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### **HARDWARE AND MANUFACTURING METHODS TO COAT ELECTRODE MATERIALS TO SUBSTRATES IN SECONDARY IONIC BATTERY MANUFACTURING PROCESS**

#### **Abstract**

A counter-rotating roller system for coating electrode materials includes a roll-to-roll apparatus including a dispensing roller that selectively dispenses a conductive substrate from a substrate roll along a first direction towards a receiving roller. A counter-rotating roller module including a counter-rotating roller is stationarily positioned at a predetermined height over the conductive substrate. The direction of the tangential velocity of the counter-rotating roller at an interface between the counter-rotating roller and the conductive substrate is 180 degrees opposite the first direction of travel of the conductive substrate. The system further includes a continuous powder application module positioned on a feed side of the counter-rotating roller to continuously deposit a volume of a dry powder mixture on the conductive substrate prior to contact with the counter-rotating roller. A contact roller scraper contacting the counter-rotating roller removes powder particles from an outer surface of the counter-rotating roller.

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## Background/Summary

CROSS REFERENCE TO RELATED APPLICATIONS [0001] The present application claims priority as a continuation of U.S. patent application Ser. No. 18/623,880 (Attorney Docket No. PB23002-US), entitled “HARDWARE AND MANUFACTURING METHODS TO COAT ELECTRODE MATERIALS TO SUBSTRATES IN SECONDARY IONIC BATTERY MANUFACTURING PROCESS” and filed on Apr. 1, 2024, which claims priority to U.S. Provisional Patent Application No. 63/456,465 entitled HARDWARE AND MANUFACTURING METHODS TO COAT ELECTRODE MATERIALS TO SUBSTRATES IN SECONDARY IONIC BATTERY MANUFACTURING PROCESS filed Apr. 1, 2023, which is incorporated herein by reference in its entirety.

### BACKGROUND

[0002] Innovation of portable electronic devices has drawn attention towards efficient energy storage as transportable systems and portable devices such as smartphones, laptops, and smart health devices, have reduced in size while their energy needs have increased. Electrochemical storage and conversion devices are expanding the capabilities of these systems in a variety of fields including portable electronic devices, air and space craft technologies, passenger and cargo vehicles, and biomedical instrumentation. Electrochemical storage and conversion devices have designs and performance attributes specifically engineered to provide compatibility with a diverse range of application requirements and operating environments.

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## Description

### BRIEF DESCRIPTION OF THE DRAWINGS

[0003] The present disclosure may be better understood, and its numerous features and advantages made apparent to those skilled in the art by referencing the accompanying drawings. The use of the same reference symbols in different drawings indicates similar or identical items.

[0004] FIG. 1 is a perspective view of a counter-rotating roller system in accordance with some embodiments.

[0005] FIG. 2 is a top-down view of a counter-rotating roller system in accordance with some embodiments.

[0006] FIG. 3 is a side view of a counter-rotating roller system in accordance with some embodiments.

[0007] FIG. 4 is a side cross section view of a counter-rotating roller module in accordance with some embodiments.

[0008] FIG. 5 is a perspective and exploded view of a continuous powder application module in accordance with some embodiments.

[0009] FIG. 6 is a block diagram of a method for coating electrode materials to conductive substrates in accordance with some embodiments.

#### DETAILED DESCRIPTION

[0010] Significant pressures exist to expand the functionality of energy storage and conversion devices, such as batteries, fuel cells, and electrochemical capacitors, in an ever-widening range of applications. Continued development has created a need for mechanically robust, high reliability, and high energy density electrochemical storage and conversion devices capable of good performance in a useful range of operating environments. Many recent advances in electrochemical storage technology are attributable to fabrication and integration of new materials for device components. Battery technology, for example, continues to rapidly develop, at least in part, due to the development of electrode and electrolyte materials for these systems.

[0011] A secondary ionic battery has multiple components of which the cathode and the anode, collectively referred to as electrodes, are the most important and expensive parts of the battery to manufacture. Conventionally, many lithium-ion battery electrodes are manufactured using a slurry coating process. In the slurry coating process, the electrode precursor materials are dispersed in a solvent to create an electrode slurry. The main component of the solids mixture in an electrode slurry is the active electrode material in powder form. The mixture may also include lesser amounts of electrically conductive additive material and polymeric binder, both also in powder form, with the active electrode material making up the majority fraction of the solids mixture.

[0012] The electrode slurry is coated onto metal current collectors before undergoing a long, slow, and energy intensive drying process to remove the solvent and create a film on the metal current collector. The solvent removal/drying process is critical as coating- or drying-related defects in the anode or cathode are known to occur at this stage. This powder-to-slurry-to-film production method yields a dry electrode film attached or bonded to a metal current collector. The final state of the electrode is a dried film or layer of electrode materials on a metal current collector. The solvent in the powder-to-slurry-to-film production method serves as a working medium for: (i) proper mixing, homogenous distribution, and easy spreading of the powder ingredients in slurry form; and (ii) binding of the solid mixture ingredients to the electrode current collector substrate. The complete removal of the solvent is required before an electrode can be used.

[0013] From an environmental standpoint, the slurry process is detrimental as it often involves harmful solvents, such as N-methyl-pyrrolidone (NMP). In processes involving NMP, a solvent recycling facility is further required in addition to drying equipment to prevent NMP from being released into the environment. This requirement to recover the solvent and prevent its release results in additional capital and operating costs when using a slurry electrode fabrication process involving NMP or other similar solvents. Accordingly, there is a need for electrode fabrication processes that avoids slurries and does not involve solvents. A solventless current collector coating process would be more energy- and cost-efficient by eliminating the energy intensive drying step needed to completely remove all the mediator solvent species.

[0014] Solventless electrode coating fabrication processes can promote a greener supply chain for energy storage devices such as lithium-ion batteries. Drying equipment occupies a large footprint; however, drying equipment would not be required in a solventless electrode fabrication process. Consequently, a solventless process requires a much smaller industrial footprint. This means not only is the solventless process efficient in terms of operating cost but would also be easier to deploy at the industrial scale for new battery manufacturers or existing manufacturers seeking to increase production.

[0015] To address these problems and improve electrode manufacturing, FIGS. 1-6 describe hardware and methods of coating electrode materials to substrates in a solventless manner by decoupling the electrode material mixing process from the binding processes that bind the solid

mixture ingredients to the electrode current collector substrate. In some embodiments, a counter-rotating roller system for coating electrode materials includes a roll-to-roll apparatus including a dispensing roller that selectively dispenses a conductive substrate from a substrate roll. The conductive substrate is directed along a first direction towards a receiving roller. Further, a counter-rotating roller module including a counter-rotating roller is stationarily positioned at a predetermined height over the conductive substrate. The direction of the tangential velocity of the counter-rotating roller at an interface between the counter-rotating roller and the conductive substrate is 180 degrees opposite the first direction of travel of the conductive substrate. The counter-rotating roller system further includes a continuous powder application module positioned on a feed side of the counter-rotating roller module to continuously deposit a volume of a dry powder mixture on the conductive substrate prior to contact with the counter-rotating roller. A contact roller scraper contacting the counter-rotating roller is configured to remove residual powder particles from an outer surface of the counter-rotating roller.

[0016] With reference to FIGS. 1-5, a counter-rotating roller system according to a first embodiment is shown therein and designated **100**. The counter-rotating roller system **100** includes a roll-to-roll apparatus **102** including a dispensing roller **104** that selectively dispenses a conductive substrate **106** from a substrate roll **108**. The conductive substrate **106** is directed along a first direction (as indicated by the left-to-right arrows of FIGS. 1-4) of travel towards a receiving roller (not shown). In various embodiments, a counter-rotating roller module **110** includes a counter-rotating roller **112** stationarily positioned at a predetermined height **114** over the conductive substrate **106**. The conductive substrate **106**, in various embodiments, is a conductive electrode substrate including graphene-coated substrates, graphite-coated substrates, carbon-coated substrates, metal foils (including copper and aluminum), conductive polymers, conductive polymer coated metal foils, carbon fiber mats, nickel foam, Polyethylene Terephthalate (PET) coated with conductive layers, stainless steel mesh, titanium foils, and the like. The counter-rotating roller **112** does not directly contact the conductive substrate **106**. According to some embodiments, the height **114** between the conductive substrate **106** and the counter-rotating roller **112** is set at a fixed height in the range from about 10 microns to 2 millimeters.

[0017] A continuous powder application module **116** is positioned on a feed side **118** of the counter-rotating roller module **110** to continuously deposit a volume **122** (e.g., a heap or charge of powder) of a dry powder mixture **124** on the conductive substrate **106** prior to contact with the counter-rotating roller **112**. In various embodiment, a heap or charge of the dry powder mixture **124** is deposited by the continuous powder application module **116** in front of the counter-rotating roller **112** (e.g., on the feed side **118** prior to particles of the dry powder mixture **124** coming into contact with the counter-rotating roller **112** due to the conductive substrate **106** traveling along the first direction).

[0018] The dry powder mixture **124** includes a mixture of intermediate dry particles, with each intermediate dry particle being a composite particle that has an engineered structure that has all the requisite constituents to make an electrode. A fully functioning battery electrode is fabricated by depositing this dry particle on a current collector (e.g., conductive substrate **106**) and applying a combination of heat and/or pressure to bind the intermediate dry particles to the current collector. For the purposes of this disclosure, an intermediate dry particle refers to a particle having all the required constituents in correct proportions as in an equivalent slurry without the solvent. Specifically, in the case where the electrode is a cathode, the intermediate dry particle has the binder and a conductive additive coated and mixed with the active electrode material. In some embodiments, the intermediate dry particle is a composite powder. In other embodiments, the intermediate dry particle may not be a composite powder.

[0019] In some embodiments, the dry powder mixture **124** includes a flow agent mixed in with the intermediate dry particles to alter the inter-particle interactions that influence the spreading on the conductive substrate **106**. The particle size distribution of the intermediate dry particles in the dry

powder mixture **124** is controlled to control the deposition and spreading on the conductive substrate **106**. In various embodiments, the conductive substrate **106** is a metal foil that is primed or pre-coated with carbon or other agent to improve bonding or performance of the electrode. For example, in one embodiment, the conductive substrate **106** is a corona treated foil that improves adhesion to the dry powder mixture **124**. In another embodiment, the conductive substrate **106** is a plasma-treated foil that improve adhesion to the dry powder mixture **124**.

[0020] In various embodiments, the counter-rotating roller **112** rotates in a direction opposite to the travel direction of the conductive substrate **106**. For example, with reference to FIG. 4, a tangential velocity direction **132** of the counter-rotating roller **112** at an interface (e.g., point of least distance) between the counter-rotating roller **112** and the conductive substrate **106** is 180 degrees opposite the first direction of travel of the conductive substrate **106**. This interface between a roller and the substrate is sometimes referred to as the nip where there is direct contact between the roller and the substrate but is referred to as the interface to clarify that there is a gap (e.g., of height **114**) underneath which the dry powder mixture **124** passes. In this manner, the counter-rotating roller **112** which is positioned at a fixed height **114** over the conductive substrate **106** spreads the heap or charge of the dry powder mixture **124** and to form a thin film layer of the dry powder mixture **124** to the conductive substrate **106**. The counter-rotation movement of the counter-rotating roller **112** helps to distribute compaction force evenly and avoids creating hollows or ridges in the material being compacted (e.g., the dry powder mixture **124**) by evenly distributing force and preventing skewing or uneven compaction.

[0021] In some embodiments, the counter-rotating roller module **110** includes flexible wipers or scrapers to prevent the dry powder mixture **124** from going on the other side. For example, as shown in FIG. 4, the counter-rotating roller module **110** includes a contact roller scraper **126** contacting the counter-rotating roller **112** that is configured to remove any residual powder particles from an outer surface **128** of the counter-rotating roller **112**. The contact roller scraper **126**, in various embodiments, is a flexible member (e.g., constructed out of rubber) that extends along approximately the entirety of the longitudinal length of the counter-rotating roller **112** and is in direct contact with the outer surface of the counter-rotating roller **112** such that any residual powder particles are scraped off the outer surface **128** and fall back to the feed side **118** of the counter-rotating roller module **110**. That is, the contact roller scraper prevents powder particles of the dry powder mixture **124** from being carried by rotational movement over the top of the counter-rotating roller and falling to the post-roller side **120** of the counter-rotating roller module **110** as uneven loose powder (as opposed to properly passing underneath the counter-rotating roller **112** and emerging on the post-roller side **120** as a uniform film layer).

[0022] The counter-rotating roller system **100** also includes an orientation maintenance module **130** designed to maintain the conductive substrate **106** in a flat orientation beneath the counter-rotating roller module **110**. In some embodiments, the orientation maintenance module **130** is a high-precision support plate or a vacuum table system that the conductive substrate **106** remains uniformly flat and does not skew off track during powder deposition and compaction processes, thereby achieving consistent layer thickness and adherence across the substrate surface. In other embodiments, the orientation maintenance module **130** is a freely rotating roller or other mechanism capable of maintaining a flat and square orientation of the conductive substrate **106** relative to the counter-rotating module **110** and counter-rotating roller **112**.

[0023] It should be noted that the systems and methods of coating electrode materials to substrates using a counter-rotating roller system **100** can be performed with or without the application of heat. For example, in some embodiments, the counter-rotating roller system **100** includes a powder heating module (not shown) configured to heat the heap of dry powder mixture **124** and/or heat the conductive substrate **106** prior to contact with the counter-rotating roller **112** for improved spreading characteristics. In some embodiments, prior to engagement with the counter-rotating roller module **110**, or a first section of the counter rotating module **110**, the dry powder mixture **124**

undergoes pre-heating using a dedicated heating unit employing established powder heating techniques to bring the powder to an optimal temperature that enhances the powder's readiness for compaction and adherence to the electrode conductive substrate **106** upon contact with the counter-rotating roller **112**. For example, in some embodiments, the powder heating module includes but is not limited to: quartz lamps, infrared heaters, lasers (e.g., CO2 laser, diode laser, ND-Yag laser), heated gaseous fluids, microwave radiation heater, and the like. In various embodiments, the dry powder mixture **124** is heated prior to contacting the counter-rotating roller **112** to temperatures ranging between 20° C. to 200° C.

[0024] Similarly, in various embodiments, the surface temperature of the counter-rotating roller **112** is controlled. The counter-rotating roller **112** is heated either directly or indirectly to control flowability of the dry powder mixture **124** while spreading and coating. For example, in some embodiments, the counter-rotating roller module **110** includes an integrated heating system configured to control a temperature of the outer surface **126** of the counter-rotating roller **112**. The integrated heating system enables precise temperature adjustments across the roller surface to control the thermal conditions of the powder mixture deposited on the conductive electrode substrate, and is designed to apply a specific range of pressures to the powder-coated substrate, aiding in achieving a uniformly compacted layer. Heat is applied to the counter-rotating roller system **100** using heated counter-rotating rollers or other heated rollers that aid in the conveying and/or tamping of the dry powder mixture **124** on the conductive substrate **106**. In various embodiments, the rollers are heated with any combination of circulating oil, gas or other fluids, infra-red heating, resistive heating, and the like. In some embodiments, heat is applied to the respective areas using varying magnetic or electric field heating. Additionally, in some embodiments, a combination of two or more heat sources is applied to the rollers. In other embodiments, the counter-rotating roller **112** is cooled to control flowability of the dry powder mixture **124** while spreading and coating. In some embodiments, the diameter of the counter-rotating roller **112** is varied to control flowability of the dry powder mixture **124** while spreading and coating. For example, in various embodiments, the counter-rotating roller **112** is heated to temperatures ranging between 20° C. to 200° C.

[0025] In other embodiments, heat is applied to the conductive substrate **106** before the application of the dry powder mixture **124**. In other embodiments, heat is applied to the conductive substrate **106** after it has been coated with the dry powder mixture **124**. In other embodiments, the dry powder mixture **124** is heated before application to the conductive substrate. In other embodiments, the dry powder mixture **124** is heated after application to the conductive substrate. In some other embodiments, the dry powder mixture **124** is heated and then applied to a conductive substrate **106** that is also heated.

[0026] In some embodiments, the surface finish of the outer surface **128** of the counter-rotating roller **112** is selected to adjust the coefficient of friction between the dry powder mixture **124** and the counter-rotating roller **112**. The material composition and surface roughness **128** of the counter-rotating roller **112** are selected to modulate frictional interactions between the dry powder mixture **124**, conductive substrate **106**, and the counter-rotating roller **112**, ensuring optimal compaction and layer consistency to achieve desired electrode layer properties. The surface finish of the outer surface **128** of the counter-rotating roller **112** is also selected to adjust the coefficient of friction between the conductive substrate **106** and the counter-rotating roller **112**. For example, in some embodiments, the surface finish of the counter rotating roller is in a range of having average surface roughness (Ra) values are between 0.1 micron Ra (smooth) to 50 micron Ra measured according to the ISO 21920 standard. The choice of material of the counter-rotating roller **112** is varied to control flowability of the dry powder mixture **124** while spreading and coating. Additionally, in some embodiments, the counter-rotating roller **112** is electrically grounded to prevent static charge formation.

[0027] In some embodiments, the counter-rotating roller system **100** further includes a doctor blade

(not shown) to assist in the spreading of the dry powder mixture **124**. The dry powder mixture **124** is heaped or spread on the uncoated side of the foil, and then the height of the doctor blade is adjusted to a sufficient height to apply a uniform coat of dry powder mixture **124** particles on the conductive substrate **106**. In some embodiments, the fixed surfaces of the doctor blade rides on the conductive substrate **106**. In other embodiments, the fixed surfaces of the doctor blade rides above the conductive substrate **106** and not touch the conductive substrate **106** in any way. In some embodiments, the doctor blade is stationary. In other embodiments, the doctor blade moves. In some embodiments, the leading and trailing profile of the doctor blade is varied to be able to control the spreading and coating of the dry powder mixture **124**. In some embodiments, the surface roughness of the leading and trailing profile of the doctor blade is varied to be able to control the spreading and coating of the dry powder mixture **124**. In some embodiments, the choice of material of the doctor blade is varied to be able to control the spreading and coating of the dry powder mixture **124**.

[0028] In one embodiment, the application and spreading of the dry powder mixture **124** is followed by the heating and pressing of the dry powder mixture **124** on the conductive substrate **106** (referred to as a roll-to-roll manufacturing process). In one embodiment, the dry powder mixture **124** including intermediate dry particles is spread evenly on the conductive substrate **106** and then heated and pressed after passing underneath the counter-rotating roller **112** to yield a fully functional dry battery electrode. For example, in various embodiments, the counter-rotating roller system **100** includes a post-processing compression module (not shown) positioned downstream of the counter-rotating roller module **110**, wherein the post-processing compression module includes one or more heated rollers configured to exert an additional compaction force on the conductive substrate **106**.

[0029] It should be recognized that FIGS. **1-5** are described above primarily in the context of presenting the conductive substrate **106** to the counter-rotating roller **112** with a roll-to-roll mechanism. However, those skilled in the art will recognize that the counter-rotating roller system **100** may present the conductive substrate **106** using various other mechanisms without departing from the scope of this disclosure. For example, in some embodiments, the conductive substrate **106** is presented to the counter-rotating roller **112** by sheet feeding, manual placement, or automated conveyor system, allowing the processing of substrates in diverse forms and sizes. In some embodiments, the conductive substrate may remain stationary with the counter-rotating roller moving relative to the conductive substrate. In some embodiments, the application, heating and pressing of the dry powder mixture **124** is performed on discrete pieces of metal foil. In some embodiments, irrespective of whether it is a roll-to-roll process or a discrete process, the dry powder mixture **124** is applied and spread and then fixed or pressed with only pressure and without the application of heat. In other embodiments, the conductive substrate **106** is fixed and the spreading mechanism (e.g., counter-rotating roller **112**) moves relative to the conductive substrate **106** instead.

[0030] With reference to FIG. **5**, a perspective and exploded view of a continuous powder application module **116** according to one embodiment is shown therein. The continuous powder application module **116** is configured to precisely control the volume and rate of dry powder mixture **124** dispensed onto the conductive substrate **106**, utilizing real-time feedback mechanisms for uniform deposition and optimal powder utilization. As shown, the powder application module **116** includes a powder hopper **502** for retaining the dry powder mixture **124**. The powder hopper **502** includes a bottom plate **504** that secures a sieve screen **506** (e.g., a mesh) to the powder hopper **502**. Vibrations are applied to the powder hopper **502** by the vibrator **508**, thereby causing particles of the dry powder mixture **124** to be dispersed from the bottom out through the sieve screen **506** and onto the conductive substrate **106** due to the mechanical vibrations. In some embodiments, the vibrating powder hopper **502** with a control mesh opening is used to provide feed material to the counter-rotating roller **112**. According to various embodiments, the sieve screen **506** has a mesh or

screen opening size in the range between US Mesh size 10 (2000 micron opening) to US Mesh size 1250 (10 micron opening). In some embodiments, the sieve screen **506** has a mesh or screen opening size in the range between US Mesh size 50 (297 micron opening) to US Mesh size 400 (37 micron opening).

[0031] In other embodiments, the vibrating powder hopper **502** provides feed material to a doctor blade. In some embodiments, the vibrating powder hopper **502** with mesh bottom is stationary. In other embodiments, the vibrating powder hopper **502** moves along one axis as the conductive substrate **106** is being coated. In yet another embodiment, the powder hopper **502** moves back and forth across the width of the conductive substrate **106**. In some embodiments, the mesh or opening size of the vibrating powder hopper **502** is varied to be able to better control the spreading and coating of the dry powder mixture **124**. In some embodiments of the hole size in the bottom of the vibrating hopper is not uniform. In some embodiments, the amplitude and frequency of mechanical vibration is varied to be able to control the spreading and coating of the dry powder mixture **124**. In some embodiments, the width of the tank in the direction of relative movement with respect to the conductive substrate **106** is varied to be able to control the spreading and coating of the dry powder mixture **124**. In other embodiments, the heap or charge of the dry powder mixture **124** is deposited or fed in front of the counter-rotating roller **112** using other apparatuses. For example, in various embodiments, the continuous powder application module **116** includes but is not limited to: a star feeder, a screw feeder, and the like.

[0032] Additionally, other apparatuses for continuously applying powder to the conductive substrate **106** include an inclined vibrating table. The surface roughness of the base of the vibrating table, the amplitude and frequency of the vibration, the height above the conductive substrate **106**, temperature of the vibrating table, the addition of a gate towards the dispensing edge of the table are parameters that are controlled to get a good spreading and coating on the foil. Similarly, in other embodiments, the continuous powder application module **116** includes a dry powder spraying system such as a high volume low pressure (HVLP) spraying system or a low volume low pressure (LVLP) system in which pressured air or any other gaseous fluid aerosolizes particles of the dry powder mixture **124** and transfers the particles to the conductive substrate **106**. In various embodiments, the diameter and shape of the nozzle, the volume and pressure of the gaseous fluid, the temperature of the gaseous fluid, the distance between the nozzle, and the substrate are parameters varied to achieve better spreading and coating on the conductive substrate **106**. In various embodiments, different combinations of feeding methods and leveling method might be used, including but not limited to multiples of feeding and multiples of leveling methods in series.

[0033] It should be recognized that FIGS. **1-4** are described above primarily in the context of a singular counter-rotating roller **112** for ease of illustration and description. However, those skilled in the art will recognize that the counter-rotating roller system **100** may include any number of rollers without departing from the scope of this disclosure. For example, in some embodiments, the counter-rotating roller module **110** includes two or more counter-rotating rollers arranged in series at different heights above the conductive substrate **106** to incrementally increase the density (e.g., by reducing the thickness of the film layer) of the electrode substrate by sequentially applying compaction forces, thereby optimizing the powder layer's density and uniformity. Additionally, those skilled in the art will recognize that the two or more counter-rotating rollers may be positioned at different heights, rotate at different speeds, have different surface finishes/textures, and/or heated to different temperatures relative to each other.

[0034] Referring now to FIG. **6**, illustrated is a block diagram of a method **600** for coating electrode materials to conductive substrates in accordance with some embodiments. For ease of illustration and description, the method **600** is described below with reference to and in an example context of the counter-rotating roller system of FIGS. **1-5**. However, the method **600** is not limited to these example contexts, but instead may be employed for any of a variety of possible configurations using the guidelines provided herein.



[0035] The method **600** begins at block **602** with depositing, at a position upstream of a counter-rotating roller, a volume of a dry powder mixture onto a surface of a conductive substrate. In various embodiments, the operations of block **602** include continuously depositing a volume of the dry powder mixture as the conductive substrate travels along a first direction towards the counter-rotating roller. For example, in the context of FIGS. **1-5**, a continuous powder application module **116** is positioned on a feed side **118** of the counter-rotating roller module **110** to continuously deposit a volume **122** of a dry powder mixture **124** on the conductive substrate **106** prior to contact with the counter-rotating roller **112**. The continuous deposition of the volume of a dry powder mixture on the substrate is performed prior to its engagement with the counter-rotating roller, with the volume deposited to achieve a predetermined thickness of the powder on the conductive substrate.

[0036] In various embodiments, the operations of block **602** also include precisely controlling the volume and rate of dry powder mixture dispensed onto the substrate using real-time feedback mechanisms. For example, in some embodiments, a rate of powder deposition from the continuous powder application module **116** is closely match to a volume of powder mixture spread by and passing underneath the counter-rotating roller **112** to prevent a gradual buildup of dry powder mixture **124** on the feed side **118** of the counter-rotating roller module **110** that would eventually overwhelm the contact roller scraper **126** and/or spill off of the conductive substrate **106** and orientation maintenance module **130**.

[0037] In various embodiment, the conductive substrate is primed or pre-coated with carbon or any other agent that improves bonding or performance of the electrode. In one embodiment, the foil can be corona treated to improve adhesion to the dry powder mixture **124**. In another embodiment, the foil can be plasma treated to improve adhesion to the dry powder mixture **124**. In various embodiments, a heap or charge of the dry powder mixture **124** is deposited by the continuous powder application module **116** in front of the counter-rotating roller **112** (e.g., on the feed side **118** prior to particles of the dry powder mixture **124** coming into contact with the counter-rotating roller **112** due to the conductive substrate **106** traveling along the first direction).

[0038] As illustrated in FIG. **5**, the powder application module **116** includes a powder hopper **502** for retaining the dry powder mixture **124** and configured to precisely control the volume and rate of dry powder mixture **124** dispensed onto the conductive substrate **106**, utilizing real-time feedback mechanisms for uniform deposition and optimal powder utilization. The powder hopper **502** includes a bottom plate **504** that secures a sieve screen **506** (e.g., a mesh) to the powder hopper **502**. Vibrations are applied to the powder hopper **502** by the vibrator **508**, thereby causing particles of the dry powder mixture **124** to be dispersed from the bottom out through the sieve screen **506** and onto the conductive substrate **106** due to the mechanical vibrations. In some embodiments, the vibrating powder hopper **502** with a control mesh opening is used to provide feed material to the counter-rotating roller **112**.

[0039] In other embodiments, the vibrating powder hopper **502** provides feed material to a doctor blade. In some embodiments, the vibrating powder hopper **502** with mesh bottom is stationary. In other embodiments, the vibrating powder hopper **502** moves along one axis as the conductive substrate **106** is being coated. In yet another embodiment, the powder hopper **502** moves back and forth across the width of the conductive substrate **106**. In some embodiments, the mesh or opening size of the vibrating powder hopper **502** is varied to be able to better control the spreading and coating of the dry powder mixture **124**. In some embodiments of the hole size in the bottom of the vibrating hopper is not uniform. In some embodiments, the amplitude and frequency of mechanical vibration is varied to be able to control the spreading and coating of the dry powder mixture **124**. In some embodiments, the width of the tank in the direction of relative movement with respect to the conductive substrate **106** is varied to be able to control the spreading and coating of the dry powder mixture **124**. In other embodiments, the heap or charge of the dry powder mixture **124** is deposited or fed in front of the counter-rotating roller **112** using other apparatuses. For example, in various

embodiments, the continuous powder application module **116** includes but is not limited to: a star feeder, a screw feeder, and the like.

[0040] Additionally, in other embodiments, an inclined vibrating table continuously applies powder to the conductive substrate **106**. The surface roughness of the base of the vibrating table, the amplitude and frequency of the vibration, the height above the conductive substrate **106**, temperature of the vibrating table, the addition of a gate towards the dispensing edge of the table are parameters that are controlled to get a good spreading and coating on the foil. Similarly, in other embodiments, the continuous powder application module **116** includes a dry powder spraying system such as a high volume low pressure (HVLP) spraying system or a low volume low pressure (LVLP) system in which pressured air or any other gaseous fluid aerosolizes particles of the dry powder mixture **124** and transfers the particles to the conductive substrate **106**. In various embodiments, the diameter and shape of the nozzle, the volume and pressure of the gaseous fluid, the temperature of the gaseous fluid, the distance between the nozzle, and the substrate are parameters varied to achieve better spreading and coating on the conductive substrate **106**. In various embodiments, different combinations of feeding methods and leveling method might be used, including but not limited to multiples of feeding and multiples of leveling methods in series.

[0041] In other embodiments, heat is applied to the conductive substrate **106** before the application of the dry powder mixture **124**. In other embodiments, heat is applied to the conductive substrate **106** after it has been coated with the dry powder mixture **124**. In other embodiments, the dry powder mixture **124** is heated before application to the conductive substrate. In other embodiments, the dry powder mixture **124** is heated after application to the conductive substrate. In some other embodiments, the dry powder mixture **124** is heated and then applied to a conductive substrate **106** that is also heated. For example, in some embodiments, the counter-rotating roller system **100** includes a powder heating module (not shown) configured to heat the heap of dry powder mixture **124** and/or heat the conductive substrate **106** prior to contact with the counter-rotating roller **112** for improved spreading characteristics. In some embodiments, prior to engagement with the counter-rotating roller module **110**, or a first section of the counter rotating module **110**, the dry powder mixture **124** undergoes pre-heating using a dedicated heating unit employing established powder heating techniques to bring the powder to an optimal temperature that enhances the powder's readiness for compaction and adherence to the electrode conductive substrate **106** upon contact with the counter-rotating roller **112**. For example, in some embodiments, the powder heating module includes but is not limited to: quartz lamps, infrared heaters, lasers (e.g., CO<sub>2</sub> laser, diode laser, ND-Yag laser), heated gaseous fluids, microwave radiation heater, and the like. In various embodiments, the dry powder mixture **124** is heated prior to contacting the counter-rotating roller **112** to temperatures ranging between 20° C. to 200° C.

[0042] The method **600** continues at block **604** with feeding the conductive substrate along a first direction towards the counter-rotating roller. With respect to FIGS. **1-4**, in various embodiments, the counter-rotating roller system **100** includes a roll-to-roll apparatus **102** including a dispensing roller **104** that selectively dispenses a conductive substrate **106** from a substrate roll **108**. The conductive substrate **106** is directed along a first direction (as indicated by the left-to-right arrows of FIGS. **1-4**) of travel towards a receiving roller (not shown). In some embodiments, the operations of block **604** further include adjusting the unwinding operation of the conductive substrate from the substrate roll **108** with both continuous and intermittent feed at variable speeds.

[0043] In various embodiments, the operations of block **604** also include maintaining the conductive substrate in a flat orientation beneath the counter-rotating roller. For example, with reference to FIG. **4**, in various embodiments, the counter-rotating roller system **100** also includes an orientation maintenance module **130** designed to maintain the conductive substrate **106** in a flat orientation beneath the counter-rotating roller module **110**. In some embodiments, the orientation maintenance module **130** is a high-precision support plate or a vacuum table system that the conductive substrate **106** remains uniformly flat and does not skew off track during powder

deposition and compaction processes, thereby achieving consistent layer thickness and adherence across the substrate surface. In other embodiments, the orientation maintenance module **130** is a freely rotating roller or other mechanism capable of maintaining a flat and square orientation of the conductive substrate **106** relative to the counter-rotating module **110** and counter-rotating roller **112**.

[0044] In other embodiments, rather than utilizing a roll-to-roll mechanism, feeding the conductive substrate to the counter-rotating roller includes sheet feeding, manual placement, or automated conveyor system, allowing the processing of substrates in diverse forms and sizes. In some embodiments, the conductive substrate may remain stationary with the counter-rotating roller moving relative to the conductive substrate. In some embodiments, the application, heating and pressing of the dry powder mixture **124** is performed on discrete pieces of metal foil.

[0045] The method **600** continues at block **606** with spreading, based at least in part on a counter-rotational movement of the counter-rotating roller, the volume of the dry powder mixture onto the surface of the conductive substrate, wherein a direction of the tangential velocity of the counter-rotating roller at an interface between the counter-rotating roller and the conductive substrate is 180 degrees opposite the first direction of travel of the conductive substrate. With respect to FIG. **4**, in various embodiments, a counter-rotating roller module **110** includes a counter-rotating roller **112** stationarily positioned at a predetermined height **114** over the conductive substrate **106**.

[0046] The counter-rotating roller **112** rotates in a direction opposite to the travel direction of the conductive substrate **106** to form a constant layer thickness and density of powder across the conductive substrate. For example, with reference to FIG. **4**, a tangential velocity direction **132** of the counter-rotating roller **112** at an interface (e.g., point of least distance) between the counter-rotating roller **112** and the conductive substrate **106** is 180 degrees opposite the first direction of travel of the conductive substrate **106**. This interface between a roller and the substrate is sometimes referred to as the nip where there is direct contact between the roller and the substrate but is referred to as the interface to clarify that there is a gap (e.g., of height **114**) underneath which the dry powder mixture **124** passes. In this manner, the counter-rotating roller **112** which is positioned at a fixed height **114** over the conductive substrate **106** spreads the heap or charge of the dry powder mixture **124** and to form a thin film layer of the dry powder mixture **124** to the conductive substrate **106**. The counter-rotation movement of the counter-rotating roller **112** helps to distribute compaction force evenly and avoids creating hollows or ridges in the material being compacted (e.g., the dry powder mixture **124**) by evenly distributing force and preventing skewing or uneven compaction. Further, counter rotation speed of the counter-rotating roller **112** is adjustable to be able to control the spreading and coating of the dry powder mixture **124**.

[0047] It should be noted that operations of block **606** for coating electrode materials to substrates using counter-rotational movement can be performed with or without the application of heat. For example, in some embodiments, the surface temperature of the counter-rotating roller **112** is controlled. The counter-rotating roller **112** is heated either directly or indirectly to control flowability of the dry powder mixture **124** while spreading and coating. For example, in some embodiments, the counter-rotating roller module **110** includes an integrated heating system configured to control a temperature of the outer surface **126** of the counter-rotating roller **112**. The integrated heating system enables precise temperature adjustments across the roller surface to control the thermal conditions of the powder mixture deposited on the conductive electrode substrate, and is designed to apply a specific range of pressures to the powder-coated substrate, aiding in achieving a uniformly compacted layer. Heat is applied to the counter-rotating roller system **100** using heated counter-rotating rollers or other heated rollers that aid in the conveying and/or tamping of the dry powder mixture **124** on the conductive substrate **106**. In various embodiments, the rollers are heated with any combination of circulating oil, gas or other fluids, infra-red heating, resistive heating, and the like. In some embodiments, heat is applied to the respective areas using varying magnetic or electric field heating. Additionally, in some

embodiments, a combination of two or more heat sources is applied to the rollers. In other embodiments, the counter-rotating roller **112** is cooled to control flowability of the dry powder mixture **124** while spreading and coating. In some embodiments, the diameter of the counter-rotating roller **112** is varied to control flowability of the dry powder mixture **124** while spreading and coating. For example, in various embodiments, the counter-rotating roller **112** is heated to temperatures ranging between 20° C. to 200° C.

[0048] In other embodiments, the counter-rotating roller system **100** further includes a doctor blade (not shown) to assist in the spreading of the dry powder mixture **124**. The dry powder mixture **124** is heaped or spread on the uncoated side of the foil, and then the height of the doctor blade is adjusted to a sufficient height to apply a uniform coat of dry powder mixture **124** particles on the conductive substrate **106**. In some embodiments, the fixed surfaces of the doctor blade rides on the conductive substrate **106**. In other embodiments, the fixed surfaces of the doctor blade rides above the conductive substrate **106** and not touch the conductive substrate **106** in any way. In some embodiments, the doctor blade is stationary. In other embodiments, the doctor blade moves. In some embodiments, the leading and trailing profile of the doctor blade is varied to be able to control the spreading and coating of the dry powder mixture **124**. In some embodiments, the surface roughness of the leading and trailing profile of the doctor blade is varied to be able to control the spreading and coating of the dry powder mixture **124**. In some embodiments, the choice of material of the doctor blade is varied to be able to control the spreading and coating of the dry powder mixture **124**.

[0049] The method **600** continues at block **608** with scraping, based on a contact roller scraper contacting the counter-rotating roller, residual powder particles from an outer surface of the counter-rotating roller. With respect to FIG. **4**, in various embodiments, the counter-rotating roller module **110** includes flexible wipers or scrapers to prevent the dry powder mixture **124** from going on the other side. For example, as shown in FIG. **4**, the counter-rotating roller module **110** includes a contact roller scraper **126** contacting the counter-rotating roller **112** that is configured to remove any residual powder particles from an outer surface **128** of the counter-rotating roller **112**. The contact roller scraper **126**, in various embodiments, is a flexible member (e.g., constructed out of rubber) that extends along approximately the entirety of the longitudinal length of the counter-rotating roller **112** and is in direct contact with the outer surface of the counter-rotating roller **112** such that any residual powder particles are scraped off the outer surface **128** and fall back to the feed side **118** of the counter-rotating roller module **110**. That is, the contact roller scraper prevents powder particles of the dry powder mixture **124** from being carried by rotational movement over the top of the counter-rotating roller and falling to the post-roller side **120** of the counter-rotating roller module **110** as uneven loose powder (as opposed to properly passing underneath the counter-rotating roller **112** and emerging on the post-roller side **120** as a uniform film layer).

[0050] In some embodiments, the operations of block **608** are followed by the heating and/or pressing of the dry powder mixture **124** on the conductive substrate **106** (referred to as a roll-to-roll manufacturing process), thereby compacting the powder-coated substrate to form an electrode layer. In one embodiment, the dry powder mixture **124** including intermediate dry particles is spread evenly on the conductive substrate **106** and then heated and pressed after passing underneath the counter-rotating roller **112** to yield a fully functional dry battery electrode. For example, in various embodiments, the counter-rotating roller system **100** includes a post-processing compression module (not shown) positioned downstream of the counter-rotating roller module **110**, wherein the post-processing compression module includes one or more heated rollers configured to exert an additional compaction force on the conductive substrate **106**. In some embodiments, irrespective of whether it is a roll-to-roll process or a discrete process, the dry powder mixture **124** is applied and spread and then fixed or pressed with only pressure and without the application of heat.

[0051] It should be recognized that FIGS. **1-6** are described above primarily in the context of a

singular counter-rotating roller **112** for ease of illustration and description. However, those skilled in the art will recognize that the counter-rotating roller system **100** may include any number of rollers without departing from the scope of this disclosure. In some embodiments, post-processing compression pressure is applied using two rollers similar to the traditional rolling mill after the conductive substrate is coated with the dry powder mixture **124**. The diameter of the rollers, the surface finish and roughness of the rollers, material choice of the rollers, speed of the rollers, any coatings on the rollers (nitriding, anti-slip, non-stick, anti-corrosion), temperature of the rollers, and the gap between the rollers are important to control to get the appropriate final electrode thickness. [0052] For example, in some embodiments, the counter-rotating roller module **110** includes two or more counter-rotating rollers arranged in series at different heights above the conductive substrate **106** to incrementally increase the density (e.g., by reducing the thickness of the film layer) of the electrode substrate by sequentially applying compaction forces, thereby optimizing the powder layer's density and uniformity, sometime referred to as laminating. Additionally, those skilled in the art will recognize that the two or more counter-rotating rollers may be positioned at different heights, rotate at different speeds, have different surface finishes/textures, and/or heated to different temperatures relative to each other.

[0053] In some embodiments, there are successive pairs of rollers that successively compress the foil with the coated dry powder mixture **124**. This is achieved by having decreasing spacing between the two rollers in each successive roller pair. In some embodiments, there is only one roller and instead of the other roller it can be a fixed object/plate/table/curved surface where the foil slides over. In some embodiments, the angle of wrap is varied between the foil and roller for improved binding of the dry powder mixture **124** to the conductive substrate **106**. In some embodiments, the roller is completely replaced by an inclined upper belt and a flat lower belt where the gap successively decreases along the length of the belts. The belt can be constructed with a variety of materials and the foil coated with the dry powder mixture **124** will move through the belt. In another embodiment, the upper belt and lower belt are both inclined. These belts could be heated or not heated.

[0054] Accordingly, as discussed herein, FIGS. **1-6** describe techniques that improve electrode manufacturing for use in electrochemical energy storage devices such as batteries and more specifically lithium-ion based batteries by helping to reduce capital and operating costs, energy usage and eliminate or minimize the use of toxic and environmentally hazardous materials. This disclosure can be applied to both the anode electrode manufacturing and cathode electrode manufacturing of the secondary ionic battery such as a lithium-ion battery, a sodium-ion battery, and the like.

[0055] Throughout this disclosure, the numerical values represent approximate measures or limits to ranges to encompass minor deviations from the given values and embodiments having about the value mentioned as well as those having exactly the value mentioned. Other than in the working examples provided at the end of the detailed description, all numerical values of parameters (e.g., of quantities or conditions) in this specification, including the appended claims, are to be understood as being modified in all instances by the term “about” whether or not “about” actually appears before the numerical value. “About” indicates that the stated numerical value allows some slight imprecision (with some approach to exactness in the value; approximately or reasonably close to the value; nearly). If the imprecision provided by “about” is not otherwise understood in the art with this ordinary meaning, then “about” as used herein indicates at least variations that may arise from ordinary methods of measuring and using such parameters. For example, “about” may comprise a variation of less than or equal to 5%, optionally less than or equal to 4%, optionally less than or equal to 3%, optionally less than or equal to 2%, optionally less than or equal to 1%, optionally less than or equal to 0.5%, and in certain aspects, optionally less than or equal to 0.1%. In addition, disclosure of ranges includes disclosure of all values and further divided ranges within the entire range, including endpoints and sub-ranges given for the ranges.

[0056] Note that not all of the activities or elements described above in the general description are required, that a portion of a specific activity or device may not be required, and that one or more further activities may be performed, or elements included, in addition to those described. Still further, the order in which activities are listed are not necessarily the order in which they are performed. Also, the concepts have been described with reference to specific embodiments. However, one of ordinary skill in the art appreciates that various modifications and changes can be made without departing from the scope of the present disclosure as set forth in the claims below. Accordingly, the specification and figures are to be regarded in an illustrative rather than a restrictive sense, and all such modifications are intended to be included within the scope of the present disclosure.

[0057] Benefits, other advantages, and solutions to problems have been described above with regard to specific embodiments. However, the benefits, advantages, solutions to problems, and any feature(s) that may cause any benefit, advantage, or solution to occur or become more pronounced are not to be construed as a critical, required, or essential feature of any or all the claims. Moreover, the particular embodiments disclosed above are illustrative only, as the disclosed subject matter may be modified and practiced in different but equivalent manners apparent to those skilled in the art having the benefit of the teachings herein. No limitations are intended to the details of construction or design herein shown, other than as described in the claims below. It is therefore evident that the particular embodiments disclosed above may be altered or modified and all such variations are considered within the scope of the disclosed subject matter. Accordingly, the protection sought herein is as set forth in the claims below.

## Claims

1. A system, comprising: a roll-to-roll apparatus including a dispensing roller that selectively dispenses a conductive substrate from a substrate roll, wherein the conductive substrate is directed in an upwardly inclined direction towards a first curved edge of a stationary orientation maintenance module before transitioning to feeding the conductive substrate over a horizontal surface of the stationary orientation maintenance module; a counter-rotating roller module including a counter-rotating roller stationarily positioned at a predetermined height over the conductive substrate, wherein a tangential velocity direction of the counter-rotating roller at an interface between the counter-rotating roller and the conductive substrate is 180 degrees opposite the first direction of travel of the conductive substrate; a continuous powder application module positioned on a feed side of the counter-rotating roller module to continuously deposit a volume of a dry powder mixture on the conductive substrate prior to contact with the counter-rotating roller; and a contact roller scraper contacting the counter-rotating roller and configured to remove residual powder particles from an outer surface of the counter-rotating roller.
2. The system of claim 1, wherein the continuous powder dispenser includes a vibrating hopper sieve.
3. The system of claim 1, wherein the counter-rotating roller module includes two or more counter-rotating rollers arranged in series.
4. The system of claim 1, further comprising: a post-processing compression module positioned downstream of the counter-rotating roller module, wherein the post-processing compression module includes one or more heated rollers configured to exert an additional compaction force on the conductive substrate.
5. The system of claim 1, wherein the conductive substrate includes materials selected from the group consisting of graphene-coated substrates, graphite-coated substrates, carbon-coated substrates, copper foils, aluminum foils, metal foils, conductive polymers, conductive polymer coated metal foils, carbon fiber mats, nickel foam, Polyethylene Terephthalate (PET) coated with conductive layers, stainless steel mesh, and titanium foils.

- 6.** The system of claim 1, further comprising: an orientation maintenance module configured to maintain the conductive substrate in a flat orientation beneath the counter-rotating roller module.
- 7.** The system of claim 1, wherein the counter-rotating roller module further includes an integrated heating system configured to adjust temperatures across the outer surface of the counter-rotating roller.
- 8.** An apparatus, comprising: a counter-rotating roller module including a counter-rotating roller stationarily positioned at a predetermined height over a conductive substrate, wherein a direction of the tangential velocity of the counter-rotating roller at an interface between the counter-rotating roller and the conductive substrate is 180 degrees opposite a first direction of travel of the conductive substrate; a continuous powder application module positioned on a feed side of the counter-rotating roller module to continuously deposit a volume of a dry powder mixture on the conductive substrate prior to contact with the counter-rotating roller; and a contact roller scraper contacting the counter-rotating roller and configured to remove residual powder particles from an outer surface of the counter-rotating roller.
- 9.** The apparatus of claim 8, wherein the continuous powder dispenser includes a vibrating hopper sieve.
- 10.** The apparatus of claim 8, wherein the counter-rotating roller module includes two or more counter-rotating rollers arranged in series.
- 11.** The apparatus of claim 8, further comprising: a post-processing compression module positioned downstream of the counter-rotating roller module, wherein the post-processing compression module includes one or more heated rollers configured to exert an additional compaction force on the conductive substrate.
- 12.** The apparatus of claim 8, wherein the conductive substrate includes materials selected from the group consisting of graphene-coated substrates, graphite-coated substrates, carbon-coated substrates, copper foils, aluminum foils, metal foils, conductive polymers, conductive polymer coated metal foils, carbon fiber mats, nickel foam, Polyethylene Terephthalate (PET) coated with conductive layers, stainless steel mesh, and titanium foils.
- 13.** The apparatus of claim 8, further comprising: an orientation maintenance module configured to maintain the conductive substrate in a flat orientation beneath the counter-rotating roller module.
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