CHAPTER I
INTRODUCTION

There has been an enormous amount of research on the creation, miniaturization and rapid operation of novel dielectric materials in recent years due to the advancement of electronic information technology (Tian, 2003). The material, at microwave frequencies, has to meet various required properties such as moderate relative permittivity and low loss tangent, which are governable and predictable; low moisture uptake; near-zero shrinkage; appropriate flexibility; as well as good dimensional stability (JS and LokeNgai, 2011). To fulfill all these requirements, the combination of dissimilar components or polymer-ceramic composites is a major candidate for this application because they possess good processability, chemical stability, and mechanical properties with excellent dielectric properties of ceramics, resulting in obtaining superior electrical characteristics with low processing temperatures and operating costs (Dang, *et al.*, 2012). In order to mass produce in electronic industries, a 0–3 connectivity (individual ceramic particles with a three-dimensionally connected polymer phase) of the composites is mostly attractive due to simply combining and shaping (Dias and Das-Gupta, 1996). A new class of thermosetting polybenzoxazines is considered suitable as a polymer matrix in composites due to their excellent mechanical and physical properties. In the past, dielectric materials were employed in several composite materials. Nevertheless, there have been very few studies of polybenzoxazines on electrical applications. In 2006, Panomsuwan and co-workers frabricated diamine-based polybenzoxazine/$BaTiO_3$ composites with 0-3 connectivity as a new type of dielectric material. The results revealed that this novel composite showed good dielectric characteristics in the frequency range of 1 kHz to 10 MHz. The dielectric constant and loss tangent of the composite with 70 wt.% of BST were 13.8 and 0.013, respectively. Krueson, *et al.* later developed the material by introducing other kinds of fluorine- and amine-based polybenzoxazine and doped barium titanate with strontium in order to shift the Curie temperature to obtain a higher dielectric constant at room temperature. This research displayed the improvement in dielectric

properties of the composites. At 80 wt.% BST loading, the dielectric constant and loss tangent at 1 kHz were 40 and 0.120, respectively.

However, limitations of polybenzoxazines in this area of research are their rigidity and brittleness. There are various alternatives to improve their drawbacks. Blending with flexible resins such as urethane elastomer (Takeichi, *et al.,* 2000), epoxy (Rimdusit, *et al.*, 2008), and polydimetylsiloxane (Ardhyananta, *et al.*, 2009) is a approach to improving toughness and extending processing capability (Ishida and Allen, 1996). Among these resins, benzoxazine-urethane copolymers are attractive because they are easy to prepare (Rimdusit, *et al.,* 2011). As another solution, preparation of high molecular weight polybenzoxazine precursor is an excellent way to overcome the shortcomings of the usual monomeric benzoxazines and so on. Until now, the study of toughen polybenzoxazine composites applied to high frequency applications has not been reported. Therefore, the challenge is to firstly prepare the feasible dielectric composites that can be used in the microwave region. In this work, amine-based (BA-a) polybenzoxazine was combined with poly(propylene glycol), tolylene 2,4-diisocyanate terminated (PPG-TDI) as urethane prepolymer to widen processing window. The effects of urethane content were observed based on dielectric properties. Additionally, high molecular weight methylenedianiline- (BA-mda) and hexamethylenediamine-based (BA-hda) polybenzoxazines would be prepared as a polymer matrix and studied dielectric behaviors as well. For enhancing dielectric constant, ceramic powder-barium strontium tinanate in the form of $Ba_{0.3}Sr_{0.7}TiO_3$ was synthesized via high purity technique, and the sol-gel method to formulate the composition. The dielectric properties under temperature and frequency variations of composites on the amount of BST loading were investigated and discussed.