

## CHAPTER I

### INTRODUCTION

Early in 1940s, ferroelectricity aspects of the barium titanate polycrystalline ceramics ( $\text{BaTiO}_3$ ) were studied regarding to the possession of very high dielectric constant. Within those discoveries have led to many industrialized electronic productions, such as, capacitors, electrical transducers, etc. (Haertling, 1999). More recently, the modified perovskite structure ( $\text{ABO}_3$ ) of barium titanate such as barium strontium titanate ( $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$  or BST) (Alexandru *et al.*, 2004), the effect of strontium substitution are reduction of ferroelectric transition temperature, suppression of dielectric and loss tangent for high and low frequency range, and reduction of unit cell volume. Another idea to suppress dielectric constant and loss tangent is to dope magnesium dopant in barium strontium titanate ( $\text{Ba}_{1-x-y}\text{Sr}_x\text{Mg}_y\text{TiO}_3$ ) (Berbecaru *et al.*, 2008) in order to improve the temperature dependent ferroelectric behavior in the microwave tunable devices. The nature of ceramic materials are very brittle, difficult to process and they have low electrical breakdown strength. Therefore, polymeric materials has been introduced to overcome these limitations (Yu *et al.*, 2011). The polymer themselves are flexible, easy to process and they have high electrical breakdown strength. However, the disadvantage of polymeric materials is the possession of very low dielectric constant compare to those of ceramic materials (Muralidhar *et al.*, 1987).

Therefore, composite materials were introduced in the electronic industry as a result of combination between the merits of two or more phases instead of using the best single phase materials (Newnham *et al.*, 1978). The composite materials consist of filler, which can be either fibers, flakes or particles, dispersed in a matrix composed of metal, glass or polymer (Dias *et al.*, 1996). By optimizing the materials parameters, the advantage of the ceramic which is the magnificent dielectric properties can be combined with polymeric material to enhance the flexibility and feasibility of so-called polymer-ceramic composite. Not only selecting the proper materials for a specific usage, but also concerning about the connectivity, each phase of composite material is required to be zero, one, two or three dimension self-connected (Newnham *et al.*, 1978). There are many combinations of connectivity in multiphase system. Although, the 0-3 connectivity in diphasic system is commonly

used due to the ease of fabrication and attractiveness in producing thin-film capacitor and embedded sensor (Fang *et al.*, 2009). The various types of polymer matrix phase including thermoset, thermoplastic and elastomer in the 0-3 composites were examined (Sebastian *et al.*, 2010). However, these polymers are not environmentally friendly. Since there are various concerns of global warming, biodegradable polymers are introduced in order to reduce the amount of plastic wastes.

The biodegradable polymers are commonly used in packaging and medical purposes. Although, the electrical applications of biodegradable polymer have been slightly studied. Some biodegradable polymers were investigated their utilization as electrical insulators regarding to the conductivity, dielectric properties and breakdown strength compared to those of low density polyethylene (LDPE) (Ohki *et al.*, 2007). Among those biodegradable polymer, poly(butylene succinate) (PBS) has adequate physical and electrical properties besides it has lower glass transition temperature than that of poly-L-lactic acid (PLLA) and polyethylene terephthalate succinate (PETS), which can be implied that it would fulfill the flexibility, feasibility, and more importantly, bring biodegradability to the dielectric 0-3 composites.

In this thesis work, PBS semicrystalline aliphatic polyester were used as the matrix phase. PBS is biodegradable polymer with the promising processability, flexibility and strength which can be compared to those of low density polyethylene (LDPE) (Edlund *et al.*, 2003). Moreover, the barium strontium titanate ( $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ , BST) powder were used as particulate filler to introduce dielectric properties of the matrix phase. Generally, the ceramic powder were obtained by the conventional mixed oxide method (solid state reaction), although the fabrication of ceramic by sol-gel method has been used not often (Wodecka-Duś *et al.*, 2007). The merits of this sol-gel method are the ease of compositional control, moderate processing temperature and fine-sized particle (Sharma *et al.*, 1993).

Part of this work is aimed to study the effects of strontium ( $\text{Sr}^{2+}$ ) and magnesium ( $\text{Mg}^{2+}$ ) content in the barium titanate particles synthesized by using low temperature sol-gel method. The magnesium-doped barium strontium titanate ( $\text{Ba}_{1-x-y}\text{Sr}_x\text{Mg}_y\text{TiO}_3$ ) particles with strontium content of  $x = 0.3, 0.4, 0.5$  and magnesium dopant content of 0, 0.5, 1.0 and 2.0 mol% were investigated the effect of strontium substituents and magnesium dopant on the phase formation, frequency-

dependent and temperature-dependent dielectric properties. Furthermore, high dielectric constant BST particles were used as filler phase which dispersed in poly(butylene succinate) as matrix phase in polymer-ceramic composite using 0-3 connectivity method in the composition from 0 wt% to 50 wt%. Finally, the mechanical properties, dielectric properties as the function of the filler content, frequency and temperature of the PBS-composite thin-film were investigated along with the dispersion state of PBS-composite.