

*FIG. 1*



FIG. 2

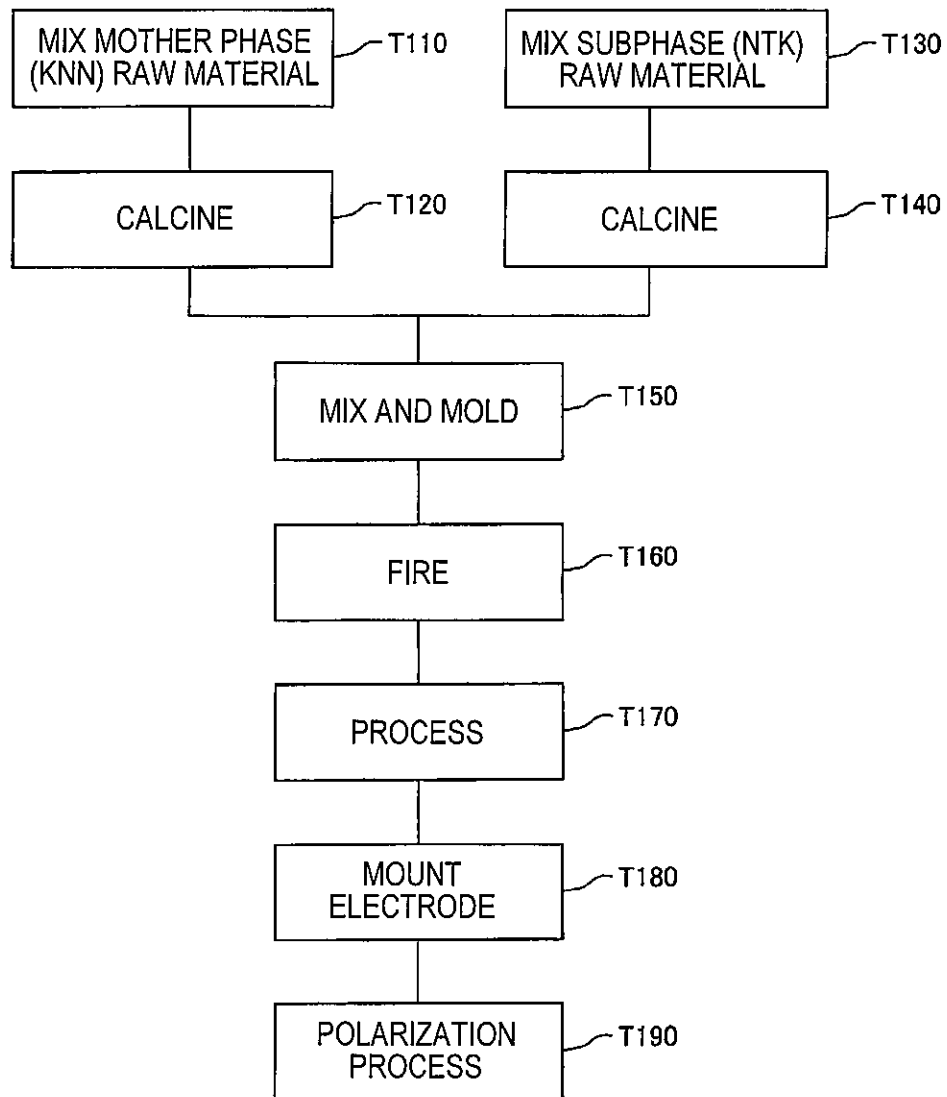


FIG. 3

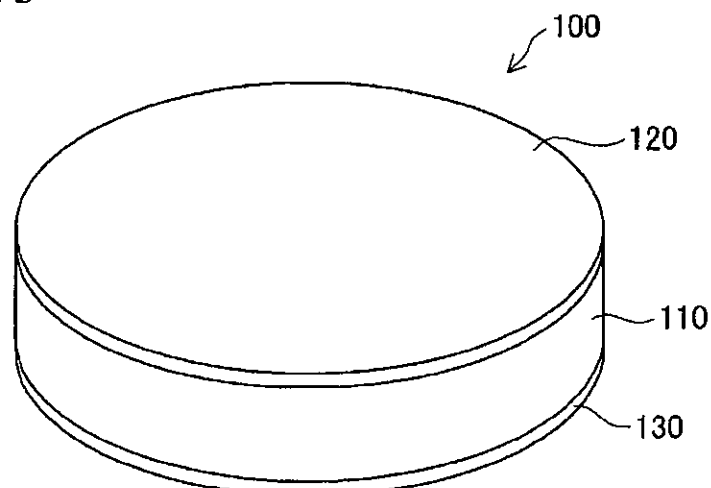


FIG. 4

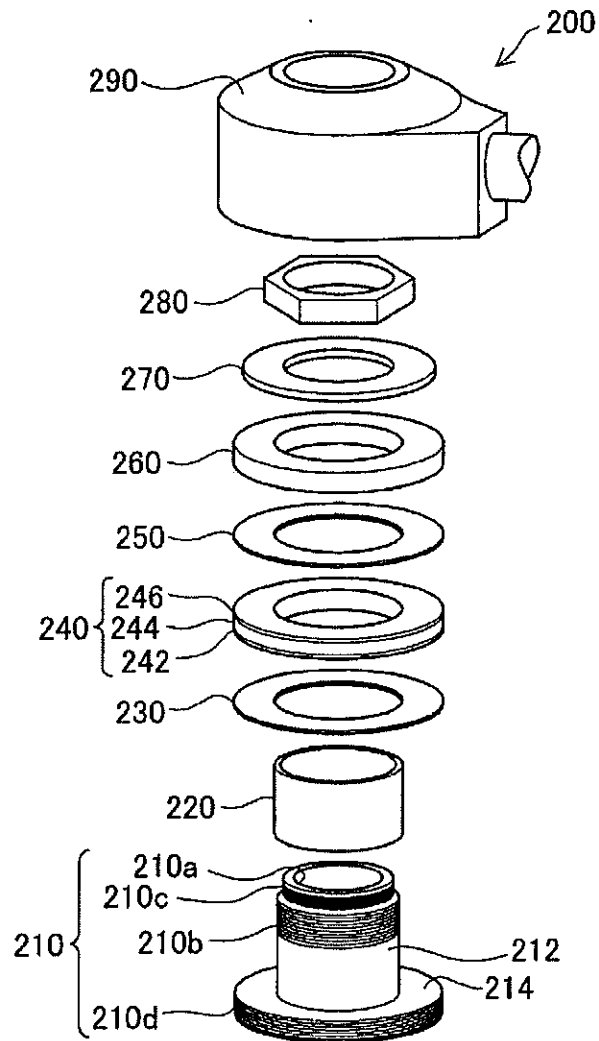


FIG. 5

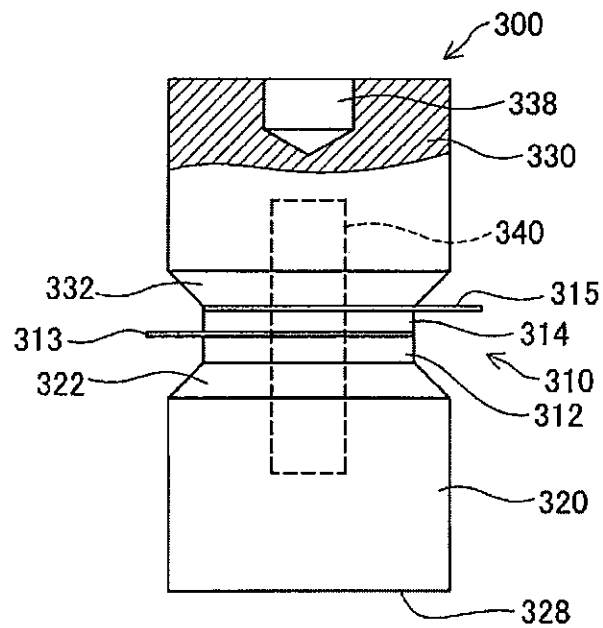


FIG. 6

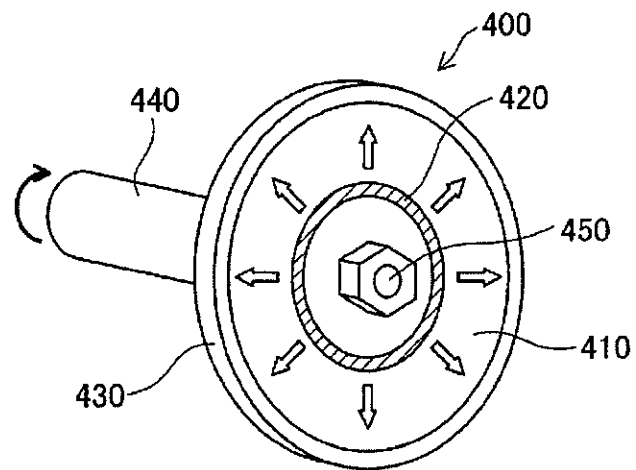


FIG. 7

5/24

| Sample | First crystal phase (mother phase) |           |       |       |       |       |      | Second crystal phase (subphase) |           |           | Subphase content (mol%) | Relative permittivity $\epsilon_{33}/T/\epsilon_0$ | Piezoelectric constant $d_{33}$ (pC/N) | Electromechanical coupling coefficient $k_r$ |
|--------|------------------------------------|-----------|-------|-------|-------|-------|------|---------------------------------|-----------|-----------|-------------------------|----------------------------------------------------|----------------------------------------|----------------------------------------------|
|        | Element E                          | Element D | a     | b     | c     | d     | e    | f                               | Element A | Element B | x                       |                                                    |                                        |                                              |
| S01    | -                                  | -         | -     | -     | -     | -     | -    | -                               | K         | Nb        | 0                       | 500                                                | -                                      | -                                            |
| S02    | -                                  | -         | -     | -     | -     | -     | -    | -                               | K         | Nb        | 0.15                    | 500                                                | -                                      | -                                            |
| S03    | -                                  | Nb        | 0.500 | 0.500 | 0     | 0     | 1.00 | 3.00                            | -         | -         | -                       | 430                                                | 90                                     | 0.30                                         |
| S04    | -                                  | Nb        | 0.490 | 0.490 | 0.020 | 0     | 1.00 | 3.00                            | -         | -         | -                       | 430                                                | 95                                     | 0.31                                         |
| S05    | -                                  | Nb        | 0.490 | 0.490 | 0.020 | 0     | 1.00 | 3.00                            | K         | Nb        | 0                       | 1080                                               | 154                                    | 0.36                                         |
| S06    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 1100                                               | 120                                    | 0.30                                         |
| S07    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 1110                                               | 164                                    | 0.32                                         |
| S08    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 1130                                               | 170                                    | 0.43                                         |
| S09    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 1050                                               | 163                                    | 0.35                                         |
| S10    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 1000                                               | 120                                    | 0.31                                         |
| S11    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 950                                                | 105                                    | 0.30                                         |
| S12    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 900                                                | 60                                     | 0.20                                         |
| S13    | Ca                                 | Nb        | 0.455 | 0.474 | 0.022 | 0.049 | 1.07 | 3.06                            | K         | Ta        | 0                       | 1250                                               | 153                                    | 0.35                                         |
| S14    | Ca                                 | Nb        | 0.455 | 0.474 | 0.022 | 0.049 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 1200                                               | 155                                    | 0.38                                         |
| S15    | Ca, Sr                             | Nb        | 0.420 | 0.519 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0                       | 920                                                | 120                                    | 0.31                                         |

First crystal phase (mother phase):  $(K_aNa_bLi_cE_d)_fDO_f$

(E is at least one of Ca, Sr, and Ba; D is at least one of Nb and Ta;  $a+b+c+d=1$ ; e is any value; f is any value that forms a perovskite)

Second crystal phase (subphase):  $A_{1-x}Ti_{1-x}B_{1+x}O_5$

(A is at least one alkali metal; B is at least one of Nb and Ta; x is any value)

FIG. 8

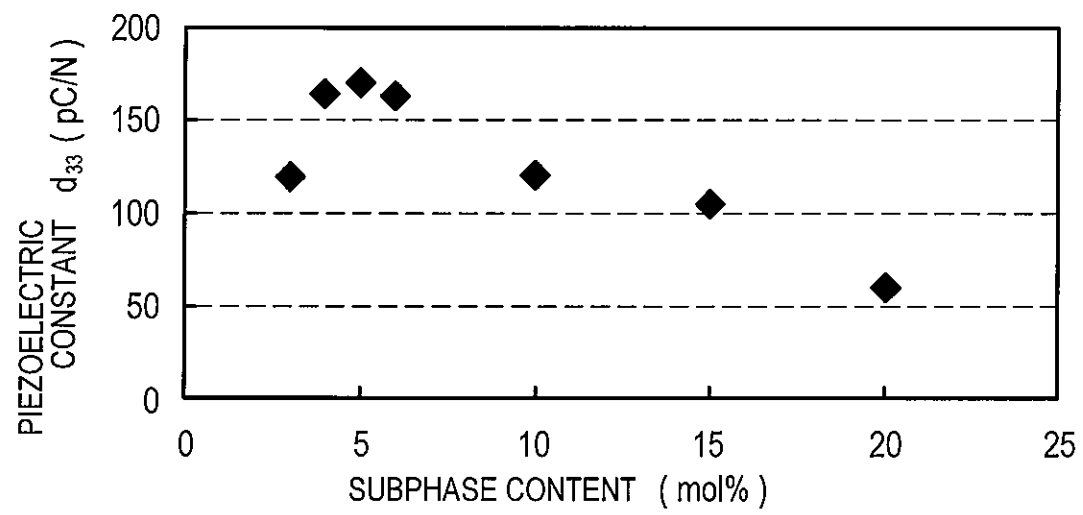


FIG. 9

| Sample | First crystal phase (mother phase) |           |       |       |       |       |      | Second crystal phase (subphase) |           |           | Subphase content (mol%) | Curie point T <sub>c</sub> (°C) | Phase transition at room temperature |
|--------|------------------------------------|-----------|-------|-------|-------|-------|------|---------------------------------|-----------|-----------|-------------------------|---------------------------------|--------------------------------------|
|        | Element E                          | Element D | a     | b     | c     | d     | e    | f                               | Element A | Element B | x                       |                                 |                                      |
| S01    | -                                  | -         | -     | -     | -     | -     | -    | -                               | K         | Nb        | 0                       | -                               | -                                    |
| S02    | -                                  | -         | -     | -     | -     | -     | -    | -                               | K         | Nb        | 0.15                    | -                               | -                                    |
| S03    | -                                  | Nb        | 0.500 | 0.500 | 0     | 0     | 1.00 | 3.00                            | -         | -         | -                       | 400                             | Present                              |
| S04    | -                                  | Nb        | 0.490 | 0.490 | 0.020 | 0     | 1.00 | 3.00                            | -         | -         | -                       | 410                             | Present                              |
| S05    | -                                  | Nb        | 0.490 | 0.490 | 0.020 | 0     | 1.00 | 3.00                            | K         | Nb        | 0                       | 340                             | Absent                               |
| S06    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 350                             | Absent                               |
| S07    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 350                             | Absent                               |
| S08    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 350                             | Absent                               |
| S09    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 350                             | Absent                               |
| S10    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 350                             | Absent                               |
| S11    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 350                             | Absent                               |
| S12    | Ca                                 | Nb        | 0.421 | 0.518 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 350                             | Absent                               |
| S13    | Ca                                 | Nb        | 0.455 | 0.474 | 0.022 | 0.049 | 1.07 | 3.06                            | K         | Ta        | 0                       | 300                             | Absent                               |
| S14    | Ca                                 | Nb        | 0.455 | 0.474 | 0.022 | 0.049 | 1.07 | 3.06                            | K         | Nb        | 0.15                    | 350                             | Absent                               |
| S15    | Ca, Sr                             | Nb        | 0.420 | 0.519 | 0.022 | 0.039 | 1.07 | 3.06                            | K         | Nb        | 0                       | 340                             | Absent                               |

First crystal phase (mother phase):  $(K_aNa_bLi_cE_d)_fDO_f$

(E is at least one of Ca, Sr, and Ba; D is at least one of Nb and Ta;  $a+b+c+d=l$ ; e is any value; f is any value that forms a perovskite)

Second crystal phase (subphase):  $A_{1-x}Ti_xB_{1+x}O_5$

(A is at least one alkali metal; B is at least one of Nb and Ta; x is any value)

FIG. 10

8/24

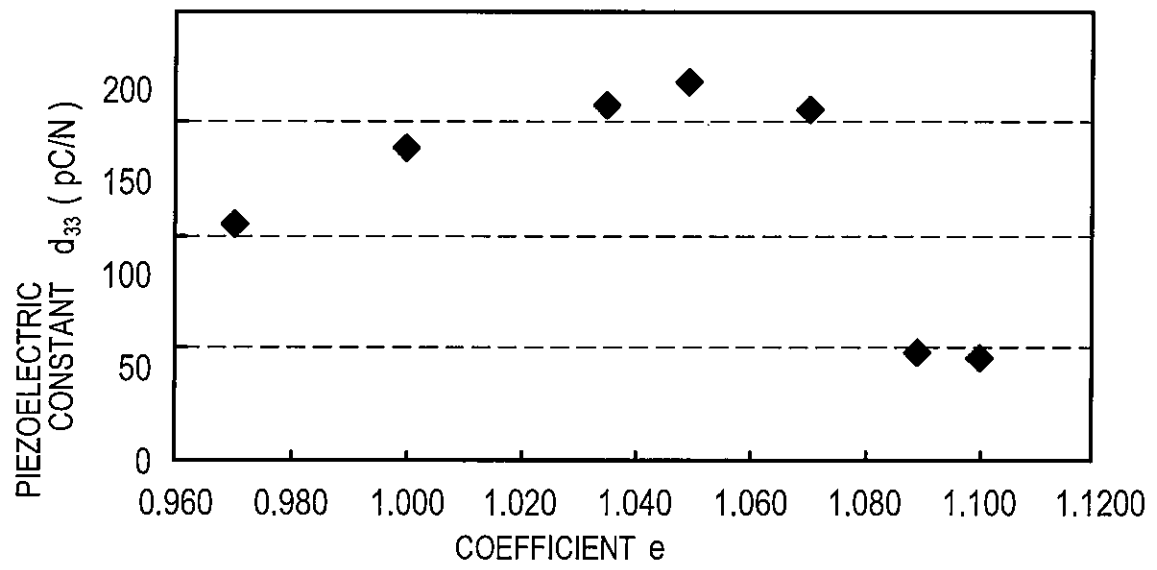
| Sample    | First crystal phase (mother phase) |       |       |       |       | Second crystal phase (subphase) |           |           | Subphase content (mol%) | Relative permittivity $\epsilon_{33} T/\epsilon_0$ | Piezoelectric constant $d_{33}$ (pC/N) | Electromechanical coupling coefficient $K_r$ |
|-----------|------------------------------------|-------|-------|-------|-------|---------------------------------|-----------|-----------|-------------------------|----------------------------------------------------|----------------------------------------|----------------------------------------------|
|           | a                                  | b     | c     | d     | e     | f                               | Element A | Element B | x                       |                                                    |                                        |                                              |
| S04       | 0.490                              | 0.490 | 0.020 | 0     | 1.00  | 3.00                            | -         | -         | -                       | 430                                                | 95                                     | 0.31                                         |
| S21       | 0.455                              | 0.474 | 0.022 | 0.049 | 0.970 | 3.01                            | K         | Nb        | 0                       | 880                                                | 105                                    | 0.29                                         |
| S22       | 0.455                              | 0.474 | 0.022 | 0.049 | 1.000 | 3.02                            | K         | Nb        | 0.15                    | 1040                                               | 139                                    | 0.36                                         |
| S23       | 0.455                              | 0.474 | 0.022 | 0.049 | 1.035 | 3.04                            | K         | Nb        | 0.15                    | 1110                                               | 157                                    | 0.40                                         |
| S24       | 0.455                              | 0.474 | 0.022 | 0.049 | 1.049 | 3.05                            | K         | Nb        | 0.15                    | 1060                                               | 167                                    | 0.42                                         |
| S25 (S14) | 0.455                              | 0.474 | 0.022 | 0.049 | 1.070 | 3.06                            | K         | Nb        | 0.15                    | 1200                                               | 155                                    | 0.38                                         |
| S26       | 0.455                              | 0.474 | 0.022 | 0.049 | 1.089 | 3.07                            | K         | Nb        | 0.15                    | 1100                                               | 47                                     | 0.12                                         |
| S27       | 0.455                              | 0.474 | 0.022 | 0.049 | 1.100 | 3.08                            | K         | Nb        | 0.15                    | 1090                                               | 45                                     | 0.12                                         |

First crystal phase (mother phase):  $(K_a Na_b Li_c Ca_d)_e NbO_f$   
 ( $a+b+c+d=1$ ; e is any value; f is any value that forms a perovskite)

Second crystal phase (subphase):  $A_{1-x} Ti_{1-x} B_{1+x} O_5$   
 (A is at least one alkali metal; B is at least one of Nb and Ta; x is any value)

9/24

FIG. 11



| Sample    | First crystal phase (mother phase) |       |       |       |       |      | Second crystal phase (subphase) |           | Subphase content (mol%) | Additional metal |          | Relative permittivity $\epsilon_{33} T/\epsilon_0$ | Piezo-electric constant $d_{33}$ (pC/N) | Electro-mechanical coupling coefficient $K_r$ |
|-----------|------------------------------------|-------|-------|-------|-------|------|---------------------------------|-----------|-------------------------|------------------|----------|----------------------------------------------------|-----------------------------------------|-----------------------------------------------|
|           | a                                  | b     | c     | d     | e     | f    | Element A                       | Element B |                         | metal            | mol%     |                                                    |                                         |                                               |
| S04       | 0.490                              | 0.490 | 0.020 | 0     | 1.00  | 3.00 | -                               | -         | 0                       | -                | 0        | 430                                                | 95                                      | 0.31                                          |
| S31       | 0.500                              | 0.500 | 0     | 0     | 1.000 | 3.00 | -                               | -         | 0                       | Cu               | 1        | 235                                                | 95                                      | 0.37                                          |
| S32 (S08) | 0.421                              | 0.518 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | -                | 0        | 1130                                               | 170                                     | 0.43                                          |
| S33       | 0.455                              | 0.484 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Cu               | 0.5      | 1050                                               | 186                                     | 0.46                                          |
| S34       | 0.455                              | 0.484 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Cu               | 1        | 985                                                | 138                                     | 0.38                                          |
| S35       | 0.455                              | 0.484 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Cu, Fe           | 0.5, 0.5 | 1042                                               | 200                                     | 0.48                                          |
| S36       | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Fe               | 0.5      | 1119                                               | 199                                     | 0.48                                          |
| S37       | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Cu, Ni           | 0.5, 0.5 | 1150                                               | 170                                     | 0.44                                          |
| S38       | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Ag               | 0.5      | 1060                                               | 171                                     | 0.42                                          |
| S39       | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Zr               | 0.5      | 1121                                               | 166                                     | 0.42                                          |
| S40       | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Zn               | 0.5      | 1221                                               | 175                                     | 0.42                                          |
| S41       | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Sc, Bi           | 0.5, 0.5 | 1213                                               | 172                                     | 0.41                                          |
| S42       | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 5                       | Mn               | 0.5      | 976                                                | 156                                     | 0.42                                          |
| S43       | 0.455                              | 0.474 | 0.022 | 0.049 | 1.070 | 3.06 | K                               | Nb        | 5                       | Co               | 0.5      | 1080                                               | 180                                     | 0.45                                          |

First crystal phase (mother phase):  $(K_aNa_bLi_cCa_d)_eNbO_f$

( $a+b+c+d=e$ ;  $e$  is any value;  $f$  is any value that forms a perovskite)

Second crystal phase (subphase):  $A_{1-x}Ti_{1-x}B_{1+x}O_5$

( $A$  is at least one alkali metal;  $B$  is at least one of Nb and Ta;  $x$  is any value)

FIG. 13

| Sample | First crystal phase (mother phase) |       |       |       |       |      | Second crystal phase (subphase) |           |      | Subphase content (mol%) | Additional metal |          | Allowable voltage (kV/mm) |
|--------|------------------------------------|-------|-------|-------|-------|------|---------------------------------|-----------|------|-------------------------|------------------|----------|---------------------------|
|        | a                                  | b     | c     | d     | e     | f    | Element A                       | Element B | x    |                         | metal            | mol%     |                           |
| S03    | 0.500                              | 0.500 | 0     | 0     | 1.00  | 3.00 | -                               | -         | -    | 0                       | -                | 0        | 3                         |
| S04    | 0.490                              | 0.490 | 0.020 | 0     | 1.00  | 3.00 | -                               | -         | -    | 0                       | -                | 0        | 3                         |
| S08    | 0.421                              | 0.518 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 0.15 | 5                       | -                | 0        | 7                         |
| S035   | 0.455                              | 0.484 | 0.022 | 0.039 | 1.070 | 3.06 | K                               | Nb        | 0.15 | 5                       | Cu, Fe           | 0.5, 0.5 | 9                         |

First crystal phase (mother phase):  $(K_aNa_bLi_cCa_d)_cNbO_f$   
 ( $a+b+c+d=1$ ; e is any value; f is any value that forms a perovskite)

Second crystal phase (subphase):  $A_{1-x}Ti_{1-x}B_{1+x}O_5$   
 (A is at least one alkali metal; B is at least one of Nb and Ta; x is any value)

FIG. 14

Second crystal phase mixed as  $\text{KTiNbO}_5$  phase

| Sample        | First crystal phase (mother phase) |       |       |       |       |      | Subphase content (mol%) | Additional metal |          | NTK phase (XRD analysis) | NTK phase (TEM-EDS) |
|---------------|------------------------------------|-------|-------|-------|-------|------|-------------------------|------------------|----------|--------------------------|---------------------|
|               | a                                  | b     | c     | d     | e     | f    |                         | metal            | mol%     |                          |                     |
| S06 (Fig. 7)  | 0.421                              | 0.518 | 0.022 | 0.039 | 1.070 | 3.06 | 3                       | -                | 0        | -                        | 1115                |
| S08 (Fig. 7)  | 0.421                              | 0.518 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | -                | 0        | 1115                     | 1115                |
| S10 (Fig. 7)  | 0.421                              | 0.518 | 0.022 | 0.039 | 1.070 | 3.06 | 10                      | -                | 0        | 1115                     | 1115                |
| S12 (Fig. 7)  | 0.421                              | 0.518 | 0.022 | 0.039 | 1.070 | 3.06 | 20                      | -                | 0        | 1115                     | 1115                |
| S33 (Fig. 12) | 0.455                              | 0.484 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | Cu               | 0.5      | -                        | 1115                |
| S35 (Fig. 12) | 0.455                              | 0.484 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | Cu, Fe           | 0.5, 0.5 | 1319                     | 1319                |
| S36 (Fig. 12) | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | Fe               | 0.5      | 1319                     | 1319                |
| S40 (Fig. 12) | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | Zn               | 0.5      | 1115                     | 1115                |
| S42 (Fig. 12) | 0.464                              | 0.474 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | Mn               | 0.5      | 1115, 1319               | 1115, 1319          |

First crystal phase (mother phase):  $(\text{K}_a\text{Na}_b\text{Li}_c\text{Ca}_d)_e\text{NbO}_f$   
 (a+b+c+d=1; e is any value; f is any value that forms a perovskite)

Second crystal phase (subphase):

NTK phase: 1115      $\text{KTiNbO}_5$  phase

NTK phase: 1319      $\text{KTi}_3\text{NbO}_9$  phase

FIG. 15

13/24

Second crystal phase mixed as  $\text{KTi}_3\text{NbO}_9$  phase

| Sample | First crystal phase (mother phase) |       |       |       |       |      | Subphase content (mol%) | Additional metal |          | NTK phase (XRD analysis) | NTK phase (TEM-EDS) |
|--------|------------------------------------|-------|-------|-------|-------|------|-------------------------|------------------|----------|--------------------------|---------------------|
|        | a                                  | b     | c     | d     | e     | f    |                         | metal            | mol%     |                          |                     |
| S51    | 0.421                              | 0.518 | 0.022 | 0.039 | 1.070 | 3.06 | 3                       | -                | 0        | -                        | 1319                |
| S52    | 0.421                              | 0.518 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | -                | 0        | 1319                     | 1319                |
| S53    | 0.455                              | 0.484 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | Cu               | 0.5      | -                        | 1319                |
| S54    | 0.455                              | 0.484 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | Cu, Fe           | 0.5, 0.5 | 1319                     | 1319                |
| S55    | 0.421                              | 0.518 | 0.022 | 0.039 | 1.070 | 3.06 | 5                       | Fe               | 0.5      | 1319                     | 1319                |
| S56    | 0.421                              | 0.508 | 0.022 | 0.049 | 1.070 | 3.07 | 5                       | Zn               | 0.5      | 1319                     | 1319                |
| S57    | 0.416                              | 0.503 | 0.033 | 0.049 | 1.080 | 3.07 | 5                       | Mn               | 0.5      | 1319                     | 1319                |

First crystal phase (mother phase):  $(\text{K}_a\text{Na}_b\text{Li}_c\text{Ca}_d)_e\text{NbO}_f$   
 ( $a+b+c+d=1$ ; e is any value; f is any value that forms a perovskite)

Second crystal phase (subphase):

NTK phase: 1115      $\text{KTiNbO}_5$  phase  
 NTK phase: 1319      $\text{KTi}_3\text{NbO}_9$  phase

| Sample        | First crystal phase (mother phase) |       |       |       |       |       |       |      |       | Subphase content (mol%) | Additional metal |                  | Relative permittivity $\epsilon_{33}T/\epsilon_0$ | Piezoelectric constant $d_{31}$ (pC/N) | Electromechanical coupling coefficient $k_T$ |
|---------------|------------------------------------|-------|-------|-------|-------|-------|-------|------|-------|-------------------------|------------------|------------------|---------------------------------------------------|----------------------------------------|----------------------------------------------|
|               | Elements E1, E2                    | a     | b     | c     | d1    | d2    | e     | f    | Metal |                         | mol%             |                  |                                                   |                                        |                                              |
| S04 (Fig. 12) | -                                  | 0.490 | 0.490 | 0.020 | 0     | 0     | 1.00  | 3.00 |       | 0                       | -                | 0                | 430                                               | 95                                     | 0.31                                         |
| S31 (Fig. 12) | -                                  | 0.500 | 0.500 | 0.000 | 0     | 0     | 1.00  | 3.00 |       | 0                       | Cu               | 1                | 235                                               | 95                                     | 0.37                                         |
| S61           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Zr               | 1                | 1207                                              | 189                                    | 0.42                                         |
| S62           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Cu, Zr           | 0.5, 1           | 1370                                              | 224                                    | 0.50                                         |
| S63           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Fe, Zr           | 0.5, 1           | 1405                                              | 185                                    | 0.42                                         |
| S64           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Zn, Zr           | 0.5, 1           | 1414                                              | 203                                    | 0.46                                         |
| S65           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Cu, Fe, Zr       | 0.5, 0.5, 1      | 1480                                              | 234                                    | 0.52                                         |
| S66           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Cu, Fe, Zr       | 0.6, 0.3, 1      | 1502                                              | 243                                    | 0.52                                         |
| S67           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Fe, Zn, Zr       | 0.5, 0.5, 1      | 1445                                              | 205                                    | 0.47                                         |
| S68           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1550                                              | 253                                    | 0.51                                         |
| S69           | Ca                                 | 0.449 | 0.478 | 0.022 | 0.050 | 0     | 1.08  | 3.07 |       | 5                       | Cu, Fe, Zr       | 0.5, 0.5, 1      | 1390                                              | 217                                    | 0.48                                         |
| S70           | Ca, Sr                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Cu, Fe, Zr       | 0.5, 0.5, 1      | 1278                                              | 211                                    | 0.47                                         |
| S71           | Ca, Ba                             | 0.396 | 0.532 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Cu, Fe, Zr       | 0.5, 0.5, 1      | 1317                                              | 205                                    | 0.46                                         |
| S72           | Ca                                 | 0.396 | 0.532 | 0.022 | 0.050 | 0     | 1.08  | 3.07 |       | 5                       | Ni, Cu, Zr       | 0.5, 0.5, 1      | 1329                                              | 189                                    | 0.43                                         |
| S73           | Ca, Sr                             | 0.396 | 0.532 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 5                       | Ni, Cu, Zr       | 0.5, 0.5, 1      | 1285                                              | 183                                    | 0.41                                         |
| S74           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.012 | 0.98  | 3.01 |       | 5                       | Cu, Fe, Zr       | 0.5, 0.5, 1      | 904                                               | 83                                     | 0.24                                         |
| S75           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.012 | 1.047 | 3.05 |       | 5                       | Cu, Fe, Zr       | 0.5, 0.5, 1      | 988                                               | 121                                    | 0.33                                         |
| S76           | Ca                                 | 0.396 | 0.537 | 0.022 | 0.045 | 0     | 1.08  | 3.06 |       | 5                       | Ni, Cu, Zr       | 0.5, 0.5, 1      | 1070                                              | 142                                    | 0.37                                         |
| S77           | Ca, Sr                             | 0.396 | 0.537 | 0.022 | 0.034 | 0.011 | 1.08  | 3.06 |       | 5                       | Ni, Cu, Zr       | 0.5, 0.5, 1      | 1300                                              | 195                                    | 0.45                                         |
| S78           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 0.98  | 3.01 |       | 5                       | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1360                                              | 174                                    | 0.39                                         |
| S79           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.09  | 3.07 |       | 5                       | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1500                                              | 205                                    | 0.41                                         |
| S80           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.12  | 3.09 |       | 5                       | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | Not densified                                     |                                        |                                              |
| S81           | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08  | 3.07 |       | 0                       | -                | 0                | Not densified                                     |                                        |                                              |

First crystal phase (mother phase):  $(K_aNa_bLi_cE1_dE2_{d2})_eNbO_f$   
(E1 and E2 are at least one of Ca, Sr, and Ba;  $a+b+c+d1+d2=1$ ; e is any value; f is any value that forms a perovskite)  
Second crystal phase (subphase):  $K_{0.85}Ti_{0.85}Nb_{1.15}O_5$

Results of thermal cycle evaluation test

| Sample        | First crystal phase (mother phase) |       |       |       |       |       |      |      | Subphase content (mol%) | Additional metal |      | Electromechanical coupling coefficient Kr |                     |
|---------------|------------------------------------|-------|-------|-------|-------|-------|------|------|-------------------------|------------------|------|-------------------------------------------|---------------------|
|               | Elements E1, E2                    | a     | b     | c     | d1    | d2    | c    | f    |                         | Metal            | mol% | Initial value                             | After thermal cycle |
| S04 (Fig. 12) | -                                  | 0.490 | 0.490 | 0.020 | 0     | 0     | 1.00 | 3.00 | -                       | 0                | 0.31 | 0.10                                      | -67.7               |
| S31 (Fig. 12) | -                                  | 0.500 | 0.500 | 0     | 0     | 0     | 1.00 | 3.00 | Cu                      | 1                | 0.37 | 0.12                                      | -67.6               |
| S32 (Fig. 12) | Ca                                 | 0.421 | 0.518 | 0.022 | 0.039 | 0     | 1.07 | 3.06 | -                       | 0                | 0.43 | 0.32                                      | -25.6               |
| S61 (Fig. 16) | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 | Zr                      | 1                | 0.42 | 0.32                                      | -23.8               |
| S62 (Fig. 16) | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 | Cu, Zr                  | 0.5, 1           | 0.50 | 0.40                                      | -20.0               |
| S63 (Fig. 16) | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 | Fe, Zr                  | 0.5, 1           | 0.42 | 0.36                                      | -14.3               |
| S64 (Fig. 16) | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 | Zn, Zr                  | 0.5, 1           | 0.46 | 0.34                                      | -26.1               |
| S65 (Fig. 16) | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 | Cu, Fe, Zr              | 0.5, 0.5, 1      | 0.52 | 0.44                                      | -15.4               |
| S67 (Fig. 16) | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 | Fe, Zn, Zr              | 0.5, 0.5, 1      | 0.47 | 0.37                                      | -21.3               |
| S68 (Fig. 16) | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 | Cu, Fe, Zn, Zr          | 0.5, 0.5, 0.5, 1 | 0.51 | 0.45                                      | -11.8               |
| S69 (Fig. 16) | Ca                                 | 0.449 | 0.478 | 0.022 | 0.050 | 0     | 1.08 | 3.07 | Cu, Fe, Zr              | 0.5, 0.5, 1      | 0.48 | 0.43                                      | -10.4               |

First crystal phase (mother phase):  $(K_aNa_bLi_cE1_{d1}E2_{d2})_xNbO_f$ (E1 and E2 are at least one of Ca, Sr, and Ba;  $a+b+c+d1+d2=1$ ; e is any value; f is any value that forms a perovskite)Second crystal phase (subphase):  $K_{0.85}Ti_{0.85}Nb_{1.15}O_5$

| Sample | First crystal phase (mother phase) |       |       |       |       |       |      | Subphase content (mol%) | Additional metal |                  | Relative permittivity $\epsilon_{33}T/\epsilon_0$ | Piezo-electric constant $d_{33}$ (pC/N) | Electro-mechanical coupling coefficient $K_r$ | Porosity vol% | Dielectric breakdown voltage kV/mm |
|--------|------------------------------------|-------|-------|-------|-------|-------|------|-------------------------|------------------|------------------|---------------------------------------------------|-----------------------------------------|-----------------------------------------------|---------------|------------------------------------|
|        | Elements E1, E2                    | a     | b     | c     | d1    | d2    | e    | f                       | Metal            | mol%             |                                                   |                                         |                                               |               |                                    |
| S90    | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07                    | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1150                                              | 105                                     | 0.36                                          | 1             | 6                                  |
| S91    | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07                    | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1200                                              | 160                                     | 0.38                                          | 0.5           | 7                                  |
| S92    | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07                    | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1500                                              | 233                                     | 0.49                                          | 0             | 9                                  |
| S68    | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07                    | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1550                                              | 253                                     | 0.51                                          | 0             | 9                                  |
| S93    | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07                    | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1550                                              | 240                                     | 0.49                                          | 0             | 9                                  |
| S94    | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07                    | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1400                                              | 130                                     | 0.3                                           | 0             | 5                                  |
| S95    | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07                    | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1350                                              | 120                                     | 0.28                                          | 0             | 4                                  |
| S96    | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07                    | Cu, Fe, Zn, Zr   | 0.5, 0.5, 0.5, 1 | 1350                                              | 110                                     | 0.27                                          | 0             | 2                                  |

First crystal phase (mother phase):  $(K_aNa_bLi_cE1_dE2_{d2})_eNbO_f$

(E1 and E2 are at least one of Ca, Sr, and Ba;  $a+b+c+d1+d2=1$ ; e is any value; f is any value that forms a perovskite)

Second crystal phase (subphase):  $K_{0.85}Ti_{0.85}Nb_{1.15}O_5$

FIG. 19

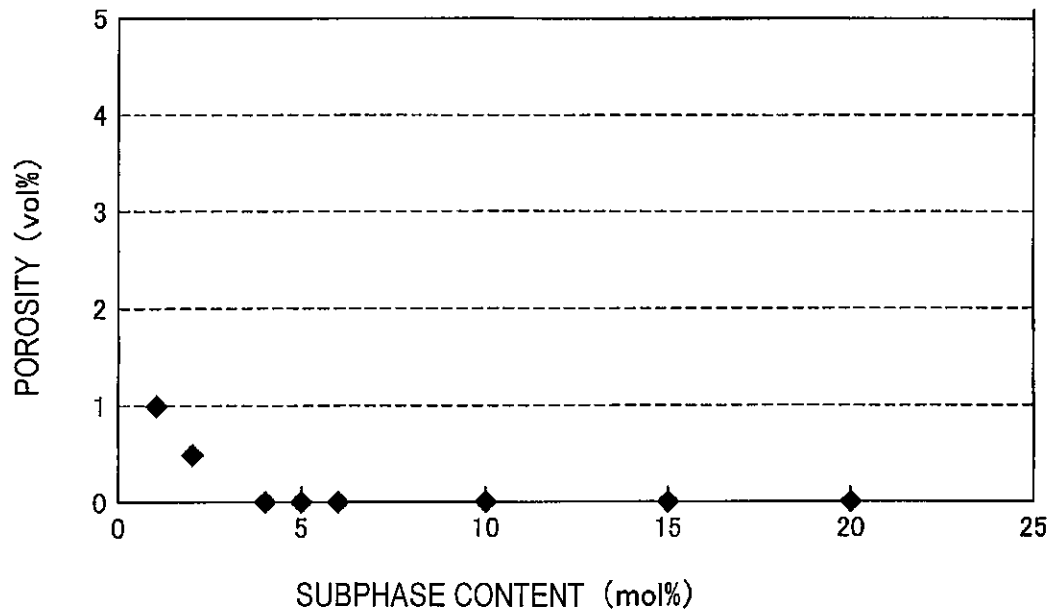


FIG. 20

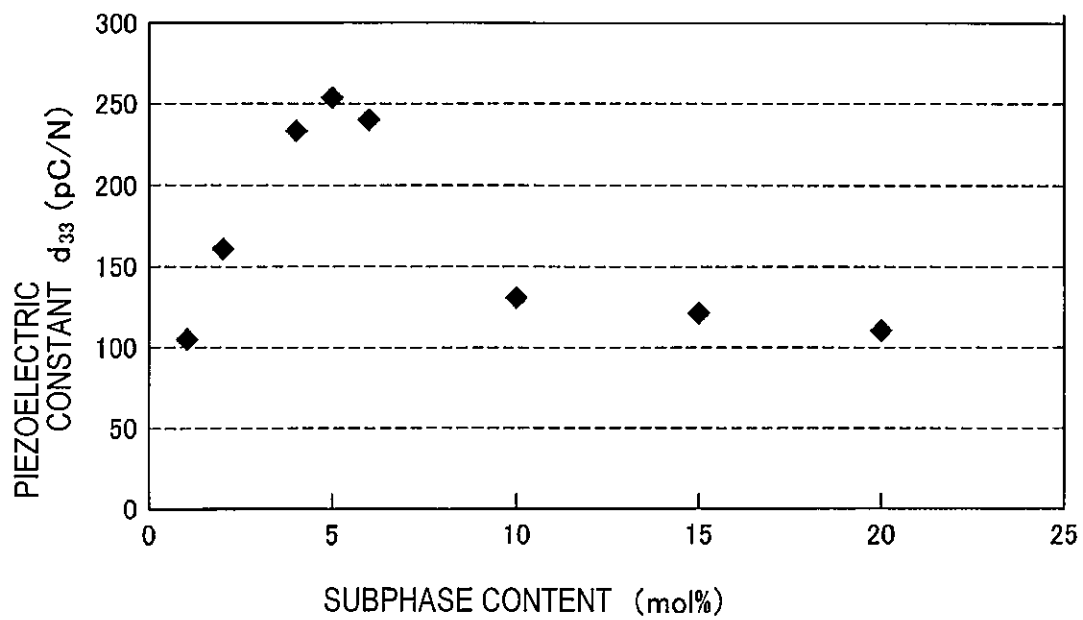
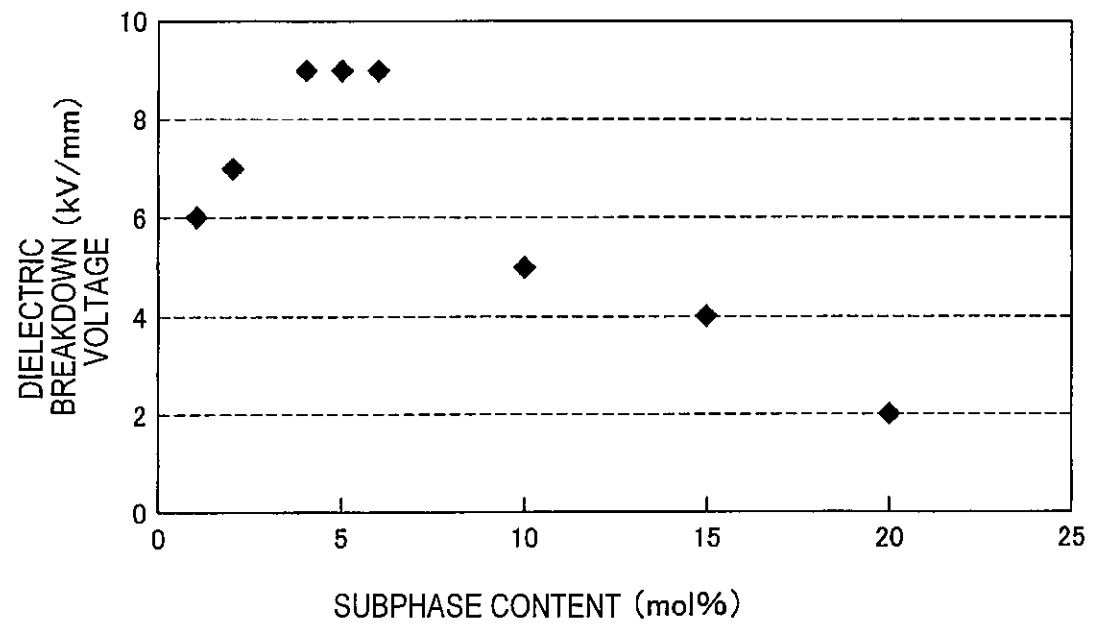
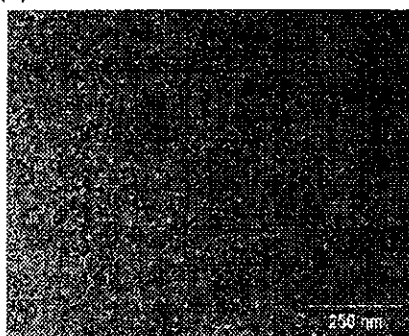


FIG. 21

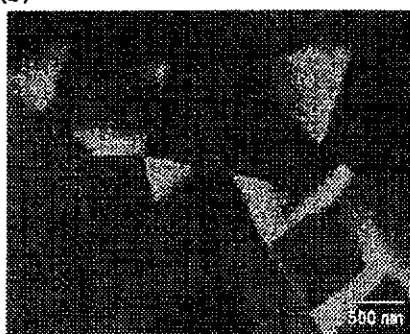


*FIG. 22*

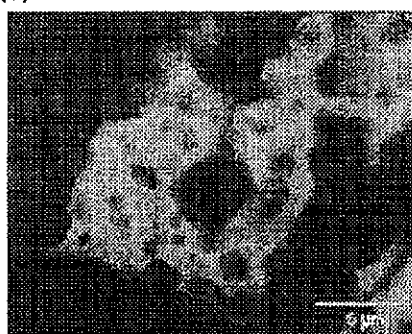
(a)



(b)



(d)



(c)

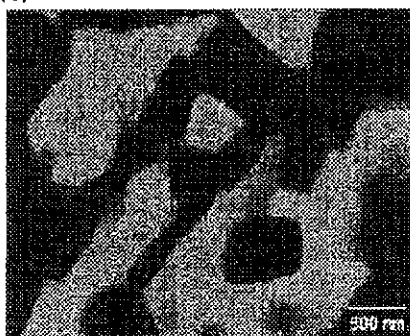


FIG. 23

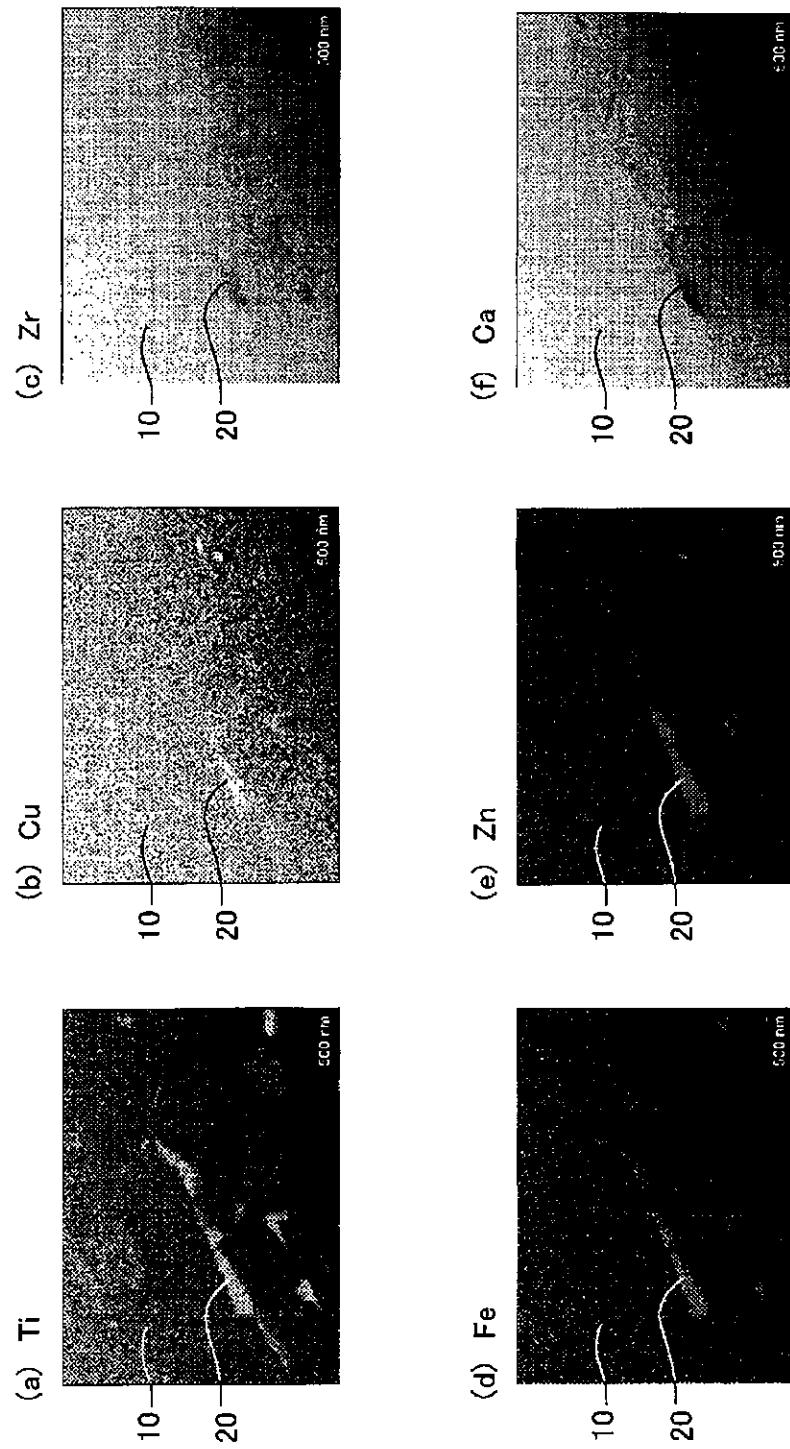
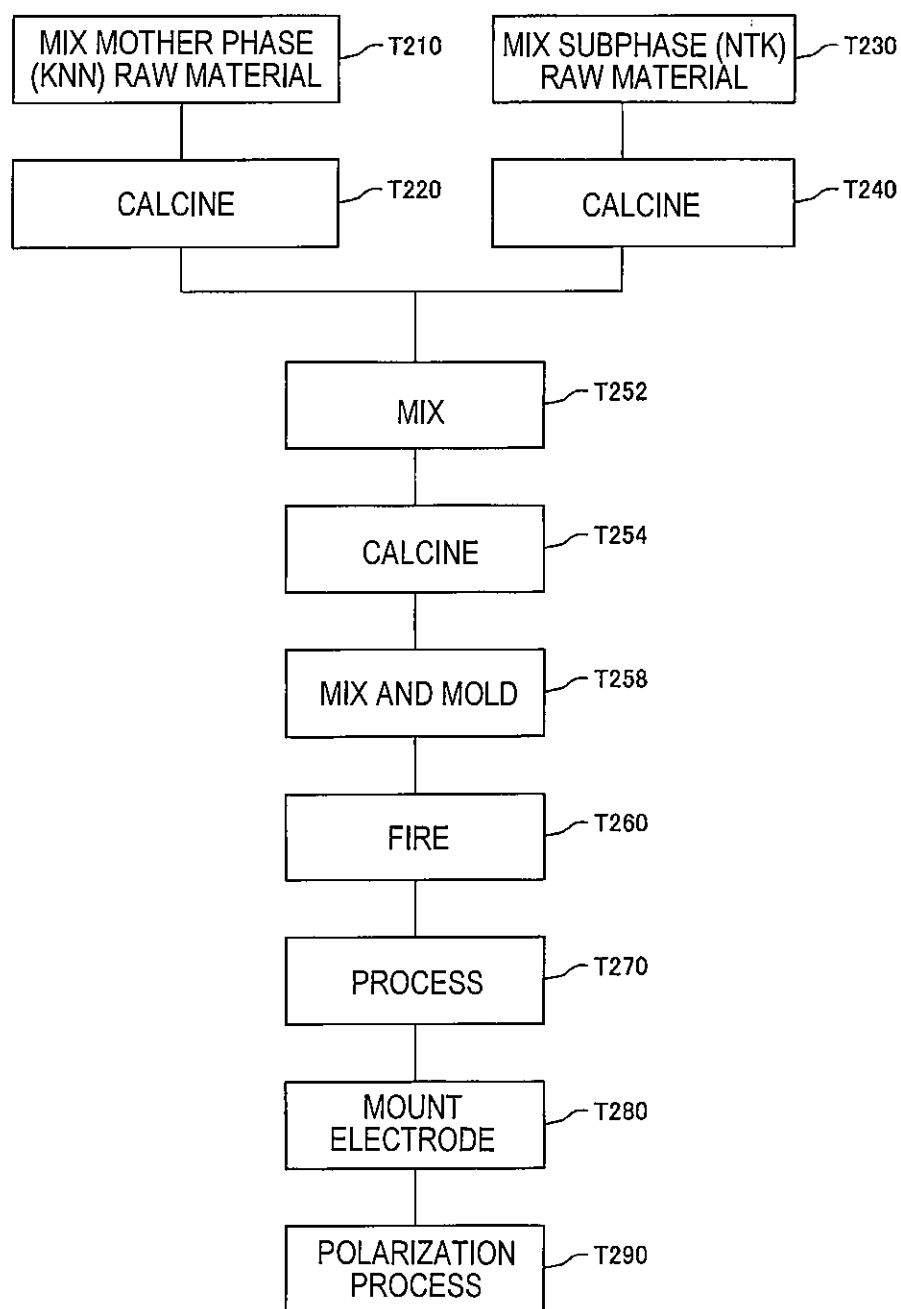


FIG. 24



| Sample        | First crystal phase (mother phase) |       |       |       |       |       |      |      |  |    | Subphase content (mol%) | Additional metal |                  | Relative permittivity $\epsilon_{33}/\epsilon_0$ | Compliance $\epsilon_{33}^*$ (pm <sup>2</sup> /N) | Piezoelectric constant $d_{31}$ (pC/N) | Electromechanical coupling coefficient $k_r$ | Calcining after mixing mother phase and subphase |
|---------------|------------------------------------|-------|-------|-------|-------|-------|------|------|--|----|-------------------------|------------------|------------------|--------------------------------------------------|---------------------------------------------------|----------------------------------------|----------------------------------------------|--------------------------------------------------|
|               | Elements E1, E2                    | a     | b     | c     | d1    | d2    | e    | f    |  |    |                         | Metal            | mol%             |                                                  |                                                   |                                        |                                              |                                                  |
| S04 (Fig. 12) | -                                  | 0.490 | 0.490 | 0.020 | 0.000 | 0.000 | 1.00 | 3.00 |  | 0  |                         | -                | 0                | 430                                              | 4                                                 | 95                                     | 0.31                                         | Absent                                           |
| S101          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 |  | 5  |                         | Co, Zn, Zr       | 0.5, 0.5, 1      | 1427                                             | 14                                                | 243                                    | 0.49                                         | Absent                                           |
| S102          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 |  | 1  |                         | Co, Zn, Zr       | 0.5, 0.5, 1      | 1023                                             | 5                                                 | 105                                    | 0.31                                         | Present                                          |
| S103          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 |  | 3  |                         | Co, Zn, Zr       | 0.5, 0.5, 1      | 1140                                             | 8                                                 | 153                                    | 0.36                                         | Present                                          |
| S104          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 |  | 5  |                         | Co, Zn, Zr       | 0.5, 0.5, 1      | 1490                                             | 15                                                | 256                                    | 0.52                                         | Present                                          |
| S105          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 |  | 8  |                         | Co, Zn, Zr       | 0.5, 0.5, 1      | 1520                                             | 12                                                | 200                                    | 0.44                                         | Present                                          |
| S106          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 |  | 15 |                         | Co, Zn, Zr       | 0.5, 0.5, 1      | 1610                                             | 7                                                 | 120                                    | 0.32                                         | Present                                          |
| S107          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.05 | 3.05 |  | 5  |                         | Co, Zn, Zr       | 0.5, 0.5, 1      | 1520                                             | 15                                                | 262                                    | 0.52                                         | Present                                          |
| S108          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.022 | 1.08 | 3.08 |  | 5  |                         | Co, Zn, Zr       | 0.5, 0.5, 2      | 1720                                             | 12                                                | 276                                    | 0.49                                         | Present                                          |
| S109          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 |  | 5  |                         | Co, Zr           | 0.5, 1           | 1334                                             | 10                                                | 190                                    | 0.4                                          | Present                                          |
| S110          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.004 | 1.08 | 3.06 |  | 5  |                         | Co, Zn           | 0.5, 0.5         | 1110                                             | 9                                                 | 177                                    | 0.4                                          | Present                                          |
| S111          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.004 | 1.08 | 3.06 |  | 5  |                         | Co, Fe           | 0.5, 0.5         | 1244                                             | 9                                                 | 181                                    | 0.37                                         | Present                                          |
| S112          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.004 | 1.08 | 3.06 |  | 5  |                         | Co, Cu           | 0.5, 0.5         | 1053                                             | 8                                                 | 188                                    | 0.41                                         | Present                                          |
| S113          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 |  | 5  |                         | Co, Cu, Zr       | 0.5, 0.5, 1      | 1405                                             | 10                                                | 222                                    | 0.42                                         | Present                                          |
| S114          | Ca, Ba                             | 0.449 | 0.478 | 0.022 | 0.039 | 0.011 | 1.08 | 3.07 |  | 5  |                         | Co, Zn, Fe, Zr   | 0.5, 0.5, 0.5, 1 | 1504                                             | 11                                                | 225                                    | 0.42                                         | Present                                          |

First crystal phase (mother phase):  $(K_aNa_bLi_cFe_{1-d1}E_{2-d2})NbO_f$

(E1 and E2 are at least one of Ca, Sr, and Ba;  $a+b+c+d1+d2=1$ ; e is any value that forms a perovskite)

Second crystal phase (subphase):  $K_{0.85}Ti_{0.85}Nb_{1.15}O_5$

FIG. 26

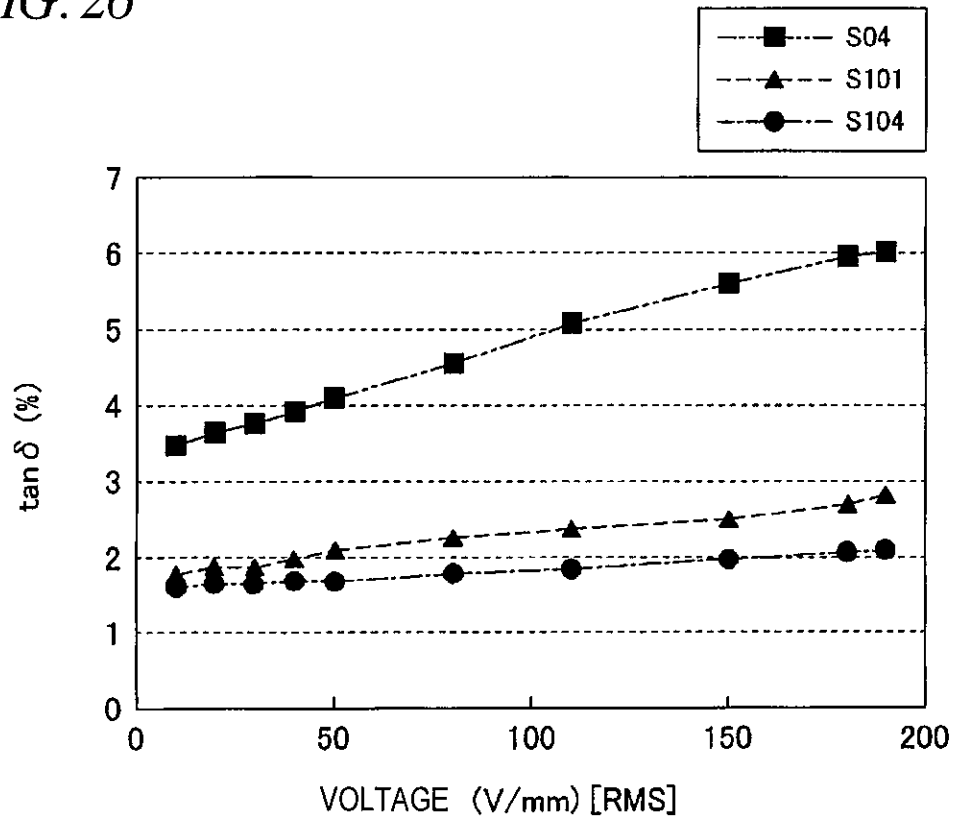


FIG. 27

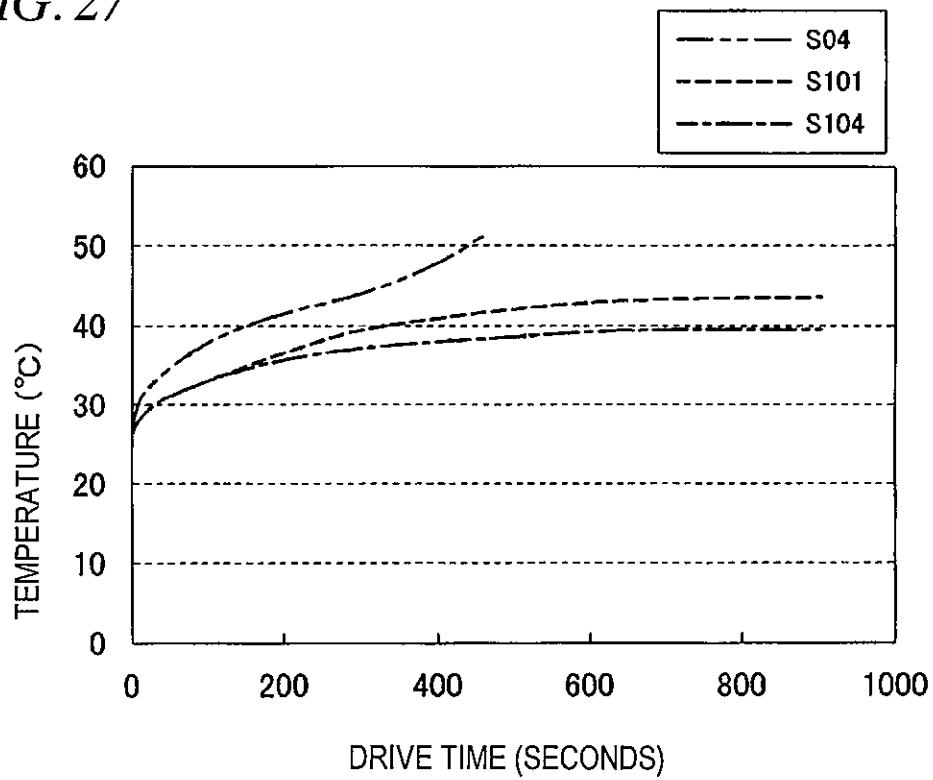


FIG. 28

