The Incorporation of Ceramic Membranes in MBR Systems for Wastewater Treatment: Advantages and Patented New Developments

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Abstract: The membrane biological reactor (MBR) configuration has proven to be optimal for treatment of many industrial wastewaters, especially when optimized treatment efficiencies are an important consideration. Ceramic MBR is being consolidated as a reliable technology for industrial applications, due to the benefits of the system, while ceramic membranes may be an adequate alternative for anaerobic MBR application, due to evidence of low membrane fouling rate and the ability to rigorously clean their surface without at the same time reducing the effective membrane life. This review summarizes Recent Patents on Engineering that disclose key data for both the functioning of MBRs and the use of ceramic membranes. Specifically, an advanced control system for a membrane bioreactor wastewater treatment plant is disclosed, while an integrated biological treatment coupled to membrane filtration is proposed for advanced nutrients removal and reuse of treated waters. Recent studies have been focused on the production of a specific bioceramic membrane with selective adsorption effects and simultaneous digestion ability. Bioceramic membranes may provide effective carbon sources and required minerals to activated sludge microorganisms, while they can be used as biological media filter material for air filtration and wastewater treatment. Furthermore, bioceramics may adsorb phosphate ions and remove oxygen rich components such as nitrates from aqueous solutions.

Keywords: Bioceramic, ceramic membranes, control system, MBR systems, water treatment, wastewater treatment.

1. INTRODUCTION

Membranes can be manufactured from various materials, such as ceramics, organics or metals [1] Table **1**. Organic membranes are most commonly applied in water treatment, including a wide variety of membrane materials, pore sizes, pore size distributions, membrane configurations and production processes. The main benefit for the application of organic membranes is associated with the manufacturing costs. Ceramic membranes are about 10 times more expensive than organic membranes [2]. Other differences in the characteristics of ceramic and organic membranes can be found in resistivity against cleaning agents, hydrophobic/hydrophilic properties, mechanical strength etc; for example, ceramic membranes have a significant resistivity against very high trans-membrane pressures and temperatures. Organic membranes, like cellulose acetate membranes, are usually sensitive to oxidizing agents or the biological activities of the medium where they are immersed.

The membrane structure can be isotropic or anisotropic. Isotropic membranes have a uniform composition and structure throughout. Anisotropic (or asymmetric) membranes consist of a number of layers, each one having different structure and permeability [1].

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The Membrane Bioreactor combines the biological activated sludge process with a membrane filtration step for the separation of activated sludge solids from the water phase. Ceramic MBR can be a very good alternative for industrial wastewaters attributed to its robustness and reliability, as well as due to indirect but important benefits linked to the reduction of the bioreactor volume and to lower sludge production [4].

2. MBR SYSTEMS

2.1. MBR Technology

MBR technology is considered the most advanced technology for wastewater treatment, while Membrane Bioreactor systems have become a promising wastewater treatment technique combining activated sludge and membrane separation; it is a process resulting in a high quality effluent independent of settling characteristics of the biomass. Membrane bioreactors have several advantages over the conventional activated sludge systems, including stable and high effluent quality, easy operation and complete removal of bacteria. However, membrane bioreactors may have several drawbacks due to membrane fouling, which therefore result to higher operation and maintenance costs than conventional systems [5].

The membranes can be incorporated in the biological process in two ways:

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Table 1. Typical materials used for the manufacture of membranes [3].

Organic membranes		Ceramic membranes	
Cellulose acetate	CA	Titanium oxide (Titania)	TiO_2
Polyetherimide	Ultem	Zircon oxide (Zirconia)	ZrO_2
Polyacrylonitrile	PAN	Aluminiumoxide (Alumina)	$\gamma\text{-}Al_2O_3$
Polyethersulphone	PES		
Teflon			
Polyvinylidenefluoride	PVDF		
Polyethylene	PE		

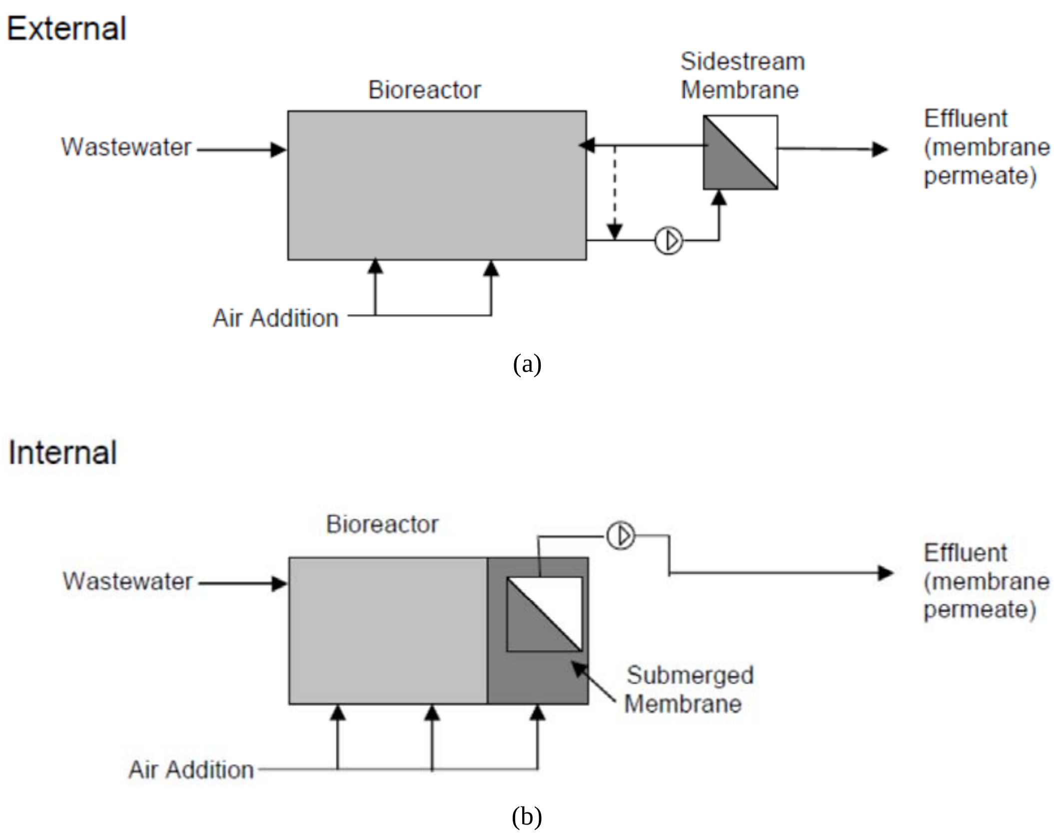

Fig. (1). Different configurations of the MBR process: (**a**) internal and (**b**) external membrane modules [4].

Internal

The membranes are submerged in an aerated tank and permeation takes place under vacuum application, towards the interior of the membrane Fig. (**1a**). Commonly used membrane configurations are hollow-fiber and plate and frame modules.

External (Side Stream)

The membranes are placed external to the reactor and sludge is recirculated through the (usually tubular) membrane elements, where permeation (separation of water) takes place inside-out Fig. (**1b**) [4].

The strict distinction between internal and external MBR is not maintained in practice, because in several applications there is a separate membrane compartment, with its own aeration and a circulation flow [6, 7]. A circulation through the membrane compartment, combined with aeration is more effective for filtration performance than aeration alone [8].

2.2. Recent patents on MBR Engineering

Membrane bioreactors combine membrane filtering technology and activated sludge biodegradation processes for the treatment of wastewater. In a typical membrane bioreactor system, immersed or external membranes are used to filter

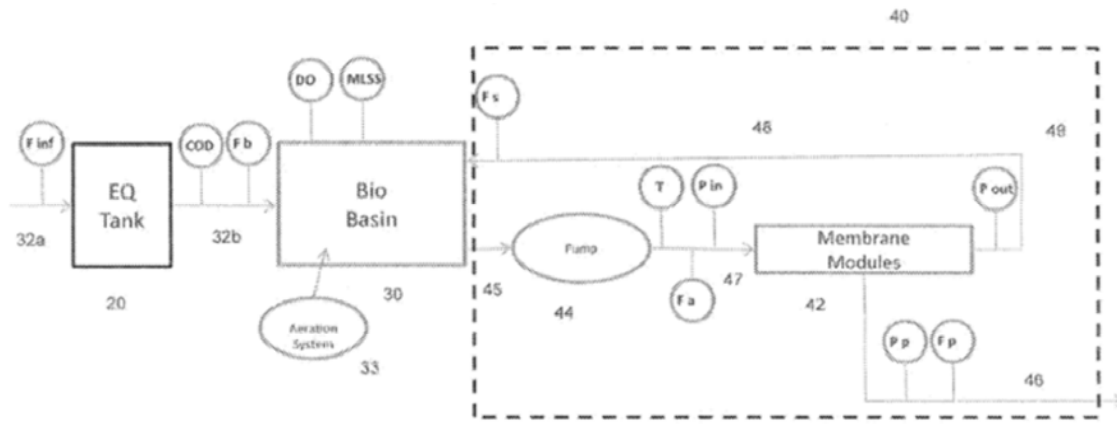

Fig. (2). Wastewater treatment operation with an external membrane bioreactor (eMBR) system adapted to employ or use microprocessor control systems [12].

an activated sludge stream from a bioreactor to produce a high quality effluent, as generally described for example, in U.S. Patents Nos. 7,879,229 and 8,114,293 for "Method of operating a water treatment plant with immersed membranes" [9, 10].

A recent patent with International Publication Number WO 2012/082967 A2 (21 June of 2012) entitled "Integrated biological treatment and membrane filtration for nutrient removal and advanced reuse" [11], is related to systems and methods for wastewater treatment and, in particular, for treating wastewater utilizing biological treatment and a post-treatment or polishing stage. The latter stage is dedicated to the reduction or elimination of membranes fouling potential.

In addition to organic carbon removal and nitrification, denitrification of the secondary effluent is foreseen, prior to filtering. Nevertheless, reduction of total phosphorous in the treated filtered effluent is provided by the addition of certain coagulants; the introduction of the appropriate chemical may take place either to the influent or to the secondary effluent. Furthermore, chlorine disinfection of the effluent may be carried out, resulting to a high quality effluent. A reverse osmosis unit may be employed for advanced filtration of the effluent; suspended solids concentration of the effluent is expected to be less than 15 mg/L.

Another recent patent with International Publication Number WO 2012/173988 A1 (20 December of 2012) entitled "Advanced control system for wastewater treatment plants with membrane bioreactors" [12], discloses an advanced control system for a membrane bioreactor based wastewater treatment plant. The control system comprises a membrane bioreactor (MBR) system and a microprocessor controller that receives signals corresponding to selected measured MBR parameters; therefore, one or more MBR operation parameters are calculated or estimated including Membrane Conductivity (Fxc) (the degree of permeability of a cellular membrane to certain ions; the reciprocal of the membrane resistance); and/or Oxygen Uptake Rate (OUR) (a parameter that can be used to evaluate the rate at which metabolic processes take place in activated sludge treatment processes with suspended sludge) for efficient monitoring of system performance. The microprocessor based controller compares one or more calculated or estimated MBR parameters to prescribed set-points or desired ranges controlling thus, the operation of one or more pumps and valves in the MBR system. This method aims to the adjustment of the cleaning cycle in the MBR system, the MBR flows in the MBR system, or the influent flow to the biological basin in response thereto. The present application claims priority from U.S. provisional patent application Ser. No. 61/496,275 filed Jun. 13, 2011, the disclosure of which is incorporated for reference herein.

Figure **2** is a schematic representation of a wastewater treatment operation with an external membrane bioreactor (eMBR) system adapted to employ or use the proposed control systems.

The microprocessor based controller functions, include the following: (i) receipt of signals corresponding to the measured MBR parameters from the plurality of sensors; (ii) calculation of the Membrane Conductivity (Fxc); (iv) comparison of the calculated membrane conductivity (Fxc) to prescribed setpoints; and (iv) start-up of a membrane cleaning cycle when membrane conductivity falls below minimum setpoint. The measured parameters include the temperature of the stream fed to the membrane modules or units; the flow rate of the stream into the membrane modules or units; the flow rate of the sludge stream wasted from the membrane modules or units; the flow rate of the permeate stream sucked out of the membrane modules or units; the pressure of the influent to the membrane modules or units; the pressure of the permeate residual from the membrane modules or units; and the pressure of the permeate flow out of the membrane modules or units.

3. CERAMIC MEMBRANES

3.1. Membrane Characteristics

Ceramic membranes offer significant advantages over polymeric or metallic membranes in several applications working under extreme operating conditions due to their intrinsic properties i.e., rigid porous structure, high-temperature resistance, high-chemical resistance to aggressive aqueous and organic media and insensitiveness to biological attack.

Table 2. Parameters of a ceramic membrane element, module and sealing [13].

Specification	Available Choice
Membrane material	Alumina, Zirconia, Titania, Silicon Carbide
Membrane area	from 0.0055 m^2 to 0.418 m^2
Shape of channel	cylindrical; flower-like; honeycomb-shaped
Membrane pore size	from 0.45 KD to 1.2 µm
Element length	from 250 mm to 1200 mm
Element diameter	12 mm to 40 mm
Module material	Stainless steel, Titanium and plastic
Sealing material:	Rubber, Silicon

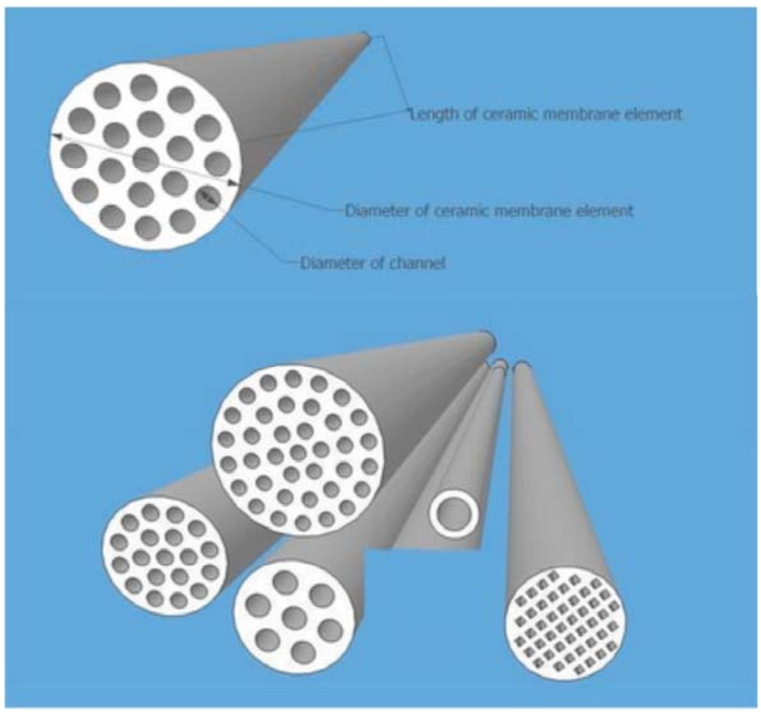

Fig. (3). Conceptual diagram of a ceramic membrane element, module and sealing [13].

Applications with a high potential for the implementation of ceramic membranes include: treatment of waste liquids and gases; liquid processing including drinking water, domestic water, food beverages; and product recovery in various industries ranging from micrometer-sized species (mineral particles, microorganisms, macromolecules, etc.), to nanometer-sized species (viruses, colloids, molecules, ions), up to filtration pretreatment before the application of other separation techniques including filtration through polymer membranes.

According to the particular conditions of an application, ceramic membrane elements come up with tailored parameters to meet specific requirements, such as: membrane material; membrane area; membrane pore size; element dimension; channel shape; the materials of module and sealing.

The characteristic parameters of a ceramic membrane element, module and sealing and the corresponding available materials are listed in the following Table **2** and are shown in Fig. (**3**) [13].

Typically, ceramic membrane can work under temperatures up to 300 ^{o}C (572 ^{o}F), pressures up to 2.6 MPa and pH ranges from 1 to 14.

Ceramic membrane elements are made up of a macroporous (>50 nm) inorganic material (with a tubular, multichannel, or monolithic geometry) supporting a multilayer porous ceramic structure exhibiting a non-deformable porosity with pore sizes ranging from macropores (>50 nm) to micropores (< 2 nm) as shown in Fig. (**4**) [13]. Fouling has been proved to be mainly affected by membrane's microstructure, surface roughness and pore sizes. Ceramic membrane with

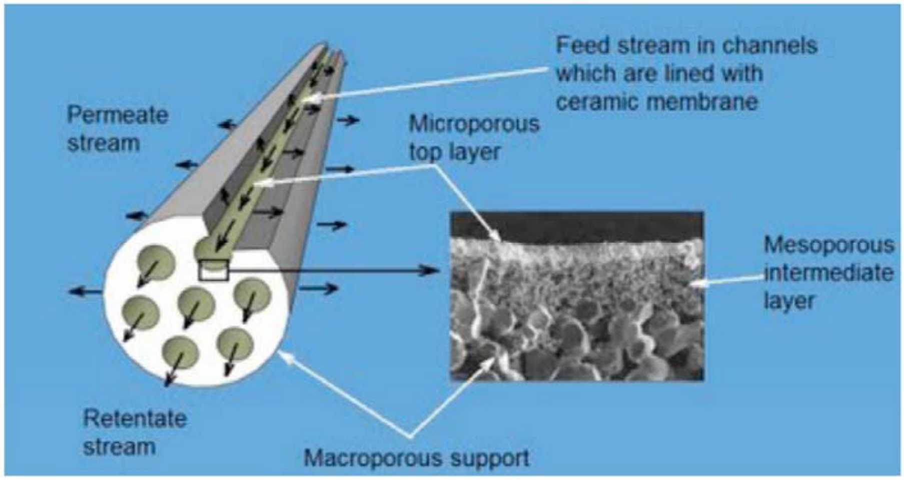

Fig. (4). Porous structure of a ceramic membrane element [13].

the roughest surface and biggest pore size (300 nm), presented the highest fouling potential with respect to the TMP profile [14].

Ceramic membranes are well-known to offer unique advantages due to their robustness, but their higher cost has limited their use in cost sensitive processes, such as the environmental related applications. Nevertheless, the cost reduction of the membrane modules and the corresponding housings, the need to accomplish stringent regulations and to give response to competitive markets have led the industrial sector to incorporate this technology for the treatment of wastewater in increasing rate. The ceramic membrane bioreactor technology is frequently an adequate alternative selection for high strength industrial waters of low flowrates, as it guarantees the reliability, robustness and stability of the process generally required by industrial applications [15].

The advantages of ceramic membranes have been recognized a long time ago. Process stability and minimum need for support and maintenance resources are the main features often asked by plant engineers and operators. As a result, the benefits offered by ceramic membrane systems such as flexibility in chemical cleaning, ease of use and general robustness and reliability, outweigh the energy penalty for many low-flow applications.

The major advantages of ceramic membranes can be summarized as follows:

- Resistance to extreme pH (1-14) and temperature (100 ºC) ranges: this stability is sometimes underestimated for MBR application since biological systems work at neutral pH and moderate temperature. However, extreme chemical cleaning can be crucial in industrial applications, where severe fouling can be often found requiring the implementation of aggressive cleaning chemicals.
- Chemically inert: ceramic membranes are resistant to chemical attack and are not affected by solvents, oxidants or corrosion.
- Long and reliable life time: average membrane-life can be much longer than organic membranes and therefore the membrane replacement cost is reduced considerably.
- Narrow and well controlled pore size distribution, in comparison to organic membranes, due to the specific fabrication process, based on the sintering of homogeneous size powders.
- Mechanical strength, which provides the operational advantage of not being subjected to compaction under relatively high pressures.
- High porosity and hydrophilic surface, resulting in high permeability.
- Negligible interaction between ceramic material and biomass: thus, the biofouling rate is minimized.
- Geometric simplicity: thanks to the diameter of the channels, membranes can handle highly viscous fluids, with high concentration of suspended solids, minimizing the need for extensive pre-treatment and security pre-filters. The ability to operate under a wide range of solids concentration (from low values up to 30 g/L) is especially important during the startup of the membrane bioreactor since there is not a requirement to apply low fluxes when the suspended solids content is quite low.
- Membrane cost/lifetime ratio competitive with organic membranes.

In addition to these advantages, the side-stream configuration has several implications that favor the installation of these membranes:

- Compactness and modularity: the intensive operation leads to lower membrane surface and more compact systems. The accessibility to the membranes is significantly better and the modules can be easily removed or opened facilitating thus the monitoring and maintenance daily operations.
- Cleaning in place (CIP): the chemical cleaning is easier, as the filtration circuit is easily isolated from the biological system. As a result, all maintenance operations are simplified.
- In those applications where operation is seasonal or when there is an intermittent process flow, the maintenance of the membranes is easier when the system is shut down [15, 16].

Ceramic membranes, despite their intrinsically superior properties, have not been widely adopted due to their high manufacture cost and rigid structures: their relatively high cost is due to the utilization of expensive raw materials for the fabrication of a complex multilayer system. Specific costs of ceramic membranes vary in a wide range, depending on module type and the pore size. In addition to cost, other drawbacks include the brittle nature of ceramic membrane material making them quite sensitive to mechanical shock conditions and the lower flexibility than polymers. Different thermal expansion of ceramic membrane and the module housing may cause problems with the sealing. Therefore, attention should be considered for choosing an appropriate gasket between the ceramic membrane and the housing. Furthermore, the weakness of ceramic membranes arises mainly from the manufacturing process, which makes it difficult to achieve a reproducible final product quality. Costs of organic membranes showed a sharp decrease in recent years leading to the assumption that a similar development for ceramic membranes may occur in the future. Moreover, higher fluxes for ceramic membranes will decrease the required membrane area for a given water flow. Longer membrane life time is another factor which may compensate the higher investment costs compared to organic membranes [17].

3.2. Ceramic Membranes in MBR Applications

In 1982, Dorr Oliver introduced an anaerobic MBR system for the treatment of industrial wastewaters [18]. Pilot study results indicated that the particular technology was promising for the treatment of high strength wastewaters assuming that appropriate improvements should be made regarding membrane efficiency. Since that time, a number of anaerobic MBR research and development studies have been completed [19]. The results of these studies and a recent work completed under a Water Environment Research Federation project [20] imply that ceramic membranes might be a good alternative process in the conventional anaerobic MBR application, due to the evidence of less membrane fouling problems and the ability to rigorously clean the membranes without deteriorating the membrane life.

The analysis of the operation of industrial MBR systems has shown that more than 60% of total daily cost of operation and maintenance works is due to sludge dewatering and management. Ceramic MBR is able to support high variations of MLSS, leading to weekly or even monthly sludge drainage and dewatering if necessary, so MBR operation becomes more flexible. The ceramic MBR, may operate at very low F/M ratio, resulting to almost zero sludge production under certain conditions.

In general, the optimum scenario for the application of ceramic MBR comprises one or more of the following situations:

- High water quality needs: as a result of the reliability on pore size distribution and the robustness of the filtration system, ceramic MBR guarantees high effluent quality and reuse possibilities [21].
- Difficult (to be treated, mainly industrial) wastewaters: MBRs are usually considered as the best technology for difficult wastewaters. Ceramic membranes make the MBR more robust to chemicals, solvents, temperature and other industrial attacks. That is especially important, when treating leachates, oily wastewater and slowly biodegradable wastewaters or biorefractory compounds.
- Small-medium installations: the robustness, minimum sludge production, compactness and much longer membrane life compensates for the higher energy requirements, expected for MBR systems
- Medium-high organic loads: in MBR systems operating costs are mainly linked to the volumetric loading, while in conventional systems, organic loading determines the required air supply. When treating small fluxes of high loaded waters, the additional energy input for filtration is not significant compared to aeration basin needs, and ceramic MBR technology becomes competitive.
- Decentralized urban wastewater treatment: small MBRs for the treatment and reuse of municipal wastewater in small communities require a robust process so that limited maintenance is required while, when maintenance is necessary, it is not need to be performed by highly qualified personnel. External ceramic membranes cover these pre-assumptions better than polymeric ones due to the longer lifetime, less likelihood for mechanical failure and the ability to withstand aggressive chemical cleaning conditions. Although currently decentralized systems do not represent the main market for ceramic MBRs, it is an interesting niche application for this technology [15].

A rather recent application of ceramic membranes is the dewatering of wasted activated sludge without the requirement for polymer addition. Handling and management of sludge is a rather costly process in activated sludge systems; membrane filtration coupled to biological treatment appears as an effective, simple alternative option requiring negligible space. For small wastewater treatment plants, ceramic membranes can be applied successfully for sludge treatment simultaneously to the biological stage [15].

3.3. Recent Patents on Ceramic Membranes Engineering

Excess sludge produced during the biological degradation of organic compounds in activated sludge processes, is usually wasted from the secondary sedimentation tanks and subjected to a number of treatment steps such as thickening, followed by anaerobic digestion, and dewatering. However, the final sludge product contains a large number of pathogens, heavy metals, residual odor and other harmful compounds requiring therefore, the implementation of appropriate advanced treatment measues and adequate management options for the disposal or utilization with minimum environmental impacts.

A recent patent with International Publication Number WO 2012/055257 A1, (5 March of 2012) entitled “Bioceramic useful as biological media filter material for air filtration and wastewater treatment, comprises sludge, kaolin and metal M or its oxide” [22], aims to deal with the problem of wasted sludge management providing a biological process where selective adsorption and digestion functions are taking place simultaneously in the same method.

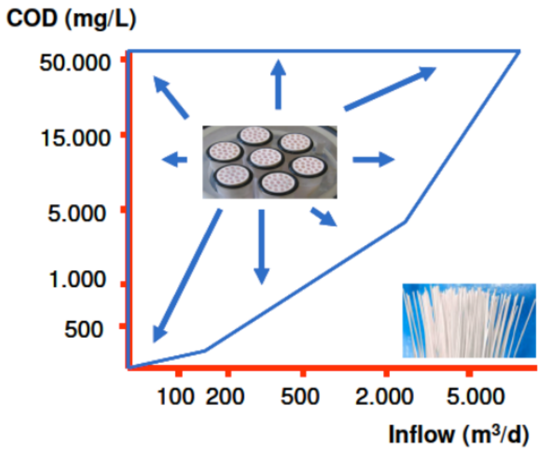

Fig. (5). Optimum scenario for the Ceramic Membrane Bioreactor process [15].

Towards that target, a bioceramic module with particular properties is proposed: the bioceramic is manufactured by mixing the following raw materials, by weight: 30-70 parts of certain sludge, 2-15 parts of kaolin, 1-5 parts of certain metal M or oxide of metal M. The bioceramic has a structure of amorphous Si-CM and a specific surface area of 400-500 m^2/g. According to the proposed method, sludges from paper-making wastewater, food industry and wastewater treatment plants and kaolin might be used as raw materials, followed by the addition of crop husk. After the mixing of the raw materials, activation of the porous bioceramic particles or powders is carried out by the utilization of oxygen-free sintering microwave process to obtain the final bioceramic membrane. The so produced material has the ability of adsorbing benzene, phenol, multiple hydrocarbons, dimethyl sulfide and thioether malodorous substances; in addition, the bioceramic product may adsorb phosphate ions and oxygen rich compounds such as nitrates, nitrites from the aqueous phase, contributing therefore to the efficient nutrient removal from wastewater. In adition, the bioceramic material presents such characteristics due to its unique manufacturing method that can provide the required carbon and mineral doses to the activated sludge microorganism, enhancing thus the bacteria activities towards pollutants removal. Due to its properties, the proposed bioceramic can be used as biological media filter material for air filtration and wastewater treatment.

An additional apparatus and method for alternative aeration using a ceramic membrane have been reported, allowing the biological treatment to be performed in an intermittent aeration tank. The method consists in the combination of an anaerobic tank with a number of intermittent aeration tanks and the selective influent feed according to the operation status of the intermittent aeration tanks.

The above disclosure with International Publication Number US 2013/0015124 A1 (17 January of 2013) entitled “Apparatus and method for alternative aeration-effluent wastewater treatment using ceramic membrane” [23], foresees an alternative aeration-effluent wastewater treatment, which allows the aeration of influent and the continuous discharge of effluent treated water through two intermittent aeration tanks. In each intermittent aeration tank, ceramic modules are placed providing two independent lines: an air injection line and a treated water discharge line, as shown in Fig. (**6**) and Table **3**.

The proposed ceramic membrane system includes:

- An anaerobic tank for influent phosphorus (P) removal and denitrifying nitrite-nitrogen and nitrate-nitrogen;
- Two intermittent aeration tanks, operating under subsequent anaerobic and anoxic conditions according to the following schedule: under the aerobic condition, organic nitrogen and ammonia nitrogen are converted into nitrite

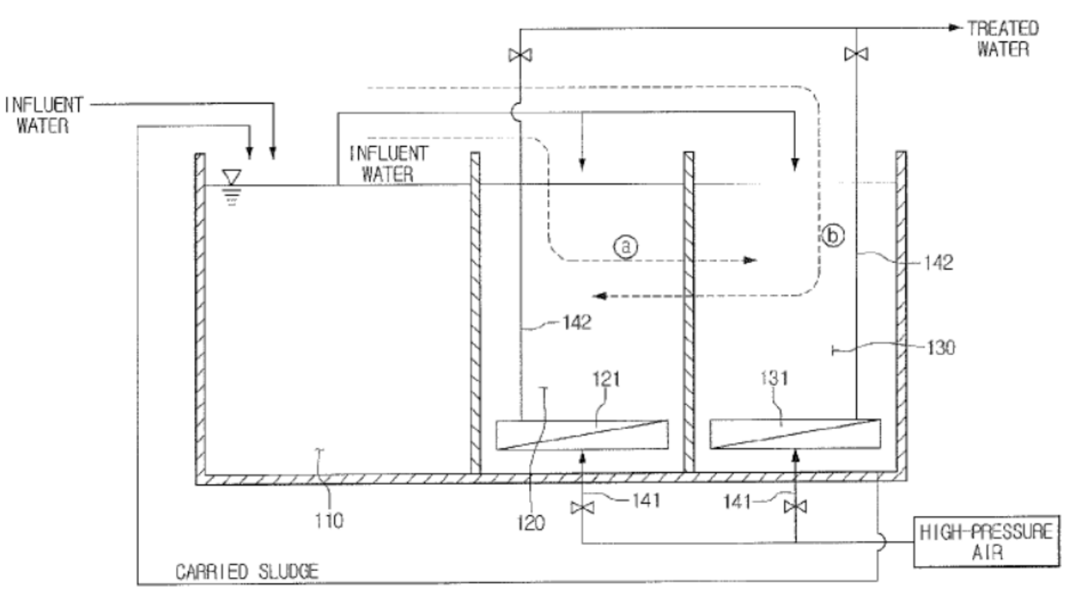

Fig. (6). Apparatus for alternative aeration-effluent wastewater treatment using ceramic membrane; **a**) Flow of Influent Water when first intermittent aeration tank (120) is under aerobic condition, **b**) Flow of Influent Water when second intermittent aeration tank (130) is under aerobic condition [23].

Table 3. Detailed Discription of Main Elements [23].

Main Elements	
110: anaerobic tank	120: first intermittent aeration tank
121: first ceramic membrane	130: second intermittent aeration tank
131: second ceramic membrane	141: air injection line
142: treated water discharge line	

and nitrate nitrogen while influent phosphorus is taken by phosphorus-storing bacteria; under anoxic condition, nitrate nitrogen is reduced into nitrogen gas;

- Two ceramic membrane modules placed at the lower part of the two intermittent aeration tanks producing high quality treated water.

4. CURRENT & FUTURE DEVELOPMENTS

The membrane biological reactor (MBR) configuration has proven to be an efficient and convenient alternative to conventional activated sludge for the treatment of several industrial wastewaters when treatment performance is an important consideration. In a typical membrane bioreactor system, immersed or external membranes are used to filter the mixed liquor from a bioreactor delivering a high quality effluent.

Polymeric organic membranes are today most commonly applied in water and wastewater treatment applications, with a wide variety of membrane materials, pore sizes, pore size distributions, membrane configurations and production processes. The main benefit for the implementation of organic membranes is associated to manufacturing costs. However, Ceramic Membranes offer significant advantages over polymeric or metallic membranes in several applications working under extreme operating conditions due to their intrinsic properties.

MBR systems utilizing ceramic membranes are being consolidated as a reliable technology for industrial applications due to the intrinsic benefits, such as less membrane fouling rate and the ability to rigorously clean the membranes using aggressive chemicals without affecting the membrane life. Moreover, ceramic membranes may be a good alternative in the anaerobic MBR applications. Furthermore, the maintenance of all the properties of the system and, at the same time, the minimization of the manufacturing cost and the optimization of the operational conditions for minimizing energy consumption represent an ambitious challenge towards their future applications

This review summarizes Recent patents on Engineering that disclose key data for both the functioning of MBRs through the incorporation of ceramic membranes.

Specifically, an advanced control system for a membrane bioreactor based wastewater treatment plant is disclosed, comprising a microprocessor based controller that receives signals corresponding to selected measured MBR parameters and calculates or estimates one or more MBR parameters such as Membrane Conductivity, Oxygen Uptake Rate (OUR) aiming to the efficient control of MBR performance.

Furthermore, an integrated biological treatment coupled to membrane filtration for nutrient advanced removal and effluent reuse is proposed utilizing a post-treatment or polishing stage. The proposed process may provide a reduction or elimination of fouling rate of membranes due to the incorporation of the post-treatment stage.

An important and promising technique is the manufacturing of a bioceramic material with selective adsorption and digestion properties; this material may be useful as a biological media filter material for air filtration and wastewater treatment. The incorporation of this bioceramic module is useful in treatment plants fed by industrial wastewaters, while it can contribute to the production of lower amounts of wasted sludge.

Moreover, a method for alternative aeration-effluent wastewater treatment using ceramic membrane is proposed, allowing the biological stage to be carried out in an intermittent aeration tank; the process is based on the combination of an anaerobic tank with a number of intermittent aeration tanks. In the proposed technique, influent flow is selectively directed to the reactors, according to the operation status of the intermittent aeration tanks.

Overall, the proposed methods represent new and recent patented developments for the utilization of ceramic membranes in MBR systems for efficient wastewater treatment, overcoming the well-known problems of polymeric membranes; nevertheless, the full scale implementation of these processes represents a challenge for their extended application in various processes, over a wide range of influent properties and operation status.

CONFLICT OF INTEREST

The authors confirm that this article content has no conflicts of interest.

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