sintering of thin films, these films tend to crack, warp, or otherwise have surface deteriorations.

[0005] There is therefore a series of problems in the relevant field related to casting green tapes of ceramics, such as but not limited to garnets, and to sintering these green tapes to prepare high density garnet thin films. What is needed in the relevant field is, for example, improved materials and processes for casting green tapes with high density.

SUMMARY

[0006] The instant disclosure sets forth such materials and processes, in addition to making and using the same, and other solutions to problems in the relevant field.

[0007] There is provided a process for making a high density green tape is provided, the process comprising:

(a) providing a slurry comprising at least one 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 density >2.9 g/ml.

In certain examples, each non-reactive environment is unique with regard to temperature, pressure, or atmosphere composition. In certain examples, each non-reactive environment is the same non-reactive environment.

[0007a] In one aspect, there is provided 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,

wherein the non-reactive environment comprises nitrogen gas or argon gas, or a combination thereof, and a humidity at -10^{o}C to -20^{o}C, at -20^{o}C to

-30ºC, at -30ºC to -40ºC, at -40ºC to -50ºC, or at -50ºC to -60 ºC dew point,

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,

wherein at least one source has a particle size distribution d_{50} of 100 nm - 200 nm, 200 nm - 300 nm, 300 nm - 400 nm, 400 nm - 500 nm, 500 nm - 600 nm, 600 nm - 700 nm, 700 nm - 800 nm, 800 nm - 900 nm, 900 nm - 1 µm, 1 µm - 2 µm, or 2 µm - 3 µm,

wherein the process further comprises milling at least one source powder in an anhydrous aprotic solvent,

wherein the aprotic solvent is selected from the group consisting of: benzene, toluene, xylene, ethyl acetate, tetrahydrofuran, dioxane, and 1,2-dimethoxyethane; and

further comprising milling the source powder until the source powder has a particle size distribution d_{50} of 100 nm-200 nm, 200 nm-300 nm, 300 nm-400 nm, 400 nm-500 nm, 500 nm-600 nm, 600 nm-700 nm, or 700 nm-750nm.

[0008] In some examples, the at least one source powder is calcined in a non-reactive environment to achieve density >4.7 g/ml as measured by geometric density. In some examples, the amount of the at least one source powder in the green tape is at least 50%, 55%, 55%, 60%, 65%, 70%, 80%, 85%, or 90% by weight. In some examples, 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. In some examples, the lithium-stuffed garnet is a material selected from the group consisting of: $Li_ALa_BM'_cM''_DZr_EO_F$, 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. In some examples, the particle size d_{50} is about 100 nm-200 nm, about 200 nm-300 nm,

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,

   wherein the non-reactive environment comprises nitrogen gas or argon gas, or a combination thereof, and a humidity at -10^{o}C to -20^{o}C, at -20^{o}C to -30^{o}C, at -30^{o}C to -40^{o}C, at -40^{o}C to -50^{o}C, or at -50^{o}C to -60 ^{o}C dew point,

   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,

   wherein at least one source has a particle size distribution d_{50} of 100 nm - 200 nm, 200 nm - 300 nm, 300 nm - 400 nm, 400 nm - 500 nm, 500 nm - 600 nm, 600 nm - 700 nm, 700 nm - 800 nm, 800 nm - 900 nm, 900 nm - 1 µm, 1 µm - 2 µm, or 2 µm - 3 µm,

   wherein the process further comprises milling at least one source powder in an anhydrous aprotic solvent,

   wherein the aprotic solvent is selected from the group consisting of: benzene, toluene, xylene, ethyl acetate, tetrahydrofuran, dioxane, and 1,2-dimethoxyethane; and

   further comprising milling the source powder until the source powder has a particle size distribution d_{50} of 100 nm-200 nm, 200 nm-300 nm, 300 nm-400 nm, 400 nm-500 nm, 500 nm-600 nm, 600 nm-700 nm, or 700 nm-750nm.

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%, 55%, 60%, 65%, 70%, 75%, 80%, 85%, or 90% by weight.

4. The process of claim 1, wherein the lithium-stuffed garnet is a material selected from the group consisting of: $Li_ALa_BM'_cM''_DZr_EO_F$, 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.

5. The process of any one of claims 1 to 4, further comprising, prior to step (c) or step (d), mixing the slurry 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), polyvinylpyrrolidine (PVP), poly(ethyl methacrylate) (PEMA), acrylic polymers, binders from the acrylic copolymer family of resins, binders from the polyvinyl butyral family of resins, and combinations thereof.

6. The process of any one of claims 1 to 5, further comprising, in step (b), milling the slurry, with a dispersant selected from the group consisting of fish oil, fatty acids of degree C_8– C_{20}, alcohols of degree C_8– C_{20}, alkylamines of degree C_8– C_{20}, 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 polyoxyethylene fatty ether family of surfactants, the polymeric family of dispersants, the styrene maleic anhydride family of dispersants, the polysorbate family of surfactants, and the sorbitan oleate family of surfactants.

7. The process of claim 6, wherein the fatty acids of degree C_8– C_{20} are selected from dodecanoic acid, oleic acid, stearic acid, linolenic acid, and/or linoleic acid.

8. The process of claim 6, wherein the alcohols of degree C_8– C_{20} are selected from dodecanol, oleyl alcohol, stearyl alcohol, and combinations thereof.

9. The process of claim 6, wherein the alkylamines of degree C_8– C_{20} are selected from dodecylamine, oleylamine, stearylamine, and combinations thereof.

10. The process of claim 6, wherein the phospholipids are selected from phosphatidylcholine, lecithin, and combinations thereof.

11. The process of any one of claims 1 to 10, further comprising, prior to step (c) or step (d), mixing the slurry of the source powder with a plasticizer selected from dibutyl phthalate, dioctyl phthalate, and benzyl butyl phthalate.

12. The process of any one of claims 1 to 11, 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.

13. The process of any one of claims 1 to 12, wherein the slurry when dried comprises the source powder at 80% wt/wt.

14. The process of any one of claim 1 to 13, 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.

15. The process of any one of claim 1 to 14, wherein the green tape comprises particles of a lithium-stuffed garnet, or wherein the green tape has a density between 2.9 g/cm^3 and 3.2 g/cm^3 as measured by geometric density.

16. The process of any one of claim 1 to 14, wherein the green tape has a density between 3.0 g/cm^3 and 3.2 g/cm^3 as measured by geometric density.