Mechanical and Bioactive Properties of Mullite Reinforced Pseudowollastonite Biocomposite

(Sifat Mekanik dan Kebioaktifan Biokomposit Pseudowolastonit Diperkuat dengan Mulit)

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ABSTRACT

Bioactive composites consist of pseudowollastonite and mullite synthesized from natural resources was developed for bone implant applications. To realize such applications, a mechanical test of these composites and in vitro bioactivity in SBF solution were studied. The present paper reports pseudowollastonite synthesized from the rice husk ash and limestone reinforced with 10, 20 and 30 wt. % of mullite. Influence of sintering temperature, phase composition, morphology towards mechanical properties of various pseudowollastonite-mullite (PSW-M) composites was examined prior to the bioactivity test. It was found that pseudowollastonite with the addition of 20 wt. % of mullite sintered at 1150°C gave the best result for diametral tensile strength (DTS) and hardness with the value of 8.8 ± 0.15 MPa and 3.79 ± 0.13 GPa, respectively. The obvious increment in the mechanical strength was due to the formation of liquid phase $CaAl_2O_3$ during sintering at 1150°C. In addition, the formation of fibrous apatite (HA) layer of amorphous calcium phosphate (ACP) with Ca/P ratio 1.8 on PSW20M sample confirmed the good bioactivity of the composite.

Keywords: Bioactivity; mechanical; mullite; pseudowollastonite

ABSTRAK

Komposit bioaktif yang terdiri daripada pseudowolastonit dan mulit disintesis daripada sumber semula jadi telah dibangunkan untuk aplikasi implan tulang. Untuk merealisasikan aplikasi ini, ujian mekanik dan kebioaktifan secara in vitro dalam larutan badan tersimulasi (SBF) bagi komposit ini telah dijalankan. Kajian pada kali ini melaporkan pseudowolastonit yang disintesis daripada abu sekam padi dan batu kapur yang diperkuat dengan 10, 20 dan 30 % bt. kandungan mulit. Pengaruh suhu sinteran, komposisi, fasa dan morfologi terhadap sifat mekanik komposit pseudowolastonit-mulit (PSW-M) telah dianalisis sebelum ujian kebioaktifan dijalankan. Keputusan mendapati pseudowolastonit dengan penambahan 20 % bt. mulit yang disinter pada suhu 1150°C memberikan hasil yang terbaik untuk kekuatan regangan diameter (DTS) dan kekerasan masing-masing dengan nilai 8.8 ± 0.15 MPa dan 3.79 ± 0.13 GPa. Peningkatan kekuatan mekanik yang ketara ini adalah disebabkan oleh pembentukan fasa cair $CaAl_2O_3$ semasa proses sinteran pada suhu 1150°C. Di samping itu, pembentukan lapisan apatit (HA) jenis kalsium fosfat amorfus (ACP) dengan nisbah (Ca / P: 1.8) bagi sampel PSW20M pada hari ke-7 membuktikan kebioaktifan yang baik daripada komposit ini.

Kata kunci: Kebioaktifan; mekanik; mulit; pseudowolastonit

INTRODUCTION

The development of bioactive ceramics has moved toward having a bioceramic material that can form a direct bond with bone and tissue as well as providing good mechanical properties (Best et al. 2008). Over the past few years, calcium silicates have been considered for low-load bearing and hard tissue replacement in biomedical application. Calcium silicate ($CaSiO_3$) or wollastonite has received much attention due to its high biocompatibility and bioactivity in the human body (Anjaneyulu & Sasikumar 2014; De Aza et al. 2006; De La Casa-Lillo et al. 2011). Calcium silicate could be synthesized using synthetic and natural sources as a precursor. Some of them were used as a filler in bone implants (Engqvist et al. 2006), a coating on inert implants such as titanium alloy (Cannillo et al. 2009; Liu & Ding 2002) or used as a bulk. However, because of many constraints and problems aroused from the coating of implants after a certain time period, the performance of bioceramics is better to be improved as a bulk.

Recent work has focused on biocomposite materials because of their associated bioactivity properties and good mechanical performances. Researchers have struggled to develop material with good biocompatibility and superior mechanical properties. Several attempts to develop biocomposite using numerous types of reinforcement including foreign oxides and metallic dispersions were reported in the literature. Several studies have reported that the integration of Ti into $CaSiO_3$ sintered at high sintering

temperature reduced the dissolution rate. Numerous researchers have explored the role and interaction of bioinert ceramic reinforcement towards hydroxyapatite (HAp) and calcium phosphate (CaP). Hardness and fracture toughness of HAp-ZrO_2 and HAp-Al_2O_3 were much improved as compared to the HAp itself after the addition of zirconia (ZrO_2) and alumina (Al_2O_3) (Kim et al. 2003; Silva et al. 2001; Zhang et al. 2006). For the bioceramics material, the brittleness possess a challenge. Therefore, it is combined with other materials to obtain good physical properties for load-bearing implant applications.

As far as the authors are aware, very little work has been carried out on the ceramic reinforcement like alumina with calcium silicate. A study has reported that, after sintering process, $CaSiO_3/Al_2O_3$ composite formed an alumina-rich calcium aluminates whereby a better hardness and toughness of the composite was attained (Shirazi et al. 2014). Recently, the application of calcium silicate and mullite as ceramic reinforcement for the biomedical application also has been highlighted by several researchers. The mullite addition into HAp favored the decomposition of HA into α/βTCP and supported the osteoconduction after 12-week implantation *in vivo*, thus became an appropriate composite for bone cell adhesion and proliferation (Nath et al. 2011). Based on the reviewed by Park and Bronzino (2002), mullite could be categorized as bioinert material and has been successfully made as a synthetic cellular solid in the form of foamed mullite which mimics the cancellous bone.

Mullite is one of the ceramic materials which are stable at the atmospheric pressure but rarely found as a mineral in nature. It is the refractory material that generally used for high-temperature applications which constitutes the alumina and silica phase. Mullite is a solid solution compound with stoichiometries of either silica-rich $3Al_2O_3 \cdot 2SiO_2$ (3:2 mullite) or alumina-rich $2Al_2O_3 \cdot SiO_2$ (2:1 mullite). Mullite has a density (3.2 g/cm³) much lower than zirconia (5.6 g/cm³) and alumina (3.96 g/cm³), hardness (15GPa) greater than ZrO_2 (12GPa), and an adequate fracture toughness (2.5 MPam0.5) (Schneider et al. 2008). The physical and mechanical properties of the calcium phosphate-mullite composite were enhanced significantly after mullite additions (Nath et al. 2012). Therefore, calcium silicate-mullite biocomposite can be developed using mullite as dispersions to produce a lightweight material with good mechanical and bioactive properties. Consequently, in this study, the effects of mullite addition on the mechanical properties and the bioactivity of the pseudowollastonite-mullite (PSW-M) biocomposites were studied.

MATERIALS AND METHODS

SYNTHESIS OF MATERIAL

The synthesis process of calcium silicate was conducted using the solid-state method. Limestone ($CaCO_3$) and rice husk ash (SiO_2, 98%) were used as a precursor for CaO and SiO_2, respectively. Calcium oxide (CaO) was obtained from the calcination of limestone and silicon dioxide (SiO_2) from the burned rice husk ash with the ratio of 55:45 as described in the previous study by Hamisah et al. (2013). The CaO-SiO_2 mixture was dried and sintered at the temperature of 1250°C for 1 h at 5°C/min which then transformed the mixture into pseudowollastonite (α-$CaSiO_3$) phases. XRD of calcined calcium silicates at 1250°C showed the pure pseudowollastonite peak. Another raw material, andalusite powder from Terengganu, Malaysia, was sintered at 1450°C for 4 h at 5°C/min using the High-Temperature Naberterm Furnace to produce a pure mullite phase. This mullite acts as the reinforcement in the pseudowollastonite matrix. The starting material pseudowollastonite and mullite have an average particle size of 26.5 and 11.11 μm, respectively. Table 1 shows some of the physical properties of pseudowollastonite and mullite used in this experiment.

TABLE 1. Physical properties of pseudowollastonite and mullite

Properties	Pseudowollastonite	Mullite
Colour	White	brownish
Average size (μm)	26.7	11.11
Melting Point (°C)	≥1300	≥1700
Density g/cm³	2.98	3.17

In the present experiments, pseudowollastonite powder is ball milled with mullite at different compositions using a rotational speed of 500 rpm for 15 min in acetone to break the agglomerates and have a homogenous, as well as fine structure. The amount of mullite in the composite is varied by (10, 20 and 30 wt. %) as stated in Table 2. Pseudowollastonite and pseudowollastonite-mullite composite powder were moulded into the stainless steel mould under a pressure of 2.5T to form a pellet with size ±13 mm diameters. Both pure pseudowollastonite and biocomposite pellet were sintered at low temperature 800°C, 1000°C and the highest temperature is 1150°C. Different sintering temperatures were used in pellet sintering and changes in mechanical properties with respect to sintering temperature was observed. The maximum sintering temperature used was 1150°C because the pseudowollastonite-mullite composites pellet will melt and deformed at 1200°C. After that, the density of the compact was measured by the Archimedes' method using water as the fluid. The samples were weighed in air and water, and the density was measured using the equation:

$$\rho = \frac{m\ water}{m\ air - m\ water}(\rho\ water = \rho\ air) + \rho\ air$$

where ρ is the density of the material; m water is the weight in grams (g) of the specimen in water; m air is the weight in grams (g) of the specimen in air; and ρ water is the density of water. The relative density was then calculated by

TABLE 2. Sample designation for different composition of PSW-mullite composite

Sample designation	Composition (%)		Sintering condition
	Pseudowollastonite	Mullite	
PSW	100	0	800°C, 1000°C, 1150°C for 2 h
PSW10M	90	10	800°C, 1000°C, 1150°C for 2 h
PSW20M	80	20	800°C, 1000°C, 1150°C for 2 h
PSW30M	70	30	800°C, 1000°C, 1150°C for 2 h

comparing the measured data and the theoretical densities ($CaSiO_3$ = 2.905g/cm^3, Mullite = 3.2 g/cm^3).

CHARACTERIZATION OF SINTERED BIOCOMPOSITE

The element composition of the synthesized pseudowollastonite and mullite was examined using X-Ray Fluorescence (XRF). Phase compositions of prepared samples were identified by X-ray diffraction (XRD) (BRUKER, D8 ADVANCE) (Karlsruhe, Germany) with a CuKa radiation (k = 0.154 nm) and a Ni filter, at a working voltage of 40 kV and a working current of 30 mA.

Microstructural characterizations were carried out using field emission scanning electron microscope (FESEM) (Merlin) accompanied with the energy dispersive spectroscopy (EDAX).

MECHANICAL ASSESSMENT OF THE PSW/MULLITE BIOCOMPOSITE

There are many mechanical tests to measure the tensile strength of specimen especially bioceramics. For instance, four-point bending test, flexural test and diametral compression test (DCT). In diametral compression test, the result of tensile strength for a brittle material is more consistent as compared to the common method such as the four-point bending test (Hsu et al. 2007; Osendi & Baudı́n 1983). This is because, in DCT method, the bulk material is exposed to the tensile stresses. Whereas in four-point bending test, only the maximum tensile stresse is subjected on the surface of the specimen. Thus, the strength values may be greatly affected by the surface imperfection if any (Marghussian & Sheikh-Mehdi Mesgar 2000). According to Bieniawski and Hawkes (1978), the value obtained from the DCT method is much lower than that from the flexural test, but higher than that from the direct tensile test. Thus, lower strength values in DCT test is anticipated. According to Harabi and Chehlatt (2013), the relationship between measured tensile strength (σ_t) value and its equivalent flexural (bending) strength (σ_f) is given in (1);

$$\sigma_f(MPa) = 2.7\sigma_t \ (MPa) \tag{1}$$

In this experiment, the tensile strength of the material is examined using diametral compression test (DCT). The reliability and efficiency of this method for the fracture test of bioceramics have been confirmed by Elghazel et al. (2015) using a standard method for disc shape concrete or ceramics ASTM C 496/C 496M – 04 (2011). INSTRON 8874

with the load cell 5kN and loading speed of 0.075 mm/min is used. In diametral compression test, a disc sample is used to determine the tensile strength by subjecting the specimen to two opposite sides of the normal load. The applied load develops across the diametrical plane of the disc parallel to the loading direction. Figure 1 illustrates the diametral compression test. Fracture occurs along the vertical plane that passes through the center of the disc. Here, the tensile stress is directly proportional to the compressive load applied. Such diametral tensile strength is calculated using (2).

$$\sigma_t = \frac{2P}{\pi DT} \tag{2}$$

where σ_t is the maximum tensile strength (MPa); P is the maximum applied load at fracture (Newton); D is the diameter of the samples measured from the center (mm); and T is the thickness (mm) of the disc. To calculate the stress using (2), the fracture must be initiated at the center of the disc across the disc. In order to assure the validity of the test, the contact width of the sample must be less than 20% of the sample diameter and a near-vertical fracture occurred (Pilliar et al. 2001). The crushing of the disc sample at the loading points that occasionally happen in brittle ceramic materials will cause invalid test results.

Microhardness testing machine was used to measure the Vickers hardness values of the samples. In Vickers indentation technique, well-polished specimens were subjected to the Vickers indenter: 10 kg load at 15 s was applied onto the specimen surface by diamond pyramid tip. For each composition, four to five specimens were tested for diametral tensile strength and microhardness and the results were then averaged.

BIOACTIVITY TEST IN SIMULATED BODY FLUID

The *in vitro* bioactivity of the PSW/Mullite composite was studied by soaking all samples in the SBF solution prepared by the method proposed by Kokubo and Takadama (2006). The samples were soaked in the polyethelyne PE holder and keep in the incubator at 36.7°C for 1, 3, 7 and 14 days. The SBF solution was changed every 3 days to maintain the ion in the solution. After soaking in the SBF solution for every respective day, the sample was taken out and immersed in acetone for 2 h to stop the reaction, rinsed with distilled water and left dried in the incubator for 24 h. The *in vitro* bioactivity assessment is the prior test before proceeding to the *in vivo* evaluations.
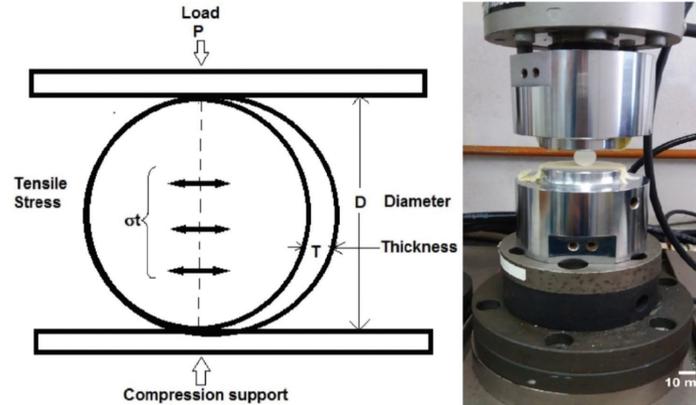
FIGURE 1. The illustration of diametral compression test measured using
INSTRON 8874 with the load cell 5kN

RESULTS AND DISCUSSION

PHASE COMPOSITION AND DENSITY

The XRD results showed the spectra of pseudowollastonite-mullite composite for different compositions sintered at different temperatures. Figure 2 shows that the peak intensity of mullite increases as the content of mullite in the composite increased. A very intense and sharp peak could be seen at every sintering temperature. Based on the XRD spectra in Figure 2, the increasing temperature from 800°C - 1000°C caused no significant changes of phases for different amount of mullite additions at 10-30 wt. %. Furthermore, it could be seen obviously that, the new phase of calcium aluminium oxide existed for different mullite concentration at 1150°C sintering temperature. Shirazi et al. (2014) reported that the formation of new phase $CaAl_2O_4$ (ref. no.JCPDS 001-0888) at peak angles of 30, 35.5 and 37.2 (Figure 2(g)- 2(i)) is due to the reaction between alumina and calcium silicate. The formation of calcium aluminum oxide at the temperature of 1150°C is based on the following reaction.

$$CaSiO_3 + Al_2O_3 \longrightarrow CaAl_2O_4 + SiO_2$$

In addition, the presence of gehlenite phase could also be observed at peak angles of 31.4, 53.1 and 29.1 The formation of softer sintered phase gehlenite is a common for pressureless sintering of mullite (Nath et al. 2008). The relationship between calcium aluminum oxide and gehlenite formation with mechanical properties of the material will be further discussed in the next section.

In general, the sintering temperature is the crucial factor that affects the densification of the composite. The density profile of the sintered component in Figure 3 shows that densification assisted by sintering. Based on the graph, the density for all samples were significantly increases as we increase the sintering temperature from 800°C to 1150°C. A drastic change of density could be observed on PSW samples when the temperature increased
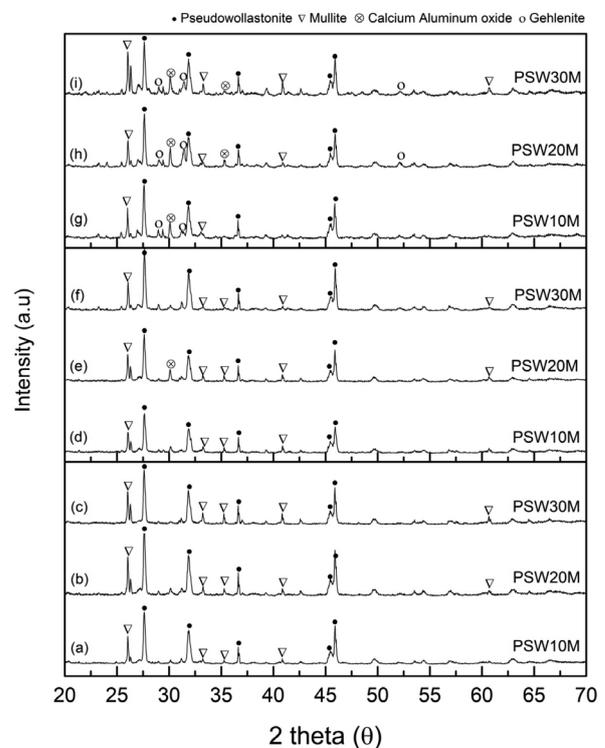

FIGURE 2. XRD patterns of the composite discs with different compositions i.e. PSW10M, PSW20M, PSW30M after sintering at 800°C (a,b,c), 1000°C (d,e,f) and 1150°C (g,h,i) for 2 h

from 800°C to 1000°C. Whereas for PSW20M composite, the density was sharply improved at the range between 1000°C and 1150°C. The highest relative density of 2.895 g/cm³ shows in Figure 3 is pseudowollastonite with 20 wt. % mullite addition. At the same sintering temperature with greater amount of mullite (30 wt. %) additions, the density is decrease (2.72 g/cm³). Further additions of mullite seems like do not assist a better densification which might due to unnecessary amount of mullite additions that will cause excessive large grain of mullite-mullite contact in the sintered body thus, pore elimination occur less efficiently (Nath et al. 2008).
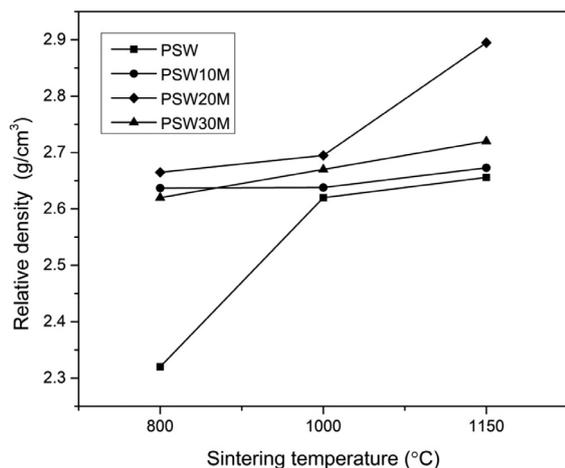
FIGURE 3. Relative density vs porosity of the samples with different firing temperatures

EFFECT OF MULLITE ADDITIONS AND SINTERING
TEMPERATURE ON MECHANICAL PROPERTIES

Figure 4 shows the relationships between the diametral tensile strength (DTS) and the sintering temperature corresponded to different pseudowollastonite-mullite compositions. The DTS increased along with the sintering temperature. The highest strength of 100%PSW sample without any reinforcement is 5.04 ± 0.20 MPa. Additions of 20 wt. % mullite reinforcement increased the strength to 8.8 ± 0.15 MPa at the sintering temperature of 1150°C. For the reason that, most of the micro pores are eliminated at 1150°C (Figure 5). However, further increased of mullite content to 30 wt. % reduced the strength to 6.32 ± 0.15. Shirazi et al. (2014) remark that the presence of alumina particle at 15 wt. % concentration in calcium silicate matrix enhances the hardness and fracture toughness of calcium silicate. However, at 25 wt. % alumina concentration, the mechanical properties of $CaSiO_3$-Al_2O_3 decrease. The decrease in the mechanical strength at higher alumina content is attributing to the incomplete reaction between calcium silicate and alumina at low sintering temperature. Therefore, increasing mullite content in this study over 20 wt. % also decreases the DTS value in reason. Nevertheless, the DTS value of sintered pseudowollastonite (α-$CaSiO_3$) after mullite additions in this study is much improved than that of synthesized CaO-SiO_2 cements with few other methods such as sol gel techniques that have diametral strength about 2.2-2.4 MPa when sintered at 1000°C (Chen et al. 2015) and hydrothermal method for 2.9 MPa (Ding et al. 2009), respectively.

One of the factors that have significant effects on the mechanical properties of material is the liquid phase sintering (LPS) mechanism that happened during sintering. During LPS reaction, the atoms become more mobilized and re-arranged. In addition, the grain growth was promoted through the movement of particle among larger granule (Maitra et al. 2006). The formation of new phase $CaAl_2O_3$ occurred at temperature 1150°C (Figure 2). Such

liquid phase enhances the densification thus becomes the important reason in improving the mechanical properties. Several researches also highlighted that the existence of calcium aluminium oxide $CaAl_2O_4$ (Figure 2) will act as the hardener phase that enhances the strength of the HA composite (Gautier et al. 1997; Horng & Min 1994; Ji & Marquis 1992). In addition, Nath et al. (2010) also point out the benefit of using mullite particle as reinforcement because of good densification that could be achieved due to the liquid-phase sintering. The study on ternary phase diagram CaO-Al_2O_3-SiO_2 system prove the formation of gehlenite, anorthite and calcium aluminum oxide ($CaAl_2O_3$) for calcium silicate containing mullite composite which is dependable on the sintering temperature (Zhao et al. 2007).

The same trend is perceived for hardness test (Figure 4). As would be expected, the hardness increased as the temperature increases from 800°C - 1150°C. The vickers hardness value of about 2.31 ± 0.14 GPa is obtained for pseudowollastonite pellet when sintered at 1150°C. Consequently, the best hardness value was 3.79 ± 0.14 GPa given by PSW20M sample when sintered at this highest temperature. This result is comparable to other work that attained the micro hardness value of 2.6 GPa for synthesized pseudowollastonite sintered at 1150°C from local raw material $CaCO_3$ which then improved up to 3.1 GPa with the addition of 5 wt. % of B_2O_3 (Harabi & Chehlatt 2013). Other research also reported that HA(20 wt. %)- wollastonite (80 wt. %) composite sintered at 1000°C for 2 h reached the hardness of 3.92 GPa. Back to this study, as the mullite content increased to 30 wt. %, both of DTS and hardness decreased. Such results are in line with the earlier results
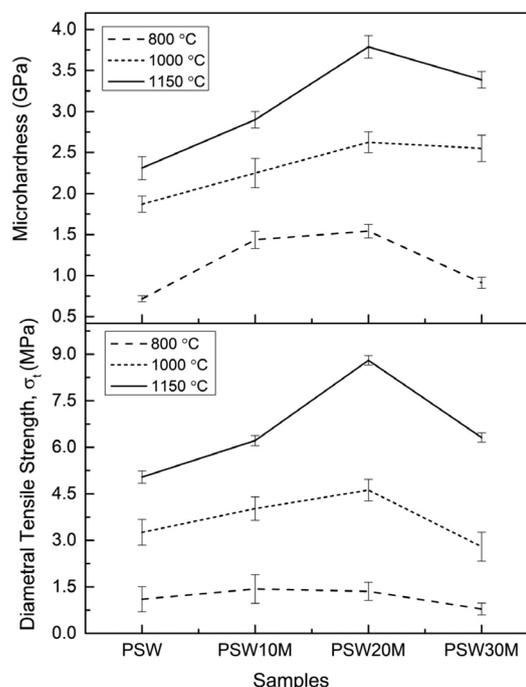

FIGURE 4. Microhardness and diametral tensile strength of sintered pseudowollastonite-mullite (PSW-M) composite plotted against different compositions

reported in literature (Nath et al. 2012) which the highest strength of HAp achieved with not more than 20 wt. % addition of mullite. Thus, it can be possibly deduced that, 20 wt. % of mullite reinforcement caused the optimum amount of $CaSiO_3$-mullite contact which leads to the far-reaching sintering reaction.

The mechanical properties of the materials are also interrelated with the microstructural features. The FESEM micrograph in Figure 5 shows the surface microstructure of pseudowollastonite-20% mullite disc sintered at different sintering temperatures with distinct and interconnected pores. High porosities reduced the fracture toughness and strength of the materials. More abundant pores could be seen for the PSW20M composite sintered at 800°C compared to the higher sintering temperature at 1150°C (Figure 5) which supports our results that high DTS is attained for samples sintered at higher temperature. A much improved compactness could be seen for PSW20M sample sintered at 1150°C as compared to the pseudowollastonite (Figure 5(c) & 5(d)). Eventually, during sintering at 1150°C, the particles are distorted hence abruptly eliminates the micro pores at the grain boundaries. Other samples with different weight percentage of mullite have insignificant differences in terms of morphology with this pseudowollastonite-20% mullite composite. This condition happened because the presence of liquid phase during sintering at high temperature enhanced the mullite grain growth hence assists the pore closure as reported by Bakr (2012). As a result of capillary action during liquid phase sintering (LPS), liquid flow leads to particle rearrangement. This caused larger voids and micro pores reduced and contribute to densification. The particle rearrangement, diffusion of mass into pores and grain growth are some of the phenomenon that normally occurred during sintering.

The mullite reinforced composites up to 20 wt. % shows the greatest diametral tensile strength (8.8 ± 0.15 MPa) and hardness (3.79 ± 0.13 GPa) compared to the non-reinforced wollastonite (5.04 ± 0.20 MPa) and (2.31 ± 0.14 GPa), respectively. Ultimately, based on the comparisons of different mullite compositions and sintering temperatures, it can be concluded that the best mechanical strength which includes DTS and hardness is given by PSW20M sintered at 1150°C that has the lowest porosity and a good densification. Therefore, the following study of the *in vitro* bioactivity in SBF solutions has focused on the investigation of the PSW20M sample.

APATITE FORMATION ON PSW20M COMPOSITES

The formation of the apatite phase on the surface of the material was monitored during the *in vitro* test in SBF solution. According to Kokubo and Takadama (2006), the ability of the implanted biomaterial to form interfacial bonds with tissues and bone *in vivo* could be predicted by the examination of apatite formation on material in the SBF solution. Figure 6 shows the PSW20M sample before immersion and after immersion up to 14 days of immersion in the SBF solution. The decreased in intensity of the sharp peaks of pseudowollastonite and mullite could be clearly observed from day 0 to day 14. This shows that the dissolution has occurred. At day 7, the presence of a broad peak at $2\theta = 32\text{-}34°$ indicates the formation of amorphous HA started to form. This amorphous peak remains and increases its intensity until day 14. During the immersion of PSW20M samples in the SBF solution, the ion exchange from the material surface Ca^{2+} into the solution and H^+ ion from the SBF solution occurred. The negatively charged surface with the functional group (Si-O)
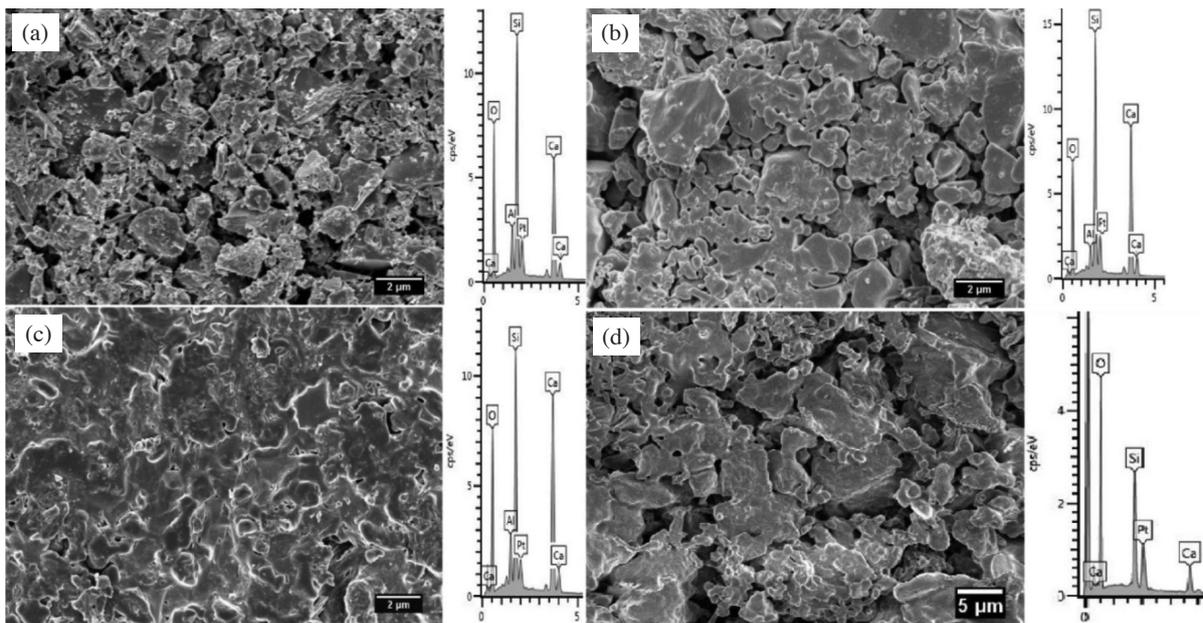

FIGURE 5. The morphology of PSW/20%M disc surface shows the disc composite at 5000× magnification (in lens mode) sintered at (a) 800°C, (b) 1000°C and (c) 1150°C and pseudowollastonite disc sintered at 1150°C
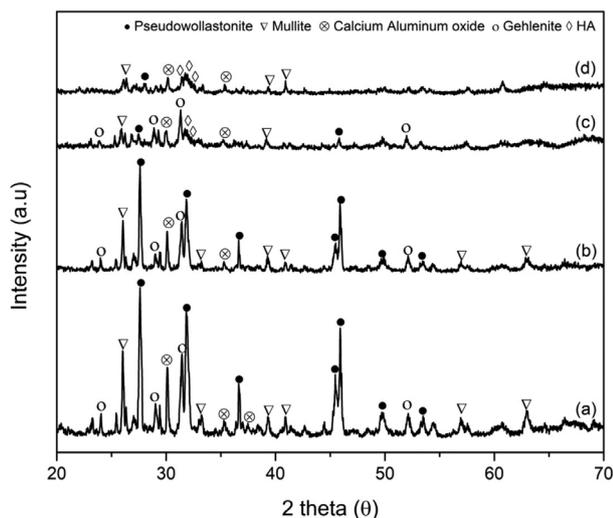
FIGURE 6. XRD patterns of the PSW20M samples sintered at 1150°C soaking in SBF solutions for (a) 0, (b) 3, (c) 7, and 14 days

FIGURE 7. FTIR spectra of the PSW20M samples sintered at 1150°C and soaked in SBF solutions for (a) 0, (b) 3, (c) 7 and (d) 14 days

is then formed on the sample surface. This Si-O- induces apatite nucleation (Hamisah et al. 2016). Based on the FTIR spectrum in Figure 7, the bending band for the silicon ion (Si-O-) could be observed at the wavenumber 1067.18 cm^{-1}. In addition, after 7 days, the presence of PO groups could be observed at the wavenumber 1020 cm^{-1} which demonstrates the apatite formation.

FESEM images of PSW20M samples before and after immersion in SBF solutions for 0, 7 and 14 days are displayed in Figure 8. The rough and fibrous layers deposited after 7 days of immersion is the apatite layer that formed on the samples. These results are confirmed by EDS analysis which shows the presence of Ca/P ratio is 1.8 after 7 days of immersion in SBF solution. This ratio indicates the formation of amorphous calcium phosphate (ACP). Dorozhkin (2007) highlighted that range of ACP must be in the range of 1.2 to 2.2. Thus, it may be concluded that pseudowollastonite-mullite composite exhibit good bioactivity properties.

CONCLUSION

Pseudowollastonite containing mullite was successfully prepared and sintered at various temperatures. The role of mullite as reinforcement was proven capable of enhancing the mechanical properties of the pseudowollastonite. The liquid phase sintering reaction which occurred at 1150°C promotes grain growth thus assisted densification. The optimum amount of mullite addition to pseudowollastonite at 20 wt. % gave the best hardness and diametral tensile strength. The strength of composite will be deteriorated as the amount of mullite increased. The presence of HA peak from XRD and the apatite morphology from FESEM after immersion in SBF solutions for 7 days showed that PSW20M composite was bioactive. Therefore, this finding suggested that pseudowollastonite-mullite composite synthesized
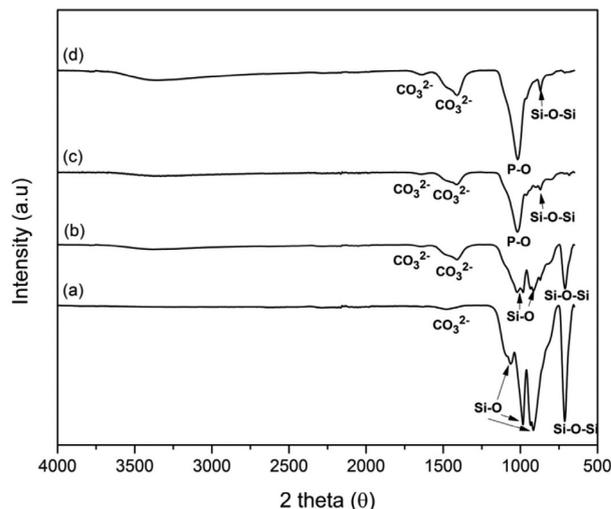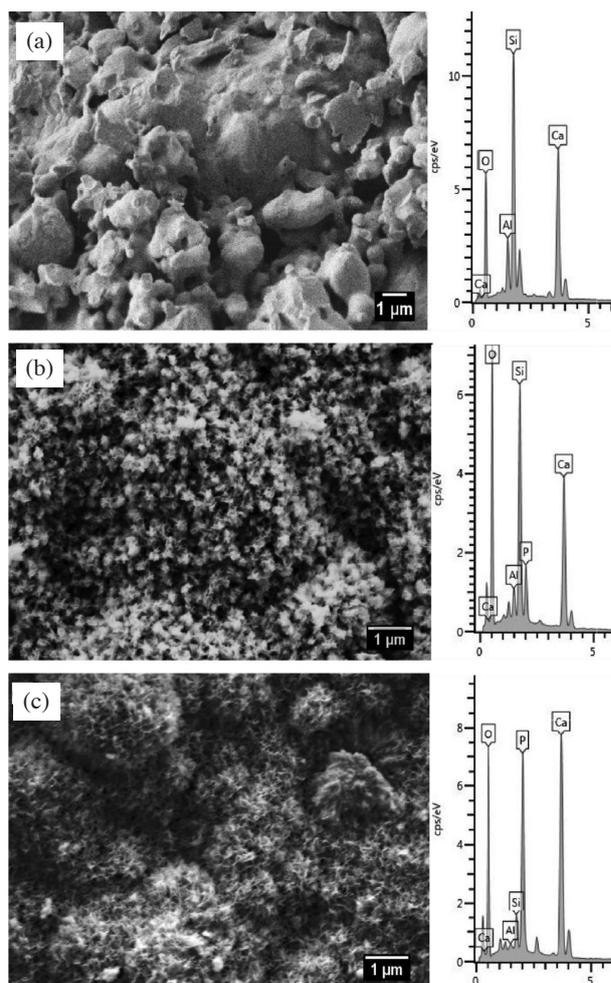

FIGURE 8. SEM morphology of the PSW20M composite sintered at 1150°C after soaking for (a) 0 and (b) 7 and (c) 14 days in the SBF solution

from the natural resources of rice husk ash limestone and natural mullite have the promising mechanical properties as well as good bioactivity which have the potential for implant in biomedical application. In future, cytotoxicity and live-dead assays will be performed on human cells and behavior of this composite *in vivo* will be examined.

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