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Comparison of the method of classes and the quadrature of moment for the modelling of Neodymium Oxalate Precipitation

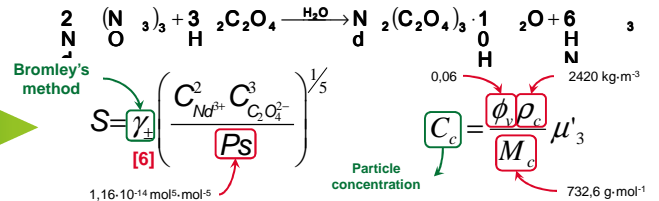
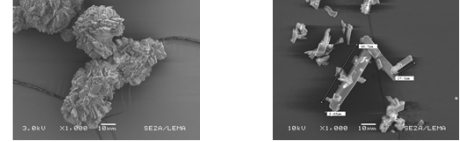
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Introduction

- Oxalic precipitation :
 - to deal with radioactive waste and recover the actinides lanthanides → To facilitate the development of experimental methods
- Modelling approach:
 - Experimentation → Thermodynamics + kinetics & numerical methods [1] [2] [7] [4] [5]

Nd₂(C₂O₄)₃·10 H₂O obtained for precipitation different conditions



Thermodynamics et kinetic laws

- Supersaturation ratio (S) → The driving force of the precipitation process
- Kinetic laws of nucleation, cristal growth and agglomeration = f (S)

Homogeneous primary nucleation

$$R_N = 3 \cdot 10^{31} \exp\left[-\frac{67600}{RT}\right] \exp\left[-\frac{187}{(LnS)^2}\right]$$

S > 50 293K < T < 333K

[1] [8] [9]

Crystal growth

$$G = 2,9 \cdot 10^{-6} \exp\left[-\frac{14000}{RT}\right] (Ps)^{1/5} (S-1)$$

293K < T < 333K

[2] [3]

Agglomeration

$$\beta = 2,55 \cdot 10^{-7} \cdot I^{-0,70} \cdot S \cdot \dot{\gamma}^{-0,24} \exp\left[-\frac{40900}{RT}\right]$$

S > 61 293K < T < 333K [7]

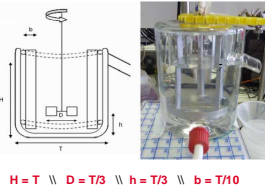
45s⁻¹ < $\dot{\gamma}$ < 1024s⁻¹ $\dot{\gamma} = \sqrt{\frac{\bar{E}}{V}} \bar{E} = \frac{Np \cdot N^3 \cdot D^5}{V}$

600mol·m⁻³ < I < 2100mol·m⁻³

Experimental Study

MSMPR

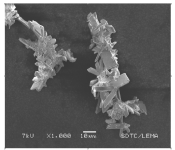
- Continuous precipitation until steady state
 - V = 200 ml
 - Temperature = 20° C
 - four stainless steel baffles
 - stainless steel four 45° pitched blade turbine → Np = 1,5



Continuous experiments

- Short mean residence time ≈ 1 min → high S
- Constant agglomeration kernel : β
- Scanning Electron Microscopy → crystal size distributions

MEB observations of Loose agglomerates



	N tr/min	C _{Nd,0} mol·m ⁻³	C _{Ox,0} mol·m ⁻³	s ⁻¹	L _{4,3} μm
Run 1	1000	142,2	213,7	362	65
Run 2	2000	142,2	213,7	1024	41

Two population balances

Method of classes

$$\frac{dN'_k}{dt} + \frac{N'_k}{\tau} = 0^k R_N + \frac{1}{\Delta L_k} [F(G_{k+1/2}) - F(G_{k-1/2})]$$

Elementary particles 3rd order scheme of Koren [4]

$$\frac{dN_k}{dt} + \frac{N_k}{\tau} = \frac{N'_k}{\tau} + B_k - D_k$$

Loose agglomerates scheme of Litster [5]

Quadrature of Moments

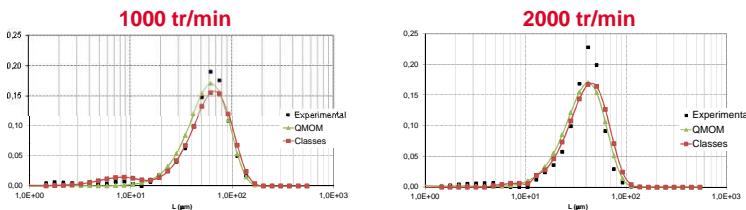
$$\frac{d\mu'_k}{dt} + \frac{\mu'_k}{\tau} = 0^k R_N + k G \mu'_{k-1}$$

for k = 0, 1, 2, ..., 2n-1 [10]

$$\frac{d\mu_k}{dt} + \frac{\mu_k}{\tau} = \frac{\mu'_k}{\tau} + \frac{\beta}{2} \sum_{i=1}^n w_i \sum_{j=1}^n w_j (L_i^2 + L_j^2)^{k/3} - \frac{\beta}{2} \sum_{i=1}^n w_i L_i^k \sum_{j=1}^n w_j$$

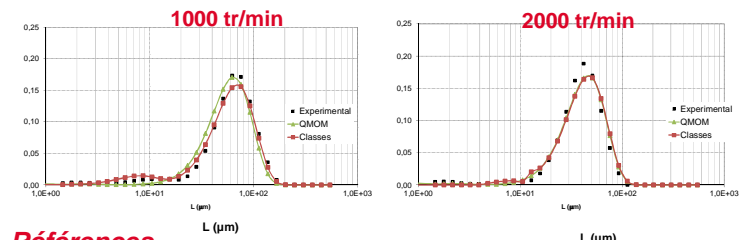
Main results

- Comparison of volume fractions at 5 τ



- Crystal size distributions → Experimental vs. Predicted

- Comparison of volume fractions at steady state



Conclusions

- Kinetic laws (R_N, G et β) and loose agglomerates from experimental runs
- Two population balance models : solved by the Method of classes and QMOM
- Both methods compared well with experimental data during transient and at steady state
- QMOM required much less computational effort and is preferentially used with the reconstruction method detailed in [10]

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