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Compression Behaviour of Pseudowollastonite-Mullite Biocomposite

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ABSTRACT. In this work, the mechanical properties of pseudowollastonite-mullite (PSW-M) composite were investigated over a range of sintering temperature. The amount of mullite was varied at different percentage 10, 20 and 30 wt.% relative to the composite content to assess the optimum amount of compressive strength. The pseudowollastonite was produced by sol gel method using calcium oxide from limestone and silica from the raw rice husk ash as a precursor. Next, pseudowollastonite and mullite was ball milled for 15 min with acetone. All different composition of PSW-M was sintered at 800 ºC, 1000 ºC, and 1150 ºC to examine the sintering effect. The phase composition of the pellet was analysed using XRD and the compression strength was measured through the universal material testing machine INSTRON 8874. It was found that, the sintering temperature had the greatest influence on the compressibility when measured over a range of mullite composition during compression test. PSW-M sample with 20 wt.% of mullite addition and sintered at temperature 1150 ºC was found to be the highest compressive strength, 30 MPa.

Keywords: *Compression, Pseudowollastonite, Mullite;*

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1. INTRODUCTION

In the quest of ideal bioactive bone implant material, significant efforts have been devoted entirely to produce bioceramic composites with better mechanical strength as well as biocompatible with the human body. Several biomaterials that were commonly used in biomedical applications as scafffold are hydroxyapatite (HA), calcium phosphate (CaP), and calcium silicate ($CaSiO_3$) based composites. Over the past few years, a stable forms of calcium silicate which also known as pseudowollastonite above temperature ~1125 ºC (α-$CaSiO_3$) has been proved as bioactive and has an excellent osteogenic properties which has been considered for dental and load bearing applications [1,2]. One of the biggest features that indicate the excellent bioactivity of the material is the ability to have a quick direct integration with bone [3]. Several study reported that, pseudowollastonite was able to stimulate the apatite formation on their surface within short induction period [4,5]. However, calcium silicate ceramics has a low mechanical strength such as fracture toughness, low compressibility and hardness which has become one of the biggest challenges [6]. Therefore, it is sensible to modify the calcium silicate in order to enhance the mechanical strength and realise

the full potential of the composite. Numerous types of polymer [7,8], metal [9], and ceramics reinforcement such as alumina [10] and zirconia [11] has been used as reinforcement to form a better mechanical strength of the biocomposite. In this study, mullite $3Al_2O_3.2SiO_2$ was selected as the reinforcing phase to enhance the mechanical properties of calcium silicate. Mullite is the refractory material that commonly used for high temperature applications which consists of alumina and silica. Mullite has much lower density (3.05 g/cm^3) than zirconia (6.1 g/cm^3) and alumina (3.95 g/cm^3), but higher hardness (15 GPa) than ZrO_2 (12 GPa), with tolerable fracture toughness (3 MPa.m$^{0.5}$). Thus, a new combination of pseudowollastonite with a high strength potential of mullite was prepared to enhance the compressibility property of the biocomposite. Besides compression test, the phase composition and morphology of the composite were examined using X-ray diffraction (XRD, D8 Advance Bruker, Germany) and field emission scanning electron microscopy (FESEM, Gemini, Zeiss Supra Series, Germany).

2. MATERIALS AND METHOD

In this study, pseudowollastonite was synthesized from the rice husk ash and limestone using the sol gel method with the ratio of CaO: SiO_2 45:55. The CaO and SiO_2 powder were mixed in distilled water and autoclaved at 135 ºC for 8 hours, dried in the oven, and the powder was sintered at temperature 1250 ºC for 1 hour to produce pseudowollastonite phase. The detail procedure of synthesizing wollastonite has been described in the previous work [12]. Mullite was simply synthesized by the calcination of the andalusite at the temperature 1450 ºC for 4 hours. The synthesized pseudowollastonite was ball milled at 400 rpm with mullite powder at 10, 20 and 30 wt.% composition for 15 min with acetone. Then, a cylindrical pellet was formed by mixing the composite powder with phosphate buffer as a lubricant using magnetic stirrer and pressed manually with the dimension of 12.5 mm height and 6 mm diameter. Next, for densification purposes, all samples were sintered at varied temperature 800, 1000 and 1150 ºC. The uniaxial compression strength of the composite was determined using 5 kN load cell of the universal material testing machine Instron 8874 at a crosshead speed of 0.5 mm min^{-1}. At least 7 specimens were tested on each group. The compressive strength was obtained using Eq. 1.

$$\sigma = \frac{F}{A} \quad (1)$$

Where F is the maximum load at failure (N) and A is the pressure area (mm^2)

3. RESULTS AND DISCUSSION

XRD analyses of the pseudowollastonite-mullite (PSW-M) system were carried out to compare the phases after sintering at different temperature (Fig. 1). Based on the XRD patterns, the sintered sample for 20 wt.% of mullite additions into pseudowollastonite identified the main crystalline phase of mullite at peak angle 26.2 and pseudowollastonite at (2θ = 27.5º, 45.6º, and 32º). However, XRD indicates the presence of gehlenite when sintered at 800 ºC and 1000 ºC. Nath et al. [13] found that the presence of gehlenite ($Ca_2Al_2SiO_7$) and CaO will cause reduction of calcium phosphate-mullite composite hardness and young modulus. Based on the XRD spectra in Fig. 1, gehlenite phase were presented at 800 ºC and 1000 ºC. Nevertheless, at 1150 ºC, only mullite and pseudowollastonite phase was present. Sintering at 1150 ºC is the maximum temperature for PSW-M composite since sintering over 1150 ºC will results in the melting and crystallizations of the composite.

The result of average compressive strength measured over a range of composition (10, 20, 30 wt.% of mullite) sintered at (800, 1000, 1150 ºC) respectively is depicted in Fig. 2. Samples sintered at temperature 800 ºC have the lowest compressive strength with only in range of 0.2-3.3 MPa across all different composition. A slight increase on the strength could be seen for PSW/M composite when sintered at 1000 ºC. However, when sintered at 1150 ºC, the obvious changes in strength could be observed for 20 and 30 wt.% of

mullite addition which abruptly increases the compressive strength up to 30 MPa and elastic modulus of 4.37 GPa.

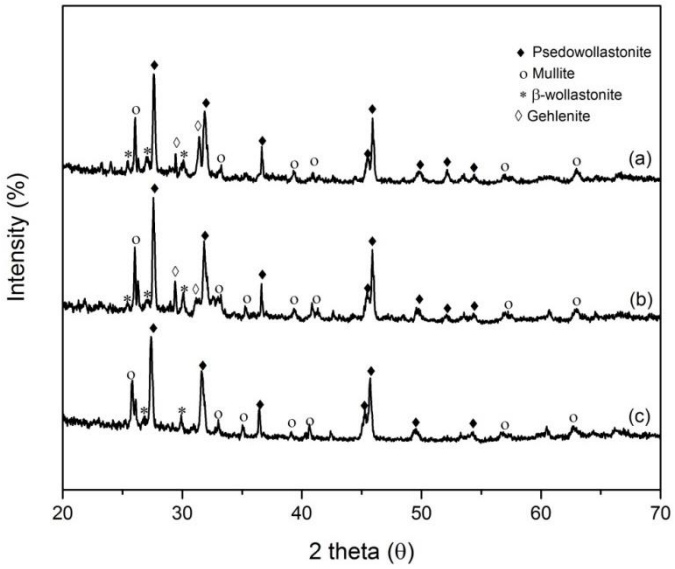

Fig. 1 XRD patterns of pseudowollastonite-20% mullite pellet sintered at (a) 800 ºC, (b) 1000 ºC, and (c) 1150 ºC

The results revealed that PSW-20M composite sintered at temperature 1150 ºC with about 92.74% of the theoretical density has the highest compression strength. Mullite has turned out to be the strengthening agent to improve the mechanical properties of the pseudowollastonite. Besides, a similar trend could be observed in other study on calcium phosphate-mullite composite whereby 30% of mullite additions has far better compressive strength compared to the pure hydroxyapatite [14]. Fig. 3 displays the agglomerated microstructure of the PSW-20% mullite pellet sintered at 800 ºC, 1000 ºC and 1150 ºC. Fig. 3 reveals that, at 800 ºC sintering, abundant of micro-porosities could be seen which were considerably reduced after sintered at higher temperature. Based on the FESEM image, the dominant process that occurred after sintering over 1000 ºC was the coarsening of particle.

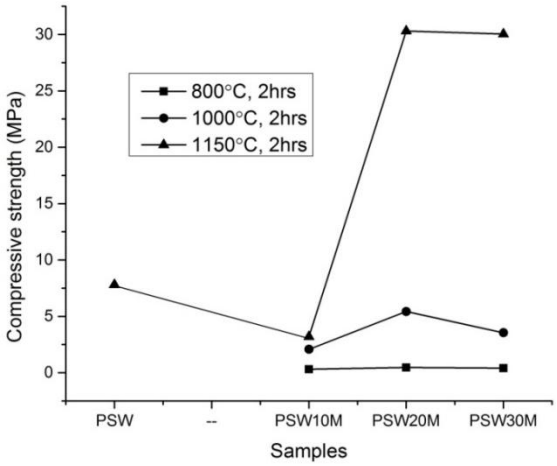

Fig. 2 Compressive strength of sintered pseudowollastonite-mullite (PSW-M) composite plotted against different compositionsThe density was measured using Archimedes principle (Table 1). As the sintering

Table 1 Densification and elastic modulus of biocomposites at different sintering temperature

Sample/ Sintering temperature (Time: 2 h)	Densification (%)	E.Modulus (MPa)
PSW, 1150 ºC	89	150.40
800 ºC		
PSW10M	78.7	16.03
PSW20M	82.8	25.02
PSW30M	81	20.75
1000 ºC		
PSW10M	79.34	160.17
PSW20M	84.94	330.21
PSW30M	85.5	179.86
1150 ºC		
PSW10M	85.06	407.74
PSW20M	92.74	4370.00
PSW30M	90.04	2870.00

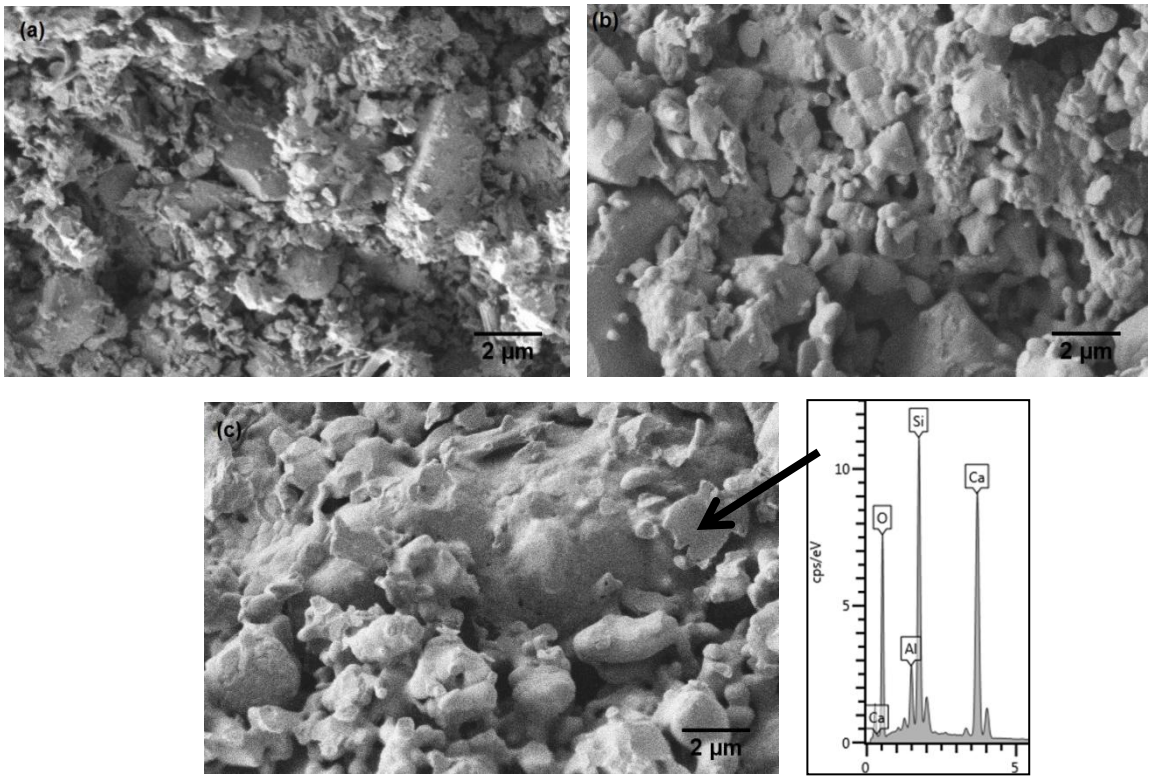

Fig. 3 The SEM micrographs and EDAX Spectrum of PSW20M sintered at temperature (a) 800 ºC, (b) 1000 ºC and (c) 1150 ºC

temperature increases, the density also increases due to the shrinkage that occur thus reduced porosity. Theoretically, as the porosity decrease, the mechanical property increases. Some research reported on the correlation of porosity rate, grain arrangement and the mechanical strength which supports this theory. The grain coarsening happen and at the meantime, the porosity rate were reduced [15]. Based on table 1, each sample represents relatively low dense. The maximum achievable density was only 92% ρ_{th} for sample P20M sintered at temperature 1150 ºC. This could be one of the possible reasons that lead to a moderately low compressive strength compared to other sintered ceramics (up to 350 MPa) [14]. Nevertheless, a great sudden increased in compressive strength after been sintered at 1150 ºC proves that porosities and densification after sintering has some influence on the mechanical properties. Thus, the optimization of sintering parameters and phases existed after sintering was part of the major concern that may affect the mechanical strength of the composite.

4. SUMMARY

The compressive strength of the developed biocomposite PSW-M has been evaluated. The increments of sintering temperature for different compositions enhance the compressive strength. The addition of 20% of pseudowollastonite-mullite composite mixture sintered at temperature 1150 ºC lead to the highest strength of biocomposite.

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