

**UNITED STATES DISTRICT COURT
FOR THE EASTERN DISTRICT OF TEXAS
MARSHALL DIVISION**

NINGDE AMPEREX TECHNOLOGY
LIMITED,

Plaintiff,

v.

ZHUHAI COSMX BATTERY CO., LTD.,

Defendant.

Civil Action No. 2:22-CV-00232-JRG

DECLARATION OF DR. SETH MILLER

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I. EXPERIENCE AND QUALIFICATIONS

1. I, Seth Miller, Ph.D., submit this declaration at the request of Zhuhai CosMX Battery Co. Ltd., as an expert in the fields of chemistry, polymers (plastics), polymer-particle composites, porous materials, and materials characterization techniques (including scattering and spectroscopies) in relation to lithium-ion batteries. My qualifications in these areas, as well as other areas, are established below and by my curriculum vitae, which is attached as Appendix A and includes a list of my publications.

2. My background includes a Ph.D. in Chemistry from the California Institute of Technology in 1998, and postdoctoral research at the University of California, Berkeley. My work in academic labs included developing organic magnetic and conductive materials, including small molecules and polymers. These small molecules included nitrile and multi-nitrile materials. During this research I applied electrochemical techniques including bulk electrolysis, and characterized the resulting thin films

3. From 2000 to 2005 I worked in the packaging group of Digital Light Products at Texas Instruments, with a primary focus on ensuring that the packaging environment and surface chemistry of this micro-machined device was stable during long term device use. Part of this work included application of high surface area, porous inorganic materials as sorbents.

4. From 2005 to 2008 I worked at a startup called Zettacore developing new electrolyte for a single-molecule thick battery for application as a memory device. In that role I created new thin film and free-standing electrolyte and separator materials; built thin film battery cells; and tested those cells for initial functionality and long term performance. The electrolytes were a composite material comprising organic ionic liquid salts, nanoparticles, and optionally polymer precursors and organic electrolyte co-solvents. Among the characterization techniques I

used for these materials were dynamic light scattering, SEM, and AFM. During this period, I also developed porous inorganic films as separator materials for these thin film batteries, and applied organic-inorganic composite films for membrane gas separation. These thin composite films for gas separations were commonly laminated on a porous support.

5. Starting in 2008 I became an independent consultant, and have since consulted with dozens of companies, investors, and academics in the field of batteries including battery materials development, novel architectures, cost modeling, and applicability for scale-up, including an appointment as research faculty at the University of Colorado, Boulder to formally advise a graduate student in the development of a nanostructured battery. I have worked with two Series A venture firms to evaluate their potential investments in the battery market, and act as a paid consultant to companies evaluating market opportunities for next generation energy storage devices. I am author or co-author on four issued patents for battery materials and architectures.

6. As an independent consultant, I have served as an inventor on numerous patents for my clients, including work on membrane gas separation materials. This included application of materials and coatings on porous supports.

7. I have served as an expert in litigation in seventeen cases outside of this one, including a battery patent infringement hearing before the ITC, during which I conducted extensive analysis of battery separator materials, including consideration of their coatings. My role in this and other matters was to coordinate chemical and physical analysis, and opine on technical matters related to their function.

8. My background and qualifications are more fully set out in my *curriculum vitae*, attached as Appendix A.

II. DOCUMENTS AND INFORMATION CONSIDERED IN FORMING OPINIONS

9. I have been asked to provide my opinion on various terms and phrases in the claims of U.S. Patent No. 10,964,987 (“the ’987 patent”) as of the asserted priority date of the ’987 patent, April 11, 2018, and U.S. 10,833,363 (“the ’363 patent”) as of the asserted priority date of the ’363 patent, September 21, 2018, based on the knowledge and understanding of a person of ordinary skill in the art, and to respond to certain proposed claim interpretations of Plaintiff.

10. In forming my opinions, I have reviewed the ’987 patent and related prosecution history and the ’363 patent and related prosecution history. I also have considered the materials listed in Appendix C.

11. I additionally have based my opinions on my professional and academic experience in the areas of chemistry and particle size characterization and analysis techniques, generally, and as to separators and lithium-ion batteries, as well as electrolytes. I reserve the right to testify about my experience and the materials I considered in forming my opinions, as listed in Appendix C. To the extent I am provided additional documents or information, including any declarations produced by Plaintiff, I may offer further opinions. In addition to these materials, I may consider additional documents and information in forming any rebuttal opinions.

III. LEGAL PRINCIPLES

12. I have no formal legal training, but I have been advised by Defendant’s counsel about general guidelines and rules that I should apply relating to reviewing patent claims. I reserve the right to supplement my declaration to consider any modifications of these standards if I am informed of any such modifications.

13. I have been informed that a patent includes a specification containing the written description of one or more embodiments of the invention, drawings, and figures, as well as one or more claims that point out and claim the subject matter of the invention. I understand that the claims define and measure the patent's scope. Each claim defines a separate invention. I have also been informed and understand that a dependent claim incorporates each and every element of the claim from which it depends.

14. I have been informed and understand that the patent's claims are construed to determine their meaning and scope.

15. I have been informed and understand that the proper construction of the patent claims, and their terms and phrases, is a matter of law for the Court to decide. I understand that the Court construes the patent claims according to their ordinary and customary meaning to a person of ordinary skill in the art at the time of invention. I understand that interpreting a term or phrase contained in a patent claim involves reading the language of the claim and considering other materials relevant to the claim, including the patent specification and the prosecution history. I understand that evidence extrinsic to the patent and its prosecution history may also be consulted, provided it is not inconsistent with the claim language, the patent specification, or the prosecution history.

16. I have been informed and understand that a patent's claim, viewed in light of the specification and prosecution history, must inform a person of ordinary skill of the scope of the invention with reasonable certainty. I understand that if it does not, it is indefinite.

17. I have been informed and understand that to be found not indefinite, a claim must provide objective boundaries for a person of ordinary skill in the relevant field. I understand that

a claim may be indefinite when it contains words or phrases with unclear meanings or meanings that fail to define the scope of the claimed invention with reasonable certainty.

18. I have been informed and understand that when a claimed characteristic depends on a measurement for which different measuring methods lead to different results, the patent specification or prosecution history must guide a skilled artisan on which method to use. I understand that without providing guidance, the claim may be indefinite. I understand that a patent specification presenting specific working examples of the method used to measure the claimed characteristic may provide an objective standard of the claim's scope.

19. I also understand that a "subjective" claim term having a scope that depends on a particular person's opinion (and may vary from person to person) may be indefinite.

20. I have been informed and understand that a claim may be indefinite when (1) skilled artisans are aware of different known methods for measuring a claimed characteristic, (2) the specification and the prosecution history do not suggest any of the known methods in particular, and (3) applying the different known methods results in such varying outcomes that a product may infringe the claim when employing one known method but the same product may not infringe the claim when employing another known method.

IV. STATEMENT OF OPINIONS EXPRESSED AND BASES AND REASONS THEREFORE

A. Background of the '987 Patent and Asserted Claims

21. I understand that this litigation involves the '987 patent, which relates to "a separator and energy storage device," such as a lithium-ion battery. '987 patent, Abstract; *see, e.g.*, 2:17-18. The separator includes a porous layer that "comprises inorganic particles and a binder, and a ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.3 to 3.0." '987 patent, Abstract.

1. The '987 Patent Claims

22. Claim 1 of the '987 patent reads as follows:

A separator, comprising: a porous substrate; and a porous layer arranged on a surface of the porous substrate, wherein the porous layer comprises inorganic particles and a binder, and a ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.3 to 3.0.

23. Claim 2 is dependent on claim 1 and specifies that “the porous layer has a thickness of 0.2 μm to 10 μm .”

24. Claim 6 is dependent on claim 1 and specifies that “the inorganic particles are one or more selected from the group consisting of alumina, silica, magnesia, titanium oxide, hafnium dioxide, tin oxide, zirconia, cerium dioxide, nickel oxide, zinc oxide, calcium oxide, boehmite, aluminum hydroxide, magnesium hydroxide, calcium hydroxide, and barium sulfate.”

25. Claim 7 is dependent on claim 1 and specifies that “the binder is one or more selected from the group consisting of polyvinylidene fluoride, vinylidene fluoride-hexafluoropropylene copolymer, polyamide, polyacrylonitrile, polyacrylate, polyacrylic acid, polyacrylates, sodium carboxymethylcellulose, polyvinylpyrrolidone, polyvinyl ether, polymethyl methacrylate, polytetrafluoroethylene and polyhexafluoropropylene.”

26. Claim 8 is dependent on claim 1 and recites “[a]n energy storage device, comprising the separator according to claim 1.”

27. Claim 9 is dependent on claim 8 and specifies that “the energy storage device is a lithium-ion battery.”

2. The Specification of the '987 Patent

28. The specification of the '987 patent describes a separator having a porous layer arranged on a surface of a porous substrate. '987 patent, 2:47-56; 3:57-66. The porous layer, as described by the specification, "comprises inorganic particles and a binder." '987 patent, 2:47-56; 3:57-66. The specification further discloses that the inorganic particles of the described separator "are one or more selected from the group consisting of alumina, silica, magnesia, titanium oxide, hafnium dioxide, tin oxide, zirconia, cerium dioxide, nickel oxide, zinc oxide, calcium oxide, boehmite, aluminum hydroxide, magnesium hydroxide, calcium hydroxide, and barium sulfate." '987 patent, 2:1-6. These materials include types of ceramics that were commonly used or studied for use in separators at the time of the asserted priority date in 2018. It further states that "the inorganic particles contained in the pores in the porous layer can serve to support the pores, and therefore the pores can be well protected from being destroyed." '987 patent, 2:63-66. "In the above-described separator, the pores have an average pore size of 0.3 μm to 20 μm ." '987 patent, 1:62-63.

29. The inorganic particles span a distribution of sizes, and the specification refers to the Dv90 of the inorganic particles, which it defines as "a particle size which reaches 90% the cumulative volume from the side of small particle size in the granularity distribution on a volume basis." '987 patent, 2:47-56; 3:57-66; 4:22-25. I understand that ATL chose to define Dv90 with this wording, which is somewhat awkward. In my opinion, ATL's definition is consistent with how an ordinary artisan would understand Dv90. An ordinary artisan would generally understand that Dv90 to represent the 90th percentile of the particle size distribution by particle volume. For example, Merkus writes that D90 represents "90% point in the cumulative undersize PSD,"

where PSD stands for the particle size distribution. Henk. G. Merkus, Particle Size Measurements: Fundamentals, Practice, Quality, 23 (2009) [hereinafter Merkus].

30. As noted above, claim 1 of the '987 patent requires “a ratio of Dv90 of the inorganic particles to the thickness of the porous layer [to be] in a range from 0.3 to 3.0.”

31. The specification details that the thickness of the porous layer is measured “on a thickness gage . . . using a 5 mm flat bottom probe with a speed of 50 mm/min and a pressure of 0.01N.” '987 patent, 13:35-39. The porous layer is measured with a “Model VL-50 LITEMATIC from Naitutoyo company” thickness gauge. '987 patent, 13:35-39. The specification further describes that “[e]ach of the separator samples provided with a porous layer is measured for 60 thickness points, and the average thickness is taken as the measured value.” '987 patent, 13:39-41. “The thickness of the porous layer is equal to the thickness of the separator minus the thickness of the porous substrate.” '987 patent, 13:41-43. Additionally, the specification details that when “there are two porous films, the thickness is divided by 2 to obtain the thickness of each porous layer.” '987 patent, 13:43-45.

32. Unlike the measurement details for the thickness parameter, the specification does not provide a measurement technique for the Dv90 of the inorganic particles. Nor does it provide the details that a POSA would need to implement any of the known possible measurement techniques for the Dv90 of the inorganic particles at the precision of Dv90 required by the reported ratio values in the '987 patent, such as the measurement principle, sample source and lot number, and any additional numerical analyses or models used to process the data. *Compare* ASTM E1617-09 (Reapproved 2014) at 2-3; *and* '987 patent, 13:35-45 (disclosing the instrument, measurement technique, number of samples, and numerical analysis used to measure

the thickness of the porous layer); *with* '987 patent, 1:35-16:23 (disclosing no such information about the Dv90 measurement).

33. The exact value of this ratio is stated to be critical for battery performance. “[T]he ratio of Dv90 of the inorganic particles to the thickness of the porous layer has an effect on the rate performance and cycle performance of the lithium-ion battery.” '987 patent, 15:26-30. The '987 patent's specification further discloses that, for improved rate performance and cycle performance, the ratio of the Dv90 of the inorganic particles to the thickness of the porous layer must be between 0.3 and 3.0. '987 patent, 2:47-56;15:32-36. The specification discloses that “the porous layer has a thickness of 0.2 μm to 10 μm .” '987 patent, 1:55-58. Accordingly, the Dv90 of the inorganic particles for the porous layer could range from 0.06 μm -30 μm . *See infra* ¶106, Table 1, n.5.

34. The specification reports test results showing the rate performance and cycle performance for fifteen examples consistent with the invention, examples 1-15. '987 patent, 10:6-12, Table 2. For example 1, “the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.3.” '987 patent, 11:22-27, Table 1. The specification reports the rate performance and cycle performance for example 1 at 79.50% and 85.70%, respectively. '987 patent, Table 2. Table 2 reports the thickness of the porous layer as 2 μm , therefore, the Dv90 of the inorganic particles in example 1 is 0.6 μm . '987 patent, Table 2. For example 7, “the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 3.0.” '987 patent, 11:22-27, Table 1. The specification reports the rate performance and cycle performance for example 7 at 80.40% and 82.70%, respectively. '987 patent, Table 2. Table 2 reports the thickness of the porous layer as 2 μm , therefore, the Dv90 of the inorganic particles in example 7 is 6 μm . '987 patent, Table 2.

35. The specification discloses that lithium-ion battery rate performance and cycle performance are negatively impacted “[i]f the ratio of Dv90 of the inorganic particles to the thickness of the porous layer is too low.” ’987 patent, 15:49-59. When the ratio gets too low, “the inorganic particles cannot be effectively exposed on the surface of the porous layer.” ’987 patent, 15:49-59. This, in turn, “does not facilitate an exchange of the first solvent in the coating solution and the third solvent in the coagulating solution in the preparation process of the porous layer.” ’987 patent, 15:49-59. Additionally, a too-low ratio “cannot enhance diffusion [and] is not advantageous for the development of the pores.” ’987 patent, 15:49-59.

36. The specification also discloses that performance is negatively impacted “[i]f the ratio of the Dv90 of the inorganic particles to the thickness of the porous layer is too high.” ’987 patent, 15:36-49. When the ratio gets too high, “the pore size distribution of the porous layer tends to be non-uniform.” ’987 patent, 15:36-49. Additionally, “the surface of the porous layer is heterogeneous, the porous layer is easily detached from the surface of the porous substrate, and defects are formed in the [lithium-ion battery].” ’987 patent, 15:36-49. A too-high ratio negatively affects “the safety performance of the [lithium ion battery].” ’987 patent, 15:36-49.

37. To illustrate these points, the specification reports test results showing the rate performance and cycle performance for comparative examples that are examples of separators and cells that are not part of the invention. ’987 patent, 15:26-36, Tables 1-2. For comparative example 1, “[t]he ratio of Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.2.” ’987 patent, 10:60-61, Table 1. The specification reports the rate performance and cycle performance for comparative example 1 at 69.80% and 70.50%, respectively. Table 2 reports the thickness of the porous layer as 2 μm , therefore, the Dv90 of the inorganic particles in comparative example 1 is 0.4 μm . ’987 patent, Table 2.

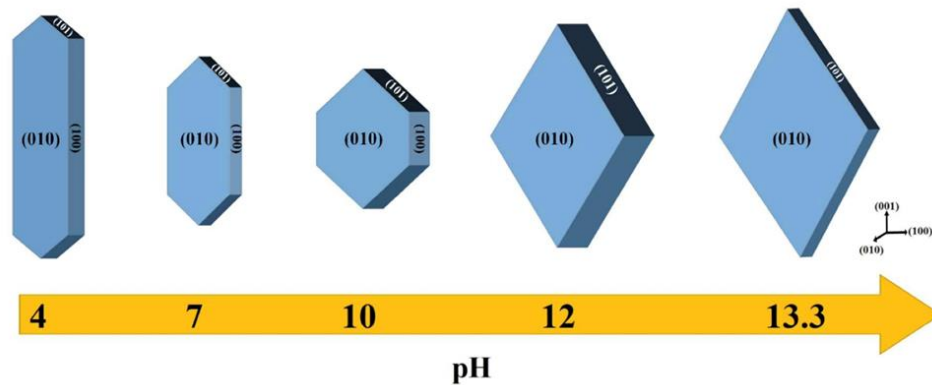
38. For comparative example 2, “the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 3.2.” ’987 patent, 11:10-13, Table 1. The specification reports the rate performance and cycle performance for comparative example 1 at 71.30% and 72.60%, respectively. Table 2 reports the thickness of the porous layer as 2 μm , therefore, the Dv90 of the inorganic particles in comparative example 2 is 6 μm . ’987 patent, Table 2. Because the specification provides no measured Dv90 of the inorganic particles, a person of ordinary skill in the art would need to calculate the Dv90 from the measurements provided and would be limited by the reported precision of those measurements. An ordinary artisan would understand that the value of “2 μm ” should be interpreted to contain one significant figure, following standard practice among chemists. Specifically, an ordinary artisan would understand that “[w]hen significant figures are used, the last digit is understood to be uncertain,” Raymond Chang, General Chemistry 14 (5th ed. 2008) [hereinafter Chang], such that the value should be interpreted as $2 \pm 1 \mu\text{m}$, or in the range of 1.5-2.5 μm . More specifically, an ordinary artisan would understand that the ’987 patent provides no guidance to expect that all films reported with a thickness of “2 μm ” are 2.0 μm , and do not have a thickness of 2.1 μm , or 1.9 μm , or 2.4 μm . When handling significant figures in calculations, a POSA would further understand that “the number of significant figures in the final product or quotient is determined by the original number that has the *smallest* number of significant figures.” Chang at 16. Thus when multiplying film thickness by the Dv90-to-thickness ratios reported in the patent, the calculated Dv90 can possess only one significant figure if the film thickness possesses only one significant figure. I have used such standard scientific practice in my discussions above and below. The specification reports the precision of the claimed ratio to two significant figures and the claimed thickness to one significant figure. Because the product of two measured values is

reliable only insofar as it has no more significant figures than either measured value, a person of ordinary skill in the art would reliably calculate that the Dv90 of the inorganic particles in comparative example 2 is 6 μm .

39. Nonetheless, if a person of ordinary skill in the art did not consider the reported number of significant figures of the thickness measurement (which a person of ordinary skill in the art would not do) the calculated Dv90 of the inorganic particles in comparative example 2 would be 6.4 μm .

40. The figures in the '987 patent do not provide enough information to evaluate the particle size distribution of the respective samples, and a skilled artisan cannot draw reliable conclusions from these images, including because of the limited number of particles and limited resolution, which inconclusively show some cubic, some rectangular, and possibly some rounded inorganic particles. Figs. 1-4. Indeed, as discussed below, there is no requirement that the boehmite particles as used in Example 3 to produce the images in Figures 1 and 2 will possess any particular morphology. In particular, is it known boehmite may include elongated, hexagon, cubic, or plate-like particles depending on the manner of synthesis, as discussed in Zhang 2018, which reflects the knowledge of the ordinary artisan at the asserted priority date of the '987 patent.¹ See generally Zhang et al., *Size and Morphology Controlled Synthesis of Boehmite Nanoplates and Crystal Growth Mechanisms*, 18 *Crystal Growth & Design* 3596 (2018) [hereinafter Zhang 2018]. The array of particle morphologies is illustrated from Scheme 1 of Zhang 2018, shown below.

¹ Zhang 2018 was published on April 25, 2018, which is two weeks after the asserted priority date of April 11, 2018. This accurately reflects that a person of ordinary skill in the art understood at and before the asserted priority date that boehmite could have various morphologies.



41.

Zhang 2018 at Scheme 1.

42. The '987 patent does not discuss particle morphology. Figures 1-4 are of sufficiently low resolution that I find it difficult to interpret what particle morphologies are present in the sample, and it is my opinion that an ordinary artisan would be similarly challenged.

3. The Prosecution History of the '987 Patent

43. During prosecution of U.S. 16/013,887, which led to the '987 patent, the Applicant acknowledged that adjusting particle size or inorganic particles in a separator was known in the art at the time of the invention and that there are many parameters by which a skilled artisan can measure particle size (e.g., Dv10, Dv50, "mode diameter," etc.).

ATL_COSMX_00001987, April 10, 2020 Response to Non-Final Office Action, 3. The Applicant argued that a person of ordinary skill in the art would not conceive of the ratio of the Dv90 of the inorganic particles to the thickness of the porous layer. ATL_COSMX_00001987, June 20, 2018 Response to Non-Final Office Action, 3.

44. The ratio of the Dv90 of the inorganic particles to the thickness of the porous layer was indicated as the reason for allowance of the '987 patent. ATL_COSMX_00001862, Notice of Allowance, 4. During prosecution, the Applicant stressed the importance of measuring

both the thickness of the porous layer and the Dv90 of the inorganic particles to provide the claimed ratio. ATL_COSMX_00001913, November 4, 2020 Amendment and Response to Non-Final Office Action, 7 (“Note, a person of ordinary skill in the art would not be able to provide the ratio from the parameter of Dv90 or thickness of the porous layer alone one would need both parameters to provide the ratio.”).

45. The prosecution history for the '987 patent does not provide any guidance on the technique to use to measure the Dv90 of the inorganic particles. The specification provides the measurement technique for the thickness of the porous layer. However, the specification and the prosecution history are silent on the measurement technique for the Dv90 of the inorganic particles. The prosecution history of the '987 patent fails to offer any description, examples, or explanation of appropriate techniques to measure the Dv90 of the inorganic particles.

B. Level of Ordinary Skill in the Art

46. I have been advised that the claim terms of the '987 patent are to be ascribed the meanings that they would have in the context of the patent as a whole to a person of ordinary skill in the field of the invention as of the time of filing. I understand that the '987 patent may have a priority date as early as April 11, 2018 (the filing date of Chinese Application 201810321779.5A). I, therefore, have considered the qualifications of a person of ordinary skill in the art before and around April 11, 2018, for the '987 patent.

47. I understand that in a related proceeding before the patent office, a level of the person of ordinary skill in the art for the '987 patent was offered, such that the person of ordinary skill in the art at the time of the alleged priority date in 2018 would have had at least a bachelor's degree or equivalent degree from an accredited institution in chemistry, chemical engineering, materials science, or a similar discipline covering relevant principles of organic and inorganic

chemistry, polymer processing, ceramic materials, and ceramic processing, as well as three or more years of experience in electrochemical energy storage devices, lithium-ion battery technology, separators, characterizations and analyses of particle size, film thickness, and/or materials compositions, battery performance characterization, or other applications of polymer chemistry, ceramic chemistry, and ceramic processing. More education could substitute for less experience, and vice versa.

48. I agree with this proposed level of ordinary skill in the art and have applied it in my analysis herein. It is consistent with my review of the patent, which concerns electrochemical energy storage devices and separators and the chemistry, chemical engineering, and materials science principles of organic and inorganic chemistry, polymer processing, ceramic materials, ceramic processing, characterizations and analyses of particle size and film thickness, materials compositions, and the characterization of battery performance. It is also consistent with the discussion of the invention in the prosecution history of the '987 patent and with references in the field, such as those discussed herein. This level of skill is also confirmed by my experience of the level of ordinary skill in the art from working and supervising those in the industry.²

² A higher level of skill would not change my opinions expressed herein, as individuals with greater levels of skill also understand the limitations and requisites of conducting particle size distribution analysis and measuring Dv90 of inorganic particles for separators. Likewise, a less experienced artisan would also not change my opinions, as that individual would still face the same problems in being unable to determine if a particular test result falls within or outside of the claim scope at the precision required by the '987 patent, even if that person does not understand *why* the variation between techniques and instruments exists.

C. Measurement Techniques for Particle Size Characterization and Distribution Techniques Can Provide Significantly Variable Results Preventing Comparisons Across Techniques and Instruments

49. Knowledge of particle sizes and size distributions is “a prerequisite” for most production and processing operations because particle sizes and size distributions can have a significant effect on the mechanical strength, density, electrical, and thermal properties of a finished object, such as a separator for lithium-ion batteries. Ajit Jillavenkatesa et al, NIST Recommended Practice Guide: Particle Size Characterization 1 (Spec. Publ. 960-1, 2001) [hereinafter NIST]. Particle size distributions are reported in various parameters, including Dv10, Dv50, and Dv90, to inform people of ordinary skill in the art of the characteristic particle sizes of the particles under evaluation. NIST 135-36. Particle sizes and particle size distributions can be measured by numerous commercially available instruments, which use different underlying physical principles. NIST 1-3. Dv90 is a parameter used for describing the particle size distribution of a material, including particle size distributions for inorganic particles used in separators for use in lithium-ion batteries.

50. The National Institute of Standards and Technology (NIST) published the NIST Recommended Practice Guide on Particle Size Characterization, noting that “[i]t is important to understand that as different instruments are based on different physical principles, there are bound to be differences in the size results obtained from these instruments. Furthermore, even when using instruments based on the same physical principle, manufacturers’ use of proprietary algorithms, components and variations in adaptations of the same basic physical principle can give rise to significant variations in the measured size. Thus, comparison of size and size distribution results from different instruments should be conducted with **extreme caution**.” NIST 3 (emphasis added).

51. To compare particle size and size distribution measurements across instruments, protocols may be developed by performing extensive analysis of suitable standards on each of the instruments. Many people of ordinary skill in the art “consider particle size and size distribution results as relative measurements, where results from one run can be compared with those from another, obtained on the same instrument under similar conditions.” NIST 3. Even if seeking to consider particle size and particle size distribution results as absolute measurements, the reliability of the measurements can be affected by, for example, the number of particles that are counted, the representative nature of the particles included in the analysis, the shape of the particles, the state of dispersion, the specimen preparation technique followed, the use of primary and secondary standards for calibration, and the material used as standards. NIST 3.

52. An ordinary artisan would have understood that the measurement techniques disclosed for particle size distributions, including Dv90 measurements and those techniques discussed in section D below, have significant variations in results between techniques and instruments. *See, e.g.,* Merkus at 14-15 (“In the real world, most particles are not spherical, but have different shapes and often rough surfaces. Materials of different chemical composition have different properties (e.g., density, conductivity, refractive index) and, even when the bulk composition is the same, different crystal structures or impurities may lead to different properties. These differences have a different influence in the principles that are responsible for appearance and behavior of particles, for example color and flow properties, but also behavior during sieving, sedimentation or light scattering. Consequently, **different PSDs will often result from different techniques.**”) (emphasis added). Accordingly, an ordinary artisan would understand comparing Dv90 results requires extensive research to develop protocols to allow for

an appropriate comparison. NIST 3. Indeed, “the same collection of particles may and often will yield different PSD’s coming from different measurement techniques.” Merkus 16.

53. An ordinary artisan in April 2018 would be aware of multiple measurement methods used for evaluating the Dv90 of inorganic particles in separators for use in lithium-ion batteries, including laser light diffraction techniques, centrifugal sedimentation optical and x-ray techniques, quasi-elastic light scattering techniques, scanning electron microscopy, light microscopy, etc. See NIST 1-2, Table 1.1; M. Li et al., *Comparison of Particle Size Distributions Measured Using Different Techniques*, 23 Particulate Science and Technology 265, 265 (2005) [hereinafter Li 2005]; X. Jia & E. Garboczi, *Advances in shape measurement in the digital world*, 26 Particuology, 19-20 (2016) [hereinafter Jia 2016].

Table 1.1. Particle Size Analysis Instruments Based on Different Physical Principles			
Instrumental Technique	Physical Principle	Volume Fraction Range (%)	Size Range(μm)
Acoustic Attenuation Spectroscopy (N) [Ultrasonic Attenuation Spectroscopy]	ultrasonics	> 1	0.05 to 10
Centrifugal Sedimentation-Optical (E)	sedimentation	ID	0.01 to 30
Centrifugal Sedimentation-X-Ray (E)	sedimentation	ID	0.01 to 100
Electrical Resistance Zone Sensing (E) [Particle Counting, Coulter Counter]	particle counting	< 0.1	0.4 to 1200
Electroacoustic Spectroscopy (N) [Electrokinetic Sonic Amplitude]	ultrasonics	> 1	0.1 to 10
Gas Absorption Surface Area Analysis (E) [BET Absorption]	surface area analysis	NA	NA
Laser Light Diffraction (E) [Static Light Scattering, Mie Scattering, Elastic Light Scattering]	electromagnetic wave interaction and scattering	0.01 to 5	0.04 to 1000
Light Microscopy (E)	particle counting	NA	> 1.0
Quasi-Elastic Light Scattering (E) [Dynamic Light Scattering, Photon Correlation Spectroscopy, Optical Beating Spectroscopy]		<0.005	0.003 to 6
Scanning Electron Microscopy (E)	particle counting	NA	>0.1
X-Ray Gravimetric Sedimentation (E)	sedimentation	ID	0.5 to 100
Colloid Vibration Current (N) [Single Frequency]	ultrasonics	> 1	<10
Electrokinetic Sonic Amplitude (N) [single frequency]	ultrasonics	> 1	<10
Microelectrophoresis (E) [Laser Light Scattering, Quasi-Elastic Light Scattering]	electromagnetic wave interaction and scattering	<0.001	0.1 to 1
Sieving (E)	sieving	NA	5 to 100,000
E: Established Technique N: Emerging Technique ID: Instrument Dependent Information compiled by V. A. Hackley, Ceramics Division, NIST			

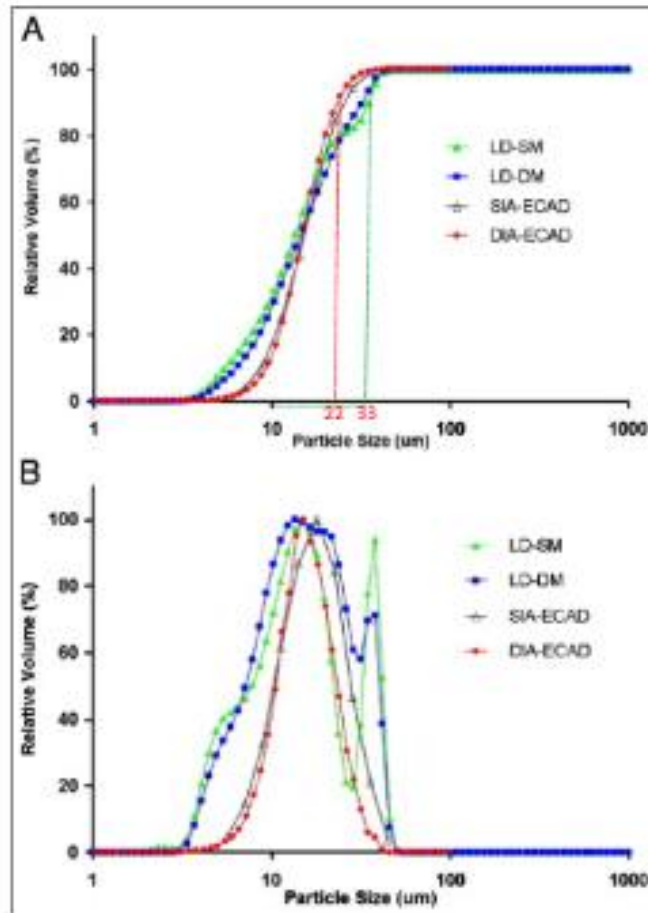
54.

NIST 2 at Table 1.1.

55. An ordinary artisan would have understood that these test methods are known to produce inconsistent results, with variations in results depending on several factors. Indeed, an ordinary artisan considers particle size distributions, including Dv90, as “relative” values as they require utilizing the same method, instrument, and parameters to be able to compare results. Even today, it is recommended “that users are consistent with whatever method is chosen for powder characterization, as there will be variation in data, regardless of the method type selected.” J. Grubbs et al, *Comparison of laser diffraction and image analysis techniques for particle size-shape characterization in additive manufacturing applications*, 391 Powder Technology 20, 33 (2021) [hereinafter Grubbs 2021]; see Whiting et al., *A comparison of particle size distribution and morphology data acquired using lab-based and commercially available techniques: Application to stainless steel powder*, 396 Powder Technology, 648, 648 (2022) [hereinafter Whiting 2022] (“It is, therefore, crucial to accurately know the PSD and morphology of these powders. There exist several measurement techniques for these quantities, but since each method is based on different physical phenomena, which are sensitive to different aspects of a particle's shape and size, **it is unclear how the measured PSDs and morphology compare to one another.**” (emphasis added)).

56. Materials with bimodal particle size distributions or irregular particle shapes, for example, can skew Dv90 results in significant manners. An ordinary artisan is aware of these limitations, see NIST 112, 128, 130, 135-36; Jia 2016 19-20, and reports details of particle size distribution testing, such as the testing method and instrument, to allow a person of ordinary skill in the art to obtain comparable results. See, e.g., NIST 125 (citing ASTM E1617-97) (standard requiring reporting all pertinent information relating to the material being analyzed, the instrument being used, and the methodology used for generating data, and the statistical

approaches for evaluating the data, where even NIST recognizes that the lowest Level I reporting requires “information about the basic measurement principle of the instrument used for analysis (physical principle such as sedimentation, laser light scattering or optical analysis)” to minimize errors that may arise during data interpretation (NIST 126-127)); (citing ASTM E1617-97) (requiring in 4.4.4 the reporting of principle of measurement for Level 1 and noting that “[o]ften, the measurement principle can be defined by describing the instrumentation or apparatus used for the analysis”; recognizing in 3.2.3 that “Level 3 provides detailed written procedures to allow duplication of the measurement”; and including “a complete description of any instrument operational and calibration procedures not specified by a published procedure” for Level 3 reporting in 4.6.3). For example, research on laser diffraction and image analysis techniques demonstrated a significant difference in Dv90 between the techniques and highlighted that rectangular particles can be detected as bimodally distributed by light scattering, while unimodal by imaging techniques.



57.

A.P. Tinke et al., *Particle*

shape and orientation in laser diffraction and static image analysis size distribution analysis of micrometer sized rectangular particles, 186 Powder Technology 154, Figure 8 (2008)

[hereinafter Tinke 2008] (annotations providing the approximate Dv90 of 22 and 33 added for comparison purposes, showing laser diffraction results in a Dv90 approximately 50% higher than that image analysis, largely because it detects the presence of a second distribution).

58. Given the significant differences in results between different measurement techniques, researchers have undertaken extensive research over many years in an attempt to correlate results—and such correlations require an understanding of the particular techniques used and reflect the underlying principle that a measurement from one particle size distribution cannot be carried over to another technique without applying an appropriate correlation, See,

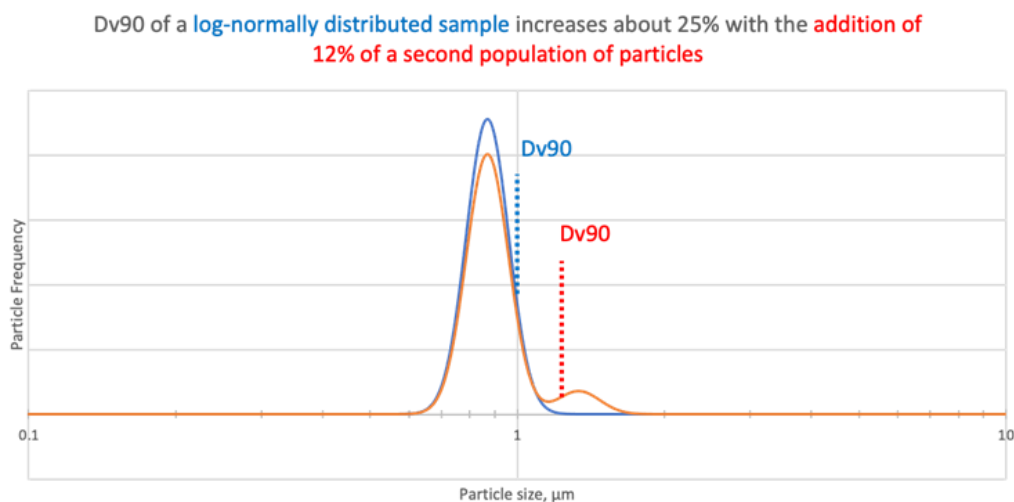
e.g., Tinke 2008 at 165 (“The equivalent circular area diameter (ECAD) is traditionally used in the comparison of LD and SIA data. However, based on the various examples discussed in this paper, we conclude that for micrometer-sized rectangular particles the scattering intensity weighted diffraction diameter (SIWDD) is a better basis for the correlation of SIA and LD data.”).

59. Thus, when inorganic particle materials have a bimodal distribution or irregular shape, which is not uncommon for the inorganic particle materials specified by the '987 patent, the reported Dv90 can be extremely sensitive to the technique and tool used. NIST 132-37; Tinke 2008 at 158-65. Furthermore, the Dv90 is inherently biased to a small fraction of material when there is not a unimodal distribution, and it is well established that different instruments and techniques calculate particle size inconsistently for non-spherical particles. NIST 132-37; Tinke 2008 at 154, 158-65; Li 2005 at 282 (“It is shown that particle shape strongly affects the PSDs measured by different techniques for nonspherical particle measurements. There is no consistent PSD result from different particle measurement techniques due to the influence of shape.”)³; *see* Grubbs 2021 at 27-28 (“It is known that LD-derived PSDs are greatly influenced by particle morphology, particularly when the typical particle shape deviates from the ideal spherical shape; as the particle’s morphology becomes more irregular, the LD system is more likely to center the PSD at slightly larger particle sizes, thus differing from the SIA measurements.”). However,

³ I note that an ordinary artisan would understand that the Dv90s reported in Table 5 were taken *after* the silica flake particles were sieved (and thereby subjected to the effects of sieving irregularly shaped particles discussed above), which account for some similarities in Dv90s in Table 5 as well as significant differences between others. I agree with the authors’ conclusion that there was no consistent particle size distribution result (including Dv90 results) from different particle measurement techniques.

significant differences between particle size distributions can also result from using different techniques for measuring spherical particles as well. Li 2005 at 277, Table 3.

60. Bimodal distributions, like those common for inorganic particles, can make using Dv90 challenging. To illustrate, I prepared the below figure showing a unimodal lognormal distribution⁴ (in blue) and bimodal distribution (in red). Both curves have the same total volume of particles, but they are distributed differently. The red bimodal distribution includes a small amount (12% volume fraction) of slightly larger particles, but has a significantly larger Dv90 (about 25% bigger). Thus, an ordinary artisan would understand that bimodal distributions can add uncertainty into Dv90 measurement techniques.



⁴ Log-normal is one of several size distribution functions commonly found for particles, and particle mixtures can also follow the form of a Rosin-Rammler Distribution or a Gaudin-Schuhmann (GS) Distribution. NIST 162. The choice to use one particular model over another can also impact the calculated values of Dv90 from a data set.

D. Measurement Techniques for Dv90 of Inorganic Particles

1. Laser Diffraction

61. Laser diffraction techniques are one class of particle size measurement techniques. Laser diffraction instruments fall within the category of non-imaging type of instruments, and the employed non-imaging methods of size determination can further be classified into numerous groups based upon the physical principle describing the interaction between the light beam and the particle and the information that can be deduced from that interaction. NIST 93-94.

62. The underlying assumption in the design of laser diffraction instruments is that the scattered light pattern formed at the detector of the instrument is a summation of the scattering pattern produced by each particle that is being measured in the sample. NIST 94. The deconvolution of the resultant pattern can generate information about the particle sizes and distributions based on the scattering pattern produced by each particle. NIST 94.

63. These instruments are suitable for a wide range of particle sizes and materials, including ceramics (such as the inorganic particles in the '987 patent) and ranges from 0.04-1000 microns. See NIST Table 1.1. Different mathematical solutions, or models, are used to solve the scattering problems and have different size limits associated. For example, Mie model typically can be used to analyze particles down to 0.1 microns, while the Fraunhofer model for size analysis typically has a lower size limit of 2 microns. NIST 94.

64. There are known limitations and errors associated with laser diffraction measurement techniques. For example, instrument performance and operation are highly dependent on instrument design (e.g., laser sources of different wavelengths, differing numbers and positions of detectors), such that comparison of results from different instruments may be

“misleading.” NIST 95. Laser diffraction measurement techniques may require a precise knowledge of optical properties of the sample being measured, cannot distinguish between dispersed particles and agglomerates, and “[s]ignificant error and bias may be introduced in the particle size distribution results, if powder particle shapes deviate from spherical configurations.” NIST 95.

65. Indeed, one study of ceramic particles found an almost 2x difference in reported Dv90s between different laser diffraction instruments (showing a difference of almost 2 μm). Naito et al., *Effect of particle shape on the particle size distribution measured with commercial equipment*, 100 Powder Technology 52, Fig. 2 (1998) [hereinafter Naito 1998] (annotations in red added).

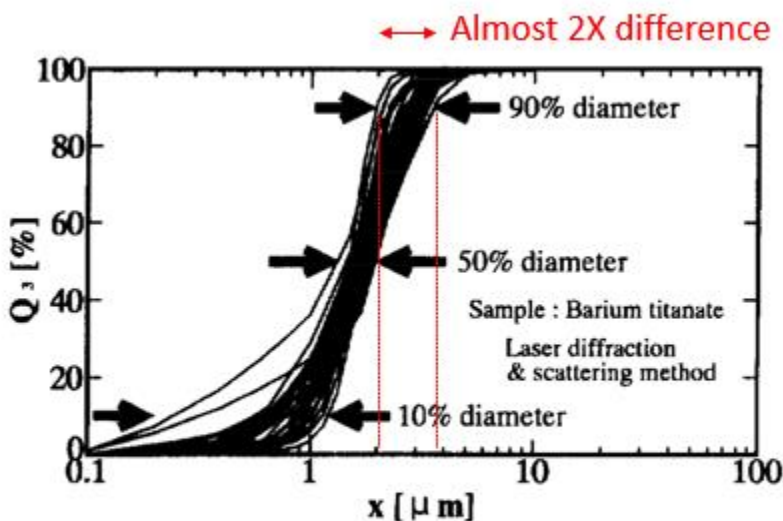


Fig. 2. Results of round robin for barium titanate measured by laser diffraction and scattering method. Eighty-four data measured by eight kinds of models are included in these results.

66. Another study demonstrated that different laser diffraction analysis instruments produced significantly different results on particles with elongated, rectangular geometries. Kelly et al, *Commercial Reference Shape Standards Use in the Study of Particle Shape Effect on Laser Diffraction Particle Size Analysis*, 7(2) AAPS PharmSciTech E1, Table 2 (2006) [hereinafter

Kelly 2006]. From the data reported on then-commercially available standards, it can be seen that variations of over 70% in reported Dv90 can be identified simply by changing measurement tool, and a variation of over 30% can be seen when using the same laser diffraction tool in a different mode (AEA 1003, Malvern Mastersizer). Kelly 2006 at Table 2. Thus, a person of ordinary skill in the art would have understood laser diffraction particle size analysis can produce significantly different results between instruments when considering nonspherical particles, or within the same instrument when changing methods. An ordinary artisan would have been especially cautious about making precise comparisons between laser diffraction particle size analysis techniques and measurement results from other particle size analysis techniques.

Table 2. Intermanufacturer LD psa AEA 1001 to AEA 1003 Measurement Results Comparison*

Sample ID	AEA 1001				AEA 1002				AEA 1003			
Physical dimensions (μm)	3 × 1.7 × 1				7.5 × 1.7 × 1				12 × 1.7 × 1			
Equivalent spherical volume diameter (μm)	2.14				2.90				3.39			
Equivalent circular area diameter (μm)	2.55				4.03				5.10			
Statistics: Volume Diameters (μm)												
LD psa system ID conditions	d10	d50	d90	d(4/3)	d10	d50	d90	d(4/3)	d10	d50	d90	d(4/3)
Beckman Coulter LS 13 320; RI = 1.54 for 3 μm	1.68	2.46	3.76	2.61	1.83	4.00	6.53	4.78	1.58	5.33	13.31	6.51
Horiba LA-920	2.29	2.55	3.26	NC	3.34	4.05	5.31	NC	NM	NM	NM	NM
Malvern Mastersizer2000 general purpose mode	1.78	2.94	5.59	3.36	NM	NM	NM	NM	2.23	4.60	10.75	5.66
Malvern Mastersizer 2000 single narrow mode (spherical)	NM	NM	NM	NM	2.02	5.23	6.76	4.71	2.07	5.34	12.71	6.48
Malvern Mastersizer 2000 single narrow mode (spherical) flow stop	NM	NM	NM	NM	NM	NM	NM	NM	2.25	5.26	9.93	5.69
Malvern Mastersizer 2000 multiple narrow mode	NM	NM	NM	NM	NM	NM	NM	NM	2.00	4.96	12.42	6.13
Malvern Mastersizer S polydisperse	NM	NM	NM	NM	1.49	4.07	9.43	5.21	1.53	3.90	11.48	5.40

67.

Kelly 2006 at Table 2 (annotations added).

68. The authors also noted that “The study has also highlighted the need for additional shape standards of varying geometry that are affordable and available in a variety of monosize and polydisperse wet and dry preparations to allow the simulation studies required in the case-by-case determination of the appropriateness of the quality control application of LD psa systems,” which further underscores the need of the skilled artisan to know the technique and

precise standard materials used in order to compare results between different instruments and different techniques. Kelly 2006 at E11.

2. Image Analysis Techniques

69. Microscopy-based techniques, including via light microscopes (optical light microscopes), scanning electron microscopes (SEM), and transmission electron microscopes (TEM) for particle size characterization are another class of techniques for particle size distribution measurements. These techniques involve direct observation of particles and the resultant determination of size based on a measure of the particle diameters. “Typically, the calculated sizes are expressed as the diameter of a sphere that has the same projected area as the projected image of the particle,” and these techniques provide the ability to determine the particle shape, in addition to making a direct measurement of size. NIST 69. Numerous commercial instruments and software packages are available for particle size and size distribution analysis, including some commercial instruments that are sold as complete packages including the microscope, accessories, dedicated computer systems and software algorithms for analysis and some packages that are available as software add-ons to existing microscopes. NIST 69. Different instrument packages have various functionality, where some are limited to size determination of well-dispersed particles, and others have the ability to discern individual particle sizes and shapes from clusters and agglomerates. NIST 69.

70. These techniques and instruments are suitable for a wide range of particle sizes and materials, including ceramics (such as the inorganic particles in the '987 patent). See NIST 70; *see also* NIST Table 1.1. Optical microscopes are typically more limited in magnification and resolution, which limits most reproducible analysis by optical microscopy to approximately 1 μm particles with a theoretical resolution limit of about 0.2 μm (dependent upon wavelength of

illumination source). “Scanning electron microscopes enable examination of details at higher magnification and resolution, and are thus suitable for particles in the size range of about 0.1 μm to 1000 μm (the lower limit depending upon the quality of the instrument being used),” and “Transmission electron microscopes enable analysis in the size range of 0.01 μm to 10 μm , retaining suitable resolution to enable distinguishing surface characteristics.” NIST 70.

71. There are limitations and errors associated with microscopy-based measurement techniques. For example, size analysis by microscopy-based techniques can lead to erroneous results if sampling and sample preparation does not result in a sample representative of the bulk powder being measured. NIST described it as “absolutely essential that the sample being analyzed is representative of the bulk of the powder” and explained that, while sampling error needs to be avoided in any size analysis technique, “it has an even greater implication, when sample size gets smaller,” as is the case with microscopy-based measurement techniques. Indeed, “any bias towards either fine particles or coarse particles in the observed field will tend to shift the calculated size and size distribution in that direction,” and these shifts are a significant problem for most real-world systems, where particles tend to have a broad size distribution. NIST 72. NIST further recognized that “Often, particle settling or agglomeration may cause particles of a particular size fraction to settle at the bottom, or to be present in a disproportionate amount in the specimen, leading to a bias in the counting and thus a skewed result.” NIST 72. Accordingly, the sample must be well dispersed, the sample must be representative of the material being studied, and a large number of particles should be counted to control statistical errors due to inadequate representation. NIST 72.

72. The statistical errors typically encountered when using microscopy-based techniques are due to counting a disproportionately larger fraction of coarse or fine particles than

is actually present in the material as a whole, giving rise to a bias towards that fraction. To avoid these errors and biases, a certain number of particles need to be counted, and that number of particles can be estimated by mathematical theories to keep the statistical error within a desired range. As one example, the NIST authors estimated for a powder with certain specified powder distribution properties that approximately 3400 particles need to be analyzed if it is desired that 99% of the data should be included in the relative error of $\pm 5\%$. NIST 75-76. The mathematical theories provide some guidelines for estimating the number of particles that need to be analyzed in order to have results with some degree of confidence, but the assumptions that have been considered in developing the mathematical theories “do not always hold good for all ‘real’ powder distributions and the user should have a good estimate of the values of terms such as [the value of the non-zero exponential constant defining the powder distribution] and [the variance of the powder distribution] of the powder distribution.” NIST 75-76.

73. Other commonly encountered sources of error for microscopy-based techniques include (1) errors associated with specimen/sample preparation (e.g., nonrepresentative sampling of powders, inadequate or improper dispersion of powders during specimen preparation, agglomeration or fracture of primary particles during specimen preparation), (2) errors associated with instrumentation/equipment and image analysis (e.g., counting of inadequate number of particles, improper calibration of microscope magnification, microscope operation under less than ideal parameters, charging of specimen surface while observing under an electron beam, operating microscope in regions of “empty magnification” where there is no gain in resolution beyond a certain magnification limit, improper calibration of image analysis hardware/software, errors in establishing gray scale threshold in image analysis techniques), and (3) errors due to human and other factors (e.g., operator fatigue, operator skill level and experience in use of

equipment and techniques associated with analysis, lack of understanding of equipment and software/hardware capabilities and limitations, errors due to inadequate or incorrect statistical analysis. NIST 85-86.

74. A thorough understanding of these various sources of common errors “is essential in order to be able to identify the occurrence of these and develop procedures to minimize their impact on the final results.” NIST 85. NIST recognized that “[i]t is also necessary to understand that while these errors cannot be eliminated, implementation of standard procedures and protocols can help characterize the errors and the magnitude of their influence on the measured results” and that successful procedures are “simple and lead to reproducible results.” NIST 85-86.

75. A person of ordinary skill in the art in April 2018 would also have understood that “[o]ther important concerns arise when analyzing irregularly shaped particles or particles with a significant aspect ratio” because “[i]rregularly-shaped particles may come to rest on the sample stub in a preferred orientation and may thus bias the analysis.” If such orientation of particles is unavoidable, “procedures should be designed to factor in such a bias in the results.” NIST 86.

76. Error associated with microscopy-based analysis measurement techniques results in significantly different values for Dv90 results, including for irregularly-shaped particles (such as irregularly-shaped inorganic particles in the '987 patent) and where parameters are not set to ensure reproducibility. NIST 86. For example, one study found substantial differences in calculated Dv90 between dynamic (Camsizer) and static (MIA) optical microscopy analysis, as shown in the table below. Kumar Patchigolla and Derek Wilkinson, *Crystal Shape Characterisation of Dry Samples using Microscopic and Dynamic Image Analysis*, 26 Particle & Particle Systems Characterization 171, 176-78 (2009) [hereinafter Patchigolla 2009].

Table 2: Median size and span of comparison using microscopic and dynamic image analysis.

Material	Sieve range		MIA	Camsizer
α -glutamic acid	$180 < d \leq 250 \mu\text{m}$	$D_{50}, \mu\text{m}$	280.7	322
		$D_{10}, \mu\text{m}$	215.8	254
		$D_{90}, \mu\text{m}$	305.1	441
		Span	0.32	0.579
		Aspect ratio	1.18	1.39
β -glutamic acid	$90 < d \leq 180 \mu\text{m}$	$D_{50}, \mu\text{m}$	345.8	393
		$D_{10}, \mu\text{m}$	231.8	177
		$D_{90}, \mu\text{m}$	430	767
		Span	0.57	1.501
		Aspect ratio	6.25	2.53
MSG	$180 < d \leq 250 \mu\text{m}$	$D_{50}, \mu\text{m}$	423.5	410
		$D_{10}, \mu\text{m}$	303	274
		$D_{90}, \mu\text{m}$	516.9	632
		Span	0.51	0.873
		Aspect ratio	3.36	1.91

Patchigolla 2009 at Table 2 (annotations added).

77. The challenge of collecting statistically relevant populations of particle sizes from SEM data is not generally solved. For example, see the discussion of “Workflow towards automated segmentation of agglomerated, non-spherical particles from electron microscopy images using artificial neural networks” by Rühle et al., which states “electron microscopy techniques yield images from which the shape and size of individual nanoparticles (or at least their 2D projections) can be extracted directly to yield a number-based particle size distribution. This advantage is however counterbalanced by the reduced number of particles that can be imaged simultaneously, and especially the tedious and usually manually performed extraction of the size and shape descriptors of the nanomaterials from these images. To mitigate these drawbacks, it is highly desirable to use automated image analysis, rather than a tedious manual

measurement of individual particles that is prone to errors and operator bias in order to get representative particle size and shape descriptors for a statistically relevant number of nanoparticles.” 11 Scientific Reports 4942 (2021) [hereinafter Rühle 2021]. The paper represents the state of the art of such techniques in 2021, after the priority date of the '987 patent. Even at this time, such technology was not yet standardized for industrial use, merely demonstrated in a research lab.

3. Gravitational and Centrifugal Sedimentation Techniques

78. Another class of measurement techniques, gravitational and centrifugal sedimentation techniques, including optical and x-ray techniques, determine particle size by measuring the velocity at which particles settle in a fluid, after applying gravitational or centrifugal forces. NIST 49-52, Table 1.1. These techniques, and the instruments used to apply them, are some of the most commonly used in industrial and research applications to determine particle size and particle size distribution. NIST 49.

79. These techniques are suitable for a wide variety of powder systems and rely on knowing the density of the particles in the powder system. NIST 50-51. One requirement for this measurement technique is that the particles in the powder system have the same density. NIST 50-51.

80. X-ray centrifugal sedimentation techniques use an x-ray beam to determine the concentration of particles at a given depth in the fluid based on x-ray absorption. NIST 50. Optical centrifugal sedimentation techniques determine the concentration of particles at a given depth in the fluid based on the scattering of a light beam. NIST 50. These techniques use the concentration of particles at a given depth and the known particle density to determine the

settling velocities of the particles. NIST 50. Particle size by diameter is then calculated from the corresponding particle settling velocities. NIST 50.

81. These instruments are suitable for a wide range of particle materials and sizes, including ceramics (such as the inorganic particles in the '987 patent) and ranges from 0.01 μm - 100 μm . See NIST 50-52, Table 1.1.

82. There are known limitations and error associated with gravitational sedimentation measurement techniques. For example, instruments based on centrifugal sedimentation optical and x-ray techniques report particle sizes and distributions in terms of the equivalent spherical diameter. NIST 51. This means that particle sizes and distributions reported by the instrument become increasingly nonrepresentative as the aspect ratio of non-spherically shaped particles increases. NIST 51-52, 58-61; Naito 1998 at Fig. 5. These techniques should not be relied upon alone unless the sample contains only spherical particles. NIST 61.

83. Additional limitations include the need to use samples with particles of uniform density having a density significantly different from the density of the dispersion medium and the sensitivity and accuracy depending on the particle mass absorption coefficient (for x-ray techniques) and optical constants used (for optical techniques).

84. Error associated with gravitational sedimentation measurement techniques results in significantly different values for Dv90 results for irregularly shaped particles, such as for those irregularly-shaped inorganic particles of the '987 patent. Naito 1998 at Fig. 5.

4. Sieving

85. "Sieving is one of the oldest techniques of powder classification (i.e., separation based on size or some other physical characteristic) that is still in use today. It is among the most

widely used and least expensive techniques for determination of particle size distribution over a broad size range.” NIST 27.

86. “The method, in the most general sense, consists of shaking (agitating) the sample through a stacked series of sieves with decreasing mesh size. The mesh with the largest aperture is at the top, and that with the smallest aperture is at the bottom of the stack. Size distribution is reported as the mass of the material retained on a mesh of given size, but may also be reported as the cumulative mass retained on all sieves above a mesh size or as the cumulative mass fraction above a given mesh size.” NIST 27.

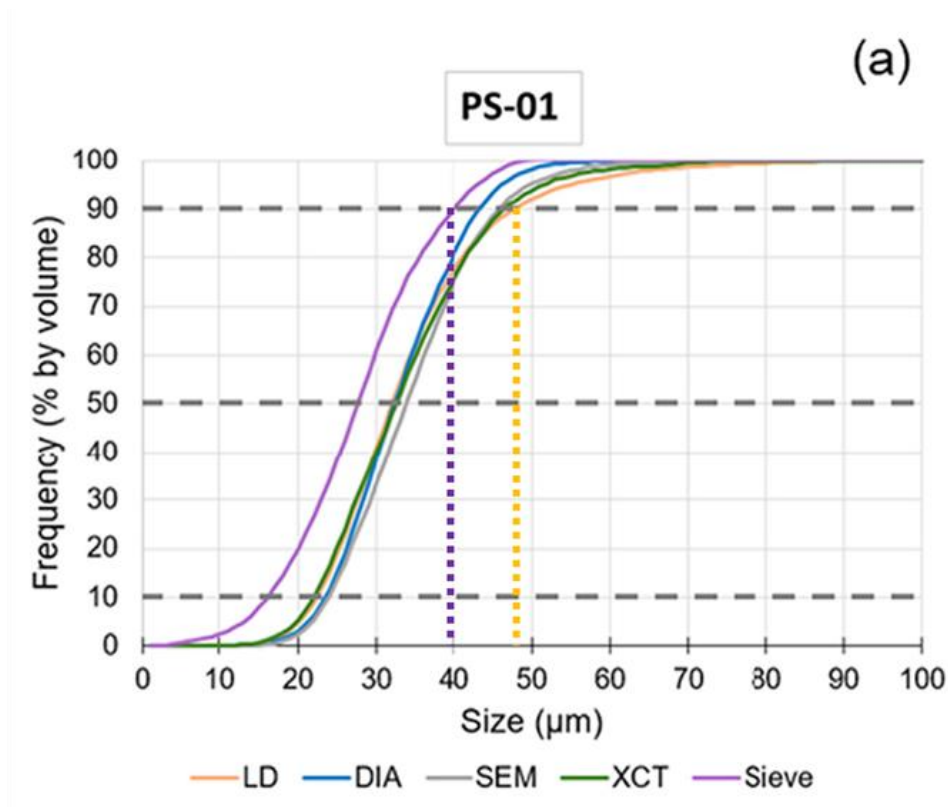
87. These instruments are suitable on a wide range of particle sizes and materials, including ceramics (such as the inorganic particles in the '987 patent) and ranges from about 125 mm to 20 μm . See NIST 29, Table 1.1.

88. There are known limitations and error associated with sieving measurement techniques, and “[i]t has been observed that the coefficient of variance may vary between 10 % for sieves with small aperture widths and 5% for sieves with larger aperture widths.” NIST 28. For example, sources of error and variations can arise during the sieving process due to inadequate and/or improper maintenance of sieves, during/due to material transfer to and from sieves, and due to powder characteristics that may cause particles to agglomerate or fracture during analysis. “Significant bias may be introduced during analysis of acicular [(needle shaped)] particles.” NIST 29.

89. NIST recognized that “repeatability and reproducibility of results” may be affected by the mechanical motion during sieving, and that “[w]hen particles have high aspect ratios (e.g., needle shaped particles), PSDs are susceptible to large uncertainties due to measurement of smallest diameter by this method.” NIST 29; *see also* Merkus 14-18, Table 2.1.

90. Sieve diameters for nonspherical particles with aspect ratios close to 1 generally underestimate particle size because particles with aspect ratios close to 1 pass through the sieve by their smaller dimension. Merkus 14-18, 229. For nonspherical particles with significant aspect ratios, “it is much more difficult, if not impossible, to reach such optimum positions” and the largest dimension may determine the measurement reported by sieving, overestimating particle size. Merkus 229; *see also* Merkus Table 2.1.

91. Error associated with sieving analysis measurement techniques results in significantly different values for Dv90 results for irregularly-shaped particles, such as for those irregularly-shaped inorganic particles of the '987 patent. Elongated particles in particular produce results when using sieves that differ substantially from results from other techniques, as the particles are able to fall through the holes in the sieve along the elongation axis, akin to rice falling through the holes of a colander. Whiting 2022, for example, reported Dv90 results for irregular particles that were about 20% lower than the results from laser diffraction.



92.

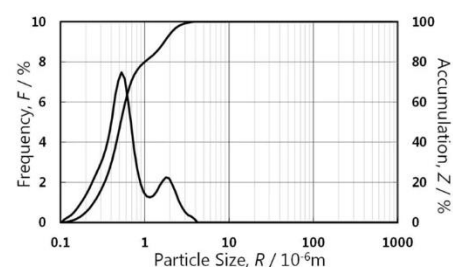
Whiting 2022 at Fig. 7(a) (annotated with purple and yellow dotted lines).

5. Inorganic Particle Materials

93. Inorganic materials may be unimodal, bimodal, and have irregular shapes that impact Dv90 measurements. For example, commercially available magnesium oxide particles may exhibit a bimodal distribution with cubic geometry, and have a Dv90 of about 1.5 μm .

(B) Primary particle

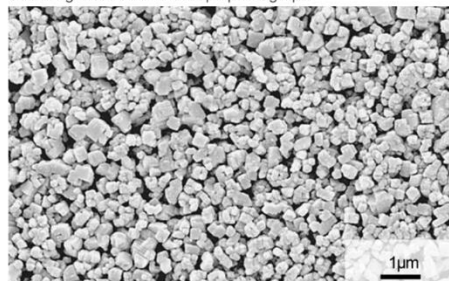
Particle Size Distribution (Typical)



Measurement method: Laser diffraction scattering method [Strong Dispersion]

*The properties are the typical values and not the guaranteed values.

Scanning Electron Microscope photograph

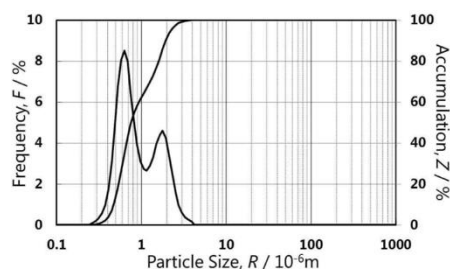


<https://tateho-chemical.com/products/puremag.html>.

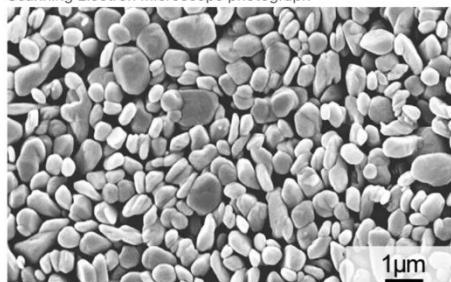
94. For another example, commercially available magnesium hydroxide particles may exhibit a bimodal distribution with plate geometry and a Dv90 of 2 μm .

(B) Primary particle

Particle Size Distribution (Typical)



Scanning Electron Microscope photograph

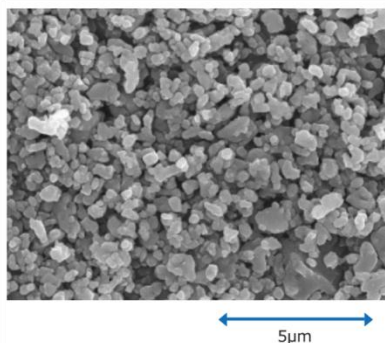
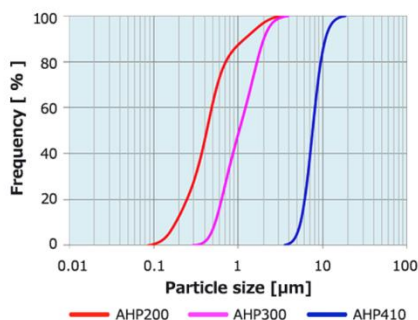


Measurement method: Laser diffraction scattering method [Strong Dispersion]

*The properties are the typical values and not the guaranteed values.

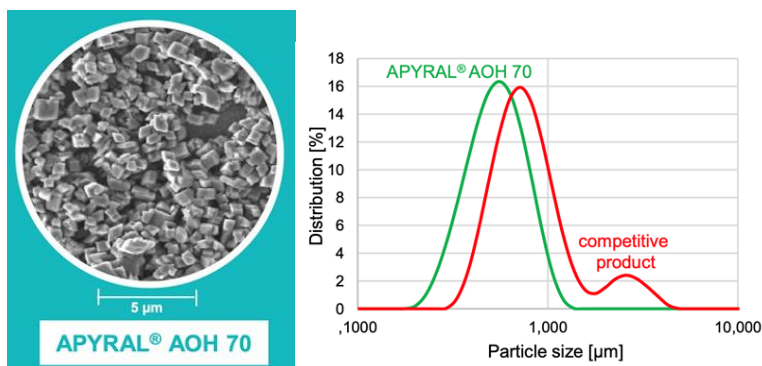
<https://tateho-chemical.com/products/puremag-fnm.html>.

95. For another example, commercially available alumina particles may have irregular geometry and a Dv90 of e.g., 1 μm .



<https://www.nikkeikin.com/highpurityalumina/ahp200.html>.

96. Commercial boehmite (aluminum oxide hydroxide) particles come in a variety of morphologies, even from the same vendor. For example, the German company Nabaltec offers boehmite powder for battery separator applications of a specific grade Apyral AOH 70. The company specifically advertises that their material is unique in not having a bimodal particle distribution, which they consider an advantage over other competing commercial products.



https://nabaltec.de/fileadmin/user_upload/03_produkte/03-3_boehmit/TDS_Boehmite_LIBS.pdf.

97. Method of preparation of the inorganic particles can impact the particle geometry or shape. For example, aluminum hydroxide may have various plate-like geometries or rod-shaped geometry.

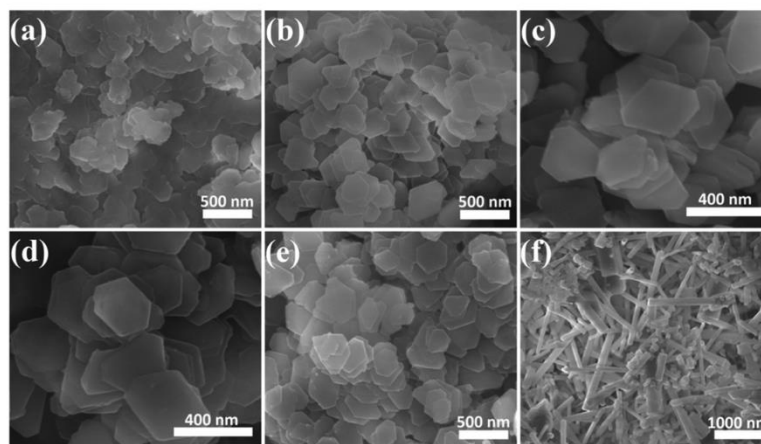


Figure 5. SEM images of as-prepared gibbsite at different pH values: (a) 1; (b) 3; (c) 5; (d) 10; (e) 12; and (f) 13.5.

Zhang et al., *Fast Synthesis of Gibbsite Nanoplates and Process Optimization using Box-Behnken Experimental Design*, 17 (12) *Crystal Growth & Design* 6801, 6803, 6806 (2017) [hereinafter Zhang 2017].

98. Additionally, in my experience as one in the art at the time, commercially available products before the asserted priority date had various morphologies as is still the case in commercially available products today. In 2018, a person of ordinary skill in the art would

have understood that the inorganic particles in the '987 patent could have various geometries, including plate-like, rod-like, and irregular shapes, and have bimodal distributions wherein small changes to a product can dramatically impact Dv90.

E. “Dv90,” “Dv90 of Inorganic Particles,” and “Ratio of Dv90 of the Inorganic Particles to the Thickness of the Porous Layer” Render the '987 Claims Indefinite

99. I have been asked to consider whether the '987 patent claims, when read in light of the patent's specification and its prosecution history, “inform with reasonable certainty those skilled in the art about the scope of the invention.” For the reasons summarized in this declaration, after analyzing the '987 patent, its claims, the specification, and its prosecution history (including the prior art references cited therein), it is my opinion that uncertainty in how to measure “Dv90,” “Dv90 of inorganic particles,” and “a ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.3 to 3.0” prevents the claims from informing those skilled in the art of the claim's scope with reasonable certainty. The claims are, accordingly, indefinite.

100. The '987 claims provide no method for measuring Dv90. Claim 1, for example, recites that “a ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.3 to 3.0” but specifies no measurement methodology, testing standard, or instrument for determining Dv90. As discussed above, ordinary artisans at the time of the '987 filing would be aware that different test methodologies and instruments could lead to differing Dv90 measurements. See NIST 112, 128, 130, 135-36. For this reason, discussions of particle size distributions typically report how testing was performed, such as the testing method and instrument, to allow a person of ordinary skill in the art to obtain comparable results. *See, e.g.*, NIST 125 (“Standards for the Reporting of Particle Size Data”). Industry standards, like the

ASTM E1617-97 standard, require reporting all pertinent information relating to the material being analyzed, the instrument being used, the methodology used for generating data, and the statistical approaches for evaluating the data. *See* ASTM E1617-97 3.2.3 (“Level 3 provides detailed written procedures to allow duplication of the measurement”); *see also* ASTM E1617-09 (2014) (requiring same); ASTM E1617-09 (2019) (requiring same). And even the lowest Level I reporting requires “information about the basic measurement principle of the instrument used for analysis (physical principle such as sedimentation, laser light scattering or optical analysis)” to minimize errors that may arise during data interpretation. NIST 126-127; ASTM E1617-97 (“4.4.4 The report shall include the principle of measurement, such as sieving, sedimentation, light blockage, electronic zone sensing, laser light scattering, and so forth. Often, the measurement principle can be defined by describing the instrumentation or apparatus used for the analysis.”); *see also* ASTM E1617-09 (2014) (requiring same); ASTM E1617-09 (2019) (requiring same). The ’987 claims provide no such information. This lack of guidance in the ’987 claim language would, in my opinion, leave the ordinary artisan unable to determine the scope of the claims, particularly at the upper and lower ends of the claimed range of the ratio of Dv90 of the inorganic particles to the thickness of the porous layer.

101. The ’987 specification does not cure these deficiencies. The specification defines “Dv90,” but does not tell the ordinary artisan how it should be measured. In three places, the specification repeats that “[t]he Dv90 of the inorganic particles refers to a particle size which reaches 90% the cumulative volume from the side of small particle size in the granularity distribution on a volume basis.” ’987 patent, 1:53-56, 3:63-66, 5:22-25. In my opinion, this definition does not tell the ordinary artisan how to assess the scope of the claims. The definition does not specify a measurement technique or commercially available instruments for performing

the measurement, as commonly done by those in the art so that “experiments conducted to generate this information can be reproduced when necessary.” NIST 125. The ’987 definition also fails to provide the basic information required by the ASTM and NIST-recognized standards described above. NIST 126-127. Knowing that the different measurement techniques can lead to “significant variations in the measured size,” the ordinary artisan would require more information to assess the scope of the ’987 claims, and particularly the claims’ boundaries.

102. The ’987 specification provides no other description of the definition or measurement of “Dv90.” Nor does it report any measured Dv90 values, let alone the techniques used to make these measurements. This again departs from best practice in the scientific literature relating to particle-size distribution: NIST confirmed that “It is a good practice to represent the results of the particle size distribution as both a cumulative finer than and fraction distribution graph as a function of the observed particle size. This enables the user to observe the nature of the distribution (unimodal, bimodal, etc.) the fraction of the distribution in a particular size range of interest, and be able to determine the size corresponding to a particular fraction (d10, d50 or d90, etc.).” NIST 111. The ’987 patent fails to provide this information.

103. As summarized in the chart below, the absence of information provided by the ’987 patent regarding particle size measurements contrasts starkly with the detail it provides regarding the measurement of thickness. This illustrates, in my opinion, the inadequacy of specification’s description of Dv90 measurement techniques:

	’987 Disclosure for Thickness Measurement	’987 Disclosure for Dv90 Measurement
Instrument:	“Model VL-50 LITEMATIC from Naitutoyo company”	None
Measurement technique:	“using a 5 mm flat bottom probe with a speed of 50 mm/min and a pressure of 0.01N”	None
Number of Samples:	“[e]ach of the separator samples . . . is measured for 60 thickness points”	None

Numerical Analysis:	“average thickness is taken as the measured value” and “thickness of the porous layer is equal to the thickness of the separator minus the thickness of the porous substrate”	None
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104. As summarized in the chart above, for thickness, the '987 patent specification describes a commercially available instrument used to make the measurement, the measurement speed and probe characteristics, the number of samples taken, and the numerical analysis performed to derive a measurement. '987 patent 13:35-45. The specification also details test protocols, instrumentation, and numerical analysis for gas permeability and porosity measurements. '987 patent 13:46-14:3. But the '987 patent provides ***no corresponding information*** about the measurement of Dv90. This omission, in my opinion, would leave the ordinary artisan at the time of the '987 patent's asserted priority date in 2018 with little guidance as to how Dv90 was to be measured when assessing the patent's claims.

105. Multiple measurement techniques would be available to the ordinary artisan for determining the particle size distribution. *See supra* Sections IV.C-D. By virtue of the disclosed ranges of exemplary thickness and exemplary ratios of Dv90 to thickness, the '987 patent discloses that exemplary Dv90s may be in a range of 0.06 μm to 30 μm . *See supra* ¶133, '987 patent, claims 1, 6; '987 patent, 2:1-6, 4:30-36. Reference materials available to those in the art identify multiple techniques appropriate for particles this size. NIST 1-2, Table 1.1; Merkus 142-46. These include, for example: laser light diffraction techniques, centrifugal sedimentation optical and x-ray techniques, quasi-elastic light scattering techniques, scanning electron microscopy, light microscopy, etc. NIST 1-2, Table 1.1; Merkus 142-46. The ordinary artisan would know, in my opinion, that these different categories of particle size measurement techniques, based on different principles, will give different reported values. Indeed, even instruments based on the same principle would give significant variations in the measured sizes

due to different adaptations of the same principles by different manufacturers. Naito 1998 at Fig. 2. Thus, as explained by publications by the National Institute of Standards and Technology, particle size is best presented as “relative measurements, where results from one run can be compared with those from another [] obtained on the same instrument under similar conditions.” NIST 3. The ’987 patent’s lack of basic information about measurement techniques prevents this best-practice approach.

106. It is my opinion that the ’987 patent’s lack of guidance on measurement techniques would make it difficult for ordinary artisans to determine whether particular product samples were within or outside of the patent’s claims. The ’987 patent’s specification discloses 17 examples, 15 of which are described as practicing the claims and two of which the inventors identify as “comparative.” The two comparative examples have ratios of Dv90 to porous layer thickness **just outside** of the claimed range of 0.3-3.0. The specification distinguishes these comparative examples—just outside of the claimed range—from those just within the range, stating that “[a]s can be seen from a comparison among examples 1-7 and comparative examples 1-2, the ratio of Dv90 of the inorganic particles to the thickness of the porous layer has an effect on the rate performance and cycle performance of the lithium-ion battery.” ’987 patent, 15:26-30. The ’987 patent provides that if “the ratio of Dv90 of the inorganic particles to the thickness of the porous layer is too high (for example, comparative example 2), the pore size distribution of the porous layer tends to be non-uniform, the surface of the porous layer is heterogeneous, the porous layer is easily detached from the surface of the porous substrate, and defects are formed in the energy storage device (such as a lithium-ion battery).” ’987 patent, 15:36-43. The specification also provides that “[i]f the ratio of Dv90 of the inorganic particles to the thickness of the porous layer is too low (for example, comparative example 1), the inorganic particles

cannot be effectively exposed on the surface of the porous layer . . . [which] is also not advantageous for the improvement of the rate performance and cycle performance.” ’987 patent, 15:49-59. In my opinion, an ordinary artisan would encounter trouble distinguishing the practicing examples from the comparative examples. Comparative example 1 has a Dv90 of 0.4 μm , just 0.2 μm smaller than the Dv90 of practicing example 1. Comparative example 2 has a Dv90 of 6 μm , the same as the Dv90 of practicing example 7 when accounting for significant figures. Even if significant figures are disregarded, comparative example 2 has a Dv90 of 6.4 μm , just 0.4 μm larger than the Dv90 of practicing example 7.

Table 1 – Dv90 calculation based on measurements disclosed for the ratios and thicknesses

	Example 1	Comparative Example 1	Example 7	Comparative Example 2
Ratio of the Dv90 of the inorganic particles to the thickness of the porous layer	0.3	0.2	3.0	3.2
Thickness of porous layer (μm)	2	2	2	2
Dv90 of the inorganic particles (μm) ⁵	0.6	0.4	6	6

107. When comparing example 7 (with measurements asserted to be inside the claimed ranges) and comparative example 2 (with measurement asserted to be outside the claimed ranges), the use of different measurement techniques, sampling techniques, or instruments could result in errors significantly greater than the difference between the reported results. Indeed, as shown in Table 1 above, an ordinary artisan would calculate the Dv90 for example 7 and comparative example 2 to *both* be 6 due to the level of precision of the thickness measurement. Even if significant figures are disregarded, the difference between calculated Dv90s for these examples would likely be less than variations that occur between different Dv90 techniques. For

⁵ Calculated Dv90 values using the claimed ratio and thickness where:

Ratio of the Dv90 of the inorganic particles to the thickness of the porous layer = (Dv90 of the inorganic particles) / (thickness of the porous layer);

Dv90 of the inorganic particles = (Ratio of the Dv90 of the inorganic particles to the thickness of the porous layer) * (thickness of the porous layer);

Thus, e.g., Dv90 of the inorganic particles_{example 1} = 0.3 * 2 = 0.6

example, the deviation in Dv90s of 6.7% (and only 0.4 μm total difference) between Dv90s of 6.0 μm in example 7 and 6.4 μm in comparative example 2 (i.e., $(6.4-6.0)/6.0$) is significantly less than the almost 2x difference reported between different laser diffraction instruments in the Naito study on plate-like inorganic materials, the approximately 50% difference between laser diffraction and static image analysis conducted by Tinke on those rectangular particles, or the approximately 20% difference between spherical silica glass beads that were in the 20-36 μm by sieving by Li. Naito 1998 at Fig. 2; Tinke 2008 at Fig. 8; Li at 277, Table 3. Thus, a person of ordinary skill in the art measuring the Dv90 of the inorganic particles in example 7 or comparative example 2 would not know whether the particles fall inside or outside of the claim depending on the measurement techniques used. In my opinion, a purported 6.7% deviation (if significant figures are disregarded, otherwise there is no reported deviation) between particle sizes that would render a ratio inside or outside of the claims seems small and outside the range of what an optimistic operator would suggest is real. Indeed, even a 10% error between measurement techniques could bring a result from a certain example inside or outside of the claim (e.g., Dv90s inside at examples 1 and 7, or outside at the comparative examples 1 and 2). As discussed above, different measurement techniques, and even different instruments utilizing the same general technique or the same instrument run in different modes, can exhibit variations in particle size distributions and Dv90 up to 2x (100%).

Table 2 – recreated based on Tables 1 and 2 of the '987 patent

		Example 7	Comparative Example 2	Example 1	Comparative Example 1
Solid content in the coating solution		15%	15%	15%	15%
Types of inorganic particles		boehmite	boehmite	boehmite	boehmite
Ratio of the Dv90 of the inorganic particles to the thickness of the porous layer		3.0	3.2	0.3	0.2
Volume ratio of the inorganic particles to the binder		0.8	0.8	0.8	0.8
Coagulating solution	second solvent	NMP	NMP	NMP	NMP
	content of second solvent	40%	40%	40%	40%
Porosity of porous layer		57%	60%	48%	45%
Gurley value of porous layer (s/100 cc)		18	16	53	70
Thickness of porous layer (μm)		2	2	2	2
2C / 0.2 C rate performance		80.40%	71.30%	79.50%	69.80%
Capacity retention rate after 200 cycles		82.70%	72.60%	85.70%	70.50%

108. In contrast, literature in the field, both before and since the filing of the '987 patent, discloses the measurement methods and techniques, particle shapes, measurement procedures (such as dispersion solvent and method), and the measurement data of Dv90. Grubbs 2021 at 21-22; Li 2005 at 282-83; Tinke 2008 at 154-65; Naito 1998 at 58-61; Kelly 2006 at E11; Whiting 2022 at 654-62; NIST 111. For example, Saeki, cited by the examiner during prosecution discloses several measurement methods and techniques, including using the

Microtrac MT 3300 EX laser particle size distribution analyzer and “using a particle diameter measuring device (MICROTRACTMUPA 150 manufactured by Leed & Northrup Co.) based on a light scattering method.” U.S. Patent Application No. 2019/0305278 A1 to Saeki et al. at ¶ [0302]. Likewise, patent publications⁶ routinely disclose the measurement techniques and instruments used or suitable for characterizing particle size results. *See, e.g.*, Iwai at 15:3-6 (“laser diffraction particle size distribution measuring apparatus (MASTERSIZER 2000, produced by Sysmex Corporation)”); Akiike at ¶ [0195] (“The porous membrane slurry was measured by a laser diffraction particle size distribution measuring apparatus (“SALD-2000” manufactured by Shimadzu Corp.) to obtain D10, D50, and D90 of the non-conductive particles.”); Ueki at 12:43-46 (“a particle size distribution meter of a laser diffraction and scattering type (manufactured by Nikkiso Co.)”); Nishikawa at 12:55-59 (“a laser diffraction particle size distribution measuring apparatus (Mastersizer 2000, produced by Sysmex Corporation)”); Katayama at ¶ [0043] (“a laser diffraction particle size analyzer (e.g., LA-920 manufactured by Horiba, Ltd.) by dispersing the heat-resistant fine particles in a medium”); Schmitz at ¶ [0112] (“a laser scattering method in accordance with ISO 13320-1. An example of a suitable instrument for particle size analysis is a Microtrac S 3500.”). The lack of any such disclosure in the ’987 patent leaves the ordinary artisan unclear about the scope of the invention.

109. The recitation of “inorganic particles” in the ’987 patent’s claims further complicates the task facing the ordinary artisan. The ’987 patent does not limit its inorganic particles to a particular compound or shape. The patent lists varied compounds—“one or more selected from the group consisting of alumina, silica, magnesia, titanium oxide, hafnium dioxide,

⁶ I considered these prior art references solely for the purposes of confirming what details were disclosed in the particle size measurement.

tin oxide, zirconia, cerium dioxide, nickel oxide, zinc oxide, calcium oxide, boehmite, aluminum hydroxide, magnesium hydroxide, calcium hydroxide, and barium sulfate”—and says they may (or may not) be modified by sintering or other shaping techniques. ’987 patent 4:22-36. This leaves the ordinary artisan with the understanding that the claimed particles are not limited to a particular geometry, and instead could be spherical, shaped like plates, cylindrical rods, needle structures, or some other shape. As one example, aluminum hydroxide—one of the compounds identified in the patent—may have various plate-like geometries or rod-shaped geometry depending on how it is processed. Zhang 2017 at 6803, 6806. And different measurement techniques and instruments will provide different results for irregular shaped particles. Li 2005 at 278-83; NIST 86, 95; Merkus 143. Particle shape strongly affects the particle size distributions measured by different techniques for non-spherical particles. Li 2005 at 278-83; NIST 86, 95; Merkus 143. Significant differences between particle size distributions can result from using different techniques for measuring spherical particles as well. Li 2005 at 277, Table 3. Thus, without information related to the particle shapes and measurement techniques and conditions, an ordinary artisan may not know the conversion between different methods.

110. The prosecution of the ’987 patent does not change my analysis. During prosecution, the ratio of the Dv90 of the inorganic particles to the thickness of the porous layer was indicated to be the reason for allowance. ATL_COSMX_00001862, Notice of Allowance, 4. But the Applicant provided no guidance on how to determine Dv90, without which, a POSA cannot possibly get a “ratio of Dv90 of the inorganic particles to the thickness of the porous layer.” ATL_COSMX_00001862, Notice of Allowance, 4.

111. The ’987 patent fails to disclose any of the above measurement techniques or parameters for Dv90, which an ordinary artisan understands significantly impact Dv90 test

results. Furthermore, the specification's disclosed precision between what falls inside and outside the claims (a difference in Dv90 between 0 μm and 0.4 μm), would make it extremely difficult for an ordinary artisan to determine whether or not a particular product or sample would infringe the claims as written, especially at the edges of the claimed ranges. For at least these reasons and those further discussed above, it is my opinion that the '987 patent claims, when read in light of the patent's specification and the prosecution history do not "inform with reasonable certainty those skilled in the art about the scope of the invention." It is my opinion that the uncertainty in how to measure "Dv90," "Dv90 of inorganic particles," and "a ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.3 to 3.0"—when analyzed in view of the claims, the specification, and its prosecution history (including the prior art references cited therein)—would prevent the ordinary artisan from ascertaining the claims' scope with reasonable certainty. Therefore, I believe the claims of the '987 patent are indefinite.

F. "Dinitrile" and "Trinitrile" Compounds in the '363 Patent Have a Plain and Ordinary Meaning to an Ordinary Artisan

112. I also considered the '363 patent, its prosecution history, and the plain and ordinary meaning of the phrases "dinitrile compound" and "trinitrile compound" to a person of ordinary skill in the art. For my opinions on the '363 patent, I considered a person of ordinary skill in the art to be someone having a Ph.D. or a similar advanced degree in chemistry, chemical engineering, materials science, or a related field, and two or more years of experience related to the design, research, evaluation, preparation, and/or manufacture of electrochemical energy storage devices. This level of skill is approximate, and more experience would compensate for less formal education, and vice versa. It is consistent with the field provided by the '363 patent, which relates to "the technical field of energy storage technologies, in particular to an electrolyte

and an electrochemical device containing the electrolyte.” See ’363 patent, 1:12-14. A lower level of skill in the art, including an undergraduate or high school student taking chemistry courses, would not change my opinions offered herein.

113. In my opinion, the plain and ordinary meaning of “dinitrile compound” is “a compound with two cyano groups” and the plain and ordinary meaning of “trinitrile compound” is “a compound with three cyano groups.” Under basic chemistry naming conventions, known as nomenclature, an ordinary artisan understands that dinitrile compounds are compounds with *two* nitriles and trinitrile compounds are compounds with *three* nitriles, and not some other number of nitriles. See, e.g., Hawley’s Condensed Chemical Dictionary (16th ed. 2016) 168, 428, 974, 1358 (defining “di-” as a “[p]refix meaning two,” recognizing that “di- is preferred in chemical nomenclature” over the prefix “bi-,” applying “tri-” in chemical names to mean three); Raymond Chang, General Chemistry 46-47 (5th ed. 2008) (applying di and tri prefixes to mean two and three, respectively, in “straightforward” naming of molecular compounds).

114. An ordinary artisan likewise understands that nitrile compounds are organic compounds with cyano groups, i.e., “-C≡N grouping, e.g., acrylonitrile $\text{H}_2\text{C}=\text{CHC}\equiv\text{N}$ and acetonitrile ($\text{CH}_3\text{C}\equiv\text{N}$).” Hawley’s (2016) 974 (recognizing hazards of nitriles and providing examples: “[s]ome organic cyanide compounds are flammable (acetonitrile, acrylonitrile). Many organic cyanide compounds are toxic (acetonitrile, acrylonitrile).”) This is consistent with the specification of the ’363 patent, which states, for example, that “An embodiment of the present application provides an electrolyte and an electrochemical device containing the electrolyte, wherein the electrolyte comprises a compound comprising two cyano groups (herein also referred to as ‘a dinitrile compound’), a compound comprising three cyano groups (herein also referred to as ‘a trinitrile compound’), and propyl propionate.” ’363, patent, 1:37-56. It is further

consistent with the remainder of the claim language, which requires separate accounting of dinitrile compounds and trinitrile compounds. *See, e.g.*, claim 1. Additionally, nothing in the prosecution history departs from this plain and ordinary meaning.

115. I understand that ATL has not disagreed with anything in the proposed plain and ordinary meanings of these phrases, and I reserve the right to respond to any positions ATL later raises disputing this understanding of the plain and ordinary meaning of these phrases.

V. COMPENSATION

116. I am being compensated at my normal consulting rate of \$650 per hour for the time I spend on this matter. My compensation in no way depends on the outcome of this proceeding or the content of my testimony, and I have no other interest in this proceeding.

VI. SUPPLEMENTATION OF REPORT

117. I reserve the right to supplement or amend my opinions in response to additional opinions expressed by Plaintiff's expert(s) or in light of any additional evidence, testimony, discovery, or other information that may be provided to me after the date of this report. I also reserve the right to prepare and rely on visual aids or demonstratives that demonstrate my opinions at any hearing or trial.

I declare under penalty of perjury under the laws of the United States of America that the foregoing is true and correct.

Dated: May 23, 2023



Seth Miller, Ph.D.

Appendix A

Seth Adrian Miller, Ph.D.

1025 Neon Forest Circle, Longmont, CO 80504
(720) 432-4332 seth@heronscientific.com

MISSION

Through Heron Scientific, I offer technology consulting, IP consulting, and expert services, leveraging my expertise in developing new materials and device technologies with an emphasis on batteries and nanostructured materials.

EXPERIENCE SUMMARY

Business-focused technical leader with excellent analytical, communication and leadership skills. Technical expertise on the applications of materials science and device technologies to lithium batteries, novel semiconductor technologies, and medical devices. Led the development of multiple technologies from innovation to the marketplace.

SELECTED ACCOMPLISHMENTS

- Averted >\$100M in potential recall liability to TI by identifying a previously undiagnosed failure mechanism in a MEMS display. Led team to design replacement parts, shipping in <9 months.
- Acted as lead technical expert retained by SK Innovation in patent litigation against LG Energy Solutions. This and two related cases were collectively settled for \$1.8 billion.
- Author on 93 issued US patents, across a wide range of fields including batteries, nanomaterials, surface chemistry, and electronic and optical systems

PROFESSIONAL EXPERIENCE

Heron Scientific Inc., Longmont, CO
President

2008 – Present

Over the last decade, I have acted as a technology and innovation consultant to startups and Fortune 500 clients, and served as a testifying expert in litigation at the ITC, US District Court, and California State Court. Examples include:

- Consulting to lithium ion, sodium ion, and zinc-bromine battery companies on performance and cost improvement
- Consulting to a lithium ion battery startup to improve a solid electrolyte to enable safe, dendrite-free lithium metal batteries. Engagement included experimental design, data review, and fundraising through grant writing.
- For a large multinational, developed roadmaps for next generation energy storage and harvesting technology for wearables. This included analyzing performance, form factor, materials supply chain, and capability to scale.
- Analysis of the future of non-destructive test tech for a \$700M public company, reporting directly to the CEO
- Diligence for a Series A venture funds on energy storage, transportation, energy harvesting, micromachines and opto-electronic materials including technical assessment, scalability, and fit with intended markets.
- Retained as technical expert in litigation, including patent infringement, trade secret theft, false advertising, breach of contract, and personal libel (around technical claims). Deposed and testified at court/hearings six times.

In IP matters I have over a decade of experience in expert witness testimony, prior art and invalidity searching, IPR, performing diligence for investors, reviews of portfolios for large IP owners, and IP and technology landscapes.

Cairn Energy Research Associates
Electrochemistry Research Manager

2016-2021

Cost modeling, performance benchmarking, and risk assessment of new battery and supercapacitor technologies

Xinova, Seattle, WA
Managing Director, Innovation Banking and Development

2018 - 2020

Xinova helped companies find and scale technology growth through open requests to an inventor network; strategic planning to help clients take advantage of technology disruption; and access to capital to fund product development

Exhibit 2011

Ningde Amperex Technology Ltd.

IPR2023-00587, U.S. Patent No. 10,964,987

Page 56 of 107

Technology Reserve, Inc., Vancouver, BC, Canada 2014 - 2016
Chief Technologist / Technology Advisor
Helped multinational organizations out-license technologies to small and medium-sized businesses (SMEs).

ClearMark Systems, LLC, Longmont, CO 2013 – 2015
CEO
Co-founded a DARPA-funded software company developing anti-counterfeiting and forensics analysis software. The technology uses machine vision to capture a microscopic image of tooling-specific characteristics of a product, and by comparing these images against a database, an automated algorithm confirms authenticity. Raised \$1.1 M.

Rocky Mountain Section of the MRS, Longmont, CO 2008 – 2015
Founder, President
Founded and led non-profit that hosts technical talks, short courses, and networking events for the materials science community in the Front Range of Colorado.

University of Colorado, Boulder 2009 - 2014
Research Faculty
Collaboration with Prof. Doug Gin, Chemical Engineering. Consultant to Prof. Steve George, Chemistry. Consultant to Prof. Jill DuPre, ATLAS Institute.

Fluonic, Inc, Boulder, Colorado 2008 – 2013
Chief Scientific Officer
Led development of a microfluidic flow sensor for medical infusion. The sensor passed a short burst of electricity through a medical fluid to oxidize/reduce water, creating a localized concentration of acid/base which was sensed downstream.

Zettacore, Inc., Englewood, CO 2005 – 2008
Principal Polymer Chemist
Developed a new class of high strength gelled liquid electrolyte for a semiconductor memory device that stored bits in the redox states of porphyrin monolayers and multilayers. Raised \$500,000 in grant support for Zettacore's successor company, eSionic, to develop the technology further.

Texas Instruments Inc., DLP Products, Dallas, TX 2000 – 2005
Process Development, Packaging Development, Technical Manager
In 2001 elected as Group Member, Technical Staff, fastest in division history. Developed new technical solutions to dramatically improve the optical contrast ratio of DLP displays. Identified an unanticipated photodegradation mechanism in fielded devices, and led the team to develop a remedy that shipped as a "free upgrade" <9 months from project inception. Led teams of up to 20 engineers in integration and qualification of new anti-adhesion coatings.

EDUCATION

University of California, Berkeley, Berkeley, CA 1998-2000
Post-Doctoral Researcher, Dept of Chemistry, Lab of Prof. Doug Gin
Synthesis and testing of nanostructured organic polymers used as chemical catalysts

California Institute of Technology, Pasadena, CA 1992-1998
Ph. D., Chemistry, Lab of Prof. Denis Dougherty
Design, synthesis, and test of stable organic radicals (small molecules) which crystallize into magnets. Design and synthesis of new semiconducting and conducting polymers.

Carleton College, Northfield, MN B.A., 1988-1992
Chemistry, with distinction

LITIGATION HISTORY (AS TECHNICAL EXPERT)

- I have been retained as an expert eighteen times, and testified at trial or at an arbitration hearing six times.
- My testimony includes twice at the ITC (patent cases), one false advertising case (NY District Court), one trade secret matter (NJ Superior Court), one contract arbitration hearing (WIPO), and once in a personal libel case (California state).

Case: *Ningde Amperex Technology Limited v. Zhuhai CosMX Battery Co., Ltd., 2:22-cv-00232-JRG (E.D. Tex.)*
Parties: <Same>
Lead Atty: Jeffrey Totten
Firm: Finnegan
Year: 2023
Role: Testifying expert
Outcome: Ongoing

Case: Cellgard vs Shenzhen Senior Material Technology
Parties: <Same>
Lead Atty: Thomas Yeh
Firm: Latham & Watkins
Year: 2023
Role: Testifying expert
Outcome: Ongoing

Case: Oxygenator Water Technologies, Inc. v. Tennant Company
Parties: <Same>
Lead Atty: Nathan Louwaige
Firm: Carlson Caspers
Year: 2022
Role: Testifying expert
Outcome: Ongoing

Case: <IPR, display technology, Confidential>
Parties: <Confidential>
Lead Atty: Philip Irwin
Firm: Covington Burlington
Year: 2022
Role: Testifying expert
Outcome: Petition abandoned

Case: <IPR, display technology, Confidential>
Parties: <Confidential>
Lead Atty: Kaveh Saba
Firm: Covington Burlington
Year: 2022
Role: Testifying expert
Outcome: Petition abandoned

Case: <Patent litigation, optical materials, Confidential>
Parties: <Confidential>
Lead Atty: Matt Holohan
Firm: Sheridan Ross
Year: 2022
Role: Testifying expert
Outcome: Settled

Case: <Arbitration, breach of contract, Confidential>
Parties: <Confidential>
Lead Atty: Alicia Hickok
Firm: Faegre Drinker
Year: 2022
Role: Testifying expert
Outcome: Judgment, confidential

Case: <IPR, optical materials Confidential>
Parties: <Confidential>
Lead Atty: Cody Reeves
Firm: Covington
Year: 2021-2022
Role: Consulting expert
Outcome: Petition abandoned

Case: <Business defamation, Confidential>
Parties: <Confidential>
Lead Atty: Colin Cabral
Firm: Proskauer Rose
Year: 2021-2022
Role: Consulting expert, performed and coordinated analyses
Outcome: Ongoing

Case: Certain pouch-type battery cells, battery modules, and battery packs, components thereof, and products containing the same, ITC 337-1179
Parties: SK Innovation v LG Chemical
Lead Atty: Heng Gong
Firm: Covington
Year: 2019-2021
Role: Testifying expert, coordinated physical analysis, deposed, testified at trial
Outcome: Settled
LINK: https://www.usitc.gov/press_room/news_release/2019/er100411175.htm

Case: <Confidential>
Parties: As stated
Lead Atty: Baldassare Vinti
Firm: Proskauer Rose
Year: 2020
Role: Testifying expert, coordinated chemical analysis
Outcome: Settled

Case: Absorption Pharmaceuticals v. Reckitt Benckiser
Parties: As stated
Lead Atty: Colin Cabral
Firm: Proskauer Rose
Year: 2018-2022
Role: Testifying expert, coordinated chemical analysis, deposed, testified at jury trial
Outcome: [Jury decision for Reckitt](#)

Case: Prem Bikkina v Jagan Mahadevan
Parties: As stated
Lead Atty: Jeremy Tissot
Firm: Tissot Law
Year: 2017
Role: Testifying expert on accuracy of published research
Outcome: Judgement in favor of Bikkina

Case: Proposed class action suit, false advertising, Clorox
Parties: <class> v Clorox
Lead Atty: Mark Dearman
Firm: RGRD Law
Year: 2014
Role: Retained expert
Outcome: Case dismissed on legal grounds

Case: Unnamed, patent infringement
Parties: University of Michigan v Honda
Lead Atty: Patrick Neidemeier
Firm: Proskauer Rose
Year: 2013
Role: Retained expert, coordinated chemical analysis
Outcome: Case dismissed on legal grounds

Case: Unnamed, patent infringement
Parties: Analog Devices v Knowles
Lead Atty: Nathan Harris
Firm: Proskauer Rose
Year: 2012
Role: Retained expert on case stemming from "Certain MEMS Devices and Products Containing Same", above
Outcome: Settled

Case: Church & Dwight Co. v. The Clorox Company
Parties: Church & Dwight v Clorox
Lead Atty: Alexander Kaplan
Firm: Proskauer Rose
Year: 2011
Role: Testifying expert on technical subject matter, the effectiveness of carbon as an odor sorbent
Outcome: Judgment in favor of Church & Dwight, injunction against Clorox
LINK: <https://casetext.com/case/church-dwight-co-v-clorox-co>

Case: Certain MEMS Devices and Products Containing Same, ITC 337-700

Parties: Analog Devices v Knowles
Lead Atty: Colin Cabral
Firm: Proskauer Rose
Year: 2010
Role: Participated from inception; testified at deposition and on stand as lead expert, based on my expertise in MEMS stiction
Outcome: Judgment in favor of Analog Devices
LINK: <https://www.usitc.gov/publications/337/Pub4294.pdf>

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Appendix B



US010964987B2

(12) **United States Patent**
Yi et al.

(10) **Patent No.:** **US 10,964,987 B2**

(45) **Date of Patent:** **Mar. 30, 2021**

(54) **SEPARATOR AND ENERGY STORAGE DEVICE**

(71) Applicant: **Ningde Amperex Technology Limited**,
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(72) Inventors: **Jianjian Yi**, Fujian (CN); **Xinzhi Zhang**,
Fujian (CN); **Zengbin Wei**, Fujian (CN); **Xinghua Tao**,
Fujian (CN)

(73) Assignee: **Ningde Amperex Technology Limited**,
Ningde (CN)

(*) Notice: Subject to any disclaimer, the term of this
patent is extended or adjusted under 35
U.S.C. 154(b) by 79 days.

(21) Appl. No.: **16/013,887**

(22) Filed: **Jun. 20, 2018**

(65) **Prior Publication Data**

US 2019/0319243 A1 Oct. 17, 2019

(30) **Foreign Application Priority Data**

Apr. 11, 2018 (CN) 201810321779.5

(51) **Int. Cl.**

H01M 2/16 (2006.01)

H01M 2/14 (2006.01)

H01M 50/449 (2021.01)

H01M 10/0525 (2010.01)

H01M 50/403 (2021.01)

H01M 50/411 (2021.01)

H01M 50/446 (2021.01)

(52) **U.S. Cl.**

CPC **H01M 50/449** (2021.01); **H01M 10/0525**
(2013.01); **H01M 50/403** (2021.01); **H01M**
50/411 (2021.01); **H01M 50/446** (2021.01)

(58) **Field of Classification Search**

None

See application file for complete search history.

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Primary Examiner — Osei K Amponsah

(74) *Attorney, Agent, or Firm* — Taylor English Duma
LLP

(57) **ABSTRACT**

The application provides a separator and an energy storage device. The separator comprises: a porous substrate; and a porous layer arranged on a surface of the porous substrate, wherein the porous layer comprises inorganic particles and a binder, and a ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.3 to 3.0. Excellent adhesion exists between the separator and the electrode according to the present application, which ensures that the energy storage device has good safety performance. Moreover, the rate performance and cycle performance of the energy storage device can be greatly improved due to the existence of inorganic particles in the separator.

17 Claims, 2 Drawing Sheets

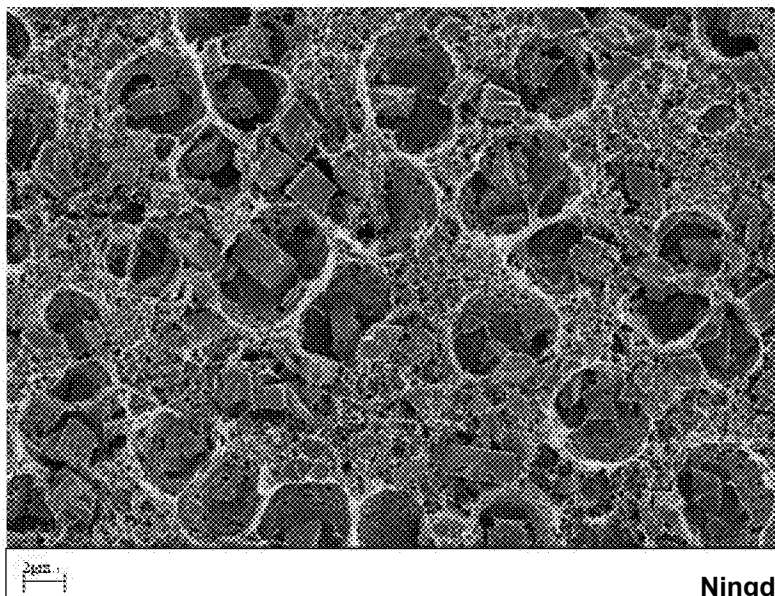


Exhibit 2011

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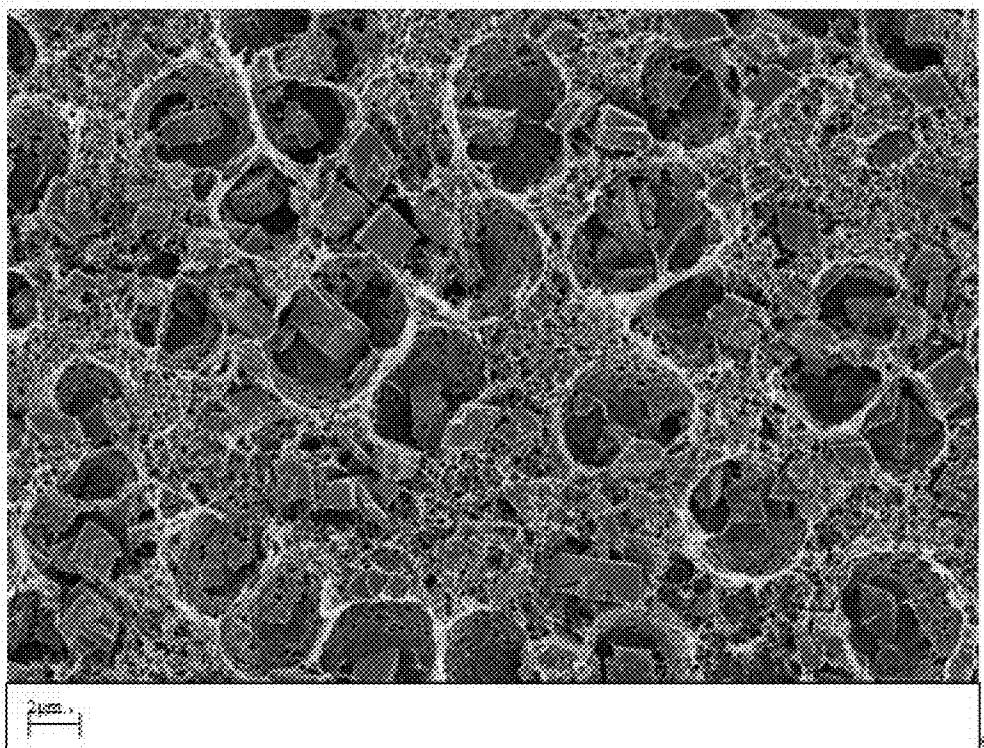


FIG. 1

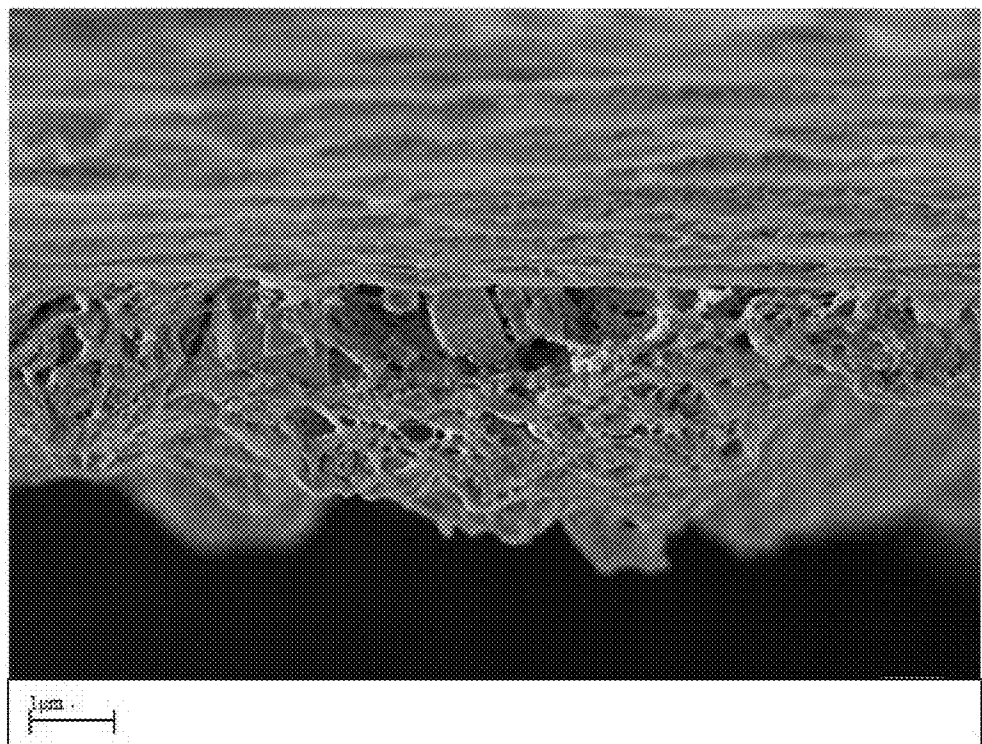


FIG. 2

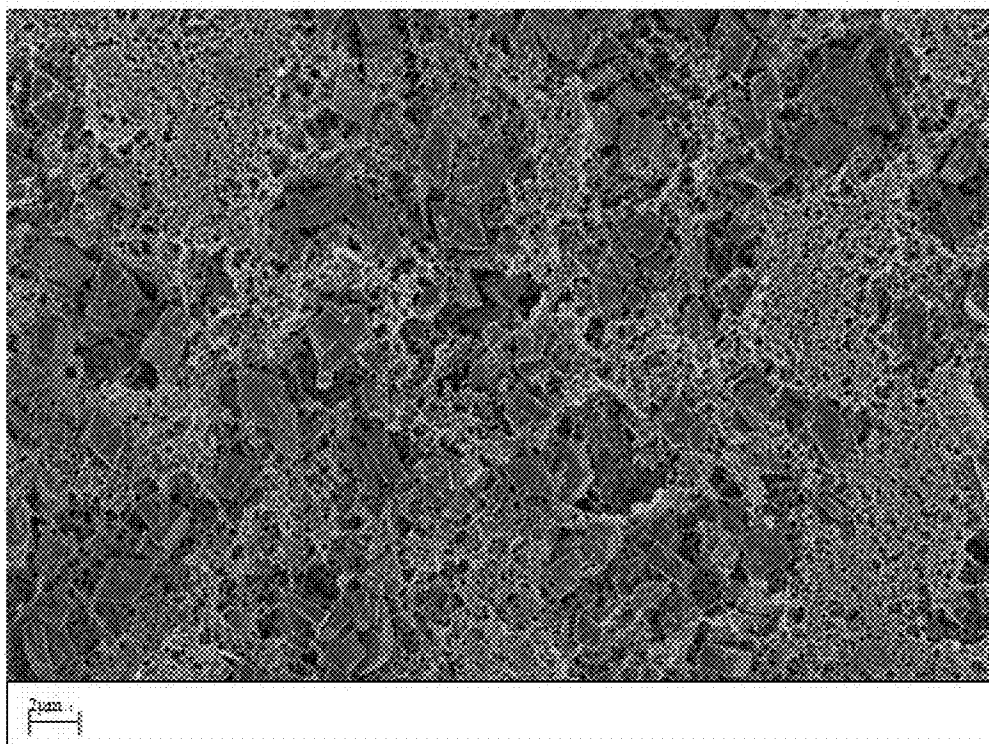


FIG.3

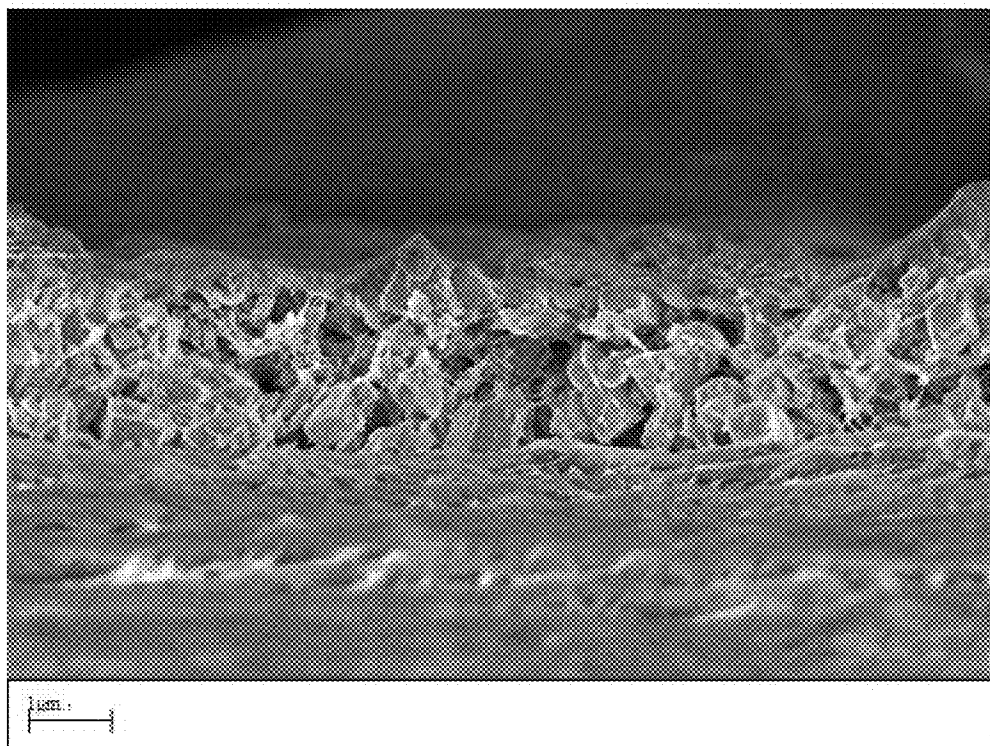


FIG.4

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SEPARATOR AND ENERGY STORAGE DEVICE

CROSS REFERENCES TO RELATED APPLICATIONS

This application claims the benefit of Chinese Patent Application No. 201810321779.5 filed on Apr. 11, 2018. The entire contents of the above application are hereby incorporated by reference in their entirety.

FIELD OF THE APPLICATION

The application relates to the field of energy storage devices, and in particular, to a separator and an energy storage device.

BACKGROUND OF THE APPLICATION

Non-aqueous secondary batteries, particularly lithium-ion batteries, are widely used in portable electronic devices such as notebook computers, digital cameras, camcorders, and cellphones, due to their high energy density and good rate performance. In recent years, the application of lithium-ion batteries as the power supply for electrical vehicles, is also promoting the development of lithium-ion battery technology. In the cycle process of the lithium-ion battery, with the progress of charging and discharging, there will be a gap between the electrode and the separator, resulting in a reduction of the cycle capacity of the lithium-ion battery and thereby having an influence on its service life. Therefore, there is an urgent need for a technical solution to solve the problem of the gap between the separator and the electrode so as to improve the service life of the lithium-ion battery.

SUMMARY OF THE APPLICATION

A separator and an energy storage device are provided according to the present application. With the separator provided by the application, not only the problem of the gap between the separator and the electrode can be solved, but also excellent adhesive force between the separator and the electrode can be maintained; moreover, the rate performance and cycle performance of the energy storage device can be improved due to the pore structures maintained in the separator of the application.

A separator is provided according to an example of the present application. The separator comprises: a porous substrate; and a porous layer arranged on a surface of the porous substrate, wherein the porous layer comprises inorganic particles and a binder, and a ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range of 0.3 to 3.0. The Dv90 of the inorganic particles refers to a particle size which reaches 90% the cumulative volume from the side of small particle size in the granularity distribution on a volume basis.

In the above-described separator, the porous layer has a thickness of 0.2 μm to 10 μm .

In the above-described separator, the porous layer has pores formed by the binder, and the inorganic particles are distributed in the pores.

In the above-described separator, the pores have an average pore size of 0.3 μm to 20 μm .

In the above-described separator, the volume ratio of the inorganic particles to the binder is in a range from 0.2 to 3.0.

In the above-described separator, the porous layer has a porosity of 20% to 90%.

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In the above-described separator, the inorganic particles are one or more selected from the group consisting of alumina, silica, magnesia, titanium oxide, hafnium dioxide, tin oxide, zirconia, cerium dioxide, nickel oxide, zinc oxide, calcium oxide, boehmite, aluminum hydroxide, magnesium hydroxide, calcium hydroxide, and barium sulfate.

In the above-described separator, the binder is one or more selected from the group consisting of polyvinylidene fluoride, vinylidene fluoride-hexafluoropropylene copolymer, polyamide, polyacrylonitrile, polyacrylate, polyacrylic acid, polyacrylates, sodium carboxymethylcellulose, polyvinylpyrrolidone, polyvinyl ether, polymethyl methacrylate, polytetrafluoroethylene and polyhexafluoropropylene.

An energy storage device is further provided according to the present application, which comprises the above-described separator.

In the above-described energy storage device, the energy storage device is a lithium-ion battery.

A method for preparing a separator is further provided according to the present application, the method comprising the steps of:

mixing inorganic particles with a binder to generate a mixture, and adding a first solvent into the mixture and stirring the mixture with the first solvent to obtain a uniform coating solution; coating the uniform coating solution onto a surface of a porous substrate to form a wet film; immersing the wet film into a coagulating solution for phase transformation; and drying the phase-transformed wet film to obtain the separator.

In the above-described method, the solid content in the coating solution is in a range of 7% to 25%.

In the above-described method, the coagulating solution comprises a second solvent and a third solvent, and the second solvent has a mass percentage of 20% to 60%.

In the above-described method, the first solvent is one or more independently selected from the group consisting of N-methylpyrrolidone (NMP), dimethylacetamide (DMAC) and dimethylformamide (DMF).

In the above-described method, the porous substrate comprises a polymer film, a multilayer polymer film, or a non-woven fabric formed of any one or more of the following polymers: polyethylene, polypropylene, polyethylene terephthalate, polyphthaloyl diamine, polybutylene terephthalate, polyester, polyacetal, polyamide, polyimide, polyetheretherketone, polyaryletherketone, polyetherimide, polyamide imide, polybenzimidazole, polyethersulfone, polyphenylene oxide, cycloolefin copolymer, polyphenylene sulfide, and polyethylene naphthalene.

In the above-described method, the polyethylene is at least one component selected from the group consisting of high density polyethylene, low density polyethylene, and ultrahigh molecular weight polyethylene.

In the above-described method, the second solvent is one or more independently selected from the group consisting of N-methylpyrrolidone, dimethylacetamide and dimethylformamide, and the third solvent is one or more selected from the group consisting of deionized water, ethanol, propanol, acetone, dimethyl carbonate and diethyl carbonate.

An excellent adhesion exists between the separator and the electrode according to the present application, thus ensuring that the energy storage device has good safety performance. Moreover, the porous layer is arranged on a surface of the separator, and the inorganic particles contained in the pores in the porous layer can serve to support the pores, and therefore the pores can be well protected from being destroyed. As such, the probability of pore blockage due to compression and swelling in the separator is reduced.

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the separator would exhibit high ionic conductivity, and the rate performance and cycle performance of the energy storage device are greatly improved.

BRIEF DESCRIPTION OF THE ACCOMPANYING DRAWINGS

FIG. 1 is an electronic microscope image (3000 times magnification) of pores of the lower surface of the porous layer (i.e., the surface of the porous layer that is away from the porous substrate) in example 3;

FIG. 2 is an electronic microscope image (10,000 times magnification) of the cross section of the porous layer in the thickness direction in example 3;

FIG. 3 is an electronic microscope image (3000 times magnification) of pores of the lower surface of the porous layer (i.e., the surface of the porous layer that is away from the porous substrate) in comparative example 2; and

FIG. 4 is an electronic microscope image (10,000 times magnification) of the cross section of the porous layer in the thickness direction in comparative example 2.

DETAILED DESCRIPTION OF THE PREFERRED EXAMPLES

Exemplary examples will be described in details. While these exemplary examples may be implemented in various forms, the applications should not be construed as limited to the examples of the application set forth herein. Rather, these examples are provided with the purpose of making the disclosure of the application thorough and complete and fully conveying the scope of the application to those skilled in the art.

A separator and a preparation method thereof as well as an energy storage device according to the present application will be described in details hereinafter.

In a first aspect of the application, the separator comprises a porous substrate and a porous layer. The porous layer is arranged on a surface of the porous substrate. Specifically, the porous layer is arranged on one surface of the porous substrate, or the porous layer is arranged on both surfaces of the porous substrate, or the porous layer is arranged on part of the surface of the porous substrate. The porous substrate comprises a polymer film, a multilayer polymer film, or a non-woven fabric formed of any one or more of the following polymers: polyethylene, polypropylene, polyethylene terephthalate, polyphthaloyl diamine, polybutylene terephthalate, polyester, polyacetal, polyamide, polyimide, polyetheretherketone, polyaryletherketone, polyetherimide, polyamide imide, polybenzimidazole, polyethersulfone, polyphenylene oxide, cycloolefin copolymer, polyphenylene sulfide, and polyethylene naphthalene. The above-mentioned polyethylene is at least one component selected from the group consisting of high density polyethylene, low density polyethylene, and ultrahigh molecular weight polyethylene.

In an example of the application, the porous layer arranged on the porous substrate comprises a binder and inorganic particles. The porous layer has pores formed by the binder, and the pores at least comprises a part of the inorganic particles. A ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.3 to 3.0. The Dv90 of the inorganic particles refers to a particle size which reaches 90% the cumulative volume from the side of small particle size in the granularity distribution on a volume basis. The binder is one or more selected from the group consisting of polyvinylidene fluoride, vinylidene fluo-

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ride-hexafluoropropylene copolymer, polyamide, polyacrylonitrile, polyacrylate, polyacrylic acid, polyacrylates, sodium carboxymethylcellulose, polyvinylpyrrolidone, polyvinyl ether, polymethyl methacrylate, polytetrafluoroethylene and polyhexafluoropropylene. The binder can provide a sufficient binding interface for the electrode, thus ensuring a high adhesive force between the separator and the electrode, and enabling the energy storage device (such as a lithium-ion battery) to have a high safety performance.

The pores in the porous layer allow the separator to have good electrolyte diffusion and absorption capabilities, improving the ionic conductivity of the separator, and thereby improving the rate performance of the energy storage device (such as a lithium-ion battery). The average pore size of the porous layer is not less than 0.3 μm , which makes the separator have good electrolyte diffusion and absorption capabilities, improves the ionic conductivity of the separator, reduces the polarization reaction, and thus can improve the rate performance of the energy storage device (such as a lithium-ion battery). The porous layer may have a porosity of 20% to 90%.

The inorganic particles distributed in the pores of the porous layer provide a good mechanical support for the porous layer, preventing the porous layer from compressing and collapsing during processing the energy storage device (such as a lithium-ion battery), and preventing the porous layer from being compressed and destroyed. Inorganic particles refer to a class of inorganic materials obtained from natural or synthetic compounds through the process of shaping and high-temperature sintering or the like. The inorganic particles are one or more selected from the group consisting of alumina, silica, magnesia, titanium oxide, hafnium dioxide, tin oxide, zirconia, cerium dioxide, nickel oxide, zinc oxide, calcium oxide, boehmite, aluminum hydroxide, magnesium hydroxide, calcium hydroxide, and barium sulfate. The inorganic particles may contain polar functional groups such as a hydroxyl group. The surface of the inorganic particles containing polar functional groups can be more easily combined with the non-solvent (third solvent) in the coagulating solution in the preparation process, which facilitates the diffusion of the non-solvent (third solvent) into the porous layer along the surface of the inorganic particles so that larger pores are formed in the vicinity of the inorganic particles, and the rate performance of the energy storage device (such as a lithium-ion battery) is thereby improved. The non-solvent (third solvent) in the coagulating solution will be described below.

In an example of the present application, the volume ratio of the inorganic particles to the binder in the porous layer is in a range from 0.2 to 3.0. An exchange between the first solvent in the coating solution and the third solvent in the coagulating solution can be increased by increasing the volume ratio of the inorganic particles to the binder, and the porosity of the porous layer is increased; conversely, if the volume ratio of the inorganic particles to the binder is reduced, the porosity of the porous layer will be decreased. If the volume ratio of the inorganic particles to the binder is too low, the average pore size of the porous layer will be decreased, and the porosity of the porous layer will be decreased; moreover, if the content of inorganic particles is decreased, the mechanical strength and heat resistance of the porous layer is decreased. If the volume ratio of the inorganic particles to the binder is too high, the adhesive force of the porous layer will be reduced, and the porous layer is easily detached from the surface of the porous substrate, resulting in deterioration of the safety performance of the energy storage device (such as a lithium-ion battery).

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The porous layer has a thickness of 0.2 μm to 10 μm . If the thickness of the porous layer is too high, the gas permeability of the porous layer will be deteriorated, and the rate performance of the energy storage device (such as a lithium-ion battery) will be decreased. If the thickness of the porous layer is too low, the adhesive force of the porous layer will be reduced, a gap will easily occur between the separator and the electrode, and the hardness of the energy storage device (such as a lithium-ion battery) will be decreased, resulting in deterioration of the safety performance of the energy storage device (such as a lithium-ion battery).

In addition, in an example of the present application, pores having a larger average pore size can be obtained by controlling the ratio of Dv90 of the inorganic particles to the thickness of the porous layer. In the electrolyte environment, after the binder (such as polyvinylidene fluoride) in the porous layer swells, more through-holes can be retained, which avoids swelling and pore blockage, ensures a channel for ion transportation, reduces polarization reactions, and improves the rate performance of the energy storage device (such as a lithium-ion battery). The Dv90 of the inorganic particles refers to a particle size which reaches 90% the cumulative volume from the side of small particle size in the granularity distribution on a volume basis.

In some examples of the present application, the ratio of the Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.4 to 1.5. By increasing the ratio of Dv90 of the inorganic particles to the thickness of the porous layer, the mechanical strength of the porous layer is increased, and the porous layer is prevented from undergoing pore blockage due to compression in the preparation or cycle process of the energy storage device (such as a lithium-ion battery), thereby increasing the rate performance of the energy storage device (such as a lithium-ion battery). Conversely, the mechanical properties of the porous layer may be reduced by reducing the ratio of Dv90 of inorganic particles to the thickness of the porous layer, leading to pore blockage of the porous layer due to compression in the preparation and cycle processes of the energy storage device (such as a lithium-ion battery), and the rate performance of the energy storage device (such as a lithium-ion battery) is reduced. In addition, if the ratio of the Dv90 of the inorganic particles to the thickness of the porous layer is too high, the pore size distribution of the porous layer tends to be non-uniform, resulting in a non-uniform surface of the porous layer, which further makes the porous layer easy to detach from the surface of the porous substrate and forms defects in the energy storage device (such as a lithium-ion battery). Moreover, if the ratio of Dv90 of the inorganic particles to the thickness of the porous layer is too high, the adhesive force of the porous layer is reduced, affecting the safety performance of the energy storage device (such as a lithium-ion battery). If the ratio of Dv90 of the inorganic particles to the thickness of the porous layer is too low, the inorganic particles cannot be effectively exposed on the surface of the porous layer, which does not facilitate an exchange between the first solvent in the coating solution and the third solvent in the coagulating solution in the preparation of the porous layer, therefore cannot enhance diffusion, and is not advantageous for the development of the pores.

The method for preparing the separator according to the present application is described below. Firstly, the inorganic particles are mixed with the binder, and then the first solvent is added into the mixture and the mixture is stirred to obtain a uniform coating solution, wherein the binder dissolves in the first solvent. The first solvent is one or more selected

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from the group consisting of N-methylpyrrolidone (NMP), dimethylacetamide (DMAC) and dimethylformamide (DMF). The porous layer formed using N-methylpyrrolidone (NMP) has the largest average pore size and the highest porosity; the porous layer formed using dimethylformamide (DMF) has the smallest average pore size and the lowest porosity; and the porous layer formed using dimethylacetamide (DMAC) has an average pore size and a porosity between those of the porous layer formed using N-methylpyrrolidone (NMP) and the porous layer formed using dimethylformamide (DMF). The temperature of the coating solution may be in a range from 15 degrees Celsius to 30 degrees Celsius. The solid content of the coating solution may be in a range from 7% to 25%. Within this range, if the solid content of the coating solution is increased, the viscosity of the coating solution is increased, the exchange rate between the third solvent in the coagulating solution and the first solvent in the coating solution becomes lower, and the average pore size and porosity of the formed porous layer are reduced; conversely, if the solid content of the coating solution is reduced, the average pore size and porosity of the porous layer are increased. However, if the solid content of the coating solution is too high and exceeds this range, the gas permeability of the porous layer does not meet the requirements and cannot satisfy the use for an energy storage device (such as a lithium-ion battery). If the solid content of the coating solution is too low, which is below this range, the strength of the porous layer is decreased, and it is difficult to form a film on the surface of the porous substrate.

Next, the coating solution is uniformly coated on the surface of the porous substrate to form a wet film. The coating method of the coating solution is dip coating. Meanwhile, any other suitable coating process may also be used.

Thereafter, the porous substrate with the wet film is immersed into a coagulating solution for phase transformation, wherein the coagulating solution may include a second solvent and a third solvent, and the second solvent and the third solvent are soluble with each other. The second solvent is one or more selected from the group consisting of N-methylpyrrolidone (NMP), dimethylacetamide (DMAC) and dimethylformamide (DMF), and the third solvent is one or more selected from the group consisting of deionized water, ethanol, propanol, acetone, dimethyl carbonate, and diethyl carbonate. The mass percentage of the second solvent in the coagulating solution may be in a range from 20% to 60%. The average pore size and porosity of the porous layer can be increased by decreasing the content of the second solvent in the coagulating solution. However, if the content of the second solvent is too low, a dense surface film may be formed on the surface of the porous layer that is away from the porous substrate, resulting in deterioration of the gas permeability of the porous layer. If the content of the second solvent is too high, complete pores cannot be formed, or the formed porous layer has a very low average pore size. The temperature of the coagulating solution may be in a range from 15 degrees Celsius to 30 degrees Celsius. The period for phase transformation may be in a range from 10 s to 90 s.

Finally, after the completion of the phase transformation, the wet film is dried to obtain a separator, wherein a porous layer is arranged on the surface of the porous substrate, and inorganic particles are distributed in the pores of the porous layer. The drying temperature may be in a range from 50 degrees Celsius to 70 degrees Celsius, and the drying period may be in a range from 10 minutes to 40 minutes.

According to some examples of the present application, a porous layer is prepared based on a phase transformation (i.e., non-solvent induced phase separation, NIPS) principle, wherein the first solvent in the coating solution is an organic solvent, and the binder (such as PVDF) can be dissolved in the first solvent. The coagulating solution contains a third solvent (deionized water), and the binder is insoluble in the third solvent (deionized water). Therefore, after the coating solution is coated on the porous substrate and then immersed into the coagulating solution, the first solvent in the coating solution is extracted into the third solvent (deionized water) in the coagulating solution, and the binder in the coating solution is solidified and precipitate to form a porous layer.

An energy storage device including the above-described separator is further provided according to the present application, such as a lithium-ion battery. In the application, the lithium-ion battery is only an illustrative example of the energy storage device, and the energy storage device may also comprise other suitable devices. The lithium-ion battery further comprises a positive electrode, a negative electrode, and an electrolyte, and the separator of the present application is placed between the positive electrode and the negative electrode. The positive current collector may be aluminum foil or nickel foil, and the negative current collector may be copper foil or nickel foil.

In the lithium-ion battery described above, the positive electrode comprises a positive electrode material capable of intercalation and deintercalation of lithium (Li) (hereinafter sometimes referred to as "positive electrode material capable of intercalation/deintercalation of lithium (Li)"). Examples of the positive electrode material capable of intercalation/deintercalation of lithium (Li) may include one or more of lithium cobaltate, lithium nickel cobalt manganate, lithium nickel cobalt aluminate, lithium manganate, lithium iron manganese phosphate, lithium vanadium phosphate, lithium vanadium oxide phosphate, lithium iron phosphate, lithium titanate, and lithium-rich manganese-based materials.

In the above-mentioned positive electrode material, the chemical formula of lithium cobaltate may be expressed as $\text{Li}_x\text{Co}_a\text{M1}_b\text{O}_{2-c}$, wherein M1 represents at least one selected from the group consisting of nickel (Ni), manganese (Mn), magnesium (Mg), aluminum (Al), boron (B), titanium (Ti), vanadium (V), chromium (Cr), ferrum (Fe), copper (Cu), zinc (Zn), molybdenum (Mo), tin (Sn), calcium (Ca), strontium (Sr), tungsten (W), yttrium (Y), lanthanum (La), zirconium (Zr), and silicon (Si), and the values of x, a, b, and c are respectively within the following ranges: $0.8 \leq x \leq 1.2$, $0.8 \leq a \leq 1$, $0 \leq b \leq 0.2$, $-0.1 \leq c \leq 0.2$.

In the above-mentioned positive electrode material, the chemical formula of lithium nickel cobalt manganate or lithium nickel cobalt aluminate may be expressed as $\text{Li}_y\text{Ni}_d\text{M2}_e\text{O}_{2-f}$, wherein M2 represents at least one selected from the group consisting of cobalt (Co), manganese (Mn), magnesium (Mg), aluminum (Al), boron (B), titanium (Ti), vanadium (V), chromium (Cr), ferrum (Fe), copper (Cu), zinc (Zn), molybdenum (Mo), tin (Sn), calcium (Ca), yttrium (Sr), tungsten (W), zirconium (Zr), and silicon (Si), and the values of y, d, e, and f are respectively within the following ranges: $0.8 \leq y \leq 1.2$, $0.3 \leq d \leq 0.98$, $0.02 \leq e \leq 0.7$, $-0.1 \leq f \leq 0.2$.

In the above-mentioned positive electrode material, the chemical formula of lithium manganate is expressed as $\text{Li}_z\text{Mn}_{2-g}\text{M3}_g\text{O}_{4-h}$, wherein M3 represents at least one selected from the group consisting of cobalt (Co), nickel (Ni), magnesium (Mg), aluminum (Al), boron (B), titanium (Ti), vanadium (V), chromium (Cr), ferrum (Fe), copper

(Cu), zinc (Zn), molybdenum (Mo), tin (Sn), calcium (Ca), strontium (Sr), and tungsten (W), and the values of z, g and h are respectively within the following ranges: $0.8 \leq z \leq 1.2$, $0 \leq g \leq 1.0$, and $-0.2 \leq h \leq 0.2$.

The negative electrode comprises a negative electrode material capable of intercalation and deintercalation of lithium (Li) (hereinafter, sometimes referred to as "negative electrode material capable of intercalation/deintercalation of lithium (Li)"). Examples of the negative electrode material capable of intercalation/deintercalation of lithium (Li) may include a carbon material, a metal compound, an oxide, a sulfide, a nitride of lithium such as LiN_3 , lithium metal, a metal which formed an alloy with lithium, and a polymer material.

Examples of carbon materials may include low graphitized carbon, easily graphitized carbon, artificial graphite, natural graphite, mesocarbon microbeads, soft carbon, hard carbon, pyrolytic carbon, coke, glassy carbon, organic polymer compound sintered body, carbon fiber and active carbon. Coke may include pitch coke, needle coke, and petroleum coke. The organic polymer compound sintered body refers to materials obtained by calcining and carbonizing a polymer material such as a phenol plastic or a furan resin at a suitable temperature and, and some of these materials are classified into low graphitized carbon or easily graphitized carbon. Examples of polymeric materials may include polyacetylene and polypyrrole.

Among these negative electrode materials capable of intercalation/deintercalation of lithium (Li), further, materials whose charge and discharge voltages are close to the charge and discharge voltages of lithium metal are selected. This is because of the fact that the lower the charge and discharge voltages of the negative electrode material are, the more easily the battery can have a higher energy density. The carbon material can be selected as the negative electrode material, since the crystal structure of the carbon material has only small changes during charging and discharging. Therefore, good cycle characteristics and high charge and discharge capacities can be obtained. In particular, graphite can be selected, since it can provide a high electrochemical equivalent and energy density.

In addition, the negative electrode material capable of intercalation/deintercalation of lithium (Li) may include elemental lithium metal, metal elements and semi-metal elements capable of forming an alloy together with lithium (Li), alloys and compounds including such elements, etc. In particular, they are used together with the carbon material, since good cycle characteristics and high energy density can be obtained in this case. In addition to alloys comprising two or more metal elements, alloys used herein further include alloys comprising one or more metal elements and one or more semi-metal elements. The alloys may be in the following forms of solid solutions, eutectic crystals (eutectic mixtures), intermetallic compounds, and mixtures thereof.

Examples of metal elements and semi-metal elements may include tin (Sn), lead (Pb), aluminum (Al), indium (In), silicon (Si), zinc (Zn), antimony (Sb), bismuth (Bi), cadmium (Cd), magnesium (Mg), boron (B), gallium (Ga), germanium (Ge), arsenic (As), silver (Ag), zirconium (Zr), yttrium (Y), and hafnium (Hf). Examples of the above-described alloys and compounds may include a material expressed as a chemical formula: $\text{Ma}_s\text{Mb}_t\text{Li}_u$, and a material expressed as a chemical formula: $\text{Ma}_p\text{Mc}_q\text{Md}_r$. In these chemical formulas, Ma represents at least one of metal elements and semi-metal elements capable of forming alloy with lithium, Mb represents at least one of these metal elements and semi-metal elements other than lithium and

Ma, Mc represents at least one of the non-metal elements, Md represents at least one of these metal elements and semi-metal elements other than Ma, and s, t, u, p, q, and r satisfy $s > 0$, $t \geq 0$, $u \geq 0$, $p > 0$, $q > 0$, and $r \geq 0$, respectively.

In addition, an inorganic compound that does not include lithium (Li) may be used in the negative electrode, such as MnO_2 , V_2O_5 , V_6O_{13} , NiS, and MoS.

The lithium-ion battery described above further comprises an electrolyte, which may be one or more of a gel electrolyte, a solid electrolyte, and an electrolyte. The electrolyte comprises a lithium salt and a non-aqueous solvent.

The lithium salt is one or more selected from the group consisting of LiPF_6 , LiBF_4 , LiAsF_6 , LiClO_4 , $\text{LiB}(\text{C}_6\text{H}_5)_4$, LiCH_3SO_3 , LiCF_3SO_3 , $\text{Li}(\text{SO}_2\text{CF}_3)_2$, $\text{Li}(\text{SO}_2\text{CF}_3)_3$, LiSiF_6 , LiBOB , and lithium difluoroborate. For example, LiPF_6 is used as a lithium salt, since it can provide high ionic conductivity and improve cycle performance.

The non-aqueous solvent may be a carbonate compound, a carboxylic acid ester compound, an ether compound, other organic solvents or combinations thereof.

The carbonate compound may be a chain carbonate compound, a cyclic carbonate compound, a fluorinated carbonate compound or combinations thereof.

Examples of chain carbonate compounds include diethyl carbonate (DEC), dimethyl carbonate (DMC), dipropyl carbonate (DPC), methylpropyl carbonate (MPC), ethyl propyl carbonate (EPC), methyl ethyl carbonate (MEC) and combinations thereof. Examples of the cyclic carbonate compounds include ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate (BC), vinyl ethylene carbonate (VEC), and combinations thereof. Examples of the fluoro-carbonate compound include fluoroethylene carbonate (FEC), 1,2-difluoroethylene carbonate, 1,1-difluoroethylene carbonate, 1,1,2-trifluoroethylene carbonate, 1,1,2,2-tetrafluoroethylene carbonate, 1-fluoro-2-methylethyl carbonate, 1-fluoro-1-methyl-ethylene carbonate, 1,2-difluoro-1-methylethylene carbonate, 1,1,2-trifluoro-2-methylethyl carbonate, trifluoromethyl ethylene carbonate, and combinations thereof.

Examples of carboxylic acid ester compounds include methyl acetate, ethyl acetate, n-propyl acetate, tert-butyl acetate, methyl propionate, ethyl propionate, γ -butyrolactone, decanolactone, valerolactone, mevalonolactone, caprolactone, methyl formate, and combinations thereof.

Examples of ether compounds include dibutyl ether, tetraethylene glycol dimethyl ether, diethylene glycol dimethyl ether, 1,2-dimethoxyethane, 1,2-diethoxyethane, ethoxy methoxy ethane, 2-methyltetrahydrofuran, tetrahydrofuran, and combinations thereof.

Examples of other organic solvents include dimethyl sulfoxide, 1,2-dioxolane, sulfolane, methyl sulfolane, 1,3-dimethyl-2-imidazolidinone, N-methyl-2-pyrrolidone, formamide, dimethylformamide, acetonitrile, trimethyl phosphate, triethyl phosphate, trioctyl phosphate, phosphate esters, and combinations thereof.

The positive electrode, the separator and the negative electrode are sequentially wound, stacked or folded into an electrode assembly, which is then placed into a packaging shell (for example, an aluminum plastic film), an electrolyte is injected, and the chemical conversion and packaging processes are performed to prepare a lithium-ion battery.

It should be understood by those skilled in the art that the above-described method for preparing a lithium-ion battery is only an example. Other methods commonly used in the art can be used without departing from the disclosure of the present application.

Since the separator is wound or stacked together with the positive electrode and the negative electrode, in a case that the above-described porous layer is formed on the surface of the positive electrode or the negative electrode, a corresponding technical effect can also be achieved.

Some specific examples and comparable examples are listed below to better illustrate this application. In the following examples, reagents, materials, and instruments used are commercially available unless otherwise specified. Some of the parameters used in the examples and comparative examples are shown in Table 1 below.

Comparative Example 1

(1) Preparation of Negative Electrode

The negative electrode active material (artificial graphite), the binder (styrene butadiene rubber), and the conductive agent (conductive carbon black (Super P)) are mixed uniformly with the solvent (deionized water) at a mass ratio of 92:3:5 to prepare a negative electrode slurry, then the negative electrode slurry is coated uniformly on both sides of the negative electrode current collector (copper foil), then a negative electrode active material layer is formed by drying the coated negative electrode current collector (copper foil) at 85 degrees Celsius, then cold pressing, slitting and cutting processes are performed and a negative electrode tab is welded so as to obtain a negative electrode.

(2) Preparation of Positive Electrode

The positive electrode active material (lithium cobaltate (LiCoO_2)), the binder (polyvinylidene fluoride (PVDF)), the conductive agent (conductive carbon black (Super P)) are dissolved in the solvent N-methylpyrrolidone (NMP) in a mass ratio of 97:1.5:1.5. A positive electrode slurry is prepared by uniformly stirring the mixture, then the positive electrode slurry is coated uniformly on both sides of the positive electrode current collector (aluminum foil), then a positive electrode active material layer is formed by drying the coated positive electrode current collector (aluminum foil) at 85 degrees Celsius, and then cold pressing, slitting, cutting processes are performed and a positive electrode tab is welded so as to obtain a positive electrode.

(3) Preparation of Separator

Boehmite is mixed with the binder (polyvinylidene fluoride), then the solvent N-methylpyrrolidone is added and the mixture is uniformly stirred to obtain a coating solution, and the volume ratio of the inorganic particles to the binder is 0.8. The coating solution is coated on the porous substrate (polyethylene) by means of dip coating to form a wet film. The porous substrate (polyethylene) with the wet film is immersed into the coagulating solution containing deionized water (third solvent) and N-methylpyrrolidone (second solvent) for phase transformation, with both the coating solution and the coagulating solution being at a temperature of 25 degrees Celsius. After being immersed in the coagulating solution for 30 seconds, the porous substrate (polyethylene) with the wet film is dried in an oven at 60 degrees Celsius to obtain a separator with a porous layer. The mass content of N-methylpyrrolidone (second solvent) in the coagulating solution is 40%. The ratio of Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.2.

(4) Preparation of Electrolyte

A solution prepared with lithium salt LiPF_6 and a non-aqueous organic solvent (ethylene carbonate (EC):diethyl carbonate (DEC):ethyl methyl carbonate (EMC):vinyl carbonate (VC)=8:85:5:2, by a mass ratio) by a mass ratio of 8:92 is used as the electrolyte of the lithium-ion battery.

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(5) Preparation of Lithium-Ion Battery

An electrode assembly is obtained by winding the positive electrode, the separator, and the negative electrode, and then sealing, injection of the electrolyte, forming, and suction molding processes are performed to obtain the lithium-ion battery.

Comparative Example 2

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 3.2 in comparative example 2. As can be seen in FIGS. 3-4, FIG. 3 is an electronic microscope image (3000 times magnification) of pores of a lower surface of the porous layer in comparative example 2, FIG. 4 is an electronic microscope image (10,000 times magnification) of the cross section of the porous layer in the thickness direction in comparative example 2.

Example 1

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.3 in Example 1.

Example 2

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.5 in Example 2.

Example 3

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.8 in Example 3. As can be seen in FIGS. 1-2, FIG. 1 is an electronic microscope image (3000 times magnification) of pores of a lower surface of the porous layer in example 3, FIG. 2 is an electronic microscope image (10,000 times magnification) of the cross section of the porous layer in the thickness direction in example 3.

Example 4

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 1.0 in Example 4.

Example 5

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 1.5 in Example 5.

Example 6

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 2.0 in Example 6.

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Example 7

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 3.0 in Example 7.

Example 8

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.8, and the volume ratio of the inorganic particles to the binder is 0.2 in Example 8.

Example 9

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.8, and the volume ratio of the inorganic particles to the binder is 0.5 in Example 9.

Example 10

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.8, and the volume ratio of the inorganic particles to the binder is 1.2 in Example 10.

Example 11

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.8, and the volume ratio of the inorganic particles to the binder is 2 in Example 11.

Example 12

The preparation method is the same as that of comparative example 1, except that the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.8, and the volume ratio of the inorganic particles to the binder is 3 in Example 12.

Example 13

The preparation method is the same as that of comparative example 1, except that the inorganic particles used in Example 13 are alumina, and the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.8 in Example 13.

Example 14

The preparation method is the same as that of comparative example 1, except that the inorganic particles used in Example 14 are magnesium hydroxide, and the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.8 in Example 14.

Example 15

The preparation method is the same as that of comparative example 1, except that the inorganic particles used in

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Example 15 are titanium oxide, and the ratio of the Dv90 of the inorganic particles in the porous layer to the thickness of the porous layer is 0.8 in Example 15.

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is measured. Then, the true volume V2 of the porous layer is equal to $V2=V20-V0$, and the porosity of the porous layer is equal to $1-V2/V1$.

TABLE 1

	Solid content in the coating solution	Types of inorganic particles	Ratio of the Dv90 of the inorganic	Volume ratio	Coagulating solution	
			particles to the thickness of the porous layer	of the inorganic particles to the binder	second solvent	content of second solvent
Example 1	15%	boehmite	0.3	0.8	NMP	40%
Example 2	15%	boehmite	0.5	0.8	NMP	40%
Example 3	15%	boehmite	0.8	0.8	NMP	40%
Example 4	15%	boehmite	1.0	0.8	NMP	40%
Example 5	15%	boehmite	1.5	0.8	NMP	40%
Example 6	15%	boehmite	2.0	0.8	NMP	40%
Example 7	15%	boehmite	3.0	0.8	NMP	40%
Example 8	15%	boehmite	0.8	0.2	NMP	40%
Example 9	15%	boehmite	0.8	0.5	NMP	40%
Example 3	15%	boehmite	0.8	0.8	NMP	40%
Example 10	15%	boehmite	0.8	1.2	NMP	40%
Example 11	15%	boehmite	0.8	2	NMP	40%
Example 12	15%	boehmite	0.8	3	NMP	40%
Example 3	15%	boehmite	0.8	0.8	NMP	40%
Example 13	15%	alumina	0.8	0.8	NMP	40%
Example 14	15%	magnesium hydroxide	0.8	0.8	NMP	40%
Example 15	15%	titanium oxide	0.8	0.8	NMP	40%
Comparative Example 1	15%	boehmite	0.2	0.8	NMP	40%
Comparative Example 2	15%	boehmite	3.2	0.8	NMP	40%

Next, the performances of separator and lithium-ion battery are tested.

(1) Thickness Test of Porous Layer

Separator samples are placed on a thickness gage (Model VL-50 LITEMATIC from Naitutoyo company) using a 5 mm flat bottom probe with a speed of 50 mm/min and a pressure of 0.01N. Each of the separator samples provided with a porous layer is measured for 60 thickness points, and the average thickness is taken as the measured value. The thickness of the porous layer is equal to the thickness of the separator minus the thickness of the porous substrate. In a case that there are two porous films, the thickness is divided by 2 to obtain the thickness of each porous layer.

(2) Gas Permeability Test of Porous Layer

A 100 mm×100 mm separator sample provided with a porous layer is cut and tested using a US Gurley 4110N Densometers with 100 cc test gas, and the period that the test gas passes completely through the separator sample provided with the porous layer is recorded as a Gurley value. The Gurley value of the porous layer is equal to the Gurley value of the separator provided with the porous layer minus the Gurley value of the separator without the porous layer (i.e., a pure porous substrate).

(3) Porosity Test of Porous Layer

The length, width, and thickness of a separator sample provided with a porous layer are measured, and the thickness of the porous layer is obtained by subtracting the thickness of the porous substrate (polyethylene) from the thickness of the separator, and the apparent volume V1 of the porous layer is obtained by calculation. The true volume V20 of the separator sample provided with the porous layer is measured using a true density meter (AccuPyc II Model 1340 Gas Pycnometer, Micromeritics Company), and the true volume V0 of the porous substrate (polyethylene) of the same area

(4) Rate Performance Test of Lithium-Ion Battery

At 25 degrees Celsius, the lithium-ion battery is discharged to 3.0 V at a constant current of 0.2 C, rested for 10 minutes, then is charged to 4.4 V at a constant current of 0.7 C, then is charged to 0.02 C at a constant voltage of 4.4 V, rested for 10 min, and is further discharged at a constant current of 0.2 C until the voltage reaches 3.0 V. The discharge capacity at this time is measured and recorded as Q1. Then, the lithium-ion battery is charged to 4.4V at a constant current of 0.7 C, and then charged to 0.02 C at a constant voltage of 4.4 V, rested for 10 minutes, and then is discharged at a constant current of 2 C until the voltage is 3.0 V. The discharge capacity at this time is measured and recorded as Q2.

2 C/0.2 C rate performance (%) of the lithium-ion battery is equal to $Q2/Q1 \times 100\%$.

(5) Cycle Performance Test of Lithium-Ion Battery

At 25 degrees Celsius, the lithium-ion battery is charged to 4.4 V at a constant current of 0.7 C, then charged to 0.02 C at a constant voltage of 4.4 V, rested for 10 minutes, and discharged to 3.0 V at a constant current of 1 C, rested for 10 minutes. The discharge capacity at this time is measured and recorded as Q3. The above-described steps are regarded as one cycle of charge and discharge. 200 cycles are performed, and the discharge capacity after 200 cycles is recorded as Q4.

The capacity retention rate (%) of the lithium-ion battery after 200 cycles is equal to $Q4/Q3 \times 100\%$.

The test results of examples and comparative examples are shown in Table 2 below.

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TABLE 2

	Porosity of porous layer	Gurley value of porous layer (s/100 cc)	Thickness of porous layer (μm)	2 C/0.2 C rate performance	Capacity retention rate after 200 cycles
Example 1	48%	53	2	79.50%	85.70%
Example 2	50%	42	2	83.70%	92.80%
Example 3	52%	30	2	87.50%	95.60%
Example 4	53%	28	2	84.60%	93.10%
Example 5	54%	25	2	82.50%	91.20%
Example 6	56%	20	2	81.70%	85.20%
Example 7	57%	18	2	80.40%	82.70%
Example 8	46%	57	2	81.10%	87.20%
Example 9	49%	43	2	82.80%	91.70%
Example 3	52%	30	2	87.50%	95.60%
Example 10	54%	25	2	83.10%	91.30%
Example 11	56%	21	2	81.50%	88.80%
Example 12	59%	16	2	79.70%	83.80%
Example 3	52%	30	2	87.50%	95.60%
Example 13	50%	43	2	84.40%	92.90%
Example 14	52%	31	2	86.90%	94.30%
Example 15	51%	36	2	84.10%	91.30%
Comparative Example 1	45%	70	2	69.80%	70.50%
Comparative Example 2	60%	16	2	71.30%	72.60%

As can be seen from a comparison among examples 1-7 and comparative examples 1-2, the ratio of Dv90 of the inorganic particles to the thickness of the porous layer has an effect on the rate performance and cycle performance of the lithium-ion battery, and the probability of pore blockage due to compression swelling in the porous layer can be reduced effectively by controlling the ratio of the Dv90 of the inorganic particles to the thickness of the porous layer to be in a range from 0.3 to 3.0, so that the rate performance and cycle performance of the lithium-ion battery can be effectively improved. If the ratio of Dv90 of the inorganic particles to the thickness of the porous layer is too high (for example, comparative example 2), the pore size distribution of the porous layer tends to be non-uniform, the surface of the porous layer is heterogeneous, the porous layer is easily detached from the surface of the porous substrate, and defects are formed in the energy storage device (such as a lithium-ion battery), which is not advantageous for the improvement on rate performance and cycle performance. In addition, if the ratio of Dv90 of the inorganic particles to the thickness of the porous layer is too high, the adhesive force of the porous layer is reduced relatively, affecting the safety performance of the energy storage device (such as a lithium-ion battery). If the ratio of Dv90 of the inorganic particles to the thickness of the porous layer is too low (for example, comparative example 1), the inorganic particles cannot be effectively exposed on the surface of the porous layer, which does not facilitate an exchange of the first solvent in the coating solution and the third solvent in the coagulating solution in the preparation process of the porous layer, cannot enhance diffusion, is not advantageous for the development of the pores, and is also not advantageous for the improvement of the rate performance and cycle performance.

As can be seen from the comparison among Example 3 and Examples 8-12, the volume ratio of inorganic particles to the binder has an effect on the rate performance and cycle performance of the lithium-ion battery. An increase of the volume ratio is advantageous for an exchange of the third solvent in the coagulating solution and the first solvent in the coating solution and the increase of the porosity of the porous layer. If the volume ratio of inorganic particles to the

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binder is too low, the porosity of the porous layer is decreased, the gas permeability of the separator deteriorates, and the rate performance of the lithium-ion battery is not significantly improved, which is not advantageous for the improvement of the rate performance and cycle performance. If the ratio of the volume ratio of the inorganic particles to the binder is too high, the adhesive force of the porous layer is reduced, gaps are easily formed between the separator and the electrodes, the hardness of the lithium-ion battery is reduced, and thus the rate performance and the cycle performance are not significantly improved.

As can be seen from the comparison among Example 3 and Examples 13-15, the use of various inorganic particles can improve the rate performance and cycle performance of the lithium-ion battery, and the types of inorganic particles used have less effect on the rate performance and cycle performance of the lithium-ion battery.

It should be understood by those skilled in the art that the above-described examples are only illustrative examples and should not be construed limiting the application. The various changes, substitutions, and alterations could be made to the application without departing from the spirit and scope of the application.

What is claimed is:

1. A separator, comprising: a porous substrate; and a porous layer arranged on a surface of the porous substrate, wherein the porous layer comprises inorganic particles and a binder, and a ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.3 to 3.0.
2. The separator according to claim 1, wherein the porous layer has a thickness of 0.2 μm to 10 μm .
3. The separator according to claim 1, wherein the pores have an average pore size of 0.3 μm to 20 μm .
4. The separator according to claim 1, wherein a volume ratio of the inorganic particles to the binder is in a range from 0.2 to 3.0.
5. The separator according to claim 1, wherein the porous layer has a porosity of 20% to 90%.
6. The separator according to claim 1, wherein the inorganic particles are one or more selected from the group consisting of alumina, silica, magnesia, titanium oxide, hafnium dioxide, tin oxide, zirconia, cerium dioxide, nickel oxide, zinc oxide, calcium oxide, boehmite, aluminum hydroxide, magnesium hydroxide, calcium hydroxide, and barium sulfate.
7. The separator according to claim 1, wherein the binder is one or more selected from the group consisting of polyvinylidene fluoride, vinylidene fluoride-hexafluoropropylene copolymer, polyamide, polyacrylonitrile, polyacrylate, polyacrylic acid, polyacrylates, sodium carboxymethylcellulose, polyvinylpyrrolidone, polyvinyl ether, polymethyl methacrylate, polytetrafluoroethylene and polyhexafluoropropylene.
8. An energy storage device, comprising the separator according to claim 1.
9. The energy storage device according to claim 8, wherein the energy storage device is a lithium-ion battery.
10. A method for preparing the separator of claim 1, wherein the method comprises steps of: mixing inorganic particles with a binder to generate a mixture; adding a first solvent into the mixture; stirring the mixture with the first solvent to obtain a uniform coating solution; coating the uniform coating solution onto a surface of a porous substrate to form a wet film.

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the coating solution is in a range from 7% to 25%; and wherein a ratio of Dv90 of the inorganic particles to the thickness of the porous layer is in a range from 0.3 to 3.0.

11. The method according to claim 10, wherein the solid content in the coating solution is in a range from 7% to 25%.

12. The method according to claim 10, wherein the coagulating solution comprises a second solvent and a third solvent, and the second solvent has a mass percentage of 20% to 60%.

13. The method according to claim 10, wherein the first solvent is one or more independently selected from the group consisting of N-methylpyrrolidone, dimethylacetamide and dimethylformamide.

14. The method according to claim 10, wherein the porous substrate comprises a polymer film, a multilayer polymer film, or a non-woven fabric formed by any one or more of the following polymers: polyethylene, polypropylene, polyethylene terephthalate, polyphthaloyl diamine, polybutylene terephthalate, polyester, polyacetal, polyamide, polyimide,

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polyetheretherketone, polyaryletherketone, polyetherimide, polyamide imide, polybenzimidazole, polyethersulfone, polyphenylene oxide, cycloolefin copolymer, polyphenylene sulfide, and polyethylene naphthalene.

15. The method according to claim 14, wherein the polyethylene is at least one selected from the group consisting of high density polyethylene, low density polyethylene, and ultrahigh molecular weight polyethylene.

16. The method according to claim 12, wherein the second solvent is one or more independently selected from the group consisting of N-methylpyrrolidone, dimethylacetamide and dimethylformamide, and the third solvent is one or more selected from the group consisting of deionized water, ethanol, propanol, acetone, dimethyl carbonate and diethyl carbonate.

17. The separator according to claim 1, wherein the porous layer has pores formed by the binder, at least a part of the inorganic particles are embedded in the pores.

* * * * *

Appendix C

List of Materials Considered

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U.S. Patent No. 10,964,987

Prosecution History for U.S. Patent No. 10,964,987

U.S. Patent No. 10,833,363

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Appendix D

(12) **United States Patent**
Wang et al.

(10) **Patent No.:** **US 10,833,363 B2**
(45) **Date of Patent:** **Nov. 10, 2020**

(54) **ELECTROLYTE AND ELECTROCHEMICAL DEVICE**

(71) Applicant: **NINGDE AMPEREX TECHNOLOGY LIMITED**, Ningde (CN)

(72) Inventors: **Kefei Wang**, Ningde (CN); **Qiao Zeng**, Ningde (CN); **Liangzhen Xiao**, Ningde (CN); **Fei Wu**, Ningde (CN)

(73) Assignee: **NINGDE AMPEREX TECHNOLOGY LIMITED**, Ningde (CN)

(*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 110 days.

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(22) Filed: **Dec. 6, 2018**

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H01M 10/0525 (2010.01)

H01M 4/13 (2010.01)

(52) **U.S. Cl.**

CPC ... **H01M 10/0566** (2013.01); **H01M 10/0525** (2013.01); **H01M 2300/0025** (2013.01)

(58) **Field of Classification Search**

CPC H01M 10/05; H01M 10/0566; H01M 10/0545

See application file for complete search history.

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(57) **ABSTRACT**

The present application relates to an electrolyte and an electrochemical device comprising the electrolyte. The electrolyte comprises a dinitrile compound, a trinitrile compound, and propyl propionate, wherein, based on the total weight of the electrolyte, the content X of the nitrile compound and the content Y of the trinitrile compound meet the conditions represented by Formula (1) and Formula (2): {about 2 wt % $\leq$ (X+Y) $\leq$ about 11 wt % . . . (1), about $0.1 \leq (X/Y) \leq$ about 8 . . . (2)}. The electrolyte of the present application is capable of effectively inhibiting the increase in DC internal resistance of an electrochemical device so that the electrochemical device has excellent cycle and storage performance.

16 Claims, 3 Drawing Sheets



Electrode C

(56)

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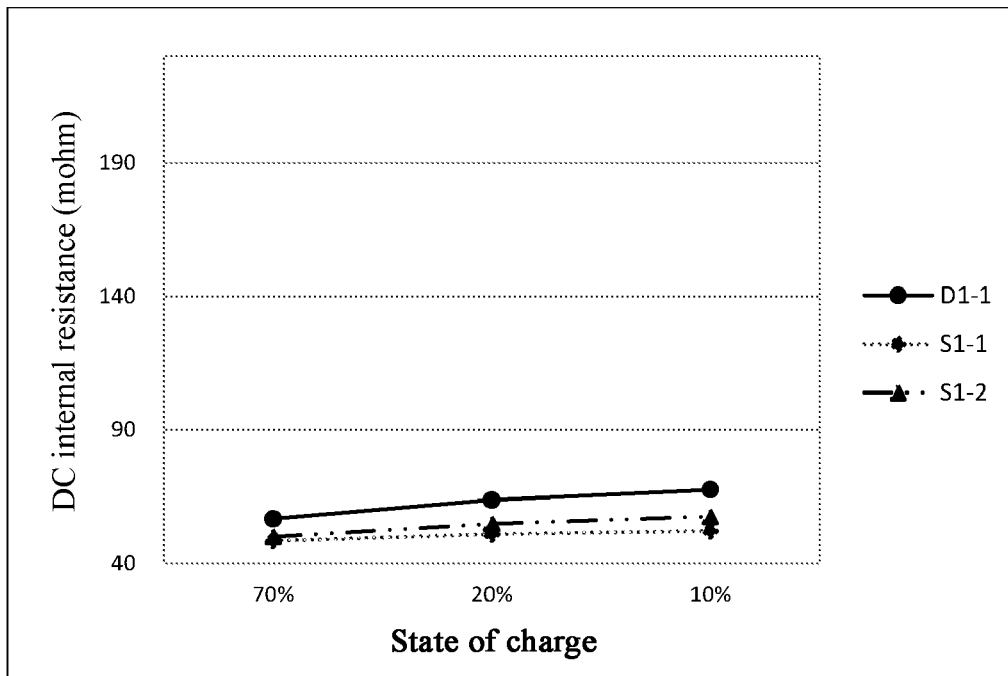


FIG. 1a

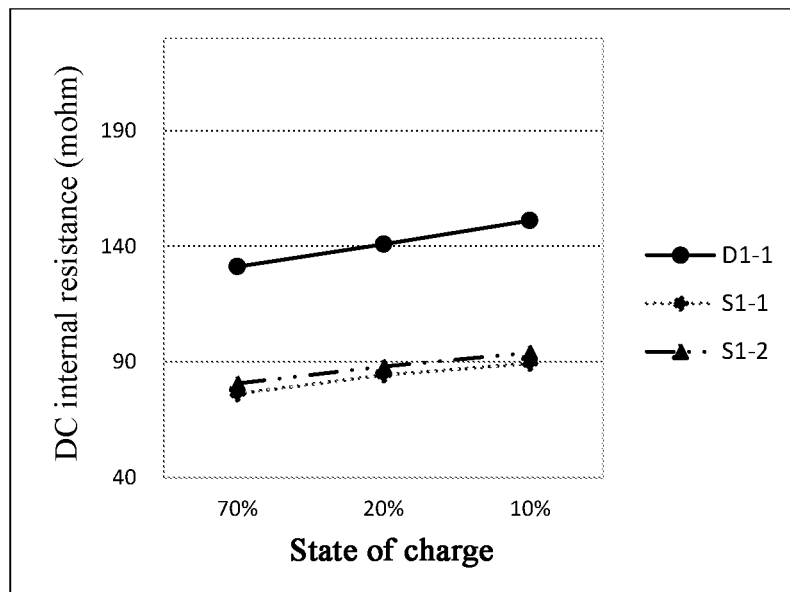


FIG. 1b

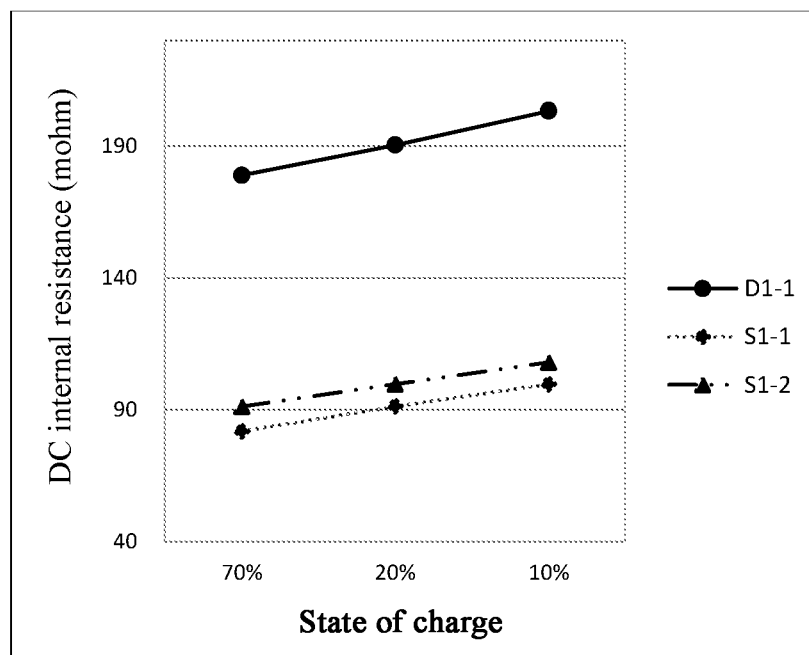


FIG. 1c

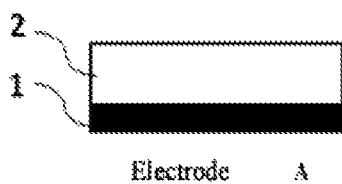


FIG. 2

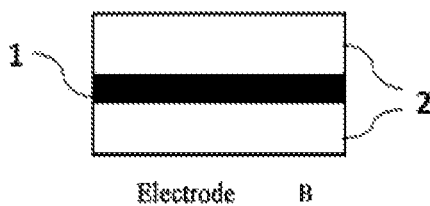


FIG. 3

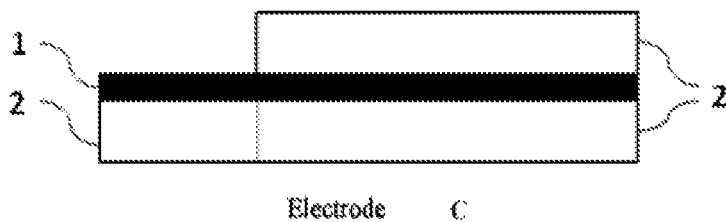


FIG. 4

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ELECTROLYTE AND ELECTROCHEMICAL
DEVICE

BACKGROUND OF THE INVENTION

The present application claims the benefit of priority from China Patent Application No. 201811108529.X, filed on 21 Sep. 2018, the disclosure of which is hereby incorporated by reference in its entirety.

1. Field of the Invention

The present disclosure relates to the technical field of energy storage technologies, in particular to an electrolyte and an electrochemical device containing the electrolyte.

2. Description of the Related Art

With the rapid development of electronic devices and electric vehicles, the requirement for high capacity electrochemical devices has grown. In order to increase capacity density of electrochemical devices, there is a need for effective methods to develop high-voltage electrochemical devices.

At present, electrochemical devices with a working voltage above 4.4V have become a hot research area in many research institutes and enterprises. However, at high voltages, the oxidation activity of the positive electrode material increases, and the stability decreases, which makes the electrolyte decompose on the surface of the positive electrode easily or cause deterioration of the battery material, resulting in a decrease in battery capacity. In order to solve the above problems, it is definitely necessary to provide an improved electrochemical device.

SUMMARY OF THE INVENTION

An embodiment of the present application provides an electrolyte and an electrochemical device containing the electrolyte, wherein the electrolyte comprises a compound comprising two cyano groups (herein also referred to as “a dinitrile compound”), a compound comprising three cyano groups (herein also referred to as “a trinitrile compound”), and propyl propionate. The dinitrile compound can form a protective film on the cathode of the electrochemical device, so as to inhibit the decomposition of the solvent in the electrochemical device. However, since the protective film itself is decomposed on the surface of the cathode at a high potential, the effect of inhibiting decomposition of the solvent cannot be sustained for a long time. The present inventors unexpectedly found that by using a mixture of a dinitrile compound, a trinitrile compound and propyl propionate, a firm protective film which is not easily decomposed on the surface of the cathode at a high potential can be formed. The electrolyte according to the embodiment of the present application can effectively inhibit the increase in DC internal resistance of the electrochemical device.

In an embodiment, the present application provides an electrolyte. The electrolyte comprises a dinitrile compound, a trinitrile compound, and propyl propionate, wherein, based on the total weight of the electrolyte, the weight percentage (X) of the dinitrile compound and the weight percentage (Y) of the trinitrile compound based on the total weight of the electrolyte solution meet the conditions represented by Formula (1) and Formula (2):

$$\text{about } 2 \text{ wt } \% \leq (X+Y) \leq \text{about } 11 \text{ wt } \% \quad (1); \text{ and} \quad 65$$

$$\text{about } 0.1 \leq (X/Y) \leq \text{about } 8 \quad (2).$$

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According to an embodiment of the present application, the weight percentage (Y) of the trinitrile compound and the weight percentage (Z) of the propyl propionate based on the total weight of the electrolyte meet the condition represented by Formula (3):

$$\text{about } 0.01 \leq (Y/Z) \leq \text{about } 0.3 \quad (3).$$

According to an embodiment of the present application, the weight percentage of the dinitrile compound is X, and X is about 0.01-10 wt %; the weight percentage of the trinitrile compound is Y, and Y is about 0.01-10 wt %; and the weight percentage of the propyl propionate is Z, and Z is about 5-50 wt %. The electrolyte according to the embodiments of the present application can effectively inhibit the increase in DC internal resistance of the electrochemical device, thereby achieving a better effect.

According to an embodiment of the present application, the electrolyte further comprises a fluoroether, wherein based on the total weight of the electrolyte, the content of fluoroether is about 0.01-10 wt %. The fluoroether can form a better protective film with the trinitrile compound, thereby improving the DC internal resistance and the storage performance of the electrochemical device.

According to an embodiment of the present application, the electrolyte further comprises a cyclic phosphonic anhydride, wherein, based on the total weight of the electrolyte, the content of the cyclic phosphonic anhydride is about 0.01-10 wt %. The addition of the cyclic phosphonic anhydride can further inhibit the increase in DC internal resistance during the cycle.

According to an embodiment of the present application, the electrolyte further comprises one selected from the group consisting of: a cyclic carbonate ester having a carbon-carbon double bond, a fluorinated chain carbonate ester, a fluorinated cyclic carbonate ester, a compound having a sulfur-oxygen double bond, and any combination thereof. These compounds can form a firm protective film which is not easily to be decomposed at the electrode interface with the dinitrile compound, the trinitrile compound and propyl propionate, thereby further inhibiting the side reactions in the electrochemical device and reducing the voltage drop during storage of the electrochemical device, so as to improve the long-term storage performance and reliability of the electrochemical device.

In another embodiment, the present application provides an electrochemical device and an electronic device using the electrochemical device. The electrochemical device comprises electrodes and an electrolyte which is any of the electrolyte described above.

According to an embodiment of the present application, the electrode includes a current collector and a coating on the current collector. The coating includes a single-sided coating, a double-sided coating, or a combination thereof. The single-sided coating is a coating formed by applying a slurry on one surface of the current collector. The double-sided coating is a coating formed by applying a slurry on two surfaces of the current collector. The electrode with the single-sided coating has an electrode compaction density D1, and the electrode with the double-sided coating has an electrode compaction density D2, wherein D1 and D2 meet the following relationship: about $0.8 \leq D1/D2 \leq \text{about } 1.2$.

According to an embodiment of the present application, the electrode s include a cathode and an anode. In some embodiments, when the electrode is an cathode, about $3.5 \text{ g/cm}^3 \leq D2 \leq \text{about } 4.3 \text{ g/cm}^3$. In some other embodiments, when the electrode is an anode, about $1.2 \text{ g/cm}^3 \leq D2 \leq 1.8 \text{ g/cm}^3$.

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In the electrochemical device according to the embodiments of the present application, when the electrolyte of the electrochemical device comprises a compound comprising two cyano groups, a compound comprising three cyano groups, and propyl propionate, and the electrode compaction densities meet the above relationships, the electrode can have good electrical conductivity, the effects of the cathode and anode active materials are well exerted, which is important to controlling the expansion of electrochemical devices. Therefore, the electrochemical device according to the embodiments of the present application achieves high capacity density and has excellent cycle and storage performances.

In another embodiment, the present application provides an electronic device including the electrochemical device.

Additional aspects and advantages of the embodiments of the present application will be partially described, illustrated or explained by way of examples in the description as follows.

BRIEF DESCRIPTION OF THE DRAWINGS

The invention will be described according to the appended drawings in which:

The drawings needed for describing the embodiments of the present application or the prior art will be briefly described below to facilitate the description of the embodiments of the present application. Obviously, the drawings in the following description are only a portion of the embodiments of the present application. For those skilled in the art, the drawings of other embodiments can be obtained according to the structures illustrated in the drawings without creative efforts.

FIG. 1a shows the DC internal resistances of fresh batteries of Examples S1-1 and S1-2 of the present application and Comparative Example D1-1 at different charge states (10% SOC, 20% SOC, and 70% SOC).

FIG. 1b shows the DC internal resistances of batteries of Examples S1-1 and S1-2 of the present application and Comparative Example D1-1 at different charge states (10% SOC, 20% SOC, and 70% SOC) after 200 cycles of charge and discharge.

FIG. 1c shows the DC internal resistances of batteries of Examples S1-1 and S1-2 of the present application and Comparative Example D1-1 at different charge states (10% SOC, 20% SOC, and 70% SOC) after 400 cycles of charge and discharge.

FIG. 2 is a view schematically showing the structure of an electrode A with a single-sided coating of the present application.

FIG. 3 is a view schematically showing the structure of an electrode B with a double-sided coating of the present application.

FIG. 4 is a view schematically showing the structure of an electrode C with a single-sided coating and a double-sided coating of the present application.

PREFERRED EMBODIMENT OF THE PRESENT APPLICATION

Embodiments of the present application will be described in detail below. Throughout the specification of the present application, the same or similar components and components having the same or similar functions are denoted by similar reference numerals. The embodiments described herein with respect to the figures are explanatory, and illustrative, and are provided to facilitate the basic under-

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standing of the application. The embodiments of the present application should not be interpreted as limitations to the present application. Unless otherwise expressly indicated, the following terms used herein have the meanings indicated below.

As used herein, the term “about” is used to describe and depict minor variations. When used in connection with an event or circumstance, the term may refer to an example in which the event or circumstance occurs precisely, and an example in which the event or circumstance occurs approximately. For example, when used in connection with a value, the term may refer to a range of variation less than or equal to $\pm 10\%$ of the stated value, such as less than or equal to $\pm 5\%$, less than or equal to $\pm 4\%$, less than or equal to $\pm 3\%$, less than or equal to $\pm 2\%$, less than or equal to $\pm 1\%$, less than or equal to $\pm 0.5\%$, less than or equal to $\pm 0.1\%$, or less than or equal to $\pm 0.05\%$. In addition, amounts, ratios, and other values are sometimes presented in a range format in this application. It is to be understood that such a range format is provided for convenience and simplicity, and should be understood flexibly to include not only the numerical values that are explicitly defined in the range, but also all the individual values or sub-ranges that are included in the range, as if each value and sub-range are explicitly specified.

As used herein, “hydrocarbyl” covers alkyl, alkenyl, and alkynyl groups. For example, the hydrocarbyl may be a straight-chain hydrocarbon structure having 1 to 20 carbon atoms. The hydrocarbon group also may be a branched or cyclic hydrocarbon structure having 3 to 20 carbon atoms. When a hydrocarbon group having a specific number of carbon atoms is defined, it may cover all geometric isomers having the carbon number. The hydrocarbon group herein may also be a hydrocarbon group having 1 to 15 carbon atoms, a hydrocarbon group having 1 to 10 carbon atoms, a hydrocarbon group having 1 to 5 carbon atoms, a hydrocarbon group having 5 to 20 carbon atoms, a hydrocarbon group having 5 to 15 carbon atoms or a hydrocarbon group having 5 to 10 carbon atoms. Additionally, the hydrocarbon group may be optionally substituted. For example, the hydrocarbon group may be substituted by halo including fluorine, chlorine, bromine, and iodine, an alkyl group, an aryl group or a heteroaryl group.

As used herein, the “alkyl group” may be a linear saturated hydrocarbon structure having 1 to 20 carbon atoms. The alkyl group also may be a branched or cyclic hydrocarbon structure having 3 to 20 carbon atoms. For example, the alkyl group may be an alkyl group having 1-20 carbon atoms, an alkyl group having 1-10 carbon atoms, an alkyl group having 1-5 carbon atoms, an alkyl group having 5-20 carbon atoms, an alkyl group having 5-15 carbon atoms, or an alkyl group having 5-10 carbon atoms. When an alkyl group having a specific number of carbon atoms is defined, it may cover all geometric isomers having the carbon number. Therefore, for example, “butyl” means n-butyl, sec-butyl, isobutyl, tert-butyl and cyclobutyl; and “propyl” includes n-propyl, isopropyl and cyclopropyl. Examples of the alkyl group include, but are not limited to, methyl, ethyl, n-propyl, isopropyl, cyclopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, cyclobutyl, n-pentyl, isoamyl, neopentyl, cyclopentyl, methylcyclopentyl, ethylcyclopentyl, n-hexyl, isohexyl, cyclohexyl, n-heptyl, octyl, cyclopropyl, cyclobutyl, norbornyl and so on. Additionally, the alkyl group may be optionally substituted.

As used herein, the term “alkylene group” means a linear or branched divalent saturated hydrocarbon group. For example, the alkylene group may be an alkylene group

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having 1-20 carbon atoms, an alkylene group having 1-15 carbon atoms, an alkylene group having 1-10 carbon atoms, an alkylene group having 1-5 carbon atoms, an alkylene group having 5-20 carbon atoms, an alkylene group having 5-15 carbon atoms, or alkylene group having 5-10 carbon atoms. Representative alkylene group includes (for example) methylene, ethane-1,2-diyl ("ethylene"), propane-1,2-diyl, propane-1,3-diyl, butane-1,4-diyl, pentane-1,5-diyl and the like. Additionally, the alkylene group may be optionally substituted.

As used herein, the term "alkenylene group" covers both linear and branched alkenylene groups. When an alkenylene group having a specific number of carbon atoms is defined, it may cover all geometric isomers having the carbon number. For example, the alkenylene group may be an alkenylene group having 2-20 carbon atoms, an alkenylene group having 2-15 carbon atoms, an alkenylene group having 2-10 carbon atoms, an alkenylene group having 2-5 carbon atoms, an alkenylene group having 5-20 carbon atoms, an alkenylene group having 5-15 carbon atoms, or alkenylene group having 5-10 carbon atoms. Representative alkenylene group includes (for example) ethenylene, propenylene, butenylene and the like. Additionally, the alkenylene group may be optionally substituted.

As used herein, the term "aryl" encompasses both monocyclic and polycyclic systems. A polycyclic ring may have two or more rings in which two carbons are shared by two adjacent rings (where the rings are "fused"), in which at least one of the rings is aromatic and other rings may be for example, a cycloalkyl group, a cycloalkenyl group, an aryl group, a heterocyclyl group and/or a heteroaryl group. For example, the aryl group may be a C_6 - C_{50} aryl group, a C_6 - C_{40} aryl group, a C_6 - C_{30} aryl group, a C_6 - C_{20} aryl group, or a C_6 - C_{10} aryl group. Representative aryl group includes (for example) phenyl, methylphenyl, propylphenyl, isopropylphenyl, benzyl and naphthalen-1-yl, naphthalen-2-yl and the like. Additionally, the aryl group may be optionally substituted.

As used herein, the term "heteroaryl group" covers a monocyclic heteroaromatic group which may include one to three heteroatoms, for example, pyrrole, furan, thiophene, imidazole, oxazole, thiazole, triazole, pyrazole, pyridine, pyrazine, pyrimidine, and the like. The term heteroaryl group also includes a polycyclic heteroaromatic system having two or more rings in which two atoms are shared by two adjacent rings (where the ring is "fused"), in which at least one of the rings is a heteroaryl group, and other rings may be a cycloalkyl group, a cycloalkenyl group, an aryl group, a heterocyclyl group and/or a heteroaryl group. The heteroatom in the heteroaryl group may be for example O, S, N, Se, and so on. For example, the heteroaryl group may be a C_2 - C_{50} heteroaryl group, a C_2 - C_{40} heteroaryl group, a C_2 - C_{30} heteroaryl group, a C_2 - C_{20} heteroaryl group, or a C_2 - C_{10} heteroaryl group. Additionally, the aryl group may be optionally substituted.

When the above substituents are substituted, the substituent is selected from the group consisting of halogen, an alkyl group, a cycloalkyl group, an alkenyl group, an aryl group, and a heteroaryl group.

As used herein, the content of each component is obtained based on the total weight of the electrolyte.

I. Electrolyte

An embodiment of the present application provides an electrolyte, which comprises an electrolyte and a solvent in which the electrolyte is dissolved. It is one of the main characteristics of the electrolyte of the present application that the electrolyte comprises a dinitrile compound, a trini-

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trile compound, and propyl propionate, wherein, based on the total weight of the electrolyte, the weight percentage of the is X and the weight percentage of the trinitrile compound is Y, and X and Y meet the conditions represented by Formula (1) and Formula (2):

$$\text{about } 2 \text{ wt } \% \leq (X+Y) \leq \text{about } 11 \text{ wt } \% \quad (1); \text{ and}$$

$$\text{about } 0.1 \leq (X/Y) \leq \text{about } 8 \quad (2).$$

In some embodiments, X and Y meet about $2 \text{ wt } \% \leq (X+Y) \leq \text{about } 8 \text{ wt } \%$ and about $0.1 \leq (X/Y) \leq \text{about } 6$ simultaneously. In some embodiments, X and Y meet about $3 \text{ wt } \% \leq (X+Y) \leq 6 \text{ wt } \%$ and about $0.2 \leq (X/Y) \leq \text{about } 5$ simultaneously. In some embodiments, X and Y meet about $4 \text{ wt } \% \leq (X+Y) \leq \text{about } 5 \text{ wt } \%$ and about $0.3 \leq (X/Y) \leq \text{about } 4$ simultaneously. In some embodiments, X and Y meet about $2 \text{ wt } \% \leq (X+Y) \leq \text{about } 5 \text{ wt } \%$ and about $0.1 \leq (X/Y) \leq \text{about } 1$ simultaneously.

In some embodiments, based on the total weight of the electrolyte, the weight percentage of the trinitrile compound is Y and the weight percentage of the propyl propionate is Z, and Y and Z meet the condition represented by Formula (3):

$$\text{about } 0.01 \leq (Y/Z) \leq \text{about } 0.3 \quad (3).$$

In some embodiments, Y and Z meet about $0.01 \leq (Y/Z) \leq \text{about } 0.2$. In some embodiments, Y and Z meet about $0.02 \leq (Y/Z) \leq \text{about } 0.1$.

In some embodiments, the weight percentage of the dinitrile compound is about 0.01-10 wt %. In some embodiments, the weight percentage of the dinitrile compound is not less than about 0.01 wt %, not less than about 0.1 wt %, not less than about 0.3 wt %, or not less than about 0.5 wt %. In some embodiments, the weight percentage of the dinitrile compound is not greater than about 10 wt %, not greater than about 8 wt %, or not greater than about 6 wt %.

In some embodiments, the weight percentage of the trinitrile compound is about 0.01-10 wt %. In some embodiments, the weight percentage of the trinitrile compound is not less than about 0.01 wt %, not less than about 0.1 wt %, not less than about 0.3 wt %, or not less than about 0.5 wt %. In some embodiments, the weight percentage of the trinitrile compound is not greater than about 10 wt %, not greater than about 8 wt %, not greater than about 5 wt %, not greater than about 4 wt %, or not greater than about 3 wt %.

In some embodiments, the weight percentage of the propyl propionate is about 5-50 wt %. In some embodiments, the weight percentage (Z) of the propyl propionate is about 5-40 wt %, about 10-40 wt %, about 10-30 wt %, about 20-50 wt %, about 20-40 wt %, about 20-30 wt %, or about 25-30 wt %.

Compound Comprising Two Cyano Groups (Dinitrile Compound)

In some embodiments, the dinitrile compound according to the present application includes a compound of Formula [4] or [5]:



or a combination thereof,

wherein in Formula [4] or [5], R_1 , R_2 , R_3 and R_4 are each independently an alkylene having 1-20 carbon atoms, for example, an alkylene group having 1-15 carbon atoms, an alkylene group having 1-10 carbon atoms, an alkylene group having 1-5 carbon atoms, an alkylene group having 5-10 carbon atoms, an alkylene group having 5-20 carbon atoms, an alkylene group having 5-15 carbon atoms, an alkylene group having 2-20 carbon atoms, an alkenylene group

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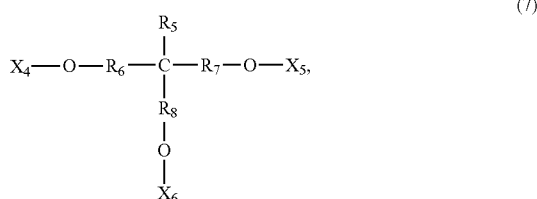
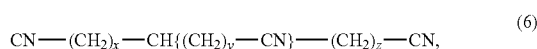
having 2-15 carbon atoms, an alkenylene group having 2-10 carbon atoms, an alkenylene group having 2-5 carbon atoms, an alkenylene group having 5-20 carbon atoms, an alkenylene group having 5-15 carbon atoms, or an alkenylene group having 5-10 carbon atoms, and n is an integer from 0 to 5.

In some embodiments, the dinitrile compound according to the present application is a compound of Formula [4] or [5].

In some embodiments, the dinitrile compound of the present application includes, but is not limited to, butanedinitrile, glutaronitrile, adiponitrile, 1,5-dicyanopentane, 1,6-dicyanohexane, 1,7-dicyanoheptane, 1,8-dicyanooctane, 1,9-dicyanononane, 1,10-dicyanodecane, 1,12-dicyanododecane, tetramethylbutanedinitrile, 2-methylglutaronitrile, 2,4-dimethylglutaronitrile, 2,2,4,4-tetramethylglutaronitrile, 1,4-dicyanopentane, 2,6-dicyanoheptane, 2,7-dicyanooctane, 2,8-dicyanononane, 1,6-dicyanodecane, 1,2-dicyanobenzene, 1,3-dicyanobenzene, 1,4-dicyanobenzene, 3,5-dioxapimelonitrile, 1,4-bis(cyanoethoxy)butane, ethylene glycol bis(2-cyanoethyl)ether, diethylene glycol bis(2-cyanoethyl)ether, triethylene glycol bis(2-cyanoethyl)ether, tetraethylene glycol bis(2-cyanoethyl)ether, 3,6,9,12,15,18-hexaoxaecicosanoic dinitrile, 1,3-bis(2-cyanoethoxy)propane, 1,4-bis(2-cyanoethoxy)butane, 1,5-bis(2-cyanoethoxy)pentane, ethylene glycol bis(4-cyanobutyl) ether, 1,4-dicyano-2-butene, 1,4-dicyano-2-methyl-2-butene, 1,4-dicyano-2-ethyl-2-butene, 1,4-dicyano-2,3-dimethyl-2-butene, 1,4-dicyano-2,3-diethyl-2-butene, 1,6-dicyano-3-hexene, 1,6-dicyano-2-methyl-3-hexene, or 1,6-dicyano-2-methyl-5-methyl-3-hexene. The dinitrile compounds may be used alone or in combination of two or more thereof.

Compound Comprising Three Cyano Groups (Trinitrile Compound)

In some embodiments, the trinitrile compound according to the present application includes a compound of Formula [6] or [7]:



or a combination thereof,

wherein in Formula [6], x, y, and z represents an integer from 0 to 5 and x, y, and z are not 0 at the same time; and

in Formula [7], R₅ may be hydrogen or an alkyl group having 1-20 carbon atoms, for example, an alkyl group having 1-10 carbon atoms, an alkyl group having 1-5 carbon atoms, an alkyl group having 5-20 carbon atoms, an alkyl group having 5-15 carbon atoms, or an alkyl group having 5-10 carbon atoms; R₆, R₇, and R₈ each independently may be an alkylene group having 1-20 carbon atoms, for example, an alkylene group having 1-15 carbon atoms, an alkylene group having 1-10 carbon atoms, an alkylene group having 1-5 carbon atoms, an alkylene group having 5-20 carbon atoms, an alkylene group having 5-15 carbon atoms, or an alkylene group having 5-10 carbon atoms; and X₄, X₅

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and X₆ each independently may be —R₉—CN, wherein R₉ may be an alkylene group having 1-20 carbon atoms, for example, an alkylene group having 1-15 carbon atoms, an alkylene group having 1-10 carbon atoms, an alkylene group having 1-5 carbon atoms, an alkylene group having 5-20 carbon atoms, an alkylene group having 5-15 carbon atoms, or an alkylene group having 5-10 carbon atoms.

In some embodiments, the trinitrile compound according to the present application is a compound of Formula [6] or [7].

In some embodiments, the trinitrile compound of the present application includes, but is not limited to, 1,3,5-pentanetricarbonitrile, 1,2,3-propanetrinitrile, 1,3,6-hexanetricarbonitrile, 1,2,6-hexanetricarbonitrile, 1,2,3-tris(2-cyanoethoxy)propane, 1,2,4-tris(2-cyanoethoxy)butane, 1,1,1-tris(cyanoethoxymethylene)ethane, 1,1,1-tris(cyanoethoxymethylene)propane, 3-methyl-1,3,5-tris(cyanoethoxy)pentane, 1,2,7-tris(cyanoethoxy)heptane, 1,2,6-tris(cyanoethoxy)hexane, or 1,2,5-tris(cyanoethoxy)pentane. The trinitrile compounds may be used alone or in combination of two or more thereof.

Fluoroether Compound

In some embodiments, the fluoroether compound of the present application includes at least one of the compounds of Formula [8], Formula [9], Formula [10], or Formula [11]:



where in Formulae [8], [9], [10], and [11], Rf1 and Rf2 are each independently a linear or branched C₁ to C₁₂ fluoroalkyl group at least one hydrogen atom of which is replaced with fluorine, R is a linear or branched C₁ to C₁₂ alkyl group, and R' is a linear or branched C₁ to C₅ alkylene group, and n is an integer from 1 to 5.

In some embodiments, the fluoroether compound of the present application is a compound of Formula [8], Formula [9], Formula [10], or Formula [11].

In some embodiments, Rf1 or Rf2 is each independently a fluoroalkyl group selected from the group consisting of HCF₂—, CF₃—, HCF₂CF₂—, CH₃CF₂—, CF₃CH₂—, CF₃CF₂—, (CF₃)₂CH—, HCF₂CF₂CH₂—, CF₃CH₂CH₂—, HCF₂CF₂CF₂CH₂—, HCF₂CF₂CF₂CF₂CH₂—, CF₃CF₂CH₂—, CF₃CFHCF₂CH₂—, HCF₂CF(CF₃)CH₂—, and CF₃CF₂CH₂CH₂—.

The fluoroether can form a better protective film with the trinitrile compound, thereby further improving the DC internal resistance and the storage performance of the battery.

In some embodiments, the fluoroether of the present application includes, but is not limited to:

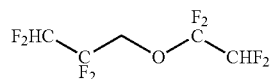
HCF₂CF₂CH₂OCF₂CF₂H (FEPE), (CF₃)₂CFCF(CF₂CF₃) (OCH₃) (TMMP), CF₃CHFCF₂CH(CH₃)OCF₂CHFCF₃ (TPTP), HCF₂CF₂CH₂OCF₂CF₂CF₂H, HCF₂CF₂OCF₂CF₃, HCF₂CF₂OCH₂CH₂OCF₂CF₃H, HCF₂CF₂OCH₂CH₂OCF₂CF₂H, HCF₂CF₂CH₂OCF₂CF₂CF₂H, HCF₂CF₂OCH₂CH₂OCF₂CF₂CF₂H, HCF₂CF₂OCH₂CH₂OCF₂CF₂CF₂CF₂H, CH₃OCH₂CH₂OCH₂CH₂F, CH₃OCH₂CH₂OCH₂CF₃, CH₃OCH₂CH(CH₃)OCH₂CH₂F, CH₃OCH₂CH(CH₃)OCH₂CF₃, FCH₂CH₂OCH₂CH₂OCH₂CH₂F, FCH₂CH₂OCH₂CH(CH₃)OCH₂CH₂F, CF₃CH₂O(CH₂CH₂O)₂CH₂CF₃ or CF₃CH₂OCH₂CH(CH₃)OCH₂CF₃.

The fluoroether may be used alone or in combination of two or more thereof.

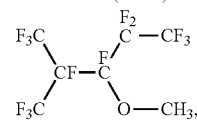
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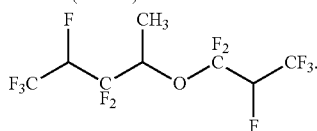
The structural formulas of FEPE, TMMP, and TPTP are shown below:



1,1-difluoro-2,2-difluoroethyl-2',2'-difluoro-3',3'-difluoropropyl ether,
(FEPE)



2-trifluoromethyl-3-methoxyperfluoropentane,
(TMMP)

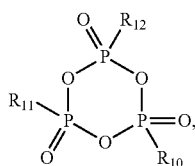


2-(trifluoro-2-fluoro-3-difluoropropoxy)-3-difluoro-4-fluoro-5-trifluoropentane,
(TPTP)

In some embodiments, based on the total weight of the electrolyte, the content of the fluoroether is not less than about 0.01 wt %, not less than about 0.1 wt %, or not less than about 0.5 wt %. In some embodiments, the content of the fluoroether is not greater than about 5 wt %, not greater than about 4 wt %, not greater than about 3 wt %, or not greater than about 2 wt %.

Cyclic Phosphonic Anhydride Compound

In some embodiments, the cyclic phosphonic anhydride according to the present application includes one or more of the compounds of Formula [12]:



wherein in Formula [12], R_{10} , R_{11} , and R_{12} each independently may be hydrogen; each independently may be an alkyl group having 1-20 carbon atoms, for example, an alkyl group having 1-15 carbon atoms, an alkyl group having 1-10 carbon atoms, an alkyl group having 1-5 carbon atoms, an alkyl group having 5-20 carbon atoms, an alkyl group having 5-15 carbon atoms, and an alkyl group having 5-10 carbon atoms; and each independently may be an aryl group having 6-50 carbon atoms, for example, an aryl group having 6-30 carbon atoms, an aryl group having 6-26 carbon atoms, an aryl group having 6-20 carbon atoms, an aryl group having 10-50 carbon atoms, an aryl group having 10-30 carbon atoms, an aryl group having 10-26 carbon atoms, or an aryl group having 10-20 carbon atoms, wherein R_{10} , R_{11} , and R_{12} may be different from or the same as each other, or any two of them are the same.

In some embodiments, the cyclic phosphonic anhydride compound of the present application includes, but is not

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limited to, compounds of following formulae, which may be used alone, or in combination of two or more thereof:

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(TM3P)

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(TE3P)

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(T3P)

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[12]
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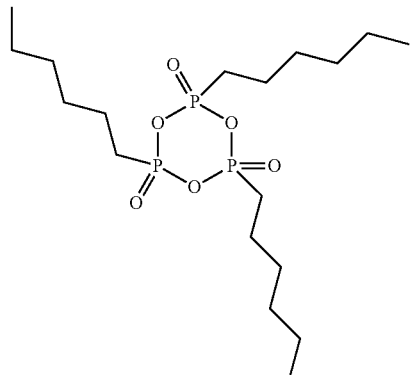
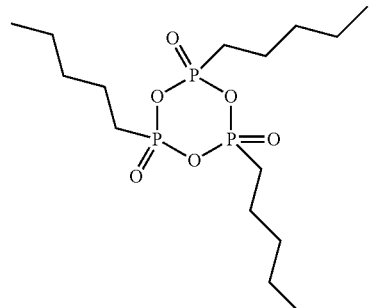
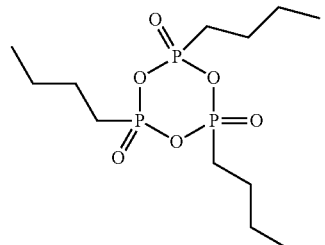
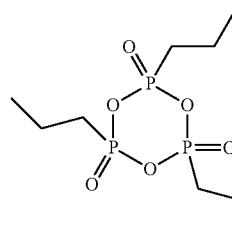
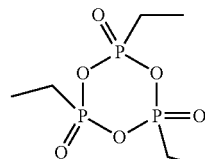
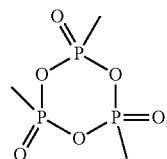


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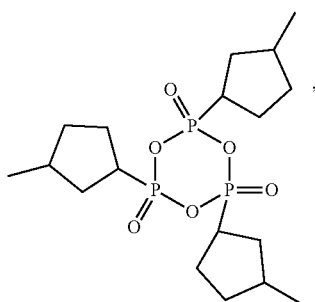
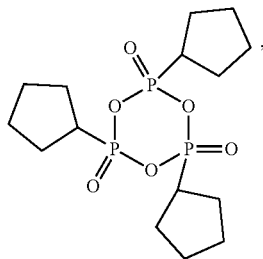
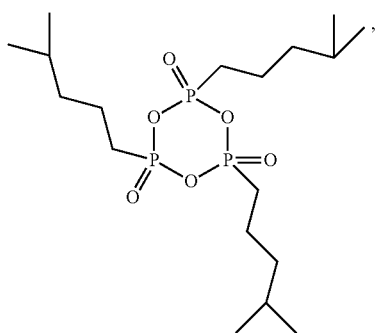
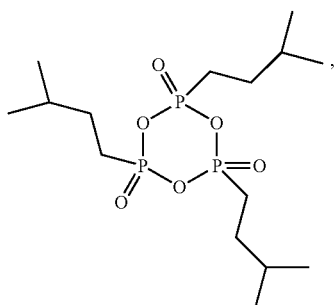
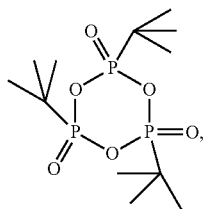
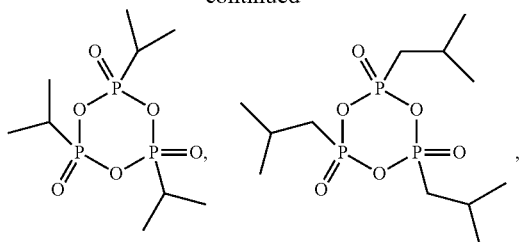
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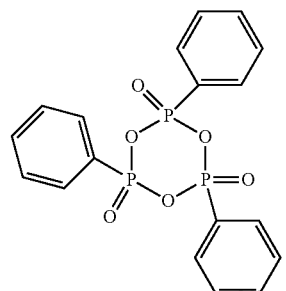
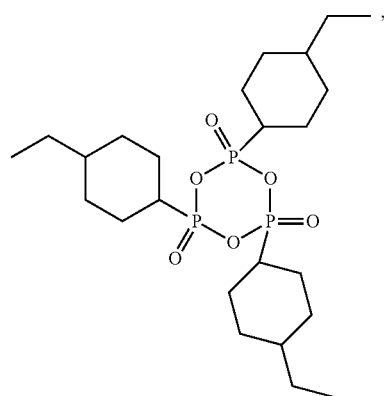
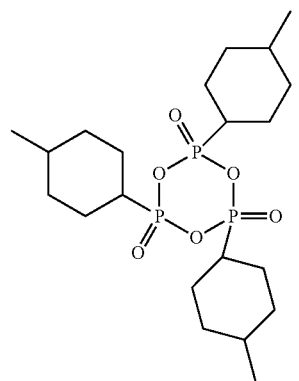
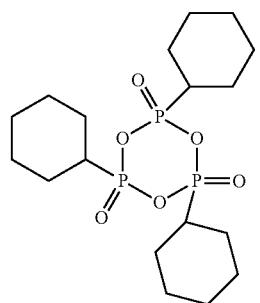
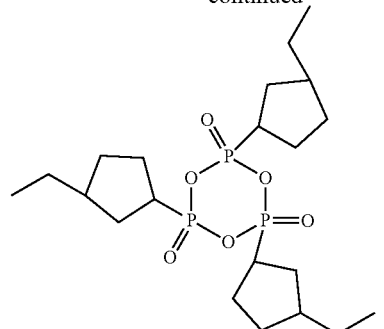
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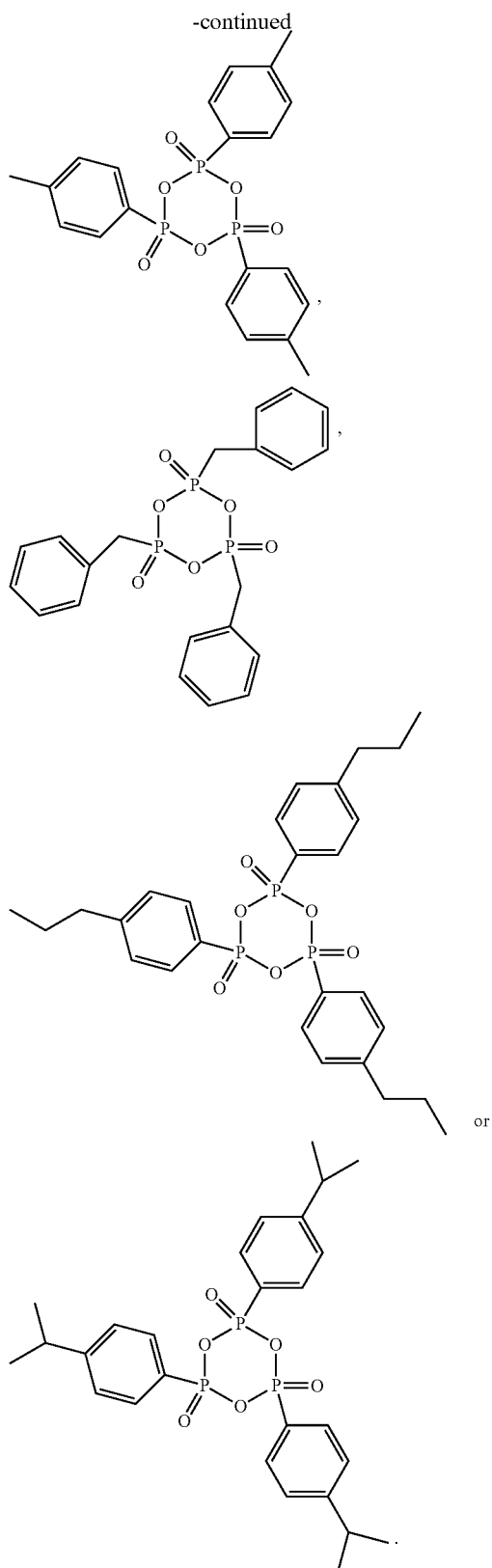
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In some embodiments, based on the total weight of the electrolyte, the content of the cyclic phosphonic anhydride is not less than about 0.01 wt % or not less than about 0.1 wt %. In some embodiments, the content of the cyclic

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phosphonic anhydride is not less than about 0.3 wt % or not less than about 0.5 wt %. In some embodiments, the content of the cyclic phosphonic anhydride is not greater than about 4 wt % or not greater than about 3 wt %.

5 Other Additives

The electrolyte of the present application may further comprises one or more selected from the group consisting of: a cyclic carbonate ester having a carbon-carbon double bond, a fluoro chain carbonate ester, a fluorinated cyclic carbonate ester, or a compound having a sulfur-oxygen double bond.

In some embodiments, the cyclic carbonate ester having a carbon-carbon double bond useful in the present application includes, but is not limited to: vinylene carbonate, methyl vinylene carbonate, ethyl vinylene carbonate, 1,2-dimethyl vinylene carbonate, 1,2-diethyl vinylene carbonate, fluorovinylene carbonate, and trifluoromethylvinylene carbonate; vinyl ethylene carbonate, 1-methyl-2-vinylethylene carbonate, 1-ethyl-2-vinylethylene carbonate, 1-n-propyl-2-vinylethylene carbonate, 1-methyl-2-vinylethylene carbonate, 1,1-divinylethylene carbonate, and 1,2-divinylethylene carbonate; and 1,1-dimethyl-2-methylene ethylene carbonate, and 1,1-diethyl-2-methylene ethylene carbonate.

25 The cyclic carbonate ester having a carbon-carbon double bond may be used alone or in combination of two or more thereof.

In some embodiments, based on the total weight of the electrolyte, the content of the cyclic carbonate ester having a carbon-carbon double bond is not less than about 0.01 wt %, not less than about 0.1 wt %, or not less than about 0.3 wt %. In some embodiments, the content of the cyclic carbonate ester having a carbon-carbon double bond is not less than about 0.5 wt %. In some embodiments, the content of the cyclic carbonate ester having a carbon-carbon double bond is not greater than about 5 wt %, not greater than about 3 wt %, or not greater than about 1 wt %.

In some embodiments, the fluorinated chain carbonate ester according to the present application includes, but is not limited to: fluoromethyl carbonate, difluoromethyl carbonate, trifluoromethyl carbonate, trifluoroethyl carbonate, or bis(trifluoroethyl) carbonate. The fluorinated chain carbonate ester may be used alone or in combination of two or more thereof.

45 In some embodiments, based on the total weight of the electrolyte, the content of the fluorinated chain carbonate ester is not less than about 0.01 wt % or not less than about 0.1 wt %. In some embodiments, the content of the fluorinated chain carbonate ester is not less than about 0.3 wt %.

50 In some embodiments, the content of the fluorinated chain carbonate ester is not less than about 0.5 wt %. In some other embodiments, the content of the fluorinated chain carbonate ester is not greater than about 3 wt % or not greater than about 5 wt %. In some other embodiments, the content of the fluorinated chain carbonate ester is not greater than about 1 wt %.

55 In some embodiments, the fluorinated cyclic carbonate ester according to the present application includes, but is not limited to, fluoroethylene carbonate, 4,4-difluoroethylene carbonate, 4,5-difluoroethylene carbonate, 4-fluoro-4-methylethylene carbonate, 4,5-difluoro-4-methylethylene carbonate, 4-fluoro-5-methylethylene carbonate, 4,4-difluoro-5-methylethylene carbonate, 4-(fluoromethyl)-ethylene carbonate, 4-(difluoromethyl)-ethylene carbonate, 4-(trifluoromethyl)-ethylene carbonate, 4-(fluoromethyl)-4-fluoroethylene carbonate, 4-(fluoromethyl)-5-fluoroethylene carbonate, 4-fluoro-4,5-dimethylethylene carbonate, 4,5-difluoro-4,5-dimethylethylene carbonate, and 4,4-difluoro-4,5-dimethylethylene carbonate.

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5-dimethylethylene carbonate. The fluorinated cyclic carbonate esters may be used alone or in combination of two or more thereof.

In some embodiments, the content of the fluorinated cyclic carbonate ester is not less than about 0.1 wt % based on the total weight of the electrolyte. In some embodiments, the content of the fluoro cyclic carbonate ester is not less than about 0.5 wt %. In some embodiments, the content of the fluorinated cyclic carbonate ester is not less than about 2 wt %. In some embodiments, the content of the fluorinated cyclic carbonate ester is not less than about 4 wt %. In some embodiments, the content of the fluorinated cyclic carbonate ester is not greater than about 15 wt %. In some embodiments, the content of the fluorinated cyclic carbonate ester is not greater than about 10 wt %. In some embodiments, the content of the fluorinated cyclic carbonate ester is not greater than about 8 wt %.

In some embodiments, the compound having a sulfur-oxygen double bond according to the present application includes, but is not limited to: a cyclic sulfate ester, a chain sulfate ester, a chain sulfonate ester, a cyclic sulfonate ester, a chain sulfite ester, a cyclic sulfite ester, a chain sulfone, a cyclic sulfones, and so on. The compound having a sulfur-oxygen double bond may be used alone or in combination of two or more thereof.

In some embodiments, based on the total weight of the electrolyte, the content of the compound having a sulfur-oxygen double bond is not less than about 0.01 wt %. In some embodiments, the content of the compound having a sulfur-oxygen double bond is not less than about 0.1 wt %. In some embodiments, the content of the compound having a sulfur-oxygen double bond is not less than about 0.3 wt %. In some embodiments, the content of the compound having a sulfur-oxygen double bond is not less than about 0.5 wt %. In some other embodiments, the content of the compound having a sulfur-oxygen double bond is not greater than about 5 wt %. In some embodiments, the content of the compound having a sulfur-oxygen double bond is not greater than about 4 wt %. In some embodiments, the content of the compound having a sulfur-oxygen double bond is not greater than about 3 wt %.

In some embodiments, the cyclic sulfate ester according to the present application includes, but is not limited to: 1,2-ethylene glycol sulfate, 1,2-propylene glycol sulfate, 1,3-propylene glycol sulfate, 1,2-butyleneglycol sulfate, 1,3-butyleneglycol sulfate, 1,4-butyleneglycol sulfate, 1,2-pentylene glycol sulfate, 1,3-pentylene glycol sulfate, 1,4-pentylene glycol sulfate, and 1,5-pentylene glycol sulfate. The cyclic sulfate ester may be used alone or in combination of two or more thereof.

In some embodiments, the chain sulfate ester according to the present application includes, but is not limited to: a dialkyl sulfate such as dimethyl sulfate, methyl ethyl sulfate, and diethyl sulfate. The chain sulfate ester may be used alone or in combination of two or more thereof.

In some embodiments, the chain sulfonate ester according to the present application includes, but is not limited to: methyl fluorosulfonate, ethyl fluorosulfonate, methyl methanesulfonate, ethyl methanesulfonate, butyl dimesylate, methyl 2-(methylsulfonyloxy)propionate, ethyl 2-(methylsulfonyloxy)propionate, methyl methylsulfonyloxyacetate, ethyl methylsulfonyloxyacetate, phenyl methylsulfonate, and pentafluorophenyl methylsulfonate. The chain sulfonate ester may be used alone or in combination of two or more thereof.

In some embodiments, the cyclic sulfonate ester according to the present application includes, but is not limited to:

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1,3-propanesultone, 1-fluoro-1,3-propanesultone, 2-fluoro-1,3-propanesultone, 3-fluoro-1,3-propanesultone, 1-methyl-1,3-propanesultone, 2-methyl-1,3-propanesultone, 3-methyl-1,3-propanesultone, 1-propene-1,3-sultone, 2-propene-1,3-sultone, 1-fluoro-1-propene-1,3-sultone, 2-fluoro-1-propene-1,3-sultone, 3-fluoro-1-propene-1,3-sultone, 1-fluoro-2-propene-1,3-sultone, 2-fluoro-2-propene-1,3-sultone, 3-fluoro-2-propene-1,3-sultone, 1-methyl-1-propene-1,3-sultone, 2-methyl-1-propene-1,3-sultone, 3-methyl-1-propene-1,3-sultone, 1-methyl-2-propene-1,3-sultone, 2-methyl-2-propene-1,3-sultone, 3-methyl-2-propene-1,3-sultone, 1,4-butane sultone, 1,5-pentane sultone, methylene methanedisulfonate, and ethylene methanedisulfonate. The cyclic sulfonate ester may be used alone or in combination of two or more thereof.

In some embodiments, the chain sulfite ester according to the present application includes, but is not limited to: dimethyl sulfite, methyl ethyl sulfite, and diethyl sulfite. The chain sulfite ester may be used alone or in combination of two or more thereof.

In some embodiments, the cyclic sulfite ester according to the present application includes, but is not limited to: 1,2-ethylene glycol sulfite, 1,2-propylene glycol sulfite, 1,3-propylene glycol sulfite, 1,2-butyleneglycol sulfite, 1,3-butyleneglycol sulfite, 1,4-butyleneglycol sulfite, 1,2-pentylene glycol sulfite, 1,3-pentylene glycol sulfite, 1,4-pentylene glycol sulfite, and 1,5-pentylene glycol sulfite. The cyclic sulfite ester may be used alone or in combination of two or more thereof.

In some embodiments, the chain sulfone ester according to the present application includes, but is not limited to: a dialkyl sulfone compound such as dimethyl sulfone, and diethyl sulfone.

In some embodiments, the cyclic sulfone according to the present application includes, but is not limited to: sulfolane, methyl sulfolane, 4,5-dimethyl sulfolane, and sulfolene. The cyclic sulfone may be used alone or in combination of two or more thereof.

Electrolyte

The electrolyte used in the electrolyte according to the embodiments of the present application may be an electrolyte known in the prior art, including, but not limited to: an inorganic lithium salt, for example LiClO_4 , LiAsF_6 , LiPF_6 , LiBF_4 , LiSbF_6 , LiSO_3F , and $\text{LiN}(\text{FSO}_2)_2$; a fluorine-containing organic lithium salt, for example LiCF_3SO_3 , $\text{LiN}(\text{FSO}_2)(\text{CF}_3\text{SO}_2)$, $\text{LiN}(\text{CF}_3\text{SO}_2)_2$, $\text{LiN}(\text{C}_2\text{F}_5\text{SO}_2)_2$, cyclic lithium 1,3-hexafluoropropane disulfonimide, cyclic lithium 1,2-tetrafluoroethane disulfonimide, $\text{LiN}(\text{CF}_3\text{SO}_2)(\text{C}_4\text{F}_9\text{SO}_2)$, $\text{LiC}(\text{CF}_3\text{SO}_2)_3$, $\text{LiPF}_4(\text{CF}_3)_2$, $\text{LiPF}_4(\text{C}_2\text{F}_5)_2$, $\text{LiPF}_4(\text{CF}_3\text{SO}_2)_2$, $\text{LiPF}_4(\text{C}_2\text{F}_5\text{SO}_2)_2$, $\text{LiBF}_2(\text{CF}_3)_2$, $\text{LiBF}_2(\text{C}_2\text{F}_5)_2$, $\text{LiBF}_2(\text{CF}_3\text{SO}_2)_2$, and $\text{LiBF}_2(\text{C}_2\text{F}_5\text{SO}_2)_2$; and a lithium salt containing a dicarboxylic acid complex, for example, lithium bis(oxalato)borate, lithium difluoro(oxalato)borate, lithium tris(oxalato)phosphate, lithium difluoro-bis(oxalato)phosphate, and lithiumtetrafluoro(oxalato)phosphate. In addition, the electrolyte may be used alone or in combination of two or more thereof. For example, in some embodiments, the electrolyte includes a combination of LiPF_6 and LiBF_4 . In some embodiments, the electrolyte includes a combination of an inorganic lithium salt such as LiPF_6 or LiBF_4 and a fluorine-containing organic lithium salt such as LiCF_3SO_3 , $\text{LiN}(\text{CF}_3\text{SO}_2)_2$, or $\text{LiN}(\text{C}_2\text{F}_5\text{SO}_2)_2$. In some embodiments, the concentration of the electrolyte is in the range of about 0.8-about 3 mol/L, for example, about 0.8-about 2.5 mol/L, about 0.8-about 2 mol/L, about 1-about 2 mol/L, for example, 1 mol/L, 1.15 mol/L, 1.2 mol/L, 1.5 mol/L, 2 mol/L or 2.5 mol/L.

Solvent

The solvent used in the electrolyte according to the embodiments of the present application may be any non-aqueous solvent known in the art as a solvent for an electrolyte.

In some embodiments, the non-aqueous solvent useful in the present application includes, but is not limited to: a cyclic carbonate ester, a chain carbonate ester, a cyclic carboxylate ester, a chain carboxylate ester, a cyclic ether, a chain ether, a phosphorus-containing organic solvent, a sulfur-containing organic solvent, an aromatic fluorine-containing solvent or any combination thereof.

In some embodiments, the cyclic carbonate ester according to the present application generally has 3-6 carbon atoms, and includes, but is not limited to: for example, ethylene carbonate, propylene carbonate, butylene carbonate, and other cyclic carbonates.

In some embodiments, the chain carbonate ester according to the present application includes, but is not limited to: a chain carbonate ester such as dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, methyl n-propyl carbonate, ethyl n-propyl carbonate, and di-n-propyl carbonate; and a fluorinated chain carbonate ester, for example, bis (fluoromethyl) carbonate, bis(difluoromethyl) carbonate, bis (trifluoromethyl) carbonate, bis(2-fluoroethyl) carbonate, bis(2,2-difluoroethyl) carbonate, bis(2,2,2-trifluoroethyl) carbonate, 2-fluoroethylmethyl carbonate, 2,2-difluoroethylmethyl carbonate, and 2,2,2-trifluoroethylmethyl carbonate.

In some embodiments, the cyclic carboxylate ester according to present application includes, but is not limited to: γ -butyrolactone, γ -valerolactone, and the like, and a cyclic carboxylate ester some hydrogen atoms of which are substituted with fluorine.

In some embodiments, the chain carboxylate ester according to the present application includes, but is not limited to: methyl acetate, ethyl acetate, propyl acetate, isopropyl acetate, butyl acetate, sec-butyl acetate, isobutyl acetate, tert-butyl acetate, methyl propionate, ethyl propionate, propyl propionate, isopropyl propionate, methyl butyrate, ethyl butyrate, propyl butyrate, methyl isobutyrate, ethyl isobutyrate, methyl valerate, ethyl valerate, methyl pivalate and ethyl pivalate or the like, and a chain carboxylate ester some hydrogen atoms of which are substituted with fluorine. The fluorinated chain carboxylate ester includes, but is not limited to: methyl trifluoroacetate, ethyl trifluoroacetate, propyl trifluoroacetate, butyl trifluoroacetate and 2,2,2-trifluoroethyl trifluoroacetate.

In some embodiments, the cyclic ether according to the present application includes, but is not limited to: tetrahydrofuran, 2-methyltetrahydrofuran, 1,3-dioxolane, 2-methyl-1,3-dioxolane, 4-methyl-1,3-dioxolane, 1,3-dioxane, 1,4-dioxane and dimethoxypropane. In some embodiments, the chain ether according to the present application includes, but is not limited to: dimethoxymethane, 1,1-dimethoxyethane, 1,2-dimethoxyethane, diethoxymethane, 1,1-diethoxyethane, 1,2-diethoxyethane, ethoxymethoxymethane, 1,1-ethoxymethoxyethane and 1,2-ethoxymethoxyethane.

In some embodiments, the phosphorus-containing organic solvent according to the present application includes, but is not limited to: trimethyl phosphate, triethyl phosphate, dimethyl ethyl phosphate, methyl diethyl phosphate, ethylene methyl phosphate, ethylene ethyl phosphate, triphenyl phosphate, trimethyl phosphite, triethyl phosphite, triphenyl phosphite, tris(2,2,2-trifluoroethyl) phosphate and tris(2,2,3,3,3-pentafluoropropyl) phosphate.

In some embodiments, the sulfur-containing organic solvent according to the present application includes, but is not limited to: sulfolane, 2-methylsulfolane, 3-methylsulfolane, dimethyl sulfone, diethyl sulfone, ethyl methyl sulfone, methyl propyl sulfone, dimethyl sulfoxide, methyl methanesulfonate, ethyl methanesulfonate, methyl ethanesulfonate, ethyl ethanesulfonate, dimethyl sulfate, diethyl sulfate and dibutyl sulfate, as well as a sulfur-containing organic solvent some hydrogen atoms of which are substituted with fluorine.

In some embodiments, the aromatic fluorine-containing organic solvent according to the present application includes, but is not limited to: fluorobenzene, difluorobenzene, trifluorobenzene, tetrafluorobenzene, pentafluorobenzene, hexafluorobenzene, and trifluoromethylbenzene.

In some embodiments, the non-aqueous solvent of the present application may be used alone, or in combination of two or more thereof.

II. Electrochemical Device

The electrochemical device of the present invention includes any device in which an electrochemical reaction takes place, and specific examples include all kinds of primary batteries, secondary batteries, fuel cells, solar cells, or capacitors. In particular, the electrochemical device is a lithium secondary battery including a lithium metal secondary battery, a lithium ion secondary battery, a lithium polymer secondary battery or a lithium ion polymer secondary battery. In some embodiments, the electrochemical device of the present application is an electrochemical device having a cathode having a cathode active material capable of absorbing and releasing metal ions; an anode having an anode active material capable of absorbing and releasing metal ions, and characterized by comprising any of the electrolytes of the present application.

Electrolyte

The electrolyte used in the electrochemical device of the present application is any of the aforementioned electrolytes according to the present application. Moreover, the electrolyte used in the electrochemical device of the present application may include other electrolytes falling within the scope of present application.

Anode

The anode material used in the electrochemical device according to the embodiments of the present application can be prepared using materials, construction and manufacturing methods well known in the art. For example, the anode of the present application can be prepared using the technique described in U.S. Pat. No. 9,812,739B, the entire contents of which are incorporated herein by reference.

In some embodiments, the anode active material is any substance capable of electrochemically absorbing and releasing a metal ion such as lithium ion. In some embodiments, the anode active material includes a carbonaceous material, a silicon-carbon material, an alloy material or a lithium-containing metal composite oxide material. In some embodiments, the anode active material includes one or more of those described above.

In some embodiments, the anode can be formed by adding a binder and a solvent to, and if necessary, adding a thickener, a conductive material, a filler, or the like the anode active material, to prepare a slurry, coating the slurry to a current collector, drying, and then pressing.

In some embodiments, when the anode includes an alloy-based material, a thin film layer (anode active material layer) as an anode active material can be formed by vapor deposition, sputtering, or plating.

In some embodiments, when the anode includes lithium metal, an anode active material layer is formed from, for

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example, a conductive skeleton of twisted spherical shape and metal particles dispersed in the conductive skeleton, wherein the conductive skeleton of twisted spherical shape may have a porosity of about 5% to about 85%, and a protective layer may be further disposed on the anode active material layer of lithium metal.

Cathode

The cathode material used in the electrochemical device of the present application can be prepared using materials, construction and manufacturing methods well known in the art. In some embodiments, the cathode of the present application can be prepared using the technique described in U.S. Pat. No. 9,812,739B, the entire contents of which are incorporated herein by reference.

In some embodiments, the cathode active material includes, but is not limited to: a sulfide, a phosphate salt compound and a lithium-transition metal composite oxide. In some embodiments, the cathode active material includes a lithium-transition metal compound which has a structure capable of removing and intercalating lithium ions. In some embodiments, the composition of the lithium-transition metal compound can refer to the technical content described in U.S. Pat. No. 9,812,739B.

In some embodiments, the cathode is prepared by forming a cathode material from a cathode active material layer including a lithium-transition metal compound powder and a binder on a current collector.

In some embodiments, the cathode active material layer can generally be produced by: dry mixing a cathode material and a binder (and a conductive material and a thickener, if necessary) to form flakes, pressing the obtained flakes on a cathode current collector or dissolving or dispersing the material in a liquid medium to form a slurry, coating the slurry on a cathode current collector, and drying. In some embodiments, the material of the cathode active material layer includes any material known in the art. In some embodiments, the cathode active material layer includes materials described in U.S. Pat. No. 9,812,739B.

Electrode Compaction Density

The electrochemical device of the present application comprises a cathode and an anode, a separator and an electrolyte. The electrode includes a current collector and a coating on the current collector. The coating includes a single-sided coating, a double-sided coating, or a combination thereof. The single-sided coating is a coating formed by applying a slurry on one surface of the current collector. The double-sided coating is a coating formed by applying a slurry on two opposite surfaces of the current collector.

In some embodiments, as shown in FIG. 2, an electrode A includes a current collector 1 and a coating 2 on the current collector 1. As shown in FIG. 2, the coating 2 is only present on one surface of the current collector, and such coating is a single-sided coating.

In some embodiments, as shown in FIG. 3, an electrode B includes a current collector 1 and a coating 2 on the current collector. As shown in FIG. 3, the coating 2 is present on two opposite surfaces of the current collector, and such coating is a double-sided coating.

In some embodiments, as shown by an electrode C in FIG. 4, one surface of a portion of a current collector 1 is coated with a slurry to form a coating 2 and the two opposite surfaces of the other portion of the current collector 1 are coated with a slurry to form a coating 2. Such coating includes both a single-sided coating and a double-sided coating.

In a wound electrochemical device, the cathode and the cathode are usually each formed by winding an elongated

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electrode, so that both a single-sided coating and a double-sided coating are present on the elongated electrode. In a laminated electrochemical device, the cathode and the anode are usually formed by laminating-shaped electrode s, and there is only a single-sided coating or a double-sided coating on the same electrode. In an electrochemical device in which the wound and laminated electrode s are assembled in combination, the cathode and anode generally comprise an elongated electrode having both a single-sided coating and a double-sided coating, and a -shaped electrode having only a single-sided coating or a double-sided coating.

The electrode has an electrode compaction density. The electrode compaction density is obtained by: measuring the thickness of an electrode using a precise measurement tool, such as a ten-thousandths micrometer; then taking the electrode of a certain area and accurately measuring the area and weight; and calculating the electrode compaction density by a formula below:

$$\text{Electrode compaction density} = (\text{Weight of electrode} - \text{Weight of current collector}) / (\text{Area of electrode} / (\text{Thickness of electrode} - \text{Thickness of current collector}))$$

A lower compaction density makes the porosity higher, making some of the particles in an insulating state, and unable to participate in charge and discharge, thereby resulting in a low specific discharge capacity, thus affecting the performance of the electrochemical device. A too high compaction density may cause difficulty in infiltrability of the electrolyte and decrease in solution retention, such that the cycle and rate performances cannot be guaranteed. Properly controlling the electrode compaction density of the single-sided and double-sided coating is important to obtain electrochemical devices with high capacity density, and excellent cycle and storage performances.

In some embodiments, the electrode with a single-sided coating has an electrode compaction density D1, and an electrode with a double-sided coating has an electrode compaction density D2, and D1 and D2 meet the relationship: about $0.8 \leq D1/D2 \leq$ about 1.2, under this circumstance, the effects of the cathode and anode active materials are well exerted, so that the electrode can obtain good electrical conductivity, which is also important to control the expansion of electrochemical devices. Therefore, the obtained electrochemical device has high capacity density and excellent cycle and storage performances.

In some embodiments, D1 and D2 meet the relationship: about $0.9 \leq D1/D2 \leq$ about 1.1. Under this circumstance, the performance of the electrochemical device can be further improved.

In some embodiments, D1 and D2 meet the relationship: about $0.9 \leq D1/D2 \leq$ about 1.1.

In some embodiments, D1 and D2 meet the relationship: about $0.95 \leq D1/D2 \leq$ about 1.05. Under this circumstance, the distributions of pores and pore sizes in the single-sided coating and the double-sided coating are more uniform, the distributions of the conductive agent and the binder are more uniform, the contact resistance and charge exchange resistance of the electrode are lowered, and the active area capable of participating in the reaction is increased, thereby significantly improving the electrochemical performance of the material and further improving the performances of the electrochemical device.

In some embodiments, the electrode may be a cathode or an anode. When the electrode is a cathode, about $3.5 \text{ g/cm}^3 \leq D2 \leq$ about 4.3 g/cm^3 . When D2 is within this range, the cathode can have good electrical conductivity, and the effect of the cathode active material can be well exerted.

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When the electrode is an anode, about $1.2 \text{ g/cm}^3 \leq D2 \leq$ about 1.8 g/cm^3 . When D2 is within this range, the anode can have a higher breaking strength, thereby preventing the electrode particles from falling off during the cycle.

Separator

In some embodiments, the electrochemical device of the present application is provided with a separator between the positive electrode and the negative electrode to prevent short circuit. The material and shape of the separator used in the electrochemical device of the present application are not particularly limited, and may be any of the techniques disclosed in the prior art. In some embodiments, the separator includes a polymer or an inorganic substance or the like formed from a material which is stable to the electrolyte of the present application.

For example, the separator may include a substrate layer and a surface treatment layer. The substrate layer is a non-woven fabric, film, or composite film having a porous structure, and the material of the substrate layer is at least one selected from polyethylene, polypropylene, polyethylene terephthalate, and polyimide. Particularly, a porous polypropylene film, a porous polyethylene film, a polypropylene nonwoven fabric, a polyethylene nonwoven fabric, or a porous polypropylene-polyethylene-polypropylene composite film may be used.

At least one surface of the substrate layer is provided with a surface treatment layer, which may be a polymer layer or an inorganic layer, or a layer formed by mixing a polymer and an inorganic substance.

The inorganic layer comprises inorganic particles and a binder. The inorganic particles are one or more selected from alumina, silica, magnesia, titanium oxide, hafnium dioxide, tin oxide, cerium dioxide, nickel oxide, zinc oxide, calcium oxide, zirconia, yttria, silicon carbide, boehmite, aluminum hydroxide, magnesium hydroxide, calcium hydroxide and barium sulfate. The binder is one or more selected from polyvinylidene fluoride, a copolymer of vinylidene fluoride-hexafluoropropylene, a polyamide, polyacrylonitrile, a polyacrylate ester, polyacrylic acid, a polyacrylate salt, polyvinylpyrrolidone, polyvinyl ether, polymethyl methacrylate, polytetrafluoroethylene, and polyhexafluoropropylene. The polymer layer contains a polymer, and the material of the polymer includes at least one of a polyamide, polyacrylonitrile, a polyacrylate ester, polyacrylic acid, a polyacrylate salt, polyvinylpyrrolidone, polyvinyl ether, polyvinylidene fluoride or poly(vinylidene fluoride-hexafluoropropylene).

III. Application

At a high voltage, the oxidizability of the cathode material is increased, and the stability is lowered, which makes the electrolyte easily decompose on the surface of the positive electrode or results in deterioration of the materials of the electrochemical device, so that the capacity of the electrochemical device is decreased. Prior to the present application, the primary solution is to add a film-forming additive to the electrolyte. However, doing so will cause an increase in the DC internal resistance of the battery, thereby resulting in a decrease in the cycle performance and a decrease in the capacity retention rate.

The electrolyte according to the embodiments of the present application can be used to inhibit the increase in DC internal resistance of an electrochemical device, and thus applicable to an electronic device comprising the electrochemical device.

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The use of the electrochemical device according to the present application is not particularly limited, and can be used in various known applications, such as notebook computers, pen-input computers, mobile computers, e-book players, portable phones, portable fax machines, portable copiers, portable printers, head-mounted stereo headphones, video recorders, LCD TVs, portable cleaners, portable CD players, Mini discs, transceivers, electronic notebooks, calculators, memory cards, portable recorders, radios, backup power sources, motors, vehicles, motorcycles, motorbicycle, bicycles, lighting apparatus, toys, game consoles, clocks, electric tools, flashing light, cameras, large batteries for household use, or lithium ion capacitors.

EXAMPLES

Hereinafter, the present application will be specifically described by way of examples and comparative examples; however, the present application is not limited thereto as long as they do not deviate from the spirit of the present application.

1. Preparation of Lithium-Ion Battery

(1) Preparation of an Anode

Graphite, conductive carbon black (Super-P), styrene-butadiene rubber, and sodium carboxymethyl cellulose (CMC) were mixed at a weight ratio of about 95:2:2:1 in deionized water as a solvent, and stirred uniformly, to obtain an anode slurry. The slurry was coated on a copper foil having a thickness of about $12 \mu\text{m}$, dried, cold pressed, and cut, and then a tab was welded, to obtain an anode.

(2) Preparation of a Cathode

Lithium cobalt oxide (LiCoO_2), conductive carbon black (Super-P), and polyvinylidene fluoride (PVDF) were mixed at a weight ratio of about 95:2:3 in N-methylpyrrolidone as a solvent, and stirred uniformly, to obtain a cathode slurry. The slurry was coated on an aluminum foil having a thickness of about $12 \mu\text{m}$, dried, cold pressed, and cut, and then a tab was welded, to obtain a cathode.

(3) Preparation of an Electrolyte:

Under a dry argon atmosphere, EC, PC, and DEC (at a weight ratio of about 1:1:1) were mixed, and then LiPF_6 was added and mixed uniformly, to form a basic electrolyte, in which the concentration of LiPF_6 is 1.15 mol/L . Different amounts of substances shown in Tables 1-1 to 9 were added to the basic electrolyte to obtain the electrolytes of different examples and comparative examples, wherein the contents of each substance in the electrolyte described below were obtained based on the total weight of the electrolyte.

(4) Preparation of a Separator

A porous PE polymer film was used as a separator.

(5) Preparation of Lithium-Ion Battery

The obtained cathode, anode and separator were wound in sequence and placed in the outer packaging foil, with a liquid injection port being left. The electrolyte was injected via the liquid injection port, encapsulated, formed and capacity graded to obtain a lithium ion battery. The fresh battery herein refers to a battery which is obtained through the aforementioned preparation process and is ready for shipping.

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2. Test Methods of Battery Performance

(1) Test Method for Change in DC Internal Resistance at 20% SOC

At 25° C., the battery was charged to 4.45 V at a constant current of 1 C (nominal capacity) and then charged to a current of ≤ 0.05 C at a constant voltage of 4.45 V, allowed to stand for 5 min, and discharged to a cut-off voltage of 3 V at a constant current of 1 C. The actual discharge capacity was recorded. The battery was adjusted to 50% of the full charge capacity required with the actual capacity, continuously discharged for 10 s at a current of 0.3 C. The DC internal resistances (DCIRs) (average of the measurements from 15 batteries) after one cycle, 200 cycles and 400 cycles was obtained by dividing the current with the difference between the voltage before discharge and the voltage at the end of discharge.

(2) Test Method for Storage Performance at High Temperature of 60° C.

The battery was allowed to stand at 25° C. for 30 minutes, charged to 4.45 V at a constant current of 0.5 C and then charged to 0.05 C at a constant voltage of 4.45 V, allowed to stand for 5 minutes, and then stored at 60° C. for 21 days. The thickness expansion rate of the battery was measured and calculated by a formula below:

$$\text{Thickness expansion rate} = \frac{(\text{Thickness after storage} - \text{Thickness before storage})}{\text{Thickness before storage}} \times 100\%$$

(3) Test Method for Battery Capacity Retention Rate

At 45° C., the battery was charged to 4.45 V at a constant current of 1 C and then charged to a current of 0.05 C at a constant voltage, and discharged to 3.0 V at a constant current of 1 C, the above being the first cycle. Multiple cycles were performed on the battery under the above conditions. The capacity retention rate of the fresh battery and the battery after 200 cycles and 400 cycles was calculated respectively. The capacity retention rate after cycles is calculated by a formula below:

$$\text{Capacity retention rate after the cycle} = \frac{(\text{Discharge capacity after corresponding cycles} / \text{Discharge capacity after the first cycle}) \times 100\%}{}$$

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(4) Test Method for Voltage Drop

At 25° C., the battery was charged to 4.45 V at a constant current of 1 C and then charged to a current of 0.05 C at a constant voltage, discharged to 3.2 V at a constant current of 1 C, and allowed to stand for 5 minutes. Then the voltage was tested, and after 24 h's storage at 85° C., the voltage was tested again. Voltage drop = Voltage before storage - Voltage after storage.

(5) Intermittent Cycle Test Method

At 50° C., the lithium ion battery was charged to 4.45 V at a constant voltage of 0.5 C and then to a cut-off current of 0.05 C at a constant current, allowed to stand for 20 h, and then discharged to 3.0 V at a constant current of 0.5 C. Repeating charge/discharge as above, the capacity retention rate of the fresh battery, and the battery after 30, 50, and 100 cycles was calculated respectively. Capacity retention rate after cycles = (Discharge capacity after corresponding cycles / Discharge capacity of the first cycle) $\times 100\%$

3. Test Results

The abbreviations for the chemical materials used in the examples of present application are shown in the following two tables:

Material	Abbreviation	Material Name	Abbreviation
Ethylene carbonate	EC	1,3-propanesultone	PS
Propylene carbonate	PC	Ethylene sulfate	DTD
Ethyl methyl carbonate	EMC	Butanedinitrile	SN
Dimethyl carbonate	DMC	Adiponitrile	ADN
Diethyl carbonate	DEC	1,4-dicyano-2-butene	DCB
γ -butyrolactone	GBL	1,3,6-Hexanetricarbonitrile	HTCN-1
γ -valerolactone	VL	1,2,6-Hexanetricarbonitrile	HTCN-2
Ethyl propionate	EP	1,3,5-Pentanetricarbonitrile	PTCN
Propyl propionate	PP	Ethylene glycol bis(2-cyanoethyl) ether	EDN
		1,1-difluoro-2,2-difluoroethyl-2',2'difluoro-3',3'-difluoropropyl ether	FEPE
		2-trifluoromethyl-3-methoxyperfluoropentane	TMMP
		2-(trifluoro-2-fluoro-3-difluoro)-3-difluoro-4-fluoro-5-trifluoropentane	TPTP
Vinylene carbonate	VC	1-Propylphosphonic cyclic anhydride	T3P
Fluoroethylene carbonate	FEC	1-Methylphosphonic cyclic anhydride	TM3P
		1-Ethylphosphonic cyclic anhydride	TE3P
		1,2,3-tris(2-cyanoethoxy)propane	TCEP

Dinitrile compound	A	A ₁ : SN; A ₂ : ADN; A ₃ : EDN; A ₄ : DCB
Trinitrile compound	B	B ₁ : HTCN-1; B ₂ : HTCN-2; B ₃ : PTCN; B ₄ : TCEP
Propyl propionate (PP)	C	
Fluoroether	D	D ₁ : FEPE; D ₂ : TMMP; D ₃ : TPTP
Cyclic phosphonic anhydride	E	E ₁ : T3P E ₂ : TM3P; E ₃ : TE3P
Other additives	F	F ₁ : VC; F ₂ : PS; F ₃ : DTD
Cyclic carboxylate ester	H	H ₁ : GBL; H ₂ : VL
Single- and double-side compaction density	G	

(1) Different amounts of a compound comprising two cyano groups, a compound comprising three cyano groups and/or propyl propionate as shown in Tables 1-1 and 1-2 were added to the basic electrolyte. The electrolyte was

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injected into a battery prepared according to the above method. The DC internal resistance at 20% SOC of the batteries was tested. The test results are shown in Tables 1-1 and 1-2.

TABLE 1-1

	A ₁ (X wt %)	B ₁ (Y wt %)	C (Z wt %)	X + Y	X/Y	DC internal resistance (mΩ) at 20% SOC		
						Fresh Battery	After 200 cycles	After 400 cycles
S1-1	4	1	30	5	4	50.8	84.3	91.2
S1-2	4	2	30	6	2	54.4	88.0	98.6
S1-3	4	3	30	7	1.3	55.7	88.9	99.4
S1-4	4	0.5	30	4.5	8	57.8	92.0	101.0
S1-5	3	0.5	30	3.5	6	57.1	91.2	93.5
S1-6	4	0.4	30	4.2	10	55.9	99.4	112.8
S1-7	3	2	30	5	1.5	52.3	84.4	91.5
S1-8	2	2	30	4	1	51.9	84.2	91.4
S1-9	1	2	30	3	0.5	52.3	86.5	92.4
S1-10	0.5	2	30	2.5	0.25	54.8	79.4	102.5
S1-11	0.2	2	30	2.2	0.1	59.2	91.8	105.3
S1-12	1	5	30	6	0.2	58.5	97.0	104.1
S1-13	7	3	30	10	2.3	60.4	92.9	107.3
D1-1	0	0	0	0	0	63.8	140.9	190.4
D1-2	0	1	0	1	0	62.3	138.5	188.2
D1-3	4	0	0	4	0	61.8	136.5	184.2
D1-4	0.1	1	0	1.1	0.1	63.8	120.9	126.4
D1-5	4	0.1	0	4.1	40	68.3	109.8	124.9
D1-6	7	5	0	12	1.4	69.2	112.3	133.0
D1-7	1	11	0	12	0.09	63.8	100.9	139.3
D1-8	11	1	0	12	10	72.0	116.3	135.4

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TABLES 1-2

	A (X wt %)	B (Y wt %)	C (Z wt %)	X + Y	X/Y	DC internal resistance (mΩ) at 20% SOC		
						Fresh Battery	After 200 cycles	After 400 cycles
S1-14	A ₂ (4)	B ₂ (3)	30	7	1.3	55.5	88.1	98.7
S1-15	A ₂ (4)	B ₂ (0.5)	30	4.5	8	57.2	91.6	100.6
S1-16	A ₂ (3)	B ₂ (0.5)	30	3.5	6	55.9	89.4	98.8
S1-17	A ₃ (3)	B ₃ (2)	30	5	1.5	51.1	82.6	90.7
S1-18	A ₃ (2)	B ₃ (2)	30	4	1	51.2	83.3	90.5
S1-19	A ₃ (1)	B ₃ (2)	30	3	0.5	51.3	85.1	91.1
S1-20	A ₃ (0.5)	B ₄ (2)	30	2.5	0.25	54.3	79.1	90.1
S1-21	A ₂ (0.2)	B ₂ (2)	30	2.2	0.1	58.5	91.2	104.8
S1-22	A ₂ (1)	B ₂ (5)	30	6	0.2	58.1	96.5	103.3
S1-23	A ₂ (7)	B ₂ (3)	30	10	2.3	60.2	92.3	106.5
S1-24	A ₄ (2)	B ₃ (2)	30	4	1	57.9	86.1	93.4

As shown in Table 1-1, in Comparative Examples D1-1 to D1-8, butanedinitrile or 1,3,6-hexanetricarbonitrile was added alone, and no propyl propionate was added. The DC internal resistance of the battery was significantly increased after cycles.

When butanedinitrile, 1,3,6-hexanetricarbonitrile and propyl propionate were used in Examples S1-1 to S1-13 simultaneously, the increase in DC internal resistance of the battery after cycles was obviously inhibited.

When the propyl propionate content is kept unchanged, and the relation between the butanedinitrile content X and the 1,3,6-hexanetricarbonitrile content Y is adjusted, it is found that when X and Y meet the conditions represented by both Formula (1) and Formula (2): {about 2 wt % ≤ (X+Y) ≤ about 11 wt % . . . (1); and about 0.1 ≤ (X/Y) ≤ about 8 . . . (2)}, the inhibition effect on the increase in DC internal resistance of the battery after cycles is remarkable. When X

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and Y meet about 2 wt % ≤ (X+Y) ≤ about 8 wt % and about 0.1 ≤ (X/Y) ≤ about 6, the combination of the additives can exhibit synergistic effect and can sufficiently suppress the increase in DC internal resistance.

FIG. 1a shows the DC internal resistances of fresh batteries of Examples S1-1 and S1-2 of the present application and Comparative Example D1-1 at different charge states (10% SOC, 20% SOC, and 70% SOC). FIG. 1b shows the DC internal resistances of batteries of Examples S1-1 and S1-2 of the present application and Comparative Example D1-1 at different charge states (10% SOC, 20% SOC, and 70% SOC) after 200 cycles of charge and discharge. FIG. 1c shows the DC internal resistances of batteries of Examples S1-1 and S1-2 of the present application and Comparative Example D1-1 at different charge states (10% SOC, 20% SOC, and 70% SOC) after 400 cycles of charge and discharge. It can be seen that the increase in DC internal resistance during cycles in Examples S1-1 and S1-2 is significantly improved compared with Comparative Example D1-1.

As shown in Table 1-2, when the dinitrile compound is adiponitrile or ethylene glycol bis(2-cyanoethyl) ether, and

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the trinitrile compound is 1,2,6-hexanetricarbonitrile, 1,3,6-Hexanetricarbonitrile, or 1,2,3-tris(2-cyanoethoxy)propane, the effect of inhibiting the increase in DC internal resistance can also be achieved. When the dinitrile compound is ethylene glycol bis(2-cyanoethyl)ether, or the trinitrile compound is 1,2,3-tris(2-cyanoethoxy)propane, the effect of inhibiting the increase in DC internal resistance of the battery after cycles is more significant.

(2) Different amounts of a compound comprising two cyano groups, a compound comprising three cyano groups and/or propyl propionate as shown in Table 2 were added to the basic electrolyte. The electrolyte was injected into a battery prepared according to the above method. The DC internal resistance at 20% SOC of the battery was tested. The test results are shown in Table 2.

TABLE 2

	C (Z wt %)	A ₁ (X wt %)	B ₁ (Y wt %)	Y/Z	DC internal resistance (mΩ) at 20% SOC		
					Fresh battery	After 200 cycles	After 400 cycles
S1-1	30	4	1	0.03	50.8	84.3	91.2
S2-1	5	4	1	0.2	68.7	85.2	97.6
S2-2	10	4	1	0.1	60.3	83.1	95.7
S2-3	20	4	1	0.05	56.1	81.6	93.9
S2-4	40	4	1	0.025	47.5	78.2	87.2
S2-5	50	4	1	0.02	56.3	82.6	98.3
S2-6	10	4	3	0.3	64.3	89.5	97.7
S2-7	50	4	0.5	0.01	53.2	84.3	94.1
D2-1	4	4	1	0.25	72.1	108.5	118.9
D2-2	60	4	1	0.017	42.0	100.1	113.9
D2-3	10	4	5	0.5	62.3	85.4	111.3
D2-4	50	4	0.1	0.0025	53.2	89.3	109.4

As shown in Table 2, when the content of propyl propionate is within the range of 5 wt % to 50 wt %, the DC internal resistance of the fresh battery decreases continuously with the propyl propionate content increasing. In addition, when the content of the propyl propionate is less than 5 wt % or greater than 50 wt %, the DC internal resistance of the battery is greatly increased after 200 cycles, and the performance is deteriorated. The combination of additives can form a cathode protection film so as to reduce the side reactions, thereby effectively controlling the polarization and side reactions of the battery. The ratio of the content of the trinitrile compound to the content of propyl propionate has great effect on the change in DC internal resistance of the battery. When Y/Z is within the range of 0.01-0.3, a better inhibition effect on the increase in DC internal resistance is achieved.

(3) To the basic electrolyte, 30 wt % of propyl propionate, 4 wt % of butanedinitrile (A1), 1 wt % of 1,3,6-hexanetricarbonitrile (B1) and a fluoroether of different contents as shown in Table 3 were added. The obtained electrolyte was

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injected to a battery prepared according to the above method. The DC internal resistance at 20% SOC and the 21-day thickness expansion rate at 60° C. were tested. The test results are shown in Table 3.

TABLE 3

	D (content, wt %)	DC internal resistance (mΩ) of fresh battery at 20% SOC	21-day thickness expansion rate at 60° C.
S1-1	0	50.8	7.6%
S3-1	D ₁ (0.1)	50.2	6.2%
S3-2	D ₁ (0.5)	49.8	6.0%
S3-3	D ₁ (1)	48.6	5.7%
S3-4	D ₁ (1.5)	48.5	5.1%
S3-5	D ₁ (2)	48.4	4.8%
S3-6	D ₁ (2.5)	48.3	4.9%
S3-7	D ₁ (3)	48.1	5.6%
S3-8	D ₁ (5)	49.3	6.8%
S3-9	D ₂ (1)	49.2	6.1%

TABLE 3-continued

	D (content, wt %)	DC internal resistance (mΩ) of fresh battery at 20% SOC	21-day thickness expansion rate at 60° C.
S3-10	D ₃ (1)	49.4	5.7%
D3-1	D ₁ (6)	51.3	8.5%

As can be seen from Table 3, with the addition of fluoroether (0.1-5 wt %), the DC internal resistance and the thickness expansion rate of the fresh battery are improved to a certain extent, and the storage performance is slightly decreased when the addition amount is large, but it is still within the desired range. When the fluoroether content is more than 5 wt %, the DC internal resistance and the thickness expansion rate after storage at 60° C. of the fresh battery are deteriorated.

(4) To the basic electrolyte, 30 wt % of propyl propionate, 4 wt % of butanedinitrile (A1), 1 wt % of 1,3,6-hexanetricarbonitrile (B1) and a cyclic phosphonic anhydride of different contents as shown in Table 4 were added. The obtained electrolyte was injected to a battery prepared according to the above method. The DC internal resistance at 20% SOC and the capacity retention ratio of the battery were tested. The test results are shown in Table 4.

TABLE 4

E (content, wt %)	DC internal resistance (mΩ) at 20% SOC			Capacity retention rate		
	Fresh battery	After 200 cycles	After 400 cycles	Fresh battery	After 200 cycles	After 400 cycles
S1-1 0	50.8	84.3	91.2	100%	93.5%	86.3%
S4-1 E ₁ (0.1)	48.3	51.5	88.2	100%	95.7%	90.1%
S4-2 E ₁ (0.5)	45.4	48.2	82.3	100%	96.3%	91.4%
S4-3 E ₁ (1)	44.6	47.1	80.5	100%	96.1%	90.7%
S4-4 E ₁ (2)	45.6	47.3	81.6	100%	95.2%	89.6%
S4-5 E ₁ (3)	45.9	49.1	89.4	100%	94.7%	88.5%
S4-6 E ₂ (1)	45.2	48.3	88.3	100%	95.3%	91.0%
S4-7 E ₃ (1)	45.3	48.7	89.5	100%	95.1%	90.8%
D4-1 E ₁ (4)	46.1	58.6	105.3	100%	92.1%	85.3%

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It can be seen from Table 4 that with the addition of cyclic phosphonic anhydride (0.1-3 wt %), the DC internal resistance of the fresh battery and the battery after cycles is improved to a certain extent, which may be attributed to the relatively stable structure of the composite protective film formed by the combined additives; and the cycle performance is improved first and then deteriorated with the addition of the cyclic phosphonic anhydride. When the content is more than 3 wt %, the cycle performance is affected, which may be caused by the decomposition of the cyclic phosphonic anhydride.

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(5) To the basic electrolyte, 30 wt % of propyl propionate, 4 wt % of butanedinitrile (A1), 1 wt % of 1,3,6-hexanetrinitrile (B1) and a fluoroether and a cyclic phosphonic anhydride of different contents as shown in Table 5 were added. The obtained electrolyte was injected to a battery prepared according to the above method. The DC internal resistance at 20% SOC, the 21-day thickness expansion rate at 60° C., and the capacity retention rate of the battery were tested. The test results are shown in Table 5.

TABLE 5

	D ₁ (content: wt %)	E ₁ (content: wt %)	DC internal resistance (mΩ) at 20% SOC			21-day thickness expansion rate at 60° C.	Capacity retention rate		
			Fresh battery	After 200 cycles	After 400 cycles		Fresh battery	After 200 cycles	After 400 cycles
S1-1	0	0	50.8	84.3	91.2	7.6%	100%	93.5%	86.3%
S3-1	0.1	0	50.2	83.1	90.4	6.2%	100%	94.2%	87.9%
S4-2	0	0.5	45.4	48.2	82.3	6.7%	100%	96.3%	91.4%
S5-1	0.1	0.5	44.7	47.2	80.8	6.0%	100%	96.4%	91.6%
S5-2	1	0.5	43.8	46.3	80.5	5.3%	100%	96.8%	91.7%
S5-3	1	0.3	44.1	48.2	79.3	4.3%	100%	97.1%	92.3%
S5-4	1	0.1	46.3	50.5	86.2	4.5%	100%	97.6%	92.9%
S5-5	3	2	43.5	47.1	81.3	4.0%	100%	94.8%	89.4%

As can be seen from Table 5, with the addition of the fluoroether and the cyclic phosphonic anhydride, the DC internal resistance, the storage performance and the cycle performance of the fresh battery and the battery after cycles are further improved.

(6) To the basic electrolyte, 30 wt % of propyl propionate, 4 wt % of butanedinitrile (A1), 1 wt % of 1,3,6-hexanetrinitrile (B1) and other additives of different contents as shown in Table 6 were added. The obtained electrolyte was injected to a battery prepared according to the above method. The voltage drop after storage at 3.2V and 85° C. for 24 h of the battery were tested. The test results are shown in Table 6.

TABLE 6

	VC (content: wt %)	VEC (content: wt %)	FEC (content: wt %)	PS (content: wt %)	DTD (content: wt %)	Voltage drop after storage at 3.2 V and 85° C. for 24 hrs
S6-1	0.5	—	—	—	—	0.35 V
S6-2	0.5	—	—	3	—	0.32 V
S6-3	0.5	—	—	3	0.5	0.23 V
S6-4	1	—	—	3	—	0.3 V
S6-5	1	—	3	3	—	0.20 V
S6-6	—	0.5	—	3	0.5	0.25 V
D6-1	—	—	—	—	—	0.4 V

Exhibit 2011

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The addition of the film-forming additives VC, VEC, FEC, PS, DTD can further improve the stability of the solid electrolytic interface (SEI) film of the battery. Using a combination of various additives is more conducive to improve the stability of the battery, which is beneficial to the long-term storage of the battery and thus improves the battery reliability.

(7) To the basic electrolyte, 30 wt % of propyl propionate, 4 wt % of butanedinitrile (A1), 1 wt % of 1,3,6-hexanetri-carbonitrile (B1) were added. The obtained electrolyte was injected to a battery prepared according to the above method. The electrode compaction density ratio (D1/D2) of the battery was changed according to Table 7, and the capacity retention rate of the battery was tested. The test results are shown in Table 7.

TABLE 7

	Compaction density ratio (D1/D2)	Capacity retention rate		
		Fresh battery	After 200 cycles	After 400 cycles
S7-1	0.8	100%	93.8%	89.3%
S7-2	0.9	100%	95.3%	90.1%
S7-3	0.95	100%	95.9%	90.8%
S7-4	1.0	100%	96.8%	91.4%
S7-5	1.05	100%	95.8%	90.7%
S7-6	1.1	100%	95.2%	90.2%
S7-7	1.2	100%	93.7%	88.8%
D7-1	1.3	100%	91.2%	83.6%
D7-1	0.7	100%	90.5%	82.7%

As can be seen from Table 7, the electrode compaction density ratio (D1/D2) of the lithium ion battery has a significant effect on the cycle performance of the lithium ion battery. A too large or too small D1/D2 will damage the cycle performance of lithium-ion batteries. It can be seen that when D1/D2 is within the range of 0.8-1.2, a better cycle performance is obtained, possibly because the combined additives facilitate the reduction of the electrode interface resistance and the reduction of the cell polarization.

(8) To the basic electrolyte, 30 wt % of propyl propionate, 4 wt % of butanedinitrile (A1), 1 wt % of 1,3,6-hexanetri-carbonitrile (B1) and a cyclic phosphonic anhydride of different contents as shown in Table 8 were added. The obtained electrolyte was injected to a battery prepared according to the above method. The electrode compaction density ratio (D1/D2) was changed according to Table 8, and the capacity retention rate of the obtained battery was tested. The test results are shown in Table 8.

TABLE 8

	D ₁ (content:)	E ₁ (content:)	G	Capacity retention rate		
				Fresh battery	After 200 cycles	After 400 cycles
S8-1	1	—	0.8	100%	94.9%	90.1%
S8-2	1	—	1.0	100%	97.3%	92.5%
S8-3	1	—	1.2	100%	94.8%	89.9%
S8-4	—	0.3	0.8	100%	94.3%	89.6%
S85	—	0.3	1.0	100%	97.9%	92.2%
S8-6	—	0.3	1.2	100%	95.0%	90.5%
S8-7	1	0.3	1.0	100%	98.1%	93.3%

With the addition of the fluoroether or the cyclic phosphonic anhydride, the cycle performance of the lithium ion battery is further improved when the electrode compaction density of the battery is within the range of 0.8-1.2.

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(9) To the basic electrolyte, 30 wt % of propyl propionate, 4 wt % of butanedinitrile (A1), 1 wt % of 1,3,6-hexanetri-carbonitrile (B1) and a cyclic carboxylate ester of different contents as shown in Table 9 were added. The obtained electrolyte was injected to a battery prepared according to the above method. The capacity retention rate after intermittent cycle of the obtained battery was tested. The test results are shown in Table 9.

TABLE 9

	H (content: wt %)	Capacity retention rate after intermittent cycle			
		Fresh battery	After 30 cycles	After 50 cycles	After 100 cycles
S1-1	0	100%	73.80%	64.30%	54.50%
S9-1	H ₁ (1)	100%	82.50%	74.60%	64.20%
S9-2	H ₁ (10)	100%	87.20%	81.40%	77.20%
S9-3	H ₁ (20)	100%	86.20%	81.30%	78.00%
S9-4	H ₁ (30)	100%	88.30%	83.30%	79.80%
S9-5	H ₁ (40)	100%	87.50%	80.60%	75.20%
S9-6	H ₁ (50)	100%	86.30%	74.40%	63.20%
S9-7	H ₁ (60)	100%	85.20%	71.60%	57.60%
S9-8	H ₂ (1)	100%	81.80%	73.20%	61.80%
S9-9	H ₂ (10)	100%	86.50%	80.80%	76.10%
S9-10	H ₂ (20)	100%	85.30%	81.30%	77.10%
S9-11	H ₂ (30)	100%	88.10%	83.00%	79.30%
S9-12	H ₂ (40)	100%	87.10%	80.10%	74.80%
S9-13	H ₂ (50)	100%	85.20%	73.70%	61.90%
S9-14	H ₂ (60)	100%	82.20%	70.10%	55.90%

The capacity retention rate after intermittent cycle of the battery increases with the increase of the amount of the cyclic carboxylate ester, possibly because the cyclic carboxylate forms a passivated film on the surface of the cathode. However, when the cyclic carboxylate ester content is close to 40 wt %, the intermittent cycle performance deteriorates to a certain extent, which is mainly caused by a side reaction between LiPF₆ and the cyclic carboxylate ester. Therefore, the amount of the cyclic carboxylate ester is preferably moderate and should not be too large.

References throughout the specification to “some embodiments”, “partial embodiments”, “one embodiment”, “another example”, “example”, “specific example” or “partial examples” mean that at least one embodiment or example of the application includes specific features, structures, materials or characteristics described in the embodiments or examples. Thus, the descriptions appear throughout the specification, such as “in some embodiments”, “in an embodiment”, “in one embodiment”, “in another example”, “in an example”, “in a particular example” or “for example”, are not necessarily the same embodiment or example in the application. Furthermore, the particular features, structures, materials or characteristics herein may be combined in any suitable manner in one or more embodiments or examples.

While the illustrative embodiments have been shown and described, it will be understood by those skilled in the art that the embodiments are not to be construed as limiting the present invention, and modifications, substitutions and changes can be made to the embodiments without departing from the spirit and scope of the present application.

The above-described embodiments of the present invention are intended to be illustrative only. Numerous alternative embodiments may be devised by persons skilled in the art without departing from the scope of the following claims.

The above-described embodiments of the present invention are intended to be illustrative only. Numerous alternative embodiments may be devised by persons skilled in the art without departing from the scope of the following claims.

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What is claimed is:

1. An electrolyte, comprising a dinitrile compound, a trinitrile compound, and propyl propionate, wherein, based on a total weight of the electrolyte, a weight percentage of the dinitrile compound is X and a weight percentage of the trinitrile compound is Y, where X and Y meet conditions represented by Formula (1) and Formula (2):

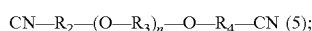
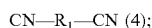
$$\text{about } 2 \text{ wt } \% \leq (X+Y) \leq \text{about } 11 \text{ wt } \% \quad (1); \text{ and}$$

$$\text{about } 0.1 \leq (X/Y) \leq \text{about } 8 \quad (2),$$

wherein, based on the total weight of the electrolyte, a weight percentage of the propyl propionate is Z, where Y and Z meet a condition represented by Formula (3):

$$\text{about } 0.01 \leq (Y/Z) \leq \text{about } 0.3 \quad (3).$$

2. The electrolyte according to claim 1, wherein the dinitrile compound comprises a compound of Formula (4) or (5):

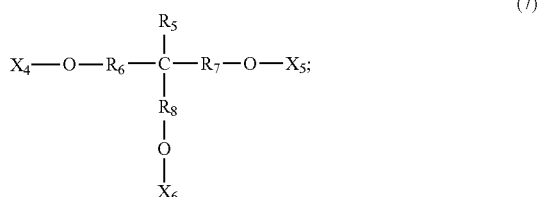
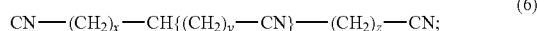


or a combination thereof,

where R_1 , R_2 , R_3 and R_4 are each independently an alkylene group having 1-5 carbon atoms or an alkynylene group having 2-5 carbon atoms, and n represents an integer from 0 to 5.

3. The electrolyte according to claim 1, wherein the dinitrile compound is one selected from the group consisting of butanedinitrile, glutaronitrile, adiponitrile, 1,5-dicyanopentane, 1,6-dicyanohexane, 1,7-dicyanoheptane, 1,8-dicyanooctane, 1,9-dicyanononane, 1,10-dicyanodecane, 1,12-dicyanododecane, tetramethylbutanedinitrile, 2-methylglutaronitrile, 2,4-dimethylglutaronitrile, 2,2,4,4-tetramethylglutaronitrile, 1,4-dicyanopentane, 2,6-dicyanoheptane, 2,7-dicyanooctane, 2,8-dicyanononane, 1,6-dicyanodecane, 1,2-dicyanobenzene, 1,3-dicyanobenzene, 1,4-dicyanobenzene, 3,5-dioxapimelonitrile, 1,4-bis(cyanoethoxy)butane, ethylene glycol bis(2-cyanoethyl)ether, diethylene glycol bis(2-cyanoethyl)ether, triethylene glycol bis(2-cyanoethyl)ether, tetraethylene glycol bis(2-cyanoethyl)ether, 3,6,9,12,15,18-hexaoxaicosanoic dinitrile, 1,3-bis(2-cyanoethoxy)propane, 1,4-bis(2-cyanoethoxy)butane, 1,5-bis(2-cyanoethoxy)pentane, ethylene glycol bis(4-cyanobutyl)ether, 1,4-dicyano-2-butene, 1,4-dicyano-2-methyl-2-butene, 1,4-dicyano-2-ethyl-2-butene, 1,4-dicyano-2,3-dimethyl-2-butene, 1,4-dicyano-2,3-diethyl-2-butene, 1,6-dicyano-3-hexene, 1,6-dicyano-2-methyl-3-hexene, 1,6-dicyano-2-methyl-5-methyl-3-hexene, and any combination thereof.

4. The electrolyte according to claim 1, wherein the trinitrile compound comprises a compound of Formula (6) or (7):



or a combination thereof,

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where in Formula (6), x, y, and z represent an integer from 0 to 5, and x, y, and z are not 0 at the same time; and

in Formula (7), R_5 represents hydrogen or an alkyl group having 1-5 carbon atoms, R_6 , R_7 and R_8 are each independently an alkylene group having 1-5 carbon atoms, and X_4 , X_5 and X_6 are each independently $-\text{R}_9-\text{CN}$, in which R_9 represents an alkylene group having 1-5 carbon atoms.

5. The electrolyte according to claim 1, wherein the trinitrile compound is one selected from the group consisting of: 1,3,5-pentanetricarbonitrile; 1,2,3-propanetrinitrile; 1,3,6-hexanetricarbonitrile; 1,2,6-hexanetricarbonitrile; 1,2,3-tris(2-cyanoethoxy)propane; 1,2,4-tris(2-cyanoethoxy)butane; 1,1,1-tris(cyanoethoxymethylene)ethane; 1,1,1-tris(cyanoethoxymethylene)propane; 3-methyl-1,3,5-tris(cyanoethoxy)pentane; 1,2,7-tris(cyanoethoxy)heptane; 1,2,6-tris(cyanoethoxy)hexane; 1,2,5-tris(cyanoethoxy)pentane; and any combination thereof.

6. The electrolyte according to claim 1, wherein X is about 0.01-10 wt %, Y is about 0.01-10 wt %, and a weight percentage Z of the propyl propionate is about 5-50 wt %.

7. The electrolyte according to claim 1, further comprising a fluoroether comprising at least one of the compounds of Formula (8), Formula (9), Formula (10) or Formula (11):



wherein Formulae (8), (9), (10), and (11), Rf1 and Rf2 are each independently a linear or branched C_1 to C_{12} fluoroalkyl group having at least one hydrogen atom substituted with fluoro, R is a linear or branched C_1 to C_{12} alkyl group, and R' is a linear or branched C_1 to C_5 alkylene group, and n is an integer from 1 to 5.

8. The electrolyte according to claim 7, wherein Rf1 or Rf2 is each independently a fluoroalkyl group selected from the group consisting of HCF_2- , CF_3- , HCF_2CF_2- , CH_3CF_2- , CF_3CH_2- , CF_3CF_2- , $(\text{CF}_3)_2\text{CH}-$, $\text{HCF}_2\text{CF}_2\text{CH}_2-$, $\text{CF}_3\text{CH}_2\text{CH}_2-$, $\text{HCF}_2\text{CF}_2\text{CF}_2\text{CH}_2-$, $\text{HCF}_2\text{CF}_2\text{CF}_2\text{CH}_2-$, $\text{CF}_3\text{CF}_2\text{CH}_2-$, $\text{CF}_3\text{CF}_2\text{CH}_2-$, $\text{CF}_3\text{CFHCF}_2\text{CH}_2-$, $\text{HCF}_2\text{CF}(\text{CF}_3)\text{CH}_2-$, and $\text{CF}_3\text{CF}_2\text{CH}_2\text{CH}_2-$.

9. The electrolyte according to claim 7, wherein the fluoroether is one selected from the group consisting of: $\text{HCF}_2\text{CF}_2\text{CH}_2\text{OCF}_2\text{CF}_2\text{H}$, $(\text{CF}_3)_2\text{CF}(\text{CF}_2\text{CF}_3)(\text{OCH}_3)$, $\text{CF}_3\text{CH}_2\text{CF}_2\text{CH}(\text{CH}_3)\text{OCF}_2\text{CH}_2\text{CF}_3$, $\text{HCF}_2\text{CF}_2\text{CH}_2\text{OCF}_2\text{CF}_2\text{CF}_2\text{H}$, $\text{HCF}_2\text{CF}_2\text{OCH}_2\text{CF}_3$, $\text{HCF}_2\text{CF}_2\text{OCH}_2\text{CH}_2\text{OCF}_2\text{CF}_2\text{H}$, $\text{HCF}_2\text{CF}_2\text{OCH}_2\text{CH}_2\text{OCF}_2\text{CF}_2\text{H}$, $\text{HCF}_2\text{CF}_2\text{OCH}_2\text{CH}_2\text{OCF}_2\text{CF}_2\text{H}$, $\text{HCF}_2\text{CF}_2\text{OCH}_2\text{CH}_2\text{OCF}_2\text{CF}_2\text{H}$, $\text{CH}_3\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{F}$, $\text{CH}_3\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CF}_3$, $\text{CH}_3\text{OCH}_2\text{CH}(\text{CH}_3)\text{OCH}_2\text{CH}_2\text{F}$, $\text{CH}_3\text{OCH}_2\text{CH}(\text{CH}_3)\text{OCH}_2\text{CF}_3$, $\text{FCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{F}$, $\text{FCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{F}$, $\text{CF}_3\text{CH}_2\text{O}(\text{CH}_2\text{CH}_2\text{O})_2\text{CH}_2\text{CF}_3$, $\text{CF}_3\text{CH}_2\text{OCH}_2\text{CH}(\text{CH}_3)\text{OCH}_2\text{CF}_3$, and any combination thereof.

10. The electrolyte according to claim 1, further comprising a cyclic phosphonic anhydride, based on the total weight of the electrolyte, a content of cyclic phosphonic anhydride is about 0.01-10 wt %.

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11. The electrolyte according to claim 1, further comprising one selected from a group consisting of: a cyclic carbonate ester having a carbon-carbon double bond, a fluorinated chain carbonate ester, a fluorinated cyclic carbonate ester, a compound having a sulfur-oxygen double bond, and any combination thereof. 5

12. The electrolyte according to claim 1, further comprising a cyclic carboxylate ester, including γ -butyrolactone or γ -valerolactone or a combination thereof.

13. An electrochemical device, wherein the electrochemical device comprises electrodes and an electrolyte comprising a dinitrile compound, a trinitrile compound, and propyl propionate, wherein, based on the total weight of the electrolyte, the weight percentage of the dinitrile compound is X and the weight percentage of the trinitrile compound is Y, where X and Y meet the conditions represented by Formula (1) and Formula (2):

$$\text{about } 2 \text{ wt } \% \leq (X+Y) \leq \text{about } 11 \text{ wt } \% \quad (1); \text{ and}$$

$$\text{about } 0.1 \leq (X/Y) \leq \text{about } 8 \quad (2). \quad 20$$

14. The electrochemical device according to claim 13, wherein the electrode comprises a current collector and a coating on the current collector, the coating comprising a single-sided coating and a double-sided coating, wherein: the single-sided coating is a coating formed by applying a slurry on one surface of the current collector; and the

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double-sided coating is a coating formed by applying a slurry on two opposite surfaces of the current collector; and

the electrode with the single-sided coating has an electrode compaction density D1, and the electrode with the double-sided coating has an electrode compaction density D2, wherein D1 and D2 meet the relationship: about $0.8 \leq D1/D2 \leq \text{about } 1.2$.

15. The electrochemical device according to claim 14, wherein the electrode s comprise a cathode and a anode, wherein when the electrode is a cathode, $3.5 \text{ g/cm}^3 \leq D2 \leq 4.3 \text{ g/cm}^3$; or when the electrode is an anode, $1.2 \text{ g/cm}^3 \leq D2 \leq 1.8 \text{ g/cm}^3$.

16. An electronic device, comprising an electrochemical device that includes electrodes and an electrolyte comprising a dinitrile compound, a trinitrile compound, and propyl propionate, wherein, based on the total weight of the electrolyte, the weight percentage of the dinitrile compound is X and the weight percentage of the trinitrile compound is Y, where X and Y meet the conditions represented by Formula (1) and Formula (2):

$$\text{about } 2 \text{ wt } \% \leq (X+Y) \leq \text{about } 11 \text{ wt } \% \quad (1); \text{ and}$$

$$\text{about } 0.1 \leq (X/Y) \leq \text{about } 8 \quad (2). \quad 25$$

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