A REVIEW ON LITHIUM — ION POLYMER ELECTROLYTE BATTERIES

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Battery power as a portable energy generator stands as a competitor to IC engines in view of its inherent capability to provide energy for multifarious applications without atmospheric pollution. Among all the existing power sources, the recently conceived polymer electrolyte concept in the place of conventional electrolytes has paved way for the new era of Lithium-ion polymer batteries. Due to the outstanding performance of these batteries, the lithium-ion polymer battery is the future battery of the Universe. An extensive literature survey has been made in this regard and the output is presented in nutshell.

Keywords: Polymer electrolytes, plasticiser & conductivity.

INTRODUCTION

After 25 years in the doldrums, with relatively uneventful market performance based on Nickel Cadmium and Lead acid batteries, the rechargeable battery market has erupted in the last five years. The driving force for this rejuvenation is the demand for higher energy / power sources for cellular phones and laptop computers.

The lithium-ion polymer battery system, has the same theoretical high performance level as the liquid electrolyte system along with light weight, due to the use of foil-lined plastic materials [1]. Polymer matrix, an immobile electrolyte with no free liquid in the cell is viewed as an increased safety factor by some researchers. The lower manufacturing cost, easy fabrication, and its possible usage in series and parallel strings to meet most power requirement raise the status of lithium-ion polymer batteries to an impeccable level. Therefore the lithium-ion polymer battery with the same basic cell chemistry and theoretical energy storage capability as the lithium-ion cells with liquid electrolyte, is the most probable candidate for next millennium. The introduction of " smart " battery packs make the battery pack market to undergo a major shift in chemistries with the smart chemistries (lithium-ion & nickel-metal hydride) displacing the dumb chemistries (nickel - cadmium) [2] and evidently this is the supporting evidence for the same.

Comparison of various battery technologies

In association with the rapid progress in size and in thickness reduction of electronic devices and development of multimedia industries in recent years demand has been increasing for small size electronic devices including cellular phones and PDA. With this situation as the background the battery industry has a very important role to play. Replacement of conventional batteries with nickel-metal hydride and lithium-ion types is in progress, particularly in cellular phones and the like, while a primary lithium battery which is small in size and large in capacity is drawing attention. The lithium metal battery system is not yet a developed technology because of its poor cycle life. Zinc - air and rechargeable alkaline batteries were not included for discussion in the present study because of their limited availability, restricted marketing acceptance due to low cycle life and requirement of specific recharging conditions like mechanical recharging. Lithium-ion batteries currently available in cylindrical or prismatic shape are using non-aqueous solvent based electrolytes where the electrochemical window is limited up to only 4 volts. On the other hand studies on the polymer electrolyte started after its discovery by Wright *et al.* [3] in 1973. Its application as a battery electrolyte has become active in recent years as a result of proposals made by Armand *et al.* [4]. The conventional lithium-ion batteries with electrolyte solutions have problems with long term reliability [5] and the polymer electrolytes with excellent thermal resistance have ultimately won the battle in terms of numerous advantages. Hence it is worthwhile to compare the various potential electric vehicle batteries with respect to energy and power density, cycle life and cost (Table I).

Smart chemistry vs marketing

Smart chemistry (Lithium-ion and part of NiMH) views the smart battery pack market in terms of three assumptions (1) Within a few years, Lithium-ion (both non-aqueous &

polymer) will be the dominant chemistry for portable electronics applications. (2) Ni- Cd chemistry may decline in all application areas in due course. (3) Ni-MH is currently popular, but it will decline in market share as Lithium-ion (non-aqueous & polymer, preferably polymer) displaces it.

The battery market forecast by NRI clearly says that the Lithium- ion production is expected to exceed Ni-MH cell production by the turn of the century. The low cost of the Ni-Cd system makes it the candidate of choice for most low-end devices. Therefore the growth in the rechargeable battery market will be met with Ni-MH and Lithium-ion cells. Because of its lighter weight & cost equivalence, the Lithium-ion is expected to capture the larger portion of the growth. The Lithium-ion polymer battery, just now appearing in the market and opening new horizons for battery applications has proved its superiority over non-aqueous system after the announcement of "world's thinnest" Mitsubishi computer with a polymer battery of Ultralife in October 1997.

Salient features of Lithium-ion polymer batteries

* Advanced rechargeable technology
* Inherent packaging flexibility
* Shaping flexibility to unusual configurations
* Higher energy density (3 times that of Ni-MH)
* Wide Electrochemical window upto 5 V
* Perfect safety & Reliability
* Longer cycle life with lesser capacity fade
* Good volume utilisation
* Vast applications [viz. MEMS, EV & Space (Teledesic communication satellites)]

General performance of Lithium-ion polymer batteries

Operating Voltage	: 3.8 V
Specific energy	- 130 Wh/Kg
Energy density	- 246 Wh/l
Cost (cell level)	- \$ 2.5 - 3.5/Wh
Cycle life	- > 300 cycles
Self discharge	- < 0.1% per month
Operating temperature	- 253 - 328 K
Charge rate	- 80% capacity in 1 hour
	100% capacity in 3 hours
Environmental concerns	- non-toxic

Reported combinations of cathode and anode

Anode	Cathode
Graphite	- $LiCr_{0.02}Mn_{1.98}O_4$
Natural graphite	- $LiCoO_2$ & $Li_4Ti_5O_{12}$
Plasticized graphite	- $LiNiO_2$ & $LiMn_2O_4$
Carbon	- $Li_{1.05}Mn_2O_4$ & $LiMn_2O_4$
Lithiated carbon	- Lithiated metal oxides
Lithiated graphite (Li_xC_6)	- $Li_xMn_2O_4$, $LiMn_2O_4$
	$Li_{1-x}Mn_2O_4$, $LiNiO_2$, $LiCoO_2$

The present Lithium-ion polymer cell performance is approaching 150 Wh/Kg. The improvement result from higher capacity cathode materials produced by replacing a portion of the cobalt with higher energy nickel in the oxide structure. All the present producers have announced service improvements in addition to an expectation of approaching 200 Wh/Kg in the near future [1].

As price becomes more important, a shift away from cobalt and nickel oxide cathode to lower cost manganese cathode materials can be expected. Moli Energy is the first to make this transition. Two more driving forces behind this transition are

* Manganese oxide spinel is stable above 4.2 V and can tolerate charging to higher voltages than can the cobalt-nickel oxide cathode cells. Therefore sophisticated voltage control circuitry is not required, thus gives a considerable cost saving.

* Manganese has large reliable supply of low cost raw materials, despite the fact that cobalt & nickel resources are limited and their raw materials are more expensive.

Even performance wise, $LiMn_2O_4$ is favoured compared to those of $LiCoO_2$ & $LiNiO_2$ as is evident from Table II.

TABLE I: Comparison of potential electric vehicle batteries

Battery systems	Energy density (Wh/Kg)	Power density (Wh/Kg)	Service life cycles	Cost \$
Lead-acid	33-50	80-300	400-1000	150
Ni-Cd	40-60	125-300	100-1500	450-750
Ni-MH	55-70	100-200	600-1200	30
Li-polymer	130	100	300	100-500
* Zinc air	150-200	60-220	1500	100
# Zinc Br$_2$	70-80	35-60	300-500	150-300

* requires mechanical recharge, # hazardous in nature

Highlights of certain proven systems based on different polymer electrolytes

Polymer electrolyte-based Lithium-ion batteries offer many advantages over those containing liquid electrolytes. For example, they can be compacted in plastic materials to yield higher energy densities, are amenable to inexpensive manufacturing employing the lamination and packaging processes used commonly in the plastics industry, and are potentially safer due to the absence of highly volatile liquids. Polymer electrolyte batteries also have the ability to be geometrically compliant, a feature which allows them to be fabricated in the shapes required by a wide variety of products such as cellular phones, pagers, power drills etc. Practical batteries based on polymer electrolytes have not yet become a reality because of the unavailability of electrolytes with high enough ambient temperature conductivity. The conventional polymer electrolytes composed of complexes of Lithium salts with long-chain polyethers, such as Poly (ethylene oxide) (PEO), have conductivities in the range of 10^{-8}-10^{-5} S/cm at room temperature, significantly lower than 10^{-3} S/cm, which is required for the development of practical batteries capable of discharging at moderate to high rates, i.e., the C/10 to C rates. The preparation of polymer electrolytes with conductivities of this magnitude required a non-conventional approach. Abraham et al. (6-9) have shown that dimensionally stable polymer electrolytes with conductivities of $> 10^{-3}$ S/cm can be prepared by encapsulating organic liquid electrolytes in a polymer matrix, A typical electrolyte is polyacrylonitrile (PAN) plasticised by a solution of $LiPF_6$ dissolved in a mixture of EC and PC. Other polymers such as Poly (-Vinyl Chloride) [10,11], Poly (Vinylidene Fluoride) [12,13], Poly (Vinyl Sulphone) [14,15], Poly (ethylene glycol diacrylate) [6,7] and Poly (vinyl pyrrolidinone) [6,7] can be used along with solutions of $LiPF_6$, $LiN(SO_2CF_3)_2$, $LiBF_4$, $LiAsF_6$ and $LiSO_3CF_3$ in EC/PC. Electrochemical studies have validated the usefulness of these electrolytes for the development of Lithium and Lithium-ion polymer batteries [16]. PAN based electrolytes are especially suitable for high voltage cathodes such as spinel $LiMn_2O_4$.

The Bellcore technology (Patented for valence lithium ion polymer battery) uses poly (vinylidene difluoride) co-polymer for the electrode binding agent as well as for the polymer electrolyte base. The copolymer allows the electrode active masses and the polymer electrolyte to be formed in to flexible, self-supporting films using a solvent casting technique.

Solid polymer electrolyte discovered by Wright et al. would make the fabrication of highly safe batteries possible, allow use of printing techniques, and fabrication of a thin film battery feasible. However SPE has indicated a problem of low ionic conductivity at room temperature which is quite different from the conventional electrolytes. It is known that lithium ion movements depend on segmental motion of polymers [17], so improving movement of the polymer segment is one of the problems to be solved.

Methods to solve this problem include methods to cross link polymers [18] and to introduce polymers into side chains to make them a comb shape [19]. Watanabe and co-workers [20] have achieved 10^{-4} S/cm at 1073 K with a co-polymer of 2-(2-methoxy ethoxy) ethyl glycidyl ether and ethylene oxide with LiTFSI.

Propositions to add plasticizers have been made by a large number of researchers. Organic electrolyte itself has been used as a plasticizer, and it is referred to as a gel electrolyte. Such research has been carried out since 1973 [21]. The SPE prepared through casting methods uses organic solvents as plasticizers possesses improved ionic conductivity ranging from 10-6 to 10^{-3} S/cm at room temperature. A number of reports have been given recently on electrolytes using PAN [22-24], PMMA [25-27] and PVDF [28] as the main chains. Such research is actively elucidating ionic migration mechanisms, as well as improving the conductivity of the electrolyte and its stability under a battery driving condition.

The next important step in lithium ion polymer battery technology will be the production of prototypes having a full plastic configuration, which assures modularity in design and reduction in fabrication costs. Several attempts to reach this goal are presently underway. Many types of highly conducting polymer electrolytes have been developed today and characterised [29-30]. The recently reported gel type electrolyte membrane [31] follows the synthesis of PAN based membranes [30] for its preparation.

Recent reports of Abraham et al. discuss about the conductivities of some PAN electrolytes at room temperature from which PAN electrolytes can be identified as ideal electrolytes for the fabrication of lithium-ion polymer battery (Table III).

Other attractive feature of these electrolytes includes good mechanical strength and excellent electrochemical stability, besides having a simplified preparation. A detailed summary

TABLE II: Comparative performance chart

Type	Capacity Ah Kg^{-1}	Voltage (at mid discharge) V	Energy Wh Kg^{-1}
$LiCoO_2$ – C	22	3.6	78
$LiMn_2O_4$ – C	25	3.7	91
$LiNiO_2$ – C	25	3.2	80

TABLE III: Conductivities of PAN electrolytes

Electrolyte composition (wt%)	Conductivity (S/cm x 10^{-3} at 303 K)
14% PAN : 39% EC : 39% PC : 8% $LiPF_6$	2.4
13% PAN : 59.2% EC : 14.8% PC : 13% $LiPF_6$	4.1
13% PAN : 77.5% EC : 9.5% $LiPF_6$	4.6
13.5% PAN : 37.5% EC : 37.5% PC : 11.5% $LiAsF_6$	2.0
13% PAN : 75% EC : 12% $LiAsF_6$	3.6
14.2% PAN : 39.3% EC : 39.3% PC : 7.2% $LiN(SO_2CF_3)_2$	2.1

of the properties of these electrolytes has been reported elsewhere also [32].

A list of selective polymer electrolytes can be arranged according to their conductance as showin in Table IV.

The temperature dependence of the conductivities of PAN - EC/PC - $LiPF_6$ electrolytes by varying the ratio of EC and PC and the excellent cycling efficiency of PAN electrolyte were studied by Abraham *et al.* Also, the cycling results of these electrolytes when considered for first 18 cycles, include 5 formation cycles and 13 additional cycles in which the rate/cap. behaviour was evaluated. The importance of polymer electrolytes in terms of cycle life and capacity can be understood from Table V.

The construction of a lithium ion polymer battery showed by Brodd *et al.* (1) is a card form of battery with a thickness of 0.45 mm. The electrodes and electrolytes can be made easily because a printing process can be used for fabrication. With respect to this shape, the area can be either reduced or increased easily.

Similarly, the basic building of the valence polymer battery can be depicted in a configuration, termed BI-cell, which is the basic building block of all valance batteries. Polymer films with positive and negative mass are heat laminated together in a single unit with the incorporation of polymer electrolyte separators. The Li-cell building blocks are stacked to give the bi-cell the desired capacity & height.

Regarding the cell assembly, the preliminary engineering data required for the lithium-ion polymer batteries, used in cellular phone & computers are listed below.

Preliminary engineering data

Cellular phones batteries

Designation	Size, mm	Ah	Wt, g
IMP 360965	36.0 x 65.0 x 9.0	1.35	36
IMP 340448	34.1 x 48.0 x 4.1	0.35	13

TABLE IV: List of promising electrolytes — Vital participants

Name of the electrolyte	Conductivity (S/cm)	Reference
GPC (Glass polymer composite) 0.4GeS_2, 0.3 $Li_2S_{0.3}$ LiI and $P(EO)_8.LiN(CF_3SO_2)_2$	4.5 x 10^{-5} (room temp) 8 x 10^{-4} at 353 K	31
PAN - PVDF - PVS based plasticized electrolytes	10^{-4} - 10^{-3} (room temp)	32
PVDF - PEO - $LiClO_4$ electrolyte membrane	2.62 x 10^{-5} at 303 K	33
PVDF - EC - PC - LiX electrolyte [X = SO_3CF_3, PF_6, $N(SO_2CF_3)_2$]	2.2 x 10^{-3} (room temp)	34
PVDF - PC - DMF - LiX [X = AsF_6, $N(CF_3SO_2)_2$ $C(CF_3SO_2)_3$]	3 x 10^{-4} at 298 K	35
PAN - EC - DEC - $LiClO_4$ (19:53.5:23:4.5)	4 x 10^{-3}	36
PAN - EC/PC-$LiPF_6$ PAN - EC/DPC - $LiPF_6$	2 x 10^{-3} - 4.6 x 10^{-3} at room temp	37
2-(2-methoxyethoxy) ethylglycidylether-EO-LiTFSI	10^{-4}	30
$LiPF_6$ - EC - DEC and PVC based electrolytes	——	38
PAN-$LiClO_4$-EC-DEC	——	37
Insitu polymer electrolyte	——	39
PAN-LiTFSI gel or PAN - EC based electrolytes	——	40
BEG - 1 solvents	——	41
PEG based electrolytes	——	42

((— value not reported)

Computer batteries

4 x 4 x 4*	101.6 x 101.6 x 4.0	2.90	87
4 x 4 x 6*	101.6 x 101.6 x 6.0	4.60	122
4 x 4 x 8*	101.6 x 101.6 x 8.0	6.20	164

* Temporary designation

CONCLUSION

Lithium ion polymer batteries using different polymer electrolytes showed excellent charge-discharge and cycling

TABLE V: Highlights of certain proven systems

System	Cycles	Capacity
Graphite/PAN/LiMn$_2$O$_4$	>500	120 Wh/Kg
Disordered LiC$_6$/SPE (In situ)/		
MO (Metal Oxide)	500	80%
C/GPC (Glass plymer		(retention)
composite)/ Li$_{1.05}$Mn$_2$O$_4$	50	110-100
Composite C/PVC based		mAh/g
electrolyte/ composite cathode	20	Improved
C/CPE (composite polymer		sp.capacity
electrolyte)/LiNiO$_2$	120	100%

characteristics in addition to high safety. These polymeric electrolytes are attractive for use as separator cum ion conductive media for the fabrication of solid state lithium -ion polymer batteries. Based on the utility of cathodes & anodes, change of ratio of the electrolytes, change of organic solvents, with a suitable cell assembly may lead to excellent outputs. As a battery design (of Ultralife) has been realised that has a thickness as thin as 0.45 mm, it is possible to arrange such a battery in spaces within electronic device casings or in the liquid crystal panel in notebook type personal computers. This battery could certainly lead to an expectation that electronic devices will in future be further reduced in weight, thickness and length.

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INVESTIGATIONS ON MAGNESIUM POLYMER ELECTROLYTES

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Synthesis and characterization of Polyethylene Oxide (PEO) blended with Magnesium salts viz, $MgCl_2$, $Mg(ClO_4)_2$ are presented in this paper. Microstructural characterization was done using Fourier Transform Infra red spectroscopy (FTIR). AC impedance Technique was used for electrical characterization of the prepared polymer electrolytes. Discharge behaviour of Magnesium button cells assembled using the above polymer electrolytes are discussed in terms of capacity output. Preliminary investigations reveal the possibility of fabricating Magnesium Polymer batteries.

Keywords: Polyethylene oxide, magnesium salts, $MgCl_2$ and $Mg(ClO_4)_2$

INTRODUCTION

Magnesium (Mg) is an interesting anode battery material with more advantages like high standard potential (-2.37 V), low cost, good low temperature performance due to their exothermic corrosion reaction during discharge etc. On the other hand, solid polymer electrolytes have many advantages· in comparison to conventional organic, inorganic or liquid electrolytes. These polymer electrolytes have advantages like

* Thin film forming property

* Good processability

* Flexibility

* Light Weight

* Elasticity

* Transparency

* High ionic conductivity

* Wide potential window

* Kinetically stable

* Moderate temperature (343 to 423 K)

Hence, it is provoking to fabricate and study the performance of a battery system combining magnesium and solid polymer electrolyte like PEO blended with magnesium salts. In the present study we want to look at the effect of solid polymer electrolyte on the performance of the solid state magnesium battery. We report herein a comprehensive investigation carried out on polyethylene oxide (PEO) with a magnesium salt as a possible solid polymer electrolyte in solid state magnesium batteries. In this work the synthesis, structural and electrical characterization are described regarding new polymeric electrolytes for magnesium batteries on the basis of polyether blend polyethylene oxide (PEO).

EXPERIMENTAL

Preparation of anode

Mg AZ 31 alloy sheets with 1.5 mm thickness was used as anode. The Mg sheets were cut into pieces to get 2×4 cm² size of flat anode. Then, a hole is made by drillers on one corner of the Flat electrode. Then the hole is riveted with silver rivet which acts as current collecting points. A flexible copper wire is soldered over the rivet. The rivets as well as the riveted area of the anode was marked with araldite so as to eliminate the bimetallic corrosion of magnesium. The anode was cleaned before it is used.

Preparation of cathode

The cathode mixture containing Manganese oxide, Acetylene black and Teflon (binder) of appropriate proportions are mixed uniformly. The cathode mixture was spread over the mesh and covered with tissue paper and pressed at an optimized pressure.