Multi-compartmental simulation of actinides and lanthanides oxalic precipitation in a vortex reactor in the nuclear energy context

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Highlights

- The vortex hydrodynamics is described by a multi-compartmental model
- The steady state population balance equation is solved by a discretization method
- Nucleation, size-independent growth and size-dependent agglomeration are included
- The methodology is validated from neodymium and uranium precipitation experimental values

1. Introduction

Oxalic precipitation is a crucial step on the actinide recycling process since it defines the morphology and the physical characteristics of the raw materials to the MOX fuel fabrication process. One of the narrowest constraints concerns the size of the particles produced since it determines the proper operation of the entire solid treatment chain. Continuous precipitation of actinides is conducted in a vortex reactor. The internal flow pattern produces two macro mixing zones and the concentration of the reagents strongly depends on the spatial position in the vessel.

The population balance equation (PBE) is a well-known statement used to predict crystal size distribution in precipitation processes. Nowadays many PBE numerical solution strategies are reported in the literature due to the lack of a general methodology adapted to any context.

This work focuses on the solution of the population balance equation at steady state in presence of nucleation, crystal growth and loose agglomeration, by combining hydrodynamics and crystallization principles in order to predict the characteristics of both solid and liquid phases leaving the vortex reactor.

2. Methods

The first step consisted in studying independently (i) mixing and (ii) precipitation. CFD simulations were performed at the macro scales to study the local mixing conditions and to reproduce the flow behavior. Due to a strongly anisotropic turbulence originating from the dominance of the tangential velocity, a Large Eddy Simulation (LES) approach was adopted [1]. As result, the flow is schematically represented as a combination of several interconnected well-mixed zones. Crystallization mechanisms were studied separately in order to determine supersaturation expressions and nucleation, growth and agglomeration kinetics.

In a first time a tool to model the behavior of one compartment is developed: the nucleation, growth and agglomeration kinetics are included in the same model to verify their coherence. The model developed allows algorithm solution, critical variables and convergence criteria to be identified. Following this approach, the vortex reactor modelling is decomposed in three steps: (i) The simulation of a Mixed Suspension Mixed Product Removal (MSMPR) reactor, (ii) the study of the interaction between two elementary blocks (MSMPR) and (iii) the implementation and solution of the multi-compartment model.

3. Results and discussion

A numerical methodology to solve the steady state population balance including nucleation, size-independent growth and loose agglomeration is developed. It is based on a discretization method and

the fixed-point algorithm coupled with convergence acceleration methods [2]. In a first time, the oxalic precipitation of neodymium and uranium in a well-mixed reactor is simulated [3]. The results demonstrated to fit the experimental measurements in a wide range of operational conditions. Also, the crossed secant acceleration method is performant to solve the agglomeration population balance equation whether the agglomeration kernel is size dependent or not.

The same methodology is suitable to the simulation of the multi-compartmental model. Again, an accelerated fixed-point algorithm is implemented in order to solve the recycling streams. The population balance equation algorithm is integrated in the 5 compartments model describing the vortex reactor. As result, local properties of the liquid and solid phase are predicted. As an example, the local crystal size distribution in depicted in Figure 1.

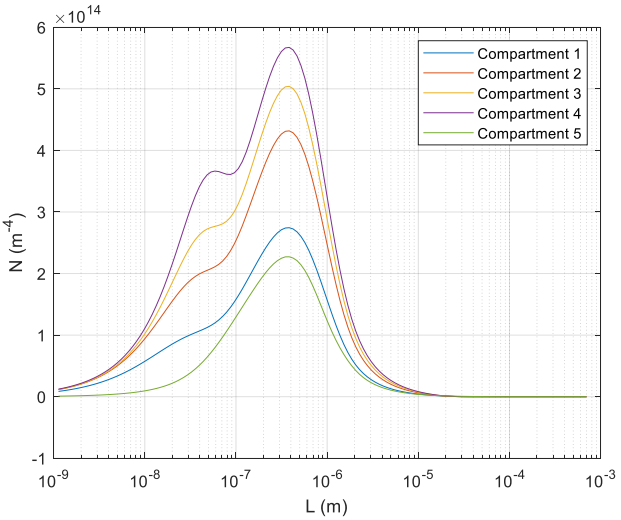

Figure 1. Crystal size distribution evolution in the vortex reactor.

4. Conclusions

The simulation of the vortex reactor is performed by the implementation of looped accelerated fixed-point algorithms. Detailed information of both, liquid and solid phase can be approached: supersaturation, concentration, crystal and monoparticles size distribution are determined in every compartment describing the flow pattern in the vortex reactor. Robustness and accuracy are ensured by modifying the operational conditions and the species to be precipitated.

References

[1] E Saikali, MG Rodio, G Bois, U Bieder, N Leterrier, M Bertrand and Y Dolias. Validation of the hydrodynamics in a turbulent un-baed stirred tank: A necessity for vortex-reactor precipitation studies. Chemical Engineering Science, vol 214, pp 115426, 2020.

[2] I. Ramière and T. Helfer, “Iterative residual-based vector methods to accelerate fixed point iterations,” Computers & Mathematics with Applications, Nov. 2015, doi: 10.1016/j.camwa.2015.08.025.

[3] Ruiz Vasquez, Cristian Camilo, Noureddine Lebaz, Isabelle Ramière, Sophie Lalleman, et Murielle Bertrand. 2023. « Steady State Population Balance Modelling of Precipitation Processes: Nucleation, Growth and Size-Dependent Agglomeration ».. https://doi.org/10.1016/j.jcrysgro.2023.127258.

Keywords
Vortex reactor, Population balance, Oxalic precipitation, Actinides.