Determination of Sintering Temperature in Synthesis of Gelcasted Porous Ceramic

[1]SuriatiEka Putri, [2]Diana Eka Pratiwi, [3]Rachmat Triandi, [4]Diah Mardiana, [5]Iwan Dini

[1,2,3,4,5]Lecturer
[1,2,5]Department of Chemistry, *Faculty of Mathematics and Natural Science*
[1,2,5]Universitas Negeri Makassar, Makassar, Indonesia
[3,4]Department of Chemistry, *Faculty of Mathematics and Natural Science*
[3,4]Universitas Brawijaya, Malang, Indonesia

***Abstract*—The aim of this study was to determine the temperature of gelcasted ceramic to obtain maximum pore. Pore formation occurs when the polymer system in ceramic body is being degraded. Polymer used was acrylamide (AM) with crosslinker methylen bis acrylamide (MBAM) and the raw material was natural clay. The method of synthesis porous ceramic was gelcasting method which produce polymer in the slurry as a green body ceramic. Determination of sintering temperature using TGA / DTA method. Based on the analysis, the sintering process of gelcastedcrude ceramic body was carried out at the temperature of 28 °C - 1100 °C with a certain treatment of temperature and time. The sintering process using heating rate of 50 °C/15 minutes with 60 minutes detention at the temperature of 100 °C, 200 °C, 500 °C, and 1100 °C.**

***IndexTerms*—Clay, Ceramics, Gelcasting, Sintering**

I. INTRODUCTION

There are so many benefit of porous material in chemical study either as adsorbent or assupport catalyst. Porous ceramic is one example of porous material that commonly used. According [1], the method of synthesis porous ceramic is currently being developed. Porous ceramics are very widely used in several field as support catalysts[2], in the health sector as a proponent of artificial bone, and in the food sector as media immobilization of bacteria in the manufacture of yeast. Some products in Indonesia were obtained from abroad. Thus, it is necessary to improve the technology of synthesis porous ceramics in Indonesia by using natural materials.

The natural materials may used as raw material in ceramic synthesis are materials contained alumina-silica as main components. One of the materials is natural clay. Traditional methods have been developed in the manufacture of porous ceramic, such as dry-pressing [3], extrusion [4], and slip casting. However, themethods needs high pressure, porous molds, and the methods are fairly complex [2].

One example of ceramic synthesis method that has not been developed in Indonesia is gelcasting method. The basic principle of this method is in situ polymerization, in which the monomers and crosslinkers undertake a polymerization reactions within the premix slurry solution. This method has several advantages; the technique need low cost with fairly easy to use, produce high porosity, and the mold used can be varied. However, it is difficult to produce a homogeneous pore size using the gelcasting method [1].

The gelcasting method use monomer, crosslinker, initiator and catalyst with the principle of in situ polymerization in a ceramic slurry. Based on the research result [5] the high porosity in ceramic body mayproduced using high purity chemical compound, e.g. monomer acrylamide (AM). Acrylamide is one kind of monomer which has one double bond, therefore it just produce a linear polymer. Linear polymer would be difficult to act as pore template, so that crosslinkers are needed. One kind of crosslinkeris methylenbisacrilamide (MBAM) which produce gel if react with aquades. Crosslinker needed if the organic monomer used only contain one double bond until produce linear polymer chain [6]. The reaction between AM and MBAM to produce chain polymer called addition polymerization reaction. So that, it need initiator to form a radical ion from monomer. An example of initiator can be usedalso polymerization is ammonium persulfate (APS) with a tetrametyldiamin (TEMED) as a catalyst.

The amount of polymer used about 40 % of green body ceramic [2]. The pore formation process occurs when the polymer isbeing degraded from the ceramic body, namely the sintering process. Therefore, the sintering temperature is very important to be assessed in the synthesis of porous ceramics. Also, sintering is also influential on hardness and morphology of ceramics [7].

II. RESEARCHMETHODS

The specifications of the materials used in this work are natural clay which similar to that of Ref. [8][9].The natural clay is dried and then homogenized. After that, it is meshed up to 40-80 mesh size. The sample used is the sieved powder. Natural clay of 2.8 g is added into premix solution that contains of AM monomers and MBAM crosslinker (40%: 2%) with the solids loading of 50 vol % [10][11] and mole ratio of AM monomer and MBAM crosslinker is 12: 1. The resulting suspension is homogenized for approximately 20 minutes, then initiator and catalyst are added; 40 µL of 10% APS and 20 µLTEMED 99 %. Then, the dough is stored in a mold until the dough dry. When the dough is dry then it can be removed from the mold.

The green body of gelcasted ceramic is dried for two days in the air. The crude gelcasted ceramics further analyzed using TGA/DTA with the heating rate of 10 ^{o}C/minutes and the initial weight of ceramics is 62.3 mg observed the change of ceramic weight to the temperature changes.

III. Result and Discussion

In the sintering process, the ceramics are burnedafter the molding process toobtain stronger and more solid ceramic products. Sintering temperature is determined so the release of organic compounds are expected to become the pore template is complete. In this study polyacrylamide acts as pore template which is crosslinked with MBAM.

In this study, the sintering temperature is determined after the thermal analysis of the gelcastedraw ceramic using TGA / DTA. Temperature measurement data showed a weightreduction of the ceramic and the phase changesof some mineralsin clay A sample. Thermal analysis results of gelcastedcrude ceramicare shown in Figure 1.

Based on Fig. 1, in the sintering process, detention is held for 60 minutes at the temperature of 100 °C, 200 °C and 500 °C. At a temperature of 100 °C dehydration (release of water molecules) occur with a weight reduction of 6.31% of the initial weight of the ceramic. At a temperature of 200 ^{o}C, the phase changes from quartz into trydinitis predicted, while at a temperature of 500 ^{o}C some chemical reactions occur, for example curing (hardening) and oxidation with a weight reduction of 34.99% of the initial weight of the ceramic. At the temperature of 600 °C to 1100 °C ceramics weight reduction has been constant so the heating is carried out atthe temperature of 550 °C to 1100 °C with a heating rate of 50^{o}C/15minutes. When the temperature of 1100 ^{o}Cis reached, detainedis carried out again for 60 minutes.

The heating is carried out to a temperature of 1100 ^{o}C due to the phase transformation of γ-Al_2O_3 into α-Al_2O_3 occurs at temperatures above 1000 °C which will produce microstructures with a fairly high degree of porosity [6].The percentage of mass reduction is 51,77 %.

In the sintering process, at the temperature range of 200 ^{o}C to 250 ^{o}C pungent odor is produced; it is predicted as the releasing reaction of NH_3 gas in polymer systems. This process is reinforced by the mass reduction in the temperature range 200-250 °C in the TGA curve which is equal to 1,925 mg.

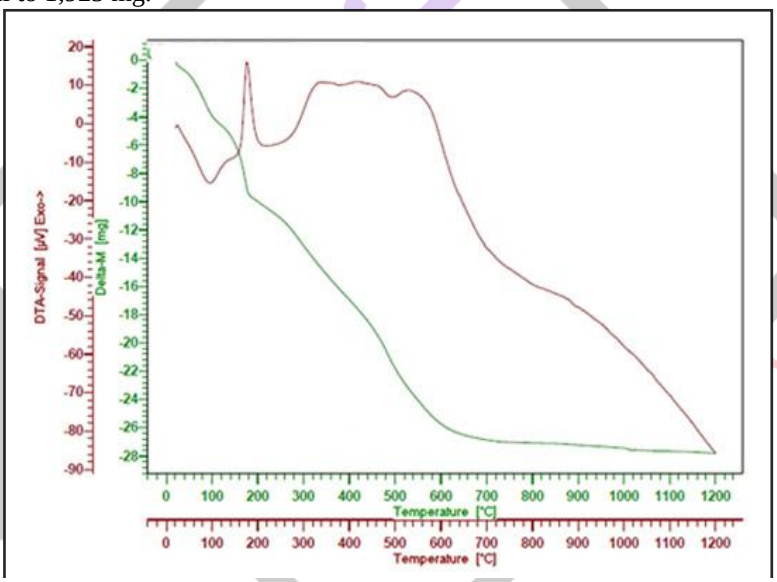


Fig 1: Thermogram of gelcastedcrude ceramic thermal analysis using TGA / DTA

There are two stages of degradation of polyacrylamide which is at a temperature of 230 °C and 400 °C respectively the release of NH_3 gas and CO_2 gas [12]. The release of $NH_3$and CO_2respectively are shown in the Fig. 2.The oxidation process at a temperature of about 500 °C is marked by changing color into black ceramic body and at a temperature of 600 °C to be red, it is predicted the metal oxidation of Fe^{2+} into Fe_2O_3. Weight reduction at a temperature of 500 ^{o}Cis predictedas the releasing reaction of CO_2 gas in a polymer system. At the temperature of 600 °C to 1200 °C by DTA curve indicating a decline in the occurrence of decomposition (degradation of the polymer system).

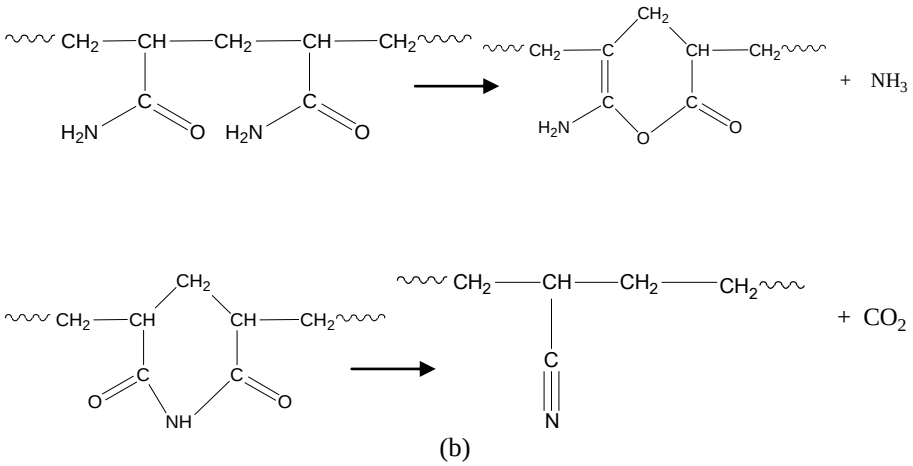


Fig 2 : Degradation of polyacrylamide (a) release of NH_3 and (b) release of CO_2

The data is supported based on IR absorption data. Based on Fig. 3 IR spectra absorption occurs at the 1645 cm^{-1}indicate the presence of the primary amide group (-$CONH_2$-) and on the absorption at 3454 cm^{-1} indicate the presence of secondary amide group (-CONH-). It indicates NH_3 released from green ceramic body and produce the pore.The absorption of inorganic compound from raw material of ceramic occur at 1100 cm^{-1} to 300 cm^{-1}. Therefore the absorption above 1100 cm^{-1}indicates the organic compound from degradation of polymer chain.

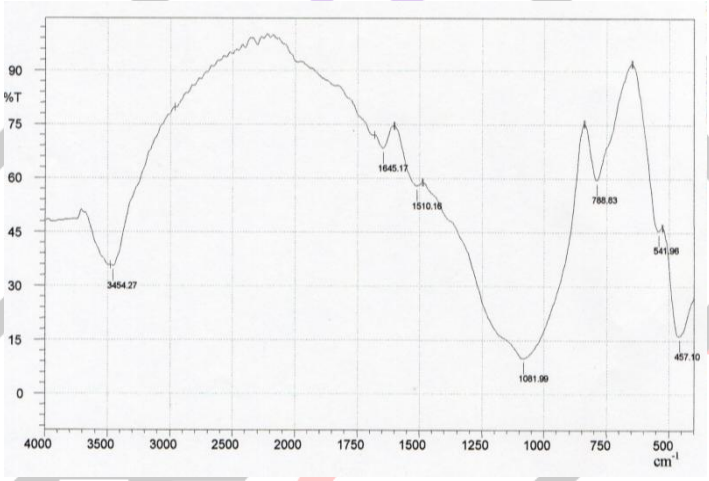
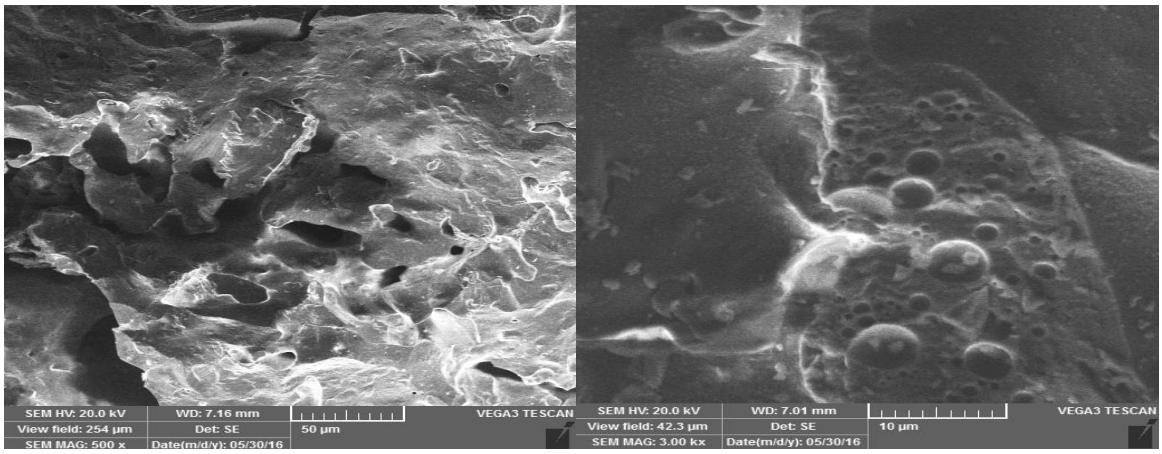


Fig 3: IR Spectra of sintered porous ceramic

Ceramic character which the sintering temperature produce the hardness of porous gelcasted ceramic 97 HB. It is higher than sintering temperature without detention at 100 ^oC, 200 ^oC, 500 ^oC and 1100 ^oC, it produce the hardness of porous ceramic 70 HB [8]. Morphology of porous ceramic at the sintering temperature shown at the Fig. 3a and the morphology of porous ceramic according TGA/DTA results shown at the Fig. 4b.

The pore distribution and pore types at Fig. 4a is not homogeny whereas Fig. 3b use the temperature based on TGA/DTA results produce the pore distribution is more homogeny and the surface area is larger. It indicate the degradation of polymer chain is completed at the sintered temperature. The size and surface area should be analyzed by BET at the next research.

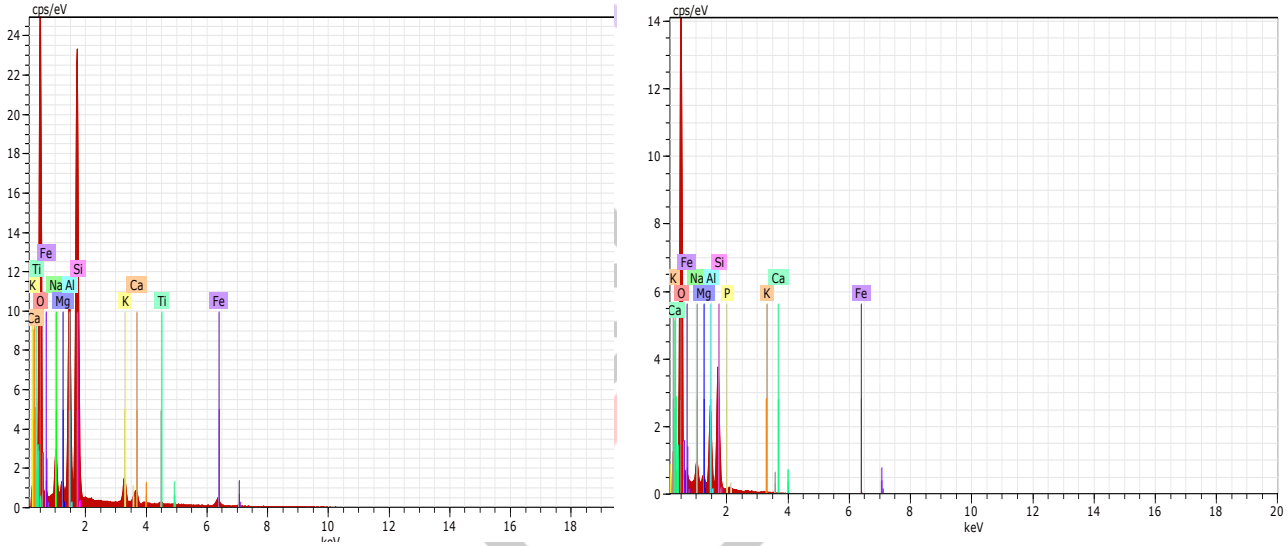
The pore distribution of Fig. 4a and Fig.4b is not homogeny, due to the polymerization rate in green body ceramic was not controlled. Types of pore on gelcasted ceramic consist of opened pore and closed pore. Closed pore are a cavity trapped in solids and no access to the surface meanwhile opened pore has access to the surface even though the cavity is in the middle of solid.



(a) (b)

Fig 4:Morphology of porous ceramic: (a) without detention; (b) based on TGA/DTA results

Based on Fig.5, the amount of oxygen gas increase from 28.30 % to 58.23 %. It indicate the gas was trapped in pores of gelcasted ceramic body. The content of ion Fe^{2+} decreased from 2.80 % to 0.77 %, it indicated this ion has been oxidized at sintering process. The high content of alumina-silica before and after sintered showed that clay as a raw materials was not degraded on sintering process,i.e. Al_2O_3 of 25.73 % and SiO_2 of 55.10 %. Some minerals content on ceramic body was degraded on sintering process, the content of Na_2O decreased from 12.23 % to 7.04 %; the content of MgO decreased from 6.83 % to 2.80 %; the content of K_2O decreased from 3.13 % to 0.78 %. Thus, it suspected these minerals also contribute to the formation of pores. However, the released of CO_2 and NH_3 gases from gelcasted ceramic body cannotbe observed through SEM-EDX data.



(a)(b)

Fig 5: Spectrum of porous ceramic by SEM-EDX (a) before sintered; (b) after sintered

IV. CONCLUSION

Based on the analysis using TGA / DTA, the sintering process of the gelcasted crude ceramic body is carried out at the temperature of 28°C - 1100 °C with a certain treatment of temperature and time. Heating is carried out at a heating rate of 50 °C / 15 minutes with 60 minutes detention at the temperature of 100 °C, 200 °C, 500 °C, and 1100 °C.

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