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Study on the agglomeration kinetics of uranium peroxide

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Abstract

Considering the previous study dealing with thermodynamic and kinetic phenomena (nucleation and crystal growth) during the uranium peroxide precipitation, this work focuses on the agglomeration mechanism. It provides the results obtained from the experiments carried out in a MSMPR reactor operating at steady state. The influence of the operating parameters on the uranium peroxide agglomerates was studied in order to identify the agglomeration kernel. The method is based on the resolution of the population balance equation using the method of moments and the experimental particle size distributions. The results lead to a size-independent kernel directly proportional to the crystal growth rate. Under the stirring conditions studied, the agglomeration appears to be significantly reduced by mixing which results in a kernel inversely proportional to the average shear rate. The agglomeration kinetic law obtained in this study will be used for the process modelling in a further study.

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1. Introduction

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

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extraction or 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 purified uranyl sulfate solution by using several chemical reagents. The suspension filtration gives yellow cake, which is packaged and transported for further nuclear-related processing. The production of yellow-cake for the manufacture of nuclear fuel is a major step in the nuclear fuel cycle. This is the last step operating near the mining area. Among the reagents for the precipitation of yellow-cake, hydrogen peroxide presents the best decontamination factor towards certain impurities contained in uranium ores[1]. The reaction is performed by mixing a uranyl sulfate solution with hydrogen peroxide at room temperature according to the following equilibrium:

$$UO_2SO_4 + H_2O_2 \overset{H_2O}{\longleftrightarrow} UO_2(O_2)\cdot 4H_2O + H_2SO_4 \quad (1)$$

Planteur et *al*.[2] published both thermodynamic and kinetic study describing the crystal growth rate as well as the model for calculating supersaturation ratio. The objective of this work is to continue the phenomenological study of the precipitation of uranium peroxide by focusing on agglomeration phenomena in a wide range of operating conditions.

Nomenclature

b	baffle width m
B(L)	birth function in terms of size $m^{-4}.s^{-1}$
D	stirrer diameter m
D(L)	death function in terms of size $m^{-4}.s^{-1}$
G	crystal growth rate $m.s^{-1}$
H	reactor height m
L	particle size m
L_C	nuclei critical size m
$d_{4,3}$	mean particle size m
N	stirring speed rpm
n(L)	agglomerate number density m^{-4}
R_N	nucleation rate $m^3.s^{-1}$
T	reactor diameter m
Y	height of the mixer relative to the bottom of the reactor m
β	agglomeration kernel $m^3.s^{-1}$
γ	shear rate s^{-1}
δ	Dirac delta function m^{-1}
λ	particle size m
τ	mean residence time s

2. Theory

2.1. Agglomeration process

Agglomeration is the mechanism by which solid particles collide and adhere to each other by forming physical bonds called crystalline bridges. The agglomerate so formed behaves as a single particle. This mechanism, highly complex, can be described as the result of two steps, which depend simultaneously of many physico-chemical and hydrodynamic parameters:

- The first step consists in a binary collision: during contacting, the short-range electrostatic forces are involved. These are the van der Waals attraction forces and repulsive forces coming from the existence of an electrical