

## Review Article

# Firing-Associated Recycling of Coal-Fired Power Plant Fly Ash

**Vu Thi Ngoc Minh** <sup>1</sup>, **Vuong-Hung Pham**,<sup>2</sup> **Vu Hoang Tung**,<sup>1</sup> **Cao Tho Tung**,<sup>1</sup>  
and **Nguyen Thi Hong Phuong** <sup>1</sup>

<sup>1</sup>*School of Chemical Engineering, Hanoi University of Science and Technology (HUST), No. 01, Dai Co Viet Road, Hanoi, Vietnam*

<sup>2</sup>*Advanced Institute for Science and Technology (AIST), Hanoi University of Science and Technology (HUST), No. 01, Dai Co Viet Road, Hanoi, Vietnam*

Correspondence should be addressed to Vu Thi Ngoc Minh; [minh.vuthingoc@hust.edu.vn](mailto:minh.vuthingoc@hust.edu.vn)

Received 22 June 2022; Revised 18 September 2022; Accepted 7 October 2022; Published 27 February 2023

Academic Editor: Tien Duc Pham

Copyright © 2023 Vu Thi Ngoc Minh et al. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

Coal-fired power plant fly ash is a global environmental concern due to its small particle size, heavy metal content, and increased emissions. Although widely used in concrete, geopolymer, and fly ash brick production, a large amount of fly ash remains in storage sites or is used in landfills due to inadequate raw material quality, resulting in a waste of a recoverable resource. Therefore, the ongoing need is to develop new methods for recycling fly ash. The present review differentiates the physiochemical properties of fly ash from two coal combustion processes: fluidized bed combustion and pulverized coal combustion. It then discusses applications that can consume fly ash without strict chemical requirements, focusing on firing-associated methods. Finally, the challenges and opportunities of fly ash recycling are discussed.

## 1. Introduction

Since the late 19th, coal-fired power has continuously grown and has been an essential source of global electricity. As of 2019, coal-fired power accounts for 36.7% of the world's total electricity production and 22.4% of the total electricity of countries in the organization for economic cooperation and development (OECD) [1]. Due to environmental concerns and the need for sustainable development, coal-fired power has gradually been replaced by other energy sources, such as wind, solar, natural gas, biomass, and combustible waste power. However, under the pressure of economic development, despite the decrease in proportion, the total capacity of coal-fired power continues to grow, especially in Asian countries such as China, India, and Southeast Asia, threatening net-zero targets by 2050. The IEA predicts that coal-fired power generation from 2021 to 2024 will increase by 4.1% in China, 11% in India, and 12% in Southeast Asia [2].

Environmental concerns arise from fuel extraction operations, the coal power generation process, and emission treatment. The primary discharge of coal combustion includes acidic gases such as SO<sub>2</sub>, NO<sub>x</sub>, and CO<sub>2</sub>; heavy metal vapors such as mercury; and solid combustion residues such as dust, fly ash, and bottom ash. Strongly acidic gases, including NO<sub>x</sub> and SO<sub>2</sub>, go through chemical processes depending on the combustion system. Conventional boilers use low-ash coal grades and the combustion chamber temperature is usually above 1400°C. At this temperature, nitrogen gas oxidizes, forming NO<sub>x</sub> in the flue gas. The flue gas is directed to an adsorption tower that performs selective catalytic reduction (SCR) to convert NO<sub>x</sub> into nitrogen gas and water. The SO<sub>2</sub> gas formed by the oxidation of sulfur-containing substances is absorbed in a wet flue gas desulfurization (WFGD) system with lime/limestone slurry to produce calcium sulfate. Fluidized bed combustion boilers often use high-ash coal grades and the temperature of the combustion chamber usually does not exceed 900°C

to limit the generation of  $\text{NO}_x$ . In this system, limestone can be fed into the combustion chamber to absorb  $\text{SO}_2$  gas [3].

Coal fly ash is also a primary environmental concern due to its small particle size and heavy metal contents. Continued increases in coal-fired power generation have resulted in a continued increase in global coal ash emissions. While a large amount of coal fly ash is used in nonfiring construction materials such as concrete [4–6], fly ash bricks [7, 8], and geopolymer [9, 10], a significant amount remains at the storage site or is used for landfills. A recent review of fly ash usage in China shows that 56% of fly ash is used in construction, 35% is used in landfills, and 9% is used in other applications [11].

For safety reasons, fly ash used as pozzolanic additives in concrete and cement has stringent quality requirements. The American standard ASTM C618-19 classifies fly ash used for concrete into classes F and C by chemical composition, loss-on-ignition, physical properties, and pozzolanic activity [12]. Chemically, the most significant difference between Class C and Class F is in the CaO content and the total  $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$  content. Due to its high CaO content, often associated with lime and calcium sulfate, Class C fly ash features self-adhesive properties [13]. Class F fly ash contains a higher  $\text{SiO}_2$  content than Class C and shows pozzolanic activity in mortar and concrete. Ones that do not belong to Class C and Class F are not considered pozzolanic additives. Due to the quality requirements mentioned above, a large portion of fly ash piles up in storage sites by the power plant or is used for landfills and mine backfills [14, 15].

Under environmental stress, intensive studies have focused on thoroughly treating and utilizing fly ash in more value-added applications. In India, 32.87% of fly ash was left untreated during 2017–2018 [16], which decreased to 7.59% by 2020–2021 [17]. During the year 2020–2021, ten modes of fly ash utilization were effective: cement (25.81%), mine filling (6.20%), bricks and tiles (12.98%), reclamation of low-lying area (15.59%), ash dyke raising (7.94%), roads and flyovers (15.04%), agriculture (0.03%), concrete (0.83%), hydropower sector (0.03%), and others (7.97%) [17]. In this example, the main distribution of fly ash is in construction, of which the portion used for landfill and foundation reclamation accounts for a greater proportion than that used in cement, concrete, brick, and tiles. The environmental concerns associated with such applications include wind-blown dust [18] and the dissolution of toxic metals into groundwater [19, 20]. Therefore, methods of using fly ash to mitigate these risks are still under study.

Due to its porous structure, fly ash has a large specific surface area, which is suitable as a substrate in waste gas and wastewater treatment. In gas treatment, fly ash is mixed with calcium hydroxide to adsorb  $\text{SO}_2$  in flue gas [21, 22]. This mixture can also adsorb toxic organic vapors such as toluene [23] and m-xylene [24]. In wastewater treatment, fly ash improves the precipitation of heavy metals [25], boron [26], and phosphate [27] in the presence of calcium hydroxide. In addition, fly ash can adsorb phenolic compounds [28, 29], dyes [30, 31], and pesticides [32]. Although economically

and technically efficient to some extent, these applications accumulate toxic substances on the fly ash, making post-adsorption treatment of fly ash even more complicated.

Other applications of fly ash include raw materials for glass [33, 34], glass ceramics [35, 36], sintered bricks [37, 38], and ceramic tiles [39, 40]. This review covers the physical and chemical properties of coal fly ash and focuses on the firing-associated recycling of this material. The main objective of the review is to provide evidence that coal fly ash can be used as ceramic raw material without violating regulations on the release of heavy metals into the environment during use. The review also proves that this method can recycle a large amount of fly ash remaining in the storage sites because it does not place any limit on the origin and quality of fly ash.

## 2. Methodology

Most of the documents cited in this review paper are publications from Google Scholar and Web of Science databases. The statistical data from the International Energy Agency (IEA) and the India Central Electricity Authority (CEA) and test methods of the American Society for Testing and Materials (ASTM) and the International Standards Organization (ISO) are also cited.

As a supplement to the review, coal fly ash samples were collected and characterized. The samples were collected from four coal-fired power plants in Vietnam: Dong Trieu, Mong Duong I, Uong Bi, and Ninh Binh. Dong Trieu and Mong Duong I power plants apply the fluidized bed combustion (FBC) technique, whereas Uong Bi and Ninh Binh power plants employ the pulverized coal combustion (PCC) technique. The fly ash was dried overnight at  $115^\circ\text{C}$  before being characterized by X-ray diffractometry, scanning electron microscopy, and laser scattering particle size analysis.

A Bruker X-ray Diffractometer model D2 Phaser was used for phase analysis. Care was taken to keep the top surface of the sample mass flat and on the plane of the top of the sample holder to avoid possible displacement errors. The copper radiation source of the instrument was operated at 30 kV and 20 mA while scanning between 10 and 60 degrees  $2-\theta$ . The data collected from the diffractometer were analyzed using Crystal Impact's Match software using the COD database.

The microstructural properties of fly ash were investigated by a JOEL 6360LV scanning electron microscope. Fly ash samples were sprinkled onto PELCO Tabs™ carbon conductive tabs adhered to the SEM sample holders, which were then gently tapped to remove nonadherent particles. Before being placed in the SEM instrument, the sample holders were placed in a sputter coater to coat the fly ash particles with a thin layer of platinum for conductive purposes. The SEM ran under a vacuum.

The fly ash particle size distribution was analyzed using a HORIBA Partica Mini LA350 instrument, which used a laser source with a wavelength of 650 nm. The instrument works on the principle of laser diffraction and Mie's light scattering theory.

### 3. Properties of Coal-Fired Power Plant Fly Ash

Coal combustion techniques significantly influence the morphology, particle shape, and particle size distribution of fly ash. Figure 1 presents the scanning electron microscopy image of fly ash from four coal-fired thermal power plants. Due to high-temperature ( $>1400^{\circ}\text{C}$ ) combustion, the flux agents in the PCC fly ash melted, stuck to unmelted particles, and rounded under surface tension, forming spherical particles upon cooling. On the other hand, FBC fly ash particles have irregular shapes as they were created from a low-temperature ( $<900^{\circ}\text{C}$ ) combustion process and thus did not have sufficient liquid phase to form spherical particles.

Fly ash is a collection of fine particles that have harmful effects on human health and ecology. Significantly, the wind can blow fine fly ash particles long distances, making fly ash a global health and environmental concern [41, 42]. Most coal fly ash has particle sizes under 100 micrometers [43–45]. Figure 2 shows the particle size distribution curves of four types of fly ash. They present the frequency distribution ( $q$  in %) of particles within a specific size range. Although the particle size distribution curves are different and the combustion methods show no trend in these curves, they all have one thing in common: 90% of each sample (D90) is smaller than 100 micrometers and the median particle size (D50) is in the range of 12.4–17.6 micrometers.

The chemical composition of fly ash is quite complicated, depending on the coal origin and the combustion technique. By weight percentage, dominant in fly ash is  $\text{SiO}_2$ , followed by  $\text{Al}_2\text{O}_3$ , and other oxides such as  $\text{Fe}_2\text{O}_3$ ,  $\text{CaO}$ ,  $\text{K}_2\text{O}$ ,  $\text{Na}_2\text{O}$ , and  $\text{TiO}_2$  (Table 1). The introduction of limestone to the FBC boilers adds  $\text{CaO}$  to the solid combustion residues. Fly ash is also a source of heavy metals, including but not limited to V, Cr, Mn, Co, Ni, As, and Hg [46, 48, 49]. In addition, there are distinct differences between the chemical composition of different size fractions of fly ash from a boiler. Significantly higher concentrations of total carbon and heavy metals are found in smaller particles. Slightly higher concentrations of major elements, Si, Al, Fe, Ca, K, Na, Mg, Mn, and Ti, are present in coarser ones [46, 48]. Fortunately, most heavy metals are locked in the glass phase of fly ash [50]. However, the ability to release heavy metals and rare Earth elements from fly ash in landfills is always a community concern. The allowable limits for the disposal of coal combustion residuals vary between countries and regions but may be tightened in the future [51, 52].

The mineral composition of fly ash depends on the coal type and combustion techniques. X-ray diffraction (XRD) analysis of a series of fly ash showed a significant variation in the glass phase content, from 45 to 80% [53]. Besides the glass phase, quartz is the most popular crystalline phase. Its diffraction peaks stand out in the XRD patterns of all types of coal fly ash. Since PCC boilers operate at temperatures above  $1400^{\circ}\text{C}$ , high enough for mullite crystallization, its fly ash often contains mullite ( $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ ) [54, 55]. The mineral composition of mullite is heterogeneous due to the substitution of impurities at the aluminum sites, forming solid solutions. XRD and nuclear magnetic resonance analysis on a bituminous coal fly ash showed a mullite average chemical

composition of  $\text{Al}_{4.61}\text{Fe}_{0.05}\text{Ti}_{0.02}\text{Si}_{1.32}\text{O}_{9.66}$  [56]. FBC boilers operate at approximately  $850^{\circ}\text{C}$ , which is not high enough for mullite formation; thus, no diffraction peaks of mullite can be observed. Instead, hematite, phengite and anhydrite gehlenite might be present in the XRD patterns of FBC fly ash [4, 55, 57]. Figure 3 presents the XRD patterns of four types of fly ash. PCC fly ash shows only two crystal phases, quartz and mullite, while FBC fly ash shows three crystalline phases, including quartz, phengite, and hematite.

In addition to oxide solids, fly ash may contain significant amounts of unburnt carbon (UC), a result of incomplete combustion. The UC content depends on the combustion technology, operating conditions, and coal type. It is assessed by the loss-on-ignition (LOI) test [58]. Thermal analysis of fly ash confirms the presence of UC with a large exothermic peak incorporated with weight loss. The starting point of UC combustion in TG-DTA analysis varies with the type of coal fly ash [55, 59, 60]. Typically, the amount of UC in fly ash is higher than that in bottom ash [61].

Fly ash UC is an inexpensive source of activated carbon for adsorbing harmful components of flue gas such as  $\text{NO}_x$  [62], mercury vapor [63, 64], and organic substances [65]. However, UC is a factor hindering the use of fly ash in many construction applications. From the concrete perspective, UC adsorbs air-entraining admixture, reducing the physical properties of concrete [66]. As specified by ASTM C618 - 19 standard, the LOI content of fly ash used as pozzolanic additives in concrete should be under 6% with some tolerance to the Class F fly ash [12].

The leaching test performed on fly ash by the EN 12457-4 test method shows that the heavy metal concentrations in the leachate are below the inert waste limits specified by the European Landfill Directive [67]. However, the long-term leaching test indicates a potential risk to soil and groundwater [68]. As has been demonstrated in various studies, it is possible to immobilize heavy metals of solid wastes from wastewater treatment plants [4, 69], red sludge from bauxite processing plants [70], and blast furnace slag [71–73] in the structure of glass [74], glass ceramics [75], and ceramic materials [76]. Likewise, introducing fly ash to ceramic production is a potential solution to lock the heavy metals of fly ash in the ceramic network.

Due to high  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  contents, fly ash is suitable as a precursor for aluminosilicate crystalline phases, such as mullite [77, 78] and cordierite [79–82]. In addition, the high  $\text{CaO}$  content in certain types of fly ash, especially those from FBC boilers using limestone to absorb  $\text{SO}_2$ , makes them suitable as a raw material for glass ceramics that contain calcium silicate and calcium aluminosilicate crystalline phases [83–86]. Furthermore, due to its high glass phase content, fly ash is also considered a fluxing agent that promotes sintering in ceramics.

### 4. Firing-Associated Recycling of Fly Ash

**4.1. Bricks.** Fly ash can replace up to 80% of clay in clay bricks, but some adjustments should be made. The presence of a large amount of fly ash significantly reduces the raw mix plasticity index. Therefore, it is necessary to add plastic

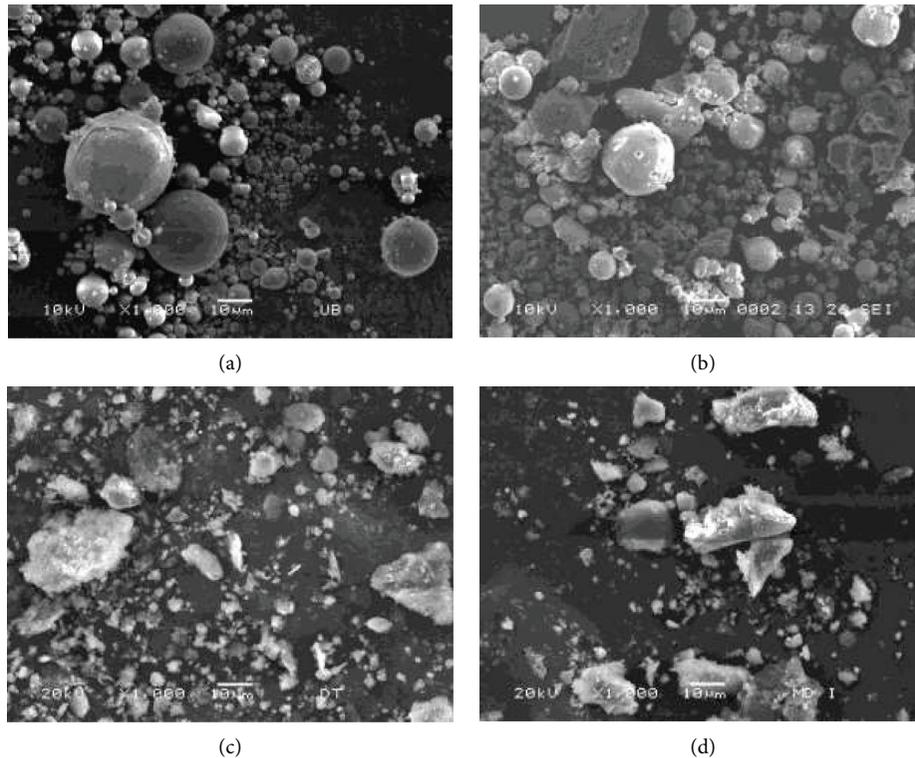


FIGURE 1: Scanning electron microscopy images of fly ash: (a) Uong Bi PCC, (b) Ninh Binh PCC, (c) Dong Trieu FBC, and (d) Mong Duong I FBC.

additives to facilitate extrusion. In addition, the porous structure of fly ash particles impedes the sintering process, increasing the porosity of the sintered product even at low degrees of replacement [87]. Hence, it may be necessary to grind the fly ash to break down its porous structure and increase the contact surface area to speed up the sintering process [37]. Along with the high porosity, the high refractoriness of fly ash requires an increase of the firing temperature by 50 to 100°C so that the physical properties of the sintered products meet those of conventional clay bricks. Most reports indicate that when using large amounts of fly ash, the firing temperature is 1050°C or higher [37, 88]. A test at the industrial scale reveals a number of obstacles that should be overcome: brick swelling and deformation due to local melting and the occurrence of black core and cracks [89].

It is possible to manufacture bricks with 100% of the solid material being fly ash. In this case, shaping requires a temporary binder because most fly ash lacks adhesive capacity. Kayali reported that although the apparent density of the fired product is 28% lower than that of conventional clay bricks, its compressive strength is 24% higher, and brick-to-mortar bond strength is 44% higher than those of the best local clay bricks [8].

It is noteworthy that heavy metals are immobilized in the ceramic structure of the brick. Sutcu et al. [88] showed that the release of heavy metals, including Cr, Mn, Ni, Cu, Zn, As, Cd, Ba, Hg, and Pb, was substantially lower than the maximum allowable limits regulated by U.S. Environmental Protection Agency (EPA) for hazardous solid waste and the

threshold limits of EPA Victoria (Australia) for industrial solid waste. A study by Leiva et al. also shows that the leaching contents are below the limits of the European Landfill Directive for granular waste, the Italian national regulation for the reuse of waste in construction material, and the Dutch Soil Quality Decree for bound or shaped materials [47].

**4.2. Ceramic Tiles.** Fly ash with high flux ( $\text{Fe}_2\text{O}_3 + \text{Na}_2\text{O} + \text{K}_2\text{O}$ ) contents can be used to replace feldspar in ceramic tile production. Olgun et al. were able to improve the modulus of rupture of wall tiles by replacing potassium feldspar in the raw mix with fly ash and borax solid waste without having to change the sintering temperature [90]. As the porous structure of fly ash hinders sintering, resulting in high porosity, it is mainly used as a raw material for wall tiles, equivalent to Group BIII as classified by EN 14411. This type of ceramic tile is not subject to the loading force in use but requires high water absorption for good mortar adhesion, allowing higher water absorption and lower flexural strength than those of the floor tiles [91].

Similar to that observed in clay bricks, the disruption of porous fly ash particles also improves the sintering of the ceramic tiles [92]. When high alumina fly ash is used and a floor tile is the desired product, the firing temperature must be increased to achieve the required physical properties. In particular, the sintering process becomes more difficult when the  $\text{Al}_2\text{O}_3$  content is up to 40%, equivalent to fireclay refractory bricks. Wang et al. showed that for producing

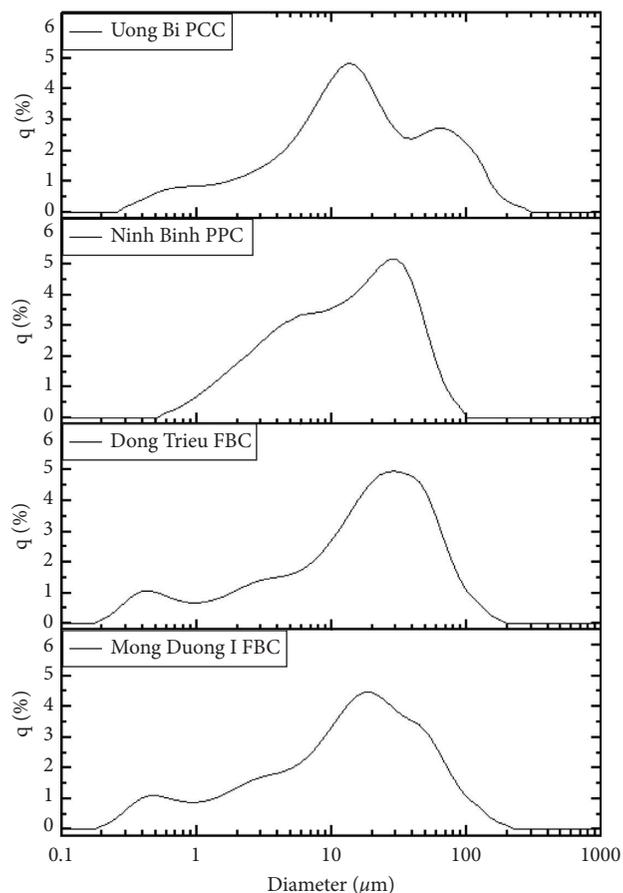


FIGURE 2: Fly ash particle size distribution.

TABLE 1: Chemical composition of fly ash.

Chemical composition	Ninh Binh, Vietnam <sup>a</sup>	Mong Duong I, Vietnam <sup>a</sup>	Seyitomer, Turkey [7]	Tennessee, US [15]	XianYang, China [33]	Bokaro, India <sup>b,d</sup> [46]	Bokaro, India <sup>c,d</sup> [46]	Spain <sup>d</sup> [47]
SiO <sub>2</sub> (wt.%)	45.20	52.16	57.21	49.54	54.39	39.57	51.94	45.02
Al <sub>2</sub> O <sub>3</sub> (wt.%)	19.99	23.65	20.39	19.11	23.21	22.13	26.41	25.18
Fe <sub>2</sub> O <sub>3</sub> (wt.%)	6.86	5.97	10.89	14.79	11.50	3.21	5.21	16.52
CaO (wt.%)	1.52	1.94	2.75	6.23	4.77	0.42	0.49	8.53
MgO (wt.%)	1.29	1.09	4.96	0.42	1.73	0.19	0.27	0.97
K <sub>2</sub> O (wt.%)	3.13	3.30	1.36	—	1.32	0.93	0.95	2.14
Na <sub>2</sub> O (wt.%)	0.32	0.19	0.40	—	0.37	0.13	0.16	0.43
TiO <sub>2</sub> (wt.%)	0.57	0.91	0.81	—	—	2.08	2.09	—
SO <sub>3</sub>	—	—	—	0.34	—	1.45	0.50	—
LOI (wt.%)	20.15	9.46	0.94	2.43	13.38	29.8	9.67	0.7
Cd (ppm)	—	—	—	—	—	—	—	0.9
Pb (ppm)	—	—	79.0	—	—	31.06	63.42	58
Zn (ppm)	—	—	112.6	—	—	47.2	76.6	309
Cu (ppm)	—	—	98.8	—	—	73.8	97.7	-
Cr (ppm)	—	—	454.5	—	—	140.7	137.4	149
Ni (ppm)	—	—	1975.9	—	—	77.5	65.7	74
Mn (ppm)	—	—	790.4	—	—	117.8	268.4	-

<sup>a</sup> Present work, by chemical analysis. <sup>b</sup> Fraction retained by Sieve No. 150 BS (+104 μm). <sup>c</sup> Fraction passed through by Sieve No. 300 B.S and retained by Sieve No 350 BS (-53 μm to +45 μm). <sup>d</sup> Incomplete list of the analyzed heavy metals and rear Earth elements from the cited sources.

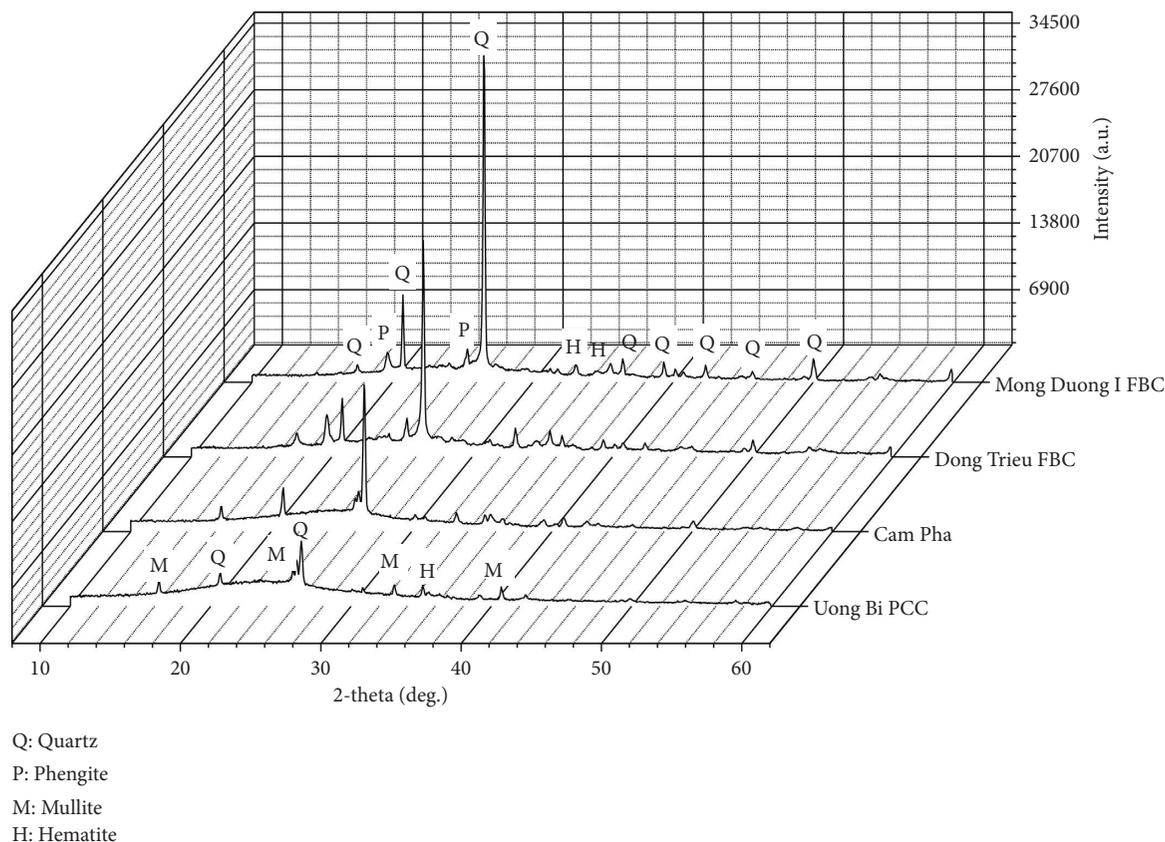


FIGURE 3: The X-ray diffraction patterns of fly ash samples.

ceramic tiles with water absorption below 0.5% from a raw mix with 70% high alumina fly ash, the sintering temperature is as high as 1300°C [93]. Solid wastes with high flux contents, such as glass waste [94, 95], boron waste [90, 96], or lithium mine tailings, are viable additives that help lower the sintering temperature [97].

Wang et al. used high alumina content fly ash (40%  $\text{Al}_2\text{O}_3$ ) and waste glass to make insulating ceramic tiles, consisting of a layer of foam and a layer of dense material. The raw mix of the foam layer consists of waste glass (50%), fly ash (30%), clay (15%), and feldspar (5%). The raw mix of the dense layer consists of fly ash (60–80%), clay (15%), and quartz (5–25%). With single-loading pressing and calcination at 1200°C, the foam layer performs insulation capacity with an average pore diameter of 300–500 micrometers. In addition, the bi-layer ceramic shows a flexural strength of 31.5 MPa. This way, over 70 weight percent of the tile originates from industrial waste [94].

Most fly ash has a  $\text{Fe}_2\text{O}_3$  content of 4–10% (Table 1). The presence of a remarkably high  $\text{Fe}_2\text{O}_3$  content gives a clay brick color to fly ash-based ceramic bodies [8]. On the other hand, the combination of  $\text{Fe}_2\text{O}_3$  and UC causes the black core in bricks [98]. Therefore, if masking the ceramic body color is desired, an engobe glaze with high opacity and whiteness should be applied on the surface of the tiles before the decoration is finished.

Although a test method for determining lead and cadmium leaching from ceramic tiles is available [99], there is no harmonized requirement for hazardous element

concentrations in the leachate of ceramic tiles. However, one can infer from the studies on bricks that ceramic tiles are also capable of immobilizing hazardous elements, even better than bricks because of their denser structure formed by higher sintering temperatures.

#### 4.3. Insulating Materials and Lightweight Concrete Aggregates.

Insulating materials and lightweight concrete aggregates feature porous structures constructed by closed pores. The sintering of these materials requires a simultaneous generation of gas and molten phases with sufficient quantity and viscosity. The molten phase entraps the gas, forming closed pores upon cooling. Fly ash is an excellent candidate for this application due to its UC and high glass phase contents. The UC oxidizes at elevated temperatures releasing carbon dioxide gas while the glass phase is ready to melt. However, if fly ash is the only raw material, the firing temperature should be up to 1300–1400°C for bloating [100]. Furthermore, UC might be low in certain fly ash sources (Table 1). Therefore, fluxes and gas-forming agents are required for (1) generating gases, (2) reducing the firing temperature, and (3) adjusting the quantity and viscosity of the liquid phase for entrapping the generated gases. The fluxes of interest include waste glass [101, 102], limestone [103, 104], and synthetic chemicals such as sodium salts [105, 106]. Some fluxes, including limestone and soda, also play the role of gas-forming agents.

Since most fly ash is not self-adhesive, a temporary binder is required to form the green pellets. On a laboratory

scale, organic binders such as polyvinyl alcohol have been tested [100]. On a larger scale, clay is a popular binder due to its availability and low cost [103, 106]. Besides, bentonite [102, 104] and ordinary Portland cement [104] have been used.

**4.4. Mullite and Cordierite Ceramics.** Fly ash is also a raw material for the production of mullite ceramics due to its high  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  contents. Low alumina fly ash often requires an alumina augmenting agent such as aluminum oxide [107], bauxite [77, 108], or high alumina industrial wastes [78, 109]. Unlike FBC fly ash, mullite is usually available in PPC fly ash. The presence of mullite makes the mullite crystallization in PCC fly ash-based ceramics easier than in those using FBC fly ash.

Although fluxes are available in fly ash and start forming a liquid phase at relatively low temperatures, fly ash-based mullite ceramics are often sintered at high temperatures, usually above  $1400^\circ\text{C}$ , because mullite is a refractory phase. Making dense mullite ceramics from PCC fly ash and bauxite, Dong et al. showed that the solid-state reaction between cristobalite and corundum occurs at temperatures below  $1300^\circ\text{C}$ , followed by the dissolution of the corundum into the liquid phase at higher temperatures where secondary crystallization occurs. The formation and recrystallization of mullite lead to volume expansion which is slightly dominant over the shrinkage of the sintering process. At  $1600^\circ\text{C}$ , the material achieves a relative density of 93.94% with spherical pores and fracture strength of 186.19 MPa [77]. Jung et al. combined aluminum oxide with PCC fly ash, from which UC was removed, to produce mullite ceramics at temperatures of 1400, 1500, and  $1600^\circ\text{C}$ . Although the pellet is cold isostatic pressing, the density of the fired product was relatively low (63%). This low density is due to the exaggerated grain growth of needle-shaped mullite crystals incorporated with voids formation [107].

The introduction of the supplements alters the sintering mechanism either by preventing excessive grain growth or by creating new phases, thereby improving the physical properties of the sintered products. The presence of 3Y-PSZ inhibits the crystal growth of mullite, leading to an improved fracture strength [107]. Magnesia effectively promotes sintering, significantly above  $1450^\circ\text{C}$ . It slightly reduces the linear thermal expansion coefficient (LTEC) at  $1300^\circ\text{C}$  by forming low thermal expansion  $\alpha$ -cordierite. However, it slightly increases the LTEC above  $1400^\circ\text{C}$  due to the formation of high expansion corundum and the spinel ( $\text{MgAl}_2\text{O}_4$ ) [108]. SiC enables the growth of mullite needle-shaped crystals out of the saturated glass phase, resulting in better bonding of the crystals, reducing the true porosity but increasing the closed-pore porosity. As a result, thermal conductivity and cold crushing strength are improved [110].  $\text{TiO}_2$  hinders the sintering process at low temperatures but promotes sintering above  $1300^\circ\text{C}$ . This phenomenon is useful for the unsaturated sintering of porous mullite ceramic membrane supports [111].

The porous structure of fly ash particles and the recrystallization of mullite favor the fabrication of porous mullite ceramics for insulation or filtration purposes. Chen

et al. fabricated porous materials from coal mine waste kaolin and spherical hollow fly ash, using bentonite as the binder and calcium iodate ( $\text{Ca}(\text{IO}_3)_2 \cdot 6\text{H}_2\text{O}$ ) as a flocculant. After firing at  $1550^\circ\text{C}$ , the resulting product has a porosity of 44.73–46.12%, with 99% of the porosity being closed pores, and mullite is the main crystalline phase. The freeze-gel casting/polymer sponge technique can produce a porous mullite ceramic with a porosity of 66.1% and an average compressive strength of 45 MPa [112].

Fly ash is also suitable as a raw material for cordierite ceramics, a material well known for its low LTEC. Cordierite crystallizes in the orthorhombic system. Its dimorphism is hexagonal indialite which is more refractory than cordierite [113–115]. Although the IMA formula of cordierite is  $2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$ , it appears as a solid solution in practice [116, 117]. Thanks to its low LTEC, cordierite ceramic is highly resistant to thermal shock. This material also possesses high chemical resistance and a low dielectric constant. The synthesis of cordierite from fly ash requires MgO supplementing materials such as talc [118], magnesia [119], magnesite [120], or dolomite [121]. Unlike mullite, the sintering temperature of cordierite ceramics is usually below  $1200^\circ\text{C}$ . In addition to indialite, fly ash-based cordierite ceramics may contain mullite, cristobalite, periclase, and spinel [118, 120]. Porous cordierite ceramics from fly ash can be used for microfiltration membranes [82], catalytic substrates [118], and membrane supports [121].

**4.5. Foam Glass and Glass Ceramics.** Thanks to its high silicate glass content, fly ash can be used as a raw material for glass and glass-ceramic synthesis. Erol et al. produce glass by melting fly ash at  $1500^\circ\text{C}$ , glass ceramics by annealing the obtained glass at  $1150^\circ\text{C}$ , and ceramics by firing green pellets at  $1200^\circ\text{C}$ . The only phase in the glass products is amorphous, the crystalline phase in the glass-ceramic products is augite ( $\text{Ca}(\text{Mg}, \text{Fe}^{3+}, \text{Al}) (\text{Si}, \text{Al})_2\text{O}_6$ ), and in the ceramic is quartz, mullite, and enstatite ( $(\text{Mg}, \text{Fe})\text{SiO}_3$ ) [34]. However, making glass and glass ceramics products from fly ash can be difficult due to high  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  (network formers) contents and insufficient fluxes (network modifiers). Therefore, it is necessary to add flux ingredients to lower the melting temperature.

One of the most attentive applications of fly ash-based glass is glass foam, which is considered a low-cost insulation material in construction. Besides fly ash, glass foam raw mixes often have flux and foaming agents. Most of them are industrial wastes. The commonly used flux is waste glass. It is used in large quantities comparable to fly ash [33, 122]. Alternatively, borax [122] and soda [123] can be added to the raw mix as network former and modifier, respectively. Foaming agents can be obtained from a variety of sources, including calcium carbonate [122], soda [123], and SiC [33, 124]. For instance, Bai et al. produced glass foam by melting a raw mix of fly ash, waste glass, and SiC waste at  $950^\circ\text{C}$ . The resulting product expanded 5.81 times compared to the green body [33].

Another fascinating application of fly ash-based glass is glass ceramics because of their high mechanical strength and thermal shock resistance. The crystalline phase in fly ash-

based glass ceramics is controlled by adjusting the composition of the raw mix. The two glass ceramic systems of great interest are SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-CaO [125, 126] and SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-MgO [79, 127]. When a high amount of Fe<sub>2</sub>O<sub>3</sub> is available in fly ash, the system SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-Fe<sub>2</sub>O<sub>3</sub>-CaO is of interest [128]. Depending on the chemical composition of the mix, the crystalline phases in the SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-CaO glass-ceramics include diopside (Ca(Mg, Al)(Si, Al)<sub>2</sub>O<sub>6</sub>) [125], augite (Ca(Mg, Fe)Si<sub>2</sub>O<sub>6</sub>) [34, 125], and wollastonite (CaSiO<sub>3</sub>) [129]. SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-MgO glass ceramics are of the most interest because they contain cordierite, a mineral with low LTEC, and are suitable for thermal shock applications [79, 127].

**4.6. Portland Cement Clinker.** In cement production, fly ash often serves as a pozzolanic additive, but high UC fly ash is not suitable for this application. Another possibility of using fly ash in cement production is to replace clay in the raw mixes of Portland cement clinker [130–133], belite cement clinker [134], and belite-sulfoaluminate cement clinker [135]. This perspective has been investigated from the laboratory to the industrial scale. Laboratory studies showed that the use of fly ash reduced the sintering temperature of Portland cement clinker [130, 131]. Commercial demonstration on high carbon fly ash showed that the obtained fly ash-based Portland cement performs a higher compressive strength than the normal ones at all ages, despite a lower fineness [132]. The use of high carbon high fly ash has the additional benefit of fuel-saving [133].

Unlike ceramic firing, SO<sub>2</sub> formed in the firing process of clinker can be reabsorbed by calcium oxide. The resulting calcium sulfate helps reduce the gypsum amount required for cement setting control [136]. Therefore, using fly ash as raw material for cement clinker brings valuable economic, technical, and environmental benefits.

## 5. Challenges and Opportunities

Although it is demonstrated that fly ash-based glass and ceramic products can immobilize the heavy metals and rare elements of fly ash in their oxide networks, they are not suitable as food and drug containers. This restriction is mainly due to the stringent regulations applied to these products [137–139]. On the other hand, SO<sub>x</sub> emissions from burning fly ash are also of concern because the SO<sub>3</sub> content in certain types of fly ash may be high. High sulfur fly ash is often associated with FBC boilers that introduce limestone into the furnace to absorb SO<sub>x</sub> gas. The SO<sub>3</sub> content in fly ash can be up to 14% [140]. A dramatic increase in the sulfur dioxide content in flue gases during the firing of FBC fly ash-based ceramic tiles was observed. [92]. Unlike ceramic firing, SO<sub>2</sub> formed in the firing process of cement clinker can be reabsorbed and becomes a valuable source of sulfate for cement setting control. Therefore, characterizing fly ash before recycling in these applications is critical in preventing the redistribution of toxic SO<sub>2</sub> into the environment.

## 6. Conclusion

Coal fly ash is a global concern due to its small particle size and heavy metal contents with increasing emissions. Faced with concerns about wind-blowing dust and heavy metal leakage from fly ash landfills and storage sites, efforts to use fly ash for safe and value-added applications are underway. Notable among them are firing-associated measures, including the production of ceramics, lightweight concrete aggregates, glass, and glass ceramics. There is almost no restriction for fly ash in these applications. Most notably, the recycling of fly ash and other industrial solid wastes can be combined in these applications without violating regulations on heavy metals released during use. Besides, the unburnt carbon content can alleviate the heat required for firing. The firing temperature, product phase composition, and physical properties of the products can be controlled by supplementing agents. However, the sulfur content might be high in certain types of fly ash and go into the flue gas as sulfur dioxide. This problem can be solved in cement kilns but should be addressed in other applications.

## Data Availability

All data generated and analyzed to support the findings of this study are available within the article.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

## Authors' Contributions

Conceptualization and supervision were done by Vu Thi Ngoc Minh. Vu Thi Ngoc Minh reviewed the article. Data accuracy was tested by Pham Hung Vuong. Pham Hung Vuong reviewed the article. Conceptualization and data accuracy were done by Vu Hoang Tung. Data analysis was performed by Cao Tho Tung. Cao Tho Tung reviewed the article. Nguyen Thi Hong Phuong performed data analysis.

## Acknowledgments

This work was financially supported by the Vietnam Ministry of Training and Education under grant number B2020-BKA-17. The authors wish to thank Mr. Mai Van Vo and Ms. Bui Thi Thu Ha for their valuable technical support.

## References

- [1] IEA, "Electricity information: overview," 2022, <https://www.iea.org/reports/electricity-information-overview>.
- [2] IEA, "Coal 2021: analysis and forecast to 2024," 2021, <https://www.iea.org/reports/coal-2021>.
- [3] P. Breeze, *Coal-fired Generation*, Academic Press, Cambridge, MA, USA, 2015.
- [4] M. Zahedi and F. Rajabipour, "Fluidized bed combustion (FBC) fly ash and its performance in concrete," *ACI Materials Journal*, vol. 116, no. 4, 2019.
- [5] V. Malhotra, "High-performance high-volume fly ash concrete," *Concrete International*, vol. 24, no. 7, pp. 30–34, 2002.

- [6] P. K. Mehta, "High-performance, high-volume fly ash concrete for sustainable development," in *Proceedings of the International Workshop on Sustainable Development and Concrete Technology*, Ames, IA, USA, 2004.
- [7] T. Cicek and M. Tanriverdi, "Lime based steam autoclaved fly ash bricks," *Construction and Building Materials*, vol. 21, no. 6, pp. 1295–1300, 2007.
- [8] O. Kayali, "High performance bricks from fly ash," in *Proceedings of the World of Coal Ash Conference*, Lexington, Kentucky, 2005.
- [9] M. Amran, S. Debbarma, and T. Ozbakkaloglu, "Fly ash-based eco-friendly geopolymer concrete: a critical review of the long-term durability properties," *Construction and Building Materials*, vol. 270, Article ID 121857, 2021.
- [10] İ. İ. Atabey, O. Karahan, C. Bilim, and C. D. Atiş, "The influence of activator type and quantity on the transport properties of class F fly ash geopolymer," *Construction and Building Materials*, vol. 264, Article ID 120268, 2020.
- [11] Y. Luo, Y. Wu, S. Ma, S. Zheng, Y. Zhang, and P. K. Chu, "Utilization of coal fly ash in China: a mini-review on challenges and future directions," *Environmental Science and Pollution Research*, vol. 28, no. 15, pp. 18727–18740, 2021.
- [12] ASTM C618-19 Standard Specification for Coal Fly, Ash and Raw or Calcined Natural Pozzolan for Use in Concrete, ASTM, West Conshohocken, PA, USA, 2019.
- [13] A. Arman and A. Arman, "Class C fly ash as a full or partial replacement for portland cement or lime," *Transportation Research Record*, vol. 1219, 1989.
- [14] M. Li, J. Zhang, A. Li, and N. Zhou, "Reutilisation of coal gangue and fly ash as underground backfill materials for surface subsidence control," *Journal of Cleaner Production*, vol. 254, Article ID 120113, 2020.
- [15] Y. Yao and H. Sun, "A novel silica alumina-based backfill material composed of coal refuse and fly ash," *Journal of Hazardous Materials*, vol. 213–214, pp. 71–82, 2012.
- [16] CEA, *Report on Fly Ash Generation at Coal/Lignite Based Thermal Power Stations and its Utilization in the Country for the Year 2017-2018*, Central Electricity Authority, Ministry of Power, Government of India, New Delhi, India, 2018.
- [17] CEA, *Report on Fly Ash Generation at Coal/Lignite Based Thermal Power Stations and its Utilization in the Country for the Year 2020-2021*, Central Electricity Authority, Ministry of Power, Government of India, New Delhi, India, 2021.
- [18] UEP Agency, *Human and Ecological Risk Assessment of Coal Combustion Wastes*, RTI International NC, Raleigh, NC, USA, 2007.
- [19] P. Sandeep, S. K. Sahu, P. Kothai, and G. G. Pandit, "Leaching behavior of selected trace and toxic metals in coal fly ash samples collected from two thermal power plants, India," *Bulletin of Environmental Contamination and Toxicology*, vol. 97, no. 3, pp. 425–431, 2016.
- [20] S. Zhao, Y. Duan, J. Lu et al., "Chemical speciation and leaching characteristics of hazardous trace elements in coal and fly ash from coal-fired power plants," *Fuel*, vol. 232, pp. 463–469, 2018.
- [21] A. Al-Shawabkeh, H. Maisuda, and M. Hasatani, "Comparative reactivity of treated FBC-and PCC-Fly ash for SO<sub>2</sub> removal," *Canadian Journal of Chemical Engineering*, vol. 73, no. 5, pp. 678–685, 1995.
- [22] H. Tsuchiai, T. Ishizuka, H. Nakamura, T. Ueno, and H. Hattori, "Removal of sulfur dioxide from flue gas by the absorbent prepared from coal ash: effects of nitrogen oxide and water vapor," *Industrial & Engineering Chemistry Research*, vol. 35, no. 3, pp. 851–855, 1996.
- [23] M. Rovatti, A. Peloso, and G. Ferraiolo, "Susceptibility to regeneration of fly ash as an adsorbent material," *Resources, Conservation and Recycling*, vol. 1, no. 2, 1988.
- [24] S. J. Rothenberg, G. Metzler, J. Poliner, W. E. Bechtold, A. F. Eidson, and G. J. Newton, "Adsorption kinetics of vapor-phase m-xylene on coal fly ash," *Environmental Science and Technology*, vol. 25, no. 5, pp. 930–935, 1991.
- [25] Q. Chen, Z. Luo, C. Hills, G. Xue, and M. Tyrer, "Precipitation of heavy metals from wastewater using simulated flue gas: sequent additions of fly ash, lime and carbon dioxide," *Water Research*, vol. 43, no. 10, pp. 2605–2614, 2009.
- [26] N. Öztürk and D. Kavak, "Adsorption of boron from aqueous solutions using fly ash: batch and column studies," *Journal of Hazardous Materials*, vol. 127, no. 1-3, pp. 81–88, 2005.
- [27] J. Chen, H. Kong, D. Wu, X. Chen, D. Zhang, and Z. Sun, "Phosphate immobilization from aqueous solution by fly ashes in relation to their composition," *Journal of Hazardous Materials*, vol. 139, no. 2, pp. 293–300, 2007.
- [28] Z. Aksu and J. Yener, "The usage of dried activated sludge and fly ash wastes in phenol biosorption/adsorption: comparison with granular activated carbon," *Journal of Environmental Science and Health, Part A*, vol. 34, no. 9, pp. 1777–1796, 1999.
- [29] M. Ugurlu, A. Gurses, M. Yalcin, and C. Dogar, "Removal of phenolic and lignin compounds from bleached kraft mill effluent by fly ash and sepiolite," *Adsorption*, vol. 11, no. 1, pp. 87–97, 2005.
- [30] S. Wang, Y. Boyjoo, A. Choueib, and Z. Zhu, "Removal of dyes from aqueous solution using fly ash and red mud," *Water Research*, vol. 39, no. 1, pp. 129–138, 2005.
- [31] N. Dizge, C. Aydinler, E. Demirbas, M. Kobya, and S. Kara, "Adsorption of reactive dyes from aqueous solutions by fly ash: kinetic and equilibrium studies," *Journal of Hazardous Materials*, vol. 150, no. 3, pp. 737–746, 2008.
- [32] V. K. Gupta and I. Ali, "Removal of DDD and DDE from wastewater using bagasse fly ash, a sugar industry waste," *Water Research*, vol. 35, no. 1, pp. 33–40, 2001.
- [33] J. Bai, X. Yang, S. Xu, W. Jing, and J. Yang, "Preparation of foam glass from waste glass and fly ash," *Materials Letters*, vol. 136, pp. 52–54, 2014.
- [34] M. Erol, S. Küçükbayrak, and A. Ersoy-Mericboyu, "Comparison of the properties of glass, glass-ceramic and ceramic materials produced from coal fly ash," *Journal of Hazardous Materials*, vol. 153, no. 1-2, pp. 418–425, 2008.
- [35] D. H. Vu, K.-S. Wang, J.-H. Chen, B. X. Nam, and B. H. Bac, "Glass-ceramic from mixtures of bottom ash and fly ash," *Waste Management*, vol. 32, no. 12, pp. 2306–2314, 2012.
- [36] J. Zhang, W. Dong, J. Li, L. Qiao, J. Zheng, and J. Sheng, "Utilization of coal fly ash in the glass-ceramic production," *Journal of Hazardous Materials*, vol. 149, no. 2, pp. 523–526, 2007.
- [37] X. Lingling, G. Wei, W. Tao, and Y. Nanru, "Study on fired bricks with replacing clay by fly ash in high volume ratio," *Construction and Building Materials*, vol. 19, no. 3, pp. 243–247, 2005.
- [38] M. S. Başpınar, E. Kahraman, G. Görhan, and İ. Demir, "Production of fired construction brick from high sulfate-containing fly ash with boric acid addition," *Waste Management & Research: The Journal for a Sustainable Circular Economy*, vol. 28, no. 1, pp. 4–10, 2010.
- [39] S. Yürüyen and H. Ö. Toplan, "The sintering kinetics of porcelain bodies made from waste glass and fly ash," *Ceramics International*, vol. 35, no. 6, pp. 2427–2433, 2009.

- [40] Y. Luo, S. Ma, C. Liu, Z. Zhao, S. Zheng, and X. Wang, "Effect of particle size and alkali activation on coal fly ash and their role in sintered ceramic tiles," *Journal of the European Ceramic Society*, vol. 37, no. 4, pp. 1847–1856, 2017.
- [41] N. L. Rose, V. J. Jones, P. E. Noon, D. A. Hodgson, R. J. Flower, and P. G. Appleby, "Long-range transport of pollutants to the Falkland Islands and Antarctica: evidence from lake sediment fly ash particle records," *Environmental Science and Technology*, vol. 46, no. 18, pp. 9881–9889, 2012.
- [42] M. Whiteside and J. M. Herndon, "Aerosolized coal fly ash: risk factor for neurodegenerative disease," *Journal of Advances in Medicine and Medical Research*, vol. 25, no. 10, pp. 1–11, 2018.
- [43] J. Han, H. Fang, and K. Wang, "Design and control shrinkage behavior of high-strength self-consolidating concrete using shrinkage-reducing admixture and super-absorbent polymer," *Journal of sustainable cement-based materials*, vol. 3, no. 3-4, pp. 182–190, 2014.
- [44] S. H. Lee, H. J. Kim, E. Sakai, and M. Daimon, "Effect of particle size distribution of fly ash-cement system on the fluidity of cement pastes," *Cement and Concrete Research*, vol. 33, no. 5, pp. 763–768, 2003.
- [45] A. Sarkar, R. Rano, K. Mishra, and I. Sinha, "Particle size distribution profile of some Indian fly ash—a comparative study to assess their possible uses," *Fuel Processing Technology*, vol. 86, no. 11, pp. 1221–1238, 2005.
- [46] A. Sarkar, R. Rano, G. Udaybhanu, and A. Basu, "A comprehensive characterisation of fly ash from a thermal power plant in Eastern India," *Fuel Processing Technology*, vol. 87, no. 3, pp. 259–277, 2006.
- [47] C. Leiva, M. Rodriguez-Galan, C. Arenas, B. Alonso-Farinas, and B. Peceno, "A mechanical, leaching and radiological assessment of fired bricks with a high content of fly ash," *Ceramics International*, vol. 44, no. 11, pp. 13313–13319, 2018.
- [48] C. Lanzerstorfer, "Fly ash from coal combustion: dependence of the concentration of various elements on the particle size," *Fuel*, vol. 228, pp. 263–271, 2018.
- [49] R. Blissett, N. Smalley, and N. Rowson, "An investigation into six coal fly ashes from the United Kingdom and Poland to evaluate rare earth element content," *Fuel*, vol. 119, pp. 236–239, 2014.
- [50] L. D. Hulett, A. J. Weinberger, K. J. Northcutt, and M. Ferguson, "Chemical species in fly ash from coal-burning power plants," *Science*, vol. 210, no. 4476, pp. 1356–1358, 1980.
- [51] E. Commission, "Council directive 1999/31/EC of 26 april 1999 on the landfill of waste," *Official Journal, L*, vol. 182, pp. 1–19, 1999.
- [52] EPA, *Hazardous and Solid Waste Management System: Disposal of Coal Combustion Residuals from Electric Utilities; Amendments to the National Minimum Criteria (Phase One, Part One)*, EPA, Washington, DC, USA, 2018.
- [53] C. R. Ward and D. French, "Determination of glass content and estimation of glass composition in fly ash using quantitative X-ray diffractometry," *Fuel*, vol. 85, no. 16, pp. 2268–2277, 2006.
- [54] P. W. D. Plessis, T. V. Ojumu, and L. F. Petrik, "Waste minimization protocols for the process of synthesizing zeolites from South African coal fly ash," *Materials*, vol. 6, no. 5, pp. 1688–1703, 2013.
- [55] W. Wu, F. Matalkah, A. Darsanasiri, and P. Soroushian, "Fluidized bed combustion coal fly ash: comparative evaluation for potential use in alkali-activated binders," *International Journal of Coal Preparation and Utilization*, vol. 42, no. 1, pp. 51–66, 2019.
- [56] S. Gomes and M. François, "Characterization of mullite in silicoaluminous fly ash by XRD, TEM, and <sup>29</sup>Si MAS NMR," *Cement and Concrete Research*, vol. 30, no. 2, pp. 175–181, 2000.
- [57] D. Jozić, J. Zelić, and I. Janjatović, "Influence of the coarse fly ash on the mechanical properties of the cement mortars," *Ceramics-Silikáty*, vol. 54, no. 2, pp. 144–151, 2010.
- [58] ASTM D7348-21 Standard Test Methods for Loss on Ignition, (LOI) of Solid Combustion Residues, ASTM, West Conshohocken, PA, USA, 2021.
- [59] R. Kaur and D. Goyal, "Mineralogical comparison of coal fly ash with soil for use in agriculture," *Journal of Material Cycles and Waste Management*, vol. 18, no. 1, pp. 186–200, 2016.
- [60] W. Wons, K. Rzepa, M. Reben, and P. Murzyn, "Thermal studies of fly ashes expansion," *Journal of Thermal Analysis and Calorimetry*, vol. 143, no. 4, pp. 2883–2891, 2021.
- [61] L. Bartoňová, Z. Klika, and D. Spears, "Characterization of unburned carbon from ash after bituminous coal and lignite combustion in CFBs," *Fuel*, vol. 86, no. 3, pp. 455–463, 2007.
- [62] G. Lu and D. Do, "Adsorption properties of fly ash particles for NO<sub>x</sub> removal from flue gases," *Fuel Processing Technology*, vol. 27, no. 1, pp. 95–107, 1991.
- [63] J. C. Hower, H. L. Clack, M. M. Hood, S. G. Hopps, and G. H. Thomas, "Impact of coal source changes on mercury content in fly ash: examples from a Kentucky power plant," *International Journal of Coal Geology*, vol. 170, pp. 2–6, 2017.
- [64] S. D. Serre and G. D. Silcox, "Adsorption of elemental mercury on the residual carbon in coal fly ash," *Industrial & Engineering Chemistry Research*, vol. 39, no. 6, pp. 1723–1730, 2000.
- [65] M. Melih Demirkan, A. H. Aydilek, E. A. Seagren, and J. C. Hower, "Naphthalene and o-xylene adsorption onto high carbon fly ash," *Journal of Environmental Engineering*, vol. 137, no. 5, pp. 377–387, 2011.
- [66] C. Jolicoeur, T. C. To, É. Benoît, R. Hill, Z. Zhang, and M. Pagé, "Fly ash-carbon effects on concrete air entrainment: fundamental studies on their origin and chemical mitigation," in *Proceedings of the 9th ACI International Conference on Superplasticizers and Other Chemical Admixtures in Concrete*, Seville, Spain, 2009.
- [67] C. Leiva, C. Arenas, B. Alonso-farinas et al., "Characteristics of fired bricks with co-combustion fly ashes," *Journal of Building Engineering*, vol. 5, pp. 114–118, 2016.
- [68] C. N. Lange, M. Flues, G. Hiromoto, M. E. G. Boscov, and I. M. C. Camargo, "Long-term leaching of As, Cd, Mo, Pb, and Zn from coal fly ash in column test," *Environmental Monitoring and Assessment*, vol. 191, no. 10, pp. 602–612, 2019.
- [69] P. de Faria Busch, M. T. Marvila, G. C. Girondi Delaqua, and C. M. F. Vieira, "Recycling of waste glass extracted from a WTP into ceramic materials," *Journal of Material Cycles and Waste Management*, vol. 24, no. 2, pp. 763–774, 2022.
- [70] V. Agrawal, R. Paulose, R. Arya et al., "Green conversion of hazardous red mud into diagnostic X-ray shielding tiles," *Journal of Hazardous Materials*, vol. 424, Article ID 127507, 2022.
- [71] H. Liu, H. Lu, D. Chen, H. Wang, H. Xu, and R. Zhang, "Preparation and properties of glass-ceramics derived from blast-furnace slag by a ceramic-sintering process," *Ceramics International*, vol. 35, no. 8, pp. 3181–3184, 2009.

- [72] H. Gao, X. Liu, J. Chen, J. Qi, Y. Wang, and Z. Ai, "Preparation of glass-ceramics with low density and high strength using blast furnace slag, glass fiber and water glass," *Ceramics International*, vol. 44, no. 6, pp. 6044–6053, 2018.
- [73] L. Ding, W. Ning, Q. Wang, D. Shi, and L. Luo, "Preparation and characterization of glass-ceramic foams from blast furnace slag and waste glass," *Materials Letters*, vol. 141, pp. 327–329, 2015.
- [74] O. D. Ozdemir, A. K. Figen, and S. Piskin, "Utilization of galvanic sludge as raw material for production of glass," *International Conference on Chemistry and Chemical Process IPCBEE*, vol. 10, pp. 45–49, 2011.
- [75] J. S. McCloy and A. Goel, "Glass-ceramics for nuclear-waste immobilization," *MRS Bulletin*, vol. 42, no. 03, pp. 233–240, 2017.
- [76] J. J. Castañeda, E. Espejo, and G. I. Cubillos, "Evaluation of industrial shaping processes and firing cycles for the encapsulation of galvanic sludge in ceramics," *Boletín de la Sociedad Española de Cerámica y Vidrio*, Elsevier, Amsterdam, Netherlands, 2022.
- [77] Y. Dong, X. Feng, X. Feng, Y. Ding, X. Liu, and G. Meng, "Preparation of low-cost mullite ceramics from natural bauxite and industrial waste fly ash," *Journal of Alloys and Compounds*, vol. 460, no. 1-2, pp. 599–606, 2008.
- [78] S. Chen, W. H. Cai, J. M. Wu et al., "Porous mullite ceramics with a fully closed-cell structure fabricated by direct coagulation casting using fly ash hollow spheres/kaolin suspension," *Ceramics International*, vol. 46, no. 11, pp. 17508–17513, 2020, <https://doi.org/10.1016/j.ceramint.2020.04.046>.
- [79] Y. He, W. Cheng, and H. Cai, "Characterization of  $\alpha$ -cordierite glass-ceramics from fly ash," *Journal of Hazardous Materials*, vol. 120, no. 1-3, pp. 265–269, 2005.
- [80] S. Kumar, K. K. Singh, and P. Ramachandrarao, "Synthesis of cordierite from fly ash and its refractory properties," *Journal of Materials Science Letters*, vol. 19, no. 14, pp. 1263–1265, 2000.
- [81] H. Shao, K. Liang, F. Zhou, G. Wang, and F. Peng, "Characterization of cordierite-based glass-ceramics produced from fly ash," *Journal of Non-Crystalline Solids*, vol. 337, no. 2, pp. 157–160, 2004.
- [82] Y. Dong, X. Liu, Q. Ma, and G. Meng, "Preparation of cordierite-based porous ceramic micro-filtration membranes using waste fly ash as the main raw materials," *Journal of Membrane Science*, vol. 285, no. 1-2, pp. 173–181, 2006.
- [83] F. Peng, K.-m. Liang, and A.-m. Hu, "Nano-crystal glass-ceramics obtained from high alumina coal fly ash," *Fuel*, vol. 84, no. 4, pp. 341–346, 2005.
- [84] Z. Karoly, I. Mohai, M. Toth, F. Wéber, and J. Szépvölgyi, "Production of glass-ceramics from fly ash using arc plasma," *Journal of the European Ceramic Society*, vol. 27, no. 2-3, pp. 1721–1725, 2007.
- [85] J. M. Kim and H. S. Kim, "Processing and properties of a glass-ceramic from coal fly ash from a thermal power plant through an economic process," *Journal of the European Ceramic Society*, vol. 24, no. 9, pp. 2825–2833, 2004.
- [86] J.-M. Kim and H.-S. Kim, "Glass-ceramic produced from a municipal waste incinerator fly ash with high Cl content," *Journal of the European Ceramic Society*, vol. 24, no. 8, pp. 2373–2382, 2004.
- [87] G. Cultrone and E. Sebastián, "Fly ash addition in clayey materials to improve the quality of solid bricks," *Construction and Building Materials*, vol. 23, no. 2, pp. 1178–1184, 2009.
- [88] M. Sutcu, E. Erdogmus, O. Gencel, A. Gholampour, E. Atan, and T. Ozbakkaloglu, "Recycling of bottom ash and fly ash wastes in eco-friendly clay brick production," *Journal of Cleaner Production*, vol. 233, pp. 753–764, 2019.
- [89] R. M. Vasić, Ž. Lalić, Z. Radojević, and L. Nešić, "Possibilities of producing solid bricks from mixtures of plastic brick clay and fly ash," *Key Engineering Materials*, vol. 264, 2004.
- [90] A. Olgun, Y. Erdogan, Y. Ayhan, and B. J. C. I. Zeybek, "Development of ceramic tiles from coal fly ash and tincal ore waste," *Ceramics International*, vol. 31, no. 1, pp. 153–158, 2005.
- [91] BS EN 14411, *Ceramic Tiles. Definition, Classification, Characteristics, Assessment and Verification of Constancy of Performance and Marking*, BS EN, London, UK, 2016.
- [92] R. Sokolar and L. Vodova, "The effect of fluidized fly ash on the properties of dry pressed ceramic tiles based on fly ash-clay body," *Ceramics International*, vol. 37, no. 7, pp. 2879–2885, 2011.
- [93] H. Wang, M. Zhu, Y. Sun, R. Ji, L. Liu, and X. Wang, "Synthesis of a ceramic tile base based on high-alumina fly ash," *Construction and Building Materials*, vol. 155, pp. 930–938, 2017.
- [94] H. Wang, Y. Sun, L. Liu, R. Ji, and X. Wang, "Integrated utilization of fly ash and waste glass for synthesis of foam/dense bi-layered insulation ceramic tile," *Energy and Buildings*, vol. 168, pp. 67–75, 2018.
- [95] P. Chindaprasirt, A. Srisuwan, C. Saengthong, S. Lawanwadeekul, and N. Phonphuak, "Synergistic effect of fly ash and glass cullet additive on properties of fire clay bricks," *Journal of Building Engineering*, vol. 44, Article ID 102942, 2021.
- [96] A. Christogerou, T. Kavas, Y. Pontikes, S. Koyas, Y. Tabak, and G. Angelopoulos, "Use of boron wastes in the production of heavy clay ceramics," *Ceramics International*, vol. 35, no. 1, pp. 447–452, 2009.
- [97] P. N. Lemougna, J. Yliniemi, H. Nguyen et al., "Utilisation of glass wool waste and mine tailings in high performance building ceramics," *Journal of Building Engineering*, vol. 31, Article ID 101383, 2020.
- [98] N. Kockal, "Utilisation of different types of coal fly ash in the production of ceramic tiles," *Boletín de la Sociedad Española de Cerámica y Vidrio*, vol. 51, 2012.
- [99] ISO, *ISO 10545-15:2021 Ceramic tiles - Part 15: Determination Of Lead And Cadmium Given Off By Tiles*, ISO, Geneva, Switzerland, 2021.
- [100] R. Chen, Y. Li, R. Xiang, and S. Li, "Effect of particle size of fly ash on the properties of lightweight insulation materials," *Construction and Building Materials*, vol. 123, pp. 120–126, 2016.
- [101] Y.-L. Wei, S.-H. Cheng, and G.-W. Ko, "Effect of waste glass addition on lightweight aggregates prepared from F-class coal fly ash," *Construction and Building Materials*, vol. 112, pp. 773–782, 2016.
- [102] N. U. Kockal and T. Ozturan, "Optimization of properties of fly ash aggregates for high-strength lightweight concrete production," *Materials & Design*, vol. 32, no. 6, pp. 3586–3593, 2011.
- [103] T. Y. Lo, H. Cui, S. A. Memon, and T. Noguchi, "Manufacturing of sintered lightweight aggregate using high-carbon fly ash and its effect on the mechanical properties and microstructure of concrete," *Journal of Cleaner Production*, vol. 112, pp. 753–762, 2016.

- [104] K. Ramamurthy and K. Harikrishnan, "Influence of binders on properties of sintered fly ash aggregate," *Cement and Concrete Composites*, vol. 28, no. 1, pp. 33–38, 2006.
- [105] Y.-L. Wei, S.-H. Cheng, W.-J. Chen, Y.-H. Lu, K. Chen, and P.-C. Wu, "Influence of various sodium salt species on formation mechanism of lightweight aggregates made from coal fly ash-based material," *Construction and Building Materials*, vol. 239, Article ID 117890, 2020.
- [106] J. G. Otero, F. Blanco, M. Garcia, and J. Ayala, "Manufacture of refractory insulating bricks using fly ash and clay," *British Ceramic Transactions*, vol. 103, no. 4, pp. 181–186, 2004.
- [107] J. S. Jung, H. C. Park, and R. Stevens, "Mullite ceramics derived from coal fly ash," *Journal of Materials Science Letters*, vol. 20, no. 12, pp. 1089–1091, 2001.
- [108] Y. Dong, S. Hampshire, J.-e. Zhou, Z. Ji, J. Wang, and G. Meng, "Sintering and characterization of flyash-based mullite with MgO addition," *Journal of the European Ceramic Society*, vol. 31, no. 5, pp. 687–695, 2011.
- [109] C. T. Foo, M. A. M. Salleh, K. K. Ying, and K. A. Matori, "Mineralogy and thermal expansion study of mullite-based ceramics synthesized from coal fly ash and aluminum dross industrial wastes," *Ceramics International*, vol. 45, no. 6, pp. 7488–7494, 2019.
- [110] B. Ma, C. Su, X. Ren et al., "Preparation and properties of porous mullite ceramics with high-closed porosity and high strength from fly ash via reaction synthesis process," *Journal of Alloys and Compounds*, vol. 803, pp. 981–991, 2019.
- [111] Y. Dong, S. Hampshire, Je Zhou et al., "Recycling of fly ash for preparing porous mullite membrane supports with titania addition," *Journal of Hazardous Materials*, vol. 180, no. 1–3, pp. 173–180, 2010.
- [112] J. H. Lee, H. J. Choi, S. Y. Yoon, B. K. Kim, and H. C. Park, "Porous mullite ceramics derived from coal fly ash using a freeze-gel casting/polymer sponge technique," *Journal of Porous Materials*, vol. 20, no. 1, pp. 219–226, 2013.
- [113] A. Miyashiro, T. Iiyama, T. Miyashiro, and M. Yamasaki, "The polymorphism of cordierite and indialite," *American Journal of Science*, vol. 253, no. 4, pp. 185–208, 1955.
- [114] G. Gibbs, "The polymorphism of cordierite I: the crystal structure of low cordierite," *American Mineralogist: Journal of Earth and Planetary Materials*, vol. 51, no. 7, pp. 1068–1087, 1966.
- [115] E. P. Meagher and G. V. Gibbs, "The polymorphism of cordierite: II. The crystal structure of indialite," *The Canadian Mineralogist*, vol. 15, pp. 43–49, 1977.
- [116] T. A. Bulbak and S. V. Shvedenkova, "Solid solutions of (Mg, Fe<sup>2+</sup>)-cordierite: synthesis, water content, and magnetic properties," *Geochemistry International*, vol. 49, no. 4, pp. 391–406, 2011.
- [117] W. Shi, B. Liu, X. Deng, J. Li, and Y. Yang, "In-situ synthesis and properties of cordierite-bonded porous SiC membrane supports using diatomite as silicon source," *Journal of the European Ceramic Society*, vol. 36, no. 14, pp. 3465–3472, 2016.
- [118] R. Goren, C. Ozgur, and H. Gocmez, "The preparation of cordierite from talc, fly ash, fused silica and alumina mixtures," *Ceramics International*, vol. 32, no. 1, pp. 53–56, 2006.
- [119] M. Senthil Kumar, M. Vanmathi, G. Senguttuvan, R. V. Mangalaraja, and G. Sakthivel, "Fly ash constituent-silica and alumina role in the synthesis and characterization of cordierite based ceramics," *Silicon*, vol. 11, no. 6, pp. 2599–2611, 2019.
- [120] S. Wang, H. Wang, Z. Chen, R. Ji, L. Liu, and X. Wang, "Fabrication and characterization of porous cordierite ceramics prepared from fly ash and natural minerals," *Ceramics International*, vol. 45, no. 15, pp. 18306–18314, 2019.
- [121] J. Liu, Y. Dong, X. Dong et al., "Feasible recycling of industrial waste coal fly ash for preparation of anorthite-cordierite based porous ceramic membrane supports with addition of dolomite," *Journal of the European Ceramic Society*, vol. 36, no. 4, pp. 1059–1071, 2016.
- [122] M. Zhu, R. Ji, Z. Li, H. Wang, L. Liu, and Z. Zhang, "Preparation of glass ceramic foams for thermal insulation applications from coal fly ash and waste glass," *Construction and Building Materials*, vol. 112, pp. 398–405, 2016.
- [123] Q. Ma, Q. Wang, L. Luo, and C. Fan, "Preparation of high strength and low-cost glass ceramic foams with extremely high coal fly ash content IOP Conference Series: Materials Science and Engineering," *IOP Conference Series: Materials Science and Engineering*, vol. 397, no. 1, Article ID 012071, 2018.
- [124] H. R. Fernandes, D. U. Tulyaganov, and J. M. F. Ferreira, "Production and characterisation of glass ceramic foams from recycled raw materials," *Advances in Applied Ceramics*, vol. 108, no. 1, pp. 9–13, 2009.
- [125] S.-D. Yoon, J.-U. Lee, J.-H. Lee, Y.-H. Yun, and W.-J. Yoon, "Characterization of wollastonite glass-ceramics made from waste glass and coal fly ash," *Journal of Materials Science & Technology*, vol. 29, no. 2, pp. 149–153, 2013.
- [126] L. Barbieri, I. Lancellotti, T. Manfredini, I. Queralt, J. Rincon, and M. Romero, "Design, obtainment and properties of glasses and glass-ceramics from coal fly ash," *Fuel*, vol. 78, no. 2, pp. 271–276, 1999.
- [127] S. Wang and K. Liang, "High infrared radiance glass-ceramics obtained from fly ash and titanium slag," *Chemosphere*, vol. 69, no. 11, pp. 1798–1801, 2007.
- [128] H. Chen, H. Lin, P. Zhang et al., "Immobilisation of heavy metals in hazardous waste incineration residue using SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-Fe<sub>2</sub>O<sub>3</sub>-CaO glass-ceramic," *Ceramics International*, vol. 47, no. 6, pp. 8468–8477, 2021.
- [129] L. Barbieri, A. M. Ferrari, I. Lancellotti, C. Leonelli, J. M. Rincón, and M. Romero, "Crystallization of (Na<sub>2</sub>O-MgO)-CaO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> glassy systems formulated from waste products," *Journal of the American Ceramic Society*, vol. 83, no. 10, pp. 2515–2520, 2004.
- [130] M. Komljenović, L. Petrašinović-Stojkanović, Z. Baščarević, N. Jovanović, and A. Rosić, "Fly ash as the potential raw mixture component for Portland cement clinker synthesis," *Journal of Thermal Analysis and Calorimetry*, vol. 96, no. 2, pp. 363–368, 2009.
- [131] H. Hassanien Mohamed Darweesh, "A low temperature manufactured Portland cement clinker from pulverized waste of fly ash," *International Journal of Materials Science and Applications*, vol. 9, no. 2, p. 34, 2020.
- [132] J. I. Bhatti, J. Gajda, and F. Miller, "Commercial demonstration of high-carbon fly ash technology in cement manufacturing," *International Journal of Materials Science and Applications*, 2003.
- [133] J. Bhatti, "Use of high carbon fly ash as a component of raw mix for cement manufacture," 1998, <https://www.epri.com/research/products/TR-110808>.
- [134] A. Guerrero, S. Goñi, I. Campillo, and A. Moragues, "Belite cement clinker from coal fly ash of high Ca content. Optimization of synthesis parameters," *Environmental Science and Technology*, vol. 38, no. 11, pp. 3209–3213, 2004.
- [135] M. Mirković, L. Kljajević, S. Nenadović et al., "Fly ash as a raw material for low-carbon cement clinkers and its

- radiological properties,” *Journal of Radioanalytical and Nuclear Chemistry*, vol. 328, no. 3, pp. 1391–1398, 2021.
- [136] D. E. Macphee and E. E. Lachowski, “3. Cement components and their phase relations,” in *Lea’s Chemistry of Cement and Concrete*, P. C. Hewlett, Ed., Elsevier Science & Technology Books, Amsterdam, Netherlands, 2004.
- [137] ISO, *ISO 4531: 2018 Vitreous And Porcelain Enamels - Release From Enamelled Articles In Contact With Food - Methods Of Test And Limits*, ISO, Geneva, Switzerland, 2018.
- [138] ISO, *ISO 6486-1:2019 Ceramic Ware, Glass Ceramic Ware And Glass Dinnerware In Contact With Food - Release Of Lead And Cadmium - Part 1: Test Method*, ISO, Geneva, Switzerland, 2019.
- [139] EC Regulation, “1935/2004 of the European Parliament and of the Council of 27 October 2004 on materials and articles intended to come into contact with food and repealing Directives 80/590/EEC and 89/109/EEC,” *Official Journal of the European Union L*, vol. 338, no. 4, 2004.
- [140] A. Ríos, M. González, C. Montes, J. Vásquez, and J. Arellano, “Assessing the effect of fly ash with a high  $\text{SO}_3$  content in hybrid alkaline fly ash pastes (HAFAPs),” *Construction and Building Materials*, vol. 238, Article ID 117776, 2020.