Synthesis of Zeolites from Coal Fly Ash for the Removal of Harmful Gaseous Pollutants: A Review

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ABSTRACT

SO_x, NO_x, CO_x, volatile organic compounds (VOCs), and Hg vapor directly and indirectly harm the atmospheric environment and human health by contributing to the formation of photochemical smog, acid rain, and haze and posing risks of potential toxicity (e.g., carcinogenicity). Therefore, effectively controlling and reducing the pollution caused by these chemicals is critical, and to that end, several methods have been developed, among which adsorption is one of the most common and effective techniques. As adsorption materials, zeolites show great potential in reducing air pollution. Moreover, coal fly ash (CFA)-based zeolite synthesis enables the simultaneous treatment of air and solid waste pollution. In this study, the progress of recent years in research on zeolite synthesis by CFA is reviewed, and the challenges besetting this method are discussed. In addition, we examine the application of CFA-based zeolites in removing or minimizing harmful gases. As we enter an era of utilizing disposed waste, developing efficient and low-cost materials for the removal of harmful gases may require improving the synthesis of high-purity, high-performance fly ash zeolite materials.

Keywords: Coal fly ash; Zeolite; Synthesis; Gaseous pollutant; Removal.

INTRODUCTION

Coal is the second largest fuel in the global energy mix, and it accounted for 27% of the global energy consumption in 2018 (Fig. 1(a)) (BP, 2019). China remains to be the world's largest energy consumer, and it contributed 34% to the growth of the global energy demand in 2018 (Fig. 1(b)) (BP, 2019). In addition, coal is still China's dominant fuel. Its share of total energy consumption was 58% in 2018 (Fig. 2(a)). Fig. 2(b) shows the trend of coal consumption, which increased slightly in 2018 compared with the value in 2017 (BP, 2019).

Coal fly ash (CFA), a by-product of coal combustion, is one of the most abundant industrial solid wastes, and the emission of CFA increases annually. CFA contains heavy metals, polycyclic aromatic hydrocarbons, silica, and other toxic substances (Tanaka *et al.*, 2008b; Pandey *et al.*, 2011; Bartoňová, 2015). High CFA emissions can take over land resources, pollute the air, and damage human health (Delkash *et al.*, 2015). Therefore, proper disposal of CFA is urgently required. At present, CFA is mainly used as building materials in the form of concrete (Dilmore and Neufeld, 2001), cement

(Bouzoubaa and Lachemi, 2001), and hollow blocks (Kumar, 2003) (Fig. 3). This material is also utilized in backfilling treatments (Yao *et al.*, 2015). However, these applications have low utilization and low added value. Therefore, utilizing the advantages of CFA and transforming this material into a high-value-added product have become research hotspots. The main components of CFA are Si- and Al-containing compounds, which are similar to zeolite. Hence, CFA is conducive to the synthesis of zeolite (Cardoso *et al.*, 2015).

Zeolite has broad application prospects due to its potential molecular sieving, high specific surface area, and good thermal and chemical stability (Wang *et al.*, 2010; Jiang *et al.*, 2011; Charkhi *et al.*, 2012). Zeolite is a kind of aluminosilicate molecular sieve crystal with uniform pores, and its skeleton contains Al, Si, and O (Andaç *et al.*, 2005). Cations and water exist in the pores (Vuong and Do, 2009). As depicted in Fig. 4, tetrahedral TO₄ (T = Si, Al) is the most basic structural unit of the zeolite framework, and interconnected tetrahedral TO₄ requires additional cations to balance the negative charge generated by Al ions (Belviso, 2018).

The polyhedral rings that are formed by sharing O atoms and connected in different ways are called *secondary building unit* (SBU). Several common SBU are illustrated in Fig. 5. *Composite building unit* (CBU) is formed by the connection of SBU, and they form different kinds of zeolite skeleton structures through additional connections (Fig. 6) (Smith, 1988). Several common zeolite structures are illustrated in

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Fig. 1. (a) Shares of worldwide primary energy consumption and (b) contributions from different regions to primary energy growth in 2018.

Fig. 2. (a) Shares of China primary energy consumption and (b) China's coal consumption in 2010–2018.

Fig. 3. Specific utilization of coal fly ash.

Fig. 4. Aluminum replaces silicon in the tetrahedron to create a negative charge in the zeolite tetrahedral skeleton (adapted from Belviso, 2018).

Fig. 5. Secondary building units and their symbols in framework structure of zeolite (adapted from Smith, 1988).

Fig. 7, and Table 1 presents their framework structures and related materials. Different zeolite structures result in distinct channels (Fig. 8). Benefitting from these structural features, zeolites have the characteristics of adsorption selectivity, high specific surface, and high ion exchange capacity (Hasan *et al.*, 2012; Mousavi *et al.*, 2013). Consequently, zeolites have enormous potential for adsorption and catalytic applications.

Sustained and rapid economic development and industrialization would inevitably increase the consumption of energy and other materials and could cause a series of environmental pollution problems, such as pollution by SO_x (Wang *et al.*, 2018a; Carter *et al.*, 2018; Yang *et al.*, 2019b), NO_x (Gaskin *et al.*, 2019), and volatile organic compounds (VOCs) (Aziz and Kim, 2017; Difallah *et al.*, 2017; Wang *et al.*, 2018b). Atmospheric aerosol is one of the main pollutants that affect the urban environment and human health, and it directly or indirectly causes haze, photochemical smog, acid

rain, and other climate changes (Li *et al.*, 2016). This pollutant contains many toxic and harmful substances and serves as a carrier of other pollutants, which cause great harm on human health and the ecological environment. Depending on the formation mechanism, atmospheric aerosols can be divided into primary and secondary aerosols (Seinfeld and Pankow, 2003). Primary aerosols refer to aerosol particles that are directly discharged into the atmosphere, and secondary aerosols are particles produced by the oxidation of certain gaseous pollutants (e.g., SO_x , NO_x , and hydrocarbons) in the atmosphere (e.g., from SO_2 to SO_4^{2-}) (Pöschl, 2005).

Organic aerosols, most of which belong to secondary aerosols, account for 20–90% of the aerosol mass concentration in terms of $\text{PM}_{2.5}$ (particle size ≤ 2.5 nm) (Riipinen *et al.*, 2012). VOCs are important precursors for the formation of organic aerosols (Guenther *et al.*, 2006). Therefore, reducing and controlling VOC emission are necessary.

Fig. 6. Some common composite building units in framework structure of zeolite (adapted from Smith, 1988).

Fig. 7. Several common zeolite framework structures: (a) LTA (Yordanov *et al.*, 2014), (b) FAU (Lim *et al.*, 2009), (c) SOD (Leung *et al.*, 2015), (d) GIS (Kunecki *et al.*, 2018), (e) EDI (Harrison, 2001), (f) CHA (Smith *et al.*, 2000).

Zeolites have been widely used for air pollution control due to their excellent performance. In recent years, the synthesis of zeolites with considerable properties (e.g., wide availability and low cost) from CFA has elicited much interest from researchers. Bukhari *et al.* (2015) introduced the progress of research on and the application of converting CFA into zeolites through microwave and ultrasonic energies. Mallapur and Oubagaranadin (2017) reviewed the progress of research on synthetic zeolites from fly ash,

municipal solid waste, agricultural waste, industrial kaolin waste, and other hazardous inorganic waste. Belviso (2018) comprehensively analyzed the characteristics and application prospects of coal ash and biomass ash and discussed the key factors affecting the synthesis of zeolites from coal ash and the shortcomings of various synthetic methods. The present work reviews the synthesis methods of various zeolites from CFA and highlights the application progress of CFA-based zeolites in gas pollution control.

Table 1. Several common zeolite framework structures and their related materials (from Structure Commission of the International Zeolite Association).

Type	Cell shape	Channel	Unit cell dimension			CBU	Related materials	Ref.
			a (Å)	b (Å)	c (Å)			
LTA	Cubic	3-D	4.21	4.21	4.21	d4r, sod, lta	Zeolite A, SAPO-42, UZM-9, ITQ-29	(Lok <i>et al.</i> , 1984; Corma <i>et al.</i> , 2004; Lewis <i>et al.</i> , 2004; Bouizi <i>et al.</i> , 2007)
FAU	Cubic	3-D	7.35	7.35	7.35	d6r, sod	NaX, NaY, ZSM-20, [Ga-Si-O]-FAU	(Newsam <i>et al.</i> , 1989; Occelli <i>et al.</i> , 2000; Greiser <i>et al.</i> , 2016; Schroeder <i>et al.</i> , 2018)
GIS	Tetragonal	3-D	3.32	3.32	3.32	gis	NaP1, [Be-P-O]-GIS, SAPO-43, garronite	(Baerlocher and Meier, 1972; Helliwell <i>et al.</i> , 1993; Artioli and Marchi, 1999; Zhang <i>et al.</i> , 2001)
SOD	Cubic	3-D	2.53	2.53	2.53	sod	Sodalite, AlPO-20, SIZ-9	(Löns and Schulz, 1967; Wilson <i>et al.</i> , 1982; Parnham and Morris, 2006)
EDI	Tetragonal	3-D	3.2	3.2	3.44	nat	[Zn-As-O]-EDI, K-F, Zeolite N, Linde F	(Tambuyzer and Bosmans, 1976; Sherman, 1977; Zeolite, 1997; Feng <i>et al.</i> , 2001)
CHA	Trigonal	3-D	3.72	3.72	3.72	d6r, cha	DAF-5, chabazite, UiO-21, SSZ-62	(Sankar <i>et al.</i> , 1998; Kongshaug <i>et al.</i> , 2000; Zhang <i>et al.</i> , 2008; Itakura <i>et al.</i> , 2011)

Fig. 8. Channel structure of (1) MCM-41 (Jabbari *et al.*, 2015), (2) ZSM-5 (Flanigen *et al.*, 1978), (3) faujasite (Johnson *et al.*, 1999).

SYNTHESIS OF ZEOLITES FROM CFA

The research on and understanding of the crystallization mechanism of zeolites provide theoretical guiding significance for the synthesis of zeolites with specific structures and functions and have far-reaching importance for the process and technology of industrial production. Throughout the development history of zeolites, the discussion of their synthesis mechanism has consistently been a controversial topic. The formation of zeolites is a complicated process that involves the transformation of silicates and aluminates, the formation and dissolution of aluminosilicate, the continuous change in the gel's solid and solution phases, and the formation and growth of zeolite nucleation. On the basis of the changes in solid and liquid phases during synthesis, solid-phase, liquid-phase, and bidirectional transformation mechanisms

have been proposed (Cundy and Cox, 2003). With the incessant progress of experimental and characterization methods, the understanding of the crystallization mechanism of zeolites has been constantly improved.

The principle of the solid-phase transformation mechanism is that no solid-phase dissolution should exist in the crystallization process, nor should the liquid phase be directly involved in the nucleation and crystal growth of zeolites. Only the structural rearrangement of the aluminosilicate skeleton generated by the gel solid under certain conditions is involved, resulting in the nucleation and crystal growth of zeolites (Tsapatsis *et al.*, 1996). Breck (1964) investigated the relationship between zeolite crystallization and time through X-ray diffraction. The crystallization process was divided into two stages, namely, induction and growth. The solid-phase transformation mechanism was comprehensively

examined via electron microscope analysis. Guth and Caullet (1986) verified that in extreme cases where only a large number of amorphous gels is present, the liquid phase does not participate in the crystallization process. Instead, the gel is directly rearranged in situ to synthesize zeolites.

Kerr (1966) proposed the liquid-phase transformation mechanism of zeolite crystallization. They found that the gel was initially dissolved in NaOH solution and produced dissolving derivatives, which in turn yielded the zeolite in the solution. The liquid-phase transformation mechanism also considers an equilibrium state between the solid and liquid phases. The growth of the crystal nucleus consumes the silicate and aluminate ions in the solution, and the consumption of the liquid-phase components in the crystallization process leads to the continued dissolution of the solid phase (Grizzetti and Artioli, 2002).

Improvements in characterization methods gave way to the development of the bidirectional transformation mechanism. Gabelica *et al.* (1984) studied the crystallization process of ZSM-5 through a dual system and found that the crystallization process of one system has many similarities with the liquid-phase mechanism, whereas the other high-silicon system belongs to the solid-phase mechanism. Using the small-angle neutron scattering technique, Iton *et al.* (1992) confirmed that the crystallization mechanism of ZSM-5 varies with different silicon sources. Van Grieken *et al.* (2000) reported that the crystallization process of ZSM-5 zeolite is due to the simultaneous existence of solid- and liquid-phase mechanisms.

Theoretically, given that the main components of CFA are Si and Al, zeolite can be synthesized from CFA. Many domestic and foreign scholars have conducted studies on this field (Yaping *et al.*, 2008). The synthesis methods for zeolite include direct hydrothermal, two-step hydrothermal,

alkaline fusion-assisted hydrothermal, and microwave-assisted methods. This section focuses on the analysis of the process and parameters of each method for the formation of zeolite types from CFA and the advantages and limitations of each approach. Fig. 9 displays the preparation process of zeolites with CFA as the raw material, and Table 2 shows information on the preparation methods and their products.

Direct Hydrothermal Method

The direct hydrothermal method involves dissolving CFA in an alkaline solution and subsequently synthesizing zeolite products through thermal treatment. Querol *et al.* (1999) investigated the influence of 14 kinds of Spanish CFA with different physical and chemical properties on zeolite synthesis. The results showed that CFA with the highest content of aluminum-silicate glass has the greatest potential for industrial application. The researchers also noted that in the process of synthesizing zeolites from CFA, the main active substances are glass and quartz, whereas mullite has low activity (Querol *et al.*, 1995). Izidoro *et al.* (2012) discussed the synthesis of zeolites from five kinds of Brazilian CFA using the hydrothermal method and discovered that zeolite made from CFA containing more than 70% SiO_2 and Al_2O_3 has a high ion exchange capacity. The results also suggest that the element composition of CFA is one of the important factors that influence the synthesis of zeolites. Inada *et al.* (2005a) analyzed the effect of the Si/Al molar ratio and alkaline condition on the type of synthetic zeolite. The authors showed that it is beneficial to the formation of Zeolite P with silica-rich CFA at a low alkali concentration, whereas the addition of a Si source can promote the formation of Zeolite P at a high alkali concentration. This finding indicates that the addition of Si or Al sources can control the type of

Fig. 9. Several typical flowcharts of preparation of zeolites from coal fly ash.

Table 2. Main methods for synthesis of zeolites from coal fly ash.

Method	Reagents	Preparation condition	Zeolites	Remarks	Ref.
Direct hydrothermal	NaOH, NaAlO ₂	Heating: 85°C, 72 h	NaA, NaX	Simple operation, low cost, long time, low purity, low yield	(Tanaka et al., 2002)
	NaOH	Heating: 150°C, 48 h	NaP1, sodalite, analcime, gmelinite		(Querol et al., 1997b)
Two-step hydrothermal	NaOH, KOH	Heating: 150°C, 336 h	NaP1, nosean	High purity, complex operation, low yield	(Querol et al., 1995)
	NaOH	Heating: 100°C, 24 h	analcime, merlinoite		(Izidoro, 2012)
	NaOH, SiO ₂ , Al ₂ O ₃	Heating: 100°C, 24 h	NaX, NaP1, sodalite, gmelinite		(Inada, 2005a)
	NaOH	Aging: 150°C, 4 h; Heating: 80°C, 96 h	NaP1, sodalite		(Watek, 2008)
	NaOH	Aging: 24 h; Heating: 117°C, 48 h	NaP1		(Tauanov et al., 2018)
Alkaline fusion-assisted hydrothermal	NaOH, Na ₂ SiO ₃ , 9H ₂ O	Heating: 60°C, 28 d	Zeolite	High yield, high cost, high energy	(De Rossi, 2019)
	NaOH, NaAlO ₂	Aging: 90°C, 6 h; Heating filtrate: 90°C, 48 h; Heating residue: 90°C, 24 h	NaP1		(Hollman, 1999)
	NaOH, NaAlO ₂	Aging: 80°C, 2 h; Heating filtrate: 100°C, 190 min; Heating residue: 100°C, 370 min	NaP1, NaX		(Wang et al., 2008)
	NaOH	Aging: 47°C, 48 h; Heating filtrate: 140°C, 48 h	NaA		(Musyoka, 2012)
	NaOH	CFA:NaOH = 1:1.2; Calcination: 500°C, 1 h; Aging: 12 h; Heating: 100°C, 6 h	NaP		(Shigemoto, 1993)
Microwave assisted	NaOH	CFA:NaOH = 1:1.2; Calcination: 600°C, 1 h; Aging: 8 h; Heating: 100°C, 10 h	NaY, mullite	Short time, low yield, low purity	(Rayalu et al., 2000)
	NaOH, CTAB, NH ₄ OH	CFA:NaOH = 1:1.2; Calcination: 550°C, 1 h; Aging: 24 h; Heating filtrate: 97°C, 98 h	NaY		(Kumar et al., 2001)
	NaOH	CFA:NaOH = 1:1.2; Calcination: 550°C, 1 h; Aging: 24 h; Heating: 120°C, 4 h	MCM-41		(Kazemian et al., 2010)
	LiOH·H ₂ O	CFA + 25% LiOH·H ₂ O; Calcination: 980°C, 1 h; Heating: 180°C, 12 h	NaP		(Yao et al., 2009)
	NaOH, Na ₂ CO ₃	NaOH:Na ₂ CO ₃ = 1:2.8; Calcination: 760°C; Aging: 4 h; Heating: 90°C, 4 h	Li-ABW		(Yang et al., 2019a)
Microwave assisted	NaOH	CFA:NaOH = 1:1.2; Calcination: 500°C, 1 h; Aging: 25°C, 16 h; Heating: 100°C, 10 h	NaA	Short time, low yield, low purity	(Sivalingam, 2018)
	NaOH	CFA:NaOH = 1:1; Calcination: 600°C, 2 h; Heating: 80°C, 24 h	NaX		(Feng et al., 2018)
	NaOH, NaAlO ₂	Aging: 85°C, 24 h; Microwave: 600 W, 1 h	NaA		(Tanaka et al., 2008a)
	NaOH, NaAlO ₂	Aging: 60°C, 12 h; Microwave: 300 W, 0.5 h	NaA, quartz		(Behin, 2015)
	NaOH	Microwave: 100°C, 2 h	Phillipsite		(Fukui et al., 2006)
Microwave assisted	NaOH, NaAlO ₂	Microwave: 450°C, 0.25 h; Heating: 90°C, 12 h	Faujasite	Short time, low yield, low purity	(Li et al., 2018)
	NaOH	Aging: 90°C, 6 h; Microwave (filtrate): 250 W, 105°C, 30 min	NaP, quartz, calcite, mullite		(Aldahri et al., 2017)
	NaOH	Microwave (filtrate): 250 W, 30 min	NaP, LTA		(Al-Dahri et al., 2018)

synthetic zeolites. Walek *et al.* (2008) studied the influence of different solid/liquid (S/L) ratios on the synthesis of zeolites from CFA. They discovered that a low S/L ratio is favorable to the dissolution of CFA. A single-phase NaP1 zeolite was obtained at conditions of 2 M NaOH solution, 104°C solution temperature, and 80°C crystallization temperature. The relationship between the synthesis conditions and the products was clearly described at a low S/L ratio. The S/L ratio usually used is 50–150 g dm⁻³, which greatly limits the dissolution of CFA. But the low S/L ratio (4 g dm⁻³) reached 85 wt% in 4 hours. Murayama *et al.* (2002) described the effect of alkali on the mechanism for the synthesis of fly ash-based zeolites. They discovered that the OH⁻ in the alkaline solution greatly promotes the dissolution of Si and Al ions, and Na ions promote the formation of Zeolite P, which can selectively capture K⁺. Using biomass fly ash as the raw material, De Rossi *et al.* (2019) synthesized Zeolite P with a specific surface area of 56.35 m² g⁻¹ through an in situ method at 60°C.

The direct hydrothermal method has a simple operation and low production cost. However, the method still has several disadvantages, including long hydrothermal time, high temperature, high energy consumption, low product purity, low yield, and many by-products.

Two-step Hydrothermal Method

Hollman *et al.* (1999) were the first to extend the one-step method to two-step synthesis of pure zeolites from CFA. The Si in CFA can be extracted by adding NaOH solution to synthesize pure zeolite (> 99%). In addition, the solid residue from the reaction can be converted into zeolite via the traditional method. The cationic exchange capacity of pure zeolite is approximately two times that of the blend zeolite/residual CFA, which renders the former more suitable for wastewater treatment than the latter. Moriyama *et al.* (2005) described the effect of high-temperature processing and drainage during operation on the synthesis of zeolites from CFA. They found that the appropriate synthesis condition involves 2.5–3.5 mol L⁻¹ of NaOH and 0.88–1.10 L kg⁻¹ of liquid/solid. The zeolite obtained by a pilot plant has a higher cation exchange capacity than that produced by the traditional method. Wang *et al.* (2008) discussed the effects of NaOH concentration on the dissolution of silica in CFA and synthesized different types of zeolites through the two-step method. The findings revealed that due to the increase in NaOH concentration, Zeolite A with a narrow particle size distribution can be obtained within a short crystallization time. Submicron Zeolite A was synthesized from CFA for the first time. Musyoka *et al.* (2012) optimized the synthesis parameters of pure-phase Zeolite P synthesized from three kinds of CFA by using the two-step method. They reported that the quality of synthetic zeolites is largely affected by the CFA source and can be improved by adjusting the experimental conditions. Moreover, the cation exchange capacity of pure-phase Zeolite P can reach 4.11 meg g⁻¹.

The Si and Al ions in the waste liquor of the direct hydrothermal synthesis process can be fully utilized in the two-step synthesis process. Zeolites with high purity can be further obtained by adding a Si or Al source. However, this

method entails a complex operation, long reaction cycle, low yield of pure zeolite, and high cost.

Alkaline Fusion-assisted Hydrothermal Method

The alkaline fusion-assisted hydrothermal method was introduced to improve the yield and quality of zeolite synthesis. This method comprises two main parts. The first part involves melting the mixture of CFA and alkali at a high temperature. In the second part, the alkali melt product is processed through hydrothermal treatment.

Shigemoto *et al.* (1993) were the first to introduce the alkaline fusion-assisted hydrothermal process for the synthesis of Zeolite X with a crystallinity of 62%. Mullite was proven to be an inert substance that limits the quality and yield of zeolite synthesized from CFA. The mechanism of this method was also analyzed by Bergaut and Singer (1996), who proved that the formation of sodium aluminosilicates occurs through an endothermic reaction at 170–180°C. Aluminosilicate gel can be formed and further converted into zeolite by mixing with water. The researchers also discovered that mullite can be fully hydrolyzed and converted into zeolite by adding a small amount of water to the mixture of CFA and NaOH before melting. Rayalu *et al.* (2000) analyzed the effects of NaOH concentration on the conversion of CFA and types of zeolites. They showed that a low NaOH concentration leads to a small extraction amount of aluminum silicate in CFA, which is not conducive to the formation of zeolite. However, excessive NaOH enrichment results in the conversion of zeolite to hydroxysodalite. Zeolites synthesized through the alkali fusion method usually have a higher specific surface area, cation exchange capacity, and crystallinity than those synthesized using the traditional hydrothermal method (Molina and Poole, 2004). Wajima and Sugawara (2010) reported that the amount of calcium in CFA has an important effect on the type of zeolite, and a low calcium content is conducive to the synthesis of zeolite. Yang *et al.* (2019a) proved that the synthesis of zeolite with the double alkali method is more effective than that with the single alkali approach. Using response surface methodology, Sivalingam and Sen (2018) synthesized Zeolite X with a maximum crystallinity of 76% and a maximum yield of 88%. The crystal violet adsorption capacity is 99.62%, which is better than that of commercial Zeolite X. Feng *et al.* (2018) quantified the transfer of heavy metals from fly ash to produce zeolite and wastewater and realized high conversion of the main elements (98.2% Al and 96.5% Si) to synthesize zeolite and zero secondary solid waste.

Many studies have proven that zeolites synthesized through the alkaline fusion-assisted hydrothermal method have high conversion and quality. This method can be successfully applied to different kinds of CFA, but it requires calcination, which increases the cost of preparation and needs further study.

Microwave-assisted Method

Querol *et al.* (1997a) proposed the synthesis of zeolites from CFA through the microwave-assisted method, which is a synthesis process that involves changing the heating mode from electric heating to microwave heating. Compared with

the traditional hydrothermal method, the microwave method can obtain zeolites with the same quality but within a shorter time. Tanaka *et al.* (2008a) proved that the microwave method can shorten the crystallization time and obtained single-phase Zeolite A from CFA through this technique. Inada *et al.* (2005b) described the effect of microwave irradiation on the yield of zeolite synthesized from CFA and analyzed the crystallization mechanism of zeolite. They demonstrated that microwave irradiation can effectively improve zeolitization, continuous microwave radiation can inhibit the formation of zeolite crystals, and early microwave heating is beneficial to the formation of zeolite. Behin *et al.* (2015) conducted an experimental study on the microwave synthesis of Zeolite A. Zeolite Linde Type A with high crystallinity and adsorption capacity were synthesized from CFA and industrial wastewater within a short time (10–30 min) at a relatively low-power microwave irradiation (100–300 W).

Related studies indicate that the major advantages of the microwave-assisted method include increasing the crystallization rate, shortening the crystallization time, and reducing production costs, all of which provide the possibility for the industrialized production of zeolites from CFA. However, due to disadvantages, such as low zeolite conversion rate, numerous by-products, and certain difficulties in amplification engineering equipment, this method has been investigated only at the laboratory scale. Further studies are necessary before applying this method at the industrial scale.

In addition to these usual methods, other techniques,

including molten salt method (Park *et al.*, 2000; Choi *et al.*, 2001), dialysis method (Tanaka *et al.*, 2006), ultrasound-assisted method (Musyoka *et al.*, 2011; Ojumu *et al.*, 2016), and supercritical water technology (Jin *et al.*, 2010), have also been used for the synthesis of zeolites from CFA. Different preparation methods and conditions can form different kinds of zeolites. Fig. 10 depicts the morphology of CFA and several typical zeolites obtained from CFA.

APPLICATION OF CFA ZEOLITES FOR THE REMOVAL OF GASEOUS POLLUTANTS

Air pollution due to NO_x (Zhang *et al.*, 2019b), CO_x (Pillai *et al.*, 2010; Younas *et al.*, 2016; Zhang *et al.*, 2017), SO₂ (Zhang *et al.*, 2019a), Hg (Qi *et al.*, 2015), and VOCs (Lee *et al.*, 2004; Bhatia *et al.*, 2009; Cosseron *et al.*, 2013) is detrimental to human beings and the environment. The main sources and hazards of these pollutants are listed in Table 3. Effectively controlling and reducing air pollution are important issues for scientists today.

At present, the common methods for treating gas pollutants include adsorption (Kang *et al.*, 2018), biodegradation (Zhu *et al.*, 2013), direct combustion (Pan *et al.*, 2018), and catalytic oxidation (Saqer *et al.*, 2011); adsorption and catalytic oxidation are considered the two most effective methods. The adsorption method is widely used due to its advantages, including simple equipment, easy operation, convenient economy, and high efficiency, and its mechanism has been widely studied.

Fig. 10. SEM images of (a) coal fly ash (Missengue *et al.*, 2017), (b) Zeolite A (Qian and Li, 2015), (c) Zeolite X (Hu *et al.*, 2017), (d) Zeolite Y (Padhy *et al.*, 2015), (e) Zeolite ZSM-5 (Anuwattana *et al.*, 2008), (f) hydroxysodalite (Hong *et al.*, 2017).

Table 3. Primary sources and effects of gaseous pollutants.

Pollutants	Primary sources	Environmental hazards	Ref.
CO ₂	Fossil fuel combustion	Global warming, extreme weather	(Pillai <i>et al.</i> , 2010;
CO	Iron works, fertilizer plants and petrochemical industry	Harmful to the brain, cardiovascular system and even fatal	Younas <i>et al.</i> , 2016; Zhang <i>et al.</i> , 2017)
NO _x	Fossil fuel combustion	Photochemical smog, acid rain	(Zhang <i>et al.</i> , 2019b)
SO ₂	Coal combustion	Acid rain, asthma	(Zhang <i>et al.</i> , 2019a)
Hg	Volcanic activity, fossil fuel combustion	Air and water pollution, insanity, pneumonia	(Qi <i>et al.</i> , 2015)
VOCs	Benzene	Petroleum, adhesives, Carcinogen, damage the ozone layer,	(Lee <i>et al.</i> , 2004;
	Toluene	lacquers, pesticides, photochemical smog	Bhatia <i>et al.</i> , 2009;
	Ethylbenzene	gasoline, paints, combustion sources	Cosseron <i>et al.</i> , 2013)
	Carbon tetrachloride	Chemical extractants, paints, Acute toxicity, greenhouse effect,	
	Chlorobenzene	adhesive, polymer syntheses damage the ozone layer	
	Acetone	Varnishes, adhesives, paint Central nervous system depression,	
		thinners headache, nausea	
	Formaldehyde	Decorative, cosmetics, Nasal tumors, irritation of the throat,	
		adhesives, biomass burning eyes, and skin	
	Methanol	Preservative Central nervous system depression,	
		irritation of the throat and eyes	
	Methylene chloride	Flooring materials, furniture, Carcinogen, irritation of skin, harm	
		fumigants, coatings and to liver and kidney	
		paints	

Many achievements in the removal of gaseous pollutants have been obtained through the combination of the abovementioned methods and mechanisms. Table 4 presents a summary of the research results on removing gaseous pollutants using zeolites synthesized by CFA.

Removal of S Compounds

SO₂ and SO₃ are the major components of acid rain, and they can form sub-micron acid particles that are absorbed by the human body (Yang and Zhang, 2011; Hui *et al.*, 2015). Srinivasan and Grutzeck (1999) used Class F CFA as a raw material for the synthesis of zeolites and investigated the influencing factors of SO₂ adsorption. The results showed that the amount of removed SO₂ depends on the type of zeolite, degree of drying, and zeolite content. Regardless of the CFA source, the sample (150°C) containing analcime and sodalite could adsorb 6–7 mg SO₂ per gram, whereas the sample containing X, Y, and Na-P1 had significantly low adsorption efficiency. Suchecki *et al.* (2004) proved that fly ash zeolite (FAZ) is an effective SO₂ adsorbent and has a higher adsorption capacity than CFA (Fig. 8). In addition, thermal activation increases the SO₂ adsorption capacity of FAZs by two to three times, reaching up to 38 mg SO₂ per gram of zeolite.

Izquierdo *et al.* (2013) prepared Ca-exchanged Zeolite Y from CFA and performed corresponding SO₂ adsorption experiments. They reported that the adsorption ability of SO₂ is strongly associated with the presence or absence of water in the flue gas. In the presence of water, H₂SO₄ and CaSO₄ are formed in the adsorption process, which seriously changes the structure of zeolite and reduces its activity. Czuma *et al.* (2016) investigated the effect of different CFA-based zeolites obtained by changing the melting reaction

parameters on the adsorption capacity of SO₂. The authors stated that the yield and type of zeolites can be influenced by the alkali/ash ratio, alkali melting temperature, and adsorption capacity of SO₂. They also observed that the adsorption capacity decreases after the first cycle, whereas it remains approximately constant during the second and third cycles.

Removal of CO₂

The rapid increase in the concentration of CO₂ in the atmosphere has elicited people's concern about global climate change and CO₂ emission reduction. Liu *et al.* (2011) used the alkaline fusion hydrothermal method to form Zeolite A and A + X mixtures from CFA, which were then used in a CO₂ adsorption test. Compared with Zeolite X synthesized at a high temperature of 90°C, the two zeolites have better performance in capturing CO₂ from flue gas and are more selective for CO₂ than N₂. Lee and Jo (2010) studied the adsorption behavior of Ca ion-exchanged Na-P1 and Na-A zeolites prepared from CFA at a low CO₂ concentration. Their results indicated that the best CO₂ adsorption can be mainly attributed to the electrostatic behavior of Ca ions and the enhancement of acid-base interactions. Soe *et al.* (2016) and Muvumbu (2015) emphasized that Ca ion-exchanged zeolites have excellent CO₂ adsorbability. Sarmah *et al.* (2013) performed CO₂ adsorption experiments on CFA-based zeolite modified by monoethanolamine/*N,N*-dimethylaniline and diethylamine/*N,N*-dimethylaniline, and the adsorption capacity reached 6.89 and 5.99 mmol g⁻¹ respectively. In addition, the modified zeolite had a lower cost and higher adsorption efficiency than commercial zeolite. Kalvachev *et al.* (2016) investigated the influence of preparation conditions on the conversion of CFA into zeolite and CO₂ adsorption.

Table 4. Removal of gaseous pollutants by zeolites synthesized from CFA.

Adsorbate	Adsorbent	BET (m ² g ⁻¹)	Temperature (°C)	Flow rate (mL min ⁻¹)	Sorption capacity	Ref.
CO ₂	Zeolite A		25	30	145 mg g ⁻¹	(Soe et al., 2016)
	Zeolite X	800	25	30	220 mg g ⁻¹	
CO ₂	Zeolite X	643	25	100	225 mg g ⁻¹	(Zhang et al., 2017)
CO ₂	MCM-41-60% PEI		75	100	13.31%	(Panek et al., 2017)
CO ₂	Zeolite X-25% PEI-600	14.2	70	0.5	26 mg g ⁻¹	(Dindi et al., 2017)
CO ₂	Zeolite X		22	30	60 mg g ⁻¹	(Kalvachev et al., 2016)
SO ₂	Merlinoite, perialite	102.4	Room temperature	100	46 mg g ⁻¹	(Pedrolo et al., 2017)
SO ₂	Zeolite X, Zeolite A		25		1.68 mmol g ⁻¹	(Czuma et al., 2016)
SO ₂	Zeolite Y- Ca	426	100	25	6.9 mg g ⁻¹ (50% breakthrough)	(Izquierdo et al., 2013)
SO ₂	Zeolite P			1000	38 mg g ⁻¹	(Suchecki et al., 2004)
SO ₂	Analcime, sodalite			600	6–7 mg g ⁻¹	(Srinivasan and Grutzeck, 1999)
N ₂ O	Analcime, Na-PI				64.77%	(Maiti et al., 2018)
NO					24.60%	
NO ₂					10.63%	
NO _x			292		65%	
NO	Zeolite X-Ag					(Karthikeyan and Saravanan, 2013)
	Zeolite Y-Cu	536	200–210		100%	(Izquierdo et al., 2016)
	Zeolite Y-Fe	182			100%	
VOCs	Na-PI, quartz, mullite, calcite	94.49	45		1.44 µg g ⁻¹	(Bandura et al., 2016)
					10.52 µg g ⁻¹	
					26.22 µg g ⁻¹	
					25.96 µg g ⁻¹	
					20.50 µg g ⁻¹	
	NaX, quartz, mullite, calcite	157.4	45		383.67 µg g ⁻¹	(Bandura et al., 2016)
					525.03 µg g ⁻¹	
					545.26 µg g ⁻¹	
					563.82 µg g ⁻¹	
					582.91 µg g ⁻¹	
					69.2%	
	NaPI	22.08	Room temperature		63.32%	(Zhou et al., 2014)
	NaX, NaPI, NaA	34.06				(Wei et al., 2013)
	NaX, zeolite SAPO					(Zhang et al., 2012)
	Cancrinite, sodalite	48.5	Room temperature	110	151 mg g ⁻¹	(Wang et al., 2015)
Hg	Ag-NaX	178	100	470	75%	(Wdowin et al., 2015)
Hg	Ag-NaX	203	150	1416	98%	(Wdowin et al., 2015)
Hg	Ag-NaX	53	30	80	37.46% (5% breakthrough)	(Wdowin et al., 2014)
	Ag-NaPI		30	80	4.35% (5% breakthrough)	
Hg	Cancrinite	11.9	200–210	1000	80%	(Ma et al., 2019)

They reported that Zeolite X can be obtained when the alkali/ash ratio is 2, the roasting temperature is 550°C, and the hydrothermal reaction occurs at 90°C for 2 h. The adsorption capacity of CO₂ can reach 60 mg g⁻¹ at 22°C. Dindi *et al.* (2017) studied the effects of different polyethylenimine (PEI: MW-600 and MW-1200) loadings (15%, 20%, and 25%) on CO₂ adsorption. The results showed that PEI with a low molecular weight is better than that with a high molecular weight. The adsorption performance improves as the PEI load increases, but an excessive load hinders dispersion and degrades performance. At a loading of 25%, PEI-600 achieves the maximum CO₂ adsorption energy (26 mg g⁻¹) at 70°C.

Removal of N Compounds

NO_x are common air contaminants that cause environmental issues, such as acid rain. López-Salinas *et al.* (1997) formed Zeolite P or faujasite-type zeolites from CFA and investigated the catalytic performance of 3.6 wt.% Cu-impregnated zeolites in the CO reduction of NO. At 505 K, the conversions of NO for Cu-loaded Zeolite P and faujasite-type zeolite were 47% and 36%, respectively, whereas the untreated CFA exhibited almost no activity in the reaction. Bopaiah and Grutzeck (2000) examined the synthesis of zeolites from CFA, ground glass particles, and cement kiln dust and their adsorption of NO₂. The hydrothermal reaction was executed at 90°C for 56 days. The zeolite yield was about 30%, and it was able to capture 88 mg of NO₂ per gram of zeolite. Karthikeya and Saravanan (2013) compared the catalytic removal of NO_x using ion-exchange CFA-based X-zeolite and commercial 13-X zeolite. The authors stated that the catalytic performance of Ag-X, Fe-X, and Ni-X catalysts prepared from CFA is better than that of the commercial 13-X zeolite catalyst. Furthermore, Ag and Fe zeolite catalysts have the best NO_x reduction performance at high and low temperatures, respectively, and no obvious deactivation occurs after 50 h of reaction. Izquierdo *et al.* (2016) investigated the selective catalytic reduction of NO with fly ash-derived Zeolite Y exchanged with Cu²⁺ or Fe²⁺. At temperatures below 200°C and in the presence of water, the difference in NO reduction is caused by the different temperatures of active substances Cu²⁺ and Fe³⁺ in the ion-exchanged zeolites and Zeolite LY-Cu with a large surface area and high reactivity. The entire equation can be expressed as $4\text{NO} + 4\text{NH}_3 + \text{O}_2 \leftrightarrow 4\text{N}_2 + 6\text{H}_2\text{O}$, where water is a product of the reaction. The presence of water vapor in the airflow can upset the equilibrium, increase the concentration of NO + O₂, further promote the oxidation of Cu⁺, and increase a large number of active sites because the oxidation of Cu⁺ to Cu²⁺ below 325°C is due to NO + O₂. Maiti *et al.* (2018) applied X-ray photoelectron spectroscopy to examine the adsorption of harmful gaseous pollutants in CFA-based zeolite containing Na-P1 and analcime phases. Their findings revealed that synthetic zeolite has acid and thermal resistivity and can be used for internal and external environmental conditions. The highest adsorption of N₂O, NO, CO, and NO₂ on the zeolite surface reached 64.77%, 24.60%, 12.94%, and 10.63%, respectively. Therefore, CFA-based zeolite could be used to purify the most common air contaminants in indoor and outdoor environments.

Removal of Hg Vapor

Hg is one of the most prominent pollutants in the environment due to its high volatility, high toxicity, and biological enrichment. Wdowin *et al.* (2014) investigated the adsorption of Hg from flue gases by activated carbon with bromine (AC/Br), a natural zeolite, and zeolites (Na-X and Na-P1) synthesized from CFA. The authors stated that untreated synthetic zeolite has almost no adsorption for Hg. The Hg removal efficiency of Na-X/Ag obtained through the ion exchange method was higher than that obtained through the melting method and superior to AC/Br and natural zeolite. The increased Hg adsorption efficiency of Na-X/Ag was attributed to the highest specific surface area of the zeolite treated with Ag and the highest combination of microporosity and medium porosity. In a subsequent study, Wdowin *et al.* (2015) indicated that the removal efficiency of Hg is related to the structure of the adsorbent (powder or particle), flue gas flow, contact time, and experimental temperature. According to the experimental results, Ag-impregnated CFA-based Zeolite X reduces the Hg content in flue gas and obtains a long-term Hg breakthrough of 15–40%. The effect of granular materials on Hg capture is preferable to that of powder materials. Wang *et al.* (2015) studied the adsorption performance of Hg by zeolite achieved from CFA by using the supercritical hydrothermal method. The CFA-based zeolite obtained at an S/L ratio of 15, NaOH concentration of 1 mol L⁻¹, supercritical water temperature of 400°C, and reaction time of 5 min exhibited a satisfactory Hg removal performance. The authors emphasized that a low Si/Al ratio and low Ca and Fe content are beneficial to the synthesis of zeolite. The sodalite and cancrinite in the product were the main active crystals, and at 100°C reaction for least 450 min, the removal efficiency of Hg exceeded 75% and 60%, respectively. Ma *et al.* (2019) used fly ash and Fe-containing waste as raw materials to synthesize magnetite through the one-step supercritical hydrothermal method. After modification using HCl, the removal rate of the elemental Hg (Hg⁰) in the flue gas by zeolite exceeded 80% within the temperature range of 50–300°C.

Removal of VOCs

VOCs have attracted increasing attention due to their contribution to the creation of photochemical smog and harmful effects on the atmosphere and human health. Zhang *et al.* (2012) reported the adsorption of benzene on CFA-based Zeolite X modified using 20 wt.% HCl and 10 M NaOH. During their tests, they observed that the maximum adsorption capacity of Zeolite X on benzene is 151 mg g⁻¹, which is higher than that of commercial activated carbon. The results showed that the adsorption capacity is influenced by the preparation temperature and time. In a subsequent study, the researchers discovered that Zeolite X produced from CFA at a hydrothermal temperature of 140°C for 8 h has an Si/Al molar ratio of 7.9 and an adsorption rate of 66.51% on benzene gas (Wei *et al.*, 2013). Zhou *et al.* (2014) generated CFA-based zeolite by alkali mixture (NaOH and KOH) modification and used it for benzene vapor adsorption. The results showed that single-phase zeolite NaP1 can be obtained in the 8 M mixed alkali solution, and the adsorption

energy of benzene vapor can reach 69.2%, which is similar to that of commercial benzene vapor adsorbents. Rayalu *et al.* (2006) utilized surface-modified Zeolite Y formed from CFA as an adsorbent for VOCs. Their results indicated that Zeolite Y modified by hexadecyltrimethylammonium bromide increases the hydrophobicity and adsorption capacity of VOCs. This material is stable at 250°C, which is higher than the desorption temperature. In addition, the product has strong adsorption selectivity toward VOCs, good stability, and regeneration possibility. Bandura *et al.* (2016) demonstrated that Na-X zeolite made from CFA has a higher adsorption capacity (383.67–582.91 $\mu\text{g g}^{-1}$) for benzene, toluene, and xylene in flue gas compared with the CFA-based zeolite Na-P1 (1.44–20.50 $\mu\text{g g}^{-1}$), natural clinoptilolite (0.68–2.84 $\mu\text{g g}^{-1}$), and diatomite (0.20–0.96 $\mu\text{g g}^{-1}$). This was because micropores play a major role in the structure of faujasite with large-scale channels. The adsorption capacity of the experimental materials was in the order of xylene > toluene > benzene. Another study experimented on the use of synthetic zeolites from high-alumina CFA in the adsorption of formaldehyde, benzene, and toluene (Yuan *et al.*, 2015). The products were amorphous silicate materials, crystalline calcite, and some impurities. In addition, the adsorption performance of the products on VOCs was similar to that of commercial activated carbon and stronger than that of commercial diatomite. The order of adsorption was toluene > benzene > formaldehyde. The Fourier transform infrared spectroscopy results indicated that a weak hydrogen bond interaction exists between the products and the three volatile organic compounds.

CHALLENGES AND PERSPECTIVES

Coal fly ash, the solid waste generated by coal-fired power plants, is one of the most innovative renewable energy sources. However, due to the annual increase in its cumulative emission, the resource utilization of CFA has become a serious issue. At present, the primary method of comprehensively utilizing this substance is to produce cement, concrete, and other building materials. However, numerous studies have emphasized that transforming CFA into zeolite products is an effective way to utilize and add high value to it. Our review summarizes the four major ways of synthesizing zeolites from CFA, namely, the direct hydrothermal, two-step hydrothermal, alkaline fusion-assisted hydrothermal, and microwave-assisted methods. Each method exhibits major advantages and disadvantages, so further research is required. Exploring novel synthetic methods or combining them with other techniques should also be considered. Because only a few types of zeolites can be synthesized currently from CFA, additional types should be developed and synthesized, and their purity, crystallinity, and conversion rates should be improved. Accelerating the industrialization of CFA-based zeolite synthesis is also an important research goal. Related literature suggests that the application of CFA-based zeolites in removing harmful gases mainly focuses on ion exchange and adsorption; Therefore, there is still much room for research on catalytic oxidation of these zeolites to remove harmful gases. Furthermore, research should be conducted to increase their

efficiency in filtering exhaust gas and regenerating.

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