Materials Research Bulletin 61 (2015) 32–35

Contents lists available at ScienceDirect

Materials Research Bulletin

journal homepage: www.elsevier.com/locate/matresbu

Sintering, microstructures and magnetic properties of low temperature co-fired NiCuZn ferrites with B_2O_3 and WO_3 additions

Haikui Zhu [a,*], Yu Jin [a], Haiou Zhu [b], Wei Shen [a], Yanqiu Xu [a], Hongqing Zhou [a]

[a] College of Materials Science and Engineering, Nanjing Tech University, Nanjing 210009, PR China
[b] Jiangsu Entry-Exit Inspection and Quarantine Bureau, Nanjing 210001, PR China

ARTICLE INFO

Article history:
Received 27 December 2013
Received in revised form 26 July 2014
Accepted 19 September 2014
Available online 10 October 2014

Keywords:
A. Electronic materials
B. Crystal growth
D. Magnetic properties
D. Microstructure

ABSTRACT

Sintering behaviors and magnetic properties of the NiCuZn ferrites with different B_2O_3 and WO_3 additions are investigated. The initial permeability values of the ferrites with 0.03 wt% B_2O_3 go up firstly from 788.77 to 913.46 as WO_3 concentration increases from 0.3 to 0.7 wt%, then drop to 458.48 for the composition with 0.9 wt% WO_3. But too much B_2O_3 or WO_3 will damage the magnetic properties. The appropriate additions of mixed B_2O_3 and WO_3 not only can efficiently increase the initial permeability (μ_i) and the saturation flux density value (B_s), but also can reduce the remanent flux density value (B_r) and the coercive filed strength (H_c) of the NiCuZn ferrites sintered at 900 °C. The ferrite sample with 0.03 wt% B_2O_3 and 0.7 wt% WO_3 possesses optimum magnetic properties: μ_i = 913.46 at 100 kHz, B_s = 284.36 mT, B_r = 103.96 mT and H_c = 38.39 A m^{-1}.

1. Introduction

Nowadays, surface-mounting devices have been rapidly developed for electronic applications, such as multilayer chip inductors (MLCIs) which are produced by the ferrites using Ag electrode [1–3]. Low temperature sintered NiCuZn ferrites have been gained popularity in the field of microelectronics due to their relatively low sintering temperature and high resistivity with good performances in the high frequency range [4,5].

In order to inhibit the interfacial reaction between the NiCuZn ferrites and Ag electrode, several ways have been developed to reduce the sintering temperature of the ceramics under the melting point of Ag electrode, such as liquid phase sintering aids, low melting glass additions, smaller size material usage and chemical processing [6–8]. Among these methods, liquid phase sintering is lower in cost and easier to process than the others [9,10]. For this purpose, some additions such as B_2O_3, Bi_2O_3, PbO and glass [11–13] are introduced into NiCuZn ferrites to lower the sintering temperature and promote the magnetic properties. B_2O_3 and WO_3 have been reported as efficient aids for the sintering process of ceramics. Kavanloui and Hashemi [14] found that 1 wt% B_2O_3 addition had a significantly effect on the densification and magnetic properties of Li–Zn ferrites sintered at 1000 °C. Hui Zhong et al. [15] reported that the substitution of WO_3 substitution on NiCuZn ferrites could improve not only electromagnetic properties but also microstructures of sintered ceramic.

It is known that the magnetic properties of NiCuZn ferrites are very sensitive to the material crystal microstructure, such as porosity and grain size. The single sintering aid can even decrease the sintering temperature of NiCuZn ferrites by form liquid phase. Nevertheless, it sometime also deteriorates the initial permeability and other magnetic properties of the ferrites. So how to choose special additions to synthetically achieve low sintering behavior and high magnetic properties is a very actual problem.

The effects of B_2O_3 and WO_3 mixed additions on NiCuZn ferrites are rarely dealt with in the open reports. Therefore, different B_2O_3 and WO_3 contents are added to NiCuZn ferrites as sintering aids and the effects of additions on densification, microstructures and magnetic properties are discussed detailedly in this study.

2. Experimental procedures

A NiCuZn ferrite with composition of $Ni_{0.2}Cu_{0.2}Zn_{0.6}Fe_2O_4$ was prepared by the conventional solid state reaction method. Reagent grade raw materials of NiO, CuO, ZnO and Fe_2O_3 with purities higher than 99 wt% were chosen as the starting materials and weighed according to the formula. After wet mixed uniformly in a ball mill and dried, the powders were calcined at 800 °C for 4 h in air. Different amounts of B_2O_3 and WO_3 additions were then added to the calcined powders and the mixtures were remilled for 12 h. The powders were mixed with an amount of polyvinyl alcohol (PVA, 10 wt%) and then uniaxially pressed into toroidal and disk-shaped samples with

* Corresponding author. Tel.: +86 25 83587762.
E-mail address: Zhuhaikui@eyou.com (H. Zhu).

http://dx.doi.org/10.1016/j.materresbull.2014.09.062

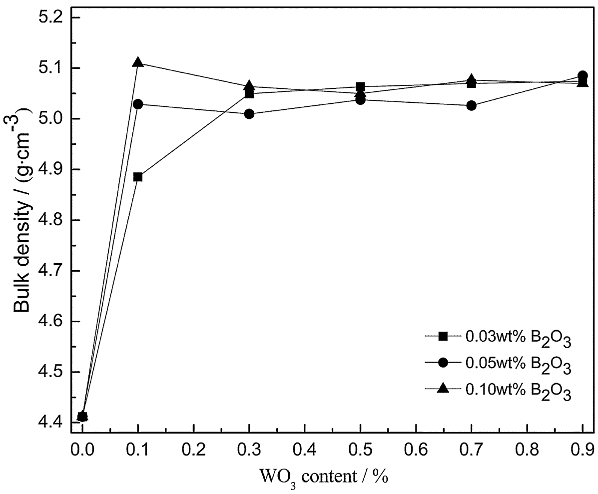

Fig. 1. Bulk densities of the ferrites as a function of different amounts of B_2O_3 and WO_3 additions.

21 mm outside diameter, 9.2 mm inside diameter and 5 mm height. These samples were sintered at 900 °C for 3 h in air.

The bulk densities were measured by Archimedes' method. Microstructures were studied on the fracture surfaces of sintered samples using a scanning electron microscope (SEM JSM-5900). The permeability and Q-factor spectra were measured at room temperature using Agilent 4294 A impedance analyzer in the frequency range of 1 kHz–20 MHz. The temperature dependence of the permeability was obtained at a constant frequency (100 kHz) over the temperature range from 25 to 150 °C. The hysteresis graph was measured using IWATSU SY-8216 B-H analyzer.

3. Results and discussion

Little addition or low sintering temperature can not improve the sintering properties of the NiCuZn ferrites, whereas too much addition in the matrix or too high sintering temperature will also damage the properties of the samples. It is found in our previous experiments that 900 °C for 3 h and less than 1 wt% (B_2O_3 + WO_3) additions are preferable to achieve the better properties of the NiCuZn ferrites. Fig. 1 shows the bulk densities of the ferrites as a function of different amounts of B_2O_3 and WO_3 additions. According to Fig. 1, small amount of the B_2O_3 and WO_3 additions dramatically improve the sinterability of the green compacts. The

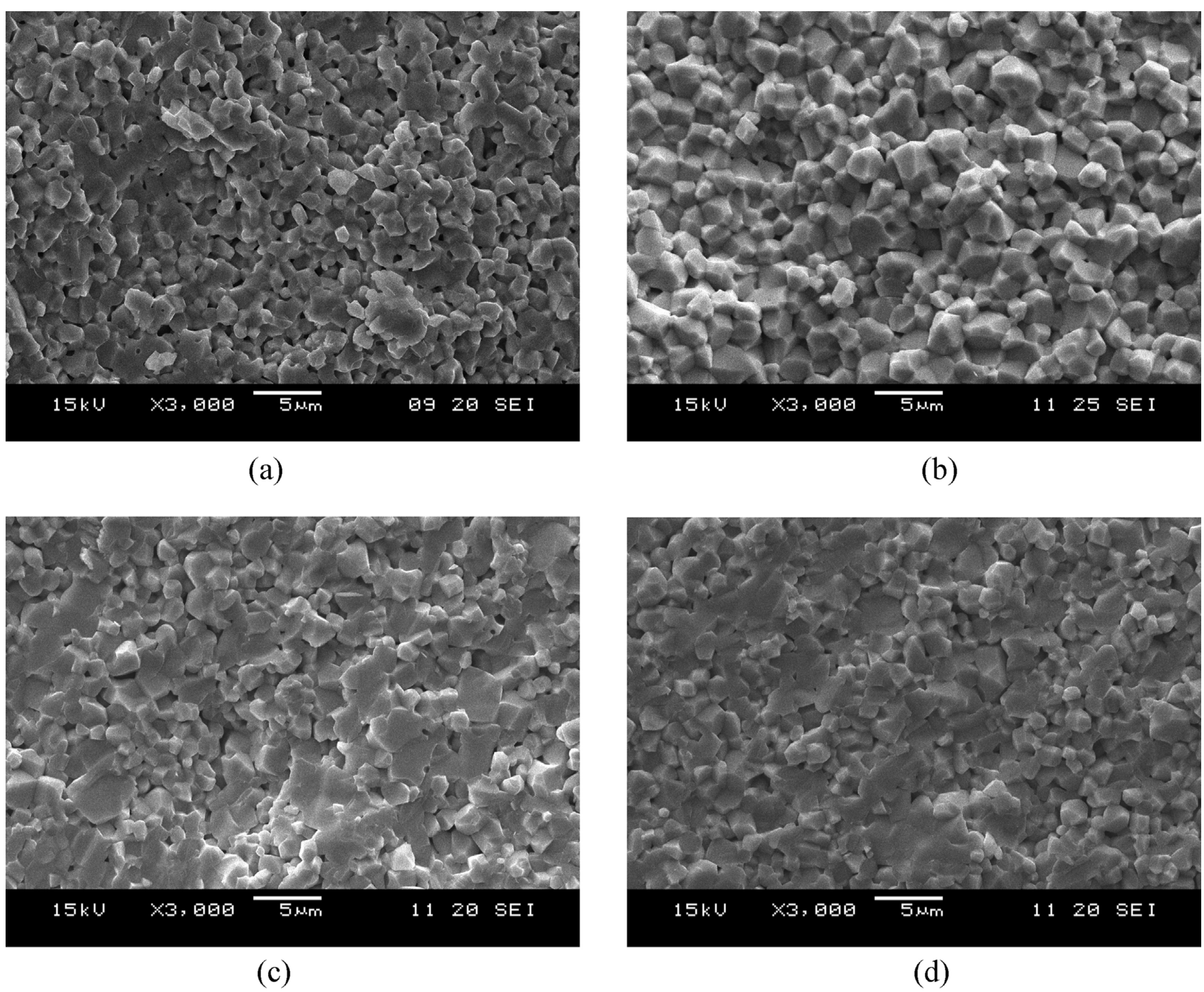

Fig. 2. SEM micrographs of the ferrites with different amount of B_2O_3 and WO_3 additions: (a) pure, (b) 0.03 wt% B_2O_3 and 0.5 wt% WO_3, (c) 0.05 wt% B_2O_3 and 0.7 wt% WO_3, (d) 0.1 wt% B_2O_3 and 0.9 wt% WO_3.