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## Dynamic modelling and simulation of eutectic freeze crystallization process for recovery of ammonium sulphate from aqueous solutions

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## ABSTRACT

The advancement of industrial sectors generates large amount of wastewater containing ammonium sulphate (AS) ions, yet the current AS recovery technologies are energy-extensive and produce excessive sludge. In this study, a dynamic model for a batch crystallizer was developed to investigate the eutectic freeze crystallization process of AS in aqueous solutions using the method of moment (MoM). The variables affecting the crystal size distribution of AS and ice were described in terms of zeroth moment ( $\mu_0$ ), first moment ( $\mu_1$ ), second moment ( $\mu_2$ ) and third moment ( $\mu_3$ ). The  $\mu_0$  of ice crystals increased significantly until 2 s and slowed down thereafter while  $\mu_0$  of AS crystal increased steadily and reached a constant value of  $1/m^3$ s after 1 s. The  $\mu_1$ ,  $\mu_2$ , and  $\mu_3$  of the AS and ice crystals depicted a similar increasing trend. Through process optimization using a 3-D plot on the initial concentration of the system and initial temperature of cooling fluid which had been identified as the significant parameters via iterative plot using MATLAB software, the AS and ice reached maximum crystallization at 255 K which was close to the eutectic point of AS-water system at 254 K. The dynamic model which comprised of population, mass and energy balance equations established in this study demonstrated effectiveness in the prediction of AS in aqueous solutions.

## 1. Introduction

The increasing human activities and development such as industrial, agricultural and municipal works are the main contributors to water pollution. It has been estimated that the wastewater produced daily as a by-product of industrial processes made up to 48 % of the total global wastewater discharged to the environment without any treatment [1]. The presence of ammonium sulphate (AS) in wastewater leads to eutrophication and poses negative impacts on water quality and human health [2-4]. Wastewater containing AS or its ions form is conventionally treated using biological methods [5]. In most cases, the removal of ammonium and sulphate ions in wastewater is focused on separately. The recovery of sulphate ions is commonly performed by reducing them to sulphide using sulphate-reducing bacteria (SRB) under anaerobic conditions [6]. However, biological treatment exhibits a challenge which inhibits the microorganism activities along with the treatment process. Sustainable and biodegradable ammonium removal processes via biological treatments have also been successfully implemented in recent years, nevertheless, these types of methods consume high energy in which the aeration of the nitrification process alone makes up half of the total energy consumption and 60 % of the operation cost comes from the wastewater treatment [7,8]. Thus, there is a trigger in research about the recovery of AS in the context of wastewater treatment using a better separation technique which has a lower energy consumption and at the same time able to produce high-purity AS.

Recent literatures have reported the application of eutectic freeze crystallization (EFC) in salt recovery from various type of wastewater, such as produced water [9], mining wastewater [10], pharmaceutical wastewater [11], dairy effluent [12], seawater brine [13], spent alkaline pulping liquor [14], brine streams [15] and petrochemical brine [16], however, limited study has focused on the removal of AS using EFC. In contrast to the conventional biological methods, EFC is an innovative technique that can separate inorganic salts from their respective liquid state by solidifying both the salt and ice concurrently. This can be achieved by reducing the temperature of the solution below the eutectic temperature without the need for any additional chemicals [17]. Unlike

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