

# Electrochemical performance of lithium/sulfur cells with three different polymer electrolytes

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## Abstract

Charge and discharge characteristics of lithium/polymer electrolyte/sulfur cells are presented. Three different electrolytes were studied, and cells were operated at temperatures ranging from ambient to about 100°C. The effects of the sulfur electrode composition and cycling regimen on both the potential profiles and the capacity fade rate were investigated. Cells prepared with poly(ethylene oxide) (PEO) and operated at 90–100°C could be discharged to nearly the full theoretical 1672 mA h/g active material but with a high rate of capacity fade. Reducing the depth of discharge to 30% or less increased the cell lifetime. Room-temperature cells with poly(ethylene glycol) dimethyl ether could be discharged to about 45% utilization of the sulfur and showed a much lower capacity fade rate after the second cycle. Several possible explanations for the high rate of capacity fade and the effect of the depth of discharge on this rate are presented. © 2000 Elsevier Science S.A. All rights reserved.

**Keywords:** Rechargeable lithium batteries; Lithium; Sulfur; Polysulfide; Capacity fade; Charge/discharge

## 1. Introduction

Rechargeable lithium batteries are being developed for portable power applications such as electric vehicles, partly because of their high specific energies in the range 100–150 W h/kg (and theoretical specific energies in the range 425–890 W h/kg), as shown in Table 1. Further increases in battery specific energy have been limited by the extent of lithium intercalation into transition metal oxides, resulting in capacities in the range 100–150 mA h/g of active material. A battery based on the lithium/(elemental) sulfur redox couple has, in contrast, a theoretical specific capacity of 1672 mA h/g of active material and a theoretical specific energy of 2600 W h/kg, assuming complete reaction to the product Li<sub>2</sub>S. Therefore, there is a strong incentive to develop Li/S batteries.

A Li/S battery with a 100% sulfur positive electrode is impossible to discharge at low temperatures because sulfur is both ionically and electronically resistive. A practical Li/S cell must therefore incorporate well-distributed electronically conducting and lithium ion-conducting phases in the positive electrode. Prior studies of Li/S cells with liquid electrolytes faced the serious problems of low active material utilization and poor rechargeability, due to the insulating nature of sulfur and Li<sub>2</sub>S, and to the loss of active material in the form of soluble polysulfide reaction products. The incomplete reversibility of the reactions to lower-order sulfides (e.g. Li<sub>2</sub>S<sub>2</sub>, Li<sub>2</sub>S) was also a factor. In addition, degradation of the Li electrode in liquid organic electrolytes limited the performance of such cells [1–4].

DeGott [5] investigated solid-state Li/S cells using a poly(ethylene-oxide)-based electrolyte and obtained very low utilization of the active material at cycling temperatures of 70°C. A recent patent [6] describes PEO-based Li/S cells with a capacity of more than 80% of the theoretical value for a single discharge and a cell lifetime of 400 cycles at 200 W h/kg of positive electrode. Higher operating temperatures were also used. Capacity fade is a

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Table 1  
Characteristic of several types of rechargeable lithium cells

Li battery couple	Theoretical specific energy (Wh/kg)	Practical specific energy (Wh/kg total cell)	Theoretical specific capacity (mAh/g active material)	Practical specific capacity (mAh/g total cell)
Li/Li <sub>x</sub> Mn <sub>2</sub> O <sub>4</sub>	428	120	285 ( <i>x</i> = 2)	100–120
LiC <sub>6</sub> /Li <sub>x</sub> CoO <sub>2</sub>	570	180	273 ( <i>x</i> = 1)	136
Li/Li <sub>x</sub> V <sub>6</sub> O <sub>13</sub>	890	150	412 ( <i>x</i> = 8)	309
Li/Li <sub>x</sub> TiS <sub>2</sub>	480	125	225 ( <i>x</i> = 1)	58
Li/S	2600	–	1672	> 200 <sup>a</sup>

<sup>a</sup>Based on positive electrode only, with 50% sulfur. Capacity data is for cycle regimen yielding the longest cycle.

persistent problem, however, in all Li/S cell configurations.

In the present work, Li/S cells were prepared for operation at ambient and elevated temperature with three different electrolytes. The effects of electrode composition and cell cycling regimen on cell performance were determined and possible reasons for capacity fade are presented.

## 2. Experimental procedures

### 2.1. Electrolyte and electrode preparation

#### 2.1.1. Poly(ethylene oxide)-based electrolyte

Poly(ethylene oxide)/lithium (bis)trifluoro-methanesulfonate imide, Li(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>N (PEO/LiTFSI) electrolyte was prepared by dissolving PEO (average mass MW 5 × 10<sup>6</sup>, Aldrich), and LiTFSI (used as received from 3 M) in acetonitrile at a mass ratio 49:1. The solution was stirred and then cast onto a glass dish, dried in air and cut to size with a cork borer. Sulfur electrodes were prepared by mixing sulfur (Brite-Life, sublimed), carbon (Shawinigan black, 50% compressed, H.M. Royal of California), and electrolyte, and adding acetonitrile and a small amount of methanol to improve dispersion. This suspension was stirred for several days or longer until it appeared to be homogeneous, and then cast into glass rings on stainless-steel current collectors. Electrodes were dried in air overnight. The final electrodes contained 50 wt.% sulfur, 16 wt.% carbon and the balance polymer electrolyte, and weighed 1 to 3 mg.

#### 2.1.2. Poly(ethylene-methylene oxide)-based electrolyte

Poly(ethylene-methylene oxide) (PEMO), a polymer similar to PEO which is, however, amorphous at room temperature, was originally developed by Nicholas et al. [7]. We synthesized this polymer by mixing 40 ml of poly(ethylene glycol) (average MW ~ 400), 100 ml of dichloromethane (CH<sub>2</sub>Cl<sub>2</sub>), and 40 g of potassium hydroxide (KOH) under a nitrogen atmosphere in an ice bath. Dichloromethane reacted with the hydroxyl end groups of polyethylene glycol to form a bridge between chains and yielded potassium chloride, which was removed with any

excess KOH using de-ionized water. The polymer was fractionated with heptane and toluene to remove small chains. The average molecular weight of the final product was ~ 180,000 g/mol as measured by gel-permeation chromatography.

Films were prepared by mixing a solution of PEMO and LiTFSI (20:1) in acetonitrile and casting in glass rings on a Teflon<sup>®</sup> substrate inside a glove box (with oxygen concentration maintained below 5 ppm). The films were dried in vacuo at 60°C overnight. The final self-standing film was sticky and translucent. Electrodes of composition 50 wt.% sulfur, 15 wt.% Shawinigan Black, and 35 wt.% PEMO/LiTFSI (20:1) were prepared as above but were cast inside a glove box. These electrodes were also dried in vacuo at 60°C overnight. The electrode weight averaged ~ 3 mg.

#### 2.1.3. Poly(ethylene-glycol) dimethyl ether-based electrolyte

Poly(ethylene-glycol) dimethyl ether (PEGDME) is a short-chained PEO polymer capped at both ends with methyl groups. Khan et al. [8] first used PEGDME as a separator by mixing it with fumed silica and butyl methacrylate to form a composite electrolyte. Detailed discussions of the surface modification of the fumed silica were described by Hou and Baker [9]. We obtained fumed silica (Aerosil A-200) as a gift from Degussa, Frankfurt, Germany. The composite fumed silica electrolyte was synthesized by mixing 1 g of modified fumed silica, 1 g of butyl methacrylate, 10 mg of AIBN (2,2'-Azobisisobutyronitrile (Aldrich)) initiator, and 8 g of PEGDME (average MW = 250) /LiTFSI (O:Li ratio 30:1) in a blender. The resulting mixture was heat-polymerized overnight in an oven at 90°C under a nitrogen atmosphere.

Positive electrodes (cathodes) prepared with PEGDME did not contain the methacrylate-modified fumed silica and butyl methacrylate because their use was intended only to improve the mechanical properties of the electrolyte separator and reduce its susceptibility to reactions with the lithium electrode. A small amount of PEO (average MW 8 × 10<sup>6</sup>) was added as a binder.

Two electrode compositions were used: (i) 50 wt.% sulfur, 15 wt.% Shawinigan black, 30 wt.% PEGDME/

LiTFSI (30:1), 5 wt.% PEO; and (ii) 75 wt.% sulfur, 7.5 wt.% Shawinigan black, 15 wt.% PEGDME/LiTFSI (30:1), 2.5 wt.% PEO. The sulfur electrodes were prepared as described above. The electrode weight averaged  $\sim 2$  mg.

## 2.2. Cell assembly and testing

Cells were assembled in stainless-steel cell holders made from Swagelok<sup>®</sup> union pipe fittings with polypropylene ferrules. Sulfur electrodes were placed in the cell chambers and layered with a polypropylene spacer ring and a polymer electrolyte separator. For cells using fumed silica, the electrolyte was a yellow paste and was applied with a spatula within the center of a 76- $\mu\text{m}$  thick polypropylene spacer.

The partially assembled cells were then dried in vacuo overnight at approximately 50–60°C and placed in a glove box without exposing it to air. A Li foil disk was placed on top of the cell sandwich, and the cell chamber was closed tightly before removing it from the glove box. PEO-electrolyte cells were tested in a convection oven at 90–105°C. PEMO-electrolyte cells were tested at ambient temperature and at 60°C. PEGDME-electrolyte cells were tested at ambient temperature. Galvanostatic cycling was performed with an Arbin Battery Testing system, and cyclic voltammetry experiments were carried out with a PAR 273 controlled by M270 software. Conductivity measurements on the polymer electrolytes were performed with blocking electrodes using the Solartron Instruments AC Impedance system (model 1286).

## 3. Results

The properties and results of Li/polymer/S cells cycled between 1.5 and 2.7 V with three different electrolytes used in this investigation are summarized in Table 2. The PEO-electrolyte cells exhibited the highest first discharge capacity and total capacity delivered. On the other hand, the room-temperature PEO electrolyte exhibited the lowest conductivity, and the cells prepared with it showed the poorest performance. During discharge, the

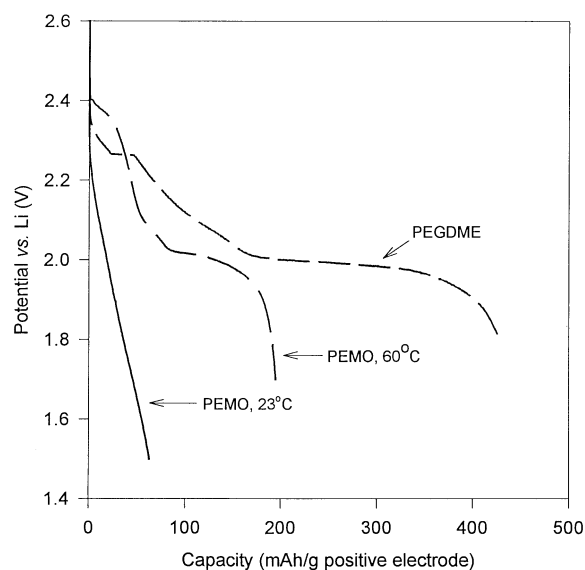


Fig. 1. First discharge profiles of three low-temperature cells: PEGDME-electrolyte cell: 0.050 mA/cm<sup>2</sup>, 23°C, 1.8 V cutoff; PEO cell at 23°C: 0.025 mA/cm<sup>2</sup>, 1.5 V cutoff; PEO cell at 60°C: 0.05 mA/cm<sup>2</sup>, 1.7 V cutoff.

voltage dropped quickly, and no plateau was observed, as shown in Fig. 1. However, when these cells were heated to 60°C, the maximum discharge capacity increased substantially, due primarily to the threefold increase in conductivity. When lithium was cycled in symmetrical Li/PEMO-LiTFSI/Li cells, a constant overpotential was observed at 60°C. When the temperature was reduced to 23°C, the cell polarized severely, as shown in Fig. 2. The overpotential-time behavior of the room-temperature Li/Li cell resembles that of the Li/PEMO/S cell during discharge.

The limiting factor for the room-temperature cells appears to be polarization in the polymer electrolyte, whereas the limiting factor for the 60°C cells is the incomplete utilization of the sulfur electrode. Table 2 shows a slower rate of capacity decline for the room-temperature PEO cells compared to the same cell at 60°C, although the initial capacity of the latter is significantly higher.

The cell with a composite fumed silica/PEGDME electrolyte performed significantly better, yielding close to 45% utilization during its first cycle. In addition, unlike

Table 2  
Li/S cell properties and capacities

Polymer electrolyte	PEO	PEMO	PEMO	Composite electrolyte
Temperature	90°C	23°C	60°C	23°C
Conductivity	4.9e – 4 S cm <sup>-1</sup>	4.2e – 5 S cm <sup>-1</sup>	1.2e – 4 S cm <sup>-1</sup>	1.5e – 3 cm <sup>-1</sup>
Discharge capacity <sup>a</sup> , Cycle 1 (A h/g of electrode)	0.722	0.044	0.243	0.376
Cycle 2	0.573	0.023	0.189	0.207
Cycle 5	0.423	0.023	0.050	0.157
Cycle 10	0.270	0.025	0.030	0.135

<sup>a</sup>The capacity is an average of three cells.

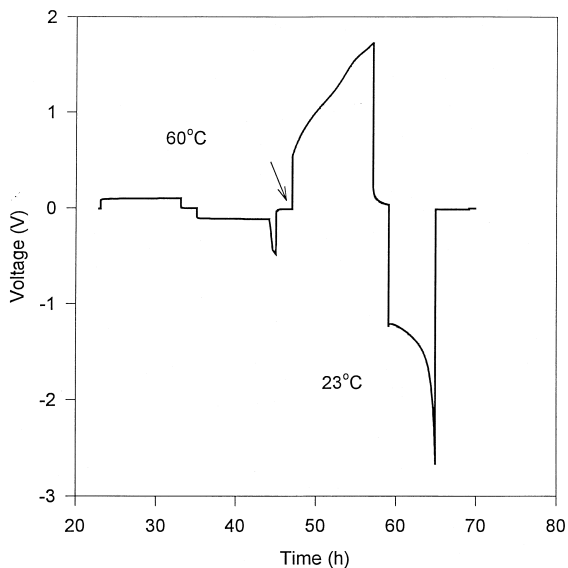


Fig. 2. Cell potential of a Li/PEMO-LiTFSI/Li cell operated at  $0.05 \text{ mA/cm}^2$ . The arrow shows where the temperature was decreased from  $60^\circ\text{C}$  at  $23^\circ\text{C}$ .

the PEMO cells, a steady plateau was observed at room temperature indicating small polarization. Fig. 3 compares the average capacity delivered for a cell with sulfur loading of 50 wt.% to one with 75 wt.% sulfur. Although the discharge capacities during the first few cycles are similar, the 75 wt.% sulfur electrode showed slightly better discharge capacity retention. The percentage of active material utilization was lower in the 75 wt.% sulfur cell, but the specific capacity of the 75 wt.% sulfur electrode was greater than that of the 50 wt.% sulfur electrode after the first few cycles.

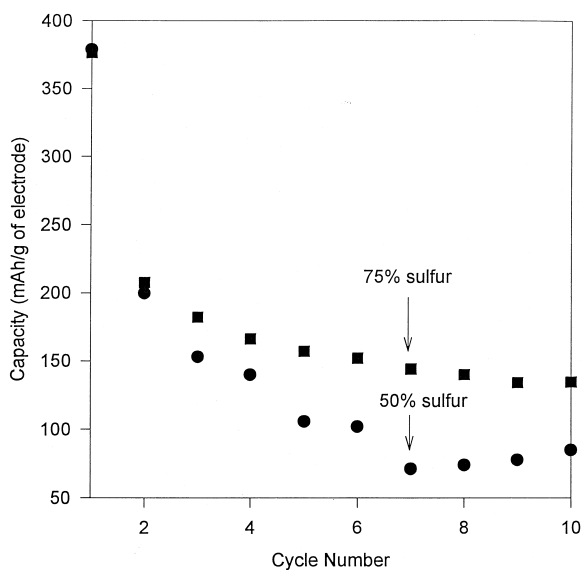


Fig. 3. Capacity vs. cycle number for two PEGDME-electrolyte cells: 50 wt.% S and 75 wt.% S in the positive electrode. Both curves are averages of three tests at  $0.05 \text{ mA/cm}^2$  and  $23^\circ\text{C}$  with 1.8 V cutoffs.

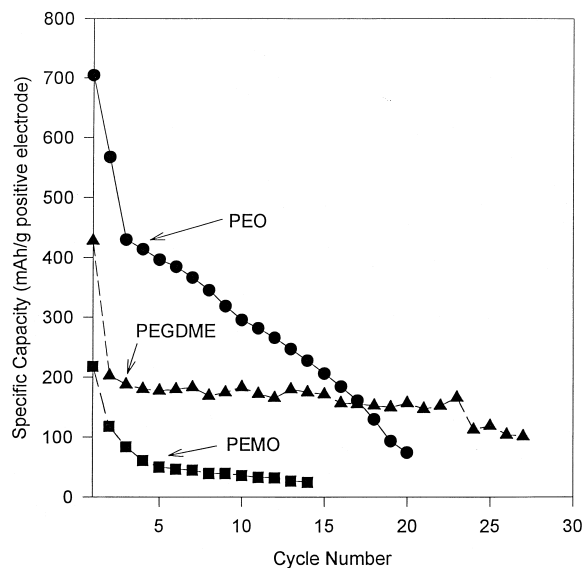


Fig. 4. Comparison of cycle lives of cells with the three different electrolytes. PEO cell (50 wt.% S):  $0.1 \text{ mA/cm}^2$  discharge, 1.5 V cutoff,  $104^\circ\text{C}$ . PEGDME cell (75 wt.% S):  $0.05 \text{ mA/cm}^2$ , 1.8 V,  $23^\circ\text{C}$ . PEMO cell (50 wt.% S):  $0.025 \text{ mA/cm}^2$ , 1.5 V,  $60^\circ\text{C}$ .

Fig. 4 shows the cycle life performance for cells of each type. All cells were cycled to 100% depth of discharge, and all cells showed a high first discharge capacity followed by a sharp capacity decline over the next few cycles. After these first few cycles, the capacity fade rate diminished substantially. Capacity fade rates were calculated starting from the cycle at which this dramatic slope change occurred. The PEO cell exhibited the highest first-cycle capacity but also showed a high capacity fade rate (2.8%/cycle for the third and higher cycles, based on the first cycle capacity). The PEGDME cell showed the next-

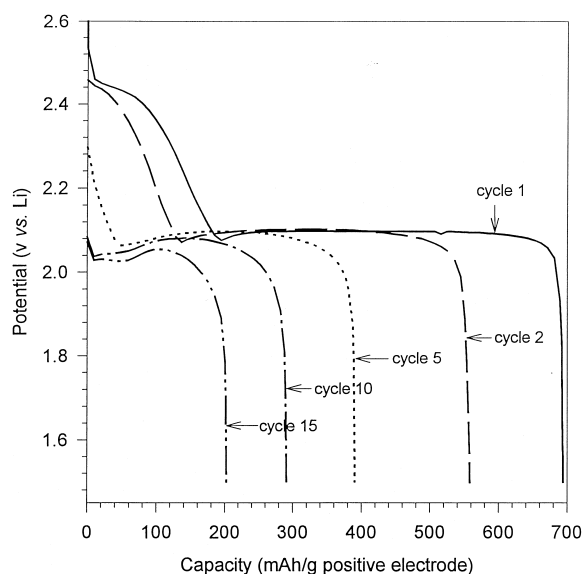


Fig. 5. Selected discharge profiles at  $0.1 \text{ mA/cm}^2$  for a PEO-electrolyte cell cycled at  $104^\circ\text{C}$  with a 1.5 V cutoff potential.

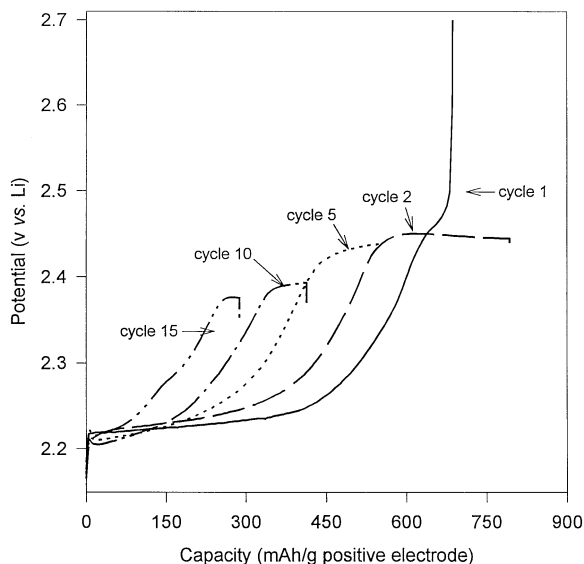


Fig. 6. Selected charge profiles at  $0.05 \text{ mA/cm}^2$  of a PEO-electrolyte cell cycled at  $104^\circ\text{C}$  with a cutoff of 2.7 V or 140% of the previous discharge capacity.

highest maximum discharge capacity and a fade rate of  $0.9\%/ \text{cycle}$  for the 2nd through 26th cycles, based on the first cycle capacity. The PEMO cell performance is the poorest based on maximum discharge capacity although the fade rate is only  $1.2\%/ \text{cycle}$ , based for the 5th and higher cycles, based on the first cycle capacity.

Voltage profiles for selected cycles of these cells are shown in Figs. 5–7. In general, during the early cycles, a short upper discharge plateau (at about 2.45 V) was followed by a much longer lower plateau (at about 2.05–2.1 V). However, in some cases with PEO cells, two distinct lower plateaus can be seen. In addition, the shapes of the

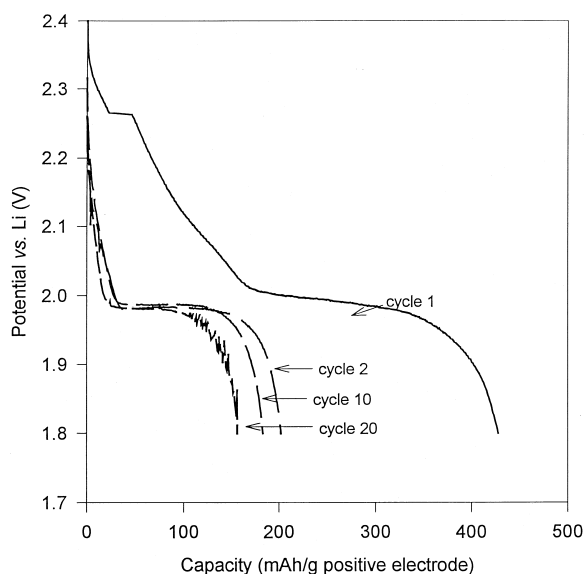


Fig. 7. Selected discharge curves of a PEGDME-electrolyte cell cycled at  $0.05 \text{ mA h/cm}^2$  and  $23^\circ\text{C}$  with a 1.8 V potential cutoff.

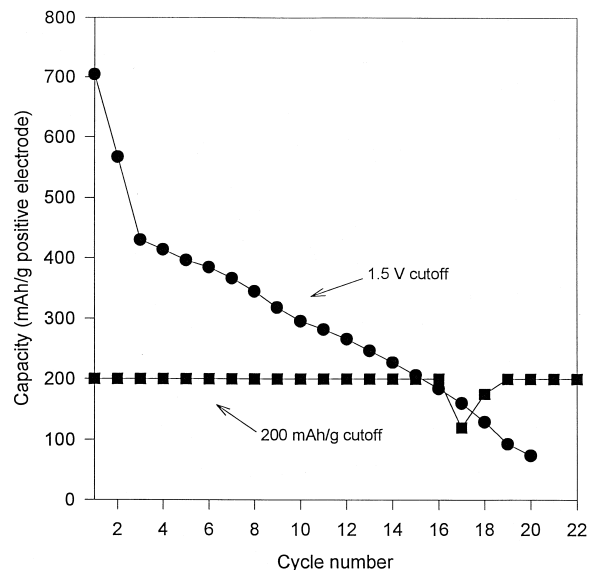


Fig. 8. Comparison of cycle life PEO-electrolyte cell discharged to 1.5 V cutoff ( $0.1 \text{ mA/cm}^2$ ,  $104^\circ\text{C}$ ) vs. one discharged to 200 mA h/g positive electrode cutoff ( $0.1 \text{ mA/cm}^2$ ,  $90^\circ\text{C}$ ).

lower discharge plateaus and the upper charge plateaus sometimes show a significant degree of convexity. Another feature of interest is the small dip, which is often observed between the upper and lower discharge plateaus (see Fig. 5).

During extended overcharges, cells frequently remain on the higher plateau for long periods. Cells discharged fully were therefore charged to a capacity limit of 140–150% of the previous discharge capacity. Some PEO cells were, however, discharged to a capacity limit as well (rather than the usual potential cutoff of 1.5 V). In Fig. 6,

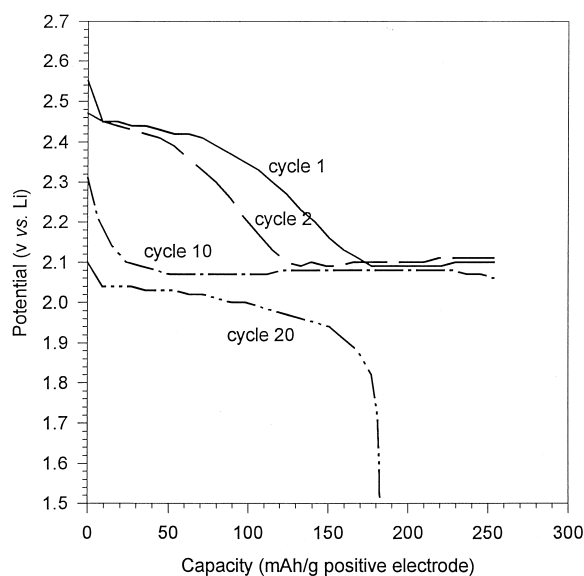


Fig. 9. Selected discharge profiles at  $0.1 \text{ mA/cm}^2$  of a PEO-electrolyte cell cycled at  $104^\circ\text{C}$  with a capacity cutoff of 251 mA h/g positive electrode.

the overcharge plateaus have ended due to capacity cut-offs.

Fig. 8 shows that a longer cycle life is attained when the cell discharge capacity is limited to 24% of the theoretical value. A comparison of the potential profiles of PEO cells discharged to 1.5 V vs. those discharged to 30% or less of the theoretical capacity showed that the upper plateau was retained longer for cells cycled at a lower depth of discharge. Fig. 5 shows the near-total loss of this upper plateau on the second discharge, whereas Fig. 9 shows that this plateau may diminish over several cycles at a low depth of discharge. For PEGDME cells cycled at room temperature, the upper plateau disappeared completely during the second and subsequent discharges (Fig. 7), corresponding to a sharp drop in discharge capacity between the first and second cycles.

In several cases, lithium dendrites limited charging capacity. In addition, leaks in cell chambers sometimes shortened the lives of cells when the cells were cycled in an oven outside the glove box. Though data from these cells could not be used in comparisons of cycle life under different conditions, it was used in analyzing changes in the discharge profiles in early cycles.

From the voltage profiles, it is clear that the phase behavior of the transformation between sulfur and  $\text{Li}_2\text{S}$  is complex. To examine this phenomenon further, slow-sweep cyclic voltammetry (0.005 mV/s) was carried out on selected PEO-electrolyte cells at 90°C. The cyclic voltammograms shown in Fig. 10 exhibit a set of peaks at  $\sim 2.45$ – $2.48$  V corresponding to the upper plateau. The first sweep shows a positive/negative area ratio of  $> 0.8$  and a separation of  $\sim 29$  mV). On the other hand, the

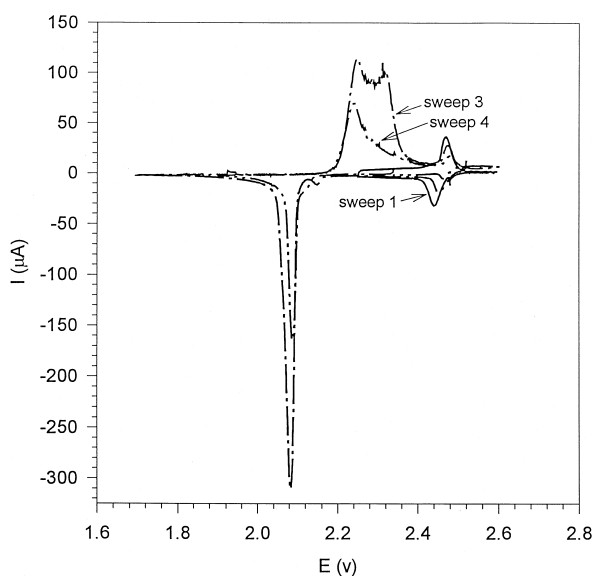


Fig. 10. Cyclic voltammograms at 90°C showing the first, third, and fourth sweeps. The first and third sweep rates were 0.005 mV/s and the fourth was 0.003 mV/s.

peaks which correspond to the 2.2 V plateau, which are observed in the full sweeps, 3 and 4, are more widely separated at about 2.08 and 2.25 V, indicative of significant irreversibility although the peak area ratios are also  $> 0.8$ . The capacity of higher-voltage process of the first sweep is about 20% that of the lower and diminishes in later sweeps. In some cases, more than one peak is visible within the main anodic peak. A third (small) cathodic peak may also be present in some cases.

Some of the cells were dismantled for component examination after testing. In addition, some uncycled PEO-electrolyte cells were observed as they sat at room temperature and at 100°C. The polymer electrolyte of all cycled cells was discolored (usually with a red or reddish-brown color) in the region facing the electrode as well as in the region outside the current path, where applicable. In addition, the uncycled PEO-electrolyte cells that sat at 100°C exhibited this discoloration.

#### 4. Discussion

Polymer ionic conductivity is an important factor in the performance of Li/S cells. The PEMO electrolyte, though it is amorphous at room temperature and has one of the highest conductivities among high-molecular-weight polymers, is too resistive to permit the utilization of most of the active material at room temperature. The significant overpotential of the PEMO-electrolyte cells at 23°C shown in Fig. 2 supports this conclusion. Other PEO-like polymers that are amorphous at room temperature are now under investigation. Unfortunately, their conductivity is not significantly higher than PEMO.

The PEGDME-electrolyte cells at room temperature performed significantly better than the PEMO-electrolyte cells at both room temperature and 60°C. The molecular weight of PEGDME is significantly lower than that of polymer (250 vs. 50,000+). When combined with a lithium salt, its conductivity is much higher than high-molecular-weight polymers, because the smaller chains are much more mobile for lithium ions to move through. However, the conductivity of a composite electrolyte using liquid solvents such as ethylene carbonate, dimethyl carbonate, diethyl carbonate and propylene carbonate instead of PEGDME have been demonstrated to be higher [10]. The high viscosity of PEGDME, compared with liquid electrolytes, is important in the design of the sulfur electrode. It allows the sulfur electrode to remain intact with adequate mechanical strength. Liquid solvents are not sufficiently viscous to form a stable thin-film electrode structure.

The PEO cell, despite its lower conductivity at 90°C, performed best in terms of initial discharge capacity. The higher conductivity of PEGDME when compared to PEO shows that there are factors other than conductivity, which

determine the maximum utilization attainable and the rate of capacity fade.

The highly colored lithium polysulfides are probable intermediates for the reaction of sulfur with lithium [11]. The upper voltage plateau may be a signature of the formation of some of these polysulfide intermediates in both the electrolyte phase and the sulfur electrode phase. Several polysulfides are known to exhibit reduction potentials in the 2-V range, consistent with this upper voltage plateau [12]. Other investigators have observed such compounds in liquid systems and have proposed reaction schemes involving disproportionation and dissociation reactions, including reactions that produce zero-valent  $S_8$  (elemental sulfur) [11,13]. Irreversible chemical reactions could explain the disappearance of the upper plateau as cycling continues. This may also be due to partial irreversibility of the final reaction to lithium sulfide ( $Li_2S$ ). The retention of the upper plateau in low-depth-of-discharge cycling may be due to remaining elemental sulfur. The complete disappearance of this upper voltage plateau during room-temperature cycling after the first cycle suggests that the reversibility may be temperature-dependent.

The convex shape of the lower plateau may be a reflection of a changing overpotential as equilibria between the different polysulfides shift. Shifting equilibria may also explain the shape of the potential profile in the region between the two plateaus in Fig. 5. The small dip observed in discharge plateaus in Fig. 5 might reflect the nucleation of crystalline lithium sulfide phases.

Lithium polysulfides are soluble in the polymer electrolyte, as demonstrated by the discoloration of the electrolyte in the region outside the current path. These intermediates are charged, and, as such, may migrate to and from the lithium electrode via a shuttle mechanism, which may result in the large overcharge capacity observed experimentally. However, as recent evidence in symmetrical Li/polymer/Li cells indicates [14], these long overcharges may also result from soft shorts produced on the lithium electrode. If polysulfides reach the negative electrode and react with metallic lithium, results from three-electrode cells demonstrate that they must form an ionically conductive or porous passivating layer.

A high rate of capacity fade was observed for all electrolytes studied. Three plausible explanations may be given. One is the irreversibility of some of the polysulfide reactions. Another is a loss of polysulfides into the electrolyte. A third is the loss of electrical contact during cycling. The last factor may be due to several phenomena, including the formation of large particles of highly resistive sulfur or lithium sulfide, the movement of polysulfides away from the carbon phase, and the agglomeration of sulfur or carbon particles as a result of the pressure exerted on the cell.

Our data on the improvement of the cycle lives of PEO cells discharged to less than 30% of the theoretical capacity suggests that the capacity fade may be due, at least in

part, to irreversible reactions in the latter part of the discharge. At a depth of discharge of 10–12%, each elemental sulfur molecule could be converted to  $Li_2S_8$ . Discharging to twice this depth of discharge could produce  $Li_2S_4$ . These two polysulfides could then disproportionate and electrochemically react to partially irreversible products, leading to an increased rate of capacity fade.

Another plausible explanation for this phenomenon is the variation in the loss of electrical contact in cycling at a different depths of discharge. In shallow-depth-of-discharge cycling, smaller resistive sulfur and  $Li_2S$  particles may form, thereby providing more active material accessible for subsequent charge or discharge.

Optimization of the system will require a determination of the maximum total capacity attainable as a function of depth of discharge. In addition, sulfur loading will be an important factor in this optimization. As Fig. 3 shows, the realizable capacity of the 75 wt.% sulfur/PEGDME cell was superior to that of the 50 wt.% sulfur cell even though a lower percentage utilization of sulfur was achieved. This demonstrates the importance of balancing the amount of sulfur and supporting materials (e.g. PEGDME, LiTFSI, Carbon, PEO) to achieve the highest capacity per unit electrode weight.

## 5. Conclusions

Lithium/polymer-electrolyte/sulfur cells possess a high realizable specific energy of 1665 W h/kg and capacity of over 1600 mA h/g sulfur. Although more information is necessary to determine the cause of the rapid capacity fade, which limits useful cell lifetime to about 20 cycles with high depth of discharge, Li/S cells show great promise for both consumer and electric vehicle applications.

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