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Review of Glass-Ceramics Containing Fly Ash

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ABSTRACT

The combustion of coal produces fly ash, a waste product that contains valuable amounts of silica, alumina, and other oxides. The disposal of fly ash poses economic and environmental challenges due to the toxicity and magnitude of ash produced. To reduce environmental hazards and minimize costs associated with disposal, the various applications of fly ash have been extensively studied. Alternatives to disposal are numerous and include the utilization of fly ash in road construction, cement production, agriculture, and waste water treatment. Due to expanding coal markets in developing countries, global fly ash production is projected to increase from 400 million tons to 1 billion tons by 2030. This increase will require new utilization methods to mitigate environmental concerns associated with fly ash disposal. Fly ash use as a precursor material in glass ceramics is an under-utilized but promising technique. Glass ceramics are specialized materials with a wide range of possible applications, including armor, biomaterials, thermal shock resistant bakeware, and tiling. The most common glass ceramics are made from aluminosilicates and transition metal oxide nucleating agents, which are the main components of fly ash. Using fly ash in the manufacture of glass ceramics instead of new materials creates less expensive glass ceramics with lower environmental impacts. This paper is a report on fly ash, glass ceramics, and fly ash-derived glass ceramics written with the purpose of understanding the potential for commercial fly ash glass ceramics.

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1. INTRODUCTION

Human activities have caused between 0.8 to 1.2 °C worth of global warming. This value is likely to increase to 1.5 °C before 2050 without significant changes to resource consumption. A 2018 report published by the Intergovernmental Panel on Climate Change (IPCC) indicated that coal combustion needs to be reduced to just 0 – 2% of the global energy portfolio in order to prevent the 1.5 °C increase from occurring [1]. However, in the time since the report was published, global coal consumption has continued to increase and passed 8 billion tons consumed in one year for the first time ever in 2022 [2]. This is due partly to the increasing coal markets in China and India, though 28% of the global energy market is coal-based [3].

Coal is an essential energy source that will continue to have a large presence in the next few decades. Pre-treatment processes to remove ash content from coal and emission control technologies in power stations can reduce the impacts of coal combustion [4]. However, the utilization of coal combustion residues (CCRs) in various applications mitigates health and environmental risks associated with disposal, reduces reliance on new materials, and furthers the responsible consumption of coal. Of the coal combustion residues, fly ash represents the largest market. The silica and alumina content of fly ash makes it valuable in many industries, including ceramics [5].

Glass ceramics emerged in the 1950s and have unique characteristics, like high strength and thermal shock resistance. They are most frequently manufactured by nucleating an aluminosilicate ternary glass to produce an end product that has both a crystal and glass phase [6]. Fly ash is comprised of aluminosilicates and transition metal oxides that can act as nucleating agents [7]. Therefore, fly ash is a material of interest for the production of glass ceramics. Fly ash is also less expensive than standard

high-grade reagents used in glass ceramic production, making it an economically feasible substitute [5]. Fly ash-derived glass ceramics can both improve the sustainability of coal consumption through waste valorization and lower the environmental impact of glass ceramics by reducing the need to process virgin materials.

2. FLY ASH

2.1 Introduction: Fly Ash

Coal combustion residuals (CCRs) are one of the world's largest sources of industrial waste. They form during the combustion of coal and include fly ash, a fine powder that rises upwards during combustion [7, 8, 9]. The US produces around 100 million metric tons of CCRs annually, of which fly ash represents around 60% [4, 8].

Fly ash is disposed of in landfills or ponds. As of 2015, the US had over 1,000 active fly ash disposal facilities [8]. The disposal of fly ash consumes land and poses environmental and health risks. Rather than being disposed, fly ash can instead be collected and utilized for structural fill, soil reclamation, wastewater treatment, cement and ceramic production, and other applications [10, 11]. The current global average utilization rate is 25%, though it varies between countries [9, 12, 13].

In 2021, the global fly ash market was valued at \$12 billion USD. It is projected to become a nearly \$17 billion industry by 2028. As global coal consumption increases to keep up with the growing global energy demand, it will become a necessity to increase the utilization of fly ash to maximize resources [12]. Fly ash utilization is a significant and economically viable field for sustainable progress and responsible consumption. Restrictions on landfilling in the US and policies supporting sustainable

usage of waste in Asia have resulted in an increase of motivation to increase fly ash utilization and commercialization [5, 12]. Utilization of fly ash provides an economical and sustainable approach to reducing reliance on virgin resources. The usage of fly ash also mitigates health and environmental risks associated with ash disposal and frees up land for other uses [8].

2.2 Composition

The composition of fly ash depends on plant operational factors and coal characteristics including rank, mineral composition, and calorific value [4, 7, 9, 14]. As such, the fly ash composition is highly variable. Three hundred sixteen minerals and one hundred eighty-eight mineral groups have been identified in fly ashes, though it is likely that this list is not complete [4].

Fly ash is composed primarily of fine spherical particles (both solid spheres and cenospheres) and unburnt carbon [11, 15]. The color of fly ash can range from tan to red to brown depending on the amount of iron and unburnt carbon present [9, 10]. It can be either acidic or basic and consists mostly of silica in addition to alumina, iron oxide, other oxides, and varying amounts of unburned carbon [5]. It contains both amorphous and crystal aluminosilicates. Major phases observed in fly ash include mullite, hematite, and quartz [7, 9, 10].

Table 1 - Properties of Fly Ash

	Value	Description
<i>Particle size:</i>	10 - 100 microns	Small
<i>Bulk density:</i>	1.01 - 1.43 g/cm ³	Low
<i>Surface area:</i>	300 - 500 m ² /kg	High
<i>Plastic Index:</i>	0 - 4	Not plastic to slightly plastic
<i>Specific Gravity:</i>	1.6 - 3.1 g/cm ³	Low
<i>LOI (unburnt carbon):</i>	1 - 20%	Depends on furnace efficiency

The geological conditions and time of formation of each coal creates varying ranks of coal with different qualities. The high rank coals, bituminous and anthracite, have higher carbon compositions and higher heating values than the low rank coals, lignite and sub-bituminous [3, 15]. Similarly, differences in geological conditions creates coals with different ash content based on the amount of inorganic mineral matter present [7]. Coal ash generally comprises between 5 – 20 wt % of feed coals, of which fly ash is 85 – 95 wt % [5]. However, some coals in India can have an ash content up to 45% [9, 10].

Of the components in fly ash, leachable trace elements pose the greatest environmental hazards [3, 5]. Due to the high surface area of fly ash, the trace elements are deposited on the surface of ash particles during condensation following combustion. They appear at levels 4 – 10 times higher in fly ash than in parent coal [11]. Trace elements include arsenic, cadmium, and chromium [16].

2.21 Class C vs Class F

The American Society for Testing and Materials (ASTM) classifies fly ash based on its calcium oxide content. ‘Class C’ fly ashes are cementitious and have 15% or more calcium oxide while ‘Class F’ ashes are pozzolanic and contain less than 5% calcium oxide [5]. Class C results from the combustion of low rank lignite and sub-bituminous coals, Class F originates from the burning of high rank coals [9, 12].

Table 2 - Differences Between Class C and Class F

	Class C	Class F
<i>Calcium oxide percent:</i>	> 15 %	< 5%
<i>Percent of fly ash production:</i>	45.50%	54.50%
<i>Properties:</i>	Cementitious	Pozzolanic
<i>Origin coals:</i>	Low rank	High rank
<i>Uses:</i>	Road construction, land reclamation, mine backfilling	Concrete production

Pozzolans are siliceous and often aluminous materials that react with water and calcium oxide at ambient temperatures to form cement [14]. Pozzolanic Class F fly ash can be used as a cement replacement in concrete to improve workability and durability. The classification of Class C and Class F fly ashes has been criticized for being too broad, as fly ash is remarkably inconsistent and different particle sizes, mineral phases, and morphology can have an effect on the cementitious and pozzolanic capabilities of ashes that cannot be described by a distinction based on calcium oxide content alone [4].

2.3 Disposal

Fly ash is disposed of in either landfills as a powder or in lagoons as a slurry. Disposal facilities are usually located on-site and adjacent to the source power station [16]. Poor disposal techniques can result in potential harm to the environment and health risks for individuals living near disposal sites. Fly ash particulate matter suspended in air poses inhalation dangers and can induce ophthalmic and respiratory irritation. It also has a potential to leach into groundwater or drinking water sources and create heavy metal contamination [5, 8, 10, 15].

In 1997, improper disposal at the Seyitomer power plant in Turkey resulted in a large movement of ash that covered a 1,950,000 m² area of agricultural land. The ash also entered a stream used to irrigate these fields. The ash spill resulted in copper and lead pollution in the soil. The levels of chromium and cobalt were elevated in the stream but were still within regulatory limits recommended for irrigation waters [17].

In December 2008, a coal ash dredge cell released over 4 million cubic meters of coal ash at the Tennessee Valley Authority (TVA) Kingston coal-fired power plant in Tennessee. The ash was released into tributaries that led to a drinking water source downstream. Sampling conducted after the spill indicated that mercury levels in the soil and fish exceeded the threshold standards [18]. Following this failure, the US EPA launched the Coal Ash Surface Impoundment Integrity Assessment Program, a holistic evaluation of the condition and safety of the nation's fly ash disposal facilities. The evaluation revealed that over 150 sites were in poor condition as of 2009. To reduce risks and prevent future failures, the EPA subsequently released new requirements for landfills that involve mandatory groundwater monitoring, certain liner specifications, required closing of high hazard ponds, and restrictions on possible locations that new sites could be built. Dust mitigation techniques, like water

spraying, construction of covers and wind barriers, and halting release during wind events, prevent possible air quality degradation [8].

2.4 Utilization

2.41 Construction Uses

In the US, 60% of the portion of utilized fly ash is used by the construction industries for bricks, soil stabilization, and concrete production [16]. Bricks made from fly ash do not require binders or additives and possess greater strength than bricks made from conventional clays [12]. Fly ash is often used as a soil stabilizer, filler, and foundation material because it decreases soil plasticity and does not swell [9, 10].

The concrete industry is the most established fly ash consumer. For over 50 years, pozzolanic Class F fly ash has been used as an economically favorable cement replacement in concrete. When added to Portland cement, fly ash creates a resulting concrete that is stronger, more durable, denser, and more resistant to weathering than conventional concrete [12]. Fly ash tends to be added to concrete at values between 15 – 35 wt %, though up to 70 wt % is possible for pavements [5]. Additionally, heavy metals in fly ash are immobilized in the cement matrix and have a negligible leachability in end products [9].

The addition of fly ash not only improves the qualities of bricks and concrete, but it also lowers the carbon footprint of these products relative to production from conventional materials [12].

2.42 Agricultural Applications

When added to soil, fly ash can increase porosity, water retention, aeration, percolation, and available nutrients [10]. These characteristics can be attributed to the low bulk density of ash. Alkaline fly ash can also be used as a substitute for lime to increase and buffer soil pH. Usage of fly ash for this application is more sustainable, as the distribution of lime to fields releases CO₂ over time. Fly ash can also be used as an insecticide due to its abrasive quality [5, 15].

Fly ash contains both macro and micro nutrients vital to plant growth, including potassium, phosphorous, calcium, magnesium, sulfur, iron, manganese, and zinc [15]. Fly ash has been used to increase crop yield in various applications, including in Korean paddy fields, which conventionally require the addition of siliceous fertilizers. A study performed by Lee et al. [19] applied raw fly ash in quantities of 0, 40, 80, and 120 megagrams per hectare to the wetland soils. The addition of fly ash increased soil pH and the amount of available silicon and phosphorous. The field that received treatment of 80 Mg/ha resulted in the highest rice yield [19].

However, though fly ash does contain nutrients, it also can contain heavy metals like selenium, molybdenum, arsenic, cobalt, and others. These can negatively influence the growth of plants that are not tolerant to heavy metals [5, 15]. While the paddy fields did not show evidence of heavy metal uptake into the plants, increased presence of heavy metals has been observed in other studies [19].

The Yamuna River receives discharges of supernatant water from the lagoon facilities of two coal-burning power stations near Delhi, India. In a several year-long study performed on the soil and vegetation near the discharge points, water hyacinth populations along the river declined from being common and in good health (except near the discharge points) to being extinct from the surveyed area. Prior to its disappearance, samples of hyacinth revealed elevated levels of cadmium, cobalt, copper, nickel, and zinc. It is likely that the metal toxicity affected the health of the hyacinth population. The study did not analyze the health or nutritional value of the floodplain soils, though other plants had elevated levels of heavy metals and maintained good health [16].

2.43 Other Functions

Fly ash is valuable in the ceramics industry for its small particle size and high content of silica, alumina, calcium oxide, and iron oxide. Glass, glass ceramics, and ceramics have been synthesized from fly ash [20]. Fly ash has been used as an inexpensive feedstock for lithium aluminosilicate glass ceramics in quantities up to 68 wt % that possess qualities that exceed that of the conventional feedstock. Fly ashes high in unburned carbon has been successfully used as an adsorption agent to remove metal ions from contaminated water with a 95% removal rate [5].

2.44 Barriers to Utilization

The utilization of fly ash is affected by economic, technical, and environmental barriers. Transportation and storage of fly ash can be expensive due to the small particle size that poses environmental concerns [11, 12]. Wet storage of fly ash degrades the natural pozzolanic properties of fly ash, preventing it from being salvaged for the concrete industry. Additionally,

the inconsistent composition of fly ashes, even amongst ashes derived from the same mine or produced at the same power station, can limit the utilization for specific products. Concrete and cement production has a high tolerance to subtle changes in composition per batch, but products that require more specific control over composition, like glass ceramics, are less tolerant to these variances [13]. Advancements in research will lower technical and economic barriers through the introduction of new commercial opportunities [11].

2.5 Future of Fly Ash

Fly ash is an abundant resource that will continue to be produced in excess until Asia reduces their dependency on coal power. The share of coal in the global energy portfolio is decreasing despite coal consumption increasing due to expansion of coal markets in China and India to keep up with their growing energy demands. In 2022, global coal consumption surpassed 8 billion metric tonnes for the first time in history. Values slightly over 8 billion are expected to be observed through 2025 barring any unforeseen dramatic political or economic changes [2]. As the global energy demand continues to increase, fly ash production and utilization will increase proportionately [12].

Expansion of commercial utilization of fly ash will provide opportunities to re-use ash rather than disposing of it. Utilization has expanded rapidly in India, growing from less than 10% in 1996 to 60% in 2012 [9]. The US presently utilizes around 50% of produced fly ash, EU uses 90%, and China utilizes between 30 – 70% [5]. The future of fly ash is a future where all countries have a 100% utilization rate.

3. GLASS CERAMICS

3.1 Introduction: Glass Ceramics

Glass ceramics were first synthesized in 1953 when a researcher at Corning Glass Works overheated a furnace to 900 °C while attempting to anneal lithium disilicate glass precipitated with silver. Expecting to find a molten puddle of glass, the researcher opened the furnace to instead reveal a nondeformed material that was resistant to fracturing when dropped on the floor [6].

Due to their exemplary strength and low coefficient of thermal expansion (CTE), commercial glass ceramics were introduced in the late 1950s as radomes to protect radar equipment in the nosecones of spaceships. While glass ceramics continue to be used in nosecones of high-performance aircraft, commercial applications have significantly expanded in the past half decade [6]. However, due to their large production costs, commercial glass ceramic products are most successful in markets where high performance is prioritized over more economical options [21].

3.2 Production

Broadly, glass ceramics are microcrystalline solids produced by the controlled devitrification, or crystallization, of suitable glasses using a nucleating agent [22]. They are composed of a residual glassy phase and one or two crystalline phases [24]. Crystallinity generally occurs in quantities 30 – 70 vol. % but may be as low as 1 ppm and as high as 99.5 vol. % [6]. The interaction between crystal and glassy phases is what results in the unique physical and chemical characteristics observed in glass ceramics [23].

Glass ceramics are traditionally formed in two stages. The process begins with glass formation and melting. Following cooling, the glass is subjected to heat treatment above the transition temperature to facilitate controlled crystallization [21, 22, 23]. Crystallization is usually performed in a kiln but can also be induced by laser, UV radiation, or electron beam technologies and is facilitated by nucleating agents that form active sites for crystals to grow [24].

3.3 Properties

Glass ceramics have a low coefficient of thermal expansion and are resistant to thermal shock. They tend to be non-porous and have high strength and durability. The average fracture strength is between 100 – 250 MPa and average toughness factor is around 1 – 2.5 MPa m^{1/2}, a significant improvement over normal glass [21]. Additionally, unlike single crystals, glass ceramics can be formed in complex shapes using glass shaping techniques like pressing and blowing [6, 22, 25].

Glass ceramics can be either opaque or transparent. To exhibit transparency, a glass ceramic needs to have a crystal size smaller than the wavelength of visible light (200 nm) and possess low birefringence between the refractive indexes of the glass and crystal phases [6].

The precursor composition determines physical and chemical properties of the material while the formed microstructure determines mechanical and optical qualities [22, 23]. Therefore, materials can be tailored to different specifications by altering the composition and microstructure [25]. For example, the negative coefficient of thermal expansion contrasts with the positive coefficient of thermal expansion to effectively cancel each other out and produce a desirably low or zero CTE. Adjusting the proportions of glassy and crystal phases can produce a range of CTEs for different applications [6, 21].

3.4 Other Manufacturing Processes

In an alternative manufacturing process, glass ceramics are synthesized from the sintering of glass particles that have been compacted into forms. In this procedure, nucleating agents are not needed because the relict surfaces of the glass grains act as nucleation sites. This method allows for powder processing, though can result in increased porosity. Hot-pressing and other densification techniques implemented prior to crystallization can reduce porosity [6, 23, 25].

Neoparium, a pore-free calcium silicate glass ceramic aesthetically reminiscent to marble or granite, is produced by Nippon Electric Glass using the sintering method for use as exterior and interior walls and counter tops [22, 23]. Other uses of sinter-produced glass ceramics include as electronic packaging and bioactive materials. The sintering method has been used to create low-cost glass ceramics from recycled materials including incineration ashes, biomass ashes, blast furnace slag, and steel slag [6].

In addition to sintering, glass ceramics can also be produced using a sol-gel preparation method, in which gel-derived glasses are used as a precursor to lower the melting temperature and speed of glasses that would otherwise be too inefficient to produce glass ceramics from. This is used to produce celsian phase crystals in the $\text{BaO-Al}_2\text{O}_3\text{-2SiO}_2$ (BAS_2) system [25].

3.5 Silica-Containing Glass Ceramics

Silica-containing glass ceramics, such as silicates, aluminosilicates, and fluosilicates, are the most commercially successful glass ceramics [22].

3.51 Silicates and Fluosilicates

Lithium silicates are most commonly nucleated with either P_2O_5 or colloidal silver. They have a wide range of applications, including as hermetic seals, connectors, cellular display screens, lens arrays, and plates for ink jet printing. Calcium silicate glass ceramics are produced through sintering and are used as wall and floor tile. Fluosilicates are used to produce tableware and mugs for Corning's Corelle line of products, magnetic memory disc substrates, seismograph bobbins, and electrical insulators [22]. Lithium disilicate is more durable than most other glass ceramics, having a strength between 350 – 400 MPa and toughness between 2.3 – 2.9 MPa m^{1/2}. It is used for both ballistic armor and in dental applications as tooth replacement [6].

3.52 Aluminosilicates

As with other glass ceramics, aluminosilicate-derived glass ceramics are resistant to thermal shock and have a low coefficient of thermal expansion (CTE). The highest performance glass ceramics originate from the $Li_2O-Al_2O_3-SiO_2$ (LAS) ternary system [6, 22]. Other aluminosilicate ternaries, such as $MgO-Al_2O_3-SiO_2$ (MAS), $ZnO-Al_2O_3-SiO_2$ (ZAS), and $BaO-Al_2O_3-2SiO_2$ (BAS₂), produce glass ceramics significant in research and commercial applications [25]. The MAS system includes the cordierite phase, which is distinguished by its excellent dielectric properties [22].

3.53 LAS System

The LAS ternary produces ultrafine grain glass ceramics with the lowest thermal expansion coefficient. Firing agents As_2O_5 and SnO_2 and various oxides may be added in different compositions to finetune the end qualities [25]. LAS glass ceramics are most commonly nucleated with either ZrO_2 or TiO_2 [22]. The main crystalline phases are either a β -quartz or β -spodumene solid solution, though cordierite may also be desired [6]. Reinforcing LAS glass ceramics with silicon carbide fibers can increase the strength beyond 800 MPa [25].

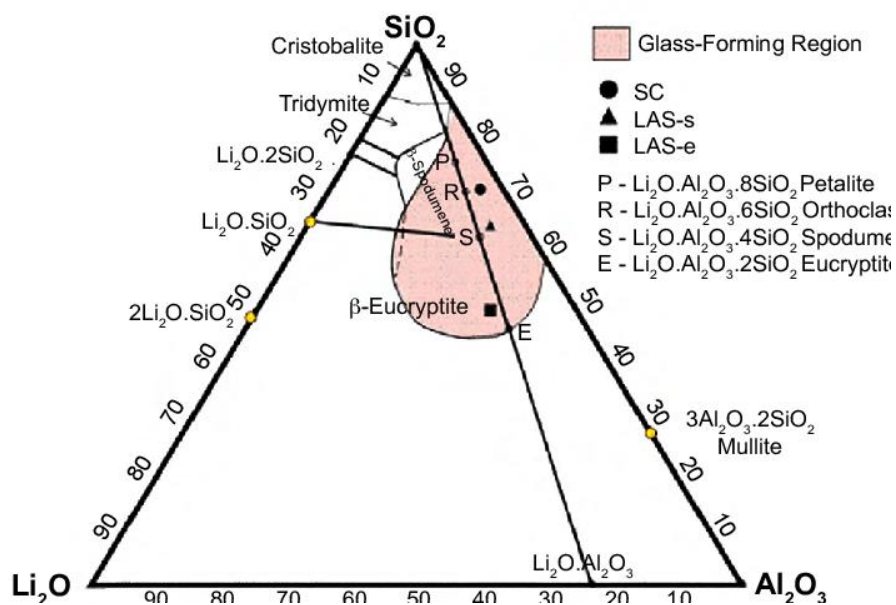


Figure 1 - LAS Ternary System

β -Quartz Solid Solution

Glass ceramics precipitated with β -quartz solid solution crystals are the most common commercial glass ceramic [21, 22]. They are often transparent due to possessing a crystal size smaller than 50 nm and low birefringence. β -quartz-based glass ceramics have the most negative CTEs and are used in cookware, telescope mirrors, windows, and range tops [6, 22, 23, 25].

β -Spodumene Solid Solution

At formation temperatures above 900 °C, β -quartz breaks down into β -spodumene. The transformation between the two phases results in a grain size increase 5 – 10 times larger than β -quartz crystals. Therefore, β -spodumene solid solution glass ceramics are opaque [25]. They have a low CTE, though not as low as β -quartz, and are used as cookware and components in turbine engines [22].

Table 3 - Commercial Glass Ceramics wt % Compositions

<i>Product name:</i> <i>Manufacturer:</i> <i>Use:</i>	β-QUARTZ			β-SPODUMENE		CORDIERITE
	Vision Corning Transparent cookware	Zerodur Schott Telescope mirrors	Narumi Nippon Electric Rangetops	Corning Ware Corning Cookware	Cercor Corning Heat exchangers	9606 Corning Radomes
Crystal Phase Oxides						
SiO ₂	68.8	55.5	65.1	69.7	72.5	56.1
Al ₂ O ₃	19.2	25.3	22.6	17.8	22.5	19.8
Li ₂ O	2.7	3.7	4.2	2.8	5.0	-
MgO	1.8	1.0	0.5	2.6	-	14.7
ZnO	1.0	1.4	-	1.0	-	-
P ₂ O ₅	-	7.9	1.2	-	-	-
F	-	-	0.1	-	-	-
Glass Phase Oxides						
Na ₂ O	0.2	0.5	0.6	0.4	-	-
K ₂ O	0.1	-	0.3	0.2	-	-
BaO	0.8	-	-	-	-	-
CaO	-	-	-	-	-	0.1
Nucleant						
TiO ₂	-	-	-	3.7	-	8.9
ZrO ₂	-	-	-	0.1	-	-
Fining Agent						
As ₂ O ₃	0.8	0.5	1.1	0.6	-	0.3
Colorant Oxide						
Fe ₂ O ₃	0.1	0.03	0.03	0.6	-	0.1

3.6 Biomedical Applications

Some glass ceramics are significant in the medical industry due to their bioactivity and osteo-productivity [21]. In addition to being used for magnetic drug targeting, magnetic bioactive glass ceramics can also be used as a method of treating cancer via hyperthermia. Glass ceramics implanted near tumorous bone tissue and then subsequently exposed to an alternating magnetic field can reach appropriate temperatures to destroy cancer cells. When implanted, bioactive glass ceramics form a layer of the mineral phase of bone and teeth, hydroxycarbonate apatite. Upon doing so, the glass ceramic implant can bond to bone or teeth and even stimulate bone regeneration [26]. As of 2009, over 50,000 successful bone implants had been performed using Cerabone from the Nippon Electric Glass company [6].

3.7 Future of Glass Ceramics

As of 2010, 2,400 US patents, 1,500 European patents, and 2,700 Japanese patents relating to glass ceramics had been filed. Companies including Corning, IBM, and Nippon Electric Glass Company contribute significantly to the advancements in glass ceramics research and industry development [6].

The full potential of glass ceramics is not yet realized. In the 70 years since their creation, only a few dozen glass ceramics have reached the consumer market [6]. However, these products are lucrative and serve functions that would not be possible using other materials. The industrial and domestic applications of glass ceramics are projected to grow as glass ceramic research expands and lowers the production costs [25]. Using siliceous wastes, like fly ash, as a feedstock for glass ceramics could generate lower cost and more sustainable glass ceramics.

4. FLY ASH-DERIVED GLASS CERAMICS

4.1 Introduction: Fly Ash-Derived Glass Ceramics

The composition of fly ash aligns with aluminosilicate glass ternary systems, allowing it to be efficiently turned into glass ceramics. Transition metal oxides, usually added to glass ceramics as nucleating agents, occur naturally in fly ash. Glass formers silica and alumina are also present in high quantities in fly ash. Furthermore, fly ash has a small particle size that is easily mixed as a powder with other glass ceramic additives with very little pretreatment or size reduction required. Heavy metals present in fly ash are incorporated into the crystal matrix, where they are less likely to leach out. Fly ash is an ideal economical and sustainable material for substitution of expensive high-grade reagents using in standard glass ceramic production [28].

4.2 Production

The process of producing glass ceramics derived from fly ash is very similar to the process of producing standard glass ceramics and ceramics [28, 29]. Fly ash is first sieved to collect the desired sizes [31, 32]. The ash may be placed in a ball mill for further size reduction, though fly ash particles are sufficiently small for use as a glass ceramic precursor without crushing [33]. Magnetic separation is a necessary step for ashes high in iron, which forms phases with low melting points during sintering. These phases degrade the performance of the end product [34, 35]. Ashes with low iron contents do not require this step. If additives or nucleating agents are being mixed with the ash, a secondary milling is required for homogenization. The resultant powder is humidified and uniaxially compacted into green bodies using pressure up to 100 MPa [30, 31].

Calcination may be needed to liberate carbonates and avoid degassing [33, 34, 35]. Nucleation is performed around 650 – 850 °C for varying time based on nucleation characteristics [32]. The material is then transferred to a crucible, placed in an electric furnace, and subjected to heat treatment between 900 – 1500 °C for 2 – 4 hours [33, 35, 36]. During this process, the dominant phases in fly ash, usually mullite and quartz, are reduced as the aluminosilicate crystal phase becomes dominant [30, 31, 32, 34].

Following heat treatment, the products are either annealed between 500 - 600 °C for 1 – 2 hours or allowed to air cool [32, 36]. Quenching may also be used to return glass ceramics to an ambient temperature [31]. The resultant forms can be crushed for analysis via x-ray diffraction, used to determine phase assemblage; scanning electron microscopy (SEM), used to identify structures and grain size; and differential thermal analysis (DTA), used to determine transition, crystallization, and melting temperatures through controlled heating [30, 33, 35]. The glasses can also be crushed and reheated in the same process to ensure homogenization [36]. Variations in procedure depend on the composition of fly ash, need for additives, and desired end characteristics.

4.3 Aluminosilicate Ternary Examples

The composition of fly ash lends itself to utilization in aluminosilicate-based glass ceramics, which are also the highest performing and most commercialized glass ceramics.

Table 4 - Comparison of Fly Ash-Derived Glass Ceramics

	Erol et al	Yao et al	Lopez-Badillo et al	He et al
<i>Origin of Ash</i>	Cayirhan Thermal Power Plant, Turkey	Hangzhou Thermal Power Plant, China	Jose Lopez Portillo Power Plant, Mexico	Wuhan Power Plant, China
<i>Ternary of Glass Ceramic</i>	CAS	LAS	BAS	MAS
<i>Fly Ash Class</i>	C	F	F	F
<i>% of Fly Ash in Precursor</i>	100%	80%	43.59%	64 - 68%
<i>Additives</i>	None	LiOH-H ₂ O	BaCo ₃ , Al ₂ O ₃	MgO, Al ₂ O ₃
<i>Coefficient of Thermal Expansion</i>	Not reported	-2.16 x 10 ⁻⁶ /C	Not reported	1.43 x 10 ⁻⁶ /C
<i>Crystallization Temperature</i>	914 C	980 C	1200 C	1200 C
<i>Composition, wt %:</i>				
<i>SiO₂</i>	42.82	50.5	61.77	57.99
<i>CaO</i>	16.8	4.6	2.13	2.49
<i>MgO</i>	5.85	-	0.78	-
<i>Fe₂O₃</i>	7.01	5.3	5.56	4.33
<i>Al₂O₃</i>	13.36	35.9	28.58	28.98
<i>Na₂O</i>	5.06	-	0.24	0.27
<i>K₂O</i>	1.83	1.3	0.21	1.31
<i>SO₃</i>	6.47	-	-	-
<i>TiO₂</i>	-	1.3	0.77	1.42

4.31 CAS

Three different fly ashes with varying compositions were used by Barbieri et al. [33] to create glass ceramics in the CAS ternary. Fly ash content ranged from 10 – 50 wt % of a mixture with pulverized glass cullet. The cullet introduced sufficient levels of silica to produce glass and served as a glass former. When subjected to heat treatment, crystallization occurred rapidly at 800 °C in 30 minutes. The most successful glass ceramics had precursors composed of 40 – 50 wt % fly ash. Adding 10 – 20 wt % fly ash did not dilute the cullet enough, and the end product was too high in silica, limiting the workability [33]. Erol et al. [36] also produced CAS glass

ceramics, though they did not add nucleates or glass formers. They found that fly ash can crystallize around 900 °C without any additives [36].

4.32 BAS

Lopez-Badillo et al. [30] tested the mechanical activation of precursor materials when milled for five minutes, four hours, eight hours, and twelve hours to produce a glass ceramic in the BAS ternary. Fly ash (44%) was mixed with 12% alumina, a glass former, and 44% barium carbonate, a nucleating agent. Surface area of the powder increased with increasing mill time up to 8 hours, at which point it began to decrease due to agglomeration. Longer milling time resulted in increased reactivity.

BAS systems often experience conversion difficulties between hexacelsian to celsian, the preferred stage. Full conversion was not achieved by Lopez-Badillo et al. [30], though they did achieve higher conversion than similar methods using standard reagents. This is due to the presence of transition metal oxides in fly ash, which are linked to higher conversion rates [30].

4.33 MAS

Fly ash can be used to produce cordierite in the MAS system [32]. Alpha cordierite phase glass ceramics were produced using 64 and 68% fly ash combined with alumina and magnesium oxide. The resultant product had a compressive strength of 50 MPa and coefficient of thermal expansion (CTE) of $1.51 - 1.43 \times 10^{-6}/^{\circ}\text{C}$. Cordierite appeared at 1125 °C and became the dominant phase at 1200 °C, at which point quartz and mullite were nondetectable [31].

4.34 LAS

Dilithium dialuminium trisilicate was prepared by adding lithium hydroxide monohydrate to Class F coal fly ash in qualities of 10, 15, 20, and 25 wt %. Lithium oxide released from $\text{LiOH}\cdot\text{H}_2\text{O}$ interacts with silica and alumina to form the $\text{Li}_2\text{Al}_2\text{Si}_3\text{O}_{10}$ phase. TiO_2 and Al_2O_3 were present at high enough quantities in the fly ash to serve the purpose of a nucleating agent and glass former, respectively. A 20% $\text{LiOH}\cdot\text{H}_2\text{O}$ additive heated at 980 °C for an hour produced the best results, including a coefficient of thermal expansion smaller than commercial glass ceramics [34].

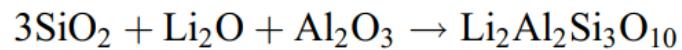
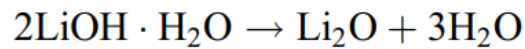


Figure 2 - Lithium Hydroxide Interactions

Kniess et al. [35] synthesized glass ceramics in the LAS ternary system using bottom ash, producing a glass ceramic with a coefficient of thermal expansion 18% smaller than that observed in commercial lithium glass ceramics. Crystallization was achieved in just ten minutes [35].

5. CONCLUSION

Coal fly ash is a valuable waste that can be exploited for its mineral content and properties. However, only a quarter of fly ash produced globally is utilized in beneficial applications. The remaining three-quarters is stored in disposal facilities that can cause heavy metal contamination of water systems or dust release. Fortunately, coal ash utilization is a rapidly expanding field. Utilization of fly ash reduces the amount of land consumed by fly ash disposal sites and reduces the reliance on virgin materials in industries where fly ash is a comparable substitute. The use of fly ash for cement production also decreases energy consumption by removing the need to process virgin feedstocks.

Similarly, the glass ceramics field is expanding as new applications are explored. The use of fly ash in aluminosilicate-based glass ceramics can produce materials that have better qualities than those being commercially produced. Fly ash naturally contains minerals that act as glass formers and nucleates, which are usually necessary additives to commercial precursors for glass ceramics. Fly ash is a convenient and inexpensive solution to finding a more sustainable feedstock for glass ceramics.

Fly ash is a notoriously inconsistent material that varies in composition and properties based on the geology of the source coal and combustion conditions. This inconsistency is one of the most substantial barriers to increasing fly ash utilization. Fly ash is most successful as a material in the concrete industry because concrete has a high tolerance for variance in composition. However, small differences in composition can dramatically affect glass ceramics. The literature reviewed in this report did not discuss potential issues related to commercialization of fly ash-derived glass ceramics. If fly ash feedstocks can be made more homogenous, commercial fly ash-derived glass ceramics could capitalize on the growth in both the fly and glass ceramics markets.

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Hannah Chop

Education

Master of Science, Energy and Mineral Engineering, May 2024

The Pennsylvania State University

Certificate: Accounting Foundations

Bachelor of Science, Environmental Systems Engineering, May 2023

The Pennsylvania State University

Schreyer Honors College

Work Experience

Environmental Engineering Intern, Turquoise Ridge Complex, Nevada Gold Mines

May – August 2022

- Performed an internal review of the reclamation bonds for two Tier 1 assets with a cumulative value of \$210 million and identified a potential bond reduction of \$680,000
- Analyzed data in permits and archives to identify current disturbances then calculated the variance between actual disturbed acreage and bonded disturbed acreage
- Utilized ArcGIS to build a map of 700+ disturbances over 10,000 acres for use by the environmental department in locating bond items
- Presented bond revision summary to site leadership and was 1 of 10 interns out of 100+ invited to present to corporate leadership team

Environmental Engineering Intern, Antelope Mine, Navajo Transitional Energy Company

May – August 2021

- Became familiar with mining regulations through support of the permit renewal process
- Plotted localized drainage basins using topographical maps to identify areas at risk of stormwater pollution and then proposed prevention methods
- Gained experiential knowledge of risk mitigation through conducting various inspections relating to SPCC and SWPPP
- Worked with a local company to develop permit boundary signage that met relevant MSHA requirements
- Conducted drone flights to collect information for surveying and soil health projects

Teaching Experience

Teaching Assistant, *Sampling and Monitoring of the Geo-Environment*, Penn State

January – May 2022

- Coordinated with other TA to grade and provide feedback on weekly assignments
- Held 4 hours of office hours per week to provide course support for 29 students

Teaching Assistant, *Exploring and Communicating Water Science*, Penn State

August – December 2020

- Led 10 students in weekly discussions pertaining to water science and sustainability rhetoric
- Graded 22 assignments and provided feedback within 36 hours of submittal

Leadership Experience

College of Earth and Mineral Sciences Student Council

University Park, PA

President

April 2022 – April 2023

- Oversee and coordinate council consisting of 100 members, 23 chairpersons, and 6 executives
- Lead weekly executive meetings and biweekly council body meetings
- Created equity-focused chairperson position
- Introduced new model to maximize the budget
- Orchestrated a gala event at a professional conference center attended by over 300 students, faculty, and staff

Vice President

April 2021 – April 2022

- Led programming to connect students to faculty/ staff, deans, and alumni
- Coordinated the College of Earth and Mineral Sciences 125th Anniversary Homecoming Events
- Organized the highest-fundraising Relay for Life team at Penn State

Secretary

April 2020 – April 2021

- Oversaw communications and 3 social media accounts
- Managed email list with 500+ recipients
- Developed a 'JEDI' executive position focused on justice, equity, diversity, and inclusion

Society for Mining, Metallurgy, and Exploration (SME): Penn State Chapter

University Park, PA

Vice-President

April 2022 – April 2023

- Support the coordination of events and speakers at weekly meetings

EMS Connect

University Park, PA

Assistant Director

April 2022 – April 2023

- Lead events to connect campus Penn State students to main campus Penn State students

Honors, Grants, and Awards

Laureate, College of Earth and Mineral Sciences (EMS) Academy for Global Experience

- Highest honors in program for fostering experiential and global competence

Ellen Steidle Achievement Award, 2022 & 2023

- Awarded for service to the College of Earth and Mineral Sciences

Student Engagement Network (SEN) Grant, 2021

- Awarded to study systems thinking in the Powder River Basin of Wyoming

Celebration of Undergraduate Engagement (CUE) Best Second-Year Digital Story, 2021: '*Analysis of the Diamond Industry*'

Celebration of Undergraduate Engagement (CUE) Best First-Year Digital Story, 2020: '*Canonsburg Water Quality Concerns*'

Dean's List: 4/6 eligible semesters

Service

Assessment of the Living, Learning, and Working Environment (ALLWE), Penn State August 2022 – April 2023

- Committee focused on increasing equity in the College of Earth and Mineral Sciences

EMS Ambassadors, Penn State April 2021 – April 2023

- Tour guide for the College of Earth and Mineral Sciences

Total Engagement with Earth and Mineral Sciences (TEEMS) Mentor, Penn State August 2022

- Mentored incoming college freshman at a four-day camp-based orientation

Theater Department Volunteer, Carnegie Science Center June 2018 – March 2020

- Volunteered 5 – 10 hrs/week performing science demonstrations and 30-minute scripted shows for visitors to the Carnegie Science Center in Pittsburgh

Outdoor School Counselor, Shaver's Creek Environmental Center October 2019

- Introduced 5th-graders to sustainability and environmental systems thinking at a week-long overnight camp

Conferences

SME MINEXCHANGE, 2023 Denver, Colorado

Awards Received: MMSA-SMEF Presidential Scholarship, Coal & Energy Division Scholarship

Presentation: *3D Printing with Fly Ash*

SME/PCMIA, 2022 Pittsburgh, Pennsylvania

National Association of State Land Reclamationists (NASLR), 2022 Virtual

Awards Received: NASLR Student Grant

Presentation: *3D Printing with Mine Tailings*

SME MINEXCHANGE, 2022 Salt Lake City, Utah

SME/PCMIA, 2021 Pittsburgh, Pennsylvania

Awards Received: Mike Kotch Memorial Scholarship Award, Claude A. Goode Memorial Scholarship Award

Student Engagement Network (SEN) Engagement Expo, 2021 University Park, Pennsylvania

Presentation: *Life in Wyoming's Powder River Basin: Exploring Social and Environmental Responsibility in the US's Largest Coal-Producing Region*