



Review

A comprehensive review of phosphorus recovery from wastewater by crystallization processes



Lihong Peng^{a, b, 1}, Hongliang Dai^{a, b, c, 1}, Yifeng Wu^a, Yonghong Peng^{a, b}, Xiwu Lu^{a, b, *}

^a School of Energy and Environment, Southeast University, No. 2 Sipailou Road, Nanjing 210096, China

^b ERC Taihu Lake Water Environment (Wuxi), No. 99 Linghu Road, Wuxi 214135, China

^c School of Environmental and Chemical Engineering, Jiangsu University of Science and Technology, Zhenjiang 212018, China

H I G H L I G H T S

- Crystallization processes for P recovery from wastewater are reviewed.
- Natural and synthetic materials are available for P recovery from wastewater.
- The application and economic feasibility of P recovery products are assessed.
- Prospects and challenges of crystallization processes for P recovery are presented.

A R T I C L E I N F O

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The presence of phosphorus (P) in discharged wastewater can lead to water pollution events and eutrophication. Given the increasing consumption of phosphate (PO_4^{3-}) rocks, wastewater containing large quantities of P is deemed as a potential source of P recovery. Crystallization of P is an ideal way to recover P because of its simple design, ease of operation, high efficiency, and limited environmental impact. This paper provides a comprehensive review of P recovery by crystallization processes with respect to the mechanisms involved, operational parameters that influence the quality of the crystal, and available seed materials for inducing crystallization. Various operational parameters including pH, molar ratio of participating ions, mixing intensity, reactor type, and seeding conditions, were detailedly investigated. Different kinds of seeds were reviewed critically with regard to their principal properties, application, and long-term prospects. Crystallized products with a high P content can be used directly as slow-release fertilizers for agricultural production, and some test methods have been developed to determine their efficiency as a fertilizer and to evaluate their availability for plants. Further, the feasibility of P recovery by crystallization was evaluated in terms of economic benefits and environmental sustainability. This work serves as a basis for future research of P recovery by crystallization processes and responses to the increasingly stringent problems of eutrophication and the growing depletion of P resources.

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* Corresponding author. School of Energy and Environment, Southeast University, No. 2 Sipailou, Nanjing 210096, China.

E-mail addresses: penglihong1023@163.com (L. Peng), daihongliang103@163.com (H. Dai), 101011433@seu.edu.cn (Y. Wu), pengyonghong809@126.com (Y. Peng), xiwulu@seu.edu.cn, xiwuluseu1@163.com (X. Lu).

¹ These authors contributed equally to this work and should be considered co-first authors.

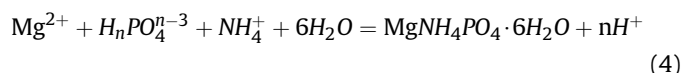
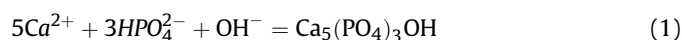
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1. Introduction

The discharge of phosphorus (P) into the aquatic environment via wastewater can lead to water pollution events because P can accelerate eutrophication, with detrimental consequences for aquatic life as well as the domestic and industrial water supply (Hanhoun et al., 2011). Phosphorus is also a key ingredient in fertilizer in the current food production and consumption system. The main source of P is phosphate (PO_4^{3-}) rock, a nonrenewable resource rapidly being depleted as a result of the increasing exploitation of PO_4^{3-} and sedimentation through the natural P cycle (Fig. 1) (Cordell et al., 2011). The global population increase has resulted in a growing demand for food production and consequently an increasing need for P to support agricultural activity (Heckenmüller et al., 2014). Because the most accessible and high-quality PO_4^{3-} rocks are being depleted, low-grade minerals with high levels of impurities, low P content, and poor accessibility in terms of cost–benefit ratio are increasingly being exploited (Desmidt et al., 2015). The result is higher mining and production costs compared to the use of high-quality minerals. There is an urgent need for alternative and renewable sources of P. Wastewater containing large quantity of P is one such potential source.

Many physical, biological, and chemical approaches to recovering P from wastewater have been developed, including chemical precipitation, crystallization, adsorption and ion exchange processes, membrane processes, electrochemical processes, and biological processes (Hutnik et al., 2013a; Loganathan et al., 2014; Luo

et al., 2015; Tarayre et al., 2016; Ichihashi and Hirooka, 2012). Most of these approaches are problematic because they produce low-purity products, have high costs, or are operationally complex. However, crystallization is an extensively researched approach with a relatively high recovery rate and considerable economic efficiency; it produces valuable products with few environmental risks (Tarayre et al., 2016; Dai et al., 2016, 2017). These features indicate the potential for P recovery by crystallization. Two major crystallization processes, the calcium phosphate (Ca-P) and struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$ -MAP) processes (as presented in Eqs. (1)–(4)), have been developed to recover P from wastewater. Ca-P or Struvite formation can be described by the following chemical reaction.



where $n = 0, 1, 2$, etc., and corresponds to the solution pH.

In recent decades, many studies have been conducted to recover P from various wastewaters, such as fertilizer industry wastewater (Hutnik et al., 2013a; Yu et al., 2013), municipal landfill leachate

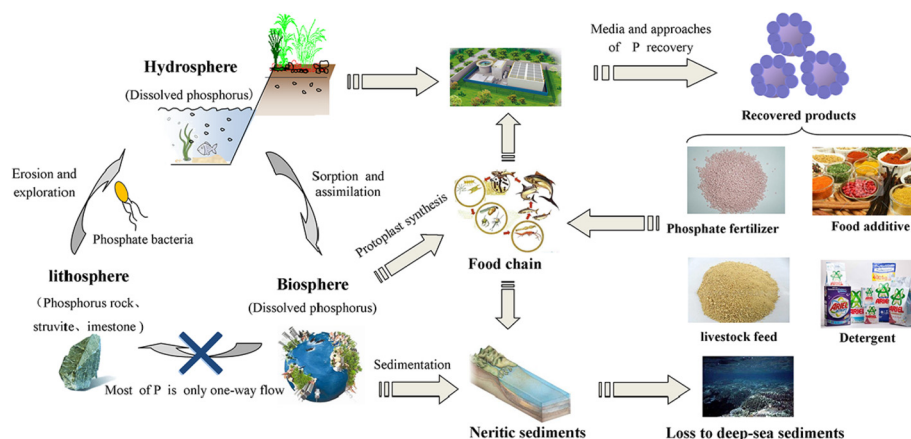


Fig. 1. The schematic diagram of phosphorus cycle in nature.

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