

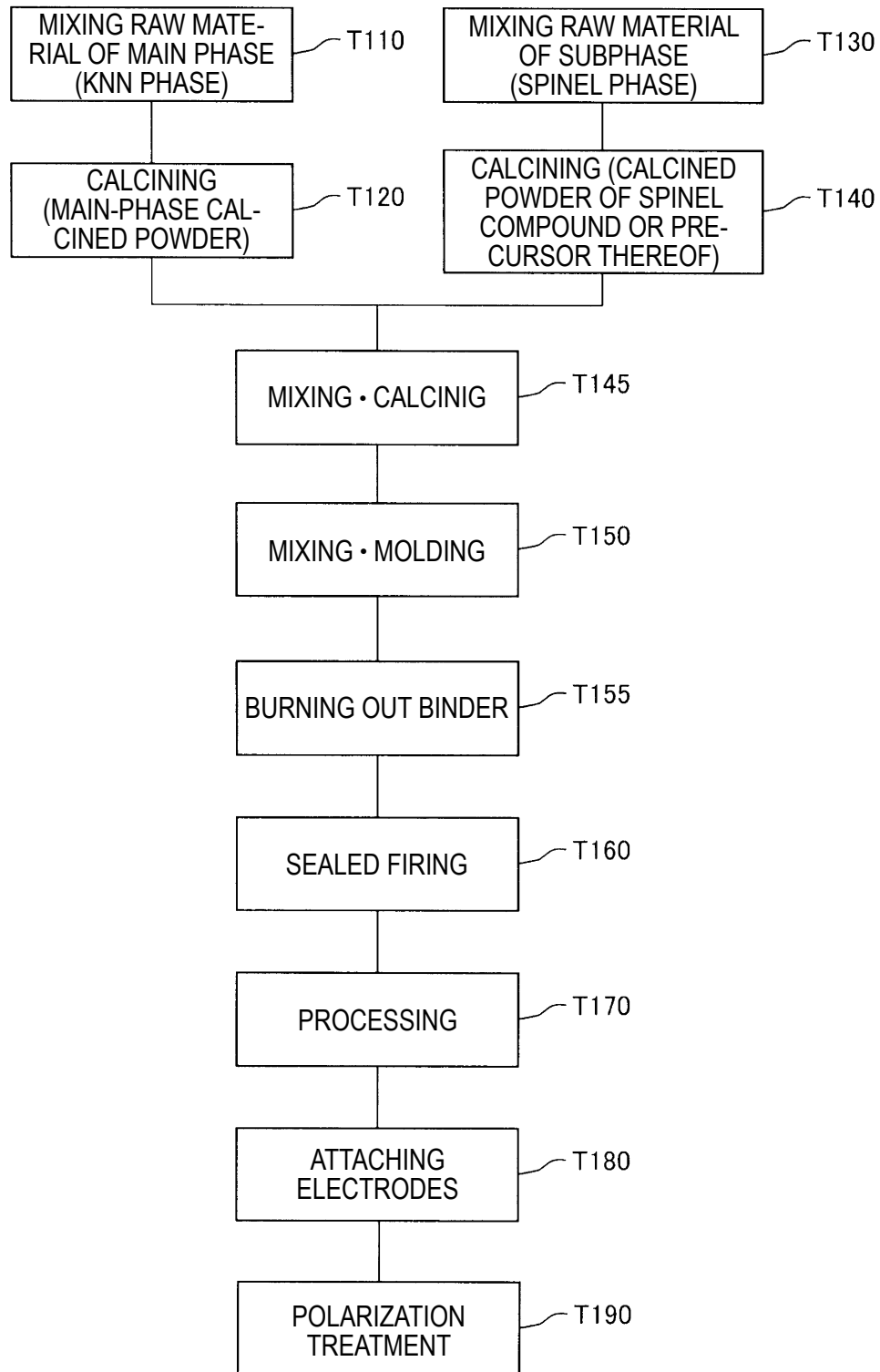
FIG. 1

FIG. 2

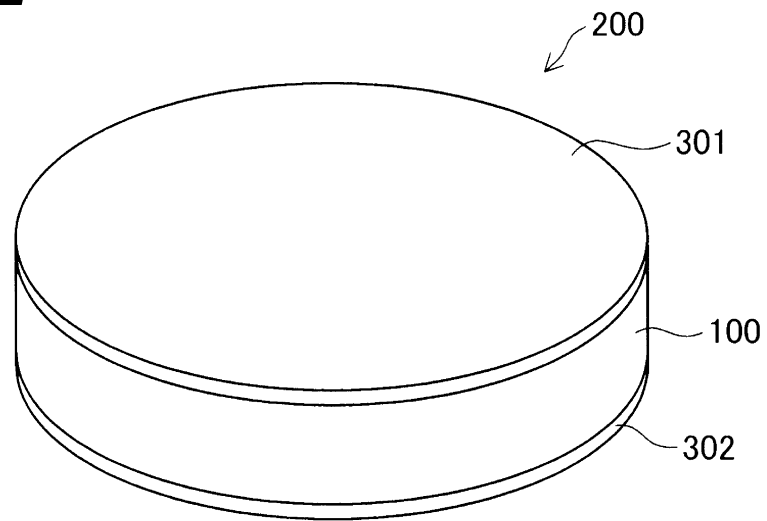


FIG. 3

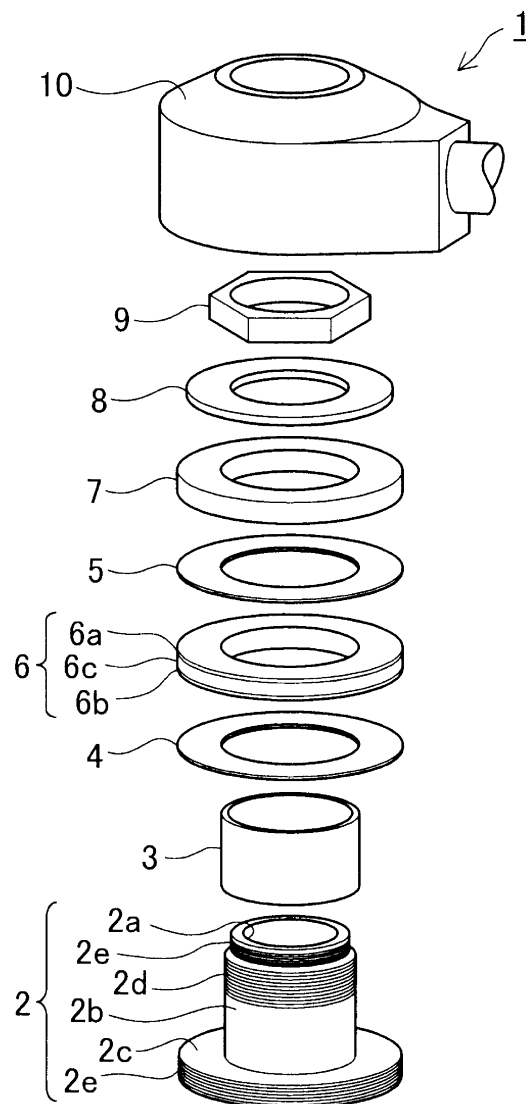


FIG. 4

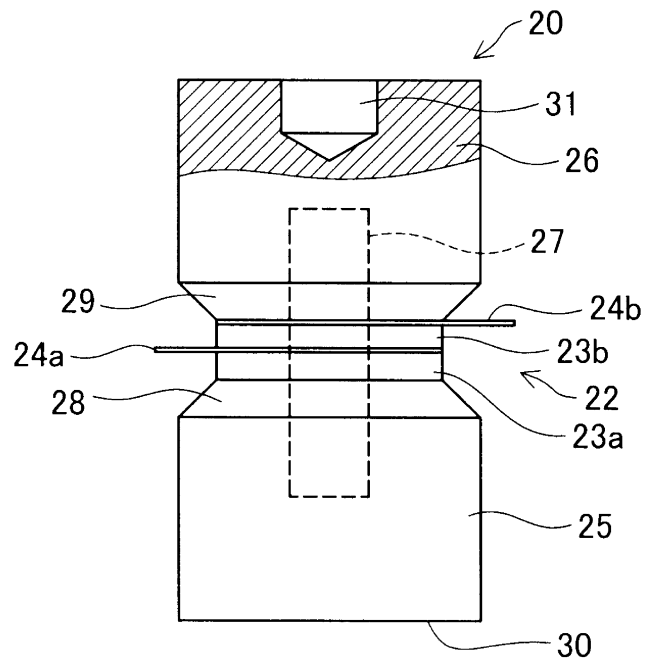


FIG. 5

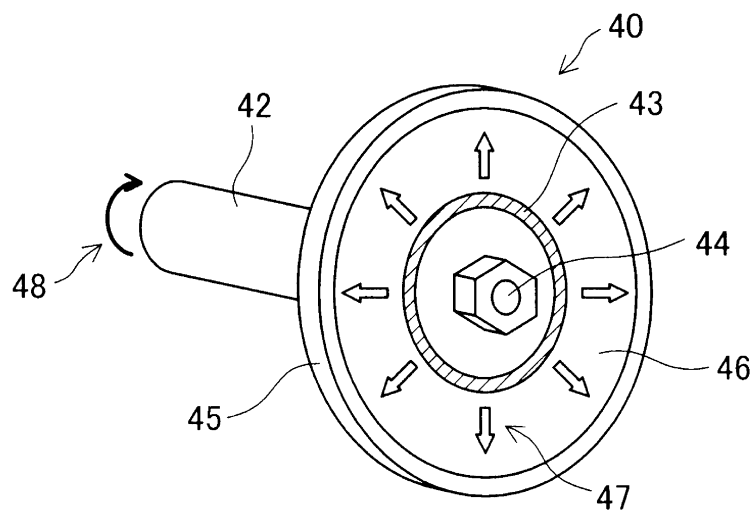


FIG. 6

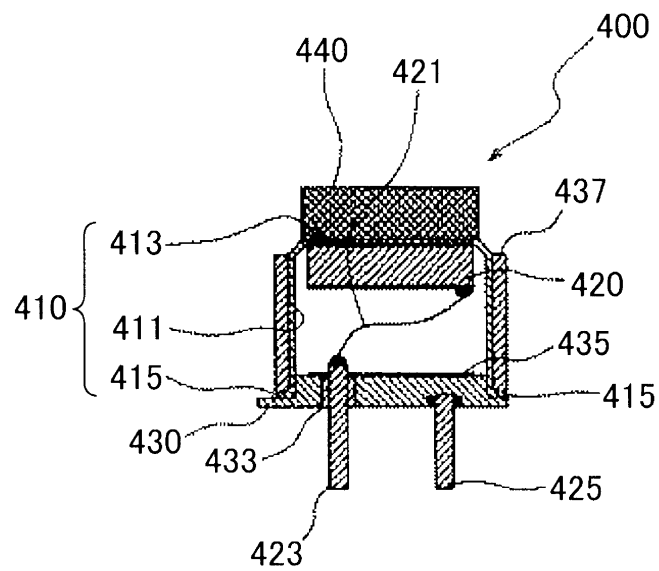
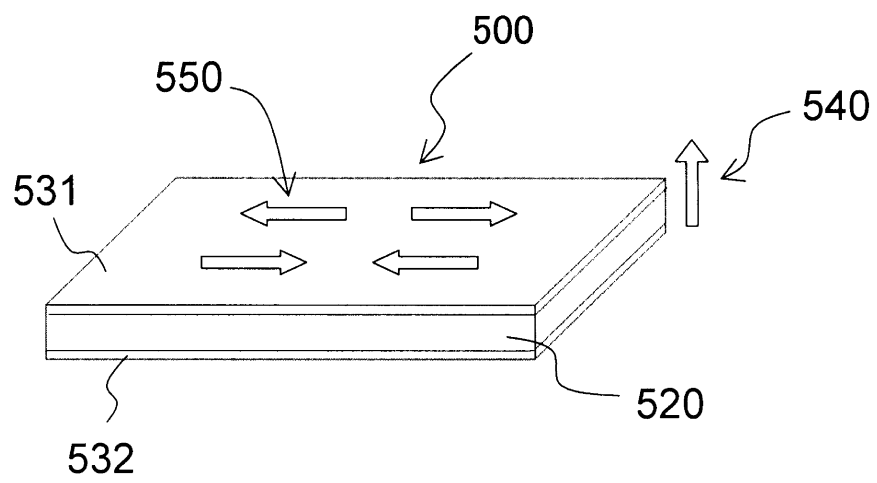


FIG. 7

Influence of subphase rate on piezoelectric properties

Sample	First crystal phase (Main phase)													Second crystal phase (Subphase)			
	Element C	Element D	Element E	a	b	c	Coefficient d of element C		e	Coefficient f of element D			Coefficient g of element E			Subphase rate (Vol%)	Element M
							d1	d2		f1	f2	f3	g1	g2	g3		
S01(*)	-	Nb	-	0.500	0.500	0	0	0	1.00	1.000	0	0	0	0	0	-	
S02(*)	-	Nb	-	0.490	0.490	0.020	0	0	1.00	1.000	0	0	0	0	0	-	
S03	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S04	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S05	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S06	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S07	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S08	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S09	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S10	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S11	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S12	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	

First crystal phase (Main phase): $(K_aNa_bLi_cCd)_e(DiFg)O_h$

(where the element C includes one or more elements selected from the group consisting of Ca, Sr, and Ba, the element D includes one or more elements selected from the group consisting of Nb, Ta, Ti, and Zr while including at least Nb or Ta, and the element E includes 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.)

Second crystal phase (Subphase): M_xTiO_y

(where the element M includes one or more elements selected from the group consisting of Li, Mg, Al, Sc, Cr, Mn, Fe, Co, Ni, Zn, Ga, Y, and Zr, and where the coefficients x and y are relative values with respect to the Ti content, $0.5 \leq x \leq 5.0$, and the coefficient y is an arbitrary value that forms spinel.)

(*) indicates a comparative example.

FIG. 8B

Influence of subphase rate on piezoelectric properties

Sample	First crystal phase (Main phase)	Second crystal phase (Subphase)			Relative permittivity	Piezoelectric constant	Electro- mechanical coupling coefficient	Room- temperature phase transition point
	Composition	Subphase rate (Vol%)	Element M	Composition				
S01(*)	FIG. 8A	0	-	-	430	90	0.30	Present
S02(*)	do.	0	-	-	430	95	0.31	Present
S03	do.	0.44	Co, Zn	CoZnTiO ₄	1470	91	0.21	Absent
S04	do.	0.80	Co, Zn	CoZnTiO ₄	1520	208	0.42	Absent
S05	do.	1.24	Co, Zn	CoZnTiO ₄	1554	255	0.46	Absent
S06	do.	1.33	Co, Zn	CoZnTiO ₄	1700	298	0.53	Absent
S07	do.	1.42	Co, Zn	CoZnTiO ₄	1761	288	0.52	Absent
S08	do.	1.90	Co, Zn	CoZnTiO ₄	1650	245	0.49	Absent
S09	do.	2.43	Co, Zn	CoZnTiO ₄	1600	211	0.41	Absent
S10	do.	3.54	Co, Zn	CoZnTiO ₄	1560	194	0.40	Absent
S11	do.	4.42	Co, Zn	CoZnTiO ₄	1530	190	0.37	Absent
S12	do.	5.30	Co, Zn	CoZnTiO ₄	1500	122	0.24	Absent

First crystal phase (Main phase): $(K_aNa_bLi_cCd)_e(DrEg)O_h$
 (where the element C includes one or more elements selected from the group consisting of Ca, Sr, and Ba, the element D includes one or more elements selected from the group consisting of Nb, Ta, Ti, and Zr while including at least Nb or Ta, and the element E includes 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.)

Second crystal phase (Subphase): M_xTiO_y
 (where the element M includes one or more elements selected from the group consisting of Li, Mg, Al, Sc, Cr, Mn, Fe, Co, Ni, Zn, Ga, Y, and Zr, and where the coefficients x and y are relative values with respect to the Ti content, $0.5 \leq x \leq 5.0$, and the coefficient y is an arbitrary value that forms spinel.)

(*) indicates a comparative example.

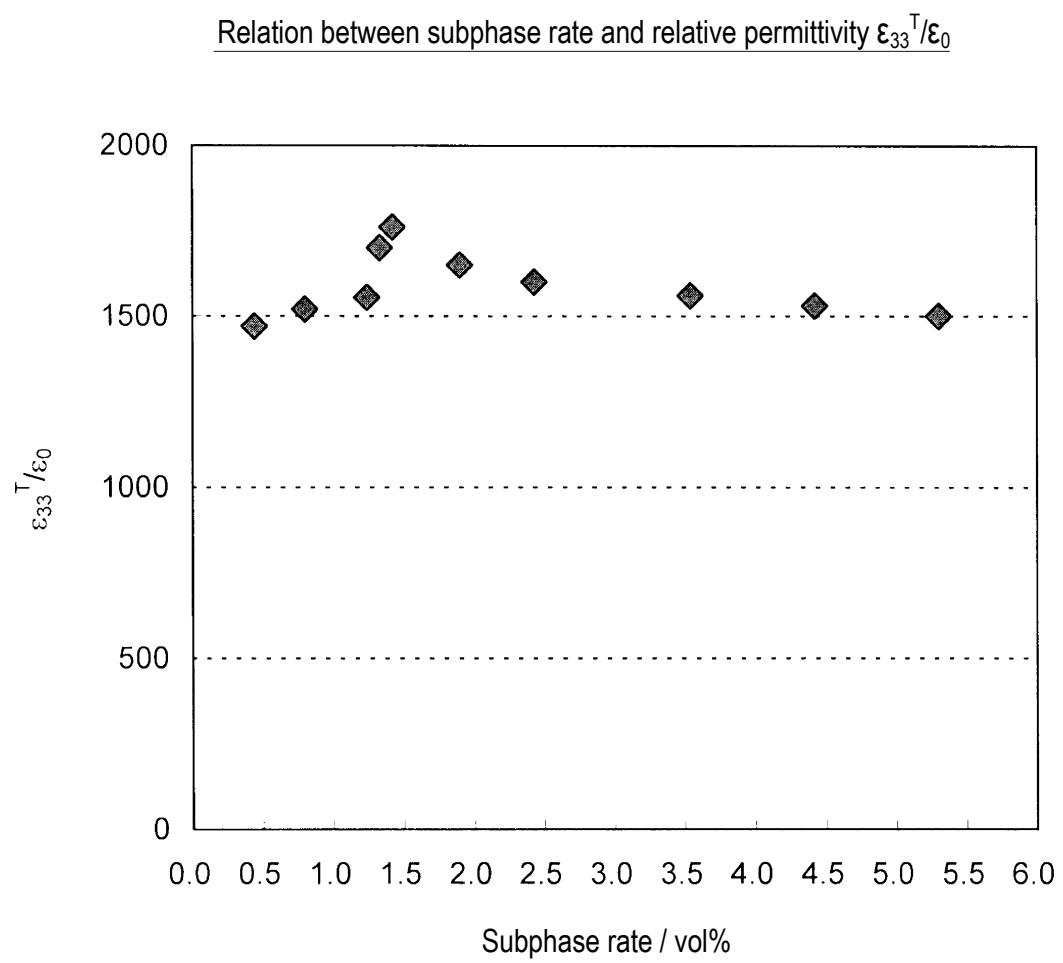
FIG. 9A

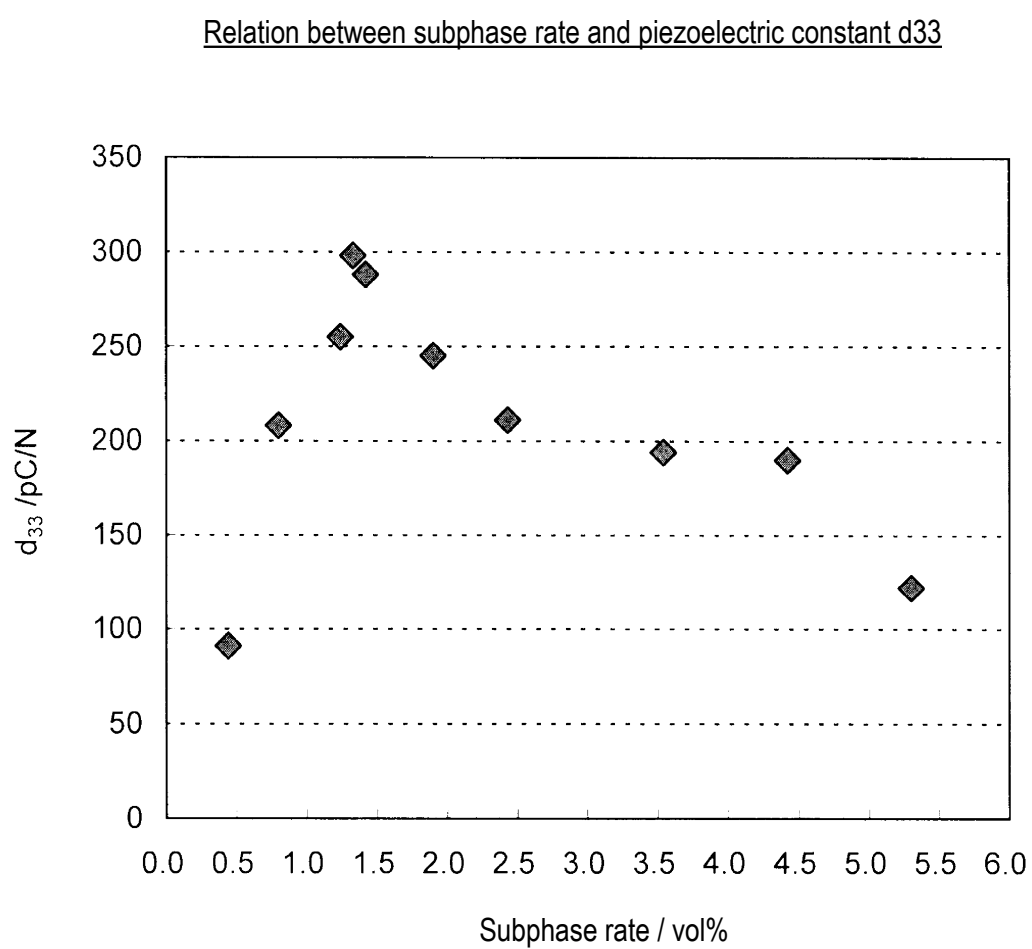
FIG. 9B

FIG. 9C

Relation between subphase rate and electromechanical coupling coefficient k_r

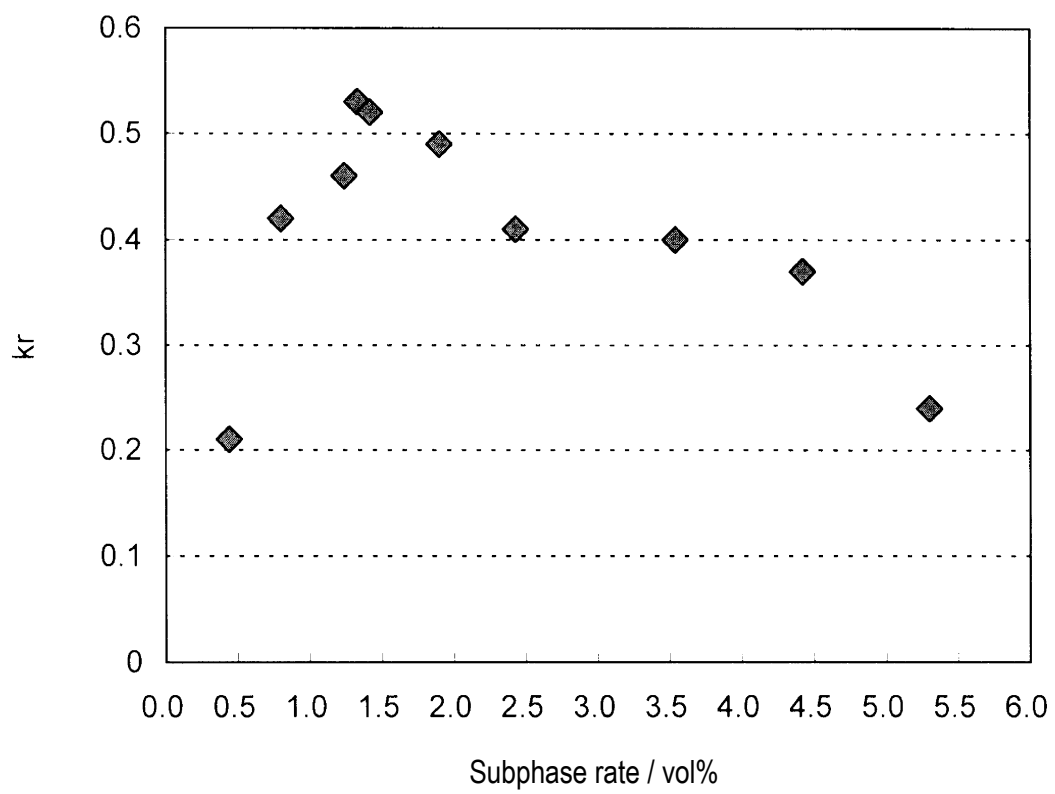
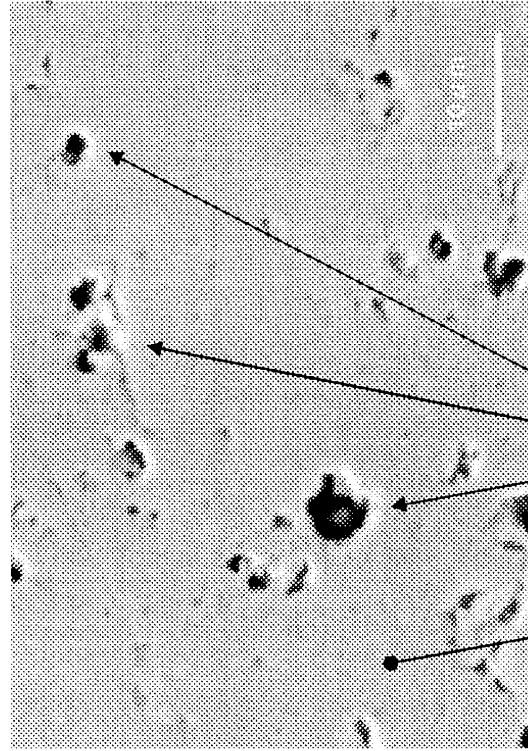
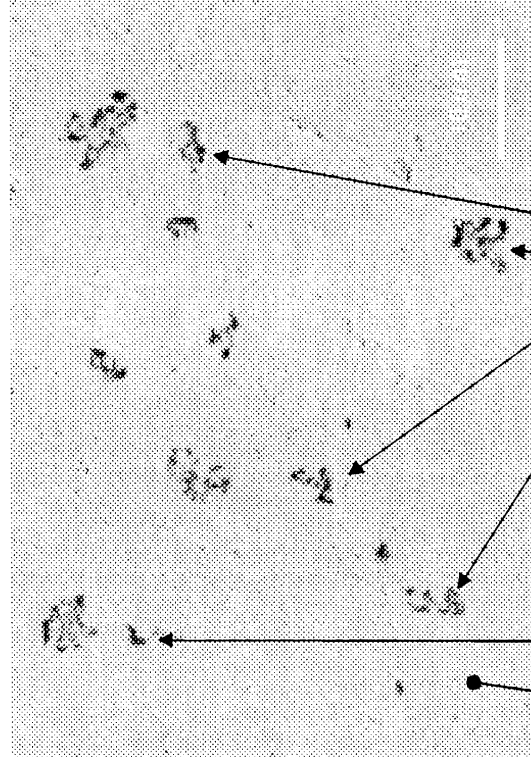


FIG. 10

(A) REFLECTED ELECTRON IMAGE OF SAMPLE S01



(B) REFLECTED ELECTRON IMAGE OF SAMPLE S06

FIRST CRYSTAL PHASE
(PEROVSKITE PHASE)

HOLES

SECOND CRYSTAL PHASE
(SPINEL PHASE)FIRST CRYSTAL PHASE
(PEROVSKITE PHASE)

HOLES

FIG. 11A

Influence of type of metallic element M of subphase (No. 1)

		First crystal phase (Main phase)													Second crystal phase (Subphase)		
Sample	Element C	Element D	Element E	a	b	c	Coefficient d of element C		e	Coefficient f of element D			Coefficient g of element E			Subphase rate (Vol%)	Element M
							d1	d2		f1	f2	f3	g1	g2	g3		
S06	Ca, Ba	Nb, Ti, Zr	Co, Zn	0.435	0.487	0.022	0.021	0.035	1.01	0.945	0.021	0.025	0.005	0.005	0	Co, Zn	
S13	Ca, Ba	Nb, Ti, Zr	Co	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.022	0.025	0.008	0	0	Co	
S14	Ca, Ba	Nb, Ti, Zr	Zn	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.022	0.025	0.008	0	0	Zn	
S15	Ca, Ba	Nb, Ti, Zr	Fe	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.022	0.025	0.008	0	0	Fe	
S16	Ca, Ba	Nb, Ti, Zr	Al	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.022	0.025	0.008	0	0	Li, Al	
S17	Ca, Ba	Nb, Ti, Zr	Mn	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.022	0.025	0.008	0	0	Mn	
S18	Ca, Ba	Nb, Ti, Zr	Mg	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.025	0.025	0.008	0	0	Mg	
S19	Ca, Ba	Nb, Ti, Zr	Ni	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.025	0.025	0.008	0	0	Ni	
S20	Ca, Ba	Nb, Ti, Zr	-	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.022	0.033	0	0	0	Li, Zr	
S21	Ca, Ba	Nb, Ti, Zr	Fe, Zn	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.025	0.025	0.007	0.002	0	Fe, Zn	
S22	Ca, Ba	Nb, Ti, Zr	Fe, Zn, Co	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.025	0.025	0.007	0.002	0.008	Fe, Zn, Co	
S23(*)	Ca, Ba	Nb, Ti, Zr	Sr	0.488	0.409	0.027	0.026	0.050	1.00	0.953	0.022	0.025	0.008	0	0	Sr	

First crystal phase (Main phase): $(K_aNa_bLi_cCd)_e(DrFe_g)O_h$
 (where the element C includes one or more elements selected from the group consisting of Ca, Sr, and Ba, the element D includes one or more elements selected from the group consisting of Nb, Ta, Ti, and Zr while including at least Nb or Ta, and the element E includes 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.)

Second crystal phase (Subphase): M_xTiO_y
 (where the element M includes one or more elements selected from the group consisting of Li, Mg, Al, Sc, Cr, Mn, Fe, Co, Ni, Zn, Ga, Y, and Zr, and where the coefficients x and y are relative values with respect to the Ti content, $0.5 \leq x \leq 5.0$, and the coefficient y is an arbitrary value that forms spinel.)

(*) indicates a comparative example.

(In sample S23, the second crystal phase did not form a spinel compound but formed a perovskite phase.)

FIG. 11B

Influence of type of metallic element M of subphase (No. 1)

Sample	First crystal phase (Main phase)	Second crystal phase (Subphase)		Relative permittivity	Dielectric loss	Piezoelectric constant	Electro- mechanical coupling coefficient	Electro- mechanical coupling coefficient	Mechanical quality factor	Curie point
	Composition	Subphase rate (Vol%)	Element M	Composition	$\tan \delta$	$d_{33}(\text{pC/N})$	k_r	k_t	Q_m	$T_c(^{\circ}\text{C})$
S06	FIG. 11A	1.33	Co, Zn	CoZnTiO_4	1700	0.018	298	0.53	0.49	315
S13	do.	1.40	Co	Co_2TiO_4	1772	0.022	283	0.51	0.47	323
S14	do.	1.40	Zn	Zn_2TiO_4	1652	0.025	257	0.51	0.40	322
S15	do.	1.40	Fe	Fe_2TiO_4	1667	0.024	301	0.54	0.49	310
S16	do.	1.40	Li, Al	LiAlTiO_4	1799	0.023	270	0.51	0.45	320
S17	do.	1.40	Mn	Mn_2TiO_4	1620	0.016	233	0.46	0.37	322
S18	do.	1.40	Mg	Mg_2TiO_4	1810	0.027	270	0.52	0.55	321
S19	do.	1.40	Ni	Ni_2TiO_4	1757	0.034	295	0.54	0.46	330
S20	do.	1.40	Li, Zr	$\text{Li}_{1.33}(\text{Zr}, \text{Ti})_{1.67}\text{O}_4$	1330	0.026	256	0.49	0.40	320
S21	do.	1.40	Fe, Zn	FeZnTiO_4	1847	0.022	299	0.56	0.47	315
S22	do.	1.40	Fe, Zn, Co	$(\text{Fe}, \text{Zn}, \text{Co})_2\text{TiO}_4$	1780	0.021	274	0.52	0.47	340
S23(*)	do.	1.40	Sr	SrTiO_3	1320	0.037	116	0.28	0.28	300

First crystal phase (Main phase): $(\text{K}_a\text{Na}_b\text{Li}_c\text{Cd})_e(\text{DfEg})\text{O}_h$
 (where the element C includes one or more elements selected from the group consisting of Ca, Sr, and Ba, the element D includes one or more element selected from the group consisting of Nb, Ta, Ti, and Zr while including at least Nb or Ta, and the element E includes 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.)

Second crystal phase (Subphase): M_xTiO_y
 (where the element M includes one or more elements selected from the group consisting of Li, Mg, Al, Sc, Cr, Mn, Fe, Co, Ni, Zn, Ga, Y, and Zr, and where the coefficients x and y are relative values with respect to the Ti content, $0.5 \leq x \leq 5.0$, and the coefficient y is an arbitrary value that forms spinel.)

(*) indicates a comparative example.

(In sample S23, the second crystal phase did not form a spinel compound but formed a perovskite phase.)

FIG. 12A

Influence of type of metallic element M of subphase (No. 2)

Sample	First crystal phase (Main phase): (K _a Na _b Li _c C _d)e(DrE _g)O _h														Second crystal phase (Subphase)			
	Element C	Element D	Element E	a	b	c	Coefficient d of element C		e	Coefficient f of element D				Coefficient g of element E			Rate (Vol%)	Element M
							d1	d2		f1	f2	f3	f4	g1	g2	g3		
S100	Ca, Ba	Nb, Ti, Zr	Ni, Fe	0.491	0.422	0.021	0.044	0.023	1.00	0.944	0.021	0.025	0	0.005	0.005	0	Ni, Fe	
S101	Ca, Ba	Nb, Ti, Zr	Mg, Fe	0.490	0.422	0.021	0.042	0.024	0.99	0.942	0.022	0.025	0	0.008	0.004	0	Mg, Fe	
S102	Ca, Ba	Nb, Ti, Zr	Fe, Al	0.487	0.422	0.021	0.045	0.025	1.06	0.954	0.019	0.016	0	0.005	0.005	0	Fe, Al, Li	
S103	Ca, Ba	Nb, Ti, Zr	Mn, Al	0.490	0.423	0.021	0.044	0.022	1.06	0.954	0.019	0.016	0	0.005	0.005	0	Mn, Al, Li	
S104	Ca, Ba	Nb, Ti, Zr	Mg, Mg	0.486	0.427	0.021	0.042	0.024	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Mg, Mn	
S105	Ca, Ba	Nb, Ti, Zr	Ni, Mg	0.532	0.383	0.021	0.041	0.022	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Ni, Mg	
S106	Ca, Ba	Nb, Ti, Zr	Ni	0.481	0.430	0.021	0.043	0.025	1.05	0.949	0.019	0.026	0	0.005	0	0	Ni, Zr	
S107	Ca, Ba	Nb, Ti, Hf	Ni	0.481	0.430	0.021	0.043	0.025	1.05	0.949	0.019	0.026	0	0.005	0	0	Ni, Hf	
S108	Ca, Ba	Nb, Ti, Zr	Ni, Zn	0.486	0.427	0.021	0.042	0.024	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Ni, Zn	
S109	Ca, Ba	Nb, Ti, Zr	Fe, Ni	0.486	0.427	0.021	0.042	0.024	1.04	0.939	0.019	0.021	0	0.016	0.005	0	Fe, Ni	
S110	Ca, Ba	Nb, Ti, Zr	Sc	0.486	0.427	0.021	0.042	0.024	1.06	0.954	0.019	0.021	0	0.005	0.000	0	Sc, Li	
S111	Ca, Ba	Nb, Ti, Zr	Co, Mg	0.488	0.409	0.027	0.026	0.050	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Co, Mg	
S112	Ca, Ba	Nb, Ti, Zr	Co, Ni	0.488	0.409	0.027	0.026	0.050	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Co, Ni	
S113	Ca, Ba	Nb, Ti, Zr	Mg, Zn	0.488	0.409	0.027	0.026	0.050	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Mg, Zn	
S114	Ca, Ba	Nb, Ti, Zr	Co, Fe	0.488	0.409	0.027	0.026	0.050	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Co, Fe	
S115	Ca, Ba	Nb, Ti, Zr	Co, Fe	0.435	0.487	0.022	0.021	0.035	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Co, Fe	
S116	Ca, Ba	Nb, Ti, Ta	Co, Fe	0.435	0.487	0.022	0.021	0.035	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Co, Fe	
S117	Ca, Sr	Nb, Ti, Zr	Co, Fe	0.435	0.487	0.022	0.021	0.035	1.05	0.949	0.019	0.021	0	0.005	0.005	0	Co, Fe	
S118	Ca, Ba	Nb, Ti, Zn, Sn	Zn	0.435	0.487	0.022	0.021	0.035	1.04	0.935	0.019	0.021	0.021	0.005	0.000	0	Zn, Sn	
S119	Ca, Ba	Nb, Ti, Zn, Sb	Zn	0.435	0.487	0.022	0.021	0.035	1.04	0.935	0.019	0.021	0.021	0.005	0.000	0	Zn, Sb	
S120	Ca, Ba	Nb, Ti, Zn, Si	Zn	0.435	0.487	0.022	0.021	0.035	1.05	0.944	0.019	0.021	0.010	0.005	0.000	0	Zn, Si	

FIG. 12B

Influence of type of metallic element M of subphase (No. 2)

Sample		First crystal phase (Main phase): (K _a Na _b Li _c C _d)e(D _f E _g)O _h													Second crystal phase (Subphase)				
		Element C	Element D	Element E	a	b	c	Coefficient d of element C		e	Coefficient f of element D				Coefficient g of element E			Rate (Vol%)	Element M
								d1	d2		f1	f2	f3	f4	g1	g2	g3		
S121		Ca, Ba	Nb, Ti, Zr	Fe	0.435	0.487	0.022	0.021	0.035	1.06	0.954	0.019	0.021	0	0.005	0.000	0	1.80	Fe
S122		Ca, Ba	Nb, Ti, Zr	Ni	0.435	0.487	0.022	0.021	0.035	1.06	0.954	0.019	0.021	0	0.005	0.000	0	1.80	Ni
S123		Ca, Ba	Nb, Ti, Zr	Mg	0.435	0.487	0.022	0.021	0.035	1.06	0.954	0.019	0.021	0	0.005	0.000	0	1.80	Mg
S124		Ca, Ba	Nb, Ti, Zr	Zn	0.435	0.487	0.022	0.021	0.035	1.06	0.954	0.019	0.021	0	0.005	0.000	0	1.80	Zn
S125		Ca, Ba	Nb, Ti, Zr	Mn	0.435	0.487	0.022	0.021	0.035	1.06	0.954	0.019	0.021	0	0.005	0.000	0	1.80	Mn
S126		Ca, Ba	Nb, Ti, Zr	Co	0.435	0.487	0.022	0.021	0.035	1.06	0.954	0.019	0.021	0	0.005	0.000	0	1.80	Co
S127		Ca, Ba	Nb, Ti, Zr	Fe, Zn Co	0.507	0.403	0.023	0.043	0.024	1.04	0.947	0.028	0.021	0	0.002	0.002	0.003	1.50	Fe, Zn, Co
S128		Ca, Ba	Nb, Ti, Hf	Fe, Zn Co	0.500	0.405	0.023	0.045	0.026	1.03	0.947	0.028	0.022	0	0.002	0.002	0.002	1.60	Fe, Zn, Co
S129		Ca, Ba	Nb, Ti, Zr	Fe, Zn Co	0.498	0.407	0.023	0.047	0.026	1.02	0.947	0.027	0.022	0	0.002	0.002	0.002	1.70	Fe, Zn, Co
S130		Ca, Ba	Nb, Ti, Zr	Fe, Zn Co	0.426	0.507	0.019	0.029	0.018	1.00	0.956	0.022	0.020	0	0.001	0.001	0.001	1.60	Fe, Zn, Co
S131		Ca, Ba	Nb, Ti, Si	Fe, Zn Co	0.429	0.509	0.019	0.031	0.012	1.00	0.961	0.024	0.013	0	0.001	0.002	0.001	1.50	Fe, Zn, Co

FIG. 12C

Influence of type of metallic element M of subphase (No. 2)

Sample	First crystal phase (Main phase)		Second crystal phase (Subphase)			Third crystal phase (Subphase)		Relative permittivity	Dielectric loss	Piezoelectric constant	Electro-mechanical coupling coefficient	Electro-mechanical coupling coefficient	Mechanical quality factor	Curie point
	Composition	Rate (Vol%)	Element M	Composition	Rate (Vol%)	Composition	Rate (Vol%)	$\epsilon_{33}^T/\epsilon_0$	$\tan\delta$	$d_{33}(\text{pC/N})$	kr	kt	Qm	$T_c(^{\circ}\text{C})$
S100	FIG. 12A	1.80	Ni, Fe	NiFeTiO ₄	0.1 or less	(Ba, Na, K) ₃ (Nb, Ni, Fe) ₅ O ₁₅	1720	0.021	0.021	317	0.55	0.48	62	317
S101	do.	1.80	Mg, Fe	MgFeTiO ₄	Absent	-	1650	0.022	0.022	319	0.55	0.47	60	318
S102	do.	1.80	Li, Fe, Al	LiFe _{0.5} Ti _{1.5} O ₄	Absent	-	1800	0.024	0.024	285	0.52	0.45	74	319
S103	do.	1.80	Li, Mn, Al	LiMn _{0.5} Ti _{1.5} O ₄	Absent	-	1700	0.014	0.014	245	0.50	0.44	146	327
S104	do.	1.80	Mg, Mn	MgMnTiO ₄	Absent	-	1620	0.013	0.013	257	0.51	0.45	143	325
S105	do.	1.80	Ni, Mg	MgNiTiO ₄	Absent	-	1680	0.023	0.023	299	0.53	0.47	65	323
S106	do.	1.80	Ni, Zr	Ni ₂ (Ti, Zr)O ₄	0.3	(Ba, Na, K) ₃ (Nb, Ni, Zr) ₅ O ₁₅	1720	0.020	0.020	316	0.56	0.49	53	328
S107	do.	1.80	Ni, Hf	Ni ₂ (Ti, Hf)O ₄	0.3	(Ba, Na, K) ₃ (Nb, Ni, Hf) ₅ O ₁₅	1800	0.022	0.022	300	0.54	0.46	60	320
S108	do.	1.80	Ni, Zn	ZnNiTiO ₄	Absent	-	1560	0.027	0.027	277	0.52	0.45	67	324
S109	do.	1.80	Fe, Ni	Ni _{1.5} FeTi _{0.5} O ₄	0.1 or less	(Ba, Na, K) ₃ (Nb, Ni, Fe) ₅ O ₁₅	1540	0.020	0.020	314	0.54	0.46	60	319
S110	do.	1.80	Sc, Li	LiScTiO ₄	Absent	-	1750	0.020	0.020	292	0.54	0.45	55	316
S111	do.	1.80	Co, Mg	CoMgTiO ₄	Absent	-	1680	0.021	0.021	318	0.54	0.47	66	324
S112	do.	1.80	Co, Ni	CoNiTiO ₄	0.1 or less	(Ba, Na, K) ₃ (Nb, Co, Ni) ₅ O ₁₅	1710	0.022	0.022	288	0.54	0.47	59	321
S113	do.	1.80	Mg, Zn	ZnMgTiO ₄	Absent	-	1690	0.021	0.021	293	0.53	0.46	62	326
S114	do.	1.80	Co, Fe	CoFeTiO ₄	Absent	-	1715	0.021	0.021	295	0.55	0.48	60	316
S115	do.	1.80	Co, Fe	CoFeTiO ₄	Absent	-	1650	0.020	0.020	320	0.55	0.48	54	315
S116	do.	1.80	Co, Fe	CoFeTiO ₄	Absent	-	1830	0.024	0.024	282	0.53	0.46	52	307
S117	do.	1.80	Co, Fe	CoFeTiO ₄	Absent	-	1880	0.025	0.025	300	0.54	0.46	55	318
S118	do.	1.80	Zn, Sn	Zn ₂ (Ti, Sn) ₄	Absent	-	1820	0.018	0.018	301	0.52	0.45	49	323
S119	do.	1.80	Zn, Sb	Zn ₂ (Ti, Sb) ₄	Absent	-	1720	0.024	0.024	278	0.52	0.46	58	309
S120	do.	2.00	Zn, Si	Zn ₂ (Ti, Si) ₄	Absent	-	1710	0.022	0.022	290	0.52	0.45	51	300

FIG. 12D

Influence of type of metallic element M of subphase (No. 2)

Sample	First crystal phase (Main phase)	Second crystal phase (Subphase)			Third crystal phase (Subphase)		Relative permittivity	Dielectric loss	Piezoelectric constant	Electro-mechanical coupling coefficient	Electro-mechanical coupling coefficient	Curie point
	Composition	Rate (Vol%)	Element M	Composition	Rate (Vol%)	Composition	$\epsilon_{33}/\epsilon_{00}$	$\tan\delta$	$d_{33}(\text{pC/N})$	kr	kt	$T_c(^{\circ}\text{C})$
S121	FIG. 12B	1.80	Fe	Fe_2TiO_4	0.2	(Ba, Na, K) ₃ (Nb, Fe) ₅ O ₁₅	1730	0.021	263	0.52	0.46	310
S122	do.	1.80	Ni	Ni_2TiO_4	0.1 or less	(Ba, Na, K) ₃ (Nb, Ni) ₅ O ₁₅	1760	0.018	270	0.50	0.45	319
S123	do.	1.80	Mg	Mg_2TiO_4	Absent	-	1745	0.018	271	0.51	0.46	321
S124	do.	1.80	Zn	Zn_2TiO_4	0.2	(Ba, Na, K) ₃ (Nb, Zn) ₅ O ₁₅	1600	0.030	250	0.51	0.44	322
S125	do.	1.80	Mn	Mn_2TiO_4	0.2	(Ba, Na, K) ₃ (Nb, Mn) ₅ O ₁₅	1590	0.011	222	0.45	0.41	322
S126	do.	1.80	Co	Co_2TiO_4	Absent	-	1700	0.022	262	0.49	0.41	323
S127	do.	1.50	Fe, Zn, Co	(Fe, Zn, Co) ₂ TiO ₄	0.1 or less	(Ba, Na, K) ₃ (Nb, Ni, Zn) ₅ O ₁₅	1760	0.018	295	0.52	0.47	324
S128	do.	1.60	Fe, Zn, Co	(Fe, Zn, Co) ₂ TiO ₄	0.1 or less	(Ba, Na, K) ₃ (Nb, Ni, Hf) ₅ O ₁₅	1510	0.022	290	0.51	0.46	328
S129	do.	1.70	Fe, Zn, Co	(Fe, Zn, Co) ₂ TiO ₄	0.1 or less	(Ba, Na, K) ₃ (Nb, Ni, Zn) ₅ O ₁₅	1340	0.024	280	0.51	0.44	330
S130	do.	1.60	Fe, Zn, Co	(Fe, Zn, Co) ₂ TiO ₄	0.1 or less	(Ba, Na, K) ₃ (Nb, Ni, Fe) ₅ O ₁₅	1630	0.015	285	0.50	0.46	320
S131	do.	1.50	Fe, Zn, Co	(Fe, Zn, Co) ₂ TiO ₄	0.1 or less	(Ba, Na, K) ₃ (Nb, Ni, Zn) ₅ O ₁₅	1845	0.018	310	0.56	0.49	318

FIG. 13

Evaluation result on insulation properties

Sample	Subphase rate	d ₃₃	Polarization condition (polarization time: 30 min.)		
	(Vol%)	(pC/N)	25°C_7kV/mm	40°C_7kV/mm	80°C_9kV/mm
S01(*)	0	90	NG	NG	NG
S03	0.44	61	OK	OK	NG
S06	1.33	298	OK	OK	OK
S08	1.90	245	OK	OK	OK

Direct voltage was placed and a breakage state of element was evaluated.

(*) indicates a comparative example.

FIG. 14

Evaluation on high-temperature endurance

Sample	d ₃₃ (pC/N)		Degradation rate (%)
	Initial	After aging	
S01(*)	90	60	-33.3
S22	274	264	-3.6

Piezoelectric properties after aging at 200°C for 10 h were compared.

(*) indicates a comparative example.

FIG. 15

Result on thermal cycle test

Sample	d ₃₃ (pC/N)		
	Initial	After thermal cycle	Degradation rate (%)
S02(*)	95	63	-66
S129	280	276	-1.4
S130	285	279	-2.1
S131	310	299	-3.5

(*) indicates a comparative example.

FIG. 16A

Influence of coefficient e of first crystal phase (main phase) (No.1)

Sample	First crystal phase (Main phase)										Second crystal phase (Subphase)				
	Element C	Element D	Element E	a	b	c	Coefficient d of element C		e	Coefficient f of element D			g	Subphase rate (Vol%)	Element M
							d1	d2		f1	f2	f3			
S01(*)	-	Nb	-	0.500	0.500	0	0	0	1.00	1.000	0	0	0	0	0
S24	Ca, Ba	Nb, Ti, Zr	Co	0.488	0.409	0.027	0.026	0.050	0.800	0.953	0.022	0.025	0.008	1.40	Co
S25	Ca, Ba	Nb, Ti, Zr	Co	0.488	0.409	0.027	0.026	0.050	0.880	0.953	0.022	0.025	0.008	1.40	Co
S26	Ca, Ba	Nb, Ti, Zr	Co	0.488	0.409	0.027	0.026	0.050	0.940	0.953	0.022	0.025	0.008	1.40	Co
S27	Ca, Ba	Nb, Ti, Zr	Co	0.488	0.409	0.027	0.026	0.050	0.980	0.953	0.022	0.025	0.008	1.40	Co
S28(S13)	Ca, Ba	Nb, Ti, Zr	Co	0.488	0.409	0.027	0.026	0.050	1.000	0.953	0.022	0.025	0.008	1.40	Co
S29	Ca, Ba	Nb, Ti, Zr	Co	0.488	0.409	0.027	0.026	0.050	1.030	0.953	0.022	0.025	0.008	1.40	Co
S30	Ca, Ba	Nb, Ti, Zr	Co	0.488	0.409	0.027	0.026	0.050	1.070	0.953	0.022	0.025	0.008	1.40	Co
S31	Ca, Ba	Nb, Ti, Zr	Co	0.488	0.409	0.027	0.026	0.050	1.100	0.953	0.022	0.025	0.008	1.40	Co

First crystal phase (Main phase): $(K_aNa_bLi_cCd)_e(DfEg)O_h$
(where the element C includes one or more elements selected from the group consisting of Ca, Sr, and Ba, the element D includes one or more elements selected from the group consisting of Nb, Ta, Ti, and Zr while including at least Nb or Ta, and the element E includes one or more elements selected from the group consisting of Co, Zn, Fe, Al, Mn, Mg, Ni, Y, Sc, and Ga, where $a + b + c + d = 1$, e is arbitrary, $f + g = 1$, and h is an arbitrary value that forms perovskite.)

Second crystal phase (Subphase): M_xTiO_y
(where the element M includes one or more elements selected from the group consisting of Li, Co, Zn, Mg, Ni, Fe, Al, Y, Sc, Ga, Mn, and Zr, and where the coefficients x and y are relative values with respect to the Ti content, $0.5 \leq x \leq 5.0$, and the coefficient y is an arbitrary value that forms spinel.)

(*) indicates a comparative example.

FIG. 16B

Influence of coefficient e of first crystal phase (main phase) (No.2)

Sample	First crystal phase (Main phase)		Second crystal phase (Subphase)		Relative permittivity	Piezoelectric constant	Electro- mechanical coupling coefficient
	Composition	e	Subphase rate (Vol%)	Element M			
S01(*)	FIG. 14A	1.00	0.0	-	430	90	0.30
S 24	do.	0.80	1.40	Co	890	103	0.28
S 25	do.	0.88	1.40	Co	1400	205	0.40
S 26	do.	0.94	1.40	Co	1430	220	0.42
S 27	do.	0.98	1.40	Co	1560	255	0.48
S28(S13)	do.	1.00	1.40	Co	1772	283	0.51
S 29	do.	1.03	1.40	Co	1620	244	0.46
S 30	do.	1.07	1.40	Co	1540	232	0.45
S 31	do.	1.10	1.40	Co	1500	98	0.26

First crystal phase (Main phase): $(K_aNa_bLi_cCo)_e(DfEg)O_h$
(where the element C includes one or more elements selected from the group consisting of Ca, Sr, and Ba, the element D includes one or more elements selected from the group consisting of Nb, Ta, Ti, and Zr while including at least Nb or Ta, and the element E includes one or more elements selected from the group consisting of Co, Zn, Fe, Al, Mn, Mg, Ni, Y, Sc, and Ga, where $a + b + c + d = 1$, e is arbitrary, $f + g = 1$, and h is an arbitrary value that forms perovskite.)

Second crystal phase (Subphase): M_xTiO_y
(where the element M includes one or more elements selected from the group consisting of Li, Co, Zn, Mg, Ni, Fe, Al, Y, Sc, Ga, Mn, and Zr, and where the coefficients x and y are relative values with respect to the Ti content, $0.5 \leq x \leq 5.0$, and the coefficient y is an arbitrary value that forms spinel.)

(*) indicates a comparative example.

FIG. 17