

Cofiring behavior and interfacial structure of NiCuZn ferrite/PMN ferroelectrics composites for multilayer LC filters

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Abstract

The cofiring behavior, interfacial structure and cofiring migration between NiCuZn ferrite and lead magnesium niobate (PMN)-based relaxor ferroelectric materials were investigated via thermomechanical analyzer (TMA), X-ray diffractometer (XRD), scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS). Mismatched sintering shrinkage between NiCuZn ferrite and PMN was modified by adding an appropriate amount of sintering aids, Bi₂O₃, into NiCuZn ferrite. Pyrochlore phase appeared in the mixture of NiCuZn ferrite and PMN, which is detrimental to the final electric properties of LC filters. EDS results indicated that the interdiffusion at the heterogeneous interfaces in the composites, such as Fe, Pb, Zn, existed which can strengthen combinations between ferrite layers and ferroelectrics layers. © 2005 Elsevier B.V. All rights reserved.

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1. Introduction

Recently, multilayer chip LC filters have been developed as a promising surface mounting device. They are made with cofired multilayer structure of ferrite, dielectric and internal conductors. The properties of devices depend on the electromagnetic properties of the sintered ferrites, dielectrics and the microstructure of the interface. On the one hand, interfacial diffusion is inevitable, which will affect electromagnetic properties near the interface. On the other hand, the mismatch in sintering process between ferrite and dielectric may cause residual stresses or cracks. Therefore, the research on the cofiring behaviors between different ferrite and ferroelectric ceramic materials is of importance for the development of multilayer LC filters [1,2].

During the past few years, much research work was dedicated to studying the cofiring behaviors of multilayer chip LC filters based on different systems, such as ZnTiO₃-TiO₂/Co₂Z (Ba₃Co₂Fe₂₄O₄₁), BaTiO₃/Ni-Zn ferrite, PNN-PT-PZW/NiCuZn, TiO₂/NiCuZn [3–6].

However, the cofiring properties and interfacial interactions between NiCuZn ferrite and PMN ferroelectrics have not been studied. NiCuZn ferrite, which has excellent magnetic properties and can be sintered at low temperature with an appropriate amount of sintering aids, is an important material for producing MLCI [7,8]. Lead magnesium niobate (PMN)-based relaxor ferroelectrics have become one of the most important material systems in MLCC applications owing to their valuable properties, such as low sintering temperatures and high dielectric constants [9–11]. Both are promising candidates for multilayer chip LC filter applications.

In spite of many advantages, interfacial problems, much more complex than in conventional filters, may become impedance to the commercial application of the new process. In order to succeed in cofiring the composite monolithic structures, it is essential to fully consider the compatibility of the different layers in the thermal, mechanical, chemical properties and so on. Moreover, interfacial interdiffusion also has a great impact on the electronic characteristics of multilayer LC filters. Accordingly, the investigation of potential interfacial diffusion has become a crucial issue for multilayer LC filters.

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In this work, the multilayer composites were prepared by cofiring NiCuZn ferrite (abbreviated as NiCuZn) and $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ (PMN) ferroelectrics. The sintering characteristics of single ferrite and ferroelectrics were analyzed firstly. In addition, the cofiring properties, including chemical reaction, interfacial diffusion and interface microstructure of ferrite/ferroelectrics composite were investigated.

2. Experimental procedure

2.1. Specimen preparation

The NiCuZn ferrite ($\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.6}\text{Fe}_{1.96}\text{O}_4$) was prepared by the traditional solid-state reaction method. The analytical grade NiO, CuO, ZnO and Fe_2O_3 provided by Beijing Beihua Fine Chemicals Co. LTD were used as raw materials. These raw materials were mixed according to stoichiometric composition and then calcined at 700°C . In order to modify the densification properties of the NiCuZn ferrite and lower the sintering temperature, an appropriate amount of Bi_2O_3 (Beijing Beihua Fine Chemicals Co. Ltd.) was added to the NiCuZn ferrite, and then mixed in ball mill for 24 h and dried. The $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ relaxor ferroelectric ceramic powders were provided by Fenghua Electronic Ceramic Company (Guangdong, China). The NiCuZn ferrite and PMN powders were separately added with PVA for granulation, shaped into pellets (10 mm in diameter and 1.5 mm in height) and then cold-isostatic pressed at 200 MPa. The di-layer composites of NiCuZn/PMN were prepared by first pressing NiCuZn ferrite powder in a mould, then adding PMN powder and pressing again at 1.5 MPa and the resulting pellets were finally processed by cold-isostatic pressing at 150 MPa for 3 min. The resulting samples were sintered at 950°C for 4 h.

2.2. Characterization

Shrinkage curves were determined by a thermomechanical analyzer (TMA, SETARAM). The phase structure for the specimens was identified using X-ray diffractometer (XRD, RIGAKU). The microstructure and element distribution on the cross section of the composites were examined by scanning electron microscopy (SEM, JEM-6301F) in secondary-electron (SE) and back-scattering (BS) modes and energy dispersive X-ray spectroscopy (EDS), respectively.

3. Results and discussion

3.1. Shrinking behavior

Fig. 1 shows the comparison of the sintering behaviors between NiCuZn and PMN, both fired at a heating rate of $10^\circ\text{C}/\text{min}$. It can be seen that there are evident differences in the sintering shrinkage behaviors between NiCuZn and

PMN. The onset temperature of NiCuZn ferrite ($\sim 720^\circ\text{C}$) is much lower than that of PMN, and it sinters gradually and finishes up with a linear shrinkage of approximately 15%, which is larger than that of PMN, a final linear shrinkage of nearly 12%. Once PMN begins to densify, the densification is completed at a higher densification rate over a narrow temperature range compared with the shrinking process of NiCuZn. As shown in Fig. 1b, the maximum densification rate for pure PMN is much higher than that of NiCuZn. Obvious differences exist, involving initial sintering temperature, sintering densification rate, and total shrinkage. The mismatched sintering shrinkage would establish interfacial stresses and causes the warping deformation of multilayer configuration.

From the analyses above, a suitable modification of line shrinkage profiles is a prerequisite for a successful cofiring system. The sintering behavior of ferrite was favorably changed by the addition of Bi_2O_3 . Curves of NiCuZn added with 1.0 wt.% Bi_2O_3 in Fig. 1 shows that the addition of sintering aids makes the sintering behavior of NiCuZn much similar to that of PMN. The initial densification temperature of NiCuZn was slightly moved from 720 to 800°C when doped by 1.0 wt.% Bi_2O_3 and the corresponding maximum shrinkage rate was increased a little from 9.0×10^{-4} to $2.1 \times 10^{-3}^\circ\text{C}^{-1}$. A comparison between the densification behaviors of NiCuZn with 1.0 wt.% Bi_2O_3 and the pure one reveals that the sintering additives can favor-

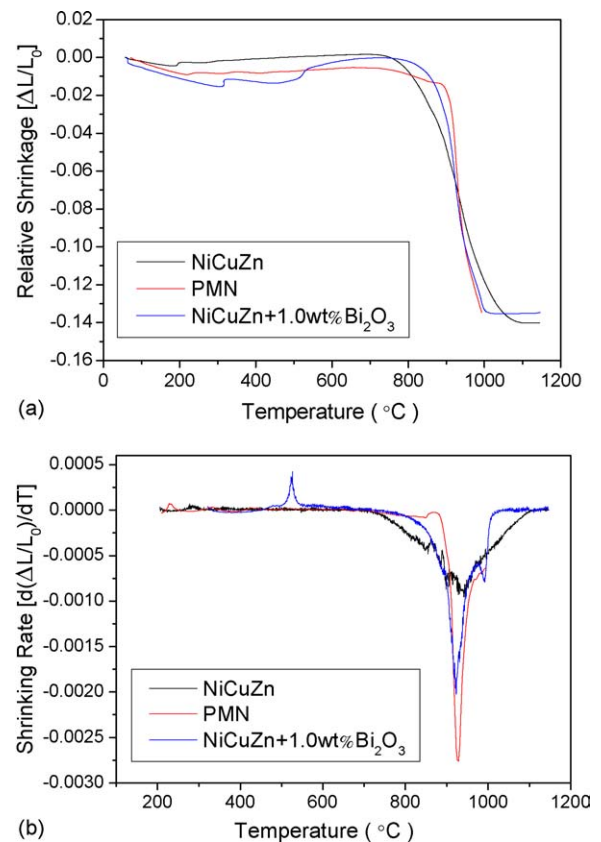


Fig. 1. Densification characteristics of NiCuZn, PMN and NiCuZn added with 1.0 wt.% Bi_2O_3 : (a) shrinkage; (b) shrinking rate.

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