Advanced thermal management for temperature homogenization in high-power lithium-ion battery systems based on prismatic cells

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***Abstract*—In order to extend the lifetime of lithium-ion batteries, an advanced thermal management concept is investigated. In battery modules, different cell temperatures lead to higher efforts in cell balancing and reduce the system´s lifetime. Especially when battery systems with phase change material operate outside the phase transition range high temperature gradients can occur that result in different ageing speeds of the cells. The effect of temperature dependent ageing of the battery cells is further investigated. A battery module concept is developed with focus on temperature homogenization by optimization of the module design and material characteristics. The module design combines several approaches including optimized interface pads, thermal storage materials and anisotropic multilayer graphite sheets. Numerical simulations with material and geometrical models are used for the evaluation of the concept with reference models. In addition, a battery cell model is set up, which describes the reversible and irreversible heat generation rate. Using model-order-reduction, the simulations are accelerated by reduction of the calculation time. In order to optimize the material parameters, the simulations are analyzed with design exploration techniques. As a result, the overall temperature differences in the module are minimized and the temperature distribution is homogenized with new developed interface pads. In combination with high thermally conductive synthetic graphite sheets the pads also compensate the insulating behavior of thermal storage material, which is used for temperature peak reduction and to smooth temperature changes.**

Keywords: lithium-ion batteries, lifetime increase, passive thermal management, temperature homogenization, heat generation, FEM simulation, design optimization, model-order-redcution

I. Introduction

Due to their high energy density, lithium-ion batteries are used for various applications in a fast growing global market. One main challenge to meet the expectations in the market is to increase the lifetime of battery systems, which results from the state of health (SOH) of the individual cells. The ageing of the cells appears in capacity fading and power capability drift after a certain amount of operating hours. Since these effects are mainly temperature driven, inhomogeneities in temperature distribution lead to different ageing speeds of the individual cells. Drifting SOHs of the cells reduce lifetime and capacity of the overall battery pack and cell voltage balancing is required to compensate differences in cell capacities. In summary, a higher temperature difference in a battery module results in more balancing efforts, shorter lifetime and a reduction of the overall power performance of the system.

Consequently an effective thermal management is needed to homogenize the temperature distribution. In this paper a concept is introduced, in which passive thermal management is realized and integrated in the mechanical module design. The potential of passive thermal management is already presented in phase change material (PCM) based battery concepts with benefit in reduction of temperature peaks and smoothing of the temperature distribution [1]-[3]. However, the effect of PCM regarding homogenization is limited to the range of phase transition of the material. Outside this range the uniformity is reduced especially for higher charge or discharge currents.

In the present paper, the thermal management is done in a passive way and demonstrated on a module concept with prismatic battery cells and liquid cooling. Different approaches in module design are combined to meet the target of temperature homogenization (temperature difference < 5 K). The focus is on the materials with specific thermal characteristics. With finite element method (FEM) simulations, the concept is further investigated, optimized and evaluated. Design exploration techniques are helping to define material requirements and to adapt the module construction. Simulation models with according heat generation rates of the battery cells are created. The simulation model development is accelerated with model-order-reduction (MOR), which reduces calculation time and efforts.

Finally, the simulation results are discussed and the improvement potential is explained with reference models. These investigations provide a basis for the development of a smart battery system with advanced passive thermal management.

II. Influences on Ageing of Battery Cells

The cell ageing has an impact on cell capacity (e.g., reducing the overall range of a battery electric vehicle) but also may affect the state of function (SOF) by reducing the maximum specified discharge current (e.g., worsen the driving dynamics of the electric vehicle). A lot of research has been done on identifying cell ageing mechanisms [4][5]. Mainly, cell aging is influenced by cycling and temperature besides calendaric ageing. It is commonly found in literature, that various chemical and physical processes lead to capacity fade or increased series resistance caused by primary loss of cyclable lithium and secondary loss of active material [6]. Cyclable lithium may be lost during side reactions inside the battery or during growth of the solid electrolyte interphase (SEI). In addition, usable active material may be reduced by mechanical effects (e.g., cracks of the graphite anode), or passive layers (e.g., lithium plating at low temperatures) [6]. Further it is shown, that battery cell ageing is accelerated with rising temperature [4]. In addition, Waldmann, et al. [7] found a linear correlation between cycling temperature of lithium-ion batteries and their ageing rate.

As ageing caused by cycling and calendaric ageing affects all series connected cells equally, only temperature dependent ageing may cause inhomogeneous cell ageing inside one battery pack. While the overall performance of the battery pack is limited by the worst single battery cell, the battery system design focuses on temperature and cell ageing homogenization in order to achieve an improved long-term performance.

In comparison to state-of-the-art approaches using active cell balancing, the solution proposed in this paper reduces costs, as the active balancing electronics has a high bill of materials (BOM) and must be individually adapted to every change in cell configuration. A purely passive thermal management approach is highly modular and thus reducing development time and costs.

III. MODULE CONCEPT WITH PRISMATIC CELLS

As a demonstrator, a liquid cooled module with 24s1p configuration and prismatic battery cells is selected. Plated aluminum bus-bars are laser beam welded and connect the cells in series. The bended cooling sheets with plain bearings are attached to tie rods as pictured in Figure 1:

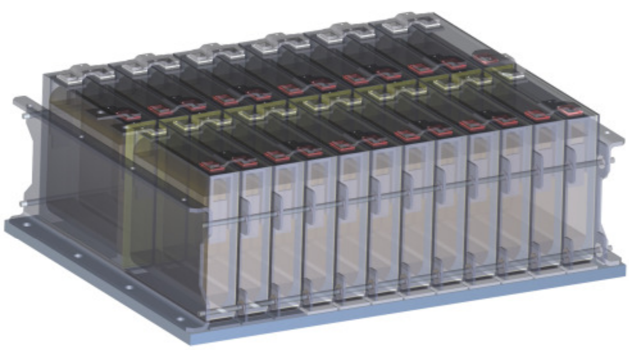

Figure 1: Battery module concept (24s1p) in transparent view

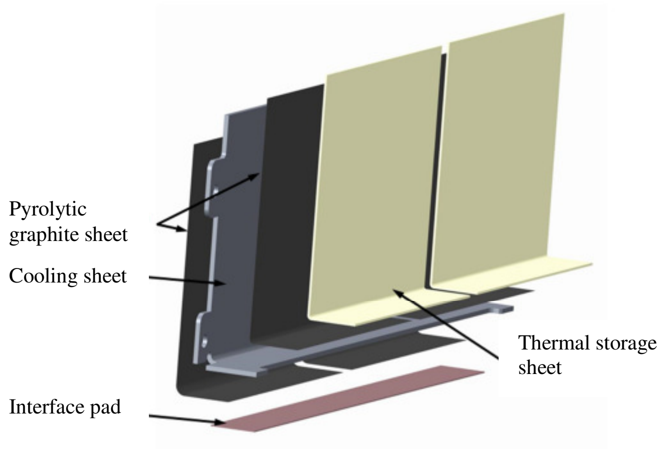

Figure 2: Cooling sheet assembly in exploded view

Figure 2 describes the cooling sheet assembly including interface pads, thermal storage sheets (TSS) and graphite sheets. The interface pads as conformable materials ensure the thermal connection between battery cell housings and cooling plate by compensating geometrical tolerances of the components. Thermal resistances due to air gaps and interfacial resistances are reduced. Common gap filler pads are based on silicone elastomers filled with ceramics to improve the thermal conductivity. In comparison to the other cooling parts made of aluminum, they have a lower thermal conductivity but are necessary to reduce the contact resistances. To resolve this contradiction, their functionality is expanded by using pads with different adjustable thermal resistances for a homogenous temperature distribution. With the thermal conductivities as parameters, an optimal set for a homogenized module temperature distribution has to be identified by parametrical FEM simulations. The weight compared to a cooling sheet with standard materials is increased by 28 g per cooling sheet corresponding to 2.8% of cell weight on module level.

Thermal storage materials, for example paraffin or salt hydrates based phase change materials (PCM), are selected among other criteria by melting point, specific heat, volume change, cycling stability and latent fusion heat at phase transition. To counter the disadvantage of a low heat conductivity, these materials have to be combined with heat conducting materials, for example an expanded graphite carrier matrix or aluminum foam. In battery systems, potential designs with PCM are matrix blocks or sheets. In this paper the focus on module level is set on sheet design, as matrix blocks have the disadvantage of inhomogeneous heat distribution and a disadvantageous behavior regarding heat dissipation. For this application PCM with a phase transition temperature of 40°C and a latent heat of 120 J/g is selected and combined with a heat spreading graphite layer to reduce temperature fluctuations. The PCM layer with 1 mm thickness is adhered to the inner surface of the cooling sheets in direct contact to the prismatic cell housing.

Synthetic pyrolytic graphite sheets (PGS) are made of graphite in a crystalline structure that is produced by heat decomposition of a graphite polymeric film. In comparison to natural graphite sheets synthetic materials have an improved in-plane thermal conductivity that is 2-5 times the value of copper. PGS is available in different sheet thicknesses; thinner sheets have an increased orthotropic thermal conductivity but allow a lower heat flux. To overcome this issue, several PGS layers are combined by adhesive films to a multi-layer sheet. It should be noted that multi-layer graphite sheets have lower through-plane conductivities and the bendability is limited due to bending stiffness and detaching of the single films. While providing potential in heat transport and spreading in battery modules, graphite as an electrical conductor has to be combined with electrically insulating materials. For this application a 4-layer system with a thickness of 17 µm each with an in-plane conductivity of 1850 W/(mK) is selected. Combined with an electrically insulating adhesive film it is attached to the outer surface of the cooling sheet. It provides a thermal connection between cooling plate and cell housing.

IV. THEORETICAL CONSIDERATIONS

Basis of the simulation is the heat balance equation of the battery cell including reversible and irreversible heat generation, convection and radiation [8]:

$$mc_p \frac{dT}{dt} = V_t I(t) - \frac{I(t) T \Delta S(t)}{nF} - hA(T - T_a) - \sigma \varepsilon A(T^4 - T_a^4) \tag{1}$$

where m is the mass of the cell [g], c_p the specific heat [J/g], T the temperature [K], t the time [s], V_t the over voltage [V], I the current [A], ΔS the change in entropy [J/K] and n is the number of the transferred electrons in the electrochemical reaction. F is the Faraday constant, h the heat transfer coefficient [W/m²K], T_a the ambient temperature [K], σ the Boltzmann constant, ε the emissivity coefficient and A the surface area [m²].

In this thermal model, the rise in temperature is generated by internal resistances and thermodynamic processes on the electrode. While the irreversible heat generation rate is dependent on state of charge (SOC) and temperature, the reversible contribution due to the entropy changes depends on the electrode materials. The over voltage is defined as the voltage difference between the terminal voltage V(t) and the cell equilibrium voltage $V_{eq}(t)$ of the relaxed cell:

$$V_t(t) = V(t) - V_{eq}(t) \tag{2}$$

The equilibrium voltage shows a hysteresis behavior and is asymmetrical for charging and discharging. The internal resistances are caused by charge transfer reactions, diffusion of ions in electrolyte and electrodes, and ohmic resistances. Another contribution to the heat balance is the entropic heat TΔS due to lithium intercalation in cathodes and anodes [9]. Further contributions in the heat balance equation are heat emission due to convection set by the heat transfer coefficient and radiation depending on the surface material.

The heat generation rate is determined by cycling tests of a battery cell at different temperatures and discharge rates, where the equilibrium voltage is calculated by extrapolation to a zero discharge current. The entropic change is dependent on the temperature derivative of the equilibrium voltage referred to the SOC.

$$\Delta S(t) = nF \left(\frac{dE_{eq}(t)}{dT} \right)_{SOC} \tag{3}$$

Besides thermal considerations, simple material models have to be developed. Multilayer materials such as the electrode stack are simplified to equivalent anisotropic homogenous models. Their volume averaged properties are calculated based on validated literature values or determined by measurements. The specific heat c_p [J/g] result with

$$\rho C_p = \frac{\sum_i \rho_i c_{p\,i} d_i}{\sum_i d_i} \tag{4}$$

where ρ_i, d_i and c_{pi} are the density [kg/m³], the thickness [m] and the specific heat [J/g] of the layers. Another example for modeling is the heat store material which is implemented as a two layer body. The thermal behavior of the PCM layer is modeled as a temperature dependent change in specific heat. The PGS layer is divided in several parts with orthotropic thermal conductivities which are linked to local coordinate systems in the simulation set up.

V. SIMULATION METHOD

Besides the SOC and temperature dependent heat generation rate, ambient conditions and material data, the thermal simulation set up requires a FEM capable CAD model. A suitable CAD model is able to reproduce the thermal behavior despite having relatively simple model geometry. Complex models will result in meshing problems and long calculation times, especially in transient simulations. Simplifications have to be made which are mainly based on experience and trial and error. The development of these models is an iterative process of optimization to find a trade-off between model complexity and accordance to measurements.

The simulation model of the battery cell containing the theoretical heat generation rate and the geometrical and material parameters is verified with a test set-up as seen in Figure 3. The temperature progression of a 4C discharge cycle is recorded by thermocouples attached to the battery cell housing.

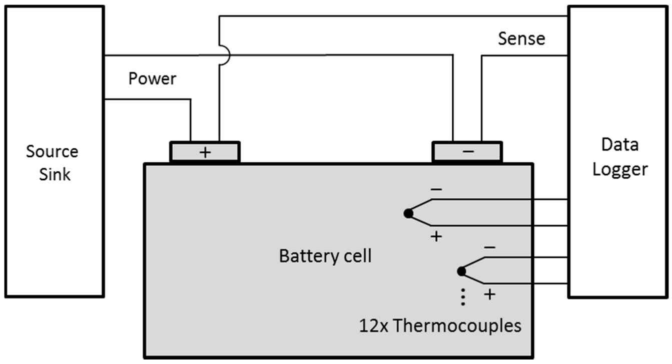

Figure 3: Test set-up for simulation cell model verification

Figure 4 shows the arithmetic averaged absolute deviations at different timesteps between the measured values and the results of a transient simulation of one battery cell.

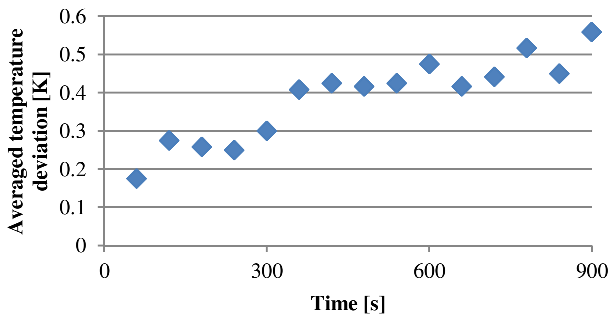

Figure 4: Arithmetic averaged deviation between measurements and simulated data

Software used for the simulation model set up is PTC CREO 2.0, ANSYS Mechanical 15.0, MOR for ANSYS and Python 2.5. The solver runs on a machine with 2x Intel(R) Xeon E5-2667v2 CPU @ 3.40GHz with CentOS 6.5 64 Bit and 512 GB RAM. All simulation set ups include transient thermal simulations of the model, which are further used for design exploration and parametrical optimizations. Aims of the thermal simulations are the validation of the design concept and the material property optimizations with focus on temperature homogenization. The electro thermal coupling of a simulation model describing a high capacity lithium-ion battery system is introduced in [10].

Design exploration techniques as iterative processes demand a high simulation effort and lead to long calculation times. MOR is used to speed up the development of the simulation models [11]. MOR is a mathematical method, used for numerical simulations in different fields of application, aiming at finding an approximated system of lower order and reducing the simulation computing time. If a model is reduced with MOR, the description of this dynamic system, represented in state space formulation of linear first order differential equations, is reduced to a system with lower dimension that can be solved in a shorter time.

In order to determine the thermal parameters of the interface pads optimization techniques are used to achieve a homogenized temperature distribution over battery module. Constraints are set to define a design space of the thermal conductivity values of the pads between a theoretical minimum of 0 W/(mK) and a maximum of 15 W/(mK). Within this limited design space sampling points are located by the design of experiments (DOE) algorithm central composite design (CCD). Then a response surface, the approximation of the system response that is later used for the optimization, is derived by nonparametric regression. When the response surface is validated by comparison with simulated design points, it is further investigated by a goal driven optimization. The optimization method multi objective genetic algorithm (MOGA) is applied, that supports several objectives and constraints in search for a global optimum. In this case two equally weighted objectives are defined, first the minimization of the maximum temperature and secondly the minimization of the temperature difference. As result, a parameter set for a homogeneous temperature distribution is obtained.

VI. Results

A. *Optimized interface pads*

For evaluation, a reference model is set up, in which the optimized pads are replaced by common high thermal conductive gap filler pads. Due to the double symmetry of the battery module only six cells are considered for a more clearly evaluation. In Figure 5, the temperature distributions for both models are shown along a construction path through the models at the last time step after heat generation, which is corresponding to a 4C discharge process. This path passes the hot spots of the battery cells which are located above the center of the cells between the terminals.

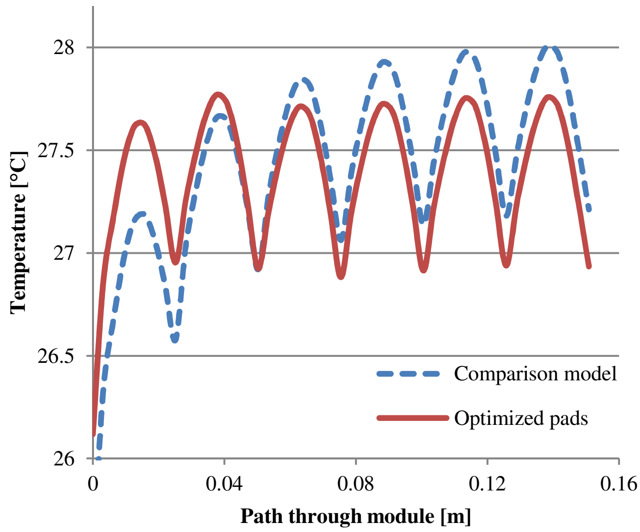

Figure 5: Temperature graph through the battery module after discharging

The optimized pads increase the temperature of the outer cells, while reducing it for the cells in the middle of the module. The maximum temperature difference along the path is reduced by 34% in simulation. In Figure 6 the temperature distribution of the optimized and reference model are illustrated. The internal cell stacks are highlighted and show a more uniform temperature distribution for the optimized model.

expected, PCM leads to a transient temperature homogenization but it slightly increases the maximum temperature difference over the module. The second effect is reduced by the optimized interface pads and the PGS.

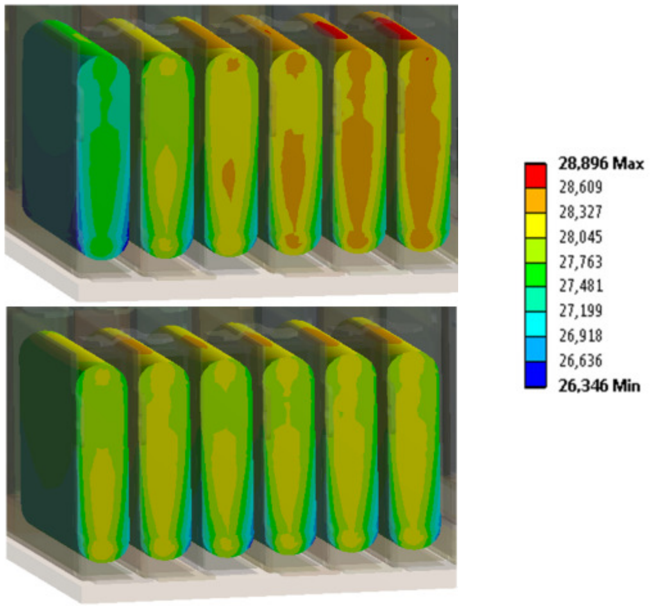

Figure 6: Comparison of temperature distribution [°C] in the cell stacks without (top) and with optimized interface pads (bottom)

B. Thermal storage sheets (TSS)

PCM is used to smooth the temperature curves and reduce peaks. As previously mentioned, the temperature range besides phase transition is critical for temperature homogenization. Therefore the simulation considers the operating range below the phase transition temperature of 40°C. As PCM sheets are also providing a thermal connection between cell housing and cooling sheets, a reference model with foam sheets instead of the thermal storage material is created. A heat conducting micro cellular flame retardant polyurethane foam is applied, as used in common energy storage applications. The discharge process is followed by a cool down phase without heat generation or liquid cooling. In Figure 7 the development of the minimum and maximum temperatures of the cell stacks are figured for both models.

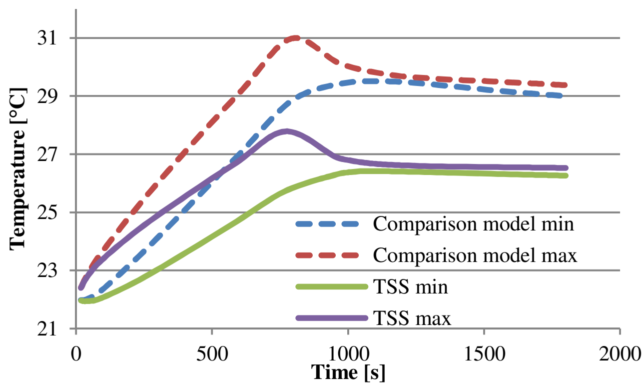

Figure 7: Influence of thermal storage sheets on temperature development

The average temperature and the entire temperature range are significantly reduced by the thermal storage material. As

C. Pyrolytic graphite sheets (PGS)

In this simulation, the influence of PGS on temperature homogenization is investigated. In Figure 8 the maximum temperature difference of the battery module is plotted over time. Again a structurally identical reference model, but without PGS, serves as a reference. The internal heat generation is applied to the cell stacks and corresponds to a 4C and 8C discharge current.

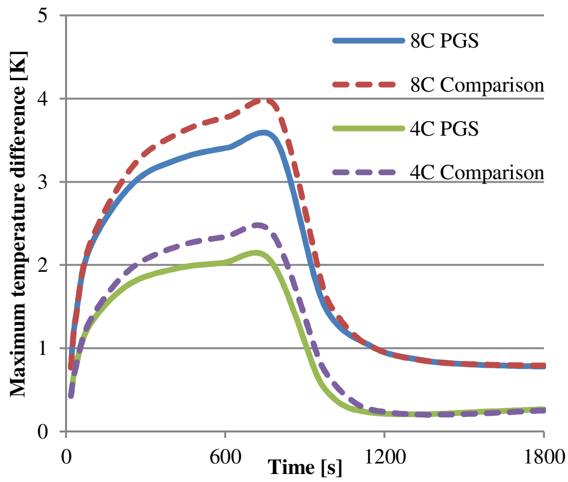

Figure 8: Maximum temperature difference of the cell stacks

PGS increases the heat flow along the cooling sheets and homogenize the temperature distribution of the cell housings and the thermal storage sheets. Temperature differences between the cell stacks are reduced. Higher current rates lead to increased temperature differences. The maximum temperature can be reduced with PGS in both discharge simulations.

VII. CONCLUSION

A concept based on enhanced cooling sheets with optimized interface pads, heat store material and multilayer pyrolytic graphite sheets has been developed to homogenize the temperature distribution in battery modules. Several FEM simulation models were set up including CAD and material models to evaluate and optimize the concept. The heat generation rate for a prismatic cell, corresponding to the reversible and irreversible heat generation, was calculated from the extrapolated, SOC and temperature dependent equilibrium voltage. By the use of model-order-reduction the iterative process of the simulation model development was shortened. With exploration techniques the design was optimized with regard to a uniform temperature distribution. The effects on temperature homogenization were evaluated with reference models on simulation level. It was shown, that the investigated design improvements have a positive influence on temperature distribution and thereby a beneficial effect on the lifetime of the battery module. The transient temperature development was homogenized with phase-change-material, while the temperature distribution over the module is minimized with optimized interface pads and pyrolytic graphite sheets. As next steps, the simulation results have to be evaluated by measurements on a prototype with accordingly materials. The discharge cycles are substituted by a driving cycle and a test set-up for measuring the temperature distribution will be developed. Furthermore the investigations of passive thermal management are expanded to module concepts with other cell types.

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