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Hybrid LES–multizonal modelling of the uranium oxalate precipitation

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ABSTRACT

Oxalic precipitation is usually applied to process radioactive wastes or to recover actinides from a multicomponent solution. Due to their later handling and use, actinide precipitates have to satisfy strict requirements. In nuclear environment where experiments are limited, simulation may be the only way to test different industrial configurations in order to adjust industrial campaigns. In this manuscript, the modelling of the tetravalent uranium oxalate precipitation in an unbaffled reactor is achieved using a hybrid multizonal/CFD model. The population balance is solved according to the moment method in a multizonal model entirely parameterised through computational fluid dynamics calculations based on large-eddy simulation (LES) and a sub-grid model of micromixing. The modelling is applied to show how the specificity of flow field in an unbaffled reactor can have a noticeable effect on the uranium oxalate properties.

1. Introduction

Oxalic precipitation is usually applied in nuclear industry to process radioactive wastes or to recover actinides from a multi-component solution. Due to their later handling and use, actinide oxalate precipitates have to meet very strict requirements. However precipitation reactions are well known to be highly sensitive to many operation parameters such as hydrodynamics, the presence of impurities, ionic strength, temperature, etc. That is why computer simulation appears to be a very effective tool to predict the evolutions of the system subjected to various operating conditions, especially in nuclear environment where experiments are limited. The generation of waste and the radioactive material requirements may make it impossible to perform experiments at industrial scale. In such cases, the simulation is the only way to test different industrial configurations and to adjust industrial campaigns.

This study focuses on the modelling of the precipitation process of tetravalent uranium oxalate in a vortex precipitator. This latter consists in an unbaffled rod-stirred tank reactor operating in the spent nuclear fuel reprocessing industry (Auchapt and Ferlay, 1981; Mahmud et al., 2009). According to Rankine's combined vortex description, the flow generated in such a reactor is characterised by two macromixing zones: the liquid near the axis rotates as a solid cylinder with an angular velocity closed to the agitator one, whereas the outside liquid behaves as a free vortex. The vortex formation has a significant effect on the flow and mixing processes. Consequently, the reactor cannot be assumed as well-mixed and a hydrodynamic model is required to simulate the precipitation process.

As there is little information available in the literature relating to unbaffled reactors especially when they are stirred with a magnetic rod, a hydrodynamic modelling has been developed using computational fluid dynamics (CFD). Due to the absence of baffles, the circulation flow around the impeller axis is predominant so that the turbulence is strongly anisotropic. The classical statistical turbulence models (Reynolds-Average Navier–Stokes (RANS)-like models) commonly used in chemical engineering cannot be then applied and the turbulence transport simulation has been achieved using the large-eddy simulation (LES) approach (Lamarque et al., 2010). In LES, the large scales are solved while the small ones are modelled. Through this model, the effects of sub-grid scales affect the larger scale evolutions. The unsteady flow field can be reached taking into account the real movement of the stirrer. LES appears to be a very promising alternative to RANS techniques and direct numerical simulations (DNS) (Hartmann et al., 2004; Murthy and Joshi, 2008; Delafosse et al., 2008). The first ones present many limitations especially when solving complex flows with highly anisotropic turbulence while the latter one remains still far too expensive for industrial applications. Recently several chemical engineering studies based

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on the large-eddy simulation approach have been published in the literature, in particularly to describe flows in unbaffled stirred tank reactor (Alcamo et al., 2005; Yeoh et al., 2005; Yoon et al., 2009). Results obtained are overall very satisfactory.

Currently a key issue for modelling a precipitation process arises from turbulence and chemistry coupling. Different methodologies can be applied ranging from hybrid methods based on multizonal models (Zhou et al., 1998; Zauner and Jones, 2000; Rigopoulos and Jones, 2003; Kagoshima and Mann, 2006; Bermingham et al., 2007) to a fully coupling. Rigopoulos (2010) proposes a complete review on the methods employed to attack the problem of coupled fluid and population dynamics, in both homogeneous and heterogeneous systems. Bezzo et al. (2004) provide a general methodology to construct hybrid multizonal/CFD models based on a bidirectional coupling between the population balance and the CFD model. In this multiscale modelling, the multizonal model that comprises several well mixed compartments is parameterised through CFD. In other works, the population balance equations have been directly implemented into a CFD code (Wei et al., 2001; Jaworski and Nienow, 2003; Oncul et al., 2006; Zucca et al., 2006). Hybrid methods offer a practical compromise between simplified models based on idealised mixing description and the computationally expensive CFD solution.

Moreover, turbulence–precipitation reaction coupling results in equations with unclosed terms arising from the reaction source terms. When the characteristic time scales associated with the particle formation reaction overlap with the turbulence time scales, fluctuations due to the turbulence cannot be neglected and models are needed to obtain closure (Flamelet, Eddy Dissipation Concept, PDF, etc.).

The objective of this study is to investigate the potential of a multizonal approach in which hydrodynamic information is entirely extracted from LES calculations, to simulate the oxalic precipitation of tetravalent uranium. The suitable zones to describe mixing on the vortex precipitator according to the operating parameters are not set a prior, but optimised from CFD simulations for each operating conditions. A one-way coupling between multizonal and CFD models has been developed since, in the operating conditions considered here, the Stokes number for particles is about 10^{-3} so that the fluid flow properties can be assumed not to be affected by the presence of solid phase. To our knowledge it is the first attempt to apply LES to the modelling of a precipitation process.

The first part of this paper is concerned with the LES simulation of vortex precipitator. The specificity of mixing in such a reactor is highlighted by simulating the injection of a tracer in different zones. Section 3 describes the population balance resolution for the uranium oxalate precipitation using moment approach in a multizonal model. The oxalic precipitation of tetravalent uranium is then simulated in the industrial reactor using the hybrid multizonal/LES model. Computations are performed to see what the impact of the vortex presence has on the properties of actinide precipitates formed.

2. LES modelling of vortex precipitator

Owing to the manipulation of radioactive materials at large scale, nuclear industry has to develop unusual reactor geometries. In particularly, the most commonly impellers, such as paddle or propeller stirrers, are often not suitable and other particular devices have to be used. The aim is, on the one hand, to have as little internal equipment as possible in order to avoid radioactive accumulation and, on the other hand, to minimise the maintenance operations. The industrial precipitator considered in this study (Auchapt and Ferlay, 1981; Mahmud et al., 2009) is an unbaffled cylindrical glass vessel stirred by a cylindrical magnetic rod.

2.1. Calculation parameters

The LES calculations are performed using trio_U, a parallel CFD platform developed at the French Atomic Energy Commission (Calvin and Emonot, 1997; Calvin et al., 2002; Benarafa et al., 2006; www-trio-u.cea.fr).

The computational field is based on the geometry shown in Fig. 1. The tank is supposed to be filled with water initially at rest.

The ratios between the tank diameter D, the tank height H and the rod length L_r are defined as follows (Auchapt and Ferlay, 1981):

$$L_r/D = 0.47 \quad (1)$$

$$H/D = 1.5 \quad (2)$$

The flow is fully turbulent as the impeller Reynolds number Re_a is greater than 10^4 (Perry and Chilton, 1973):

$$Re_a = \frac{ND^2}{\nu} \approx 7 \times 10^4 \quad (3)$$

where N is the stirrer speed and ν the kinematic viscosity.

An unstructured grid is realised with about 650,000 tetrahedral elements (see Fig. 2).

The free surface is modelled using the Discontinuous Front Tracking (DFT) method (Mathieu et al., 2003) and is defined by a moving Lagrangian grid independent from the Eulerian finite

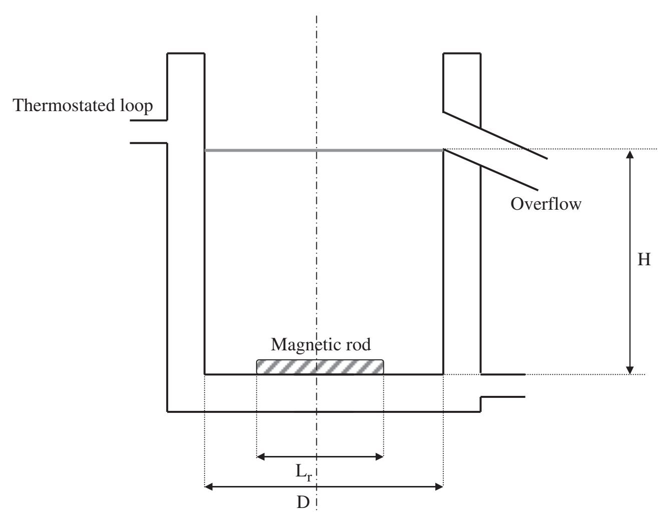

Fig. 1. Precipitator scheme.

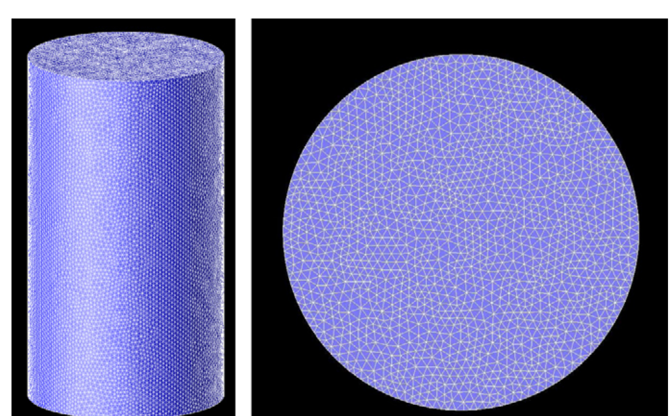

Fig. 2. Unstructured grid of the precipitator with tetrahedral elements.