# HEAT LOSS MEASUREMENTS ON AN ENCLOSURE FOR HIGH TEMPERATURE BATTERIES

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### Summary

Heat loss measurements are reported for an enclosure which is suitable for high temperature batteries operating between 300 and 470 °C. The enclosure is rectangular and has a load-bearing vacuum insulation. The heat loss measurements are used to determine the thermal conductivity of the insulation. At 450 and 300 °C, a thermal conductivity of 3.2 and 2.0 mW/(m K), respectively, was determined. These values are smaller by a factor of about 20 than can be obtained with conventional (non-evacuated) thermal insulations. This is an important step towards the realization of electric vehicles driven by high temperature batteries.

# 1. Introduction

The two most promising candidates for advanced electric vehicle batter ies are the Na/S [1] and the LiAl/FeS [2] systems. They operate at 330 and 470 °C, respectively. In order to avoid unnecessary energy dissipation, the insulation of such a high temperature battery should be as efficient as possible. At the same time, the weight and volume of an enclosure have to be minimized. Conventional insulation materials such as Cerawool (a product of Johns Manville, Denver) have relatively high thermal conductivities,  $\lambda$  (about 60 mW/(m K)), thus requiring thick walls and heavy casings. Vacuum insulations (powder or multifoil) result in  $\lambda$ -values which are about one decade lower [3]. This would allow wall thicknesses of 3 cm and less for a heat loss of only 200 to 300 W for a 50 kW h vehicle battery. However, for engineering reasons, the use of evacuated insulations has so far been mainly limited to cylindrical enclosures, which are not very favourable for volume utilization by batteries. Therefore, efforts were made to manufacture a rectangular container with vacuum insulation capable of withstanding the atmospheric pressure acting on its shells ("load-bearing").

The Linde Division of Union Carbide Corp. [4, 5] succeeded in manufacturing such a casing by inserting a glass fiber board between the two shells. After several test runs, the casing was shipped to Argonne National Laboratory for testing. In the following, some of the thermal measurements will be reported.

The measured total heat losses can be used to determine the total thermal conductivity by calculation of all the different heat loss components through the wall material and the plug, from a measured distribution of temperatures inside and outside the casing. The method will be discussed in detail in ref. 6 and is applied in the present paper to the Linde vacuum insulation. Compared with standard methods which usually simulate an external load when measuring the  $\lambda$  of load-bearing insulations, the procedure applied here has the advantage that the atmospheric load is neither altered with time, if the insulation filling should be elastic, nor with temperature.

#### 2. Experimental

Figure 1 shows the casing and its dimensions. The weight is about 28 kg and the distance between the two shells is 25 mm. In order to maintain a good vacuum, even if small leaks occur, the unit is gettered with barium powder. The pressure at the beginning of the measurement was 2.8  $\mu$ mHg. During initial heat-up at Linde, small buckles formed in the inner shell. Although this would probably be of no consequence, a 450 mm plug made of Cerawool was inserted in order to prevent these buckles from enlarging. This resulted in the utilizable volume,  $V_0$ , being reduced to about 66 liters and the inner surface to 1 m<sup>2</sup>.

A heater was placed inside the enclosure, and the temperatures  $T_1$  (wall temperatures inside  $V_0$ ), and  $T_2$  at the outer shell were measured with thermocouples. Measurements were made when the enclosure had reached thermal equilibrium, which was attained after heating for more than 48 h. Data were taken as integrated values over a minimum time of 1 h.



Fig. 1. Linde battery enclosure (schematic). Dimensions are given in mm.  $\hat{Q}_0$  denotes the net heat loss from the heated area.  $\dot{Q}_1$ ,  $\dot{Q}_2$  and  $\dot{Q}_3$  are heat loss components through the wall material, the plug, and the thermal insulation of the enclosure in the neighbourhood of the plug, respectively.

Measurements of the total heat loss,  $\dot{Q}_{T}$ , were carried out at  $T_1 = 449$ , 361 and 300 °C. The vacuum dropped during the operation period (about 7 days) from 2.8 to 2.3  $\mu$ mHg.

In order to calculate the thermal conductivity,  $\lambda$ , we need the net heat loss,  $\dot{Q}_0$ , from the heated area of the inner shell to the outer shell of the battery container. From  $\dot{Q}_0$ ,  $\lambda$  can be extracted as a mean value averaged over the entire heated shell surface. Figure 1 shows, schematically, the heat loss  $\dot{Q}_1$  through the wall material surrounding the plug, the heat loss  $\dot{Q}_2$ through the plug material, and the heat loss  $\dot{Q}_3$  through the thermal insulation of the battery container in the neighbourhood of the plug. From these contributions, we have

$$\dot{Q}_0 = \dot{Q}_{\rm T} - (\dot{Q}_1 + \dot{Q}_2 + \dot{Q}_3).$$

Note that  $\dot{Q}_3$  is a function,  $\dot{Q}_3 = f(\lambda)$ , of the unknown thermal conductivity, which must be taken as  $\lambda(T'_m)$  at an appropriate mean temperature,  $T'_m$ , of the insulation in the neighbourhood of the plug. We also have

$$\dot{Q}_0 = \frac{\lambda(T_m'')D}{(F_m \,\Delta T_0)}$$

where D,  $F_{\rm m}$ ,  $T_{\rm m}''$  and  $\Delta T_0$  denote the thickness of the insulation, the mean surface of the heated area ( $V_0$ ), the mean temperature of the insulation within  $F_{\rm m}$ , and the temperature difference between  $T_1$  and  $T_2$ , respectively. From both relations for  $\dot{Q}_0$ ,  $\lambda$  is obtained at a temperature  $T_{\rm m}$ , which is the weighted mean of the temperatures  $T_{\rm m}'$  and  $T_{\rm m}''$  with respect to the corresponding mean surfaces. Using the  $\dot{Q}_{\rm T}$  measured at different  $T_1$ ,  $T_2$  and  $T_{\rm m}$ ,  $\lambda$ is determined as a function of temperature.

### 3. Results and discussion

In Table 1, the measured values of the total heat loss,  $\dot{Q}_{T}$ , are given for the different temperatures,  $T_1$ , of the inner shell within the heated area.  $\dot{Q}_1$ ,  $\dot{Q}_2$  and  $\dot{Q}_3$  were calculated from the measured temperature distributions of the inner and outer shells and of the ends of the plug. Using the relations for  $\dot{Q}_0$  given above,  $\dot{Q}_0$  and  $\lambda$  were determined and are listed in Table 1.

TABLE 1 Measured  $(T_1, \dot{Q}_T)$  and calculated data for the Linde enclosure

T <sub>1</sub> (°C)	Q <sub>T</sub> (W)	<b>Q</b> <sub>1</sub> (W)	<b>.</b> (W)	<b>Q</b> 3 (W)	<b>Q̂</b> ₀ (₩)	λ (mW/(m K))
449	88	10.9	7.0	20.1	50.0	3.2
361	54	8.7	5.6	11.7	28.0	2.3
300	40	7.1	4.6	8.0	20.3	2.0



Fig. 2. Heat loss of the Linde casing (1) and of a battery container with a load-bearing powder insulation (2) as a function of the difference between wall temperatures  $T_1$  and  $T_2$ .

Figure 2 shows experimental heat losses,  $\dot{Q}_0/A$  (A denotes unit area), obtained for the Linde vacuum insulation (curve 1) and, for comparison, those of a battery container of  $0.395 \times 0.344 \times 1.200 \text{ m}^3$  utilizable inner volume (curve 2) which was insulated with a microporous, evacuated powder filling (load-bearing, pressed plates from Grünzweig & Hartmann und Glasfaser AG., Ludwigshafen, FRG, prepared from a mixture of fumed silica, FeTiO<sub>3</sub> as opacifier, and glass fibers, using a pressure of about  $6 \times 10^5$  Pa). Taking into account the less thick Linde insulation, the specific heat losses of the Linde vacuum jacket are at least a factor of 3 less than those measured with the load-bearing, pressed plates.

In the diffusion approximation (see standard literature on radiative transfer, e.g., ref. 7), the function  $\lambda$  can be written as the algebraic sum of the solid conduction component,  $\lambda_{sc}$ , and the radiation part,  $\lambda_{Rad}$ , as a linear expression of the variable  $T^{*3}$ ;

$$\lambda = \lambda_{\rm SC} + aT^{*3} \tag{1}$$

where  $\lambda_{SC}$  is usually assumed to be independent of temperature and where  $T^*$  is a radiation temperature  $(T^{*3} = (T_1^2 + T_2^2)(T_1 + T_2))$ . This is valid because the fibers are highly dispersed, and the temperature difference between two neighbouring fibers is very small compared with the total  $\Delta T_0$ , and because the optical thickness of a compacted fiber board is usually high (see below). The component  $\lambda_{SC}$  is, in the  $(\lambda, T^{*3})$  plane, the intercept of  $\lambda$  with the ordinate at  $T^{*3} = 0$  (which implies that  $T_1 = 0$  and  $T_2 = 0$ ). If we measure  $\lambda$  at various  $T_1$  and  $T_2$ ,  $\lambda_{SC}$  can be determined from an extrapolation of  $\lambda$  can introduce serious errors only if the solid conduction of the material is very low or the transparency is high. However, in the case considered here, we have nearly equal amounts of solid conduction and radiation parts in the total  $\lambda$ , and the coefficient of total extinction, E, is high (see below).

Since the second term in eqn. (1) is the radiation part (assuming the real index of refraction n = 1)

$$\lambda_{\text{Rad}} = aT^{*3} = \frac{4\sigma T^{*3}}{3E}$$
(2)

we can extract not only the relative and absolute parts of solid conduction and radiation in the total  $\lambda$  but also the total extinction coefficient, E ( $\sigma$ denotes the radiation constant).

Figure 3 contains the data for  $\lambda$  measured by the heat flow meter method [5] (open circles) which follow, in the  $(\lambda, T^{*3})$  plane, a straight line, as required by the diffusion approximation. This finding demonstrates not only the applicability of the diffusion approximation for the radiative part of the total  $\lambda$ , but also justifies the assumption of a constant  $\lambda_{sc}$ . While data for  $\lambda$  obtained with a heat flow meter are usually of high accuracy concerning relative values, the absolute amount of  $\lambda$  is, however, subject to uncertainties in the calibration constants, the heat contact between samples and the meter, etc. Note that the data taken in ref. 5 were obtained using a rather small testing apparatus. Experimental errors in measurements of  $\lambda$ using a heat flow meter are thus frequently larger than 10% of the absolute values.

The  $\lambda$  values extracted from the net heat loss,  $\dot{Q}_0$ , of a closed surface do not suffer from calibration uncertainties but depend entirely on the easily detectable temperature distributions on the hot and cold shells and the geometry. The  $\lambda$  values determined by this method are given for the Linde vacuum insulation in Fig. 3 (crosses) for comparison. Assuming experimental errors of 10% in the Linde data and the  $\dot{Q}_T$ , it is seen from this Figure that the absolute amount of the Linde data is roughly confirmed by the  $\lambda$  value extracted from  $\dot{Q}_0$ . Note that the slope  $d\lambda/dT^{*3}$  is nearly equal for both measurements.



Fig. 3. Thermal conductivity as a function of the radiation temperature  $T^{*3}$ . O, Linde data obtained by the heat flow meter method for a glass fiber board (density 0.35 g/cm<sup>3</sup>, see ref. 5); —, least squares fit to the data; —, hypothetical error of the Linde data of 10%; X,  $\lambda$  values obtained from the net heat losses  $\dot{Q}_0$  of the Linde battery enclosure. Error bars are total errors estimated from Gauss' law of propagation of individual uncertainties  $\Delta \dot{Q}_{T} \Delta T_1$ , etc., using  $\Delta \dot{Q}_T = 10\%$ .

From the Linde or Argonne data, we have a total E of about  $2 \times 10^4$ 1/m and an optical thickness,  $\tau_0 = ED$ , of about 500. From both measurements, the solid conduction part,  $\lambda_{SC}$ , of the total  $\lambda$  can be estimated to  $\leq 1.5$  mW/(m K). The *E* value is of the order of those extinction coefficients which can be found for strongly opacified powder insulations of low density [3]. Its high value can be explained by the large backscattering cross sections usually found for glass fibers if the fiber diameter is between 1 and 5  $\mu$ m (see ref. 8), and by the higher density (about  $0.35 \text{ g/cm}^3$ ). Following the first order approximation for the total heat flux by conduction and radiation in an absorbing and anisotropically scattering medium made in ref. 9, a large ratio of back scattering to forward scattering reduces the radiative heat transfer by about 25% compared with isotropic scattering if the optical density of the system is high. A possible explanation of the small  $\lambda_{sc}$  value could be given by considering the method of preparation of the glass fiber board. If the glass fiber paper is heated to the strain point of the fiber material, which is higher than the operating temperature of the insulation, the fibers lose their internal stresses so that the potential stress energy reaches a minimum. When the temperature is lowered, the geometry of the fiber arrangement (several hundreds of layers aligned mostly perpendicular to the temperature gradient between the hot and cold shell) remains fixed, with a high Young's modulus, M, against a further deformation (compression). Since the thermal conductivity,  $\lambda_{Ct}$ , at the points of contact between fibers is, according to ref. 10, proportional to  $\lambda_s/M^{1/3}$  (where  $\lambda_s$  is the thermal conductivity of the solid material),  $\lambda_{Ct}$  and thus  $\lambda_{SC}$  decrease with increasing M. Accordingly, we obtain with a glass fiber board the low  $\lambda$  values typical of loose glass fibers even if the board is subject to a high external load.

For a 50 kW h Na/S battery, a box of dimensions  $0.36 \times 0.4 \times 2.5 \text{ m}^3$  (about 4.1 m<sup>2</sup> surface) will be needed. According to the  $\dot{Q}_0$  values given in Table 1, the heat loss at 360 °C for such a battery will be about 220 W. Therefore, if the temperature is maintained by discharging the battery for auxiliary heating, only about 10% of the stored electric energy would be consumed within 24 h. This is a value which is acceptable for practical applications.

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