

## X-ray Diffraction Phase Analyses of Mullite Derived from Rice Husk Silica

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### Abstract

In this study, mullite synthesized from aluminum nitrate hydrate  $[(Al(NO_3)_3 \cdot 9H_2O)]$  and silica sol from rice husk was subjected to sintering treatment at temperatures of 900, 1000, 1100, 1200, and 1300 °C, and characterized using x-ray diffraction (XRD), followed by Rietveld refinement, and differential thermal analysis (DTA). The results indicated that in the sample sintered at 900 °C, no mullite phase was identified, but crystoballite and alumina were well detected. The formation of mullite started at temperature of 1000 °C and continued to grow at higher temperatures, resulted in increased weight percentage (wt%) from 62.62 to 92.29%, while crystoballite and alumina decreased from 22.42 to 1.25% and from 77.58 to 6.46 % respectively. A good correlation was found between the calculated and observed unit cells. For mullite phase, the unit cell dimensions are  $a = 7.545$  nm,  $b = 7.689$  nm and  $c = 2.884$  nm, for crystoballite  $a = b = 0.5531$  nm and  $c = 0.6923$  nm, and for alumina  $a = b = 0.5026$  nm, and  $c = 1.2808$  nm. The DTA analyses revealed that in the untreated sample, only alumina and silica were detected, while in the sintered samples we found the existence of mullite, alumina, and crystoballite.

### Abstrak

**Analisis Fasa Mullite dari Silika Sekam Padi dengan Metode Difraksi Sinar-X.** Pada penelitian ini, *mullite* yang disintesis dari aluminum nitrat hidrat  $[(Al(NO_3)_3 \cdot 9H_2O)]$  dan sol silika sekam padi dikenakan pada perlakuan *sintering* dengan suhu 900, 1000, 1100, 1200, dan 1300 °C, kemudian dikarakterisasi dengan difraksi sinar-x (XRD) dilanjutkan dengan penghalusan menggunakan metode *Rietveld*, dan analisis termal diferensial (DTA). Hasil penelitian menunjukkan bahwa dalam sampel yang disintering pada suhu 900 °C, tidak ditemukan fasa *mullite*, tetapi fasa kristobalit dan alumina terdeteksi dengan jelas. Pembentukan *mullite* baru terjadi pada suhu 1000 °C dan bertumbuh dengan peningkatan suhu ditandai dengan kenaikan persen berat (wt%) dari 62,62 menjadi 92,29%, disertai penurunan persen berat kristobalit dari 22,42 menjadi 1,25% dan penurunan persen berat alumina dari 77,58 menjadi 6,46%. Hasil penghalusan menunjukkan korelasi yang baik antara unit sel hasil perhitungan dan hasil pengamatan, dengan dimensi unit sel adalah  $a = 7,545$  nm,  $b = 7,689$  nm dan  $c = 2,884$  nm untuk mullite,  $a = b = 0,5531$  nm, dan  $c = 0,6923$  nm untuk kristobalit, dan  $a = b = 0,5026$  nm, dan  $c = 1,2808$  nm untuk alumina. Hasil analisis dengan DTA menunjukkan bahwa dalam sampel yang tidak disintering, hanya terdapat alumina dan silika, sementara dalam sampel yang disintering terdapat *mullite*, alumina, dan kristobalit.

*Keywords: mullite, rice husk, Rietveld, structure, sintering*

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### 1. Introduction

Mullite ( $3Al_2O_3 \cdot 2SiO_2$ ) is composed solely of silicon, aluminum and oxygen, and is the only crystalline phase of  $Al_2O_3$ - $SiO_2$  system stable at atmospheric pressure [1]. Due to its thermal stability and excellent mechanical properties such as hardness, strength and fracture

toughness [2-3], mullite has become an important material in ceramic industries. These interesting properties are contributed by the crystal structure of mullite which is known as orthorhombic with the space group *Pbam* and unit cell dimensions of  $a = 7.540$  nm,  $b = 7.680$  nm and  $c = 2.885$  nm for stoichiometric composition [4]. In this structure, the chains of  $AlO_6$

octahedral are located at the edges and centre of unit cell, running parallel to the *c*-axis. These chains are connected by  $(Al, Si)O_4$  tetrahedral forming double chains of Al-O and Si-O tetrahedral, in which substitution of silicon for aluminum atoms takes place with the simultaneous formation of oxygen vacancies [5-6]. According to Sundaresan and Aksay [7], mullite can be formed by thermal treatment, in which alumina phase dissolves in the silica phase. The crystal structure of mullite demonstrated that increasing the number of oxygen vacancies per unit cell causes the tetrahedral double chains to become gradually disarranged by removal of bridging oxygen atoms, leading to the formation of new  $AlO_4$  [8-9]. The characteristics and performance of mullite depend on the size, the crystallinity, and the purity of the phase. All of them vary with sintering temperature, initial raw material, and processing condition. In an attempt to produce dense mullite ceramics from various raw materials, many routes have been explored. In previous studies, synthesis of mullite from kaolinite has been reported, in which both mullite and spinel phases are simultaneously formed at 980 °C [10], but only mullite was identified at 1000 °C [11]. Both studies also described that mullite formation occurred by nucleation and growth within the aluminosilicate phase resulted from the reaction between  $Al_2O_3$  and  $SiO_2$ , and the growth rate is controlled by the dissolution of  $Al_2O_3$ . Another route that has been investigated for the same purpose is sol-gel approach [12], assuming that densification could be achieved more easily to produce high purity and homogeneous materials. For example, several studies [13-14] reported the synthesis of mullite from mixture of aluminum sulfate and silica, in which the formation of mullite took place at temperature of 1275 °C. It was proposed that at high temperature, mullite will nucleate and grow at the point contact between  $Al_2O_3$  and  $SiO_2$  particles, by diffusion of  $Al^{+3}$   $Si^{+4}$  through the crystal lattice.

Despite progressive studies on mullite, only limited numbers of investigations have been carried out to understand and evaluate quantitative phase composition of mullite prepared from rice husk using x-ray diffraction with Rietveld method. As commonly acknowledged, phase composition is closely related to the characteristics of materials. For this reason, Rietveld analysis is an integral part of material investigation of powder diffraction data [15], which allows the weight fraction of each phase in unknown to be calculated. This analysis is particularly important when the diffraction patterns contain many overlapping reflections due to low symmetry phases or very large unit cell [16]. The method has been applied to gain significant structural information of various ceramic materials such as, metastable tetragonal zirconia [17], crystal structure of mullite [18-19] and cordierite [20]. Another important advantage of Rietveld method is elimination of the effect of impurities on the

diffractogram, therefore more accurate data regarding phase composition can be achieved. This feature is very helpful in investigating phase composition of ceramics derived from rice husk silica considering the presence of various natural impurities in the husk. Many authors investigating rice husk have reported that in addition to silica, which accounts for 87-97% [21], rice husk contains several other inorganic components which combine with the silica when the silica is extracted with alkali solution. The most commonly found are Al, Fe, Ca, Mg, Mn, K, and Na [22-24]. When the silica is subjected to thermal treatment, these elements are transformed into oxides and become impurities to silica which suppress the accuracy of analysis using traditional XRD method. It is then clear that XRD analysis of materials derived from rice husk silica should include the Rietveld processing to compensate the effect of the above impurities in order to produce more accurate data.

The central stage of rietveld process is refinement of the model using the least squares fitting of a calculated diffraction pattern to the measured pattern. This step is aimed to minimize the difference between the observed and calculated diffraction patterns together with the background function, from which structural information of crystal such as atomic position, weight percent, and unit cell could be extracted. Principle calculations of the mathematics of general crystallographic refinement are based on the least square equation developed by Sourt and Jenson [25] and Prince [26]. According to the equation, the model is presumed to be optimum when the sum of the squares of the differences between the observed and calculated patterns is minimum. In practice, the agreement between the observed and calculated patterns is defined in term of agreement indices (*R*), which is divided into four specific terms as described by Wiles and Young [27]. According to Wiles and Young, the expected profile *R* for a perfect fit and parameter is known as the goodness of fit (GOF), and is used to define the success and completeness of the refinement.

The purpose of this present study was to evaluate the formation of mullite derived from rice husk silica, with the emphasis to quantify mullite as the main phase, and to evaluate the secondary phases simultaneously formed as a function of sintering temperature. In addition to scientific interest, utilization of rice husk is a significant innovation toward generation of value added from this agriculture waste abundantly available in Indonesia.

## 2. Experiment

Raw materials used for the experiments consists of rice husk as sources of silica and  $[(Al(NO_3)_3 \cdot 9H_2O)]$  (Merck, kGaA, Damstadt, Germany) as sources of alumina. Preparation of rice husk silica sol was carried out following the procedure previously described [28].

Mullite ceramics were prepared using the sol-gel method suitable proportion of  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}:\text{SiO}_2$  to produce the composite with the molar ratio of  $\text{Al}_2\text{O}_3$  to  $\text{SiO}_2$  of (3:2) [29-30]. Alumina sol was prepared by dissolving 44 grams of  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  in 100 mL ethanol under magnetic stirring for 1 hour. Both of silica and alumina sols were mixed and stirred at 110 °C for 1 hour to get dry gel. Dry gel was sintered at temperatures of 900, 1000, 1100, 1200, and 1300 °C, with a heating rate of 5 °C /min and holding time at peak temperature for 5 hours. Sample for x-ray powder diffraction analyses was prepared by grinding the sample with mortar and pestle to obtain powder with the size of 200 mesh. The x-ray diffraction pattern was recorded with an automated Shimadzu XD-610 X-ray diffractometer equipped with a scintillation counter. The conditions for XRD analysis applied are as follows: source  $\text{CuK}_\alpha$  ( $\lambda = 0.15418$ ), 40 kV and 30 mA, spot size,  $0.02 \times 8 \text{ mm}^2$ , with a  $0.15^\circ$  receiving slit. Patterns were recorded over goniometer ( $2\theta$ ) from  $10$ - $100^\circ$  with a step size  $0.02^\circ$ , counting time per step, 1 s, and scintillation counter beam monochromator with NaI detector. Rietveld analysis [31] was carried out using Rietica software for Windows 95/98/NT version 1.70 [32]. The global optimized parameters were background polynomial coefficients, lattice parameters, zero-shift error, unit cell, scale factor, peak shapes, occupancies and preferred orientation. Pseudo-Voigt function and March models were used to refine peak profile and preferred orientation, respectively [33]. The models of crystal structure for mullite, crystoballite, and alumina, for refinement of the sample were taken from [34-36], respectively. Differential thermal analysis (DTA) was carried out using DTA Merck Setaram Tag 24 S. The thermogram was produced by scanning the sample at temperature range of 30 to 1000 °C.

### 3. Results and Discussion

Based on our investigations in 2010 [29-30], qualitative XRD analysis was conducted by comparing the diffraction lines of the samples with those in standard from The Powder Diffraction File (PDF) [37] files and showed that the major phase obtained for the sample is mullite (PDF-15-0776), with secondary phases of crystoballite (PDF-39-1425), and alumina (PDF-46-1212) as shown in Table 1. The crystoballite and alumina phases originated from rice husk and aluminum nitrate as raw materials are due to the thermal treatment. The results showed that in the sample sintered at 900 °C, only crystoballite and alumina phases are evidently detected. Mullite formation occurred at 1000 up to 1300 °C, accompanied by the secondary phases of crystoballite and alumina. These results also confirmed that the prepared mullite has high homogeneity, which was in accordance with the results by Messing and Hong [38], who concluded that mullite was formed at temperature from 1000 up to 1350 °C. The crystallization

**Table 1. Phase Identification in the Samples Sintered at Different Temperatures for 5 Hours**

Temp (°C)	$2\theta$	Phase
900	21.4	Crystoballite
	35.4	Alumina
1000	21.5	Crystoballite
	29.5	Mullite
	35.4	Alumina
1100	21.3	Crystoballite
	29.5	Mullite
	35.3	Alumina
1200	21.6	Crystoballite
	29.3	Mullite
	35.3	Alumina
1300	21.4	Crystoballite
	29.2	Mullite
	35.4	Alumina

of crystoballite and alumina obtained depends on the temperature, which decreased with increasing temperature. The presence of alumina within the mullite matrix could be explained in term of transformation of mullite that can be viewed as decomposition according to the reaction of  $2(2\text{Al}_2\text{O}_3 \cdot \text{SiO}_2) \rightarrow 3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 + \alpha\text{Al}_2\text{O}_3$  [39]. This result seems to be different with the finding of Septawendar, *et al.* [40] who reported that transformation of mullite followed the steps of  $\gamma\text{-Al}_2\text{O}_3 + \text{SiO}_2$  amorphous  $\rightarrow$  mullite +  $\gamma\text{-Al}_2\text{O}_3$  + amorphous of  $\text{SiO}_2$ . Meanwhile, mullite and spinel were identified for mullitization route of aluminum nitrate unhydrate and TEOS [41]. This varied phases observed reflect the role of raw materials in governing the formation of phases in mullite. According to the results of this current study, it is proposed that mullite start to form at 1000 °C most likely driven by the formation of crystoballite from amorphous silica, because crystoballite has been known as very reactive phase.

It is clear that mullite, crystoballite and alumina phases are formed. To investigate the structural features of these phases, Rietveld analysis was used to obtain quantitative information when more than one phases are present in the sample, therefore it could be used to distinguish the phase composition of the samples sintered at different temperatures. Rietveld analysis was satisfied by figure-of-merits (FoMs) which are for the sintered samples, tabulated in Table 2. These results demonstrate that the crystal structure data used as model together with other refinement parameters to simulate all diffractions achieve high agreements between observed and calculated patterns. According to Kisi (1998) [42], all FoMs are quite acceptable because the goodness-of fit (GoF) values is less than 4 and  $R_{\text{wp}}$  less than 20%. Also, the fitting plots indicates the

accomplishment of the Rietveld refinement, in which different plots between calculated and observed patterns show minor fluctuation (Figure, 1 (a), (b), (c), (d) and (e)). These results also show that the output of the refinement can be used to provide further information such as, phase composition, lattice parameter, and atomic position.

Table 3 illustrates the changes in weight percentage from Rietveld refinement with XRD data for the samples sintered at temperatures of 900, 1000, 1100, 1200 and 1300 °C for 5 hours. For crystalline sample sintered at 900 °C, a two-phase refinement was carried out and indicates the presence of crystoballite and alumina, with the weight percentage of about 22.42 wt% for crystoballite and 77.58 wt% for alumina. The weight percentage of mullite phase was found to increase when the sample sintered from 1000 up to 1300 °C. The increase of mullite is probably due to the polymorphic aluminosilicate which crystallized to mullite, where at these temperatures some of the silica has reacted with alumina to form mullite, crystoballite and alumina simultaneously. These reactions caused the weight percentage of mullite increased and both crystoballite alumina decreased. Evidently, with increasing temperature, the weight percentage for the mullite phase increased markedly from 62.62 wt% to 92.29 wt%, and crystoballite decreased from 22.42 wt% to 1.25 wt%, while alumina decreased from 77.58 wt% to 6.36 wt%.

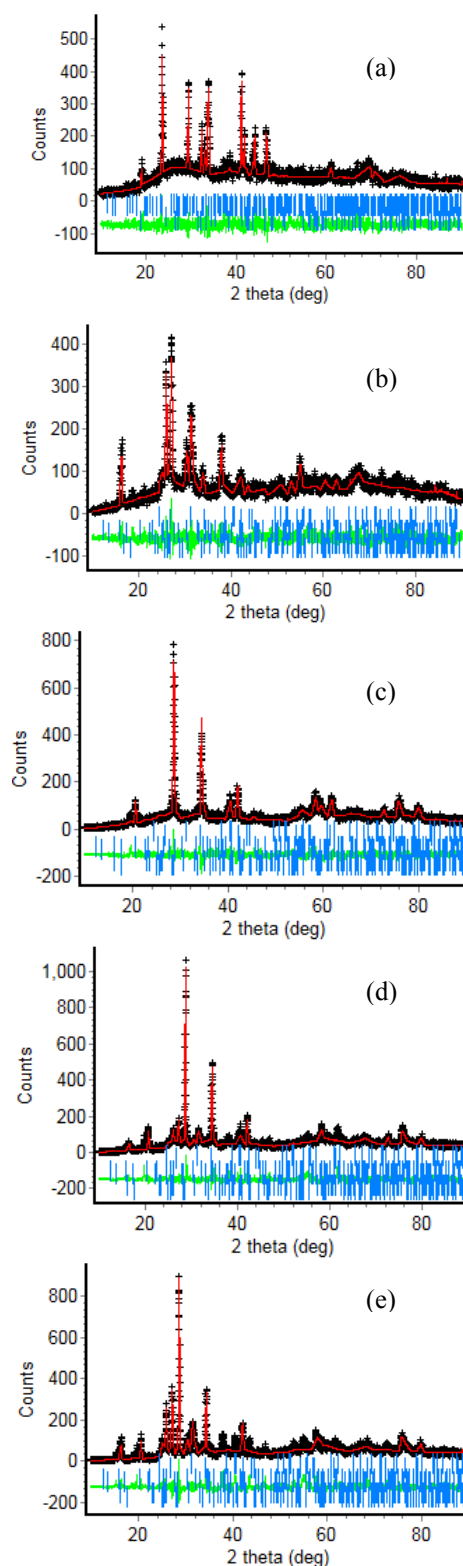
**Table 2. Figures-of Merit (Foms) from Refinement of XRD Data for the Samples Sintered at Different Temperatures for 5 Hours**

Temp (°C)	R <sub>exp</sub>	R <sub>wp</sub>	GoF
900	11.22	12.93	1.32
1000	12.63	15.19	1.44
1100	13.45	16.40	1.48
1200	13.37	18.76	1.96
1300	13.29	19.43	2.54

**Table 3. Weight Percentage (wt %) from Refinement of XRD Data for the Samples Sintered at Different Temperatures for 5 hours. Estimated Errors for the Least Significant Digits are Given in Parentheses**

Temp (°C)	M	C	A
900	-	22.4[2]	77.5[8]
1000	62.6[2]	5.2[1]	32.1[7]
1100	64.0[2]	4.6[7]	31.3[1]
1200	80.6[1]	4.4[5]	14.9[5]
1300	92.2[9]	1.2[5]	6.4[6]

M, Mullite; C, Crystoballite; A, Alumina



**Figure 1. XRD Rietveld Plot for the Sintered Samples. The Observed Data are Shown by the (+) Sign, and the Calculated Data by a Solid Line. The Vertical Line (Blue) Represents the Point Series HKL. The Green Line below the Vertical Lines is the Difference Profile, (a) 900 °C, (b) 1000 °C, (c) 1100 °C, (d) 1200 °C and (e) 1300 °C**

Table 4 shows the parameter cell of model for refined phase, and Rietveld output of the parameter cell for all samples is shown in Table 5. The lattice constants of phase for mullite in the sintered samples agree well for all experimental conditions, but for crystoballite and alumina some differences should be acknowledged. Despite these differences, the results are still in the range of acceptable values. Deviation for crystoballite and alumina observed in this study is most likely resulted from the presence of impurities, which might interfere the analysis.

The XRD analyses clearly indicated that existence of mullite as expected, however this method provides no information regarding the mechanism of the formation of mullite phase. Therefore, to gain some insights on how the process leading to mullite formation took place, the samples were analysed using differential thermal analysis (DTA). Each of the sample was subjected to DTA in the temperature range of 50-1000 °C, producing thermograms compiled in Figure 2.

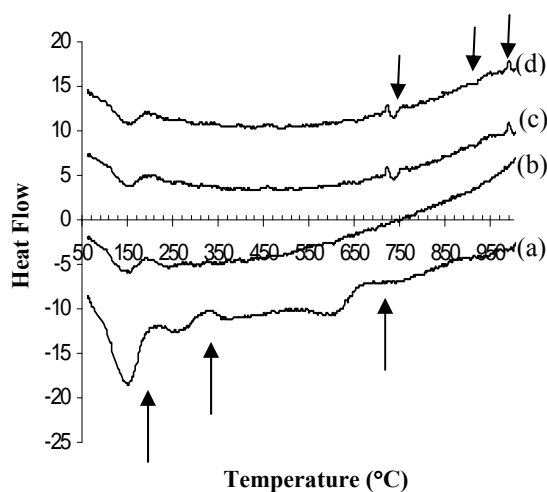
Figure 2a is the thermogram of the sample without sintering treatment, showing the existence of three peaks located at around 50-150, 250-360, and 650-800 °C (indicated by arrows), respectively. The peak in the range of 50-150 °C is most likely due to the evaporation of water and residual organics probably present during the preparation of sample. The peak at around 250-360 °C could be assigned to the dehydration of silica and aluminum hydroxide which is in agreement with the results of others [43]. The peak around 750-800 °C is assigned to the formation of crystoballite and alumina, based on the results obtained from XRD analyses previously described (Table 1).

**Table 4. Parameter Cells of the Model Used for Refinement of the Samples (Unit Cell, Space Groups)**

Mullite	Crystoballite	Alumina
a (nm) = 7.5433	a (nm) = 0.4971	a (nm) = 0.4754
b (nm) = 7.6917	b (nm) = 0.4971	b (nm) = 0.4754
c (nm) = 2.8840	c (nm) = 0.6923	c (nm) = 1.2982
orthorhombic/Pbam	Tetragonal/P4 <sub>1</sub> 2 <sub>1</sub> 2	Hexagonal/R-3C

**Table 5. Parameter Cells of the Rietveld Out Put for the Samples (Unit Cell, Space Groups)**

Mullite	Crystoballite	Alumina
a (nm) = 7.5450	a (nm) = 0.5531	a (nm) = 0.5026
b (nm) = 7.6890	b (nm) = 0.5531	b (nm) = 0.5026
c (nm) = 2.8840	c (nm) = 0.6923	c (nm) = 1.2808
orthorhombic/Pbam	Tetragonal/P4 <sub>1</sub> 2 <sub>1</sub> 2	Hexagonal/R-3C



**Figure 2. DTA Scan of the Sample (a) without Sintering, (b) 1100, (c) 1200 and (d) 1300 °C**

In the sintered samples (Figure 2b to 2d) it can be seen that the peak associated with evaporation of water is much smaller compared to that observed in the sample without sintering, suggesting that sintered samples contain less water and are more dense. In addition, it is quite evident that phase transformation has taken place as indicated by increased thermogram lines from 750 to 1000 °C, indicating that crystoballite, alumina, and mullite have been produced. The formation of these three phases can be seen more evidently in the samples sintered at 1200 and 1300 °C, as indicated by the peaks at around 750, 900, and 1000 °C (by arrows), which are assigned to crystoballite, alumina, and mullite, respectively. Based on these results of DTA, it can be concluded that sintering process plays an important role in facilitating the formation of mullite.

#### 4. Conclusions

Successful application of the Rietveld method for quantitative phase analysis and for structure refinement of mullite prepared from rice husk by sol-gel process was achieved in this study. Application of the method was found to enable the elimination of the interference of natural impurities in the rice husk silica. The results revealed that the formation of mullite is as a function of sintering temperatures. The refined data of XRD results show that the weight percentage of the mullite phase increased with increased temperature, and secondary phases (crystoballite and alumina) decreased with increased temperature. From the results of DTA analyses it can be seen that sintering treatment promotes the formation of mullite. Overall, the results indicated that rice husk silica was a potential starting material for synthesis of mullite, in particular, and silica based materials in general.

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