CHAPTER I
INTRODUCTION

Recently, the electronic industries are corresponding to the increasing consumer demand in telecommunication and microelectronic devices which are operated at high frequency for cost effective product miniaturization. The passive electronic devices require high performance with enhance reliability, lower cost, lower size and weight that are significant efforts in research and development. But now it is possible to gain optimum properties by integrating the different materials. Current researches have prepared composite materials made from ceramics and polymers for electronic application, such as dielectric material in capacitors, because ceramics own high dielectric performance and polymers often are low cost and easily processed. Therefore, polymer–ceramic materials have stimulant much attention for using in microelectronics packaging (Castro *et al.*, 2005). As the dielectric properties are strongly influenced by the ceramic phase but it is difficult to process (Sebastian *et al.*, 2007). So, it is necessary to use polymers with low viscosity to increase the relaxation phenomena and dielectric losses (Cho *et al.*, 2004).

The piezoelectric effect is a phenomenon that related to the electric polarization (i.e. charges) that polymerization generate the noncentrosymmetric crystals when applied the stress onto the materials that usually found in quartz material (SiO_2) and poled material which has polarization properties and usually found in ceramics and some polymers such as polyvinylidene fluoride. On the other hand, it can induce strain by applied electric field (converse piezoelectric effect). The piezoelectric materials have a ferroelectric property as a ferromagnetic property. The advantages of piezoelectric materials are green energy, resistance from severe damage shock and using well at wide temperature range (Tressler, J.F. *et al.*, 1998). Ferroelectric materials with optimal dielectric constants and dissipation factors have received considerable attention, since their growing use in various electronic devices (L.C. Costa, 2010). Especially, in recent year the microelectronics devices usually made from ferroelectric ceramic–polymer composites. These materials are good candidates for embedded capacitors, capacitively coupled electrical solutions and other integrated high-frequency electronic devices. Ferroelectric ceramic–polymer composites made of barium strontium titanates $Ba_{1-x}Sr_xTiO_3$ (BST) with different polymers, such as epoxy, polyvinylidene fluoride (PVDF), polyimide, cyanoethylated cellulose polymer, etc., have been investigated for microwave substrate application (Tao *et al.*, 2007).

Materials that are used in microelectronic packaging have to simultaneously fulfill diverse requirements, such as low dielectric loss, moderate relative permittivity, moisture absorption resistance, low coefficient of thermal expansion (CTE), high dimensional stability and high mechanical stiffness (Vilkman *et al.*, 2007). Hence, polymers filled with low loss ceramics are suitable for electronic packaging for substrates device.

In this work, the attractive polymer matrix is polybenzoxazine that is the new member in the family of phenolic resins derived from the ring-opening polymerization of corresponding benzoxazine monomers. These novel types of phenolic resins have not only the characteristics of traditional phenolic resins such as excellent thermal properties and flame retardance (Ning, X., and Iahida, H., 1994) but also unique characteristics such as molecular design flexibility, low moisture absorption, near zero shrinkage upon polymerization, low melt viscosity, low coefficient of thermal expansion (CTE) and good dielectric properties. Furthermore, benzoxazines are able to polymerize simply through heating without strong acids catalyst, and without producing harmful byproducts during the cure process (Shen S. B. and Ishida H., 1996). Thus, polybenzoxazines overcome the disadvantages of the traditional phenolic resins, which leads to a novel and promising candidate for high performance thermosetting resins (Tsutomu *et al.*, 2005) that usually used as polymer substrate in electronic application (Rimdusit S. and Ishida H., 2000).

Barium strontium titanate ($Ba_{1-x}Sr_xTiO_3$ BST) is used to the ceramic filler (Heli *et al.*, 2007). The lead- free BST is a versatile ferroelectric ceramic that is related compound of barium titanate ($BaTiO_3$), formed by the substitution of strontium for barium, enabling BST to have a high dielectric constant, low dielectric loss at room temperature and high frequencies, compared with $BaTiO_3$ (Lu *et al.*,2003). Besides, it is good electrical properties and low cost (Costa *et al.*, 2010). Thus, BST is widely used in microwave devices application.

The sol-gel process has many attractive advantages which are more adjustability and a lot of possibility to obtain high purity and homogeneity materials (shaped as monolithic blocks, powders or thin layers), the composition of these can be controled perfectly and easily to process in narrow-size distribution(Yang *et al.*, 2002). So, it is the method that appropriate for prepare the powder BST in this research.

For the composites of ceramic and polymer, the dielectric constant of the composites is an important parameter for application. The dielectric constant should be as high as possible. On the other hand, the dielectric loss should be as low as possible. A Polybenzoxazine/BST nanocomposite was synthesized and characterized their dielectric properties by the research works of Panomsuwan *et al.* (2006) and Kureson *et al.* (2008). Their found that molecular structure has effect to the dielectric properties of polymer matrix (at 1 kHz) which the dielectric constant of aliphatic diamine-based polybenzoxazine (3.81) lower than aniline based polybenzoxazine and fluorine based polybenzoxazine (4.49 and 4.54, respectively). While the BST ceramic with a strontium fraction of 0.7 shows the lowest dielectric loss (Panomsuwan *et al.*,2006). According to the result of Kureson *et al.*, The higher ceramic content leads to a higher composite dielectric constant. The composite between aniline based polybenzoxazine and 80 wt% of sol-gel $Ba_{0.3}Sr_{0.7}TiO_3$ powder provides the dielectric constant as high as 39 at 1 kHz. Moreover, the surface modification process could improve the dielectric properties; the composite treat with silane coupling could showed the highest dielectric constant but the composite treat with benzoxazine monomer and phthalocyanine could showed the lower in dielectric loss. But all of their works were measured at low frequency while nowadays the telecommunication applications need to use at high frequency (microwave frequency).

The main objective of this research is to improve the dielectric properties of the polybenzoxazine composite for using at microwave frequency. In this work, the benzoxazine monomers were synthesized as aniline based and fluorinate based monomers. BST powders were prepared from the sol-gel method, followed by surface treatment in order to prevent the ceramic agglomeration. Finally the dielectric properties of polybenzoxazine-BST composite were measured at microwave frequencies.