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Sustainable saline wastewater treatment using eutectic freeze crystallization: Recent advances, challenges and future prospects

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ABSTRACT

The increase in global population, industrialization and urbanization has resulted in a corresponding increase in saline wastewater, causing escalating environmental and health concerns. The increasing volume of saline wastewater generated by various industries poses significant disposal challenges, and regulatory pressures and public awareness are driving the need for innovative and environmentally friendly treatment techniques. In response to these challenges, eutectic freeze crystallization (EFC) has emerged as a promising innovative solution, owing to its energy-efficient nature and ability to recover salts. This comprehensive review provided insights into the bibliometric analysis of EFC technology, offering an overview of the current state-of-the-art research in the EFC process for saline wastewater treatment. In comparison between EFC and other existing saline wastewater treatment processes, EFC holds remarkable advantages in terms of liquid waste minimization, valuable resource recovery and without salinity restriction. Besides, the advancement and applications of EFC in the treatment of various type of saline wastewater were also discussed, indicating a growing interest among researchers due to its high versatility. While successful applications of EFC have been highlighted, the challenges such as ice scaling issue and complexity of multi-component wastewater treatment using EFC still persist. These challenges bring directions for future prospects in EFC research, underscoring the need for more research and development efforts to enhance the effectiveness and economic benefits of EFC technology for its widespread application in treating industrial saline wastewater.

1. Introduction

Water is a natural resource abundant on Earth, with 75% of the planet's surface covered by water. However, 97.5% of surface water is saline, with only 0.3% of fresh water is available for direct human use [1]. Human population growth, industrialization, urbanization and economic development have led to a substantial increase in water demand, resulting in a global water crisis. As the demand for water continues to rise and the availability of freshwater remains limited, our planet is headed towards a state of greater water scarcity. By 2025, the human population is expected to reach 8.2 billion and is projected to approach 10 billion by 2050, with two-thirds of these individuals likely to experience water stress and the remainder facing complete water scarcity [2]. Undeniably, huge amount of industrial wastewater generation, which is the main source of water pollution, has intensified the global water crisis. In order to ensure a sustainable source of clean water, stringent laws and regulations are enforced by authorities on industries to ensure proper wastewater treatment is employed prior to the disposal into waterbodies.

Saline wastewater refers to the wastewater with high dissolved salt content, typically exceeding the levels that are considered safe for discharge into the environment or for use in various industrial or agricultural processes. This type of wastewater can be generated from a variety of sources, including oil and gas production [3,4], mining operations [5], desalination processes [6,7], seafood processing [8], dairy effluent [9], textile [10], pharmaceutical [11] and agricultural industries [12]. Improper treatment and disposal of saline wastewater result in severe water pollution by disrupting the salinity balance, pH and temperature of the ecosystem [13,14]. In addition to the adverse impact on aquatic life, high salt content in wastewater also leads to

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eutrophication, harms the surrounding soil and vegetation, and contaminates groundwater [15]. Thus, it is essential for industries to employ effective treatment processes to mitigate the environmental and health risks associated with saline wastewater.

Evaporative crystallization (EC) process is conventionally used in treating saline wastewater, however it has significant drawbacks. This method uses heat to vaporize the water content of the wastewater, leaving highly concentrated brine as the product which requires further separation or appropriate disposal [16]. The common brine disposal methods include solar evaporation ponds, surface water discharge and deep well injection [17–19]. Additionally, the large amount of wastewater involved in the evaporation process makes it an expensive and environmentally unfavourable approach due to its high energy consumption [9,20]. Eutectic freeze crystallization (EFC) is a promising technology for overcoming the drawbacks encountered by EC process. This process operated under eutectic condition allows for simultaneous ice and salt separation from saline wastewater without additional chemicals. The separated product of ice and salt crystal can be easily attained by gravity due to their significant difference in density. EFC is highly energy-efficient due to the low heat of fusion of ice (6.01 kJ/mol) which results in an estimated energy saving of up to 70% as compared to conventional EC process [21]. According to Randall et al. [16], EFC has the potential to achieve cost savings of over 80% in comparison to EC process. This is attributed to the significantly lower latent heat of fusion of ice, which is approximately 13% of the latent heat of evaporation of water. Additionally, the life cycle assessments have indicated that EFC process is capable of considerably decreasing the energy usage and carbon footprint as compared to EC process [22]. Besides, EFC process also exhibits minimal corrosion rate and fouling issues [23,24]. EFC offers a promising solution to resource recovery with the development of zero liquid discharge technologies, making it a sustainable technology in the long term. The recovered salts from the EFC process can be utilized as fertilizers, providing market value that helps to offset the cost of crystallization and eliminates the disposal expenses [5,25]. EFC also offers the advantage of recovering both pure salts and water, allowing for their reuse in the industrial process, which not only helps in resource conservation but also leads to a reduction in the overall cost of raw materials [26,27].

From the literature, most of the previous studies reported on the application of EFC in treating various types of wastewaters have been conducted at the laboratory scale. The implementation of EFC in real industrial applications has been limited, and the technology is still considered emerging and immature. However, given its high potential and the numerous benefits it offers as a sustainable approach for saline wastewater treatment, there is a need for a comprehensive review to assess the current research progress in the field. This review presented the bibliometric analysis to demonstrate the current state-of-the-art research on EFC technology for sustainable treatment of saline wastewater. Apart from that, the comparison between EFC and other available physicochemical and biological treatment methods was also discussed to highlight the potential of EFC as a sustainable and effective approach for treating saline wastewater. This review also explored the diverse applications of EFC in different types of wastewater treatment, as well as addressing the challenges and future prospects of EFC technology. Lastly, the potential research directions for further advancement of EFC technology to facilitate its widespread implementation in industrial practice were also discussed.

2. Bibliometric analysis of EFC research

A bibliometric analysis was performed using VOSviewer software version 1.6.19, incorporating 63 articles extracted from the Scopus database. These articles covered the period from 1998 to December 2023 and were retrieved based on the initial keyword search of "eutectic freeze crystallization". The article selection process involved a manual review of abstracts to ensure that only relevant articles were included in order to maintain representativeness within the field of EFC. This careful screening process helped to ensure that the chosen articles were aligned with the research focus and objectives of the bibliometric analysis, thereby enhancing the reliability and relevance of the analysis. By employing co-occurrence analysis of the keywords extracted from these articles as depicted in Fig. 1, the analysis aimed to uncover the primary research themes, concepts and emerging areas within the field of EFC technology. This analysis not only contributed to a comprehensive understanding of the current state of knowledge but also aided in identifying the research gaps and exploring the potential directions for future investigations. The results of the co-occurrence analysis indicated that EFC has remained an emerging technology in the realm of saline wastewater treatment. The research focus primarily revolved around fundamental aspects of EFC, including "crystallization kinetics", “growth rate” and "nucleation rate." This highlighted the ongoing efforts to comprehend the underlying fundamental mechanisms dictating crystal formation and growth during EFC, demonstrating a commitment to advancing researchers’ understanding of the process. Furthermore, the keywords associated with "phase diagram", "eutectic temperature" and "eutectic points" signified a keen interest in investigating the phase behaviour of systems involving EFC. Such knowledge is pivotal for optimizing the process efficiency and control. Additionally, the presence of keywords such as “purification”, “separation”, "desalination", and "wastewater treatment" emphasized the exploration of EFC as a potential method for facilitating the removal of contaminants from various type of wastewater. Furthermore, the inclusion of the keyword "recovery", “salts”, “sodium sulfate” and “sodium chloride” underscores the goal of resource recovery in EFC research. This suggested a growing focus on achieving sustainable saline wastewater treatment processes, where EFC plays a significant role by enabling efficient resource recovery. Moreover, the emergence of keywords "heat transfer" and "ice scaling" pointed to researchers actively investigating the thermal aspects of EFC and addressing the challenges related to heat transfer and scaling phenomena. These aspects are crucial for optimizing process design and operation, ensuring efficient heat exchange and mitigating scaling issues that may hinder the process performance. Overall, the co-occurrence analysis of keywords has provided insights into the current research landscape of EFC in saline wastewater treatment. The emphasis on the fundamental aspects, feasibility studies and addressing specific challenges demonstrated that EFC has remained an evolving technology with substantial potential.

3. Operation principles of EFC

The simplified flow diagram of EFC process is illustrated in Fig. 2. EFC process begins with the feed brine solution being fed into a pre-cooler to lower the temperature of the brine solution in preparation for the subsequent crystallization process. The pre-cooled brine solution is then directed into the crystallizer, where further cooling beneath eutectic temperature leads to the formation of ice and salt crystals. From the top of the crystallizer, the ice product is directed to a wash column where it undergoes washing with cold water [28,29]. This washing process effectively purifies the ice, yielding pure ice as the final product. Meanwhile, the filtrate outlet from the wash column, which consists of water and any dissolved impurities removed during the washing process, is being recycled back to the crystallizer. At the bottom of the crystallizer, the output salt slurry is directed to a belt filter for further separation of the salt crystals from the mother liquor, producing salt cake as the output [28,29]. The filtrate from the belt filter is also recycled back to the crystallizer for EFC process.

The operation principles of EFC could be explained using a phase diagram. A phase diagram provides information about the presence of different phases in a system at varying temperatures and compositions. Analyzing the solution concentration and temperature using a phase diagram is essential in EFC technology as it helps to understand the solubility and predict the composition of the crystalline product [6].