CHAPTER 1: INTRODUCTION AND OBJECTIVES

1.1 Introduction to Forsterite

Ceramic is made of inorganic elements that comprises of atoms with ionic and covalent bonds. Ceramics are regarded as the oldest artificial materials with evidence of artifacts made of clay that can be traced back to 24,000 BC (Carter & Norton, 2013). Ancient Greeks used the term keramos to describe hardware that was made by materials which includes clay that requires heat treatment such as bricks, sanitary ware and tableware. Ceramics are sintered in a furnace. The porosity can be reduced by applying specific layer of coating on the ceramic surface. Ceramics are used in our daily application including construction industry, interior design industry and in the field of art.

Advanced ceramics are developed continuously for specific applications such as aerospace, nuclear, electronics and biomedical application. Highly refined raw materials with rigorous controlled composition are put through strict regulated forming and sintering process to meet the specific high mechanical strength, stiffness and enhanced toughness. Some of the commonly used advanced ceramics are silicon carbide (SiC), hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$) and silicon nitride (Si_3N_4). Si_3N_4 and SiC are used in the automotive industry due to their characteristics of low density, low thermal conductivity, low coefficient for thermal expansion and high temperature resistance. They are also commonly used in the manufacturing of turbochargers and performance disc brakes for sports cars (Riley, 2000; Xu et al., 2014) which are better options compared to other materials such steel, cast iron and aluminum alloys which generally lack high temperature strength.

Hydroxyapatite or HA is one of the advanced ceramic that received considerable attention as suitable bioceramic due to the similarity of inorganic element of the bone matrix plus the high biocompatibility with natural bone (Zhou & Lee, 2011, Lombardi et al., 2011). The biocompatibility studies on human hard tissues were carried out 30 years ago and the results generally indicated intimate bonding between human bones and HA occurred after implantation for between 4 months to 9 years. Examples of the mentioned studies include the successful implantation of human middle ear (Blitterswijk et al., 1985), successful binding of HA and human bones without damaging the fibrous tissues (Oguchi et. al., 1995), successful integration throughout the pores of HA orbital implant (Sires & Benda, 2000) and carbon matrices coated with HA which enhanced the corneal cells growing process without any inflammation problem (Sandeman et al., 2009).

HA has been proven biocompatible with human bone but the ceramic could not be employed for load bearing application including artificial knee and hip joints due to the relatively low mechanical properties of the material such as inherent brittleness, poor fatigue resistance and strength (Fathi & Hanafi, 2007). In addition, HA has low fracture toughness value between 0.7 $MPam^{1/2}$ to 1.2 $MPam^{1/2}$ compared to natural bone between 2.2 $MPam^{1/2}$ to 4.6 $MPam^{1/2}$ (LeGeros & LeGeros, 1993). The degradation of fracture toughness has been reported to be associated with the formation of secondary phases and porosity (Kobayashi et al., 2006; Thangamani et al., 2002). Studies have also revealed decreasing fracture toughness and hardness with increasing grain size because of high sintering temperature (Veljovic et al., 2009; Rodriguez-Lorenzo et al., 2002).

Due to this limitation, the search for an alternative material has focused on forsterite which shows similar potential as HA for biomedical application. Forsterite (Mg_2SiO_4) was named after the German scientist Johann Forster. Forsterite has orthorhombic structure and the end member within the olivine family (McDonnell et al., 2002). Forsterite was discovered to have improved mechanical properties compared to hydroxyapatite and bioglass which are calcium phosphate based ceramics (Fathi & Kharaziha, 2009a; Ni et al., 2007). Forsterite has higher fracture toughness of 4.9 $MPam^{1/2}$ compared with 1.2 $MPam^{1/2}$ for HA (Hench, 2005).

One of the main determining factor discovered that affects the stability of forsterite phase was the synthesized forsterite’s starting particle size (Ni et al. 2007). The long heat treatment process during powder processing phase usually caused grain growth as the powder consolidated to establish solid entity in the form of ceramic compacts. However, there was no thorough and systematic investigation done yet on the micro structural properties during the sintering and densification phase.

It is important to prevent abnormal grain growth during sintering to obtain high dense bioceramic body. The density of forsterite would be determined by the onset of grain growth during the final densification phase. In conventional pressureless sintering under air atmosphere in a box furnace, the heating process is normally slow typically lasting for more than 24 hours. During this period, the grain boundary diffusion process helped to remove the intergranular pores. At the same time, some grains start to grow very fast up to the point of consuming smaller grains. Densification is practically impossible because of the trapped pores among the large grains (Kingery et al., 1976).

The convetional pressureless sintering in air was used mostly in previous studies to produce forstertie. This consolidation phase using this sintering technique require slow heating rate at high sintering temperature and long dwell time resulting in dense body with large grain size and low mechanical properties (Kharaziha & Fathi, 2010). There are various techniques established to synthesize forsterite including the solid-state reaction of silica (SiO_2) perilcase and magnesium oxide (MgO) (Chen et al., 2015), the polymer precursor method (Tsai, 2002) and the sol-gel method (Sanosh et al., 2010). However in recent years researchers added some innovation into the forsterite synthesis method to improve the mechanical properties. Some of them include mixture of bioactive glass ($CaO–P_2O_5–SiO_2$) and nanocrystalline forsterite to form bioceramic via sol–gel processing method (Kamalian et al., 2012) and adding kaolin to enhance the properties of forsterite refractory (Bhargavi et al., 2014).

Among others, one critical step that will influence the mechanical properties of forsterite is the synthesis method which requires the combination of several steps during processing. These steps include the ball milling time, holding time during sintering and whether to apply heat treatment before the final sintering phase. Previous studies have shown forsterite powder with heat treatment followed by ball milling duration of 40 and 100 hours still shown the presence of secondary phase (Tavangarian & Emadi, 2009; 2010c). The long duration of ball milling is not somewhat feasible. Therefore a more optimized forsterite synthesis method and applying the conventional pressureless sintering method should be developed without compromising the mechanical properties.

Hence, this research is dedicated to carry out a systematic study on improving the steps during the synthesis process to produce well-defined powders that will subsequently exhibit improved sintering via the conventional method and enhanced mechanical characteristics.

1.2 Aims and Objectives of Current Research

The objective of this research is to develop a dense forsterite ceramic via conventional presureless sintering method. The research entails the formulation synthesis of forsterite composition and optimization of powder processing conditions. The investigation on the formulated forsterite will include the sintering characteristics and mechanical properties within a range of temperatures. The specific objectives of this research are as follows. Firstly, to optimize the synthesis method via the solid state route to produce a single phase forsterite ceramic powder using the conventional pressureless sintering method. Secondly, to evaluate the mechanical properties of forsterite ceramic synthesized using the optimized synthesis method. The final objective is to elucidate the sintering mechanism and grain size effect on the mechanical properties of forsterite ceramic.

1.3 Structure of the Thesis

A thorough literature review to establish the current status of the development of forsterite ceramic, particularly focusing on powder processing and sintering methods is given in Chapter 2. The experimental procedures, forsterite powder processing, sintering studies, body characterization and evaluation are presented in Chapter 3. The results and discussion comprised of two sections in Chapter 4. The first part of the chapter discussed the densification behavior of the synthesized forsterite and comparison with previous results using the conventional pressureless sintering. The second part of the chapter discussed microstructural and grain size analysis of the synthesized forsterite. The different parameters compared include different temperatures, schedule and holding time. The particle morphology, agglomeration and phase retention characteristics are deliberated as well. Sintering effect on the mechanical properties of forsterite is also described. Microstructural evaluation of the powder produced and grain morphology are discussed. The forsterite sintering process is also discussed. Lastly, Chapter 5 presents the current research results and summarizes the research findings and some suggestions for further work.