Lithium Oxide in the $\text{Li}(\text{Si})/\text{FeS}_2$ Thermal Battery System

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Abstract

The formation of lithium oxide (Li₂O) in Li(Si)/FeS₂ thermal batteries during the required shelf life of twenty-five years has been identified in previous work as a reaction deleterious to thermal battery performance. This paper gives the results of a study designed to determine performance degradation caused by Li₂O and to determine an acceptable level of Li₂O that can be used to define required dryness of battery parts and allowable leak rates.

Pellets preconditioned with Li₂O were used in single cells or in batteries. Their performance was compared with discharges made using pellets with no Li₂O added. The actual Li₂O present in anode pellets at various stages during fabrication was determined by using 14 Mev neutron activation analysis.

The significant results can be summarized as follows:

- Li₂O increases the "wetness" of separator pellets. This effect reaches a plateau at about 2.5 wt. % Li₂O in a separator pellet which is 65 wt. % (LiCl · KCl eutectic) blended with 35 wt. % MgO.
- Li₂O blended with 44 wt. % Li(Si) causes an abrupt end-of-life on that part of the discharge corresponding to Li₂₁Si₈ discharging to Li₂Si. This effect always occurs if the Li₂O weight percentage is as high as 30, and was observed for Li₂O weight percentage as low as 15.
- A layer of Li₂O on anode pellets surfaces facing the current collectors causes the same abrupt end-of-life if that layer represents 6-8 wt. % Li₂O in the anode pellet.

This work shows that thermal battery production controls should be designed in such a manner that not more than 15 wt. % of the Li(Si) is oxidized at the end of the desired self life. Furthermore, the formation of a Li₂O layer equivalent to the oxidation of 6.0 wt. % of the anode on the surface facing the current collector must be prevented. Battery designers must allow for a drop in coulombic efficiency as the Li(Si) reacts, and the effect on performance of Li₂O in the separator must be considered.

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Lithium Oxide in the Li(Si)/FeS₂ Thermal Battery System

Introduction

Thermal batteries developed by Sandia National Laboratories have a shelf life requirement of twenty-five years. They must function within a narrow specific voltage range with a high reliability (typically, a success probability of 0.995 is required). The technology being developed at Sandia National Laboratories for the Li(Si)/FeS₂ system has been described in detail elsewhere, and some familiarity with this technology is assumed.^{1,2} To specify manufacturing conditions and material properties that insure that the batteries meet stated requirements, it is necessary to predict effects on performance caused by any chemical reactions occurring during battery construction and storage.

In previous work the reaction of residual water with lithium to form lithium oxide was identified as the only observed deleterious reaction during storage.^{3,4} (The referenced works excluded organic potting materials; another aging test using a variety of organic materials is currently in progress.) The reaction of Li(Si) with water to form Li₂O may form LiOH in an intermediate step, but lithium is very mobile in Li(Si) and can react with LiOH to form Li₂O. In earlier work, no LiOH was observed in pellets exposed to water vapor and analyzed several days later with various surface analytical instruments.⁵ In an ongoing study, substantial hydrogen has been observed in the over-gas sealed units. Apparently the hydrogen formed in the reaction of lithium with water vapor remains in the battery as hydrogen gas.⁶ Water is present in newly constructed batteries because it adheres to the surfaces of almost all materials. Furthermore, any leakage through seals and welds during the self life of a battery introduces additional water. In order to determine acceptable levels of water and to define allowable leak rates, it is necessary to define the effect of Li₂O on performance.

The general approach in this work was to precondition anode and separator pellets with Li₂O, analyze a representative sample for oxygen content, and test the pellets for performance using a single cell tester.

Some battery tests confirmed that battery performance was at least as good as that predicted from single cells tests. Previous work indicated that Li₂O formed aging was concentrated on the anode surface next to the separator.^{3,4} Since Li₂O is soluble in the LiCl·KCl electrolyte,⁷ effects of Li₂O could be manifested in either anode or separator; for this reason, both separators and anodes were preconditioned.

During the course of this work, it was discovered that small quantities of Li₂O caused the pellets to be more fluid (or less rigid) at operating temperature. The particular MgO originally used as a separator material in this technology was considered inadequate when small percentages of Li₂O were dissolved in the electrolyte. For this reason, MgO from a number of vendors was screened for use as separator materials. The results of that survey are also included in this report.

Experimental Section

Two techniques were used to precondition pellets with Li₂O. In one series of tests (involving both anodes and separators) Li₂O was blended with powder before pellets were pressed. In a second series of experiments involving anodes only, flat pellets were positioned on a flat stainless steel pan and exposed to water vapor in a constant humidity chamber until a desired weight gain was reached. Most of these anodes were conditioned with only one surface exposed to the water vapor, but some had both faces exposed. Since earlier work indicated that water vapor at ambient temperatures reacts with Li(Si) at an appreciable rate to form Li₂O only on exposed surfaces, and since only Li₂O was identified, it was assumed that the weight gain represented Li₂O.⁵

Anode pellets were analyzed for oxygen content using fast neutron activation analysis. This technique has been described elsewhere, and will not be detailed here.⁸ The estimated error in the oxygen analysis

reported here is less than 10%. Because of experimental difficulties, it was not possible to do single cell experiments using the same anodes that were analyzed for oxygen. Analysis was done on a representative number of pellets from a particular lot, and it was assumed that the other pellets in that lot contained the same quantity of oxygen.

Preconditioned anode pellets were discharged in single cells or in batteries. The single cell testers used at Sandia have been described elsewhere, and will not be discussed here.⁹ Discharges were made in a dry room (water vapor <300 ppm) and in an argon glove box (water vapor and oxygen <10 ppm). All single cell tests utilized a force of 2.2 psi on the top platen and all were discharged at a constant current of 100 mA/cm² with the platens at 520°C. All cells used 304 stainless steel current collectors, and some single cell discharges used a reference electrode (Ag/Ag⁺ 0.1M) inserted into the separator layer. Most single cell test used 10.0 cm² pellets with these weights: anode, 1.0 gram; separator, 2.0 grams; cathode, 3.0 grams. Some tests used heavier pellets, as indicated below. Battery tests used larger pellets and a design similar to one previously published, but with different weight pellets.¹⁴

Separator pellets were tested for performance in single cell discharges; mechanical tests were made to determine changes in physical integrity. Mechanical tests included pellet breaking strength at ambient temperature and the thickness reduction of a pellet between 520°C platens under a constant force. The determination of thickness reduction used a method developed by R. A. Guidotti and F. W. Reinhardt (both SNLA Organization 2523) and used heated platens positioned identically to a single cell tester. The top platen was loaded with a weight to give the desired force. A micrometer with a digital read-out was positioned on top of the weight. A pellet was placed between the platens (stabilized at 520°C) and thickness reduction was indicated by the digital readout. In practice, it required about a minute for the pellets to stop deforming and stabilize at a given thickness.

Materials used in this work were prepared in the same manner as those in earlier reports.⁴ Anode material was prepared by grinding Li(Si) (44 ± 1 wt.% Li) in an argon atmosphere to produce a powder that was -40 + 200 mesh (US standard sieve size). Separator material was prepared using a multistep process. MgO was first dried for 4 hours at 600°C and then blended with granulated LiCl·KCl eutectic using a Freon® blend technique. After evaporating the Freon®, the resulting powder was fused at 400°C for sixteen hours,

then cooled, granulated, and sieved through a 60 mesh screen. Cathode powder was prepared by blending FeS₂ (-60 mesh) with granulated electrolyte and an electrolyte-binder mix in a ball mill. The FeS₂ was 95% pure (with SiO₂ as the major impurity). The electrolyte binder used in the cathode consisted of 88 wt.% LiCl·KCl and 12 wt.% Cab-o-sil® (SiO₂). The weight ratio of FeS₂/electrolyte/electrolyte-binder was 64/16/20. The weight ratio of the electrolyte/MgO in the separator was either 65/35 or 70/30.

Powders used for pressing pellets preconditioned by blending in Li₂O were prepared by adding the desired amount of Li₂O and blending in a ball mill. Two grades of Li₂O were used. Most of the work utilized a product from Alpha Ventron labeled 95% pure, but when analyzed using a nuclear magnetic resonance technique, it was found to contain as much as 14% LiOH¹⁰. Subsequently, a 99% pure grade of Li₂O from Cerac Pure was used to repeat certain experiments.

Results

Evaluation of Various MgO Products

Seven MgO products were evaluated as separator materials. Powders were made with 70 wt.% LiCl·KCl and 30 wt.% MgO. Two gram, ten cm² pellets were formed using 25 tons of pressure. Table 1 lists data characterizing the different MgO products; Table 2 lists results of mechanical tests used to evaluate the pellets. Also given in Table 1 is a qualitative ranking of the effectiveness of the various MgO products as thermal battery separator materials. The qualitative ranking was based on the data in Tables 1 and 2 as well as of the sharpness of pellet edges and the tendency to develop cracks during the pelletizing procedure.

In single cell tests, all but the Meller material performed acceptably. It is likely that all the materials (except Meller) could be used in batteries, but the optimum ratio of MgO to electrolyte would be different for each product. The manufacturers of Merck Maglite S #3331 reported a change in processing, and supplied us with a sample from a pre-production lot labeled Merck Maglite S #3334. In preliminary tests the #3334 material appeared to have an effectiveness similar to Merck Maglite A in separator pellets. (See Table 1 for rating.)

Table 1. Properties of Various MgO Samples with a Qualitative Ranking of the Effectiveness of the Different Products in Separator Pellets. (1 = very good, 5 = very poor).

Supplier	Bet Surface Area (m ² /g)	Sedigraph Particle Size Range* (Eq. Spher. Dia. μ m)	Impurities (%) (EMM. Spect.)	Effectiveness Ranking
Baker	81	2-0.3 (50%)	0.3	2
Merck Maglite S #3331	28	4-0.3 (75%)	1.0	1
Merck Maglite A	17	6-0.3 (25%)	2.2	3
Morton 90	76	4-0.3 (30%)	1.5	3
Morton 170	58	4-0.3 (35%)	1.2	3
Meller	53	20-0.3 (30%)	---	5
Mallinckrodt	21	5-0.6 (100%)	0.4	4

*The balance of the material had particles smaller than 0.3 μ m equivalent spherical diameter.

Table 2. Characterization of Separator Pellets Made With MgO From Different Suppliers.*

MgO Supplier	Pellet Density (g/cm ³)	Breaking Force (g)	Reduction In Thickness (%)
Baker	2.00	713	15.8
Merck, Maglite S #3331	1.84	914	12.7
Merck, Maglite A	1.97	833	14.5
Morton 90	1.98	715	17.1
Morton 170	1.96	430	27.6
Meller	1.89	944	----

*The thickness-reduction tests used a pressure of 170 gm/cm² (2.41 psi) and a platen temperature of 520°C. Pellets were made using 25 tons pressing force.

Effect of Li₂O in Separator Pellets

Most of the early development work at Sandia with the Li(Si)/FeS₂ thermal battery system used a particular lot of MgO purchased from the Baker Chemical Co. When 3.0 wt.% Li₂O was added to a separator pellet consisting of 30 wt.% Baker MgO, the pellet became so fluid that it extruded out in single cell experiments. (The same separator with no Li₂O added performed very well in single cell tests.) In

similar tests using Merck Maglite S #3331, the separator maintained its integrity when Li₂O was added. All following data use Maglite S #3331.

Separator powder consisting of weight ratios of electrolyte to MgO of 65/35 and 70/30 were evaluated. Li₂O was mixed with the two powders in the desired weight ratios and 2.0 gram pellets were pressed and tested. Figure 1 gives a plot of time to 1.5 volts for single cell discharges using pellets made from the 65/35 mix with various amounts of Li₂O added. Also

Figure 1. Effect of Lithium Oxide on the Discharge Time to 1.5 Volts for Single Cell Experiments in an Argon Atmosphere.

plotted on this figure is an average value for a number of cells with a 70/30 EB with no Li_2O added. That 65/35 pellets with Li_2O added discharge in a manner very similar to 70/30 pellets (as shown in Figure 1) suggests that Li_2O makes the 65/35 pellets “wetter” or more fluid. The data on Figure 1 were generated from discharges in an argon glove box. Similar data have been generated using discharges in dry air. In the dry air data (not given here) a trend identical to that shown on Figure 1 was observed, except that the time to 1.5 volts became constant at a Li_2O content of about 0.5 wt.% instead of 3.0 wt.%. It is likely that this difference results from $\text{Li}(\text{Si})$ reacting with ambient oxygen to form additional Li_2O during dry-air discharges.

Effect of Li_2O in Anode Pellets

Anode pellets containing various amounts of Li_2O were used in single cell experiments in both dry air and argon. The major impact on functionality caused by the Li_2O was an abrupt end of life as illustrated on Figure 2. The voltage change on Figure 2 at about 25 minutes results from an anode transition. Although data are not reported here, reference electrodes in several cells confirmed that the abrupt end of life was associated with the anode.

The amount of Li_2O necessary to cause the abrupt end of life was greater for those discharges done in argon than for those done in air. In both cases, the time for the abrupt end of life was somewhat dependent on the amount of Li_2O added (more Li_2O caused an earlier end of life). All discharges exhibiting the

abrupt end of life performed well through the first transition of the $\text{Li}(\text{Si})$ (see Reference 11 and 12 for discussion of $\text{Li}[\text{Si}]$ transitions exhibited by discharging 44 wt.% $\text{Li}[\text{Si}]$), with the abrupt end of life occurring at some point on the curve where the $\text{Li}_{21}\text{Si}_8$ phase was discharging. Some discharges were run with electrolyte added to the anode to determine if performance would be improved. These experiments seemed to reduce the frequency of the abrupt end of life phenomenon, but did not eliminate it. Tables 3 and 4 summarize the observed data for these discharges.

Figure 2. Representative Discharge Curves for Normal Cells (solid line) and for Cells With Anodes Containing Li_2O . The approximate weight percentages giving the traces are indicated for discharge in air and argon.

Table 3. Data Summary for Single Cell Discharges With Anodes Containing Li₂O Blended With the Li(Si) Powder

Number of Discharges	Wt.% Li ₂ O Added	Average Time to 1.5 V (min)	Standard Deviation For Time to 1.5 V (min)	Number of Cells With Abrupt End of Life	Average Time to Abrupt End of Life
Discharges in Air					
8	.5	23.9	1.9	0	--
4	1.0	26.5	1.0	0	--
5	1.5	27.8	2.28	3	33.3
10	2.5	26.8	2.44	4	33.3
4	3.0	23.25	1.26	4	29.5
5	5.0	24.4	2.07	5	27
2	10.0	27.5		1	33
9	15.0	22.1	1.79	9	25.2
Discharges in Argon					
5	0	23.9	3.13	0	--
3	2.5	24.7	0.58	0	--
3	2.0	29.3	2.9	0	--
8	15.0	24.4	2.0	1	49.0
4	17.5	24.5	1.0	4	58.5
8	30	27.7	3.24	8	34.7
*5	30	25.4	1.34	5	50.6

*discharge used 99% Li₂O

Table 4. Data Summary for Single Cell Discharges In Argon With Both Electrolyte and Li₂O Added to the Anodes.

Anode Wt. Ratio of Li(Si)/Electrolyte/Li ₂ O	Number of Discharge	Average Time To 1.5 V (min)	Standard Deviation For Time to 1.5 V (min)	Number Of Cells With Abrupt End of Life	Average Time To Abrupt End of Life
77.7/19.4/2.9	5	32.4	3.85	3	38.7
80/15/5	9	29.1	1.36	4	32.5
80/20/0	4	33.5	5.9	1	45.0
65/30/15 (1.5 gm. anodes)	3	20.5	4.09	3	22.3

Anode pellets preconditioned by exposure to water vapor in a controlled humidity chamber ($RH = 11\%$) were discharged in an argon atmosphere. The extent of the reaction of these pellets was determined by measuring the weight gain of each individual pellet. Pellets with the reacted side facing the separator layer had normal discharges. Apparently, this case is essentially identical to the earlier data, where Li_2O was blended with the $Li(Si)$ powder. Pellets with the reacted side facing the current collector gave quite different results. An abrupt end of life analogous to that shown on Figure 2 occurred in these pellets at a weight gain as low as 3 wt. %. To further define this phenomenon, anode pellets with equal area, but with two different weights (1.0 ± 0.1 and 2.0 ± 0.1 grams) were preconditioned and discharged in cells with 6.0 gram cathodes. If the assumption is allowed that the thickness of the layer is proportional to the weight gain, the data indicate that the relative amount, rather than the thickness of the layer, determined the quantity of Li_2O in the pellet that caused failure. Figure 3 summarizes data observed for those discharges using 1 gram anodes. The cells used to generate the data on Figure 3 were anode limited when a 1 gram anode was used, but were cathode limited for the 2 gram anodes. This caused the cells with 2 gram anodes to have fewer amp-minutes/gram $Li(Si)$; however, the break in both curves occurs at 3-4 wt. % gain.

Figure 3. Plot of Amp-Min/Gm. Anode for Cells With Varying Amounts of Weight Gain (Li_2O) in a Layer Facing the Current Collector. Data points for a 1 gm. anode are indicated by circle; data points for a 2 gm. anode are indicated by squares.

Effect of Higher Purity Li_2O

Most of the Li_2O data described earlier used a grade of Li_2O that was only 95% pure and had to be ground. Subsequently, a -100 mesh 99% pure grade was purchased. A number of single cell tests were

repeated using the higher purity grade, and battery tests discussed in a later paragraph used the purer grade. No change in the data was observed that could be attributed to the different grades of Li_2O .

Battery Impact

The weight ratios of cathode to anode in Sandia-designed batteries are such that the batteries are very cathode limited. In fact, no impact was observed in batteries using pellets with weights analogous to present designs because of cathode-limited performance, even with Li_2O content as high as 30 wt. %. To demonstrate that single cell test data are representative of real battery applications, a ten cell battery was designed. (The ten cell battery used a long-life battery design.^{2,3}) This battery had 2.0 gm anodes and 10 gm cathodes. The battery was discharged through constant load of 3.12 ohms to drain the anode to the point at which the Li_2O effect could be observed. Figure 4

Figure 4. Plot of Voltage Versus Time for Three Ten Cell Batteries Using Ten gm Cathodes and Two gm Anodes. Battery load was 3.12 ohms. Battery "A" anodes were normal 44% $Li(Si)$; Battery "B" anodes contained 30 wt. % Li_2O ; and Battery "C" anodes had a 6.0 wt. % layer of oxide (12 wt. % Li_2O) facing the current collectors.

gives voltage versus time for three discharges: a normal battery, one with 30 wt.% Li_2O and one with anodes having a layer of Li_2O . The layer of Li_2O on these anodes represented an average of 6.0 wt.% with a maximum of 6.4 wt.% and a minimum of 5.7 wt.%. Three earlier batteries, built with anode pellets reacted to about 4 ~ 5 wt.% in the layer next to the current collectors, did not give the abrupt end of life.

Discussion

Source of Li_2O

Lithium oxide is present in the initial $\text{Li}(\text{Si})$ powder procured from vendors. Additional quantities are added by exposure and by the pelletizing process. The actual Li_2O content of pellets will vary with handling procedures and with different facilities. Once a battery is constructed, most residual water (and perhaps all oxygen) in the battery reacts with $\text{Li}(\text{Si})$. An accelerated aging study currently underway indicates that the residual gas in aged batteries is completely depleted of oxygen and water vapor. Considerable hydrogen is also present in the gas. It is not known at this time whether all oxygen reacts with the $\text{Li}(\text{Si})$ or some other species (such as FeS_2). It appears to be a conservative estimate for the purpose of, for example, leak rate calculations, to assume that all oxygen does react with $\text{Li}(\text{Si})$.

In the course of this work, the oxide content of $\text{Li}(\text{Si})$ has been repeatedly measured at various stages in the process of building a battery. The oxide content of sample pellets from lots made of $\text{Li}_2\text{O} + \text{Li}(\text{Si})$ blends has also been determined. All pellets used in this work were made with lots of $\text{Li}(\text{Si})$ powder that contained about 0.3 wt.% oxygen (0.6% Li_2O). The pelletizing process increased the oxygen wt.% to a range of roughly 0.6 - 1.4 wt.%. The pelletizing process was closely monitored and the oxide content was determined for pellets made at various times during a particular pellet-making session. The physical act of compacting the pellet increased the oxide content. It is postulated that localized hot spots formed by the crushing and breaking of particles substantially increase the reaction rate of $\text{Li}(\text{Si})$ with oxygen (and nitrogen) in the vicinity of the hot spots.

As a result of the oxide initially present in the powder and the oxide formed during the pelletizing process, all pellets used in this work contained from 0.6 to 1.5 wt.% more oxygen than indicated by weight gain or by the amount of Li_2O added. Furthermore, variability in the blending process added uncertainty. Oxygen contents determined for pellets from various

samples are given in Table 5. Calculated values are given assuming the blended Li_2O as the only source of oxygen.

Implications of this Work for Battery Design and Production

The results of this work suggest several points that are relevant to battery design and production. The ability of magnesium oxide to immobilize the electrolyte varies widely depending on the particular product used. No correlation between the data on Figures 1 and 2 and the effectiveness of MgO as a binder is apparent. More work needs to be done to define the properties of MgO which control the integrity of an EB pellet before specifications can be written to define adequately MgO as a separator/binder for thermal batteries. Unless and until such work is completed, designers should ensure that they have an adequate supply of MgO identical to the material used during development.

Battery performance can change during the shelf life because of build-up of lithium oxide at the separator-anode interface. This change can improve battery performance; however, the change in separator integrity may be significant enough to cause failure under conditions of severe shock loading. It is suggested that designers with extreme shock requirements either adopt a separator with Li_2O added to approximate the performance change, or check the design under the most severe conditions with Li_2O added to the separator.

Production and procurement controls should be defined in a manner such that $\text{Li}(\text{Si})$ does not oxidize to the extent that performance is affected at the end of the expected shelf life of the battery. At the present time, it is assumed that all oxygen and water vapor leaking into the battery reacts with $\text{Li}(\text{Si})$. The total possible oxygen in the pellets of a completed battery is equal to the sum of the oxygen present in the pellets at the time of battery assembly, the oxygen in the water absorbed by the battery parts, the free oxygen sealed in the battery when it is sealed, and the oxygen that leaks into the battery (as water and free oxygen) during shelf-life. It is suggested that this quantity not exceed 8 wt.% of the total amount of $\text{Li}(\text{Si})$ in the battery (15% of the available lithium reacted).

Production process controls should be defined in such a manner that a layer of Li_2O equivalent to 3 wt.% oxygen (6% of the $\text{Li}(\text{Si})$ reacted) does not form on a $\text{Li}(\text{Si})$ pellet. Such a layer if placed next to the current collector, can conceivably cause failure. Exposure times necessary to produce such a layer can be estimated from the work detailed in Reference 6.

Table 5. Oxygen Content of Anode Pellets Made From Blends of Li₂O and Li(Si)

Wt.% Li ₂ O Added	Calculated Wt.% Oxygen	Measured Oxygen Wt.% For Representative Pellets
1.0	0.53	1.58, 1.30, 1.23, 1.15, 1.16
1.5	0.80	2.09, 1.86, 1.81, 1.57, 1.81
2.0	1.06	3.45, 3.01, 2.42, 2.52, 2.41
15	8.0	9.7, 9.3, 8.1
17.5	9.33	11.00, 10.8, 10.5
20	10.6	11.9, 11.9, 11.5
30	15.9	16.4, 17.1, 17.2

Thermal Batteries Using Li₂₁Si₈

Previous Li(Si)/FeS₂ thermal battery designs have utilized a 44 wt.% Li(Si) alloy which is largely the Li₁₅Si₄ phase (see Reference 11), and the batteries have not been required to discharge past this phase. It is feasible to build a battery using the Li₂₁Si₈ phase, and some performance advantages as well as superior pelletizability of this material make it attractive. However, all observed failures in this work occurred while this phase was discharging. Some single cell tests using this phase blended with Li₂O (15 wt.%) failed immediately. It appears that Li₂₁Si₈ (or the discharge product phase Li₂Si) is somehow involved in the failure mechanism. Any production battery utilizing the Li₂₁Si₈ phase would probably require tighter processing controls in order to insure functionality at the end of shelf life.

Conclusions

A build-up of Li₂O in Li(Si)/FeS₂ thermal batteries can cause performance changes and even battery failure. Lithium oxide in the separator pellet increases the fluidity or "wetness" of the pellet. This effect reaches a plateau at about 2.5 wt.% Li₂O in the separator powder. Excessive amounts of Li₂O in the anode or in a layer on the anode facing the current collector can cause a premature end-of-life for a discharging cell or battery. This effect was observed for greater than 15 wt.% Li₂O blended with Li(Si) and for reaction layers with greater than ~3.5 wt.% Li₂O facing the current collectors.

The effectiveness of MgO as a separator/binder is variable depending on the source of the material. This variability may be a function of physical parameters,

but no correlations are possible with data from this study. More work is necessary before conclusions can be made.

The data presented in this work are not sufficient to define a mechanism for the abrupt end-of-life or for the change in pellet fluidity caused by Li₂O. The data are complete enough to be used in specifying production procedures and leak rates. It appears that the abrupt end-of-life is caused by the formation of some non-conductive layer, and the current collector may or may not be involved. More work must be done before any conclusions about the failure mechanism are possible.

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