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(54) **Electrolyte for batteries and battery packs**

Elektrolyt für Batterien und Batterieblöcke

Électrolyte pour batteries et blocs-batteries

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EP-A2- 0 825 663 **WO-A-2008/109734**
US-A- 4 869 977 **US-A1- 2005 221 168**
US-A1- 2007 166 609 **US-A1- 2007 178 370**

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Description

FIELD OF THE INVENTION

5 **[0001]** The embodiments of the present invention relate to lithium batteries, more specifically, to an electrolyte for lithium batteries and battery packs.

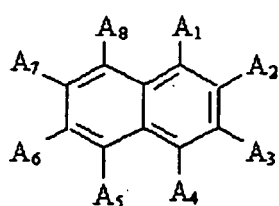
BACKGROUND

10 **[0002]** Because of its high voltage and high energy density, lithium secondary batteries are widely used in various mobile electronic devices and electric tools. However, with rapidly demanding performance of portable devices on the rise, lithium-ion batteries having higher capacity, longer life and higher energy density remains challenging. Furthermore, the increasing demand places additional requirement on safety, security and performance of these batteries.

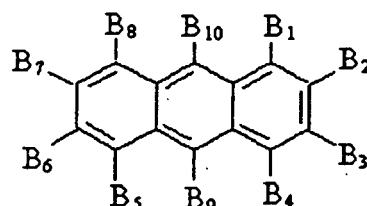
15 **[0003]** From WO 2008/109734 A, US 2007/166609 A1 and US 2007/178370 A1 different kinds of electrolytes for lithium-ion batteries are known. US 2005/221168 A1 in particular discloses electrolytes for lithium-ion batteries employing additives comprising aromatic fused ring compounds and fused heterocyclic compounds and alkoxy-substituted aromatic compounds in order to increase cycle life even under overcharge conditions.

20 **[0004]** Accordingly, one embodiment of the invention discloses an electrolyte for lithium-ion batteries according to claim 1. The electrolyte comprises: a lithium salt; a non-aqueous solvent; and an additive, the additive comprising substances A, B and C, wherein: substance A includes one or more fused ring compounds and fused heterocyclic compounds; substance B includes an alkoxy aromatic compound; and substance C includes halogenated borane-based salt.

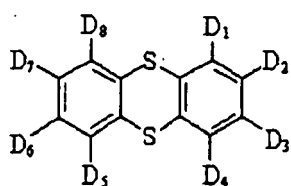
25 **[0005]** Moreover, the fused ring compounds of substance A include one or more members selected from chemical structures (1) and (2), and the fused heterocyclic compounds of substance A include one or more members selected from chemical structures (3), (4) and (5); and



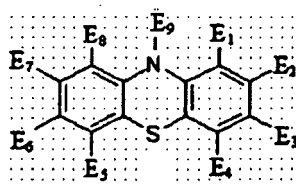
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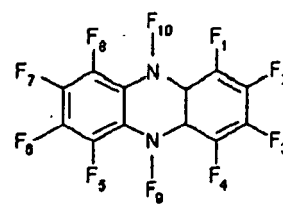
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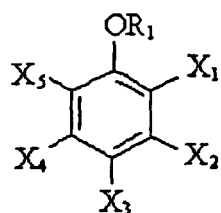
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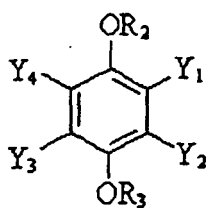
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50 wherein A₁-A₈, B₁-B₁₀, D₁-D₈, E₁-E₉ and F₁-F₁₀ can be selected from members consisting of hydrogen, halogens, alkyl groups having 1-20 carbon atoms and acyl groups having 1-15 carbon atoms wherein weight ratio of substances A:B:C can range from 1: 0.05:0.05 to 1:15:15. In one example, the weight ratio of the fused ring compounds to the fused heterocyclic compounds of substance A can range from about 1:5 to about 10:1.

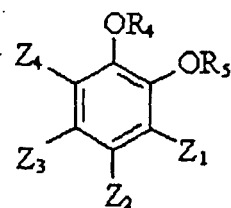
55 **[0006]** In another embodiment, the alkoxy aromatic compounds of substance B include one or more members selected from chemical structures (6), (7) and (8); and



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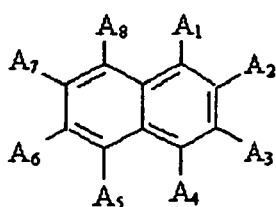


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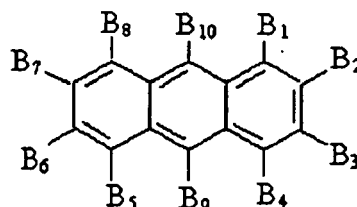
wherein R_1 - R_5 can be selected from members consisting of alkyl groups having 1-15 carbon atoms, and X_1 - X_5 , Y_1 - Y_4 and Z_1 - Z_4 can be selected from members consisting of hydrogen, halogens, and alkyl groups having 1-15 carbon atoms.

[0007] In yet another embodiment, the halogenated borane-based salt includes $Li_2B_{12}F_mH_{12-m}$, wherein $12 \geq m \geq 1$. In one example, the amount of additive can be about 1 to 20 % of the electrolyte by weight. In another example, the lithium salt includes one or more members of lithium hexafluorophosphate, lithium tetrafluoroborate, lithium hexafluoroarsenate, lithium perchlorate, lithium trifluoromethylsulfonate, lithium perfluorobutane sulfonate, lithium aluminate, lithium chloroaluminate, fluorinated lithium sulfonimide, lithium chloride and lithium iodide. In yet another example, the non-aqueous solvent include one or more members of gamma-butyrolactone, methyl ethyl carbonate, methyl propyl carbonate, dipropyl carbonate, anhydride, N-methyl pyrrolidone, N-dimethylformamide, N-methyl acetamide, acetonitrile, N,N-dimethylformamide, sulfolane, dimethyl sulfoxide, diethyl sulfite, and other unsaturated cyclic organic esters having fluorine and sulfur.

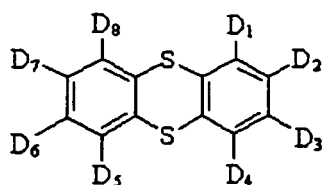
[0008] In another embodiment, an electrolyte for lithium-ion batteries, the electrolyte comprising: a lithium salt; a non-aqueous solvent; and an additive, the additive comprising two or more members selected from the group consisting of substances A, B and C, wherein: substance A includes one or more fused ring compounds and fused heterocyclic compounds, wherein the fused ring compounds include one or more members selected from chemical structures (1) and (2) and the fused heterocyclic compounds include one or more members selected from chemical structures (3), (4) and (5); and



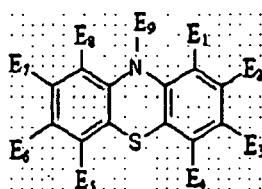
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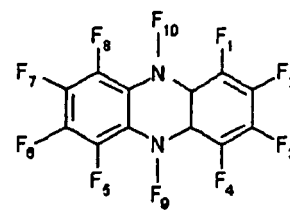
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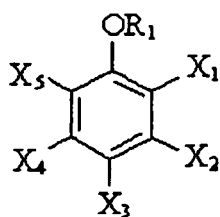


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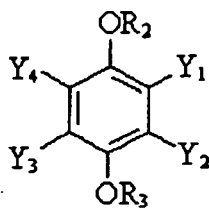


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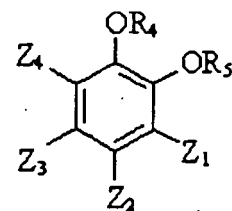
wherein A_1 - A_8 , B_1 - B_{10} , D_1 - D_8 , E_1 - E_9 and F_1 - F_{10} can be selected from members consisting of hydrogen, halogens, alkyl groups having 1-20 carbon atoms and acyl groups having 1-15 carbon atoms; substance B includes an alkoxy aromatic compound, wherein the alkoxy aromatic compound includes one or more members selected from chemical structures (6), (7) and (8); and



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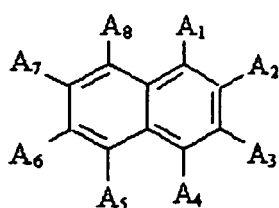


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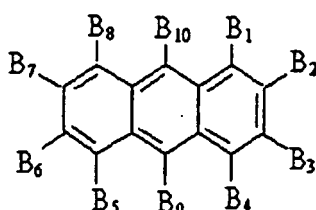
wherein R_1 - R_5 can be selected from members consisting of alkyl groups having 1-15 carbon atoms, and X_1 - X_5 , Y_1 - Y_4 and Z_1 - Z_4 can be selected from members consisting of hydrogen, halogens, and alkyl groups having 1-15 carbon atoms; and substance C includes halogenated borane-based salt, wherein the borane-based salt includes $Li_2B_{12}F_mH_{12-m}$, wherein $12 \geq m \geq 1$.

[0009] In one embodiment, the additive includes two members selected from the group consisting of substances A, B and C, wherein weight ratio of the two members can range from about 1:15 to 15:1. In another embodiment, the additive includes three members selected from each of substances A, B and C, wherein weight ratio of substances A: B:C can range from about 1:0.05:0.05 to about 1:15:15. In yet another embodiment, the weight ratio of the fused ring compounds to the fused heterocyclic compounds of substance A can range from about 1:5 to about 10:1. In one instance, the amount of additive can be about 1 to 20 % of the electrolyte by weight.

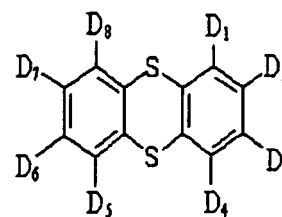
[0010] In another embodiment, an electrolyte for lithium-ion batteries, the electrolyte comprising: a lithium salt selected from one or more members consisting of lithium hexafluorophosphate, lithium tetrafluoroborate, lithium hexafluoroarsenate, lithium perchlorate, lithium trifluoromethylsulfonate, lithium perfluorobutane sulfonate, lithium aluminate, lithium chloroaluminate, fluorinated lithium sulfonimide, lithium chloride and lithium iodide; a non-aqueous solvent selected from one or more members consisting of gamma-butyrolactone, methyl ethyl carbonate, methyl propyl carbonate, dipropyl carbonate, anhydride, N-methyl pyrrolidone, N-dimethylformamide, N-methyl acetamide, acetonitrile, N,N-dimethylformamide, sulfolane, dimethyl sulfoxide, diethyl sulfite, and other unsaturated cyclic organic esters having fluorine and sulfur; and an additive being about 1 to 20 % of the electrolyte by weight, the additive comprising two or more members selected from the group consisting of substances A, B and C, wherein: substance A includes one or more fused ring compounds and fused heterocyclic compounds, wherein the fused ring compounds include one or more members selected from chemical structures (1) and (2) and the fused heterocyclic compounds include one or more members selected from chemical structures (3), (4) and (5); and



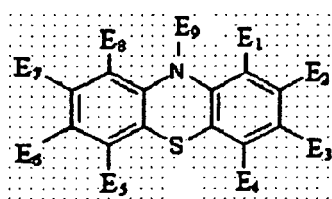
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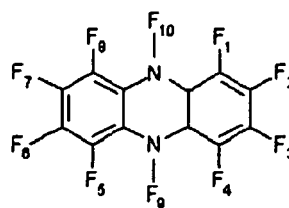
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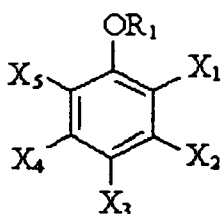


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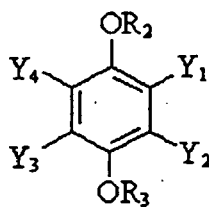


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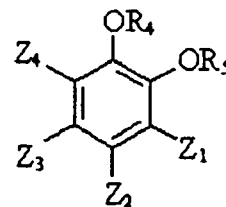
wherein A_1 - A_8 , B_1 - B_{10} , D_1 - D_8 , E_1 - E_9 and F_1 - F_{10} can be selected from members consisting of hydrogen, halogens, alkyl groups having 1-20 carbon atoms and acyl groups having 1-15 carbon atoms; substance B includes an alkoxy aromatic compound, wherein the alkoxy aromatic compound includes one or more members selected from chemical structures (6), (7) and (8); and



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wherein R_1 - R_5 can be selected from members consisting of alkyl groups having 1-15 carbon atoms, and X_1 - X_5 , Y_1 - Y_4 and Z_1 - Z_4 can be selected from members consisting of hydrogen, halogens, and alkyl groups having 1-15 carbon atoms; and substance C includes halogenated borane-based salt, wherein the borane-based salt includes $Li_2B_{12}F_mH_{12-m}$, wherein $12 \geq m \geq 1$.

[0011] In other embodiments, lithium-ion batteries and battery packs may be produced using the electrolyte as described in the previously disclosed embodiments.

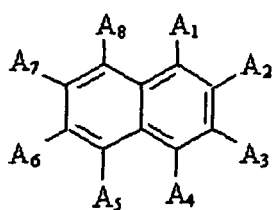
[0012] Other variations, embodiments and features of the present invention will become evident from the following detailed description, drawings and claims.

DETAILED DESCRIPTION

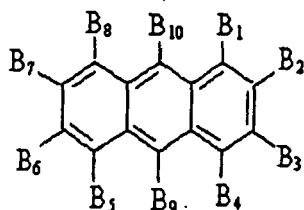
[0013] The presently disclosed embodiments are therefore considered in all respects to be illustrative and not restrictive.

[0014] A first embodiment discloses an electrolyte for lithium-ion batteries, the electrolyte consisting of a lithium salt, a non-aqueous solvent and an additive. In this embodiment, the additive includes two or more members selected from substances A, B and C, wherein substance A includes one or more fused ring compounds and fused heterocyclic compounds, substance B includes alkoxy aromatic compounds, and substance C includes halogenated borane-based salt.

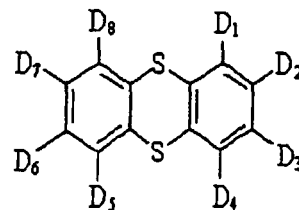
[0015] Substance A includes one or more fused ring compounds and fused heterocyclic compounds, the fused ring compounds being one or more members selected from chemical structures (1) and (2), and the fused heterocyclic compounds being one or more members selected from chemical structures (3), (4) and (5).



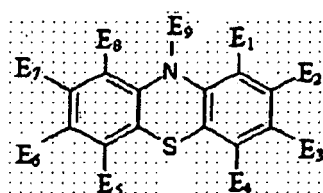
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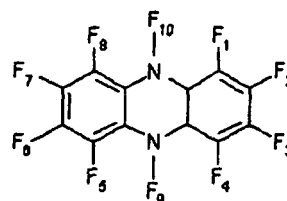
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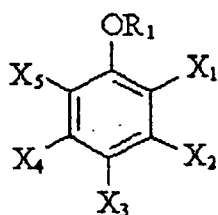


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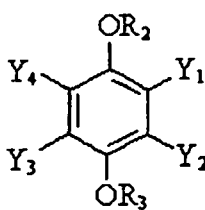
In these embodiments, A₁-A₈, B₁-B₁₀, D₁-D₈, E₁-E₉ and F₁-F₁₀ can be selected from members consisting of hydrogen, halogens (e.g., fluorine, chlorine and bromine), alkyl groups having 1-20 carbon atoms and acyl groups having 1-15 carbon atoms.

[0016] In one embodiment, substance A includes a mixture of one or more fused ring compounds (1) or (2), and one or more fused heterocyclic compounds (3), (4) or (5), wherein the ratio of the fused ring to heterocyclic compounds by weight can range from about 1:5 to about 10:1. In other embodiments, the ratio of fused ring compound to fused heterocyclic compound can vary from about 1:1 to about 5:1 by weight. The mixing of the fused ring compounds and the fused heterocyclic compounds increases the solubility of substance A and can facilitate dissolution of substance C, which has enhanced voltage limiting properties but lowered solubility within the electrolyte. The addition makes minimal alterations to the ion conductivity of the electrolyte allowing the battery to exhibit enhanced characteristics including low temperature operation, improved storage and cycle time, and voltage uniformity for single batteries and battery packs.

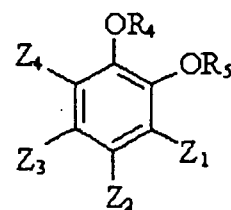
[0017] Substance B includes one or more alkoxy aromatic compounds selected from chemical structures (6), (7) and (8).



(6)



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[0018] In these embodiments, R₁-R₅ can be selected from members including alkyl groups having 1-15 carbon atoms, and X₁-X₅, Y₁-Y₄ and Z₁-Z₄ can be selected from members including hydrogen, halogens (e.g., fluorine, chlorine and bromine) and alkyl groups having 1-15 carbon atoms.

[0019] Substance C includes one or more halogenated borane-based salt including without limitation Li₂B₁₂F_mH_{12-m}, wherein 12 ≥ m ≥ 1.

[0020] In one instance, the amount of additive can be about 1 to 20 % of the electrolyte by weight. In other instances, the amount of additive can be about 1 to 10 %.

[0021] When the additive includes any two of substances A, B or C, the ratio between them can vary from about 1:15

to about 15:1 by weight. In other embodiments, the ratio can vary from about 1:6 to about 6:1 by weight. When the additive includes each member of substances A, B and C, the ratio of A:B:C by weight can vary from about 1:0.05:0.05 to about 1:15:15. In other embodiments, the ratio of A:B:C by weight can vary from about 1:0.3:0.3 to about 1:3:3.

[0022] The lithium salt for the electrolyte can be various lithium salts in the art including one or more members of lithium hexafluorophosphate, lithium tetrafluoroborate, lithium hexafluoroarsenate, lithium perchlorate, lithium trifluoromethylsulfonate, lithium perfluorobutane sulfonate, lithium aluminate, lithium chloroaluminate, fluorinated lithium sulfonimide, lithium chloride and lithium iodide. The concentration of the lithium salt in the electrolyte can be from about 0.3 to 4.0 mol/L. In other instances, the concentration of the lithium salt in the electrolyte can be from about 0.5 to 2.0 mol/L.

[0023] The non-aqueous solvent for the electrolyte can be various non-aqueous solvents in the art including one or more members of gamma-butyrolactone, methyl ethyl carbonate, methyl propyl carbonate, dipropyl carbonate, anhydride, N-methyl pyrrolidone, N-dimethylformamide, N-methyl acetamide, acetonitrile, N,N-dimethylformamide, sulfolane, dimethyl sulfoxide, diethyl sulfite, and other unsaturated cyclic organic esters having fluorine and sulfur.

[0024] The presently disclosed electrolyte can be prepared by methods of preparing electrolyte for lithium-ion batteries. For example, the lithium salt, organic solvent and additive can be mixed and prepared using the parameters described below.

[0025] A lithium secondary battery includes a battery core and an electrolyte, wherein the electrolyte includes the electrolyte embodiments disclosed above for lithium-ion batteries. There are no limitations to other parts and structures of the lithium-ion battery. In one instance, the battery core includes a cathode, anode and separator between the cathode and anode. The separator, situated between the cathode and anode, has the ability to insulate and maintain liquids within, and can be tightly sealed within the battery core together with the cathode and anode. The types of separator include modified polyethylene fiber, modified polypropylene fiber, ultra-thin glass fiber, vinyl fiber or nylon fiber with wetted polyolefin microporous membrane formed by welding or bonding of composite membrane.

[0026] In one example, the cathode includes a current collector and cathode material coated onto or filled within the current collector. The types of current collector include without limitation aluminum foil, copper foil or steel strip with nickel plating. The types of cathode material include without limitation LiFePO_4 , $\text{Li}_x\text{Ni}_{1-y}\text{CoO}_2$ ($0.9 \leq x \leq 1.1$ and $0 \leq y \leq 1.0$), $\text{Li}_m\text{Mn}_{2-n}\text{B}_n\text{O}_2$ ($0.9 \leq m \leq 1.1$, $0 \leq n \leq 1.0$, and B being transition metals), $\text{Li}_{10a}\text{M}_b\text{Mn}_{2-b}\text{O}_4$ (wherein $-0.1 \leq a \leq 0.2$, $0 \leq b \leq 1.0$, and M being one or more elements select from Li, B, Mg, Al, Ti, Cr, Co, Fe, Ni, Cu, Zn, Ga, Yt, F, I, S).

[0027] The cathode material further includes an adhesive, the adhesive being a mixture of hydrophilic and hydrophobic components. The ratio of hydrophilic component to hydrophobic component can be about 0.3 to 1 but need not be limited. The adhesive can come in aqueous, emulsion or solid forms. The concentration of the hydrophilic adhesive solution and the hydrophobic adhesive emulsion can be adjusted accordingly for viscosity and preparation of the anode and cathode slurry coating. In one example, the concentration of the hydrophilic adhesive solution can be about 0.5 to 4.0 % by weight while the concentration of the hydrophobic adhesive emulsion can be about 10 to 80 % by weight. The hydrophobic adhesive includes polytetrafluoroethylene (PTFE), styrene-butadiene rubber (SBR), or mixture of PTFE and SBR while the hydrophilic adhesive includes hydroxypropyl methylcellulose (HPMC), sodium carboxymethylcellulose, hydroxyethylcellulose, polyvinyl alcohol (PVA), or mixtures thereof. The amount of adhesives within the cathode material can be about 0.01 to 8 % by weight. In other embodiments, the amount of adhesives can be about 1 to 5 % by weight.

[0028] The cathode material as described above further includes a conductive agent for enhancing the conductivity of the electrode and reducing the internal resistance of the battery. In one example, the conductive agent can be about 0 to 15 % by weight of the cathode material. In other examples, the conductive agent can be about 0 to 10 % by weight. The conductive agent includes one or more members selected from the group consisting of conductive carbon black, acetylene black, nickel powder, copper powder and conductive graphite.

[0029] In one example, the anode includes a conductive base and an anode active material coated onto or filled within the conductive base. The types of conductive base include without limitation aluminum foil, copper foil, steel strip with nickel plating and steel strip with punched holes. The types of anode active material include without limitation natural graphite, artificial graphite, petroleum coke, organic cracked carbon, mesocarbon microspheres, carbon fiber, tin alloy and silicon alloy. The anode active material further includes an adhesive being selected from one or more members of polyvinyl alcohol, polytetrafluoroethylene (PTFE), hydroxymethyl cellulose (CMC) and styrene-butadiene rubber (SBR). In one instance, the amount of adhesive can be about 0.5 to 8 % by weight of the anode active material. In other instances, the amount of adhesive can be about 2 to 5 %. In one embodiment, lithium foil can function as the anode.

[0030] The solvent for preparing the cathode and anode slurry include one or more members of N-methyl pyrrolidone (NMP), dimethylformamide (DMF), diethylformamide (DEF), dimethyl sulfoxide (DMSO), tetrahydrofuran (THF), water, alcohol and mixtures thereof. The amount of solvent to be used can be enough for coating the slurry onto the base and collector. In other examples, the amount of solvent can be about 100 to 150 % by weight of the cathode or anode active materials.

[0031] The method of preparing the lithium secondary battery includes situating the separator between the cathode and anode to form an electrode group, which can be housed within the battery core. The presently disclosed embodiments of electrolyte can be injected within the battery core and sealed to form the lithium secondary battery. The method of

preparing the cathode includes mixing coating slurry of cathode active material, adhesive and conductive agent on the current collector, drying, pressing or rolling, and slicing to form the cathode. In one instance, the drying process can be at temperatures of about 50 to 160 °C. In other instances, the drying process can be at temperatures of about 80 to 150 °C. The method of preparing the anode can be similar in all respect to that of preparing the cathode with the exception that anode active material and adhesive are utilized instead. Presently disclosed embodiments further include a battery pack having a plurality of single batteries wherein each battery includes a lithium secondary battery incorporating the previously disclosed electrolyte embodiments. The single batteries can be connected in series or in parallel with various methods of coupling between the electrode terminals of the batteries in forming the serial or parallel connections. In one example, the single batteries can be coupled together by joining the electrode terminals of the batteries. In one instance, the electrode terminals can be soldered or welded together. After coupling the single batteries and forming a battery pack, the battery pack can be securely fastened and heat dissipation structures can be disposed between the single batteries. The single batteries can be further secured to minimize movement.

[0032] The following provides additional details of the electrolyte embodiments of the present invention.

Comparative EXAMPLE 1

[0033] At room temperature and within a glove box, ethylene carbonate, dimethyl carbonate and ethyl methyl carbonate are mixed in a ratio of 1 : 1 : 1. LiPF_6 can subsequently be added to the mixture to provide a solution with a concentration of 1 mol/L. An additive, in an amount of about 5 % by weight of the total final electrolyte product, can be added to the solution. The composition of the additive includes naphthalene, thianthrene and 3-chloro anisole (Sigma-Aldrich Co.) at a weight ratio of 2 : 1 : 1. The electrolyte can be labeled as A1.

COMPARATIVE EXAMPLE 2

[0034] Substantially similar in all respect to that of Example 1 except the additive can be about 2 % by weight of the total final electrolyte product and its composition includes thianthrene and 4-Br-1,2-dimethoxybenzene (Sigma-Aldrich Co.) at a weight ratio of 2 : 1. The electrolyte can be labeled as A2.

COMPARATIVE EXAMPLE 3

[0035] Substantially similar in all respect to that of Example 1 except the additive can be about 10 % by weight of the total final electrolyte product and its composition includes 3-chloro anisole and $\text{Li}_2\text{B}_{12}\text{FH}_{11}$ (Sigma-Aldrich Co.) at a weight ratio of 1 : 1. The electrolyte can be labeled as A3.

COMPARATIVE EXAMPLE 4

[0036] Substantially similar in all respect to that of Example 1 except the additive can be about 1 % by weight of the total final electrolyte product and its composition includes naphthalene, thianthrene and $\text{Li}_2\text{B}_{12}\text{F}_3\text{H}_9$ (Sigma-Aldrich Co.) at a weight ratio of 1 : 1 : 4. The electrolyte can be labeled as A4.

EXAMPLE 5

[0037] Substantially similar in all respect to that of Example 1 except the composition of the additive includes anthracene, N,N'-dimethyldihydrophenazine (Sigma-Aldrich Co.), 4-Cl-1,2-dimethoxybenzene (Sigma-Aldrich Co.) and $\text{Li}_2\text{B}_{12}\text{F}_2\text{H}_{10}$ (Sigma-Aldrich Co.) at a weight ratio of 1 : 0.2 : 3 : 1. The electrolyte can be labeled as A5.

COMPARATIVE EXAMPLE 6

[0038] Substantially similar in all respect to that of Example 1 except the additive can be about 2 % by weight of the total final electrolyte product and its composition includes thianthrene, 2,5-butyl-1,4-dimethoxybenzene (Sigma-Aldrich Co.) and $\text{Li}_2\text{B}_{12}\text{F}_{10}\text{H}_2$ (Sigma-Aldrich Co.) at a weight ratio of 2 : 1 : 1. The electrolyte can be labeled as A6.

COMPARATIVE EXAMPLE 7

[0039] Substantially similar in all respect to that of Example 1 except the additive can be about 5 % by weight of the total final electrolyte product and its composition includes 2,7-acetyl-thianthrene (Sigma-Aldrich Co.), 3-chloro anisole and $\text{Li}_2\text{B}_{12}\text{F}_6\text{H}_6$ (Sigma-Aldrich Co.) at a weight ratio of 1 : 1 : 1. The electrolyte can be labeled as A7.

REFERENCE 1

5 [0040] At room temperature and within a glove box, ethylene carbonate, dimethyl carbonate and ethyl methyl carbonate are mixed in a ratio of 1 : 1 : 1. LiPF₆ can subsequently be added to the mixture to provide a solution with a concentration of 1 mol/L. The electrolyte can be labeled as C1.

REFERENCE 2

10 [0041] At room temperature and within a glove box, ethylene carbonate, dimethyl carbonate and ethyl methyl carbonate are mixed in a ratio of 1 : 1 : 1. LiPF₆ can subsequently be added to the mixture to provide a solution with a concentration of 1 mol/L. An additive, in an amount of about 3 % by weight of the total final electrolyte product, can be added to the solution. The composition of the additive includes vinylene carbonate, biphenyl, cyclohexylbenzene, 1,3-propanesultone and vinylsulfurylbenzene at a weight ratio of 1 : 1 : 2 : 1 : 1. The electrolyte can be labeled as C2.

15 [0042] The following describes methods of preparing lithium batteries incorporating the electrolyte embodiments of the present invention.

PREPARATION OF THE CATHODE

20 [0043] Add 100 % by weight of LiFePO₄ (FMC Corp.), 3.5 % by weight of polyvinylidene fluoride adhesive (Atofina Chemical, Inc., 761 # PVDF), and 4.5 % by weight of acetylene black to 70 % by weight of NMP, mix and churn in vacuum mixer to form cathode slurry. Apply cathode slurry coating onto 20 micron thick aluminum foil, dry for 1 hour at a temperature of about 130 °C, and press to provide the cathode having a thickness of about 165 microns.

PREPARATION OF THE ANODE

25 [0044] Add 100 % by weight of black lead (Sodiff Advanced Materials Co., LTD, DAG84), 1 % by weight of carbon black, 3.5 % by weight of styrene-butadiene rubber (SBR) adhesive latex (Nantong Shenhua Chemical Industry Co., LTD., TAIPOLE 1500E), and 1 % by weight of carboxymethyl cellulose (Jinangmen Quantum High-Tech Biological Co., LTD, CMC1500) to 120 % by weight of deionized water, mix and churn in a vacuum mixer to form anode slurry. Apply
30 anode slurry coating onto 12 micron thick copper foil, dry for 1 hour at a temperature of about 140 °C, and press to provide the anode having a thickness of about 105 microns

PREPARATION OF THE BATTERIES

35 [0045] Take the cathode and anode from above, together with separator of about 20 micron thick, are wound into thick a square-shaped lithium-ion battery core. The electrolyte (about 3.8 g) from each of Examples 1-7 are subsequently injected into the battery core and air-sealed to provide seven 053450A type lithium-ion batteries labeled B1-B7. Likewise, the electrolyte from each of References 1-2 are subsequently injected into the battery core and air-sealed to provide
40 two 053450A type lithium-ion batteries labeled D1-D2.

[0046] In other embodiments, three batteries from each of B1-B7 can be connected in series to provide battery packs labeled E1-E7. For example, three B1 batteries can be coupled in series to provide battery pack E1, three B2 batteries can be coupled in series to provide battery pack E2, three B3 batteries can be coupled in series to provide battery pack E3, and so forth. Likewise, three batteries from each of D1-D2 can be connected in series to provide battery packs F1 and F2, respectively.
45

BATTERY TESTING

1. Overcharging performance testing

50 [0047] At room temperature, batteries B1-B7 and D1-D2 are overcharged to 0.075 A and 4.6 V. The physical appearance and duration of the battery's ability to maintain the overcharge are shown in Table 1.

2. Capacity retention performance testing

55 [0048] At room temperature, batteries B1-B7 and D1-D2 are charged at 0.75 A constant current to 3.8 V, at constant voltage to cut-off current 0.01 A, and discharged at 0.75 A constant current to 2.0 V to produce an initial capacity. The thickness of the batteries are also noted. The charging /discharging cycle can be repeated 500 times to provide the battery's capacity after 500 cycles. Each battery's capacity retention rate after 500 cycles and thickness changes are

shown in Table 1. The capacity retention rate can be calculated using the following equation:

$$\text{Capacity retention rate} = (\text{capacity after 500 cycles} / \text{initial capacity}) \times 100 \%$$

Table 1. Overcharging and Cycling Performances of Batteries B1-B7 and D1-D2.

| No. | Overcharge Time (hour) | State | Capacity Retention Rate after 500 Cycles (%) | Change in Thickness (mm) |
|-----|------------------------|--------------------------------|--|--------------------------|
| B1 | 2.5 | Inflated | 98.5 | 0.02 |
| B2 | 2.5 | Inflated | 98.1 | 0.03 |
| B3 | 2.5 | Inflated | 98.4 | 0.03 |
| B4 | 2.5 | Inflated | 98.7 | 0.02 |
| B5 | 2.5 | Inflated | 98.7 | 0.02 |
| B6 | 2.5 | Inflated | 99.2 | 0.01 |
| B7 | 2.5 | Inflated | 99.6 | 0.01 |
| D1 | 0.5 | Exploded | 95.6 | 0.1 |
| D2 | 1.5 | Anti-Exploding Valve Activated | 92.1 | 0.7 |

3. Low temperature discharge performance testing

[0049] At room temperature, batteries B1-B7 and D1-D2 are charged at 0.75 A constant current to 3.8 V, at constant voltage to cut-off current 0.01 A, and discharged at 0.75 A constant current to 2.0 V to produce an initial capacity. The batteries are again charged at 0.75 A constant current to 3.8 V, and then at constant voltage to cut-off current 0.01 A. The batteries are maintained at -10 °C and -20 °C for 120 minutes, removed, and discharged again at 0.75 A constant current to 2.0 V. Each battery's capacity ratio at different temperatures and ending resistance value are shown in Table 2. The capacity ratio can be calculated using the following equation:

$$\text{Capacity ratio} = (\text{discharge capacity} / \text{initial capacity}) \times 100\%$$

Table 2. Low Temperature Discharge Performance of Batteries B1-B7 and D1-D2.

| No. | -10 °C | | -20 °C | |
|-----|-----------------------------------|------------------------|-----------------------------------|-----------------------|
| | -10 °C / 25 °C Capacity Ratio (%) | Ending resistance (mΩ) | -20 °C / 25 °C Capacity Ratio (%) | Final resistance (mΩ) |
| B1 | 52.9 | 45.0 | 42.5 | 54.9 |
| B2 | 52.1 | 45.8 | 41.9 | 55.7 |
| B3 | 52.4 | 45.2 | 41.8 | 55.7 |
| B4 | 52.7 | 44.9 | 42.1 | 54.5 |
| B5 | 53.0 | 44.7 | 44.3 | 54.0 |
| B6 | 53.5 | 44.2 | 46.2 | 53.8 |

(continued)

| No. | -10 °C | | -20 °C | |
|-----|-----------------------------------|------------------------|-----------------------------------|-----------------------|
| | -10 °C / 25 °C Capacity Ratio (%) | Ending resistance (mΩ) | -20 °C / 25 °C Capacity Ratio (%) | Final resistance (mΩ) |
| B7 | 54.3 | 43.8 | 46.6 | 53.1 |
| D1 | 52.2 | 46.1 | 40.5 | 55.9 |
| D2 | 41.7 | 58.3 | 29.1 | 70.1 |

4. High temperature storage performance testing

[0050] At room temperature, batteries B1-B7 and D1-D2 are charged at 0.75 A constant current to 3.8 V, at constant voltage to cut-off current 0.01 A, and discharged at 0.75 A constant current to 2.0 V to produce an initial capacity. The thickness, resistance and voltage of the batteries are also noted. The batteries are again charged at 0.75 A constant current to 3.8 V, maintained at 85 °C for 48 hours, and discharged again at 0.75 A constant current to 2.0 V. The thickness, resistance and voltage of the batteries are again noted. Results are shown in Table 3.

Table 3. High Temperature Storage Performance of Batteries B1-B7 and D1-D2.

| No. | Capacity Retention (%) | Capacity Recovery (%) | Changes in Thickness (mm) | Changes in Resistance (%) | Voltage Drop □V□ |
|-----|------------------------|-----------------------|---------------------------|---------------------------|------------------|
| B1 | 94.8 | 97.5 | 0.15 | 11.9 | 0.048 |
| B2 | 94.4 | 97.2 | 0.16 | 12.5 | 0.057 |
| B3 | 94.2 | 97.3 | 0.15 | 12.8 | 0.051 |
| B4 | 95.0 | 97.5 | 0.15 | 12.0 | 0.047 |
| B5 | 95.4 | 98.2 | 0.15 | 11.7 | 0.043 |
| B6 | 96.7 | 98.9 | 0.14 | 11.4 | 0.041 |
| B7 | 97.0 | 98.8 | 0.13 | 11.1 | 0.041 |
| D1 | 94.1 | 96.4 | 0.20 | 12.7 | 0.053 |
| D2 | 90.1 | 92.3 | 0.96 | 50.3 | 0.133 |

5. Battery pack capacity retention performance testing

[0051] At room temperature, battery packs E1-E7 and F1-F2 are charged at 0.75 A constant current to 11.4 V, at constant voltage to cut-off current 0.01 A, and discharged at 0.75 A constant current to 6.0 V to produce an initial capacity for the entire battery pack. The initial capacity for each single battery within the battery pack can also be acquired from the cathode and anode ends. The charging / discharging cycle can be repeated 300 times to provide the battery pack's capacity after 300 cycles. The battery packs' capacity retention rates after 300 cycles are shown in Table 4. The capacity retention rate can be calculated using the following equation:

$$\text{Capacity retention rate} = (\text{capacity after 300 cycles} / \text{initial capacity}) \times 100 \%$$

Table 4. Capacity Retention Performance of Battery Packs E1-E7 and F1-F2.

| No. | Capacity Retention (%) | Charging Voltage | | | | Discharging Voltage | | | |
|-----|------------------------|------------------|-------------------------|------------------|-------------------------|---------------------|-------------------------|------------------|-------------------------|
| | | Initial Cycle | | 300 Cycles | | Initial Cycle | | 300 Cycles | |
| | | Pack Voltage (V) | Single Voltage (V) | Pack Voltage (V) | Single Voltage (V) | Pack Voltage (V) | Single Voltage (V) | Pack Voltage (V) | Single Voltage (V) |
| E1 | 97.3 | 11.410 | 3.807 3.797 3.806 | 11.410 | 3.803 3.803 3.804 | 6.010 | 2.012 2.003 1.995 | 6.003 | 2.005 2.007 1.991 |
| E2 | 96.6 | 11.410 | 3.807 3.802 3.801 | 11.414 | 3.806 3.800 3.808 | 6.007 | 1.993 2.006 2.008 | 6.010 | 1.998 2.012 2.000 |
| E3 | 96.5 | 11.413 | 3.800 3.809 3.804 | 11.415 | 3.806 3.810 3.799 | 6.001 | 2.000 2.003 1.998 | 6.009 | 2.005 2.005 1.999 |
| E4 | 97.5 | 11.401 | 3.799 3.801 3.801 | 11.411 | 3.801 3.807 3.803 | 6.003 | 2.006 1.995 2.002 | 5.998 | 2.006 1.997 1.995 |
| E5 | 97.8 | 11.409 | 3.806 3.805 3.798 | 11.410 | 3.805 3.799 3.806 | 6.000 | 1.998 2.001 2.001 | 6.006 | 1.998 2.003 2.005 |
| E6 | 98.0 | 11.408 | 3.802 3.799 3.807 | 11.411 | 3.806 3.801 3.804 | 6.006 | 1.996 2.010 2.000 | 6.008 | 1.992 2.110 2.106 |
| E7 | 98.3 | 11.407 | 3.807 3.797 3.803 | 11.409 | 3.804 3.799 3.805 | 6.005 | 2.005 2.002 1.998 | 6.000 | 2.007 1.998 1.995 |
| F1 | 94.3 | 11.427 | 3.814 3.805 3.808 | 12.251 | 4.077 4.077 4.097 | 6.028 | 1.992 2.021 2.015 | 5.991 | 0.280 2.855 2.856 |
| F2 | 85.7 | 11.425 | 3.810 3.803 3.812 | 12.011 | 3.622 4.145 4.244 | 5.989 | 2.006 1.981 2.002 | 6.012 | 2.180 1.309 2.523 |

[0052] Based on the results of Table 1, batteries containing the electrolyte embodiment of the present invention did not explode when overcharged, and exhibited excellent cycling (charging / discharging) performance. And based on the results of Tables 2 and 3, the batteries exhibited enhanced low temperature discharge capacities and high temperature storage properties. Last but not least, based on the results of Table 4, battery packs containing single batteries incorporating the electrolyte embodiment of the present invention exhibited minimal voltage changes or differences.

[0053] Lithium secondary batteries containing the non-aqueous electrolyte embodiment of the present invention are capable of delivering enhanced overcharging and cycling (charging / discharging) characteristics, improved low temperature discharge and high temperature storage properties, and added safety performance when provided in battery packs.

[0054] Although the invention has been described in detail with reference to several embodiments, additional variations and modifications exist within the scope of the invention as described and defined in the following claims.

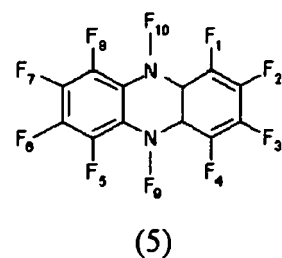
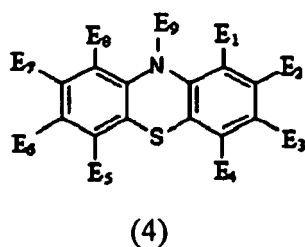
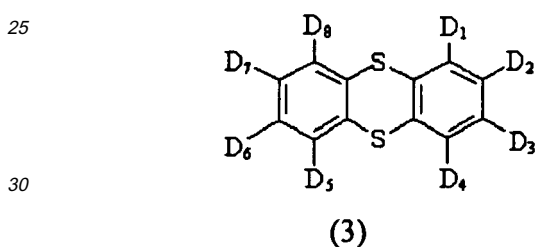
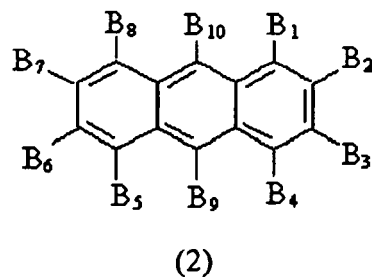
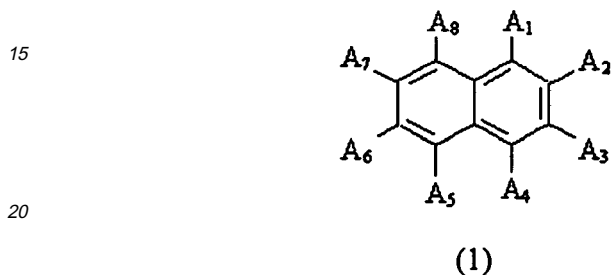
Claims

1. An electrolyte for lithium-ion batteries, the electrolyte comprising:

a lithium salt;
 a non-aqueous solvent; and
 an additive, the additive comprising substances A, B and C, wherein:

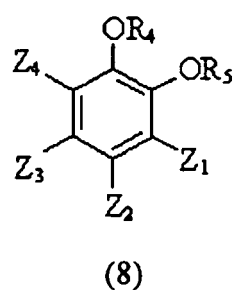
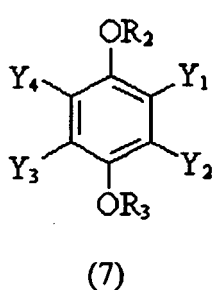
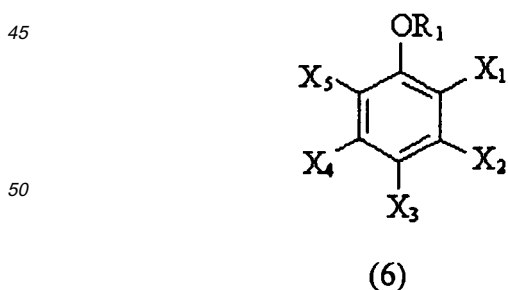
- 5 substance A includes one or more fused ring compounds and fused heterocyclic compounds;
 substance B includes an alkoxy aromatic compound; and
 substance C includes halogenated borane-based salt,

10 wherein the fused ring compounds of substance A include one or more members selected from chemical structures (1) and (2), and the fused heterocyclic compounds of substance A include one or more members selected from chemical structures (3), (4) and (5); and



wherein A₁-A₈, B₁-B₁₀, D₁-D₈, E₁-E₉ and F₁-F₁₀ can be selected from members consisting of hydrogen, halogens, alkyl groups having 1-20 carbon atoms and acyl groups having 1-15 carbon atoms, wherein weight ratio of substances A : B : C can range from 1 : 0.05 : 0.05 to 1 : 15 : 15.

- 40 2. The electrolyte of any one of claim 1, wherein weight ratio of the fused ring compounds to the fused heterocyclic compounds of substance A can range from 1 : 5 to 10 : 1.
- 45 3. The electrolyte of any one of claim 1 or 2, wherein the alkoxy aromatic compounds of substance B include one or more members selected from chemical structures (6), (7) and (8); and



wherein R₁-R₅ can be selected from members consisting of alkyl groups having 1-15 carbon atoms, and X₁-X₅, Y₁-Y₄ and Z₁-Z₄ can be selected from members consisting of hydrogen, halogens, and alkyl groups having 1-15 carbon atoms.

4. The electrolyte of any one of claims 1 to 3, wherein the halogenated borane-based salt includes $\text{Li}_2\text{B}_{12}\text{F}_m\text{H}_{12-m}$, wherein $12 \geq m \geq 1$.
5. The electrolyte of any one of claims 1 to 4, wherein the amount of additive can be 1 to 20 % of the electrolyte by weight.
6. The electrolyte of any one of claims 1 to 5, wherein the lithium salt includes one or more members of lithium hexafluorophosphate, lithium tetrafluoroborate, lithium hexafluoroarsenate, lithium perchlorate, lithium trifluoromethylsulfonate, lithium perfluorobutane sulfonate, lithium aluminate, lithium chloroaluminate, fluorinated lithium sulfonimide, lithium chloride and lithium iodide.
7. The electrolyte of any one of claims 1 to 6, wherein the non-aqueous solvent include one or more members of gamma-butyrolactone, methyl ethyl carbonate, methyl propyl carbonate, dipropyl carbonate, anhydride, N-methyl pyrrolidone, N-dimethylformamide, N-methyl acetamide, acetonitrile, N,N-dimethylformamide, sulfolane, dimethyl sulfoxide, diethyl sulfite, and other unsaturated cyclic organic esters having fluorine and sulfur.
8. A lithium-ion battery comprising a battery core and electrolyte, the electrolyte comprising the electrolyte of any one of claims 1 to 7.
9. A battery pack comprising a plurality of single lithium-ion batteries coupled in series or parallel, each lithium-ion battery comprising the electrolyte of any one of claims 1 to 7.

Patentansprüche

1. Elektrolyt für Lithiumionenbatterien, wobei der Elektrolyt umfasst:

ein Lithiumsalz;

ein nicht wässriges Lösungsmittel; und

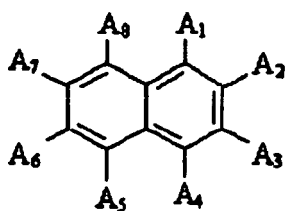
ein Additiv, wobei das Additiv die Substanzen A, B und C umfasst, wobei:

die Substanz A eine oder mehrere fusionierte Ringverbindungen und fusionierte heterocyclische Verbindungen umfasst;

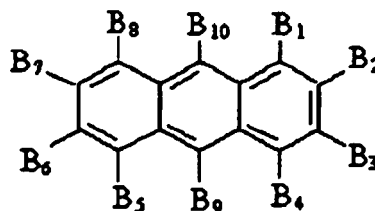
die Substanz B eine aromatische Alkoxyverbindung umfasst; und

die Substanz C ein halogeniertes auf Boran basierendes Salz umfasst,

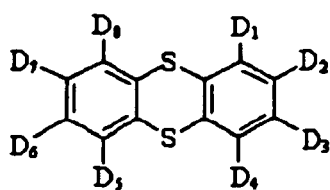
wobei die fusionierten Ringverbindungen der Substanz A ein oder mehrere Mitglieder umfassen, die aus den chemischen Strukturen (1) und (2) ausgewählt sind, und die fusionierten heterocyclischen Verbindungen der Substanz A ein oder mehrere Mitglieder umfassen, die aus den chemischen Strukturen (3), (4) und (5) ausgewählt sind; und



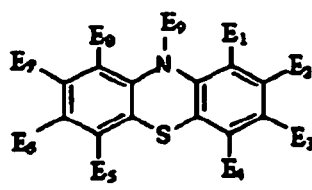
(1)



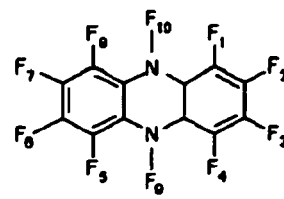
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(3)



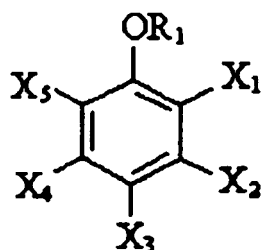
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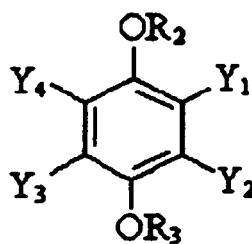
(5)

wobei A_1 - A_8 , B_1 - B_{10} , D_1 - D_8 , E_1 - E_9 und F_1 - F_{10} aus Mitgliedern ausgewählt sein können, die aus Wasserstoff, Halogenen, Alkylgruppen mit 1-20 Kohlenstoffatomen und Acylgruppen mit 1-15 Kohlenstoffatomen bestehen, wobei das Gewichtsverhältnis der Substanzen A : B : C im Bereich von 1 : 0,05 : 0,05 bis 1 : 15 : 15 liegen kann.

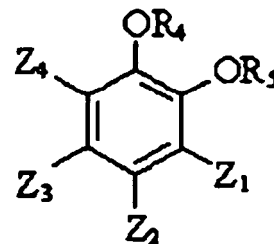
2. Elektrolyt nach Anspruch 1, wobei das Gewichtsverhältnis der fusionierten Ringverbindungen zu den fusionierten heterocyclischen Verbindungen der Substanz A im Bereich von 1 : 5 bis 10 : 1 liegen kann.
3. Elektrolyt nach einem von Anspruch 1 oder 2, wobei die aromatischen Alkoxyverbindungen der Substanz B ein oder mehrere Mitglieder umfassen, die aus den chemischen Strukturen (6), (7) und (8) ausgewählt sind; und



(6)



(7)



(8)

wobei R_1 - R_5 aus Mitgliedern ausgewählt sein können, die aus Alkylgruppen mit 1-15 Kohlenstoffatomen bestehen, und X_1 - X_5 , Y_1 - Y_4 und Z_1 - Z_4 aus Mitgliedern ausgewählt sein können, die aus Wasserstoff, Halogenen und Alkylgruppen mit 1-15 Kohlenstoffatomen bestehen.

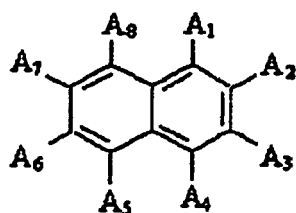
4. Elektrolyt nach einem der Ansprüche 1 bis 3, wobei das halogenierte auf Boran basierende Salz $Li_2B_{12}F_mH_{12-m}$ umfasst, wobei $12 \geq m \geq 1$.
5. Elektrolyt nach einem der Ansprüche 1 bis 4, wobei die Menge des Additivs 1 bis 20 % des Elektrolyten auf das Gewicht bezogen sein kann.
6. Elektrolyt nach einem der Ansprüche 1 bis 5, wobei das Lithiumsalz ein oder mehrere Mitglieder von Lithiumhexafluorophosphat, Lithiumtetrafluorborat, Lithiumhexafluorarsenat, Lithiumperchlorat, Lithiumtrifluormethylsulfonat, Lithiumperfluorbutansulfonat, Lithiumaluminat, Lithiumchloraluminat, fluoriertes Lithiumsulfonimid, Lithiumchlorid und Lithiumjodid umfasst.
7. Elektrolyt nach einem der Ansprüche 1 bis 6, wobei das nicht wässrige Lösungsmittel ein oder mehrere Mitglieder von Gammabutyrolacton, Methylethylcarbonat, Methylpropylcarbonat, Dipropylcarbonat, Anhydrid, N-Methylpyrrolidon, N-Dimethylformamid, N-Methylacetamid, Acetonitril, N,N-Dimethylformamid, Sulfolan, Dimethylsulfoxid, Diethylsulfid und andere ungesättigte cyclische organische Ester mit Fluor und Schwefel umfasst.
8. Lithiumionenbatterie mit einem Batteriekern und einem Elektrolyten, wobei der Elektrolyt den Elektrolyten nach einem der Ansprüche 1 bis 7 umfasst.

9. Batteriestapel mit mehreren einzelnen Lithiumionenbatterien, die in Reihe oder parallel gekoppelt sind, wobei jede Lithiumionenbatterie den Elektrolyten nach einem der Ansprüche 1 bis 7 umfasst.

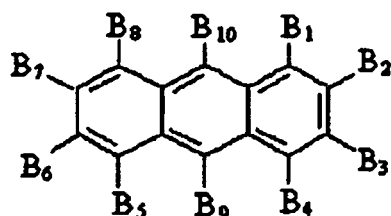
5 **Revendications**

1. Electrolyte pour batteries au lithium-ion, lequel électrolyte comprend:

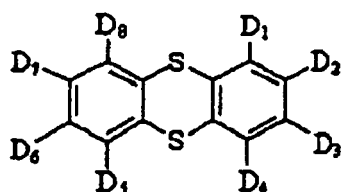
un sel de lithium;
 un solvant non aqueux; et
 un additif, l'additif comprenant des substances A, B et C, où
 la substance A comprend un ou plusieurs composés cycliques condensés et composés hétérocycliques condensés;
 la substance B comprend un composé alcoxy aromatique; et
 la substance C comprend un sel à base de borane halogéné,
 dans lequel les composés cycliques condensés de la substance A comprennent un ou plusieurs membres choisis parmi les structures chimiques (1) et (2), et les composés hétérocycliques condensés de la substance A comprennent un ou plusieurs membres choisis parmi les structures chimiques (3), (4) et (5):



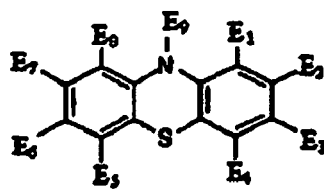
(1)



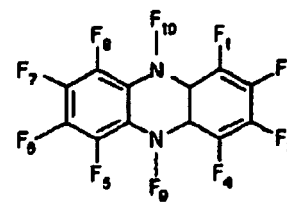
(2)



(3)



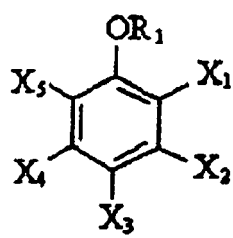
(4)



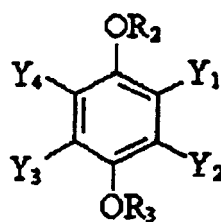
(5)

où A₁ à A₈, B₁ à B₁₀, D₁ à D₈, E₁ à E₉ et F₁ à F₁₀ peuvent être choisis parmi les membres constitués par l'hydrogène, les halogènes, les groupes alkyle ayant de 1 à 20 atomes de carbone et les groupes acyle ayant de 1 à 15 atomes de carbone,
 dans lequel le rapport en poids des substances A : B : C peut être situé dans la plage allant de 1 : 0,05 : 0,05 à 1 : 15 : 15.

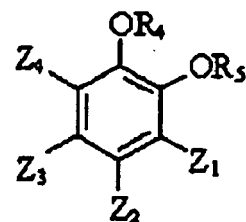
2. Electrolyte selon la revendication 1, dans lequel le rapport en poids des composés cycliques condensés aux composés hétérocycliques condensés de la substance A peut être situé dans la plage allant de 1 : 5 à 10 : .1.
3. Electrolyte selon l'une quelconque des revendications 1 et 2, dans lequel les composés alcoxy aromatiques de la substance B comprennent un ou plusieurs membres choisis parmi les structures chimiques (6), (7) et (8):



(6)



(7)



(8)

où R_1 à R_5 peuvent être choisis parmi les membres constitués par les groupes alkyle ayant de 1 à 15 atomes de carbone, et X_1 à X_5 , Y_1 à Y_4 et Z_1 à Z_4 peuvent être choisis parmi les membres constitués par l'hydrogène, les halogènes, et les groupes alkyle ayant de 1 à 15 atomes de carbone.

4. Electrolyte selon l'une quelconque des revendications 1 à 3, dans lequel le sel à base de borane halogéné comprend $\text{Li}_2\text{B}_{12}\text{F}_m\text{H}_{12-m}$ où $12 \geq m \geq 1$.
5. Electrolyte selon l'une quelconque des revendications 1 à 4, dans lequel la quantité d'additif peut être de 1 à 20 % en poids de l'électrolyte.
6. Electrolyte selon l'une quelconque des revendications 1 à 5, dans lequel le sel de lithium comprend un ou plusieurs membres choisis parmi l'hexafluorophosphate de lithium, le tétrafluoroborate de lithium, l'hexafluoroarsénate de lithium, le perchlorate de lithium, le trifluorométhylsulfonate de lithium, le perfluorobutanesulfonate de lithium, l'aluminate de lithium, le chloroaluminate de lithium, le lithium-sulfonimide fluoré, le chlorure de lithium et l'iodure de lithium.
7. Electrolyte selon l'une quelconque des revendications 1 à 6, dans lequel le solvant non aqueux comprend un ou plusieurs membres choisis parmi la γ -butyrolactone, le carbonate de méthyléthyle, le carbonate de méthylpropyle, le carbonate de dipropyle, un anhydride, la N-méthylpyrrolidone, le N-diméthylformamide, le N-méthylacétamide, l'acétonitrile, le N,N-diméthylformamide, le sulfolane, le diméthylsulfoxyde, le sulfite de diéthyle, et d'autres esters organiques cycliques insaturés contenant du fluor et du soufre.
8. Batterie au lithium-ion comprenant un coeur de batterie et un électrolyte, l'électrolyte comprenant l'électrolyte de l'une quelconque des revendications 1 à 7.
9. Bloc de batteries comprenant une pluralité de batteries au lithium-ion unitaires couplées en série ou en parallèle, chaque batterie au lithium-ion comprenant l'électrolyte de l'une quelconque des revendications 1 à 7.

REFERENCES CITED IN THE DESCRIPTION

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