

Study on uranium peroxide precipitation in a continuous fluidized-bed reactor with mechanical stirring



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HIGHLIGHTS

- Precipitation in the fluidized bed leads to a high-quality of solid particles.
- Uranium peroxide precipitation shows a strong influence of the stirring rate.
- We report the technical feasibility to increase the uranium peroxide production.
- All information provided by this study will be used for the process modeling.

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ABSTRACT

This study provides the results obtained from the experiments carried out in a specially adapted continuous fluidized-bed reactor equipped with a multi-impeller stirring device. The influence of operating parameters on yellow cake precipitation as uranium peroxide was studied. The effects of stirring speed and feed flow rate were investigated. An increased stirring speed decreased the agglomerate mean size, mainly due to the strong negative influence of the shear rate on the agglomeration kinetics. In addition, the nominal capacity of the fluidized-bed reactor was doubled, while the uranium concentration level was strictly respected at the outlet. The experimental results obtained in this study will be used for process modeling in a further study.

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1. Introduction

Crystallization processes are widely used in different fields: pharmaceuticals, microelectronics, the environment, water treatment, the food industry, and especially in chemistry. However, the crystals have to meet certain product characteristic requirements such as morphology, particle size distribution, density and humidity. Hence, the main problem often encountered in these processes lies in matching the reactor process conditions to the desired product quality. Here, the focus is on the control and optimization of a precipitation process in the nuclear fuel industry.

The uranium production methodology consists in treating uranium ore with a solution of sulfuric acid in order to obtain a raw diluted uranyl sulfate solution. This solution is then purified and concentrated by liquid–liquid extraction or an ion exchange resin process, from which a pure uranyl sulfate solution containing around 50 g/L of uranium is obtained. Uranium can be precipitated from the pure uranyl sulfate solution by using many chemical reagents in different types of precipitation reactor. The suspension filtration gives yellow cake, which is packaged and transported for further nuclear-related processing.

The fluidized-bed reactor (FBR) is widely used in industrial precipitation processes. A fluidized-bed precipitator with mechanical stirring was developed in the Reaction and Process Engineering Laboratory, Nancy, France (Sellami et al., 2005), and has been employed to many industrial precipitation processes in which it is valued due to its process robustness and good product quality. For example, excellent results were obtained in the extremely high purification of concentrated sodium chloride solutions before

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Notation

Roman symbols

d	particle size (μm)
D	reactor diameter (cm)
H	reactor height (cm)
N	stirring speed (min^{-1})
R	recirculation ratio (–)
S	supersaturation ratio (–)
t_0	initial time (h)
t_f	final time (h)

Greek symbols

β	agglomeration kernel ($\text{m}^3 \text{s}^{-1}$)
$\dot{\gamma}$	shear rate (s^{-1})

Abbreviations

FBR	fluidized-bed reactor
PSD	particle size distribution
SP	sampling point

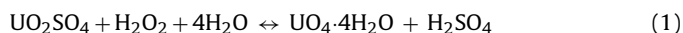
electrolysis (Dandeu, 2006), and in the precipitation of struvite from urban wastes patented by Naskeo Environment (Goy and Frederic, 2010).

In the nuclear field, a continuous uranium precipitation process using hydrogen peroxide in this type of FBR was patented by AREVA (Courtaud et al., 2010). The advantage of this process is the high quality of the solid uranium peroxide particles produced, in terms of granulometry, morphology, purity and density. This improves filtering operations, as well as downstream powder handling processes. In this work, the objective is to study the influence, at pilot scale, of the main operating parameters on product quality and

productivity in order to adapt the FBR with mechanical stirring to industrial processes under different operating conditions, which in turn depend on a wide diversity of ores. In addition, the experimental data will be further used for validation of process modeling of uranium peroxide precipitation in this type of fluidized-bed plants.

2. Experimental procedure

The uranium peroxide precipitation was performed at room temperature by mixing a uranyl sulfate solution with hydrogen peroxide according to the precipitation reaction equation below (Eq. (1)). The hydrogen peroxide (35%) was added to reach a hydrogen peroxide-to-uranium ratio of 1.5:1.



The reaction is performed in acid media, at a pH of around 3. According to Eq. (1), the acidity increases during the precipitation reaction as sulfuric acid is formed, with a corresponding decrease in the quantity of the precipitated uranium peroxide (the reaction reaches the equilibrium). Sodium uranates are formed over pH higher than 5. Monitoring and control of the pH is of capital importance due to its high influence on the precipitation kinetics (Planteur et al., 2013; Gupta et al., 2004). The pH was adjusted to remain around 3 by adding a sodium hydroxide solution with a concentration of 200 g/L. pH was continuously monitored with a pH probe (set up at the top of the settler), to ensure the pH remains within the range of 2.8–3.5.

2.1. Reactor design

The experimental set up is shown in Fig. 1. The fluidized-bed reactor was divided into two zones. The fluidization zone (Zone 1) consists of a column with an H/D ratio = 60 (column height to diameter). A transparent PVC column was used in order to see the mixing

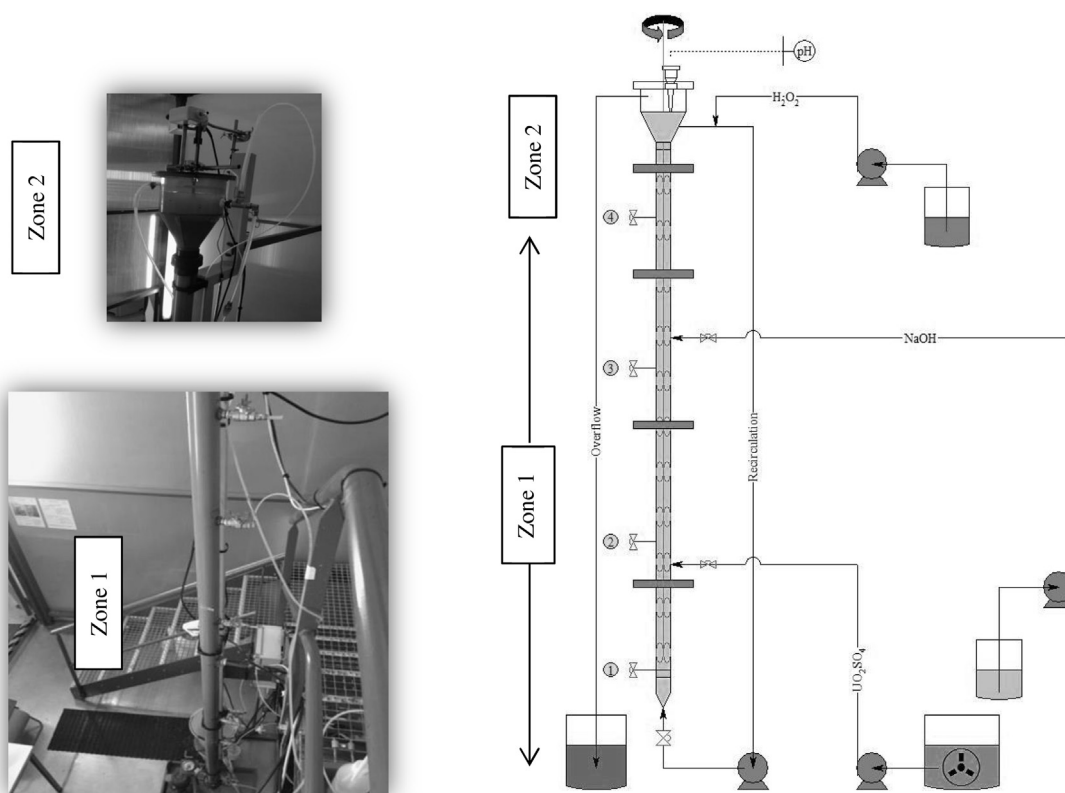


Fig. 1. Experimental set up.

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