[0016] 2 The conductivity of solid polymer electrolyte membrane is relatively low at room temperature.

[0017] 3 The properties of the gel-type polymer electrolyte membrane are:

[0018] 3.1 Weak mechanical strength, and easy to be soften at high temperature;

[0019] 3.2 Not suitable for forming membrane due to low viscosity;

[0020] 3.3 Inferior liquid electrolytes absorbability at high temperature;

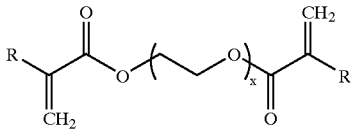
[0021] 3.4 Complex membrane forming process;

[0022] 3.5 Hardly adhesive to electrode

SUMMARY OF THE INVENTION

[0023] A primary object of the present invention is to provide a composite gel-type polymer electrolyte membrane that absorbs liquid electrolytes by PAN gel-type electrolytes, which are copolymerized by acrylonitrile (AN) monomer and crosslinkable monomer with two terminal acrylic acid ester function group. The crosslinkable monomer with two terminal acrylic acid ester function group chemically links to ethylene glycol bond which can transfer lithium ion and have superior compatible to the liquid electrolytes; thereby can absorb liquid electrolytes volume via 6-fold greater than main polymer. It can add PVdF polymer which molecule weight is over 5000, or PVdF containing over 80% PVdF-co-HFP polymer to enhance mechanical strength.

[0024] The liquid electrolytes absorbed in the interpenetrating network (IPN), which consists of a mix of PAN and PVdF polymers, are made from using nonaqueous solvent, (i.e., cyclic carbonate, acyclic carbonate, amide solvent, lactone solvent, ester solvent, etc.), to dissolve alkaline-metal or alkaline-earth metal salts. The chemical formula of crosslinked gel-type PAN electrolytes is represented by Formula I:



(I)

[0025] wherein R: is selected from the group consisting of hydrocarbyl of 1 to 4 carbon atoms; x: 1~10.

BRIEF DESCRIPTION OF THE DRAWINGS

[0026] The structure and the technical means adopted by the present invention to achieve the above and other objects can be best understood by referring to the following detailed description of the preferred embodiments and the accompanying drawings, wherein

[0027] **FIG. 1** is the experimental result obtained from crosslinked PAN electrolyte membrane absorbing different kind of liquid electrolytes.

[0028] **FIG. 2** is the structure illustration of 2016 coin cell batteries with PVdF-PAN complex electrolyte membrane.

[0029] **FIG. 3** is the experiment result of the specific capacity of the battery versus cycle number of the separator that utilizing PVdF-PAN complex electrolyte membrane as the separator of polyaniline secondary batteries (polyaniline as the positive electrode) with lithium metal.

[0030] **FIG. 4** is the experiment result of specific capacity of the battery versus cycle number of the separator that utilizing PVdF-PAN complex electrolyte membrane as the separator of lithium ion batteries ($LiMn_2O_4$ as the positive electrode).

[0031] **FIG. 5** is the experiment result of specific capacity of the battery versus cycle number of the separator that utilizing PVdF-PAN complex electrolyte membrane as the separator of polyaniline secondary batteries with carbon negative electrode.

[0032] **FIG. 6** is the sectional view of three layers structure of $LiCoO_2$, positive electrode; crosslinked PVdF-PAN complex electrolytes membrane; and MCMB, carbon negative electrode.

DETAILED DESCRIPTION OF THE INVENTION

[0033] The secondary batteries of which positive electrode materials are $LiMnO_2$, $LiMn_2O_4$, $LiCiO_2$, $LiNiO_2$, etc.; and the negative electrode materials are lithium metal, alloy lithium or dopants with lithium, wherein, dopants are graphite or irregulated crystalline carbon materials. The secondary batteries of organically conductive polymer of which positive materials are doped or undoped organosulfur and organosulfide polymer, polyaniline, polypyrrole, etc., those are polymerized by chemical or electrochemical method, or formed as complexes of conductive polymer and carbon black by chemically polymerization based on the surface of carbon black or graphite.

[0034] The positive and negative electrode materials mentioned above are substantially powder type, thus, it must utilize adhesive reagent to glue active powder as a membrane during manufacturing processes of electrode. Generally, the binders are PVdF, PTFE, and so on.

[0035] The present invention, composite gel-type polymer electrolyte membrane contains PVdF polymer which is compatible to binder, while coating pre-polymerizing solution to the electrode, the polymer chains will wind between the interfaces. Thus, it leads to the electrolytes closely compacting to an electrode and the adhesive interface will not dissolve in the liquid electrolytes.

[0036] Common lithium ion batteries that are not compatible to electrode, thus, it use metal shell as a material to seal and to connect between electrode and separator. This present invention, composite gel-type polymer electrolyte membrane can closely compact to electrode natively. Besides, the liquid electrolytes are absorbed in the crosslinked polymer PAN to prevent leaking. Therefore, it can be designed as cartridge a type and also as a reduction in weight and volume of batteries.

[0037] Corresponding to valuable and practical polymer electrolyte membrane, the properties of the high absorbility to liquid electrolytes, well mechanical strength, superior compatible to electrode are most important. The high absorbility to liquid electrolytes can arise the conductivity and