Membrane Bioreactor in China: A Critical Review

Olusegun K. Abass[1,2], Xing Wu[1], Youzhi Guo[3] and Kaisong Zhang[1,*]

[1]*Institute of Urban Environment, Chinese Academy of Sciences, Xiamen, 361021, China*

[2]*University of Chinese Academy of Sciences, Beijing 100049, China*

[3]*Desalination Branch of China Water Enterprises Confederation, Beijing 100028, China*

Abstract: The application of membrane bioreactor (MBR) for wastewater treatment and reuse has received ample scientific and industrial considerations due to its potential for improved effluent quality, strong anti-shock loading capacity, less residual sludge and small footprint. Particularly in 2006, since the first operation of a large-scale MBR in China, it's acceptability for wastewater reclamation had nearly tripled in recent years. This rigorous review work focus on the overall operational chain for the MBR system with attention placed on the Chinese MBR market, fouling characteristics, energy costs, and application trend in China. Recent developments in membrane materials, compositions, and properties are summarized. Roles of cake-layer formation, extracellular polymeric substances (EPS) and hydrodynamic effect on membrane fouling are critically assessed. Lastly, future perspective regarding long term development of MBR applications in China and potential areas of fouling mitigation are identified.

Keywords: Wastewater treatment, MBR market, Industrial applications, Membrane bioreactor (MBR), Membrane fouling.

1. INTRODUCTION

The concept of submerged membranes bioreactors (MBRs) was first introduced by Kazuo Yamamoto, father of MBRs in 1990's [1]. It later became one of the best available technologies in wastewater treatment. The exponential growth of membrane bioreactor technology and market is currently creating a stir globally as the prospect for wastewater reuse and reclamation continues to build up pressure on existing facilities.

In 1994, bench work on membrane bioreactor (MBR) began in China [2] following an initial review work on MBR published by Chen [3] in 1991. Sequel to that, a group of scholars from the Dalian Institute of Chemical Physics, Chinese Academy of Sciences published the very first English journal article on membrane bioreactor using a fermenter and a flat pervaporation module for ethanol fermentation [4]. Since then, the emergence and application of MBRs across China have witnessed several landmarks, especially in the treatment of polluted surface water (such as river and rainwater runoff), municipal and industrial wastewater [5]. Starting from the early 1990s, the growth and application of MBR technology in China have undergone different maturation stages from a laboratory-scale operation capacity to large-scale applications with current production capacity reaching 10^6 m^3 /day in 2015 [6].

The rapid development of MBR in China is partly due to increasing environmental demand, policy formulation and market incentive. According to reports, the government of China proposed in its 12th National Five-Year Plan to improve the amount of wastewater reclaimed from roughly 10% in 2010 to around 15% in 2015 [7]. This projection will undeniably enhance the relevance of MBR technology which boast of cleaner effluent production and meets the high quality standard requirement recently introduced by China Environmental Protection Administration (EPA) [8]. Compared with conventional activated sludge (CAS] processes, MBR possess better configuration which allows the integration of an advanced membrane separation with an activated sludge system, thus giving rise to an independent control of the hydraulic residence time (HRT) and sludge retention time (SRT) while retaining a high concentration of sludge biomass in the reactors. In addition, the MBR process features the ability to reduce large quantities of residuals [9], smaller footprint, better effluent quality [10] and induces the potential to recover wastewater resource which is a demerit of the CAS process [11]. To date, various statutory laws and regulations have been released by the Ministry of Environmental Protection, China to heighten the input of MBR technologies for wastewater reuse and to standardize membrane module production among others [12, 13].

In this review, we attempt to present an in-depth analysis of the overall operational chain for the MBR system focusing on academic research, market development, membrane materials and engineering,

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[*]Address correspondence to this author at the Institute of Urban Environment, Chinese Academy of Sciences, Xiamen, 361021, PR China;
Tel.: +86-592-6190540; Fax: +86-592-6190540; E-mail: kszhang@iue.ac.cn

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and industrial applications of MBR in China. In the following, the fundamental aspects of MBR analysis viz-a-viz energy cost will be reviewed. The individual processes taking place in an MBR from changes in membrane type to effluent output quality will be described. Lastly, the main conclusions and future perspectives are identified.

2. MBR SITUATION IN CHINA

2.1. Academic Research Progress of MBR in China

Notable increase in MBR research outputs from Chinese authors written both in Chinese (Figure **1a**) and English (Figure **1b**) emerged about the year 1995. Since then, a huge amount of research work became available owing to the insight obtained from their publications. The databases of Web of Science, Weipu, Wanfang, China National Knowledge Infrastructure (CNKI), Elsevier Science Direct, Taylor & Francis, American Chemical Society (ACS), Wiley Interdisciplinary Reviews (WIREs), Springer Link, EI village, Pub Med, and Google Scholar were used for the analysis. Moreover, a peak in Chinese MBR publications was reached in 2010 after which the number of articles published declined. This decline is not unusual as many Chinese researchers became more attracted to English publications which in recent years have been the major trend in many academic institutions across China. Besides, the sharp increase of MBR English paper publications is simply a reflection of the potentials of MBR prospect in China.

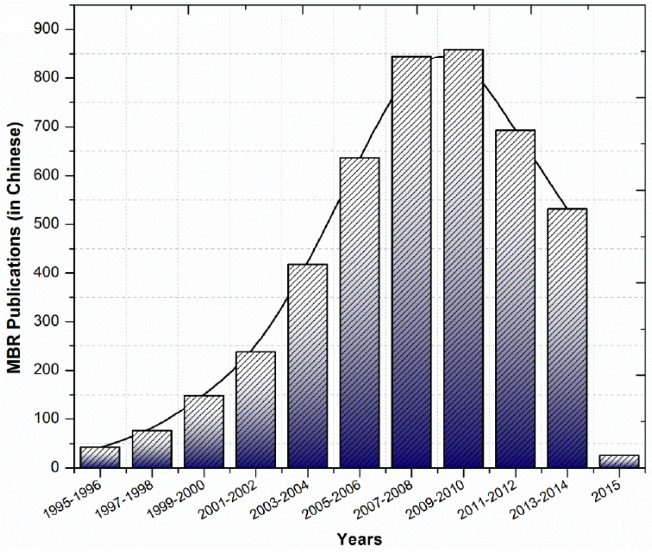

Figure 1a: Peer-reviewed articles (in Chinese) on membrane bioreactor by Chinese authors, from 1995 to 2015 (up to April 2015).

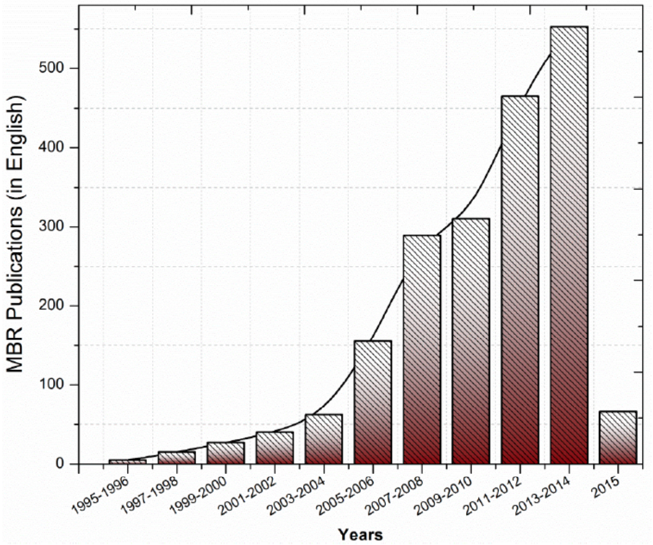

Figure 1b: Peer-reviewed articles (in English) on membrane bioreactor by Chinese authors, from 1995 to 2015 (up to April 2015).

The increasing number of international MBR patents in the last ten years has completely revolutionized the prospect of MBR technological application in China as shown in Figure **2**. This is clearly an indication of the involvement of highly trained and qualified personnel contributing their quota to the development of MBR technology. Many of these projects were initiated by the China Ministry of Science and Technology (MOST) under the national 9th [14] and 12th “Five-year-plan” [7]. In recent years, increasing growth of membrane and MBR module manufacturing companies in China generated fierce competition amongst major players stimulating the rise of novel MBR applications. Starting from year 2006, less than 20 Chinese MBR patents were available. At this time, the number of MBR researchers began increasing and government contribution to the MBR industry amplified. Also, the number of available private and government owned companies operating full-scale MBR processes were on the rise. After that period, a geometric increase in patented publications was witnessed, reaching a total number of about 250 at the end of 2013.

The decreasing trend of patents amount seen in the year 2014 could be similar to that observed between year 2010 and 2012. This argument was put forward as a result of the aggressive publication of patents within few months into the year 2015 and the constantly evolving MBR market in China as predicted by several market indices [15, 16].

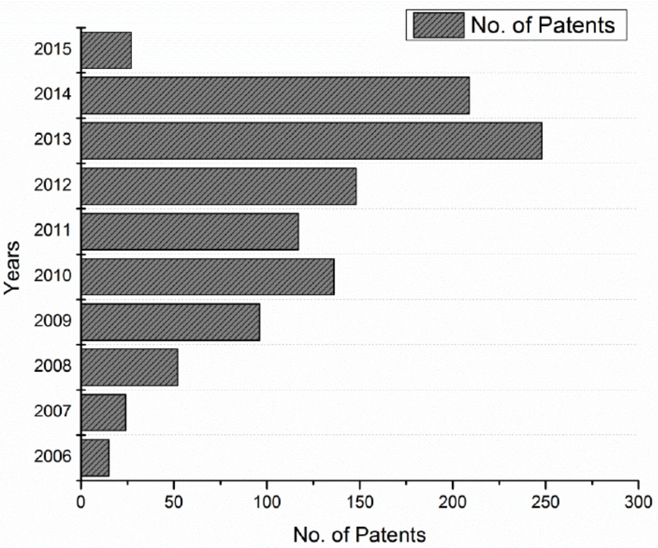

Figure 2: Internationally patented MBR publications in China since 2006 (up to April 2015).

2.2. MBR Market Analysis

2.2.1. MBR Market Share

Globally, MBR systems has been implemented in more than 200 countries with an average growth rate of approximately 12.7%, and global marketization of MBR is expected to reach US$1.3 billion in 2015 [17]. However, according to recent report by Global Industry Analyst (GIA], the MBR market growth is forecasted at US$888 million by the year 2017, using municipal and industrial end-user segments indices [18]. This decline in capital projection could be as a result of the economic recession experienced across the globe due to current fall in oil prices. Steady growth of MBR market is however expected as the economy of some major player-countries remains unaffected (*e.g* China and USA).

GIA further predict that China and Brazil will attain the fastest growth rates within the given forecast period. Sales and application of MBR have generally grown faster than the GDPs of countries installing them, as the case in China. In another related research carried out by Transparency Market Research, analysts predicted that by the end of 2019, MBR market will reach a value of US$2.5 billion [19]. The global membrane bioreactor market is currently reported to be growing at a compound annual growth rate (CAGR) of 14.5% from 2013 to 2019 with municipal wastewater treatment segment dominating at CAGR of 21.7% in terms of volume [19]. By 2012, Asia Pacific region recorded nearly 39% of the total market share while Europe held 19%.

2.2.2. MBR Installations and Market Prediction

Data of the largest MBR plants in a survey of MBR installations across the globe between year 2014 and 2017 are shown in Figure **3**. A comparatively high rate of expansion of MBR installation across the globe place China as the world's largest MBR market, with over 1.4 million m^3/day installed capacity to date and has 730,000 m^3/day of additional capacity in the planning stages. Using the data equation obtained from Business Communications Company Inc (now BCC Research) and Srinivasan [16, 20], we predict the global MBR market growth value from 2006 to 2020 as shown in Figure **4**. Moreover, we compare forecast figures from Transparency Market Research with our projection (using fitting data from Frost and Sullivan (F&S) report [15]), the analysis show significant correlation with predicted MBR market value for the year 2019. Hence, we safely predict that the global market cost of MBR will reach an estimated value of US$ 2.7 billion by the year 2020.

Report from Frost and Sullivan research service on China MBR outlook, predicted that the Chinese MBR market is expected to reach $1.35 billion in 2017 at an impressive compound annual growth rate of 28.9% [15]. China, Asia Pacific and the Middle East are anticipated to be the leading markets in the upcoming years. Municipalities which are important large buyers of MBR technology will be the driving forces in these regions.

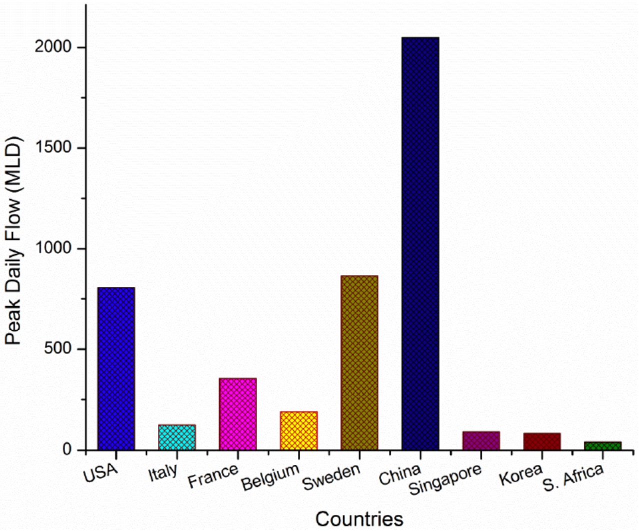

Figure 3: World's Largest MBR plants in mega liters per day [Sampled number of plant(s) in each country starting from USA include; 3, 1, 1, 1, 1, 13, 1, 1, in that order between year 2014 and 2017] (Source: The MBR Site [21]).

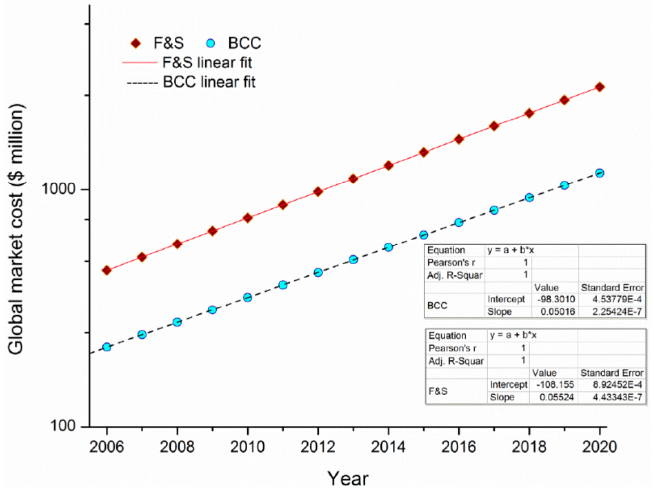

Figure 4: MBR global market value projection (data equation obtained from Frost and Sullivan (F&S), Business Communication Company report and Srinivasan, 2007).

2.2.3. MBR Market Advancement in China

In a world of rapidly changing environmental, industrial and market demands, an analysis of MBR technological advancement in a country like China will inevitably be a valuable tool. The evolution of MBR market in China began in 1998 with the construction of a municipal based wastewater treatment facility located in Liaoning, a Northeastern province of China [22]. Following this event, a rapid expansion of the technology occurred leading to development of some large scale MBR project which entered the Chinese market around 2004. The impetus for MBR market growth has been attributed to water scarcity especially in northern regions of China [23] and the promotion of novel technologies stimulating practical applications of MBR. Evidence of this was demonstrated during the popularization of MBRs utility across the eutrophic basins of Lake Dian and Tai adopted by Yunnan and Jiangsu Provinces respectively. Another factor that has contributed to MBR market growth is the geometric increase in water prices owing to newly formulated policies for water conservation [24].

According to the International Water Association (IWA) reports in 2010, MBR membrane module products is currently increasing by a factor of 3 to 5 per year, and the market itself growing exponentially at a rate between 11.5 and 13%. As novel products continue to enter the market and fiercer competition among key MBR contractors are being witnessed, the growing trend in MBR marketization is expected to be sustained in the coming years. Likewise, more stringent regulations on wastewater reuse strategies will inevitably surge up creating wider market for MBR development in China and rest of the world.

2.3. MBR Plants in China

In China, a number of municipal and industrial scale MBRs with a minimum design capacity ≥10,000 m^3/day has being in operation since 2006 [25]. The largest operating capacity of newly constructed plants is expected to reach ≥ 2 × 10^5 m^3/day between the year 2015 and 2017. The major players in this development include the Fuzhou Yangli Wastewater Treatment Plant (WWTP) Phase IV, installed by United Envirotech Ltd, Wuhan Sanjintan WWTP constructed by Origin Water Technology Co., Ltd, Macau WWTP situated in Macau Special Administrative Region and the Shunyi WWTP located in Beijing. Completed MBR project in year 2014 include; the Macau WWTP and the Chengdu Qingbaijiang WWTP (Upgrade), both having capacity of 189,000 m^3/day and 20,000 m^3/day for municipal and industrial wastewater treatment and reuse respectively. The plants were constructed by GE Water Process Technologies and Origin Water Technology Co., Ltd in that order.

As at November, 2014, a Joint Venture (JV) agreement between Beijing Drainage Group Co. Ltd (a state-owned-enterprise) and United Envirotech Ltd (UEL) was signed in the amount of RMB120 million (US$ 19.1 million) to set up a new state-of-art membrane manufacturing facility with an initial membrane area capacity of 2 ×10^6 m^2 assembled into membrane modules. Memstar Pte Ltd and UEL were the major membrane supplier and subsidiaries in the deal. Table **1** illustrates representative large-scale MBR installations for wastewater treatment in China since 2006. Approximately 56% of these plants are used for the purpose of domestic/municipal wastewater treatment while 37% are utilized in industrial wastewater treatment. The remaining 7% proportion is employed in treatment of polluted surface water. Recently, Origin Water supplied its membrane products for two prominent MBR projects in China: one for municipal wastewater treatment with a designed capacity of 200,000 m^3/day and the other for surface water treatment with a 50,000 m^3/day capacity. Interestingly, Chinese MBR providers such as Beijing Origin Water have emerged as the chief threat to GE Water's global MBR position.

Table 1: Summary Large-Scale MBR Plants and Membrane Suppliers for Wastewater Treatment in China Since 2006

Location	MBR Facility	Wastewater type	Capacity (m^3/day)	Membrane Suppliers/ MBR contractor*	Year Commissioned /(Expected)	References
Beijing	Miyun WWTP	Municipal	45,000	Mitsubishi-Rayon/Origin water	2006	[26]
Inner Mongolia	Jingqiao Power Plant WWTP	Municipal	31,000	GE Zenon /Lucency	2006	[25]
Guangdong	Xiaohu Island Petrochem. Industrial Park	Petrochemical	10,000	Asahi Kasei /Novo	2006	[27]
Guangdong	Huizhou Petrochem. Engr. Corp.	Petrochemical	25,000	Asahi Kasei /Novo	2006	[25]
Hainan	Hainan Petrochemical Engr. Corp.	Petrochemical	12,000	Asahi Kasei /Novo	2006	[25]
Beijing	Huairou WWTP	Municipal	35,000	Asahi Kasei/Origin water	2007	[28]
Beijing	Beixiaohe WWTP (Phase I)	Municipal	60,000	Siemens Memcor /Siemens	2007	[29]
Heilongjiang	Harbin Petrochemical Engineering Corporation	Petrochemical	10,000	Memstar/Novo	2007	[25]
Guangdong	Huizhou Tianxin Petrochem. Engr. Corp.	Petrochemical	15,000	Asahi Kasei/ Novo	2007	[27]
Tianjin	Tianjin Airport Waste water system	Industrial	30,000	Tianjin Motimo	2007	[25]
Beijing	Wenyu River Water Treatment plant	Polluted River water	100,000	Asahi Kasei/Origin water	2007	[25]
Beijing	Pinggu WWTP	Municipal	40,000	Asahi Kasei/Origin water	2008	[30]
Sichuan	Chengdu Banknote printing complex wastewater system	Pulp and paper mill	10,000	Mitsubishi-Rayon	2008	[25]
Sichuan	Sichuan Wenchuang WWTP	Municipal	10,000	Memstar/Novo	2009	[30]
Shanxi	Liulin WWTP	Municipal	30,000	Asahi Kasei/Beijing E&E	2009	[30]
Jiangsu	Wuxi Chengbei WWTP	Municipal	50,000	Origin water/ Origin water	2009	[31]
Jiangsu	Wuxi Shuofang WWTP	Municipal	20,000	Mitsubishi-Rayon/ Origin water	2009	[32]
Jiangsu	Wuxi Meicun WWTP	Municipal	30,000	GE/BMEDI	2009	[33]
Jiangxi	Jiujiang Petrochemical Engr. Corp.	Municipal	12,000	Asahi Kasei /CSEP	2009	[30]
Hubei	Shiyan Shending River WWTP	Municipal	110,000	Origin water /Origin water	2009	[28]
Jiangsu	Jiangsu Dafenggang WWTP	Pharmaceutical Industry	10,000	Novo/Memstar	2009	[30]
Jiangsu	Jiangsu Tianxing Binjiang WWTP (Phase II)	Municipal & Chemical	30,000	Memstar/Novo	2009	[27]
Jiangsu	Kunshan WWTP	Municipal	15,000	GE/BCEED	2010	[30]
Guangdong	Guangzhou Jingxi WWTP	Municipal	100,000	Memstar/Novo	2010	[34]
Yunnan	Kunming No.4 WWTP	Municipal	60,000	Origin water /Origin water	2010	[35]
Jiangsu	Wuxi Hudai WWTP	Municipal	21,000	Origin water /Origin water	2010	[31]
Beijing	Wenyu River Water treatment plant[Phase II)	Polluted River water	100,000	Mitsubishi-Rayon / Origin water	2010	[6]
Jiangsu	Kunshan banknote printing and minting	Pulp and paper mill	9,000	Kubota /Poten Enviro	2011	[6]
Xinjiang	China Huadian Corporation Changji power plant	Power plant	12,480	Asahi Kasei/CHEC water	2011	[6]
Yunnan	Kunming Luolonghe rainwater treatment plant	Polluted River water	50,000	Origin water /Origin water	2011	[6]

(Table 1). Contd…..

Location	MBR Facility	Wastewater type	Capacity (m3/day)	Membrane Suppliers/ MBR contractor*	Year Commissioned /(Expected)	References
Inner Mongolia	Hohhot Togtoh industrial park WWTP (upgrade)	Pharmaceutical Industry	20,000	Mitsubishi-Rayon /United water	2011	[6]
Jiangsu	Yangzhou Qingshan WWTP (Phase II)	Municipal & Chemical	40,000	Hyflux /Hyflux	2012	[6]
Zhejiang	Orient Huaqiang textile dyeing and printing	Textile dying	14,000	Tianjin Motimo/ Tianjin Motimo	2012	[6]
Beijing	Qinghe WWTP (Phase III)	Municipal	150,000	Origin water /Origin water	2012	[6]
Liaoning	Liaoyang Central District WWTP (Upgrade)	Municipal	80,000	Memstar /Novo	2012	[6]
Jiangsu	Wuxi Chengbei WWTP (phase IV)	Municipal	20,000	Kubota /CERI	2012	[31]
Ningxia	China Shenhua Ningxia Coal Industry Group	Coal chemical	36,000	Canpure/WBD	2013	[6]
Yunnan	Kunming No. 10 WWTP	Municipal	150,000	Origin water /Origin water	2013	[6]
Beijing	Daxing Huangcun WWTP (Upgrade)	Municipal	120,000	Origin water /Origin water	2013	[6]
Inner Mongolia	China Shenhua Group Erdos plant	Coal liquefaction	9,840	Toray/Poten Enviro	2013	[6]
Jiangsu	Nanjing Chengdong WWTP (Phase III)	Municipal	150,000	Origin water /Origin water	2013	[6]
Sichuan	Chengdu Qingbaijiang WWTP (Upgrade)	Fine chemicals	20,000	GE/BEWG	2014	[6]
Jiangsu	Nanjing East WWTP (Phase 3)	Municipal	150,000	Origin water /Origin water	2014	[21)
Shandong	Yantai Taoziwan WWTP	Municipal	150,000	Origin water /Origin water	2014	[21)
Jilin	Jilin WWPT (Phase 2)	Municipal	150,000	Origin water /Origin water	2014	[21)
Hunan	Changsha 2nd WWTP	Municipal	140,000	Origin water /Origin water	2014	[21)
Hunan	Zhuzhou Longquan WWTP (Phase III)	Municipal	100,000	Tianjin Motimo / Tianjin Motimo	2014	[6]
Macau	Macau WWTP	-	189,000	GE Water / GE Water	2014	[21]
Fujian	Fuzhou Yangli WWTP (Phase IV)	Municipal	200,000	Memstar /United Enviro	2015	[6]
Jilin	Jilin City WWTP (Phase I, upgrade)	Municipal	150,000	Origin water /Origin water	2015	[6]
Hubei	Wuhan Sanjintan WWTP (Upgrade)	Municipal	200,000	Origin water /Origin water	2015	[21]
Shanxi	Jinyang WWTP [Phase 1）	Municipal	120,000	Origin water /Origin water	2015	[21]
Beijing	Shunyi WWTP	-	234,000	GE Water	2016	[21]
Guangdong	Zhuhai Qianshan WWTP	-	200,000	Origin water /Origin water	2016	[21]
Macau Special Administrative Region	Macau WWTP	-	210,000	GE Water	2017	[21]

– data unavailable, WWTP – Wastewater treatment plant, Engr. Corp. – Engineering Corporation.

3. ENGINEERING APPLICATIONS OF MBR

3.1. Industrial Wastewater Treatment

Application of membrane bioreactor in the treatment of industrial wastewater is becoming popular as shown in Table **1**. Current hot topics include the application of MBR for the treatment of hospital wastewater [36], oily wastewater [37], landfill leachate wastewater [38], food-processing wastewater [39, 40], pharmaceutical wastewater [41, 42], petrochemical wastewater [43], printing and dyeing wastewater [44]. In addition, MBR technology is widely used in high concentration organic wastewater treatment, and industrial wastewater containing refractory and intractable organics. A comprehensive survey of MBR applications show that the main application areas of MBR in industry focuses on food, textile and chemical industrial wastewater all of which accounts for a proportion of 18.8% each (Figure **5**]. MBR treatment of mechanical and paper industrial wastewater are still being considered as alternative. Tianjin Airport Waste water System was the first industrial MBR project with a capacity of 30,000 m^3 /day, indicating the fast growth of MBR applications in China. The success of these projects initiated a blooming era for the application of super large-scale MBRs in China as seen in Table **1**.

Lately, the most popular direction for MBR application in China is the treatment of food industrial wastewater [45, 46]. Due to the teeming population of China, a large amount of wastewater are daily released by food processing industries which contains high concentrations of COD and suspended solid (SS) [47]. Using anaerobic membrane bioreactor (AnMBR), COD removal efficiency is generally greater than 90% considering the characteristics of high organic influent usually found in food industrial wastewater [48, 49]. This renders AnMBR a suitable treatment option. Recalcitrant pollutants present in heavy industrial discharges might be improperly removed using conventional activated sludge system leading to serious ecotoxicological and environmental damage to host organisms. Therefore, the introduction of MBR technologies in the Chinese industrial sector has become a plausible development. In recent times, the total large scale MBR design capacities installed (including municipal, industrial and surface wastewater) across China has exceeded a million cubic meter per day accounting for approximately 1.4% of the total wastewater treatment capacity in China (Figure **6**).

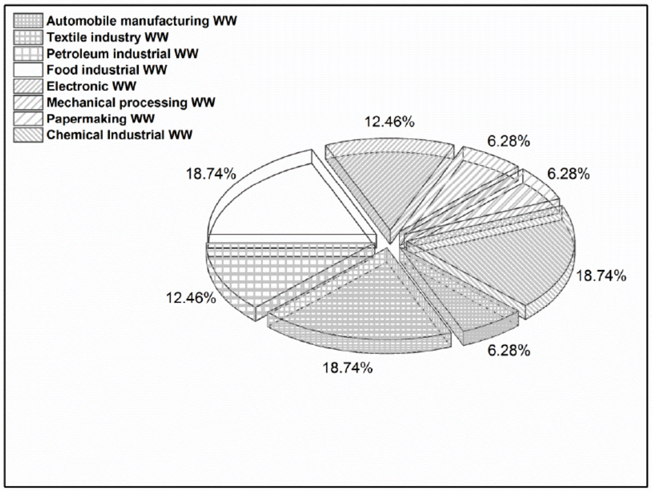

Figure 5: MBR treatment capacity of various industrial wastewaters in China (Source: China water net survey data).

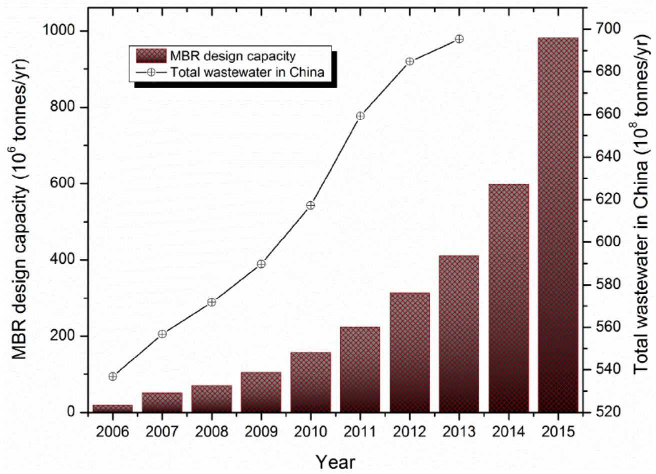

Figure 6: Full-scale capacity development of MBR applications across the wastewater treatment industry in China since 2006.

3.2. MBR Fundamental Research Designs

For any MBR design, useful knowledge about the wastewater components is of great importance due to substantial variation in wastewater characteristics [47]. Measurable features of industrial wastewaters includes chemical oxygen demand (COD), biochemical oxygen demand (BOD), suspended solids (SS), ammonium nitrogen, heavy metals levels, turbidity, pH, color, and biological parameter. Depending on the target pollutants, different types of MBR designs have been utilized for wastewater treatment some of which include; hybrid growth membrane bioreactor (HG-MBR) for treatment of mixed wastewater [50], membrane distillation bioreactor (MDBR] which has the potential to tap on waste heat generated in industries to produce high quality product water [43], and submerged membrane bioreactor (SAMBR) used especially for oily wastewater treatment [51].

Others include biofilm membrane bioreactor [BFMBR) employed to improve the process of denitrification [52], biomass separation membrane bioreactor (BSMBR) [53], and forward osmosis membrane bioreactor (FOMBR) for production of high quality permeate [54]. At the same time, two other innovative processes such as the membrane enzyme bioreactor (MEBR) for hydrolysis of special organic substances such as olive oil and whey protein and membrane pervaporation bioreactor (MPBR) for volatile organic wastewater treatment [55, 56]. Although, study of all the types of MBR has been carried out, the BSMBRs and SAMBR have successively been used in commercial applications and the remaining types of MBR are still at the level of lab-scale research.

3.3. Large Scale MBR Designs for Industrial Wastewater Treatment

Full scale MBRs are commonly run conservatively to avoid operational crisis. This is because, elaborate utilization of MBR technology are not completely immune to challenges such as increase of transmembrane pressure (TMP) (a common indicator of membrane fouling), high-energy consumption rate, inadequate understanding of the complex biological processes and membrane filtration phenomena which are characteristics low points confronting further development of the MBR process [57]. To combat these issues, a number of MBR configuration have been put to use including the submerged, external and airlift MBR design (Figure **7**).

These designs, specifically the external MBR configuration are tailored towards a direct hydrodynamic control of membrane fouling and offer the advantages of easier membrane replacement and high flux production but at the expense of high energy consumption [58]. The submerged MBR design tends to overcome this disadvantage because it involves lower operating cost and less rigorous cleaning [59]. The airlift MBR configuration modifies the deficiencies of the previously discussed designs using the side-stream air-lift principle to yield a robust framework for energy optimization [60]. In China, all three configurations have been used for treatment of municipal wastewater, landfill leachate, industrial and sewage wastewater [61, 62], though airlift configuration has been little used. Until now, the industrial design capacities of MBR applications across China continue to increase compared with the overall MBR design capacities (for municipal, industrial and surface water) which is also gaining increasing popularity as shown in Figure **8**. A steady growth in industrial MBR design by 3% is expected before the end of 2015.

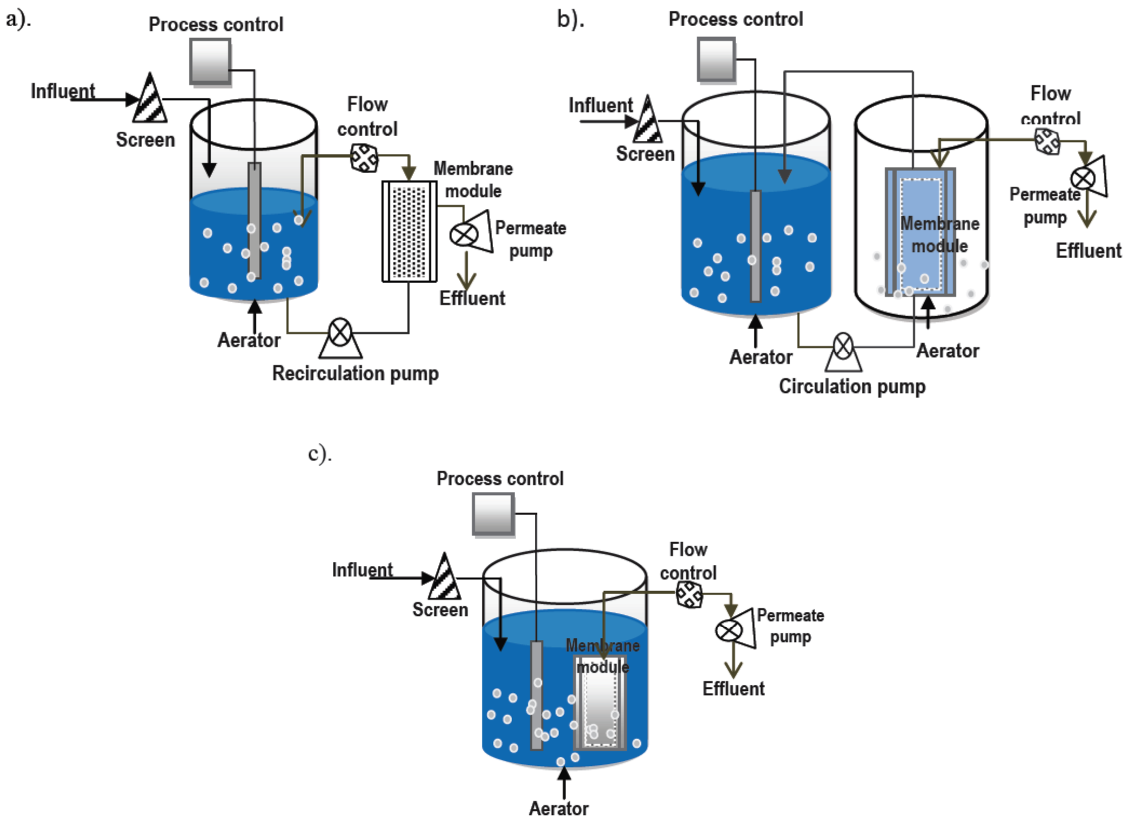

Figure 7: Schematic representation of membrane bioreactor designs applied in China (a) external design (b) airlift design (c) submerged design

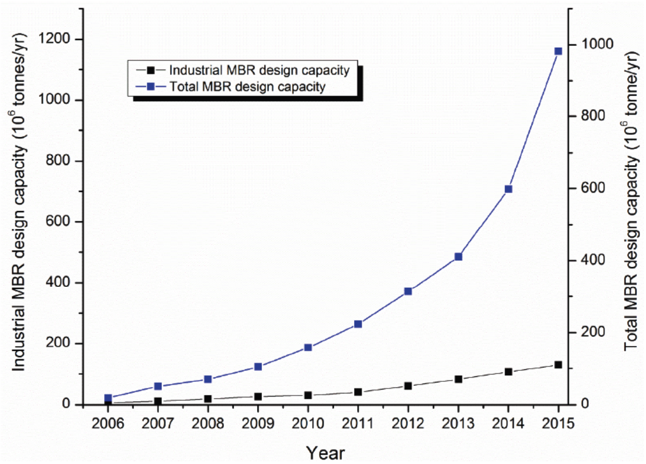

Figure 8: Full scale industrial MBR design capacity in relation to total MBR design capacities installed in China since 2006

4. MEMBRANE PROPERTIES, FUNDAMENTALS AND FOULING CHARACTERISTICS

4.1. Membrane Materials

The quest for better effluent quality and space requirement in a rapidly developing country like China have set the pace for improvement of membrane materials and design in the modern day MBR system. Determination of suitable membrane characteristics such as the hydrophobicity/hydrophilicity, permeability, nominal pore sizes, porosity, surface charges and roughness has been extensively investigated since the 1990s [63]. Other properties like oleophobicity, amphiphilicity, and specific pollutant rejection ability of membranes are parameters believed to improve MBR performance [64, 65]. Polymeric ultrafiltration membranes of Polyvinylidene fluoride (PVDF), Polyacrylonitrile (PAN) and Polyethersulfone (PES) have been found to possess different characteristics with respect to their different affinities for extra-polymeric substances (EPS) [66]. This variation in properties tend to improve the fouling resistance of the membranes with PES been the least resistant and hence, more research are currently performed to mitigate this disadvantage due to the cheaper cost of PES membranes.

Another advancement made in the development of membrane for MBR is the fabrication of a composite microfiltration membrane, prepared by blending polyvinylidene fluoride (PVDF) and hydrophilic graphene oxide (GO) nanosheets showing high permeability over a long period of time and less accumulation of extracellular polymeric substances [67]. Recent development of advance filtration membranes like the polymer-based membranes, ceramic or inorganic membranes, and nano-composite membranes for MBR applications have led to the re-structuring of the MBR industries [68, 69], and thus providing more ground for the rational use of the technology in China. Inorganic membranes, such as aluminum, zirconium, and titanium oxide, show better hydraulic, thermal, and chemical resistance when used as filters in MBR, though the cost implication is quite high.

Yet, the prevailing membrane materials used for MBR are the halogenated hydrocarbon polymers like polyvinylidene fluoride (PVDF), due to their superior properties in chemical tolerance, comparably low cost, mechanical strength and biological stability [70]. PVDF film materials are generally produced by wet spinning and thermally induced two-phase separation modes (phase-inversion method) [71, 72]. Wet spinning is a relatively mature technology, accounting for 60% of the MBR market capacity.

4.2. Membrane Module Configuration and Manufacturers

Membrane configurations including flat-sheet, tubular, and hollow-fiber which are the earliest module design have been employed in municipal and industrial wastewater treatment facilities in China (Figure **9**). Factors including high density packing, permeability and brittleness of different membrane module configurations have led to an integral development of MBR in China. The Daya Bay Guangzhou petrochemical project launched in 2000 by Asahi Kasei was the first known membrane project produced by thermally induced technique and their product subsequently entered the Chinese market in 2005. The most recent of all is the introduction of a relatively new membrane bioreactor (MBR) with membrane accommodating carrier (MAC arrier), designed by GE's China Technology Center to help industries meet stringent water discharge requirements and enable greater water reuse, while achieving cost savings and increased operating efficiency [73].

MBR with MACarrier (a carbon-based product) combines a highly efficient MA Carrier with a special type of membrane called the Zee Weed 500D membranes (produced by GE Zenon, China). This integration enhances the removal of recalcitrant organics and toxicity and can achieve a chemical oxygen demand (COD) reduction of more than 50 percent compared to a MBR without MA Carrier. One of advantage of this novel technology is the ability of the MA Carrier to be biologically regenerated in the bioreactor which helps to cut down on operational costs. Equally, increasing MBR requirement and high import costs of membrane products effectively promoted the development of native MBR manufacturers as shown in Table **2**.

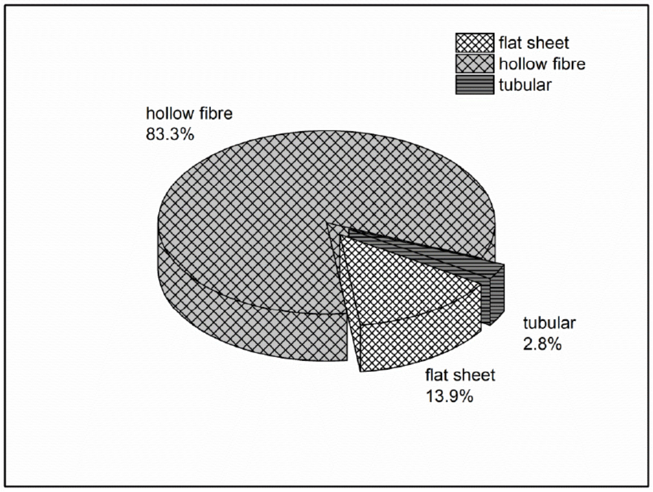

Figure 9: MBR module configuration utilize in China (Source: China water net survey data).

4.3. Energy Consumption Analysis in MBR

Given that energy demand contributes significantly to the running cost of an MBR system, it is therefore important to optimize the process energy consumption to make the technology more competitive [57]. Aeration comprises more than 50% of the total energy requirement in MBR process. Verrecht *et al*., [74], conducted a model-based experiment to determine the key parameters required for optimizing the energy demand in MBR without compromising the nutrient removal. The study revealed that decreasing membrane aeration and sludge retention time (SRT) were most beneficial for minimizing cost on energy consumption, while increasing the recirculation flow led to improved total nitrogen removal but also a gradual deterioration in total phosphorus removal. Energy consumption rates between 0.2 and 2.4 kWh/m^3 were reported for submerged operation of municipal and industrial wastewater treatments, and the aeration cost accounted for more than 80%, whereas energy consumption generated by permeate suction/permeate back-flush accounted for less than 3% of total energy consumption (75].

The most demanding apparatus regarding energy consumption is the coarse bubble aerator followed by the mixer. A trial operation was carried out on a MBR pilot plant to decipher the main energy consuming process [76], the study reveal that no significant difference was observed when the mixing and re-circulating pump time was reduced. In another study, Zhang *et al*., [77], successfully applied an experimental optimum bubbling regime in an MBR system to achieve an effective and economical means of reducing the

Table 2: Main Membrane Manufacturers in China and their Corresponding MBR Module

Manufacturer	Membrane Material*	Module	Pore Size (μm)
SINAP, China	PVDF	Flat sheet	0.1
Shanghai Zizheng Environ., China	PES	Flat sheet	0.2
Jiangsu Lantian Peier, China	PVDF	Flat sheet	0.08-0.3
Zheda Hyflux Hualv, China	PP	Flat sheet	0.06-0.14
Liaoning Univ. of Petrol. Chem. Technol., China; Toray, Japan	PVDF	Flat sheet	0.1-0.4
Jiangsu Jiuwu Hi-Tech, Nanjing Univ. of Technol., China	Ceramic	Tubular	0.02-0.5
Litree, China	PVDF & PVC	Hollow fiber	0.02
Zhaojin Motian, China	PP & PVDF	Hollow fiber	0.2
Hangzhou Creflux, China	PP	Hollow fiber	0.1-0.2
Beijing Scinor, China	PVDF	Hollow fiber	0.1
Tianjin Motimo, China	PVDF	Hollow fiber	<0.1
Origin Water, China	PVDF	Hollow fiber	0.3
Donghua Univ., China	PVC	Hollow fiber	0.2
Zheda Hyflux Hualv, China	PP	Hollow fiber	0.06-0.27

*PVDF – polyvinylidene fluoride, PVC – polyvinyl chloride, PP – polypropylene, PES – polyether-sulfone cost of energy consumption during non-aerating period to prevent fouling. The energy consumption for side-stream systems is usually two orders of magnitude higher than that of submerged systems, and aeration accounts for only about 20% of the total operation costs, regardless of whether the system is used for municipal or industrial wastewater treatments [78]. Generally, the external configuration provides more direct hydrodynamic control of fouling, and offers the advantages of easier membrane replacement and high fluxes but at the expense of frequent cleaning and high energy consumption (of the order 10 kWh/m^3) [58]. Several distinct advantages of submerged configuration are their much lower energy consumption and fewer rigorous cleaning procedures, as well as the milder operational conditions due to the lower tangential velocities.

4.4. Hydrodynamic Relations

Since about 50-70% of energy consumption in MBR operation is related to aeration, most of the recent researches now focus on improving aeration efficiency through a systematic understanding of the hydrodynamic conditions suitable for aeration reduction. In a submerged MBR, an increase in aeration produces a corresponding rise in shear stress on the membrane surface, and eliminates the need for a recirculation pump [79]. Aeration not only provides oxygen to the biomass, but also maintains the solids in suspension and scours the membrane surface to reduce membrane fouling [80]. However, increasing aeration intensity or flow velocity could increase energy cost and disrupt sludge flocs, producing small-size particles and releasing more extracellular polymeric substances (EPS) that negatively impact membrane fouling [81, 82]. In addition, under conditions of intense aeration, solutes and colloids could be incorporated onto the membrane surface becoming a major foulant. This occur because, the resistance of solutes and colloids is not effectively reduced by merely increasing the shear stress [83, 84].

With respect to hydrodynamic settings, an increase in cross flow velocity (CFV) result in increased shear force causing a resultant reduction in floc sizes. This effect creates a fluctuating increase and decrease of the critical flux below which severe fouling (removal and irremovable) in a given MBR filtration system will not occur [85]. Typically, CFV values of 2–3 m/s are sufficient to prevent the formation of reversible fouling and have no obvious effect on microbial activity in external MBRs [86].

4.5. Membrane Fouling in MBR

The complex nature of membrane foulants and activated sludge handling remains a puzzle till date. It represents one of the most challenging issues constraining the extensive applications of MBRs. Considering the complexity of activated sludge, it is not surprising that the fouling behavior in MBRs is more complicated than that in most membrane applications. Sludge characteristics are defined by certain parameters including quantity of dissolved oxygen (DO), hydraulic retention time (HRT), sludge retention time (SRT) and the food to micro-organism (F/M) ratio [87-90]. Understanding the process optimization of these parameters can assist in the modification of activated sludge and could indirectly decrease membrane fouling. All the parameters involved in the design and operation of MBR processes affect membrane fouling and this factors have hinder the fast commercialization of MBR in some sensitive wastewater producing industries.

Three fouling stages were proposed by Zhang *et al*., [91] which comprise an initial rise in transmembrane pressure (TMP), a long-time weak rise in TMP and a sharp increase in the TMP differential rate also known as TMP jump [92]. However, some studies have revealed that the main cause of fouling in MBR is related to sludge cake formation on membrane surface, absorption of colloids within/on membrane, spatial and temporal changes of foulant composition during long-term operation (for instance, the change of bacteria community and biopolymer components in the cake layer), and deposition of sludge flocs onto membrane surface [93-97].

Extracellular polymeric substances (EPS) and their sub-groups, such as transparent exopolymer particles (TEP), self-assembly gels (SAG), biopolymer clusters (BPC) and soluble microbial products (SMP) are usually present in the feed water of most wastewater treatment facilities at varying concentration depending on the pretreatment technique applied [98-102]. Factors governing membrane fouling by EPS and its precursors (TEPs) follows a complex pathway and commonly involves macromolecule–surface associations (Figure **10**). These interactions are determined by the membrane physiochemical properties (hydrophobicity, nanoscale roughness, and surface charges), water chemistry (concentrations of divalent cations, ionic strength, and pH), and hydrodynamic conditions such as the cross-flow velocity and initial permeate flux [103].

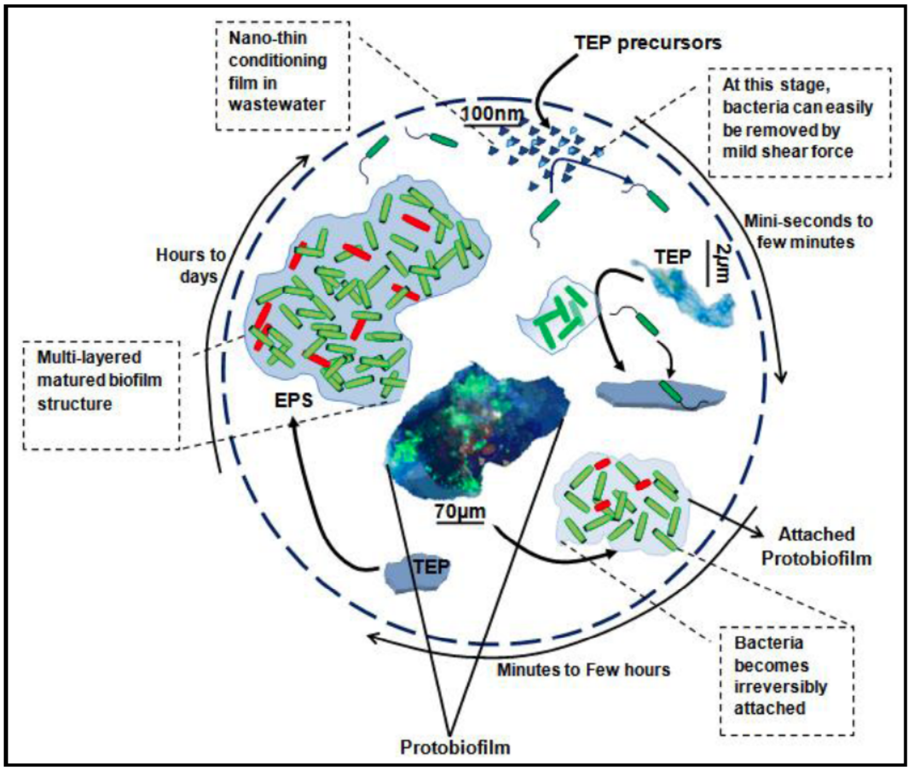

Figure 10: Illustrative pathway showing the developmental stages of transparent exopolymers (TEPs) and Extracellular polymeric substances (EPSs) leading to formation of biofilms and its implication for membrane fouling in MBRs (adapted from Bar-Zeev *et al*., [104]).

4.2.1. Fouling Process Control in MBR

Enormous research works are being conducted to combat/prevent membrane fouling in MBR. These include: (a) thorough understanding of the process optimization and operating conditions [105]. Operating parameters that significantly influence membrane fouling consist of the aeration intensity [81, 106, 107], temperature [108, 109], organic loading rate [110], ratio of suction and non suction time [111], and the filtration modes [112, 113]. (b) Improvement on mixed liquor filterability. Several chemical and biological compounds like diatomite, powdered activated carbon (PAC) and zeolite have been added to MBR to modify the filterability of mixed liquor. An investigation conducted by Yang *et al*., [114] show that diatomite addition is a reliable and effective approach for both membrane fouling mitigation and improvement in pollutants removal. The MBR system with diatomite addition of 50 mg/L enhanced the removal of COD, TN and TP by 0.9%, 6.9% and 31.2%, respectively, as compared to the control (without diatomite addition).

Similarly, reduction in membrane fouling was observed when PAC and zeolite was added to mixed liquor. Addition of PAC decreased the content of extracellular polymeric substances (EPS) in microbial cells, increased the floc-size distribution and prevented apparently high viscosity of the mixed liquor while zeolite addition resulted in increased membrane scouring and decreased the supernatant organic matters of the mixed liquor [115-119]. Other control mechanisms are (c) Early detection of membrane fouling using Surface-Enhanced Raman Spectroscopy (SERs) [120-122], and (d) Membrane modification to improve the antifouling properties [123]. To enhance the antifouling characteristics of the membrane used in MBR process, a number of means have been proposed including; plasma-induced tethering of sugar moieties [124], ammonia or carbon-dioxide plasma treatment [125, 126], surface modification by sequential, photo-induced graft polymerization of acrylic acid [127, 128], and the immobilization of poly(N-vinyl-2-pyrrolidone) on membrane surface [129-131].

However, the propensity for membrane fouling has been said to increase with increasing nanoscale roughness of thin film composite (TFC) membranes due to the lowering of the energetic barrier required for particle deposition [132]. Current research reveals a critical range of roughness scale and suggests the possibility of utilizing modified membrane surface morphology for membrane fouling mitigation [133].

4.2.2. Engineering Control of Fouling in MBR

Due to problems associated with bio films formation in MBR, it is necessary to undertake certain mechanical and chemical cleaning procedure to prevent the membrane surface or pores from being blocked by pollutants and to stabilize the water flux and rejection performance of the membrane. This assists in extending the lifespan of the membrane. In practice, applying appropriate pretreatment to feed water and chemically or biochemically modifying the mixed liquor is a key step to preventing clogging of MBR membranes. Engineering controls of membrane fouling in membrane bioreactor is in four stages: First, air scrubbing; water backwash; chemical cleaning to prevent pore blockage; and a restorative chemical cleaning technique which include sub-situ and ex-situ cleaning (Figure **11**). MBR fouling control regularly take place by pulse aeration in-situ cleaning and non-cleaning for membrane maintenance in order to achieve water quality assurance and extend membrane life. Fouling control measures mentioned above is more focus on lab-scale and pilot plant-based operation. Though, the engineering controls described here have seen few applications in large-scale MBRs in China [134]. More work are currently ongoing for transfer of pilot scale fouling controls to industrial level applications as seen in a recent work by Monclús *et al*. [135].

5. CONCLUSIONS AND PERSPECTIVES

In this paper, a critical analysis of MBR applications in China is described. Much progress has been made in the last ten years and it can be deduced that MBR technology proved to be a very efficient tool for wastewater treatment and reuse in China.

1. MBR was successful developed and proved to be one of the best available technologies for wastewater treatment and reuse in China in the last 10 years.
2. Application of MBR in China is expected to thrive in the coming years due to the prevailing market situation, tighter environmental laws and the global water stress, particularly in Northern China where water crisis has been recurring in recent years.
3. The ongoing clamp-down by government on industries and companies producing hazardous materials will stimulate actions in the systematic use of MBR to meeting demanding environmental standards. Due to these stringent regulations, a significant increase in MBR plant capacity and a widening of application areas will likely emerge in the nearest future.
4. Potential areas for MBR application include food-related wastewater like poultry, aquaculture, and other farm-reared animals. Also, more hospital/pharmaceutical wastewaters which may contain a wide variety of microbial pathogens and viruses will be areas of research interest.

However, MBR technological applications are still laced with challenges including:

1. Applications of MBRs in small city, town and countryside. With the constant urbanization in

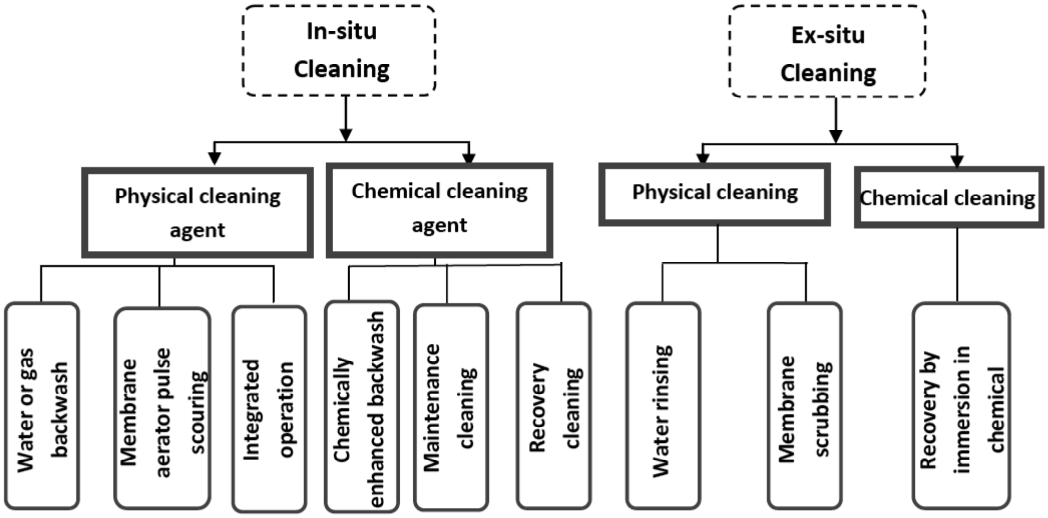

Figure 11: Schematic diagram showing classification of engineering fouling control strategies

China, the water environment gets more and more attention in town and small city, even in countryside. The applications and management of MBR, especially the long term operation of MBR should be an area of focus for sustainable development.

2. Energy consumption and conservation. Cost associated with the energy demand in MBR is quite high accounting for more than 50% of the total operating cost. This could be a hindrance to the expansion of the technology. Energy consumption could be reduced by enhancing the aeration efficiency such as decreasing membrane aeration and SRT based on specific conditions to minimize cost on energy consumption for fouling prevention. Another option is exploring different membrane material design as described earlier in this review paper.

3. Inadequate understanding of the process optimization for large-scale MBR applications. Fundamental information on issues such as running cost, energy, operational design, and manufacturing cost of MBR systems for wastewater treatments are issues that needs urgent attention. Additionally, pressure decay tests cannot be used for flat sheet membranes hence, improved process automation and control is arguably one of the key areas for the future of MBR technology.

4. Membrane fouling. The concept of membrane fouling in MBR is a critical limiting factor affecting the widespread application of MBR for wastewater reclamation. Fats, oil and grease remain a concern for MBRs due to the naturally hydrophobic nature of the membranes. High organic loadings at the MBR feed water are one of the main hurdles to overcome. Upcoming studies should focus on rigorous identification and characterization of membrane foulants (including the bacteria community of the foulants, chemical and biological components of foulants) and other areas such as comprehensive mitigation means using early fouling detection mechanisms.

5. The challenging aspects of MBRs are by far incomparable with the promising prospects it holds. Hence, long-term development of MBR will focus more on improving membrane fouling resistance, clogging of either membrane channels or aerators, cost/energy reduction and development of antifouling membrane materials. Additional effort will be seen in areas of improved membrane air scour efficiency, use of MBR for portable water production (debatable). Moreso, aspects where effluent sludge discharge handling and reclamation requirement exist, will see improvement thereby promoting the development of technologically acceptable and economically feasible treatment alternatives.

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