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Abstract:

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.
Cooling power decreased whereas cooling duration increased with increasing melting temperature. The water-based PCMs showed a >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.

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

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

Classical PCMs provide cooling by changing phase from a solid to a liquid state. While this process is going on, their temperature will remain at their phase change temperature. They usually contain water, paraffin or (biobased) oil as a main component and are available in a wide range of melting temperatures. The most well-known example is pure water, changing phase at 0°C. However, such a low melting temperature may lead to vasoconstriction and discomfort when applied directly on the skin. PCM with a higher melting point reduces those risks and requires less refrigeration power to solidify (Bendkowska et al., 2010), although at the cost of a weaker and slower cooling effect.

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

Next to the classical PCMs changing from solid to liquid, hydrated inorganic salts are able to store and release latent heat as well. The most known example in this category is Glauber's Salt ($Na_2SO_4.10H_2O$). When the salt is cooled to a temperature lower than the saturation point, a supersaturated solution with crystal growth appears (Mondal, 2008). Compared to PCMs like paraffin waxes, hydrated salts have a larger energy storage density and a higher thermal conductivity (Farid et al., 2004). In addition, they are less flammable and remain more or less flexible in a cooled state. However, they experience problems with supercooling, phase segregation, corrosiveness and instability on thermal cycling, limiting their application (Chandel & Agarwal, 2017; Safari et al., 2017). Further, the typical phase change temperature of hydrated salts (~32°C) is too high for personal cooling. It can be lowered to 18°C by using additives, but remains in a higher range of melting points than the classical PCMs.

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

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

A final point of interest was the packaging pattern. Applications with a large cooling surface (e.g., a PCM pullover) will generally consist of different compartments to ensure an even PCM distribution and to improve freedom of movement. However, this segmentation will diminish contact area and may consequently affect cooling power. Recently, the authors developed a new PCM packaging design with interconnected hexagon-shaped compartments. The hexagon shape features three axes around which a sample can bend. As a result, it improves the flexibility to adapt to body curves and movements, increasing wearing comfort in work and exercise. The interconnections between segments enable a ‘waterbed mechanism’ to reduce the risk of puncturing a compartment by high impact force. The third aim of this study was to investigate the effect of this packaging design on contact area and cooling characteristics. For that purpose, a cool pack with hexagon compartments was compared to an unsegmented cool pack with similar PCM-content. We hypothesized that the hexagon pattern would reduce cooling power in proportion to contact area (H_3).

Materials and Methods

Study Design

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

PCM Samples

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

[Place Table 1 about here]

Water. Water has a very large heat capacity, with a latent heat of 334 J/g. However, because of its low phase change temperature and subsequent high cooling intensity, vasoconstriction and even frostbite might occur when applied for too long on bare skin. Therefore, we both tested water in just a standard thin film TPU packaging (denoted as H_2O) and in TPU-packaging added with an insulating neoprene layer on top at the contact side (denoted as H_2O-neo). A 0.8 mm thick neoprene foam rubber sheet was used (Bardy et al., 2005), supplied with an adhesive layer to ensure optimal connection between the rubber and the PCM's packaging. Although it will somewhat reduce the cooling power, closed cell neoprene rubber contains the appropriate material properties for this purpose; it does not soak up water, is flexible, durable and it insulates very well (thermal conductivity of 0.05 W/mK).

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

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

IZI Flexible PCM. IZI Flexible PCM is a commercially available PCM (IZI Body Cooling, The Netherlands), consisting of a sodium sulfate decahydrate ($Na_2SO_4.10H_2O$) and additives. It is available in three different crystallization temperatures (18, 24 and

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

Sample preparation. For the first evaluation, all PCMs were packaged in square sample packs of equal size and weight. As the aluminum packaging of IZI Flexible PCM could not be reproduced by the researchers, the size (empty) and weight of all other test samples was adjusted accordingly (116 x 140 mm inner bottom surface of the empty pack, 250 g PCM). These other samples were packaged in 0.1 mm thick TPU foil and sealed using a laser cutter (Figure 1a). As the samples of H_20 and EtOH exceeded the maximum cooling power of the hotplate, two smaller packs of half the size and mass (81 x 98 mm inner bottom surface of the empty pack, 125 g) were produced, providing a similar contact pressure on the plate. These small packs were used to check the full power profile and the extent of the missing data during the standard pack measurement.

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

[Figure 1 about here]

Measurements

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

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

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

Procedures

PCM packs were frozen in a refrigerator at -21°C for at least six hours. During freezing, samples were compressed between two Perspex plates with a 1 kg weight on top, in order to create a flat surface for optimal initial contact with the hotplate.

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

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

Data Analysis

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

Results

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

The two half-sized packs had a mean PCM-mass of 123.5 ± 1.5 g. The mean bottom surface area of the empty half-sized packs was 78 cm^2, while the mean contact area with the hotplate was 43.5 cm^2. ~~So~~ Therefore, although the mass and surface area of the empty packs was nearly 50% of the standard packs, filling reduced the contact area to about 40% of the standard packs. This will have had a slight effect on cooling rate and contact pressure. Nevertheless, as the total cooling capacity is determined by the PCM-mass, the smaller packs were still assumed to have 50% of the cooling capacity of the standard packs.

Power Profiles

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

[Place Figure 2 about here]

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

Figure 3a provides an overview of the different cooling gels and salts. Two inflexible (ungelled) counterparts have been added to the figure for comparison. It is clearly visible that the phase change plateau characterizing the classical PCMs is absent for the gels and salts (EtOHgel, Inu15gel, Izi18-alu and Izi24-alu). Instead, they show an instant cooling peak followed by a gradual decrease. Nevertheless, also among the flexible PCMs, the lowest melting point (EtOHgel) results in the highest cooling power. Further, the Izi18 salt shows a higher peak power than Izi24 and Inu15gel for the first 5 minutes.

The use of aluminum and TPU foil as packaging material for Izi24 flexible PCM barely showed any difference. The cooling power patterns of Izi24_TPU en IZI24_alu never deviated more than 1.7 W and have therefore not been depicted both. It shows that the packaging materials had a negligible effect on the cooling performance and both samples are considered identical.

Figure 3b illustrates the effect of adding a layer of neoprene to the sample. The corresponding samples without neoprene layer have been added to the graph for comparison. The neoprene layer considerably lowered initial cooling power of the H_2O-sample, but increased the cooling duration by using the PCM's latent heat capacity at a lower rate. The H_2O-neo sample reached the 5W threshold at about 108 min (not shown in the graph), a factor three longer than the H_2O package without insulation. However, the mean cooling power dropped by a factor 4. For EtOHgel, these factors were much more limited (1.3 and 1.5 respectively). *[Place Figure 3 about here]*

Cooling Duration, Average Cooling Power and Cooling Capacity

Figure 4 shows the cooling capacity, cooling duration and mean cooling power of all PCM samples, for the interval above the 5 W cooling threshold. All results are averages of two tests; error bars indicate the individual values. As the upper part of the H_2O- and EtOH-profiles was missing, their capacities (123 and 87 kJ respectively) were underestimated. The additional measurement with a half-sized pack resulted in cooling capacities of 66 and 51 kJ respectively. Therefore, it can be estimated that the real capacity of the standard sized packs is approximately 131 and 103 kJ, implying an initial underestimation of about 6 and 16%. Applying these corrected capacities to the cooling duration of the original pack, leads to a 7% higher mean power for H_2O (5820 vs. 5458

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

[Place Figure 4 about here]

Packaging Pattern

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

[Figure 5 about here]

Discussion

PCM Characteristics

The primary aim of this study was to evaluate 11 cool packs with classical PCMs, cooling gels and inorganic salts on their cooling characteristics. The power profile of the water-based PCMs (H_2O and EtOH) showed a stable phase change interval at a high cooling power. Although the peak power could not exactly be determined, it is clear that both peak power and mean power are considerably higher than the oil and salt-based PCMs. The trade-off is a 25-60% shorter cooling duration than these latter PCMs. Despite the shorter cooling duration, the total cooling capacity (power*duration) of the water-based PCMs is still distinctly higher than the other samples. The addition of 17% ethanol to water (EtOH) lowers the melting point of the PCM to -9°C and keeps the PCM in a non-rigid state. However, the increased flexibility goes at the cost of some cooling potential, as both power and duration decrease. This is likely due to incomplete crystallization of the water part.

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

The cooling gels and inorganic salts demonstrated a clearly different power profile from the above-mentioned PCMs. Instead of a 20-60 minute cooling plateau, the gel and salt containing specimens showed a first intense cooling peak, followed by an immediate and steady decrease. The lack of a cooling plateau suggests the absence of a crystal-melting phase. Therefore, it could be hypothesized that the additives for structuring or stabilizing the material obstruct crystallization of the PCM and suppress the phase change. Nevertheless, comparison of the experimental heat capacity with calculations on the substance specifications suggests that the phase change cannot have been completely absent. An alternative explanation for the lack of a phase change plateau could be the reduced circulation within the viscous gels and salts. It was observed that at the end of the test, the upper part of theses sample (particularly EtOHgel) was still substantially colder than the lower part, which was in contact with the hotplate. Therefore, there does not seem to be a stable melting phase, but rather a simultaneous warming of the lower PCM layers and melting of the higher layers. During practical use in cooling garment, however, body movement may reduce this effect by stimulating the mixing of the PCM fluid.

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

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

Packaging Material

The second aim of this study was to investigate the effect of two specific adaptations in packaging material. First, the standard aluminum packaging of the IZI Flexible PCM cool packs was compared to the TPU packaging which enclosed the other samples. Results indicate that an aluminum packaging does not have a clear advantage in cooling potential to TPU, so we have to reject hypothesis H_1. However, the aluminum laminate packaging may have other advantages which were not investigated here. For example, aluminum laminate is known to effectively block water loss by evaporation, which may benefit the stability of the cooling packages. Moreover, its high conductivity may provide an optimized homogenization of the surface temperature, preventing spots with too strong cooling. Finally, it might be beneficial for the cooling perception on the skin.

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

Packaging Design

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

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

Practical Implications

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

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

Cooling duration. The best way to increase the cooling duration is by increasing the PCM mass per contact area. If it is also required to minimize the PCM mass, cooling duration can be extended considerably by attaching an insulative (neoprene) layer to the cooling pack. Although such a layer reduces the cooling power, it may provide an additional benefit in protecting the skin. Therefore, this option is particularly applicable for long-term cooling with moderate to low intensity, directly on the skin. Similar effects on cooling duration and power can be expected when a clothing layer separates cool pack and skin, or when a thicker TPU-layer is used. Finally, cooling duration can also be increased by adding a thickening agent, making the substance more viscous. The suppression of crystallization makes this solution less effective in terms of cooling power, but provides more convenience in terms of flexibility.

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

Limitations and Recommendations

Although PCM samples were of approximately similar size, slight variations in contact area were apparent within trials and between samples. Variations within trials occurred as a result of the melting process, levelling the surface roughness of the frozen pack during the first 10 min by a thin melt layer. By averaging measured contact areas in solid and fluid states (if applicable), a best guess has been made. Variations between samples were due to differences in material properties like viscosity, phase change behavior, density, etc. As a result, the contact area of the EtOHgel samples was slightly smaller than average, whereas the contact area of the Izi-alu samples was somewhat larger. Mean power values have been normalized and thus corrected for these differences. Nevertheless, small inaccuracies in contact area estimation cannot be excluded, although its effect on the main results is considered limited. Further, some condensation was inevitable during the measurement. This water could have affected the conductivity of the plate and PCM. Its evaporation could have had some additional impact.

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

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

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

Conclusions

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

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

The findings of this study can assist in making more considered choices for testing and designing specific sports, medical and occupational PCM applications, in order to alleviate heat stress more effectively. That is, it demonstrates the impact of PCM choice on the cooling intensity/duration ratio, including the opportunity to fine-tune this ratio using insulative packaging. Further, it increases understanding of the trade-off between flexibility and cooling requirements when using flexible PCM and/or segmentation, highlighting the magnitude of both the crystallizing mass and the contact area as important parameters. Human subject testing is suggested to validate the presented findings and to 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_melt = melting temperature; TPU = thermoplastic polyurethane.

PCM	Composition	T_melt (°C)	Consistency when frozen	Packaging	Short name	Manufacturer
Water	H_2O	0	Solid	TPU	H_2O	Custom made
Water	H_2O	0	Solid	TPU Neoprene	H_2O-neo	Custom made
Ethanol	C_2H_5OH 17% H_2O 83%	-9	Flexible	TPU	EtOH	Custom made
Ethanol gel	C_2H_5OH 17% H_2O 83% + CML	-9	Flexible	TPU	EtOHgel	Custom made
Ethanol gel	C_2H_5OH 17% H_2O 83% + CML	-9	Flexible	TPU Neoprene	EtOHgel-neo	Custom made
Inuteq-PAC 6.5	Bio-based oil	6.5	Solid	TPU	Inu6.5	Inuteq
Inuteq-PAC 15	Bio-based oil	15	Solid	TPU	Inu15	Inuteq
Inuteq-PAC gel	Bio-based oil 30% Additives 70%	15	Flexible	TPU	Inu15gel	Inuteq
IZI Flexible PCM 18	$Na_2SO_4.10H_2O$	18	Flexible	Aluminum	Izi18-alu	IZI BodyCooling
IZI Flexible PCM 24	$Na_2SO_4.10H_2O$	24	Flexible	Aluminum	Izi24-alu	IZI BodyCooling
IZI Flexible PCM 24	$Na_2SO_4.10H_2O$	24	Flexible	TPU	Izi24-TPU	Customized (from Izi24-alu)

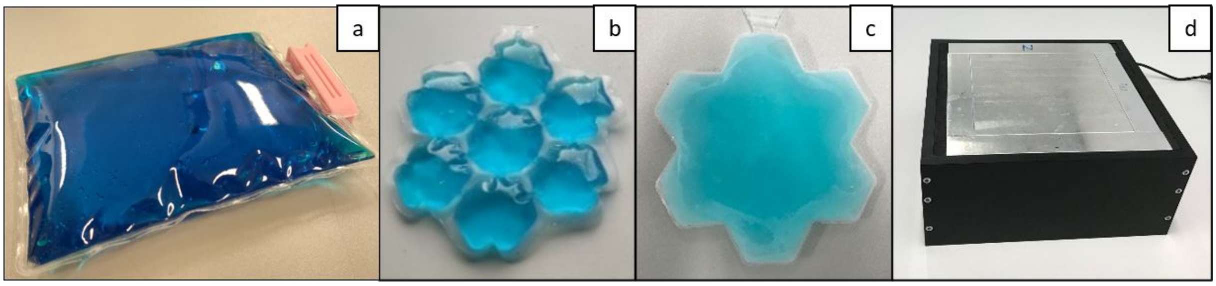

Figure 1. Test material and equipment: a)square sample pack, b)segmented hexagon pack, c)unsegmented hexagon pack and d)custom-made guarded hotplate.

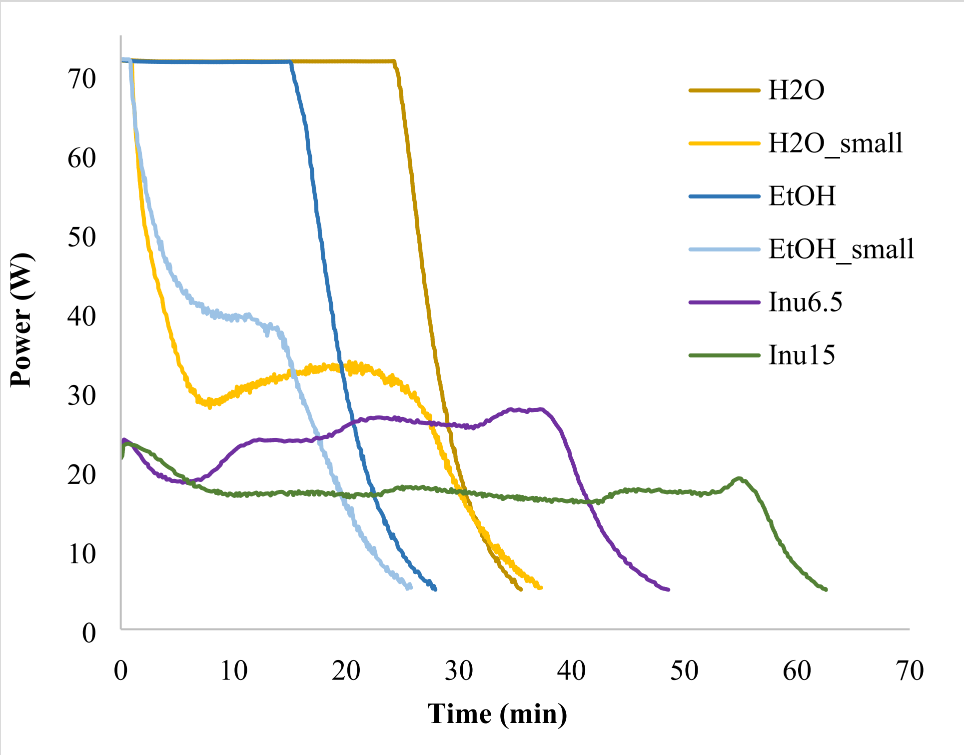

Figure 2. Cooling power profile of the PCM samples with water (H_2O), 17% ethanol + water (EtOH), Inuteq PAC-6.5°C (Inu6.5) and Inuteq PAC-15°C (Inu15). H_2O_small and EtOH_small are half-sized compared to the standard samples; these samples featured about 50% of the mass and about 40% of the contact area of the original packs.

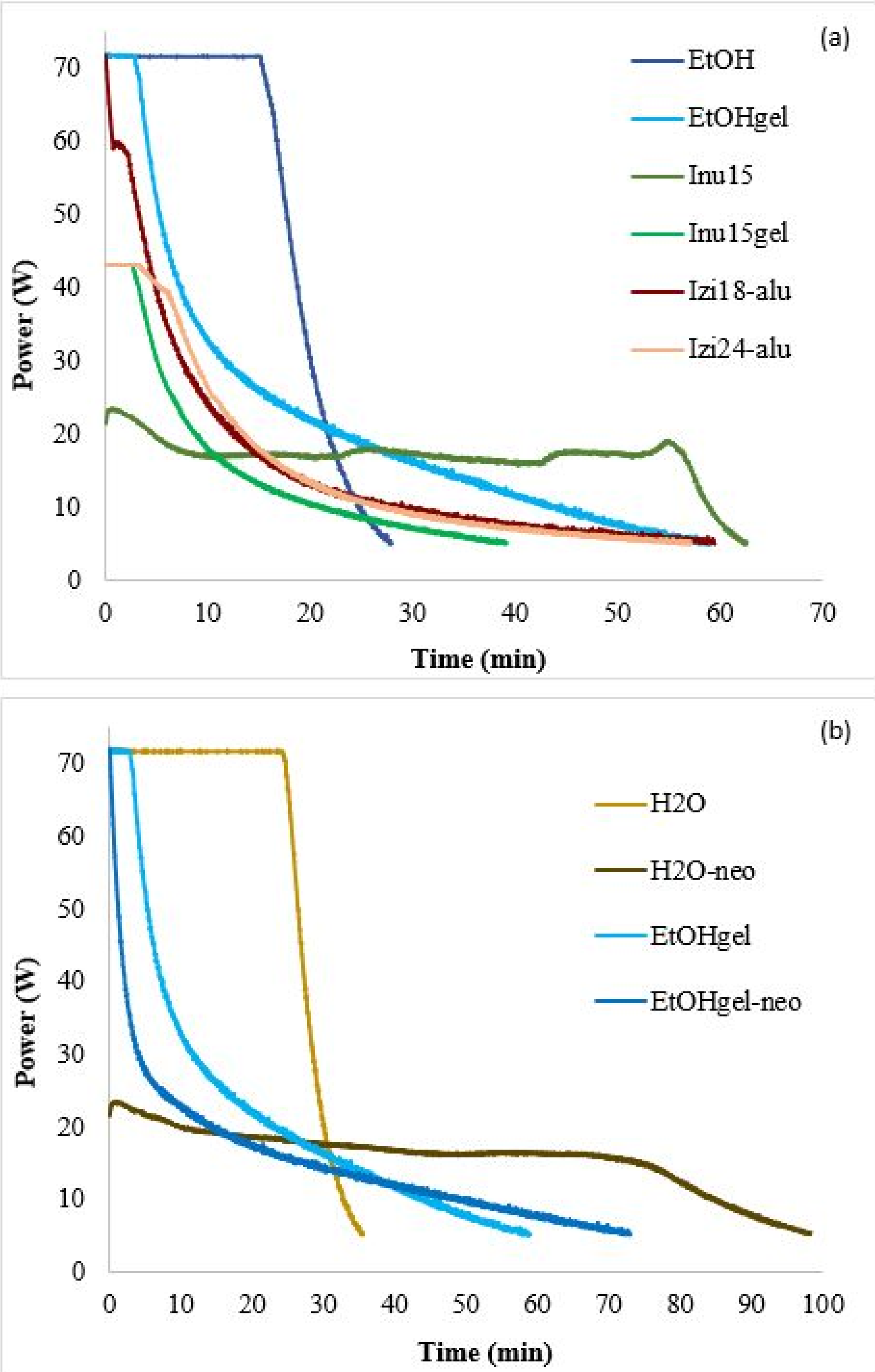

Figure 3. Cooling power profile of a) the flexible PCM samples with 17% ethanol + water (EtOH), 17% ethanol + water + cellulose gum (EtOHgel), Inuteq PAC-15°C (Inu15), Inuteq PAC-15°C gel (Inu15gel), IZI flex PCM 18°C (Izi18-alu) and IZI flex PCM 24°C (Izi24-alu). and b) the H_2O and EtOHgel samples with extra neoprene layer on their packaging (H_2O-neo and EthOHgel-neo respectively), along with their equivalents without neoprene layer.

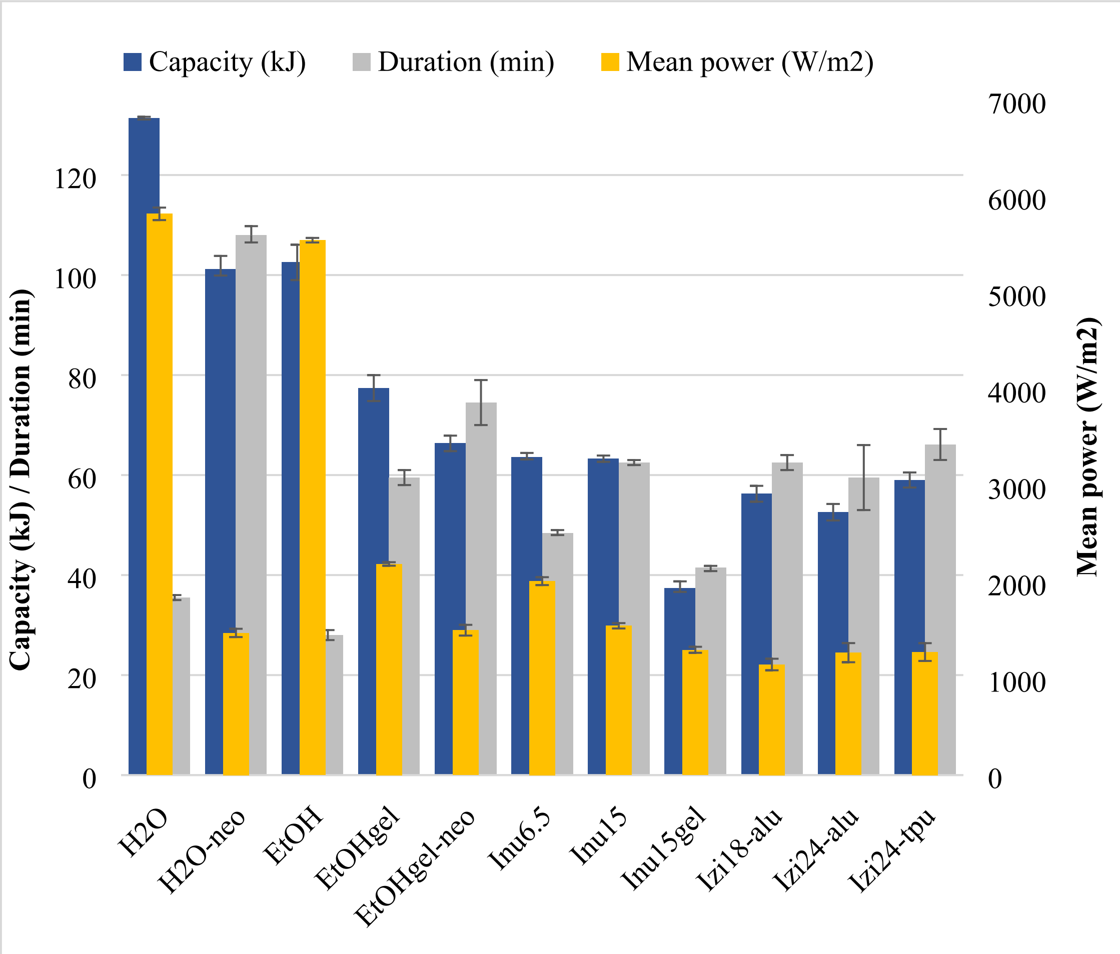

Figure 4. Cooling characteristics of all PCM samples (250 g) for the interval above the 5 W cooling threshold. Cooling capacity (kJ) and duration (min) are depicted on the left *y* axis and mean cooling power (W/m^2) on the right *y* axis. All results are averages of two tests; error bars indicate the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

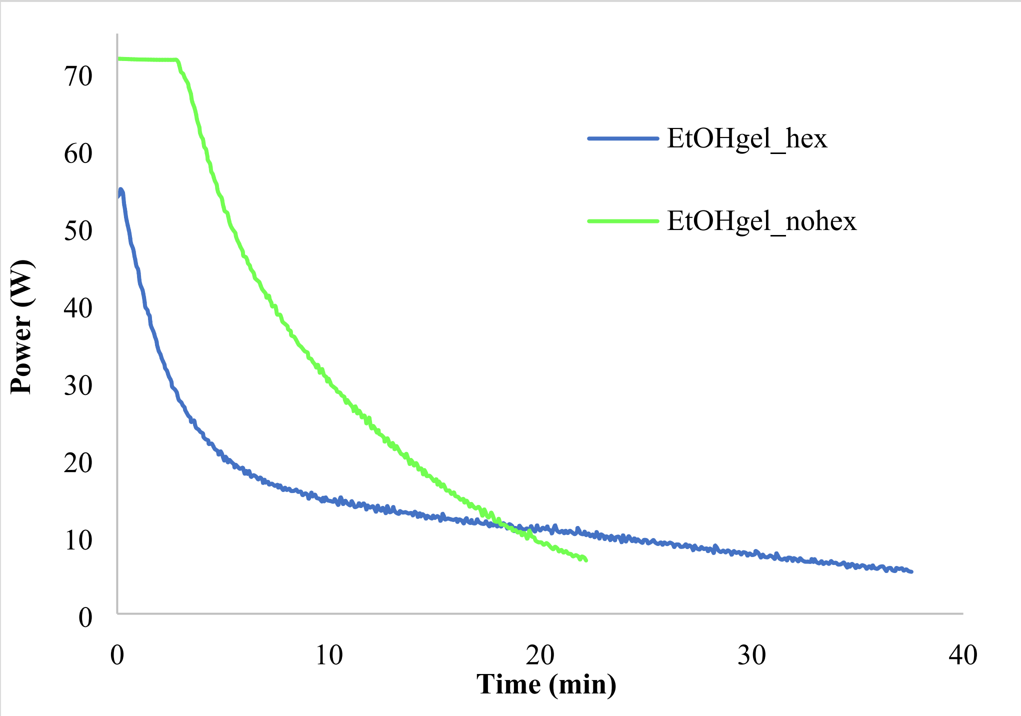

Figure 5. Cooling power profile of EtOHgel in a pack with and without hexagon segmentation.