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The utilization of Glass Waste to Synthesize Zeolite: Effect of Hydrothermal Temperature Variations

Sriatun[1], Hoki Perdananingtyas, Pardoyo

Department of Chemistry, Faculty of Science and Mathematics, Diponegoro University
Semarang Indonesia 50275
e-mail: sriatun71@gmail.com[1]

Abstract*: The utilization of household furniture glass waste as a source of silica is used to synthesize zeolite. This research aimed to obtain synthetic zeolite from glass waste and to determine the effect of hydrothermal temperature on crystallinity and zeolite type. The synthesis begins with extracting silica from glass waste using sodium carbonate. Then it is reacted with sodium aluminate to produce a gel. The hydrothermal process was carried out for 4 days at various temperatures, namely 80℃, 100℃, and 120℃. Products are analyzed FTIR, XRD, and GSA. The product at a hydrothermal temperature of 100℃ has the best crystallinity and belongs to the phillipsite mineral type where 2θ = 28.424˚; 17.814°; 21.837°; 12.052°; 33.693°, and has the largest surface area of 147.22 m^2/g.*

Keywords—glass waste; hydrothermal temperature; zeolite; crystallinity

1. INTRODUCTION

The glass comes from materials that are liquid but have a high density, and an amorphous structure. The atoms do not form an orderly network, like crystals, or what is commonly called glass. Glass is mostly made from silica (SiO_2), a mixture of sandstone and flux which produces a low viscosity and melting point, which is then mixed again with a stabilizer to make it strong.

Glass is widely used in various fields, ranging from household appliances, transportation, laboratories, buildings, and others. The wide use of glass in various aspects of life causes the emergence of potential waste from glass. Meanwhile, the inorganic materials that makeup glass cannot be decomposed by organisms, so it is necessary to make efforts to utilize this waste into more useful materials. The utilization of glass waste as a more useful material has been done quite a lot. The main component of glass is silica [1], so silica is the most easily utilized substance compared to other substances in glass waste. Generally, glass waste consists mainly of SiO_2 (>60 %) and Al_2O_3 (>10 %), and other alkaline oxides [2].

Currently, most of the processes used to recycle the collected glass waste are suitable for environmental and economic aspects. Various studies have been carried out on utilizing glass waste. Because silica is the main component of glass waste, the solution is to use it as a source of silica to synthesize zeolite. Zeolite Na-LTA was prepared hydrothermally at 60℃ for 6 days from glass waste derived from glass fluorescent tubes and pieces of aluminum [3]. The synthesis of Na-LTA was controlled hydrothermal time (2, 4, 6, and 8 days), particle size (< 63 µm: GP-63, < 125 µm: GP-125 and < 315 µm: GP-315), the mass of aluminum scrap dissolved in the aluminate solution (0.125; 0.25; 0.5; 0.75; 1.0 g) and solid /liquid ((5, 10, 15 and 20). The synthesized Na-LTA zeolite was modified with zinc cations via ion exchange to improve its anti-bacterial properties. Zn-LTA was used to develop gelatin-based biopolymer film composites [4].

Zeolite LTA also was synthesized from alum sludge and glass waste at 700℃ for 2 h via alkaline fusion preactivation followed by hydrothermal treatment at 80℃ for 5 h. Increasing the SiO_2/Al_2O_3 ratio of the feed mixture from 1 to 2.3 doubled the yield of zeolite LTA to 67% [5] [6]. Research topics that utilize waste materials as a source of silica to make zeolite or silica alumina-based materials include the use of bagasse ash [7] [8].

The conversion of glass waste to zeolite has also been carried out through microwave radiation (MWR). At lower temperatures and shorter reaction times, the MWR improves more uniform grain size compared to conventional processes. This shows that the MWR was more efficient due to the interaction of electromagnetic waves with materials, thereby enhancing the interaction process, reducing densification temperature, and improving the uniformity of the product microstructure [9]. Even though, hydrothermal synthesis of zeolite is preferred because good crystal growth requires a longer time. Temperature, hydrothermal time of 4-10 days, and other parameters have also been studied [10] [11] [12].

This study aims to obtain zeolite from glass waste and determine the effect of hydrothermal temperature on the product zeolite properties such as crystallinity structure, pore size, and surface area.

2. METHODOLOGY

2.1 Materials and equipment

The materials were used glass waste, distilled water, Na_2CO_3, NaOH, $Al(OH)_3$, and CH_3COOH. Meanwhile, the equipment and instruments used were glassware, hot plates, magnetic stirrers, pH indicators, Wathman® filter paper, Teflon vessels, ARISTON® ovens, VULCAN® furnaces, nickel crucibles, analytical balances, AAS Perkin-Elmer 26

spectrophotometer, Thermo Nicolet Avatar 360® FTIR Spectrophotometer, Shimadzu 6000® X-RD Diffractometer.

2.2 Preparation of Sodium Silica and Sodium Aluminate

The glass waste is washed and dried. After that, the glass is destructed to dry until fine glass powder is formed and sifted to 100 mesh size. The extract of silica from glass waste was prepared by 10 grams of glass powder was added 17.6 g of Na_2CO_3, mixed thoroughly, then heated at 900℃ for 5.5 hours. This process produces solid sodium silicate. The solid was dissolved in distilled water. In the next step, the mixture was filtered to obtain sodium silicate filtrate. The dissolved silica content was determined by AAS.

Sodium aluminate is prepared by dissolving 30 grams of NaOH p.a in 100 mL of distilled water, then stirring with a magnetic stirrer at 100 ℃ until boiling. Twenty grams of $Al(OH)_3$ were added to the solution little by little while stirring until dissolved. Then it was diluted to 250 mL and then allowed to stand at room temperature to obtain sodium aluminate.

2.3 Synthesize Zeolite

The volume ratio of sodium silicate: sodium aluminate is 21:4. The mixture is stirred until homogeneous and conditioned at pH 12. Then the solution is put into a Teflon vessel for the hydrothermal process for 4 days with temperature variations of 80℃, 100℃, and 120℃. The product is filtered and washed to neutral pH, then dried to constant weight. The final product is characterized by FTIR, XRD, and GSA.

3. Results And Discussions

The zeolite product obtained from this synthesis process is expected to have good crystallinity, large pore size, and large surface area. The source of silica used to synthesize zeolite comes from household glass waste. The extracted silica is only 23.19% of the mass of the glass powder. This is probably due to the high levels of additives such as alkaline oxides (Na_2O) and alkaline earth (CaO, MgO, K_2O) [13].

The formation of zeolite from sodium silicate and sodium aluminate is initiated by the formation of a transparent gel. The setting at pH=12 is related to the relatively large concentration of OH^- in the solution system causing the number of $Al(OH)^{4-}$ species to decrease due to the dehydration process to form AlO^{2-} ions or as a dimer, namely $[(OH)_3Al\text{-}O\text{-}Al(OH)_3]^{2-}$. The dehydration process will facilitate the formation of a silica-alumina framework in zeolite [14]. The pH in alkaline conditions also aims to reveal the presence of the main ion forming the silica-alumina framework, namely SiO_4^{4-} .The formation of a gel (gelation process) indicates an interaction between silicate and aluminate ions to form a long chain of silica alumina polymer [15]. The formation of a white gel is the beginning of the formation of nuclei and crystal growth which is important in the synthesis process. This process begins with a condensation reaction and is followed by polymerization of SiO_4^{4-} and AlO_4^{5-} monomers and then a saturated solution forms Si-O-Al bonds. In the condensation reaction, polymer chains with various variations, namely Si-O-Si and Si-O-Al, are formed with a by-product in the form of H_2O. The formation of an alumina-silica framework also depends on the Si/Al ratio. The excessive amount of Al causes the silicate monomers to be unable to compensate for the formation of alumina-silicate polymers.

3.1 Effect of Hydrothermal Temperature on Functional Groups in Zeolite Products

The functional groups of the zeolite products in the fingerprint region were observed at various hydrothermal temperatures, namely 80℃, 100℃, and 120℃ for 4 days. The observed absorption is wavenumber 300-4000 cm^{-1}.

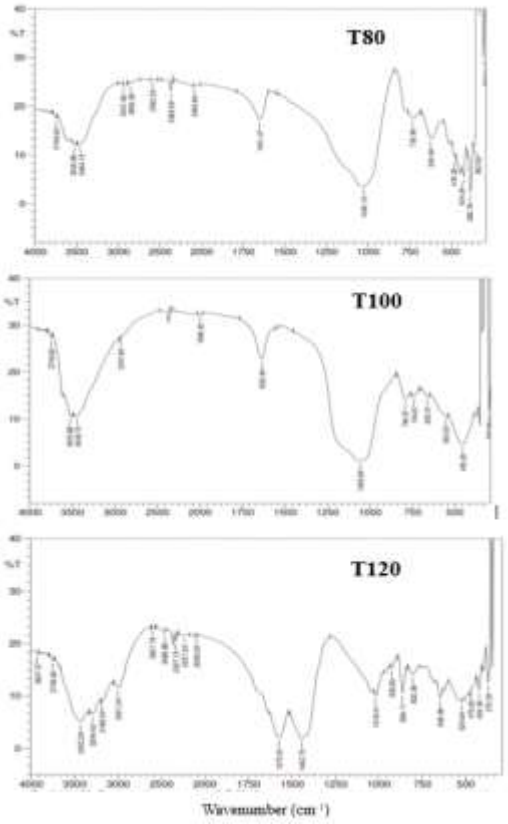

Fig. 1. *FTIR spectra of zeolite products at various hydrothermal temperatures*

Fig. 1 shows the absorption area of 400-300 cm^{-1} shows the presence of pore opening in all samples, whereas T80 (300.9 cm^{-1}), T100 (385.76 cm^{-1}), and T120 (370.33 cm^{-1}). Sample T100 showed the highest intensity. The presence of Si-O-Si bending vibrations was shown by all samples, samples T80 (455.2 cm^{-1}), T100 (424.34 cm^{-1}), T120 (424.34 cm^{-1}), where the sharpest peaks are shown in sample T80.

External linkage (double ring) also appears in all samples, T80 (555.5 cm^{-1}), T100 (624.94 cm^{-1}), and T120 (524.64 cm^{-1}). The double ring is a specific character shown with the appearance of absorption in the area of 650-500 cm^{-1}, this is an

external link between one polyhedral and another. External linkage shows the formation of a 3-dimensional framework. The best 3D framework formation is shown in the T100 sample, although the intensity is still low. The wavenumbers 800-700 cm^{-1} are Si-O stretching vibrations of Si-O-Si [16] only shown in samples T80 and T100, in samples T80 (740.67 cm^{-1}), T100 (732.95 cm^{-1}).

All samples showed the absorption of asymmetric stretching vibration O-T-O from TO_4. Sample T80 (1056.99 cm^{-1}), T100 (1026.13 cm^{-1}), and T120 (1018.41 cm^{-1}, sample T100 had the highest intensity. An absorption at wavenumbers 1700-1500 cm^{-1} also appears in all samples, this absorption indicates the O-H bending vibration of H_2O. The sharpest absorption was shown in samples T80 (1635.64 cm^{-1}), T100 (1651.07 cm^{-1}), and T120 (1573.91 cm^{-1}). The smaller the intensity of H_2O, the less OH^- was formed so more Si-O is formed. The number of Si-O bonds formed indicates the number of frameworks, while the framework indicates the crystallinity of the sample. The sample T100 shows the smallest O-H intensity, from the explanation above, it has the highest crystallinity. Absorption at wave numbers 1110-1000 cm^{-1} appears in all samples. This absorption is the strongest, originating from the asymmetric stretching vibrations of internal and external tetrahedral Si-O-Al and Si-O-S,i and is a characteristic of silica-alumina materials [16].

3.2 ZEOLITE PRODUCT CRYSTALLINITY

Each crystalline solid has a characteristic diffractogram pattern. Compounds analyzed using X-Ray Diffraction are compounds that have a crystalline structure. The X-ray diffraction method is commonly used because it does not damage the structure of the compound being analyzed.

Fig. 2 shows that samples T80, T100, and T120 have different diffractograms from each other. This is seen from the peaks that appear at the diffraction angle of 2Ɵ and different intensities. The increase in temperature does not cause a significant change in the hydrothermal product diffractogram pattern. The phenomenon that arises due to the increase in temperature increases the intensity of the crystalline phase and decreases the amorphous phase [14].

Hydrothermal temperature is an important parameter in the synthesis of zeolite because it affects crystal growth. Samples T80, T100, and T120 experienced an increase in crystallinity due to an increase in temperature, with increasing temperature, it will increase the effective collision so that it will increase the formation of zeolite crystals [15]. The crystallinity of a substance can be seen from the crystal structure that is formed.

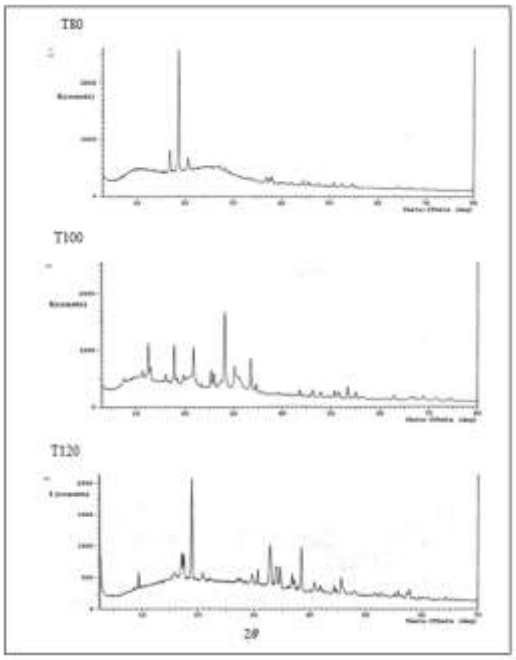

Fig. 2. *The XRD pattern of synthesized zeolite*

Identification of modified compounds aims to determine specifically the type of compound obtained from the results of synthesis with databases such as JCPDS, Ruff, or ICDD. The results of the identification of the diffraction peaks of the synthesized zeolite products with JCPDS are presented in Table 1.

Table 1. The value 2 theta zeolite synthesis product

Standard (JCPDS)			Samples		
Name	***2 Theta (°)***	***Intensity (counts)***	***Name***	***2 Theta (°)***	***Intensity (counts)***
ZSM-23 (ID 43-0015)	18.864	100	**T80**	18.698	100
	16.920	20		16.896	18
	18.104	20		18.400	14
	20.089	20		20.628	10
	37.582	50		37.991	6
Philipsite (ID 47-0162)	28.424	100	**T100**	28.123	100
	17.814	30		17.791	49
	21.837	60		21.778	48
	12.502	40		12.574	47
	33.693	60		33.504	43
Tosudite (ID 12-0231)	18.596	100	**T120**	18.886	100
	38.311	25		38.445	47
	32.295	25		32.840	43
	17.846	25		17.094	24

Table 1 shows the T80 sample is similar to ZSM 23 (ID 43-0015), the T100 sample is similar to phillipsite (ID 47-0162), and the T120 sample is similar to tosudite (ID 12-0231). Besides that, the sample also has other peaks which indicate

the synthesis product has more than one constituent component. The T100 sample contains the philipsite component and the ZSM-23 component which is indicated by the diffraction angle of 30.212°; 25.428° and 26.021°. Meanwhile, in the T120 sample, apart from the tosudite, there was also lusod at 2Ɵ = 34.876°; 33.080° and 33.940°.

3.3 THE PORE STRUCTURE OF THE SYNTHESIS PRODUCT

The pore size classification according to IUPAC is micropore (d < 20 Å), mesoporous (20 < d < 500 Å), and macropore (d > 500 Å) [17]. This classification was based on the adsorption of nitrogen gas at a normal boiling point by porous solids with a large range of pore sizes. The pore size distribution is an important aspect of adsorption and is necessary for the evaluation of porous media as well as membranes. Pore size distribution influences adsorption capacity, adsorption kinetics and adsorption selectivity, and is the basis for adsorbent synthesis. The average pore radius, specific surface area, and total pore volume of samples T80, T100, and T120 are presented in Table 2.

Table 2. The data of pore structure of zeolite synthesis product

Samples	Average of pore radius (Angstrom)	Spesific surface area (m^2/g)	Total pore Volume (cm^3/g)
T80	13.379	48.4026	32.378
T100	13.257	147.2226	97.584
T120	14.149	21.3001	15.069

Table 2 shows that the pore size of each sample, namely sample T80 (13.379 Å), T100 (13.257 Å), T120 (14.149 Å), is a microporous material with a size < 20 Å. The average pore radius of all samples also shows the pore size of the synthesized zeolite included in the microporous material. All zeolite products of synthesis have a less uniform pore size.

The temperature variations in samples T80 and T100 have almost the same average pore size but the specific surface area and total pore volume are very different. This indicates that T100 has more pores than at T80. The hydrothermal temperature at T100 is higher than T80, thus more pores are formed thereby increasing the specific surface area and total pore volume.

T120 has an average pore size larger than T80 and T100 but the surface area and total pore volume are not larger than samples T80 and T100. This indicates that although T120 has a larger pore size, not as many pores are formed as T80 and T100. The higher the hydrothermal temperature, the larger the pore size formed, this will also be followed by an increase in surface area and total pore volume. T120 has a smaller surface area and total pore volume than T100, this is due to the crystallinity of the T120 sample which is smaller than T100, because the hydrothermal temperature at T120 is greater than T100. The higher the hydrothermal temperature (further increase) will cause a decrease in crystallinity. Lower crystallinity makes the framework that is formed more brittle, causing the surface area and total pore volume to be small.

4. CONCLUSIONS

Glass waste can be used to make zeolite through the hydrothermal process. Based on the zeolite character, the resulting hydrothermal temperature of 100°C (T100) produces zeolite with high crystallinity, TO_4 group strength, and a pore size of 13.257 Å, a surface area of 147.2226 m^2/g and a total pore volume of 97.584 cm^3/g.

5. ACKNOWLEDGMENT

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