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APPLICATION IN A $\text{Li-Al/LiCl-KCl/FeS}$ BATTERY

by

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DEVELOPMENT OF POROUS SINTERED-CERAMIC SEPARATORS FOR APPLICATION IN A Li-Al/LiCl-KCl/FeS BATTERY*

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ABSTRACT

The procedure for fabrication of porous sintered-ceramic separators and the technical feasibility of using such separators in Li-Al/molten LiCl-KCl/FeS battery cells have been investigated. Processing techniques have been developed to fabricate ~1.5-2.5 mm thick, ~35-60% porous, flat, sintered Y_2O_3 and MgO separator plates with sufficient strength to allow handling prior to and during cell assembly. These separators performed successfully in laboratory-scale cells for up to ~2000 h and 283 cycles, indicating that the concept of a sintered separator is viable for Li-Al/FeS batteries. The particularly attractive features of these separators are potentially low cost, prefabricated form that allows easy cell assembly, and small pore size (average diameter 0.5-1.0 μm) which provides good particle retention. The test results from the sintered-separator cells indicate that Y_2O_3 is probably unsuitable for long-term performance in Li-Al/FeS cells because of its reaction with the positive active material. This is in agreement with the recently reported data on cells with Y_2O_3 felt and powder separators. Sintered MgO separators, however, showed good chemical and mechanical stability in the cell environment.

INTRODUCTION

High-performance Li-Al/molten LiCl-KCl/FeS_x (x = 1 or 2) secondary batteries are being developed by Argonne National Laboratory for electric-vehicle propulsion and for energy storage in electric utility systems (ref. 1). These batteries, which operate at 400-500°C, are currently of a vertically oriented prismatic design with one or more inner positive (FeS_x) electrodes separated from negative (Li-Al) electrodes by ceramic separators. Molten LiCl-KCl eutectic (melting point 352°C) acts as the electrolyte in the system. The overall reaction that takes place in the Li-Al/FeS_x battery upon discharge can be written as (ref. 2)

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(1)

or

(2)

where the composition of the negative electrode at complete discharge of a stoichiometric cell would be Li_2S and Fe. The reverse of these reactions occurs on recharging the cell.

Since the presence of lithium, lithium-aluminum, and iron sulfides makes the cell environment very reactive at elevated temperatures, the selection of suitable materials for various cell components is extremely critical for successful performance and commercialization of these batteries. The development of an inexpensive ceramic separator is the subject of the present study. The primary purpose of these separators is to maintain electrical separation between the electrodes while permitting ionic transport. This transport occurs through the pores of the separators, which are filled with the electrolyte.

The general requirements for the separators needed for Li-Al/ FeS_x cells have been outlined by Mathers et al. (ref. 3). These requirements are (1) The separator must be a good electrical insulator to prevent direct electron flow between the electrodes. (2) It should be <2 mm thick and highly porous (>50% porosity) to minimize cell weight and the resistance to ionic flow between the electrodes. (3) The pore channels in the separators should be <20 μm in diameter to prevent movement of the active powders in the electrode through the separator. (4) The separator must be chemically stable in the cell environment at the operating temperatures. (5) The mechanical strength and flexibility must be sufficient to allow easy cell assembly and to withstand the stresses that are generated in the cells during cycling. (6) The separators must be inexpensive so that the cost goal for a commercial cell can be achieved.

Based on these requirements, a number of materials such as BN and Y_2O_3 fabric (refs. 2-5), BN and Y_2O_3 felt (refs. 2,6), and ceramic powders [Y_2O_3 , MgO, AlN, $\beta\text{-Si}_3\text{N}_2$, etc. (ref. 7)] have been selected and used as separators in laboratory- and engineering-scale experiments. An alternative form of separator, porous sintered-ceramic plates (ref. 7), is being considered for use in Li-Al/iron sulfide batteries. Separators of this type have uniform structure and thickness and are amenable to low-cost mass production from any ceramic powder that is compatible with the cell environment, and their prefabricated form facilitates cell assembly. The primary concerns in the fabrication of sintered separators are achieving the necessary strength and mechanical stability while maintaining thinness and high porosity. In the present study, processing techniques

have been developed to fabricate flat Y_2O_3 and MgO plates ~ 1.5 – 2.5 mm thick, with up to $\sim 60\%$ porosity comprised mainly of extremely small (average diameter 0.5 – 0.8 μm) pores, using conventional cold-pressing and sintering techniques. These plates were subsequently tested in laboratory-scale Li-Al/FeS battery cells. Details of the fabrication method, and data on the performance and the postoperative characteristics of the test cells, are described below. The results indicate that the performance of cells containing porous sintered-ceramic separators in laboratory-scale experiments is comparable to that of cells with other candidate separators. Therefore, the sintered separators can be considered as suitable candidates for use in larger cells.

EXPERIMENTAL PROCEDURE

Fabrication of separator plates

Since the plates must be highly porous and very thin, damage in handling prior to firing and warpage after firing are often serious problems. The magnitude of these problems increases manyfold with increasing porosity and/or decreasing plate thickness, both of which are desirable for reduction of internal cell resistance. Because of these problems, several powder-processing techniques were tested for fabrication of the separators. Most of these preliminary studies were directed toward fabrication of porous Y_2O_3 plates. Starting materials such as as-received Y_2O_3 ^{*} and MgO,[†] calcined Y_2O_3 and MgO, Y_2O_3 foam,^{**} and Y_2O_3 -nitric acid plaster compositions (refs. 9 and 10) were used. The best processing technique tried thus far, which provided optimum handling characteristics of the plates before and after firing and resulted in negligible shrinkage and warpage of the plates after firing, is as follows: The starting Y_2O_3 or MgO powder[§] was calcined at a temperature higher than the sintering temperature to keep the shrinkage during subsequent sintering to a minimum. The calcined powder was then mixed with a small fraction (5 to 10 wt %) of as-received oxide powder, which provided strength to the green body prior to the cold-pressing and sintering operation and provided adequate sinterability to the cold-pressed plates. The following steps were included before and during the sintering procedure to assure a flat plate: (1) The corners of the cold-pressed green plates were

^{*}Molycorp., 99.99% Y_2O_3 .

[†]Mallinckrodt, reagent-grade MgO.

^{**}Foaming of a slurry consisting of Y_2O_3 powder, organic binders, and surface-active agents, and subsequent drying and firing of the fluid foam at high temperatures, provided the Y_2O_3 foam (ref. 8) for fabrication of separators.

[§]The starting powder can be provided in several forms. The simplest and preferred form is commercially available Y_2O_3 or MgO powder with particle sizes of <1 to 5 μm . Alternatively, the starting material can be a compound which on calcination decomposes to the desired oxides. For example, crushed Y_2O_3 foam or Y_2O_3 -nitric acid plaster compositions were successfully used for fabrication of thin, flat sintered plates. The nature of the starting material may, however, influence the pore morphology of the sintered plates.

ground off to eliminate the areas with maximum density gradients and thus reduce the tendency toward warpage. (2) The plates were carefully positioned in the furnace so as to achieve uniform heating, thus minimizing density gradients in the samples. (3) A small load ($\sim 2 \text{ g/cm}^2$) was placed on the samples to help keep the plates flat. (4) An extremely low heating rate ($\sim 60^\circ\text{C/h}$) was used to reach the sintering temperature, so as to avoid thermal stresses in the plates.

By following the above procedures, flat Y_2O_3 and MgO plates with porosities up to $\sim 60\%$ and thicknesses of $\sim 1.5\text{--}2.5 \text{ mm}$ have been fabricated. Prior to in-cell testing, the density of these plates was measured by geometric methods. Also, the cumulative pore volume and the pore-size distribution were measured by the mercury porosimeter* technique. Scanning-electron microscopy (SEM) of the fracture and as-sintered surfaces of the separator plates was also performed to examine the pore and particle-size distributions before in-cell testing.

Assembly and testing procedures for the battery cells

A schematic of the small-scale (7.6- by 12.7-cm) unsealed prismatic Li-Al/FeS cells that were used for separator testing is shown in Fig. 1 (ref. 11). This cell configuration closely approximates that of engineering-scale cells and thus allows meaningful evaluation of the separators. The sintered separators were placed between the inner positive electrodes and the outer negative electrodes. Type 304 stainless steel screens (200 mesh) welded to the electrode frames were used to prevent movement of active particles through the separators.[†] A sheet of Type 304 stainless steel or nickel was used for positive current collection at the center of the cell; the cell housing (of Type 304 stainless steel), which was in direct contact with the negative electrodes, acted as the negative current collector. The BN sleeve at the top of the cell can was used to provide insulation between the positive and negative electrodes and frames.

Four test cells (MS-1 through -4) were assembled in the semicharged state using sintered separators. Y_2O_3 separators were used in cells MS-1 and -2; MgO separators were used in cells MS-3 and -4. The positive and negative electrodes consisted of powder mixtures of FeS, Li_2S , Fe and eutectic electrolyte ($-40 +150$ size fractions) and $\text{LiAl}_{1.2}$, Al and electrolyte ($-60 +150$ size fractions), respectively, in suitable proportions and amounts,** hot-pressed at 450°C . During assembly, the electrode edges and sides were filled with calcined ceramic powder (Y_2O_3 or MgO , corresponding to the separator material) to help

*Models 900 and 910, Micromeritics Instrument Corp., Norcross, GA.

[†]As noted later, the average pore sizes in sintered separators are much smaller than the mesh sizes in the screens. Thus, use of these screens in the sintered-separator cells may not be essential.

**The proportions of various active materials determined the level of charge for the cell, whereas the amounts determined the theoretical capacity (in A-h) of the cells.

Fig. 1. Schematic of cells for testing separators.

prevent short circuiting in those areas. The Y_2O_3 separators were somewhat more wettable by the molten electrolyte than the MgO separators (ref. 12). However, maintaining the cells at $400^\circ C$ in vacuum for 1-2 h prior to cycling allowed good wetting in both Y_2O_3 and MgO separator cells. Since the cells were not sealed, it was often necessary to add electrolyte salt through the openings in the top of the cell housing to replenish losses during the course of the experiments.

The cell-performance parameters which were measured throughout the cell operation were coulombic efficiency and materials-utilization efficiency (subsequently referred to as utilization). The coulombic efficiency is a measure of reversibility of electrode processes and is calculated by dividing the number of coulombs obtained during discharge by the number of coulombs necessary to

return to charge for a particular set of conditions. A significant drop from 100% efficiency is undesirable and is indicative of side reactions, current leakage, or a short circuit in the cell. The utilization can be defined as the fraction of theoretically available material that can be effectively utilized in a cell for a given set of conditions. The ratio between the measured A-h during cell operation and the theoretical cell capacity is a measure of utilization. The sintered separator cells were cycled continuously between a discharge cutoff potential of 1.00 V and a charge cutoff potential of 1.65 V. The current density was increased in steps from 20 mA/cm² to as high as 100 mA/cm², and the cell performance parameters were measured.

The cells were sectioned in a helium-atmosphere glovebox for posttest examination. Optical microscopy, SEM, and x-ray techniques were used to identify (1) morphological changes in the separators, (2) any reactions that might have occurred between the separator and the active material and/or electrolyte, and (3) the causes of cell failures which occurred during some of the tests.

RESULTS AND DISCUSSION

Characterization of the sintered separators

As mentioned earlier, Y₂O₃ separators were used in cells MS-1 and -2, whereas cells MS-3 and -4 were assembled using MgO separators. Table 1 shows the theoretical capacities of these four cells and the processing conditions, and relevant physical characteristics of the separators used in these cells. The porosity (~50%) of the sintered separators is comparable to that of powder separators (ref. 7), but considerably less than that of felt [~90% porosity (ref. 3)]. However, note that the average pore diameter (0.50-0.76 μ m) in the sintered separators is significantly smaller than that in BN or Y₂O₃ felts [15-30 μ m (ref. 3)]. Figure 2 shows the porosimetry data* on pore size distributions in typical Y₂O₃ (~60% porous) and MgO (~45% porous) separator plates. Clearly, 100% of the pores in these plates were <8 μ m in diameter. The SEM examination of fracture surfaces from these plates (Fig. 3) confirmed the small (<5 μ m) pore size. Some agglomeration of particles could be detected in the Y₂O₃ plates (Fig. 3a), whereas the MgO samples showed uniform small (~1 μ m) particles (Fig. 3b). The small pore channels in sintered separators contribute greatly to the successful performance of the cells because of their ability to keep the active electrode particles in their respective positions.

*The pore diameters were calculated from the relation $D \approx 176.8/p$, where D is the pore diameter and p is the absolute pressure in psia needed for mercury penetration. The equation assumes a mercury contact angle of 130° and a surface tension of 474 dynes/cm, as specified in the instruction manual for the mercury porosimeter.

TABLE 1

Processing conditions and physical characteristics of the sintered separators subjected to in-cell testing

Cell no.	Theoretical capacity (A-h)	Separator plate no.	Composition	Calcination		Sintering		Porosity (%)	Average pore diameter ^a (μm)	Cumulative pore volume ^a (cm ³ /g)	Thickness (mm)
				Temp. (°C)	Time (h)	Temp. (°C)	Time (h)				
MS-1	35.7	115	Crushed Y ₂ O ₃ -nitric acid plaster and 12% binder	1250	8-1/2	1500	No hold	50.6			2.47
		125	Crushed Y ₂ O ₃ -nitric acid plaster, no binder	1250	8-1/2	1440	No hold	51.3	0.76	0.1364	2.13
MS-2	50.0	179	Calcined Y ₂ O ₃ foam mixed with 10% as-received Y ₂ O ₃	1550	13	1450	6	60.8	0.50	0.2997	1.50
		180	Calcined Y ₂ O ₃ foam mixed with 10% as-received Y ₂ O ₃	1550	13	1450	6	60.1	0.50	0.2997	1.47
MS-3	35.7	201	Calcined MgO mixed with ~5% as-received MgO	1450	2-3/4	1400	5	42.2			1.93
		202	Calcined MgO mixed with ~5% as-received MgO	1450	2-3/4	1400	5	38.5			1.88
MS-4	50.0	217	Calcined MgO mixed with 6.7% as-received MgO	1450	4	1420	3-1/4	33.8			1.78
		218	Calcined MgO mixed with 6.7% as-received MgO	1450	4	1420	3-1/4	44.2	0.74	0.2910	1.74

^aThese values were determined from plates that are similar to the separators which were actually used in-cell.

Fig. 2. Pore-size distributions in sintered separator plates.

Fig. 3. Scanning-electron micrographs of fracture surfaces from sintered (a) Y_2O_3 and (b) MgO plates.

Cell performance

The performance of cells MS-1 through -4 is shown in Fig. 4, where coulombic efficiency and percent utilization at various discharge current densities are plotted as a function of number of cycles. Cell MS-1 ran successfully for ~ 2000 h and 283 cycles; its operation was then voluntarily terminated for post-test examination. During the run, the current density of cell MS-1 was raised

Fig. 4. Coulombic efficiency and utilization of sintered-separator cells as a function of the number of cycles and discharge current density.

from 20 mA/cm² to 100 mA/cm² in steps and subsequently brought down to as low as 40 mA/cm² (Fig. 4a). Note that the utilization of MS-1 at any particular current density remained reasonably constant (within 2%) even after the cell was cycled at various current densities.

Operation of cells MS-2 and -3 was terminated after 158 cycles (~1200 h) and 65 cycles (~770 h), respectively, because of the serious loss of coulombic efficiency (Figs. 4b and c). Prior to this loss, both cells had shown good, stable performance. Cell MS-4 operated successfully for ~1440 h and 160 cycles (Fig. 4d); its run was terminated at that point because of a break in a welded joint in the positive current collector. Despite the low utilization of cell MS-4, the coulombic efficiency remained at 100% throughout its operation, except at the time when the current collector broke; the sharp drop in efficiency evident during the 150th cycle (Fig. 4d) corresponds to this event.

The utilization of the active material as a function of discharge current density for cells MS-1 through -4 is shown in Fig. 5. For comparison, this figure includes the data (ref. 7) from a BN-felt cell (SC-25, felt thickness ~ 1.25 mm) and a MgO powder-separator cell (SC-21, ~ 1.8 mm thick). The BN-felt cell exhibited better performance than any other cell shown in Fig. 5. This is probably due to the lower porosity (and therefore lower internal resistance to ionic flow) of the felt separators as compared to that of the sintered and powder separators. The cells (MS-1 and -2) with sintered Y_2O_3 separators performed better than those with MgO separators (MS-3 and -4). Again, this can be related to the lower porosity of sintered Y_2O_3 separators as compared to that of MgO separators.

Fig. 5. Comparison of performance of sintered-, BN felt- (SC-25), and MgO powder- (SC-21) separator cells.

The data presented in Fig. 5 show that the utilization of the sintered-separator cells decreased in a nonlinear fashion with increasing discharge current density. The powder-separator cell showed similar behavior, although the utilization decreased at a much more rapid rate. In contrast, the BN-felt cell was characterized by an essentially linear drop in utilization with increasing current density. The reason for these differences in performance are not known at this time.

Posttest cell examination

As mentioned earlier, the tests of cells MS-2 and -3 were terminated because of serious loss of coulombic efficiency, while cells MS-1 and -4 performed satisfactorily throughout their operation. Posttest examination of cells MS-2 and -3 identified the general areas where electrical short circuits had occurred. In cell MS-2, neither the precise region of the short nor the reason for it could be determined. However, in cell MS-3, it was evident that the short had developed at the top of the cell can and was due to contact between the positive and negative electrode frames. This short was attributed to faulty cell assembly and not to any materials limitations.

The metallographic examination of the separators in the cut cross sections of the cells after operation indicated that the Y_2O_3 separators in cells MS-1 and -2 reacted to some extent on the positive-electrode side. This is particularly evident in cell MS-2 (Fig. 6). In addition to the reacted zone on the positive-electrode side and along the cracks, an increased amount of porosity (indicating possible damage of the separator) was evident on the positive side of the separators. Although cell MS-1 was operated for a longer period than MS-2, the separators in MS-1 showed less reaction and damage than those in MS-2. This is probably because MS-1 had a lower theoretical capacity (see Table 1) and therefore less active material available for reaction.

Fig. 6. Section of the sintered Y_2O_3 separator from cell MS-2.

The reaction product on the Y_2O_3 separator was further characterized by careful x-ray diffraction analysis, which indicated the presence of Y_2O_2S and an unidentified phase on the positive side of the separators. However, the reaction products on the fracture surfaces of these separators could not be identified by SEM and x-ray dispersive analyses. A typical fracture surface from the cell-tested Y_2O_3 separator is shown in Fig. 7a. A comparison with Fig. 3a indicates that the separator structure did not change significantly during the period of cell operation. However, small hexagonal platelets (Fig. 7b) were detected in very limited areas of the separators after the test. The nature of the platelets, which also appeared in MgO separators, cannot be identified at this time. Since Y_2O_2S was also detected in a few other recently tested cells (ref. 13) that used Y_2O_3 felt and powder separators, it appears unlikely that the Y_2O_3 separators would perform satisfactorily during long-term operation of the Li-Al/iron-sulfide battery cells.

Fig. 7. Fracture surfaces from Y_2O_3 separator after operation in cell MS-2, showing (a) the majority of the separator structure and (b) hexagonal platelets that appeared in limited areas of the separator. Similar platelets also appeared on MgO separators after in-cell testing.

Although small unidentified platelets, similar to those detected in Y_2O_3 separators, were found in limited areas of the fracture surfaces of the sintered MgO separators after in-cell testing, neither metallographic examination nor x-ray diffraction showed any evidence of chemical reaction between the active materials and the sintered MgO separators in cells MS-3 and -4. A cross section of cell MS-4 showing electrodes and separators, and a magnified section of the

MgO separator after operation in the same cell, are shown in Fig. 8. The MgO separators showed good mechanical and chemical stability in these cell environments. A typical SEM fractograph of the MgO separator from cell MS-4 is shown in Fig. 9.

Fig. 8. (a) Photograph of a cross section of cell MS-4 and (b) typical section of MgO separator from cell MS-4.

The results from these tests indicate that sintered MgO separators are probably suitable for application in Li-Al/FeS battery systems. Since the porosity of the separators used in MS-3 and -4 was $\sim 40\%$, the utilization of these cells was rather low. However, one should be able to fabricate sintered MgO separators with porosities up to $\sim 60\%$ (as was obtained for Y_2O_3 plates) by means of the processing technique outlined above, with the choice of suitable processing

Fig. 9. Typical scanning-electron fractograph of MgO separator from cell MS-4.

parameters. This porosity is still considerably lower than that in BN felts; also, MgO separators with this porosity will be heavier than BN felt separators of the same thickness (the densities of MgO and BN are 3.58 and 2.20 g/cm³, respectively). Despite these drawbacks, sintered MgO separators offer the advantages of potentially low cost, prefabricated form for easy cell assembly, and excellent particle-retention capacity due to the small pore structures. However, these separators must be tested for longer time periods in larger cells, to verify their chemical and mechanical stability under more severe test conditions, before they can be considered for commercial applications.

SUMMARY AND CONCLUSIONS

1. Processing techniques have been developed to fabricate highly porous (up to ~60% porosity), thin (~1.5–2.5 mm), and flat sintered Y₂O₃ and MgO separator plates that can be used in laboratory-scale prismatic Li-Al/molten LiCl-KCl/FeS battery cells.
2. Since the average pore diameter in these sintered separator plates is 0.50–0.76 μm, and 100% of the pore diameters are <8.0 μm, the plates should effectively prevent the movement of active electrode particles from their respective positions.

3. Four sintered-separator cells (two with Y_2O_3 and two with MgO plates) were assembled and run to evaluate the performance of the separators. These cells operated for up to ~ 2000 h and 283 cycles at current densities up to 100 mA/cm^2 . The performance of these cells was slightly inferior to that of the BN-felt separator cells, but comparable to that of MgO-powder separator cells. The results clearly indicate that the concept of sintered separators for Li-Al/FeS batteries is viable.
4. Y_2O_3 separators are likely to be unsatisfactory for the Li-Al/FeS battery cells, because Y_2O_3 reacted with the positive electrode material in the present tests. However, sintered MgO separators showed good chemical and mechanical stability in the cell environment, and are thus considered potentially attractive candidates. Before they can be considered for commercial applications, sintered MgO separators must be tested in larger cells.

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