

## **DETERMINATION OF THE HEAT CAPACITIES OF LITHIUM/BCX (BROMINE CHLORIDE IN THIONYL CHLORIDE) BATTERIES**

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### **Summary**

Heat capacities of twelve different Lithium/BCX (BrCl in thionyl chloride) batteries in sizes AA, C, D, and DD were determined. Procedures and measurement results are reported here. The procedure allowed simple, reproducible and precise determinations of heat capacities of industrially important Lithium/BCX cells, without interfering with performance of the cells. Use of aluminum standards allowed the accuracy of the measurements to be maintained. The measured heat capacities were within 5% of calculated heat capacity values.

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### **Introduction**

Lithium/BCX (bromine chloride in thionyl chloride) cells are the subject of much interest because of their use as commercial electrochemical cells [1 - 4], and as power sources in implantable medical devices [5 - 8]. The addition of bromine chloride to lithium/thionyl chloride batteries has resulted in improved low temperature performance, higher open circuit potentials [2, 4, 9, 10], and improved safety features for the cells, including decreasing the risk of explosion [1] often seen for early lithium cells [11 - 14]. The potential for widespread application of these cells has led to an interest in the physical properties of the cells and the various cell components. Spectroscopic studies have been undertaken on interhalogen compounds in organic solvents [15 - 17], as well as in  $\text{SOCl}_2$  [18]. Another area of particular interest is in the thermal properties of the Lithium/BCX system [4, 19 - 22], where it has been shown that measured heat capacities of batteries correlate well with the heat generated from cells discharged in an insulated environment [19]. This is of particular importance to the usage

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of these batteries in the space program [4, 19 - 22], where it is often necessary to estimate heat evolution and maximum probable cell temperature during discharge, and to define battery thermal dissipation requirements. A critical parameter for such estimates is the cell specific heat or heat capacity ( $C_p$ ).

The need for an accurate and precise, yet simple, method for determining heat capacities has led to the development of the method reported here. This method provides precise results, and through the use of standards for calibration, accurate results are also obtained for the batteries. Here we report the procedure for the determination of heat capacities for Lithium/BCX batteries, sizes AA, C, D, and DD.

## Experimental

### *Materials*

Reagent grade methanol (J. T. Baker Chemical Company or Fisher Scientific Company) was initially distilled over reagent grade calcium oxide (Fisher Scientific or Alfa Products) and then purified over magnesium metal (J. T. Baker or Fisher), under a nitrogen atmosphere. The methanol was recycled after the measurements were made by repeating the above procedure. Pure aluminum rods (99.999%, Alfa Products and Aesar) were chosen to approximate actual cell sizes. Rods of  $0.95 \times 5$  cm,  $2.2 \times 5$  cm,  $2.2 \times 6$  cm, and  $2.2 \times 9$  or 11 cm were used to approximate AA, C, D, and DD cells, respectively.

### *Equipment*

Methanol transfer was accomplished using volumetric delivery pipets. The methanol temperature was kept uniform throughout the Dewar flask (350 and 650 ml Dewar flasks, Pope Scientific Inc.) by using a magnetic stir bar. Temperature measurements were accomplished with a Fluke model 80TK thermocouple attached to a Heathkit model IM-2215 portable digital multimeter, providing temperature readings to  $\pm 0.1$  °C. A Haake model A80 refrigerated bath and circulator was used to maintain samples at the desired temperature for the experiment. Between measurements, the samples were stored in a freezer set at approximately  $-15$  °C.

### *Cell description*

Standard size AA, C, D, and DD cells were used for heat capacity determinations. Construction of all cell sizes was similar except for the dimensions of the components used. Anodes were constructed by pressing sheets of lithium on each side of nickel expanded metal grids equipped with leads. Cathodes consisted of Teflon-bonded-carbon placed on expanded metal grids. A separator was placed between the anode and cathode. The assembly was rolled and fitted into a stainless steel can which was then fitted with a lid containing a glass-to-metal seal. After lid-to-case welding, cells were vacuum filled with electrolyte, which consisted of lithium tetra-

chloroaluminate dissolved in thionyl chloride:bromine chloride in a six-to-one molar ratio. Cells were close welded to provide hermetic seals and fitted with fuses to avoid damage during handling.

### *Procedure for measurements*

Before measurements were taken, cells were stored in a freezer at approximately  $-15^{\circ}\text{C}$  for a minimum of 12 h, then placed into a refrigerated bath and circulator, set at  $-15.0 \pm 0.3^{\circ}\text{C}$ , for a minimum of 3 h. A Dewar flask (350 ml Dewar flasks were used for the AA, C, and D cells, and 650 ml Dewar flasks for the DD cells) containing a magnetic stir bar and a thermistor taped to the inside wall was filled with a known volume of dry, distilled methanol (150 ml for AA, 200 ml for C, 250 ml for D, and 400 ml for DD cells). The Dewar was stoppered and stirred for 5 min to allow for an even temperature distribution throughout the solvent, then the initial temperature of the methanol was recorded. The AA, C, and D size cells were transferred from the bath to the Dewar flask. After transfer of the cell, the Dewar was quickly stoppered to avoid heat loss. Stirring was continued throughout the entire experiment. A constant rate of stirring was found to be crucial to the success of the experiment. No stirring, or highly erratic stirring rates, resulted in a temperature gradient in the methanol, producing artificially low temperature readings.

The final temperature used for the calculations was the most stable temperature observed, where the temperature readings remained constant for a minimum of 3 min. The heat capacities of standard aluminum samples, with the approximate dimensions of the various cell sizes, were measured using the above procedure. These measurements were compared with National Bureau of Standards heat capacities [23] as a means of calibration for the system. The calculated difference between the NBS heat capacity and the observed heat capacity was the correction term ( $C_{p, \text{corr}}$ ) which accounts for the contributions of the other components of the system. Three data points were obtained for each cell, and six data points were collected for each standard. The data and results were analyzed using the appropriate techniques [24], and the reported heat capacities are the averages of the corrected values. Theoretical calculations of the heat capacities of the various cells were also made, based on the contribution of the major components of the cells\*.

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\*Theoretical heat capacities were calculated as follows  $C_{p, \text{theo}} = \sum C_{p, i}(m_i/m_t)$ , where  $C_{p, i}$  is the known heat capacities for the  $i$  individual components,  $m_i$  is the mass of the  $i$  individual components, and  $m_t$  is the total mass of the cell. The components used in the calculations were: the case, header and terminal cap (304 stainless steel); the anode and cathode current collectors, tabs and fuse assembly (nickel metal); the anode (lithium metal); the cathode (BrCl in  $\text{SOCl}_2$ ); and the carbon cathode (Shawinigan AB, Teflon binder). These components amounted to  $\approx 95\%$  of the total cell mass. All other components were treated as having an insignificant contribution to the heat capacity.

## Results and discussion

Based on the principles of thermochemistry [25], the definition of heat capacity at constant pressure, or specific heat, is described by eqn. (1):

$$nC_p = (\partial q)/(\partial T) \quad (1)$$

where  $C_p$  is the molar heat capacity,  $n$  is the number of moles of a compound,  $q$  is the heat, and  $T$  is the temperature of the system. Rearrangement and integration of eqn. (1) (where integration is interpreted as finite changes in conditions), produces eqn. (2):

$$\Delta q_{\text{system}} = nC_p(\Delta T_{\text{system}}) \quad (2)$$

where  $\Delta q_{\text{system}}$  is equal to zero for an adiabatic system. Since the entire system involves two components (the solvent system, which includes the methanol, stir bar, thermistor, and Dewar walls, and the metal system, which is either the battery or the standard), eqn. (2) becomes:

$$\Delta q_{\text{system}} = 0 = n_m C_{p,m} \Delta T_m - (n_s C_{p,\text{sol}} + C'_{p,\text{sys}}) \Delta T \quad (3)$$

where  $n_s C_{p,\text{sol}}$  is the contribution of the methanol, and  $C'_{p,\text{sys}}$  is the contribution of the remainder of the system. Equation (3) is equivalent to eqn. (4), showing that the change in the heat for the metal system is equal to the change in the heat for the solvent system:

$$n_m C_{p,m} \Delta T_m = (n_s C_{p,\text{sol}} + C'_{p,\text{sys}}) \Delta T \quad (4)$$

where  $m$  refers to the metal system,  $\text{sol}$  refers to the solvent, and  $\text{sys}$  refers to the Dewar system. The contributions of the system are then calculated from the determination of the heat capacities of the standards. Equation (4) can then be rearranged to solve for the heat capacity of the metal system, as shown in eqn. (5):

$$C_{p,m} = n_s C_{p,s} \Delta T_s / n_m \Delta T_m + C_{p,\text{corr}} \quad (5)$$

where  $n_m$  and  $n_s$  can be replaced by mass, or any other measured extensive property producing the heat capacity in corresponding units.  $C_{p,\text{corr}}$  is the difference between the NBS and observed heat capacities, and it is used to correct for the contribution of the other system components to the measured heat capacity. By using the sample mass, the heat capacity of methanol, the appropriate  $C_{p,\text{corr}}$  and calculating  $\Delta T$  (the difference between the initial temperatures for the metal and solvent systems, and the most stable temperature recorded after combining the two systems), one can obtain the heat capacity per gram of cell mass.

The results of the experiments with the aluminum standards are contained in Table 1. Aluminum was chosen as a standard because of the small difference in heat capacity of aluminum relative to the cells (0.216 compared with 0.200), as well as the availability of a wide range of sizes, which was analogous to the actual cell sizes. The results for the standards are shown to be precise, based on the small coefficients of variance ( $CV < 6\%$ );

TABLE 1

Experimentally determined heat capacities for aluminum standards of size similar to actual cell size, in cal g<sup>-1</sup> K<sup>-1</sup>, and comparison with their National Bureau of Standards (NBS) heat capacities<sup>a</sup>

Standard size	n <sup>b</sup>	Range	Average ± CI <sup>c</sup>	$\sigma_{n-1}$ <sup>d</sup>	CV <sup>e</sup>	Error (%) <sup>f</sup>	C <sub>p,corr</sub> <sup>g</sup>
AA	6	0.195 - 0.217	0.205 ± 0.013	0.011	5.3	8.7	0.019
C	6	0.177 - 0.188	0.182 ± 0.005	0.004	2.2	15.3	0.033
D	6	0.175 - 0.180	0.178 ± 0.002	0.002	1.2	17.4	0.037
DD	12	0.182 - 0.196	0.186 ± 0.002	0.003	1.9	13.8	0.030

<sup>a</sup>National Bureau of Standards heat capacity is obtained by taking the heat capacity per mole of aluminum metal [23] and dividing it by the atomic mass of the aluminum.

<sup>b</sup>n is the number of determinations.

<sup>c</sup>Average is the average ( $\bar{x}$ ) for the determinations, and CI is the confidence interval at a 95% level of confidence =  $\bar{x} \pm (t\sigma_{n-1})/\sqrt{n}$  where  $t$  is taken from a  $t$  table with  $n - 1$  degrees of freedom.

<sup>d</sup> $\sigma_{n-1}$  is the standard deviation with  $n - 1$  degrees of freedom.

<sup>e</sup>CV is the coefficient of variance (relative standard deviation) =  $\sigma_{n-1}/\bar{x} \times 100$ .

<sup>f</sup>Percent. error = |experimental value - NBS value|/NBS value  $\times 100\%$ .

<sup>g</sup>C<sub>p,corr</sub> is the difference between the NBS value and the experimental value, which was used to correct for the contribution of the Dewar system.

there is error in these measurements, however, when compared with the NBS heat capacities. This deviation is to be expected [26] due to the real contributions of the Dewar system, which are neglected before the appropriate correction term is added to the measured value. The magnitude of the correction term (C<sub>p,corr</sub>) appears to depend on the size of the stir bar, the Dewar size, the volume of methanol used, and the length of the thermistor exposed to the methanol. These dependencies would also be expected due to the varying amounts of materials which come into contact with the methanol (including increased exposure to the surface area of the Dewar, to a larger stir bar, and to an increased surface area of the thermistor) involved with each of the cell-size systems.

By using the previously described procedure and the above calculations, we were able to ascertain the heat capacities of a series of Lithium/BCX cells with a high degree of precision. Then, with the use of the appropriate correction terms (C<sub>p,corr</sub>) from Table 1, we were able to calibrate the system. Table 2 is a compilation of data collected for the AA, C, D, and DD cells after addition of the corresponding C<sub>p,corr</sub>. A minimum of three cells was used for each determination. The most reproducible results were obtained for DD cells, where  $\sigma_{n-1}$  ( $n - 1$  degrees of freedom) = 0.001. The larger size DD cells would be expected to have smaller deviations due to the higher cell mass:methanol mass ratio. This high ratio produces a larger temperature change,  $\Delta T$ , in the methanol, which results in a smaller relative uncertainty in the temperature change (uncertainty in the temperature measurement,

TABLE 2

Experimentally determined heat capacities, in cal g<sup>-1</sup> K<sup>-1</sup>, corrected for the contribution of the Dewar system,  $C_{p,corr}$ , for Lithium/BCX batteries, sizes AA, C, D, and DD

Cell size	$n^a$	Range	Average $\pm$ CI <sup>b</sup>	$\sigma_{n-1}^c$	CV <sup>d</sup>
AA-1	3	0.190 - 0.200	0.194 $\pm$ 0.012	0.005	2.6
AA-2	3	0.193 - 0.199	0.195 $\pm$ 0.007	0.003	1.4
AA-3	3	0.194 - 0.205	0.200 $\pm$ 0.015	0.006	2.8
Total AA	9	0.190 - 0.205	0.197 $\pm$ 0.007	0.005	2.5
C-1	3	0.207 - 0.214	0.210 $\pm$ 0.010	0.004	1.8
C-2	3	0.202 - 0.211	0.206 $\pm$ 0.015	0.006	3.1
C-3	3	0.197 - 0.209	0.203 $\pm$ 0.020	0.008	4.2
Total C	9	0.197 - 0.214	0.207 $\pm$ 0.007	0.005	2.4
D-1	3	0.202 - 0.211	0.205 $\pm$ 0.012	0.005	2.5
D-2	3	0.200 - 0.203	0.202 $\pm$ 0.005	0.002	0.8
D-3	3	0.200 - 0.207	0.204 $\pm$ 0.010	0.004	1.7
Total D	9	0.200 - 0.211	0.203 $\pm$ 0.003	0.004	1.7
DD-1	3	0.199 - 0.203	0.200 $\pm$ 0.005	0.002	1.2
DD-2	3	0.199 - 0.201	0.200 $\pm$ 0.003	0.001	0.5
DD-3	3	0.200 - 0.202	0.201 $\pm$ 0.003	0.001	0.6
Total DD	9	0.199 - 0.203	0.200 $\pm$ 0.001	0.001	0.7

<sup>a</sup> $n$  is the number of determinations.

<sup>b</sup>Average is the average ( $\bar{x}$ ) for the determinations, and CI is the confidence interval at a 95% level of confidence =  $\bar{x} \pm (t\sigma_{n-1})/\sqrt{n}$  where  $t$  is taken from a  $t$  table with  $n - 1$  degrees of freedom.

<sup>c</sup> $\sigma_{n-1}$  is the standard deviation with  $n - 1$  degrees of freedom.

<sup>d</sup>CV is the coefficient of variance (relative standard deviation) =  $\sigma_{n-1}/\bar{x} \times 100$ .

0.1 °C divided by  $\Delta T$  for the particular cell). The largest deviations were observed for the AA and C size cells, where  $\sigma_{n-1} = 0.005$ . This was also confirmed by results for the standards (see Table 1) where the largest standards were the most precise, while the smallest standards (AA and C sizes) were the least precise, owing to a larger relative uncertainty in the temperature changes. The largest source of error in the measurements is the uncertainty in the temperature measurements, where a difference of 0.1 °C will produce a 7% change in the heat capacity of an AA-size cell. The heat capacities of the different cell sizes were identical within experimental error, as would be expected, due to the constant composition of the cells which were also identical within experimental error.

The experimental heat capacities were compared with the calculated heat capacities (see footnote p. 271) and the results of this comparison are shown in Table 3. The two heat capacities differed by 5% or less, and for the DD cells the two values were identical within experimental error. These small differences support the accuracy of the results obtained by this method. Some of the differences may be due to the contribution of the components which were neglected in the calculation of the theoretical heat capacity (including the shrink wrap, insulators, and nylon spacers). Differ-

TABLE 3

A comparison of calculated heat capacities<sup>a</sup> for Lithium/BCX batteries, sizes AA, C, D, and DD, with the corresponding experimentally determined heat capacities, in cal g<sup>-1</sup> K<sup>-1</sup>

Cell size	Calculated <sup>a</sup> heat capacities	Average of experimental heat capacities	Difference <sup>b</sup> (%)	Active:passive ratio <sup>c</sup>
AA	0.193	0.197 ± 0.007	2.1	0.91:1.00
C	0.197	0.207 ± 0.007	5.1	0.97:1.00
D	0.194	0.203 ± 0.003	4.6	0.87:1.00
DD	0.201	0.200 ± 0.001	0.5	1.05:1.00

<sup>a</sup>See ref. 25.

<sup>b</sup>Percent. difference = |experimental value - theoretical value|/theoretical value × 100%.

<sup>c</sup>The ratio was normalized so that passive was set equal to 1.00. Active components are: the lithium anode and the BCX/carbon cathode. All other components are passive.

ences may also result from the fact that nominal weights and dimensions were used in the calculations whereas, in the actual cells, some deviation from the nominal as a result of construction tolerances is expected. The heat capacity of the D-size cell was independently determined [19] by discharging the cell in an adiabatic environment and measuring the temperature rise during the discharge. The value obtained from this method was  $0.28 \pm 0.04$  cal g<sup>-1</sup> K<sup>-1</sup>. The value obtained by our method for the D size cells of  $0.200 \pm 0.003$  cal g<sup>-1</sup> K<sup>-1</sup>, is in relatively good agreement with the discharge method. The discharge method, however, was prone to much less precise results.

The contributions to the heat capacities were compared based on cell size, percent. composition, and active:passive ratio, as a means of determining the dominant contributor to the cell heat capacity. All the calculations were based on an analysis of representative cells for each cell size. Table 3 also contains the active:passive ratios for each of the four cell sizes, normalized to passive = 1. The essentially constant percentage of composition is reflected in the measured heat capacities, which are identical within experimental error. In each of the cell sizes, it is interesting to note that there is an almost equal fraction of total active material relative to total passive components (*i.e.*, active:passive ratio of approximately 1:1). However, there does not appear to be a strong correlation between the active:passive ratio and the deviation from the calculated heat capacity. Although the D cells have the smallest ratio (0.87:1), and the C cells have the second highest ratio (0.97:1), both sets of cells have positive deviations from the theoretical heat capacities. The largest active component and the largest contributor to the theoretical heat capacity was the BCX cathode material. The BCX was approximately 80% by mass of the active components, and approximately 40% by mass of the total cell. Therefore, due to the close agreement between the calculated and experimental heat capacities, as well as the predicted contribution of the active and passive components,

the measured heat capacities seem to reflect the composition of the cell, and the uniformity of the various cell designs, which were relatively independent of cell size.

## Conclusions

This method provides improved precision in the results obtained for heat capacities of cells over results obtained by the discharge method [19], and also provides results which agree well with the calculated heat capacities of the cells. The accuracy of the system is also insured through the calibration of the system. This method is also superior in that the heat capacities can be determined without adversely affecting the performance of the cell. These advantages, as well as the ease of use of this method, make it a better and more efficient way of obtaining the heat capacities of Lithium/BCX cells. The most accurate and precise measurements can be obtained by maximizing the observed temperature changes (either through lowering the initial temperature of the cells, or by increasing the cell mass:methanol mass ratio) or by using a more sensitive instrument for measuring the temperature changes. The heat capacities of cell sizes AA, C, D, and DD were all identical within experimental error, as would be expected by the constant composition of the cells, where the ratios of the various components and the active to passive ratios were essentially identical.

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