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Role of the Fe-substitution in dielectric behavior of the glass–ceramic cordierite $Mg_2Al_4Si_5O_{18}$ system

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ABSTRACT

In this study, $Mg_{2-x}M_xAl_4Si_5O_{18}$, where M=Fe and x=0.0, 0.20, 0.50, 1.0 and 1.5, have been synthesized using an arc-melter system. As a result of Fe-substitution, complex, deformed and multiphase samples were obtained. Microstructural analysis indicated that crystals of α-cordierite disappeared in the x=1.5 Fe-substituted sample. However, more densified samples than the unsubstituted sample were obtained. Dielectric constant increased gradually and capacitive resistance decreased with increasing the Fe-content in the system. The results suggested that the $Mg_2Al_4Si_5O_{18}$ materials obtained by the Fe-substitution are not suitable for high frequency applications.

1. Introduction

Dielectric properties are very important tools for terminal devices. Nowadays, the utilizable frequency range in wireless communication area has been expanded to millimeter-wave from micrometer-wave. Thus, materials with low dielectric constant (ε_r) and high quality factor (Q.f) must be used to minimize the delay time in the electronic signal transmission [1,2]. The increase of the dielectric constant causes the increase of the dielectric loss and so heating of terminal devices.

Mg-based cordierites ($Mg_2Al_4Si_5O_{18}$) are one of the promising ceramics in high frequency micro-electronic applications due to their low thermal expansion coefficient ($\alpha = 1\text{–}2\times10^{-6}\,°C^{-1}$), low dielectric constant (ε_r=5–6) and high specific resistivity ($\rho > 10^{12}\,\Omega\,cm$) [3,4]. However, it should be noted that the low dielectric constant makes this ceramic system not suitable material for some applications. The dielectric properties strongly depend on material preparation process, particle size, starting materials, stoichiometric composition, phase content etc.

Conventional glass–ceramic, solid-state reaction, precipitation parcel synthesis, sol–gel and combustion synthesis methods are used for fabrication of the $Mg_2Al_4Si_5O_{18}$ system [5–8]. Glass–ceramic technique has some advantages compared to other techniques; nonporous, high-density and homogeneous structure with strong grain connections can be easily achieved [9]. The compositional inhomogeneity and grain boundary effects can be reduced in the materials fabricated.

Some elements/aids are substituted/doped to improve the density of the material and to reduce the dielectric constant of the $Mg_2Al_4Si_5O_{18}$ system [1,8,10–15]. High densified materials have been obtained as a result of some substitutions/dopings.

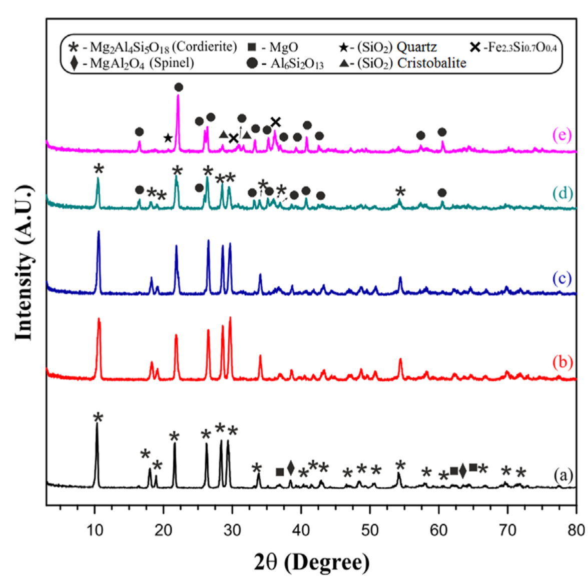

Fig. 1. XRD patterns of the $Mg_{2-x}Fe_xAl_4Si_5O_{18}$ ($0 \le x \le 2$) system heat treated at 1300 °C for 72 h. (a) x=0.0, (b) x=0.2, (c) x=0.5, (d) x=1.0 and (e) x=1.5 samples.

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It was found that the dielectric constant of the materials fabricated were lower than that of the pure $Mg_2Al_4Si_5O_{18}$ system [16].

There are reports on the effects of the substitution of Fe^{2+} for Mg^{2+} on the structural, thermal and dielectric properties of the $Mg_2Al_4Si_5O_{18}$ system in literature but few studies on the Fe^{3+} substitution for Mg^{2+} on the dielectric properties. In the present study, effects on the Fe^{3+} substitution with higher valance state than Mg on the dielectric properties in the $Mg_2Al_4Si_5O_{18}$ system were investigated in details.

2. Experimental

Glass samples with stoichiometric composition of $Mg_{2-x}Fe_xAl_4Si_5O_{18}$, where $x=0.0$, 0.20, 0.50, 1.0 and 1.5, have been prepared by melting the MgO, Al_2O_3, SiO_2 and Fe_2O_3 powders using an arc-melter system. Melting has been performed at a current of 120 A under Ar atmosphere. The undesired crystals in the glass samples were not obtained, which plays vital role for the controlled crystallization. The glass samples were heat treated at 1300 °C for 72 h with 10 °C/min heating/cooling rates in PID controlled furnace.

The X-ray diffraction (XRD) analysis of the samples was carried out using a Rigaku RadB X-ray diffractometer with Cu$K\alpha$ radiation. The scan rate was chosen as $2° \min^{-1}$ in the range of $2\theta=2–80°$. The surface morphology of the heat-treated samples was examined by JEO-Evo 40 scanning electron microscope (SEM) and their compositional analyses were performed by EDX technique. The RÖNTEC energy dispersive X-ray spectroscope was used for EDX analysis.

For determination of the dielectric constant, the samples were pelletized in 5 mm diameter and 1.5 mm thickness under 5 t. The samples were placed between two copper plates. The dielectric measurement of the samples was carried out at constant voltage of +1 V in the range of $f=100$ kHz to 1 MHz. Capacitive resistance, X_c, of the samples as a function of the frequency in the range of 100 kHz–1 MHz were measured at room temperature.

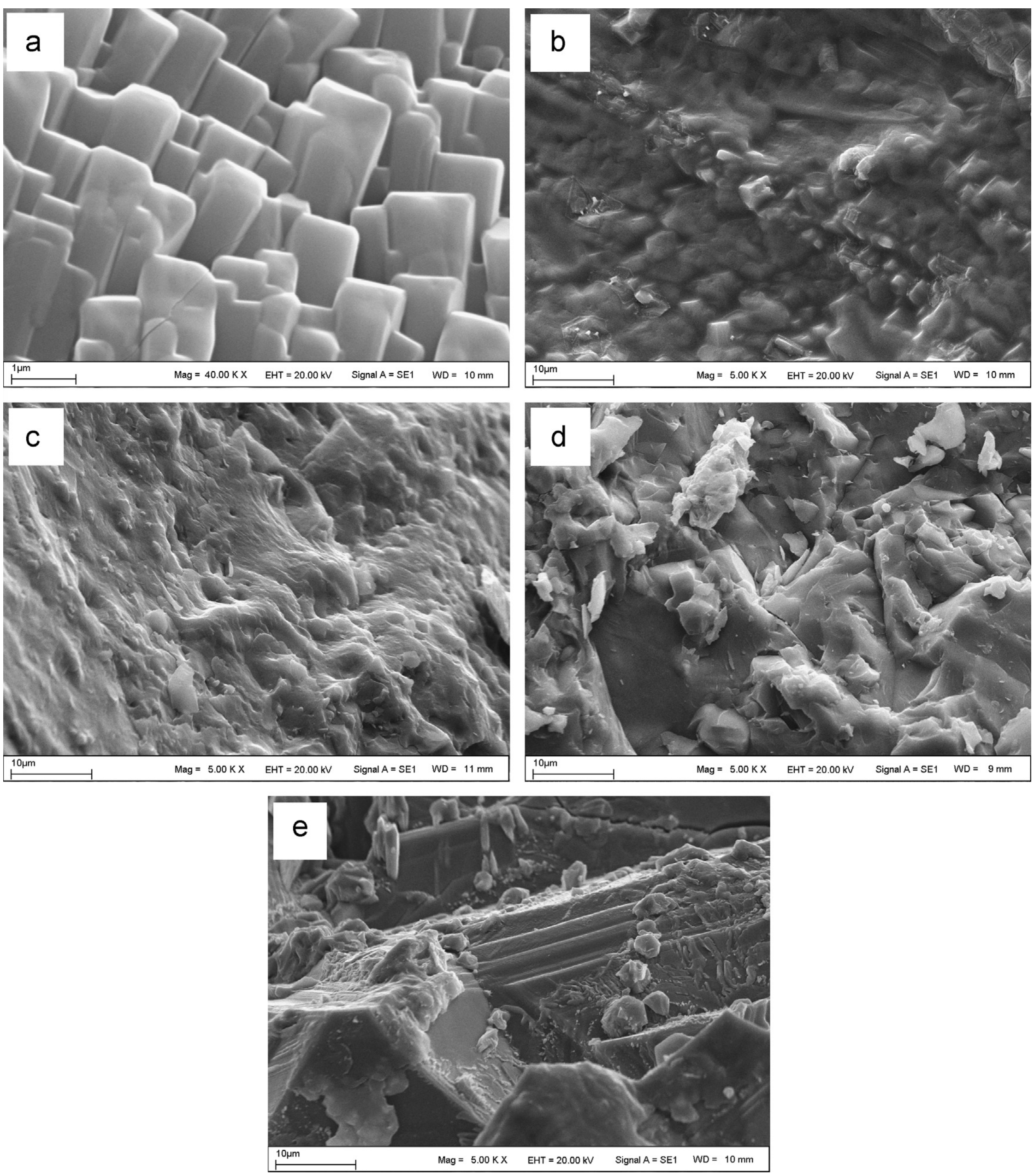

Fig. 2. SEM photographs of the $Mg_{2-x}Fe_xAl_4Si_5O_{18}$ system: (a) $x=0.0$, (b) $x=0.2$, (c) $x=0.5$, (d) $x=1.0$ and (e) $x=1.5$ samples.