



## Review article

# Application of coal fly ash based ceramic membranes in wastewater treatment: A sustainable alternative to commercial materials

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## ABSTRACT

The continued increase in the global population has resulted in increased water demand for domestic, agricultural, and industrial purposes. These activities have led to the generation of high volumes of wastewater, which has an impact on water quality. Consequently, more practical solutions are needed to improve the current wastewater treatment systems. The use of improved ceramic membranes for wastewater treatment holds significant prospects for advancement in water treatment and sanitation. Hence, different studies have employed ceramic membranes in wastewater treatment and the search for low-cost and environmentally friendly starting materials has continued to engender research interests. This review focuses on the application of coal fly ash in membrane technology for wastewater treatment. The processes of membrane fabrication and the various limitations of the material. Several factors that influence the properties and performance of coal fly ash ceramic membranes in wastewater treatment are also presented. Some possible solutions to the limitations are also proposed, while cost analysis of coal fly ash-based membranes is explored to evaluate its potential for large-scale applications.

## 1. Introduction

The incessant rise in the world's population has resulted in higher water requirements for household, farming, and industrial needs. Consequently, the quality of water is affected due to the substantial generation of wastewater. However, this wastewater can be harnessed and repurposed for various applications [1–3]. Wastewater management is crucial for sustainable development [4]. According to a study by Obaideen et al. [5], wastewater treatment helps to successfully achieve 11 out of the 17 Sustainable Development Goals. Contaminants found in wastewater include pathogens, pesticides, dyes, heavy metals, pharmaceuticals, surfactants etc [6].

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These contaminants significantly affect both animal and human health as well as aquatic ecosystems even at low concentration levels. Significant efforts have been made to overcome the challenges of wastewater in recent years and these include policy and implementation as well as the development of treatment techniques [7,8]. These techniques are classified as either conventional methods or advanced treatment methods. Advanced treatment methods include constructed wetlands, bioelectrical systems, membrane filtration technologies, adsorption, advanced oxidation processes such as Fenton oxidation processes, photocatalysis, electrochemical oxidation, and enzymatic treatment [9–11]. Conventional methods include coagulation-flocculation and ozonation. However, due to financial and technological constraints, only a limited number of the numerous and diverse treatment techniques mentioned for wastewater treatment are utilized for domestic and economic purposes [12]. To tackle the unresolved problems of global water scarcity and water pollution, there is a need for water treatment solutions that possess multiple functions, can be scaled up, are strong and reliable, do not rely on chemicals, have a minimal environmental impact, and are energy-efficient [13]. Inadequate wastewater treatment methods have led to the recent discovery of a new class of pollutants called emerging contaminants in the environment, which have not yet been completely eradicated from the ecosystem [14]. As a result, more research is needed to enhance the current wastewater treatment systems so that it complies with the established regulations to totally remove pollutants.

The use of membrane technology in wastewater treatment has drawn attention from all around the world because it uses few chemicals, effectively removes contaminants, environmentally friendly, energy-efficient, and simple [15–17]. Membranes serve as selective barriers that permit some elements pass through while keeping others [15,18]. The utilization of membranes in wastewater treatment relies on the fundamental concept of allowing water molecules to pass through selectively while trapping contaminants, achieved by applying a driving force across the membrane. Membranes are primarily categorized into organic (polymeric) and inorganic (ceramic) types [16]. Organic membranes are those that are made up of nonporous polymeric materials, such as cellulose acetate, polyethersulfone, polyimide, polysulfone, polymethylpentene, polycarbonate, polydimethylsiloxane, and polyphenylene oxide [19,20]. Ceramic membranes are fine artificial porous membranes made from sintering inorganic materials, such as alumina, zirconia oxide, titanium, silicon oxide, silicon carbide, and silicon nitride or a combination of these materials under high temperatures [21,22]. At the moment, large-scale polymeric membrane systems are widely used over inorganic membranes due to good selectivity, ease of controlling pore size during formation, high flexibility, and cost-effectiveness [23]. Most applications of polymeric membranes are in gas separation and desalination due to high permeation and selectivity. They are, however, under-utilized in areas such as wastewater treatment due to low stability towards high temperatures, chemicals and harsh conditions [24]. Consequently, inorganic membranes have rapidly received global attention in being considered as one of the potential candidates to replace available polymeric membranes [25] due to several characteristics such as high stability under a variety of operating conditions such as temperature, and toxic chemicals, high eco-compatibility and scalability [26]. This review will focus exclusively on inorganic ceramic membranes.

Ceramic membranes can be classified as either symmetric or asymmetric based on their structural properties (Fig. 1) [27]. Symmetrical structured ceramic membranes have pores that are uniform in size across the membrane's thickness as shown in Fig. 1a. Conversely, asymmetric ceramic membranes feature three layers with pore patterns that are greatest on one surface and smallest near the other surface Fig. 1b [28,29]. The membrane relies on a macroporous support that offers robustness due to its larger pores. Additionally, an intermediate layer with smaller pores compared to the support layer safeguards against material intrusion during fabrication. Lastly, the selective active layer, featuring even smaller pores than the intermediate layer, effectively rejects pollutants

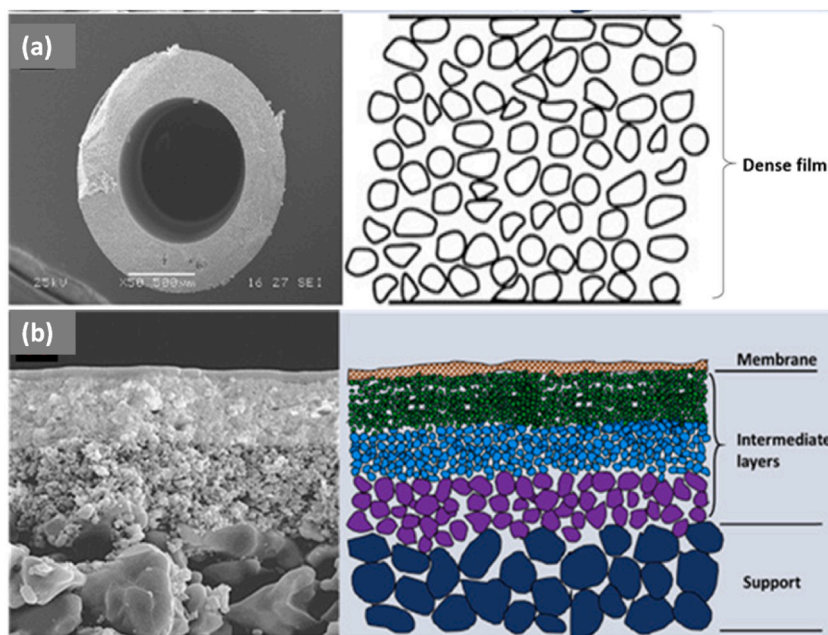


Fig. 1. The typical cross-section of (a) symmetrical ceramic membrane [35] and (b) an asymmetrical ceramic membrane [36].

[30]. Ceramic membranes are categorized into microfiltration (MF), ultrafiltration (UF), and nanofiltration (NF) based on the size of their pores. The pore sizes for each category are the ones that are greater than 50 nm, the one that ranges from 2 to 50 nm, and less than 2 nm, respectively [31].

The utilization of ceramic membrane technology in wastewater treatment is experiencing a rapid expansion owing to its extended durability, chemical stability, and minimal fouling inclination [32]. They also have excellent thermal stability which makes them useable in high-temperature operations [33]. Ceramic membranes can successfully eliminate various pollutants from wastewater through processes such as size exclusion, adsorption, and electrostatic repulsion. The efficiency of separating pollutants in wastewater using ceramic membranes depends on factors such as the membrane's pore volume, pore size, size distribution, and surface chemistry [34].

Application of ceramic membranes is mainly at the laboratory scale due to the high cost of raw materials, multi-manufacturing steps and high sintering temperatures [37,38]. The estimated cost price of ceramic membranes ranges from \$500 to \$1000 per square meter, with the price of polymeric membranes accounting for 25 % of that price [33]. In order to enhance the price competitiveness of ceramic membranes for large-scale applications, it is imperative to lower the fabrication costs associated with them [39]. The use of improved ceramic membranes for wastewater treatment appears to hold a significant prospect for future advancement in water treatment and sanitation. Several studies have employed ceramic membranes in wastewater treatment and the search for low-cost and environmentally friendly starting materials has continued to engender research interests. Therefore, solid wastes and readily available materials rich in  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  such as coal fly ash, natural clay, kaolin and sand can be used as starting material for ceramic membranes [40–43]. Studies by Hossain and Roy [44] and Sawunyama et al. [45] reported that coal fly ash and bentonite clay had significant amounts of  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  which can make them good candidates for ceramic membranes raw materials. It has been demonstrated in numerous studies that it is possible to fabricate ceramic membranes from coal fly ash. Recently, Rao et al. [46] fabricated a novel fly ash blended/potter clay ceramic membrane. Fu et al. [47] also prepared a low-cost ceramic membrane from coal fly ash as the main raw material and  $\text{AlF}_3$  as the catalyst.  $\text{SiC}$  ceramic membrane with mullite bonds were also fabricated from coal fly ash, silicon carbide and 5 %  $\text{MoO}_3$  [48]. The utilization of these affordable starting materials in large-scale membrane processes presents sustainability, environmental benefits and potential high removal efficiencies for pollutants [30,49,50]. In addition to resolving the problem of poor water quality, the use of coal fly ash in ceramic membranes in wastewater treatment represents a milestone in waste management because coal fly ash has detrimental environmental consequences [51].

According to a Scopus analysis of publications (spanning over a 20-year period) on coal fly ash-based ceramic membranes, about 175 research papers have been published, with the highest publication occurring in 2023 (Fig. 2). This is a sign of a lately popular study area that is beginning to receive significant attention from researchers. Few review articles have examined the different functional roles that coal fly ash plays in the fabrication of ceramic membranes for wastewater treatment. This review, therefore, provides up-to-date information on the use of coal fly ash as sustainable raw material for the fabrication of ceramic membrane. Finally, the causes and management of fouling in coal fly ash-based ceramic membranes are discussed, as well as the fabrication cost and future prospects. This review will contribute to the expansion of understanding on the application of coal fly ash in the fabrication of ceramic membranes.

## 2. Coal fly ash

Coal fly ash is a residue that is generated through the combustion of pulverized coal in thermal power plants [52]. It is an extremely fine, heterogeneous powder mixture of crystalline and amorphous phases with spherical geometry, and particle sizes between 10 and

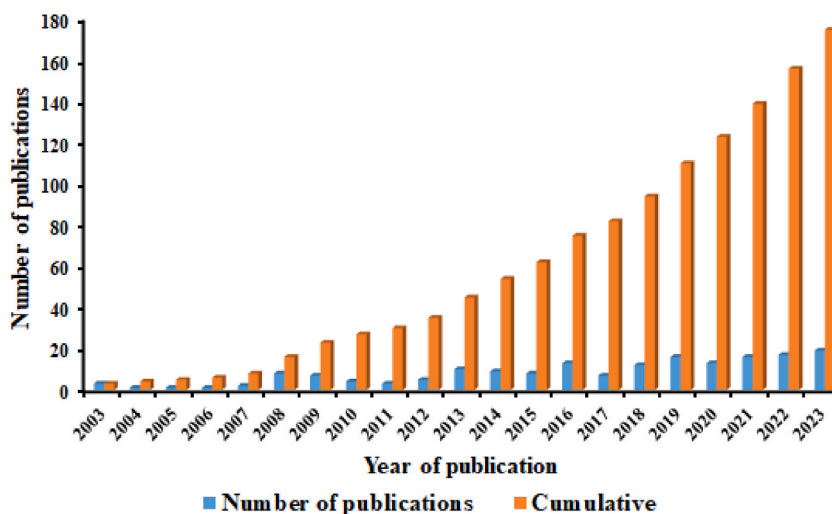


Fig. 2. Analysis of publications from Scopus database on coal fly ash based ceramic membranes over the past 20 years (search was performed using the keywords “coal fly ash + ceramic membrane”).

100  $\mu\text{m}$  [53,54]. Oxides of silicon, aluminium, calcium, iron, and calcium are the main components of coal fly ash. There are also little amounts of magnesium, calcium, potassium, sodium, titanium, and sulphur as shown in Fig. 3. Significant levels of hazardous metals such as As, Ba, Hg, Cr, Ni, V, Pb, Zn, and Se are also present in the coal fly ash [55,56]. These toxic substances may seep out of it and pollute the environment. Additionally, they may cause cancer, skin and lung infections, nausea, vomiting, and organ dysfunction in people [57].

Chemical composition, mineralogy, and coal source are used to categorize coal fly ash. The combined mass composition of silicon dioxide, aluminium dioxide, potassium oxide, titanium dioxide, and phosphorus oxide is used to classify coal fly ash in terms of its chemical composition [58,59]. Calsialic, ferrisialic, ferricalsialic, and sialic are the various classes. Based on mineralogy, coal fly ash can also be classified as magnetic spinel and mullite-quartzite [60]. Coal fly ash can be further divided into two classes: Class C and Class F, depending on where the coal came from. Class F is from bituminous and anthracite coals, which have a lower calcium concentration than Class C's high calcium content (above 20 %) lignite and sub-bituminous coals [61–63]. Compared to Class C, Class F contains a higher percentage of alumina, silica, and iron (around 70 %) [64]. High calcium content in class C results in the production of a greater amount of calcium-aluminate-silicate hydrate, whereas Class F's higher silicon content produces a greater amount of sodium-aluminate-silicate-hydrate [60,65,66]. The source of the coal, the amount of ash in the coal, the degree of coal mining and preparation, the conditions during combustion and mining, the current climate, and other pertinent aspects all affect the properties (physical and chemical) and composition of coal fly ash [67].

Around 8025 Mt of coal were used worldwide in 2022 (Fig. 2a) by several industries, including cement manufacturing, iron and steel production, and power generation. As a result, a lot of coal fly ash is being produced, but less than 30 % of them are being recycled. Consequently, disposal takes up a lot of space, which is a significant problem. Transporting the coal fly ash for disposal also comes at a considerable expense. Coal fly ash is also harmful to the environment since it can contaminate the air, water, and soil and leach hazardous compounds [69–71]. Because of this, experts are now concerned about how to properly manage, dispose of, and use coal fly ash. Even though there has been a lot of research done on the global usage of coal fly ash (Fig. 4a), the building industry as shown in Fig. 4b is currently where it is most frequently used. These applications are ascribed to shape, surface area, high strength, lightweight, and non-toxic and porosity, among other physical characteristics [72]. The most lucrative application of coal fly ash in terms of volume is as a raw material for ceramic membranes, yet little study has been done in this field.

### 3. Synthesis of coal fly ash based ceramic membranes

The fundamental components required for the manufacture of ceramic membranes include inorganic precursors, and additives such as solvents, dispersants, pore formers, binders, and plasticizers. The fabrication process involves three primary steps which are: (1) preparation of suspension or paste from precursor material and additives, (2) shaping the suspension/paste into a desired geometry such as tubular, plate or disc configuration and (3) drying, and sintering of the resultant ceramic membrane [75]. An optional step, which involves the leaching of undesired elements in the coal fly ash before the mixing of raw materials and additives is determined by the analysis of the elemental composition of the coal fly ash. The allowable concentrations of these elements are guided by the thresholds indicated in the national guidelines for heavy metal release. This is due to the possible presence of large levels of toxic metals such as Ba, As, Hg, Cr, Ni, V, Pb, Zn, and Se in coal fly ash [76,77]. The most common manufacturing techniques for ceramic

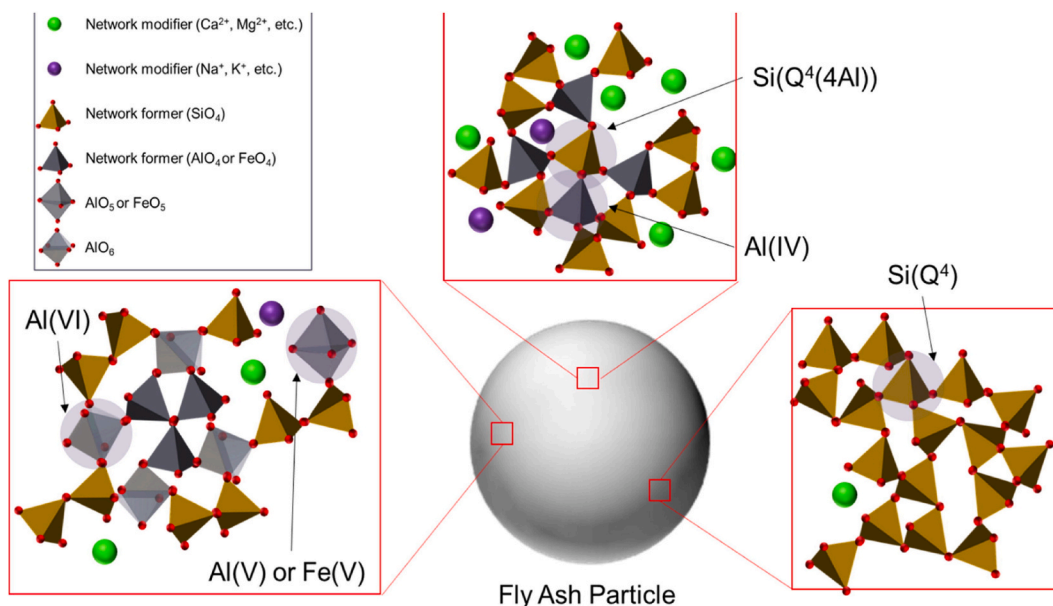


Fig. 3. Schematic Illustration of the structure of coal fly ash [68].

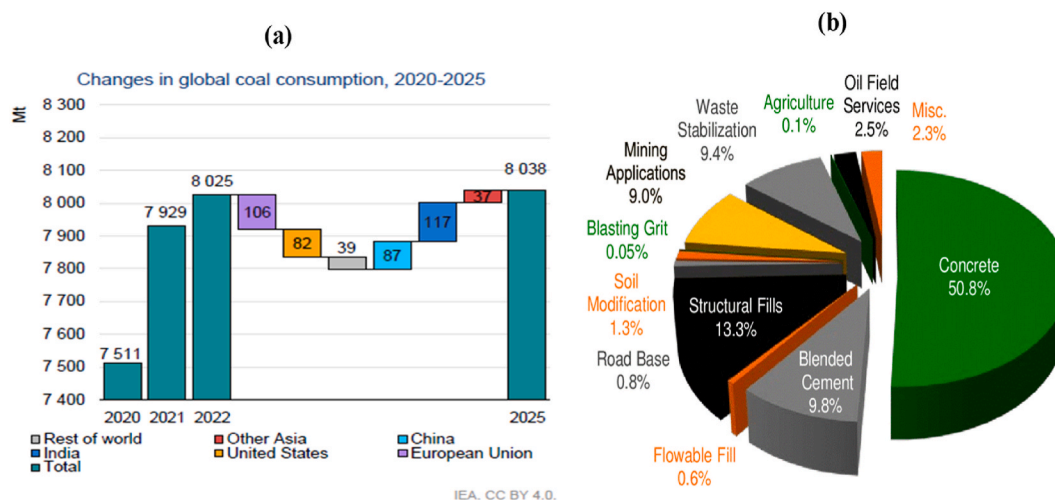


Fig. 4. (a) Changes in global coal consumption (b) various uses of coal fly ash [73,74].

membranes are freeze casting, slip casting, tape casting, pressing and extrusion. The choice of a ceramic membrane fabrication technique depends on application requirements, the desired membrane geometry, and the specific starting materials [78]. In the fabrication of ceramic membranes, the additives have a variety of important functions. The viscosity of the slurry is controlled using solvents like water and those with organic bases, such as acetone, to make it easier to spread over the carrier film and to increase the uniformity of the additives in the ceramic powder. Ceramic particles can be held together using binders such as polyvinyl alcohol [79–81], sodium metasilicate [82,83], carboxymethyl cellulose (CMC) [84], and others. The settling of the powder particles is prevented by the use of dispersants like sodium hexametaphosphate [34], sodium carbonate, and boric acid [82]. In order to make synthetic membranes more flexible, glycerine is mostly utilized as a plasticizer. The porous ceramic membrane's porosity is increased using pore-forming materials including dextrin [84,85], calcium carbonate [81,82,86], and spherical graphite [79,87] which decompose completely during the sintering process. Table 1 gives the properties, configurations, and fabrication techniques of some coal fly ash based ceramic membranes.

### 3.1. Synthesis of coal fly ash ceramic membranes by pressing method

Axial pressing and isostatic pressing are the two pressing techniques used in the fabrication of ceramic membrane supports. Axial pressing is classified as dry or wet, with dry pressing involving dry powders without water and wet pressing involving the addition of water to ceramic raw materials. The two types of isostatic pressing are cold pressing and hot pressing. Cold isostatic pressing creates membrane support using external pressure, but hot isostatic pressing creates the desired shape using heating. While isostatic pressing applies pressure from various directions to boost shape capabilities, axial pressing is more affordable and appropriate for high volume manufacture of simple geometrical ceramic membranes [88]. Apriyanti et al. [89] used the dry pressing method to fabricate flat ceramic membrane from coal fly ash. When a pressure of 1 bar and calcination temperature of 1100 °C for 7 h was applied, the resulting membrane was more durable and had a homogeneous surface free of any cracks. The coal ash membrane's major crystal structure morphology was  $Al_2O_3$ , according to XRD analysis. Due to the formation of a robust and porous ceramic membrane, the findings from the study demonstrated that coal fly ash can be used as a raw material for ceramic membranes. Liu et al. [90] fabricated a cylindrical-shaped porous ceramic membrane using wet ball milling and uniaxial pressing. Dolomite was employed as a pore-forming agent, sintering inhibitor, and reactant in addition to coal fly ash as the starting material. The findings demonstrated that the addition of dolomite effectively modified the support pore structure by increasing pores through the release of  $CO_2$  and preventing coal fly ash from sintering while supplying sufficient mechanical strength and good thermal characteristics in the finished porous ceramic membrane. In a similar study, Chathurappan et al. reported the suitability and effectiveness of dry pressing method in the fabrication of coal fly ash based ceramic membranes. The main raw materials used in the synthesis process were coal fly ash and fuller clay. The procedure involved in the fabrication is shown in Fig. 5 [91].

### 3.2. Synthesis of coal fly ash ceramic membranes by extrusion method

One of the often-employed conventional methods for fabricating multi-channel ceramic membranes that may be produced on an industrial scale is extrusion. The homogeneous, high-viscosity paste created by combining aggregate powder with additives is vacuum melted and aged. The additives form the required plastic properties for effective shape-forming without losing cohesiveness. The uniform high viscosity paste is then extruded into tubular supports by a nozzle after entering the vacuum extruder. To preserve the synthetic membrane's final structure, the remaining chemicals are evaporated. The most significant factors for process control are the applied pressure in extrusion and the rate of extrusion. The diameter of the raw material, the amount of binder, and the sintering

**Table 1**  
Techniques used in the manufacturing of coal fly ash-based ceramics and the properties of the resulting membranes.

Ceramic membrane	Fabrication method	Additives	Characteristic of the synthesized material	Ref.
Hydrophobic Coal-Fly-Ash-Based Ceramic Membrane	Extrusion	sodium hexametaphosphate of the suspension served as a dispersant and the PVA acted as a binder	Increased pore size from 0.15 $\mu\text{m}$ to 1.57 $\mu\text{m}$ , the water flux rose	[34]
Highly porous whisker-structured mullite ceramic membranes	Uniaxial pressing	MoO <sub>3</sub> as a single sintering additive Organic binder polyvinyl alcohol	Open porosity showed a significant increase from 41.65 $\pm$ 0.13 % to 58.14 $\pm$ 0.15 % as the MoO <sub>3</sub> content increased from 0 to 20 wt%, accompanied by a decrease in shrinkage and pore size in the absence of any pore-forming agent	[103]
Coal fly ash based multipurpose ultrafiltration membrane	Uniaxial pressing	–	The membrane porosity-39.8 % hydraulic pore radius - 41 nm. The membrane pure water flux increased from 2.09 to 11.31 m <sup>3</sup> /m <sup>2</sup> sec as the applied pressure increased from 69 to 483 kPa.	[91]
Low-cost microfiltration membranes from fly ash	Paste-casting	Sodium carbonate and boric acid act as dispersant Sodium metasilicate acts as binder Calcium carbonate act as pore forming agent	The membranes, which were defect-free, possessed an average pore size ranging from 1.2 to 2.3 $\mu\text{m}$ and displayed excellent chemical stability in both acidic and basic solutions. Furthermore, they had a porosity level of 35–40 %.	[82].
Porous mullite ceramic supports from high alumina fly ash	Dry pressing method	Spherical graphite as pore-forming agent. Polyvinyl alcohol (PVA) as a binder	Whose apparent porosity 55.7 %, Flexural strength 8.5 MPa and	[87]
Kaolin-fly ash-based membranes	Paste-casting method	H <sub>3</sub> BO <sub>3</sub> and na <sub>2</sub> CO <sub>3</sub> were used to provide homogeneity Calcium carbonate defines the pore morphology Sodium metasilicate (na <sub>2</sub> SiO <sub>3</sub> ·9H <sub>2</sub> O) acts as a binder.	Porosity (~34.36–39.0 %), pore size (~0.65–1.81 $\mu\text{m}$ ) flexural strength (~10–30 mpa)	[83]
SiC-Coal fly ash ceramic membranes	dry pressing method	Polyvinyl alcohol Graphite- pore former	Porosity 54.46 % exhibited good, mechanical properties	[79]
Coal fly ash ceramic membrane	extrusion method	Dolomite used as a pore-forming Carboxymethyl cellulose (cmc) used as a binder Glycerin used as a <u>plasticizer</u> ,	Large particles reduced mechanical strength of fly ash based membranes while increasing porosity.	[104]
Lumina based tubular asymmetric membranes incorporated with coal fly ash	Centrifugalcasting technique	–	Increased porosity and enhanced water permeability coincide with reduced mechanical strength and diminished linear shrinkage	[105]
High-aluminum coal fly ash ceramic membrane supports	Uniaxial cold-pressing	PVA solution as organic binder CaCO <sub>3</sub> pore forming agent	Mechanical strengths 34–90 MPa	[86]
Coal fly ash-based tubular ceramic membrane	Extrusion	Dextrin as pore-forming agent CMC, as organic binder Glycerin, to improve the plasticity	Porosity 38.9–45.9 % Mechanical strength 14.8–36.0 MPa	[84]
Fly ash cenosphere ceramic membrane	uniaxial pressing	organic binder PVA- CaCO <sub>3</sub> - pore forming agent	Porosity 59.25 % Mechanical strength 70 $\pm$ 2.58 MPa	[81]
Tubular supported ceramic microfiltration membranes from fly ash	Slip- casting	Dispersant lomard Binder dsx 3290	Average pore diameter 0.77 $\mu\text{m}$ . Pure water permeability- 1.56 $\times$ 10 <sup>4</sup> L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup> , respectively, Transmembrane- 0.1 MPa at room temperature. A porosity of 45.4 $\pm$ 0.9 % was obtained.	[97]
Mullite-whisker-structured porous ceramic membrane	Uniaxial pressing	Binder- PVA-1750 AlF <sub>3</sub> as crystallization catalyst MoO <sub>3</sub> as mineralizer		[106]
Fly ash ceramic membrane	Slip casting method	Polyvinyl alcohol	Pore size- 1.47 ( $\mu\text{m}$ ) Porosity- 39 (%) Mechanical strength 1.98 (MPa)	[107]
Tubular coal fly ash based ceramic membranes.	Extrusion method	Dextrin as pore-forming agent CMC, as organic binder Glycerin, to improve the plasticity	Porosity- 44.76–47.30 % Mechanical strength 15.16–29.11 (MPa)	[85]

temperature on the support affect the qualities and performance of the synthesized membrane [92,93]. Huang et al. synthesized coal fly ash based ceramic membrane with high efficiency for water and heat recovery. Dextrin was utilized as a binder, while carboxymethyl cellulose was used as a pore-forming agent. The procedure involved in the fabrication is shown in Fig. 6 [85]. In another study, Jedidi et al. [94] reported the fabrication of porous tubular coal fly ash based ceramic membrane using extrusion method. At a constant sintering temperature of 1125 °C, it was demonstrated that the ceramic membrane synthesized exhibited favourable mechanical and chemical resistances. Additionally, it possessed a significant average porosity of 51 % and a mean pore diameter of 4.5  $\mu\text{m}$ . The membrane's permeability was measured at 475 L h<sup>-1</sup> m<sup>-2</sup> bar.

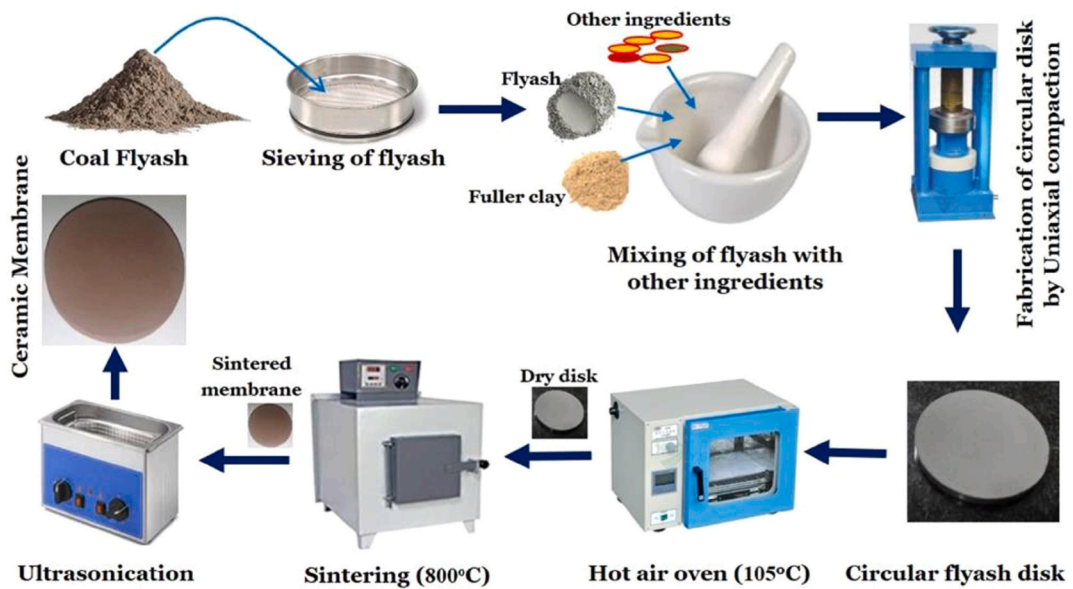


Fig. 5. Schematic illustration for the preparation of coal fly ash based ceramic fabrication by dry pressing method [91].

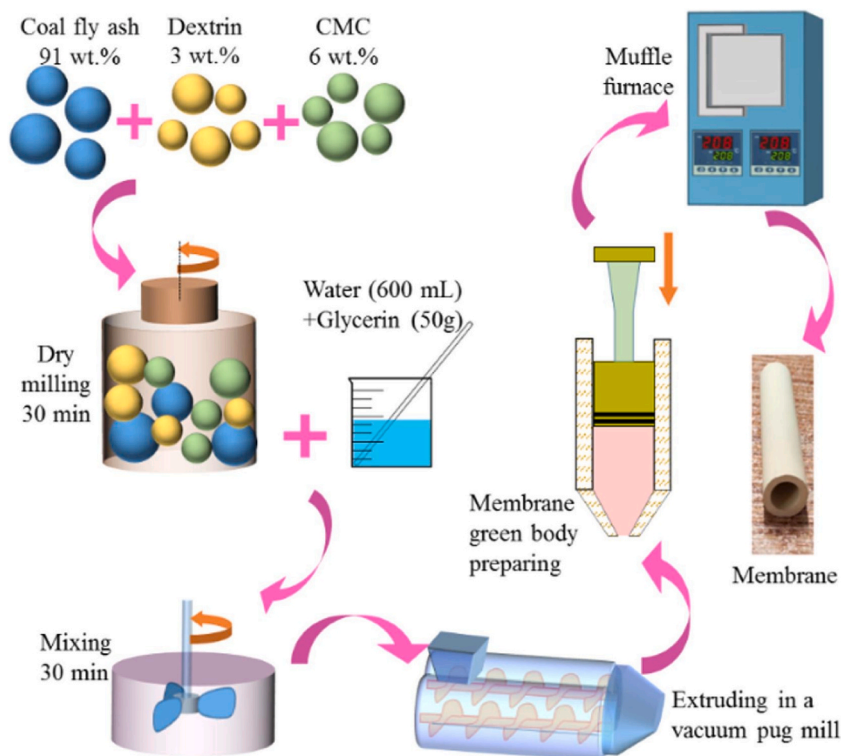


Fig. 6. Fabrication of coal fly ash based ceramic membrane via extrusion method [85].

### 3.3. Synthesis of coal fly ash ceramic membranes by slip casting method

Slip casting is an easy, affordable, and adaptable technique. It is used to fabricate ceramic membranes with complicated shapes that are non-concentric and uneven. In slip casting technique high solid content suspensions are poured into porous cast molds, allowing the solvents to seep through the pores and precipitate the particles into a layer as illustrated in Fig. 7. The resultant material is sintered after de-moulding, which is the final step in producing the desired ceramic membrane. The fabrication of ceramic membranes from

coal fly ash using the slip casting method was first reported by Jedidi et al [95]. The primary chemical components of the coal fly ash employed in the study are mainly comprised of  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ , and  $\text{Fe}_2\text{O}_3$ . The minor components were made up of a variety of alkali metals. Fly ash powder, distilled water, and polyvinyl alcohol (PVA) were mixed to create the slurry. The final membrane was subjected to a thermal process that involves 24 h of room temperature drying followed by 800 °C sintering. SEM images showed that the membrane surface was uniform and free of cracks. The results showed that the active layer's average pore diameter was 0.25  $\mu\text{m}$  and its thickness was close to 20  $\mu\text{m}$ . The fabricated ceramic membrane had a 475 L/h  $\text{m}^2$  bar permeability. A number of factors, such as sintering temperature, casting time, suspension pH, particle size, etc., have an impact on the properties of slip casted ceramic membranes [96]. Fang et al. [97] investigated the effect of slip concentration, withdrawal speed and casting time on the preparation of coal fly ash ceramic membrane fabricated using the slip casting technique. The findings showed that an increase in slip concentration causes a significant increase in membrane thickness as well as an increase in porosity because more particles adhere to one another. Additionally, it was reported that the optimum slip concentration should be utilized to prevent the development of surface flaws like cracks. The average pore size distribution and pore diameters were observed to be unaffected by casting time and pull-out speed, while the thickness of the membrane was found to be affected.

### 3.4. Synthesis of coal fly ash ceramic membranes by tape casting method

Thin, flat dense ceramic membranes made of ceramic slurry are fabricated using tape casting technique. The steps in the tape casting method are as follows: A slurry made of inorganic powder and additives such as binders, dispersants, and plasticizers are continually dispensed onto a moving substrate while a doctor blade could be adjusting the thickness (Fig. 8). Then, the cast film is dried and sintered to form the resultant ceramic membrane. The benefit of tape casting is that ceramic sheets can be configured into any desired shapes, including circular, square, and rectangular shapes and also allows adjustment of the thickness of the ceramic membrane [99]. However, the main problem with tape casting is the loss of shape accuracy caused by the deterioration of the plaster mould. Tape casting also takes a very long time when a slurry of small granules is cast [100]. Several factors, including the viscosity of the slip, the distance between the knife blade and the moving carrier, the depth of the reservoir, and the speed of the carrier, can affect the tape casting process used to fabricate the ceramic membranes [101].

## 4. Application of coal fly ash in ceramic membrane for wastewater treatment

The increasing demand for environmental protection and the reuse of waste materials, implies that the utilization of CFA in different aspects of membrane development is being continually sought. The presence of heavy metals such as chromium, vanadium and antimony in CFA is a factor that has limited its application due to the possibility of long-term leaching. This has necessitated the pre-treatment of the CFA prior to its use in ceramic membranes. Nugteren et al. [108] reported a forced leaching process using water, citrate, EDTA, oxalate and carbonate solutions. The use of cation exchange membrane assisted electrokinetic method for the removal of heavy metals from CFA has been reported by Peng et al. [109]. After the removal of heavy metals, CFA can serve the following five major purposes in the development of membranes for wastewater treatment.

### 4.1. Membrane precursor

The large amount of CFA being generated as waste in coal-fired electrical power stations, means it can be a readily available raw material for several industrial processes. Due to its high composition of  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$ , CFA can serve as an economical precursor for the synthesis of ceramic membranes for wastewater purification.

CFA has been significantly explored as raw material for the synthesis of mullite-based ceramics. Mullite is a solid solution with several unique properties, such as, low thermal conductivity, high chemical resistance and temperature stability, low thermal expansion coefficient and high flexural strength [110,111]. CFA pickling could be an important pre-treatment process in enhancing the

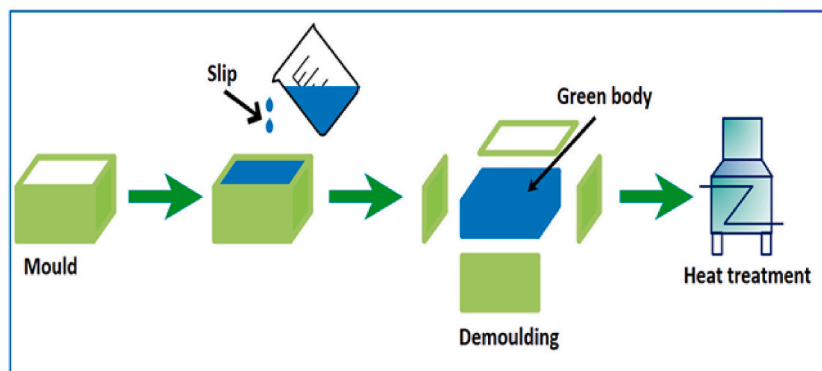


Fig. 7. Schematic illustrations of the slip casting technique for fabricating ceramic membranes [98].



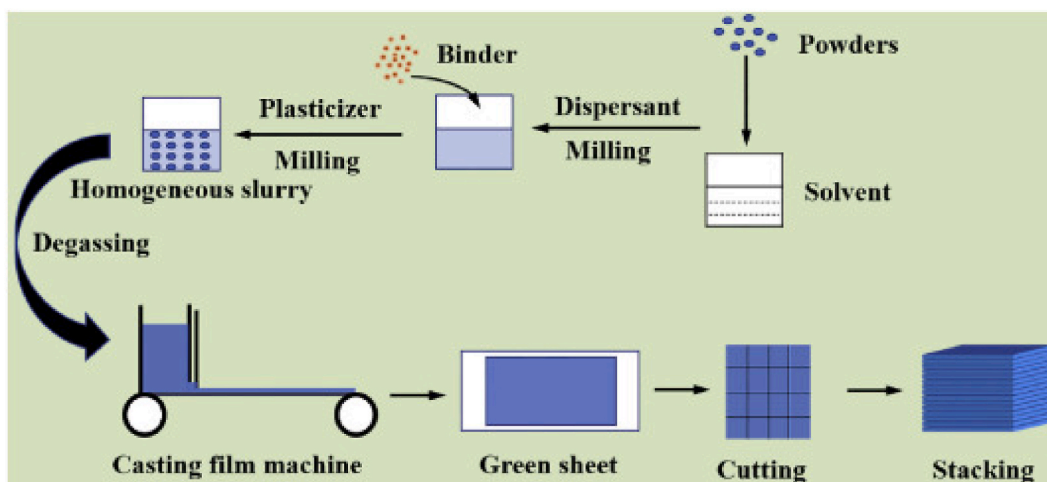


Fig. 8. Schematic illustrations of the tape casting technique for fabricating ceramic membranes [102].

flexural strength of mullite ceramics Li et al. [112]. Also, the sintering temperature and the addition of  $\text{AlF}_3$  as a whisker promoter could influence the final property of the obtained ceramic.  $\text{AlF}_3$  was observed to influence properties such as phase composition, porosity, bulk density, and aspect ratio of the obtained whiskers. Mullite obtained from CFA was employed together with multiwalled carbon nanotubes (MWCNT) to form a hierarchically structured membrane, the membrane achieved complete retention of *E. coli* and *S. Aureus* [113]. Interestingly, no pore blockage was reported for the material and fouling only occurred due to the bacterial cell deposition of the surface.

Tobermorite, a crystalline calcium silicate hydrate, is another ceramic material that has been obtained using CFA as a raw material [114]. The excellent mechanical properties and durability of tobermorite, makes it a potential material in various structural application. The high reaction activity of tobermorite has been reported to allow for a lower sintering temperature compared to traditional ceramics Luo, Ma [114]. Aluminum substitution and morphology were also observed to be two factors that influenced the mechanical property of the membrane. Tobermorite from waste materials such as newsprint recycling residue and, paper recycling ash have been reported as efficient adsorbent and cation exchanger in a few studies [115–117].

In another study, the synthesis of  $\text{Al}_2\text{O}_3$ -NaA zeolite hollow fibre membranes from CFA with high efficiency for Pb(II) ions removal in low concentration has been reported Zhu et al. [118] reported. The membrane displayed a pore diameter of 0.41  $\mu\text{m}$ , permeation flux of  $670 \pm 20.6 \text{ L/h/m}^2$  and Pb(II) ion removal reached 99.9% at 0.1 MPa. The high membrane area per volume and thin thickness of the  $\text{Al}_2\text{O}_3$  support was important in enhancing the permeation flux of the composite membrane.

#### 4.2. Sintering additive

The presence of additives often influences shrinkage, dimensional changes, grain growth and homogenization during sintering of ceramics. With the aid of appropriate additives, the sintering temperature of ceramics can be reduced through the formation of eutectic composition [119,120]. The sintering additive should enhance densification without decomposing the base materials, while also introducing a weak interface. Das et al. [121] reported the use of CFA as sintering additive in the synthesis of a mullite bonded porous SiC ceramic membrane. The obtained membrane exhibited high water flux of 5261  $\text{L/m}^2/\text{h}$  bar, mean pore size of 3.7  $\mu\text{m}$  open porosity of 44.4%. The membrane was able to achieve 91% removal of oil from a kitchen wastewater with initial oil concentration of 1657  $\text{mg/L}$ . The presence of CFA as a sintering additive, allowed for the processing of the membrane at a lower temperature, without comprising its mechanical and permeability characteristics.

#### 4.3. Membrane filler

To improve the hydrophilicity and stability of polymeric membranes, inorganic fillers have become very important in forming mixed matrix membranes. These membranes possess numerous advantages arising from the synergy between the organic and inorganic phases [122]. Several studies have explored the incorporation of ceramic metal oxides such as cerium oxide ( $\text{CeO}_2$ ), zirconium dioxide ( $\text{ZrO}_2$ ) and aluminium oxide ( $\text{Al}_2\text{O}_3$ ) as nano-fillers for ceramic membranes in wastewater treatment because of their potential to enhance the selectivity, surface area, hydrophilicity and stability of polymeric membranes [123]. Because of the large amounts of fly ash being produced by power stations and the similarity between the chemical composition of fly ash and traditional raw materials, the use of fly ash in membrane fillers as high economic and environmental value [124]. The mechanical properties of ceramics obtained using fly ash fillers, generally depends on the quantity of the reinforcing materials, component of ceramic, porosity, shape, interfacial interaction between filler and composite matrix and the presence of other reinforcements [125]. As observed by Manocha et al. [125], the porosity of a carbon-fly ash-ceramic composite was observed to increase with higher fly ash composition. This improved porosity,

however, implies that the membrane suffers from reduced coefficient of friction. This limitation could be mitigated by adding silicon carbide into the mixture, which led to an improvement in the coefficient of friction of the membrane.

The use of CFA-modified ceramics has been explored in various wastewater treatment processes because of their unique advantages. Recently, Rao et al. [46], reported the application of a novel fly ash blended/potter clay ceramic membrane in microbial fuel cell (MFC) technology for COD removal from wastewater. The hydration property of the membrane showed CFA concentration dependency, resulting from the variation in the concentration of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> in the composite as the quantity of CFA is varied. This is significant for the ceramic membrane's proton conduction efficiency and their optimal composition is highly important for improved membrane performance. The introduction of CFA as fillers for the membrane reportedly improved the thermal stability, optimized the pore size distribution and heterogeneity, enhanced water uptake, improved proton conductivity, and decreased oxygen mass transport coefficient. This improved property of the CFA-modified membranes was important in the wastewater treatment efficiency of the ceramic membrane. CFA-filled ceramic membranes were also reported as micro-electrolysis ceramic media for the pre-treatment of tetracycline wastewater. Two novel ceramics: sintering ferric-carbon (SFC) and sintering-free ferric-carbon ceramics (SFFC) based on coal ash, and scrap iron were used in tetracycline wastewater pre-treatment to improve biodegradability, removal efficiency and inhibit antibiotic-resistant bacteria growth [126].

#### 4.4. Membrane filters

Microfiltration membranes comprising of CFA active layer is another well explored application of CFA in wastewater treatment. Most fly ash membranes developed till date have been focused on the treatment of oily wastewater. However, the numerous advantages of the materials mean it is being explored also in the removal of other organic and inorganic pollutants from water. Recently, CFA-based membranes have been utilized in the treatment of effluents from textile and tin industries, domestic households, and poultry slaughterhouses. Table 2, shows the application of CFA-based membranes in the treatment of different industry generated effluents. CFA-based membranes have also been found to be effective for the removal of bacteria, phenol, and humic acid from wastewater.

The synthesis of microfiltration membranes with CFA as both microporous support and active layer has been reported Jedidi et al. [94]. The active layer obtained by slip-casting and sintering yielded a pore size of 0.25 µm and possess a water permeability of 475 L/h m<sup>2</sup> bar. The membrane showed high efficiency in the treatment of dyeing effluents from the textile industry. The stabilised permeate flux was 100 L/h/m<sup>2</sup>, the COD and color removal were 75 and 90 % respectively, with the permeate turbidity reduced to 0.5 NTU from the initial 45.5 NTU.

Modification of CFA membranes with nanostructured materials is a potential route to obtaining highly efficient filters for wastewater treatment. The incorporation of nanoparticles could influence the properties of CFA-membranes such as hydrophilicity, permeation flux, and their adsorption property [132]. Zhang et al. [132] synthesized Fe<sub>3</sub>O<sub>4</sub>@CFAS with high efficiency for Cu(II) ion removal. The incorporation of Fe<sub>3</sub>O<sub>4</sub> resulted in an enhancement of the hydrophilicity of the membrane, with no significant alteration in the permeation flux of the composite membrane observed. The improved hydrophilicity played a significant role in enhancing the adsorption property of the composite as it promotes contact and chelation of active sites with Cu(II) ion.

The use of CFA as membrane support have also reported in literatures. The synthesis of a porous mullite-whisker-structured ceramic membrane supported with CFA and bauxite as starting materials was reported by Zhu et al [106] reported, while AlF<sub>3</sub> and MoO<sub>3</sub> acted as crystallization catalyst and mineralizer, respectively. The introduction of MoO<sub>3</sub> was observed to lead to a reduction of the high temperature viscosity of liquid melts which promoted the growth of elongated mullite crystallites. The open porosity of the membrane was observed to be enhanced by the introduction of AlF<sub>3</sub>. At the optimal weight composition of MoO<sub>3</sub> and AlF<sub>3</sub> (5 and 4 wt%

**Table 2**  
Application of CFA-based membranes in the treatment of different effluent.

Description	Fabrication technique	Composition	Membrane characteristic	Wastewater type	Efficiency	Reference
Tubular microfiltration membrane	Extrusion	75 % CFA, 20 % quartz; 5 % calcium carbonate	Pore size: 0.133 µm Porosity: 40.17 %; Water permeability: $17.75 \times 10^5 \text{ (Lm}^{-2}\text{h}^{-1}\text{bar}^{-1})$	Poultry slaughterhouse wastewater	COD removal: 100 %; TSS: 100 %	[127]
Tubular membrane	Extrusion	75 % CFA, 20 % quartz; 5 % calcium carbonate	Pore size: 0.133 µm; Water permeability: $4.93 \times 10^{14} \text{ Lm}^{-2}\text{h}^{-1}\text{bar}^{-1}$ Tortuosity: 1.68	Starch-rich wastewater	COD: 100 % TOC: 100 % Turbidity: 100 %	[128]
Fly ash/alumina composite membrane	Thermal spraying	Mullite whiskers/fly ash/polyvinyl alcohol/glycerol	Pore size: 0.1 µm Permeability: $445 \text{ Lm}^{-2}\text{h}^{-1}\text{bar}^{-1}$	Oily emulsion and stannic acid rejection	TOC: 99 % Tin rejection: 99.9 %	[129]
TiO <sub>2</sub> -fly ash membrane	Hydraulic press and hydrothermal synthesis	Fly ash: 80 %; Quartz: 10 %; Calcium carbonate: 10 %	Porosity: 35 % Pore size: 1.32 µm; Permeability: $6.135 \text{ Lm}^{-2}\text{h}^{-1}\text{bar}^{-1}$	Oil-in-water emulsion	Oil rejection: 98.7 %	[130]
Kaolin/fly ash composite membrane	Sintering	AFA/kaolin/fly ash	Pore diameter: 0.32 µm; Permeability: 3650 $\text{Lm}^{-2}\text{h}^{-1}\text{bar}^{-1}$	Distillery wastewater	Oil rejection: 98.05 %	[131]

respectively), the open porosity of the membrane was  $47.3 \pm 0.6\%$ . The study further affirmed that despite the enhancement of the membrane's porosity, the mechanical strength was not compromised even though the membrane was prepared at a relatively low temperature of  $1200\text{ }^{\circ}\text{C}$ . The membrane achieved a total organic carbon removal of over  $90\%$  at  $0.2\text{ MPa}$  for oil-in-water emulsion.

Recently, the synthesis of  $\text{TiO}_2$  nanofiber membrane loaded on porous fly ash ceramic support was reported by Zhang et al. [133]. The novel composite membrane obtained from fly ash, graphite and industrial rutile was fabricated using a semi-dry press and hydrothermal process. The composite membrane showed excellent activity for the simultaneous removal of heavy metal ions and rhodamine B. The capacity of the membrane for dual functionality as both adsorbent and photocatalyst makes it a potential cost effective material for wastewater treatment.

The solute rejection efficiency at a constant flux of a choline chloride blended cellulose acetate membrane, used in CFA support, demonstrated dependence on pH. When the pH was basic, both the membrane and solute had negative charges, resulting in an increased ability to retain phenol. On the other hand, when the support membrane was coated with formaldehyde-cross-linked polyvinyl alcohol, the correlation of the rejection pattern changed considerably, leading to high rejection rates at acidic pH levels [134,135].

#### 4.5. Factors affecting the physical properties and performance of coal fly ash based ceramic membranes

The mean pore size, porosity, flexural strength, and permeability of coal fly ash based ceramic membrane can be affected significantly by several factors such as sintering temperature, dosage of the pore-forming agent, particle size of coal fly ash, and dosage of binding additives, as indicated by several studies. In contrast, numerous experiments have shown that when operating conditions change, ceramic membranes made of coal fly ash does not crack. However, the precise trend of variation of these factors with properties and performance could not be established, as a result of the involvement of other factors that influence both the performance and properties of coal fly ash based ceramic membrane [98]. Therefore, major factors that need optimization in the fabrication of coal fly ash ceramic membrane are highlighted in this section.

##### 4.5.1. Effect of sintering temperature on the properties of coal fly ash ceramic membranes

The properties of coal fly ash ceramic membranes are affected by the sintering temperature. Singh and Bulasara [82] examined how the sintering temperature affects the properties of a ceramic membrane made from coal fly ash. The membranes were sintered at  $800$ ,  $850$ ,  $900$ , and  $1000\text{ }^{\circ}\text{C}$ . The physical characteristics of the coal fly ash ceramic membrane, including color, tensile strength, crystal phase, pore size, and porosity, were observed to be impacted by changes in the sintering temperature. The results showed that the average pore size only increased for the membrane sintered at  $1000\text{ }^{\circ}\text{C}$  whereas it decreased for sintering temperatures up to  $900\text{ }^{\circ}\text{C}$  (Fig. 9a). Except for those sintered at  $1000\text{ }^{\circ}\text{C}$ , as shown in Fig. 9b, the sintered membranes all had similar colors. Fig. 9c shows that

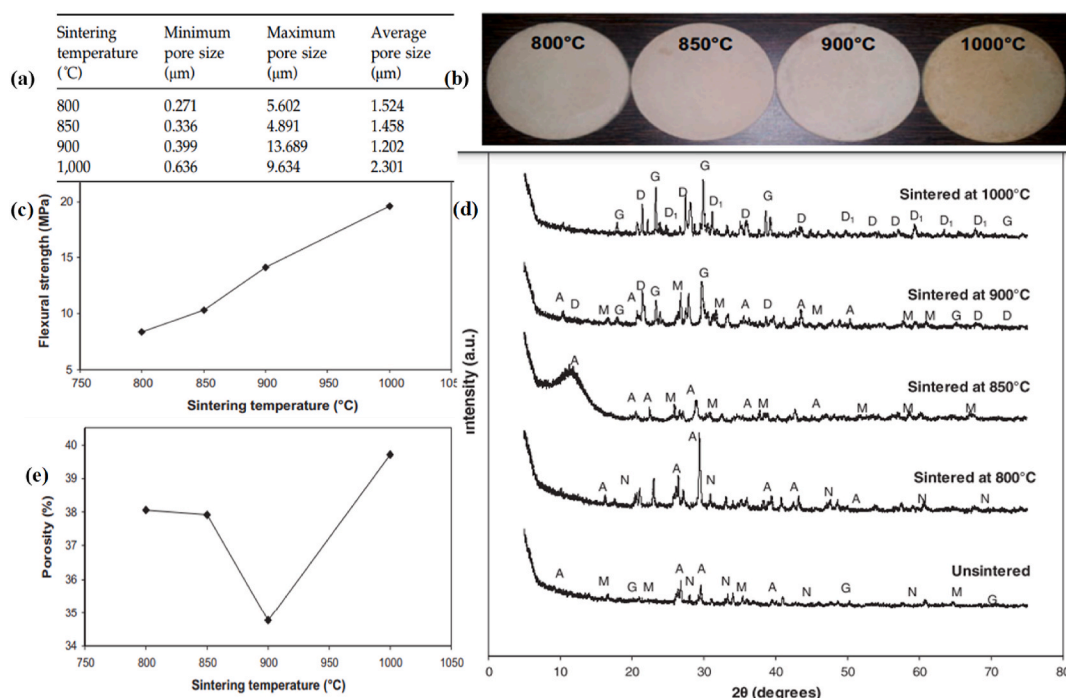


Fig. 9. (a) Variation of average pore size with sintering temperature, (b) picture of membranes sintered at four different temperatures, (c) variation of flexural strength with sintering temperature (d) XRD spectra at different sintering temperatures, and (e) variation of porosity with sintering temperature [82].

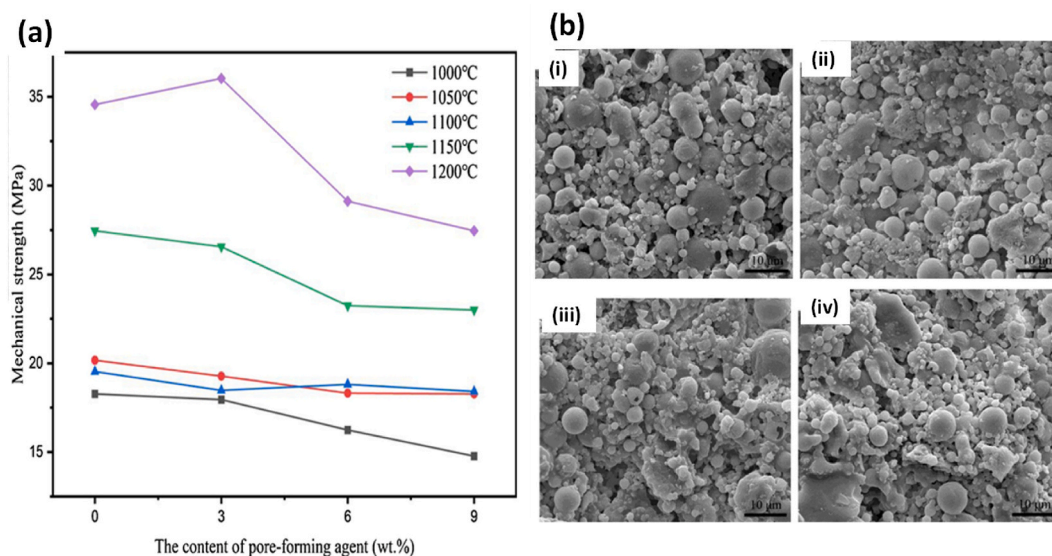
when sintering temperature increased, mechanical strength also increased. Continuous phase transitions occur during the sintering process, as shown by a comparison of the XRD patterns of various samples (Fig. 9d). The membrane sintered at 1000 °C lost anorthite, which was a significant component in the other four samples. Throughout the sintering process, other phases underwent changes and their contents changed as well. The membranes' porosity dropped from 38.1 to 34.8 % when the sintering temperature rose from 800 to 900 °C. However, the porosity of the membranes increased significantly to 39.7 % when sintered at 1000 °C Fig. 9e.

#### 4.5.2. Effect of pore forming agent dosage on the properties of coal fly ash ceramic membrane

Pore-forming agents are essential in improving the permeability of ceramic membranes by changing the morphology and porosity of the pores. These pore-forming agents can be either inorganic or organic compounds that decompose at high temperatures. Cui et al. [87] studied the effect of pore-forming agent dosage on the characteristics of coal fly ash ceramic membrane. Spherical graphite was used as a pore-forming agent in their investigation, and the dosage was altered as wt%. The results demonstrated that as the dosage increased, many pores developed and were linked together to generate holes. As a result, water absorption and permeability flow improved. Liang et al. [84] also investigated the effect of pore forming agent dosage on the properties of coal fly ash ceramic membrane. They used dextrin as the pore-forming agent, with compositions of 0, 3, 6 and 9 wt% ( $D_{50} = 7 \mu\text{m}$ ,  $C_{6n}H_{10n}O_{5n} \times H_2O$ , and  $\rho = 1.8 \text{ g/cm}^3$ ). The experimental results indicated that the pore-forming agent content and mechanical strength of the samples are essentially inversely correlated (Fig. 10a). Additionally, the results demonstrated that when the dextrin concentration increases, more pores appear in the ceramic membranes made of tubular coal fly ash, resulting in a more porous structure, as seen in the SEM micrograph in Fig. 10b. The average pore size showed a trend of initially increasing and then decreasing with increasing pore-forming agent content, reaching its maximum value when the quantity is 6 wt%. The decrease of average pore size when the pore-forming agent level reaches 9 wt% was attributed to the strong binding action of dextrin. All the aforementioned studies revealed that altering the dosage of the pore-forming agent does not affect the development of cracks in the ceramic membrane made of coal fly ash. Suresh et al. [136] also investigated the effect of pore-forming agents on mechanical strength and porosity of the fabricated membrane. In their study, coal fly ash was used as the main precursor and a mixture of corn oil, waste cooking oil and starch solution as the pore forming agent. The results showed that porosity increased with increasing dosage of the pore-forming agent which corresponded to a decrease in mechanical strength.

#### 4.5.3. Effect of particle size of coal fly ash on the properties of coal fly ash ceramic membrane

Particle size optimization of starting materials is key in the manufacturing of ceramic membrane as it helps maintain material consistency. Particle size has a significant influence on the mechanical and physical properties of ceramic membrane such as pore size, post-sintering shrinkage, bending strength, microstructural morphology, and porosity [137–140]. The effect of coal fly ash particle size on the porosity, pore size, microstructure and mechanical strength of synthesized coal fly ash based ceramic membrane has been investigated [104]. The experimental results showed that, a membrane with a denser structure and less porosity is produced when coal fly ash has a wider particle size distribution. As particle size increases, the membrane's pores get bigger while mechanical strength gets weaker. Large particles in the fly ash play a significant role in the mechanical strength of the ceramic membrane because they produce cracks in the membrane.



**Fig. 10.** (a) Relationship between the mechanical strength of the membrane and the content of the pore-forming agent as well as the temperature of sintering. (b) SEM images of Coal fly ash ceramic membrane with Wt% of dextrin (i) 0, (ii) 3, (iii) 6, and (iv) 9 sintered at 1150 °C [84].

#### 4.5.4. Effect of binder dosage on the properties of coal fly ash ceramic membrane

Binders are used to hold ceramic particles together. Goswami et al. [141] investigated how the amount of binder affected the characteristics of a ceramic membrane made from coal fly ash by extrusion technique. Fly ash was used as the basic material (75 %), together with the necessary amounts of quartz (20 %), calcium carbonate (5 %), and sodium salts of carboxymethyl cellulose acting as binders, to create tubular ceramic membranes. The results revealed that, a higher binder concentration causes agglomeration in the membrane matrix, which results in the creation of big, irregular holes. With a rise in binder concentration, the membranes' mechanical strength considerably decreased. In addition, the results showed that an increase in binder concentration had a detrimental effect on the membrane's chemical stability.

#### 4.6. Factors that affect separation efficiency of coal fly ash-based ceramic membranes

The surface characteristics of the ceramic membrane (surface wettability and surface charge), the solution properties (pH, temperature, viscosity), and the operation parameters (pressure difference, flow rate, and time) are key variables that determine the separation effectiveness of ceramic membranes [35,142]. Surface charge of coal fly ash based ceramic membranes have an impact on how pollutants interact with the membrane surface, which has an influence on the ceramic membrane's permeability and rejection capabilities [143–145]. Higher membrane flux and improved antifouling properties occur when membrane surface charge is similar to contaminants charge due to the repulsive effect of similar charge [146]. Contrarily, opposing charges cause electrostatic interactions between contaminants and membranes, which have a major negative impact on the effectiveness of separation and on membrane anti-fouling properties [147,148]. The pH of the filtration feed has a significant impact on the surface charge of the membranes [149].

The separation effectiveness of coal fly ash based ceramic membrane is also impacted by the presence of inorganic ions in the contaminants matrices. This results from these ions' ability to alter the surface charge of membrane surfaces and contaminants [150]. Alventosa-deLara et al. [151] studied the effects of the feed salt concentration on the removal of organic dyes by ultrafiltration ceramic membranes. The results showed increased overall resistance and decreased dye rejection by the ceramic membrane in the presence of NaCl. This occurred as a result of the  $\text{Na}^+$  ion neutralization of the membrane surface charge and allowing more dye particles to pass through.

Hydrophobicity and hydrophilicity of coal fly ash based ceramic membrane surfaces affect separation effectiveness of the membrane [131]. This is due to the correlation between contact angle values and ceramic membrane morphology and pore size [152]. Surface wettability has an impact on how membranes are utilized, with hydrophilic membranes frequently employed in water filtration and hydrophobic membranes excellent for membrane distillation [153]. For instance, Zhang et al. [34] fabricated hydrophobic coal fly ash based ceramic membrane for desalination. The membrane showed excellent water flux and higher salt rejection compared to aluminium based ceramic membranes. Surface geometrical structure or surface free energy can be modified to alter the wettability of membrane surfaces [34,106,154]. Zhang et al. [80] studied the effect of changing wettability of the surface of a coal fly ash based ceramic membrane. In their study the ceramic membrane was modified from hydrophilic to hydrophobic through immersion of 1H,1H,2H,2H-perfluorodecyltriethoxysilane (FC8) on the surface of the membrane. The results demonstrated that the final super hydrophobic membrane outperformed hydrophilic ones in terms of self-cleaning. The physical and chemical characteristics of the materials being separated, such as viscosity, charge, stability, and solubility, can be influenced by temperature and pH [155,156]. This could impact on the diffusion rate across the membrane pores. The fouling behaviour of the contaminants was also influenced by temperature [157].

#### 4.7. Management of fouling on coal fly ash based ceramic membranes

There have been numerous investigations on the physical and chemical deposition or adsorption of microbiological, organic, and inorganic compounds onto membrane pores or onto the membrane surface. This causes the membrane pores to contract or clog thereby reducing permeability and raising the flow resistance, which has an impact on the quantity and quality of treated effluent [158,159]. This dramatically lowers the lifespan and separation effectiveness of these membranes [160,161]. Not many reports are available on how to clean the fouled coal fly ash based ceramic membrane. Therefore, the main reasons, preventative measures, and methods for cleaning fouled ceramic membranes are all discussed in this section.

Reversible fouling, which often forms on the membrane surface, and irreversible fouling, which refers to internal fouling in the membrane pores, are the two subgroups of membrane fouling [162]. Ceramic membrane fouling is primarily a function of the properties and performance of ceramic membranes, operational conditions as well as the characteristics and nature of the filtering feed. These membrane properties include roughness, wettability, surface charge, and membrane pore size [160,163]. Foulant type (inorganic salts, metal oxides, natural organic matter, manufactured organic matter, and microbiological contaminants), concentration, pH, charge, and ionic strength are some of the properties of the feed. Fouling of ceramic membranes is influenced by a number of operating factors, including flow rate, temperature, and pressure. Coal fly ash is less likely to get irreversible fouling than organic membranes because it is predominantly composed of metallic oxides, which have strong hydrophilicity and chemical stability [163,164]. However, reversible fouling of coal fly ash or other ceramic based membranes are major obstacles to their use in wastewater treatment.

Fouling raises operational costs because it necessitates higher pressures to maintain permeate flux, uses more energy, requires more time to clean, and costs more to replace the membrane [165,166]. Therefore, specific steps must be taken to either prevent fouling or clean the fouled membrane in order to restore and maintain membrane flux, extend the cleaning cycle, and prolong the lifespan of ceramic membranes [167]. Pre-treating influents with sacrificial membranes or another type of treatment method before ceramic

membrane filtration, optimizing operational parameters, controlling the feed solution's properties, and using a pre-coat layer on the membrane are all actions that reduce fouling [162,168]. Combining different processes, such as advanced oxidation with ceramic membrane technology can also lessen fouling through changing properties of ceramic membranes and enhancing separation efficiency. Ceramic membrane surfaces that have been dip-coated with nanoparticles are an example of how the surfaces are modified [33]. Bao et al. [169] studied how to get rid of fouling by coating the ceramic membrane with cobalt oxide. The cobalt oxide was ensured not to leak into the pores of the ceramic membrane by the development of the Co–O–Al bond on its surface. The results of the study showed that ceramic membrane functionalized with cobalt oxide have antifouling properties. The pollutant's intrinsic catalytic decomposition by the metal oxide may be the cause of the antifouling qualities since it keeps the pollutant from passing untreated through the ceramic barrier.

Ceramic membrane fouling can be eliminated through cleaning procedures such as chemical, physical, electrical, and vibration. All these methods do not guarantee the complete regeneration of ceramic membranes. Chemical cleaning uses chemicals such as acids, surfactants, oxidizing agents, chelating agents, and alkali to clean ceramic membrane pores and surfaces [160,170,171]. This approach can be used to independently or in conjunction with physical cleaning techniques. Although chemical cleaning is a common cleaning approach, it has a number of negative aspects, including harm to the environment, membrane degradation, cost due to the high cost of cleaning chemicals, and the tendency to alter the nature of the foulants [162,172]. Back washing and mechanical scraping of contaminants on the membrane surface and pores are examples of physical cleaning methods. The most common of these techniques is back washing. It entails washing the membrane against the direction of the typical flux of membrane penetrates. However, according to Zielińska [173], this method can harm the ceramic membrane structure. The ideal strategy for cleaning ceramic membranes is combining cleaning techniques that work in conjunction. This is due to the fact that different ceramic membranes foulants, such as inorganic salts, metal oxides, naturally occurring organic matter, synthetically produced organic matter, and microbiological contaminants have distinct properties [174].

## 5. Coal fly ash based ceramic membrane cost analysis

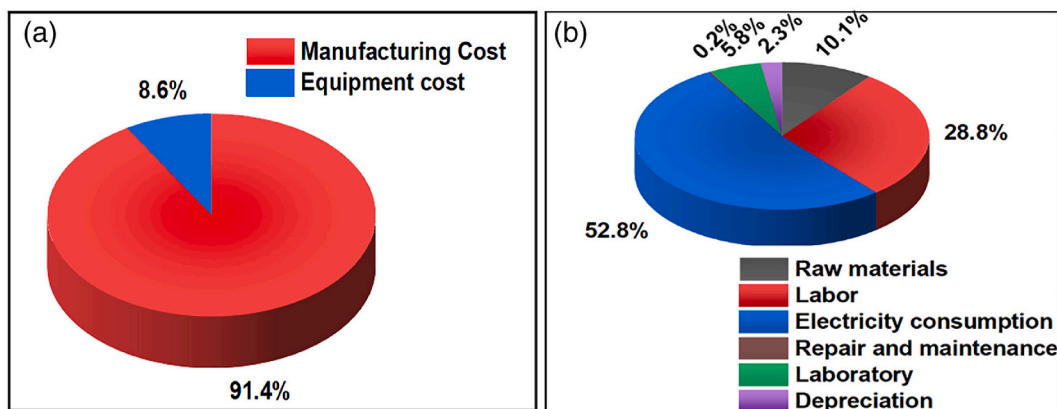
Ceramic membrane fabrication by commercial starting materials such as silicon carbide, stainless steel, aluminium, zirconium oxide and silicon nitride are very expensive. Their cost price ranges from \$500–3000/m<sup>2</sup> with the price of polymeric membranes ranging from \$20–200/m<sup>2</sup> [175,176]. The preparation of ceramic membranes from these materials also requires high sintering temperatures (1300–1700 °C) which also makes the production process costly [177]. Ceramic membranes offer lower operating costs than polymeric membranes due to their longer lifespan, simplicity of cleaning by high-temperature steam sterilization, and capacity to restore initial permeability and water flux by reverse flushing and proper cleaning [178]. One of the main factors that have made the use of coal fly ash in the production of ceramic membranes popular is their low production cost as shown in Table 3. The inexpensive production costs of coal fly ash based ceramic membranes is one of the paramount reasons why it is so common to use it in the fabrication of ceramic membranes. Therefore, the use of coal fly ash as a ceramic membrane starting material is considered a viable solution. Coal fly ash cost price ranges from \$25 to \$250/m<sup>2</sup>. The six ceramic membranes shown in Table 3 have significantly variable production cost prices because various expenses during the fabrication of the coal fly ash-based ceramic membrane were taken into account. The most complete production cost pricing should include the cost of starting materials, processing costs, maintenance expenses, labour costs, and electricity expenses [179]. The estimated production cost of a coal fly ash-based ceramic membrane fabricated by Suresh et al. [136] was USD525/m<sup>2</sup>. In their cost analysis studies, they factored in various parameters associated with the cost of coal fly ash, cost of additives, manpower, electricity and equipment. In another similar study, Goswami et al. [180] fabricated a coal fly ash ceramic membrane and the cost price of a unit membrane was estimated by considering several expenses as shown in Fig. 11. The estimated unit cost price established was USD 250/m<sup>2</sup>. Lower sintering temperatures and coal fly ash pricing are the two main factors that dramatically lower production costs [181].

## 6. Conclusions and future prospects

The use of coal fly ash in membrane technology has garnered much attention because of its low sintering temperature, high surface area, low initial cost, and significant active functional groups, which primarily consist of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>. Additionally, the use of coal fly ash marks a significant advancement in waste management. Hence, heavy metals from coal fly ash should be removed before ceramic membrane fabrication to ensure safety and environmental sustainability for long-term use. Despite the promising outcomes associated with the use of coal fly ash as membrane precursors, sintering additives, membrane supports, membrane fillers, and

**Table 3**  
Cost of different coal fly ash based ceramic membranes.

Coal fly based ceramic membrane	Production cost price (USD/m <sup>2</sup> )	Reference
Novel fly ash blended ceramic membrane in MFC	35.76	[46]
Coal fly ash ceramic membrane	25	[178]
Defective analcime/geopolymer composite membrane derived from fly ash	31.8	[182]
Fly ash-based low-cost tubular ceramic membrane	250	[180]
Low-cost microfiltration membranes from fly ash	17	[82]
Fly ash based ceramic microfiltration membranes for oil-water emulsion treatment	225	[136]



**Fig. 11.** (a) Fabrication cost of coal fly ash based ceramic membrane as percentages of manufacturing cost and equipment cost and (b) factored in manufacturing expenses [180].

membrane filters in membrane technology, their full-scale application has not been thoroughly investigated and implemented. Numerous studies have found that the use of ceramic membranes made from coal fly ash has effectively reduced turbidity, dye and heavy metal concentrations, total dissolved solids, sea desalination, and oil content of wastewater. Therefore, laboratory studies must be scaled up to explore the viability of pilot plants with full consideration of aspects that affect their efficiency and durability to advance the many uses of coal fly ash in membrane technology. To lower the fabrication cost, which is the primary factor affecting the full utilization of ceramic membranes, research should continue to look for relatively affordable and easily accessible materials rich in chemical composition that are essential in the fabrication of ceramic membranes. They could then be used as composites with coal fly ash or serve different roles in membrane preparations. The antifouling potential of ceramic membranes fabricated from coal fly ash should be enhanced to promote the effectiveness of ceramic membrane technology in the treatment of wastewater.

#### Data availability

Data used for this study will be made available upon request.

#### CRediT authorship contribution statement

**Lawrence Sawunyama:** Writing – original draft, Formal analysis. **Olalekan C. Olatunde:** Writing – original draft, Formal analysis. **Opeyemi A. Oyewo:** Writing – review & editing, Supervision. **Mokgadi F. Bopape:** Writing – review & editing, Supervision. **Damian C. Onwudiwe:** Writing – review & editing, Supervision, Conceptualization.

#### Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Damian Onwudiwe reports administrative support was provided by North-West University - Mafikeng Campus. Damian Onwudiwe reports a relationship with Elsevier B.V. that includes: Editorial board membership. Not applicable has patent Not applicable pending to Not applicable. Editorial Board Member of Heliyon-Materials If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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