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Thermodynamic and kinetic study of uranium peroxide precipitation

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In the processing of uranium ores, uranium is recovered from mill leach solutions and is transported to a processing plant. This step allows obtaining an uranium concentrate by precipitation. After drying, a solid uranium concentrate is produced (called "yellow cake" due to its color and its doughy texture at the end of the procedure), containing around 75% uranium. The "yellow cake" is sent to conversion facilities for further chemical processing.

Among the different existing precipitation devices, the continuous precipitation with hydrogen peroxide in a fluidized bed reactor leads to high-quality solid particles. The precipitation is achieved at pH ~ 3 by mixing hydrogen peroxide and uranyl sulfate solutions, according to the following equation:

$$UO_2SO_4 + H_2O_2 + 4H_2O \leftrightarrow UO_4 \cdot 4H_2O + H_2SO_4$$
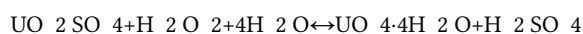
Solid particles must fulfill specific requirements, in particular concerning the size. Thus, the control of the crystal growth becomes a key parameter during the uranium peroxide precipitation. Moreover, the solubility product, K_s, is required to calculate the driving force on which the kinetic law depends. For the uranium peroxide precipitation, the supersaturation ratio, S, is linked to the activities of the uranyl and peroxide ion as:

$$S = \sqrt{(a(UO_2^{2+})\, a(O_2^{2-}))/K_s}$$
with:

$$K_s = a(UO_2^{2+})_{eq}\, a(O_2^{2-})_{eq}$$

where $a(UO_2^{2+})_{eq}$ and $a(O_2^{2-})_{eq}$ are the uranyl ion activity and the peroxide ion activity at equilibrium.

The uranyl ion activity is obtain as the product of the uranyl activity coefficient and the uranyl free concentration, which are calculated from the Specific Interaction Theory and total concentration balance, respectively. Considering the experimental data of the dissociation constants of hydrogen peroxide and knowing that it is not very dissociated in solution, the expression of the supersaturation ratio becomes:

$$S = \sqrt{(\gamma_{(UO_2^{2+})}\, [UO_2^{2+}][H_2O_2])/a(H^+)^2\, (K_{A3}K_{A4})/K_s}$$

where K_{A3} and K_{A4} are the dissociation constants of hydrogen peroxide and $a(H^+) = 10^{(-pH)}$.

In order to calculate the supersaturation ratio, the ratio $(K_{A3}K_{A4})/K_s$ is determined from the speciation study by a regression technique.

Once the thermodynamic law is established, the determination of the crystal growth rate expression for modelling the precipitation processes is performed. The experimental study is carried out in a glass baffled batch reactor equipped with a marine propeller and a wide range of experimental conditions.

Particle size distributions are measured before and after the growth tests; the results show that the distribution remains practically unchanged. Consequently, the two distributions confirm the absence of agglomeration, breakage and nucleation. In addition, processing experimental data, it is found that the crystal growth is linearly dependent on the supersaturation. Finally, crystal growth kinetics is independent from the impeller speed.

All the information obtained by this study will be used for modelling the precipitation of uranium peroxide in the fluidized bed reactor.

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