CLAIMS

[Claim 1] A lead-free piezoelectric ceramic composition, comprising:

a main phase comprising a first crystal phase comprising an alkali niobate/tantalate type perovskite oxide having piezoelectric properties; and

a subphase comprising a second crystal phase comprising an M-Ti-O spinel compound (where the element M is a monovalent to quadrivalent element).

[Claim 2] The lead-free piezoelectric ceramic composition according to claim 1,

wherein the element M comprises at least one metallic element selected from the group consisting of Li, Mg, Al, Sc, Cr, Mn, Fe, Co, Ni, Zn, Ga, Y, and Zr.

[Claim 3] The lead-free piezoelectric ceramic composition according to claim 1 or 2,

wherein the M-Ti-O spinel compound is represented by a compositional formula: M_xTiO_y (where the coefficients x and y are relative values when a content of Ti is taken as 1), and

wherein the coefficient x satisfies $0.5 \leq x \leq 5.0$.

[Claim 4] The lead-free piezoelectric ceramic composition according to claim 3,

wherein the coefficient y satisfies $2 \leq y \leq 8$.

[Claim 5] The lead-free piezoelectric ceramic composition according to any one of claims 1 to 4,

wherein the subphase fills a hole formed in the main phase.

[Claim 6] The lead-free piezoelectric ceramic composition according to any one of claims 1 to 5,

wherein the contained rate of the second crystal phase in the lead-free piezoelectric ceramic composition is any one of:

(i) 0.5 vol% to 5.0 vol%;

(ii) 0.5 vol% to 2.5 vol%; and

(iii) 1.0 vol% to 2.0 vol%.

[Claim 7] The lead-free piezoelectric ceramic composition according to any one of claims 1 to 6,

wherein the M-Ti-O spinel compound comprises two or more metallic elements as the element M.

[Claim 8] The lead-free piezoelectric ceramic composition according to any one of claims 1 to 7,

wherein the subphase comprises a third crystal phase in addition to the second crystal phase, the third crystal phase comprising an $A_3B_5O_{15}$ type compound (where the element A is a monovalent to divalent metal, and the element B is a divalent to pentavalent metal).

[Claim 9] The lead-free piezoelectric ceramic composition according to any one of claims 1 to 8,

wherein a volume rate of the second crystal phase is 50% or more when the entire subphase is taken as 100%.

[Claim 10] The lead-free piezoelectric ceramic composition according to any one of claims 1 to 9,

wherein the alkali niobate/tantalate type perovskite oxide of the first crystal phase comprises an alkali earth metal.

[Claim 11] The lead-free piezoelectric ceramic composition according to claim 10,

wherein the alkali niobate/tantalate type perovskite oxide of the first crystal phase is represented by a compositional formula: $(K_aNa_bLi_cC_d)_e(D_fE_g)O_h$ (where the element C comprises one or more elements selected from the group consisting of Ca, Sr, and Ba, the element D comprises one or more elements selected from the group consisting of Nb, Ta, Ti, Zr, Hf, Sn, Sb, and Si while comprising at least Nb or Ta, and the element E comprises one or more elements selected from the group consisting of Mg, Al ,Sc ,Mn ,Fe ,Co ,Ni ,Zn ,Ga, and Y, and where a + b + c + d = 1, e is arbitrary, f + g = 1, and h is an arbitrary value that forms perovskite).

[Claim 12] The lead-free piezoelectric ceramic composition according to claim 11,

wherein the coefficient e satisfies $0.88 \leq e \leq 1.07$.

[Claim 13] The lead-free piezoelectric ceramic composition according to any one of claims 1 to 12,

wherein the alkali niobate/tantalate type perovskite oxide comprises an alkali niobate type perovskite oxide.

[Claim 14] A piezoelectric element, comprising:

a piezoelectric ceramic comprising the lead-free piezoelectric ceramic composition according to any one of claims 1 to 13; and

an electrode mounted on the piezoelectric ceramic.

[Claim 15] A device comprising the piezoelectric element according to claim 14.

[Claim 16] The device according to claim 15,

wherein the device comprises any one of a knock sensor, an ultrasonic transducer, a cutting tool, an ultrasonic sensor, and an actuator.

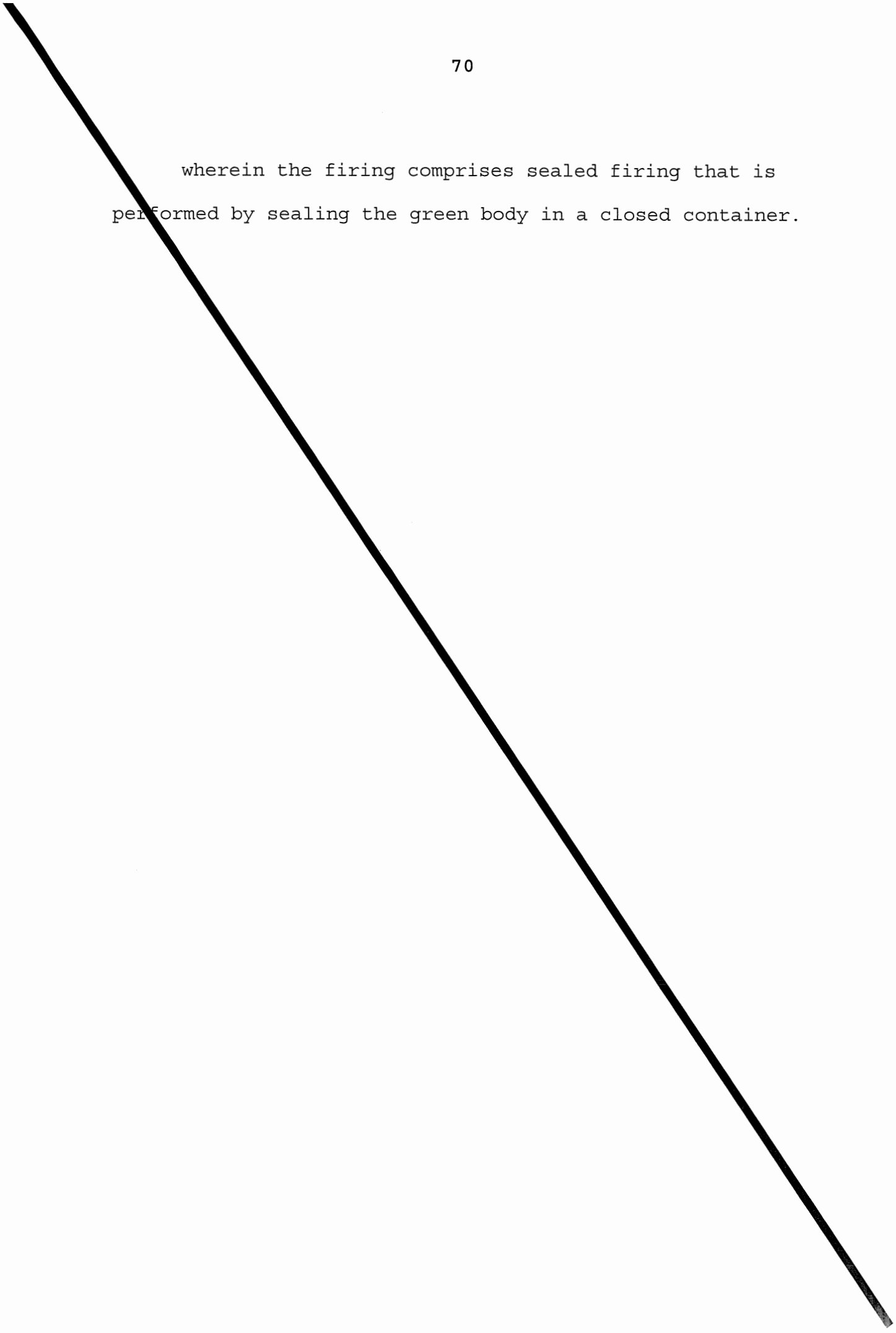
[Claim 17] A process for production of the lead-free piezoelectric ceramic composition according to any one of claims 1 to 13, the process comprising:

preparing a first powder by mixing and calcining raw materials of the first crystal phase;

preparing a second powder by mixing and calcining raw materials of the second crystal phase; and

producing the lead-free piezoelectric ceramic composition by mixing the first and second powders, molding the mixed powder material into a green body, and then firing the green body,

wherein the firing comprises sealed firing that is performed by sealing the green body in a closed container.



[Claim(s)]

[Claim 1]

A lead-free piezoelectric ceramic composition, comprising:

a main phase comprising a first crystal phase comprising an alkali niobate/tantalate type perovskite oxide having piezoelectric properties; and

a subphase comprising a second crystal phase comprising an M-Ti-O spinel compound (where the element M is a monovalent to quadrivalent element).

[Claim 2]

The lead-free piezoelectric ceramic composition according to claim 1,

wherein the element M comprises at least one metallic element selected from the group consisting of Li, Mg, Al, Sc, Cr, Mn, Fe, Co, Ni, Zn, Ga, Y, and Zr.

[Claim 3]

The lead-free piezoelectric ceramic composition according to claim 1 or 2,

wherein the M-Ti-O spinel compound is represented by a compositional formula: M_xTiO_y (where the coefficients x and y are relative values when a content of Ti is taken as 1), and

wherein the coefficient x satisfies $0.5 \leq x \leq 5.0$.

[Claim 4]

The lead-free piezoelectric ceramic composition according to claim 3,

wherein the coefficient y satisfies $2 \leq y \leq 8$.

[Claim 5]

The lead-free piezoelectric ceramic composition according to any one of claims 1 to 4,

wherein the subphase fills a hole formed in the main phase.

[Claim 6]

The lead-free piezoelectric ceramic composition according to any one of claims 1 to 5,

wherein the contained rate of the second crystal phase in the lead-free piezoelectric ceramic composition is 0.5 vol% to 5.0 vol%.

[Claim 7]

The lead-free piezoelectric ceramic composition according to any one of claims 1 to 6,

wherein the M-Ti-O spinel compound comprises two or more metallic elements as the element M.

[Claim 8]

The lead-free piezoelectric ceramic composition according to any one of claims 1 to 7,

wherein the subphase comprises a third crystal phase in addition to the second crystal phase, the third crystal phase

comprising an $A_3B_5O_{15}$ type compound (where the element A is a monovalent to divalent metal, and the element B is a divalent to pentavalent metal).

[Claim 9]

The lead-free piezoelectric ceramic composition according to any one of claims 1 to 8,

wherein a volume rate of the second crystal phase is 50% or more when the entire subphase is taken as 100%.

[Claim 10]

The lead-free piezoelectric ceramic composition according to any one of claims 1 to 9,

wherein the alkali niobate/tantalate type perovskite oxide of the first crystal phase comprises an alkali earth metal.

[Claim 11]

The lead-free piezoelectric ceramic composition according to claim 10,

wherein the alkali niobate/tantalate type perovskite oxide of the first crystal phase is represented by a compositional formula: $(K_aNa_bLi_cC_d)_e(D_fE_g)O_h$ (where the element C comprises one or more elements selected from the group consisting of Ca, Sr, and Ba, the element D comprises one or more elements selected from the group consisting of Nb, Ta, Ti, Zr, Hf, Sn, Sb, and Si while comprising at least Nb or Ta, and the element E comprises one or more elements selected from

the group consisting of Mg, Al ,Sc ,Mn ,Fe ,Co ,Ni ,Zn ,Ga, and Y, and where a + b + c + d = 1, a + b + c is not zero, e satisfies $0.80 \leq e \leq 1.10$, f + g = 1, and h is an arbitrary value that forms perovskite).

[Claim 12]

The lead-free piezoelectric ceramic composition according to claim 11,

wherein the coefficient e satisfies $0.88 \leq e \leq 1.07$.

[Claim 13]

The lead-free piezoelectric ceramic composition according to any one of claims 1 to 12,

wherein the alkali niobate/tantalate type perovskite oxide comprises an alkali niobate type perovskite oxide.

[Claim 14]

A piezoelectric element, comprising:

a piezoelectric ceramic comprising the lead-free piezoelectric ceramic composition according to any one of claims 1 to 13; and

an electrode mounted on the piezoelectric ceramic.

[Claim 15]

A device comprising the piezoelectric element according to claim 14.

[Claim 16]

The device according to claim 15,

wherein the device comprises any one of a knock sensor, an ultrasonic transducer, a cutting tool, an ultrasonic sensor, and an actuator.

[Claim 17]

A process for production of the lead-free piezoelectric ceramic composition according to any one of claims 1 to 13, the process comprising:

preparing a first powder by mixing and calcining raw materials of the first crystal phase;

preparing a second powder by mixing and calcining raw materials of the second crystal phase; and

producing the lead-free piezoelectric ceramic composition by mixing the first and second powders, molding the mixed powder material into a green body, and then firing the green body,

wherein the firing comprises sealed firing that is performed by sealing the green body in a closed container.