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Electromobility Optimized Heat Dissipation from Energy Storage Systems of Electric Vehicles

Optimized Heat Dissipation of Energy Storage Systems

The quality of the heat dissipation from batteries towards the outer casing has a strong impact on the performance and life of an electric vehicle. The heat conduction path between battery module and cooling system is realized in series production electric vehicles by means of paste-like materials. These so-called gap fillers exhibit high thermal conductivity and specific mechanical properties. The aim of an ongoing BMWi research project is to develop a new generation of gap fillers with improved thermal conductivity and reduced density and to qualify them for use in series production.

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The Audi E-tron convinces with the performance of its electric drive. While other electric vehicles electronically throttle down after short intervals with maximum power output to prevent the battery from overheating, the battery of the E-tron maintains its specific operating temperature thanks to a sophisticated cooling system. To make this possible, Audi has designed and implemented a comprehensive thermal management system.

Significance of the gap filler

The gap filler takes over the function of heat conduction and tolerance compensation between battery module and battery housing. During the assembly of the battery, the gap filler is applied into the compartments of the battery tray by a robot (*Figure 1*). The battery modules are then inserted. During this movement, the gap filler is effectively pressed against the surface to be wetted, and great care must be taken not to damage the pressure-sensitive battery cells. The flow properties of the gap filler are therefore formulated in such a way that resulting pressing forces prevent any damage to the battery. At the same time, the gap between the battery and the casing ground must be completely filled without air bubbles. This concept has already been in series production since 2018, and to date, over 50,000 vehicles have been produced and sold.

Like the combustion engine in conventional cars, the battery is a core component of electric vehicles that significantly determines its price. Due to the operational reliability, all current concepts assume that the individual battery cells can be integrated into a lightweight battery housing. The battery housing is advantageously completely integrated into the vehicle body shell lower as much as possible the center of gravity of the vehicle. For safety reasons, the water- cooling system is placed underneath the battery modules to ensure that the lithiumion cells are not exposed to water, even during a crash.

Pure polymer-based resins or pastes have an intrinsically low thermal conductivity, so they must be filled with particles of higher thermal conductivity for efficient heat dissipation. At present, ceramic particles, e.g. made of aluminum oxide, are used in particular, whereby high degrees of filling is aimed at to achieve maximum thermal conductivity.

Furthermore, pastes available today only achieve thermal conductivities of up to 3 W/($m\cdot K$), two orders of magnitude below the thermal conductivities of, for example, aluminum with 220 W/($m\cdot K$). In addition to the limited thermal conductivity, the following conflicts with the

requirements in automotive engineering arise when using current gap fillers:

- Highly filled pastes have a high density, and thus add significant weight, which conflicts with lightweight construction.
- 2. Their high flow resistance during application, due to their high viscosity, slow down the process. This contradicts a cost-efficient and fast production.
- 3. The high filler content severely wears the processing machines. Abrasion increases, and dosing machine components have to be replaced more frequently.
- 4. There is a lack of adapted concepts for automated serial processing of pastes. Previous concepts were designed for areas of several mm² in the electronics sector. In battery production, how-

ever, areas in the order of cm² must be processed.

5. Artificially synthetic spherical fillers allow for improved processability at high filling levels. However, these are currently not available at competitive prices.

The OWES research project

The OWES project (in German: Optimierte Wärmeableitung aus Energiespeichern für Serien-Elektrofahrzeuge; translated Optimized Heat Dissipation from Energy Storage Systems for Series Production Electric Vehicles), led by Audi, combines material science and production engineering research and development to focus on:

- Optimization of existing gap filler concepts through improved fillers,
- Use of alternative (cheaper, lighter) filler materials.
- New innovative heat dissipation concepts,
- Development of test and simulation methods by which materials can be assessed in terms of suitability for the manufacturing process (squeeze flow, maximum pressing forces, complete filling, thermal conductivity, abrasion, etc.),
- Adaptation of actual processes for highly filled pastes required for series production, eventually the development of new, innovative processes, such as gap filler injection.

The cooperation within the project consortium - consisting of the material manufacturers Wacker Chemie and Polytec PT, the filler supplier Quarzwerke, the equipment manufacturer Atlas Copco IAS, and the research institute Fraunhofer IFAM is sketched in Figure 2.

Challenge in the formulation of gap fillers

High thermal conductivities in gap fillers, if compared to the very low values of typical polymers ~ 0.2 W/(m·K), are achieved by adding large quantities of particles. Aluminum oxide with moderate thermal conductivity and high density (4 g/cm³) is mostly used as ceramic particles, although materials with significantly better thermal conductivity, and lower density, are available, e.g. boron or aluminum nitride. However, these fillers lead to high material costs. In order to achieve a significant thermal conductivity increase with currently available par-



Figure 1 > Before inserting the cell modules into the compartment floor, a heat-conducting gap filler is inserted.

ticles, very high filling degree must be achieved, as shown schematically in Figure 3. This makes it is necessary to reconsider density, viscosity, compressibility and thermal conductivity.

Further development of ceramic fillers

The OWES project resulted in significant further development of aluminum oxide based gap fillers. The significant improvements of thermal conductivity while largely maintaining their pressability can safely be considered as very successful that largely exceeded the targets initially set. Due to the close cooperation between the filler manufacturer and the gap filler manufacturer, thermal conductivities almost doubled from below 3 W/($m \cdot K$) at the beginning of the project to 5 $W/(m \cdot K)$ at its end. All this was achieved without negatively impacting processing properties. Cornerstones of the development were improved sphericity of the particles, a narrower size distribution, and surface coatings of the particles individually tailored to each polymer matrix. These materials are currently being comprehensively characterized at the Fraunhofer IFAM with regard to their processing properties and then processed on near-series manufacturing systems.

Aluminum based fillers

In the thermal interface materials (TIM) for high performance applications, e.g. for microelectronics, where cost plays little role, silver filler are used. For automotive applications, with significant more volumes, silver can be substituted by the much less expensive aluminum, in particular for its low density (2.7 g/cm³), and its excellent thermal conductivity that exceeds that of aluminum oxide by a factor of 7.

When aluminum powders were used, requirements regarding the processability of the pastes limited the filling degree to 60 % by volume, resulting in maximum thermal conductivity of ~ 2 W/(m·K). The project has shown that the bottleneck for good thermal conductivity is the high thermal transfer resistance of aluminum to the polymer matrix. Comprehensive simulations have shown that the use of aluminum leads to high thermal conductivities only for percolating structures, such as vertical rod structures. In these structures, the unfavorable contact resistance only occurs when entering and leaving the structure. The structures must be filigree and limited in the degree of filling to ensure the compressibility of the TIM.



Figure 2 > Cooperation in the OWES project



Figure 3 > Influence of the degree of filling on density, thermal conductivity and viscosity on conventional materials (dashed) and improved materials targeted in the project (coloured)

Carbon based fillers

By focusing on sophisticated nanotubes and graphenes, it is easy to miss that ordinary graphite also has excellent thermal conductivity, and that composite materials with very high thermal conductivities can be produced from graphite. For example, composites made of synthetic graphite in a polypropylene matrix are reported to achieve thermal conductivities of over 20 W/(m·K) at a moderate filling degree of 30 % by volume [1]. Another advantage is that graphite, like almost carbon materials, has a low density (2.3 g/cm³), limiting the weight of corresponding gap fillers.



Figure 4 > Comparison of the thermal conductivities when replacing aluminium oxide by adding graphite (top) and corresponding densities of the mixtures (bottom)

The used of nanoscale graphite as a filler leads to a very strong increase in viscosity, regardless of the low viscosity of the polymer matrix of only 200 mPa·s. Accordingly, for processability reasons, filling levels are limited to a max. of ~6.5 % by volume and only limited thermal conductivities of max. 1.8 W/($m \cdot K$) could be achieved. Substitution with spherical graphite (particle sizes of 30 µm) allowed to increase the filling ratios up to 65 vol.-%; however, the thermal conductivity remained below 1.5 W/mK. In a further approach, the aim was to produce 'hybrid systems' in existing AlOx-filled gap fillers by adding graphite, in which the heat transfer between the particles is improved.

Carbon-AlOx hybrid based filler

Graphite-AlO_x hybrid systems were prepared, whereby 0, 1 and 2 mass-% (corresponding to 0, 0.43 and 0.87 vol.-%) graphite were added to AlO_x formulations with 50, 60 and 70 vol.%. When 10 vol.-% AlO_x was substituted by 0.43 vol.-% graphite, thermal conductivity was fully retained, as can be seen in Figure 4 (top picture). The processing properties also remained almost the same. For a 50 vol.-% AlOx formulation, the thermal conductivity could be more than doubled by adding 0.87 vol.-% graphite. In addition to the lower cost of these materials, the reduction in density illustrated in Figure 4 (bottom picture) has a positive effect on the overall weight of the vehicle is also worth mentioning. Further efforts are currently being made in the OWES project using percolation effects in carbon-based fillers for improved heat transfer.

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Manufacturing properties

In the series production process at Audi, applied beads of gap fillers with high yield points are pressed when the battery is mounted. The processing properties of the gap fillers currently used are specifically adapted to this process. During assembly, the specific large-area pattern of the gap filler application is pressed by lowering the battery into small gaps. During pressing, the gap filler is squeezed through a narrowing gap. The fluid squeeze flow of the highly viscous polymer system creates a force against the lowering of the battery in form of a pressure on the bottom of the battery that peaks in the center of the contact surface. For industrial production, this process must be adjusted so that

- 1. Local pressure does not damage the pressure sensitive battery,
- 2. The pattern of the bead application leads to a complete, even, and bubblefree gap filling and
- 3. Required cycle times and tolerances are achieved.

In order to avoid costly tests on assemblies during the material selection and qualification phase, new characterization methods to determine specific material characteristics have been developed from 2015 onwards. The one developed for determining resistance to squeeze flow has now been established as a factory standard at Audi. Corresponding material parameters are gathered by Fraunhofer IFAM, Audi, or provided by the material suppliers. These characteristic values proved to be extremely helpful in the screening for gap fillers with suitable property profiles.

Simulation of squeezing flows

Squeeze flow is a well-known topic to bonding technology: it generally occurs when joining substrates. The pressure in the adhesive can increase unexpectedly when squeezing small gap heights. In investigations of the Fraunhofer IFAM on hybrid joining [2],[3],[4],[5], coupled fluid structure interaction simulations (FSI) based on the 'volume-of-fluid method' (VOF) have shown that the pressure in the adhesive can be sufficiently high to lead to plastic deformation of metallic substrates. In the FSI simulations, it has proven to be advantageous to calculate the flow of the fluid on a stationary (Euler) grid and the deformation of the joined parts on a materialfixed (Lagrange) grid. The publications on



Figure 5 > The squeeze flow experiment is used to predict the force and the pressure when squeezing gap fillers (purple).



Figure 6 > Simulation of a squeeze flow experiment on the basis of rheological data determined by a fluid structure simulation (FSI); the coloring corresponds to the degree of filling according to the 'volume-offluid method' (VOF)

hybrid joining cited above, a squeeze flow test was introduced as a suitable measuring method for determining the reaction force of the adhesive against a pressing action.

The squeeze flow experiment

In the squeeze flow experiment, a rotationally symmetrical punch with diameter D is mounted such to be axially movable (*Figure 5*). The gap between the underside of the punch and a plane surface is filled with the polymer material. In tests where the punch has been lowered at a linear speed v, the force F(h) occurring during the axial movement of the punch and the height of the gap h(t) are simultaneously measured. The movement of the punch causes a radial squeezing of the adhesive out of the gap. The maximum hydrostatic pressure in the adhesive occurs on the rotational axis (r=0). The exact parallelism of the punch and the plane surface are decisive for the measuring quality, as are the speed control and the very precise measurement of the gap height h(t).

The squeeze-flow experiment has also been established [2],[3],[4],[5] as a method to validate material models for the simulation of the gap flow during squeezing processes. Due to the simple, axial-symmetrical geometry, variants can be calcu-



Figure 7 > Experiment on the deflection of a metal sheet by the flow of a pink gap filler (left) and comparison with the simulation (right)

lated at high computing speed, and compared with experimental results. The material models for the simulation of the polymer fluid are determined based on rheological measurements. Depending on the complexity of the flow behavior of the fluid, different rheological procedures and measuring methods are used. For example, shear rate dependence, yield point, curing, temperature dependence as well as the visco-elastic and thixotropic behavior of the fluid polymer are to be determined.

Fluid structure interaction simulations

The simulation of the squeeze flow experiment can be carried out in different ways. The simulation method, and the simulation software, must be chosen in such a way that all relevant physical effects and parameters of the planned industrial application can be described adequately. For the simulation of the squeeze flow processes of gap filler material, software has to be used which offers the possibility of FSI to couple flow simulation and structure simulation. Thus, both the squeezing flow of the gap filler, by lowering the battery, and the elastic deformation of the battery by the hydrostatic pressure of the gap filler must be fully described.

Figure 6 shows a cylinder sector of a threedimensional FSI simulation based on the VOF method from the Fraunhofer IFAM. One can see the plunger (grey) lowering against the working surface (face) at gap heights of h_0 (initial), h_2 and h_3 . The fluid gap filler is shown between punch and plane surface. The gap filler is modeled on a fixed grid using the VOF method on an Euler grid. Red indicates that the respective volume cell is completely filled with gap filler, yellow and green colors indicate an incomplete filling thereof. Cells without gap fillers are blanked and border cells are cut. The illustration shows that the gap filler is being pushed out of the gap when approaching the small gap



Figure 8 > Comparison of experiment and simulation at component level

heights h_2 and h_3 . Currently, such numerical calculations for complex industrial problems require huge computational power, however, there are currently approaches [6] that aim at significantly reduced modeling effort.

Simulation of the battery assembly process

Rheological parameters and the squeeze flow test allow the comparison of different gap fillers. For the design of the production process of high-voltage batteries, however, it is necessary to know exactly the forces occurring during the setting process of the modules and to adjust the application pattern of the gap filler beads in such a way that an optimal, bubblefree filling of the gap between battery and ground is achieved. Audi uses two different simulation methods for this purpose, which relate to the following product geometries:

- bodies with medium gap height and flow paths, but complex adjacent component structure and
- cavities with extremely small gap heights and long flow paths due to the module size.

Both simulation methods are based on FSI. They describe interactions between the deformation of the structural components, and the flow forces, when the gap filler is squeezed. The mechanical simulation models stiffness and strength of the components using a variety of high-performance contact elements. The fluid simulation is based on material models resulting from the rheological measurements of the gap fillers, and uses the VOF method. With the simulations, the pressing of the gap filler – starting from the applied beads until the gap is completely filled – is exactly reproduced.

Initially, these simulation methods were successfully tested and verified on small test components (*Figure 7*). Before being transferred to real components, elaborate test setups in the scale of the battery were used for a further adjustment, and a final validation were carried out. *Figure 8* shows the setup in the size of a battery compartment with deformable structural parts, equipped with a large number of sensors to record the internal state of the cavity and the process variables. The aim of this step-by-step procedure was to detect any scale-related numerical effects, and if necessary, to exclude any sources



Figure 9 > Importance of the simulation of a complete component for industrial production

of error and inaccuracies, since the quality of the simulation methods can be evaluated in detail with the step-by-step procedure. For simulations with a high prediction quality, no 'recalibrations to fit the real world' have to be made in the validation process. The simulation method developed by Audi showed a very high prediction quality so that the release for industrial use could be successfully made at component level.

Industrial use of the simulations

Audi has been successfully using these simulations in industrial processes for several years now to determine limits for materials at an early stage. Global structures are calculated without any problems. A major advantage of the newly developed method is that there are virtually no limits to the numeric parallelizability, and thus a large number of variants can be simulated in just a few days, from which the optimum process and product parameters can be derived. The verified simulations of the total component show a very high quality of prediction.

Figure 9 summarizes the Audi simulation concept. Based upon the pressure distribution at the module bottom, it is possible to assess whether there is an impermissible load and within what time the pressure is released. The deformation of the structural components is also calculated in order to assess whether the deformation is elastic or plastic. The degree of wetting by the gap filler has a significant influence on the product properties, as this is decisive for the efficiency of the cooling. Due to the high prediction quality

and computing speed of the simulations, cycle time and limit values of the production can be predicted exactly.

Conclusions

Gap fillers are essential for temperature control of battery cells of any electrically powered vehicle. Besides thermal conductivity, cost and weight are increasingly playing an important role for the selection thereof. New approaches to optimize gap fillers are being developed and tested in the OWES project through adapted filler selection. In order to process the gap fillers in series production, the flow properties of the gap fillers must be precisely matched to the industrial process. The squeeze flow test, developed at Fraunhofer IFAM proved to be an excellent and reliable method for material qualification, as it effectively combined experimental and simulative methods to characterize and describe squeeze flow. Audi has been using coupled fluid structure simulations with high prediction quality for years to determine numerically the pressing of gap fillers during the assembly of battery cells. This full range of developed methods enables rapid and continuous optimization of material and process using sophisticated materials science and virtual tools. //

Note on funding and thanks

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References

J. King et al: Thermal Conductivity of Carbon-filled Polypropylene-based Resins. Journal of Composite Materials 44 (2010)
H. Fricke, T. Vallée: Numerical Modeling of Hybrid-bonded Joints. The Journal of Adhesion

92 (2016) 652-664

[3] H. Fricke, M. Israel: Unterschiedliche Werkstoffe prozesssicher verbinden-Simulation von Hybridfügeprozessen. Adhäsion Kleben u. Dichten 55 (2011) [7-8] 24-29

[4] M. Israel, H. Fricke: Qualitätssicherung beim Hybridfügen. Europäische Forschungsgesellschaft für Blechbearbeitung e. V. – Forschungsbericht Nr. 330 (2011)

[5] S. Niese, H. Fricke: Taschenbildung und Klebstoffverdrängung zwischen Hybridfügepunkten. Europäische Forschungsgesellschaft für Blechbearbeitung e.V. - Forschungsbericht Nr. 424 (2015)

[6] M. Müller, Y. Tong, H. Fricke, T. Vallée: An efficient numerical model for the evaluation of compression flow of high-viscosity adhesives. International Journal of Adhesion and Adhesives 85 (2018), 251-262

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