

# Conversion of Coal Fly Ash to Zeolite by Alkaline Fusion-Hydrothermal Method: A Review

Tifa Paramitha<sup>1\*</sup>

<sup>1</sup>Department of Chemical Engineering, Politeknik Negeri Bandung, Bandung, Indonesia

\*Corresponding author. Email: tifa.paramitha@polban.ac.id

## ABSTRACT

Coal fly ash is one of the industrial by-products that is produced from coal combustion. A large amount of coal fly ash is currently dumped at a landfill, while the current utilization of coal fly ash has been limited to the raw material of soil amendment production and cement concrete. Coal fly ash can be potentially utilized to be the raw materials for zeolite synthesis due to the high amount of silica and alumina content. Zeolite is a large group of minerals consisting of aluminosilicates that are balanced by calcium, sodium, or potassium. One of many methods to synthesize zeolite is the alkaline fusion-hydrothermal method. By using this method, it results in the high quality and yield of product. This study intended to discuss the procedure of zeolite synthesis from coal fly ash by alkaline fusion-hydrothermal method and to know the effect of operating conditions. The journal articles indicate that there are four steps to convert coal fly ash by alkaline fusion-hydrothermal method. These steps include pretreatment process, alkaline fusion process, aging process, and hydrothermal process. Aging process is useful for reducing the crystallization time in hydrothermal time. Before these steps, pretreatment is needed to purify the raw material. From the journal articles also indicates that the types of zeolite synthesis and these properties are determined by operating conditions and starting material.

**Keywords:** coal fly ash, fusion, hydrothermal

## 1. INTRODUCTION

Coal is defined as natural fuel in the form of combustible sedimentary rock with ash yield being less than 50 % [1]. From 2011 to 2018, total coal consumption is relatively stable for the amount of 3,700 million tonnes of oil equivalent per year [2]. Among the fossil fuel resources, coal is still dominantly used as an energy source in the world for electricity production with a percentage of 38% in 2018 [3]. This is because coal is considered to be an affordable energy source and the most plentiful fossil fuel in the world [4].

Even though coal has considerable benefits in providing energy, burning coal in coal-fired power plants generates incombustible solid residues, such as coal ash. There are two types of coal ash, namely coal bottom ash and coal fly ash. Generally, coal bottom ash is ash left from the boiler and recovered from the bottom of the boiler, while coal fly ash is recovered at the top of the boiler using dust collectors [5]. The percentage of coal fly ash is higher than of coal bottom ash in the range of 80-85% [6]. 15-18.75 tons of coal is burned results in the generation of 1 megawatt of electricity and 4.3-11 tons of coal ash.

Around 850 million tons were produced in the world per year. However, only 52.6% of coal fly ash was utilized as raw material, mainly in the production of soil amendment and cement concrete [7]. Consequently, significant quantities of coal fly ash require more area for disposal

needed. Coal fly ash accumulated in the area of landfills causes environmental problems, such as air and groundwater pollution. Therefore, it becomes an important issue and needs methods to utilize fly ash into valuable products. One of the potential applications is used as a raw material of zeolite production due to similar composition with the precursor of natural zeolite like volcanic material. Fly ash contains high amounts of silica and alumina. The applications of zeolites are adsorbents, cation exchangers, as well as molecular sieves.

Zeolites are crystalline microporous aluminosilicates with  $[AlO_4]^{4-}$  and  $[SiO_4]^{4-}$  tetrahedrons [8]. Many researchers conducted a study about the conversion of coal fly ash to zeolite by alkaline fusion-hydrothermal method. The advantages of this method are improving the yield and quality of synthetic zeolites [9]. The main purpose of this article is to discuss the procedure of zeolite synthesis from coal fly ash by alkaline fusion-hydrothermal method and to know the effects of operating conditions on the types and properties of synthetic zeolites.

## 2. METHODOLOGY

A systematic literature review was a method used in this study. A systematic literature review is defined as a literature review that identifies, assesses, and interprets all findings on the research topics, then to answer the objectives of the study. In this study, past journal articles

have been reviewed to identify the good procedure for converting coal fly ash to zeolite by alkaline fusion-hydrothermal method. Furthermore, this literature review discussed with emphasis the effect of operating conditions on the type of zeolite and its properties. The journal articles reviewed are journal articles published in standard scientific journals.

### 3. RESULT AND DISCUSSION

#### 3.1. Physical and Chemical Properties of Coal Fly Ash

The physical characteristics of coal fly ash is dominantly spherical [10] and has smooth surfaces due to amorphous glass phase covers in part of the surface of coal fly ash [11]. Figure 1 showed the size of coal fly ash is varied. As stated by Ramme-Tharaniyil [12], the size of coal fly ash is about < 1 μm to 150 μm. Besides that, coal fly ash has specific surface area of 5 m<sup>2</sup>/g [13]. The color of coal fly ash depends on its composition. According to the American Coal Ash Association [14], high CaO content can be determined by light color of coal fly ash and high iron content can be determined by brownish color. The dark gray to black is associated with high unburnt carbon content.

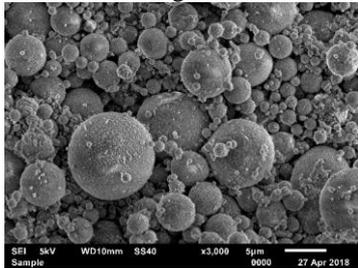


Figure 1 SEM Image of Coal Fly Ash [15]

From Figure 2, coal fly ash contains amorphous phases (silicate or aluminosilicate glass) and large crystalline phases in the form mullite (2SiO<sub>2</sub>.3Al<sub>2</sub>O<sub>3</sub>), quartz (SiO<sub>2</sub>), and small amounts of hematite and magnetite. The presence of amorphous phase is associated with the broad hump on diffraction patterns in the region of 15-35 degrees [16]. Quartz is indicated by the intense peak at 26.85 degrees and other crystalline phases are less intense peaks on the diffraction pattern [11].

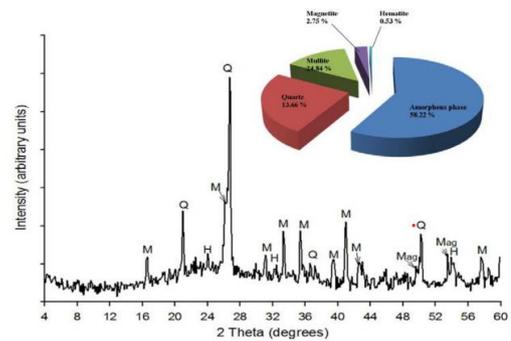


Figure 2 XRD Image of Coal Fly Ash [11]

Generally, the chemical composition of coal fly ash is varied because it depends on the types of coal (Table 1). Silica and aluminum oxides are the main component of coal fly ash, whereas the impurities consisted of metallic oxides such as Ca dan Fe. The trace elements include MgO, Na<sub>2</sub>O, K<sub>2</sub>O, and TiO<sub>2</sub>.

Table 1 Chemical Composition of Coal Fly Ash (%) [12]

Types of coal	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	Fe <sub>2</sub> O <sub>3</sub>	MgO
Lignite	15-45	10-25	15-40	4-15	3-10
Sub-bituminous	40-60	20-30	5-30	4-10	1-6
Bituminous	20-60	5-35	1-12	10-40	0-5
Types of coal	Na <sub>2</sub> O	K <sub>2</sub> O	SO <sub>3</sub>	LOI	
Lignite	0-6	0-4	0-10	0-5	
Sub-bituminous	0-2	0-4	0-2	0-3	
Bituminous	0-4	0-3	0-4	0-15	

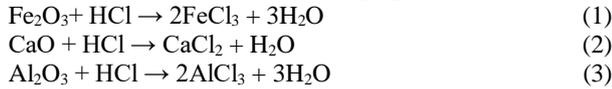
#### 3.2. Synthesis

##### 3.2.1. Pretreatment process of coal fly ash

Generally, coal fly ash consists of unburnt carbon, metallic oxides, alkaline oxides (CaO, TiO<sub>2</sub>, MgO, K<sub>2</sub>O, N<sub>2</sub>O), and others. Unburnt carbon, iron oxides, and alkaline oxides are undesirable materials for zeolite synthesis because they can interfere the process. So it needs pretreatment to reduce the composition of these materials. Unburnt carbon can reduce by high temperature of calcination process. Based on Liu et al. [17], the optimal condition of calcination process is 750 °C and 1.5 h, which resulted in a weight loss of 15.2%. The weight loss is relatively unchanged at calcination temperature > 750 °C.

Then, to reduce oxide impurities, coal fly ash is introduced to an acid solution named acid-washing pretreatment. Acid-washing pretreatment of coal fly ash has been studied by Panitchakarn et al. [18] and Kondru et al. [19] using three various acid solutions (HCl, H<sub>2</sub>SO<sub>4</sub>, HNO<sub>3</sub>). Based on

Panitchakarn et al. [20], acid-washing pretreatment using HCl solution at 20% w/w resulted in the zeolite with high purity (87% of SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and Na<sub>2</sub>O). This result is the same with experimental data of Kondru et al. [19] who reported that acid-washing pretreatment using HCl solution is most able to increase surface area and able to reduce iron and calcium oxides of coal fly ash. On the other hand, HCl solution causes dealumination of coal fly ash. These reactions are described as follows [20]:



### 3.2.2. Alkaline Fusion

Alkaline fusion is conducted by calcination process of the mixture of coal fly ash and alkaline material. Compared with KOH, sodium hydroxide (NaOH) is an effective material to extract silicon species of coal fly ash. Rios et al. [21] have conducted experiments that use KOH and NaOH as alkaline material in fusion. By using KOH showed a poor efficiency to activate coal fly ash indicated by the formation of an amorphous geopolymer as a product of zeolite synthesis. Whereas, using sodium hydroxide showed that most quartz and mullite were converted to soluble silicate and aluminate salts (Figure 3), eventually take part in the building of zeolite structure. According to Zhang et al. [22], fusion reactions are as follows.

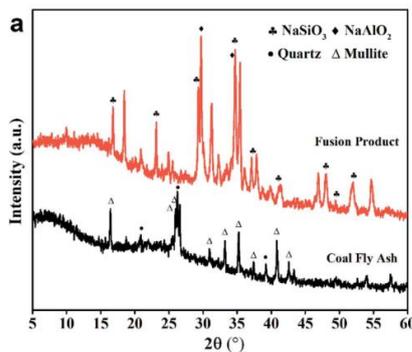
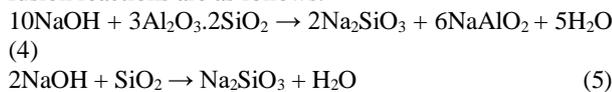


Figure 3 XRD of fused product (Ren et al., 2018)

Hu et al. [23] conducted a study to replace NaOH with low-cost alkali, Na<sub>2</sub>CO<sub>3</sub>. However, Na<sub>2</sub>CO<sub>3</sub> has low alkalinity and sluggish reaction at interface. To compensate these disadvantages, Hu et al. [23] did fusion or calcination at higher temperature (800 °C) and used tablet compression. By using tablet compression, coal fly ash and Na<sub>2</sub>CO<sub>3</sub> can contact effectively and result in most coal fly ash material could be converted to soluble materials. From this research, molar compositions of reactant to synthesize zeolite A are 1.3 Na<sub>2</sub>O: 0.6Al<sub>2</sub>O<sub>3</sub>:1SiO<sub>2</sub>:38H<sub>2</sub>O.

There are three alkaline fusion conditions that most influence zeolite formation. These factors include:

#### 1. NaOH/coal fly ash mass ratio

The amount of sodium hydroxide affects the composition of soluble silicate and aluminate in material as well as the alkalinity of the solution in the hydrothermal process. In the hydrothermal process, the presence of Na<sup>+</sup> ions contributes to the stabilization of sub-building unit of zeolite structure and fundamental in the crystallization process. Hamadi and Nabih [24] stated that the mixture of 5 grams of coal fly ash and 1 gram of NaOH resulted in reducing peak of mullite and quartz but zeolite was not formed. With the addition of 2-8 grams of NaOH, zeolite Na-PI is formed and crystallinity increase with increasing amounts of NaOH. The crystallinity of zeolite Na-P1 on the addition of NaOH 2, 4, and 8 grams were 20.1; 64.4; and 92.7. Fotovat et al. [25] stated that increasing NaOH/coal fly ash mass ratio from 1.2:1 to 2:1 resulted in a slightly increase of crystallinity from 84.7% to 88.6%. Ren et al. [26] observed that no peak of zeolite A was found at NaOH/coal fly ash mass ratio of 0.5:1. Zeolite A as main crystalline phase found at a mass ratio of 1.5:1. When increasing the mass ratio to 2:1, the peak of zeolite A decreased resulting in a low yield and CEC value. Molina and Poole [27] stated zeolite X can be produced at NaOH/coal fly ash mass ratio of 1.2:1. Further increasing the mass ratio, zeolite X transforms to hydroxy sodalite. Accordingly, a low amount of NaOH is not effective to convert coal fly ash to soluble silicate and aluminate salts as well as resulting in no zeolite due to low crystallization rate. In contrast, a high amount of NaOH produces more stable zeolite. Based on Ostwald's rule, the higher supersaturation because of higher soluble species in solution with the increase of NaOH is the better condition for synthesizing zeolite A or Na-X zeolite (metastable phase), which further recrystallize and are formed more stable zeolite (such as hydroxy sodalite) [28].

#### 2. Fusion temperature

Ruen-ngam et al. [29] have researched about the influence of fusion temperature variations on the percentage of zeolite X. The percentage of zeolite X was 40% and 67% at fusion temperature of 250 °C and 450 °C. Furthermore, the percentage of zeolite X increased slightly to 79% at 550 °C. Based on research by Kondru et al. [19], 550 °C was the maximum fusion temperature to obtain the maximum value of crystallinity and surface area. With further increase to 750 °C results in diminishes the crystallinity and surface area from 65.79% to 41.59% and 74.39 m<sup>2</sup>/g to 65.68 m<sup>2</sup>/g, respectively. Accordingly, the optimum fusion temperature is temperature above the melting point of NaOH (318 °C). However, fusion temperature above 550 °C decreases the crystallinity because non-crystalline sintered glass was formed [23, 30].

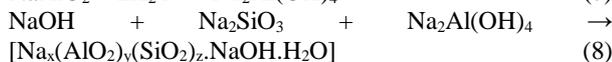
#### 3. Fusion time

Volli and Purkait [31] obtained that the mixture material of coal fly ash and sodium hydroxide was fused at 550 °C during 0.5 resulted in no peaks of the product appear. Then, the fusion time increase to 1 h resulted in zeolite X peaks

appears. Further increasing fusion time to 2 h, the peaks of zeolite X more intensified. It is clearly indicated that the minimum fusion time needed for the synthesis of zeolite X is 1 h.

### 3.2.3. Aging process

The aging process is the process to dissolve  $\text{Si}^{4+}$  and  $\text{Al}^{3+}$  and promote crystal nuclei. By aging process, it can reduce crystallization time [19]. The aging process is done at room temperature because the energy required is relatively low, which is 16 kJ/mol [32]. The aging process is carried out by dissolving and stirring the fused solids containing sodium silicate and sodium aluminosilicate with aqua DM. The reactions are explained as follows:

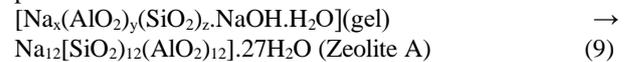


According to Kondru et al. [19], the stirred aging period affects Si/Al ratio of zeolite, crystallinity, and specific surface area. Increasing the stirred aging period from 10 h to 24 h resulted in Si/Al molar ratio of zeolite from 1.88 to 1.92. Further, increasing stirred aging period obtained Si/Al ratio of zeolite diminishes. On the other hand, the crystallinity and specific surface area increase with the increasing aging period. Based on Yao et al. [33], increasing of aging period until a certain time resulted in the increasing soluble species which then increases the supersaturation of starting material. This condition is better to nucleate metastable phases.

Some researchers added alumina source or silica source before or after aging process to control Si/Al molar ratio. Volli and Purkait [31] adjusted Si/Al molar ratio by adding  $\text{NaAlO}_2$  10-30 wt%. Based on this study, it was known that the addition of 10 wt% of  $\text{NaAlO}_2$  resulted in zeolite X with Si/Al molar ratio of 1.94. Next, increasing the amount of  $\text{NaAlO}_2$  added into a mixture of 20 wt% was obtained the formation of zeolite A with a Si/Al molar ratio of 1.67. Fotovat et al. [25] varied the amount of  $\text{NaAlO}_2$  powder added to the mixture in order to know the influence of different Si/Al molar ratios. From this research, it was obtained that Si/Al molar ratio in the mixture of lower than 2, between 2-2.4 and between 2.4-3 produced zeolite A, zeolite X, and zeolite Y, respectively. According to Ameh et al. [34], pure zeolite A can be formed at Si/Al molar ratio of mixture of < 1. By using sodium aluminate, it can provide enough amount of sodium ions into the mixture and further sodium ions stabilize the aluminosilicate during hydrothermal. Chareonpanich et al. [35] have discovered that ZSM-5 could be synthesized at  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of 20-40, crystal ZSM-5 can be formed. They adjusted the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio by adding sodium silicate which was extracted from rice husk ash.

### 3.2.4. Hydrothermal process

The hydrothermal process is carried out by heating aging products at a certain temperatures and time. In this step, the gel formed in aging process (nuclei) crystallizes to form well-structured material. Equation 9 describes the reaction of gel to form zeolite A that occur in the hydrothermal process is shown as follows:



There are three hydrothermal process conditions that most influence zeolite formation. These factors include:

#### 1. Hydrothermal temperature

Based on Hu et al. [23], synthesis zeolite at hydrothermal temperature of 60 °C and hydrothermal time of 8 hours showed wide and short peaks indicating low crystallinity and product purity. In the hydrothermal temperature between 60 and 100 °C, the pure zeolite Na-A phase is obtained. These findings are in agreement with the experimental data of Sivalingam and Sen [36]. They stated that low crystallization temperature leads to the formation of zeolite with low crystallinity percentage and crystal size. According to Zhang et al. [22], the synthesized zeolite with a high value of cation exchange capacity was obtained at a hydrothermal temperature of 90-100 °C. Whereas, at a hydrothermal temperature above 120 °C or under 90 °C resulted in the reduction of cation exchange capacity.

#### 2. Hydrothermal time

Chang and Shih [13] stated that higher hydrothermal temperatures and longer hydrothermal time generated the formation of dense and stable zeolite. Zeolite X (metastable zeolite) can transform into zeolite P at longer hydrothermal time. Liu et al. [37] and Xiao et al. [7] said, longer hydrothermal time can transform zeolite A to hydroxy sodalite phase.

#### 3. Liquid/solid ratio

Zhang et al. [22] obtained that the optimum liquid/solid ratio to get maximum CEC value is 5 ml/g. The low liquid/solid ratio caused high alkalinity of the solution in the hydrothermal process. Furthermore, it produced hydroxy sodalite with low CEC value [15, 38]. In contrast, a high liquid/solid ratio caused formation of amorphous phase or unidentified phase [39].

Many experiments conducted by researchers about the conversion of coal fly ash to zeolite by alkaline fusion-hydrothermal method and its parameters are summarized in Table 2.

**Table 2** Zeolite Synthesized by Alkaline Fusion-Hydrothermal Method

Parameter Studied	Parameter									Zeolitic phases and other synthetic products	Crystallinity (%)	CEC (meq/100 g)	Ref
	Molar ratio of starting material	Fusion				Aging		Hydrothermal					
		Alkaline source	Alkaline/Coal fly ash (g/g)	T (°C)	t (h)	L/S (ml/g)	t (h)	T (°C)	t (h)				
Alkaline source	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.1	NaOH	1.2:1	600	1	4.9	2.5	100	4	Cancrinite	*	*	[22]
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.1	KOH	1.2:1	600	1	4.9	2.5	100	4	No zeolite	*	*	
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =1.67	Na <sub>2</sub> CO <sub>3</sub>	2:1	800	2	*	1	80	6	Zeolite A	*	*	[24]
Mass ratio of NaOH and coal fly ash	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.44	NaOH	0.2:1	650	2	10	2	100	12	No Zeolite	-	*	[25]
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.44	NaOH	0.4:1	650	2	10	2	100	12	Na-PI	20.1	*	
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.44	NaOH	0.8:1	650	2	10	2	100	12	Na-PI	64.4	*	
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.44	NaOH	1.6:1	650	2	10	2	100	12	Na-PI	92.7	*	
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.4	NaOH	1.2:1	600	1.5	5	8	100	12	Faujasites	84.7	4.88	[26]
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.4	NaOH	2:1	600	1.5	5	8	100	12	Faujasites	88.6	4.18	
	Si/Al= 1	NaOH	0.5:1	650	2	5	12	85	6	No zeolite	*	*	[10]
	Si/Al= 1	NaOH	1.5:1	650	2	5	12	85	6	Zeolite A	*	*	
	Si/Al= 1	NaOH	2:1	650	2	5	12	85	6	The peaks of zeolite A decreased	*	*	
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =3.62	NaOH	1:1	650	2	5	12	85	6	Zeolite X	*	The maximum CEC value is obtained in zeolite synthesized with NaOH/coal fly ash ratio of 1.2:1	[28]
SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =3.62	NaOH	1.2:1	550	1	*	24	90	6	The peaks of zeolite X increased	*			
SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =3.62	NaOH	1.6:1	550	1	*	24	90	6	Zeolite X, Hydroxysodalite	*			
SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =3.62	NaOH	2:1	550	1	*	24	90	6	Hydroxysodalite	*			
Fusion temperature	Si/Al= 4.06	NaOH	2.25:1	250	1	*	12	90	2	% Zeolite X= 40%	*	206	[30]
	Si/Al= 4.06	NaOH	2.25:1	350	1	*	12	90	2	% Zeolite X= 45%	*	224	
	Si/Al= 4.06	NaOH	2.25:1	450	1	*	12	90	2	% Zeolite X= 67%	*	241	
	Si/Al= 4.06	NaOH	2.25:1	550	1	*	12	90	2	% Zeolite X= 79%	*	240	
	Si/Al= 1.51	NaOH	*	350	1	*	10	90-100	17	Zeolite Y	36.87	*	[20]
	Si/Al= 1.51	NaOH	*	550	1	*	10	90-100	17	Zeolite Y	65.79	*	
Fusion time	*	NaOH	*	550	0.5	10	*	*	*	No zeolite	*	With increasing of fusion time from 0.5 h to 2 h, CEC value increased from 82.6 to 192.3 meq/100 g	[32]
	*	NaOH	*	550	1	10	*	*	*	Zeolite X	*		
	*	NaOH	*	550	2	10	*	*	*	Zeolite X	*		
Aging period	Si/Al= 1.51	NaOH	*	550	1	*	10	90-100	17	*	The highest crystallinity is corresponding to aging period of 48 h	*	[20]
	Si/Al= 1.51	NaOH	*	550	1	*	24	90-100	17	*		*	
	Si/Al= 1.51	NaOH	*	550	1	*	48	90-100	17	*		*	

Si/Al molar ratio	Si/Al<2	NaOH	1.2:1	600	1.5	5	8	100	12	Zeolite A	*	*	[26]
	Si/Al= 2-2.4	NaOH	1.2:1	600	1.5	5	8	100	12	Zeolite X	*	*	
	Si/Al= 2.4-3	NaOH	1.2:1	600	1.5	5	8	100	12	Zeolite Y	*	*	
Hydrothermal temperature	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =1.67	Na <sub>2</sub> CO <sub>3</sub>	*	800	2	*	1	60	8	The peak of zeolite A are broad and short	*	*	[24]
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =1.67	Na <sub>2</sub> CO <sub>3</sub>	*	800	2	*	1	80	8	Zeolite A	*	*	
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =1.67	Na <sub>2</sub> CO <sub>3</sub>	*	800	2	*	1	100	8	Zeolite A	*	*	
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =1.67	Na <sub>2</sub> CO <sub>3</sub>	*	800	2	*	1	120	8	Zeolite A, hydroxysodalite	*	*	
Hydrothermal time	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.2	NaOH	0.5:1	650	2	2.7	1	90	3.5	Zeolite A as main product	*	The maximum CEC value is corresponding at hydrothermal time of 3.5 h	[8]
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.2	NaOH	0.5:1	650	2	2.7	1	90	4	Zeolite A transform into sodalite and philipsite	*		
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.2	NaOH	0.5:1	650	2	2.7	1	90	4.5		*		
	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =2.2	NaOH	0.5:1	650	2	2.7	1	90	5		*		
Liquid/solid ratio	Si/Al= 2	NaOH	1.2:1	550	1.5	3	12	90	15	*	*	The maximum CEC value is corresponding at liquid/solid ratio of 5	[23]
	Si/Al= 2	NaOH	1.2:1	550	1.5	5	12	90	15	*	*		
	Si/Al= 2	NaOH	1.2:1	550	1.5	10	12	90	15	*	*		
	Si/Al= 2	NaOH	1.2:1	550	1.5	15	12	90	15	*	*		

\* data is not available

#### 4. CONCLUSION

A million ton of coal fly ash is generated in the world. The higher amount of coal fly ash produced means the larger area for disposal needed. One of the potential utilization is the use of coal fly ash to synthesize zeolite. The literature review indicates that alkaline fusion-hydrothermal method is one of the methods successfully used to synthesize zeolite from coal fly ash. The steps of alkaline fusion-hydrothermal method include the pretreatment process, alkaline fusion process, aging process, and hydrothermal process. Aging process is beneficial to reduce crystallization time in hydrothermal process. Operating conditions and starting material are impact to type of zeolite synthesis and its properties. Most journal articles that have been reviewed indicate that the type of zeolites that can be synthesized using coal fly ash as raw material is zeolites with a low Si/Al molar ratio.

#### ACKNOWLEDGMENT

The authors would like to thank Unit Penelitian dan Pengabdian Masyarakat Politeknik Negeri Bandung for financial support.

#### REFERENCES

- [1] X. Wang, Y. Tang, S. Wang, and H.H. Schobert, Clean Coal Geology in China: Research advance and its future. *International Journal of Coal Science & Technology*, vol. 7 (1), 2020, pp. 299–310. DOI: <https://doi.org/10.1007/s40789-020-00321-4>
- [2] British Petroleum, 2019, BP Statistical Review of World Energy. Retrieved from British Petroleum: <https://www.bp.com/content/dam/bp/business-sites/en/global/corporate/pdfs/energy-economics/statistical-review/bp-stats-review-2019-full-report.pdf>
- [3] T. Kaberger, Progress of renewable electricity replacing fossil fuels, *Global Energy Interconnection*, vol. 1(1), 2018, pp. 48-52.
- [4] C. Zou, Q. Zhao, G. Zhang, and B. Xiong, Energy revolution: From a fossil energy era to new energy era, *Natural Gas Industry B*, vol. 3(1), 2016, pp. 1-11. DOI: <https://doi.org/10.1016/j.ngib.2016.02.001>

- [5] H. Tanaka, and A. Fujii, Effect of Stirring on the Dissolution of Coal Fly Ash and Synthesis of Pure-Form Na-A and -X Zeolites by Two-Step Process, *Advanced Powder Technology*, 2009, vol. 20, 2009, pp. 473–479.
- [6] J.D.C. Izidoro, D.A. Fungaro, J.E. Abbott, and S. Wang, Synthesis of Zeolites X and A from Fly Ashes for Cadmium dan Zinc Removal from Aqueous Solutions in Single dan Binary Ion Systems, *Fuel*, vol. 103, 2013, pp. 827–834. DOI: <https://doi.org/10.1016/j.fuel.2012.07.060>
- [7] M. Xiao, X. Hu, Y. Gong, D. Gao, P. Zhang, Q. Liu, Y. Liu, and M. Wang, Solid Transformation Synthesis of Zeolite From Fly Ash, *The Royal Society of Chemistry*, 2015, pp. 1-7. DOI: <https://doi.org/10.1039/C5RA17856H>
- [8] A. Kongnoo, S. Tontisirin, P. Worathanakul, and C. Phalakornkule, Surface Characteristics and CO<sub>2</sub> Adsorption Capacities of Acid-Activated Zeolite 13X Prepared from Palm Oil Mill Fly Ash, *Fuel*, vol. 193, 2017, pp. 385-394. DOI: <https://doi.org/10.1016/j.fuel.2016.12.087>
- [9] X. Ren, R. Qu, S. Liu, H. Zhao, W. Wu, H. Song, C. Zheng, X. Wu, and Z. Gao, Synthesis of Zeolites from Coal Fly Ash for the Removal of Harmful Gaseous Pollutants : A review, *Aerosol and Air Quality Research*, vol. 20, 2020. DOI: <https://doi.org/10.4209/aaqr.2019.12.0651>
- [10] Z.T. Yao, X.S. Ji, P.K. Sarker, J.H. Tang, L.Q. Ge, M.S. Xia, Y.Q. Xi, A Comprehensive Review on The Applications of Coal Fly Ash, *Earth-Science Reviews*, vol. 141, 2015, pp. 105-121. DOI: <https://doi.org/10.1016/j.earscirev.2014.11.016>
- [11] D. Mainganye, T.V. Ojumu, and L. Petrik, Synthesis of Zeolites Na-P1 from South African Coal Fly Ash: Effect of Impeller Design and Agitation, *Materials*, vol. 6(5), 2013, pp. 2074-2089. DOI: 10.3390/ma6052074
- [12] R. Tharaniyil, *Coal Combustion Products Utilization Handbook: Third Edition*, We Energies, USA, 2013.
- [13] H. Chang, and W. Shih, Synthesis of Zeolites A and X from Fly Ashes and Their Ion-Exchange Behavior with Cobalt Ions, *Ind. Eng. Chem. Res.*, vol. 39, 2000, pp. 4185-4191. DOI: <https://doi.org/10.1021/ie990860s>
- [14] American Coal Association, 2003, Fly Ash Facts for Highway Engineers. Retrieved from: <https://www.fhwa.dot.gov/pavement/recycling/fafacts.pdf>
- [15] W. Wulandari, T. Paramitha, J. Rizkiana, and D. Sasongko, Characterization of Zeolite A from Coal Fly Ash Via Fusion-Hydrothermal Synthesis Method, *IOP Conference Series: Materials Science and Engineering*, vol. 543, 2019. DOI: 10.1088/1757-899X/543/1/012034
- [16] O.B. Kotova, I.N. Shabalin, D.A. Shushkov, and L.S. Kocheva, Hydrothermal Synthesis of Zeolites From Coal Fly Ash, *Advances in Applied Ceramics*, vol. 115, pp. 152-157. DOI: <https://doi.org/10.1179/1743676115Y.0000000063>
- [17] Y. Liu, Q. Luo, G. Wang, X. Li, and P. Na, Synthesis and characterization of zeolite from coal fly ash. *Materials Research Express*, vol. 5(5), 2018, 055507. DOI:10.1088/2053-1591/aac3ae
- [18] P. Panitchakarn, N. Laosiripojana, N. Viriyumpikul, and P. Pavasant, Synthesis of high-purity Na-A and Na-X zeolite from coal fly ash, *Journal of the Air & Waste Management Association*, vol. 64(5), 2014, pp. 586-596 DOI: 10.1080/10962247.2013.859184
- [19] A.K. Kondru, P. Kumar, P., T. T. Teng, S. Chand, and K. Wasewar, Synthesis and Characterization of Na-Y Zeolite from Coal Fly Ash dan Its Effectiveness in Removal of Dye from Aqueous Solution by Wet Peroxide Oxidation, *Arch. Environ. Sci.*, vol. 5, 2011, pp. 46–54.
- [20] R.Y. Kurniawan and N. Widiastuti, Sintesis Zeolit-A dari Abu Dasar Batubara dengan Pemisahan Fe dan Ca, *Jurnal Teknik ITS*, vol. 6, 2017, pp. C 17- C 20.
- [21] C.A. Rios R., C.D. Williams, and C.L. Roberts, A Comparative Study of Two Methods for The Synthesis of Fly Ash-Based Sodium and Potassium Type Zeolites, *Fuel*, vol. 88, 2009, pp. 1403-1416. DOI: <https://doi.org/10.1016/j.fuel.2009.02.012>
- [22] Y. Zhang, J. Dong, F. Guo, Z. Shao, and J. Wu, Zeolite Synthesized from Coal Fly Ash Produced by a Gasification Process for Ni<sup>2+</sup> Removal from Water, *Minerals*, vol. 8 (3), 2018. DOI: <https://doi.org/10.3390/min8030116>
- [23] T. Hu, W. Gao, X. Liu, Y. Zhang, and C. Meng, Synthesis of Zeolites Na-a and Na-X from Tablet Compressed and Calcinated Coal Fly Ash, *Royal Society of Chemistry*, vol. 4, 2017, pp. 1-11. DOI: <https://doi.org/10.1098/rsos.170921>
- [24] A. Hamadi and K. Nabih, Synthesis of zeolites materials using fly ash and oil shale ash and their applications in removing heavy metals from aqueous solutions, *Journal of Chemistry*, 2018. DOI: <https://doi.org/10.1155/2018/6207910>

- [25] F. Fotovat, H. Kazemian, and M. Kazemeini, Synthesis of Na-A and Faujasitic Zeolites from High Silicon Fly Ash, *Materials Research Bulletin*, vol. 44, 2009, pp. 913–917. DOI: <https://doi.org/10.1016/j.materresbull.2008.08.008>
- [26] X. Ren, L. Xiao, R. Qu, S. Liu, D. Ye, H. Song, W. Wu, C. Zheng, X. Wu, and X. Gao, Synthesis and Characterization of Single Phase Zeolite A Using Coal Fly Ash, *Royal Society of Chemistry*, vol. 8, 2018, pp. 42200-42209. DOI: <https://doi.org/10.1039/C8RA09215J>
- [27] A. Molina and C. Poole, A Comparative Study Using Two Methods to Produce Zeolites from Fly Ash, *Mineral Engineering*, vol. 17, 2004, pp. 167-173. DOI: [10.1016/j.mineng.2003.10.025](https://doi.org/10.1016/j.mineng.2003.10.025)
- [28] R. Barrer, *Hydrothermal Chemistry of Zeolites: Chapter 5*, Academic Press, England. 1982.
- [29] D. Ruen-ngam, D. Tempuhangsuk, and R. Apiratikul, Zeolite Formation from Coal Fly Ash and Its Adsorption Potential, *Journal of the Air & Waste Management Association*, vol. 59, 2012, pp. 1140–1147. DOI: <https://doi.org/10.3155/1047-3289.59.10.1140>
- [30] N. Shigemoto, H. Hayashi, and K. Miyaura, Selective Formation of Na-X Zeolite from Coal Fly Ash by Fusion with Sodium Hydroxide prior to Hydrothermal Reaction, *Journal of Materials Science*, vol. 28, 1993, pp. 4781–4786. DOI: <https://doi.org/10.1007/BF00414272>
- [31] V. Volli and M.K. Purkait, Selective Preparation of Zeolite X and A from Flyash and Its Use as Catalyst for Biodiesel Production, *Journal of Hazardous Materials*, vol. 297, 2015, pp. 101-111. DOI: <https://doi.org/10.1016/j.jhazmat.2015.04.066>
- [32] T. Brar, P. France, and P.G. Smirniotis, Control of Crystal Size and Distribution of Zeolite A, *Ind. Eng. Res.*, vol. 40, 2001, pp. 1133-1139. DOI: <https://doi.org/10.1021/ie000748q>
- [33] Z.T. Yao, M.S. Xia, Y. Ye, and L. Zhang, Synthesis of zeolite Li-ABW from fly ash by fusion method, *Journal of Hazardous Materials*, vol. 170, 2009, pp. 639-644. DOI: <https://doi.org/10.1016/j.jhazmat.2009.05.018>
- [34] A.E. Ameh, O.O. Fatoba, N.M. Musyoka, and L.F. Petrik, Influence of aluminium source on the crystal structure and framework coordination of Al and Si in fly ash-based zeolite NaA, *Powder Technology*, vol. 306, 2017, pp. 17-25. DOI: <https://doi.org/10.1016/j.powtec.2016.11.003>
- [35] M. Chareonpanich, T. Namto, P. Kongkachuichay, and J. Limtrakul, Synthesis of ZSM-5 zeolite from lignite fly ash and rice husk ash, *Fuel Processing Technology*, vol. 85, 2004, pp. 1623-1634. DOI: [10.1016/J.FUPROC.2003.10.026](https://doi.org/10.1016/J.FUPROC.2003.10.026)
- [36] S. Sivalingam and S. Sen, Optimization of synthesis parameters and characterization of coal fly ash derived microporous zeolite X, *Applied Surface Science*, vol. 455, 2018, pp. 903-910. DOI: <https://doi.org/10.1016/j.apsusc.2018.05.222>
- [37] X.D. Liu, Y.P. Wang, X.M. Cui, Y. He, and J. Mao, Influence of synthesis parameters on NaA zeolite crystals, *Powder Technology*, vol. 243, 2013, pp. 184–193. DOI: [10.1016/J.POWTEC.2013.03.048](https://doi.org/10.1016/J.POWTEC.2013.03.048)
- [38] N. Murayama, H. Yamamoto, and J. Shibata, Mechanism of Zeolite Synthesis from Coal Fly Ash by Alkali Hydrothermal Reaction, *International Journal Mineral Processing*, vol. 64, 2002, pp. 1-17. DOI: [https://doi.org/10.1016/S0301-7516\(01\)00046-1](https://doi.org/10.1016/S0301-7516(01)00046-1)
- [39] A. Rujiwatra, M. Phueadpho, and K. Grudpan, Selective Synthesis of Zeolitic Philipsite and Hibschite Hydrogarnet from Lignite Ash Employing Calcium Hydroxide Under Mild Conditions, *Journal of Physics and Chemistry of Solids*, vol. 66, 2005, pp. 1085-1090. DOI: <https://doi.org/10.1016/j.jpcs.2005.02.004>