Chemosphere 173 (2017) 474–484

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Review

# Crystallization techniques in wastewater treatment: An overview of applications

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## HIGHLIGHTS

- Crystallization techniques utilized in wastewater resources recovery and treatment are summarized.
- They are commonly used for the reclamation of valuable salts and water.
- Their mechanisms, advantages and challenges have been discussed in detail.

## ARTICLE INFO

*Article history:*
Received 22 November 2016
Received in revised form
10 January 2017
Accepted 12 January 2017
Available online 18 January 2017

Handling Editor: Shane Snyder

*Keywords:*
Crystallization
Wastewater treatment
Desalination
Water recovery
Salt recovery

## ABSTRACT

As a by-product of industrial or domestic activities, wastewater of different compositions has caused serious environmental problems all over the world. Facing the challenge of wastewater treatment, researchers have begun to make use of crystallization techniques in wastewater treatment. Crystallization techniques have many advantages, such as high efficiency, energy saving, low costs, less space occupation and so on. In recent decades, crystallization is considered as one of promising techniques for wastewater treatment, especially for desalination, water and salt recovery. It has been widely used in engineering applications all over the world. In this paper, various crystallization techniques in wastewater treatment are summarized, mainly including evaporation crystallization, cooling crystallization, reaction crystallization, drowning-out crystallization and membrane distillation crystallization. Overall, they are mainly used for desalination, water and salt recovery. Their applications, advantages and disadvantages were compared and discussed in detail.

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http://dx.doi.org/10.1016/j.chemosphere.2017.01.070
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## 1. Introduction

Nowadays, a huge amount of wastewater is being produced everyday as a by-product of industrial or domestic activities. Wastewater from different sources, such as petroleum industry, coal industry, printing and dyeing industry, tannery, electroplating and so on, has caused serious treatment issues all around the world (Chen, 2004; Fang et al., 2017). Generally, wastewater contains complicated organic and inorganic compounds, such as high concentrations of phenolic compounds, polycyclic aromatic hydrocarbons, salts, heavy metals, ammonia, cyanide and thiocyanate (Wang et al., 2011; Lu et al., 2016). The specific compositions of wastewater are highly complex and vary greatly depending on the sources. These features make it difficult to manage and treat. Serious issues have been resulted from wastewater, including soil and water pollution, water resources shortage, wasted resource and so on. That is also the reason why wastewater treatment has been a common concerns of both governments and researchers all over the world in the past decades. The treatment of wastewater is a universal challenge due to its operation complexity and cost issues. New treatment techniques for recovering water and valuable compounds from waste water at low costs are urgently needed. In recent years, great efforts have been made to develop new techniques and significant progress have been achieved. Many new techniques, such as membrane technology, ion exchange technology, nanotechnology, have been successfully used in wastewater treatment. The performance and effectiveness of these techniques have been compared (Liang and Han, 2011; Alasfour and Abdulrahim 2011; Randall and Nathoo, 2015).

As a conventional unit operation, crystallization has been extensively studied and has found broad application in various industries all over the world. Crystallization is a solid-liquid separation technique, in which the solute crystallizes from the liquid solution and turns into a pure solid crystalline phase. This makes crystallization techniques effective for the recovery of a variety of useful and valuable substances (along with the recovery of water) from wastewater, such as recovering NaCl (Nathoo et al., 2009), $Na_2SO_4$ (Xu et al., 2010; Jiao and Jiao, 2015), $Na_2CO_3$ (Himawan et al., 2006; Spronsen et al., 2010), ammonium (Zhang et al., 2013; Huang et al., 2014), phosphates (Hutnik et al., 2013; Rahman et al., 2014), the reclamation of pure water (Williams et al., 2015), the removal of heavy metals ($Pb^{2+}$, $Mn^{2+}$, $Ni^{2+}$,$Cu^{2+}$, $Ag^{+}$, etc.) (Rubio and Tessele, 1997; Al-Tarazi, 2004) and water softening by removing $Ca^{2+}$, $Mg^{2+}$ (Suzuki et al., 2002; Rahman et al., 2011).

Comparing with other techniques, crystallization technology has many advantages, such as high recovery rate, capability of recovering both high quality water and valuable salts at the same time, no consumption of other supplementary materials (membrane, catalyst, adsorbent, ion exchange resin, oxidant or reducing agent, etc). For instance, for membrane process, which is widely used to minimize the amount of wastewater and recover pure water, large amount of membrane need to be used and need to be replaced periodically due to the blockage and pollution of the membrane. Also, in membrane process, extensive pretreatment which will consume other chemical materials area are necessary (Mickley et al., 1993). After membrane process, although the volume of wastewater is reduced, except for pure water, other valuable products, such as salts, cannot be directly recovered from the highly concentrated wastewater. Other techniques are needed to recover the salts for the wastewater. Furthermore, some disposal problems will appear because many regulations are based on concentrations rather than volume (Ahmed et al., 2000). In comparison, crystallization does not need any other supplementary materials and can recover the salts along with the recovery of pure water since crystallization is able to produce high purity products from impure solutions. Moreover, besides to good operability and stability, with a wide range of schemes to achieve energy saving and ensure high recycling efficiency, it is also widely acknowledged that crystallization could be energy saving, highly efficient and easy to scale up if appropriate design is done. (Lefebvre and Moletta 2006; Liang and Han, 2011; Liang et al., 2013; Al-Mutaz and Wazeer 2014). With so many advantages, crystallization is considered as a promising technique for wastewater treatment, especially for recovery of valuable resource. As a result, in recent years, crystallization techniques have been extensively utilized in the practical applications of wastewater treatment all over the world (Spronsen et al., 2010; Suzuki et al., 2002; Bian et al., 2011; Zhao et al., 2011; Liang and Han, 2011; Williams et al., 2013).

Many kinds of crystallization techniques have been investigated and utilized in the field of wastewater treatment. In this paper, various crystallization techniques, mainly including evaporation crystallization, cooling crystallization, reaction crystallization, drowning-out crystallization and membrane distillation crystallization, are summarized and compared. Their applications, advantages and disadvantages were discussed in detail.

## 2. Evaporation crystallization

In evaporation crystallization (EC), the solvent is detached with the solute under heat treatment and the solute crystallizes out from the solution in the form of solid state crystals. For an unsaturated solution at a given temperature, the solvent can be evaporated by supplying an external heat source. With the gradual removal of solvent, the solution finally reaches saturation. Further evaporation of solvent will lead to the crystallization of the solute from the saturated solution.

The application of EC can date back to 5000 years ago, when it was used for the production of table salt. As the most widely used crystallization method in practical production process, EC has been widely used in many fields, such as food, medicine, and so on. What is more, EC is a conventional technology in the treatment of wastewater, especially high salinity wastewater, which contains many kinds of ions such as $Cl^{-}$, $SO_4^{2-}$, $Na^{+}$, $Ca^{2+}$. At present, multi-effect evaporation crystallization is the most often used EC techniques due to its maturity and high efficiency. Meanwhile, mechanical vapor recompression crystallization (MVRC) has emerged as an alternative technique and progressed rapidly.

### 2.1. Multi-effect evaporation crystallization (MEEC)

Multi-effect evaporation (MEE) mainly consists of several single

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