Morphology dependent sintering path of nanocrystalline ThO_2

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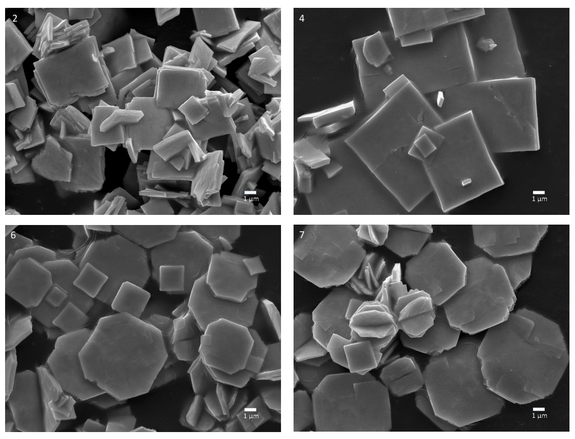
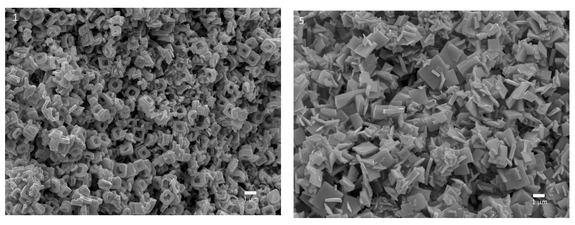
Background and objectives

Oxalates are commonly used as precursors for the preparation actinide dioxides and sesquioxides. Oxalates precipitate with high efficiency in a wide range of conditions, including at high acidity after precipitation. Unfortunately, the crystals of calcined oxide remains trapped in the shape given by the oxalate, platelets for ThO_2. We studied the effect platelet shape had on the preparation of ThO_2 pellets. We tested the performance of a variety of platelet shapes during pressing, the intermediate sintering stage and the final sintering stage

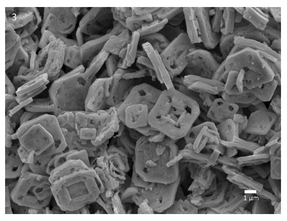
Pressing stage



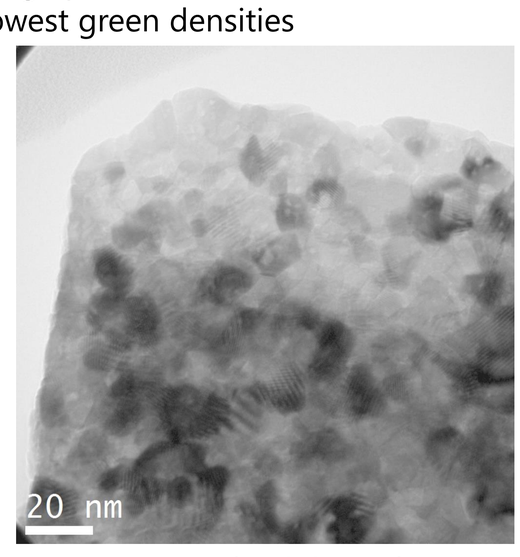
Large platelets reach good green densities



Small platelets reach lower green densities



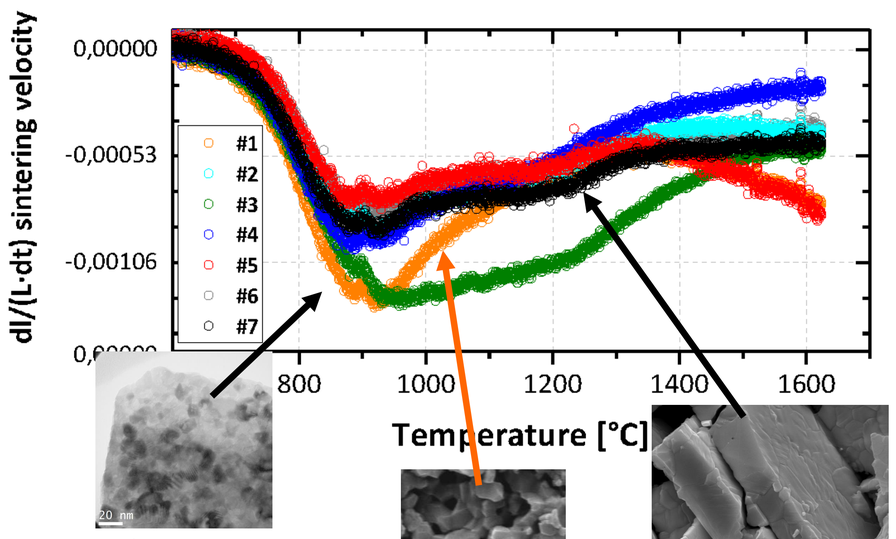
Large platelets with holes reach the lowest green densities



All platelets contain ThO_2 grains dispersed in the platelets.

Intermediate sintering stage

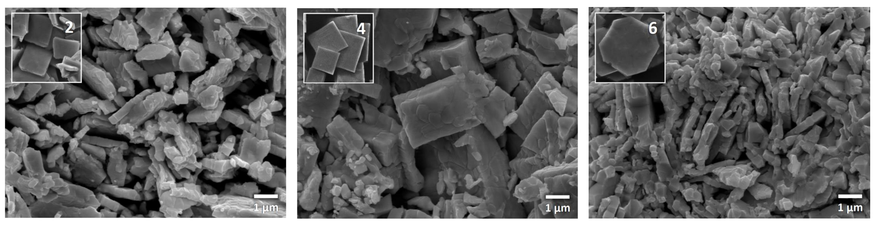
The densification of pellets from oxalate takes place in two general steps. First between ThO_2 grains in platelets. The second between platelets.



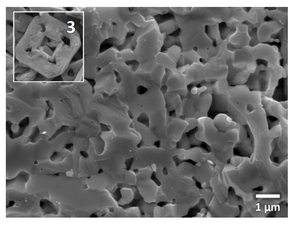
First step ThO_2 in platelets

no platelets, no second step

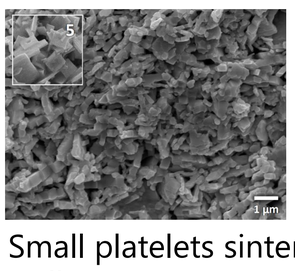
Second step Between platelets



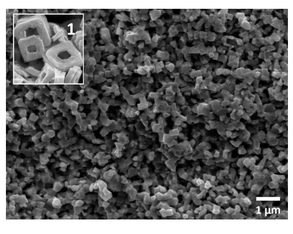
After high green densities, large platelets reach mediocre densities.



Large platelets with holes show good necking



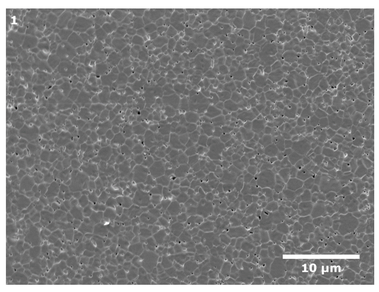
Small platelets sinter well



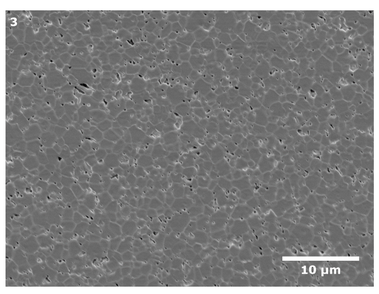
Small platelets with holes sinter to the highest intermediate densities

Final sintering stage

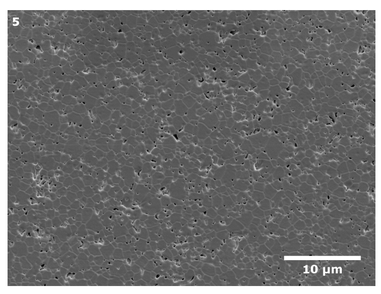
After sintering at 1750°C, further differences emerge in microstructure and density.



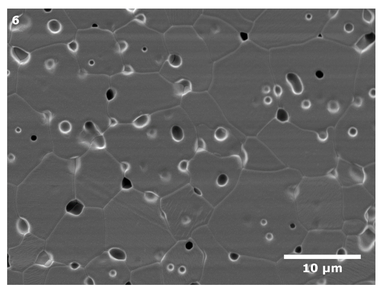
Small platelets with holes reach 97% TD.



Large platelets with holes reach 95% TD.



Intact small platelets reach 95% TD.



Intact large platelets reach 93% TD.

The lower density large platelet grade has significantly higher grain sizes and larger pores.

Conclusion and outlook

Three general groupings exist for ThO_2 derived from oxalate: large platelets, small platelets and platelets with holes.
Large platelets stack well and do not sinter well in the intermediate stage. In the final stage they do not reach sufficiently high density, but achieve good grain sizes.
Small platelets achieve lower green densities, but sinter well in the intermediate stage. In the final stage they reach sufficiently high density, but grain pinning limits grain growth to below nuclear fuel specifications.
Platelets with holes also achieve lower green densities, but sinter well in the intermediate stage. In the final stage they also reach sufficiently high density, but grain pinning limits grain growth to below nuclear fuel specifications.

No powder type achieves the correct combination of high density and large grain sizes. Since the powder preparation was not optimized, room remains to improve. Powder processing changes may be enough to sinter large platelet grades to >95% density. Alternately, a small admixture of large platelets may be sufficient to eliminate grain pinning without compromising the sinterability.

Acknowledgements: This work was cofunded by Solvay