

CLAIMS

What is claimed is:

1. A process for making a high density green tape, the process comprising:
 - (a) providing a slurry comprising a source powder;
 - (b) mixing the slurry with a binder solution in a non-reactive environment;
 - (c) casting the slurry to form a green tape in a non-reactive environment; and
 - (d) drying the green tape in a non-reactive environment to achieve a geometric density greater than 2.9 g/ml.
2. The process of claim 1, wherein the source powder is calcined in a non-reactive environment to achieve geometric density greater than 4.7 g/ml.
3. The process of claim 1, wherein the amount of source powder in the green tape is at least 50%, 55%, 60%, 65%, 70%, 75%, 80%, 85%, or 90% by weight.
4. The process of any of claims 1 to 3, wherein the at least one source powder is selected from the group consisting of lithium-stuffed garnet, chemical precursors to lithium-stuffed garnet, and lithium-stuffed garnet with aluminum oxide dopants.
5. The process claim 4, wherein the lithium-stuffed garnet is a material selected from the group consisting of: $\text{Li}_A\text{La}_B\text{M}'_C\text{M}''_D\text{ZrE}_F\text{O}_F$, wherein $4 < A < 8.5$, $1.5 < B < 4$, $0 \leq C \leq 2$, $0 \leq D \leq 2$, $0 \leq E < 2.5$, $10 < F \leq 13.5$, and M' and M'' are each, independently in each instance selected from Al, Mo, W, Nb, Sb, Ca, Ba, Sr, Ce, Hf, Rb, Ga, and Ta.
6. The process of any of claims 1 to 5, wherein at least one source has a particle size distribution d_{50} of about 100 nm - about 200 nm, about 200 nm - about 300 nm, about 300 nm - about 400 nm, about 400 nm - about 500 nm, about 500 nm - about 600 nm, about 600 nm - about 700 nm, about 700 nm - about 800 nm, about 800 nm - about 900 nm, about 900 nm - about 1 μm , about 1 μm - about 2 μm , or about 2 μm - about 3 μm .

7. The process of any one of claims 1 to 6, wherein the process further comprises milling at least one source powder in a non-reactive environment in an anhydrous aprotic solvent.
8. The process of any one of claims 1 to 7, wherein the non-reactive environment comprises nitrogen gas or argon gas, or a combination thereof, and a humidity at about -10°C to -20°C, at about -20°C to -30°C, at about -30°C to -40°C, at about -40°C to -50°C, or at about -50°C to -60 °C dew point.
9. The process of any one of claims 7 to 8, wherein the aprotic solvent is selected from the group consisting of: benzene, toluene, xylene, ethyl acetate, tetrahydrofuran, dioxane, and 1,2-dimethoxyethane.
10. The process of any of claims 1-9, wherein the milling is selected from the group consisting of dry milling, attrition milling, sonication milling, high energy milling, wet milling, jet milling, and cryogenic milling.
11. The process of claim 10, further comprising milling the source powder until the source powder has a particle size distribution d_{50} of about 100 nm-200 nm, about 200 nm-300 nm, about 300 nm-400 nm, about 400 nm-500 nm, about 500 nm-600 nm, about 600 nm-700 nm, or about 700 nm-750nm.
12. The process of any one of claims 1 to 11, further comprising, prior to step (c) or step (d), mixing the slurry of the modified source powder in a non-reactive environment with a binder selected from the group consisting of polypropylene (PP), atactic polypropylene (aPP), isotactic polypropylene (iPP), ethylene propylene rubber (EPR), ethylene pentene copolymer (EPC), polyisobutylene (PIB), styrene butadiene rubber (SBR), poly(ethylene-co-1-octene) (PE-co-PO), poly(ethylene-co-methylene cyclopentene) (PE-co-PMCP), stereoblock polypropylenes, polypropylene polymethyl pentene, polyethylene oxide (PEO), PEO block copolymers, silicone polymers and copolymers, polyvinyl butyral (PVB), poly(vinyl acetate) (PVAc), polyvinylpyrrolidone (PVP), poly(ethyl methacrylate) (PEMA), acrylic polymers, binders from the Paraloid family of resins, binders from the

Butvar family of resins, binders from the Mowital family of resins, and combinations thereof.

13. The process of any one of claims 1 to 12, further comprising, in step (b), milling the slurry of the modified source powder, with a dispersant selected from the group consisting of fish oil, fatty acids of degree C₈– C₂₀, alcohols of degree C₈– C₂₀, alkylamines of degree C₈– C₂₀, phosphate esters, phospholipids, polymeric dispersants such as poly(vinylpyridine), poly(ethylene imine), poly(ethylene oxide) and ethers thereof, poly(ethylene glycol) and ethers thereof, polyalkylene amine, polyacrylates, polymethacrylates, poly(vinyl alcohol), poly(vinyl acetate), polyvinyl butyral, maleic anhydride copolymers, glycolic acid ethoxylate lauryl ether, glycolic acid ethoxylate oleyl ether, sodium dodecyl sulfate, sodium dodecylbenzenesulfonate, cetyltrimethylammonium bromide, cetylpyridinium chloride, surfactants and dispersants from the Brij family of surfactants, the Triton family of surfactants, the Solsperse family of dispersants, the SMA family of dispersants, the Tween family of surfactants, and the Span family of surfactants.
14. The process of claim 13, wherein the fatty acids of degree C₈– C₂₀ are selected from dodecanoic acid, oleic acid, stearic acid, linolenic acid, and/or linoleic acid.
15. The process claim 13, wherein the alcohols of degree C₈– C₂₀ are at selected from dodecanol, oleyl alcohol, stearyl alcohol, and combinations thereof.
16. The process of claim 13, wherein the alkylamines of degree C₈– C₂₀ are selected from dodecylamine, oleylamine, stearylamine, and combinations thereof.
17. The process of claim 13, wherein the phospholipids are selected from phosphatidylcholine, lecithin, and combinations thereof.
18. The process of any one of claims 1 to 17, further comprising, prior to step (c) or step (d), mixing the slurry of the source powder in a non-reactive environment with a plasticizer selected from dibutyl phthalate, dioctyl phthalate, and benzyl butyl phthalate.

19. The process of any one of claims 1 to 18, wherein the filtration technique is selected from the group consisting of sieving, centrifugation, and separating particles of different size or different mass.
20. The process of any one of claims 1 to 19, wherein the slurry has a solids loading of 1 wt% to 99 wt% and wherein the solid loading refers to the amount of source powder.
21. The process of any one of claims 1 to 20, wherein the slurry when dried comprises the source powder at 80% wt/wt.
22. The process of any one of claim 1 to 21, wherein the slurry when dried comprises about 10-25% wt/wt organic content and wherein the organic content comprises slurry components other than the source powder.
23. The process of any one of claim 1 to 22, wherein the green tape comprises particles of a lithium-stuffed garnet.
24. The process of any one of claim 1 to 23, wherein the green tape has a density greater than 2.9 g/cm³ as measured by geometric density.
25. The process of any one of claims 1-24, further comprising sintering the green tape.
26. The process of any one of claims 1 to 7, wherein the non-reactive environment comprises atmospheric air at a humidity of about -10°C to -20°C, at about -20°C to -30°C, at about -30°C to -40°C, at about -40°C to -50°C, or at about -50°C to -60 °C dew point.
27. The process of any one of claims 1-26, wherein the non-reactive environment is the same non-reactive environment in each step.
28. The process of any one of claims 1-26, wherein the non-reactive environment is a different non-reactive environment in at least one step.