

Experimental Investigation on Phase Change Materials for Thermal Management of Lithium-ion Battery Packs

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Abstract—Most batteries generate a significant amount of heat during charge or discharge, which has to be dissipated by adequate cooling, as the temperature of the battery is a crucial parameter for the battery performance. An ideal battery thermal management system (BTMS) should be able to maintain a uniform temperature among all battery cells within the entire battery pack. A passive BTMS can compensate for the temperature deviations and maintain temperature uniformity in the battery pack without the use of active cooling components. The aim of this work is to investigate thermal management by a phase change material (PCM) for their feasibility and effectiveness for electric vehicle (EV) battery modules. In this type of latent heat storage, a PCM is melted by internally generated heat, which is released again during solidification on cooling. This novel form of a thermal management system could achieve the advantages of a compact, lightweight and energy-efficient system. Therefore, this work focuses on the implementation of such materials in a battery pack. After comprehensive market research, suitable PCMs that meet the requirements were identified and we studied experimentally these PCMs in a dedicated set-up. Detailed solidification and melting processes were examined and new measured PCM data is reported. During the experiments, specially developed test setups were used to check datasheets and to clarify open questions. A system of single battery cells was designed to provide the PCM with the best possible geometric spaces within the battery module. In order to certifying the thermal behaviour of battery systems with passive PCM cooling, Computational Fluid Dynamic (CFD) models of batteries and surrounding

thermal mass have been developed and could confirm the previous assumptions and calculations.

Keywords—Lithium-ion battery, Phase change material, Battery thermal management

I. INTRODUCTION

The widespread acceptance of electro mobility is strongly dependent on the performance and lifespan of the used battery technology. Lithium-ion (Li-ion) battery technology is the most promising, currently available energy storage technology for mobile applications. With the goal of reducing their weaknesses of lower performance and storage capacity at low temperatures, the use of systems for storing thermal energy is explored. The advantage of storing thermal energy in the battery system is the user behaviour and the systemic fact that vehicles are not always connected to charging stations to obtain energy for heating the battery pack. Maintaining a temperature window for the battery not only optimizes the performance and the storage capacity, but also reduces the load on the battery cells due to ambient temperature influences, which has a positive effect on lifespan. Thus, the total energy consumption of an electric vehicle in the life cycle decreases from production to recycling.

The energy efficiency and thus the actual usable energy of a Li-ion battery pack drops at winter temperatures. The charging characteristics of common Li-ion cells usually do not allow charging below freezing temperatures. [1] These two disadvantages can be drastically reduced and partially eliminated by maintaining a stable temperature window between at least 15°C and a maximum of 30°C. Some car manufacturers consider this and use battery-heating systems during charging below 0°C for EV applications. This leads to higher energy demands at the charging station. The period for maintaining the temperature window after the vehicle is parked depends on factors like ambient temperature, battery technology, insulation of the battery pack and the thermal masses of the battery. By providing an ideal temperature window, an energy efficiency increase of the energy storage system of EVs can be achieved. PCM was investigated for their suitability as thermal energy storage devices for batteries.

Since the temperature range of the PCM is determined purely by their composition, the material can be adapted to a wide variety of applications by changing the mixing ratio. This opens up new application fields for these materials, which can lead to an economic growth in these sectors. The use of PCM in combination with the requirements of battery technology new, innovative, technical approaches for latent heat storage are researched in combined systems.

II. METHODS

The system components of a battery pack were identified and suitable candidates for an energy-efficient energy storage system for EVs were listed. An overview of the most important battery cells for electro mobility was given and the thermal parameters of this battery cells were defined. Through a comprehensive market research, suitable PCMs that meet the requirements were found and we were able to study these PCMs in laboratory experiments in a dedicated set-up. Numerical calculation were performed to show the thermal behaviour of the battery system. These calculations have not yet been validated through experiments.

III. STATE OF RESEARCH

The following section introduces briefly the components required for our application, such as battery cells and PCM.

A. Battery

In the field of portable applications, the lithium-ion battery has become the most important storage technology within a very short time. A key advantage is the high energy density, which can be achieved with acceptable battery capacity [2]. There are varieties of technologies based on varying metal oxides in the cathode. Depending on the choice of material, this results in advantages in one category, which usually lead to disadvantages in others. However, none of these lithium-ion technologies is the best choice for all battery requirements.

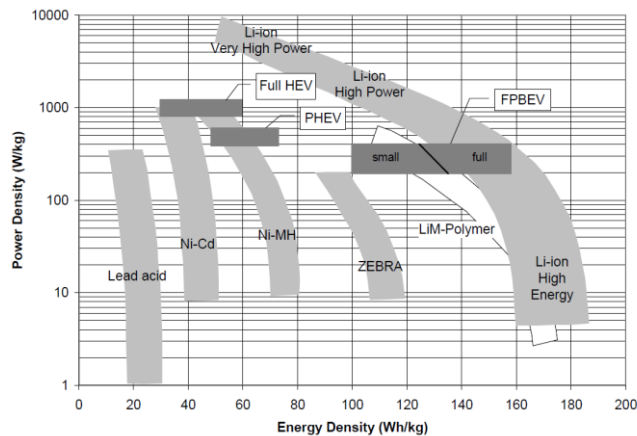


Fig. 1: Energy and power densities of battery technologies [3]

Fig. 1 shows the trade-off between energy density and power density, indicating that the accumulators have either high energy densities or high power densities. With respect to this regard, only Li-ion and lithium-polymer technologies represent promising candidates for EV battery. A power cell delivers high energy over a short time period whereas an energy cell provides lower currents over a longer period. The power of a battery is defined by cell voltage and battery impedance. The impedance depends on temperature, state-of-

charge and current. The test cell should also comply with the latest battery generation and be suitable for use in the electric powertrain. For these reasons, our choice fell on the cell in Fig. 2: Samsung SDI ICR 18650-22P.



Fig. 2: Samsung SDI ICR 18650-22P

Li-ion batteries generate heat during operation. The biggest source of heat is the cell-internal electrical resistance due to the Joule effect. There are also heat sources and sinks due to exothermic or endothermic chemical reactions [4]. Mathematical modelling of the electrochemical processes are complex and requires exact knowledge of the chemical structure of the battery. Since this work treats commercially acquired batteries whose composition is not known in detail, the effect of temperature generation related to losses, known as Joules law of heating is used to describe the heat generation of the battery cell. This law describes that an electrical current, which flows through a conductor, is producing heat. The amount of heat Q_ω can be seen as thermal energy, which is converted from electrical energy. [5]

$$Q_\omega = E_{Electrical} = P * t = \int_0^t P dt \quad (1)$$

The amount of heat energy primarily leads to a heating of the conductor by a temperature difference, which is described as:

$$\Delta v = \frac{Q_\omega}{C_v} \quad (2)$$

The term C_v is the heat capacity. Q_ω increases linearly with the time at constant power. So, also the temperature in increasing linearly until the time where it overlaps with following process. If the conductor temperature becomes warmer than its surroundings, it starts to transfer thermal energy through heat conduction, radiative or convection heat transfer. If the power supply is uniform and continuous, the conductor and the surroundings will reach an equilibrium state at a higher temperature where the emitted heat flow and the absorbed electrical power is equal.

Table 1: Nominal specification of SDI ICR 18650-22P cell [6]

Item	Specification
3.1 Typical Capacity	2150mAh (0.2C, 2.75V discharge)
3.2 Minimum Capacity	2050mAh (0.2C, 2.75V discharge)
3.3 Charging Voltage	4.2V±0.05 V
3.4 Nominal Voltage	3.62V (1C discharge)
3.5 Charging Method	CC-CV (constant voltage with limited current)
3.6 Charging Current	Standard charge: 1075mA Rapid charge : 2150mA
3.7 Charging Time	Standard charge : 3hours Rapid charge : 2.5hours
3.8 Max. Charge Current	2150mA
3.9 Max. Discharge Current	10A (Continuous discharge)
3.10 Discharge Cut-off Voltage	2.75V
3.11 Cell Weight	44.5g max
3.12 Cell Dimension	Diameter(max.): Φ 18.40 mm Height : 65mm max
3.13 Operating Temperature (Cell Surface temperature)	Charge : 0 to 45°C Discharge: -20 to 60°C
3.14 Storage Temperature	1 year : -20~25°C(1°) 3 months : -20~45°C(1°) 1 month : -20~60°C(1°)

As we can see in Table 1 the typical capacity of our test cell is 2150 mAh and nominal terminal voltage is 3.62 V. From this, we can calculate the energy requirement E for one cell charge or discharge. For a capacity comparison, we need the conversion of amp-hours in watt-hours.

In our case where Q the typical capacity of the battery and V is the nominal voltage:

$$E = Q * V \quad (3)$$

$$[Wh] = [Ah * V]$$

The equation (3) results in an energy storage requirement of approximately 8 Wh per battery cell.

B. Phase Change Material

The main task of an energy storage system is the caching of energy to balance supply and demand. In sensitive heat storage systems, heat is stored "sensible" by the temperature difference of a storage medium before and after the charging process. Decisive for the stored amount of energy is the temperature difference, the heat capacity and the mass of the storage medium. Sensitive heat storage tanks are located in almost every household as hot water boilers and buffer tanks and require good thermal insulation. In latent heat storage, the heat is stored in the "latent" rather than "sensible" storage. The charging and discharging is done by changing the state of aggregation of the medium using the respective enthalpy. These systems can store heat relatively lossless for a long time. [7] By storing heat with PCM, a large amount of energy can be stored and released again. The system works with the phase transition of the material to store energy and release it later. Some chemical processes such as melting, solidification or evaporation require energy. According to the general rules of chemistry and physics, heat is absorbed or released when the material changes from solid to liquid, and vice versa.

PCMs change their phase with a predictable amount of energy and release that energy later in a predictable time. [7] This means latent heat storage is heat storage where the thermal energy can be latently stored for a long time with low losses. It uses so-called "phase change materials" (PCM), the latent heat of fusion, heat of solution or heat of absorption is much greater than the heat that they can save due to their normal specific heat capacity.

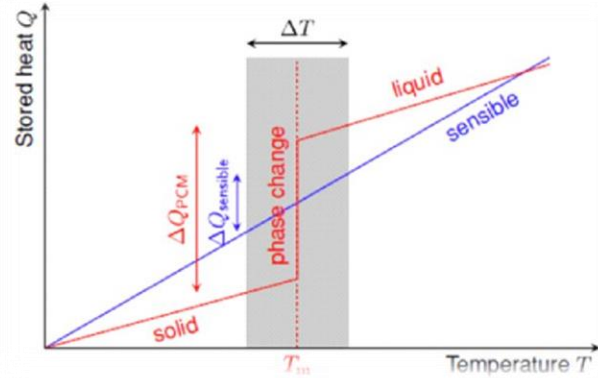


Fig. 3: Stored heat temperature diagram [8]

Fig. 3 shows the difference between sensible and latent heat storage. The advantage of greater heat storage can only be exploited in a certain area. Latent heat storage devices operate by utilizing the enthalpy of thermodynamic changes in state of a storage medium. The most commonly used principle is the use of the solid-liquid phase transition and vice versa. When charging the storage usually specific salts or paraffin is melted as storage medium that absorb a lot of heat energy. The discharge takes place as solidification, wherein the storage medium emits the previously recorded large amount of heat as solidification heat back to the environment. The amount of heat depends not only on the temperature difference of the system, but also on the mass of the material and the specific heat capacity of the substance. Specific heat capacity is a physical property of matter and indicates the amount of heat that must be supplied to one kilogram of material to increase it by one Kelvin. In the heat storage, it is advantageous if the substance has a high heat capacity because a smaller amount of the substance can be used. The specific heat capacity of the most commonly used storage media is usually limited. Depending on the temperature range, different material classes are used as latent heat storage materials. The transition solid to liquid is mainly used because the volume change is easier to handle compared to the transition solid to gaseous and the transition enthalpy is sufficiently high compared to the fixed-solid transitions. The longest and most commonly used PCM is water or ice. It meets all the necessary criteria and is very cost effective and available everywhere. In addition to water, the material classes' salt hydrates and paraffin are used most frequently [8]. The application thus determines the storage medium with the most suitable phase transition temperature. Paraffin are often used because of their chemical inertness and thus easier handling. Paraffin are a mixture of saturated hydrocarbons composed mainly of long-chain alkanes. These are obtained by distillation from petroleum. The larger the chain lengths, the higher the melting temperature. This makes it easy to set a specific melting point. Such paraffin-based PCMs are characterized in particular by

the fact that they melt congruently, are cycle-stable, ecologically harmless, are not harmful to health and are not corrosive to metallic construction materials. Super cooling also plays a role in paraffin only in very small encapsulation. Disadvantages of paraffin are their high volume change at the melting point and their very low thermal conductivity [8].

IV. EXPERIMENTS TO INVESTIGATE PCM

Since the phase change temperature is the most important criterion when selecting a PCM for a particular application, special emphasis is placed on it. In order to adapt the melting point of the paraffin to the application, it is possible to use mixtures of different materials. In most cases, however, the melting range widens and the phase transition enthalpy decreases. For cost reasons, low purity materials are used, which has similar effects. Therefore, the characterization of these PCMs will pay an important role in this project [8]. Low cost and high availability of phase change materials are very important, but there are also chemical properties that must be considered for the correct selection of a PCM like chemical stability, reversible melt cycle, no degradation after a large number of freeze / melt cycle, corrosion on the materials of the system, safety (non-toxic, non-combustible and non-explosive). There is not a single material that can have all the required properties for an ideal PCM. Therefore, the available material must be selected and additional additives added to replace the poor physical properties with a suitable system design [9]. The materials available on the market and the chosen phase transition temperatures determined the choice of the suitable PCM for a passive BTMS.

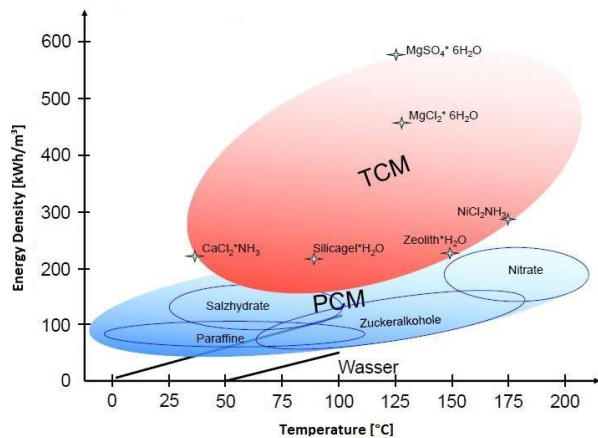


Fig. 4: Energy density of different materials [8]

In Fig. 4 we can see that for this temperature range only paraffin come into question. Since the function of the passive BTMS depends essentially on the selection of the PCM and its properties, special emphasis was placed on it. In order to verify the characteristics of PCMs own test benches were developed, which allows us to measure different PCMs simultaneously and automatically. Additionally, PCMs can be observed over a longer period. The developed PCM test bench is designed to be mobile and modular in order to enable an exchange of the test items or an extension of the heat storage unit. The exact process and the components used for this test bench were developed in a preliminary project and were described in detail in the accompanying report [10]. In the

final version of the PCM test bench, different types of salt hydrate and paraffin storage systems were selected and integrated. The experimental setup can be seen in Fig. 5.

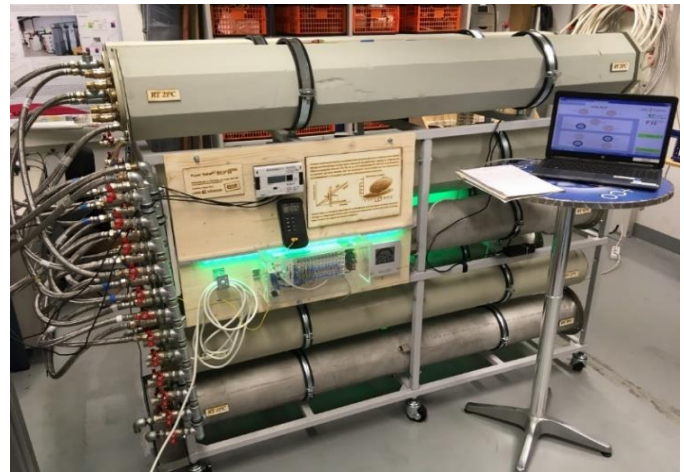


Fig. 5: Piping and control module of the heat storage unit

The control of the experimental setup was realized via a programmable logic controller (PLC). The temperature sensors were connected to the PLC via analogue input terminals. For the temperature measurement itself own measuring modules were developed. Particularly interesting for our application are the PCM tanks with a phase transition of 21°C and 26°C. The charging process of the PCM tanks, the measurement setup, and the energy supply is described in detail in the project report [10]. Fig. 6 shows the measured values such as Energy, Flow, Power and Temperature difference between supply and return-flow pipe when charging these storage tanks.

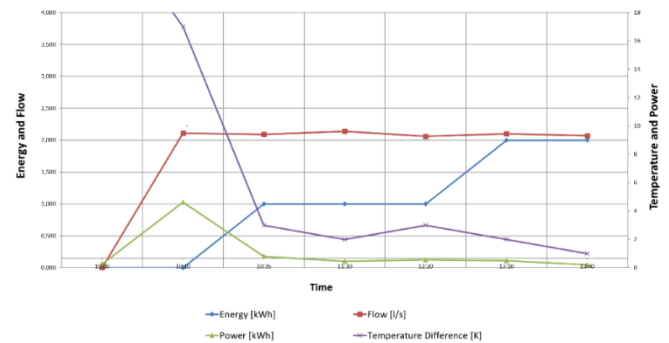


Fig. 6: Measurement data paraffin

The flow remains stable after the system has started. The differential temperature decreases with increasing charge of the storage tank. The same behaviour is also observed in the power measurement. The most important value for determining the storage capacity is represented by the energy measurement, which presents the energy content of the fully charged test samples. Since this value is mainly dependent on the amount of paraffin in the tank, it is almost identical for all PCM tank samples, regardless of the type of insulation. The last attempt in the planned measurement series is to evaluate the insulation properties of the different tank versions in order to validate the long-term memory quality of the test samples. The tanks chosen for this experiment differed only in the design of the sheaths. These were stainless steel, plastic, and

plastic insulated. As reference material, RT42 was chosen. Since this material offers the highest temperature difference to the laboratory environment, we expect the shortest possible test period. For this experiment, the tanks were first fully charged. Subsequently, the charge and discharge pumps were switched off and the temperature values of the charged storage tanks were recorded over several days in order to display the self-discharge of the individual storage tanks. The results show that the sensible heat (temperature up to the phase transition) escapes from the reservoirs within a few hours. Surprisingly, the insulated storage does not hold the heat best in our case. The explanation lies in the design of the insulation. The test samples are a small series in prototype status. The thermal insulation is not ideally designed. It still offers potential for optimization in terms of mounting and material selection. Of course, the best heat conduction to the room is the stainless steel version without insulation. From this point on, the temperature will oscillate around the phase transition point until the latent heat has escaped from the PCM tank.

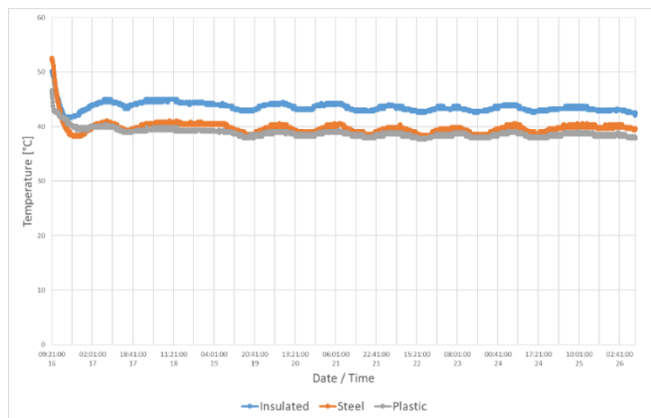


Fig. 7: self-discharging curve paraffin over 11 days

As can be seen in Fig. 7 the time when the latent heat has escaped from the PCM tank could not be reached during the data-logging period of 11 days. This means that the measured paraffin tanks, despite of non-existent thermal insulation have excellent insulation properties due to the storage material itself. This experiment confirms the good thermal insulation properties of the PCM, as well as the possibility of using the test bench for long-term test and the failure-free function of the automated data generation.

The generated measurement results matched the existing data sheets, and were able to confirm the expected insulation behaviour of the PCM. Based on the selected batteries for the battery pack and the generated measurement data of the PCMs, paraffin RT21 turns out to be the most suitable for our application.

Table 2: Area requirement of PCM

	Formula		Result
Conversion PCM Mass / Volume:		$0.20\text{kg} = 0.186\text{ ltr.} = 0.786\text{ dm}^3$	186 cm^3
Calculation Area:	$A_{\text{PCM}} = V_{\text{PCM}}/H_{\text{cell}}$	$186\text{ cm}^3/6,5\text{cm}$	28.615cm^2
	$A_{\text{cell}} = r_{\text{cell}}^2 * \pi$	$(0.92\text{cm})^2 * \pi$	2.659cm^2
	$A_{\text{Total}} = A_{\text{PCM}} + A_{\text{cell}}$	$28.615\text{cm}^2 + 2.659\text{cm}^2$	31.274cm^2

Table 3: Data sheet RT21 [11]

Rubitherm RT21			
Melting area		18 - 23	°C
Congeeing area		22 - 18	°C
Heat storage capacity	+/- 7,5%	155	kJ/kg
(latent/sensible - 13°C to 28°C)		43	Wh/kg
Specific heat capacity		2	kJ/kgK
Density solid	at 15°C	0,88	kg/l
Density liquid	at 25°C	0,77	kg/l
Heat conductivity		0,2	W/(m K)
Volume extension		12,5	%
Flashpoint		140	°C
max. Operation temperature		40	°C

The maximum heat storage capacity of this material is achieved in the temperature range of 13°C to 28°C. In this area, the heat is stored in a combined form of latent and sensible heat. As we can see in the related data sheet in Table 3 the chosen material has a heat storage capacity of 43 Wh in the selected temperature range. This results in a need for thermal energy storage material of approximately 0.20 kg per battery cell. The information on the size and weight of the battery can also be found in the data sheet. This results in an area requirement of the PCM of 28.615 mm² according to Table 2. In our concept, the amount of heat energy should be conducted over the lateral surface of the battery cell.

V. MODELLING AND SIMULATION

Many industrial applications, such as air-cooling in the coil of a conditioner or the cooling in a battery pack, can be modelled as two-dimensional periodic heat flow. The chosen system for simulation is a bank of battery cells give heat off surrounded by PCM, which should act as passive cooling. The heat generated within the cells of the battery pack is transferred to the PCM. Unlike active cooling systems, the heat transfer within the battery pack takes place by conduction only. The resulting mesh consists of a periodic module with symmetry. This results in an area requirement of the PCM of 28.615 mm² according to Table 2. The representation of active cooling vs. passive cooling can be seen in Fig. 8.

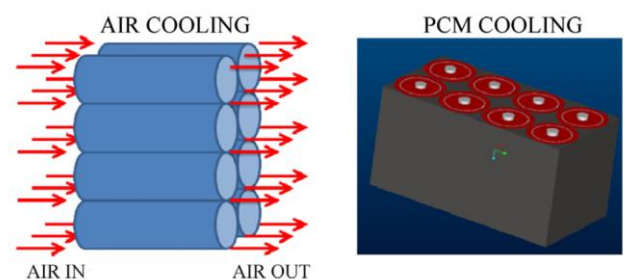


Fig. 8: Representation of active cooling vs. passive cooling [12]

Because of the symmetry of the battery stack geometry, only a portion of the battery stack model must be modelled. Since we were able to prove that the paraffin used has excellent insulating properties, we decided that at this point of development the loss of heat across the outer surface of the battery pack could be neglected. Therefore, further symmetries could be used in this model and only a portion of

the geometry will be modelled in Ansys / Fluent in order to save computer capacity and resources. The temperature distribution in the material was simulated at the maximum battery cell temperature of 45°C and ambient temperatures between 0°C and 45°C.

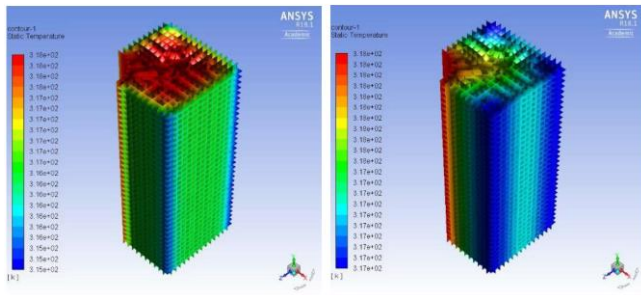


Fig. 9: PCM temperature distribution at ambient temperature of 0°C and 45°C

The heat generated by the battery cell can be completely absorbed by the PCM in these cases. From this, we conclude that the complete thermal energy of a battery cell can be absorbed by the surrounding PCM during a charge or discharge process. This means that input of thermal energy from the battery into the PCM could produce a homogeneous temperature distribution in the battery pack but for safe operation of the battery in case of overheating, additional cooling must be integrated into the concept. These calculations have not yet been validated through experiments.

VI. RESULTS

In chapter IV we show that paraffin is a suitable PCM for our application. The generated measurement results matched the existing data sheets and were able to confirm the expected insulation behaviour of the PCM. Based on the selected batteries for the battery pack and the generated measurement data of the PCMs, paraffin RT21 turns out to be the most suitable for our application. The maximum heat storage capacity of this material is achieved in the temperature range of 13°C to 28°C. In this area, the heat is stored in a combined form of latent and sensible heat. It is possible to conclude that the ideal temperature window for a permanent operation of our batteries should be between 15°C and 30°C. Using paraffin RT21, we meet this requirement. As we can see in the related data sheet in Table 3 this material has a heat storage capacity of 43 Wh in the selected temperature range. In chapter III.A we noticed an energy capacity of 8 Wh per battery cell. Based on these values, there is a need for thermal energy storage material RT21 of approximately 0.20 kg per battery cell in the battery pack.

With the assumption made in chapter IV and the corresponding calculations from III.A, the simulation was quite simple. As can be seen in Fig. 9: PCM temperature distribution at ambient temperature of 0°C and 45°C the heat generated by the battery cell can be completely absorbed by the PCM in all cases. From this, we conclude that the complete thermal energy of a battery cell can be absorbed by the surrounding PCM during a charge or discharge process according to chapter III.A and the battery pack can be supplied with a homogeneous temperature distribution.

VII. CONCLUSION

In this project, thermal management with phase change material was investigated for their feasibility and effectiveness for electric vehicle battery modules. Detailed solidification and melting processes were examined and new measured PCM data was reported. In the experiments, specially developed test setups were used to check datasheets and to clarify open questions. With the experimental test benches, we could already evaluate different latent heat storage materials and define a suitable PCM for the required temperature range of the chosen battery cell. Numerical calculation were performed to show the thermal behaviour of the battery system. These calculations have not yet been validated through experiments. The strong temperature dependency of the performance of a Li-ion battery pack could be damped by adding a high energy density thermal storage such as our PCM.

The cooling effect at ambient temperature is hardly or not at all available due to the material properties of the PCM. For safe operation of the battery in case of overheating, additional cooling because of the poor thermal conduction of the PCM must be integrated. The implementation of PCM in the vehicle battery increases the total weight of the battery and adds on the overall weight of the vehicle, which would result in an increased energy consumption. A follow-up project is planned to answer these questions sufficiently. Furthermore, the thermofluid-dynamic processes in the PCM are examined more closely in this project in order to be able to describe the phase change and heat transfer processes more precisely.

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