

Review Article

Studies on Production of Low-Cost Ceramic Membranes and Their Uses in Wastewater Treatment Processes

Gülzade Artun^{1*}, Ayşegül Aşkın²

¹ Department of Chemical Engineering, Eskisehir Osmangazi University, Eskisehir and 26480, Turkey 1, 0000-0003-1018-8679, gulzadeart@gmail.com

²Department of Chemical Engineering, Eskisehir Osmangazi University, Eskisehir and 26480, Turkey 1, 0000-0002-5781-3905, aaskin@ogu.edu.tr

* Correspondence: gulzadeart@gmail.com

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Abstract

The need for water is increasing owing to the rapidly increasing world population, urbanization, industrialization, global climate changes, and intensive agricultural activities. While the importance of water is increasing day by day, the purification and reuse of water have become more and more essential. Reducing the pollutants at the source as much as possible and if it will be given to the receiving environment, giving the wastewater treatment in accordance with the current discharge regulations, are priority processes. Of the advanced separation methods, membrane separation technology is used to reduce water resource use and control environmental pollution and has advantages such as high separation efficiency, convenient operation, compact equipment, and energy-saving. Membranes are divided into two categories according to their structures: organic and inorganic membranes (or ceramic membranes). Compared to organic membranes, inorganic membranes have many advantages such as high thermal and chemical stability, good resistance to microbial erosion, easy regeneration and cleaning, high mechanical strength, and long-term stability in harsh conditions. The high cost of ceramic membranes is an important disadvantage. In recent years, it is seen that membrane production costs are reduced by using natural minerals such as kaolin, quartz, bauxite, diatomite, clay, limestone, dolomite, zeolite, and industrial wastes instead of the commonly used ceramic membrane raw materials. By using these raw materials, besides reducing the cost of ceramic membranes, it also contributes to lowering the temperatures required for their production. In this study, studies on ceramic membranes produced from industrial wastes (waste ash, sawdust, sewage sludge, construction and demolition wastes, waste diatomite, rice husk ash, sugarcane pulp ash, corn cob ash waste etc.) were examined.

Keywords: Ceramic Membranes, Industrial Waste, Wastewater Treatment

1. Introduction

Industrialization, urbanization, and population growth, as well as excessive use of water and the consequent pollution of freshwater resources, cause increasing water scarcity worldwide [3, 10]. Since usable water resources are limited, treatment of water and wastewater, improvement of water quality, and removal of pollutants from water are important steps to be taken [28]. As a result, day by day, countries adopt strict environmental measures to ensure sustainable growth and development. In this context, innovative and affordable costs are researched in the fields of water treatment and reuse [10].

Every industry produces wastewater with different contents [42].

Industrial wastewaters may contain heavy metal ions, bacteria, organic compounds, nutrients, oil, suspended matters, dyes, pesticides, endocrine disrupting compounds, and some other toxic substances [12, 27, 32, 42]. Consequently, efficient treatment of wastewater is required to protect people, the environment, and the harmful effects on aquatic life as a receiving medium [42].

One of the wastewater types is oily wastewater. There are difficulties in the treatment of such wastewaters. Oil removal from oil-in-water emulsions is an important part of contamination control. To separate oily wastewater in treatment plants, a lot of traditional techniques are used. Skimming, gravity settling (API separator), dissolved air spinning, aggregation, and centrifugation methods are used for separating free oil from water. The membrane separation method used in the separation of oils is one of the successful methods used to separate secondary emulsions in recent years [1].

It is widely used in micro-filtration, ultra-filtration, and nano-filtration, especially in water and wastewater treatment [7]. In the membrane filtration method, no chemical additives are used to break the emulsions and membrane filtration method provides high Chemical Oxygen Demand (COD) removal. Due to such advantages, it is a very important method in the treatment of oily wastewater [1].

Membrane technology is widely and effectively used in many separation processes, including drinking water production, wastewater treatment, resource recovery, gas purification, air purification, food industries, and environmental and other industries [17, 29, 33].

2. Membrane Filters

Membrane technology, which is applied in many different industrial areas, including the separation of emulsified oil and bacteria from water, as well as processes such as membrane reactors, is defined as green and high-efficiency separation technology [25].

Membranes are effective in reducing waste disposal expenses and in the recovery and recycling of materials that provide economic advantages. Membranes have become a standard procedure in recent years due to the advantages they offer [6].

Membranes act as a physical barrier in wastewater treatment and ensure that pollutants in wastewater are removed from the water without any chemical addition and degradation [26].

Since the production of polymeric membranes is easy and has lower cost, there are many more industrial applications [12, 37]. However, porous ceramic membranes provide advantages over polymeric membranes in terms of mechanical properties, high-temperature pressure resistance and stability, good chemical stability, rugged structure, environmental friendliness, anti-fouling performance, operational durability, and long-term use [5, 12, 25, 37].

Ceramics and polymers are two main materials used in the construction of membranes for water and wastewater treatment [28]. Both polymers and ceramic materials are widely used in the manufacture of membranes for different applications.

2.1.Polymeric membranes

Polymeric membranes are more commonly used for industrial applications such as water treatment and desalination because of their low cost, scalability, and easy manufacturing processes [10, 12], 28, 36]. Since the surface of polymeric membranes is more hydrophobic, contaminants are easily adsorbed on the membrane surface due to these hydrophobic attractions [34]. The polymers used for the membrane are not biodegradable, they are polluted and may have adverse effects on the environment [10]. As a result of contamination, such systems show poor stability [3]. Contamination problems of polymeric membranes limit their long-term use.

Polymeric membranes are comparatively cheaper but rather poor in chemical, thermal, and mechanical stability, especially in industrial wastewaters [26, 41, 42].

2.2.Ceramic membranes

The reason why ceramic membranes cost more than polymer membranes is that their production is complex (It is generally used in the production of high purity Al_2O_3 powders ceramic membranes.). In addition, for these membranes sintering temperatures are high [24]. Among these expensive materials, alumina powder and a large amount of heat required for high sintering temperatures make ceramic membranes expensive [6]. It is stated that when ceramic membrane prices are provided between \$500/m² and \$1000/m², it will be possible to use it in industrial applications [31].

One study predicts a compound annual growth rate of 11.3% from 2019 to 2027 for the ceramic membrane industry. This corresponds to \$10,893.5 million. In addition to its high mechanical strength, chemical and thermal resistance, easy cleaning, antibacterial

properties, and long-lasting use of ceramic membranes are among its important advantages [10].

The use of ceramic membranes is more advantageous and effective instead of traditional steps in water treatment (coagulation, precipitation, and filtration) [6]. For this reason, studies on low-cost ceramic membranes have attracted great interest in recent years. Being low-cost ceramic membranes are generally produced from cheap raw materials that can be sintered easily such as kaolin, bauxite, clay, pozzolan, and fly ash [24].

In addition, ceramic membranes provide filtration at a wide pH range (from 1 to 14). Cleaning with aggressive chemicals, organic solvents, and water vapor is another important advantage [8].

Ceramic membranes can be broadly classified as porous and dense membranes, considering their structure. In porous membranes, pore size is one of the most important factors [6]. Dense ceramic membranes are used in gas separation processes. Especially at low temperatures, using dense ceramic membranes can provide direct separation of CO₂ from flue gases. Dense ceramic membranes' transport mechanism differs from ceramic membranes in terms of dissolution and diffusion solutes transportation [6].

The pore size of ceramic membranes is very important in determining the application areas of the membrane. As an example, microporous (>50 nm) and mesoporous (2-50 nm) ceramic membranes find wide use in many microfiltration (MF) and ultrafiltration (UF) applications. Nanofiltration (NF) membranes are used especially in micropollutants, hydrophobic macroporous membranes are used in membrane distillation (MD) applications [10].

Preparation of ceramic membranes includes forming, sintering, and surface modification steps [40]. In these steps, ceramic membranes are given the desired geometry depending on the applications, generally in flat, tubular, and hollow fiber shapes [10].

Whatever process is used in the preparation of the membrane, the most important step for the preparation of membranes with good mechanical strength is the sintering step. The sintering process consists of pre-sintering, thermolysis and as control of grain growth, relative density, shrinkage and morphology, removal of unwanted materials, which are the final sintering processes [10].

Clay material is widely used in the preparation of low-cost ceramic membranes. The chemical compositions differ depending on the origin, particle properties, and size of the clay material. Kaolin-based hollow fiber membranes are more effective in oil-water separation. The silicate-containing ash from the coal combustion and in the rice husk can

be the raw material for the mullite material. However, it is stated that the effect of both its composition and mineralogy content should be well investigated, especially in terms of their use in factory production conditions. Cost-benefit analyzes performed on an industrial scale show that the use of low-cost materials in membrane processes will provide both economic and environmental advantages [3].

In the production of ceramic membranes, fly ash, sewage sludge, pulp, waste glass, ceramic scrap, etc. are used by modifying the membrane microstructure to improve surface porosity, pore size, pore distribution, and mechanical strength. In the preparation of porous mineral-based ceramic membranes, mineral-based raw materials (kaolin, bauxite, sepiolite clay, industrial solid waste coal fly ash, etc.) are abundant in the world and their costs are low, such applications have recently attracted more attention [48].

3. Waste Materials

The increase in population, economic growth, and industrialization brings along serious environmental problems such as the continuous increase in industrial waste. These industrial wastes are released into the environment in violation of the disposal procedures, they harm the environment and cause deleterious health effects on living things [23]. When wastes are managed correctly, loss of raw materials can be prevented, natural resources and climate protection can be ensured. Solid waste recycling is another important area that can be used significantly to produce new products [6]. As a result, solid wastes can be used as ceramic materials, ceramic membranes, adsorbents, composting-fertilizer, energy, etc. [23].

The reuse of waste materials and by-products originating from industries in different sectors is an important step in the development of sustainable and environmentally friendly materials. If fly ash is not disposed of fly ash wastes in the right way, land disposal and environmental pollution problems arise. Effective recycling of waste fly ash not only reduces environmental pollution but also contributes to the production of high-value-added products. Such wastes have significant silica content. This situation is also effective in its use in many areas. Globally, fly ash, rice husk ash, and sugar cake ash are produced in large quantities and these ashes can be reused raw materials [3].

Recycling of industrial by-products or wastes to produce ceramics is investigated in terms of environmental, ecological, and economic aspects. In recent years, many research articles have been published on these topics. Though, the applications of ceramics produced from industrial wastes are still limited. Considering natural raw materials and wastes, properties of end products, continuous availability, transportation costs, fixed chemical compositions, and pretreatment applications of wastes are limited.

This situation can be overcome by increasing recycling incentives in industrial production [29].

To reduce production costs, methods such as the use of auxiliaries to lower the melting point and central casting of the production process, and one-step co-sintering processes are used. At this stage, raw materials such as natural minerals and industrial solid wastes are used.

3.1. Fly ash

Fly ash (FA) is produced in coal-fired power stations and is a type of waste whose safe disposal is problematic, although it is applied in various industries such as concrete and pavement [3, 6, 16, 17, 21]. During the combustion of coal, FA particles are usually carried away with the aid of flue gas and are trapped by electrostatic or mechanical precipitators [5].

Due to the trace elements substances in coal fly ash (CFA) content, it is considered a hazardous substance. Also, the production rate of FA is higher than consumption. For this reason, FA applications should be developed in a more comprehensive way for better disposal and use of solid waste sources [47].

Many studies have been carried out on this material since coal FA can be used as a strong, hard, and corrosion-resistant non-metallic ceramic material by containing oxygen, carbon, nitrogen, and binding compounds. Such non-metallic materials also contribute significantly to the mechanical properties of material hardness, as they are of low density, coarse-grained, not easily softened as a material, and resistant to penetration [9].

Studies in the literature indicate that waste coal FA will provide a new direction and low cost for the use of ceramic membranes [18, 19].

The diameters of spherical particle geometries of FA range from 1 to 100 microns. In addition, this material, which is easily available and has a composition rich in silica, is suitable to produce porous materials containing cordierite, mullite, and anorthite phases [3].

Studies in the literature show that FA is quite suitable to produce porous mullite membranes. Mullite phase is in $\text{SiO}_2\text{-Al}_2\text{O}_3$ binary phase. Porous mullite material has many advantages such as low coefficient of thermal expansion, good high-temperature strength, low density, and good chemical inertness [16, 21, 24]. Therefore, it is used in the preparation of ceramic supports [24].

In Liu et al. (2016) study, it was stated that in 2012, China constituted 50.2% of the world's coal consumption [36]. On the other hand, it is stated that the annual CFA production is approximately 750 million tons worldwide. CFA is widely used as a cement additive in conventional recycling. Because of the toxic trace elements in CFA content, it can cause health and environmental problems when stored in landfills [25, 36].

Treatment of oily wastewater from wastewater is very difficult. In the Agarwal et al. (2020) study, especially for the treatment of oily wastewater, thick and FA were used in the production of ceramic membranes [4]. Another example given on the use of ceramic membranes obtained from FA for the separation of oily wastewater with the microfiltration process is the study of Zou et al. (2019) [50].

In the study of Fan et al. (2021), a low-cost ceramic membrane was obtained using waste FA and bauxite, and the effects of bauxite content and sintering temperatures on the membrane support structure and performance were investigated [24]. In the study, it was stated that the porosity increased while the shrinkage amount of the membrane support decreased with the increasing bauxite contribution.

Another study carried out with FA is by Zulkifli et al. (2019). In the study, it was determined that good results were obtained in the construction of hollow membranes obtained with 45% FA [51]. With this obtained membrane, high mechanical strength was achieved at all sintering temperatures (1150°C, 1250°C, and 1350°C) and it is stated that the water flow performance is quite effective.

In the study of Apriyanti et al. (2021), the membrane properties and water treatment performance of ceramic membranes obtained using coal FA were investigated [9]. The results of the analysis of the ceramic composite membrane show that FA can be used as a raw material in the production with good porosity so that it can be used in water treatment, and as a result, a strong membrane can be produced.

In the Diana et al. (2020) study, ceramic microfiltration membrane production was carried out by using FA from cement factory wastes [19].

In the Dong et al. (2006) study, waste FA was used to prepare a cordierite-based porous ceramic membrane for microfiltration application [20].

3.2.Rice husk ash

Especially in recent years, agricultural and forestry by-products such as rice husk, rice husk ash (RHA), and wood shavings have started to be used in many new applications, especially in absorption processes. The main reasons for the use of composite materials in their content are that such materials are flexible, available in nature, biodegradable, diverse, and environmentally friendly. The use of these materials, which have very low economic values, together with the advantages they have, is becoming more common [39].

The rice husk coated with rice grains consists of 70-80% organic and 20-30% inorganic hard materials, including silica. Since this part cannot be digested by humans, the rice husk, which has a rate of 20% in rice, is separated from the rice. RHA, which makes up about 25% of the rice husk, is produced as waste by burning rice husk [3].

In Hubadillah et al. (2017) study, heavy metal removal in the water of the ceramic hollow fiber membrane obtained using RHA was investigated [30]. RHA contains significant amounts of silica and carbon.

In the study, in which the use of rice husks in the preparation of the porous ceramic membrane was investigated, the bacteria removal efficiency in water was in the range of 90-99%. In addition, information was given about the required cooking temperatures and strengths for membrane filtration of the porous membrane [22].

3.3.Sugarcane bagasse ash

In sugar and alcohol factories, sugarcane is crushed to extract its juice, resulting in residual fibrous material. This material is pulp. Sugarcane pulp ash is formed as a result of the combustion of the pulp. 250 kg of pulp fiber is formed from an average of 1000 kg of sugar cane, and then an average of 6 kg of ash is formed at the end of combustion. When evaluated according to RHA, sugarcane bagasse ash may contain up to 20% silica [3].

The ceramic membrane study obtained by using a mixture of kaolinite clay and sugar cane pulp was carried out in the study of Andrade et al. (2019) [8]. In the study, it was stated that it can be used as a low-cost forming agent in the production of ceramic membranes that can be used in microfiltration.

Sugarcane bagasse ash was used in Jamalludin et al. (2019) study [32]. In this study, the purification of oil and water mixtures originating from palm heavy mill wastewater, restaurant, and car wash by modifying the membrane, which is obtained from sugar cane wastes and has especially oil-absorbing and separating functions, was investigated. It has been stated that the obtained membrane is successful, easy, effective, and economical for oil and water separation.

In the El Maugana et al. (2022) study, anorthite-based ceramic membrane was obtained by using sugar industry wastes in order to investigate the potential of creating low-cost ceramic membranes [23]. With the optimum results obtained, it was determined that it would be appropriate to use it in industrial treatment.

3.4.Sawdust

Wood is used as a composite material with high porosity, good strength, hardness, and toughness properties with its cellulose, hemicellulose, and lignin content [14].

Sawdust, one of the agricultural and forestry by-products and one of the most attractive materials, is one of the main planting products, especially in Scandinavian countries [39]. Until now, sawdust is usually used in the manufacture of ceramic bricks

and cement. Bose and Das (2015) discussed using sawdust as a pore former to reduce the membrane fabrication cost, given the role of sawdust in membrane production [14].

In the Bose and Das (2015) study, it was investigated how the sawdust performs in membrane production, including the sintering steps [14]. In addition, the effects of controlling the chip particle size, porosity and pore size and the microstructure of the ceramic membrane obtained from the sawdust were investigated. In the study, the results of Scanning Electron Microscope (SEM) analyses of sawdust showed that it was effective in forming pores without significant changes in composition after exposure to different environments with different temperatures and pH. In addition, it is included in the study that the thermal, chemical and morphological properties of sawdust have a very important effect on membrane production.

Organic wastes (sawdust (from local carpentry factory), starch, and ion exchange resin) were used as pore-forming agents in Misrar et al. (2017) study [38]. Sawdust was supplied by a local carpentry factory. After preparing the raw materials to obtain the appropriate stoichiometry, they were brought to the desired size. Organic pore agents at the rate of 10% were added to the powder mixture that was cooked and then cooled. As a result of the study, evaluations on the effect of temperature and starch are presented. Also, in the study, it was stated that organic wastes used as performing were effective as a pore-forming agent even in very small amounts (about 2%).

3.5. Other wastes

In Abdallah et al. (2018) study, in which fine wastes with high alumina content released from the kiln cylinders were used as ceramic membrane material, it was observed that it gave successful results in salt separation from water [2].

Waste corn cobs were used together with metakaolin as membrane support on a green ceramic hollow fiber membrane in the Kamarudin et al. (2020) study [35]. Metakaolin and waste corn cobs were used as pore-forming agent. Waste corn cobs were used while raw materials were preparing. In this study, phase inversion and sintering techniques were used. The separation of this produced membrane oily wastewater was determined as 74.73%.

In the study by Tai et al. (2021), an empty fiber ceramic membrane was developed using palm oil fuel ash to be used in the treatment of oily wastewater [43]. The palm oil fuel ash used in the study was obtained from a crude oil palm mill. It has been stated that studies have been carried out on improving the purity of metal oxides in palm oil fuel ash to provide stronger mechanical strength in separation.

Diatomite sludge and halloysite clay used in a study by Yelova et al. (2020) was used for ceramic membrane construction [48]. Diatomite is used in the brewery industry to increase its brightness during final product filtration. Therefore, the brewery sludge contains diatomite material, which can be used as a pore-forming additive to ceramic membranes. Diatomite material is effective in separating even very fine particles due to its porous, hollow, and channeled structure [48].

Cordierite based materials are used in a wide variety of applications including refractories, thermal insulation, filters, membranes, heating elements, integrated circuit boards, microelectronic, thermal shock-resistant tableware, porous ceramics, microwave, and electromagnetic wave absorbers [11, 13, 15, 45, 49]. In the Wang et al. (2019) study, industrial solid waste was used as a silica channel for a cordierite-based ceramic membrane [46]. The water desalination performance of this membrane was investigated. As a result of the study, it was stated that the cordierite membrane, which is produced from low-cost raw materials (especially wastes), can be cleaned, and used in filtration and desalination processes to prevent membrane contamination. Similarly, industrial solid waste for silica source for cordierite-based ceramic membrane was used in (Teng et al 2020) study [44]. 99.9% salt rejection was obtained from the membrane obtained in the study.

4. Discussion and Conclusion

Compared to polymeric membranes, ceramic membranes have more advantages with their features such as longer life, high stability, high flow, high-temperature resistance, and low contamination.

This study focuses on waste raw materials used for low-cost ceramic membranes used in wastewater treatment. In this context, many parameters (material composition, particle size distribution, pore formers, sintering temperatures, additives) should be considered, especially for ceramic membranes.

In particular, industrial activities generate wastes which have low utilization areas or can not be disposed in a cultivated way. The use of these wastes in the production of ceramic membranes is important both in terms of environment and economy.

In this study, there are examples of waste types in different sectors in the literature. Obtaining good results from literature studies, especially on FA, is a guide for future studies. It is important that the limited studies carried out on other types of waste are expanded in the future and used as an economic value as a part of waste management.

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