Studies of Dielectric Permittivity of Y$_2$NiMnO$_6$ Ceramics for DC Bias at Various Temperatures

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**Keywords**: dielectric permittivity, composite, ceramics

**Abstract.** The dielectric permittivity ($\epsilon$) of Y$_2$NiMnO$_6$ ceramics prepared by sintering at 1400 °C over 6 to 24 hours was investigated. The response of the ceramics was measured from 1 kHz to 3 MHz, with the influence of a fixed dc bias from 0 to 1.5 V and temperature from 40 °C to -60 °C. Increasing dc bias was found to reduce $\epsilon'$ at low frequencies, while at higher frequencies the dc bias had less influence on $\epsilon'$. At 40 °C a sharp transition from high to low $\epsilon'$ occurred starting at ~100 kHz, as the temperature of the ceramic was lowered, the transition shifted to lower frequencies. This behaviour is attributed to the charge ordering of Ni$^{2+}$ and Mn$^{4+}$ ions and the thermal effect on the ions energy.

**Introduction.**

Ceramics of Y$_2$NiMnO$_6$ display magnetic and dielectric properties comparable with double perovskite exposed to an electric field and at various temperatures [1]. It is also important to note that the dielectric behaviour and activation energy of Y$_2$NiMnO$_6$ are comparable to that of charge ordered La$_2$NiMnO$_6$ [2]. In particular, the activation energy is similar to the energy needed to transfer an electron from Ni$^{2+}$ to Mn$^{4+}$ a sufficient value to substitute the direction of the polar region, indicating the conformable dielectric characteristic in Y$_2$NiMnO$_6$, which should be ascribed to the charge ordering space[3]. The charge ordered perovskite materials, including those with ferroelectric properties, have been of great interest to researchers and industry [4,5].

In this work, the Y$_2$NiMnO$_6$ ceramics sample were prepared by sintering over 6 to 24 hours [6-9]. To investigate the formation of Y$_2$NiMnO$_6$ ceramics at all conditions, dielectric measurement were used to measure dielectric permittivity ($\epsilon'$) and dielectric loss ($\epsilon''$) at various temperatures and dc bias.

**Experimental details.**

The bulk Y$_2$NiMnO$_6$ samples were synthesized using Y(CH$_3$COO)$_3$.xH$_2$O (Yttrium(III) acetate hydrate, 99.9%), Ni(CH$_3$COO)$_2$.2H$_2$O (Nickel (II)acetate tetrahydrate, 99.0%) and Mn(CH$_3$COO)$_2$.4H$_2$O (Manganese (II)acetate tetrahydrate, 99.0%). The starting precursors were mixed in deionized (DI) water with the ratio of precursors to water of 1:7.5. The mixtures were stirred at room temperature for approximately 3 hours in order to yield a homogeneous solution. Firstly, the solution was heated in air from 100 °C to 800 °C over 11 hours, and held at 800 °C for 6 hours to obtain the powder for sintering. Secondly, the temperature was ramped by 5 °C /min until the final sintering temperature of 1400 °C in air, for 6, 12, 18, and 24 hours.
In order for dielectric measurement, the ceramic samples were given electrodes by silver painting on both sides of the disk shaped samples, and they were air dried overnight at room temperature. Finally, the dielectric response of the sample was measured using the Agilent 4294A Precision Impedance Analyzer over a frequency range from 1 kHz to 3 MHz and oscillation voltage of 1.0 V. Further, the samples were subjected to an applied dc bias of 0, 0.5, 1.0 and 1.5 V.

Results and Discussion

![Graphs](image1)

**Figure 1.** Variation of the real dielectric permittivity ($\varepsilon'$) (a) 6 hours, (b) 12 hours, (c) 18 hours and (d) 24 hours with frequency of the $Y_2NiMnO_6$ ceramics at various applied dc bias.

Figure 1 demonstrates the dielectric properties that consist of dielectric permittivity of $Y_2NiMnO_6$ ceramics as a function of sintering time under applied dc bias for several frequencies. These samples were prepared by a simple thermal decomposition method at 600 °C for 6 hours followed by high sintering temperature at 1400 °C and sintering time from 6 to 24 hours (four conditions), dielectric measurements were made at 40 °C over $10^2$ to $10^7$ Hz. As seen in fig. 1(a-d), all conditions show a similar trend with $\varepsilon'$ falling as frequency increases. Samples sintered for 6, 12 and 24 hours show a steep fall in $\varepsilon'$ after $10^5$ Hz, whereas the 16 hour sample has an initial drop in $\varepsilon'$ at $10^4$ Hz followed by a plateau before falling again at $10^6$ Hz. In all cases, increasing the applied dc bias reduces the $\varepsilon'$ at frequencies below $\sim 10^4$ Hz. This affect of the dc bias on $\varepsilon'$ decays as frequency increases beyond $\sim 10^4$ Hz. Notably, samples sintered for 12 hours show high $\varepsilon'$ of 1.85 x $10^4$, 1.1x$10^4$, 1.0x$10^4$, and 9.7x$10^3$ at 0, 0.5, 1, and 1.5 V bias respectively.
Figure 2. Distinction of the real dielectric permittivity ($\varepsilon'$) at 12 hours (a) 0.0 volt, (b) 0.5 volt, (c) 1.0 volt and (d) 1.5 volt with frequency of the Y$_2$NiMnO$_6$ ceramics at various temperatures.

Figure 2 shows the dielectric permittivity ($\varepsilon'$) of Y$_2$NiMnO$_6$ ceramics sintered for 12 hours, measured at 40 °C to -60 °C with applied dc bias between $10^3$ to $10^7$ Hz. As seen in fig. 2 (a-d), a general trend of decreasing $\varepsilon'$ as frequency increases, with an initial shallow gradient followed by a steep fall. As the temperature decreases the steep fall in $\varepsilon'$ occurs at lower frequencies from $10^6$ Hz at 40 °C to $10^4$ Hz at -60 °C across all applied dc bias. Increasing the applied dc bias reduces the $\varepsilon'$ at frequencies below $\sim 10^5$ Hz across all temperatures, as it did across sintering times shown in fig. 1.
Figure 3. Difference of the dielectric loss for imaginary permittivity ($\varepsilon''$) at 12 hours (a) 0.0 volt, (b) 0.5 volt, (c) 1.0 volt and (d) 1.5 volt with frequency of the $\text{Y}_2\text{NiMnO}_6$ ceramics at various temperatures.

The imaginary part of dielectric permittivity ($\varepsilon''$) is shown in fig. 3. Samples held at 0 V bias and 40 °C display a peak in $\varepsilon''$ at $10^6$ Hz, with a local minimum of $\sim 2 \times 10^5$ at $10^5$ Hz, which rises to a local maximum of $\sim 4 \times 10^3$ at $10^3$ Hz, and then decreases at higher frequencies. This undulating trend is observed for all treatments. Lowering the temperature shifts the trend to lower frequencies, as observed with $\varepsilon'$ in fig. 2. Increasing the dc bias results in higher $\varepsilon''$ at low frequencies, with reduced influence above $\sim 10^2$ Hz. The local minimum at $10^5$ Hz increases as dc bias increases, reducing the observed peak at all temperatures. Generally, the effect of dc conduction in ceramics at low frequency on dielectric relaxation process can result in an increase in tan $\delta$ and dielectric loss. Consequently, the dielectric dispersion is very clear in both loss tangent (tan $\delta$) response and dielectric permittivity ($\varepsilon'$) response for fig. 2. This response is associated with the order-disorder type of a ferroelectric material, which is attributed to the dispersion including the motion of the ferroelectric domain boundary.

Summary

The $\text{Y}_2\text{NiMnO}_6$ ceramics measured over a temperature range 40 °C to -60 °C in order to investigate the dielectric properties and associated with heating dc bias (0.0 volt to 1.5 volt) inside materials. We found that the dielectric permittivity was higher at low frequency and low dc bias, while at high frequency and high dc bias the dielectric permittivity decreased.

Acknowledgments

The authors would like to thank the Commission of Higher Education, Ministry of Education of Thailand for the financial support. The authors also thank to Advanced Materials Physics Laboratory (Amp.), School of Physics, Institute of Science, Suranaree University of Technology, NakhonRatchasima and the division of Industrial Materials Science, Faculty of Science and Technology, Rajamangala University of Technology Phra Nakhon (RMUTP).

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