CHAPTER 1

INTRODUCTION

1.1 Overview

Ferroelectric materials have attracted considerable attention because of their possible uses in device applications such as sensors, micro-electromechanical system, nonvolatile random access memories, etc. [1]. $Pb(Zr_{1-x}Ti_x)O_3$ (PZT), having a perovskite-type structure, is an important ferroelectric material with good piezoelectric and pyroelectric properties such as high remanent polarization and high dielectric constant [1, 2]. However, PZT also Pb content has several problems such as severe polarization fatigue after bipolar switching pulses [3-5]. Bismuth layer-structured ferroelectrics are thought to be promising lead-free ferroelectric oxides materials for device applications. Among them, $Bi_4Ti_3O_{12}$ (BIT) has attracted much attention for potential utilization due to its large spontaneous polarization (P_s), low processing temperature, high Curie temperature (675°C) and high fatigue endurance [6]. The major limitation for the application of BIT is the low remanent polarization (P_r), high leakage current and high coercive field [7].

Recent studies revealed that Bi^{3+} ions in BIT structure could be substituted by donor doping in the Bi-site with La [3, 8], Dy [9, 10], Sm [11], Nd [12] or Pr [13] and in the Ti-site with Nb [14-17], V [18] or W [19] for the improvement of its ferroelectric properties. It is suggested that Bi and Ti sites doping has the different effect on the ferroelectric properties. In the case of Bi-site substitution in BIT, they exhibited enhancement of remanent polarization and excellent fatigue endurance.

However, they enhancement of remanent polarization but there was no improvement of fatigue endurance for Ti-site doping. Moreover, the doped-BIT materials generally have good fatigue endurance but low relative permittivity and ferroelectric properties lower than PZT materials [6-8].

Compounds of PZT and doped-BIT themselves are still very interesting. Many works have to be done in order to give clear understanding and bring out as much as their efficiency into their applications [20-22]. So far, many researchers have attempted to combine the fatigue-free properties of doped-BIT and superior ferroelectric and piezoelectric properties of PZT together in the form of multilayer sandwich structure thin films [23, 24]. Recently, Navavan *et al.* [25, 26] attempted to combine $Pb(Zr_{0.52}Ti_{0.48})O_3$ and La-doped BIT ($Bi_{3.25}La_{0.75}Ti_3O_{12}$; BLT) in the form of ceramics, i.e. (1-x)PZT–xBLT (when x = 0, 0.1, 0.3, 0.5, 0.7, 0.9 and 1.0) for the first time. Some improvements in ferroelectric properties were observed when a small amount of BLT was added into PZT, i.e. 0.9PZT–0.1BLT.

In this present study, extended research work on PZT–doped BIT ceramic binary system was done using different doped BIT compounds. Firstly, the Bi-site doped BIT ($Bi_{3.25}Dy_{0.75}Ti_3O_{12}$; BDT) and Ti-site doped BIT ($Bi_{3.99}Ti_{2.97}Nb_{0.03}O_{12}$; BNbT) powders were prepared and characterized. These powders were then used to prepare series of ceramics with formula (1-x)PZT–$x$doped BIT (when x = 0, 0.1, 0.3, 0.5, 0.7, 0.9 and 1.0). The ceramics will be characterized particularly in terms of phase evolution, microstructural changes, electrical properties, ferroelectric properties and their fatigue endurances. The optimum composition of PZT–doped BIT ceramic will also be reported and discussed in details. It is expected that this research would bring more understanding and give useful information on these new ceramic binary systems which can be further employed in actual applications; particularly in nonvolatile random access memories.

1.2 Objectives of this work

The main objectives of this study are to investigate physical and electrical properties of PZT–doped BIT ceramics prepared by solid state mixed oxide method. The relationships between phase evolution, microstructure, dielectric properties, ferroelectric properties and fatigue endurance of the ceramics will be investigated and discussed. The objectives of this study are as follows;

1. To prepare PZT–doped BIT (PZT–BDT and PZT–BNbT) powders and ceramics using a solid-state mixed oxide method.
2. To study the effects of processing parameters on phases, microstructural evolution, dielectric properties, ferroelectric properties and fatigue characteristics of the PZT–doped BIT ceramics.
3. To study the relationships between processing condition, phase evolution, microstructure, dielectric properties and fatigue characteristics of PZT–doped BIT ceramics.