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Research trends on desalination: zero-liquid discharge of brine (ZLD)

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ABSTRACT

Desalination provides a quality effluent for either urban or industrial use, but it produces waste that has an environmental impact. Zero-liquid discharge (ZLD) of brine aims to increase the efficiency of the desalination process, obtaining a solid that can be reused while minimizing environmental impacts. This article reviews the research on ZLD in order to measure the interest of the scientific community and evaluate the challenges of ZLD processes. A total of 238 publications were analysed. The temporal evolution of the number of published papers shows the growing interest of the scientific community in this field. Results highlight that in the future, ZLD processes need to improve several aspects, such as improving membranes to increase their performance at lower operational costs. In addition, it is necessary to reduce energy consumption and improve the control of scaling and fouling. The incorporation of more environmentally friendly energy sources could also help reduce energy costs of ZLD processes. Moreover, more suitable ways are needed to collect and separate the solids from the obtained concentrate and make better use of them.

Keywords: Bibliometric; Scopus; Brine management; Brine treatment; Brine disposal

1. Introduction

Many of the planet's natural water sources are being affected by human action [1,2]. Moreover, future planning by the water sector must reduce the extraction of natural, freshwater and promote non-conventional practices to achieve a more sustainable water use [3]. One of the proposed solutions to water scarcity is the desalination of seawater and brackish water, since a high proportion of the population lives near the coast [4]. Global desalination production is expected to exceed 200 million m^3/d by 2030 [5].

However, brine production in desalination processes is one of the key factors limiting the growth of desalination. The brine contains a high concentration of dissolved salts (>400.000 mg/L measured as total dissolved solids) with minor contributions of organic matter, metals, nutrients and pathogenic substances [6]. Its discharge has potential risks to the environment such as eutrophication, pH fluctuations and metal accumulation [7]. In order to select the best brine management technology, it is necessary to study several aspects such as the volume rejected, chemical characteristics of the concentrate, operational and capital costs, etc. [6]. Traditionally, options for brine disposal from desalination plants have been deep well injections, surface water discharge, or concentration of brines in evaporation ponds [8]. In addition, in coastal desalination plants, the costs of brine disposal to the sea ranges from 5% to 33% of total desalination costs [8].

The concept of zero liquid discharge of brine arises from the idea of maximizing the production of product water, reducing the liquid discharge to the environment, and obtaining a solid residue that can be used in other processes. The implementation of ZLD systems would make it possible to consider desalination as a technology within

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a circular economy. The ZLD concept can be achieved by various processes based on membrane and thermal technologies. On an industrial scale, only evaporators and crystallizers have been developed, however, they have high capital and operating costs [9]. Therefore, the development of new processes that improve the existing technology and increase the performance of ZLD systems is necessary.

Bibliometric analysis is one of the main tools for understanding and analysing the perspectives and interests of the scientific community on a certain topic [10]. According to Pritchard (1969) [11], it involves applying mathematical and statistical methods to analyse both the scientific literature and the authors who generate this literature. Currently, this definition has been extended to the analysis of countries, related journals, and other important aspects, such as the use of keywords [12]. In recent years, bibliometric studies have experienced exponential growth [13] attributed to the availability and accessibility of specific software and digitized scientific databases. In addition, these tools can handle large volumes of scientific data in a systematic way [14]. Bibliometric analysis is also useful for deciphering and mapping cumulative scientific knowledge and detecting innovation trends. In other words, bibliometric studies can lay a firm foundation for progress in a particular field by providing a comprehensive overview of this field [14].

The aim of this work is to examine the scientific literature on zero-liquid discharge of brine to analyse the research trends in this field. For this, a bibliometric study was carried out, analysing the temporal evolution, the countries and affiliations that have made the most contributions to this scientific field, as well as their recent lines of investigation.

2. Methodology

This analysis was carried out on the results obtained after conducting a search in the Scopus database. Scopus is one of the largest peer-reviewed bibliographic databases belonging to the Elsevier Company and has an active coverage of more than 25,000 titles from more than 5,000 international publishers in the fields of science, technology, medicine, arts, and humanities. It also has one of the oldest digitized records prior to 1970 with a total of 77.8 million contributions [15]. The search was conducted, at the end of March 2022, with a temporal limit from 1980 to 2021. To obtain representative contributions of the field of study, the use of adequate search terms is required. In this work, the search fields used were title, abstract and keywords. The words selected were "Brine" and "Zero Liquid Discharge". Fig. 1 summarizes the search equation used. Microsoft Excel was used to analyse the results and VOSviewer (version 1.6.17) was used to illustrate the co-occurrence networks. The impact index of the publication sources was analysed using the Scimago Journal and Country Rank (SJR) and CiteScore (Scopus), which makes it possible to analyse the impact of the sources and the Scopus citations. All scientific production data collected in the Scopus database with the search terms have been retrieved. Consequently, 242 results were obtained of which four documents were discarded from the bibliometric study due to lack of indexing and keyword information. Thus, 238 results have been analysed in this study.

3. Results and discussion

3.1. Evolution trends

The temporal distribution of scientific production is a key factor in analysing the rate of publications and, therefore, reveals the importance or implementation of zero liquid discharge of brine (ZLD). Fig. 2A shows the evolution for all publications related to zero liquid discharge of brine. Two periods are clearly identified: before and after 2007. Scientific production was below five publications per year before 2007. The second period (from 2007 to 2021) with more than ten publications shows a continuous exponential growth. Scientific contributions related to zero brine discharge published in 2021 reached forty-nine papers (1.4 times more than those published in 2020).

The index of authors-documents per year provides a useful measure of the interest of the scientific community in a specific field. Fig. 2B shows the temporal evolution for authors-documents ratio. The first publications on zero liquid discharge of brine were written by less than three authors. However, over time, the bibliometric analysis shows a linear growth in the number of authors interested in this topic.

To quantify the current interest in a field of study, the percentage of citations during the last five years has been determined [16]. In this case, the citations received from these papers in the last five years have been taken, that

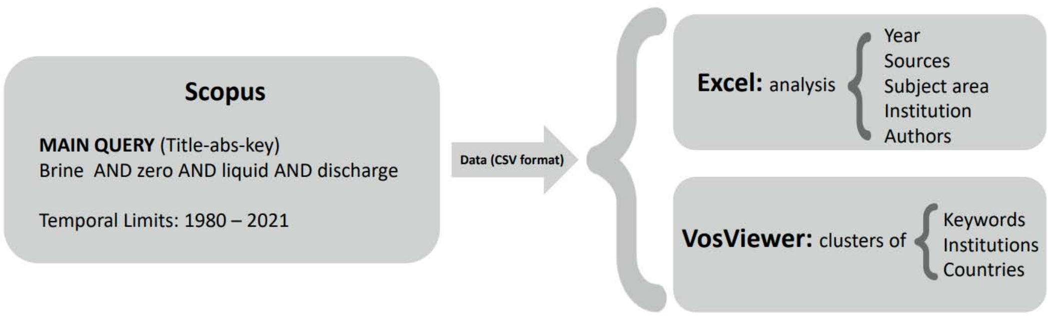

Fig. 1. Scheme of the methodology used for the bibliometric analysis.

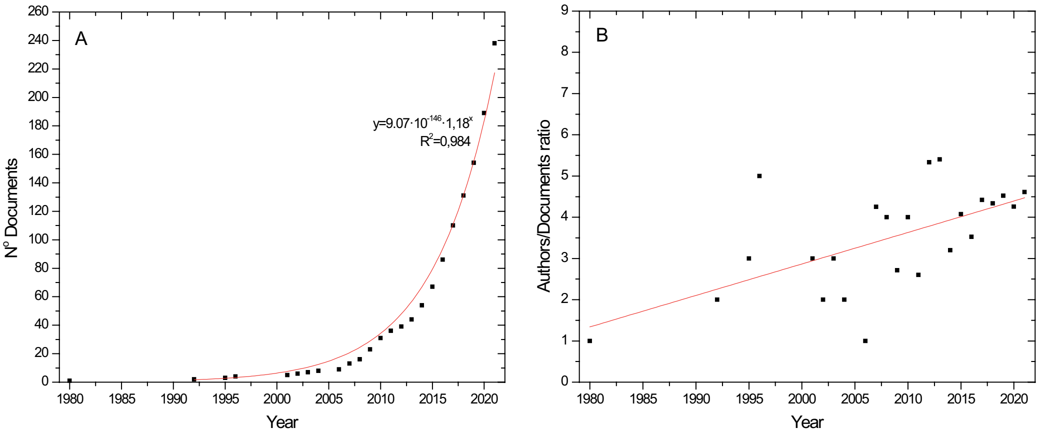

Fig. 2. Evolution of the worldwide scientific production on zero liquid discharge of brine according to (A) published documents and (B) authors and documents ratio.

is, those that have been published since 2017. All the citations received for these papers represent 48% of the total citations of the papers used in the analysis carried out in this work.

3.2. Subjects from worldwide publication

The type of publication is an indication of maturity of the technology studied [4]. A high concentration of conference publications would indicate a very new technology. In this study, most of the publications were articles and reviews in scientific journals, reaching 76% of the total number of publications. Contributions to conferences and book chapters represent a small fraction of the publications registered, 18% and 5%, respectively. Another key factor in the development of any research work is the language of publication since it significantly determines the target audience. English is the priority language for publication in the international scientific community to achieve a wider dissemination of works presented. The search results on zero brine discharge support this since more than 99% of the publications analysed are in English.

Scientific publications are grouped into scientific areas, which allow their specialization and sectorization. The classification of categories has been carried out by means of the classification system in the Scopus database and its analysis is critical to understand the way in which authors approach research in a field of study.

Fig. 3 shows the distribution and temporal evolution of scientific areas. The main areas of publication are Environmental Science (26%) and Engineering (19%). However, zero liquid discharge of brine is not analysed from a single point of view, publications in the areas of Chemical Engineering (15%), Chemistry (13%), Materials Science (10%) and Energy (8%) were found. The temporal evolution of the scientific areas is very similar in all categories. Since 2007, the growth in the number of publications has been exponential in all categories. However, each area has experienced this growth at different times. The areas related to engineering and environmental sciences have experienced continued growth since 2007. By contrast, the materials science category only started to grow in the last five years (Fig. 3).

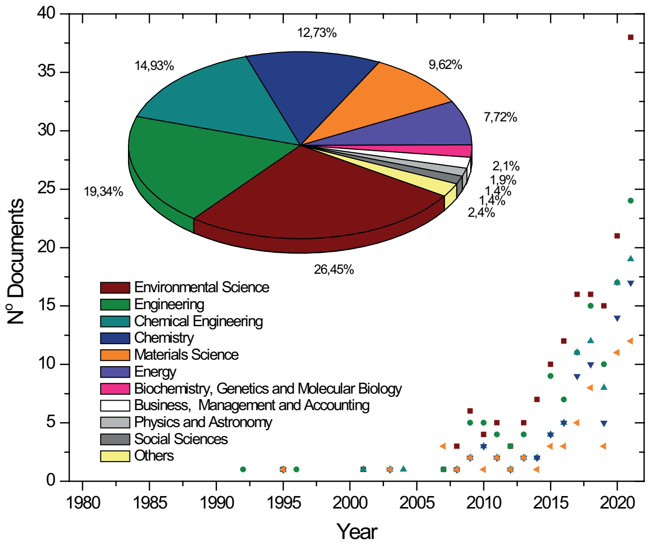

Fig. 3. Distribution and evolution of Scopus categories on zero liquid discharge of brine.

3.3. Journals

An analysis of the sources of publications on the advances carried out on a given topic contributes to identifying those publications which researchers choose for their papers and, at the same time, obtain bibliographic references for their studies. Mainly, the sources have published papers concerning zero liquid discharge of brine reported by Scopus. Specifically, 89% of the journals have only published one article in the field under study. This behaviour can be described by Lotka's Law (Price, 1976). Fig. 4 plots the number of sources vs. the number of publications on the subject under study. Lotka's Law fits the results with a high regression (greater than 0.999). The results show that the number of journals publishing papers related to zero liquid discharge of brine will rise as the number of articles increases.

Table 1 lists the top 20 Journals related to zero liquid discharge of brine. It shows that the number of publications

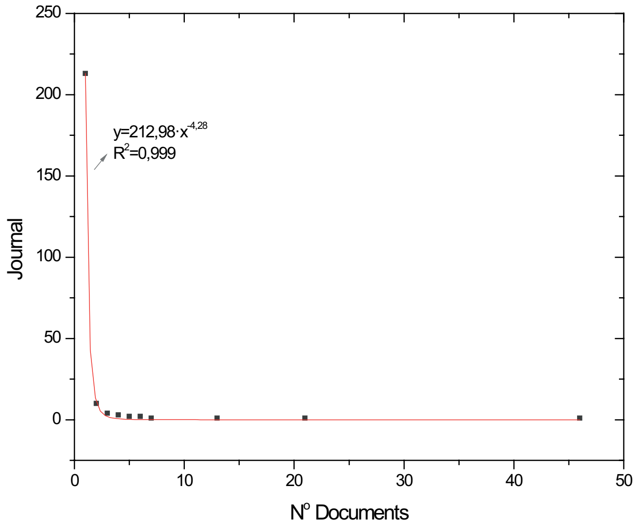

Fig. 4. Number of documents per journal conform to the Lotka's law.

(N); JCR category and its indicator, the CiteScore by Scopus was also added. The top 20 journals listed are indexed in Q1 and Q2 regardless of the area of publication, except for one journal located in the second position, which is indexed in Q4. Desalination, published by Elsevier, is a reference in all topics related to desalination processes, therefore, as expected, it occupies the first position with 46 documents and 1,090 citations. In second place, Desalination and Water Treatment edited by Desalination Publications with 21 papers but with a lower citation rate than other journals included in the ranking. This behaviour can be attributed to the greater disclosure of the journals indexed in the first quartiles. Table 2 summarizes the ten most cited papers related to zero brine discharge, which have been published in journals indexed in the first quartile of their categories.

Reviews are used by authors to reflect the latest advances on a process or technology and therefore tend to be the documents with the highest number of citations. A high number of reviews demonstrates the maturity of the subject under analysis. In the case of zero brine discharge, three of the ten most cited manuscripts found were reviews. The article with the most citations is a work in which different technologies are proposed for the treatment of brine based on its origin, also indicating those valuable compounds that can be obtained from it [17]. The second review in number of citations presents, in extensive detail, the main technologies for osmosis brine volume minimization [18]. In this paper, the treatment technologies were categorized as membrane-based, thermal-based and emerging technologies. Selection of the best treatment technology depends on several factors such as the characteristics of the RO concentrate, the extent of treatment required, whether near-ZLD or ZLD [18]. Membrane-based technologies are less energy intensive compared to thermal based technologies, but an intermediate treatment is required to decrease the concentration and minimize scale forming. By contrast, thermal-based processes have high capital costs and energy consumption. However, the combination of these technologies can achieve complete zero-liquid discharge. Emerging technologies such as forward osmosis or membrane distillation are also being developed but require more evaluation to obtain long term operational success [18].

Panagopoulus et al. (2019) proposed a ZLD system composed of three stages: preconcentration, evaporation and crystallization. In the first stage, based on membrane technologies, the volume of the brine is minimized. This step is a key to reducing the capital and operating costs of the next stages. Subsequently, the evaporation and crystallization stages are aimed at volume reduction and production of solid products.

Processes to minimize brine discharge are discussed in six of the ten most cited publications. Membrane distillation was analysed in several studies with some studies reporting high water recovery (greater than 96%) [19]. These authors observed high recoveries (>98%) when they used forward osmosis with dosages of antiscalants. By optimizing the antiscalant dose, scale control was achieved, improving the performance of both processes, membrane distillation and forward osmosis [19]. In addition, other authors combined reverse osmosis and electrodialysis and managed to achieve high effective recoveries of 97–98% using brackish water as product water [20].

Solar distillation can also be used to achieve ZLD. Shi et al. [21] analysed a 3D cup-shaped solar evaporator that produced solid salts as the only product using solar energy. Finnerty et al. [22] also reported the characterization and evaporation performance of synthetic leaf made graphene oxide, which achieved a light to vapor energy conversion of 78%.

Although ZLD results in the minimization of waste, its implementation can lead to unintentional environmental impacts. The solid products recovered are mixtures, which cannot be reused, however, it is possible to produce multiple high-purity solid salts instead of a compact mixed solid [23] for example, a production of NaCl, $CaCO_3$, Na_2SO_4 and $CaCl_2$. Jeppesen et al. [24], analysed the recovery of sodium chloride from reverse osmosis concentrate. Multi-stage flash distillation processes are the best systems for cost optimization in the extraction of sodium chloride [24]. Additionally, rubidium or phosphorus mining can represent a cost-effective option that has substantial environmental benefits, however, these applications need further investigation [24]. Ji et al. [25] evaluated water recovery and NaCl crystallization using a membrane distillation-crystallization bench-scale plant. The process achieved a water recovery greater than 90%. However, the presence of organic matter introduces negative effects in the performance process (reduction of salt crystallized and decrease of flux) in comparison with artificial brines, thus pre-treatment is necessary to remove the organic matter [25].

The possibility to recover the energy content of discharge concentrate could be implemented to reduce the overall energy consumption of reverse osmosis. The combination of direct contact membrane distillation and reverse electrodialysis was tested for both water and energy production from reverse osmosis brine thus implementing the concept of low energy and near-zero liquid discharge of brine in seawater desalination [26]. The implementation of this system requires a significant effort to optimize the net energy produced and improve the stack design [26].

Table 1
Top 20 Journals and metrics (year 2020)

Journal	N	SJR Category, Rank SJR	SJR indicator	Cite Score Scopus	Citation count
Desalination	46	Chemical Engineering 9/143 Q1 Water Resources 3/98 Q1	1.794	14.3	1090
Desalination and Water Treatment	21	Chemical Engineering 110/143 Q4 Water Resources 83/98 Q4	0.251	1.6	335
Journal of Cleaner Production	13	Environmental Engineering 6/54 Q1 Environmental Science 18/274 Q1	1.937	13.1	667
Water Research	7	Environmental Engineering 3/54 Q1 Environmental Science 6/274 Q1 Water Resources 2/98 Q1	3.099	15.6	602
Environmental Science and Technology	6	Environmental Science 20/274 Q1	2.851	13.8	308
Journal of Membrane Science	6	Chemical Engineering 11/143 Q1 Polymer Science 4/90 Q1	1.929	13.5	8
Journal of Environmental Management	5	Environmental Science 34/274 Q1	1.441	9.8	81
Separation and Purification Technology	5	Chemical Engineering 16/143 Q1	1.279	9.9	508
Energy Conversion and Management	4	Energy & Fuels 10/114 Q1 Mechanics 2/153 Q1 Thermodynamics 2/60 Q1	2.743	15.9	81
Environmental Science: water Research & Technology	4	Environmental Engineering 24/54 Q2 Environmental Science 89/274 Q2 Water Resources 20/98	1.08	5.5	145
Applied Energy	3	Energy & Fuels 9/114 Q1 Chemical engineering 6/143 Q1	3.035	17.6	115
Membranes	3	Chemistry Physical 69/162 Q2 Chemical Engineering 45/143 Q2 Material Science 125/334 Q3 Polymer Science 21/90 Q1	0.609	3.7	81
Process safety and Environmental Protection	3	Chemical Engineering 22/143 Q1 Environmental Engineering 13/54 Q1	1.173	8.9	46
Chemical Engineering and Processing Process Intensification	2	Energy & Fuels 51/114 Q2 Chemical engineering 45/143 Q2	0.828	5.9	52
Environmental Science and Pollution Research	2	Environmental Science 91/274 Q2	0.845	5.5	128
Environmental Science and Technology Letters	2	Engineering Environmental 9/54 Q1 Environmental Science 26/274 Q1	2.497	12.4	20
Journal of environmental Chemical Engineering	2	Chemical Engineering 24/143 Q1 Environmental Engineering 14/54 Q2	0.965	7.5	69
Journal of Hazardous Material	2	Engineering Environmental 4/54 Q1 Environmental Science 10/274 Q1	2.034	13.4	24
Journal of Thermal Analysis and calorimetry	2	Analytical Chemistry 18/87 Q1 Physical Chemistry 62/162 Q2 Thermodynamics 8/60 Q1	0.521	5.1	32
Journal of Water Process Engineering	2	Chemical Engineering 27/143 Q1 Environmental Engineering 16/54 Q2 Water Resources 9/98 Q1	0.901	5.2	12

The future of ZLD processes need to improve several aspects such as the development or modification of new membranes to achieve better performances at lower prices. In addition, it is necessary to decrease the energy consumption and improve control of scaling and fouling [23].

3.4. Countries and their main topics

Scientific production by country provides an analysis of the places that have made the greatest effort in the field under study. Fig. 5 shows the geographical distribution of scientific production on zero liquid discharge of brine. The

Table 2
Top 10 most cited manuscripts

Authors	Title	Year	Source	No cites
A. Pérez-González, A.M. Urtiaga, R. Ibáñez, I. Ortiz	State of the art and review on the treatment technologies of water reverse osmosis concentrates	2012	Water Research 46(2), pp. 267–283	528
C.R. Martinetti, A.E. Childress, T.Y. Cath	High recovery of concentrated RO brines using forward osmosis and membrane distillation	2009	Journal of Membrane Science 331(1–2), pp. 31–39	429
X. Ji, E. Curcio, S. Al Obaidani, G. Di Profio, E. Fontananova, E. Drioli	Membrane distillation-crystallization of seawater reverse osmosis brines	2010	Separation and Purification Technology 71(1), pp. 76–82	242
A. Subramani, J.G. Jacangelo	Treatment technologies for reverse osmosis concentrate volume minimization: a review	2014	Separation and Purification Technology 122, pp. 472–489	189
A. Panagopoulos, K.-J. Haralambous, M. Loizidou	Desalination brine disposal methods and treatment technologies – a review	2019	Science of the Total Environment 693,133545	176
C. Finnerty, L. Zhang, D.L. Sedlak, K.L. Nelson, B. Mi	Synthetic graphene oxide leaf for solar desalination with zero liquid discharge	2017	Environmental Science and Technology 51(20), pp. 11701–11709	162
Y. Oren, E. Korngold, N. Daltrophe, R. Messalem, Y. Volkman, L. Aronov, M. Weismann, N. Bouriakov, P. Glueckstern, J. Gilron	Pilot studies on high recovery BWRO-EDR for near zero liquid discharge approach	2010	Desalination 261(3), pp. 321–330	148
R.A. Tufa, E. Curcio, E. Curcio, E. Brauns, W. van Baak, E. Fontananova, G. Di Profio	Membrane distillation and reverse electrodialysis for near-zero liquid discharge and low energy seawater desalination	2015	Journal of Membrane Science 496, pp. 325–333	128
Y. Shi, C. Zhang, R. Li, S. Zhuo, Y. Jin, L. Shi, S. Hong, J. Chang, C. Ong, P. Wang	Solar evaporator with controlled salt precipitation for zero liquid discharge desalination	2018	Environmental Science and Technology 2018, 52, 20, 11822–11830	109
T. Jeppesen, L. Shu, G. Keir, V. Jegatheesan	Metal recovery from reverse osmosis concentrate	2009	Journal of Cleaner Production 17(7), pp. 703–707	95

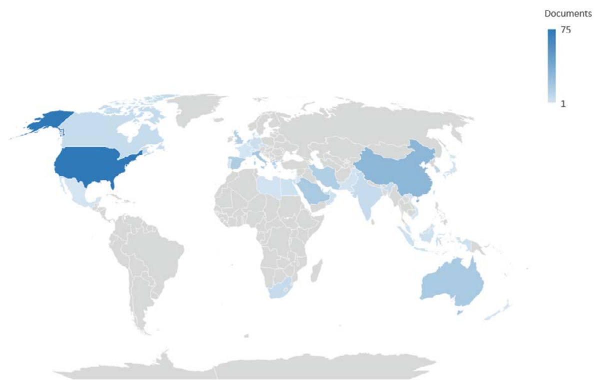

Fig. 5. Geographic distribution of the scientific production on zero liquid discharge of brine.

countries that have contributed most to the development of ZLD processes are the United States and China, with 75 and 29 publications, respectively, collected in Scopus. The results also show how countries with scarce water resources and, therefore, with high implantation of desalination processes have contributed to the development of brine minimization. Examples of these are Mediterranean and Middle Eastern countries. The analysis of international collaborations between authors also highlight the geographic interrelationships in this subject. Fig. 6 shows the clusters of geographic distribution in which the size of the coloured cloud indicates the degree of interrelationships between countries and the clouds distance shows the relationships established between each country. The United States has had the greatest number of international interrelationships. However, China, despite occupying the second position in number of publications, have not collaborated with authors from other countries. This is not the case for Spain, Iran or Italy, which although they do not occupy very high positions in the ranking of publications, have established more relationships with other countries than China. Table 3 summarizes the international collaborations of the top five countries. These data were deduced from the co-authorship of the articles. In this table, the collaborations between countries are shown, ordering them on a scale of 1 to 5. The value of 1 represents the country with which the most collaborations have been carried out and 5 the country with the least.

3.5. Affiliations and their main topics

Fig. 7 shows the top 20 institutions with scientific production in ZLD. The concentration of publications in a given centre reveals the existence of a highly focused research group in the field. For example, the institution that has contributed the most to the development of the ZLD is the National Technical University of Athens with 12 publications, and it is the only institution in this country that has done research on the topic. By contrast, there are many institutions in the USA that investigate the subject of ZLD, specifically 7 of the 20 most productive are in this country. Despite being the country that has generated the most publications, its production is highly segregated, and its institutions do not exceed five papers each, according to the terms used in the search. The second and third places in the ranking of principal institutions are shared by European research centres in Italy and Spain. The specialization of European authors in zero liquid brine discharge is reflected in the affiliation of the most productive authors. Table 4 shows the top six authors with the highest number of publications related to liquid zero brine discharge, and the overall performance of these authors is reflected by the H-index and the i-10 index. The H-index is an author-level metric that measures productivity and citation impact of publications [27,28]. The i-index represents the number of publications that

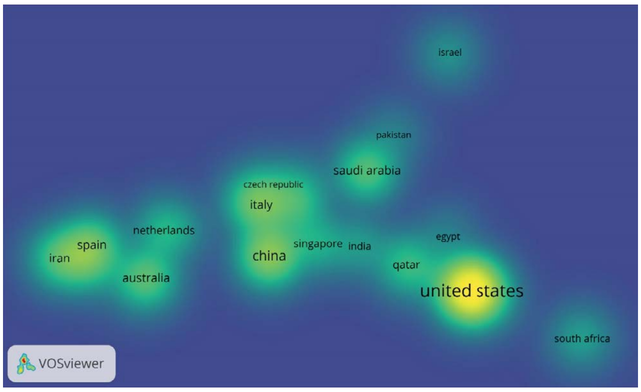

Fig. 6. Clusters of geographic distribution of the scientific production on zero liquid discharge of brine obtained using VOS viewer software.

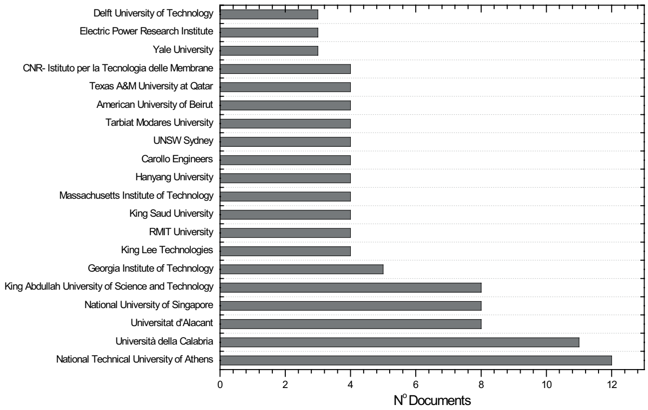

Fig. 7. Top 20 institutions with higher scientific production on the zero liquid discharge of brine topic.

Table 3
Top 5 countries and their international collaboration

Country	N	Collaboration*				
		1	2	3	4	5
United States	75	China	Saudi Arabia	Qatar	Singapore	South Africa
China	29	United States	Saudi Arabia	Australia	Singapore	Arabs Emirates
Australia	17	China	United Kingdom	South Korea	Singapore	France
Italy	17	Spain	Saudi Arabia	South Korea	Netherlands	India
Saudi Arabia	17	United States	China	Italy	Singapore	Qatar

*The value of 1 represents the country with which the most collaborations have been carried out and 5 the country with the least.

Table 4
Top 6 most productive authors based on the number of published documents

Author	Affiliation	N	H-index	i-10 Index
J.A. Caballero	Universitat d'Alacant, Alicante, Spain	8	41	96
V.C. Onishi	Edinburgh Napier University, Edinburgh, United Kingdom	8	13	16
A. Panagopoulos	National Technical University of Athens, Athens, Greece	8	12	12
E. Drioli	CNR- Istituto per la Tecnologia delle Membrane, Rende, Italy	7	93	625
E. Curcio	Università della Calabria, Rende, Italy	7	44	91
J.A. Reyes-Labarta	Universitat d'Alacant, Alicante, Spain	7	20	41

have at least ten citations [29]. Both indices are considered the best known to evaluate scientific production and performance [30].

The analysis of the priority research lines of institutions has been carried out using the top five keywords (Table 5). The National Technical University of Athens' publications have used general keywords that cover a complete study of ZLD processes. In the last two years, this institution has had a high scientific production related to this topic, with four publications per year. During 2021, Panagopoulos A. published several works that give a technical-economic vision of processes to achieve ZLD. The results showed that the process that produces a pure solid from ZLD has a higher specific energy consumption than processes where a mixed solid salt is obtained. However, if solid salt revenues were also considered, the system to obtain a pure salt would be an economically feasible option [31]. The influence of the kind of brine was also analysed techno-economically by the same author in two scenarios; brackish water and seawater systems [32] and it was found that ZLD processes for seawater had higher energy consumption. Overall, the results showed that the proposed ZLD systems can be effective in the treatment and valorisation of brine desalination and could have significant implications for industries that generate brine (e.g., textile, food and leather industries) [32]. The techno-economic analysis for different methods to minimize the volumes of brine discharge suggests that they could all be sustainable and cost-effective [33]. The author suggests that future research should focus on the improvement of membrane-based technologies and the incorporation of more environmentally-friendly energy sources to enhance efficiency and increase ZLD processes [31–33].

The second and fourth most productive affiliations are the University of Calabria and the National University of Singapore, which, like the National Technology University of Athens, use keywords that generally describe ZLD processes (Table 5). In the last five years, the research group of the University of Calabria has published three articles and a book chapter related to the topic. In 2017, they published an interesting article in collaboration with other institutions, which provided a review of membrane processes that can be used in ZLD approaches and described their principal problems that restrict development and upscaling [34]. In this study, the authors proposed additional investigation into process integration to shrink equipment size, boost plant efficiency, reduce capital costs, minimize environmental impact, as well as reduce carbon dioxide footprint and improve system robustness. All of these changes would contribute to the feasibility of the membrane ZLD process, since currently, conventional thermal processes are more cost-effective than membrane processes [34]. They evaluated an alternative system for efficient seawater desalination [35]. The innovative system combines membrane distillation and reverse electrodialysis for the simultaneous production of water and energy with a drastic reduction in the amount of brine generated [35]. Recently, the selective precipitation of calcium was evaluated for brine from a reverse osmosis process using different precipitation agents under a wide range of operating conditions [36]. These results are of interest to help our understanding of precipitation processes that allow pure salt to be obtained. Therefore, they are crucial for the design of processes that achieve zero liquid discharge of brine [36].

The National University of Singapore has used keywords that generally describe ZLD processes, but in recent years they have focused more on membrane-based processes (Table 5). In these processes, brine pre-treatment is necessary to control fouling and scaling of the membrane. The most common pre-treatment is chemical precipitation to remove hardness and control calcium precipitation. However, this process does not control the formation of silica precipitates. In addition, the presence of antiscalants from previous desalination processes limits the performance of chemical precipitation [37]. The brine silica content can be removed by electrocoagulation and organic matter can be removed by coagulation, electrocoagulation, nanofiltration or adsorption [37]. In recent years, biological processes, such as microbial fuel cell or microalgae, have experimented a growing interest in the scientific community [38]. However, these technologies have little potential as pre-treatments due to the narrow scope of treatment, low performance, and the toxicity of brine for microorganisms [37]. Nanofiltration would be a

Table 5
Top 5 affiliations and their main keywords

Affiliation	Country	N	Keywords (exclude zero-liquid discharge)				
			1	2	3	4	5
National Tech. University of Athens	Greece	12	Desalination brine	Brine treatment	Desalination	Solar energy	Brine
Università della Calabria	Italy	11	Nanofiltration	Membrane fouling	Brine concentration	Brine management	Energy efficiency
Universitat d'Alacant	Spain	8	Shale gas wastewater	Mechanical vapor recompressión	Multiple effect evaporation	Optimization	Single effect evaporation
National University of Singapore	Singapore	8	Nanofiltration	Membrane fouling	Brine concentration	Energy efficiency	Brine
King Abdullah University of Sci. and Tech.	Saudi Arabia	8	Brine reclamation	Forward osmosis	Hybrid desalination	Membrane distillation	Scaling inhibition

cost-efficient pre-treatment for ZLD processes, however, nanofiltration operations can produce severe and complex membrane fouling [39]. Organic fouling dominates the lead stage, though it can be completely removed through base cleaning. However, when recovery is increased by treatment steps, fouling by calcium carbonate precipitates increases and requires more intense cleaning. Furthermore, increased pre-treatment recovery by nanofiltration can lead to unrecoverable fouling due to the formation of silica and iron precipitates [39]. Capacitive Deionization (CDI) is an emerging desalination technique that is recognized as "electrosorption". In CDI processes, the ions are immobilized by electrical double layers [40]. The National University of Singapore evaluated the treatment of industrial brine by this technology to achieve zero-liquid discharge. There are two main challenges for the use of this technology for this type of brine: the dissolved solids content and high fouling potential [40]. Thus, additional research is required to improve cell architecture, operational strategies and fouling control [40]. The maximum hydraulic pressure of osmosis membranes limits the efficiency of ZLD processes. Researchers at the National University of Singapore evaluated two new reverse osmosis processes: osmotically assisted and low salt rejection, to overcome such limitations [41]. Low-salt-rejection reverse osmosis could be more feasible than other processes due to membrane module availability and lower capital cost [41].

The fifth most productive affiliations are from the King Abdullah University of Science and Technology that have evaluated different alternatives to achieve zero brine discharge by treating real brine from seawater desalination processes. The first treatment attempts to achieve a circular system to increase the performance of reverse osmosis by membrane distillation, dilution of the concentrate by direct osmosis and increase the performance of the crystallization process [42]. Brine from the reverse osmosis unit is heated before being fed into the distillation unit. The concentrate from this unit is then fed into a direct osmosis process, which can upgrade the effluent from industrial wastewater before entering the crystallization unit. This alternative process for a sustainable operation, although the presence of chemicals in the brine water from reverse osmosis inhibits the crystallization of salts [42]. Another alternative process developed by researchers of this institution is a hybrid system that integrates multi-effect distillation and evaporative crystallization. Chen et al. [43] present a detailed thermodynamic and economic analysis in a study where a heat transfer area of 110–340 m^2 is proposed for the treatment of a brine from a seawater desalination plant. The economic study highlights the high cost of the thermal process and shows that the presence of residual heat can reduce operating costs drastically [43]. Chen et al. [44] have also published an interesting paper on a new humidification-dehumidification process evaluated in a preliminary way which uses low energy consumption, low operation cost and reduce the potential of scaling and fouling. The process crystallization system consists of two direct-spray humidifiers, a dehumidifier/condenser, an air heater and a salt separator. The brine is mixed with the process recirculation stream and fed into a condensing unit, where preheating takes place. The preheated brine is sprayed into the first humidifier for its evaporation. The water with low salt content is used in a previous condensing unit where a distillate is obtained. The concentrated water from the first humidifier is sprayed into a second humidifier where a salt sludge is obtained and separated from the liquid stream in a salt separator. This liquid stream is used as a recirculation stream. The preliminary study demonstrates the operability of the unit to obtain salt crystals and also, the energy consumption is 56% lower than a conventional crystallizer [44]. As has been mentioned, solar crystallization is a low-cost ZLD process alternative. Researchers at King Abdullah University of Science and Technology have also evaluated an advanced solar crystallizer coupled to a salt crystallization inhibitor to eliminate highly concentrated water brine [45]. A new 3D solar crystallizer operates as a dead-end type solar driven water removal mode in which water evaporation surface and light absorption surfaces are physically separated by an aluminium sheet with high thermal conductivity [45].

The Universitat d'Alacant is third in the ranking of the most productive affiliations in the field. In this case, the research group has focused on a very specific aspect of the concept of zero brine discharge, as can be seen in the keywords used (Table 5). Shale gas extraction generates a large amount of high-salinity wastewater [46] and to lessen environmental damage, shale gas flowback water should be reclaimed to be recycled or reused in a desalination process (reverse osmosis, multistage membrane or multistage flash distillation) [47]. The research group of the Universitat d'Alacant specialises in the analysis of zero liquid brine discharge processes for the desalination of wastewater and its reuse in the production of shale gas. A mathematic model, based on multistage superstructure, has been developed [48,49]. The model can analyse the ZLD target in a solar energy process coupled to a multi-effect evaporation with mechanical vapour precompression [48]. In addition, this model together with a Monte Carlo risk analysist supports the selection of more robust ZLD desalination systems applied to shale gas flowback water [49]. In recent years, the model has been improved to reduce the uncertainty of the results [47,50,51].

3.6. Keywords from worldwide publications

Table 6 shows the twenty most used keywords. The top ten keywords related to zero liquid discharge of brine are, globally, as follows: zero liquid discharge, desalination; membrane distillation; reverse osmosis; brine; brine management; brine treatment; nanofiltration; resource recovery and brine concentration. Note that some keywords such as ZLD have been grouped with others that have the same meaning acting as synonyms. The top twenty keywords can be classified into four clusters as they are highly interrelated. The four clusters are: general description, membrane processes, thermal processes and precipitation. In the first cluster, the keywords included are zero-liquid discharge; desalination; brine; brine management; brine treatment; resource recovery; brine concentration; brine disposal; techno-economic analysis and wastewater. In the second cluster, the keywords membrane distillation, reverse osmosis, nanofiltration, forward osmosis, membrane fouling are grouped together. The keywords evaporation, renewable energy, and solar desalination make up the third cluster, and finally the fourth cluster consists of crystallization and salt recovery. Keyword analysis identifies the main challenges that must be faced for the implementation of ZLD systems to be widespread and large-scale [6,8,52].

Table 6
Top 20 keywords related to zero liquid discharge of brine

Rank	Keyword	N
1	Zero liquid discharge	79
2	Desalination	37
3	Membrane distillation	24
4	Reverse osmosis	18
5	Brine	18
6	Brine management	10
7	Brine treatment	11
8	Nanofiltration	12
9	Resource recovery	8
10	Brine concentration	7
11	Brine disposal	9
12	Forward osmosis	7
13	Crystallization	7
14	Membrane fouling	5
15	Evaporation	7
16	Renewable energy	5
17	Salt recovery	6
18	Techno-economic analysis	5
19	Wastewater	6
20	Solar desalination	6

The temporal evolution of the keywords was evaluated by means of co-occurrence analysis using VOS viewer software. Fig. 8 shows the temporal evolution of concurrences between keywords over the last five years. The analysis of keywords identifies how the interest of the scientific community has changed in recent years. Five years ago, there was a high level of interest in processes that achieve zero brine discharge, but in the last few years this interest has waned. Instead, the analysis of the process as a whole, taking into account techno-economic aspects, has been the focus of research in this field in the last two years (Fig. 8). Approximately 15% of the publications in the last two years of the time range used in our search have focused on reducing the amounts of brine discharge and achieving optimal energy consumption [53–56]. Researchers have also focused on the recovery of resources from brines to minimize discharge, and, in the case of industrial effluents, the recovery of valuable components has become more important [57–59].

4. Conclusions

A bibliometric study was conducted to analyse the status and future trends of zero liquid discharge of brines. Since 2007, scientific production has experienced exponential growth, with a Price index of 48%. The results reflect the growing interest in processes to achieve ZLD. The percentage of articles indicates the maturity of the topic analysed and the number of publications per source conforms to Lotka's Law. The countries that have contributed most to the development of ZLD processes are the United States and

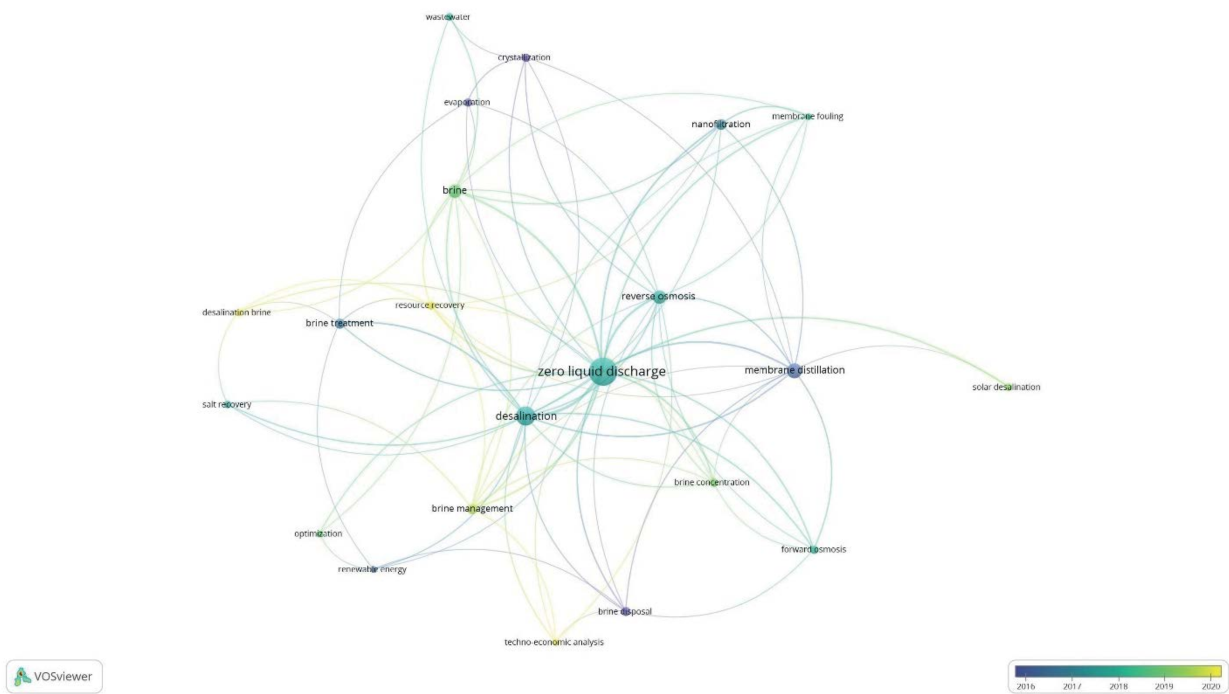

Fig. 8. Temporal evolution of keywords occurrence obtained using VOSviewer software.

China, with 75 and 29 publications, respectively. However, the main institution that has contributed to the development of the ZLD is the National Technical University of Athens with 12 articles.

The process to reach the zero liquid discharge of brine is categorized as membrane-based, thermal-based and emerging technologies. Membrane-based technologies are less energy intensive when compared to thermal based technologies, but to decrease the concentration and minimize scale forming, pre-treatment is required. Thermal-based processes have high capital costs and energy consumption. However, the combination of these technologies with membrane-based processes can achieve a complete zero-liquid discharge. Emerging technologies such as forward osmosis, membrane distillation, solar distillation or new processes are being developed but require more evaluation to obtain long-term operational confidence.

The future of ZLD processes require improvements of several aspects such as the fabrication or modification of new membranes to increase the performance with lower costs. In addition, it is necessary to reduce the energy consumption and improve the control of scaling and fouling. The incorporation of more renewable energy sources could be an alternative to help improve energy costs of ZLD processes.

In conclusion, ZLD systems can be effective in the treatment and valorisation of desalination brine. Furthermore, they can have significant implications for industries that generate brine and need to recover valuable components from it.

Author contributions

O. Díaz: Conceptualization, Methodology, Investigation, Data curation, Writing – original draft. E. Segredo-Morales: Writing – Review & Editing. Figueira, A: Investigation & Data curation. E. González: Visualization, Writing- Review & Editing.

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