REVIEW

Recent advances in lithium solid state batteries

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The most recent studies of solid state batteries, based on lithium conducting electrolytes and capable of operating at room temperature, are reviewed. This type of battery is of particular technological interest since, theoretically, it possesses high voltage and high specific energy. Unfortunately the known lithium electrolytes have low specific conductivities and consequently the cells so far developed exhibit high internal resistance. In recent years, however, there has been a great effort to overcome this shortcoming, as pointed out in this paper. The review, therefore, is intended to provide the reader with an entrée into this particular and important aspect of solid state battery technology.

Introduction

In recent years there has been a renewed interest in solid state batteries since they possess a number of typical advantages, such as miniaturization, long shelf life and rugged construction. Special attention has been given to those batteries using highly conducting electrolytes of the MAg_4I_5 type ($t_{Ag^+} = 1$) [1, 2]. These systems, however, even if favourably characterized by a relatively low internal resistance, present some disadvantages, the most serious being the low open circuit voltage (OCV) and the low specific energy density. In fact, in the case of the cells based on the $RbAg_4I_5$ electrolyte, the OCV and the energy density are, at best, 0·66 V and 4–6 Wh/kg, respectively, at 25°C [2]. These disadvantages would be overcome by the development of a solid state cell having a high OCV and a performance at least comparable with that of the cells based on silver ion conductivity. Various attempts have been made so far to achieve this goal and those utilizing lithium as anode material will be reviewed here. The review

will be limited to those cells capable of operating at room temperature.

Thin film and doped electrolyte cells

The use of lithium in solid state cells is certainly very promising, due to the high potential and the low equivalent weight of the metal. This implies the utilization of a lithium conducting electrolyte to separate it from the cathode. Some lithium ion conducting salts are known, but unfortunately none of them possesses high conductivity (see Table 1) at room temperature. This was pointed out in a recent review by Pizzini on ionic conductivity in lithium salts [3] and the reader is referred to this work for the properties of these compounds.

As far as the conductivity is concerned, it is clear from Table 1 that none of the known electrolytes may be used in large thickness to develop a high power cell. One way of trying to solve this problem is based on thin film and doping procedures. An intensive and important work in these directions has recently been under

Table 1. Specific conductivity of lithium solid electrolytes

Electrolyte	Temperature (°C)	Ionic conductivity $(\Omega \text{ cm})^{-1}$	Reference
LiI	25	$1\cdot17 \times 10^{-7}$	[4]
LiI(CaI₂ 0·2% mol) initial	28	$2\cdot3 \times 10^{-6}$	[11]
LiI(CaI₂ 1% mol) initial	28	$1\cdot2 \times 10^{-5}$	[11]
LiI(CaI₂ 0·4% mol) stable	28	$2\cdot6 \times 10^{-6}$	[11]
4LiI . NH₄ I	25	2×10^{-6}	[8]

taken at Mallory & Company Inc., in the U.S. In 1969 Liang and Bro [4] described the cell

$$\text{Li} / \text{LiI} / \text{AgI} \qquad (1)$$
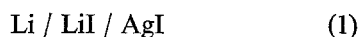
To minimize the internal resistance value, the authors adopted the solution of thin film construction. To achieve this, an AgI–H_2O slurry was spread on a silver foil and this assembly was then passed through a three-zone furnace. In this way a silver iodide film tightly bonded to the silver foil was obtained. On this AgI layer, the electrolyte, LiI, and subsequently the anode, Li, were then vacuum deposited. The vacuum deposition setup was particularly accurate and allowed the control of the deposition rate and the measurement of the film thickness [5]. Nevertheless some basic precautions were taken in order to achieve the best results. For instance the authors reported that, in order to obtain a smooth pinhole-free LiI film and consequently a good yield from the electrochemical process, the AgI substrate had to be maintained at low temperature, preferably below −20°C, and the

thickness of the LiI film had to be greater than 5μm, preferably 15 μm [5]. The OCV of cell (1) was 2·1 V, at 25°C, in agreement with the thermodynamic value related to the discharge process

$$\text{Li} + \text{AgI} = \text{LiI} + \text{Ag} \qquad (2)$$
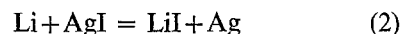
The characteristics and performances of cell (1), reported in Table 2, seemed to indicate a certain advantage and usefulness of the cell, especially in applications where high voltages and low currents are required. Unfortunately the system is not stable and failed after storage for 2–4 weeks. This, as reported by Liang [6], was due to interdiffusion of Li^+ and Ag^+ ions, which took place even at room temperature. This process resulted in a direct short circuit of the cell. To overcome this, Liang has proposed the substitution of AgI by CuI, as cathode material, in order to obtain the thin film cell

$$\text{Li} / \text{LiI} / \text{CuI} \qquad (3)$$

where the cathode was assembled by pressing a

Table 2. Characteristics and performances of cell (1) at 25°C [4, 5]

Anode thickness	$0\cdot4 \times 10^{-3}$ cm
Electrolyte thickness	$1\cdot5 \times 10^{-3}$ cm
Cathode thickness	$3\cdot0 \times 10^{-3}$ cm
Geometrical area	$2\cdot36$ cm²
OCV	2·1 V
Internal resistance*	2×10^4 Ω/cm²
Capacity (at medium rate: 25–100 KΩ load)	0·85 mA.h
Discharge efficiency	~50%
Shelf life	2 − 4 weeks

* As calculated from the reported cell polarization curve [4, 5].

powder mixture of CuI and LiI on a thin copper foil [7]. The electrolyte and the anode were vacuum deposited as in the case of cell (1). Cell (3) had an OCV of 2·06 V at 25°C, which agreed well with the thermodynamic value of 2·05 V calculated for the reaction

$$Li + CuI = LiI + Cu \qquad (4)$$
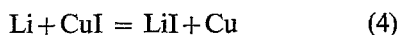
The cell was reported to be more stable than cell (1); the average self-discharge rate, at 25°C and over a period of storage of 4 weeks, was less than 0·2 $\mu A/cm^2$. Also the discharge efficiency was higher, ranging from 95%, at 5 $\mu A/cm^2$, to 60%, at 40 $\mu A/cm^2$ [7].

Another attempt to improve lithium solid state cells is reported in a recent patent by Liang and Epstein [8], where a new electrolyte of 4 LiI.NH$_4$I composition is described. The ionic specific conductivity of this salt, 2×10^{-6} $(\Omega$ cm$)^{-1}$ at room temperature, is still low, being only one order of magnitude higher than that of pure LiI. This improvement in conductivity, however, allowed the construction, by simple subsequent compression of the powdered components, of the cell

$$Li \,/\, 4LiI.NH_4I \,/\, NH_4Ag_4I_5 \qquad (5)$$

The electrolyte thickness was still very thin, as shown in Table 3 where cell characteristics and performances are reported. The OCV of the cell is 2·1 V at 25°C, in good agreement with the value calculated thermodynamically on the basis of the discharge process of the cell. This process [9] is the oxidation of Li by NH$_4$Ag$_4$I$_5$

$$4 Li + NH_4Ag_4I_5 = NH_4I + 4 Ag + 4 LiI \qquad (6)$$

It is known [10] that NH$_4$Ag$_4$I$_5$ is unstable below 32°C and decomposes into less conductive products, according to the reaction

$$2 NH_4Ag_4I_5 = (NH_4)_2AgI_3 + 7 AgI \qquad (7)$$
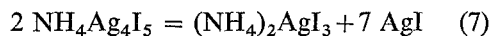
Therefore the overall cell internal resistance should increase accordingly with time. On the other hand this effect may be negligble, since the cell resistance is mainly due to the electrolyte layer. In fact the conductivity of AgI (10^{-4} Ω^{-1} cm^{-1}) is still higher than that of 4 LiI.NH$_4$I (10^{-6} Ω^{-1} cm^{-1}) at 25°C.

Finally the possibility of solving the problem of the high resistance of lithium cells by using doped electrolyte has been carefully examined by Liang and co-workers. In 1971 Schlaikjer and Liang [11] proposed LiI doped by CaI$_2$ as an improved electrolyte to use in solid state cells. It is known that divalent cation doping induces defects in the crystal lattice of LiF or LiCl or LiBr and that their conductivities increase accordingly [12]. In the case of LiI, however, it seemed that the conductivity was practically independent of the dope concentration [12, 3]. This has been recently refuted by Jackson and Young [13] and, in a way, by the cited work of Schlaikjer and Liang. These latter authors, in fact, found that the conductivity of polycrystalline LiI increased linearly with the concentration of CaI$_2$, up to 1 mol% of the latter. However it should be borne in mind that these results were obtained immediately after quenching the melt containing LiI and the dope. As the authors pointed out, measurements taken after stabilization, at 28°C, showed that, for a dope concentration higher than 0·4 mol%, the conductivity remained constant and independent of the initial CaI$_2$ added [11]. This was explained by the authors on the basis that the equilibrium solubility of CaI$_2$ in LiI at 28°C is less than 0.4 mol%.

Table 3. Characteristics and performances of cell (5) at room temperature [8]

|	
Anode thickness	0·04 cm
Electrolyte thickness	0·065 cm
Cathode thickness	0·065 cm
Surface area	1·2 cm^2
OCV	2·1 V
Internal resistance*	$\sim 3·3 \times 10^4$ Ω/cm^2
Shelf life	Extremely long

* As calculated from the reported discharge test [8].

Table 4. Characteristics and performances of cell (8) at 28°C [11]

Electrolyte composition	LiI(0·2 mol% CaI$_2$)
Electrolyte thickness	4×10^{-2} cm
Geometrical area	0·7 cm^2
OCV	2·1 V
Internal resistance*	$1·8 \times 10^4$ Ω/cm^2
Short circuit current density	0·11 mA/cm^2
Shelf life	Not reported; presumably similar to that of cell (1)

* As calculated from the reported cell polarization curve [11].

At this maximum value of CaI$_2$ solubility, the increase in the LiI conductivity is approximately one order of magnitude (see Table 1). Nevertheless Schlaikjer and Liang concluded that this improvement justified the use of the doped LiI as electrolyte in the solid state cell

$$\text{Li / LiI (CaI}_2\text{) / AgI} \qquad (8)$$
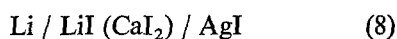
which was assembled by subsequent pressing of the powdered components into a single pellet. The OCV was 2·1 V at 28°C, as in the case of cell (1), where the discharge process is the same. The characteristics and performances of cell (8) are reported in Table 4. A comparison between the internal resistance value of cell (8) and that of cell (1) shows an improvement of the former, when differences in electrolyte thickness and cell area are considered. One has also to point out that the pressing method, used in cells (5) and (8), is a much easier procedure of assembling than the vacuum procedure, used in cell (1). In the paper of Schlaikjer and Liang no mention is made of the stability of cell (8). However it is physically similar to the cell (1) and one would expect the same degradation due to the Li$^+$ and Ag$^+$ interdiffusion process [6]. Therefore as a

first approximation, the shelf life for cell (8) is probably similar to that of cell (1) (see Table 4).

Cell (8) was subsequently greatly improved by the use of an electrolyte having at 25°C a specific resistivity two orders of magnitude less than that of pure LiI. Apparently Liang succeeded in finding doping materials having solubilities in LiI larger than that of pure CaI$_2$ and therefore capable of increasing the LiI conductivity to a greater extent. In fact in 1971 Liang reported on an electrolyte consisting of LiI doped with various additives having at 25°C a specific conductivity of 10^{-5} (Ω cm)$^{-1}$ [14]. The nature of these additives was not specified. The electrolyte was used in a solid state cell as separator between a lithium anode and a cathode consisting of a mixture of a heavy metal halide, an electronic conductor and the electrolyte. This cell may be basically indicated as

$$\text{Li / LiI (doped) / heavy metal halide} \qquad (9)$$
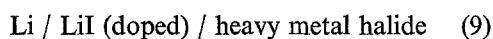
Cell (9) was assembled by pressing the components into pellets which were then hermetically sealed in a metal–ceramic container. The characteristics and performances of the cell are reported in Table 5.

Table 5. Characteristics and performances of cell (9) at 25°C [14]

Electrolyte thickness	0·018 cm
Geometrical area	1 cm^2
OCV	1·9 V
Internal resistance*	$1·8 \times 10^3$ Ω/cm^2
Capacity (at 50 A discharge)	6·5 mA.h
Discharge efficiency (1 V cutoff)	80%
Shelf life (tested)	More than 18 months
Shelf life (anticipated)	20 years

* As calculated from the reported cell polarization curve [14].

One of the most important features of the cell is certainly the relatively low internal resistance which results from the use of the LiI-doped electrolyte. The long shelf life also represents an important achievement. As the author remarked, after 18 months of storage at room temperature, the cell capacity was practically unchanged. The life test was accelerated by storing the cell for 7 months at 105°C and also in this case no variation in the cell capacity was observed [14]. Liang therefore concluded that the self-discharge rate of the cell was negligible and anticipated a shelf life of 20 years. In this cell therefore the interdiffusion process noted in cell (1) has been avoided. This is undoubtedly related to the use of a stable cathode material which has been generally indicated by Liang as a heavy metal halide [14]. Cell (9) was reported as capable of operating over a wide range of temperature. At 100°C a discharge of 1 mA/cm^2, with an efficiency of 90%, was obtained, while at −40°C the discharge current density was about 0·5 μA/cm^2, with an efficiency of 80% [14].

In summary, the extensive work so far performed by Liang and co-workers has resulted in cell prototypes having at room temperature resistances of about 2×10^3 Ω/cm^2 and outputs on average of 0·1 mA/cm^2. This is certainly a remarkable achievement. Nevertheless the electrolyte proposed has a conductivity value far inferior to that of the silver conducting electrolytes (typically 0·2 Ω^{-1} cm^{-1} at 25°C). Therefore the problem of developing a solid state lithium cell having an internal resistance value comparable with that of the silver cells is still unsolved and many workers are studying it intensively.

Charge transfer complex cells

Recently the use of charge transfer complexes as cathodes in solid state cells has been reported. A charge transfer (CT) complex may be defined as a system involving organic molecules acting as electron donors (typically pyrene or perylene or benzidine) and inorganic molecules, acting as electron acceptors (typically halogens) [15, 16]. The interesting property of these complexes is the fact that they are semiconductors with resistivities several orders of magnitude less than those

of either donor or acceptor. Several studies on the electrical properties of the CT complexes have been reported [15–20] and therefore here attention will be focused only on the behaviour of the CT complexes as electrode materials in solid state cells. In fact the use of these complexes as halogen, and in particular as iodine, electrodes is interesting for the following reasons:

(a) the CT complexes possess low resistivity and therefore they may be used as electrodes without addition of inert components, such as graphite, to assure good conductivity, as in the case of the iodine cathodes most used in solid state cells up till now. This fact should ultimately result in specific energy advantages for cells using CT complexes as electrodes;

(b) the CT complexes maintain their low resistivity for a wide range of halogen composition. For instance, in the case of the perylene–iodine complex, the resistivity only changes from 20 to 25 (Ω cm) in an iodine to perylene mole ratio range from 6 to 1 [19]. This circumstance assures the good behaviour, as iodine electrode, of the CT complex during discharge.

The CT complexes were first used as battery electrodes in 1966 at the Jet Propulsion Laboratory, in the U.S.A. At that time it was announced that studies by Hermann, Rembaum and Gutmann resulted in the development of a battery having a magnesium anode and a cathode formed by the iodine–perylene complex [21]. The cell, approximately 2·3 cm^2 area, had at room temperature an OCV of 1·5 V and outputs of a few mA/cm^2. The discharge process was the formation of magnesium iodide. The cell was described as rechargeable and with an indefinite shelf life. Subsequently the authors reported more extensively on these CT complex solid state cells [22], with the attention particularly concentrated on magnesium anodes. This work is nevertheless of interest here since magnesium can be substituted by lithium, as will be pointed out subsequently.

In the work of Gutmann and co-workers it is reported that solid state cells may be assembled coupling anodes of magnesium with cathodes consisting of CT iodine complexes. In these

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Table 6. Characteristics and performances of cell (10) at room temperature [22]

Cell diameter	1·26 cm
Cathode composition	I_2–perylene (50% in weight I_2)
Cathode thickness	0·1 cm
OCV	1·45–1·85 V
Short circuit current	1·1–16 mA

cells the electrolyte, indicated as MgI_2, was formed *in situ* during contact of both electrodes. These cells had OCV values comparable with those thermodynamically predictable from MgI_2 formation processes and exhibited discharge capabilities. The most reasonable explanation is that iodine reacts with magnesium to form a MgI_2 film which acts as the electrolyte. The most probable cell structure is therefore

$$\text{Mg} / \text{MgI}_2 / \text{I}_2\text{–CT, carbon} \qquad (10)$$
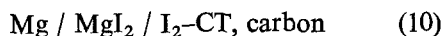
Some characteristics and performances of cell (10) are reported in Table 6. There is some controversy in the literature on the cell mechanism and on the environment in which the cell may operate, as extensively reported in a recent review by Hull [23]. Friedel found that in the complete absence of moisture, such as in a desiccator, cells similar to (10) did not operate, showing both zero OCV and no short circuit current [24]. This author therefore concluded that water participates directly in the cell reaction, forming magnesium hydroxide and hydrogen. Gutmann and co-workers reported that the complete removal of water reduced the short circuit current of cell (10) by two or three orders of magnitude and caused the OCV to drop to a value approximately 30% lower than the thermodynamic one [25]. These authors also found that the presence of vapours of high permittivity liquids improved the magnesium cell performances [25]. Finally in a recent patent by Blackburne it is reported that the addition of water to the magnesium–CT complex cells raised the short circuit current by a factor of a thousand [26].

At the present time it seems reasonable to presume that in the discharge of cells similar to cell (10), the CT complex acts as in any iodine–donor mixture, except that the necessary elec-

tronic conductivity of the cathode is achieved without the addition of an intrinsically conductive material, such as graphite. Part of the iodine of the complex reacts with the metal used as the anode to form an iodide film which serves as the electrolyte. In the case of the Mg–CT complex cells, traces of high permittivity solvents are necessary for the cell operation, probably because they partially dissolve MgI_2, thus increasing the electrolyte conductivity. The Mg–CT complex cell, therefore, may be more correctly classified as a 'dry' battery rather than a 'solid state' battery.

It is obvious that lithium may be substituted for magnesium to similarly originate a Li–CT complex cell, with immediate advantages in OCV and energy density values. Recently Schneider, Moser, Webb and Desmond have reported on a new high density solid electrolyte cell [27], having basically a lithium anode, LiI as electrolyte and 'an iodine containing cathode'

$$\text{Li} / \text{LiI} / \text{I}_2\text{-based cathode} \qquad (11)$$
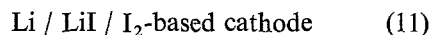
The electrolyte was obtained by exposing the anode, in the form of a lithium foil, to iodine vapours or 'simply by contacting the anode and cathode'. The cell components were maintained in adhesion by applying a small pressure, i.e. about 2 kg/cm². The authors also stated that a very thin film of electrolyte, only 1 μm thick, was sufficient to separate the anode from the cathode. This is in contrast with the findings of Liang and co-workers. As previously mentioned, these latter authors reported that to assure insulation between the electrodes of cell (1)—which uses the same electrolyte as cell (11)—the electrolyte thickness had to be greater than 5 μm [5]. This discrepancy, however, may be accounted for by the different cathode materials used in the two cases.

*Table 7. Characteristics and performances of cell (11) at 25°C [27]*

Humidity content	Less than 2%
Anode thickness	0·008–0·03 cm
Cathode thickness	0·032–0·12 cm
Initial electrolyte thickness	$\sim 10^{-4}$ cm
Initial internal resistance	150 Ω/cm^2
Internal resistance after 1 year storage	11 000 Ω/cm^2
Initial short circuit current density	20 mA/cm^2
Short circuit current density after 20 days of storage	1 mA/cm^2
Short circuit current density after 110 days of storage	0·5 mA/cm^2
OCV	2·8 V
Energy density	300 Wh/kg

Schneider and co-workers do not state which cathode material was used in cell (11), but from the reported fact that the electrolyte could be formed *in situ* by contacting anode and cathode, one may reasonably assume that the cathode was one of the iodine–CT complexes. The performances and characteristics of cell (11)—which are reported in Table 7—will therefore be discussed on the basis of this assumption.

One of the most interesting features of cell (11) is its very low internal resistance, reported as 150 Ω/cm^2 at 25°C. This figure is not in agreement, however, with that calculated on the basis of the cell dimensions (see Table 7) and of the resistivity of LiI (see Table 1). This calculation in fact would give a value of 1000 Ω/cm^2. Nevertheless this latter value would also be a good achievement since it is of the same order of magnitude as that of cell (9). Unfortunately the resistance of the cell, as reported by Schneider and co-workers, increases with time. This is apparently due to self-discharge as a consequence of the migration of iodine from cathode to anode, through LiI [27]. This produces, for a cell of 1 cm^2 of surface, an increase in internal resistance up to 11 000 Ω, after 1 year of storage at 25°C. The cell short circuit current accordingly decreases with the time of storage, as shown in Table 7.

It is interesting to notice that cell (11) was assembled in an atmosphere containing less than 2% humidity [27]. Despite this low humidity level, the cell is capable of operating, whilst this is not the case, in comparable conditions, for the magnesium–CT complex cells. In the case of lithium–CT complex cells, therefore, the presence of solvent does not seem to be necessary for the cell operation. This is confirmed by recent studies performed in our laboratory on the cell

$$\text{Li / LiI / } I_2\text{–benzidine} \qquad (12)$$
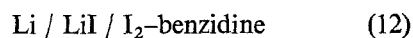
which was assembled by sandwiching a lithium pellet with an iodine-benzidine (3 to 1 mole ratio) pellet [28]. It was found that cell [12] may operate even if assembled and maintained in an atmosphere containing only a few parts per million of water. The explanation for the different behaviour of the two types of cell could be found in the fact that while LiI is a fairly good ionic conductor, this is not the case for MgI_2. Therefore the magnesium cell, as opposed to the lithium cell, needs the presence of the solvent to partially dissolve the electrolyte and consequently increase its ionic conductivity.

Conclusions

From the information reported above, it is clear that the interest in a low-resistance, high-power lithium solid state cell is enormous. Such a cell in fact would provide open circuit voltages of the order of 2–3 V. To obtain comparable voltages with the silver solid state cells, for instance with those based on the $RbAg_4I_5$ electrolyte, a combination in series of four units would be necessary [2]. This on the other hand would represent a sacrifice of the miniaturization which is one of the most important characteristics of solid state cells. Furthermore one of the

main shortcomings of the silver solid state cells, i.e. the low specific energy [2], would be completely overcome with a lithium cell, having comparable internal resistance. In fact the Li/I_2 couple, for instance, has a theoretical specific energy of 560 Wh/kg versus the 78 Wh/kg value of the Ag/I_2 couple. Finally, lithium is a much cheaper material than silver. This factor becomes particularly important when the development of primary batteries is considered, as is the case of these solid state cells.

Unfortunately, despite all the efforts so far made in this field, the development of a lithium solid state cell having, at room temperature, a performance similar to that of the silver cells has not yet been completely accomplished.

The cell developed by Liang, using LiI doped with various additives as electrolyte, is the system which so far gives the best performances. Also the cells based on charge transfer complexes as cathodes are very interesting. For these systems, however, there are still uncertainties concerning the stability and, consequently, the shelf life. Furthermore the internal resistance of these cells increases during operation because of the additional formation of LiI as discharge product. On the other hand this is also the case of cells based on the $RbAg_4I_5$ electrolyte, where poorly conductive AgI is formed upon discharge [1]. In both cases this effect is of secondary importance since it could be rather easily overcome by a suitable cell geometry. The main problem for the lithium cells definitely remains in the high resistivity of the so far known lithium solid electrolytes. Therefore most of the future efforts should be concentrated on research into new highly conductive lithium salts.

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