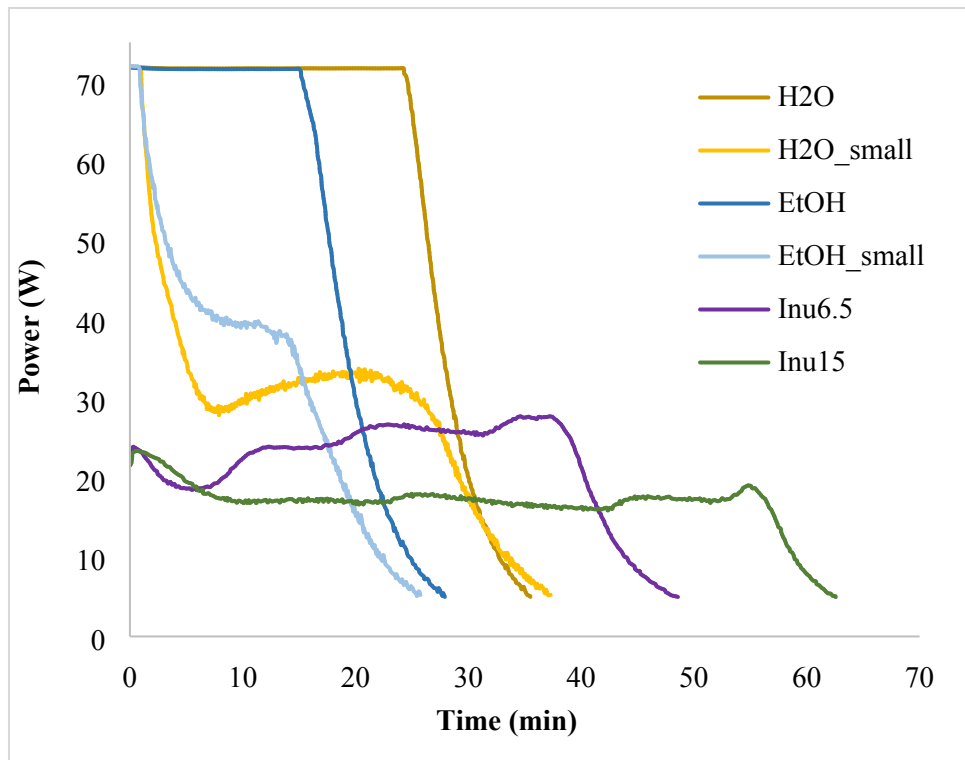


Figure 2. Cooling power profile of the PCM samples with water (H<sub>2</sub>O), 17% ethanol + water (EtOH), Inuteq PAC-6.5°C (Inu6.5) and Inuteq PAC-15°C (Inu15). H<sub>2</sub>O\_small and EtOH\_small are half-sized compared to the standard samples; these samples featured about 50% of the mass and about 40% of the contact area of the original packs.

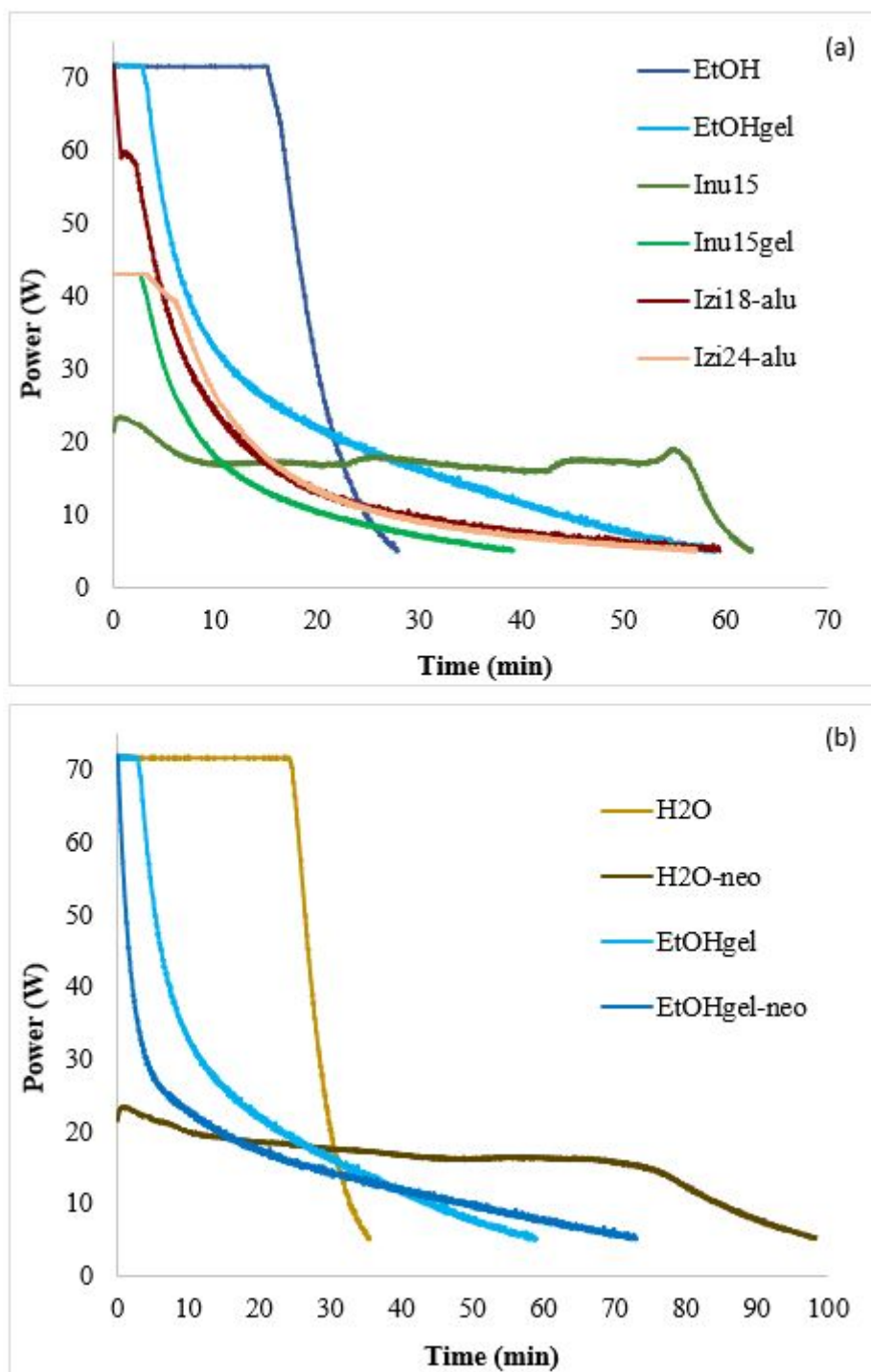


Figure 3. Cooling power profile of a) the flexible PCM samples with 17% ethanol + water (EtOH), 17% ethanol + water + cellulose gum (EtOHgel), Inuteq PAC-15°C (Inu15), Inuteq PAC-15°C gel (Inu15gel), IZI flex PCM 18°C (Izi18-alu) and IZI flex PCM 24°C (Izi24-alu). and b) the H<sub>2</sub>O and EtOHgel samples with extra neoprene layer on their packaging (H<sub>2</sub>O-neo and EtOHgel-neo respectively), along with their equivalents without neoprene layer.

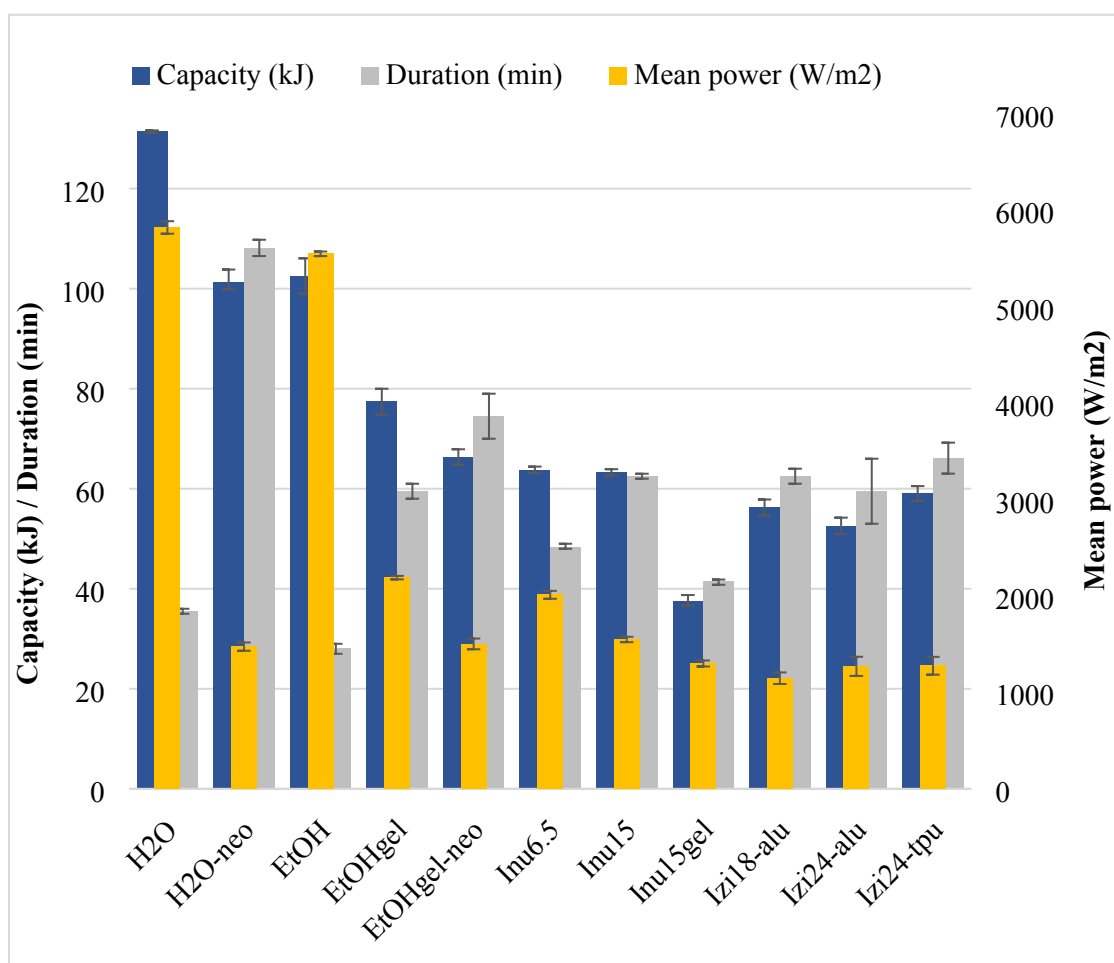


Figure 4. Cooling characteristics of all PCM samples (250 g) for the interval above the 5 W cooling threshold. Cooling capacity (kJ) and duration (min) are depicted on the left *y* axis and mean cooling power (W/m<sup>2</sup>) on the right *y* axis. All results are averages of two tests; error bars indicate the range of the two individual values, as an indication of expected variation.



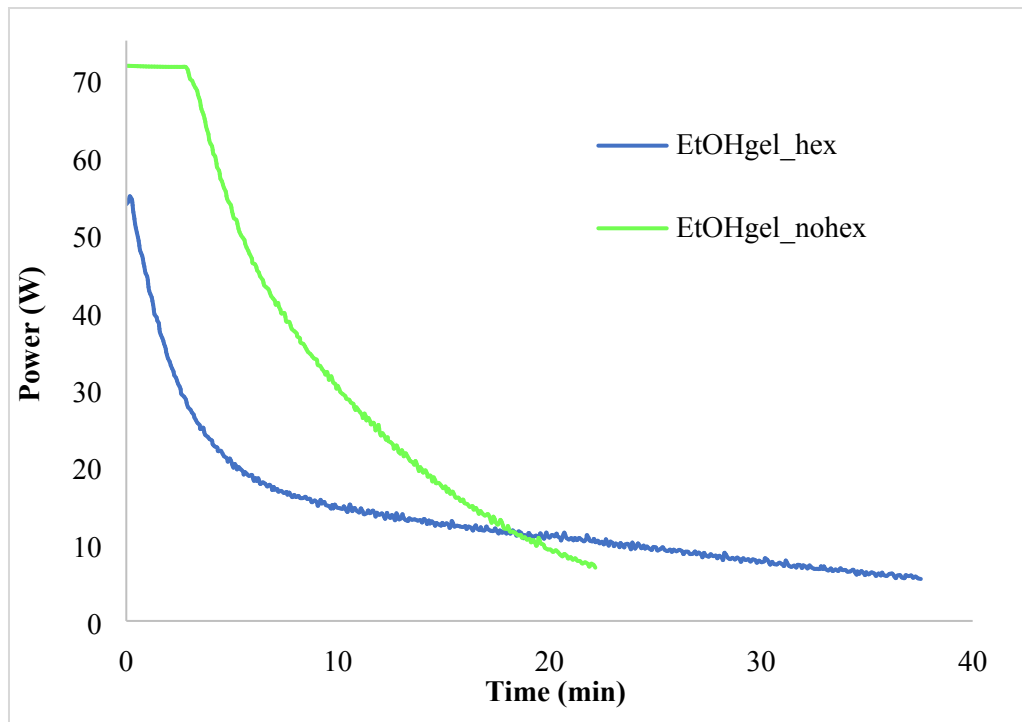


Figure 5. Cooling power profile of EtOHgel in a pack with and without hexagon segmentation.