Modelisation of the thermal decomposition of uranium oxalate

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Among the processes that could be used to synthesize actinide oxides for nuclear fuel recycling purpose, one well-known method is the thermal treatment of actinide oxalates under controlled atmosphere.

In fact, the actinide is first stabilized by a precipitation step, then the actinide oxalate is calcined in order to obtain an oxide powder to be shaped and sintered during the fabrication process. The transformation of actinide oxalate into the final oxide consists in a succession of transformations modifying the characteristics of the solid such as composition, specific surface area, granulometry, impurity content,... The thermal treatment leads to a complex evolution of the solid phase with some gaseous releases. A better understanding of gas-gas and gas-solid reactions is essential to be able to predict the final characteristics of the oxide according to the thermal parameters.

In this work we used differential scanning calorimetry and thermogravimetric analysis with evolved gas analysis in order to investigate the thermal behavior of uranium oxalate in various atmospheres. Contrary to previous studies^[1,2], a set of solids characterizations (morphological, textural and structural) was used to unambiguously indentify the reaction products. Among the seven stages involved during the global reaction, the kinetics of two of them were specifically investigated. The experimental kinetic data in isobaric and isothermal conditions are explained thanks to kinetic modelling based on the characterizations of solid and heterogeneous kinetics approach^[3]. This allows us to obtain predictive model which includes the effect of temperature and partial pressure of some gases of interest, as well as the shape and size of the solid phases.

REFERENCES

- [1] L. Duvieubourg-Garela, N. Vigier, F. Abraham et S. Grandjean, Journal of Solid State Chemistry, 181 (2008) 1899-1908.
- [2] G.K. Çılgı, H. Cetişli, et R. Donat, Journal of Thermal Analysis and Calorimetry, 115 (2014) 2007-2020.
- [3] M. Pijolat, L. Favergeon, Chapter 5: Kinetics and mechanisms of Solid-Gas Reactions, pp. 173-212 in: "The Handbook of Thermal Analysis & Calorimetry, Vol.6: Recent Advances, Techniques and Applications, 2nd ed.", Eds. S. Vyazovkin, N. Koga, C. Schick, Elsevier, (2018) 842pp.