Microwave Sintered of 10Hydroxyapatite-Yttria Stabilized Zirconia-AluminaBioceramics Composites for Biomedical Applications

by

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NUR LIYANA BINTI MOHD ROSLI
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LIST OF ABBREVIATIONS

Al_2O_3	Alumina
ASTM	American Standard Testing Method
Ca	Calcium
CaO	Calcium Oxide
$Ca_3(PO_4)_2$	Tricalcium Phosphate
$Ca_4P_2O_9$	Tetracalcium Phosphate (TTCP)
$Ca_{10}(PO_4)_2(OH)_2$	Hydroxyapatite
$CaZrO_3$	Calcium Zirconate
c-ZrO_2	Cubic Zirconia
H_2O	Water
Hv	Vickers Hardness
OH^-	Hydrogen Oxide
P	Phosphate
Pt	Platinum
Mg	Magnesium
Na_2HPO_4 $7H_2O$	Sodium Monohydrogen Phosphate Heptahydrate
SiC	Silicon Carbide
SBF	Simulated Body Fluid
SEM	Scanning electron microscope
Ti	Titanium
XRD	X-ray diffraction

YSZ	Yttria stabilized zirconia

LIST OF SYMBOLS

%	Percent
˚C	Degree Celcius
A	exposed specimen area, cm^2
g/cm^3	gram per centimeter cube
GPa	Giga Pascal
MPa	Mega Pascal
min	Minutes
wt. %	Weight Percent
µm	micrometer

PensinteranKetuharGelombangMikro10HAP-YSZ-Al_2O_3BioseramikKompositUntukAplikasiBioperubatan

ABSTRAK

Ciri-cirikeserasian bio yang minimum dankesankelonggaranimplanadalahisupentingdalamkomplikasiortopedikimplan.Keperluanutamaimplanperubatanadalahsifat-sifatmekanikal yang bolehditerima yang memberikaninteraksi yang baikdenganrangkaiansekitarnyatanpamendapattindakbalastisuyang buruk.Sifatkeserasian biohydroxyapatite (HAP) yang luarbiasadiiktirafsebagaibahanimplan yang paling praktikal.Bioaktif hydroxyapatite berhadapandenganmasalahsifat-sifatmekanikalyang lemah.Kehadiran YSZ dan Al_2O_3adalahsebagaibioseramiktidakreaktifdanfizikal yang kuatdengantahapkeserasian bio yang tinggi.Penyelidikan yang berkaitandenganbioseramikinitelahterbuktisejakbeberapatahunkebelakanganmelaluipensinterankonvensional, tetapipemprosesanbioseramikkompositinimenggunakanpemanasangelombangmikrohibridyang agaksukardidapatidalamkajianliteratur.Kajianinisecarakhususberkaitandengankesanpensinteranketuhargelombangmikrokomposit 10HAP-YSZ-Al_2O_3terhadapsuhupensinteran yang berbezadanpelbagaikomposisi YSZ dan Al_2O_3pada 10% beratHAP.Pensinteranperbandingandilakukanpadasuhu 900°C, 1000°C dan 1100°C. Komposit yang mengandungi 60% beratYSZ yang disinter olehgelombangmikropadasuhu 1000°C menunjukkansifat-sifat yang paling baik, keranapenglibatan YSZ dan Al_2O_3 yang mengatasikerapuhan yang adapada HAP. Komposit 10HAP-60YSZ-Al_2O_3menunjukkanpeningkatandalamketumpatankepada 2.88g/cm3, kekerasan Vickers dankeputusankekuatanmampatanmasing-masingadalah5.68GPa dan 36.31MPa.Optimum 30% berat Al_2O_3hadirdalamkompositberkesanmengurangkantindakbalasantara HAP dan YSZ.Kebolehsintaranlebihbaikdicapaimelaluisuhupensinteranmencukupi 1000°C menunjukkankawalanmorfologi yang lebihbaikdengansedikitjumlahkeliangansemasa proses pensinteranyang dikendalikanolehpemanasangelombangmikrohibrid. Dari segikestabilanfasadidapatisamaadadenganpeningkatanjumlah YSZ melebihi 60% atausuhupensinteransehingga 1100°C akanmempercepatkanpenguraian HAP membentukTCP bersama-samadenganpembebasanCaO. KehadiranfasaCaO yang tinggijugamenyebabkanpembentukan $CaZrO_3$dan $CaAl_2O_4$.Pembentukanfasabaruinidisebabkanolehreaksi HAP dengan ZrO_2dan Al_2O_3, menyumbangkepadakemerosotandalamsifatmekanikal. Ujianin-vitro telahdijalankanuntukmengkajitingkahlakukeserasian bio 10HAP-60YSZ-Al_2O_3komposit.Analisis XRD menunjukkankompositdalamsimulasicecairbadan (SBF) telahmenyebabkankehadiranfasa HAP yang menggalakkanpenukleusanapatit.Kajianinimenyumbangkepadakebolehlaksaanpemprosesanketuhargelompangmikrodalammenghasilkanbioseramik 10HAP-60YSZ-Al_2O_3kompositdengansifat-sifatfizikaldanmekanikal yang bolehditerimadenganadanyakeserasian biountukdigunakandalamaplikasibioperubatan.

Microwave Sintered of 10HAP-YSZ-Al_2O_3Bioceramics Composites for Biomedical Applications

ABSTRACT

The minimal biocompatibility features and consequence implant loosening are the crucial issues in orthopedic implant complication. The prime requirements of medical implant are acceptable mechanical properties which impart excellent interaction with the surrounding tissue without elicit an adverse response. The remarkable biocompatibility properties of hydroxyapatite (HAP) acknowledged as the most practical implant materials. The bioactive hydroxyapatite encountered with poor mechanical properties.The presence of YSZ and Al_2O_3 areas an inert and physically strong bioceramics with high level of biocompatibility. The research associated with this bioceramics had been proven over the past years through the conventional sintering, but processing this bioceramics composites using microwave hybrid heating is rather scarce in literature. This research is specifically concerned with the effect of microwave sintered 10HAP-YSZ-Al_2O_3 composites towardsdifferent sintering temperatures and the various compositions of YSZ and $Al_2O_3$to 10 wt. % of HAP. Comparative sintering was performed at temperatures of 900ºC, 1000 ºC and 1100 ºC. Composites containing 60 wt. % microwave sintered at temperature of 1000 ºC exhibited the greatest properties, due to incorporation of YSZ and Al_2O_3which overcome the inherent brittleness of HAP. The 10HAP-60YSZ-Al_2O_3 composites indicated an increase in density to 2.88g/cm^3, Vickers hardness and compressive strength results as 5.68GPa and 36.31MPa respectively.The optimum 30 wt. % Al_2O_3inclusion in composites effectively diminished the reaction between HAP and YSZ. Better sinterability was achieved through an adequate sintering temperature of 1000°C showed better morphology control with slight amount of porosity during the sintering process facilitate by microwave hybrid heating. In terms of phase stability it was foundthat either with increasing amounts of YSZ beyond 60 wt. % or sintering temperature up to 1100ºC will hasten the decomposition of HAP to TCP together with the releasing CaO. The substantial CaO phase also results in the formation of $CaZrO_3$ and $CaAl_2O_4$. The formation of this secondary phases corresponding to the reaction of HAP betweenZrO_2andAl_2O_3, contribute to deterioration in mechanical properties. In-vitro test has been conducted to examine the biocompatibility behavior of 10HAP-60YSZ-Al_2O_3 composites. XRD analysis indicated the composites in simulated body fluid (SBF) induced a significant presence of HAP phases which promote the apatite nucleation. This research contributes to the feasibility of microwave processing in producing 10HAP-60YSZ-Al_2O_3bioceramicscomposites with acceptable physical and mechanical properties with ordinary biocompatibility requirement for biomedical applications.

CHAPTER 1

INTRODUCTION

1.1 Research Background

There is an ongoing search on improving the development of biomaterials research as in response to the rising number of patients suffering with diseased or damaged bone. The use of biomaterials assynthetic materials that are synthesized or fabricated as a purpose of implants, scaffolds, fillers, or carriers have revolutionized to aid the patients retrieve their health problem and in some cases are promising for fully recovery (Dorozhkin, 2010). Therefore, an ideal implant material is expected to impart excellent interaction with surrounding tissue without elicit adverse response, leading to intimate apposition with living tissue (Muddugangahar et al. 2011).

Prior to the current requirement of implant material, an appropriate combination of physical properties and minimal toxicity response in the host are the only recommended criteria for implant material in the early period of medical implant development (Hench & Thompson, 2010). At that time almost all materials used for implant were single phase materials but afterwards the mechanical properties of implant material and its responsible for the growth of body tissue become the importance subject matter in the biomedical field (Arifin et al., 2014).

As with increasing demand for implant, metallic implant still predominantly used for making bone implants due to their outstanding mechanical strength (Niinomi, 2003). Despite high mechanical strength reported by metal and metal alloyed based composites, poor biocompatibility resulting to loosening related failure of implant (Yang et al., 2011). For instance, the simple interlocking bonding between interface titanium and host bone as reported by Ning & Zhou, (2008). In some cases, the subsequent corrosion has attack the metal constituent of the implant by release the toxic metallic ions such as chromium, cobalt and nickel into the human body (Poinern et al., 2013). Corrosion frequently shorten the life span of implant part which could possibly necessitate revision surgery (Patel & Gohil, 2012).

At present, the crucial issues on implant regarding the minimal biocompatibility features and consequence implant loosening have encouraged the search for better materials in term of biocompatible, biological safety and economical biomaterials to treat the orthopedic complication (Ning & Zhou, 2008). For that reason, calcium phosphate based ceramics, in particular represent as hydroxyapatite (HAP) received a great deal of attention which has been directed towards the most practical implant materials (Kupiec et al., 2013). The remarkable biocompatibility properties of HAP are identified by the chemical similarity with mineralogical composition of natural bone complemented by strong chemical bonding ability with the living tissue (Fan et al., 2009). HAP form across the implant tissue interface that mimics the body's natural repair process, which offer earlier stabilization of the implant and longer functional of life (Carter, 2007). However, bioactive hydroxyapatite (HAP) encountered with poor mechanical properties compared to other common implant material (Lim et al., 2013). Evidently, HAP found to be very sensitive towards the method of preparation and sintering conditions (Borrell et al., 2014).

Thus, numerous efforts have been made to introduce a reinforcing phase into HAP composition as to improve the mechanical properties (Nouri et al., 2012). The approach of incorporate strong ceramics materials acts as good additives to improve the inherent brittleness of HAP (Atif et al., 2012). Zirconia, especially 3 mol % yttria stabilized zirconia (YSZ) can significantly enhance the mechanical properties due to its high toughness and high compression strength in which exhibits excellent resistance against crack propagation (Velmurugan et al., 2010). Desirable properties of alumina (Al_2O_3) as an inert substance, high hardness and excellent wear resistance make it useful as a biomaterial. Furthermore, with a dispersion of Al_2O_3 in YSZ matrix is expected to suppress the low temperature degradation which prevents the abnormal grain growth by controlling the microstructure (Shufeng et al., 2012). The result demonstrated no sign rejection of implant or prolapse after certain period of implantation, thereby showed positive indication of tissue ingrowth at the implant interface (Mercioniu et al., 2012; Rogojan et al., 2012). The representative of YSZ and Al_2O_3 as inert bioceramics with their good mechanical features might be useful to overcome the inherent brittleness of HAP while maintaining its bioactivity (Maccauro et al., 2011; Oktar et al., 2007).

Previous study involved with the manufacture of HAP – ZrO_2 composites usually found that fabricating this composite complicated by the decomposition factor at elevated sintering temperature with the addition of large amount ZrO_2(Reidy, 2010). To date with the bioceramics composite manufacturing, powder processing process is significant for the parts that are required to be manufactured in powder form that pose challenges for the specific applications through the incorporation of sintering (German, 1996). The efficiency of sintering process is appraised by the quality of the properties imparts to the sintered parts at lowest processing temperature and cost (Clark et al., 2011). As a consequence, a proposed solution for this issue is to employ a different sintering regime to increase the effectiveness of sintering process which can be achieved through microwave sintering.

Microwave processing technology acknowledged much attention in recent years, as evidenced by the emerging number of research on the processing different materials by microwave for particular applications. The extensive used in microwave technology in communication, further contributed to microwave energy for materials processing applications including food processing, rubber industry, ceramics, polymer, metallic materials and composites (Manoj Gupta & Leong, 2007). The applications involved with low and high temperature (>1000˚C) utilization of microwaves. The great potential of using microwave heating on the processing ceramics was discovered over 50 years ago by Von Hippel in 1954 (Lakshmanan, 2012). Voss & Tinga, (1968) showed the feasibility of sintering ceramic-glass-ceramic seals in the mid 1960s at University of Edmonton, Canada.

A significant amount of research has been conducted on identifying and understanding the problem associated with the sintering of various types of ceramics and continuously active in this matter up to present. Most of reproducible research emphasizes the microwave energy processes was fundamentally different with conventional heating processes (Borrell et al., 2014). Microwave energy was perceived to provide rapid heating, volumetric heating, improve properties of materials, reduced environmental impact of material processing and provide approaches for processing materials that are challenging to produce microstructures that cannot be achieved by the other methods (Sharan, 2009). This has mainly led towards processing cost saving because of the reduction of processing time and energy (Mondal et al., 2010). In Malaysia, Ramesh et al., (2008) has proved the same findings regarding the short sintering cycle provided by microwave heating, without noticed any grain coarsening effecttowards the samples.

Microwave heating has been well employed for material processing, as much recognition of the heating effect of microwave joining materials (Ertugrul et al., 2014; Presenda et al., 2015). A combination of multiple materials (two or more) with different compositions by promising individual functions and properties were recognized as composites (Yoruc & Sener, 2012). The feasibility of producing composites by combining the potential materials seems to have encouraging result in the area of biomedical applications, as proven by some clinical test (Bellucci et al., 2013; Campo et al., 2014). Very limited research has been reported about microwave sintering processing to fabricate the bioceramics YSZ - Al_2O_3 added to HAP based composites (Benavente et al., 2014). Therefore, this research is conducted in order to produce bioceramics composites with acceptable physical and mechanical properties, while maintaining an ordinary biocompatibility requirement for biomedical applications.