

## SYNTHESIS AND MECHANICAL PROPERTIES OF ALUMINA SLIP CASTING FROM ALUMINIUM TRI-HYDRATE ( $\text{Al}(\text{OH})_3$ )

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### ABSTRACT

*The mechanical properties of alumina based ceramics fabricated from synthesized of aluminium tri-hydrate,  $\text{Al}(\text{OH})_3$  powder were investigated. The modification had been made on  $\text{Al}(\text{OH})_3$  precursor of to be well-suited as a substitution material in alumina slip casting process. The used of  $\text{Al}(\text{OH})_3$  in this research instead of  $\text{Al}_2\text{O}_3$  is because the cost for  $\text{Al}_2\text{O}_3$  is high, especially the one that got higher purity of  $\text{Al}_2\text{O}_3$ , so to reducing the cost in producing a high quality ceramics, synthesis process had been made on the  $\text{Al}(\text{OH})_3$  powder. Two methods to obtains  $\text{Al}_2\text{O}_3$  from  $\text{Al}(\text{OH})_3$  was used it is calcination and leaching process in the variable of sintering temperature and time. Leaching method produced higher  $\text{Al}_2\text{O}_3$  content from  $\text{Al}(\text{OH})_3$  as it increased the flexural strength and hardness of ceramics bodies. It is shown that the flexural strength and hardness is depending on the content of  $\text{Al}_2\text{O}_3$ . The microstructure of the synthesized  $\text{Al}(\text{OH})_3$  powder were grown homogenously and finer structure obtained. The grain size of pure  $\text{Al}(\text{OH})_3$  are range between 30 to 35  $\mu\text{m}$  and when the precursor was calcined, the grain size became finer by the size 24 to 27  $\mu\text{m}$ . As more synthesized process undergoes in leached method, the precursor come out with even finer grain size with a grain size ranging from 21 to 23  $\mu\text{m}$ . This research proved that the production cost of ceramics made up from alumina can be reduced by synthesized method but in the same time the quality of the product will be maintained or improved.*

Keywords: Ceramics, leaching, calcinations, aluminium hydroxide, slip casting.

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### Introduction

Alumina, ( $\text{Al}_2\text{O}_3$ ) is a high cost precursor that had been processed or extracted from aluminum tri-hydrate which it is the most applicable precursor to use in slip casting process, Alumina tri-hydrate ( $\text{Al}(\text{OH})_3$ ) was extracted from aluminium ore or known as bauxite where it is contain about 64% of alumina composition where the slurries for slip casting usually made up by 98% to 99.99% of pure alumina precursor to produce good ceramics (King, 2002). As referred to (Takao et al, 2000), (Tallon, Limacher & Franks, 2010) and (T. Hotta et al, 2005) The mechanical properties of porous ceramics related to several factors like grain bonding, purity of precursor, pore structure and pore volume fraction. There are several ways to enhanced mechanical properties of porous ceramics, one of it is by optimizing pore structure. Strong grain and high porosity  $\text{Al}_2\text{O}_3$  also can be obtained by decomposed  $\text{Al}(\text{OH})_3$  until it experience volume contraction (Z.-Y. Deng et al., 2001). The sintering time and temperature also play a role in process of ceramics forming, the sintering temperature and time can affect the microstructure, final density and sintered grain size of the green bodies (Rowley & Chu, 2002), which it can lead to the mechanical properties effect (T. Hotta et al., 2005). The decomposition of  $\text{Al}(\text{OH})_3$  formed a very fined  $\text{Al}_2\text{O}_3$  grain where it create a series of transitional  $\text{Al}_2\text{O}_3$  phases that eventually transform to a stable  $\alpha\text{-Al}_2\text{O}_3$  at high sintering temperature (Z.-Y. Deng et al., 2001).

Synthesized process can be one of the most crucial part in producing a good mechanical for ceramics, in the past study by (Y, Hotta, Banno & Oda, 2002) they used synthesized process by leaching to remove gypsum component with 0.9 M HCl acid, this leached process also had been used by (Kukolev & Karaulov, 1963) to remove iron composition that came from grinding operation which after that process, the alumina percent in the ceramics increased from 95% to 99.72%. As for synthesized process by calcinations, (Y. Hotta et al., 2002) had employed this process in his study, the green bodies was dried and calcined at 800°C for 2 hours in the air to remove the presence of deflocculant. (Kukolev & Karaulov, 1963) also applied this method and produced 95 to 97% of  $\alpha\text{-Al}_2\text{O}_3$  which it is proven that calcinations can increase the purity of  $\alpha\text{-Al}_2\text{O}_3$  composition in aluminium tri-hydrate and even the alumina precursor its own. The  $\alpha\text{-Al}_2\text{O}_3$  phase is the ultimate condition for alumina which it can be transformed from  $\gamma\text{-Al}_2\text{O}_3$  after the calcinations process were held, (Majid Farahmandjou, 2015).

In this present study,  $\text{Al}(\text{OH})_3$  precursor was used because of the cost of this precursor are five times cheaper than  $\text{Al}_2\text{O}_3$ , so the  $\text{Al}(\text{OH})_3$  were synthesized to produce a high purity  $\text{Al}_2\text{O}_3$  and a fined microstructure grain of the precursor it is to obtain an excellent mechanical properties for ceramics. Additionally, the synthesized precursor pore structure might be change after undergo the sintering process. The aim of this research was to increase the mechanical properties of ceramics by synthesizing  $\text{Al}(\text{OH})_3$  precursor and varying the sintering temperature and time for the green bodies made up from synthesized precursor.

## Experimental Procedure

### Synthesisization Process

Locally available Aluminum tri-hydrate  $\text{Al}(\text{OH})_3$  (Ipoh Ceramics, Perak, Malaysia) were used in this studies as a raw material. From the raw materials two type of precursor samples are prepared which directly calcined without any pretreatment and treated by leaching before calcination process; latter will these will be referred as sample C and LC respectively. Leaching process for LC samples are done by immersing the raw material in 0.9M HCl for 1h and filtered before washed with distilled water and leaves the precursor to dry. C and LC samples are then calcined at 1200°C for 3h with heating rate 5°C/min.

### Preparation Of Slurry And Ceramics Bodies

The preparation of slurry for both LC and C samples were started with dispersing polyacrylic acid in distilled water with solid loading of 50 vol.%. The dispersant amount were varied from 0 to 2 mass%. The slurries were ball milled for 24h with planetary mill in zircon media and balls. Gypsum mold was used to undergo a slip casting process and the green bodies detached after 10min slip. The green bodies then sintered at 1200 °C and 1500 °C for 3 and 6h as shown in Table 1 below;

**Table 1: sintering time and temperature for the green bodies**

Sample	High Temperature, 1500°C	Low Temperature, 1200 °C	Sintering Time, hours	Label
C	√		6	C15.6
		√	6	C12.6
	√		3	C15.3
		√	3	C15.6
LC	√		6	LC15.6
		√	6	LC12.6
	√		3	LC15.3
		√	3	LC15.6

### Testing And Analysing

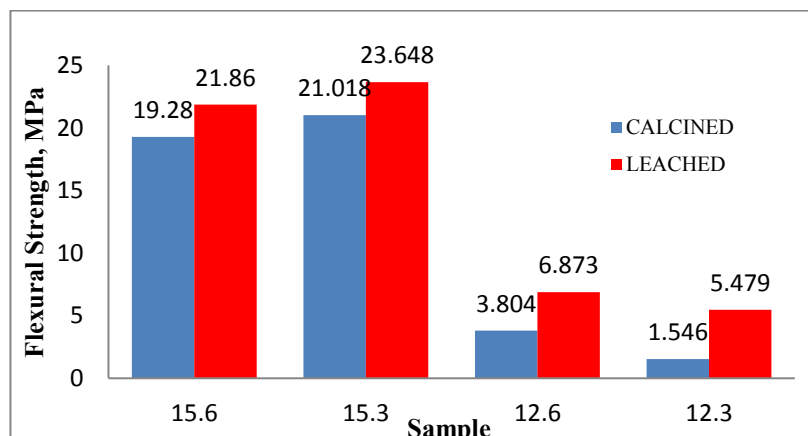
The mechanical testing for these ceramics bodies was 3 point bending test and hardness test. 3 point bending test was held with sample size 3x4x40mm by INSTRON Universal Testing Machine with 30mm span with loading speed 0.5mm/min. The hardness test was done by using Vickers hardness testing machine with the load of 50N for 10 seconds dwell time with 5 indentions for each sample. As for analyzing process, the raw material, synthesized precursor and ceramics bodies were involved. SEM and XRD were used for analyzing process.

## Result And Discussion

### Flexural Strength

Figure 1 shows sintering temperature at 1200°C for both synthesized sample had a slight different strength, C12.6 and C12.3 had lower strength compared to LC12.6 and LC12.3. This might caused by different synthesized process, because the calcined sample through synthesized process once as for leached sample, it through twice synthesized process where the decomposition of  $\text{Al}(\text{OH})_3$  are much more than calcined process.

**Figure 1: flexural strength of all samples**

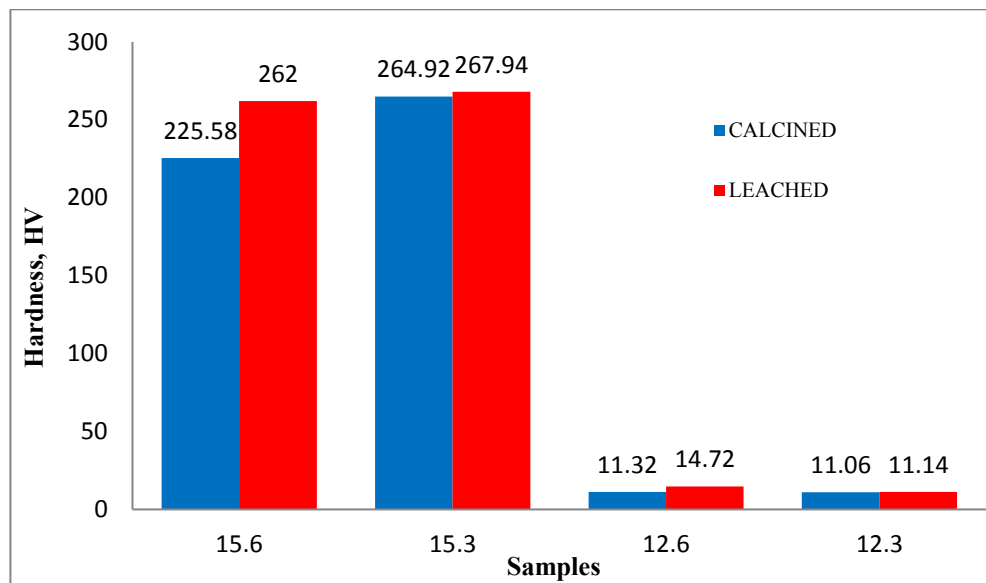


Samples sintered at 1500°C, despite the strength that higher than both C12.6 and LC12.6 it showing that the higher sintering temperature produced higher strength ceramics. However, sintering time did not contribute to the increase of strength because from the results obtained, LC15.3 sample had the highest strength, which LC15.6 sample was the one expected to hold the highest rank in term of strength. This might caused by local packing disruption where it can reduce the connectivity particles and lowering the strength of those bodies. Local packing disruption happened when the volume change during the synthesize process and the process might keep furthering as the sintering time is longer and temperature is higher (Z. Y. Deng, T. Fukasawa, M. Ando, G. J. Zhang, & T. Ohji, 2001). This can be concluded that the increased of sintering time is not practical to increased strength of the ceramics bodies but increased the sintering temperature did helped in improving the fracture strength of ceramics bodies.

### Hardness

The hardness result had shown that the issue in flexural strength also affected the hardness of the ceramics, in Figure 2 showing that ceramics that undergo 1500°C sintering temperature for 6 hours did not obtain good mechanical properties in terms of hardness, this happen for both synthesized precursors.

Figure 2: hardness value for all samples

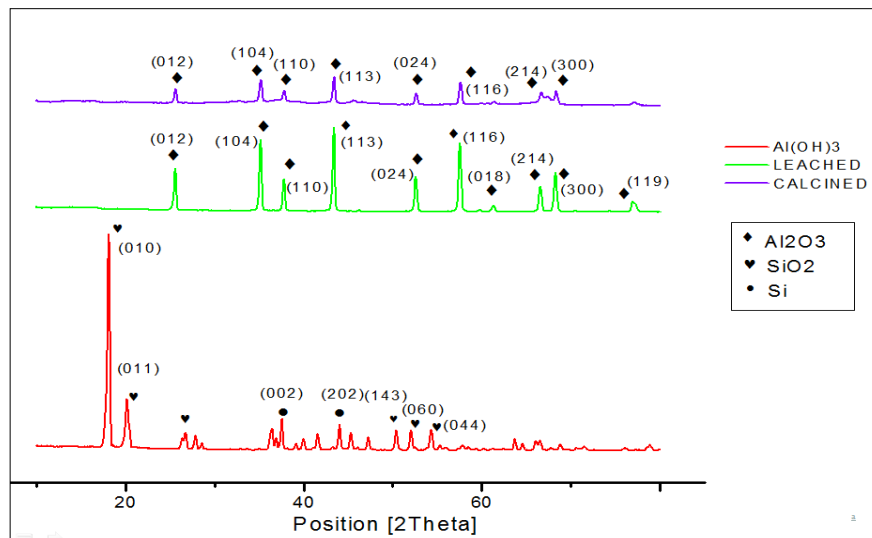


The hardness between all the samples has been compared in Figure 2, it showing that calcined and leaching sample sintered at 1200°C having slight different in term of hardness but the leaching still leading. The hardness of leaching samples is harder compare to calcined samples. As expected, among these samples that sintered for 6 hours, leached sample at 1500°C obtained the highest hardness compared to others. It is stated by (Travitzky, 2001) that the hardness test for ceramic need to be load use 100N with duration time of 15 second. However, for all of samples used in these studies are broken at the first 2 second after the indentation start.

### X-RAY DIFFRACTION (XRD)

Phase transformation occurred at synthesized precursors since both of it went through sintering process which lead to phase transformation. As can be seen in figure 3 below,  $\text{SiO}_2$  peak was removed after the synthesized process, leaving  $\text{Al}_2\text{O}_3$  conquering the high peaks in the precursors. Leached precursor contained  $\text{Al}_2\text{O}_3$  at plane (012), (104), (110), (113), (024), (116), (214) and (300) and those peaks meet the PDF card reference code 01-081-1667. Precursor that synthesized by calcinations process also showing the same result as leached but the peak is slightly lower than leached. This might caused by leaching process which the precursors need to be soak into HCl to remove other unnecessary compound and increased the  $\text{Al}_2\text{O}_3$  in the precursor before it through calcinations process for further impurity removed.

Figure 2: XRD pattern for pure  $\text{Al}(\text{OH})_3$ , leached and calcined precursor



The XRD pattern from all four samples showed in Figure 4 below, showing that the  $\text{Al}_2\text{O}_3$  phases are the only phase existed in those sample, the plane obtained for those phase were (012), (104), (110), (113), (024), (116), (214), (300) and (119) which meet the peak of PDF card reference code 01-075-0783. All four samples had almost similar pattern which proving that sintering time and temperature were not inference the phases of the ceramics bodies. The  $\text{Al}_2\text{O}_3$  phase remains even after the precursor had been process by slip casting where the dispersant was added during the process. Dispersant were removed during the sintering process of the green bodies.

Figure 3: XRD pattern from precursor synthesized by calcinations

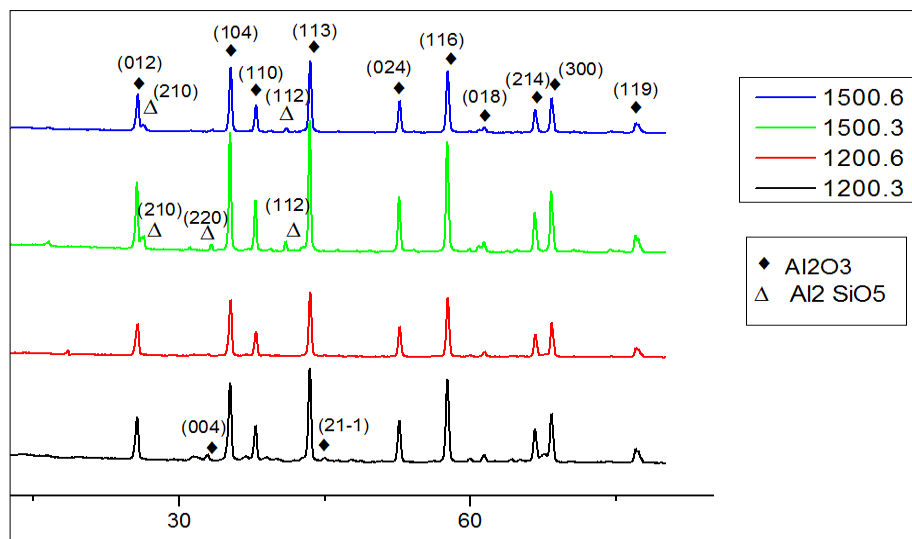
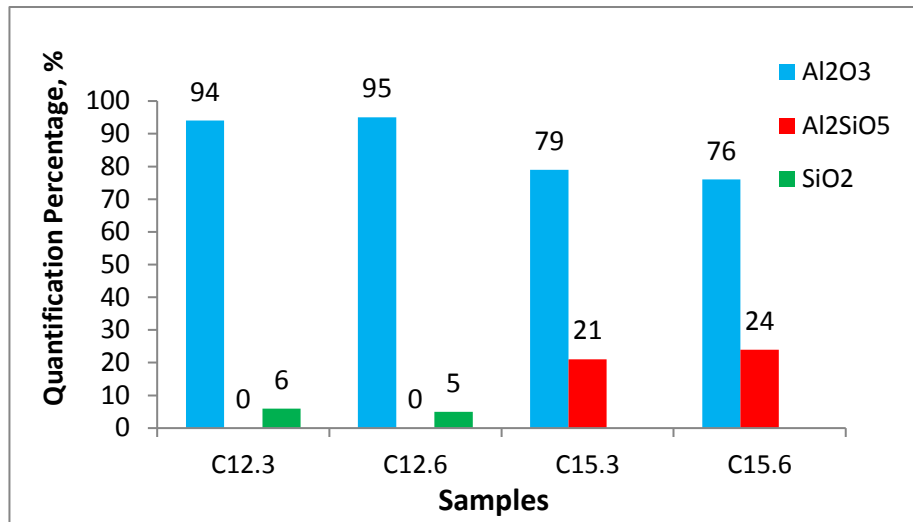
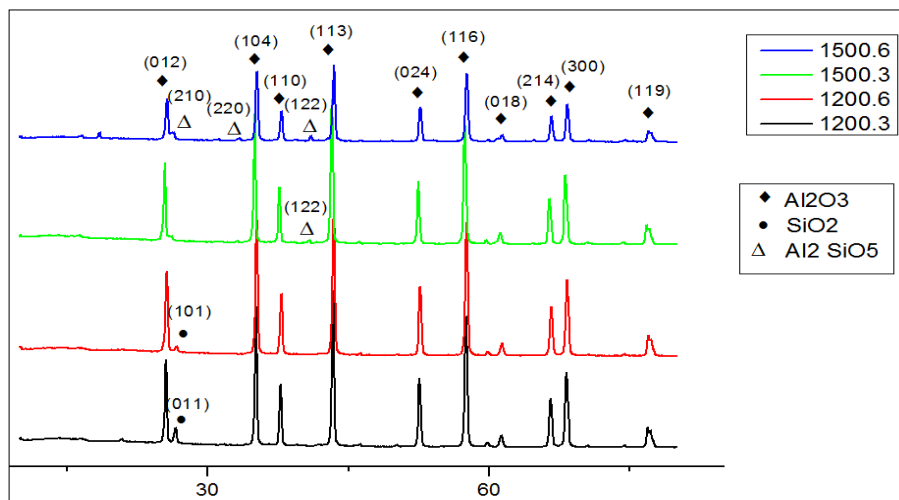


Figure 4: quantification percent, % for calcined samples



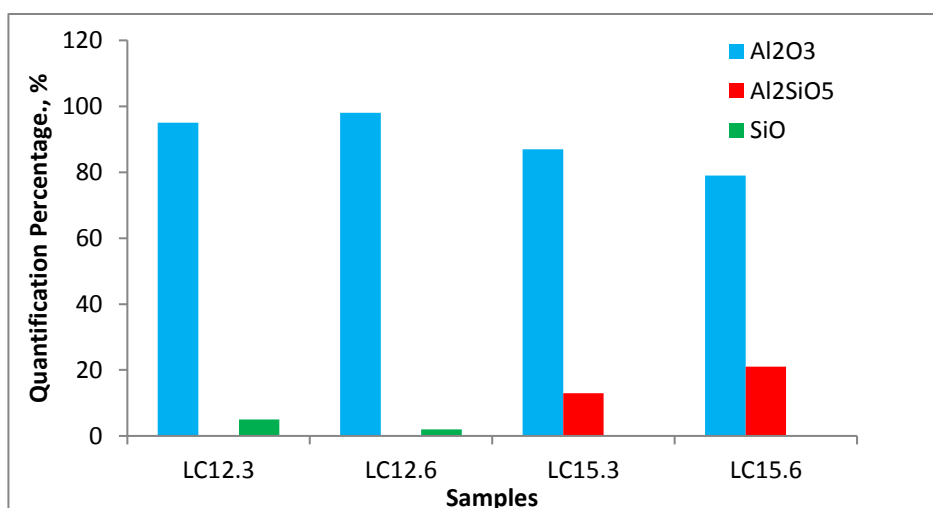
As for the quantification percent, it is showed in Figure 5 above that as the sintering time longer it will increase the purities of Al<sub>2</sub>O<sub>3</sub>. At C12.6, the purities of Al<sub>2</sub>O<sub>3</sub> higher and it produce a better mechanical properties compared to C12.3, which it proven that the higher purity of Al<sub>2</sub>O<sub>3</sub> is necessary in producing an excellent mechanical properties for ceramics bodies. The different case can be seen for sample C15.3, supposedly the purity of Al<sub>2</sub>O<sub>3</sub> increase as the temperature increase but it's not happening to this sample. The purity of Al<sub>2</sub>O<sub>3</sub> is getting low compare with C12.3, C12.6 samples, but they still produced good mechanical properties for those ceramics. This is because the Al<sub>2</sub>O<sub>3</sub> had an oxide-like combination with SiO<sub>2</sub> and produced Al<sub>2</sub>SiO<sub>5</sub> which had very good mechanical properties.

Figure 5: XRD pattern from precursor synthesized by leaching



As for samples that processed using precursor that synthesized by leaching, the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> existed in all four XRD pattern and the pattern is almost same for all four samples as shown in figure 6 above. Those patterns were detected at plane (012), (104), (110), (113), (024), (116), (214), (300) and (012), which the plane was almost the same as plane in calcined sample and meet the peak of PDF card reference code 01-075-0783.

Figure 6: quantification percent, % for leached samples.



The same trend happening to leached samples. The purity for longer sintering time is the highest, but that's not the case for L15.6. L15.6 sample had a longer sintering time but it not produced a higher purity as L12.6 and caused the mechanical properties of that sample dropped. This is because it obtained higher Al<sub>2</sub>SiO<sub>5</sub> and made the Al<sub>2</sub>O<sub>3</sub> purity decreased. Al<sub>2</sub>SiO<sub>5</sub> did helped in produced a better ceramics in term of mechanical properties, but the purity of Al<sub>2</sub>O<sub>3</sub> still played an important role in producing that type of ceramics. As showed in Figure 7, even at short sintering time, L15.3 had higher purity of Al<sub>2</sub>O<sub>3</sub> compared to L15.6 and its make that samples had better mechanical properties.

#### Unit Cell Volume

These synthesized precursor had rhombohedral crystal system where  $a=b \neq c$  as shown in table 2 below, both precursor showing that they had almost same lattice parameter (LP) but for unit cell volume (UCV), leached precursor leading with a higher value with  $254.51 \text{ \AA}^3$  compared to calcined,  $253.67 \text{ \AA}^3$ . This is showing that the synthesized by leached can increased the unit cell volume higher than synthesized by calcined.

**Table 2: summaries of the lattice parameter and unit cell volume for synthesized precursor.**

Sample	Lattice Parameter, $\text{\AA}$	Unit Cell Volume, $\text{\AA}^3$
Calcined (C)	$a = b = 4.7528$	253.67
	$c = 12.9673$	
Leached (LC)	$a = b = 4.7570$	254.51
	$c = 12.986$	

A refinement was done on four calcined samples for obtained their LP and UCV. The results were illustrate in Table 3 below, it showing that the longer sintering time will produce less UCV but it just a slight reduce by range 0.10 to 0.50 different. The LP also showing the same pattern as the UCV where longer sintering time will drop the LP slightly.

**Table 3: summaries of the lattice parameter and unit cell volume for ceramics bodies produce from calcined precursor.**

Sample	Lattice Parameter, $\text{\AA}$	Unit Cell Volume, $\text{\AA}^3$
C12.3	$a = b = 4.7510$	253.58
	$c = 12.9720$	
C12.6	$a = b = 4.7460$	253.05
	$c = 12.9700$	
C15.3	$a = b = 4.7510$	253.74
	$c = 12.9780$	

C15.6	a = b = 4.7485	253.67
	c = 12.9718	

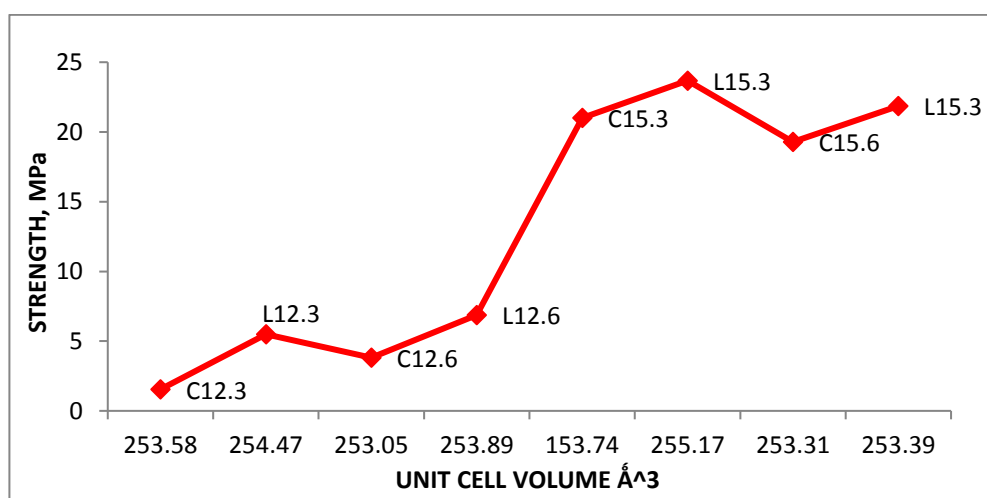
LP and UCV for four leached samples were summarized in Table 4 below. It showing that the shorter sintering time will produce more UCV but it just a slight increased by range 1.10 to 2.50 different. The LP also showing the same pattern, as the sintering time shorter, the value of LP would be increased slightly.

**Table 4: summaries of the lattice parameter and unit cell volume for ceramics bodies produce from leached precursor.**

Sample	Lattice Parameter, Å	Unit Cell Volume, Å <sup>3</sup>
L12.3	a = b = 4.7506	254.47
	c = 12.9870	
L12.6	a = b = 4.7530	253.89
	c = 12.9780	
L15.3	a = b = 4.7690	255.17
	c = 12.9500	
L15.6	a = b = 4.7490	253.39
	c = 12.9760	

The sintering time and temperature also affect UCV for each samples, it is show in figure 8 that the relationships between the strength and UCV. It showed that as the UCV increased, the strength of the ceramics bodies also increased but there is a slight different detected among those samples. Samples LC15.6 and C15.6 dropped its UCV and strength. This might cause by the effects of sintering time on those two bodies, which the longer the sintering time at higher temperature will dropped the UCV along with the strength. The relationships between the hardness and UCV also had the same pattern as strength. At samples LC15.6 and C15.6 both UCV and hardness dropped.

**Figure 8: relationships between strength and unit cell volume of ceramics bodies.**



#### SCANNING ELECTRON MICROSCOPY (SEM/EDX)



The microstructure for this precursor was obtained using Scanning Electron Microscopy (SEM). This is to identify the changes in microstructure of precursor after modification process. The SEM photograph results for  $\text{Al}(\text{OH})_3$  before and after synthesized are shown in Figure 9;

**Figure 9: SEM photograph for  $\text{Al}(\text{OH})_3$  (A) before synthesized and (B) after calcinations**

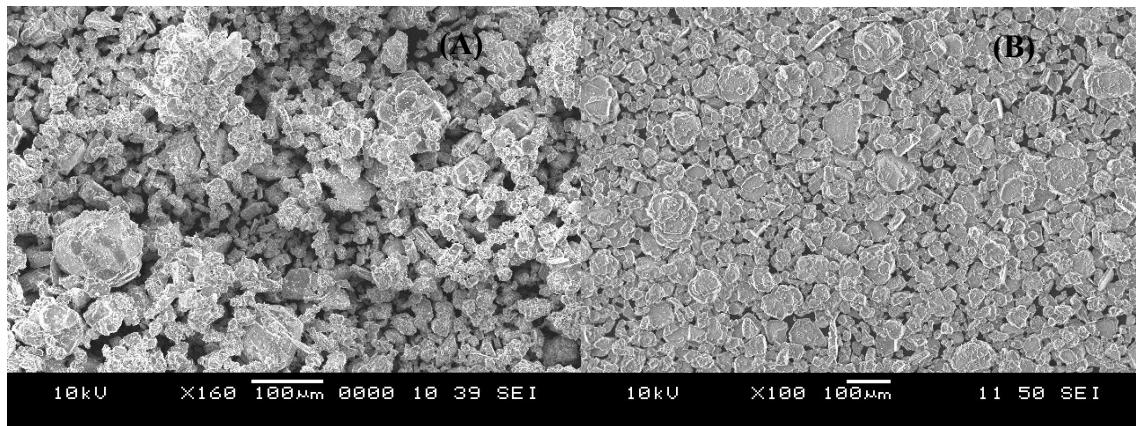
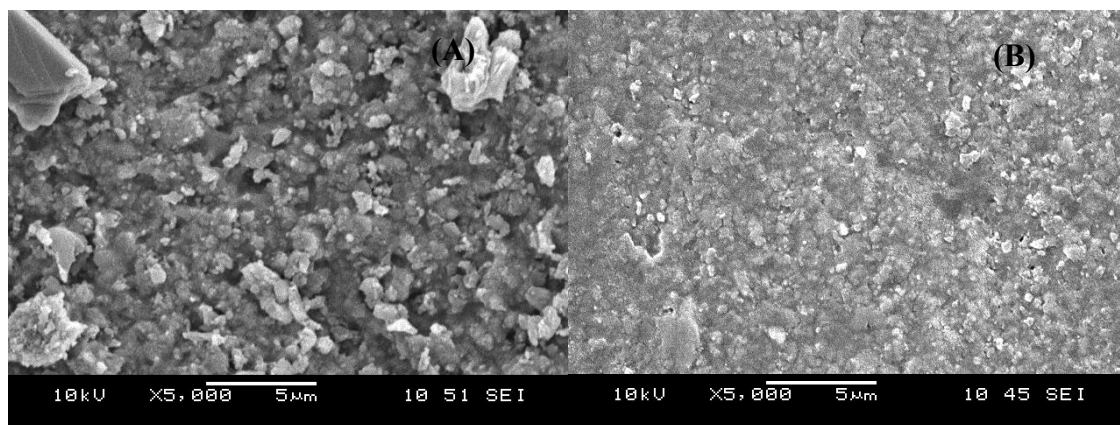


Figure 9 shows the microstructure of the precursor before and after synthesized by calcinations at  $1200^\circ\text{C}$  for 3 hours. The calcined precursor formed a very fine  $\text{Al}_2\text{O}_3$  and grew homogenously after the synthesized, in fact it can produced a good bonding between grains compare to pure  $\text{Al}(\text{OH})_3$  (Z.-Y. Deng et al., 2001). The grain size of pure  $\text{Al}(\text{OH})_3$  are range between 30 to  $35\mu\text{m}$  while for calcined precursor, the grain size are range from 24 to  $27\mu\text{m}$ , where the precursor were finer than before. Precursor obtained a fine grain as more synthesized process undergo, this is proved by the leached precursor that come out with even finer grain size compare to pure and calcined precursor with grain size ranging from 21 to  $23\mu\text{m}$ .

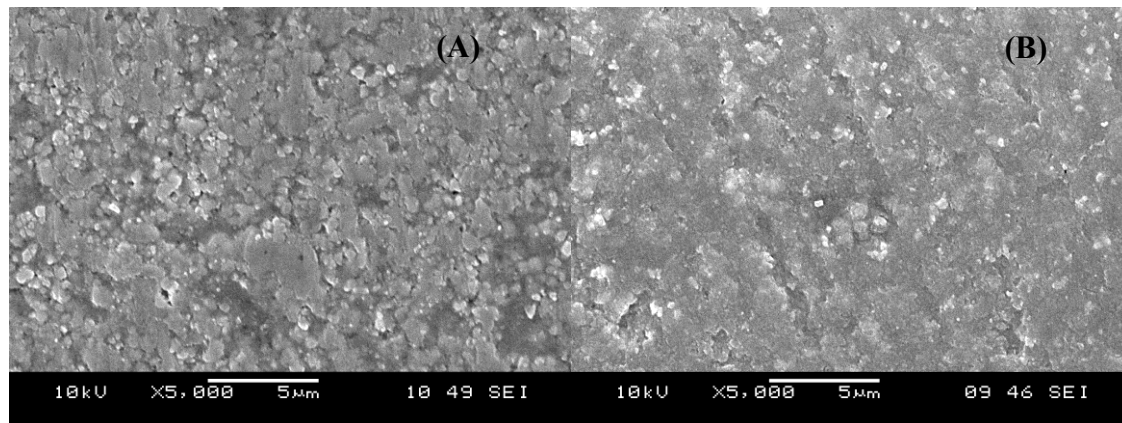
**Figure 10: SEM photograph for sample (A) C12.3 and (B) LC12.3.**



In figure 10, the different of both samples can be seen by the grain bonding, the leached sample (B) showing a great bonding as there is minimum porosity can be detected in SEM photograph, same condition faced by leached samples at different sintering temperature and time. Minimum porosity leading to a higher strength of ceramics but that is not the case for calcined sample (A), the porosity can be seen easily and the proved on this microstructure effecting the mechanical properties of the ceramics.

**Figure 11: SEM photograph for leached sample (A) LC15.3 and (B) LC15.6.**





These two samples in figure 11 were manipulated by sintering time while the temperature remain constant, as can be seen in Figure 11(B), the bonding among the grain is much closer and compact compared with sample in figure 11(A). This can be happen because of the longer sintering time that contribute to a compact grain bonding but the increased in sintering time did not increased the strength of the ceramics bodies which it might caused by local packing disruption where it can reduce the connectivity particles and lowering the strength of those bodies. Local packing disruption happened when the volume change during the synthesise process and the process might keep furthering as the sintering time is longer and temperature is higher. The grain packing is getting compact and the porosity is lower as the sintering temperature is getting higher, this changes can be seen between Figures 10(A), (B) and 11(A),(B).

## Conclusion

It is concluded that by synthesized  $\text{Al}(\text{OH})_3$  with different method can improved the mechanical properties of ceramics bodies but it will be depends on the type of synthesized process. Leaching process was found as the most applicable synthesized process since it produced better properties for ceramics and higher percentage of  $\text{Al}_2\text{O}_3$  in the synthesized precursor. A higher purity of  $\text{Al}(\text{OH})_3$  could be obtained if the molarity of HCl is increase along with a longer calcination time. Sintering give a major effect on the mechanical properties of ceramics bodies by manipulating the sintering time and temperature, increasing the sintering time did not promising in produced a better mechanical properties for ceramics bodies since it can influenced the microstructure of ceramics by generating abnormal grain growth. The Combination of  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  contribute to excellent mechanical properties of ceramics bodies, this combination occur when higher sintering temperature is applied to ceramics bodies. As for the limitation on this research, the sample size can be  $\pm$  by 0.1mm since the shrinkage happen after the sintering process, which it can affect the actual reading when undergo the mechanical testing.

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