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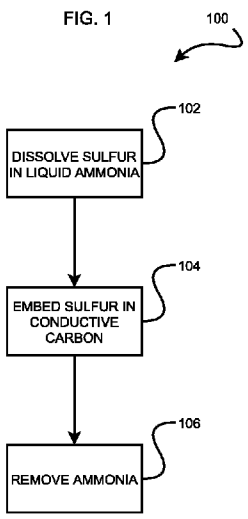
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(54) Title: SULPHUR CATHODES, SULPHUR CATHODE MATERIALS, AND APPARATUS AND METHODS FOR MAKING SAME



(57) Abstract: A method for embedding sulphur into conductive carbon is provided. Elemental sulphur is dissolved in liquid ammonia to form a sulphur-ammonia solution. Conductive carbon is soaked in the sulphur-ammonia solution to embed the conductive carbon with the dissolved sulphur. The liquid ammonia in the sulphur-ammonia solution can be removed as gaseous ammonia to yield sulphur-embedded conductive carbon. The sulphur-embedded conductive carbon can be used to manufacture sulphur cathodes. Such sulphur cathodes and batteries incorporating such sulphur cathodes are provided.

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SULPHUR CATHODES, SULPHUR CATHODE MATERIALS, AND APPARATUS AND METHODS FOR MAKING SAME

Technical Field

[0001] Some embodiments of the present invention relate to apparatus for storing and discharging energy. Some embodiments relate to metal-sulphur batteries and/or metal-sulphur electrodes. Some embodiments relate to a sulphur cathode of a metal-sulphur battery. Some embodiments relate to a sulphur cathode of a lithium-sulphur battery. Some embodiments relate to methods for embedding sulphur into activated carbon. Some embodiments relate to carbon with sulphur embedded therein. Some embodiments relate to batteries having electrodes made from carbon with sulphur embedded therein.

Background

[0002] Carbon materials such as activated carbon can provide a useful material for the manufacture of electrodes. For example, carbon-based materials can be designed as highly porous materials so as to have a high surface area. The pore sizes in the material can be quite small to provide a high surface area. Carbon materials can also offer good adsorption (i.e. adhesion of ions onto the surface of the material) and low resistance (i.e. efficient electron and ion movement at high current). Examples of potential applications for such carbon materials include batteries, including metal sulphur and lithium-sulphur batteries.

[0003] A growing area of interest in rechargeable battery technology is lithium-sulphur (Li-S) batteries. A lithium-sulphur battery has a lithium-metal anode and a sulphur cathode. Sulphur and lithium have theoretical capacities of 1672 or 1675 mA h g^{-1}, respectively. As such, a theoretical energy density of a Li-S battery is 2500 Wh kg^{-1}, which is one of the highest theoretical energy densities among rechargeable batteries. As such, lithium-sulphur batteries provide a promising electrical energy-storage system for portable electronics and electric vehicles.

[0004] Lithium-sulphur batteries operate by reduction of sulphur at the cathode to lithium sulphide: $S + 16Li \rightleftharpoons 8Li_2S$ (2.4V – 1.7V). The sulphur reduction reaction to lithium sulphide

is complex and involves the formation of various lithium polysulphides (Li_2S_x, 8<x<1, e.g. Li_2S_8, Li_2S_6, Li_2S_4, and Li_2S_2).

[0005] In the case of some lithium-sulphur batteries, the anode can be pure lithium metal (Li° oxidized to Li^+ during discharge), and in some cases the cathode can be activated carbon containing sulphur (S° reduced to S^{2-} during discharging). An ion-permeable separator is provided between the anode and the cathode, and an electrolyte used in such system is generally based on a mixture of two organic solvents such as 1,2-dimethoxyethane (DME) and 1,3-dioxolane (DOXL) containing 1 M of lithium bis(trifluoromethane sulfonyl)imide (LiN(SO2CF3)2) and 1% lithium nitrate, or the like.

[0006] Potential advantages of lithium-sulphur batteries include a high energy density (theoretically 5 times although practically 2 – 3 times more than lithium-ion), there is no requirement for top-up charging when in storage (whereas a lithium-ion battery may require 40% regular recharging to prevent capacity loss), the active materials are lighter as compared to lithium-ion, and the materials used in the manufacture of lithium-sulphur batteries are more environmentally friendly and less expensive than lithium-ion batteries (since no rare earth metals are required).

[0007] However, there are challenges for lithium-sulphur battery systems that have not yet been addressed sufficiently to make them commercially useful. For example, lithium polysulphides (Li_2S_x where x is an integer between 3 and 8) dissolve in the electrolyte and further reduce to insoluble lithium polysulphides (e.g. Li_2S_2 to Li_2S_1) that form on the anode in the battery systems. Such formations create a loss of active material, resulting in a short life cycle (i.e. fewer discharging and charge cycles).

[0008] Also, because sulphur is electronically and ionically insulating, sulphur needs to be embedded into a conductive matrix to be used in a lithium sulfur battery. Carbon is a potentially useful material for lithium-sulphur battery electrodes because it has a porous structure that supports the deposition of lithium polysulphide, and can help to minimize electrode expansion during discharge.

[0009] There remains a need for technologies that improve the capabilities of lithium-sulphur battery systems. Additionally, there is a general desire for improved apparatus and methods for making a sulphur cathode. There is also a general desire for an improved

method for embedding sulphur into a conductive matrix such as carbon, e.g. activated carbon.

[0010] The foregoing examples of the related art and limitations related thereto are intended to be illustrative and not exclusive. Other limitations of the related art will become apparent to those of skill in the art upon a reading of the specification and a study of the drawings.

Summary

[0011] The following embodiments and aspects thereof are described and illustrated in conjunction with systems, tools and methods which are meant to be exemplary and illustrative, not limiting in scope. In various embodiments, one or more of the above-described problems have been reduced or eliminated, while other embodiments are directed to other improvements.

[0012] In one aspect, a method for embedding sulphur into conductive carbon is provided. The conductive carbon is soaked in a sulphur-ammonia solution to embed the conductive carbon with at least a portion of the sulphur contained in the sulphur-ammonia solution.

[0013] In one aspect, a method for embedding sulphur into conductive carbon is provided. Elemental sulphur is dissolved in liquid ammonia to form a sulphur-ammonia solution. The conductive carbon is soaked in the sulphur-ammonia solution to embed the conductive carbon with at least a portion of the dissolved sulphur, and sulphur-embedded conductive carbon is recovered. The conductive carbon can be activated carbon. After the conductive carbon has been soaked in the sulphur-ammonia solution, ammonia can be removed as gaseous ammonia. The gaseous ammonia can be compressed and recycled for further use in the process.

[0014] The elemental sulphur can be dissolved in the liquid ammonia in a pressurized environment, for example a pressure of between about 110 psig and 135 psig at room temperature. The elemental sulphur can be dissolved in the liquid ammonia at a temperature of between -20°C and 20°C. The pressure of the pressurized environment can vary depending on the vapor pressure of ammonia at the selected temperature, e.g. the pressure of the pressurized environment can be between 45 and 55 psig if the temperature is approximate 0°C, or between 10 and 50 psig if the temperature is approximately -20°C.

The resultant sulphur-ammonia solution can contain between 5% and 30% by weight dissolved sulphur.

[0015] The step of soaking the conductive carbon in the sulphur-ammonia solution can be carried out in a pressurized environment, e.g. having a pressure of about 110 to about 135 psig at room temperature.

[0016] The sulphur-embedded conductive carbon may contain between about 30% to about 85% sulphur by weight. The sulphur-embedded conductive carbon can be used to produce a sulphur cathode, e.g. for use in a metal-sulphur battery such as a lithium-sulphur battery. A method of operating such a battery may cycle the battery only to 75% depth-of-discharge (DoD) when charging and discharging the battery to minimize formation of insoluble polysulphides.

[0017] An apparatus for embedding sulphur into conductive carbon can have a sulphur dissolving tank having a first inlet for flowing liquid ammonia into the sulphur dissolving tank, a second inlet for loading elemental sulphur into the sulphur dissolving tank, and an outlet for withdrawing a sulphur-ammonia solution from the sulphur dissolving tank; and a sulphur impregnating tank in selective fluid communication with the sulphur dissolving tank, the sulphur impregnating tank having a first inlet connected to the outlet of the sulphur dissolving tank, a second inlet for adding conductive carbon into the sulphur impregnating tank, a release valve for evaporating the liquid ammonia, and a release conduit for discharging gaseous ammonia.

[0018] In addition to the exemplary aspects and embodiments described above, further aspects and embodiments will become apparent by reference to the drawings and by study of the following detailed descriptions.

Brief Description of the Drawings

[0019] Exemplary embodiments are illustrated in referenced figures of the drawings. It is intended that the embodiments and figures disclosed herein are to be considered illustrative rather than restrictive.

[0020] FIG. 1 is a flow diagram of an example embodiment of a method for embedding sulphur into conductive carbon.

[0021] FIG. 2A is a flow diagram of a further example embodiment of a method for embedding sulphur into conductive carbon. FIG. 2B is a flow diagram of a further example embodiment of a method for embedding sulphur into conductive carbon.

[0022] FIG. 3 is a schematic diagram of an example apparatus for embedding sulphur into conductive carbon.

[0023] FIG. 4 is a schematic diagram of an example apparatus for embedding sulphur into conductive carbon using a sulphur measuring tank.

[0024] FIG. 5 is a schematic diagram of an example apparatus for embedding sulphur into conductive carbon using a high pressure metering pump.

[0025] FIG. 6 is a schematic diagram of an example apparatus for embedding sulphur into conductive carbon using a high pressure metering pump and a sulphur measuring tank.

[0026] FIG. 7 shows the profile of the discharge curve from Galvano charge and discharge tests for three cells in one example.

[0027] FIG. 8A, 8B and 8C show the discharge capacity of various LiS battery cells performed for up to 30 charge-discharge cycles.

[0028] FIG. 9 shows the discharge capacity (mAh/g) and capacity retention (%) of a representative LiS battery cell cycled in the soluble range (75% DoD) for 100 cycles of charge and discharge.

[0029] FIG. 10 shows the discharge capacity (mAh/g) and capacity retention (%) of another representative LiS battery cell cycled in the soluble range (75% DoD) for 217 cycles of charge and discharge.

[0030] FIG. 11 shows the discharge capacity (mAh/g) and capacity retention (%) of another representative LiS battery cell cycled in the soluble range (75% DoD) for 100 cycles of charge and discharge.

[0031] FIG. 12A, 12B and 12C show the determination of a minimum ratio of electrolyte to sulphur mass. FIG. 12A shows ES ratio versus capacity. FIG. 12B shows ES ratio versus initial open-circuit potential (OCP). FIG. 12C shows initial open-circuit potential (OCP) versus capacity.

[0032] FIG. 13 shows the initial two cycles of galvanostatic charge/ discharge (GCD) tested at 1C/g at 100% depth of discharge (DoD).

[0033] FIG. 14A, 14B, 14C and 14D show capacity and capacity retention for LiS batteries with sulfur impregnated lignin-based AC through 500 cycles tested at 1C/g at 100% DoD and 75% DoD, respectively. FIG 14A shows a cell cycled at a depth of discharge (DoD) of 75% (after two cycles at 100% DoD) and FIG. 14B shows a cell cycled at a depth of discharge (DoD) of 100%. FIG. 14C shows capacity and FIG. 14D shows percentage capacity retention for the 40^{th} to the 500^{th} cycles, respectively.

Description

[0034] Throughout the following description specific details are set forth in order to provide a more thorough understanding to persons skilled in the art. However, well known elements may not have been shown or described in detail to avoid unnecessarily obscuring the disclosure. Accordingly, the description and drawings are to be regarded in an illustrative, rather than a restrictive, sense.

[0035] As used in this specification, the terms “about” or “approximately” mean a value within +/- 10% of the stated value unless specified otherwise, and either one of these terms connotes that strict compliance with the numeric value recited is not critical but some variation is permissible and still within the scope of the various embodiments described herein.

[0036] As used herein, the term “conductive” means electrically conductive.

[0037] As used herein, the term “conductive carbon” means carbonaceous materials that allow the flow of electric current with little resistance. Conductive carbon has a level of conductivity (σ) of at least 100 siemens per metre (S/m), including e.g. at least 150, at least 200, at least 250, at least 500, at least 750, at least 1000, at least 1250, at least 1500, at least 1750, at least 2000, at least 2500, at least 3000, at least 3500, at least 4000, at least 4500 or at least 5000 S/m. Some examples of conductive carbon are activated carbon, graphite, graphene, carbon nanotubes, and the like, or any desired mixture thereof. Any suitable source of biomass can be used to produce carbon that can be subjected to further chemical and/or thermal processing to yield activated carbon. For example, biomass such

as coconut husk, lignin, or the like may be processed to produce biochar and/or hydrochar that can be further processed to yield activated carbon.

[0038] For example, for one form of activated carbon used in the examples described below (YPF), a conductivity of 1322 S/m is reported (N. Rey-Raap et. Al Measurement, An Electrical Conductivity Translator for Carbons, 2014, 56, 215-218). Those same authors report that graphite has a conductivity of 5023 S/m. Another source says that amorphous carbon (including activated carbon) has a conductivity of 1.25 – 2000 S/m while graphite has a conductivity of 2000 – 3300S/m (Helmenstine, ThoughtCo. Table of Electrical Resistivity and Conductivity, 608499, 27 June 2019). Fabiono reports activated carbon has a conductivity of 250 -800 S/m (Fabiano Gomes Ferreira de Paula et.al., Structural Flexibility in Activated Carbon Materials Prepared under Harsh Activation Conditions, Materials 2019, 12, 1988). A. Barroso-Bogeat et. al., Electrical conductivity of activated carbon–metal oxide nanocomposites under compression:a comparison study, Phys. Chem. Chem. Phys., 2014, 16, 25161—25175 reported that typical activated carbon has >194 S/m using a compression at 750 kPa.

[0039] The inventors have found that liquid ammonia is an effective carrier to load sulphur into conductive carbon, for example to produce a sulphur electrode. Based on this, the inventors have invented a method and apparatus for embedding sulphur into conductive carbon. The sulphur-embedded conductive carbon may be used as a sulphur cathode of a metal-sulphur battery, e.g. a lithium-sulphur battery.

[0040] FIG. 1 shows an example embodiment of a method **100** for embedding sulphur into conductive carbon.

[0041] At step **102**, sulphur, e.g. elemental sulphur, is dissolved in liquid ammonia (NH_3) to form a sulphur-ammonia solution. In some embodiments, the conditions at step **102** are selected to promote the solubility of sulphur in the liquid ammonia. In some embodiments, the liquid ammonia is anhydrous liquid ammonia.

[0042] At step **104**, conductive carbon is soaked in the sulphur-ammonia solution to impregnate/embed the conductive carbon with sulphur. In some embodiments, the conductive carbon supplied at step **104** is activated carbon.

[0043] At step **106**, liquid ammonia is removed. In some embodiments, liquid ammonia is removed as gaseous ammonia. For example, liquid ammonia may be exposed to open air

and liquid ammonia then evaporates and is released as gaseous ammonia. Removal of ammonia yields the desired conductive carbon embedded with sulphur product.

[0044] A further example embodiment of a process **200** for embedding sulphur in conductive carbon is illustrated in FIG. 2A.

[0045] At step **202**, elemental sulphur is supplied to a suitable vessel. The elemental sulphur (S°) can be any form of sulphur, including crown-shaped S_8 molecules representative of the typical stable form of sulphur. Other forms of elemental sulphur include S_6, S_7, S_9, S_{10}, S_{11}, S_{12}, S_{13}, S_{14}, S_{15}, S_{16}, S_{17}, and S_{18} rings, or linear or branched forms and are believed to also be soluble in liquid ammonia and are encompassed in various alternative embodiments. In some embodiments, the elemental sulphur is granular elemental sulphur (S°). In some embodiments, the elemental sulphur is powdered elemental sulfur. In some embodiments, granular elemental sulphur may be used to avoid potential difficulties in removing any undissolved fine powdery sulphur from the liquid ammonia solution in subsequent processing steps.

[0046] Any suitable reaction vessel can be used. In some embodiments, the reaction vessel is a pressure reactor. In some embodiments, the pressure reactor has a fine screen provided at its outlet, to facilitate separation of the resulting ammonia-sulphur solution from any undissolved sulphur. In some embodiments, the fine screen is made from an inert material such as stainless steel. In some embodiments, the screen has a mesh size that prevents particles having a diameter larger than about 25 μm from passing through the screen.

[0047] At step **204**, the pressure in the vessel containing the elemental sulphur is reduced. In some embodiments, the pressure is reduced below atmospheric pressure at step **204**. In some embodiments, step **204** is carried out under vacuum. In some embodiments, step **204** is carried out at close to zero psig. In some embodiments, the pressure level at step **204** is at least about 12 psi below an atmospheric pressure, including between about 12 psi and about 14 psi below an atmospheric pressure, including e.g. at least 12.5, 13.0, or 13.5 psi below an atmospheric pressure, or any pressure therebetween. In one example embodiment, the pressure during step **204** is reduced to approximately 12.2 psi below an atmospheric pressure. In some embodiments, a pressure feed such as a high pressure

metering pump is used for carrying out step **208** to supply liquid ammonia described below, and in such embodiments step **204** can be omitted.

[0048] In some embodiments, at step **206** the temperature of the vessel containing the elemental sulphur is reduced below room temperature to increase sulphur solubility in the liquid ammonia. In some embodiments, the temperature at step **206** is between about -20ºC and about 20ºC, including any value therebetween, e.g. -18ºC, -16ºC, -14ºC, -12ºC, -10ºC, -8ºC, -6ºC, -4ºC, -2ºC, 0ºC, 2ºC, 4ºC, 6ºC, 8ºC, 10ºC, 12ºC, 14ºC, 16ºC and 18ºC. In some embodiments, step **210** is conducted at the temperature that is set at step **206**.

[0049] As an example, the solubility of sulphur is approximately 38% (by mass) at - 20°C at 27.6 psig of vapor pressure but decreases as temperature increases, as shown in Table 1 below. Unlike other liquids such as carbon disulphide, the solubility of sulphur decreases with increasing temperature in liquid ammonia. In some embodiments, based on the data as set forth in Table 1, the solubility of sulphur (% by mass) in liquid ammonia can be predicted by Equation (1), where T is the temperature in ºC (R^2 = 0.9816):

$$\text{Solubility} = -0.344\text{T} + 32.41 \quad (1)$$

In some embodiments, based on empirically determined data as set forth in Table 1, the vapor pressure of liquid ammonia can be predicted by Equation (2) (R^2 = 0.9997):

$$\text{Vapor Pressure} = 0.0397\text{T}^2 + 2.4449\text{T} + 46.321 \quad (2)$$

Table 1. Solubility of sulfur in liquid ammonia (% by mass) at various temperatures.

Temperature (°C)	Sulfur solubility (% by mass) in LNH_3	LNH_3 vapor pressure (psig)[1]	LNH_3 vapor pressure (kPa)
35	19.65	181.1	1350
20	25.77	109.6	857.1
0	33.90	47.6	429.4
-20	38.28	12.9	190.2

[1]See https://en.wikipedia.org/wiki/Ammonia_(data_page).

[0050] At step **208**, liquid ammonia is added to the vessel containing the elemental sulphur. In some embodiments, the liquid ammonia is supplied from a pressurized reservoir of

ammonia at **209**. In some embodiments, the pressurized reservoir of ammonia at **209** is maintained in the range of about 110 to about 135 psig at room temperature, including any value therebetween, e.g. 115, 120, 125, or 130 psig. In one example embodiment, the pressurized reservoir of ammonia **209** is maintained at a pressure in the range of about 110 to about 132 psig at room temperature. Without being bound, it is believed that the pressure in reservoir **209** is determined primarily by the vapor pressure of the liquid ammonia contained therein, which varies with temperature. Thus, the pressure at which step **210** following step **209** is conducted can be the partial pressure of ammonia at the temperature at which step **210** is conducted. The pressure of the pressurized environment can vary depending on the vapor pressure of ammonia at the selected temperature, e.g. the pressure of the pressurized environment can be between 45 and 55 psig if the temperature is approximate 0°C, or between 10 and 50 psig if the temperature is approximately -20°C.

[0051] In some embodiments, the pressure differential between the vessel containing the elemental sulphur and the pressurized reservoir of ammonia drives the flow of liquid ammonia into the vessel containing the elemental sulphur. In some embodiments, a pump such as a high pressure metering pump is used at step **208** to deliver liquid ammonia to the vessel containing the elemental sulphur. Without being bound, use of such a pump may enhance the solubility of sulphur in the liquid ammonia as a higher transfer efficiency of liquid versus gaseous ammonia can be achieved.

[0052] At step **210**, the elemental sulphur is dissolved in the liquid ammonia to provide a sulphur-ammonia solution. Any suitable method of dissolution may be used, for example the sulphur-ammonia solution may be shaken or stirred to encourage the solubilization process of sulphur, sonication may be used, or a metal stirrer may be used. In some embodiments, the sulphur-ammonia solution is shaken, stirred and/or sonicated for a period of time, e.g. in the range of about 5 to about 20 minutes, including any period therebetween e.g. 10 or 15 minutes.

[0053] Without being bound by theory, various ionic forms of sulphur such as S_3^-, S_3N^-, and S_4N^- may exist in liquid ammonia. It is believed that once the solution is depressurized, e.g. at step **224** as described below with reference to this example embodiment, the liquid ammonia will self-vaporize and these sulphur species will be returned to their elemental forms.

[0054] At step **212**, any undissolved sulphur may be removed from the sulphur-ammonia solution. In some embodiments, at step **212** the sulphur-ammonia solution is transferred to a second vessel without transferring any undissolved solid sulphur to remove the sulphur-ammonia solution from the undissolved sulphur. In some embodiments, a high-pressure pump can be used to transfer the sulphur-ammonia solution. In some embodiments in which the first vessel is a pressure reactor with a screen provided at its outlet, the screen is used to allow the sulphur-ammonia solution to flow through while retaining any undissolved sulphur.

[0055] In some embodiments, at step **214**, the total amount of dissolved sulphur in the second vessel can be calculated using the gravimetric change of the first vessel relative to the second vessel. This calculation can allow for the determination of the amount of conductive carbon that can be impregnated with the dissolved sulphur.

[0056] In some embodiments, the percentage of dissolved sulphur in the sulphur-ammonia solution obtained at step **212** by weight is between about 5% and about 30%, including any value therebetween, e.g. 6%, 7%, 8%, 9%, 10%, 11%, 12%, 13%, 14%, 15%, 16%, 17%, 18%, 19%, 20%, 21%, 22%, 23%, 24%, 25%, 26%, 27%, 28% or 29% w/w.

[0057] The desired amount of conductive carbon is subsequently soaked in the sulphur-ammonia solution to impregnate/embed the conductive carbon with sulphur. In some embodiments, the desired amount of conductive carbon is directly combined with the sulphur-ammonia solution.

[0058] In the illustrated embodiment, at step **216**, the desired amount of conductive carbon is supplied in a third vessel provided. In some embodiments, the third vessel is provided with a stainless steel mesh with a mesh opening size of about 25 μm at its opening, for example to capture and retain activated carbon which might otherwise drift away by reason of air currents.

[0059] At step **218**, the pressure in the vessel containing the conductive carbon is reduced under vacuum. In some embodiments, the pressure level at step **218** is reduced to at least about 12 psi below an atmospheric pressure, including between about 12 psi and about 14 psi below an atmospheric pressure, including e.g. at least 12.0, 12.5, 13.0, or 13.5 psi below an atmospheric pressure, or any pressure therebetween. In one example embodiment, the pressure level at step **218** is reduced to about 12.2 psi below the

atmospheric pressure. In alternative embodiments, a pump such as a high pressure metering pump can be used to transfer the sulphur-ammonia solution at step **218** rather than reducing the pressure. In such embodiments, the pressure accordingly is not reduced at step **218**.

[0060] At step **220**, the sulphur-ammonia solution is supplied to the vessel containing the conductive carbon. Introduction of the sulphur-ammonia solution increases the pressure within the vessel containing the conductive carbon. In some embodiments, introduction of the sulphur-ammonia solution increases pressure in the vessel to in the range of about 110 to about 150 psig at room temperature, including any value therebetween, e.g. 115, 120, 125, 130, 135, 140 or 145 psig. In one example embodiment, the pressure at step **220** increases to in the range of about 126 to about 147 psig at room temperature. In some embodiments, step **220** is conducted at ambient or room temperature.

[0061] At step **222**, sulphur from the sulphur-ammonia solution is adsorbed onto the conductive carbon under pressure to maintain the ammonia substantially in liquid form. In some embodiments, adsorption is carried out for an adsorption period. Any suitable length of time may be used for the adsorption period. In some embodiments, the adsorption period is between about 30 seconds and about 10 minutes, including any value therebetween, e.g. 45 seconds, 1 minute, 1.5, 2, 3, 4, 5, 6, 7, 8 or 9 minutes. Longer adsorption periods could be used if desired.

[0062] At step **224**, ammonia is removed as gaseous ammonia by reducing the pressure in the vessel. In some embodiments, a needle valve is provided in fluid communication with the vessel to allow the pressure to be reduced.

[0063] In some embodiments, the output of fluid passing through a needle valve provided at the bottom of the vessel is supplied to a receiver vessel containing a suitable liquid such as water, so that bubble flow can be monitored as an indicator of gaseous ammonia flow exiting the receiver vessel. This is referred to as a blow-down or wet collection method, and drying of the resultant sulphur-impregnated activated carbon is subsequently required prior to its use.

[0064] In some embodiments, the ammonia vapor in the vessel is released by slowly opening a needle valve located at the top of the vessel, leaving the sulphur-impregnated carbon within the vessel for subsequent collection at step **226** as described below. This is

referred to as a blow-up or dry collection method, and the sulphur-impregnated activated carbon obtained by this method does not require drying prior to collection or use.

[0065] If desired, the gaseous ammonia obtained from step **224** can be recovered for subsequent re-use in the process by using a compressor to compress the recovered gaseous ammonia back to liquid ammonia.

[0066] At step **226**, the sulphur-impregnated conductive carbon is collected, for example by removal from the bottom of the vessel. In some embodiments, the vessel is a pressure reactor.

[0067] With reference to FIG. 2B, an alternative embodiment of a method **200'** is illustrated. Method **200'** is generally similar to method **200**, and like steps are illustrated with the same reference numerals and are not further described herein. Method **200'** differs from method **200** in that a pump such as a high pressure metering pump can be used to move the liquid ammonia or ammonia-sulphur solution rather than a vacuum source. Thus, step **204** is omitted, and liquid ammonia is supplied by the pump at step **208**. Similarly, step **218** is omitted and sulphur-ammonia solution is supplied directly by the pump at step **220**.

[0068] In some embodiments, the sulphur-impregnated conductive carbon collected at step **226** is further processed to manufacture electrodes, for example to provide sulphur cathodes for use in metal-sulphur batteries, e.g. lithium-sulphur batteries.

[0069] In some embodiments, the amount of conductive carbon used to absorb dissolved sulphur can be about 20% to about 100% of the weight of dissolved sulphur, including any value therebetween, e.g. 45, 50, 55, 60, 65, 70, 75, 80, 85, 90 or 95% w/w. Any desired value can be used in alternative embodiments. The final sulphur content (wt. %) in the sulphur-impregnated activated carbon can be in the range of about 30% to about 85%, including any value therebetween, e.g. 35, 40, 45, 50, 55, 60, 65, 70, 75 or 80% w/w.

[0070] Method **200** may be carried out in apparatus **300** for embedding conductive carbon with sulphur. With reference to FIG. 3, apparatus **300** has a sulphur dissolving tank **302**. In some embodiments, sulphur dissolving tank **302** is a pressure reactor.

[0071] Sulphur dissolving tank **302** is provided with suitable conduits and valves to permit the steps of method **200** to be carried out. Liquid ammonia may enter into sulphur dissolving tank **302** from a pressurized ammonia supply tank **303** through a conduit **304**.

[0072] In some embodiments, a stirrer [not shown] may be provided in sulphur dissolving tank **302** to mix the sulphur and liquid ammonia to encourage the solubilization process of the sulphur. In some embodiments, sulphur dissolving tank **302** can be uncoupled from apparatus **300** so that sulphur dissolving tank **302** can be subjected to vigorous shaking, whether by hand or by using any suitable shaking apparatus, to dissolve the sulphur in the liquid ammonia. In alternative embodiments, any suitable method such as sonication, a metal stirrer or the like may be provided to assist with dissolving sulphur in sulphur dissolving tank **302**.

[0073] Apparatus **300** has a sulphur impregnating tank **306**. Sulphur dissolving tank **302** and sulphur impregnating tank **306** are in selective fluid communication with each other. In some embodiments, sulphur dissolving tank **302** and sulphur impregnating tank **306** are connected by a pipe **310**. A suitable valve can be provided in pipe **310** to maintain the respective pressures within sulphur dissolving tank **302** and sulphur impregnating tank **306** and to control the flow of fluids therebetween.

[0074] A suitable vacuum source **308** is provided to help provide negative pressure within dissolving tank **302** and sulphur impregnating tank **306** when desired. Although a single vacuum source **308** has been shown in the illustrated embodiment, two separate vacuum sources and/or high-pressure metering pumps could be provided in alternative embodiments.

[0075] In some embodiments, a predetermined amount of conductive carbon is loaded in sulphur impregnating tank **306**, and the pressure in sulphur impregnating tank **306** is then reduced by placing sulphur impregnating tank **306** under vacuum. The solution of liquid ammonia and dissolved sulphur from sulphur dissolving tank **302** can then flow into sulphur impregnating tank **306**, which will result in sulphur impregnating tank **306** becoming pressurized, allowing the dissolved sulphur to be embedded into the conductive carbon in sulphur impregnating tank **306**.

[0076] Sulphur impregnating tank **306** has a release valve **312** to depressurize sulphur impregnating tank **306**. Once sulphur impregnating tank **306** is depressurized, liquid ammonia evaporates as gaseous ammonia and then exits from sulphur impregnating tank **306** via an exiting pipe **314**. In some embodiments, release valve **312** is located more

proximate to an upper end than a lower end of impregnating tank **306**; the illustrated view is schematic in nature only.

[0077] In some embodiments, a gas monitor **316** is provided to monitor the release of gaseous ammonia from sulphur impregnating tank **306**. In some embodiments, gas monitor **316** is a reservoir filled with suitable liquid, such as water, so that the release of ammonia from sulphur impregnating tank **306** can be monitored.

[0078] In some embodiments, sulphur-embedded conductive carbon can be collected from a lower end of sulphur impregnating tank **306** once most or all of the ammonia has been released from sulphur impregnating tank **306** and optionally dried. Sulphur-embedded conductive carbon may be used as a sulphur cathode material for use in the manufacture of a metal-sulphur battery, such as a Li-S battery.

[0079] In some embodiments, a battery containing such a sulphur-embedded conductive carbon may be cycled through charge and discharge cycles extending only to about 75% depth-of-discharge (DoD), to help prevent the formation of insoluble polysulphides, thereby prolonging the life of the battery. In some embodiments, a battery containing such a sulphur-embedded conductive carbon contains an electrolyte in a volume of between about 30 to about 80 µL/mg of sulphur, including any range or subrange therebetween, e.g. 35, 40 45, 50, 55, 60, 65, 70 or 75 µL/mg of sulphur, to help minimize the formation of insoluble polysulphides.

[0080] In some embodiments, ammonia can be collected and fed to a compressor **318** so that it can be recompressed to liquid ammonia for use again in the process conducted by apparatus **300**.

[0081] A further example embodiment of an apparatus **400** for embedding conductive carbon with sulphur is illustrated in FIG. 4. Components of apparatus **400** that are similar to apparatus **300** are illustrated with reference numerals incremented by **100** and are not further described herein (i.e. sulphur dissolving tank **402**, ammonia supply tank **403**, conduit **404**, sulphur impregnating tank **406**, vacuum source **408**, pipe **410**, release valve **412**, exiting pipe **414**, gas monitor **416** and compressor **418**).

[0082] Apparatus **400** differs from apparatus **300** in that a sulphur measuring tank **420** is provided downstream of sulphur dissolving tank **402**. After sulphur has been dissolved in liquid ammonia in sulphur dissolving tank **402**, sulphur measuring tank **420** is placed under

vacuum to draw the sulphur-ammonia solution into the interior thereof, while undissolved sulphur is retained in dissolving tank **402**, for example by positioning a suitable screen over the outlet of dissolving tank **402**. The total amount of dissolved sulphur present in sulphur measuring tank **420** is then calculated based on the gravimetric difference between tanks **402** and **420** to calculate the amount of conductive carbon to be placed in sulphur impregnating tank **406** prior to addition of the sulphur-ammonia solution from sulphur measuring tank **420**.

[0083] A further alternative example embodiment of an apparatus **300'** for embedding conductive carbon with sulphur is illustrated in FIG. 5. Apparatus **300'** is generally similar to apparatus **300**, and like components are illustrated with the same reference numeral and are not further described herein. Apparatus **300'** differs from apparatus **300** in that liquid ammonia is supplied from a liquid ammonia supply tank **303** to a high pressure metering pump **308'** that is used to supply both ammonia and ammonia-sulphur solution, rather than using a vacuum to depressurize the receiving containers. High pressure metering pump **308'** receives liquid ammonia from tank **303** via line **304-1**, and supplies liquid ammonia to dissolving tank **302** via line **304-2**. The sulphur-ammonia solution is then fed back to high pressure metering pump **308'** via line **310-1**, and to sulphur impregnating tank **306** via line **310-2**. Liquid ammonia can be compressed via compressor **318** and then fed back to liquid ammonia supply tank **303**. While a single high pressure metering pump **308'** is illustrated, in alternative embodiments separate high pressure metering pumps could be provided to facilitate the necessary fluid flow within apparatus **300'**.

[0084] A further alternative example embodiment of an apparatus **400'** for embedding conductive carbon with sulphur is illustrated in FIG. 6. Apparatus **400'** is generally similar to apparatus **400**, and like components are illustrated with the same reference numeral and are not further described herein. Apparatus **400'** differs from apparatus **400** in that liquid ammonia is supplied from a liquid ammonia supply tank **403** to a high pressure metering pump **408'** that is used to supply both ammonia and ammonia-sulphur solution. High pressure metering pump **408'** receives liquid ammonia from tank **403** via line **404-1**, and supplies liquid ammonia to dissolving tank **402** via line **404-2**. The sulphur-ammonia solution is then fed back to high pressure metering pump **408'** via line **410-1**, then to sulphur measuring tank **420** via line **410-2**, and to sulphur impregnating tank **406** via line **410-3** (or directly if desired). While a single high pressure metering pump **408'** is illustrated,

in alternative embodiments separate high pressure metering pumps could be provided to facilitate the necessary fluid flow within apparatus **400'**.

Examples

[0085] Further embodiments are more specifically described with reference to the following examples, which are intended to be illustrative and not limiting in nature.

Example 1 – Activated Carbon

[0086] Example 1 was conducted at room temperature. Granular elemental sulphur was loaded in a sulphur dissolving tank that is a pressure reactor. The sulphur dissolving tank has a fine stainless-steel screen with a sieve size of approximately 25 µm. The screen was placed at the bottom of the sulphur dissolving tank. Once the sulphur was loaded, the sulphur dissolving tank was placed under vacuum to reduce the pressure to lower than about 12.2 psi below atmospheric pressure. Alternatively, if a high-pressure metering pump is available, the high-pressure metering pump can be used rather than vacuum.

[0087] Liquid ammonia was transferred from a liquid ammonia reservoir to the sulphur dissolving tank by pressure difference between the liquid ammonia reservoir (about 110 psig at room temperature) and the sulphur dissolving tank.

[0088] Sulphur was dissolved in the liquid ammonia to form a sulphur-ammonia solution in the sulphur dissolving tank. The sulphur dissolving tank was shaken for about 10 minutes after being disconnected from the liquid ammonia reservoir to dissolve the sulphur.

[0089] A second pressure reactor was placed under vacuum to reduce the pressure to less than about 12.2 psi below atmospheric pressure. The sulphur-ammonia solution was drained from the sulphur dissolving tank into the second pressure reactor. The undissolved solid sulphur was retained in the sulphur dissolving tank by the screen placed at the bottom of the sulphur dissolving tank, while the liquid sulphur-ammonia solution was permitted to exit to the second pressure reactor, to separate undissolved sulphur from the desired sulphur-ammonia solution.

[0090] The amount of dissolved sulphur was calculated using gravimetric changes between the sulphur dissolving tank and the second pressure reactor.

[0091] A specific amount of activated carbon was weighed based on the amount of dissolved sulphur as shown in Table 2.

[0092] The specific amount of activated carbon was loaded into a sulphur impregnating tank and the sulphur impregnating tank was then placed under vacuum to reduce the pressure to less than about 12.2 psi below atmospheric pressure.

[0093] The sulphur-ammonia solution was transferred from the second pressure reactor to the sulphur impregnating tank. The dissolved sulphur is absorbed onto the activated carbon for about 1 minute at a pressure of about 110 psig.

[0094] The sulphur impregnating tank was then depressurized and gaseous ammonia was released from the sulphur impregnating tank through a needle valve. Exit of the gaseous ammonia through the needle valve was monitored by allowing the gas to bubble through a beaker filled with water.

[0095] The sulphur-impregnated activated carbon was collected from the bottom of the sulphur impregnating tank. The recovered sulphur-impregnated activated carbon can be used as a cathode material, for example for the preparation of lithium sulphur batteries.

[0096] As shown in Table 2 below, the sulphur content in the sulphur-impregnated activated carbon ranged from 48% to 67%.

Table 2. Sulphur content in tested examples.

		Trial					
Step	Description	#1	#2	#3	#4	#5	#6
202	S° (g) added for the preparation of LNH3-S° solution (Vessel R1)	5.0	5.0	5.0	4.9	5.0	5.0
208	LNH_3 (g) to dissolve S° in R1	17.7	23.5	25.1	28.7	18.3	20.7
210	Dissolved S° (g) in R1 vessel	3.6	2.0	3.7	2.7	4.9	2.2
212	Mass (g) of LNH_3 +°S° solution in R1 after undissolved S° removal	21.3	25.5	28.8	31.4	23.2	22.9
210, 212	S (%) dissolved in the LHN_3-S° solution in R1 (after undissolved S° removal)	16.9%	7.8%	12.8%	8.6%	21.1%	9.6%

214	Undissolved S° (g) in R1	1.4	3.0	1.3	2.2	0.1	2.8
212 → 220 (The LNH_3-S° solution transferred from R1 to R2)	Mass (g) of LNH_3-S° solution after undissolved S° removal and transfer(to R2 vessel which is 220)	16.9	22.6	23.8	27.5	18.0	20.4
212 → 220 Loss during the transfer stage from R1 (212) to R2(220)	Mass loss (g) of the NH_3-S° solution after transfer from 212 to 220	4.4	2.9	5.0	3.9	5.2	2.5
	Transfer eff. (%) of the LNH_3-S° solution from R1 (212) to Vessel R2 (220)	79.3%	88.6%	82.6%	87.6%	77.6%	89.1%
220	Dissolved S° (g) in in the LNH_3-S° solution in R2	2.86	1.77	3.06	2.36	3.80	1.96
222	Amount (g) of the LNH_3-S° solution and AC in Vessel R3 (which is 222)	11.2	13.2	9.2	15.1	12.5	12.9
220 → 218 Loss during the transfer stage from 220 (R2 to R3 vessel)	Mass loss (g) of the NH_3-S° solution after transfer from R2 (220) to R3 (218)	5.7	9.4	14.6	12.4	5.5	7.5
	Mass loss (g) of S° in R2 (220) after transfer from R2 (220) to R3 (218)	1.0	0.7	1.9	1.1	1.2	0.7
212 → 220	Transfer eff. (%) of the LNH_3-S° solution from R2 to R3	66.3%	58.4%	38.7%	54.9%	69.4%	63.2%
216	Type of Carbon	CAC (Commercial AC)	ABC1 Produced by inventors	ABC1	CAC-T	CBC-T-2	CBC-T-1
216	Amount (g) of AC used	1.73	0.63	1.05	1.10	1.40	0.90
222	Amount (g) of the LNH_3-S° solution and AC in R3 (which is 222)	9.6	11.9	7.9	14.6	12.4	12.5
222	Total mass of LNH_3 and S°-impregnated AC under pressure	11.3	12.5	8.9	15.7	13.8	13.4
222 (S° absorbed in AC)	S° (g) absorbed in AC	1.6	0.9	1.0	1.3	2.6	1.2
226	Mass (g) of S° impregnated AC in 226	3.4	1.6	2.1	2.4	4.0	2.1

	(vessel)						
226 (resultant S°-AC)	Sulfur content (%) in S°-impregnated AC	48.25%	59.5%	49.1%	53.3%	65.2%	57.2%

Table 2 Continued. Sulphur content in tested examples.

		Trial					
Step	Description	#7	#8	#8	#10	#11	#12
202	S° (g) added for the preparation of LNH_3-S° solution (Vessel R1)	5.1	5.0	4.9	5.0	4.9	5.0
208	LNH_3 (g) to dissolve S° in R1	20.6	19.5	23.0	19.0	21.0	13.8
210	Dissolved S° (g) in R1 vessel	2.7	2.4	2.9	2.3	1.9	2.1
212	Mass (g) of LNH_3 +°S° solution in R1 after undissolved S° removal	23.3	21.9	25.9	21.3	22.9	15.9
210, 212	S (%) dissolved in the LHN_3-S° solution in R1 (after undissolved S° removal)	11.6%	11.0%	11.2%	10.7%	8.3%	13.2%
214	Undissolved S° (g) in R1	2.4	2.6	2.0	2.7	3.0	2.9
212 → 220 (The LNH3-S° solution transferred from R1 to R2)	Mass (g) of LNH_3-S° solution after undissolved S° removal and transfer(to R2 vessel which is 220)	20.3	19.3	20.8	18.9	19.8	13.4
212 → 220 Loss during the transfer stage from R1 (212) to R2(220)	Mass loss (g) of the LNH_3-S° solution after transfer from 212 to 220	3.0	2.6	5.1	2.4	3.1	2.5
	Transfer eff. (%) of the LNH_3-S° solution from R1 (212) to Vessel R2 (220)	87.1%	88.1%	80.3%	88.8%	86.5%	84.3%
220	Dissolved S° (g) in in the LNH_3-S° solution in R2	2.35	2.12	2.33	2.02	1.64	1.77

222	Amount (g) of the LNH_3-S° solution and AC in Vessel R3 (which is 222)	15.1	14.3	16.3	14.6	13.9	12.2
220 → 218 Loss during the transfer stage from 220 (R2 to R3 vessel)	Mass loss (g) of the LNH_3-S° solution after transfer from R2 (220) to R3 (218)	5.2	5.0	4.5	4.3	5.9	1.2
	Mass loss (g) of S° in R2 (220) after transfer from R2 (220) to R3 (218)	0.6	0.5	0.5	0.5	0.5	0.2
212 → 220	Transfer eff. (%) of the LNH_3-S° solution from R2 to R3	74.4%	74.1%	78.4%	77.2%	70.2%	91.0%
216	Type of Carbon	ABC1	ABC1-2	ABC1-1	AHC1 produced by inventors	AHC1-2	AHC1-1
216	Amount (g) of AC used	0.90	0.90	1.10	1.00	0.90	0.80
222	Amount (g) of the LNH_3-S° solution and AC in R3 (which is 222)	14.9	14.1	15.2	14.2	13.7	12.2
222	Total mass of LNH_3 and S°-impregnated AC under pressure	15.8	15.0	16.3	15.2	14.6	13.0
222 (S° absorbed in AC)	S° (g) absorbed in AC	1.7	1.5	1.7	1.5	1.1	1.6
226	Mass (g) of S° impregnated AC in 226 (vessel)	2.6	2.4	2.8	2.5	2.0	2.4
226 (resultant S°-AC)	Sulfur content (%) in S°-impregnated AC	65.7%	63.2%	60.7%	60.3%	55.8%	66.8%

The various calculation steps are further summarized below:

Preparation of LNH3+°S solution

1. (Step 202) The empty mass (**M1**) of R1 is recorded.
2. (Step 202) 5g of S° is pre-charged in R1, The mass (**M2**) of R1+S° is recorded.
3. R1 is degassed (vacuumed) for the transfer of LNH_3.
4. LNH_3 is transferred. The mass (**M3**) of R1+S°+LNH_3 is recorded. In this step, R1 has LNH_3+S° solution (liquid) and an undissolved S° (solid).
5. The empty mass (**M4**) of R2 is recorded, and R2 is degassed for the transfer of the LNH_3+S° solution from R1.

6. The LNH_3+S° solution is transferred to R2 from R1. The mass (**M5**) of the R2+LNH_3+S° solution is recorded.
7. The mass (**M6**) of R1+LNH_3+S° solution + undissoved S° is recorded (the NH_3+S° solution is still left in R1). **M6** is for the calculation of the transfer efficiency of the LNH_3+S° solution.
8. (Step 212) LNH_3+S° solution (the remaining solution) in R1 is discharged in a fumehood. The mass **(M7)** of R1 + undissolved S° solution is recorded.

The determination of sulfur content in the LNH_3+°S solution

- The actual mass of S° added in R1 = M2 - M1 (the initial mass of solid S° in R1)
- The mass of LNH_3 transferred in R1= M3 - M2 (amount of LNH_3 in R1)
- The mass of LNH_3+S° solution transferred to R2 from R1 = M6 - M5 (available mass of LNH_3+S° solution)
- The mass of undissolved S° in R1 = M7 - M1 (the remaining mass of solid S° in R1)
- The mass of dissolved S° in the LNH_3+°S solution = (M2 - M1) – (M7 – M1)

Sulfur content in the LNH_3+°S solution =

$$\frac{(\mathrm{M2} - \mathrm{M1}) - (\mathrm{M7} - \mathrm{M1})}{(M2 - M1) - (M7 - M1) + (M3 - M2)}$$

[0097] Without being bound by theory, the inventors believe that if in this example metering high pressure pumps were used to transfer liquid ammonia and reactor temperature was lowered, the sulphur content in the sulphur-impregnated activated carbon may be higher. The transfer efficiency could be increased by using a high-pressure metering pump.

Example 2 – Sulphur-Embedded Activated Carbon as a Sulphur Cathode

[0098] Table 3 shows the major chemical and adsorptive properties (iodine values and surface area) of bioactivated carbon and CAC (a commercially available activated carbon YP50F obtained from Calgon used as a control). The ABC1 (activated carbon produced by the inventors from lignin as a source of carbon biomass) had 97.1% carbon, 2,919 mg/g iodine value, 2,784 m^2/g surface area, and 14.3 µm mean particle size, while CAC had 94.5% carbon, 1,796 mg/g iodine value, 1,731 m^2/g surface area, and 5.4 µm mean particle size. Other experiments performed by the inventors confirmed that the benchmark activated commercial carbon used in these examples (CAC), YP50F from Calgon, has

approximately 5-6 times higher resistance than graphite, while activated biocarbon produced by the inventors in house from lignin has approximately 2-2.5 times higher resistance than graphite.

Table 3. Chemical and adsorptive properties of carbon products used for sulfur impregnation.

Carbon ID	Carbon content (%)	Ash content (%)	Iodine value (mg/g)	Surface area (m^2/g)	Mean particle size (µm)
ABC1 produced by inventors	97.10%	0.54%	2,919	2,784	14.3
CAC	94.50%	0.35%	1,796	1,731	5.4

[0099] Table 4 summarizes test results from LiS batteries prepared using S°-impregnated biocarbon-based cathodes. ABC1 (activated carbon produced from lignin by the inventors) was impregnated with elemental sulfur at 59.46% (denoted as ABC1+60%S) and 49.08% (denoted as ABC1+50%S), respectively. CAC (commercially available control activated carbon) was also impregnated with sulfur at 48.25% (denoted as CAC+50%S).

[0100] For the fabrication method for the cathode of the LiS battery, the S°-impregnated carbon product, polyvinyl fluoride (PVF as a binder), and graphite (as a conductivity enhancing agent, given the high sulphur content of the sulphur-impregnated carbon product which reduced its conductivity) were formed into a paste using N-Methyl-2-pyrrolidone (NMP). The mass ratio of this carbon-sulfur (C-S) composite was 60:20:20, respectively. These C-S composites were coated on aluminum foil and cathodes (15 mm in diameter) were prepared for the assembly of LiS batteries (CR 2032 type) in the Ar-filled glove box.

[0101] Four cells were assembled in each group. The electrolyte (containing 1 M of lithium bis(trifluoromethane sulfonyl)imide ($LiN(SO_2CF_3)_2$) and 1% lithium nitrate in 1,2-dimethoxyethane (DME) and 1,3-dioxolane (DOXL)) was added to each cell using an electrolyte volume per 1 mg of sulfur in the cathode ranging from 100 to 115 µL. Once all LiS batteries were assembled, the initial voltage of the button cell was measured. As presented below, all button cells showed an initial voltage (> 2.3 V) with consistency except for one button cell in each of the ABC1+60%S and CAC+50%S groups. It was believed that the cathode and anode may have misaligned during assembly resulted in a direct connection which led to the malfunction of these particular LiS batteries.

Table 4. LiS batteries fabricated using S-impregnated activated biocarbon cathodes.

Carbon type	Coin cell ID(#)	Voltage (V)
ABC1+60%S (S°-impregnated ABC1 at sulfur content of 59.46%)	S8-5	2.355
	S8-6	0.000
	S8-7	2.310
	S8-8	2.370
ABC1+50%S (S° impregnated ABC1 at sulfur content of 49.08%	S9-9	2.384
	S9-10	2.388
	S9-11	2.382
	S9-12	2.419
CAC+50%S (S°-impregnated CAC at sulfur content of 48.25%)	S11-5	2.360
	S11-6	2.397
	S11-7	0.000
	S11-8	2.397

[0102] FIG. 7 shows the profile of the discharge curve from GCD (Galvano charge and discharge) tests. A representative cell from each group (S8-8 for the ABC1+60%S group, S9-11 for the ABC1+50%S group and S11-5 for the CAC+50%S group) was discharged and charged in order 3 times at full voltage range (1.4 V to 3.3V). S8-8 (ABC1+60%S) and S9-11 (ABC1+50%S) achieved a capacity of 555.9 mA/g and 614.6 mAh/g respectively, at a current density of 1675 mA/g which is a fast discharge while S11-5 (CAC+50%S) performed at a capacity of 417.6 mAh/g at a current density of 1,675 mA/g. The discharge voltage in FIG. 7 shows a typical voltage profile of the LiS battery at a constant current that has 2 plateaux. The first plateau is ground at 2.3V. The elemental sulfur (S_8) is reduced to S_2^{2-} from the 1st plateau to the end of the 2nd plateau at 2.1V. The S_2^{2-} is further reduced to S^{2-} at the 2nd transition point. This voltage profile reflects that S_8 embedded in pores of the activated carbon products is broken down to S^{2-} through the serial reduction process when the LiS battery discharges.

[0103] Of these cells, S8-8 for the ABC1+60%S group, S9-11 for the ABC1+50%S group, and S11-5 for the CAC+50%S group were continuously cycled at the full voltage range.

[0104] FIG. 8A, 8B, and 8C shows discharge capacity (mAh/g) across 30 cycles of charge and discharge. All of these LiS batteries showed lower capacity retention percentages (39.5% for S8-8 (FIG 8A), 9.7% for S9-11 (FIG 8B), and 8.6% for S11-5 (FIG. 8C)).

[0105] Table 5 shows each reduction of sulfur from S_8 to S_1. One sulfur ion at each reduction point is able to react with 2 ions of Li ionized from the Li metal anode during the discharge. The completion of this whole serial reduction includes 1 mole of elemental sulfur (S_8) using 16 moles of elemental Li. The last two reduction steps create Li_2S_2 and Li_2S, which are insoluble in the electrolyte in the LiS battery system. These insoluble polysulphides eventually solidify on the surface of the Li anode. As a result, the availability of sulphur (as an active material impregnated in activated biocarbon) lessens and the Li anode can not be ionized because of the blockage by these insoluble polysulphides (Li_2S_2 and Li_2S). Existing literature (e.g. Nazar et al., 2014, Lithium-sulfur batteries, MRS Bulletin, vol. 39; Lin and Liang, 2015, J. Materials Chemistry A, 3:919-1346; and Fotouhi, et al., 2017, Energies, 10:1937) reveals the fundamental chemistry issues in the LiS battery system.

[0106] In this study, LiS batteries were tested in a manner designed to avoid this insoluble polysulfide formation on the Li anode by using an adjusted voltage range from 3.0V to 1.96V, which is called the soluble voltage or 75% Depth of Discharge (DoD). Theoretically, 16 electrons are available at the full voltage range, while 12 electrons are available within the soluble voltage range. This means that the full capacitance will be 25% reduced within the soluble voltage range.

Table 5. Reduction of sulfur to polysulfides during discharge in the LiS battery system.

Each reduction of sulfur to polysulfides during discharge	Species of polysulfides after each reduction during discharge	Solubility in electrolyte	Soluble voltage range
$2Li^+ + S_8^- + 2e^-$	Li_2S_8	Soluble	Li_2S_2 started forming and depositing Li_2S_2 solids on the Li anode at a voltage of less than 1.960V
$2Li^+ + Li_2S_8^- + 2e^-$	Li_2S_7	Soluble	
$2Li^+ + Li_2S_7^- + 2e^-$	Li_2S_6	Soluble	
$2Li^+ + Li_2S_6^- + 2e^-$	Li_2S_5	Soluble	
$2Li^+ + Li_2S_5^- + 2e^-$	Li_2S_4	Soluble	
$2Li^+ + Li_2S_4^- + 2e^-$	Li_2S_3	Soluble	
$2Li^+ + Li_2S_3^- + 2e^-$	**Li_2S_2**	Insoluble (solids)	Li_2S_2 and Li_2S deposit on the Li anode at < 1.960 V (polysulfide shuttles)
$2Li^+ + Li_2S_2^- + 2e-$	**Li_2S_1**	Insoluble (solids)	

[0107] FIG. 9 shows discharge capacity (mAh/g) and capacity retention (%) of S8-5 (another cell representative for the ABC1+60%S group) when tested at a soluble voltage ranging from 1.95V to 3.0V. The capacity averaged 397.0 mAh/g with applied current density (1675 mA/g or 1C/g), which is a very fast discharge rate. Amazingly, the capacity retention was 98.6% (397.3 mAh/g) at the 100th cycle, indicating nearly no loss in capacitance.

[0108] FIG. 10 shows discharge capacity (mAh/g) and capacity retention (%) of S9-12 (another cell representative for the ABC1+50%S group) when also tested at a soluble voltage ranging from 1.96V to 3.0V. The capacity averaged 377.3 mAh/g when applied at a current density (1,675 mA/g or 1.0C/g) and 422.5 mAh/g when applied at a current density (838 mAh/g or 0.5C/g), which is used for most practical applications. Once again the capacity retention was high (97.97% at the 100th cycle). This high capacity retention (to 217 cycles) indicates that the formation of insoluble polysulphides was minimized by keeping the discharge within the soluble voltage range.

[0109] For the first ten cycles, capacity and retaining percentage were not stable because of an unpredictable power-cut from the test facility. For cycles 11-20, an average capacity of 377.3 mAh/g at a current density of 1675 mA/g (1.0C/g) was maintained. For cycles 21-42, capacity and retaining percentage were not stable because of several unpredictable power-cuts from the test facility. For cycles 42-217, an average capacity of 422.5 mAh/g at a current density of 838 mA/g (0.5 C/g, used for most practical applications) was maintained. At the 217th cycle, capacity was 404.8 mAh/g and capacity retained was 97.97%.

[0110] FIG. 11 shows discharge capacity (mAh/g) and capacity retention (%) of S11-6 (another cell representative for the CAC+50%S group) when tested across the soluble voltage ranging from 1.96V to 3.0V. The capacity averaged 187.7 mAh/g and was maintained over 184 mAh/g when applied at a current density of 1,675 mA/g (or 1.0C/g) and the capacity retention was 96.0% at the 100th cycle (185.4 mAh/g).

Example 3 – Optimization of Ratio of Electrolyte Volume to Sulphur Mass in Cathodes

[0111] Most previous literature has reported that a ratio of electrolyte volume to sulphur mass in the cathode strongly affects the cyclability of LiS batteries. When the electrolyte is added in an excess amount in the LiS battery, the excess amount can promote the migration of soluble polysulphides from the cathode to the anode (referred to as the shuttle effect), resulting in poor cyclability. Some literature recommended a minimum ratio ranging from 7 to 39 µL per 1 mg of sulphur in the cathode.

[0112] The inventors determined a minimum ratio of electrolyte volume (µL) to sulphur mass (mg) in the cathode, referred to as the ES ratio, using an initial open-circuit potential (OCP) (V), which is also referred to as the equilibrium potential after assembly, and capacity. The goal of this study was to determine the highest capacity at a minimum ES ratio.

[0113] FIG. 12A and FIG. 12B show the highest capacity ranging from 600 - 860 mAh/g and voltage ranging 2.3 to 2.5 V at an ES ratio of 40 - 75 (µL/mg of sulphur), respectively. FIG. 12C shows that the highest capacity of 600 - 860 mAh/g was obtained when the initial OCP was in the 2.3- 2.5V range. In FIG. 12A, the box shown in broken lines indicates the approximate capacity ranging from 600 – 860 mAh/g at an ES ratio of 40-75. In FIG. 12B, the box shown in broken lines indicates the approximate voltage ranging from 2.3 to 2.5 at an ES ratio of 40-75. In FIG. 12C, the box shown in broken lines indicates the approximate capacity ranging from 600 – 860 mAh/g with an OCP of 2.3 to 2.5 V.

Example 4 – Performance of 500 Cycled LiS Batteries with Minimum ES Ratio

[0114] FIG. 13 shows the initial two cycles of GCD from LiS batteries with sulfur-impregnated ABC1 (65.25% sulfur content) and a minimum electrolyte amount. These cells were tested at a current density of 1C/g of active sulfur or 1675 mA/g of active sulfur and 100% depth of discharge (DoD) which ranges voltage from 1.7V to 3.0V. LiS battery cells 10-3 and 10-4 with sulfur impregnated ABC1 achieved an initial capacity ranging 688 - 720 mAh/g tested 1 C/g (1,675 mA/g) at 100% DoD (1.7 – 3.0V). These cells (CR 2032, 15 mm in diameter) had the following components:

- Sulfur impregnated activated carbon (S-AC): 65.25% in S-AC

- Cathode: 60 (AC-S):20 (graphite): 20 (PVDF) coated on aluminum foil
- Electrolyte: 1 M of lithium bis(trifluoromethane sulfonyl)imide ($LiN(SO_2CF_3)_2$) and 1% lithium nitrate in 1,2-dimethoxyethane (DME) and 1,3-dioxolane (DOXL).
- Electrolyte amount used: 40 – 52 µL per 1 mg of active sulfur in the cathode
- Separator: Celgard 25 micrometer film
- Anode: 0.2 mm lithium metal

[0115] Table 6 and FIG. 14A, 14B, 14C and 14D show experimental results for LiS batteries (Cells 10-3 and 10-4) with sulphur impregnated lignin-based activated carbon which were 500 cycled at 1C/g at 100% DoD and 75% DoD, respectively (following the first two cycles conducted at 100% DoD to establish full capacity for cell 10-3 and shown for both cells in FIG. 13). FIG. 14A shows that Cell 10-3 was GCD-cycled 500 times at a current density of 1C/g and DoD of 75% (voltage ranging from 1.96 to 3.0V, 458 mAh/g) to avoid shuttle effect, while Cell 10-4 GCD-cycled 500 times at a current density of 1C/g and DoD of 100% (voltage ranging from 1.7 to 3.0V, 497 mAh/g). FIG. 14B shows the capacitance retention (%) of Cells 10-3 and 10-4 which were calculated based on the initial capacity of each cell (688 mAh/g and 67% for 10-3 (75% DoD) and 752 mAh/g and 66% for Cell 10-4 (100% DoD)).

[0116] FIGs. 14C and 14D present capacity (mAh/g) (FIG. 14C) and capacity retention (%) (FIG. 14D) from the 40th to 500th cycles, respectively. Cells 10-3 and 10-4 achieved 458 mA (67% capacity retention) at 75%DoD and 497 mAh/g (66% capacity retention) at 100% DoD after 500 cycles, respectively. A capacity decay was -0.0001% per cycle after 40 cycles of Cell 10-3 when tested at 1C and 75% DoD (1.96 – 3.0V) which is much slower than a capacity decay (-0.0005% per cycle for Cell 10-4) when tested at 1C and 100% DoD (1.7 – 3.0V). The corresponding values represented a decay of -0.0865 mAh/g per cycle for 10-3 at 75% DoD and -0.3922 mAh/g per cycle for 10-4 at 100% DoD.

Table 6. LiS batteries with sulfur impregnated lignin-based AC 500 cycled at 1C/g and 100% DoD and 75% DoD, respectively.

Cell ID	Parameters	# of cycles						
		1	40	100	200	300	400	500
10-3 (75%DoD)	Capacity (mAh/g)	687.7	487.6	522.0	488.5	514.6	473.7	457.8
	Capacity retention (%)	100%	70.9%	75.9%	71.0%	74.8%	68.9%	66.6%

10-4 (100%DoD)	Capacity (mAh/g)	751.9	668.6	649.1	624.8	547.4	531.8	496.0
	Capacity retention (%)	100%	88.9%	86.3%	83.1%	72.8%	70.7%	66.0%

[0117] Using the capacity decay constants derived from Table 6 and FIGs 14C and 14D, Table 7 summarizes the estimated capacity of cells 10-3 and 10-4 at the 1000th cycle, assuming consistent performance after the 40th cycle. The predicted capacity of cell 10-3 cycled at 75% DoD after the first two cycles would be higher than cell 10-4 cycled at 100% DoD after 1000 charge and discharge cycles.

Table 7. Estimated Capacity of Cells 10-3 and 10-4 after 1000 cycles.

Cell ID	Inputs		Capacity decay constants		Estimated values	
	Initial Cap (mAh/g)	X (# of cycles)	aX	b	Y (capacity retention %)	Capacity (mAh/g) after 1000 cycles
LC21-10-3 (75% DoD)	687.7	1000	-0.0001	0.7459	64.6%	444
LC21-10-4 (100% DoD)	751.9	1000	-0.0005	0.9136	41.4%	311

[0118] To summarize, the highest capacity ranging from 600 - 860 mAh/g with voltage ranging from 2.3 to 2.5 V was obtained when an ES ratio of 40 - 75µL/mg of sulfur in the ABC1-based cathode (or cathode with lignin-based AC) was used. Retention of capacitance can at least in some embodiments be maximized by changing to the soluble voltage range or controlling to 75% of the depth of discharge.

[0119] While a number of exemplary aspects and embodiments have been discussed above, those of skill in the art will recognize certain modifications, permutations, additions and sub-combinations thereof. It is therefore intended that the following appended claims and claims hereafter introduced are interpreted to include all such modifications, permutations, additions and sub-combinations as are consistent with the broadest interpretation of the specification as a whole.

CLAIMS:

1. A method for embedding sulphur into conductive carbon, the method comprising:
dissolving elemental sulphur in liquid ammonia to form a sulphur-ammonia solution;
soaking conductive carbon in the sulphur-ammonia solution to embed the conductive carbon with at least a portion of the dissolved sulphur; and
recovering the sulphur-embedded conductive carbon.

2. The method as defined in claim 1 or any other claim herein, further comprising, after soaking the conductive carbon in the sulphur-ammonia solution but before recovering the sulphur-embedded conductive carbon, removing ammonia from the sulphur-ammonia solution as gaseous ammonia.

3. The method as defined in any one of claims 1 or 2 or any other claim herein, wherein the elemental sulphur is dissolved in the liquid ammonia in a first pressurized environment.

4. The method as defined in claim 3 or any other claim herein, wherein the first pressurized environment has a pressure of between about 110 psig and about 135 psig at room temperature.

5. The method as defined in claim 3 or any other claim herein, wherein the first pressurized environment has a temperature of between -20°C and 20°C.

6. The method as defined in claim 3 or any other claim herein, wherein the first pressurized environment has a pressure of between 10 and 15 psig at a temperature of about -20°C.

7. The method as defined in any one of claims 1 to 6 or any other claim herein, wherein the step of dissolving elemental sulphur in liquid ammonia further comprises mixing the sulphur-ammonia solution to encourage solubilization of the elemental sulphur in the liquid ammonia.

8. The method as defined in any one of claims 1 to 7 or any other claim herein, wherein the sulphur-ammonia solution comprises between 5% and 30% by weight of dissolved sulphur.

9. The method as defined in any one of claims 1 to 8 or any other claim herein, further comprising removing any undissolved elemental sulphur from the sulphur-ammonia solution after the step of dissolving the elemental sulphur in the liquid ammonia, optionally by passing the sulphur-ammonia solution through a screen.

10. The method as defined in any one of claims 1 to 9 or any other claim herein, wherein the step of soaking the conductive carbon in the sulphur-ammonia solution is carried out in a second pressurized environment.

11. The method as defined in claim 10 or any other claim herein, wherein the second pressurized environment has a pressure of about 110 to about 135 psig at room temperature.

12. The method as defined in any one of claims 1 to 11 or any other claim herein, wherein the liquid ammonia in the sulphur-ammonia solution is removed by evaporating liquid ammonia into gaseous ammonia.

13. The method as defined in claim 12 or any other claim herein, wherein the liquid ammonia is removed by depressurizing the second pressurized environment.

14. The method as defined in either one of claims 12 or 13 or any other claim herein, wherein the ammonia is recovered through a wet collection method.

15. The method as defined in claim 14, further comprising drying the recovered sulphur-embedded conductive carbon.

16. The method as defined in either one of claims 12 or 13 or any other claim herein, wherein the ammonia is recovered through a dry collection method.

17. The method as defined in any one of claims 12 to 16 or any other claim herein, comprising recovering the gaseous ammonia and compressing the recovered gaseous ammonia back to liquid ammonia.

18. A method for embedding sulphur into conductive carbon, the method comprising: soaking conductive carbon in a sulphur-ammonia solution to embed the conductive carbon with at least a portion of the dissolved sulphur.

19. The method as defined in any one of claims 1 to 18 or any other claim herein, wherein the conductive carbon comprises activated carbon, graphite, graphene, or carbon nanotubes.

20. The method as defined in any one of claims 1 to 19 or any other claim herein, wherein the conductive carbon comprises activated carbon.

21. The method as defined in any one of claims 1 to 20 or any other claim herein, wherein the elemental sulphur comprises S_8 and/or granular elemental sulphur.

22. The method as defined in any one of claims 1 to 21 or any other claim herein, wherein the sulphur-embedded conductive carbon contains between 30% to 85% sulphur by weight, optionally between 50% and 70% sulphur by weight.

23. A sulphur-embedded conductive carbon containing between 30% to 85% sulphur by weight, optionally between 50% and 70% sulphur by weight.

24. The sulphur-embedded conductive carbon as defined in claim 23, wherein the conductive carbon comprises activated carbon.

25. Use of the sulphur-embedded conductive carbon as defined in either one of claims 23 or 24 or produced by a method as defined in any one of claims 1 to 22 as a sulphur cathode of a metal-sulphur battery.

26. The use as defined in claim 25, wherein the metal-sulphur battery is a lithium-sulphur battery.

27. A metal-sulphur battery comprising the sulphur-embedded conductive carbon as defined in either one of claims 23 or 24 or produced by a method as defined in any one of claims 1 to 22 as a sulphur cathode.

28. A lithium-sulphur battery comprising the sulphur-embedded conductive carbon as defined in either one of claims 23 or 24 or produced by a method as defined in any one of claims 1 to 22 as a sulphur cathode.

29. A metal-sulphur or lithium-sulphur battery as defined in either one of claims 27 or 28, containing electrolyte in a volume of between 30 to 80 µL/mg of sulphur, optionally between 40 to 75 µL/mg of sulphur.

30. A method of operating a battery as defined in any one of claims 27 to 29, comprising cycling the battery only to 75% depth-of-discharge when charging and discharging the battery.

31. An apparatus for embedding sulphur into conductive carbon, the apparatus comprising:

a sulphur dissolving tank comprising a first inlet for flowing liquid ammonia into the sulphur dissolving tank, a second inlet for loading elemental sulphur into the sulphur dissolving tank, and an outlet for withdrawing a sulphur-ammonia solution from the sulphur dissolving tank; and

a sulphur impregnating tank in selective fluid communication with the sulphur dissolving tank, the sulphur impregnating tank comprising a first inlet connected to the outlet of the sulphur dissolving tank, a second inlet for adding conductive carbon into the sulphur impregnating tank, a release valve for evaporating the liquid ammonia, and a release conduit for discharging gaseous ammonia.

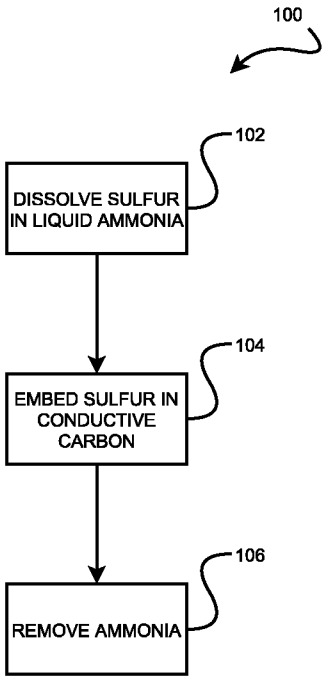

100
102
DISSOLVE SULFUR IN LIQUID AMMONIA
104
EMBED SULFUR IN CONDUCTIVE CARBON
106
REMOVE AMMONIA

FIG. 1

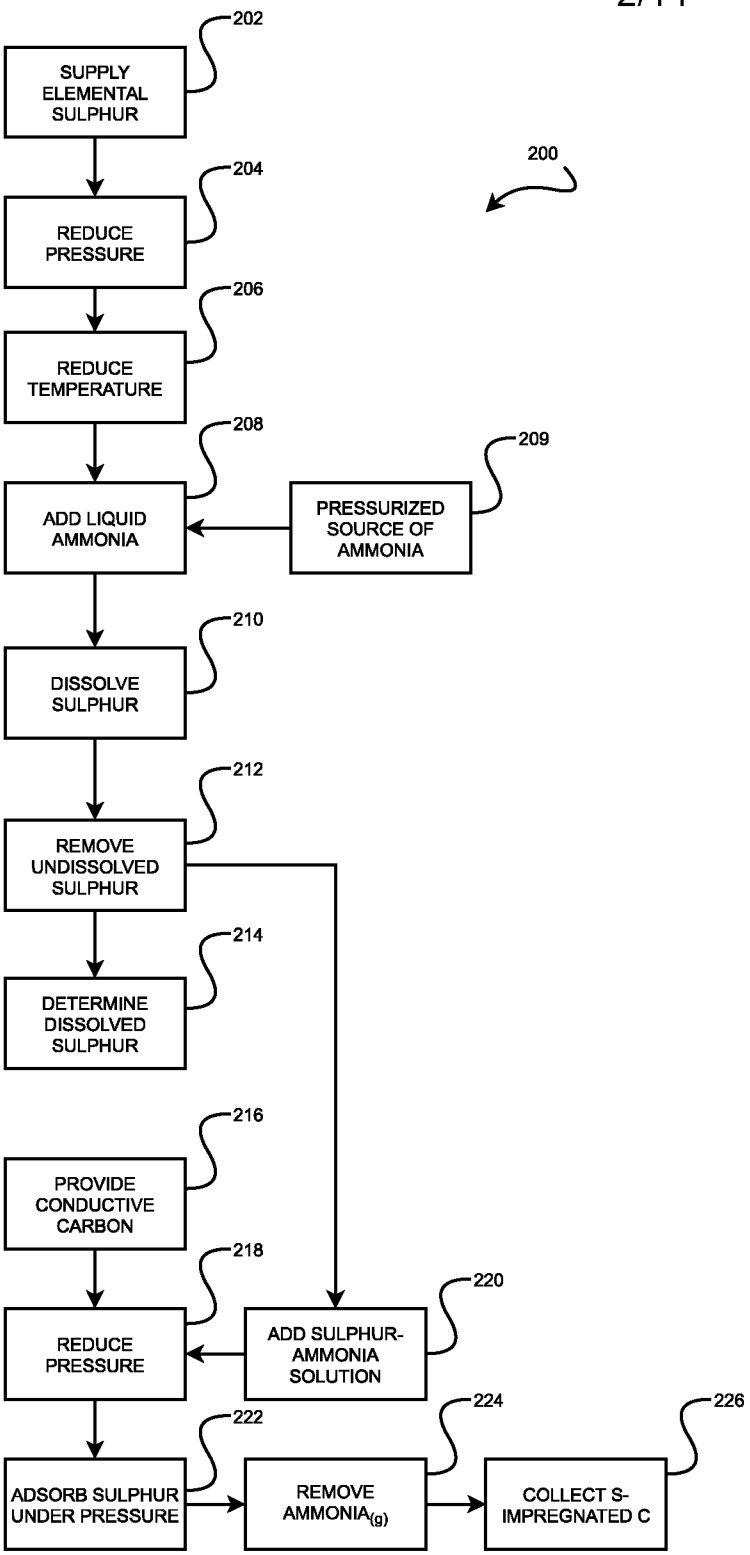

200
202
SUPPLY ELEMENTAL SULPHUR
204
REDUCE PRESSURE
206
REDUCE TEMPERATURE
208
ADD LIQUID AMMONIA
209
PRESSURIZED SOURCE OF AMMONIA
210
DISSOLVE SULPHUR
212
REMOVE UNDISSOLVED SULPHUR
214
DETERMINE DISSOLVED SULPHUR
216
PROVIDE CONDUCTIVE CARBON
218
REDUCE PRESSURE
220
ADD SULPHUR-AMMONIA SOLUTION
222
ADSORB SULPHUR UNDER PRESSURE
224
REMOVE AMMONIA(g)
226
COLLECT S-IMPREGNATED C

FIG. 2A

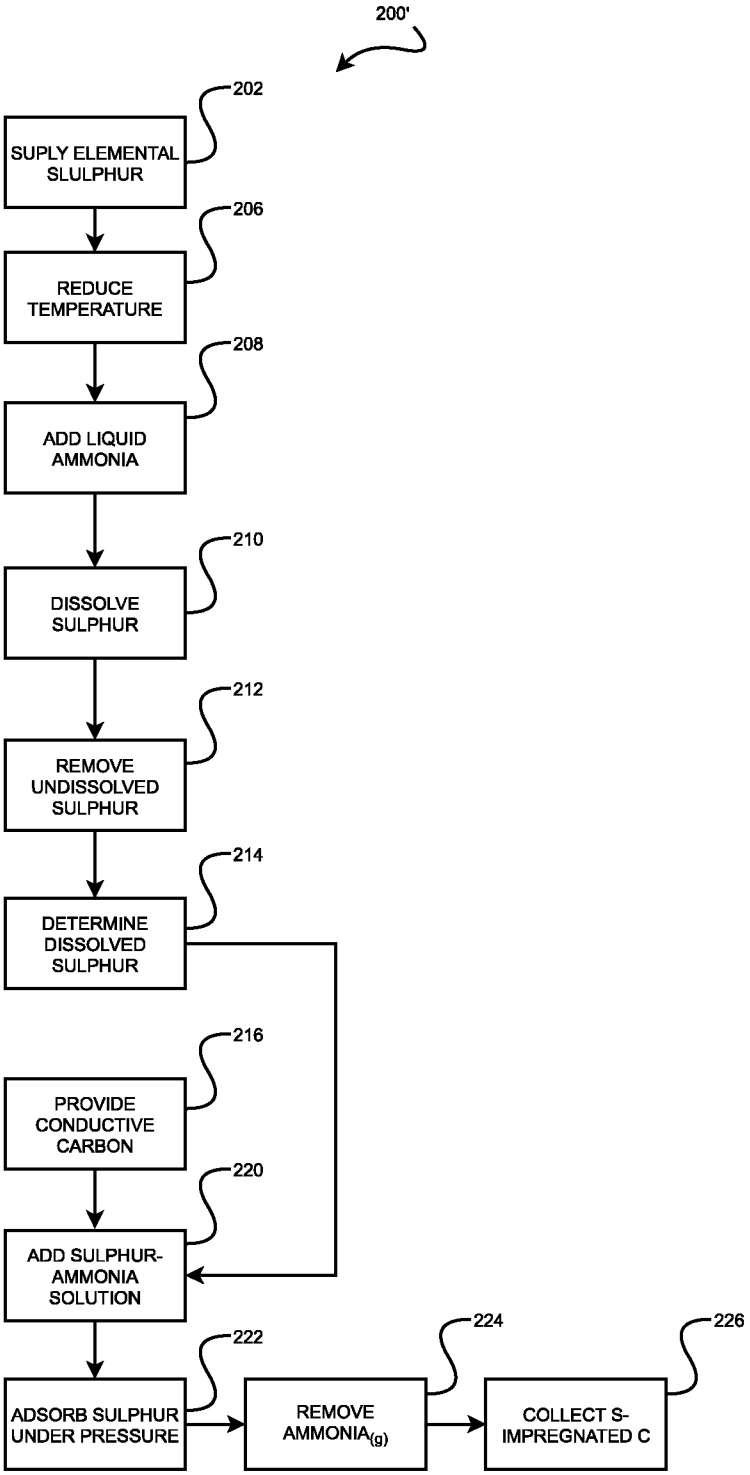

200'
202
SUPLY ELEMENTAL SLULPHUR
206
REDUCE TEMPERATURE
208
ADD LIQUID AMMONIA
210
DISSOLVE SULPHUR
212
REMOVE UNDISSOLVED SULPHUR
214
DETERMINE DISSOLVED SULPHUR
216
PROVIDE CONDUCTIVE CARBON
220
ADD SULPHUR-AMMONIA SOLUTION
222
ADSORB SULPHUR UNDER PRESSURE
224
REMOVE AMMONIA(g)
226
COLLECT S-IMPREGNATED C

FIG. 2B

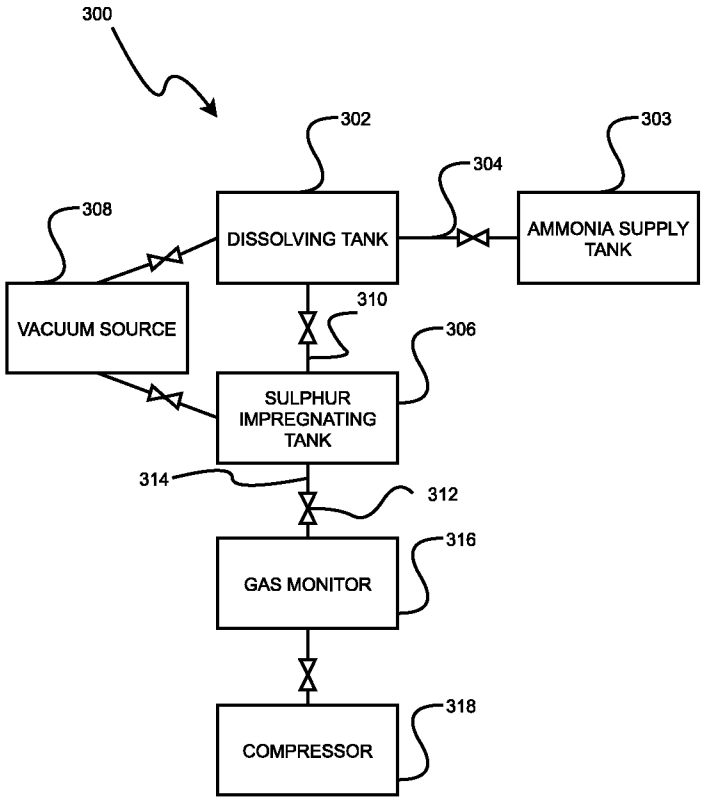

300
302
303
304
308
DISSOLVING TANK
AMMONIA SUPPLY TANK
VACUUM SOURCE
310
306
SULPHUR IMPREGNATING TANK
314
312
316
GAS MONITOR
318
COMPRESSOR

FIG. 3

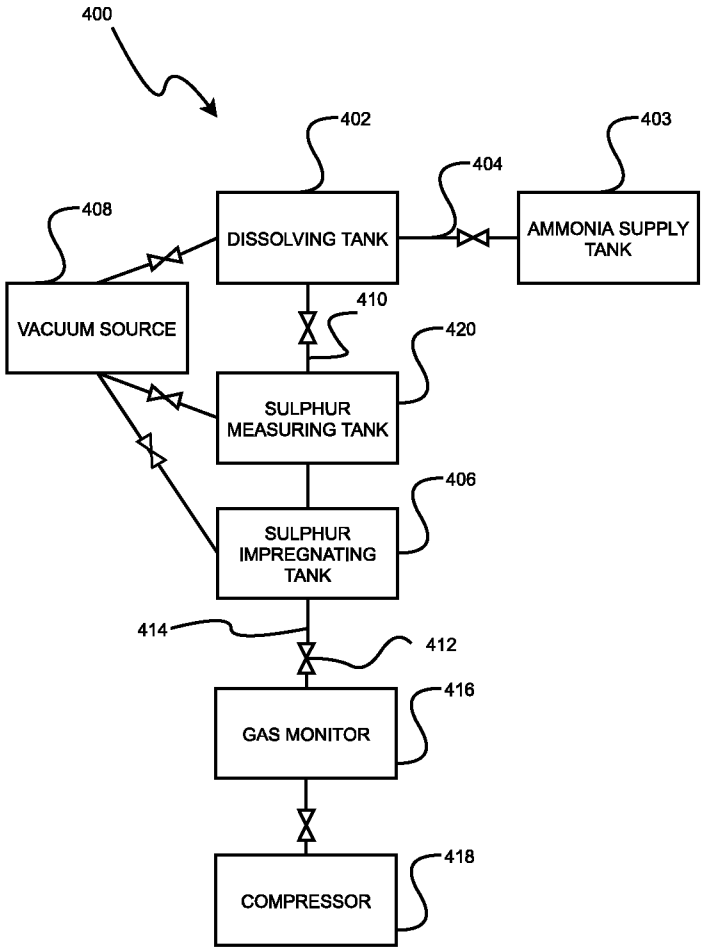

400
402
403
404
408
DISSOLVING TANK
AMMONIA SUPPLY TANK
VACUUM SOURCE
410
420
SULPHUR MEASURING TANK
406
SULPHUR IMPREGNATING TANK
414
412
416
GAS MONITOR
418
COMPRESSOR

FIG. 4

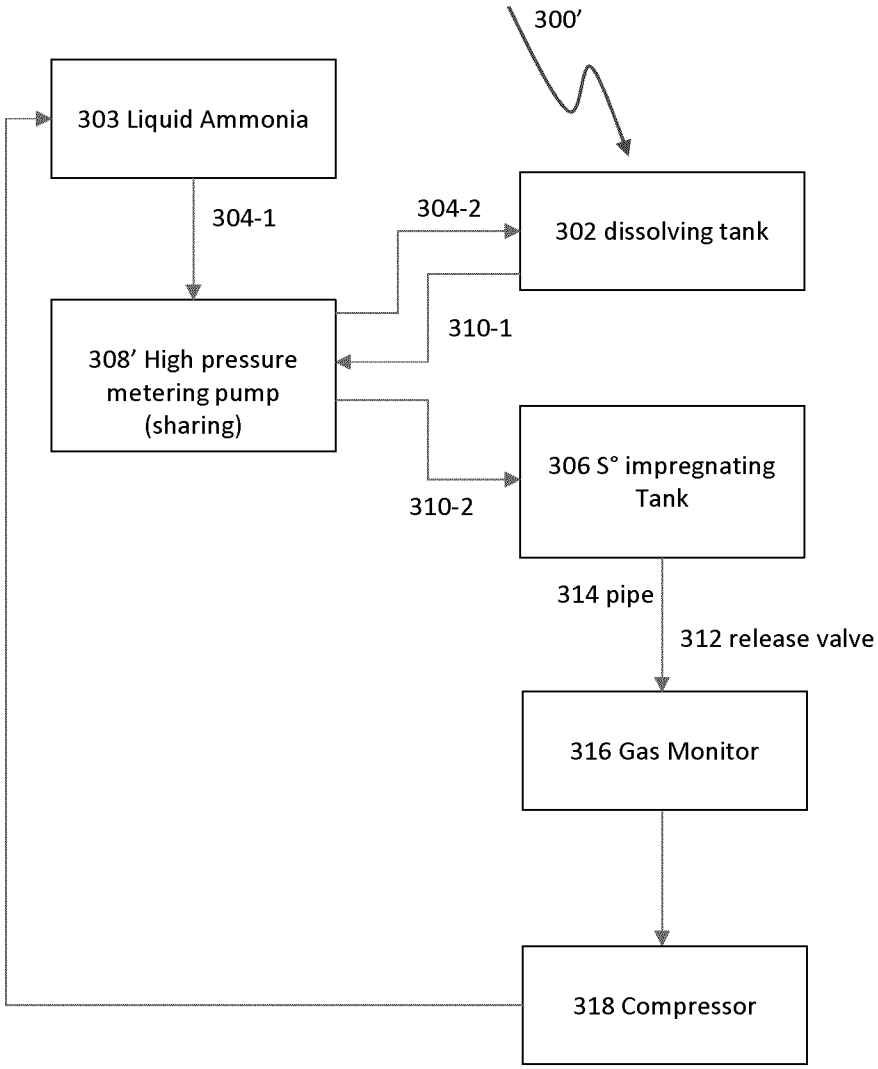

300'
303 Liquid Ammonia
304-1
304-2
302 dissolving tank
310-1
308' High pressure metering pump (sharing)
306 S° impregnating Tank
310-2
314 pipe
312 release valve
316 Gas Monitor
318 Compressor

FIG. 5

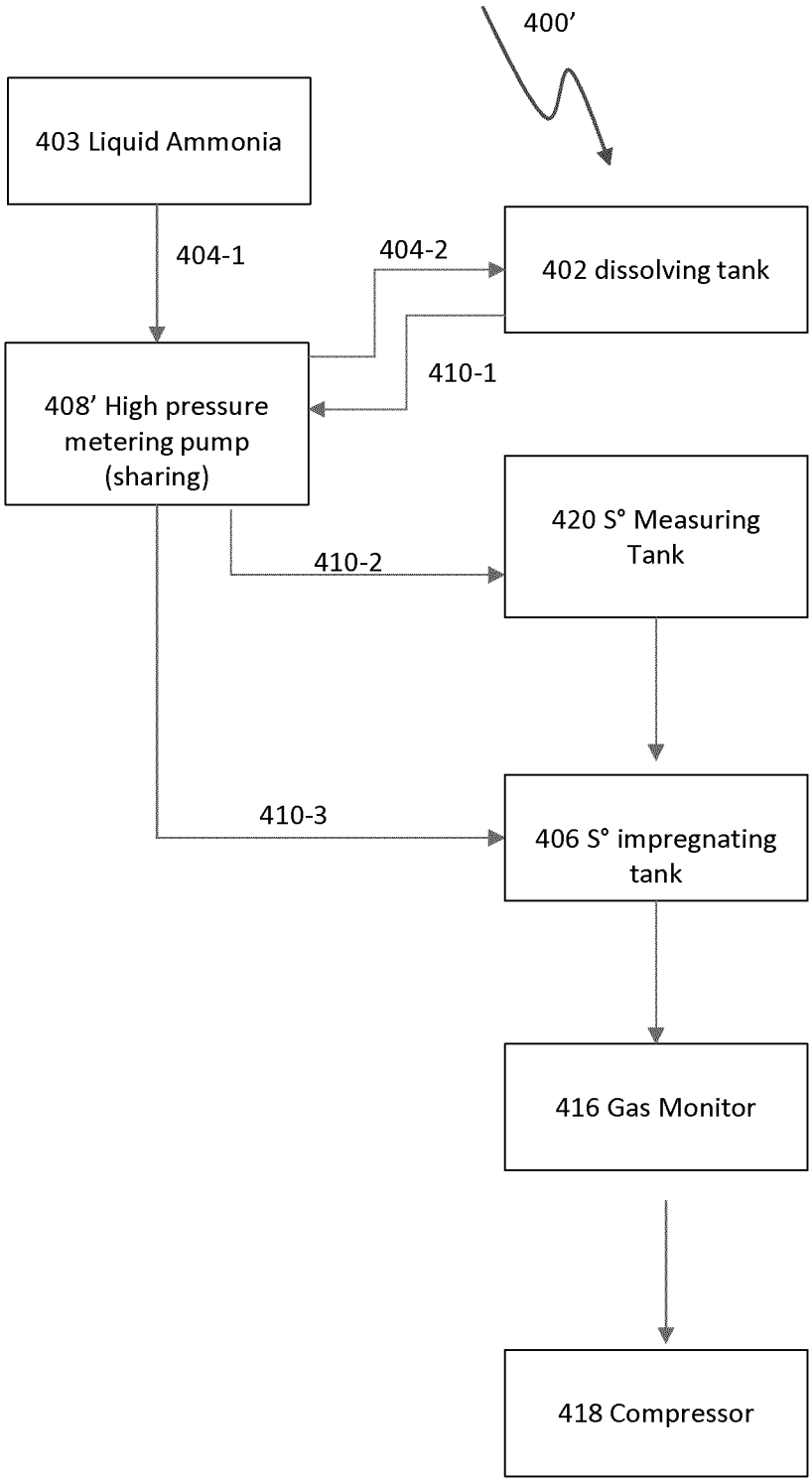

400'
403 Liquid Ammonia
404-1
404-2
402 dissolving tank
410-1
408' High pressure metering pump (sharing)
420 S° Measuring Tank
410-2
410-3
406 S° impregnating tank
416 Gas Monitor
418 Compressor

FIG. 6

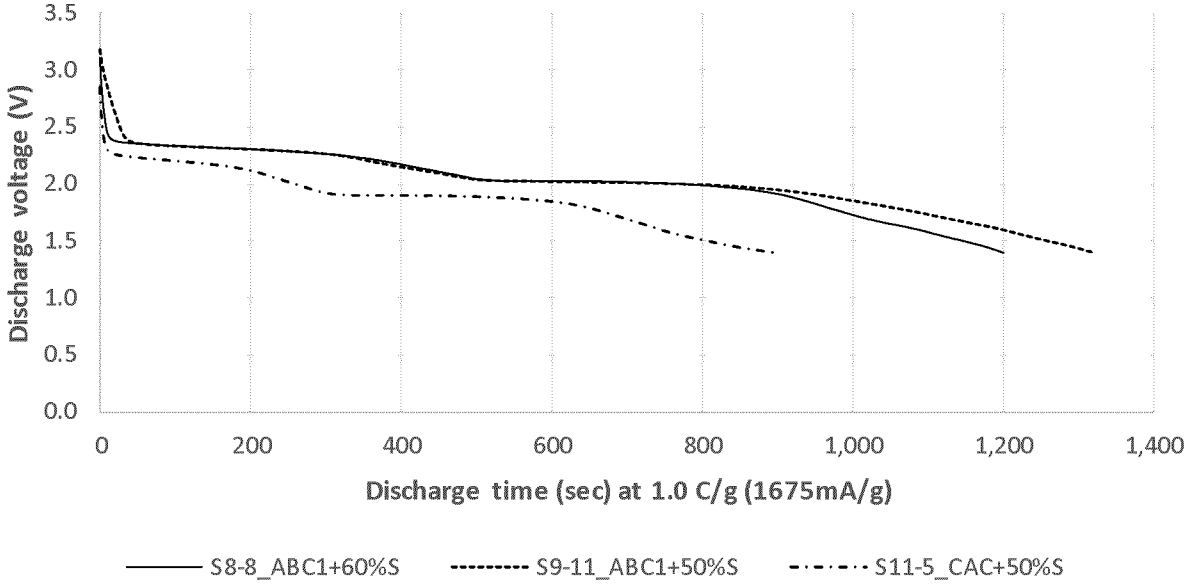

3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
Discharge voltage (V)
0
200
400
600
800
1,000
1,200
1,400
Discharge time (sec) at 1.0 C/g (1675mA/g)
S8-8_ABC1+60%S
S9-11_ABC1+50%S
S11-5_CAC+50%S

FIG. 7

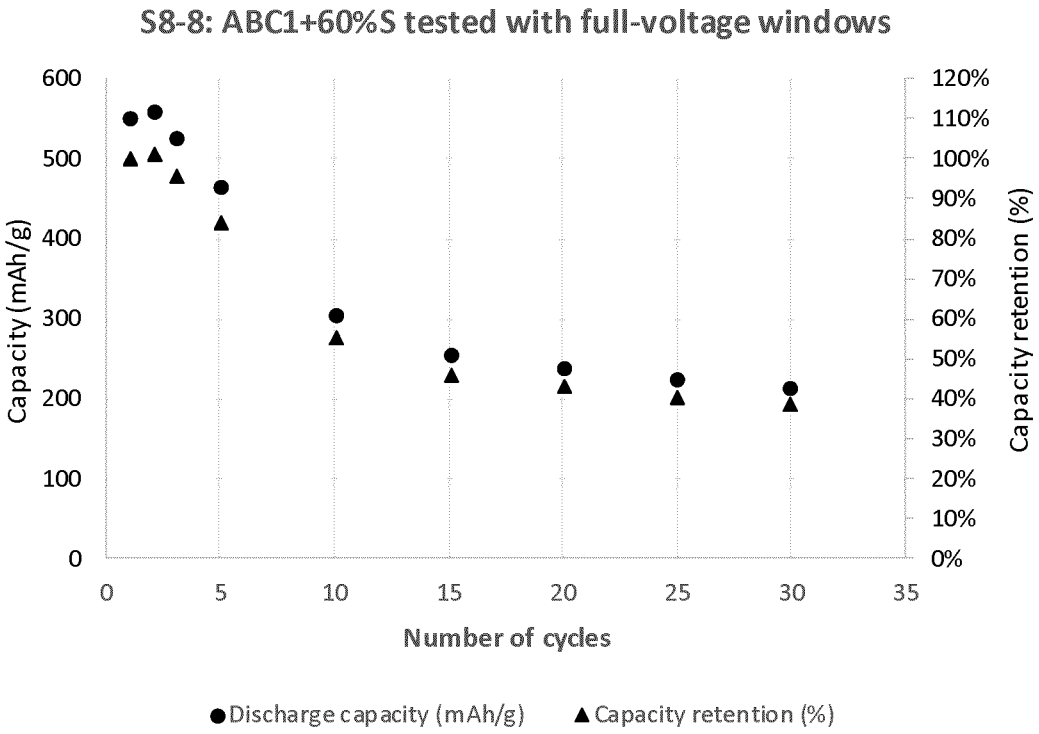

S8-8: ABC1+60%S tested with full-voltage windows
600
500
400
300
200
100
0
Capacity (mAh/g)
120%
110%
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
Capacity retention (%)
0
5
10
15
20
25
30
35
Number of cycles
Discharge capacity (mAh/g)
Capacity retention (%)

FIG. 8A

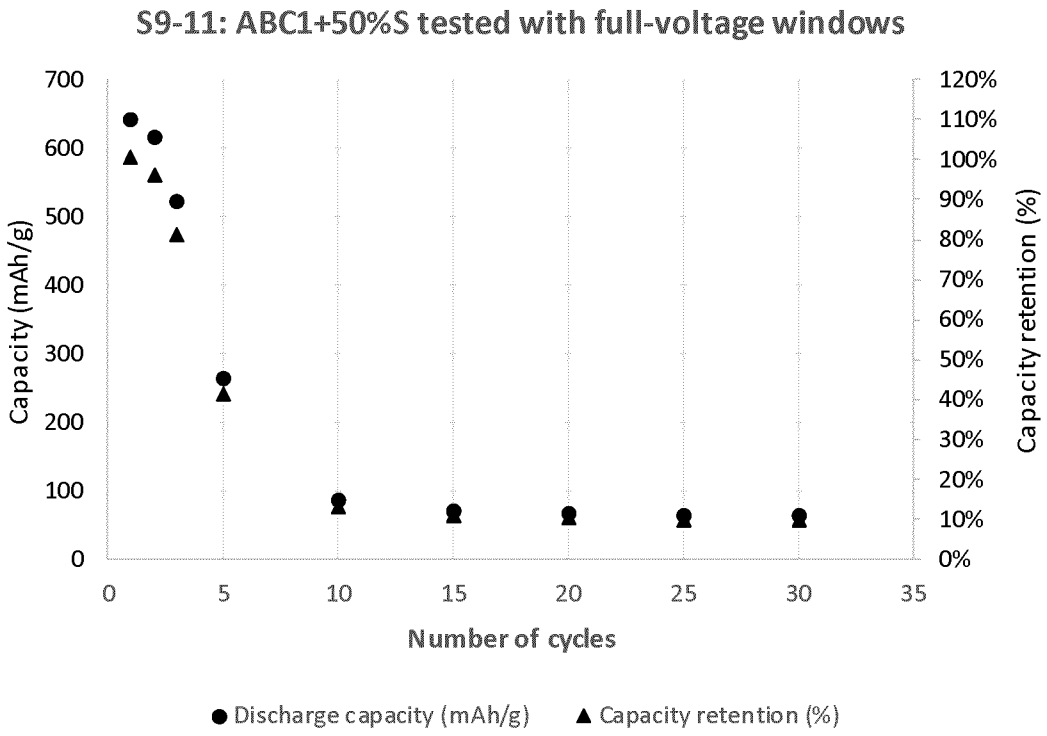

S9-11: ABC1+50%S tested with full-voltage windows
Capacity (mAh/g)
Capacity retention (%)
Number of cycles
Discharge capacity (mAh/g)
Capacity retention (%)

FIG. 8B

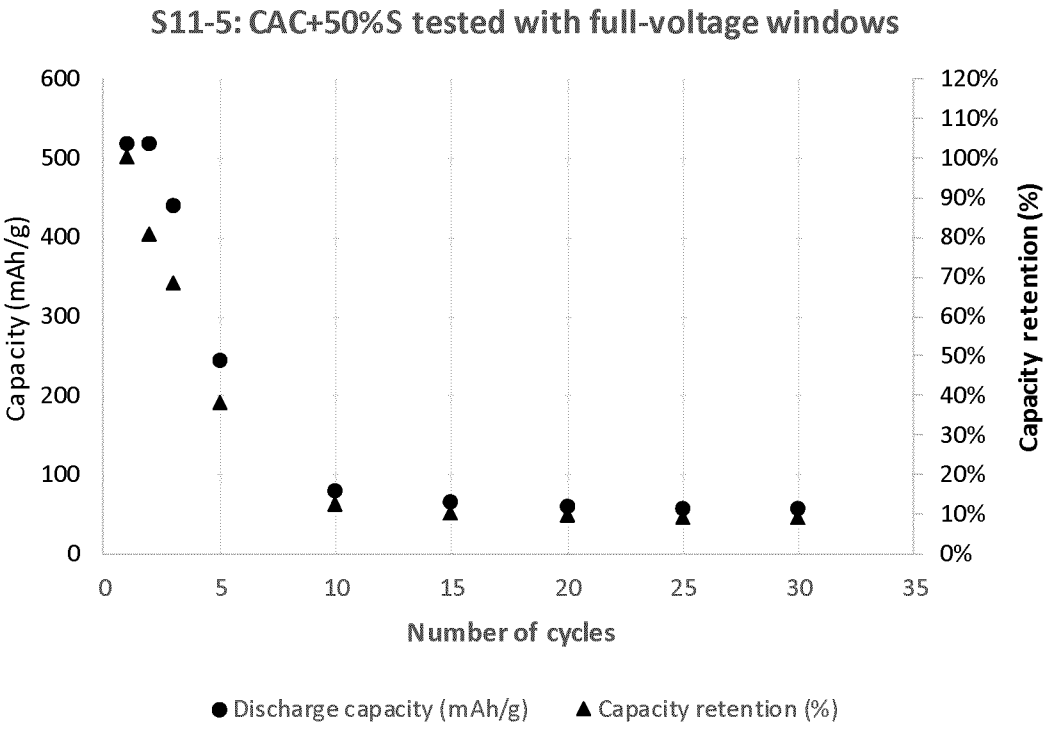

S11-5: CAC+50%S tested with full-voltage windows
Capacity (mAh/g)
Capacity retention (%)
Number of cycles
Discharge capacity (mAh/g)
Capacity retention (%)

FIG. 8C

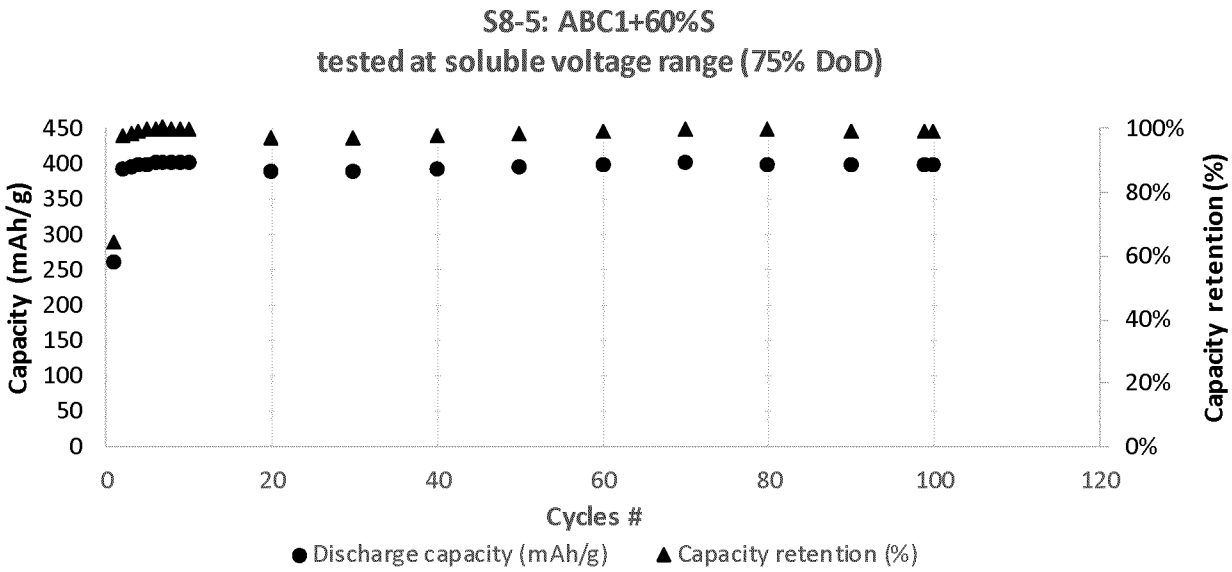

S8-5: ABC1+60%S
tested at soluble voltage range (75% DoD)
Capacity (mAh/g)
Capacity retention (%)
Cycles #
Discharge capacity (mAh/g)
Capacity retention (%)

FIG. 9

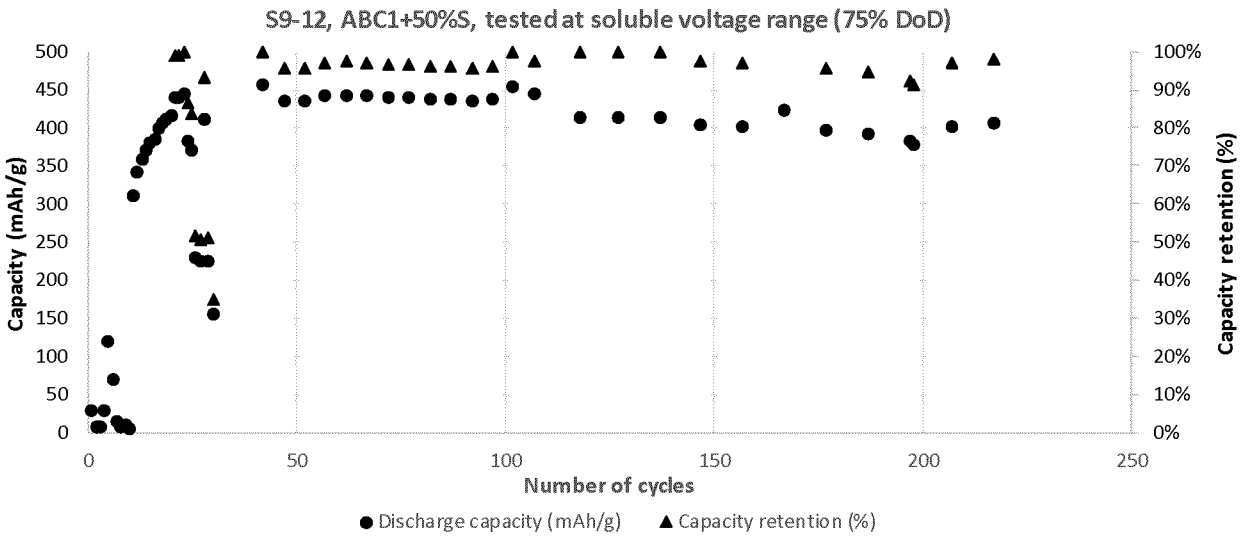

S9-12, ABC1+50%S, tested at soluble voltage range (75% DoD)
Capacity (mAh/g)
Capacity retention (%)
Number of cycles
Discharge capacity (mAh/g)
Capacity retention (%)

FIG. 10

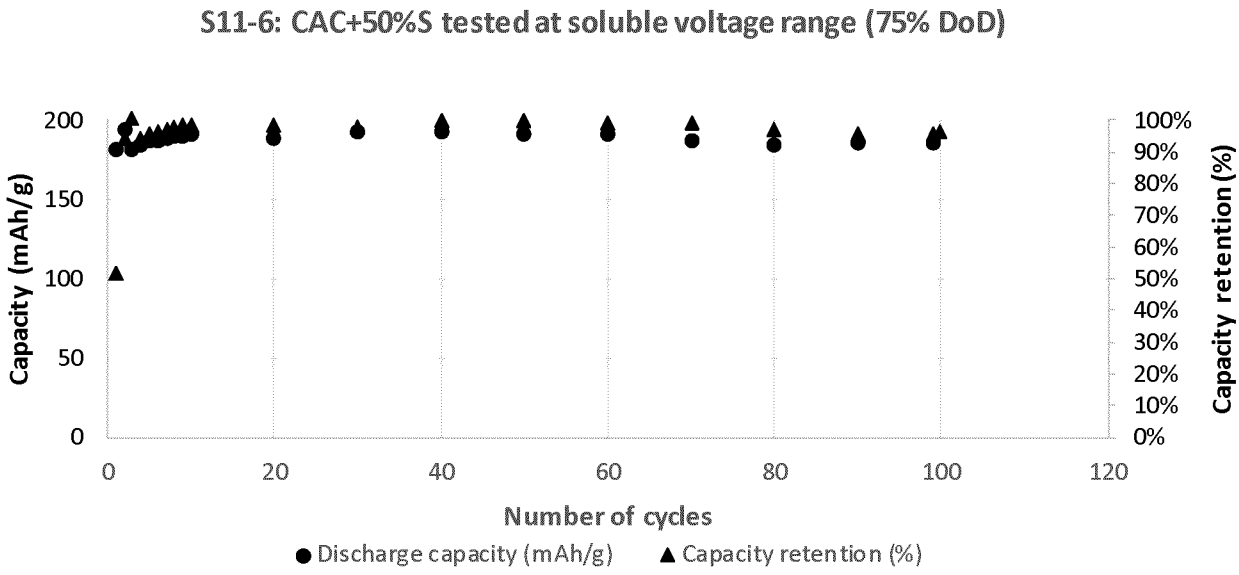


FIG. 11

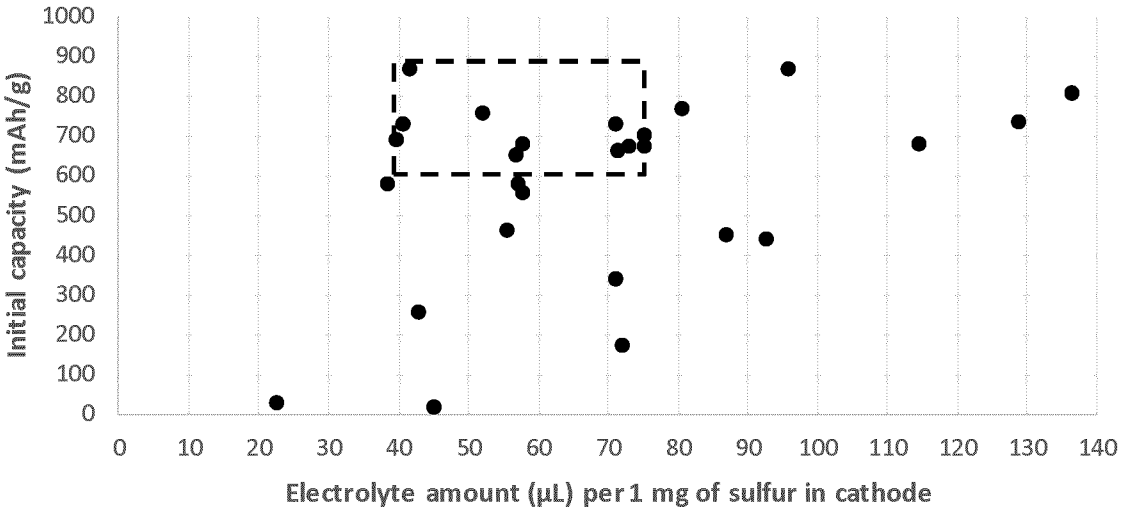


FIG. 12A

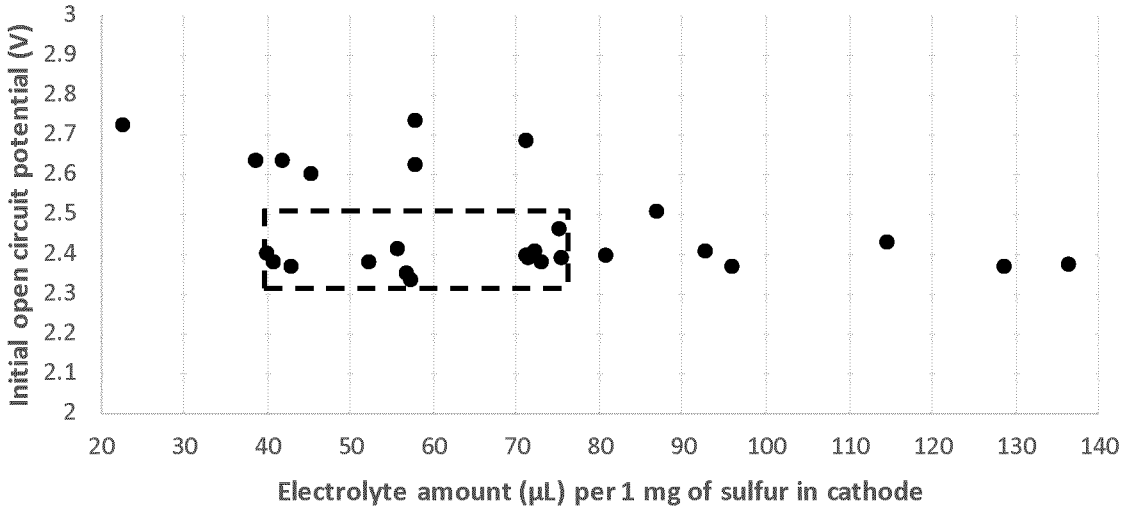


FIG. 12B

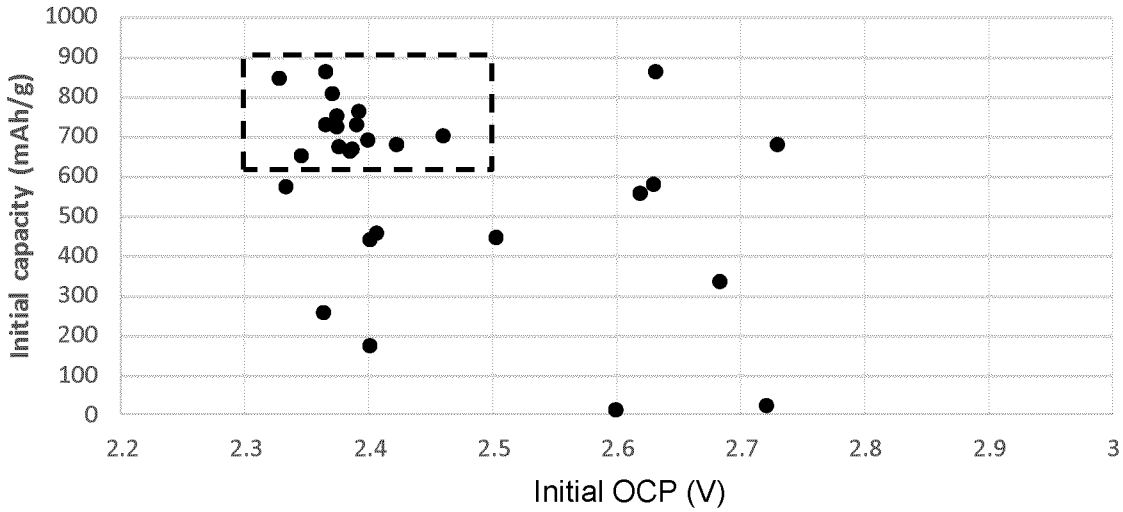


FIG. 12C

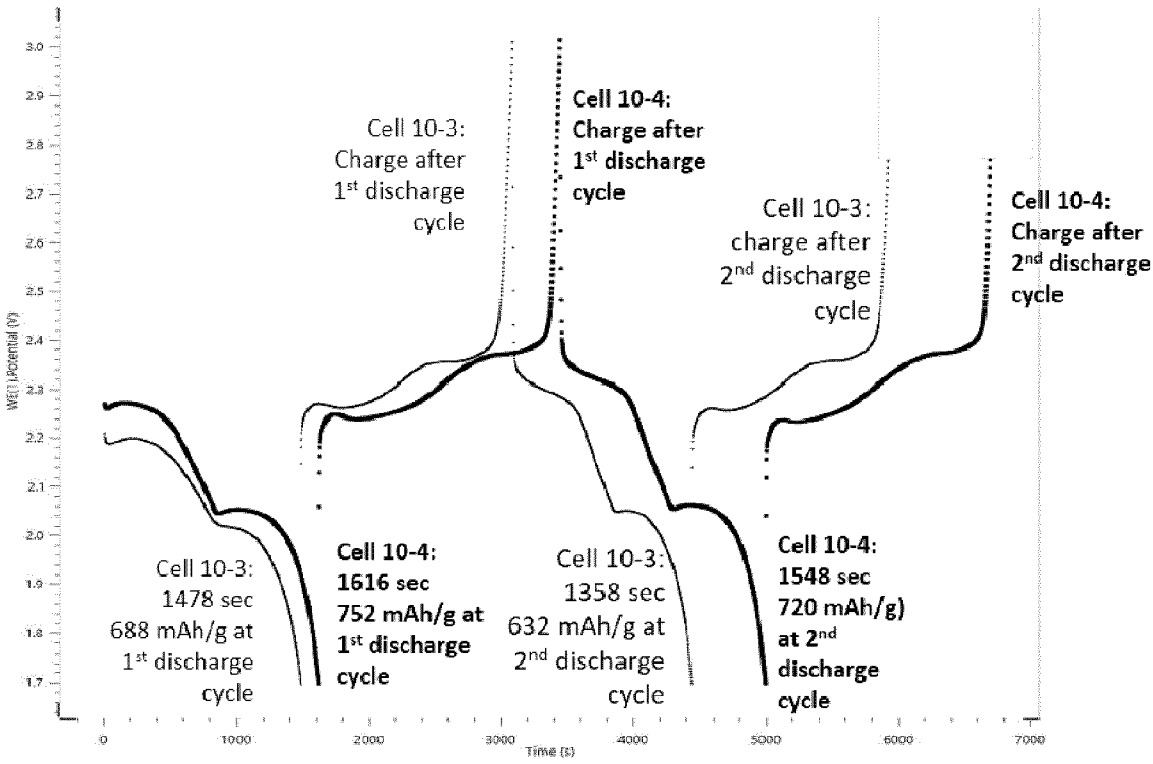


FIG. 13

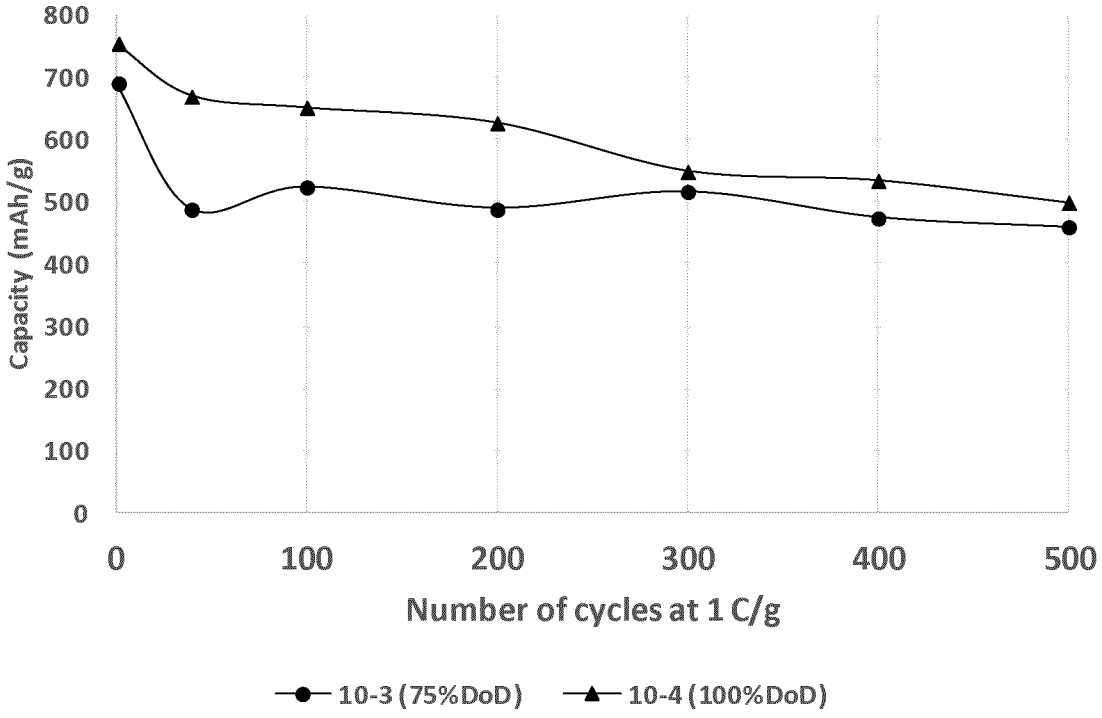


FIG. 14A

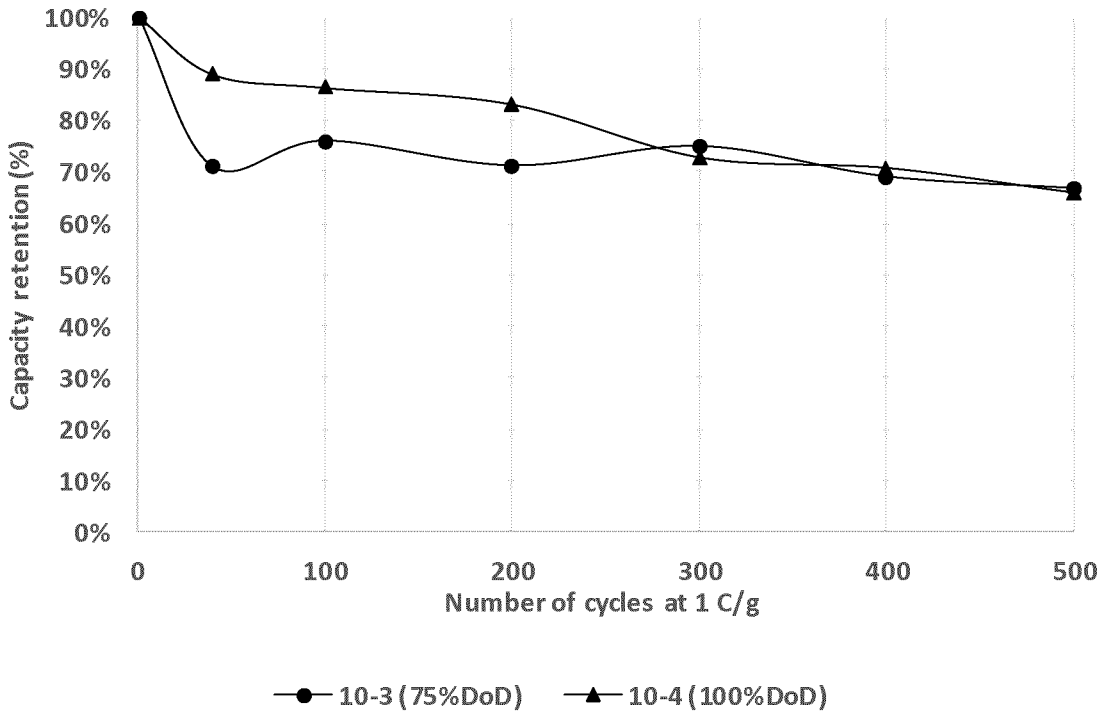


FIG. 14B

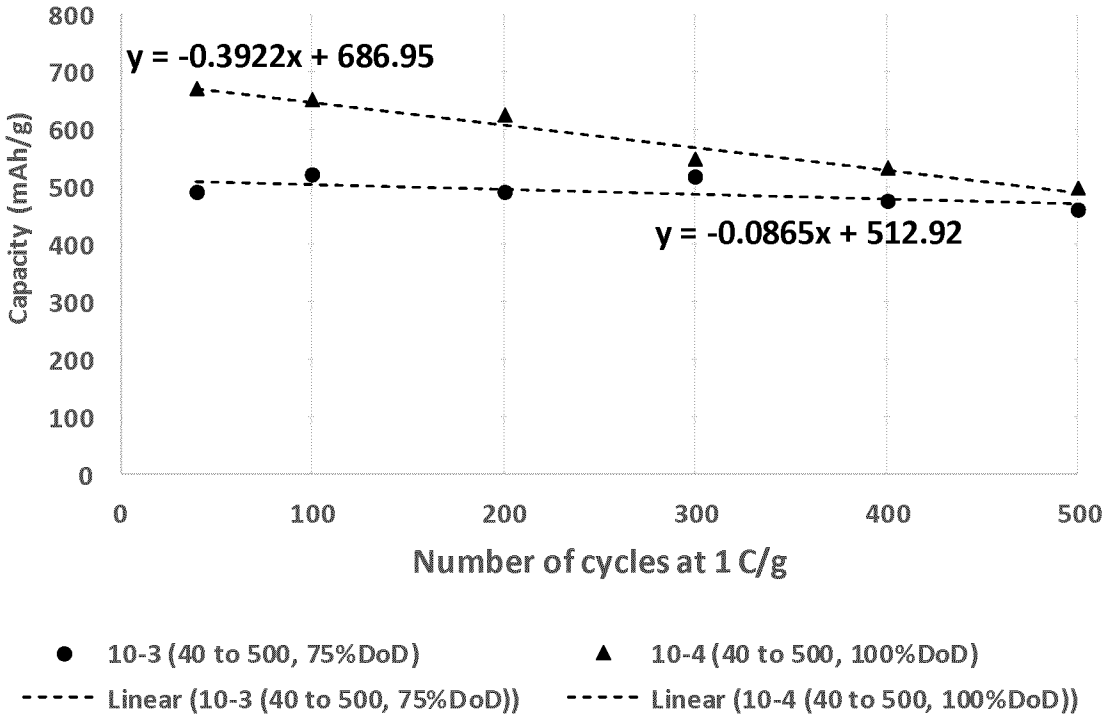


FIG. 14C

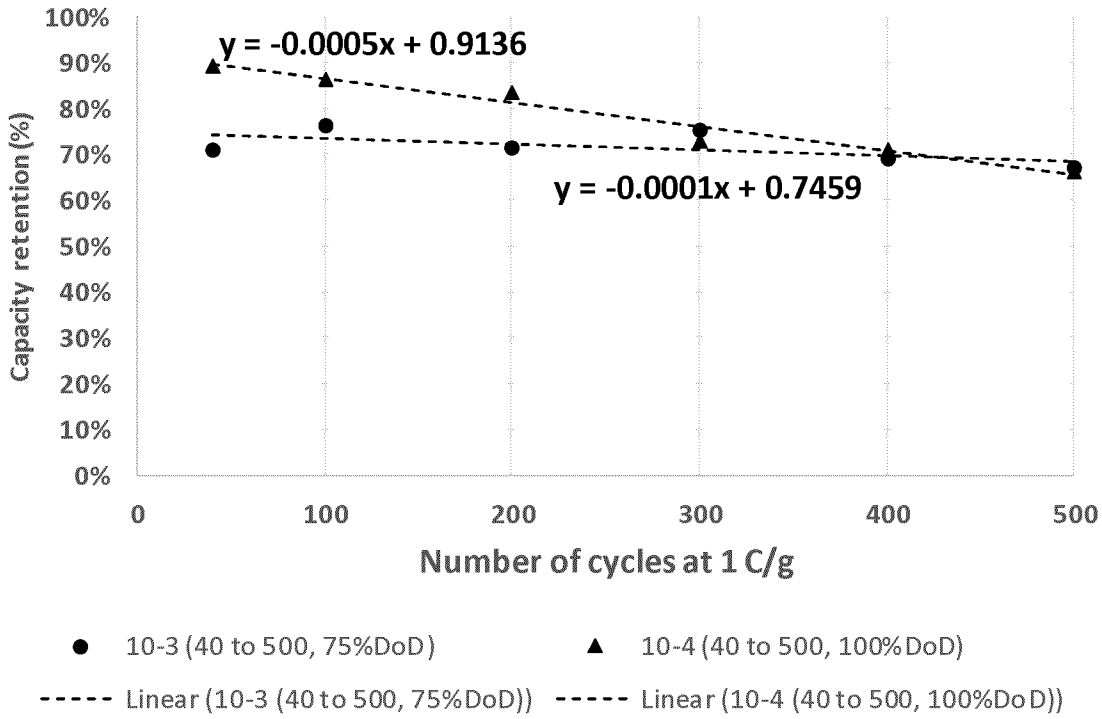


FIG. 14D

INTERNATIONAL SEARCH REPORT

International application No.
PCT/CA2021/050792

A. CLASSIFICATION OF SUBJECT MATTER
IPC: ***H01M 4/136*** (2010.01), ***H01M 10/052*** (2010.01), ***H01M 4/1397*** (2010.01), *C09C 1/44* (2006.01)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC: H01M 4/136 (2010.01), H01M 10/52 (2010.01), H01M 4/1397 (2010.01), C09C 1/44 (2006.01)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic database(s) consulted during the international search (name of database(s) and, where practicable, search terms used)
Databases: Questel-Orbit, Intellect (Canadian Patent Database), Scopus, Google Scholar, Google
Keywords: ammonia, liquid ammonia, anhydrous ammonia, carbon, activated carbon, graphene, sulfur, elemental sulfur, embed, impregnate, adsorb, sulfur impregnated carbon, sulfur embedded carbon, sulfur adsorption, lithium sulfur cathode, lithium sulfur battery, carbon disulfide

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	SUN, L. et al. Sulfur Embedded in a Mesoporous Carbon Nanotube Network as a Binder-Free Electrode for High-Performance Lithium-Sulfur Batteries. ACS Nano. December 22, 2015, Vol. 10, Issue 1, pages 1300-1308. *pg. 1307, "Fabrication of S-PCNT Composites" and "Electrochemical Measurement"*	23-30
X	GUO, J. et al. Sulfur-Impregnated Disordered Carbon Nanotubes Cathode for Lithium-Sulfur Batteries. Nano Letters. September 19, 2011, Vol. 11, Issue 10, pages 4288-4294. *pg. 4291, col. 1, lines 11 and 16, and col. 2, line 5*	23-30
X	US 2012/0088154 A1 (LIU, J. et al.) 12 April 2012 (12-04-2012) *[0017]; claims 1, 11, and 14*	23-30
A	US 4,708,853 (MATVIYA, T.M. et al.) 24 November 1987 (24-11-1987) *col. 3, lines 28-46*	1-22 and 31

☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.

*	Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A"	document defining the general state of the art which is not considered to be of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"D"	document cited by the applicant in the international application	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"E"	earlier application or patent but published on or after the international filing date	"&"	document member of the same patent family
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)		
"O"	document referring to an oral disclosure, use, exhibition or other means		
"P"	document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search 05 August 2021 (05-08-2021)	Date of mailing of the international search report 11 August 2021 (11-08-2021)
Name and mailing address of the ISA/CA Canadian Intellectual Property Office Place du Portage I, C114 - 1st Floor, Box PCT 50 Victoria Street Gatineau, Quebec K1A 0C9 Facsimile No.: 819-953-2476	Authorized officer Karlie Potts

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 8,431,106 B2 (ABRY, R.G.F. et al.) 30 April 2013 (30-04-2013) *col. 5, examples 1-3*	1-31

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/CA2021/050792

Patent Document Cited in Search Report	Publication Date	Patent Family Member(s)	Publication Date
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		CN103155243B	29 July 2015 (29-07-2015)
		EP2625735A2	14 August 2013 (14-08-2013)
		EP2625735A4	23 March 2016 (23-03-2016)
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		WO2008041132A3	03 March 2011 (03-03-2011)