

METHODS AND SYSTEMS FOR RECLAMATION OF LI-ION CATHODE MATERIALS USING MICROWAVE PLASMA PROCESSING

INCORPORATION BY REFERENCE TO ANY PRIORITY APPLICATIONS

[0001] This application claims the priority benefit under 35 U.S.C. §119(e) of U.S. Provisional Application No. 63/135,948, filed January 11, 2021, the entire disclosure of which is incorporated herein by reference.

BACKGROUND

Field

[0002] Some embodiments of the present disclosure are directed to systems and methods for reclaiming used cathode materials using microwave plasma processing.

Description

[0003] Lithium-ion batteries (LIBs) have dominated the secondary energy storage market due to their unmatched combination of energy density (150-200 W h/kg, normalized by device mass), power output (>300 W/kg), and cycle stability (~2000 cycles) coupled with lower costs due to the increasing global production capacity. Worldwide trends in mobile electrification, largely driven by the popularity of electric vehicles (EVs) has significantly increased demand for LIB production. As such, millions of metric tons of LIB waste from EV battery packs will be generated over the next several decades alone. Moreover, LIB technology is expected to play an important role in stationary energy storage systems that require high power output, enabling energy harvesting from intermittent natural sources. LIB recycling directly addresses concerns over long-term economic strains and environmental issues associated with both landfilling and raw material extraction. However, LIB recycling infrastructure has not been widely adopted, and current facilities are mostly focused on Co recovery for economic gains, rather than reuse of cathode materials.

[0004] Recycling processes to recover or reuse metals in mixed-metal LIB cathodes and comingled scrap comprising different chemistries are needed. These processes require a low environmental footprint and energy consumption. In some existing processes, a pretreatment may be used to separate the cathode materials from other battery components,

followed by entirely dissolving the active material using reductive acid leaching. A complex leachate is generated, comprising cathode metals (Li^+ , Ni^{2+} , Mn^{2+} , and Co^{2+}) and impurities (Fe^{3+} , Al^{3+} , and Cu^{2+}) from the current collectors and battery casing, which can be separated and purified using a series of selective precipitation and/or solvent extraction steps. Alternatively, the cathode can be resynthesized directly from the leachate. In other existing methods, the battery materials undergo a high-temperature melting-and-extraction, or smelting, process. Those operations are energy intensive, expensive, and operate and require sophisticated equipment to treat harmful emissions generated by the smelting process. Despite the high costs, these processes cannot recover all valuable battery materials.

[0005] It is evident that recycling infrastructure cannot primarily focus on recovering Co to maximize profits, especially given the market trends for LIB cathode chemistries driven by the EV market. Even now, cathode materials such as $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$ (NMC-111) are being substituted with $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ (NMC-622) and $\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$ (NMC-811), which comprise even smaller quantities of Co. Thus, recycling processes must handle diverse mixed-type cathodes and comingled scraps containing various cathode chemistries with high efficiency. In addition, in view for the growing demand for LIB cathode materials, recycling processes should be capable of producing usable cathode materials for LIBs through lithium supplementation, rather than simply recovering the metals, such as Co, separately.

SUMMARY

[0006] For purposes of this summary, certain aspects, advantages, and novel features of the invention are described herein. It is to be understood that not all such advantages necessarily may be achieved in accordance with any particular embodiment of the invention. Thus, for example, those skilled in the art will recognize that the invention may be embodied or carried out in a manner that achieves one advantage or group of advantages as taught herein without necessarily achieving other advantages as may be taught or suggested herein.

[0007] Some embodiments herein are directed to methods for synthesizing lithium nickel manganese cobalt oxide (NMC) powder in a microwave plasma apparatus, the method comprising: providing a feedstock to the microwave plasma apparatus, the feedstock

comprising end-of-life NMC powder, the end-of-life NMC powder having an average nickel to cobalt ratio of 5:2 or less; and introducing the feedstock into a microwave-generated plasma of the microwave plasma apparatus to synthesize an NMC powder having an average nickel to cobalt ratio greater than 5:2.

[0008] In some embodiments, the method further comprises introducing nickel containing material, manganese containing material, or cobalt containing material into the microwave-generated plasma concurrently with introducing the feedstock into the microwave-generated plasma. In some embodiments, a microstructure of the end-of-life NMC powder comprises one or more imperfections, cracks, or fissures, and wherein introducing the feedstock into the microwave-generated plasma melts the end-of-life NMC powder. In some embodiments, a microstructure of the synthesized NMC powder does not comprise the one or more imperfections, cracks, or fissures.

[0009] In some embodiments, the method further comprises introducing lithium (Li) containing material into the microwave-generated plasma concurrently introducing the feedstock into the microwave-generated plasma. In some embodiments, the end-of-life NMC powder comprises NMC-532 or NMC-111. In some embodiments, the method further comprises adding lithium (Li) containing material to the feedstock prior to introducing the feedstock into the microwave-generated plasma. In some embodiments, the end-of-life NMC powder is obtained from a used lithium-ion battery.

[0010] Some embodiments herein are directed to methods for synthesizing lithium nickel manganese cobalt oxide (NMC) powder in a microwave plasma apparatus, the method comprising: providing a feedstock to the microwave plasma apparatus, the feedstock comprising end-of-life NMC powder, the end-of-life NMC powder and having an average nickel to cobalt ratio of 5:2 or less; and introducing the end-of-life NMC powder into a microwave-generated plasma of the microwave plasma apparatus to synthesize an NMC powder having an average nickel to cobalt ratio greater than 5:2, wherein the end-of-life NMC powder is not reduced to its constituent elements prior to introducing the end-of-life NMC powder into the microwave-generated plasma.

[0011] In some embodiments, the method further comprises introducing nickel containing material into the microwave-generated plasma concurrently with introducing the feedstock into the microwave-generated plasma. In some embodiments, a microstructure of

the end-of-life NMC powder comprises one or more imperfections, cracks, or fissures, and wherein introducing the feedstock into the microwave-generated plasma melts the end-of-life NMC powder. In some embodiments, a microstructure of the synthesized NMC powder does not comprise the one or more imperfections, cracks, or fissures.

[0012] In some embodiments, the method further comprises introducing lithium (Li) containing material into the microwave-generated plasma concurrently introducing the feedstock into the microwave-generated plasma. In some embodiments, the end-of-life NMC powder comprises NMC-532 or NMC-111. In some embodiments, the method further comprises adding lithium (Li) containing material to the feedstock prior to introducing the feedstock into the microwave-generated plasma. In some embodiments, the end-of-life NMC powder is obtained from a used lithium-ion battery.

[0013] Some embodiments herein are directed to lithium nickel manganese cobalt oxide (NMC) powders produced by a method comprising: providing a feedstock to a microwave plasma apparatus, the feedstock comprising end-of-life NMC powder, the end-of-life NMC powder having an average nickel to cobalt ratio of 5:2 or less; and introducing the feedstock into a microwave-generated plasma of the microwave plasma apparatus to synthesize an NMC powder or an NMC precursor having an average nickel to cobalt ratio greater than 5:2.

[0014] In some embodiments, the end-of-life NMC powder comprises NMC-111, NMC-442, or NMC-532. In some embodiments, the NMC powder or the NMC precursor comprises NMC-611, NMC-811, or NMC-9.5.5. In some embodiments, the NMC powder or the NMC precursor has an average nickel to cobalt ratio of 5:2, 6:1, 8:1, or 18:1.

BRIEF DESCRIPTION OF THE DRAWINGS

[0015] The drawings are provided to illustrate example embodiments and are not intended to limit the scope of the disclosure. A better understanding of the systems and methods described herein will be appreciated upon reference to the following description in conjunction with the accompanying drawings, wherein:

[0016] FIG. 1 illustrates a flowchart of an example process for recycling a used solid feedstock using a microwave plasma process according to embodiments of the present disclosure.

[0017] FIG. 2 illustrates an embodiment of a top feeding microwave plasma torch that can be used in the production of recycled solid LIB precursors, according to embodiments of the present disclosure.

[0018] FIGS. 3A-3B illustrate embodiments of a microwave plasma torch that can be used in the production of recycled solid LIB precursors, according to a side feeding hopper embodiment of the present disclosure.

DETAILED DESCRIPTION

[0019] Although certain preferred embodiments and examples are disclosed below, inventive subject matter extends beyond the specifically disclosed embodiments to other alternative embodiments and/or uses and to modifications and equivalents thereof. Thus, the scope of the claims appended hereto is not limited by any of the particular embodiments described below. For example, in any method or process disclosed herein, the acts or operations of the method or process may be performed in any suitable sequence and are not necessarily limited to any particular disclosed sequence. Various operations may be described as multiple discrete operations in turn, in a manner that may be helpful in understanding certain embodiments; however, the order of description should not be construed to imply that these operations are order dependent. Additionally, the structures, systems, and/or devices described herein may be embodied as integrated components or as separate components. For purposes of comparing various embodiments, certain aspects and advantages of these embodiments are described. Not necessarily all such aspects or advantages are achieved by any particular embodiment. Thus, for example, various embodiments may be carried out in a manner that achieves or optimizes one advantage or group of advantages as taught herein without necessarily achieving other aspects or advantages as may also be taught or suggested herein.

[0020] Certain exemplary embodiments will now be described to provide an overall understanding of the principles of the structure, function, manufacture, and use of the devices and methods disclosed herein. One or more examples of these embodiments are illustrated in the accompanying drawings. Those skilled in the art will understand that the devices and methods specifically described herein and illustrated in the accompanying

drawings are non-limiting exemplary embodiments and that the scope of the present invention is defined solely by the claims. The features illustrated or described in connection with one exemplary embodiment may be combined with the features of other embodiments. Such modifications and variations are intended to be included within the scope of the present technology.

[0021] Disclosed herein are embodiments of systems and methods for recycling used solid feedstocks containing lithium powders for use in LIBs and battery cells. The powders may be Lithium Nickel Manganese Cobalt Oxide (NMC) materials. In some embodiments, the used solid feedstock can undergo a microwave plasma process to produce a newly usable, lithium supplemented solid precursor.

[0022] Specifically, disclosed herein are methodologies, systems, and apparatus for producing recycled lithium-containing particles and Li-ion battery materials from used solid feedstocks. Cathode materials for Li-ion batteries can include lithium-containing transition metal oxides, such as, for example, $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ or $\text{LiNi}_x\text{Co}_y\text{Al}_z\text{O}_2$, where $x + y + z$ equals 1 (or about 1). These materials may contain a layered crystal structure where layers of lithium atoms sit between layers of transition-metal oxide polyhedra. However, alternative crystal structures can be formed as well, such as spinel type crystal structures. As deintercalation of Li-ions occurs from the crystal structure, charge neutrality is maintained with an increase in the valence state of the transition metals. $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ or $\text{LiNi}_x\text{Co}_y\text{Al}_z\text{O}_2$ possess desirable characteristics such as relatively high energy density (mA h/g), high cyclability (% degradation per charge/discharge cycle), and thermal stability ($\leq 100^\circ\text{C}$).

[0023] In some embodiments, the used solid feedstock may comprise end-of-life NMC or other used cathode materials from used LIBs or other sources. In some embodiments, the used solid feedstock may comprise a cathode composition, including but not limited to, LiCoO_2 (LCO), LiFePO_4 (LFP), LiMn_2O_4 (LMO), $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$ (NMC-111), $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ (NMC-532), $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ (NMC-622) or $\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$ (NMC-811), or $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$ (NCA). Most preferably, the used solid feedstock comprises a form of NMC. In some embodiments, the NMC comprises NMC-532 or an NMC having an average nickel to cobalt ratio of 5:2 or less. The starting used solid cathode precursor materials are not limiting.

[0024] Various characteristics of the final newly formed solid precursor lithium-containing particles, such as porosity, particle size, particle size distribution, phase composition and purity, microstructure, etc. can be tailored and controlled by fine tuning various process parameters and input materials. In some embodiments, these can include precursor solution chemistry, plasma gas flow rates, plasma process gas choice, residence time of the used precursor within the plasma, quenching rate, power density of the plasma, etc. These process parameters can be tailored, in some embodiments, to produce micron and/or sub-micron scale particles with tailored surface area, a specific porosity level, low-resistance Li-ion diffusion pathway, a span of less than about 2 (span = $d_{90}-d_{10}/d_{50}$) and containing a micro- or nano-grain microstructure. For example, desirable NMC material properties may include a layered α - NaFeO_2 – type crystal structure with a particle size distribution (PSD) d_{50} of about 8-13 μm with a primary grain size of about 0.5 -1 μm , a surface area of less than about 0.3 m^2/g and a tap density of greater than about 2.4 g/cm^3 . In some embodiments, when using powder feedstock, the size distribution may depend on the PSD of the input material.

[0025] FIG. 1 illustrates a flowchart of an example process for recycling a used solid feedstock using a microwave plasma process according to embodiments of the present disclosure.

[0026] In some embodiments, the used solid feedstock may undergo preprocessing steps prior to introducing the used solid feedstock to a microwave plasma apparatus. In some embodiments, this preprocessing may comprise lithium replacement and/or additional changes to the chemistry of the used solid feedstock. For example, the composition of the used solid feedstock may be changed by adding component powders, such as nickel containing, manganese containing, or cobalt containing powder, to the used solid feedstock prior to microwave processing. As such, the nickel content of the used solid feedstock may be augmented in the methods described herein. In some embodiments, preprocessing may also include additional washing to remove residual electrolytes, carbon and/or contamination. Preprocessing may also include milling to break feedstock particles into the primary grains, then forming a slurry and spray drying the granules to form a solid dry powder to be fed into the plasma. Other preprocesses may include heat treatment to re-

introduce dissociated lithium back into the layered crystal structure. Also, preprocessing may include particle size classification.

[0027] In some embodiments, the used solid feedstock, which may preferably be preprocessed, is introduced to a microwave plasma environment of a microwave plasma apparatus. In some embodiments, the microwave plasma environment may comprise the exhaust or torch of the microwave plasma apparatus. In some embodiments, the microstructure of the used solid feedstock may comprise one or more imperfections, cracks, or fissures due to usage/power cycling of the used solid feedstock within a LIB. In some embodiments, introducing the used solid feedstock into the microwave plasma environment may melt the used solid feedstock. In some embodiments, melting may result in some lithium loss in the process. However, lithium may be supplemented in the final product to make up for this lithium loss. In some embodiments, during microwave plasma processing and subsequent cooling, the used solid feedstock may be reformed into electroactive material with a desired chemistry and desired crystallographic structure. Furthermore, the newly formed solid precursor may comprise a microstructure in which some or all of the one or more imperfections, cracks, or fissures are eliminated. Without being limited by theory, in some embodiments, when the used solid feedstock is melted within the microwave plasma environment and subsequently reformed with the desired chemistry, the microstructure is altered, and any cracks may be sealed or otherwise eliminated.

[0028] The used precursor material, either liquid or solid, can be introduced into a plasma for processing. U.S. Pat. Pub. No. 2018/0297122, US 8748785 B2, and US 9932673 B2 disclose certain processing techniques that can be used in the disclosed process, specifically for microwave plasma processing. Accordingly, U.S. Pat. Pub. No. 2018/0297122, US 8748785 B2, and US 9932673 B2 are incorporated by reference in its entirety and the techniques describes should be considered to be applicable to the used precursor feedstocks described herein. The plasma can include, for example, an axisymmetric microwave generated plasma and a substantially uniform temperature profile.

[0029] In some embodiments, rather than preprocessing the used solid feedstock by replacing lost lithium, lithium may be introduced into the microwave plasma simultaneously with the used solid feedstock. In some embodiments, introducing lithium

concurrently with the used solid feedstock may replace any lost lithium in the used solid feedstock upon formation of the newly formed solid precursor.

[0030] One advantage of the systems and methods herein is that breaking down of the used solid feedstock into its individual constituent elements is avoided. Rather, in some embodiments, the used solid feedstock comprises directly recycled cathode material without reducing the material to its constituent elements. In the case of NMC, the used NMC may not be reduced to elemental nickel, cobalt, and manganese. Instead, in some embodiments, the NMC may comprise a used solid feedstock to be directly introduced into a microwave plasma apparatus to form newly formed solid NMC precursor.

[0031] In some embodiments, the newly formed solid precursor (e.g., NMC) may have a different chemistry than the used solid feedstock. For example, the newly formed solid precursor may have a higher nickel content than the used solid feedstock. Specifically, in some embodiments, the used feedstock may comprise NMC-532, NMC-111, or a mixture of NMC powders having a nickel to cobalt ratio of 5:2 or less, and the newly formed solid precursor may comprise NMC-622, NMC-811, NMC-9.5.5 or another NMC powder having a nickel to cobalt ratio greater than 5:2. This is an advantage over existing processes, as the ability to change the chemistry of NMC powder is limited in an ordinary heating process, in which particles would undesirably sinter together in a crucible or furnace. As such, in previous processes, NMC would need to be reduced to its constituent elements and then re-synthesized with the desired chemistry. Using microwave plasma processing however, the chemistry of the NMC may be changed with direct recycling (i.e., without reduction to constituent elements) because of the extremely high temperature and particle interactions within the microwave plasma environment. As with the lithium replacement, the chemistry of the used solid feedstock may be altered by introducing elemental metal powders (e.g., nickel powder), metallic salts, and/or metal oxides (e.g., NiO) concurrently into the microwave plasma apparatus concurrently with the used solid feedstock.

[0032] In some embodiments, following the plasma processing, the final newly formed solid precursor, such as layered NMC crystal structures or NMC particles, are formed. Therefore, no post-processing is needed, such as calcining, which can save significant time in the production of the NMCs, such a layered NMC crystal structure.

[0033] In some embodiments, the methods described herein can be used to produce newly formed solid precursor lithium-containing materials, such as $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ (where $x \geq 0$, $y \geq 0$, $z \geq 0$, and $x + y + z = 1$). For example, $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ (NMC-532), $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ (NMC-622), or $\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$ (NMC-811) can be produced by supplementing the used solid feedstock with different proportions of lithium, nickel, manganese, and cobalt.

[0034] Some advantages of the disclosed embodiments include the ability to tailor the solid precursor chemistry and final particle morphology. Use of the plasma system also enables the use of precursor materials (i.e., NMC powder) that are impractical or impossible to directly utilize in conventional recycling operations without breaking the material down into constituent elements. The process also allows the incorporation of additional Li-content at the nano, micro, or molecular scale (in some embodiments more than one) in the used solid feedstock.

[0035] For example, newly formed NMCs formed from embodiments of the disclosure can exhibit novel morphological characteristics not seen in traditionally made NMCs. These morphological characteristics include dense/non-porous particles for maximum energy density, network porosity to enable fast ion transport in the liquid phase for high power applications, and engineered particle size and surface produced in a single processing step or with an additional calcination step.

[0036] In some embodiments, the network porosity of the NMCs can range from 0-50% (or from about 0 to about 50%), with an absence of network porosity being most desirable. The particle size can be, for example, between 1 – 50 microns (or between about 1 – about 50 microns). Additionally, a composition at the surface of the NMCs can be made different either in terms of the ratios of the primary constituents (Ni, Mn, and Co) or can be a different material entirely. For example, alumina can be used to passivate the surface.

[0037] Embodiments of the disclosed methodology also can give precise control over particle size and particle size distribution, which can be used to maximize particle packing for improved energy density. Engineered interconnected internal porosity can be created with the proper selection of used solid feedstock and process conditions, allowing electrolyte access to the interior, and thus decreasing max solid-state diffusion distances, and increasing rate capability.

[0038] Moreover, NMCs formed by embodiments of the disclosure may also exhibit well controlled size and size distribution, of what is known in the industry as secondary grain size, ranging from 1 – 150 microns (or about 1 – about 150 microns) +/- 10% (or +/- about 10%).

[0039] In some embodiments, the size distribution of the newly formed solid precursor can be a d50 of 5 – 15 μ m (or about 5 – about 15 μ m). In some embodiments, the particles can have d10 of 2 μ m (or about 2 μ m) and a d90 of 25 μ m (or about 25 μ m). However, other distributions may be advantageous for specific applications. For example, larger particles, though still in the range of <50 μ m d50 (or <about 50 μ m) can be advantageous for very low power energy storage applications. Further, smaller particles, such as 2-5 μ m d50 (or about 2 – about 5 μ m) or 0.5-5 μ m d50 (or about 0.5 – about 5 μ m) can be advantageous for very high-power applications.

[0040] Additionally, the primary grain size for the NMCs can be modified to be from 10 nm -10 microns (or about 10 nm – about 10 microns). In some embodiments, the primary grain size may be between 100 nm and 10 microns (or between about 100 nm and about 10 microns). In some embodiments, the primary grain size may be between 50 nm and 500nm (or between about 50 nm and about 500 nm). In some embodiments, the primary grain size may be between 100 nm and 500nm (or between about 100 nm and about 500 nm).

[0041] The surface area of the newly formed solid precursor material can be controlled by both material porosity and particle size distribution. For example, assuming an identical particle size distribution, an increase in either surface or network porosity leads to an increase in surface area. Similarly, when keeping the level of porosity identical, smaller particles will yield a higher surface area. The surface area of newly formed solid precursor material can be tuned within a range of 0.01 – 15 m²/g (or about 0.1 – about 15 m²/g). In some embodiments, the surface area of newly formed solid precursor material can be tuned within a range of 0.01 – 15 m²/g (or about 0.01 – about 15 m²/g). Further, the final particle size can be approximately: d50 of 5-15 μ m; d10 of 1-2 μ m; d90 of 25-40 μ m. In some embodiments, the d50 can be 2-5 microns (or about 2 microns – about 5 microns). In some embodiments, the d50 can be 0.5-5 microns (or about 0.5 microns – about 5 microns). Porosity can be modified to tailor the surface area within the desired range. In some embodiments, for NMC materials, low surface area is desired. As such, in some

embodiments, process conditions may be altered to achieve a small-surface area NMC material.

Microwave Plasma Apparatus

[0042] FIG. 2 illustrates an embodiment of a top feeding microwave plasma torch 2 that can be used in the production of recycled solid LIB precursors, according to embodiments of the present disclosure. In some embodiments, feed materials 9, 10 can be introduced into a microwave plasma torch 3, which sustains a microwave-generated plasma 11. In one example embodiment, an entrainment gas flow and a sheath, swirl, or work linear flow (downward arrows) may be injected through inlets 5 to create flow conditions within the plasma torch prior to ignition of the plasma 11 via microwave radiation source 1. The feed materials 9 are introduced axially into the microwave plasma torch 2, where they are entrained by a gas flow that directs the materials toward a hot zone 6 and the plasma 11. The gas flows can consist of a noble gas column of the periodic table, such as helium, neon, argon, etc.

[0043] Within the microwave-generated plasma, the feed materials are melted in order to repair any cracks, fissures, or imperfections in the materials. Inlets 5 can be used to introduce process gases to entrain and accelerate particles 9, 10 along axis 12 towards plasma 11. First, particles 9 are accelerated by entrainment using a core laminar or turbulent gas flow (upper set of arrows) created through an annular gap within the plasma torch. A second laminar flow (lower set of arrows) can be created through a second annular gap to provide laminar sheathing for the inside wall of dielectric torch 3 to protect it from melting due to heat radiation from plasma 11. In exemplary embodiments, the laminar flows direct particles 9, 10 toward the plasma 11 along a path as close as possible to axis 12, exposing them to a temperature within the plasma. In some embodiments, suitable flow conditions are present to keep particles 10 from reaching the inner wall of the plasma torch 3 where plasma attachment could take place. Particles 9, 10 are guided by the gas flows towards microwave plasma 11 where each undergoes thermal treatment.

[0044] Various parameters of the microwave-generated plasma, as well as particle parameters, may be adjusted in order to achieve desired results. These parameters may include microwave power, feed material size, feed material insertion rate, gas flow rates,

plasma temperature, residence time, plasma gas composition, and cooling rates. As discussed above, in this particular embodiment, the gas flows are laminar; however, in alternative embodiments, swirl flows or turbulent flows may be used to direct the feed materials toward the plasma.

[0045] FIGS. 3A-3B illustrate embodiments of a microwave plasma torch that can be used in the production of recycled solid LIB precursors, according to a side feeding hopper embodiment of the present disclosure. Thus, in this implementation, the feedstock is injected after the microwave plasma torch applicator for processing in the “plume” or “exhaust” of the microwave plasma torch. Thus, the plasma of the microwave plasma torch is engaged at the exit end of the plasma torch to allow downstream feeding of the feedstock, as opposed to the top-feeding (or upstream feeding) discussed with respect to FIG. 2. This downstream feeding can advantageously extend the lifetime of the torch as the hot zone is preserved indefinitely from any material deposits on the walls of the hot zone liner. Furthermore, it allows engaging the plasma plume downstream at temperature suitable for optimal melting of powders through precise targeting of temperature level and residence time. For example, there is the ability to dial the length of the plume using microwave power, gas flows, torch type, plasma gas composition, and pressure in the quenching vessel that contains the plasma plume.

[0046] Generally, the downstream processing method can utilize two main hardware configurations to establish a stable plasma plume which are: annular torch, such as described in U.S. Pat. Pub. No. 2018/0297122, or swirl torches described in US 8748785 B2 and US 9932673 B2, each of which is hereby incorporated by reference in its entirety. Both FIG. 3A and FIG. 3B show embodiments of a method that can be implemented with either an annular torch or a swirl torch. A feed system close-coupled with the plasma plume at the exit of the plasma torch is used to feed powder axisymmetrically to preserve process homogeneity. Other feeding configurations may include one or several individual feeding nozzles surrounding the plasma plume.

[0047] The feed materials 314 can be introduced into a microwave plasma torch 302. A hopper 306 can be used to store the feed material 314 before feeding the feed material 314 into the microwave plasma torch 302, plume, or exhaust. In alternative embodiments, the feedstock can be injected along the longitudinal axis of the plasma torch.

The microwave radiation can be brought into the plasma torch through a waveguide 304. The feed material 314 is fed into a plasma chamber 310 and is placed into contact with the plasma generated by the plasma torch 302. When in contact with the plasma, plasma plume, or plasma exhaust, the feed material melts or is otherwise altered physically or chemically. While still in the plasma chamber 310, the feed material 314 cools and solidifies before being collected into a container 312. Alternatively, the feed material 314 can exit the plasma chamber 310 while still in a melted phase and cool and solidify outside the plasma chamber. In some embodiments, a quenching chamber may be used, which may or may not use positive pressure. While described separately from FIG. 2, the embodiments of FIGS. 3A-3B are understood to use similar features and conditions to the embodiment of FIG. 2.

Additional Embodiments

[0048] In the foregoing specification, the invention has been described with reference to specific embodiments thereof. It will, however, be evident that various modifications and changes may be made thereto without departing from the broader spirit and scope of the invention. The specification and drawings are, accordingly, to be regarded in an illustrative rather than restrictive sense.

[0049] Indeed, although this invention has been disclosed in the context of certain embodiments and examples, it will be understood by those skilled in the art that the invention extends beyond the specifically disclosed embodiments to other alternative embodiments and/or uses of the invention and obvious modifications and equivalents thereof. In addition, while several variations of the embodiments of the invention have been shown and described in detail, other modifications, which are within the scope of this invention, will be readily apparent to those of skill in the art based upon this disclosure. It is also contemplated that various combinations or sub-combinations of the specific features and aspects of the embodiments may be made and still fall within the scope of the invention. It should be understood that various features and aspects of the disclosed embodiments can be combined with, or substituted for, one another in order to form varying modes of the embodiments of the disclosed invention. Any methods disclosed herein need not be performed in the order recited. Thus, it is intended that the scope of the invention herein disclosed should not be limited by the particular embodiments described above.

[0050] It will be appreciated that the systems and methods of the disclosure each have several innovative aspects, no single one of which is solely responsible or required for the desirable attributes disclosed herein. The various features and processes described above may be used independently of one another or may be combined in various ways. All possible combinations and subcombinations are intended to fall within the scope of this disclosure.

[0051] Certain features that are described in this specification in the context of separate embodiments also may be implemented in combination in a single embodiment. Conversely, various features that are described in the context of a single embodiment also may be implemented in multiple embodiments separately or in any suitable subcombination. Moreover, although features may be described above as acting in certain combinations and even initially claimed as such, one or more features from a claimed combination may in some cases be excised from the combination, and the claimed combination may be directed to a subcombination or variation of a subcombination. No single feature or group of features is necessary or indispensable to each and every embodiment.

[0052] It will also be appreciated that conditional language used herein, such as, among others, “can,” “could,” “might,” “may,” “e.g.,” and the like, unless specifically stated otherwise, or otherwise understood within the context as used, is generally intended to convey that certain embodiments include, while other embodiments do not include, certain features, elements and/or steps. Thus, such conditional language is not generally intended to imply that features, elements and/or steps are in any way required for one or more embodiments or that one or more embodiments necessarily include logic for deciding, with or without author input or prompting, whether these features, elements and/or steps are included or are to be performed in any particular embodiment. The terms “comprising,” “including,” “having,” and the like are synonymous and are used inclusively, in an open-ended fashion, and do not exclude additional elements, features, acts, operations, and so forth. In addition, the term “or” is used in its inclusive sense (and not in its exclusive sense) so that when used, for example, to connect a list of elements, the term “or” means one, some, or all of the elements in the list. In addition, the articles “a,” “an,” and “the” as used in this application and the appended claims are to be construed to mean “one or more” or “at least one” unless specified otherwise. Similarly, while operations may be depicted in the drawings in a particular order, it is to be recognized that such operations need not be performed in the

particular order shown or in sequential order, or that all illustrated operations be performed, to achieve desirable results. Further, the drawings may schematically depict one more example processes in the form of a flowchart. However, other operations that are not depicted may be incorporated in the example methods and processes that are schematically illustrated. For example, one or more additional operations may be performed before, after, simultaneously, or between any of the illustrated operations. Additionally, the operations may be rearranged or reordered in other embodiments. In certain circumstances, multitasking and parallel processing may be advantageous. Moreover, the separation of various system components in the embodiments described above should not be understood as requiring such separation in all embodiments, and it should be understood that the described program components and systems may generally be integrated together in a single software product or packaged into multiple software products. Additionally, other embodiments are within the scope of the following claims. In some cases, the actions recited in the claims may be performed in a different order and still achieve desirable results.

[0053] Further, while the methods and devices described herein may be susceptible to various modifications and alternative forms, specific examples thereof have been shown in the drawings and are herein described in detail. It should be understood, however, that the invention is not to be limited to the particular forms or methods disclosed, but, to the contrary, the invention is to cover all modifications, equivalents, and alternatives falling within the spirit and scope of the various implementations described and the appended claims. Further, the disclosure herein of any particular feature, aspect, method, property, characteristic, quality, attribute, element, or the like in connection with an implementation or embodiment can be used in all other implementations or embodiments set forth herein. Any methods disclosed herein need not be performed in the order recited. The methods disclosed herein may include certain actions taken by a practitioner; however, the methods can also include any third-party instruction of those actions, either expressly or by implication. The ranges disclosed herein also encompass any and all overlap, sub-ranges, and combinations thereof. Language such as “up to,” “at least,” “greater than,” “less than,” “between,” and the like includes the number recited. Numbers preceded by a term such as “about” or “approximately” include the recited numbers and should be interpreted based on the circumstances (e.g., as accurate as reasonably possible under the circumstances, for example

$\pm 5\%$, $\pm 10\%$, $\pm 15\%$, etc.). For example, “about 3.5 mm” includes “3.5 mm.” Phrases preceded by a term such as “substantially” include the recited phrase and should be interpreted based on the circumstances (e.g., as much as reasonably possible under the circumstances). For example, “substantially constant” includes “constant.” Unless stated otherwise, all measurements are at standard conditions including temperature and pressure.

[0054] As used herein, a phrase referring to “at least one of” a list of items refers to any combination of those items, including single members. As an example, “at least one of: A, B, or C” is intended to cover: A, B, C, A and B, A and C, B and C, and A, B, and C. Conjunctive language such as the phrase “at least one of X, Y and Z,” unless specifically stated otherwise, is otherwise understood with the context as used in general to convey that an item, term, etc. may be at least one of X, Y or Z. Thus, such conjunctive language is not generally intended to imply that certain embodiments require at least one of X, at least one of Y, and at least one of Z to each be present. The headings provided herein, if any, are for convenience only and do not necessarily affect the scope or meaning of the devices and methods disclosed herein.

[0055] Accordingly, the claims are not intended to be limited to the embodiments shown herein but are to be accorded the widest scope consistent with this disclosure, the principles and the novel features disclosed herein.

WHAT IS CLAIMED IS:

1. A method for synthesizing lithium nickel manganese cobalt oxide (NMC) powder in a microwave plasma apparatus, the method comprising:
providing a feedstock to the microwave plasma apparatus, the feedstock comprising end-of-life NMC powder, the end-of-life NMC powder having an average nickel to cobalt ratio of 5:2 or less; and
introducing the feedstock into a microwave-generated plasma of the microwave plasma apparatus to synthesize an NMC powder having an average nickel to cobalt ratio greater than 5:2.
2. The method of Claim 1, further comprising introducing nickel containing material, manganese containing material, or cobalt containing material into the microwave-generated plasma concurrently with introducing the feedstock into the microwave-generated plasma.
3. The method of Claim 1, wherein a microstructure of the end-of-life NMC powder comprises one or more imperfections, cracks, or fissures, and wherein introducing the feedstock into the microwave-generated plasma melts the end-of-life NMC powder.
4. The method of Claim 3, wherein a microstructure of the synthesized NMC powder does not comprise the one or more imperfections, cracks, or fissures.
5. The method of Claim 1, further comprising introducing lithium (Li) containing material into the microwave-generated plasma concurrently introducing the feedstock into the microwave-generated plasma.
6. The method of Claim 1, wherein the end-of-life NMC powder comprises NMC-532 or NMC-111.
7. The method of Claim 1, further comprising adding lithium (Li) containing material to the feedstock prior to introducing the feedstock into the microwave-generated plasma.
8. The method of Claim 1, wherein the end-of-life NMC powder is obtained from a used lithium-ion battery.
9. A method for synthesizing lithium nickel manganese cobalt oxide (NMC) powder in a microwave plasma apparatus, the method comprising:

providing a feedstock to the microwave plasma apparatus, the feedstock comprising end-of-life NMC powder, the end-of-life NMC powder and having an average nickel to cobalt ratio of 5:2 or less; and

introducing the end-of-life NMC powder into a microwave-generated plasma of the microwave plasma apparatus to synthesize an NMC powder,

wherein the end-of-life NMC powder is not reduced to its constituent elements prior to introducing the end-of-life NMC powder into the microwave-generated plasma.

10. The method of Claim 9, further comprising introducing nickel containing material into the microwave-generated plasma concurrently with introducing the feedstock into the microwave-generated plasma.

11. The method of Claim 9, wherein a microstructure of the end-of-life NMC powder comprises one or more imperfections, cracks, or fissures, and wherein introducing the feedstock into the microwave-generated plasma melts the end-of-life NMC powder.

12. The method of Claim 11, wherein a microstructure of the synthesized NMC powder does not comprise the one or more imperfections, cracks, or fissures.

13. The method of Claim 9, further comprising introducing lithium (Li) containing material into the microwave-generated plasma concurrently introducing the feedstock into the microwave-generated plasma.

14. The method of Claim 9, wherein the end-of-life NMC powder comprises NMC-532 or NMC-111.

15. The method of Claim 9, further comprising adding lithium (Li) containing material to the feedstock prior to introducing the feedstock into the microwave-generated plasma.

16. The method of Claim 9, wherein the end-of-life NMC powder is obtained from a used lithium-ion battery.

17. A lithium nickel manganese cobalt oxide (NMC) powder produced by a method comprising:

providing a feedstock to a microwave plasma apparatus, the feedstock comprising end-of-life NMC powder, the end-of-life NMC powder having an average nickel to cobalt ratio of 5:2 or less; and

introducing the feedstock into a microwave-generated plasma of the microwave plasma apparatus to synthesize an NMC powder or an NMC precursor having an average nickel to cobalt ratio greater than 5:2.

18. The NMC powder of Claim 17, wherein the end-of-life NMC powder comprises NMC-111, NMC-442, or NMC-532

19. The NMC powder of Claim 17, wherein the NMC powder or the NMC precursor comprises NMC-611, NMC-811, or NMC-9.5.5.

20. The NMC powder of Claim 17, wherein the NMC powder or the NMC precursor has an average nickel to cobalt ratio of 5:2, 6:1, 8:1, or 18:1.