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Abstract Submission

Submission-694

Topic : / Chemical reaction engineering / ECCE10 / Reaction mechanism and kinetics

STUDY AND MODELLING OF HETEROGENEOUS CATALYTIC REACTIONS. APPLICATION TO URANIUM DIOXIDE DISSOLUTION.

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Authors of the abstract come from :: Industry

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Dissolution is a key step in several industrial processes. It is especially a milestone of the head-end of many hydrometallurgical processes. For example, in recycling of spent nuclear fuel, the solubilization of the chemical elements is essential before performing the liquid-liquid extraction steps to separate reusable material and final waste. One of the most complex scenarios is that of heterogeneous autocatalytic reactions. Today, there are few satisfying models for these cases due to a lack of comprehension of their mechanisms.

We focus here on the dissolution of uranium dioxide in nitric medium. In order to propose optimized processes for dissolution, this study aims at better understanding the chemical, physico-chemical and hydrodynamic phenomena of such reactions. This study is also part of a modeling approach aiming, on one hand, at expressing the intrinsic reaction rates and describing the physico-chemical phenomena at interfaces and, on the other hand, at developing a general model for dissolution reactors.

Results :

Optical microscopy observation confirmed the highly autocatalytic nature of the reaction and led to measurements, for the very first time, of "true" chemical kinetics of the reaction. The acid attack of sintering-manufactured solids occurs through preferential attack sites. It develops cracks in the solids that can lead to their cleavage. This inhomogeneous attack is made possible by the establishment of bubbling in the cracks which allows periodic renewal of the reagents and thus maintains the reaction within the cracks. This point is a key component of the mechanism: a strong link among the development of cracks, bubbling through the cracks, and overall dissolution kinetics is demonstrated in this work.

A model coupling material balance to the structural evolution of the solid and liquid phase compositions, and taking into account the interfacial transport is proposed. The simulations based on this model are close to the experimental observations, and allow to replicate the effect of various reaction parameters for the very first time, such as the reduction of overall kinetics when turbulence increases.

Type of presentation :: Oral

Highlight 1: Optical microscopy is used to determine liquid-solid reactions mechanisms and kinetics.



Highlight 2: A strong link exists among development of cracks, bubbling and overall dissolution rates.

Highlight 3: A model for dissolution is proposed to evaluate the influence of reaction parameters.

Reference 1 :: P. Marc, 2014. " étude de réactions hétérogènes autocatalytiques. Application à la dissolution du dioxyde d'uranium". Thèse de l'université de lorraine.

Keywords: Chemical kinetics, Hydrodynamics, Mass transfer, Modelling, Multiphase