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Perspective

Making Waves: Zero Liquid Discharge for Sustainable Industrial Effluent Management

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Abstract: Zero liquid discharge (ZLD) aims to minimize liquid waste generation whilst extend water supply, and this industrial strategy has attracted renewed interest worldwide in recent years. In spite of the advantages such as reduced water pollution and resource recovery from waste, there are several challenges to overcome prior to wider applications of ZLD. This study will examine the main processes involved in ZLD, and analyze their limitations and potential solutions. This study also differs from past reviews on the subject, by providing a summary of the challenges that were found light of in prevalent studies. To fulfill the sustainable vision, future research that can bridge the gap between the theoretical study and industrial practice is highlighted.

Keywords: zero liquid discharge; industrial; membrane; brine; resource recovery; advanced oxidation process

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1. Introduction

The development of society and the bloom of industrial production, which consume a large number of raw materials [1], is placing increasing pressure on freshwater resources worldwide. According to the data from the Commission for Environmental Cooperation, low-income countries use 8% of their water for industrial use, while this number goes up to 59% in high-income countries [2]. The discharge of industrial effluent wastewater has posed threats to the environment and human health. To achieve a win-win situation of water conservation and pollution control, reclamation of the industrial effluents is attracting the attention from academic and industrial communities, with zero liquid discharge (ZLD) technologies that conform to this standpoint rising [3,4].

ZLD aims at eliminating any liquid waste leaving the plant or facility boundary, with the majority of water being recovered for reuse [3]. Nevertheless, this ambitious strategy has long been criticized by the high capital cost and intensive energy input because of the huge gap in the quality between the industrial effluent and reclaimed water for non-potable or recreational purposes [5]. Industrial effluents always contain various kinds of salts and refractory organic pollutants at elevated concentrations. While traditional wastewater treatment technologies such as coagulation and biological processes may contribute to decontamination, advanced treatment units are required to refine the treated water to meet the criteria for ZLD. Recent progress in reverse-osmosis (RO) has highlighted the viability of ZLD in the management of industrial effluent as compared to the early systems based on stand-alone thermal processes [4,6], and continuous innovation of technologies such as electrochemical and novel membrane-based processes provides opportunities to expand the applicability of ZLD at a reduced energy cost [7,8]. In 2020, the global ZLD market size has registered 6.37 billion USD, which is projected to reach 11.77 billion USD in 2028 [9]. The huge demand in chemical and petrochemical industries is one of the major factors driving ZLD market growth. Key players profiled in this field include GE Water & Process Technologies, Veolia Water Technologies, Aquatech International LLC, etc.

This article therefore aims to underscore the critical processes/units for ZLD deployment in sustainable industrial effluent management and to discuss the challenges that result from the mismatch between current research and demand in industrial applications. On the basis of the analyses, perspectives to facilitate the future research of industrial ZLD processes are provided.

2. Abatement of Organic Pollutants in ZLD

Abatement of organic pollutants was not an emphasis in early stage of ZLD as the industrial practice was mainly confined to the cooling system, in which the blowdown stream contains high hardness and total dissolved solids (TDS) but little organics [10]. More recently, the wastewater from core production and auxiliary units as well as domestic use has been included in the ZLD process. The dilute organic wastewater (e.g., streams from domestic use) is sent to a biological treatment system prior to the ZLD system while organic recovery and/or primary treatment units [11] would be first implemented if the feed contains organics of high concentrations. Taking coal chemical industry, an emerging niche for ZLD application, as an example, the organic wastewater contributes to 40–60% of the total [10], resulting in the installed capacity of a biological treatment system being around half that of a ZLD system. It should be noted that the biological treatment system is always termed "pretreatment unit", which is excluded from a water reuse and ZLD system (Figure 1).

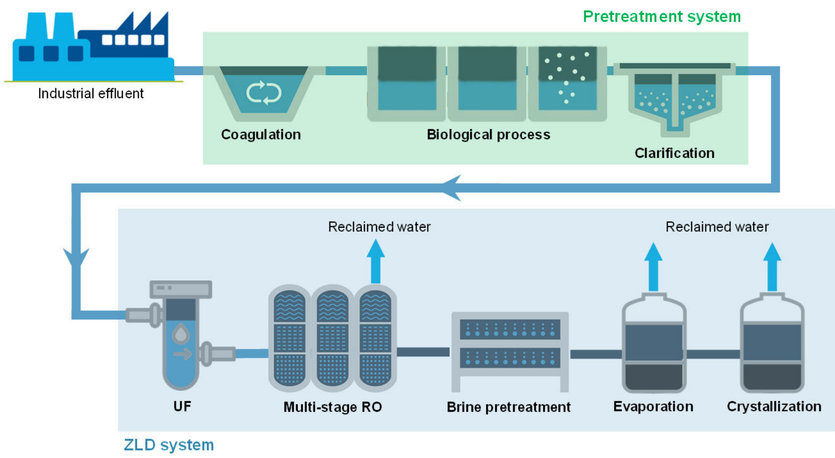

Figure 1. Typical processes to achieve zero liquid discharge (ZLD) in management of industrial effluent.

Biological processes such as activated sludge [12], biological aerated filter [13], and moving bed biofilm reactor [14] are widely used to remove the biodegradable organics from the industrial wastewater. While the effluent quality can be further improved via the integration of microfiltration and ultrafiltration in biological processes [15], the residual low molecular weight neutrals in the permeate are among the principal drivers that cause fouling of the downstream ZLD systems. Chemical precipitation/coagulation that is implemented to remove hardness ions (Ca^{2+} and Mg^{2+}) and silica (SiO_2) has shown good performance in removing residual organics from the feed of ZLD systems [11]. Recent innovations in coagulation (e.g., electrocoagulation) potentially decrease the operating cost from USD 0.5/m^3 to < USD 0.3/m^3 at a lower volume of sludge yield though more research activities are needed to address the low removal of non-polar, micromolecular substances that exhibit low affinity with the metal (oxy)hydroxide precipitates [16]. Rational adsorbents (e.g., functionalized carbon materials to induce hydrophobic and electron donor–acceptor interactions) can remove organics that have low Taft σ* constants and high steric effects [17]. With regards to the trade-off between selectivity (e.g., binding energy > 200 kJ/mol) and ease of regeneration, there has been recent interest in developing low-cost adsorbents that have high adsorption capacity whilst can be used as fuel for final disposal (e.g., lignite activated coke) [18].

Compared to the coagulation and adsorption technologies that separate and concentrate the refractory organics in a waste stream, advanced oxidation processes (AOPs) involve the use of strong oxidants (e.g., hydroxyl radicals, $^{\bullet}OH$) to break down organics, ideally to CO_2 and H_2O. The importance of AOP units in ZLD also relates to the abatement of organics in the brine from the high-pressure RO systems. Limitations in conventional pretreatment options (e.g., low biological activities in a hypersaline environment, and competitive adsorption of major ions) necessitate the use of AOPs in ZLD regime. The Fenton and Fenton-like processes that are based on the donation of electrons to H_2O_2 to produce $^{\bullet}OH$ can powerfully degrade refractory organics [12]. While the resultant coagulation by ferric (oxy)hydroxides is beneficial to removal of organics, the yield of chemical sludge is criticized. Ozonation is also widely used in degradation of organics in the brine with the mechanisms related to the direct oxidation by molecular O_3 and indirect oxidation by $^{\bullet}OH$ that are produced upon O_3 decay. The radical yield is boosted in the presence of H_2O_2 (i.e., peroxone) or catalysts (i.e., catalytic ozonation) [19]. Note that although reactive oxygen species are the main strong oxidants in Fenton and ozonation processes, the abundant Cl^- in the brine may induce the transformation of oxidants, resulting in the reactive chlorine species ($ClO^{\bullet}$, $Cl^{\bullet}$ and $Cl_2^{\bullet-}$) playing a more important role in abatement of organic pollutants [20].

3. Desalination and Water Recovery in ZLD

The core unit of a ZLD system may include multi-stage membrane processes (that are intended to recover water and to further concentrate the brine) followed by thermal treatment [12]. The feed of ZLD is typically treated by ultrafiltration (UF)/RO dual-membrane processes, in which UF is applied to decrease the turbidity and the sludge density index (SDI). A regular UF/RO dual-membrane system can recover 60–75% of the effluent from the pretreatment system, with the integration of a proprietary technology, high-efficiency RO (HERO), resulting in an overall water recovery > 90–95%. To alleviate fouling and scaling in the membrane system, the HERO process is operated at elevated pH with use of pretreatment such as softening, ion exchange and CO_2 removal [21]. The final RO brine is then sent to evaporation and crystallization units (Figure 1). Mechanical vapor compression (MVC) is commonly used in thermal concentrators and crystallizers [3]. While MVC concentrators and crystallizers are reliable to treat final brines with much higher salinity (>250 g/L) and viscosity, they are energy intensive (up to 60 kWh/m^3 of treated feedwater [22]) and require ongoing efforts to reduce the overall energy consumption in ZLD.

However, in addition to the membrane fouling/scaling concerns, the application of RO in ZLD is also constrained by an upper operating salinity range (70–75 g/L) due to hydraulic pressure limitations. Non hydraulic pressure-driven processes including electrodialysis/electrodialysis reversal (ED/EDR) and forward osmosis (FO) have been introduced to bridge the salinity gap between the RO brine and the feed of crystallizers [3]. To avoid the dilute loss when a low-salinity product water is produced from an electrolyzer, ED/EDR is used as a partial desalination process; that is, ED/EDR concentrates RO brine to a salinity of 100–200 g/L whilst EDR effluent is further desalinated by RO or partially blended with RO permeate to obtain desired product water [23]. Compared to ED/EDR that is driven by an electrical gradient, FO utilizes an osmotic pressure difference to attain water transport across a semipermeable membrane. A full-scale ZLD system installed at Changxing power plant in Zhejiang, China applies FO process (with use of NH_3/CO_2 as the thermolytic draw solute) to concentrate RO brine to >220 g/L [24], which is then fed to a crystallizer for final treatment. More recently, novel RO technologies that can overcome the hydraulic pressure limit of conventional RO are proposed, including osmotically assisted RO and low-salt-rejection RO [25]. These innovative technologies can highly concentrate industrial wastewaters under moderate operating pressures.

4. Challenges, Research Needs and Future Opportunities in ZLD

4.1. Are the Capital Expenditures (CAPEX) too High?

The CAPEX of pretreatment units for ZLD is inexpensive and is similar to that of wastewater treatment. The UF/RO dual-membrane systems can be more expensive as they are operated under conditions to yield higher water recovery. The biggest expense of ZLD is on the thermal treatment section, and it is estimated that the evaporation/crystallization block contributes to 60–70% of the total equipment cost [26]. While the CAPEX varies with the treatment options and fluctuation of the water quality, a ZLD system runs upwards of USD 0.1–1 million at a flow rate of 1 m^3/h (at an influent TDS of ~5 g/L and water recovery of 90–95%) [27,28]. This is considerably cheaper compared to that of the stand-alone thermal processes; however, the affordability of ZLD (i.e., 2–5 times CAPEX of conventional wastewater processes) is still a key concern especially in developing countries. Innovative technologies may improve the treatment efficiency but not necessarily reduce the CAPEX; for example, while use of nanofiltration-type FO in ZLD would enjoy a higher water flux and selective recovery of valuable solutes from waste streams, the water and salt revenues are compromised by the additional CAPEX [28].

4.2. How to Improve the Removal of Organics from the RO Brine?

The RO brine that enters the membrane concentrators and thermal crystallizers should have a chemical oxygen demand (COD) concentration less than 50–100 mg/L [29]. In addition to the technical requirements, the presence of dissolved organic matter (that partially originate from the biological treatment process) would influence the color of the mixed solids. Catalytic ozonation is one of the most widely investigated AOP to address this issue in ZLD, and Fe-based catalysts with abundant surface Lewis acid sits have shown high efficiency in decomposition of aqueous ozone and production of hydroxyl radicals. While there have been recent studies on the oxidative species that likely account for organics degradation in RO brine [20], our knowledge of this process is not complete. For instance, Cl^- of a high concentration influences not only the speciation of reactive oxidants but also the adsorption of organic pollutants onto the surface, which is deemed as a prerequisite for catalytic ozonation. Although the use of catalysts with high adsorption capacities improves the selectivity, this may cause the accumulation of intermediates and finally deposition of carbon on the active sites. More frequent maintenance (e.g., chemical depassivation or replenishment of catalysts) would be required. This important issue has surprisingly received limited attention.

4.3. Salt/Ammonia Recovery: Resource or Waste?

Recovery of salts from RO brine is potentially profitable to reduce the operating cost. Several salts such as NaCl, $CaCl_2$ and $MgCl_2$ (with a commercial sale price ranging from 65–400 USD/ton) can be produced depending on the feed composition [6]. However, the salt products from ZLD that always have a low grade of purity (e.g., 90–95%) are not good candidates for chlor-alkali industrial production; for example, the hardness ions in NaCl need to be removed because an excess of Mg^{2+} causes hydrogen evolution on the electrode, leading to an explosive mixture of H_2 and Cl_2 [30]. While the solid waste (or hypersaline brine) can be refined with the implementation of high-throughput and precise-selective processes (e.g., NF membranes), this inevitably increases the CAPEX and whether the final salt products offer a competitive price is open to discussion. Likewise, ammonia recovery from industrial effluent has attracted increasing interest. Acids are widely applied as the draw solutions to extract ammonia in membrane stripping [31–33]. While the market price of a more popular material, aqueous ammonia solution (USD 300/ton at 25 wt%), is more expensive than $(NH_4)_2SO_4$ (USD 150/ton, purity > 98%), the production of aqueous ammonia solution from industrial effluent is quite challenging [31].

To illustrate the under-representation of these important topics to the state-of-knowledge, we carried out an unofficial Web of Science Core Collection search of the literature on "Zero Liquid Discharge", from 2000 to 2021 (Figure 2). Among the 914 published articles retrieved from the database, a vast majority of research in this area has focused on "Water recovery", "Membranes" and "Energy cost", and there is emerging interest on "Organic removal", "Brine management" and "Salt recovery". As aforementioned, research on these topics has promoted our mechanistic and system-level knowledge to develop smart and efficient ZLD processes. According to the authors, continuous studies would be devoted to improving the energy efficiency of the membrane and thermal systems. For example, novel membrane materials (e.g., omniphobic and Janus membranes [34]) and desalination processes (e.g., solar membrane distillation [35] and capacitive deionization [36]) have been recently suggested as new paradigms for ZLD.

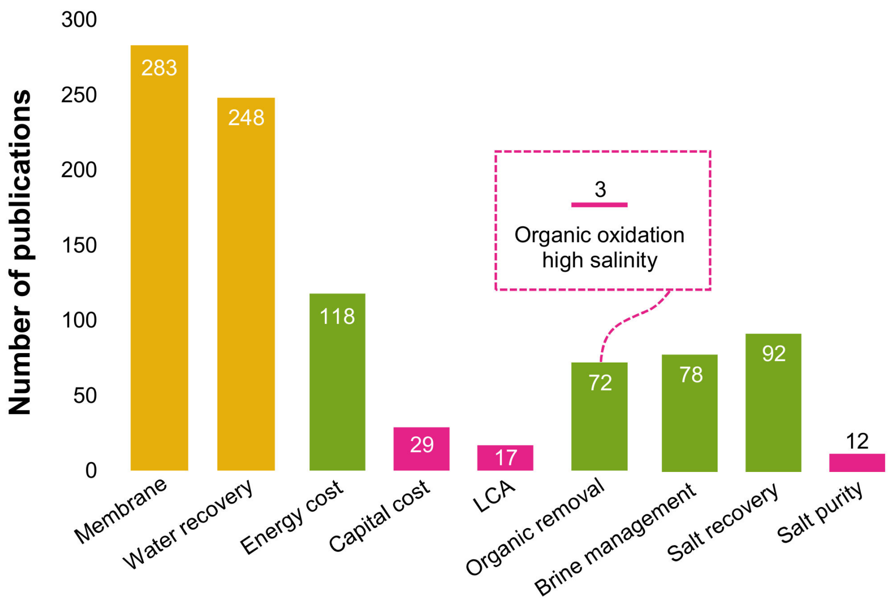

Figure 2. Literature survey of articles (Web of Science Core Collection) in topics related to "Zero liquid discharge" from 2000 to 2021 (search date: 13 September 2021).

Based on the surveyed literature, it is apparent that more work is required to address challenges: (1) development of AOPs for organic oxidation at high salinity, and (2) development of technologies to improve the purity of salts recovered. In this effort, electroactive membrane technology may meet the niche for superior selectivity and precise capture/conversion of target organic pollutants in a complex water matrix (e.g., high salinity) [37]. Our group has developed various electroactive membranes to remove refractory organics and chelated heavy metals [1,38,39]. Recycling brine waste salt for reuse requires specific chelating materials that target specific cations, thereby removing the undesired contaminants while preserving NaCl concentration in the brine. Moreover, only 29 and 17 articles in the search mentioned "Capital cost" and "Life cycle assessment (LCA)" respectively (Figure 2). While such economic and environmental analysis at bench scale may be not rigid [40], it would be a nice start towards practical evaluation of potential technologies in ZLD. In this effort, facility digital twins that mimic hydraulics, controls and water quality offer a flight simulator model to optimize the process design and to compare every life cycle stage under different scenarios.

5. Concluding Remarks

The growing worldwide population is a major driver boosting the industry water consumption in the past decade, and this trend is expected to continue in the following years. The gap between industrial water supply and demand leads to a need for ZLD that brings the idea of internal water recycling to fruition. Most existing ZLD systems reply on biological, membrane and thermal processes for pretreatment, concentration and evaporation and crystallization of industrial effluents, with an average water recovery of 90–95% registered.

A quick evaluation of the ZLD process in this study highlights the importance of development and implementation of organic abatement and desalination & salt recovery technologies to accomplish the goal. Moreover, a reprioritization of research should be considered as there is under-representation of (i) developing novel routines to produce high-quality salt products that are ready for reuse, (ii) oxidizing refractory organic pollutants in RO brine at a high selectivity and efficiency, and (iii) deploying economic and environmental-impact analyses that are essential for full-scale applications. Better understanding of these important issues will facilitate the commercialization of industrial ZLD processes towards a more sustainable future.

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