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(54) **SULFIDE ELECTROLYTE LAYER SUPPORTED DRY PROCESS ELECTRODE LAYER**

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

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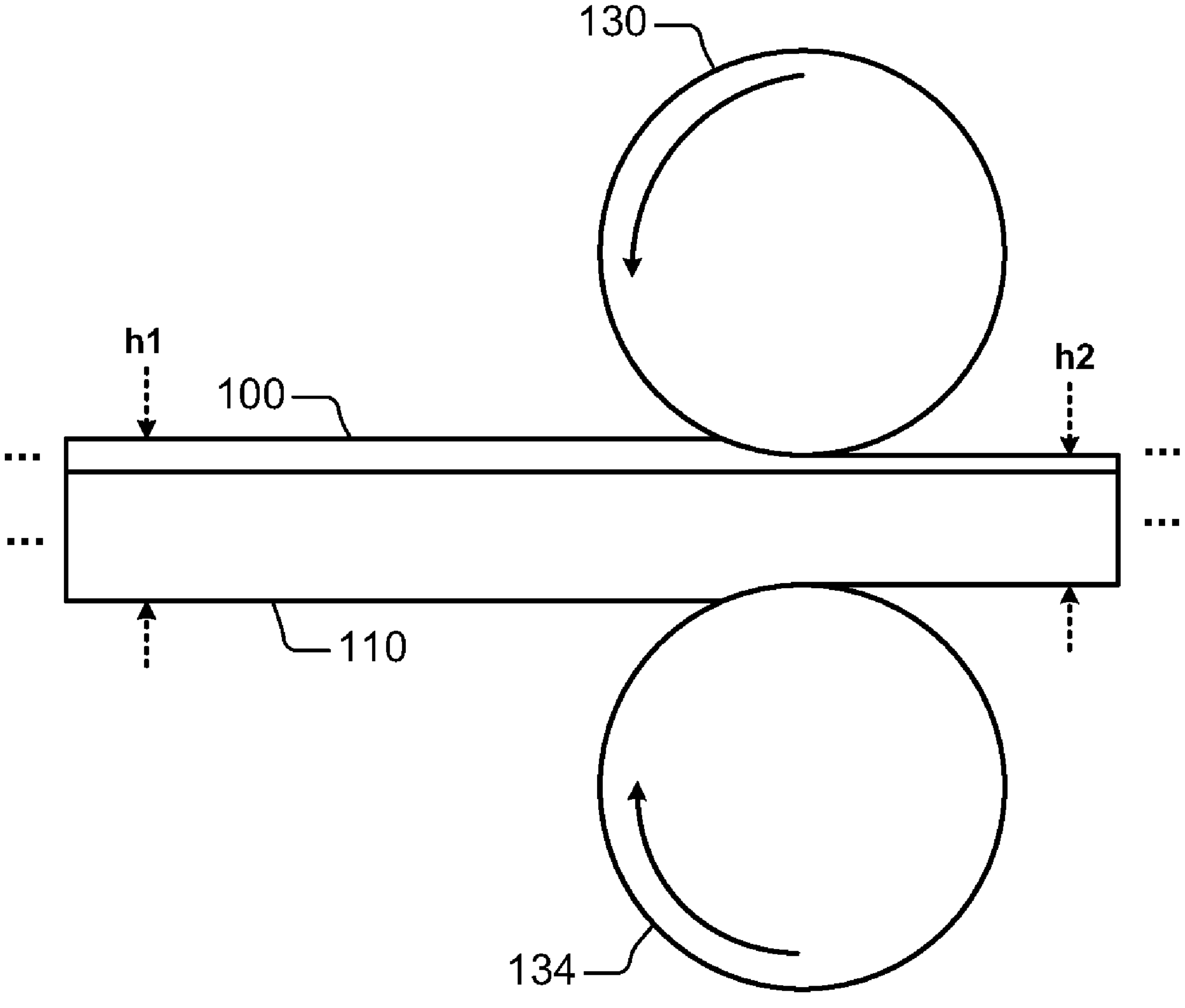
(57) **ABSTRACT**

A method for preparing an electrolyte layer supported by a dry process electrode layer, the method includes providing a sulfide electrolyte layer; providing a first dry process electrode layer; arranging a first side of the sulfide electrolyte layer adjacent to a first side of the first dry process electrode layer; and calendaring the sulfide electrolyte layer and the first dry process electrode layer to reduce a thickness of the sulfide electrolyte layer to a predetermined thickness in a range from approximately 5 micrometers (μm) to approximately 50 μm.



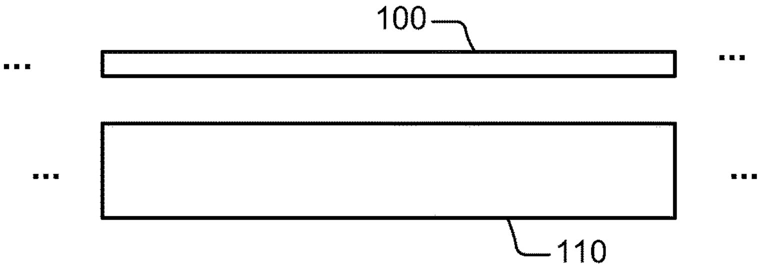


**FIG. 1**

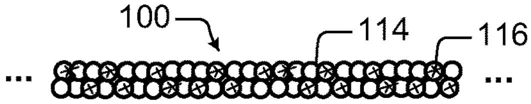


**FIG. 2A**

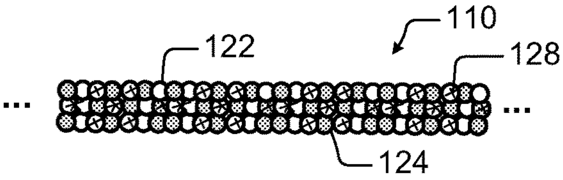
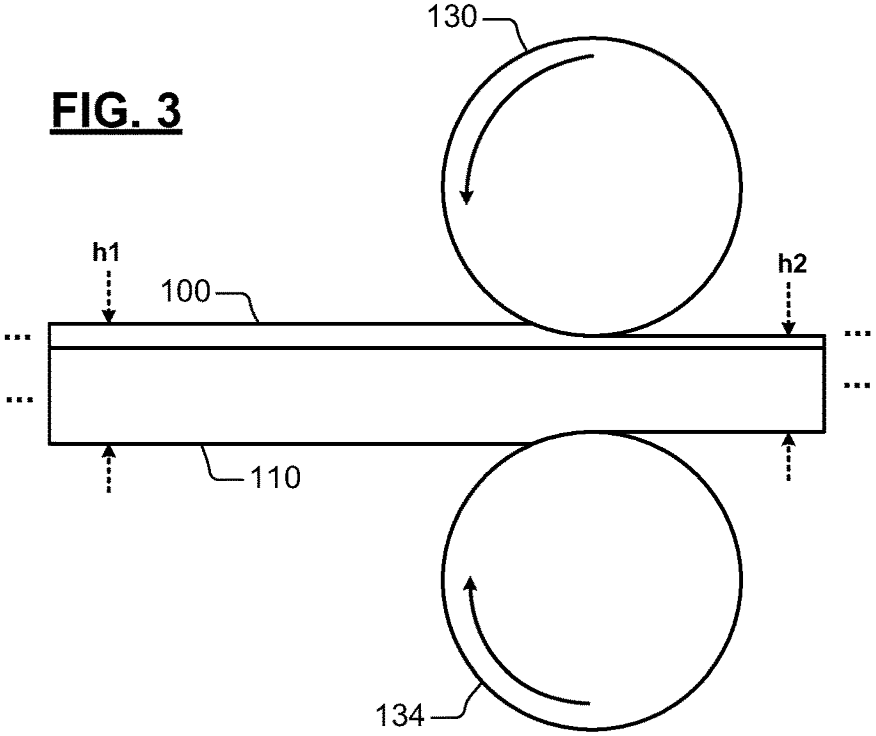


**FIG. 2B**

**FIG. 3**



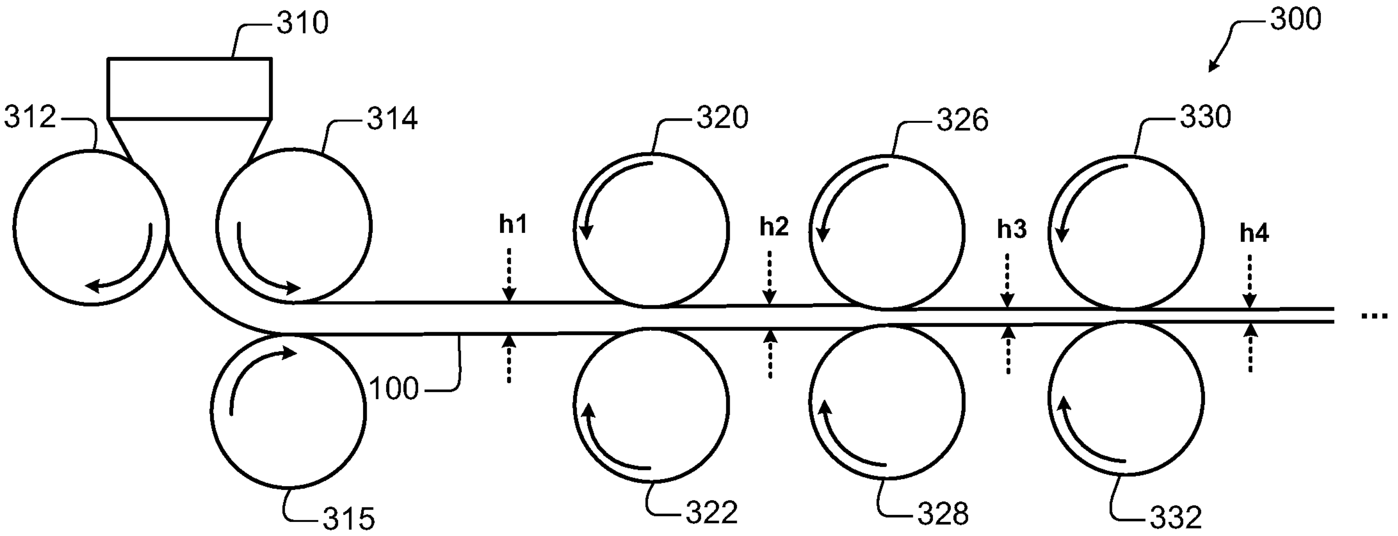

310
300
312
314
320
326
330
h1
h2
h3
h4
...
100
315
322
328
332

FIG. 4

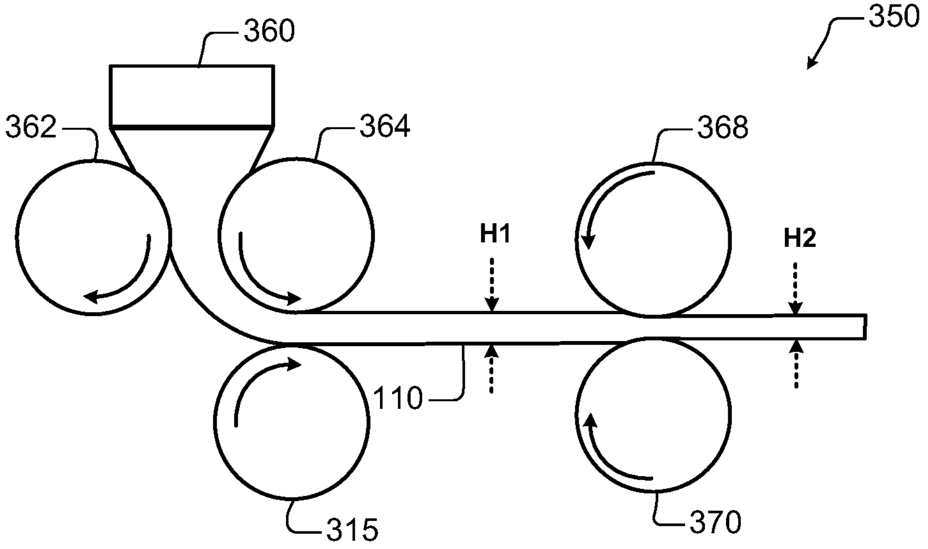

360
350
362
364
368
H1
H2
110
315
370

FIG. 5

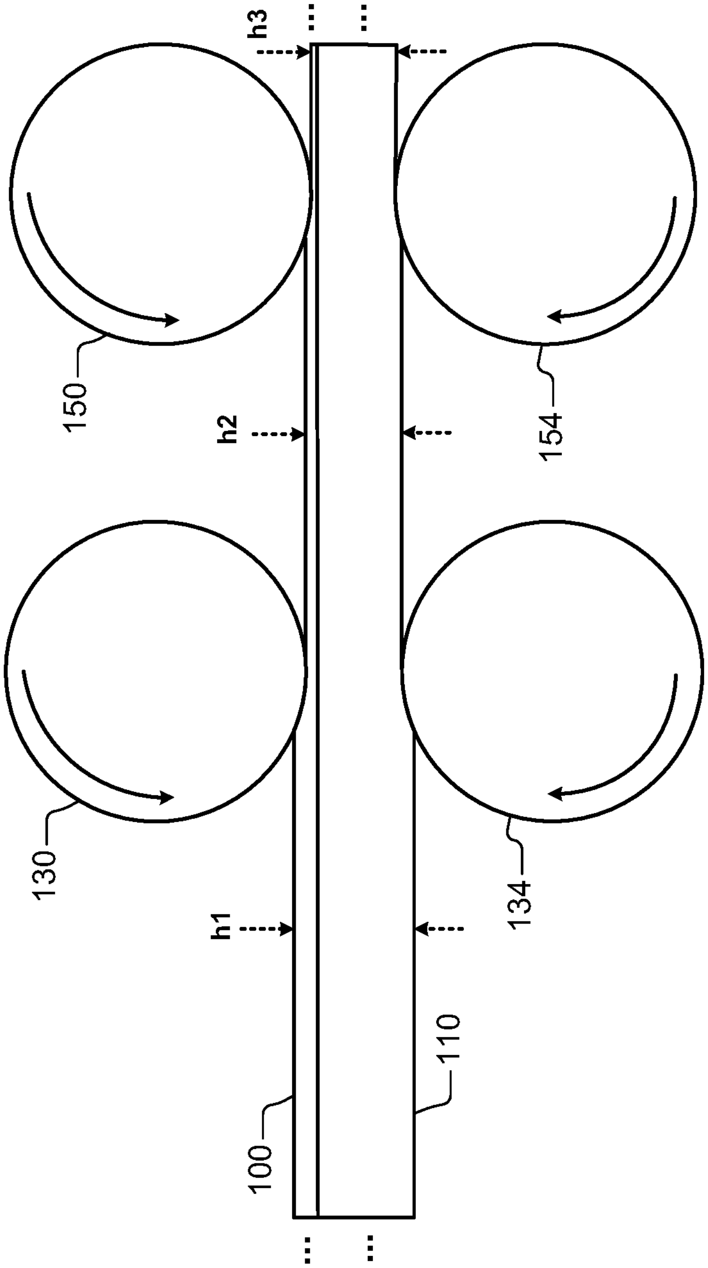


FIG. 6

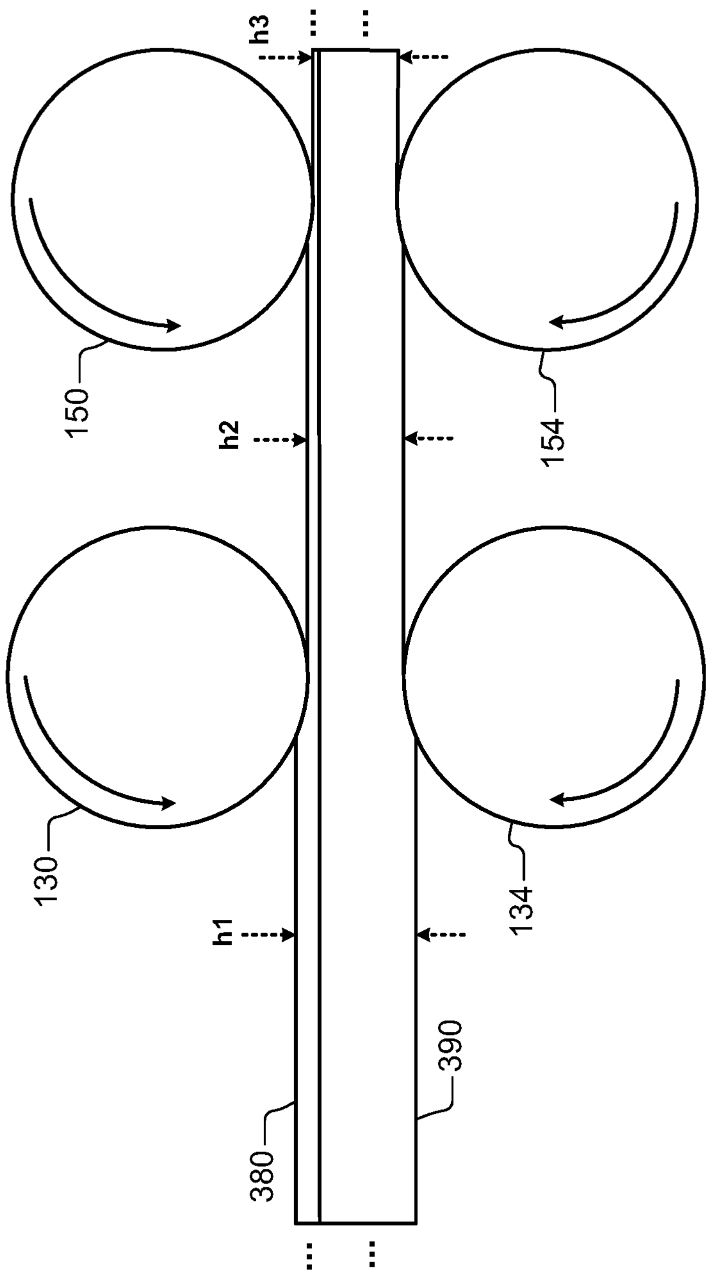


FIG. 7A

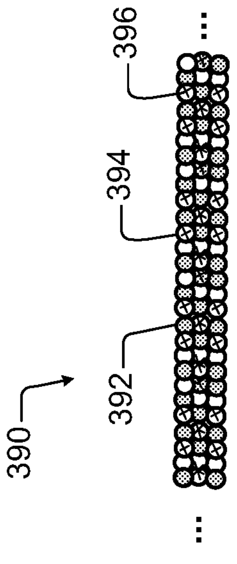


FIG. 7B

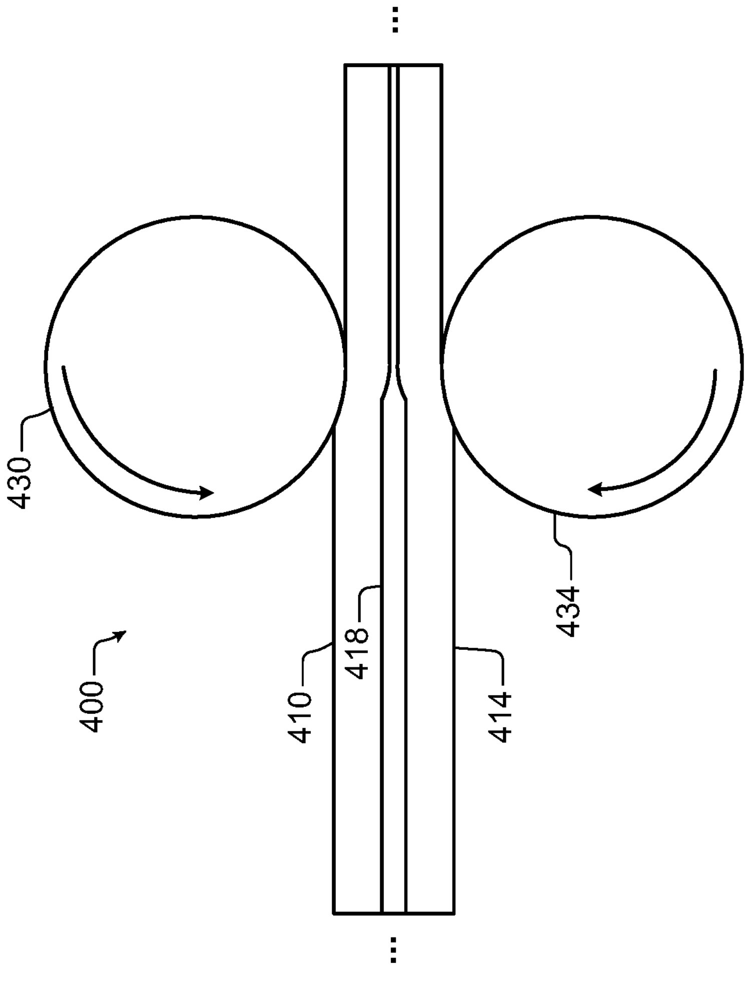


FIG. 8

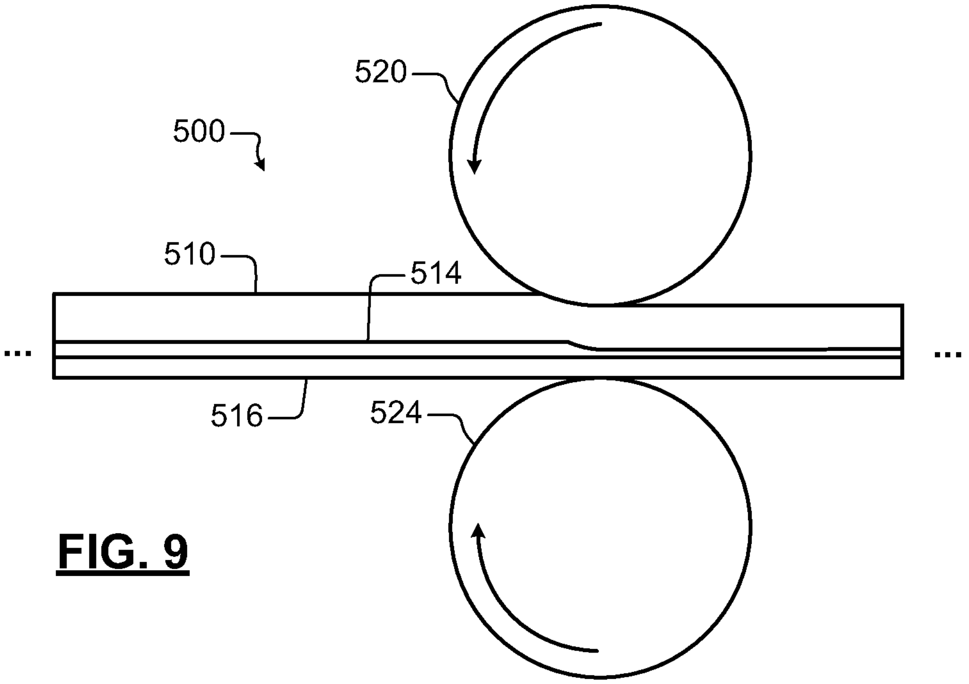


**FIG. 9**

**FIG. 10A**

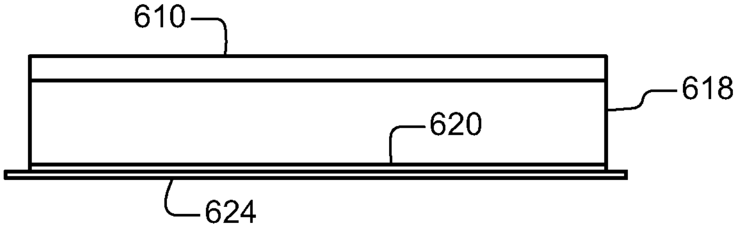
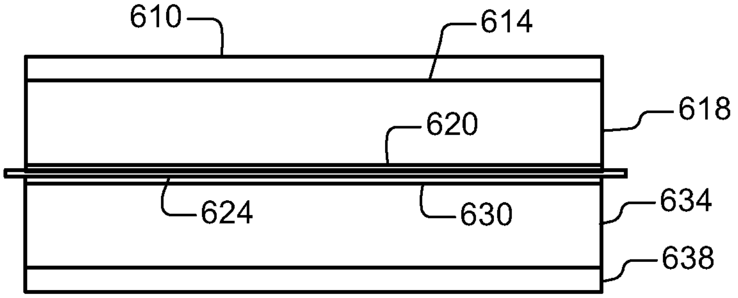


**FIG. 10B**



# SULFIDE ELECTROLYTE LAYER SUPPORTED DRY PROCESS ELECTRODE LAYER

## CROSS-REFERENCE TO RELATED APPLICATIONS

**[0001]** This application claims the benefit of Chinese Patent Application No. 202310179184.1, filed on Feb. 16, 2023. The entire disclosure of the application referenced above is incorporated herein by reference.

## INTRODUCTION

**[0002]** The information provided in this section is for the purpose of generally presenting the context of the disclosure. Work of the presently named inventors, to the extent it is described in this section, as well as aspects of the description that may not otherwise qualify as prior art at the time of filing, are neither expressly nor impliedly admitted as prior art against the present disclosure.

**[0003]** The present disclosure relates to battery cells, and more particularly to a sulfide-based electrolyte layer supported by a dry process electrode layer.

**[0004]** Electric vehicles (EVs) such as battery electric vehicles (BEVs), hybrid vehicles, and/or fuel cell vehicles include one or more electric machines and a battery system including one or more battery cells, modules and/or packs. A power control system is used to control charging and discharging of the battery system during charging and/or driving. Manufacturers of EVs are pursuing increased power density to increase the range of the EVs.

**[0005]** Solid-state batteries (SSBs) with solid electrolyte and SSBs with sulfide electrolyte have the potential to be superior to lithium-ion batteries (LIBs) in terms of abuse tolerance, working temperature range and system design.

## SUMMARY

**[0006]** A method for preparing an electrolyte layer supported by a dry process electrode layer, the method includes providing a sulfide electrolyte layer; providing a first dry process electrode layer; arranging a first side of the sulfide electrolyte layer adjacent to a first side of the first dry process electrode layer; and calendaring the sulfide electrolyte layer and the first dry process electrode layer to reduce a thickness of the sulfide electrolyte layer to a predetermined thickness in a range from approximately 5 micrometers (μm) to approximately 50 μm.

**[0007]** In other features, providing the sulfide electrolyte layer comprises preparing a mixture of a sulfide electrolyte and polytetrafluoroethylene (PTFE) binder to create the sulfide electrolyte layer; and calendaring the mixture one or more times to reduce a thickness of the sulfide electrolyte layer.

**[0008]** In other features, the sulfide electrolyte comprises 90 wt % to 99.9 wt % and the PTFE binder comprises 0.1 wt % to 10 wt % of the sulfide electrolyte layer. Providing the first dry process electrode layer comprises preparing a mixture of a sulfide electrolyte, active material, a conductive additive, and PTFE binder to create the sulfide electrolyte layer; and calendaring the mixture one or more times to reduce a thickness of the first dry process electrode layer.

**[0009]** In other features, the sulfide electrolyte comprises 10 wt % to 30 wt %, the active material comprises 50 wt % to 90 wt %, the conductive additive comprises 0 wt % to 10 wt %, and the PTFE binder comprises greater than 0 wt % and less than or equal to 10 wt % of the dry process electrode layer. The active material comprises cathode active material. The method of claim **6**, wherein the cathode active material is selected from a group consisting of rock salt layered oxide, spinel, a polyanion cathode material, lithium transition-metal oxides, and lithiated metal oxide/sulfide.

**[0010]** In other features, the active material comprises anode active material. The anode active material is selected from a group consisting of carbonaceous material, silicon, silicon and graphite, Li4Ti5O12, a transition-metal, metal oxide/sulfide, Li metal, and Li alloy. The sulfide electrolyte is selected from a group consisting of pseudobinary sulfide, pseudoternary sulfide, pseudoquaternary sulfide, halide-based solid electrolyte, and hydride-based solid electrolyte. The conductive additive is selected from a group consisting of carbon black, graphite, graphene, graphene oxide, Super P, acetylene black, carbon nanofibers, and carbon nanotubes.

**[0011]** In other features, prior to calendaring, the method includes arranging a second dry process electrode layer adjacent to a second side of the sulfide electrolyte layer. The first dry process electrode layer comprises a cathode electrode layer. The second dry process electrode layer comprises an anode electrode layer.

**[0012]** In other features, the method includes attaching the sulfide electrolyte layer and the first dry process electrode layer to a current collector using an electrically conductive adhesive. The electrically conductive adhesive comprises a polymer and an electrically conductive filler. The polymer is selected from a group consisting of epoxy, polyimide, polyester, vinyl ester, polyvinylidene fluoride (PVDF), polyamide, silicone, and acrylic. The conductive filler is selected from a group consisting of Super P, carbon black, graphene, carbon nanotubes, carbon nanofibers, and metal powder.

**[0013]** In other features, a second side of the sulfide electrolyte layer is arranged on a substrate. The substrate comprises polyethylene terephthalate (PET). The method includes attaching one side of a first one of the first dry process electrode layer and the sulfide electrolyte layer to a first side of a current collector using an electrically conductive adhesive. The method includes attaching one side of a second one of the first dry process electrode layer and the sulfide electrolyte layer to a second side of the current collector using an electrically conductive adhesive.

**[0014]** Further areas of applicability of the present disclosure will become apparent from the detailed description, the claims, and the drawings. The detailed description and specific examples are intended for purposes of illustration only and are not intended to limit the scope of the disclosure.

## BRIEF DESCRIPTION OF THE DRAWINGS

**[0015]** The present disclosure will become more fully understood from the detailed description and the accompanying drawings, wherein:

**[0016]** FIG. **1** is a side cross-sectional view of a PTFE-based solid electrolyte layer and a dry process electrode layer according to the present disclosure;

**[0017]** FIG. **2**A is a side cross-sectional view of a PTFE-based sulfide electrolyte layer according to the present disclosure;

**[0018]** FIG. **2**B is a side cross-sectional view of a dry process electrode according to the present disclosure;

**[0019]** FIG. **3** illustrates a method for calendaring the PTFE-based sulfide electrolyte layer and the dry process electrode according to the present disclosure;

**[0020]** FIG. **4** illustrates a method for preparing the PTFE-based sulfide electrolyte layer according to the present disclosure;

**[0021]** FIG. **5** illustrates a method for preparing the dry process cathode electrode according to the present disclosure;

**[0022]** FIG. **6** illustrates a method for calendaring the PTFE-based sulfide electrolyte layer and the dry process cathode electrode using multiple steps according to the present disclosure;

**[0023]** FIG. **7**A illustrates a method for calendaring a solid electrolyte layer supported by a dry process anode electrode layer using multiple steps according to the present disclosure;

**[0024]** FIG. **7**B is a side cross sectional view including an example of a dry process anode electrode layer including solid electrolyte, anode active material, and PTFE binder according to the present disclosure;

**[0025]** FIG. **8** illustrates a method for calendaring the PTFE-based sulfide electrolyte layer arranged between dry process electrodes according to the present disclosure;

**[0026]** FIG. **9** illustrates a method for calendaring the PTFE-based sulfide electrolyte layer and a dry cathode membrane supported on a substrate according to the present disclosure; and

**[0027]** FIGS. **10**A and **10**B are side cross-sectional views of single and double-sided electrodes according to the present disclosure.

**[0028]** In the drawings, reference numbers may be reused to identify similar and/or identical elements.

## DETAILED DESCRIPTION

**[0029]** While the battery cells are described herein in the context of EVs, the battery cells can be used in stationary applications, non-vehicle applications, and in other applications.

**[0030]** Sulfide electrolyte film may be prepared using a dry process sulfide electrolyte powder. The sulfide electrolyte film prepared in this manner typically has a thickness in a range from 500 micrometers (μm) to 1000 μm. While ensuring long-term cycling ability, the thickness of the sulfide electrolyte film decreases energy density and power capability.

**[0031]** When using a wet slurry process for the sulfide electrolyte film, the thickness can be reduced to 65 μm (e.g., using $Li_6PS_5Cl$ powder, polyethylene oxide (PEO) binder, $LiClO_4$ and $SiO_2$). Although the PEO binder helps to form the sulfide electrolyte film, mechanical imperfections or failures occur due its limited bonding effect. Breakage of sulfide electrolyte film may occur when microstructural defects or voids are formed during cold-pressing.

**[0032]** Free-standing sulfide-based electrolyte membranes can be produced by multiple calendaring steps with a decreasing gap. The sulfide electrolyte film has good film forming properties and a high film elongate rate due to the fibrillating polymer. However, further reducing the thickness of electrolyte membrane to less than 50 μm by calendaring may provide poor efficiency since an angle $\alpha$ of the rollers is close to zero as the thickness of the film is reduced. A resulting ultra-thin free-standing membrane may be fragile and be very hard to handle.

**[0033]** In some examples, a thin sulfide electrolyte layer (approximately 5 μm to 50 μm, e.g., approximately 25.8 μm where the term approximately means +/−10%) according to the present disclosure is achieved by a roll-to-roll calendaring of electrode dry film and sulfide electrolyte dry film. Polytetrafluoroethylene (PTFE) binder in an electrode film and a sulfide electrolyte layer may provide good film forming properties and a high film elongation rate due to the fibrillating nature of the PTFE. With the help of the supportive dry electrode layer, the thickness of the sulfide electrolyte layer can be reduced. The thin sulfide electrolyte membrane provides an effective strategy for constructing a high-power solid-state battery.

**[0034]** In some examples, a thin PTFE-based sulfide electrolyte layer (e.g., less than 30 μm) can be achieved by roll-to-roll calendaring with the help of the supportive dry cathode membrane as described herein, which increases energy density of the resulting solid-state battery. The dual layer structure shows good mechanical strength.

**[0035]** Referring now to FIGS. **1**, **2**A, and **2**B, a PTFE-based sulfide electrolyte layer **100** and a dry process cathode electrode layer **110** are shown. In FIG. **2**A, the PTFE-based sulfide electrolyte layer **100** includes a mixture of sulfide electrolyte **114** and PTFE fibrils **116**. In FIG. **2**B, the dry process cathode electrode layer **110** includes a mixture of cathode active material **122**, sulfide electrolyte **128**, and PTFE fibrils **124**.

**[0036]** Referring now to FIG. **3**, a method for calendaring the PTFE-based sulfide electrolyte layer (e.g., **100**) and the dry process electrode layer (e.g., **110**) is shown. The PTFE-based sulfide electrolyte layer **100** is arranged in contact with the dry process electrode **110**, and the laminate (combination of **100** and **110**) is compressed by rollers **130** and **134**. The combination has an initial height h1 that is reduced to a height h**2** via the rollers **130** and **134**.

**[0037]** The PTFE-based sulfide electrolyte layer **100** increases favorable ion transfer between the cathode and anode electrodes. In some examples, the PTFE-based sulfide electrolyte layer **100** has a thickness in a range from approximately 2 μm to approximately 30 μm (e.g., approximately 20 μm). In some examples, the PTFE-based sulfide electrolyte layer **100** includes or consists of sulfide electrolyte and a PTFE binder. In some examples, the sulfide electrolyte includes approximately 90 wt % to 99.9 wt %, and the PTFE binder includes approximately 0.1 wt % to 10 wt %, e.g., <2 wt %). In various implementations, the sum of wt % of the sulfide electrolyte and the PTFE binder equals 100 wt %.

**[0038]** The dry process cathode electrode layer **110** provides mechanical strength for the thin sulfide electrolyte layer to be rolled, handled, and unrolled within an electrode fabrication process. In some examples, the dry process cathode electrode layer **110** has a thickness in a range from approximately 50 μm to approximately 300 μm, (e.g., approximately 200 μm). In some examples, the dry process cathode electrode layer **110** includes or consists of sulfide electrolyte, cathode active material, conductive adhesive, and PTFE binder. In some examples, the sulfide electrolyte includes approximately 10 wt % to 30 wt %, the cathode active material includes approximately 50 wt % to 90 wt %, the conductive additives include approximately 0 wt % to 10 wt %, and the PTFE binder includes approximately greater than 0 wt % and less than or equal to 10 wt % (e.g., greater than 0% and less than 1 wt %). In various implementations, the sum of wt % of the sulfide electrolyte, the cathode active material, the conductive additives, and the PTFE binder equals 100 wt %.

**[0039]** Referring now to FIG. **4**, a multi-step calendaring process with decreasing gap is applied to reduce thickness of the PTFE-based sulfide electrolyte layer **100** gradually. In some examples, a higher calendaring temperature (heating) is used to increase the elongate rate of the combination membrane. In some examples, the calendaring temperature is in a range from 25 degrees Celsius (° C.) to 320° C. In some examples, the calendaring temperature is in a range from 80° C. to 150° C.

**[0040]** In FIG. **5**, a calendaring process may also be used to reduce the thickness of the dry process cathode electrode layer **110** as well. Then, the PTFE-based sulfide electrolyte layer **100** and the dry process cathode electrode layer **110** can be laminated together, and one or more additional calendaring steps may be performed as shown herein.

**[0041]** Referring back to FIG. **4**, a mixture of material for the PTFE-based sulfide electrolyte layer **100** is supplied by a source **310** between rollers **312** and **314**. The PTFE-based sulfide electrolyte layer is calendared using multiple steps using multiple sets of rollers. The PTFE-based sulfide electrolyte layer **100** is compressed successively by rollers **320** and **322**, rollers **326** and **328**, and rollers **330** and **332**. A height of the PTFE-based sulfide electrolyte layer **100** is successively reduced from h**1**, to h**2**, to h**3**, and to h**4**. Examples of h**1**, h**2**, h**3**, and h**4** are approximately 400 μm, approximately 200 μm, approximately 100 μm, and approximately 50 μm, respectively, although the present application is also applicable to one or more other heights.

**[0042]** In FIG. **5**, a mixture of material for the dry process cathode electrode layer **110** is supplied by a source **360** between rollers **362** and **364**. The dry process cathode electrode layer **110** is calendared using multiple steps. The dry process cathode electrode layer **110** is further compressed by one or more pairs of rollers **368** and **370**. A height of the dry process cathode electrode layer **110** is successively reduced from H**1** to H**2**. Examples of H**1** and H**2** are approximately 400 μm and approximately 200 μm, respectively, although the present application is also applicable to one or more other heights.

**[0043]** In FIG. **6**, the PTFE-based sulfide electrolyte layer and the dry process electrode are calendared using multiple steps and multiple sets of rollers. The PTFE-based sulfide electrolyte layer **100** is arranged in contact with the dry process electrode **110**, and the laminate (combination) is compressed by rollers **130** and **134** and then compressed again by rollers **150** and **154**. A height h**1** is reduced to a height h**2** and then to a height h**3**. For example, h**1** is in a range from 200 to 300 μm, h**2** is in a range from 150 to 225 μm, and h**3** is in a range from 100 to 175 μm. For example, h**1**=250 μm, h**2**=175 μm, and h**3**=125 μm.

**[0044]** Referring now to FIGS. **7**A and **7**B, a similar process can be used for a solid electrolyte layer **380** supported by a dry process anode electrode layer **390**. The dry process anode electrode layer **390** includes solid electrolyte **392**, anode active material **394**, and PTFE binder **396**.

**[0045]** The solid electrolyte layer **380** builds up favorable ion transfer between cathode and anode electrodes. In some examples, the solid electrolyte layer **380** has a thickness in a range of approximately 2 μm to approximately 30 μm (e.g., approximately 20 μm). The solid electrolyte layer **380** includes or consists of sulfide electrolyte and PTFE binder. In some examples, the sulfide electrolyte is in a range from 90 wt % to 99.9 wt % and the PTFE binder is in a range from 0.1 wt % to 10 wt % (e.g., less than 2 wt %). In various implementations, the sum of wt % of the sulfide electrolyte and the PTFE binder equals 100 wt %.

**[0046]** The dry process anode electrode layer **390** provides mechanical strength for the thin sulfide electrolyte layer **380** to be rolled, handled, and unrolled within an electrode fabrication process. In some examples, a thickness of the dry process anode electrode layer **390** is in a range from approximately 50 μm to approximately 150 μm (e.g., approximately 100 μm).

**[0047]** The dry process anode electrode layer **390** includes sulfide electrolyte, anode active material, conductive additive, and PTFE binder. In some examples, the sulfide electrolyte includes or consists of 10 wt % to 30 wt %, the anode active material comprises 50 wt % to 90 wt %, the conductive additives 0 wt % to 10 wt %, and the PTFE binder comprises greater than 0 wt % and less than or equal to 10 wt % (e.g., less than 1 wt %). In various implementations, the sum of wt % of the sulfide electrolyte, the anode active material, the conductive additives, and the PTFE binder equals 100 wt %.

**[0048]** Referring now to FIG. **8**, a method for calendaring a PTFE-based sulfide electrolyte layer **418** arranged between a dry process anode electrode layer **414** and a dry process cathode electrode layer **410** is shown. The dry process cathode electrode layer **410**, the PTFE-based sulfide electrolyte layer **418** and the dry process anode electrode layer **414** are compressed by one or more sets of rollers, such as rollers **430** and **434**.

**[0049]** In some examples, the dry process cathode electrode layer **410** has a thickness after the calendaring in a range from 50 μm to 300 μm (e.g., 200 μm). The dry process cathode electrode layer **410** may include or consist of sulfide electrolyte, cathode electrode material, conductive additives, and PTFE binder. In some examples, the sulfide electrolyte includes approximately 10 wt % to 30 wt %, the cathode active material includes approximately 50 wt % to 90 wt %, the conductive additives include approximately 0 wt % to 10 wt %, and the PTFE binder includes greater than 0 wt % and less than or equal to 10 wt % (e.g., greater than 0% and less than 1 wt %). In various implementations, the sum of wt % of the sulfide electrolyte, the cathode active material, the conductive additives, and the PTFE binder equals 100 wt %.

**[0050]** The PTFE-based sulfide electrolyte layer **418** builds up favorable ion transfer between cathode and anode electrodes. In some examples, the PTFE-based sulfide electrolyte layer **418** has a thickness in a range from approximately 2 μm to approximately 30 μm (e.g., approximately 20 μm) after the calendaring. In some examples, the PTFE-based sulfide electrolyte layer **418** includes or consists of sulfide electrolyte and PTFE binder. In an example, the sulfide electrolyte may include 90 wt % to 99.9 wt % and the PTFE binders may include 0.1 wt % to 10 wt % (e.g., 2 wt %). In various implementations, the sum of wt % of the sulfide electrolyte and the PTFE binder equals 100 wt %.

**[0051]** In some examples, the dry process anode electrode layer **414** has a thickness after the calendaring in a range from approximately 50 μm to approximately 150 μm (e.g., approximately 100 μm). The dry process anode electrode layer **414** may include a sulfide electrolyte, an anode active material, conductive additives, and PTFE binder. In some examples, the sulfide electrolyte includes or consists of 0 wt % to 30 wt %, the anode active material comprises 50 wt % to 90 wt %, the conductive additives 0 wt % to 10 wt %, and the PTFE binder comprises greater than 0 wt % and less than or equal to 10 wt % (e.g., less than 1 wt %). In various implementations, the sum of wt % of the sulfide electrolyte, the anode active material, the conductive additives, and the PTFE binder equals 100 wt %.

**[0052]** Referring now to FIG. **9**, a method **500** for calendaring a PTFE-based sulfide electrolyte layer **514** supported by a dry process electrode **510** and a substrate **516** is shown. The PTFE-based sulfide electrolyte layer **514**, the dry process electrode **510**, and the substrate **516** are compressed by one or more sets of rollers, such as rollers **520** and **524**. The dry process anode electrode layer **414** includes anode active material **420**, sulfide electrolyte **422**, and PTFE fibrils **424**.

**[0053]** The substrate **516** protects and supports the thin sulfide membrane and may have a thickness in a range from approximately 20 μm to approximately 100 μm (e.g., approximately 50 μm), although the present application is also applicable to other thicknesses. In some examples, the substrate comprises polyethylene terephthalate (PET) film.

**[0054]** Referring now to FIGS. **10**A and **10**B, single and double-sided electrodes are shown. In FIG. **10**A, a sulfide electrolyte layer **610** and a dry cathode layer **618** are attached to one side of a current collector **624**, such as using conductive adhesive **620**. In FIG. **10**B, a sulfide electrolyte layer **638** and a dry cathode layer **634** are attached to an opposite side of the current collector **624**, such as using conductive adhesive **630**.

**[0055]** In some examples, the electrically conductive adhesive layer includes polymer and conductive filler. In some examples, the conductive filler includes a carbon material Super P, carbon black, graphene, carbon nanotubes, carbon nanofibers. In some examples, the conductive filler includes metal powder such as Ag, Ni, or Al. In some examples, the polymer is configured to resist solvent and provide good adhesion. In some examples, the polymer includes epoxy, polyimide (polyamic acid), polyester, vinyl ester, thermoplastic polymers (less solvent resistant) include PVDF, polyamide, silicone, and acrylic. In some examples, a mass ratio of filler/polymer is in a range from approximately 0.1% to approximately 50%, (e.g., SP/PAA=1/3; SWCNT/PVDF=0.2%). In some examples, a thickness of the conductive adhesive is in a range from approximately 0.5 μm to approximately 20 μm.

**[0056]** In some examples, the solid electrolyte is selected from a group including or consisting of pseudobinary sulfide, pseudoternary sulfide, pseudoquaternary sulfide, halide-based solid electrolyte, and hydride-based solid electrolyte. Examples of pseudobinary sulfide include $Li_2S—P_2S_5$ system ($Li_3PS_4$, $Li_7P_3S_{11}$ and $Li_{9.6}P_3S_{12}$), $Li_2S—SnS_2$ system ($Li_4SnS_4$), $Li_2S—SiS_2$ system, $Li_2S—GeS_2$ system, $Li_2S—B_2S_3$ system, $Li_2S—Ga_2S_3$ system, $Li_2S—P_2S_3$ system, $Li_2S—Al_2S_3$ system. Examples of pseudoternary sulfide include $Li_2O—Li_2S—P_2S_5$ system, $Li_2S—P_2S_5—P_2O_5$ system, $Li_2S—P_2S_5—GeS_2$ system ($Li_{3.25}Ge_{0.25}P_{0.75}S_4$ and $Li_{10}GeP_2S_{12}$), $Li_2S—P_2S_5—LiX$ (X═F, Cl, Br, I) system ($Li_6PS_5Br$, $Li_6PS_5Cl$, $L_7P_2S_8I$ and $Li_4PS_4I$), $Li_2S—As_2S_5—SnS_2$ system ($Li_{3.833}Sn_{0.833}As_{0.166}S_4$), $Li_2S—P_2S_5—Al_2S_3$ system, $Li_2S—LiX—SiS_2$ (X═F, Cl, Br, I) system, $0.4LiI \cdot 0.6Li_4SnS_4$ and $Li_{11}Si_2PS_{12}$.

**[0057]** Examples of pseudoquaternary sulfide include $Li_2O—Li_2S—P_2S_5—P_2O_5$ system, $Li_{9.54}Si_{1.74}P_{1.44}S_{11.7}Cl_{0.3}$, $Li_7P_{2.9}Mn_{0.1}S_{10.7}I_{0.3}$ and $Li_{10.35}[Sn_{0.27}Si_{1.08}]P_{1.65}S_{12}$. Examples of halide-based solid electrolyte include $LisYCl_6$, $LisInCl_6$, $LisYBr_6$, LiI, $Li_2CdCl_4$, $Li_2MgCl_4$, $Li_2CdI_4$, $Li_2ZnI_4$, LisOCl. Examples of hydride-based solid electrolyte include $LiBH_4$, $LiBH_4—LiX$ (X═Cl, Br, or I), $LiNH_2$, $Li_2NH$, $LiBH_4$-$LINH_2$, $Li_3AlH_6$. In other examples, other solid electrolyte that possesses low grain-boundary resistance.

**[0058]** In some examples, the cathode active material is selected from a group including or consisting of rock salt layered oxide, spinel, polyanion cathode materials, lithium transition-metal oxides, and lithiated metal oxide/sulfide. Examples of rock salt layered oxides include $LiCoO_2$, $LiNi_xMn_yCo_{1-x-y}O_2$, $LiNi_xMn_yAl_{1-x-y}O_2$, $LiNi_x$Mn1-xO2, $Li_{1+x}MO_2$. Examples of spinel include $LiMn_2O_4$ and $LiNi_{0.5}Mn_{1.5}O_4$. Examples of polyanion cathode materials include $LiV_2(PO_4)_3$. Surface-coated and/or doped cathode materials mentioned above can also be used such as $LiNbO_3$-coated $LiMn_2O_4$, $Li_2ZrO_3$ or $Li_3PO_4$-coated $LiNi_xMn_yCo_{1-x-y}O_2$, and Al-doped $LiMn_2O_4$. Examples of lithiated metal oxide/sulfide include $LiTiS_2$), lithium sulfide, and sulfur.

**[0059]** In some examples, the anode active material is selected from a group including or consisting of carbonaceous material (e.g., graphite, hard carbon, soft carbon etc.), silicon, silicon mixed with graphite, $Li_4Ti_5O_{12}$, transition-metals (e.g., Sn), metal oxide/sulfide (e.g., $TiO_2$, FeS, etc.), other lithium-accepting anode materials, Li metal and Li alloy.

**[0060]** In some examples, the conductive additive is selected from a group including or consisting of carbon black, graphite, graphene, graphene oxide, Super P, acetylene black, carbon nanofibers, carbon nanotubes and other electronically conductive additives.

**[0061]** In some examples, the PTFE binder has a particle size in a range from approximately 300 um to approximately 800 um. In other examples, other fibrillating polymer binders are used (e.g., fluorinated ethylene propylene (FEP), perfluoroalkoxy (PFA), ethylene tetrafluoroethylene (ETFE), or a combination thereof).

**[0062]** In other examples, a binder polymer with film forming properties and high film elongation is used. In some examples, the binder polymer is selected from a group including or consisting of polyvinylidene fluoride-hexafluoropropylene (polyvinylidene fluoride-co-hexafluoropropylene), polyvinylidene fluoride-trichlorethylene (polyvinylidene fluoride-cotrichlorethylene), polymethylmethacrylate, Polyacrylonitrile, polyvinylpyrrolidone, polyvinylacetate, ethylene-co-vinyl acetate, polyethylene oxide, cellulose acetate, cellulose acetate butyrate, cellulose acetate propionate, cyanoethylpullulan, cyanoethylpolyvinylalcohol, cyanoethylcellulose, cyano Noethyl sucrose (cyanoethylsucrose), pullulan (pullulan), carboxyl methyl cellulose, acrylonitrile-styrene-butadiene copolymer, polyimide or mixtures thereof.

**[0063]** The foregoing description is merely illustrative in nature and is in no way intended to limit the disclosure, its application, or uses. The broad teachings of the disclosure can be implemented in a variety of forms. Therefore, while this disclosure includes particular examples, the true scope of the disclosure should not be so limited since other modifications will become apparent upon a study of the drawings, the specification, and the following claims. It should be understood that one or more steps within a method may be executed in different order (or concurrently) without altering the principles of the present disclosure. Further, although each of the embodiments is described above as having certain features, any one or more of those features described with respect to any embodiment of the disclosure can be implemented in and/or combined with features of any of the other embodiments, even if that combination is not explicitly described. In other words, the described embodiments are not mutually exclusive, and permutations of one or more embodiments with one another remain within the scope of this disclosure. Approximately as used herein may mean +/−10% of stated values.

**[0064]** Spatial and functional relationships between elements (for example, between modules, circuit elements, semiconductor layers, etc.) are described using various terms, including "connected," "engaged," "coupled," "adjacent," "next to," "on top of," "above," "below," and "disposed." Unless explicitly described as being "direct," when a relationship between first and second elements is described in the above disclosure, that relationship can be a direct relationship where no other intervening elements are present between the first and second elements, but can also be an indirect relationship where one or more intervening elements are present (either spatially or functionally) between the first and second elements. As used herein, the phrase at least one of A, B, and C should be construed to mean a logical (A OR B OR C), using a non-exclusive logical OR, and should not be construed to mean "at least one of A, at least one of B, and at least one of C."

**[0065]** In the figures, the direction of an arrow, as indicated by the arrowhead, generally demonstrates the flow of information (such as data or instructions) that is of interest to the illustration. For example, when element A and element B exchange a variety of information but information transmitted from element A to element B is relevant to the illustration, the arrow may point from element A to element B. This unidirectional arrow does not imply that no other information is transmitted from element B to element A. Further, for information sent from element A to element B, element B may send requests for, or receipt acknowledgements of, the information to element A.

What is claimed is:

**1**. A method for preparing an electrolyte layer supported by a dry process electrode layer, the method comprising:

providing a sulfide electrolyte layer;

providing a first dry process electrode layer;

arranging a first side of the sulfide electrolyte layer adjacent to a first side of the first dry process electrode layer; and

calendaring the sulfide electrolyte layer and the first dry process electrode layer to reduce a thickness of the sulfide electrolyte layer to a predetermined thickness in a range from approximately 5 micrometers (μm) to approximately 50 μm.

**2**. The method of claim **1**, wherein providing the sulfide electrolyte layer comprises:

preparing a mixture of a sulfide electrolyte and polytetrafluoroethylene (PTFE) binder to create the sulfide electrolyte layer; and

calendaring the mixture one or more times to reduce a thickness of the sulfide electrolyte layer.

**3**. The method of claim **2**, wherein the sulfide electrolyte comprises 90 wt % to 99.9 wt % and the PTFE binder comprises 0.1 wt % to 10 wt % of the sulfide electrolyte layer.

**4**. The method of claim **1**, wherein providing the first dry process electrode layer comprises:

preparing a mixture of a sulfide electrolyte, active material, a conductive additive, and PTFE binder to create the sulfide electrolyte layer; and

calendaring the mixture one or more times to reduce a thickness of the first dry process electrode layer.

**5**. The method of claim **4**, wherein the sulfide electrolyte comprises 10 wt % to 30 wt %, the active material comprises 50 wt % to 90 wt %, the conductive additive comprises 0 wt % to 10 wt %, and the PTFE binder comprises greater than 0 wt % and less than or equal to 10 wt % of the dry process electrode layer.

**6**. The method of claim **5**, wherein the active material comprises cathode active material.

**7**. The method of claim **6**, wherein the cathode active material is selected from a group consisting of rock salt layered oxide, spinel, a polyanion cathode material, lithium transition-metal oxides, and lithiated metal oxide/sulfide.

**8**. The method of claim **5**, wherein the active material comprises anode active material.

**9**. The method of claim **8**, wherein the anode active material is selected from a group consisting of carbonaceous material, silicon, silicon and graphite, $Li_4Ti_5O_{12}$, a transition-metal, metal oxide/sulfide, Li metal, and Li alloy.

**10**. The method of claim **5**, wherein the sulfide electrolyte is selected from a group consisting of pseudobinary sulfide, pseudoternary sulfide, pseudoquaternary sulfide, halide-based solid electrolyte, and hydride-based solid electrolyte.

**11**. The method of claim **5**, wherein the conductive additive is selected from a group consisting of carbon black, graphite, graphene, graphene oxide, Super P, acetylene black, carbon nanofibers, and carbon nanotubes.

**12**. The method of claim **1**, further comprising:

prior to calendaring, arranging a second dry process electrode layer adjacent to a second side of the sulfide electrolyte layer,

wherein the first dry process electrode layer comprises a cathode electrode layer, and

wherein the second dry process electrode layer comprises an anode electrode layer.

**13**. The method of claim **1**, further comprising:

attaching the sulfide electrolyte layer and the first dry process electrode layer to a current collector using an electrically conductive adhesive.

**14**. The method of claim **13**, wherein the electrically conductive adhesive comprises a polymer and an electrically conductive filler.

**15**. The method of claim **14**, wherein the polymer is selected from a group consisting of epoxy, polyimide, polyester, vinyl ester, polyvinylidene fluoride (PVDF), polyamide, silicone, and acrylic.

**16**. The method of claim **14**, wherein the conductive filler is selected from a group consisting of Super P, carbon black, graphene, carbon nanotubes, carbon nanofibers, and metal powder.

**17**. The method of claim **1**, wherein a second side of the sulfide electrolyte layer is arranged on a substrate.

**18**. The method of claim **17**, wherein the substrate comprises polyethylene terephthalate (PET).

**19**. The method of claim **1**, further comprising:

attaching one side of a first one of the first dry process electrode layer and the sulfide electrolyte layer to a first side of a current collector using an electrically conductive adhesive.

**20**. The method of claim **19**, further comprising:

attaching one side of a second one of the first dry process electrode layer and the sulfide electrolyte layer to a second side of the current collector using an electrically conductive adhesive.

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