

IN-SITU POLYMERIZED SOLID ELECTROLYTE, PREPARATION METHOD AND USE THEREOF

TECHNICAL FIELD

[0001] The present application relates to the field of lithium battery materials, particularly to an in-situ polymerized solid electrolyte, a preparation method and use thereof.

BACKGROUND ART

[0002] Secondary lithium batteries have played an important role in various fields of electronic and electrical appliances. Building energy storage facilities composed of large-scale lithium batteries can improve the efficient storage and reuse of energy. Electrolytes currently used in lithium batteries include liquid electrolytes, gel electrolytes (i.e. quasi-solid electrolytes), and solid electrolytes.

[0003] Most of the solvents of liquid electrolytes are carbonate esters (ethylene carbonate, diethyl carbonate), ethers (ethylene glycol dimethyl ether, tetrahydrofuran), carboxylate esters (methyl acetate), etc. The above substances used in liquid electrolytes are easy to leak, volatilize, decompose, and deflagrate, which makes lithium batteries easy to burn in the storage process. Therefore, flammable liquid electrolytes with poor thermal stability bring serious security risks for their large-scale storage.

[0004] In order to solve the potential safety hazard of large-scale storage of lithium batteries, the use of gel electrolytes (i.e. quasi-solid electrolyte) or solid electrolytes is one of the directions to improve the thermal stability of electrolyte and battery safety.

[0005] Among solid electrolytes, in-situ polymerized solid electrolytes have advantages of good electrode interface compatibility and high thermal stability. Polymer electrolyte derived from a cyclic ether, an olefin-based carbonate, and other polymerization monomers has high

ionic conductivity, a simple preparation method, and low cost, which has been widely concerned by researchers. However, the polymer electrolyte is poor in thermal stability and high in flammability, which makes it difficult to fundamentally improve the thermal safety performance of lithium batteries.

[0006] Ionic liquids possess nonflammability, good chemical and electrochemical stability, excellent thermal stability, and good ion conductivity at room temperature. In addition, ionic liquids exhibit good compatibility with lithium metal anodes. Solid polymer electrolytes (SPE) based on polyionic liquids are one of the most important choices for solid electrolytes with intrinsic safety characteristics.

SUMMARY

[0007] The purpose of the present application is to provide an in-situ polymerized solid electrolyte and a preparation method and use thereof in view of deficiencies of the current technology. The in-situ polymerized solid electrolyte of the present application has good thermal stability and flame retardancy, a high ionic conductivity at room temperature, and can inhibit the growth of lithium dendrites and achieve a stable interface between the electrolyte and the electrode. The all-solid-state lithium battery assembled by matching the in-situ polymerized solid electrolyte of the present application with a lithium iron phosphate cathode has good cycling stability and rate performance. The introduction of vinyl ethylene carbonate can promote the segmental motion of the polymer, increase the flexibility of the polymer, and improve the ion transport performance of the polymer solid electrolyte at room temperature. The in-situ polymerization strategy can ensure good interface contact characteristics of the polymerized solid electrolyte on the electrode surface, reduce the ion transport resistance, and improve the ion transport ability at the electrode/polymerized solid electrolyte interface. A solid polymer electrolyte based on polymer ionic liquid is prepared by simple in-situ copolymerization of ionic liquid monomers and vinyl ethylene carbonate monomers. The introduction of vinyl ethylene

carbonate units with high conductivity can decrease the crystallinity of polymer, inhibit the crystallization of polymer, promote the dissociation of lithium salt, and improve the ionic conductivity of polymer electrolyte membrane. The in-situ polymerized solid electrolyte to be prepared has good flexibility, thermal stability, flame retardancy, a wide electrochemical window, and the ability to inhibit the growth of lithium dendrites.

[0008] In a first aspect, the present application provides an in-situ polymerized solid electrolyte, which adopts the following technical solution.

[0009] An in-situ polymerized solid electrolyte including the following raw materials for preparation in parts by mass: 35-40 parts of an ionic liquid, 35-40 parts of vinyl ethylene carbonate, 25-28 parts of a lithium salt, and 0.05-0.1 parts of a photoinitiator.

[0010] By adopting the above technical solution, the ionic liquid, as a major component of the electrolyte, provides a high ionic conductivity; endows the electrolyte with non-flammability, chemical and electrochemical stability; is compatible with the lithium metal negative electrode to help form a stable electrode/electrolyte interface, and provides good thermal stability, helping to maintain the performance of the battery at high temperatures. The vinyl ethylene carbonate facilitates the segmental motion of the polymer, and increases the flexibility of the polymer; reduces the crystallinity of the polymer and inhibits the crystallization of the polymer, helping to improve the ion transport properties of the electrolyte at room-temperature; and together with the ionic liquid, improves the overall performance of the electrolyte. The lithium salt provides freely movable lithium ions, which is a key component in achieving ion conduction; interacts with a polymer matrix, helping to the dissociation of the lithium salt and further improving ionic conductivity. The photoinitiator initiates an in-situ polymerization reaction to convert monomers to a polymer; and ensures to form a uniform and dense electrolyte layer on the electrode surface during the polymerization process, thereby reducing the interfacial resistance. The ionic liquid and vinyl ethylene carbonate form a homogeneous polymer network by in-situ copolymerization, which facilitates the transport of ions while maintaining the mechanical stability and flexibility

of the electrolyte. The highly conductive vinyl ethylene carbonate units in combination with the ionic liquid optimize the ionic conductivity and electrochemical stability of the electrolyte. The uniform distribution and efficient dissociation of the lithium salt in the polymer matrix can provide sufficient lithium ions to ensure high ionic conductivity of the electrolyte. The photoinitiator enables the in-situ polymerization reaction to occur on the electrode surface, forming good interfacial contact characteristics, and reducing the ion transport resistance between the electrode and the electrolyte. In summary, the in-situ polymerized solid electrolyte of the present application achieves high ionic conductivity, good thermal stability and flame retardancy, excellent electrochemical stability, and the ability to inhibit the growth of lithium dendrites through the synergistic effect of the components, thereby improving the cycling stability and rate performance of the all-solid-state lithium battery.

[0011] Preferably, the ionic liquid is one selected from a group consisting of: 1-vinyl-3-methylimidazolium bis(fluorosulfonyl)imide, 1-vinyl-3-ethylimidazolium bis(trifluoromethylsulfonyl)imide, and 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide.

[0012] Preferably, the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide is prepared by a method including the following steps:

[0013] S31, adding 10 g of 3-chloropropylamine hydrochloride, 7.12 g of N-methylimidazole and 10 mL of tetrahydrofuran to a reactor, and stirring at 60°C for 48 h for reaction to obtain a product, after the reaction, dissolving the product in 20 mL of deionized water, and washing 3 times with 15 mL of tetrahydrofuran to obtain a 1-aminopropyl-3-methylammonium chloride solution;

[0014] S32, dissolving 10 g of lithium bis (trifluoromethane) sulfonamide in 20 mL of deionized water, then adding the 1-aminopropyl-3-methylammonium chloride solution dropwise, and stirring for 24 h for reaction, after the reaction, washing resulting mixture 3 times with

deionized water, and performing vacuum distillation at 60°C under 1000 Pa for 12 h to obtain a product A; and

[0015] S33, dissolving 10 g of the product A in 25 mL of ethanol, adding 75 mL of an ethanol solution containing 2.84 g of glycidyl methacrylate slowly, and stirring continuously at 60°C for 24 h for reaction, after the reaction, removing ethanol using a rotary evaporator to obtain a crude product, washing the crude product 3 times with n-hexane and then 3 times with ethyl acetate to obtain the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide, drying the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide under vacuum at 30°C for 24 h at 1000 Pa, and storing the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide in a refrigerator at 4°C for use.

[0016] By adopting the above technical solution, in the present application, the method for preparing the ionic liquid 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide involves multiple steps, and the effects and synergistic effects thereof are as follows. The ionic liquid, as a major component of the electrolyte, provides a high ionic conductivity, which is the basis for achieving the function of a solid electrolyte. The nonflammability and chemical stability of the ionic liquid help to improve the safety of the battery and prevent the battery from burning or exploding under abnormal conditions. The good compatibility of the ionic liquid with the lithium metal negative electrode ensures the stability of the electrolyte at the electrode surface, contributing to the formation of a stable solid electrolyte/electrode interface. The method for preparing the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide ensures the purity and performance of the ionic liquid, and improves the quality and performance of a final product

through a specific reaction step and washing process.

[0017] Preferably, the lithium salt is prepared by mixing lithium bis (trifluoromethane) sulfonamide, lithium hexafluorophosphate, and lithium bis(fluorosulfonyl)imide in a mass part ratio of 5: 3: 2.

[0018] By adopting the above technical solution, the lithium salt is a key component in the electrolyte for providing freely movable lithium ions, and these lithium ions migrate between the positive and negative electrodes during the charging and discharging process of the battery, so as to realize the charging and discharging functions of the battery. By mixing different types of lithium salts, the ionic conductivity of the electrolyte can be optimized. Different lithium salts have different degrees of dissociation and ionic conductivity characteristics, and the mixed-use of different lithium salts can achieve higher ionic conductivity at room temperature. The synergistic effect between mixed lithium salts can improve the comprehensive performance of the electrolyte. By reasonable combination, an electrolyte with both high ionic conductivity and good mechanical stability can be obtained.

[0019] Preferably, the photoinitiator is prepared by mixing phenyl bis (2, 4, 6-trimethylbenzoyl) phosphine oxide and ethyl 2, 4, 6-trimethylbenzoyl phenylphosphonate in a mass part ratio of 1: 1.

[0020] In a second aspect, the present application provides a method for preparing the in-situ polymerized solid electrolyte, which adopts the following technical solution.

[0021] As a general technical concept, the present application further provides a method for preparing the in-situ polymerized solid electrolyte described above, including the following steps:

[0022] S61, in a dry argon-filled glove with oxygen solubility < 0.1 ppm and water vapor solubility < 0.1 ppm, mixing the ionic liquid and vinyl ethylene carbonate uniformly, then adding the lithium salt, and stirring for 10-15 minutes to obtain a mixed solution; and

[0023] S62, adding the photoinitiator dropwise to the mixed solution, stirring for 3-5 minutes

to obtain a precursor mother liquid, and then irradiating the precursor mother liquid with UV light to initiate a polymerization reaction to obtain the in-situ polymerized solid electrolyte.

[0024] By adopting the above technical solution, a solid polymer electrolyte based on polymer ionic liquid is prepared by simple in-situ copolymerization of ionic liquid monomers and vinyl ethylene carbonate monomers. The introduction of vinyl ethylene carbonate units with high conductivity can decrease the crystallinity of polymer, inhibit the crystallization of polymer, promote the dissociation of lithium salt, and improve the ionic conductivity of polymer electrolyte membrane.

[0025] Preferably, in the step S62, process conditions of UV light irradiation include: a distance between an UV light source and the precursor mother liquid for photoinitiated in-situ polymerization is greater than 0 and not more than 30 cm; a time of the UV light irradiation is 5-10 minutes, and a power of the UV light is 0.1-0.2 W.

[0026] Preferably, an absorption wavelength of the UV light is 365 nm.

[0027] In a third aspect, the present application provides a use of the in-situ polymerized solid electrolyte, which adopts the following technical solution.

[0028] As a general technical concept, the present application further provides a use of the in-situ polymerized solid electrolyte as described above in a lithium battery.

[0029] Preferably, the lithium battery is assembled by the following steps: coating an active substance formed by mixing lithium iron phosphate, conductive carbon black Super P and polyvinylidene fluoride in a mass ratio of 8: 1: 1 on an aluminum foil with a thickness of 20 μm , drying the aluminum foil at 120°C under a vacuum of 1000 Pa, and then directly cutting the aluminum foil into a disc with a diameter of 12 mm to obtain a lithium iron phosphate cathode sheet for use, wherein a loading capacity of the active substance on the aluminum foil is 4 mg/cm^2 ; in an argon-filled glove box with an oxygen solubility of < 0.1 ppm and water vapor solubility of < 0.1 ppm, mixing the ionic liquid and vinyl ethylene carbonate uniformly, then adding the lithium salt, stirring for 10-15 minutes, and then adding photoinitiator dropwise,

stirring for 3-5 minutes to obtain the precursor mother liquid, coating the precursor mother liquid on a surface of the lithium iron phosphate cathode sheet, after irradiating the lithium iron phosphate cathode sheet with 365 nm UV light with a power of 0.1-0.2 W for 5-10 minutes, covering and pasting a lithium foil to assemble a CR2032 type solid-state lithium battery.

[0030] By adopting the above technical solution, the all-solid-state lithium battery assembled by matching the in-situ polymerized solid electrolyte with the lithium iron phosphate cathode of the present application has better cycling stability and rate performance.

[0031] In summary, the present application has the following beneficial effects.

[0032] 1. Increasing ionic conductivity: by using the ionic liquid having a high ionic conductivity and vinyl ethylene carbonate monomers, the in-situ polymerized solid electrolyte can achieve a high ionic conductivity at room temperature, which is critical for power output and efficiency of a battery.

[0033] 2. Improving thermal stability and flame retardancy: the nonflammability and good thermal stability of the ionic liquid help to improve the safety of the battery, preventing thermal runaway or burning under high temperatures or abnormal conditions.

[0034] 3. Inhibiting lithium dendrite growth: the in-situ polymerized solid electrolyte can effectively inhibit the growth of lithium dendrites, which helps to maintain the stability of the electrode surface, and reduce the capacity loss and safety hazards during battery cycling.

[0035] 4. Enhancing electrode/electrolyte interface stability: the in-situ polymerization strategy enables the electrolyte to form a uniform and dense layer on the electrode surface, reducing the ion transport resistance, and improving the ion transport capacity at the electrode/electrolyte interface, thereby improving the cycling stability and rate performance of the battery.

[0036] 5. Increasing flexibility: the introduction of the vinyl ethylene carbonate increases the segmental motion of the polymer and increases the flexibility of the polymer, which enables the electrolyte to better adapt to changes in the volume of the electrode and reduces the problem of

interface separation caused by mechanical stress.

[0037] 6. Expanding the electrochemical window: the in-situ polymerized solid electrolyte has a wide electrochemical window, which means that the battery can operate stably over a wider voltage range, is suitable for different types of electrode materials.

[0038] 7. Optimizing lithium salt dissociation: the introduction of vinyl ethylene carbonate units with highly conductive can decrease the crystallinity of the polymer, promote the dissociation of the lithium salt, and further improve the ionic conductivity of the electrolyte.

[0039] 8. Compatibility: the good compatibility of the ionic liquid with the lithium metal negative electrode ensures the stability of the electrolyte at the electrode surface, helping to the formation of a stable solid electrolyte/electrode interface.

DETAILED DESCRIPTION

[0040] Embodiments of the present application are described in detail below in conjunction with examples. However, those skilled in the art will understand that the following examples are only used to illustrate the present application and should not be regarded as limiting the scope of the present application. For the examples where specific conditions are not specified, operations shall be carried out in accordance with conventional conditions or the conditions recommended by the manufacturer. The reagents or instruments used without specifying the manufacturer are all conventional products that can be obtained through market procurement.

[0041] Preparation Example 1: Preparation of 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide

[0042] 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide was prepared by a method including the following steps:

[0043] S31, 10 g of 3-chloropropylamine hydrochloride, 7.12 g of N-methylimidazole, and 10 mL of tetrahydrofuran were added to a reactor. A resulting mixture was stirred at 60°C for 48 h

for reaction to obtain a product. After the reaction, the product was dissolved in 20 mL of deionized water and washed 3 times with 15 mL of tetrahydrofuran to obtain a 1-aminopropyl-3-methylammonium chloride solution;

[0044] S32, 10 g of lithium bis (trifluoromethane) sulfonamide was dissolved in 20 mL of deionized water. Then, the 1-aminopropyl-3-methylammonium chloride solution was added dropwise and stirred for 24 h for reaction. After the reaction, a resulting mixture was washed 3 times with deionized water, and vacuum distillation at 60°C under 1000 Pa for 12 h was performed to obtain a product A; and

[0045] S33, 10 g of the product A was dissolved in 25 mL of ethanol, 75 mL of an ethanol solution containing 2.84 g of glycidyl methacrylate was slowly added, and stirred continuously at 60°C for 24 h for reaction, after the reaction, ethanol was removed using a rotary evaporator to obtain a crude product; the crude product was washed 3 times with n-hexane and then 3 times with ethyl acetate to obtain the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide; the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide was dried under vacuum at 30°C for 24 h at 1000 Pa, then stored in a refrigerator at 4°C for use.

[0046] Example 1

[0047] An in-situ polymerized solid electrolyte included the following raw materials for preparation: 1-vinyl-3-methylimidazolium bis(fluorosulfonyl)imide 35 g, vinyl ethylene carbonate 35 g, lithium salt 25 g, and photoinitiator 0.05 g, the lithium salt was prepared by mixing lithium bis (trifluoromethane) sulfonamide, lithium hexafluorophosphate, and lithium bis(fluorosulfonyl)imide in a mass part ratio of 5: 3: 2; the photoinitiator was prepared by mixing phenyl bis (2, 4, 6-trimethylbenzoyl) phosphine oxide and ethyl 2, 4, 6 trimethylbenzoyl phenylphosphonate in a mass part ratio of 1: 1.

[0048] The in-situ polymerized solid electrolyte described above was prepared by a method

including the following steps:

[0049] S61, in a dry argon-filled glove with oxygen solubility < 0.1 ppm and water vapor solubility < 0.1 ppm, the 1-vinyl-3-methylimidazolium bis(fluorosulfonyl)imide and vinyl ethylene carbonate were mixed until uniform; then the lithium salt was added and stirred for 10 minutes to obtain a mixed solution; and

[0050] S62, the photoinitiator was added dropwise to the mixed solution and stirred for 3 minutes to obtain a precursor mother liquid; then the precursor mother liquid was irradiated with UV light to initiate a polymerization reaction to obtain the in-situ polymerized solid electrolyte. Process conditions of UV light irradiation included: a distance between an UV light source and the precursor mother liquid for photoinitiated in-situ polymerization was 20 cm; a time of the UV light irradiation was 5 minutes, and a power of the UV light was 0.2 W; an absorption wavelength of UV light was 365 nm.

[0051] Example 2

[0052] An in-situ polymerized solid electrolyte included the following raw materials for preparation: 1-vinyl-3-ethylimidazolium bis(trifluoromethylsulfonyl)imide 40 g, vinyl ethylene carbonate 40 g, lithium salt 28 g, and photoinitiator 0.1g; the lithium salt was prepared by mixing lithium bis (trifluoromethane) sulfonamide, lithium hexafluorophosphate, and lithium bis(fluorosulfonyl)imide in a mass part ratio of 5: 3: 2; the photoinitiator was prepared by mixing phenyl bis (2, 4, 6-trimethylbenzoyl) phosphine oxide and ethyl 2, 4, 6 trimethylbenzoyl phenylphosphonate in a mass part ratio of 1: 1.

[0053] The in-situ polymerized solid electrolyte described above was prepared by a method including the following steps:

[0054] S61, in a dry argon-filled glove with oxygen solubility < 0.1 ppm and water vapor solubility < 0.1 ppm, the 1-vinyl-3-ethylimidazolium bis(trifluoromethylsulfonyl)imide and vinyl ethylene carbonate were mixed until uniform; then the lithium salt was added and stirred for 15 minutes to obtain a mixed solution; and

[0055] S62, the photoinitiator was added dropwise to the mixed solution and stirred for 5 minutes to obtain a precursor mother liquid; then the precursor mother liquid was irradiated with UV light to initiate a polymerization reaction to obtain the in-situ polymerized solid electrolyte. Process conditions of UV light irradiation included: a distance between an UV light source and the precursor mother liquid for photoinitiated in-situ polymerization was 20 cm; a time of the UV light irradiation was 10 minutes, and a power of the UV light was 0.1 W; the absorption wavelength of UV light was 365 nm.

[0056] Example 3

[0057] An in-situ polymerized solid electrolyte included the following raw materials for preparation: 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide 38 g, vinyl ethylene carbonate 38 g, lithium salt 26 g, and photoinitiator 0.07 g; the lithium salt was prepared by mixing lithium bis (trifluoromethane) sulfonamide, lithium hexafluorophosphate, and lithium bis(fluorosulfonyl)imide in a mass part ratio of 5: 3: 2; the photoinitiator was prepared by mixing phenyl bis (2, 4, 6-trimethylbenzoyl) phosphine oxide and ethyl 2, 4, 6 trimethylbenzoyl phenylphosphonate in a mass part ratio of 1: 1.

[0058] The in-situ polymerized solid electrolyte described above was prepared by a method including the following steps:

[0059] S61, in a dry argon-filled glove with oxygen solubility < 0.1 ppm and water vapor solubility < 0.1 ppm, the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide and vinyl ethylene carbonate were mixed until uniform; then the lithium salt was added and stirred for 13 minutes to obtain a mixed solution; and

[0060] S62, the photoinitiator was added dropwise to the mixed solution and stirred for 4 minutes to obtain a precursor mother liquid; then the precursor mother liquid was irradiated with UV light to initiate a polymerization reaction to obtain the in-situ polymerized solid electrolyte.

Process conditions of UV light irradiation included: a distance between an UV light source and the precursor mother liquid for photoinitiated in-situ polymerization was 20 cm; a time of the UV light irradiation was 8 minutes, and a power of the UV light was 0.15 W; an absorption wavelength of UV light was 365 nm.

[0061] Comparative Example 1

[0062] Substantially the same as Example 3 except that the lithium salt was lithium bis(trifluoromethane) sulfonamide.

[0063] Comparative Example 2

[0064] Substantially the same as Example 3 except that the lithium salt was lithium hexafluorophosphate.

[0065] Comparative Example 3

[0066] Substantially the same as Example 3 except that the lithium salt was lithium bis(fluorosulfonyl)imide.

[0067] Application Example 1

[0068] The lithium battery was assembled by the following steps. An active substance formed by mixing lithium iron phosphate, conductive carbon black Super P, and polyvinylidene fluoride in a mass ratio of 8: 1: 1 was coated on an aluminum foil with a thickness of 20 μm , the aluminum foil was dried at 120°C under a vacuum of 1000 Pa, and then directly cut into a disc with a diameter of 12 mm to obtain a lithium iron phosphate cathode sheet for use. An loading capacity of the active substance on the aluminum foil was 4 mg/cm^2 . In an argon-filled glove box with an oxygen solubility of < 0.1 ppm and water vapor solubility of < 0.1 ppm, the precursor mother liquid of the in-situ polymerized solid electrolyte prepared in Example 1 was coated on a surface of the lithium iron phosphate cathode sheet. After the lithium iron phosphate cathode sheet was irradiated with 365 nm UV light at a power of 0.2 W for 5 minutes, a lithium foil was covered and pasted to assemble a CR2032 type solid-state lithium battery.

[0069] Application Example 2

[0070] The lithium battery was assembled by the following steps. An active substance formed by mixing lithium iron phosphate, conductive carbon black Super P, and polyvinylidene fluoride in a mass ratio of 8: 1: 1 was coated on an aluminum foil with a thickness of 20 μm , the aluminum foil was dried at 120°C under a vacuum of 1000 Pa, and then directly cut into a disc with a diameter of 12 mm to obtain a lithium iron phosphate cathode sheet for use. An loading capacity of the active substance on the aluminum foil was 4 mg/cm^2 . In an argon-filled glove box with an oxygen solubility of < 0.1 ppm and water vapor solubility of < 0.1 ppm, the precursor mother liquid of the in-situ polymerized solid electrolyte prepared in Example 2 was coated on a surface of the lithium iron phosphate cathode sheet. After the lithium iron phosphate cathode sheet was irradiated with 365 nm UV light at a power of 0.1 W for 10 minutes, a lithium foil was covered and pasted to assemble a CR2032 type solid-state lithium battery.

[0071] Application Example 3

[0072] The lithium battery was assembled by the following steps. An active substance formed by mixing lithium iron phosphate, conductive carbon black Super P, and polyvinylidene fluoride in a mass ratio of 8: 1: 1 was coated on an aluminum foil with a thickness of 20 μm , the aluminum foil was dried at 120°C under a vacuum of 1000 Pa, and then directly cut into a disc with a diameter of 12 mm to obtain a lithium iron phosphate cathode sheet for use. An loading capacity of the active substance on the aluminum foil was 4 mg/cm^2 . In an argon-filled glove box with an oxygen solubility of < 0.1 ppm and water vapor solubility of < 0.1 ppm, the precursor mother liquid of the in-situ polymerized solid electrolyte prepared in Example 3 was coated on a surface of the lithium iron phosphate cathode sheet. After the lithium iron phosphate cathode sheet was irradiated with 365 nm UV light at a power of 0.15W for 8 minutes, a lithium foil was covered and pasted to assemble a CR2032 type solid-state lithium battery.

[0073] Application Example 4

[0074] Substantially the same as Application Example 3, except that: the precursor mother liquid of the in-situ polymerized solid electrolyte prepared in Comparative Example 1 was

coated on a surface of the lithium iron phosphate cathode sheet.

[0075] Application Example 5

[0076] Substantially the same as Application Example 3, except that: the precursor mother liquid of the in-situ polymerized solid electrolyte prepared in Comparative Example 2 was coated on a surface of the lithium iron phosphate cathode sheet.

[0077] Application Example 6

[0078] Substantially the same as Application Example 3, except that: the precursor mother liquid of the in-situ polymerized solid electrolyte prepared in Comparative Example 3 was coated on a surface of the lithium iron phosphate cathode sheet.

[0079] Performance testing

[0080] The solid-state lithium batteries prepared in Application Examples 1 to 6 were sampled and placed for 24 hours, and then the stability test and long-cycle test of the solid-state lithium batteries were performed using a CT-4000 battery tester (manufactured in Shenzhen). The test results are shown in Table 1.

[0081] Capacity Testing: 2.8-3.8 V was used to test the discharge capacity at 0.2 C rate.

[0082] Long Cycle Testing: long-term charge-discharge tests were performed at 30°C at a rate of 0.2 C at 2.8-3.8 V.

[0083] Table 1 Performance testing

Item	Applicati on Example 1	Applicati on Example 2	Applicati on Example 3	Applicati on Example 4	Applicati on Example 5	Applicati on Example 6
First Discharge Capacity at a rate of 0.2 C (mAh/g)	147	149	154	152	153	152
First Discharge	90	91	94	93	94	93

Efficiency (%)						
Capacity retention after 2000 cycles at a rate of 0.2 C (%)	77	79	88	76	77	75

[0084] By analyzing the data in Table 1, it can be seen that:

[0085] 1) The in-situ polymerized solid electrolyte prepared in Examples 1-3 has excellent first discharge capacity, first coulombic efficiency, and stable cycling performance. At the same time, it shows that the in-situ polymerized solid electrolyte of the present application has good thermal stability and flame retardancy, has high ionic conductivity at room temperature, inhibits the growth of lithium dendrites, and achieves a stable electrolyte/electrode interface. An all-solid-state lithium battery assembled by matching the in-situ polymerized solid electrolyte with the lithium iron phosphate cathode of the present application has better cycling stability and rate performance. Through comparative analysis of the performances of the solid-state batteries prepared in Application Example 3 and Application Examples 1-2, it is shown that the ionic liquid 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide prepared by the present application not only provides good ionic conductivity, but also enhances the overall performance of the polymer electrolyte, including thermal stability, flexibility, and electrochemical stability, through the synergistic effect with vinyl ethylene carbonate.

[0086] 2) A comparative analysis of the performances of the solid-state batteries prepared in Application Example 3 and Application Examples 4 to 6 shows that the lithium salt can be prepared by mixing lithium bis (trifluoromethane) sulfonamide, lithium hexafluorophosphate and lithium bis(fluorosulfonyl)imide in a mass part ratio of 5: 3: 2 and that the ionic conductivity of the electrolyte can be optimized by mixing different types of lithium salts. Different lithium salts have different degrees of dissociation and ionic conductivity characteristics, and the mixed-use of different lithium salts can achieve higher ionic conductivity at room temperature.

Synergism between mixed lithium salts can improve the cycling stability of the solid-state battery.

[0087] The above embodiments are only used to explain and illustrate the technical solution of the present application, and not to limit it. Although the above examples provide specific explanations of the present application, related technical personnel should understand that modifications or equivalent substitutions can still be made to the specific embodiments of the present invention, and any modifications and equivalent substitutions that do not depart from the spirit and scope of the present application should be included in the scope of the present application.

WHAT IS CLAIMED IS:

1. An in-situ polymerized solid electrolyte, characterized by comprising the following raw materials for preparation in parts by mass: 35-40 parts of an ionic liquid, 35-40 parts of vinyl ethylene carbonate, 25-28 parts of a lithium salt, and 0.05-0.1 parts of a photoinitiator.

2. The in-situ polymerized solid electrolyte according to claim 1, characterized in that, the ionic liquid is one selected from a group consisting of: 1-vinyl-3-methylimidazolium bis(fluorosulfonyl)imide, 1-vinyl-3-ethylimidazolium bis(trifluoromethylsulfonyl)imide, and 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide.

3. The in-situ polymerized solid electrolyte according to claim 2, characterized in that, the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide is prepared by a method comprising the following steps:

S31, adding 10 g of 3-chloropropylamine hydrochloride, 7.12 g of N-methylimidazole, and 10 mL of tetrahydrofuran to a reactor, and stirring at 60°C for 48 h for reaction to obtain a product, after the reaction, dissolving the product in 20 mL of deionized water, and washing 3 times with 15 mL of tetrahydrofuran to obtain a 1-aminopropyl-3-methylammonium chloride solution;

S32, dissolving 10 g of lithium bis (trifluoromethane) sulfonamide in 20 mL of deionized water, then adding the 1-aminopropyl-3-methylammonium chloride solution dropwise, and stirring for 24 h for reaction, after the reaction, washing a resulting mixture 3 times with deionized water, and performing vacuum distillation at 60°C under 1000 Pa for 12 h to obtain a product A; and

S33, dissolving 10 g of the product A in 25 mL of ethanol, adding 75 mL of an ethanol solution containing 2.84 g of glycidyl methacrylate slowly, and stirring continuously at 60°C for 24 h for reaction, after the reaction, removing ethanol using a rotary evaporator to obtain a crude

product, washing the crude product 3 times with n-hexane and then 3 times with ethyl acetate to obtain the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide, drying the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide under vacuum at 30°C for 24 h at 1000 Pa, and then storing the 3-(3-((2-hydroxy-3-(methacryloyloxy) propyl) amino) propyl)-1-methyl-1H-imidazol-3-bis[(trifluoromethane)sulfonyl]imide in a refrigerator at 4°C for use.

4. The in-situ polymerized solid electrolyte according to claim 1, characterized in that, the lithium salt is prepared by mixing lithium bis (trifluoromethane) sulfonamide, lithium hexafluorophosphate, and lithium bis(fluorosulfonyl)imide in a mass part ratio of 5: 3: 2.

5. The in-situ polymerized solid electrolyte according to claim 1, characterized in that, the photoinitiator is prepared by mixing phenyl bis (2, 4, 6-trimethylbenzoyl) phosphine oxide and ethyl 2, 4, 6-trimethylbenzoyl phenylphosphonate in a mass part ratio of 1: 1.

6. A method for preparing the in-situ polymerized solid electrolyte according to any one of claims 1-5, characterized by comprising the following steps:

S61, in a dry argon-filled glove with oxygen solubility < 0.1 ppm and water vapor solubility < 0.1 ppm, mixing the ionic liquid and vinyl ethylene carbonate uniformly, then adding a lithium salt, and stirring for 10-15 minutes to obtain a mixed solution; and

S62, adding the photoinitiator dropwise to the mixed solution, stirring for 3-5 minutes to obtain a precursor mother liquid, and then irradiating the precursor mother liquid with ultraviolet (UV) light to initiate a polymerization reaction to obtain the in-situ polymerized solid electrolyte.

7. The method for preparing the in-situ polymerized solid electrolyte according to claim 6, characterized in that, in the step S62, process conditions of UV light irradiation comprise: a distance between an UV light source and the precursor mother liquid for photoinitiated in-situ

polymerization is greater than 0 and not more than 30 cm; a time of the UV light irradiation is 5-10 minutes, and a power of the UV light is 0.1-0.2 W.

8. The preparation method of the in-situ polymerized solid electrolyte according to claim 7, characterized in that, an absorption wavelength of the UV light is 365 nm.

9. Use of the in-situ polymerized solid electrolyte according to any one of claims 1-5 in a lithium battery.

10. The use of the in-situ polymerized solid electrolyte according to claim 9, characterized in that, the lithium battery is assembled by the following steps: coating an active substance formed by mixing lithium iron phosphate, conductive carbon black Super P, and polyvinylidene fluoride in a mass ratio of 8: 1: 1 on an aluminum foil with a thickness of 20 μm , drying the aluminum foil at 120°C under a vacuum of 1000 Pa, and then directly cutting the aluminum foil into a disc with a diameter of 12 mm to obtain a lithium iron phosphate cathode sheet for use, wherein a loading capacity of the active substance on the aluminum foil is 4 mg/cm^2 ; in an argon-filled glove box with an oxygen solubility of < 0.1 ppm and water vapor solubility of < 0.1 ppm, mixing the ionic liquid and vinyl ethylene carbonate uniformly, then adding the lithium salt, stirring for 10-15 minutes, and then adding the photoinitiator dropwise, stirring for 3-5 minutes to obtain a precursor mother liquid, coating the precursor mother liquid on a surface of the lithium iron phosphate cathode sheet, after irradiating the lithium iron phosphate cathode sheet with 365 nm UV light with a power of 0.1-0.2 W for 5-10 minutes, covering and pasting a lithium foil to assemble a CR2032 type solid-state lithium battery.