Investigation of fabrication routes of model fuels with tailored microstructures for leaching studies

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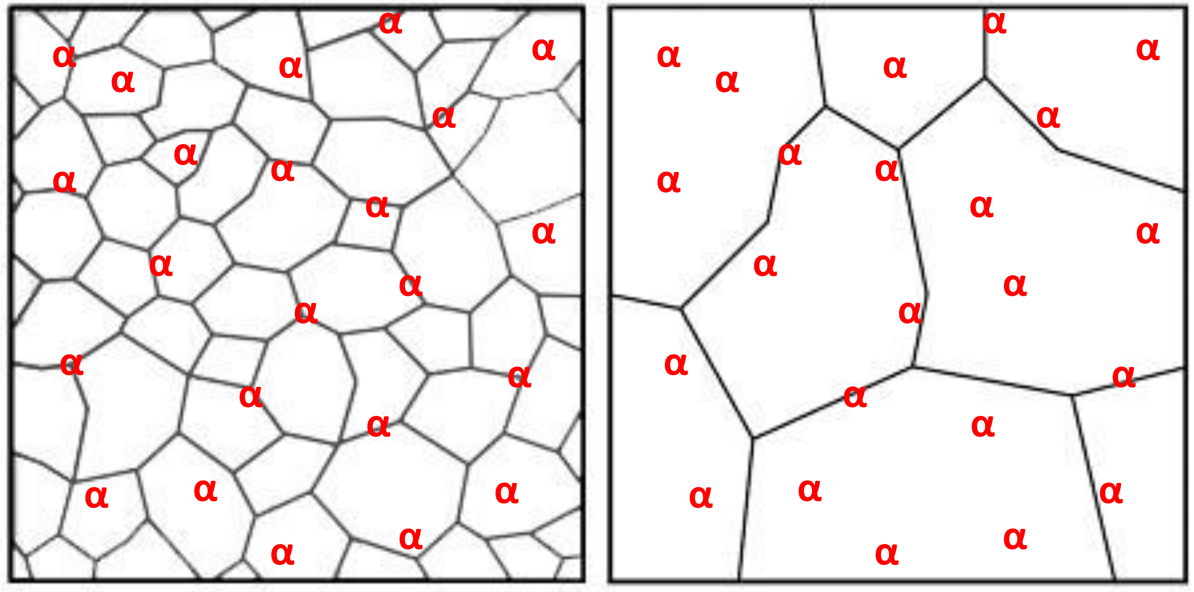
Goals

- DISCO project : prepare fuel to compare leaching behavior of alpha-doped UO2 with small and large grain microstructure

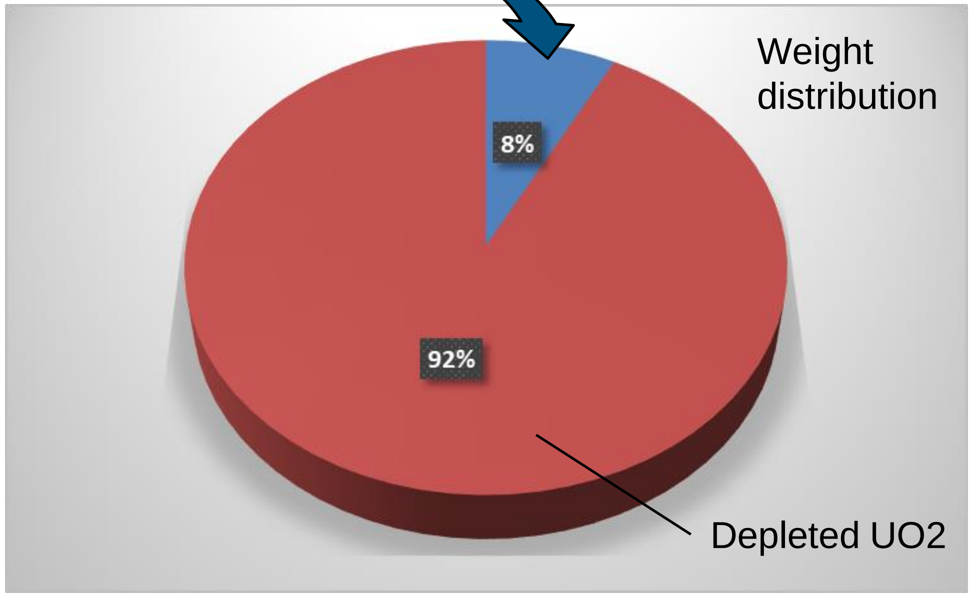


- Fuel preparation done in collaboration with FzJulich (see Philip Kegler presentation tomorrow)

^{238}Pu & ^{233}U doped U batch made by ITU

Main isotope	Specific activity	Mass		Activity	
	Bq/g	g/gUO2	%	Bq/gUO2	%
^{233}U	3.63E+08	3.69E-03	**0.42%**	1.34E+06	**0.550%**
^{234}U	2.33E+08	2.38E-04	0.03%	5.54E+04	0.023%
^{238}U	1.26E+04	8.73E-01	99.51%	1.10E+04	0.005%
^{238}Pu	6.29E+11	3.85E-04	**0.04%**	2.42E+08	**99.394%**
^{239}Pu	2.33E+09	1.53E-05	0.00%	3.57E+04	0.015%
^{240}Pu	8.51E+09	4.02E-06	0.00%	3.42E+04	0.014%

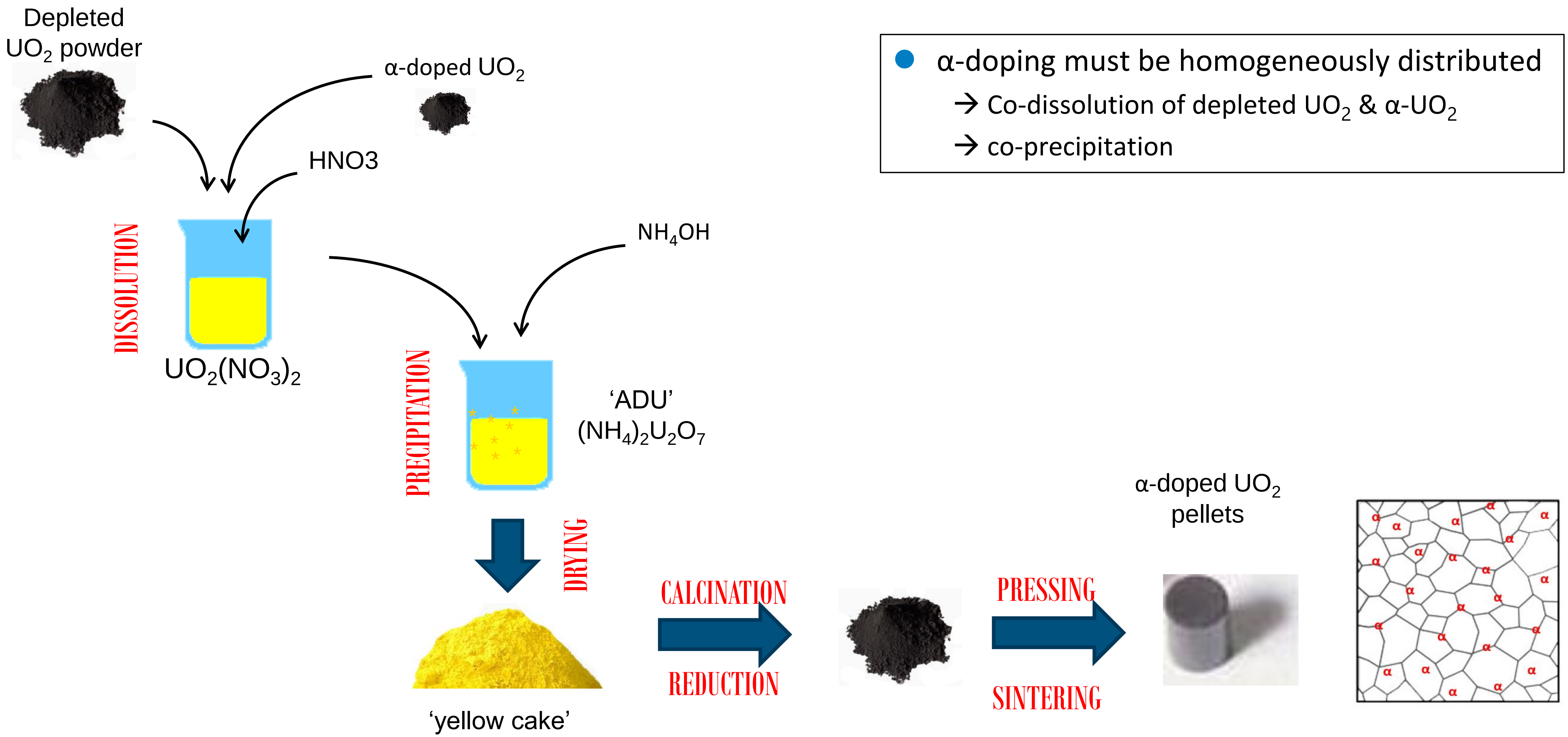
Total alpha specific activity
2.44E+08
Bq/gUO2



Dilution in depleted UO2 to reach an activity of 1.9E+08 Bq/gUO_2 = 10 000 years old simulated fuel

α-doping

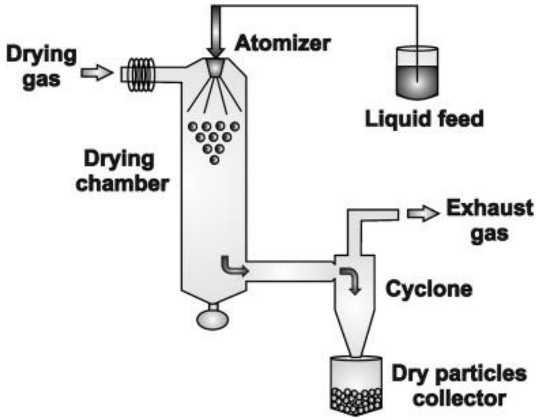
- α-doping must be homogeneously distributed
  - → Co-dissolution of depleted UO_2 & α-UO_2
  - → co-precipitation

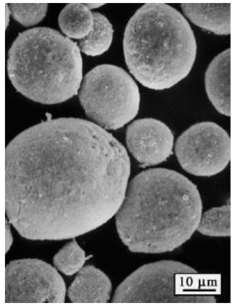


Cr-doping for grain growth

- Cr-doped fuel is now a product offered by the industry for large grain fuel
  - Both Framatome and Westinghouse have now chromia-doped fuel pellet in their line-up
  - Increase fission products retention and pellet mechanical compliance
- However, open literature on fabrication processes is scarce
- Most information comes from the seminal work of L. Bourgeois on spray-dried powder

Spray-drying of
UO2 powder suspension in
H2O + (NH4)2CrO4
L. Bourgeois seminal work
JNM 297 (2001) 313-326



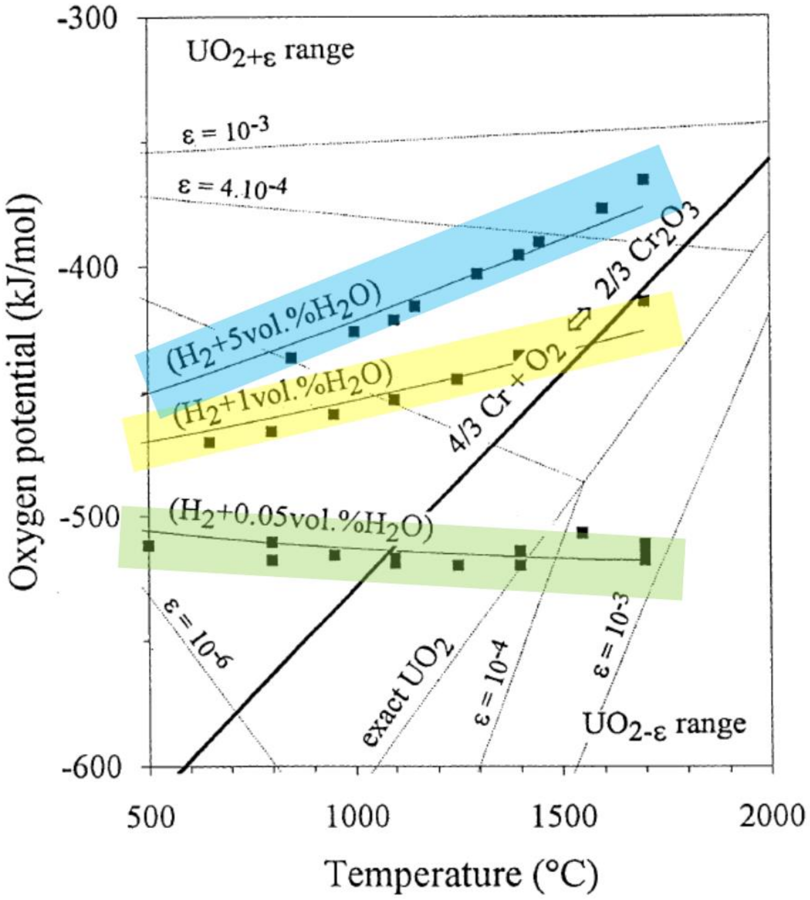


Dry-mixing of UO_2 powder and Cr2O3

→ Reference industrial process
→ Detailed study not available

Knowledge on Cr-doping from open literature

- Most of publicly available data is from L. Bourgeois, *JNM 297 (2001) 313-326* on spray dried powder
- Effect of sintering atmosphere on grain growth



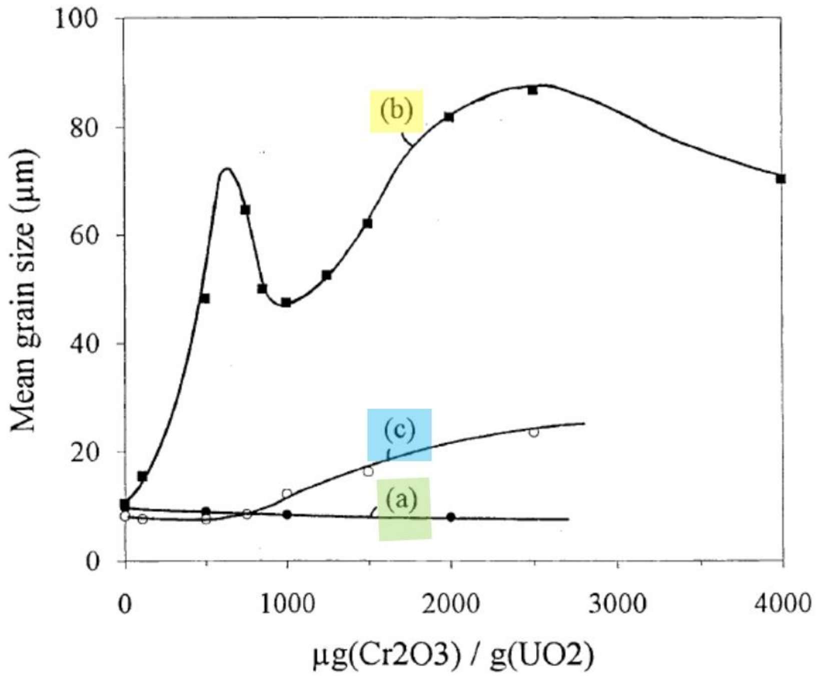
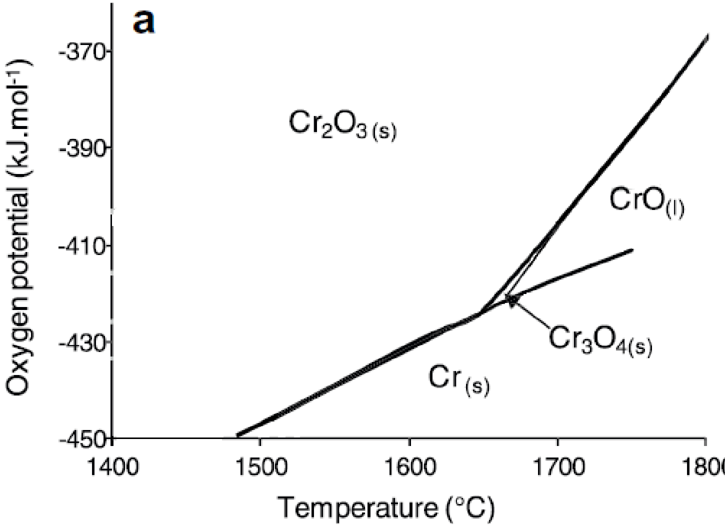


Fig. 3. Relationship between mean grain size and Cr_2O_3 content (after standard heat treatment). (a) $H_2 + 0.05$ vol.% H_2O, (b) $H_2 + 1$ vol.% H_2O and (c) $H_2 + 5$ vol.% H_2O.



Knowledge on Cr-doping from open literature

- Effect of sintering temperature

Hold temperature

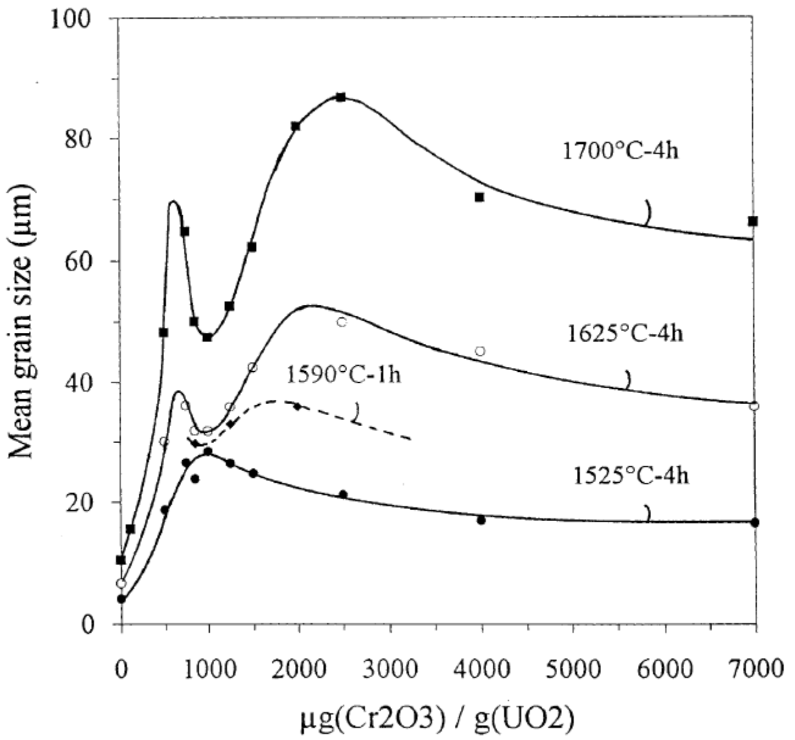


Fig. 5. Relationship between mean grain size and Cr_2O_3 content in H_2 + 1 vol.% H_2O (after standard heat treatment).

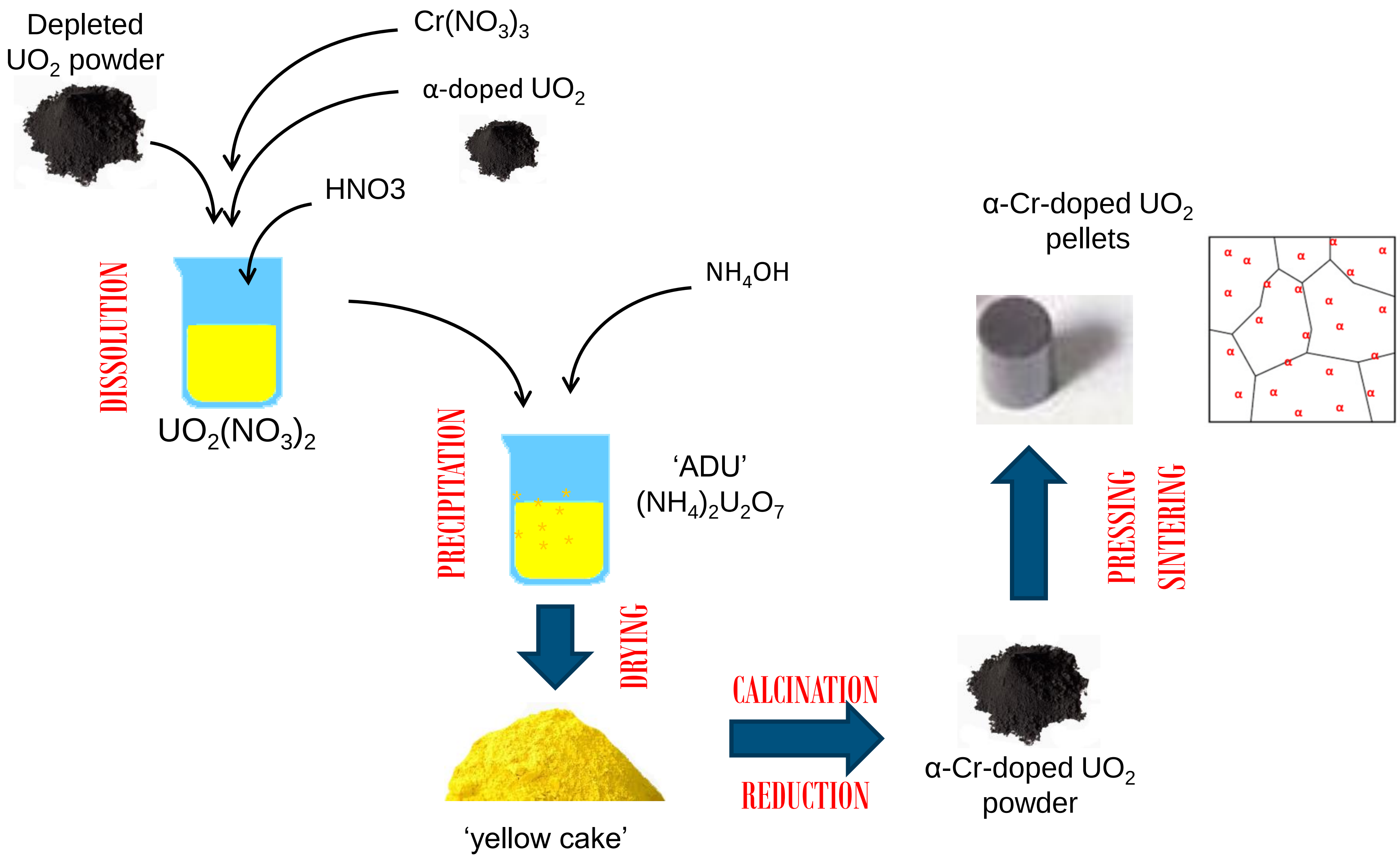
Heating rate

Influence of heating rate on grain size of Cr_2O_3-doped fuel

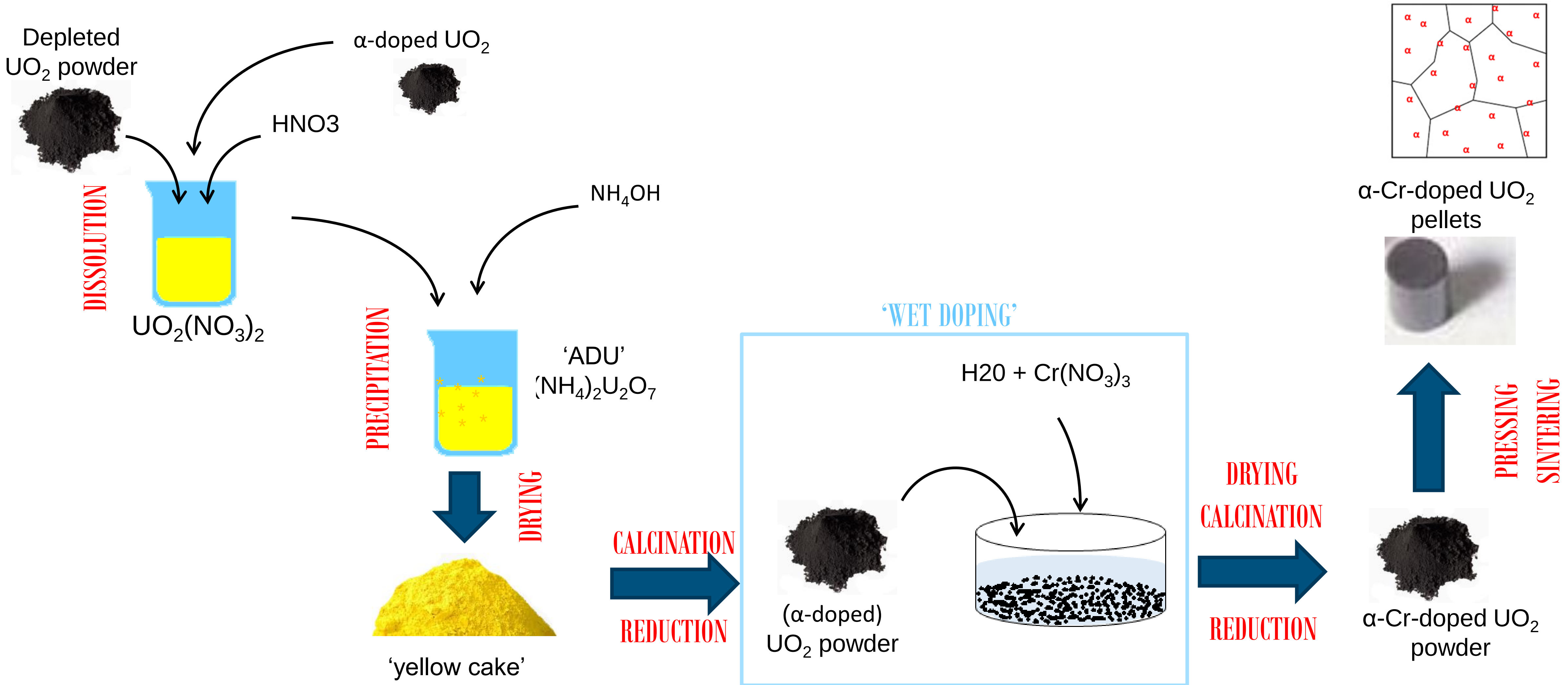
Heating rate (K h^{-1})	Grain size Relative density, ρ/ρ_{th} (%)		
	Cr_2O_3 (wt%) 0.07	Cr_2O_3 (wt%) 0.075	Cr_2O_3 (wt%) 0.2
75	49.6 µm$^{+0.7}_{-1.2}$ 99.39	45.9 µm$^{+1.2}_{-1.5}$ 99.27	73.2 µm$^{+0.6}_{-0.7}$ 99.27
100	– –	– –	68.4 µm$^{+0.8}_{-1.1}$ –
150	35.4 µm$^{+1.2}_{-1.0}$ 99.45	32.2 µm$^{+1.2}_{-1.8}$ 99.39	63.4 µm$^{+0.7}_{-0.8}$ 99.41
300	21.5 µm$^{+0.3}_{-0.5}$ 99.30	19.5 µm$^{+0.5}_{-0.4}$ 98.92	38.0 µm$^{+0.5}_{-0.6}$ 98.97
500	18.3 µm$^{+0.5}_{-0.8}$ 99.35	15.3 µm$^{+0.5}_{-0.7}$ 99.05	32.8 µm$^{+0.3}_{-0.3}$ 99.19

→ The slower the rate, the larger the grains

α + Cr doping: choice of fabrication route → co-precipitation

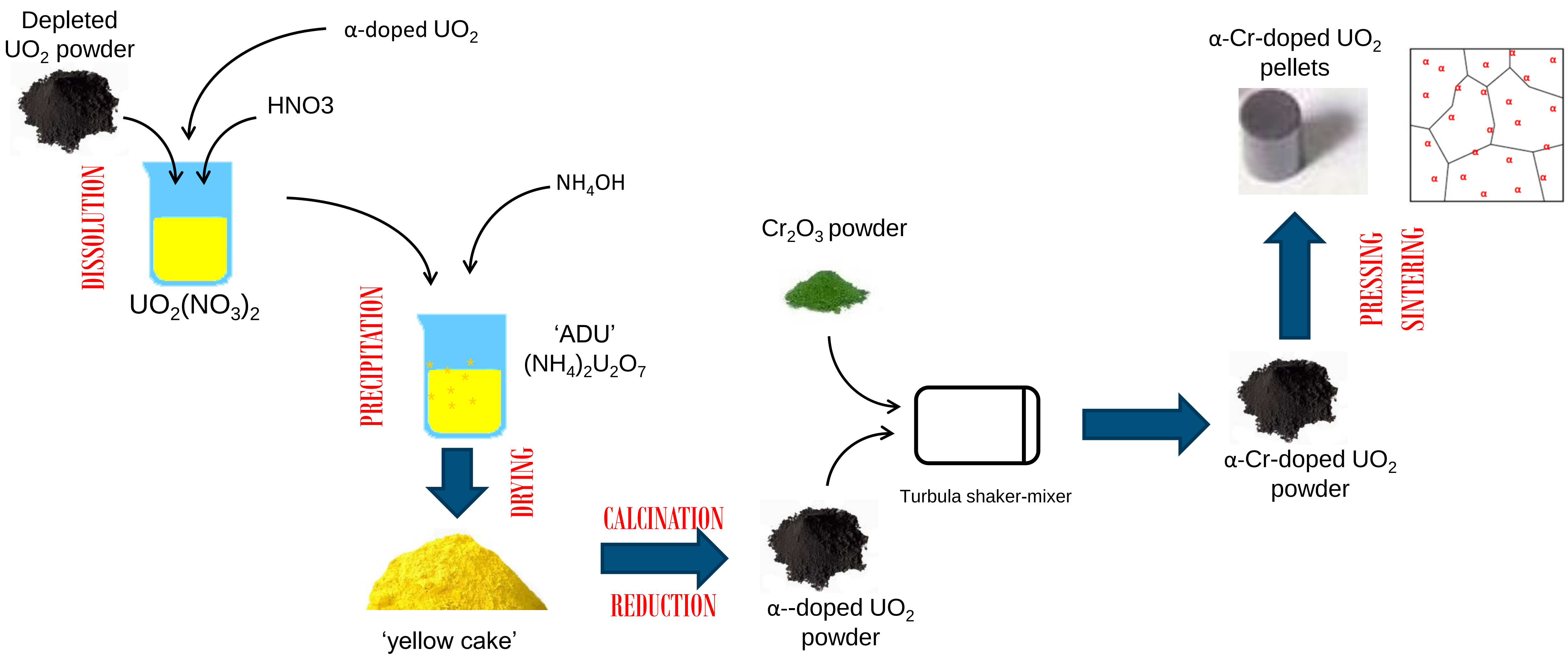


α + Cr doping: choice of fabrication route → co-precipitation + wet-coating



This step is skipped in case of Cr-doping by co-precipitation

α + Cr doping: choice of fabrication route → co-precipitation + dry-mixing

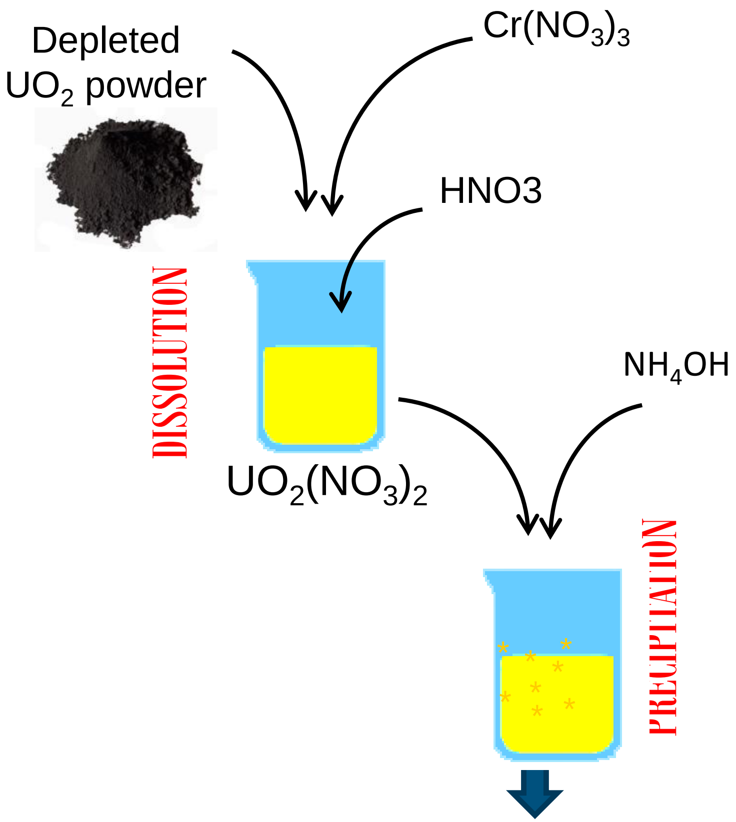


Alpha-doping in glovebox line

Testing fabrication routes for Cr-doping

Co-precipitation

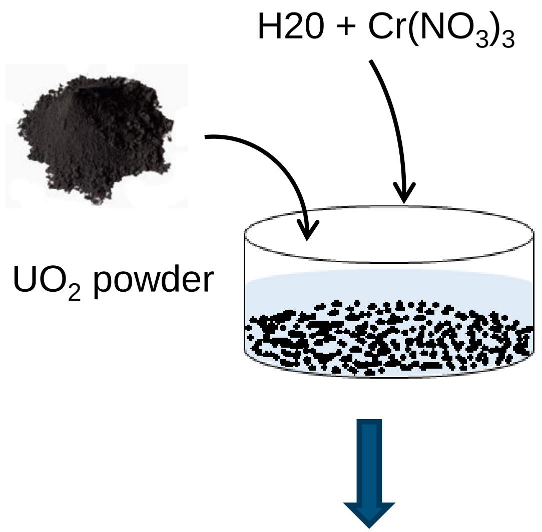
+ easiest to implement in GB



Drying
Powder processing
Pelletization / Sintering

Wet-coating

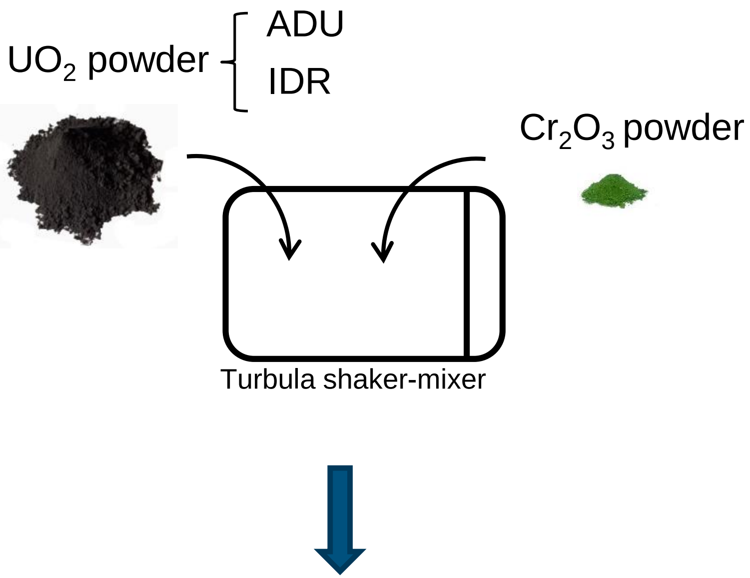
+ closest to spray-drying process for which detailed parametric studies exist



Drying
Powder processing
Pelletization / Sintering

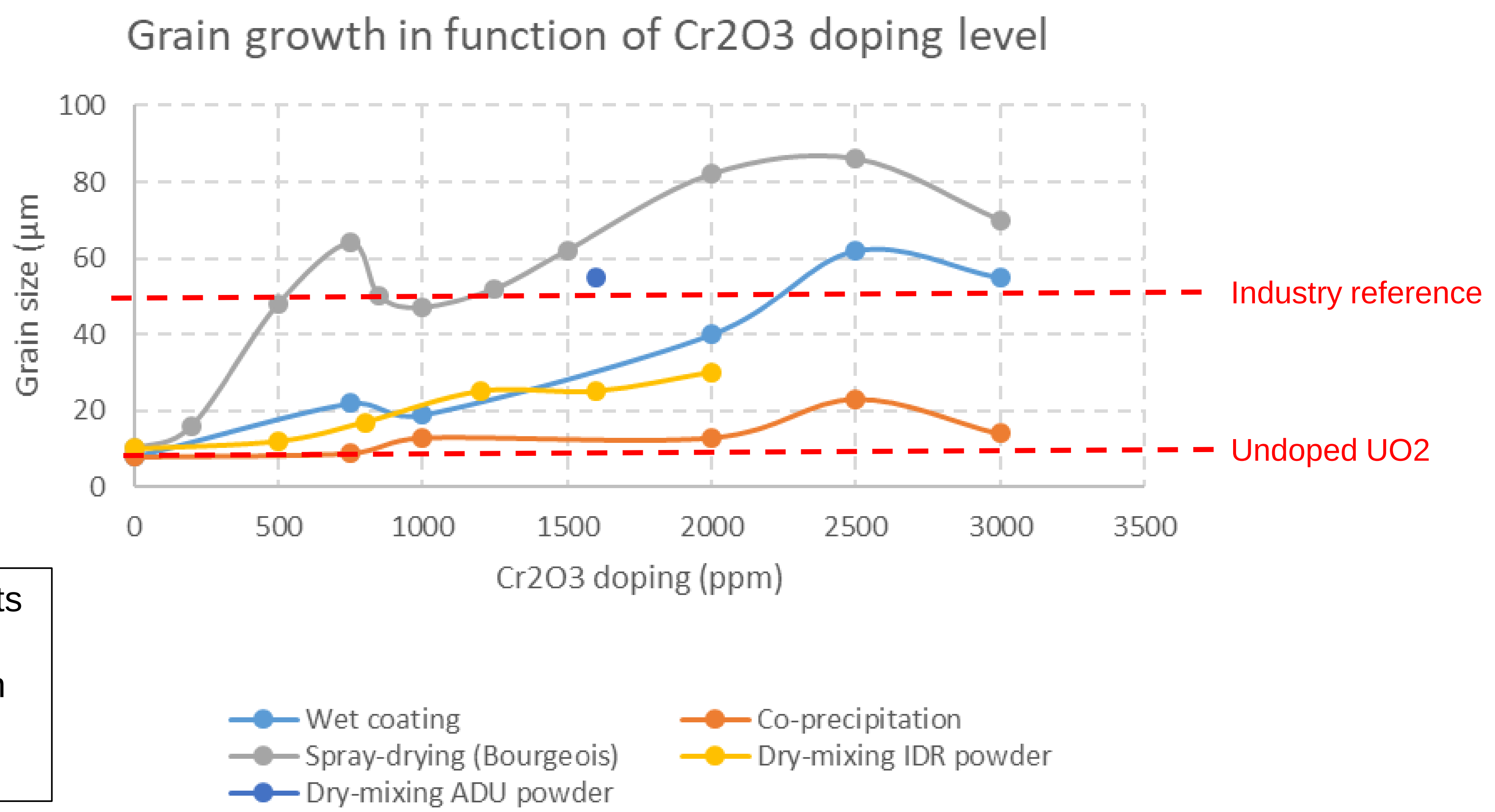
Dry-mixing

+ closest to industrial process



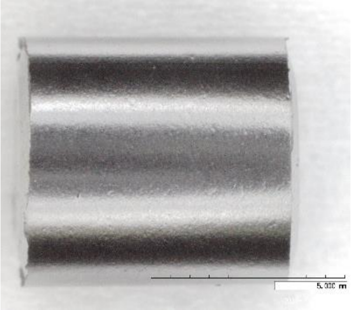
Mixing
Powder processing
Pelletization
Sintering

Results on grain growth



A series of promising results for grain growth has also been achieved at FzJulich (see presentation Philip Kegler tomorrow).

Grain size distribution not always perfectly homogeneous



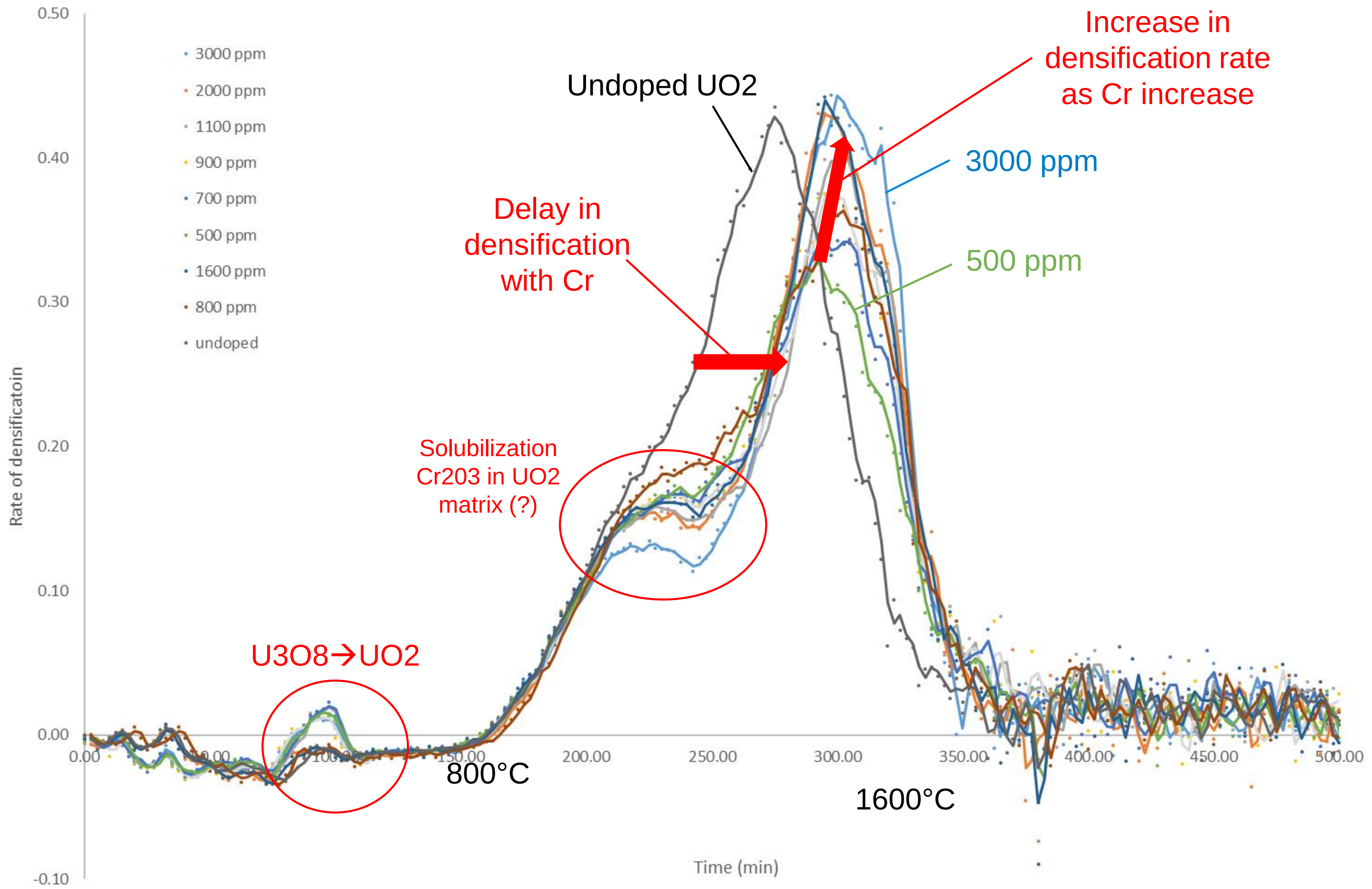
Dry-mixing
with ADU
powder

Optimized parameters

- Several additional batches are currently being analysed. They have been fabricated with 'optimized' parameters:
  - Oxygen potential @1700°C -420 kJ/mol
  - Heating rate decreased to 150°C/h in the 1200-1700°C range
  - High purity Cr_2O_3 powder
  - For ADU powder: sieving/crushing/pre-compaction+granulation help to improve pellet microstructure
  - ~15 wt. % addition of U_3O_8 is required to lower density to desired specs 95±1%TD

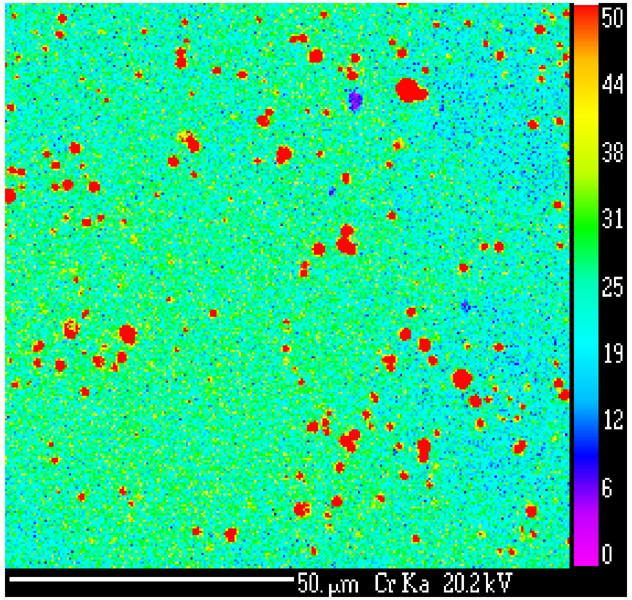
- Final production for DISCO with optimized parameters/route shall follow
  - 10 pellets reference UO_2
  - 10 Cr-doped UO_2
  - 10 α-doped UO_2
  - 10 α-Cr-doped UO_2

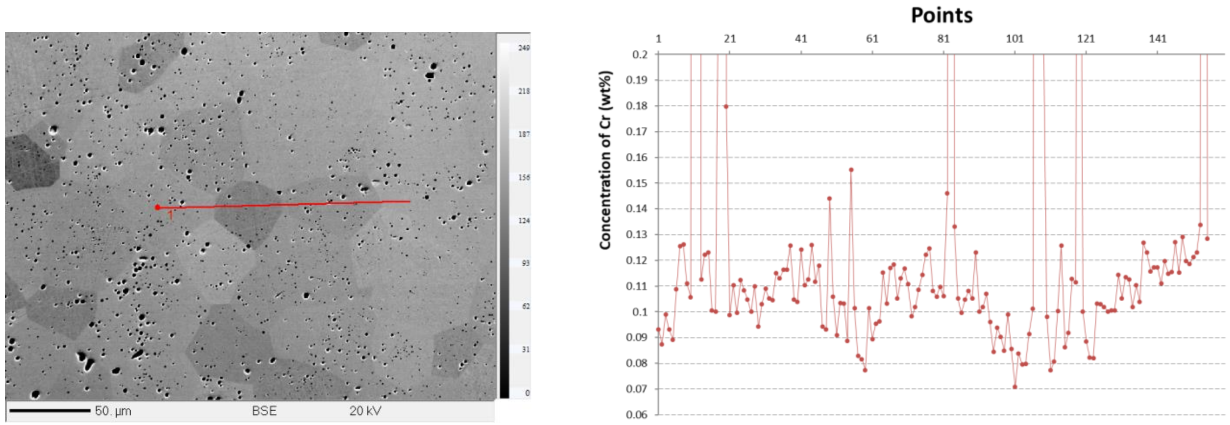
Going further: dilatometry



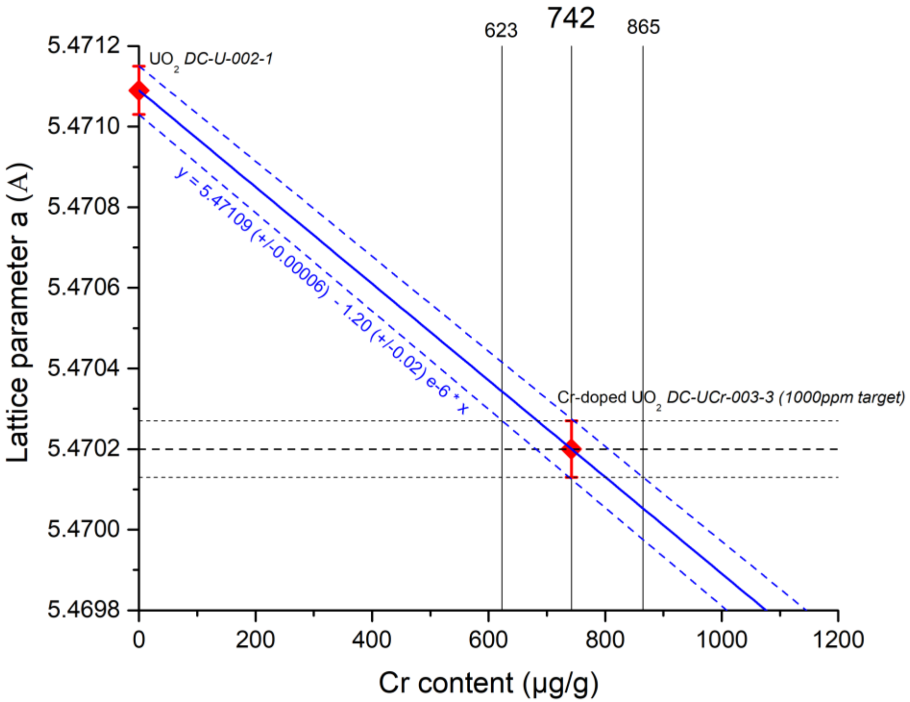
Going further: EPMA & XRD

EPMA





XRD



Oulook on doped fuels

- Approached by consortium NFIR to irradiate doped fuel discs in BR2
- A range of doped fuels shall be fabricated in our lab under guidance of NFD (Japanese Fuel Fabrication Lab) who has experience in their fabrication
- Fuels proposed:
  - Nb-doped UO_2
  - Mg-doped UO_2
  - Alumino-silicate UO_2
  - Al_2PO_4-doped UO_2
  - Kaolinite doped UO_2
  - Mo-doped UO_2