The sintering behavior and dielectric characteristics of Pb(Li_{1/4}Fe_{1/4}W_{1/2})O_3-added Pb(Mg_{1/3}Nb_{2/3})O_3 ceramics were investigated. Mixing the two end-members for 4h only formed a mixture, however, 48h-mixing mechanochromically induced the reactants to form partial perovskite solid-solutions. In the mixture specimen, the melt of Pb(Li_{1/4}Fe_{1/4}W_{1/2})O_3 acted as a reactive liquid phase to substantially enhance the densification of Pb(Mg_{1/3}Nb_{2/3})O_3, and caused grains to grow greatly and result in nonuniform microstructure. Regardless of mixing conditions, pure perovskite solid-solutions were formulated at 1000 °C when the content of Pb(Li_{1/4}Fe_{1/4}W_{1/2})O_3 was less than 10 mass%, whereas, adding excess Pb(Li_{1/4}Fe_{1/4}W_{1/2})O_3 unfavorably caused polyhedral pyrochlore phase to form in the matrix. The addition of Pb(Li_{1/4}Fe_{1/4}W_{1/2})O_3 decreased the broad maximum of dielectric permittivity of sintered specimens; nevertheless, this addition markedly suppressed the temperature dependence of dielectric permittivity. The ceramics fabricated by the mixture of two end-members possessed a larger apparent diffusiveness of dielectric permittivity than those prepared from the partial solid-solution, implying that a wider compositional fluctuation likely occurred in the former specimen.

Key-words : Pb(Mg_{1/3}Nb_{2/3})O_3, Pb(Li_{1/4}Fe_{1/4}W_{1/2})O_3, Sintering, Microstructure, Dielectric properties
position and sintering conditions were studied. Furthermore, the temperature and frequency dependencies of dielectric permittivity of sintered ceramics were investigated. A modified equation between dielectric permittivity and temperature was utilized to analyze the relaxor characteristics of obtained ceramics.

2. Experimental procedures

Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ was synthesized via a conventional mixed-oxide route after heating at 710°C for 8h. Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$ was prepared by heating the columbite MgNb$_2$O$_6$ with a stoichiometric amount of PbO at 800°C for 8h. After Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ and Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$ were prepared separately, they were weighed according to the following formula: weight of Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$: weight of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ = (1-x): x (x=5, 10 and 20%). The weighed powder was ball-milled via ZrO$_2$ balls (2 mm in diameter) with ethanol. The powder prepared by ball-milling for 4h is designated as system A, and 48h as system B. The sample codes are AP$_x$ and BP$_x$ for systems A and B, respectively, and x is the containing amount of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ (see Table 1). Following drying in a rotary evaporator, the mixed powder was uniaxially pressed into disks under 196MPa. The pressed disks were sintered in an Al$_2$O$_3$ crucible at 900 to 1100°C for 2h. The pellets were covered by Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$ powder to limit the evaporation of PbO during sintering.

The heat-treated specimens were ground to powder and analyzed via X-ray powder diffraction (XRD) analysis using Cu K$_\alpha$ radiation. The percentage of pyrochlore phase was calculated from the ratio of the intensity of the (222) diffraction peak of pyrochlore phase to the sum of the intensities of the (222) peak of pyrochlore phase and the (110) peak of Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$. The lattice parameters were determined from powder diffraction data using silicon as an internal standard. The microstructural variation in the sintered samples was studied by a scanning electron microscope (SEM). A dilatometer was utilized to examine the shrinkage behavior of pressed pellets. The percent linear change (PLC) of each specimen was calculated from the equation: PLC = ($L_T - L_0$) / $L_0$, where $L_T$ is equal to the length at temperature $T$ and $L_0$ the original length. The differential coefficient of expansion (DCE) was defined as DCE = ($L_T - L_{T-20}$) / $L_0$ / 20, where $L_{T-20}$ corresponded to the length at temperature ($T-20$)°C. The compositional analysis was semi-quantitatively carried out via an energy dispersive X-ray spectroscopy (EDS). The dielectric permittivities and dissipation factors of sintered ceramics were measured by an impedance analyzer (HP4192A). This measurement was carried out from 100 to -80°C, and automatically recorded on a personal computer.

### Table 1. Relative Amount of Pyrochlore Phase for Each Specimen at Various Conditions

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>PMN</th>
<th>AP5</th>
<th>AP10</th>
<th>AP20</th>
<th>BP5</th>
<th>BP10</th>
<th>BP20</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb(Li$<em>{0.5}$Fe$</em>{0.5}$W$_{0.5}$)O$_3$</td>
<td>0</td>
<td>5</td>
<td>10</td>
<td>20</td>
<td>5</td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td>Added Amount (mass%)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Heating Condition</th>
<th>The relative amount of pyrochlore phase (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sintered at 900°C</td>
<td>0.0  0.8  4.6</td>
</tr>
<tr>
<td>Sintered at 1000°C</td>
<td>0.0  0.8  4.6</td>
</tr>
<tr>
<td>Sintered at 1100°C</td>
<td>3.8  8.9  7.3</td>
</tr>
</tbody>
</table>

* The relative amount of pyrochlore phase was calculated from $I_{222}/(I_{222} + I_{110})$.

Fig. 1. X-ray diffraction patterns of as-milled specimens in (a) system A and (b) system B. (PMN: Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$, PLFW: Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$, and PY: pyrochlore).
tures ranging from 900 to 1100°C. The XRD results of sintered specimens are summarized in Table 1. In Pb(Mg\(_{1/3}\)Nb\(_{2/3}\))O\(_3\) and AP5, only perovskite phase was observed at all sintering temperatures. On the other hand, pyrochlore phase appeared in AP10 after sintering at 1100°C. As for AP20, pyrochlore phase was formed at sintering temperatures above 900°C, and its amount was increased with temperature. In BP5 and BP10, a small amount of pyrochlore phase was formed at 900°C. In BP20, pyrochlore phase was found at all sintering temperatures.

The results of the density of sintered specimens are listed in Table 2. The relative density was calculated on the basis of the theoretical density of Pb(Mg\(_{1/3}\)Nb\(_{2/3}\))O\(_3\) (\(a=0.40462\) nm), 5% Pb(Li\(_{1/4}\)Fe\(_{1/4}\)W\(_{1/2}\))O\(_3\)-contained Pb(Mg\(_{1/3}\)Nb\(_{2/3}\))O\(_3\) (\(a=0.40413\) nm), 10% contained Pb(Mg\(_{1/3}\)Nb\(_{2/3}\))O\(_3\) (\(a=0.40397\) nm), equal to 8.155, 8.227, and 8.289 g/cm\(^3\), respectively. As for the 20% Pb(Li\(_{1/4}\)Fe\(_{1/4}\)W\(_{1/2}\))O\(_3\) added samples, the theoretical density could not be obtained because the complete solid-solution was not formed. Table 2 indicates that the density of pure Pb(Mg\(_{1/3}\)Nb\(_{2/3}\))O\(_3\) reached only about 90% even after 1100°C-sintering. On the other hand, the maximum relative density of 97% was attained in both systems A and B when 10 mass% Pb(Li\(_{1/4}\)Fe\(_{1/4}\)W\(_{1/2}\))O\(_3\)-containing Pb(Mg\(_{1/3}\)Nb\(_{2/3}\))O\(_3\) was sintered at 1000°C. At the identical amount of Pb(Li\(_{1/4}\)Fe\(_{1/4}\)W\(_{1/2}\))O\(_3\) and the same sintering conditions, system A exhibited a higher density than system B. On sintering at 900°C, increasing the amount of Pb(Li\(_{1/4}\)Fe\(_{1/4}\)W\(_{1/2}\))O\(_3\) from zero to 20 mass% significantly increased the density from 6.11 to 7.46 g/cm\(^3\) in system A. On the other hand, in system B with 20 mass% of Pb(Li\(_{1/4}\)Fe\(_{1/4}\)W\(_{1/2}\))O\(_3\), the density of ceramics only 6.59 g/cm\(^3\). Raising the sintering temperature to 1000°C resulted in an increase of the density for all samples. The density of AP20 and BP20 became 8.06 g/cm\(^3\). When the temperature was further elevated to 1100°C, the density of pure Pb(Mg\(_{1/3}\)Nb\(_{2/3}\))O\(_3\) and AP5 and BP5 was increased; however, that of AP10, AP20, BP10, and BP20 was slightly decreased.

3.2 Thermal analysis and microstructural observation

The linear change percentage and differential coefficient of expansion curves for AP5, AP10 and AP20 are indicated in Figs. 2 (a) and (b). Two stages of shrinkage occurred in all samples, the first stage starting from about 800°C, and the second one from 900°C. From the curve of the differential coefficient of expansion, two distinct peaks were observed in the heating process which corresponded to the first and second stages of shrinkage. The peak temperature in the differential coefficient of expansion curves corresponded to the temperature where the maximum shrinkage rate took place. The first peak appeared at around 850°C, while the second one occurred between 925°C and 965°C. Results of the same experiments for BP5, BP10 and BP20 are depicted in Fig. 3. At a lower temperature range, as the amount of Pb(Li\(_{1/4}\)Fe\(_{1/4}\)W\(_{1/2}\))O\(_3\) was increased, the shrinkage percentage became increased. As shown in Fig. 3 (b), only one peak could be found at around 900°C for BP20, at around 1050°C for BP10. The results of Figs. 2 and 3 imply that the densification process of the samples in systems A and B proceeded through different sintering mechanisms.
The microstructures on the free surface of pure Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$ sintered at 900, 1000 and 1100°C are shown in Figs. 4(a), (b) and (c), respectively. After sintering even at 1100°C, the microstructure became merely slightly densified. Particle size was increased from 0.7 μm at 900°C to 1.1 μm at 1100°C. Figures 5(a) to (d) reveal the microstructures on the free surface of AP5, BP5, AP10 and BP10 samples sintered at 1000°C. AP5 and AP10 exhibited nonuniform microstructures with large grains grown to around 2 μm. Compared with system A at the same adding amount of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$, system B had a more uniform microstructure. The average grain size in BP5 was around 0.6 μm, and that in BP10 was around 0.8 μm. The microstructures of the free surface of 1100°C-sintered AP10, BP10, AP20 and BP20 are shown in Figs. 6(a) to (d). Large grains having a polyhedral shape were found to appear in the matrix of these four specimens. Besides the large polyhedral grains, small angular grains were also found in AP20, BP10 and BP20. Due to the limit of the spot size of EDS, the chemical composition of those submicron angular grains could not be determined accurately. The large polyhedral grains in either system A or B were analyzed by EDS to have a different chemical composition from Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$ grains in matrix. The composition of the polyhedral grains was primarily composed of lead (about 50 atom%) and niobium (about 40 atom%), with a trivial amount of magnesium. From the difference in morphology and chemical composition, these polyhedral grains are considered to be the pyrochlore phase.

3.3 Dielectric properties

Considering the sintered density and the content of pyrochlore phase of the samples obtained in this study, the
specimens sintered at 1000°C were chosen to measure the dielectric properties. The temperature dependence of dielectric permittivity for the specimens in system A at 10 kHz is illustrated in Fig. 7. As observed in this figure, with an increase in the content of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$, the dielectric maximum ($\varepsilon_{\text{max}}$) was decreased, the apparent Curie temperature ($T_m$) was decreased, and the peak of the dielectric permittivity became markedly broadened. Figure 8 reveals the dielectric permittivity of system B at 10 kHz. BP5 had a very low dielectric permittivity in light of its low density. As the amount of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ was increased, the temperature $T_m$ was also reduced. The dielectric maximum of systems A and B are summarized in Table 3. When the dielectric properties were measured at varied ac frequencies, all specimens displayed a strong frequency dependence. Increasing the frequency during measurement resulted in the suppression of the maximum of dielectric permittivity and the increase of the apparent Curie temperature.

### Table 3. Values of $\delta$, $\varepsilon_{\text{max}}$, $\delta/\varepsilon_{\text{max}}$ and $\gamma$ for Each Sintered Specimen at 10 kHz

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>PMN</th>
<th>AP5</th>
<th>AP10</th>
<th>AP20</th>
<th>BP5</th>
<th>BP10</th>
<th>BP20</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\delta$ (°C)</td>
<td>34.3</td>
<td>42.3</td>
<td>58.6</td>
<td>75.8</td>
<td>51.3</td>
<td>53.0</td>
<td>70.8</td>
</tr>
<tr>
<td>$\varepsilon_{\text{max}}$</td>
<td>17400</td>
<td>13200</td>
<td>7800</td>
<td>4700</td>
<td>4600</td>
<td>8800</td>
<td>4900</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>1.72</td>
<td>1.74</td>
<td>1.79</td>
<td>1.92</td>
<td>1.74</td>
<td>1.66</td>
<td>1.67</td>
</tr>
</tbody>
</table>

* $\delta$ and $\gamma$ are defined by the following equation: $1/\varepsilon = 1/\varepsilon_{\text{max}} - (T - T_m)/2\varepsilon_{\text{max}} \delta$.

### 4. Discussion

#### 4.1 Sintering and microstructural evolution

As illustrated in Fig. 1, prolonging the ball-milling to 48h induced the partial formation of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$–Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$ solid solution. This formation of solid-solution is considered to result from the mechanochemical effect. Since both Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ and Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$ exhibit perovskite structure, the migration of B-site cations into octahedral interstices in the close-packed structure seems to be accelerated by structural deformation and defects formed during intensive comminution, and possibly by local temperature rise induced by ball impacts. Similar mechanochemical influences on the formation of solid-solutions were also observed in the systems of ZnO and Al$_2$O$_3$ and $\gamma$-FeOOH and LiOH-H$_2$O$^{22,23}$.

After sintering processes a complete perovskite solid-solution without the presence of pyrochlore phase was formed in AP5 and BP5 up to 1100°C, and was formed in AP10 and BP10 up to 1000°C (see Table 1). On sintering at 1100°C, the perovskite solid-solutions of AP10 and BP10 became unstable and generated pyrochlore phase. In AP20 and BP20 pyrochlore phase appeared with perovskite phase from above 900°C. As a result, the solubility of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ in Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$ for forming complete solid-solution. As for BP20, a broad peak occurred at around 900°C. The occurrence of this peak implied that a small amount of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$, which did not form a solid-solution after ball-milling, would become to melt at elevated temperatures. On sintering at 900°C, system A exhibited higher density than system B at the identical composition. This phenomenon is ascribed to the presence of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ melt in system A. At higher temperatures, the density of specimens in system A was similar to that of the specimens in system B. The acceleration of densification in system B at a higher temperature range may be, in regard to the defective structure, formed by the substitution of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ in Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$. The mechanisms of densification process in specimens of systems A and B are considered to be different.

Increasing the Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ content from zero to 10 mass% did significantly increase the sintering density (see Table 2). However, the effect of Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ on the density after sintering at 1000 and 1100°C was found to be saturated when 20 mass% Pb(Li$_{1/4}$Fe$_{1/4}$W$_{1/2}$)O$_3$ was added in both systems. The saturation of densification seems to be relevant to the appearance of pyrochlore phase (see Fig. 6). The pyrochlore phase having a polyhedral shape was formed as isolated grains distributing themselves in the matrix. The faceted morphology and the isolated distribution state of pyrochlore phase tend to hinder the
sintering process to progress. The presence of pyrochlore phase was also found to cause the degradation of the density of AP10 and BP10 after 1100°C sintering.

The different present state of Pb(Li1/4Fe1/4W1/2)O3 in systems A and B resulted in various microstructures. System A exhibited larger average grain size than did system B (see Fig. 5) when 5 and 10 mass% Pb(Li1/4Fe1/4W1/2)O3 was added. Although a small amount of large grains (>1 μm) were also found in BP10, the fraction of large grains in AP10 was much higher than that in BP10. The grain-coarsening in system A was attributed to the formation of liquid phase through sintering. The dissolution and precipitation processes of Pb(Mg1/3Nb2/3)O3 grains are likely to occur in Pb(Li1/4Fe1/4W1/2)O3 melt. The growth of large grains during Ostwald ripening was accelerated by enhanced diffusion rates through liquid phase.24) The Pb(Mg1/3Nb2/3)O3-based system also experienced similar grain growth in the presence of PbO flux.15) The Pb(Mg1/3Nb2/3)O3 grains coarsened at the initial stage of liquid phase sintering would act as nuclei for rapid grain growth. The nonuniform microstructure in system A might be caused by the rapid grain growth for inducing preferred grain growth in part of the matrix, or by the inhomogeneous distribution of the liquid phase formed in the mixtures.

4.2 Dielectric permittivity and diffusiveness

As indicated in Figs. 7 and 8, the addition of Pb(Li1/4Fe1/4W1/2)O3 in Pb(Mg1/3Nb2/3)O3 reduced the maximum dielectric permittivity and the apparent Curie temperature Tm in both system A and system B. Although the dielectric maximum of formed solid solution is lower than that of pure Pb(Mg1/3Nb2/3)O3, the permittivity maximum of AP5, AP10 and BP10 still maintained around 13200, 7800 and 8800; in addition, the permittivity of AP5, AP10 and BP10 at room temperature is 9200, 5800 and 6000, respectively. These three specimens exhibit high magnitudes of dielectric permittivity; in addition, these specimens can be sintered at as low as 1000°C.

Not only the Pb(Li1/4Fe1/4W1/2)O3-containing amount, but also the sintering density, the microstructure, as well as the formation content of the pyrochlore phase significantly affected the dielectric maximum of the sintered ceramics. The higher sintering density of AP5 resulted in a higher dielectric maximum than did BP5 at 1000°C (see Table 2). When the Pb(Li1/4Fe1/4W1/2)O3 content was 10 and 20 mass%, no effect of density on dielectric constant should be considered since systems A and B had identical density at 1000°C. However, BP20 had a higher dielectric maximum than AP20. This fact is thought to be related with a lower content of pyrochlore phase generated in BP20. No pyrochlore phase existed in either AP10 or BP10 at 1000°C; nevertheless, the dielectric maximum of AP10 was lower than that of BP10. This difference in the dielectric maximum might result from other parameters such as the microstructure and grain size of sintered ceramics.

The dielectric permittivity of all specimens exhibited strong frequency dependence. As the frequency was raised, the maximum permittivity (εmax) decreased and Tm increased. The broad permittivity peaks at all measured frequencies depicted the behavior of diffuse phase transformation. Consequently, these phenomena revealed that all specimens possessed the relaxor ferroelectric properties. According to the previous study, the apparent diffuseness of permittivity can be used to accurately express the relation between dielectric permittivity and temperature at the range T>Tm. The modified permittivity–temperature equation is expressed as

\[ \frac{1 - \frac{1}{\varepsilon}}{\varepsilon \varepsilon_{\max}} = \frac{(T - T_m)^{\gamma}}{2 \varepsilon_{\max} \delta} \]

(1)

where ε is the value of permittivity, εmax the maximum value of permittivity, Tm the temperature at the maximum permittivity and δ the apparent diffuseness of permittivity.

For normal dielectrics, the value of γ is close to unity. On the other hand, for relaxor ferroelectrics, the value of γ is close to 2. Equation (1) can be transformed into

\[ \ln \left(1 - \frac{1}{\varepsilon} \right) = \gamma \ln (T - T_m) - (\gamma - 1) \ln (2 \varepsilon_{\max}) \]

(2)

Following Eq. (2), the values of ln(1/ε−1/εmax) for all 1000°C-sintered samples measured at 10 kHz were plotted against ln(T−Tm). The plotted data indicated that the linearity held remarkably well at the measured temperature range. This fact demonstrated the suitability of Eqs. (1) and (2) for the Pb(Li1/4Fe1/4W1/2)O3-Pb(Mg1/3Nb2/3)O3 system. The physical meaning of δ is considered to be equal to the difference between Tm and T where ε=2/3εmax, i.e.,

\[ \delta = T_m - \frac{2}{3}T_m - T \]

(3)

The value of δ is related to the broadness of the dielectric permittivity peak at the height of the peak equal to two thirds of the maximum value. The values of δ, εmax and γ for each specimen are listed in Table 3. In system A, the value of δ was significantly increased with an increase in the amount of Pb(Li1/4Fe1/4W1/2)O3. The identical trend of δ can be also found in system B. The variation of δ with composition confirmed that adding Pb(Li1/4Fe1/4W1/2)O3 in Pb(Mg1/3Nb2/3)O3 broadened the dielectric peak, i.e., reducing the temperature dependence of dielectric permittivity. The value of γ for all specimens was larger than unity, also indicating that these specimens belong to the family of ferroelectric relaxors.25) It was also found that the values of γ and δ of system A were larger than those of system B at the Pb(Li1/4Fe1/4W1/2)O3 content equal to 10 and 20%. Recalling that the raw materials in system A consisted of a mixture of two perovskites, and those of system B were formed as a partial solid-solution, the homogeneity of chemical composition in system A should be less than that in system B. Therefore, a larger microscopic composition-fluctuation will occur in the sublattices of the perovskite structure in system A. This compositional fluctuation would result in the broadening effect of the dielectric permittivity peak,20) thereby the specimens to behave more like relaxors. The similar effect of chemical homogeneity on the broadening of the dielectric permittivity was also observed in Ba(Ti0.8Zr0.2)O3 system.27) The sol-gel derived specimens exhibited better chemical homogeneity than the specimens prepared via solid-state reaction; therefore, the former specimens possessed a narrower dielectric-permittivity peak. Consequently, the synthesis processes strongly affect the chemical homogeneity in obtained ceramics, subsequently varying the broadening of dielectric peaks.

5. Conclusions

Mixing Pb(Li1/4Fe1/4W1/2)O3 with Pb(Mg1/3Nb2/3)O3 for 4h during ball-milling only formed a mixture; however, extending the mixing time to 48h resulted in the partial formation of a perovskite solid-solution. When the original Pb(Li1/4Fe1/4W1/2)O3 compound was present in the mixture, a melt was formed at elevated temperatures, which significantly enhanced the densification process and caused the Pb(Mg1/3Nb2/3)O3 grains to be coarsened. Adding 10 mass% Pb(Li1/4Fe1/4W1/2)O3 increased the density up to 97% of theoretical at 1000°C. Irrespective of the mixing conditions, a pure perovskite solid-solution was formed at 1000°C when the containing amount of Pb(Li1/4Fe1/4W1/2)O3 was less than 20 mass%. An excess amount of Pb(Li1/4Fe1/4W1/2)O3 present in Pb(Mg1/3Nb2/3)O3 induced the formation of pyrochlore phase. The pyrochlore phase exhibited a polyhedral morphology and was isolated from
matrix grains. This formation of pyrochlore phase was associated with the reduction in sintering density. The maximum of dielectric permittivity substantially depended on the adding amount of \( \text{Pb}(\text{Li}_{1/4}\text{Fe}_{1/4}\text{W}_{1/2})\text{O}_3 \), as well as the density, the microstructure and the formation amount of pyrochlore phase in sintered ceramics. The dielectric maximum was decreased with the addition of \( \text{Pb}(\text{Li}_{1/4}\text{Fe}_{1/4}\text{W}_{1/2})\text{O}_3 \); however, the temperature dependence of dielectric permittivity was markedly suppressed. The ceramics fabricated by the mixture of two end-components possessed a larger apparent diffusiveness of dielectric permittivity than those prepared from the partial solid-solution. A wider compositional fluctuation likely occurred in the sintered ceramics prepared from the former kind of specimens which had a less homogeneous distribution state of constituent components.

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References