GOBIERNO DE ESPAÑA
MINISTERIO DE CIENCIA, INNOVACIÓN Y UNIVERSIDADES

Ciemat
Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas

Alteration of advanced doped UO_2 fuel in repository conditions

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Content

1. Introduction

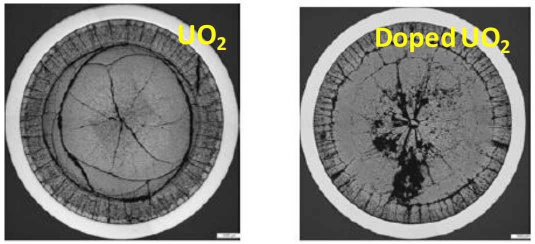
Advanced Doped Pellet Technology: Optimization in-pile behaviour

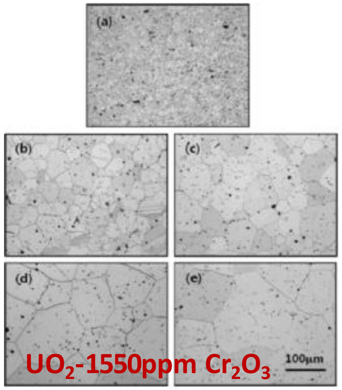
Cr_2O_3
- ↑ grain size
- ↑ R corros

⟷ synergy ⟷

Al_2O_3
- enhances the grain size enlarging effect of Cr_2O_3

- **ADOPT** (<u>Ad</u>vanced <u>DO</u>ped <u>P</u>ellet <u>T</u>echnology): **UO_2 - Cr_2O_3 + Al_2O_3** (Westinghouse)
  - Synergy between the two additives:
    - pellet densification during sintering & enlarge the pellet grain size ➔ > 35 µm
    - reduced Fission Gas Release (FGR)
    - improved Pellet Cladding Interaction (PCI) ➔ increased pellet plasticity and higher resistance against post-failure degradation
- **GAIA Cr-doped UO_2** (AREVA) 0.16 $^w/_o$
    - enhanced PCI resistance and lower risk of clad failure
    - reduction of FGR and induced rod internal pressure at the end of life
    - grain size ➔ ~ 40-50 µm
- **Gd-doped UO_2** (<10 $^w/_o$) = f(country, PWR/BWR)

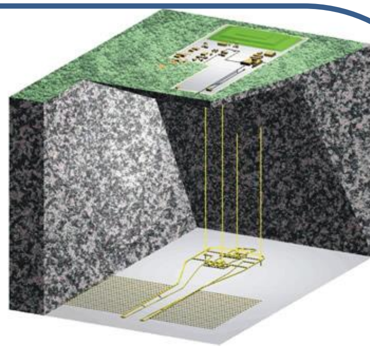


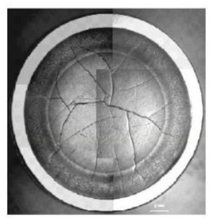


IAEA-TECDOC-1654 "Advanced Fuel Pellet Materials and Fuel Rod Design for Water Cooled Reactors " 2010
Arborelius et al. J. Nuc. Sci. tech. 43(2006) 967-976
Yang et al. Trans. of the Korean Nuc. Soc. Spring Meeting Jeju, Korea, 2009 (481-482)
Yang et al. J. Nuclear Materials 429 (2012) 25-33
2014:21 Effects of additives on U dioxide fuel behavior

1. Introduction

DGR

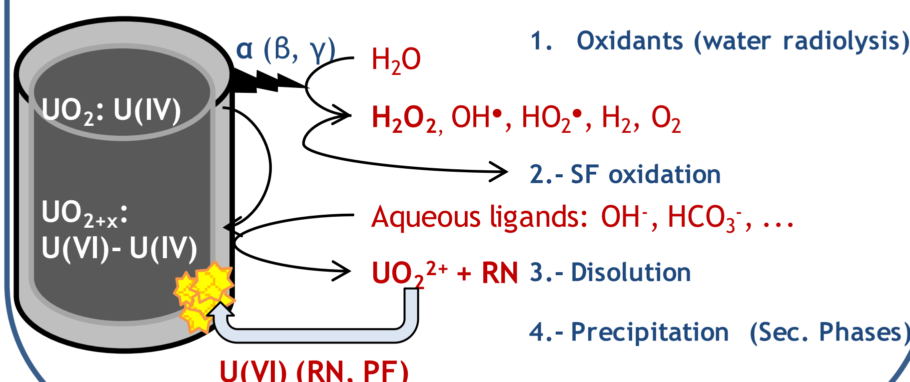




Degradation

~ 1000 y

GW-radiation interaction



Irradiated fuel in DGR

Long term behaviour

Environment conditions

pH
Eh: oxidizing/reducing
Temperature
Water: CO_3^{2-}/HCO_3^-, cations (Ca^{2+}, Si^{4+})
Gas: H_2, O_2, CO_2, ...

Irradiated fuel attributes

Pre-Oxidation(UO_{2+x}) or hydration (UO_3·xH_2O)
SE, grain size, swelling
Cladding: corrosion/hydride
PCI
Composition: oxides, metals, RN

Cement Groundwaters

- **Why?** ➔ Ubiquitous in engineering barriers
- **What?**➔ Highly-alkaline and chemically reducing environment
- Radionuclides retention / apparently lower dissolution rate of SF (vs neutral): minimise radionuclide solubility and provide sites for radionuclide sorption

U(IV) increases its redox sensitively at high pH (and relatively low Eh)*
- LWR-UO_2 SNF 50.4 GWd·$(tHM)^{-1}$ in YCW (H_2 overpressure, SHE=-0.74 V) ➔ [U] = $1.5 \cdot 10^{-9}$ M**
- YCW ➔ SCK-CEN***
  - pH = 13.5 (initial state of cement degradation)

* Ollila, K. (2008) POSIVA WR 2008-75
** Loida, A. et al. (2012) MRS 1475, 119 – 124
*** Mennecart, T. et al. (2012) NEA/RWM/R(2012)3/REV

2. Motivation

- Sample fabrication "*dry route*" & characterization

UO_2	UO_2 - Cr_2O_3	UO_2 - Cr_2O_3 - Al_2O_3	UO_2 - Gd_2O_3
	0.02 wt% *0.06 wt%*	*0.05 - 0.02 wt%*	*4.50 wt%*

- Matrix corrosion of modern LWR fuels under DGR relevant conditions for improving the predictive capability of SF corrosion models and reduction of associated uncertainties.
  - Element release & corrosion rates. Conditions: neutral-alkaline. Anoxic & reducing.
  - Characterization of the microstructural evolution upon corrosion by advanced micro-analytical tools.
  - Establishing a link between matrix corrosion behaviour of UO_2-based model systems and spent fuel.

3. Fabrication & characterization

Raw materials

↓

Pellet

↓

Characterization → **Surface morphology** / **Crystallinity** / **Densification**

samples

- UO_2 → crushed or purified (TBP)-ADU
- UO_2 - *0.02, 0.06*wt% Cr_2O_3
- UO_2 - *0.05%* Cr_2O_3-*0.02%* Al_2O_3
- UO_2 - *4.5*wt% Gd_2O_3

Techniques	Parameter	Equipment
BET	N_2-BET Surface Area	ASAP 2020 Micromeritics ISO 12800E
XRD	Crystalline structure	D8 ADVANCE Eco (Bruker) - ICD
E. Raman	Structural / speciation	HORIBA LabRam HR Evolution He-Ne 633 nm (red)
OM / SEM	Surface morphology/grain size Lineal intercept method (Heyn)	Hitachi SV6600 15 kV ASTM E 112-96 UNE-EN ISO 643:2012
Laser diffraction	Particle size	Malvern 2600 series
Archimedean imm.	Density	Satorious kit
Vickers Microhardness	Hv	Micro-Vickers hardness–MTR3 4-sided diamond pyramid

Glove box

Metal oxides (powder)

↓

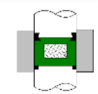
Mix blending

↓

Ball milling

Additives. (EBS $C_{38}H_{76}N_2O_2$ = 1%) - - →

↓



Pressing (uniaxial) 700 MPa

Green pellets

Rad. Lab

↓

Calcination (100, 300 & 500 °C) reducing

↓

Sintering (reducing atmosphere, 4 h, 5ºC/min, 1675 °C)

Sintered pellets

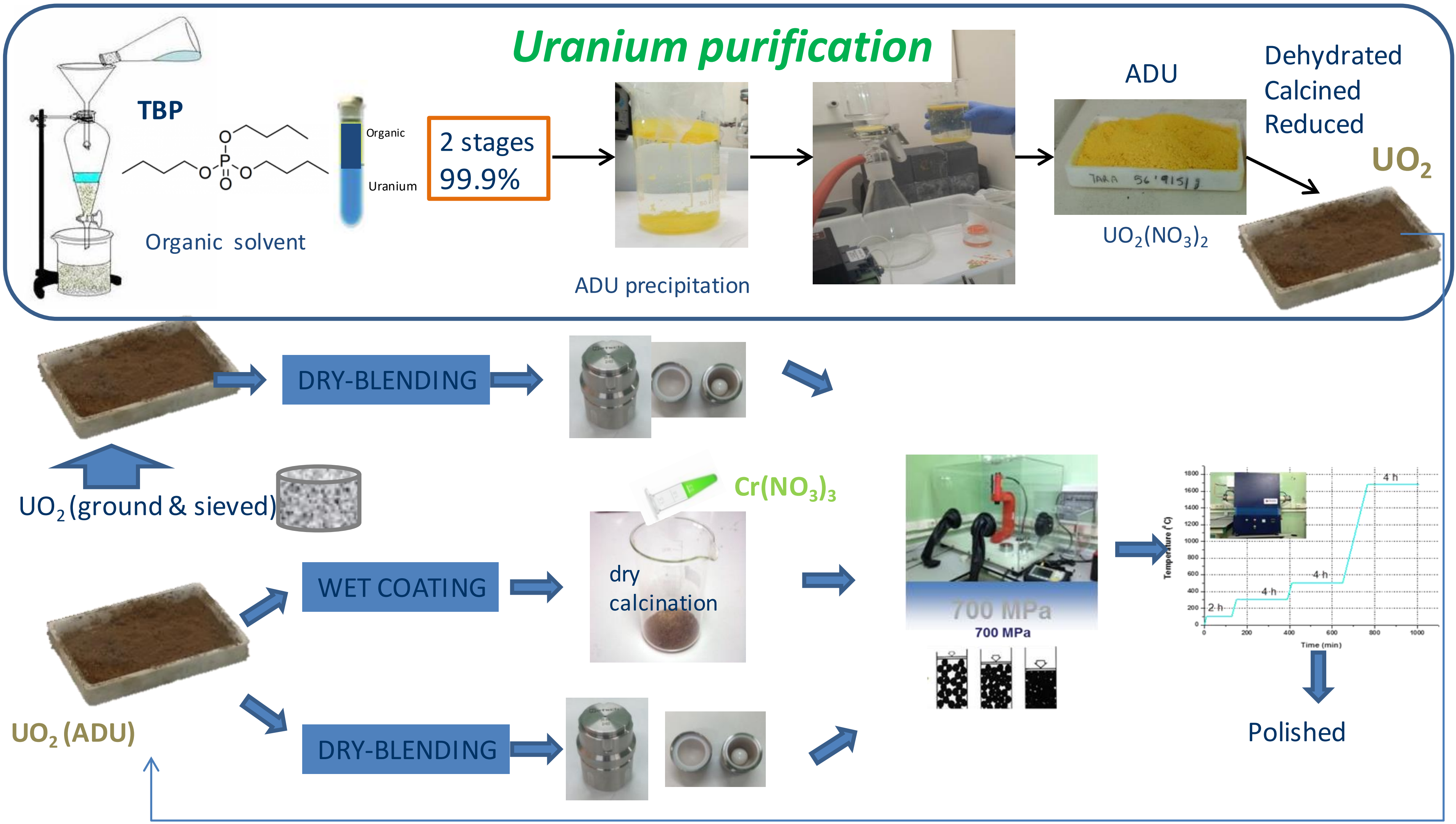
↓

Polishing

↓

Annealing (80-90% sinter.T °C, H2/N2)

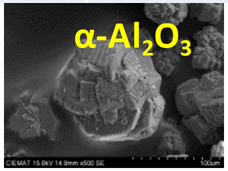
3. Fabrication & characterization

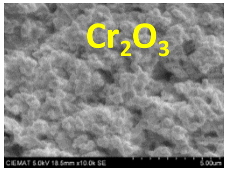


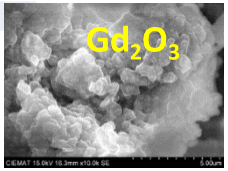
3. Fabrication & characterization

Raw

Initial powder	UO_2	UO_2 purified	α-Al_2O_3	α-Al_2O_3 calcined	Cr_2O_3	Gd_2O_3
Lattice param / nm	a = b = c = 0.546920(4)	a = b = c = 5.4712	a = 0.7944(6) b = 0.7980(4) c = 1.1722(6)	a = 0.7944(6) b = 0.7980(4) c = 1.1722(6)	a = b = 0.49586(7) c = 1.3592(2)	1.08141(4)
SSA (BET) /$m^2 \cdot g^{-1}$	0.95 ± 0.01	3.75 ± 0.04	35.26 ± 0.34	29.12± 0.25	3.47 ± 0.02	14.6 ± 0.01
Dens / $g \cdot cm^{-3}$	10.97 (TD)	10.97 (TD)	3.95	3.95	5.22	7.41
D(v, 0.5) / µm	**19.6**	**3.52**	5.49	48.61	2.09	11.8



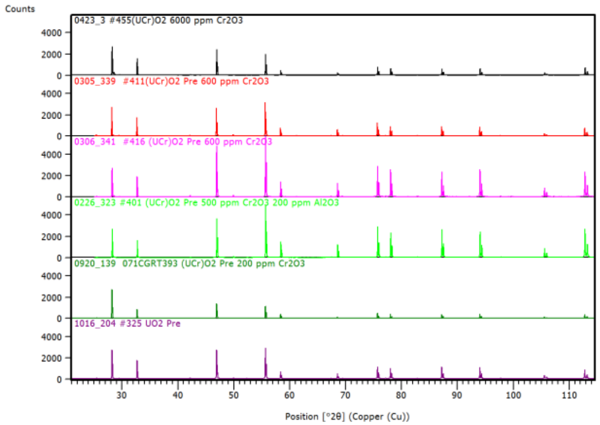


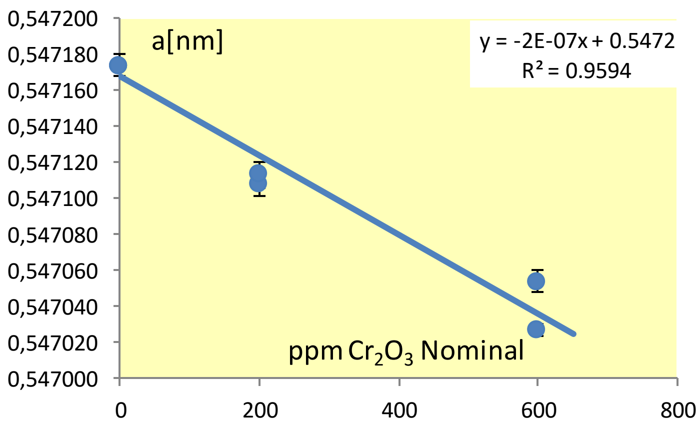


Pellet

Dry-blending (UO2 ground)	UO_2	UO_2-0.02Cr_2O_3	UO_2-0.06Cr_2O_3	UO_2-0.05Cr_2O_3-0.02Al_2O_3	UO_2-4.5Gd_2O_3
Lattice parameter / nm	a = b = c = 0.547174 (6)	0.547108 (7)	0.547113 (7)	0.547198 (5)	Phase#1 0.547 Phase#2 0.546 Phase#3 0.545
SSA (BET) /$m^2 \cdot g^{-1}$	0.36 ± 0.01	0.28 ± 0.01	0.63 ± 0.02	0.25 ± 0.01	0.6332 ± 0.01
Density / $g \cdot cm^{-3}$	8.93 ± 0.07	9.80 ± 0.12	9.83 ± 0.07	9.80 ± 0.43	9.81± 0.11

Homogeneous dissolution of dopants



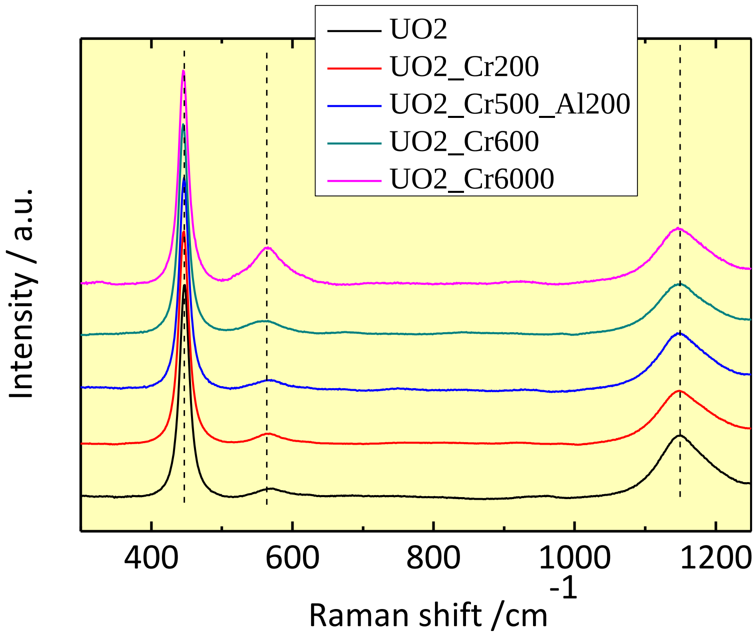


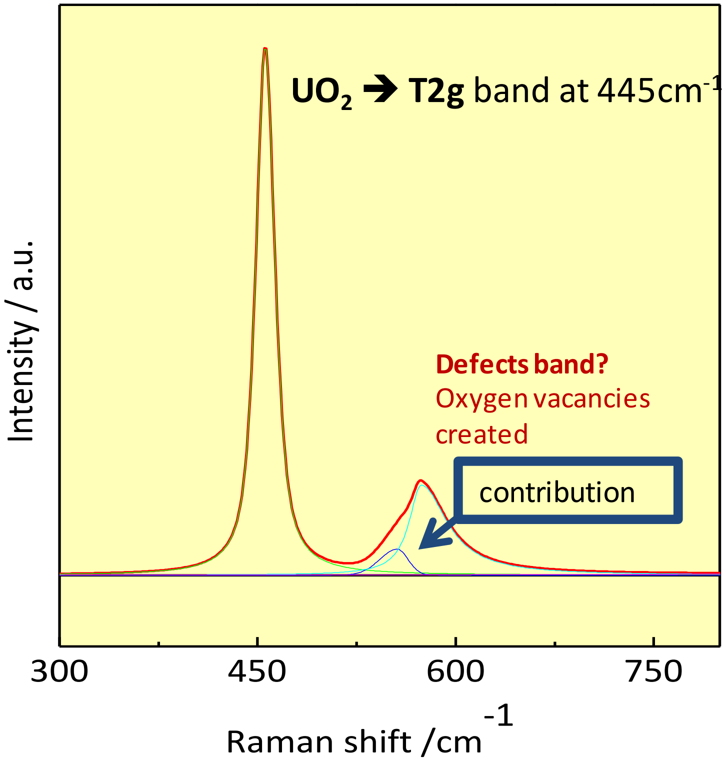
3. Fabrication & characterization: Pellet characterization. Raman

Effect of dopant (Cr)

Pellet

UO2 ground
Φ = 19.6 µm





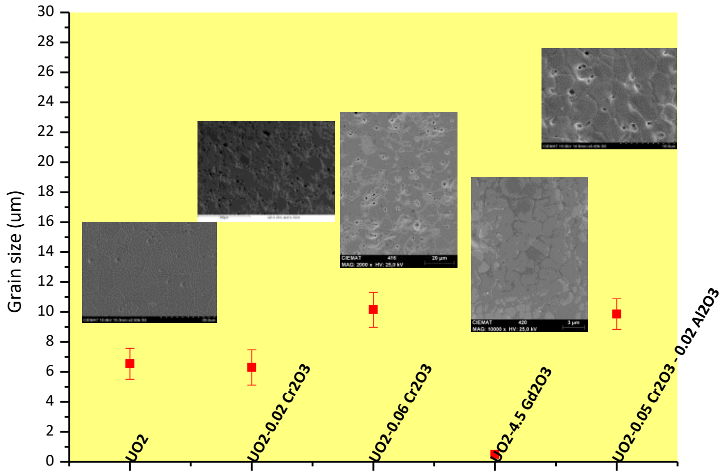
Increased [Cr] ➔ extra characteristic band (wide and asymmetric) LO ~540 cm^{-1} from 600 ppm.

Z. Talip, J. Am. Ceram. Soc.,2015,98[7], 2278-2285

3. Fabrication & characterization: Pellet characterization.

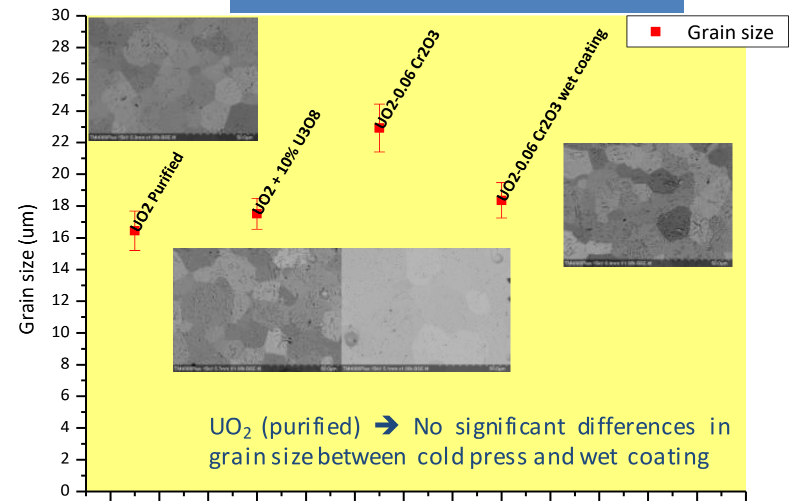
Grain size: linear intercept (LI) → SEM

UO_2 ground (Φ = 19.6 µm)

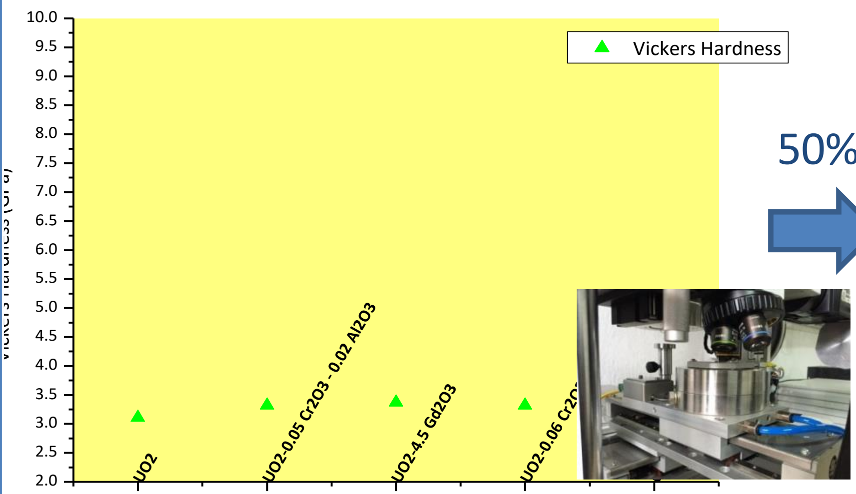


x 2

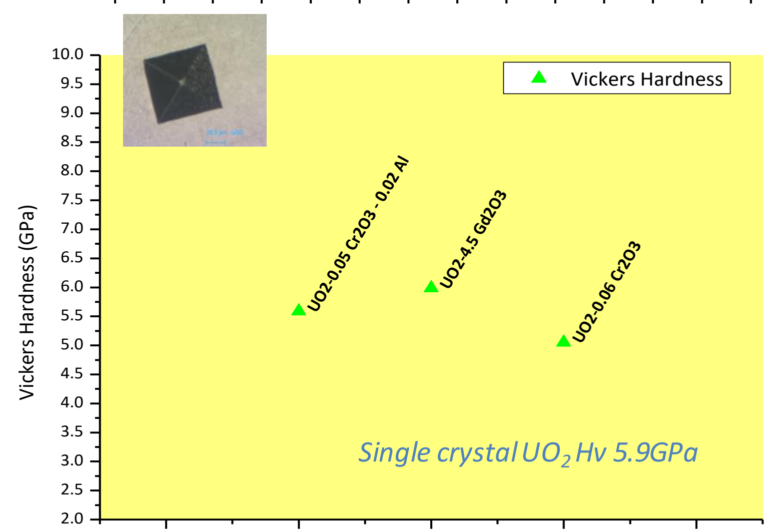
UO_2 purified (Φ = 3.52 µm)



Vickers Microhardness: Diagonal lengths of the Vickers indentation



50%



4. Dissolution tests with doped UO_2

Batch Autoclaves (7- 8 bar 4.7%H_2-N_2)

Materials

- UO_2 + 0.06% Cr_2O_3
- UO_2 + 0.05% Cr_2O_3 - 0.02% Al_2O_3
- UO_2 + 4.5% Gd_2O_3

Media - reducing (300 ml)

- ☑ PC ≡ Blank (0.02M $NaClO_4$) pH 7.2 ± 0.1
- ☑ BC ≡ Bicarbonate water (0.019M $NaHCO_3$- 0.001M NaCl) pH 8.9 ± 0.1
- ☒ YCW ≡ Cementitious water pH 13.2 ± 0.1

Static anoxic test- Batch (Ar atmosphere)

Materials

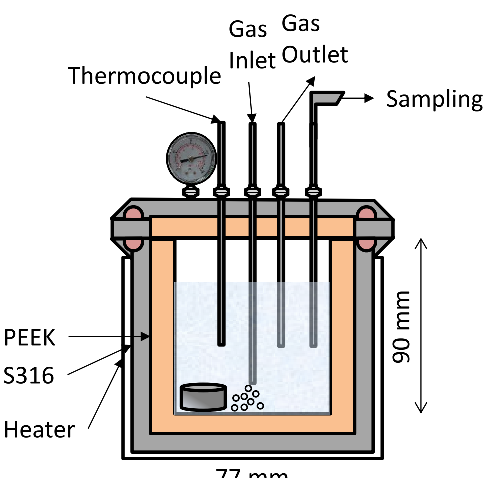
- UO_2 + 0.06% Cr_2O_3
- UO_2 + 0.05% Cr_2O_3 - 0.02% Al_2O_3

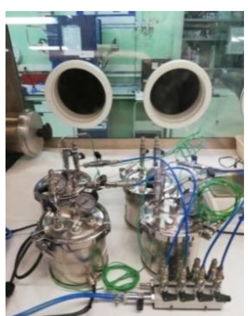
Media - anoxic (300 ml)

- YCW ≡ Cementitious water pH 13.2 ± 0.1

Experimental parameters monitoring

- [O_2] (glovebox <2ppm)
- Temperature ext.
- pH, Eh
- Concentration in solution by ICP-MS



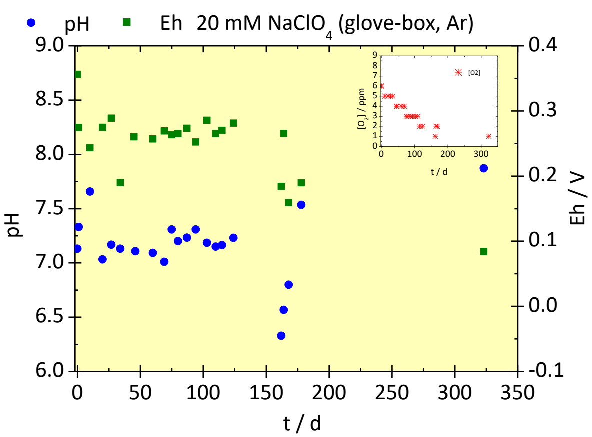


4. Dissolution tests with doped UO_2: Leachants

Monitoring: pH, Eh, [O_2], T, Element Conc. + LOD + LOQ

PC-1 $NaClO_4$ 0.02M

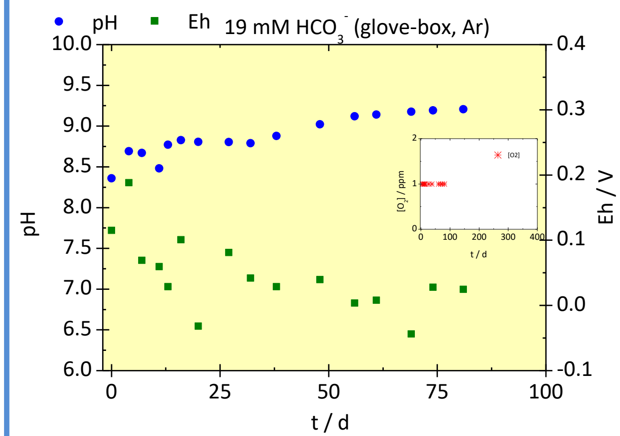
(I=0.02M; 22±2°C; pH = 7.2±0.1; Eh = 0.25±0.02 V)



Conc.	$NaClO_4$ 0.02 M
mol·l^{-1}	Conc
Na	$(2.18\pm0.03)\cdot10^{-2}$
Mg	$(2.07\pm0.02)\cdot10^{-6}$
Al	$(3.61\pm1)\cdot10^{-10}$
K	$(2.80\pm0.03)\cdot10^{-4}$
Ca	$(2.89\pm0.1)\cdot10^{-6}$
Cr	$(1.9\pm0.4)\cdot10^{-9}$
Mn	$(7.0\pm0.3)\cdot10^{-9}$
Fe	$(3.4\pm0.2)\cdot10^{-8}$
Co	$(3.9\pm0.5)\cdot10^{-10}$
Ni	$<1.7\cdot10^{-9}$
Cu	$(2.3\pm0.5)\cdot10^{-9}$
Cs	$(1.9\pm0.1)\cdot10^{-10}$
Ba	$(9.9\pm0.1)\cdot10^{-9}$
Ce	$(7.1\pm0.2)\cdot10^{-12}$
Gd	$(9\pm4)\cdot10^{-12}$
Pb	$(5.7\pm0.4)\cdot10^{-10}$
Th	$<4.3\cdot10^{-12}$
U	$(1.2\pm0.1)\cdot10^{-10}$
Cl^-	$1.02\cdot10^{-4}$
ClO4$^-$	$2.11\cdot10^{-2}$
Cond.	2 mS·cm^{-1}

BC-1 19 mM NaHCO3 - 1 mM NaCl

(I=0.02M; 23.4±0.5°C; pH = 8.9±0.1; Eh = 0.06±0.03 V)



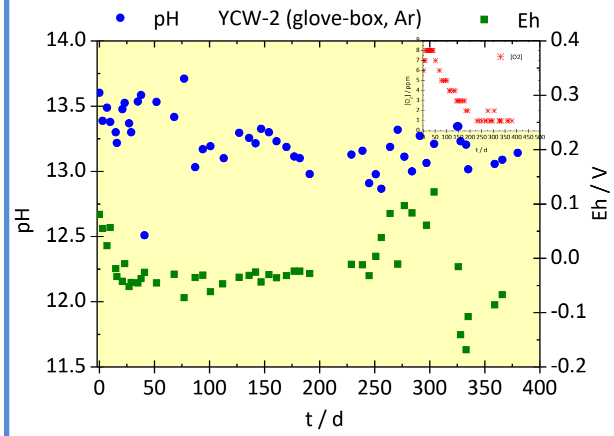
Conc.	BC-1
mol·l^{-1}	Conc
Na	$(2.10\pm0.02)\cdot10^{-2}$
Mg	$(1.48\pm0.06)\cdot10^{-6}$
Al	$(3.55\pm0.1)\cdot10^{-9}$
K	$(1.40\pm0.04)\cdot10^{-4}$
Ca	$(5.6\pm0.6)\cdot10^{-6}$
Cr	$(4\pm2)\cdot10^{-9}$
Mn	$(1.90\pm0.08)\cdot10^{-8}$
Fe	$(9\pm2)\cdot10^{-8}$
Co	$(1.43\pm0.09)\cdot10^{-9}$
Ni	$<1.7\cdot10^{-9}$
Cu	$(1.2\pm0.5)\cdot10^{-8}$
Cs	$(1.38\pm0.07)\cdot10^{-9}$
Ba	$(1.8\pm0.6)\cdot10^{-8}$
Ce	$(1\pm0.4)\cdot10^{-10}$
Gd	$(8\pm3)\cdot10^{-11}$
Pb	$(5.3\pm0.5)\cdot10^{-9}$
Th	$<4.3\cdot10^{-12}$
U	$(3.6\pm0.3)\cdot10^{-9}$
Cl^-	$1.07\cdot10^{-3}$
SO4^{2-}	$3.3\cdot10^{-5}$
Alcal.	19.4 mEq·l^{-1}
Cond.	1.64 mS·cm^{-1}

4. Dissolution tests with doped UO_2: Leachants

Monitoring: pH, Eh, $[O_2]$, T, Element Conc. + LOD + LOQ

YCW-2

(I>0.4M; 22.6±0.4°C; pH = 13.2±0.1; Eh= -0.02±0.02 V)



Conc.	YCW-2
$mol\cdot l^{-1}$	Conc
Na	$1.5\cdot10^{-1}$
Mg	$7\cdot10^{-7}$
Al	$3.3\cdot10^{-6}$
K	$3.8\cdot10^{-1}$
Ca	$4.5\cdot10^{-4}$
Cr	$<6\cdot10^{-7}$
Mn	ND
Fe	$2\cdot10^{-6}$
Co	ND
Ni	ND
Cu	ND
Cs	ND
Ba	ND
Ce	ND
Gd	$1.6\cdot10^{-9}$
Pb	ND
Th	ND
U	$2.8\cdot10^{-8}$
Cl^-	$3.9\cdot10^{-4}$
SO_4^{2-}	$2\cdot10^{-3}$
Alcal.	NM
Cond.	$85\ mS\cdot cm^{-1}$

ND = non detected
NM = not measured

4. Dissolution tests with doped UO_2: Batch Autoclaves (7- 8 bar 4.7% H_2-N_2) – 0.02M $NaClO_4$

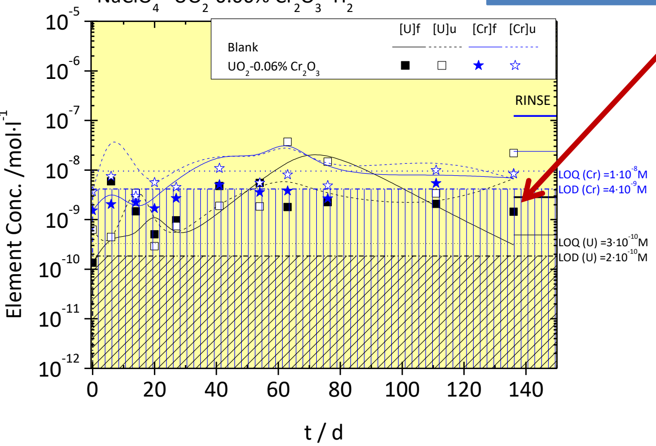
UO_2-0.06% Cr_2O_3	UO_2- 0.05% Cr_2O_3 - 0.02% Al_2O_3	UO_2-4.5% Gd_2O_3

[U] = 10^{-9} M

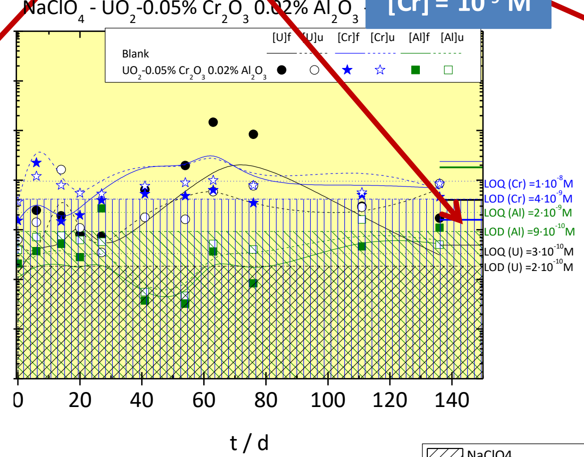
Results indicates similar trends towards dopant.

No element release detected so far / no dissolution

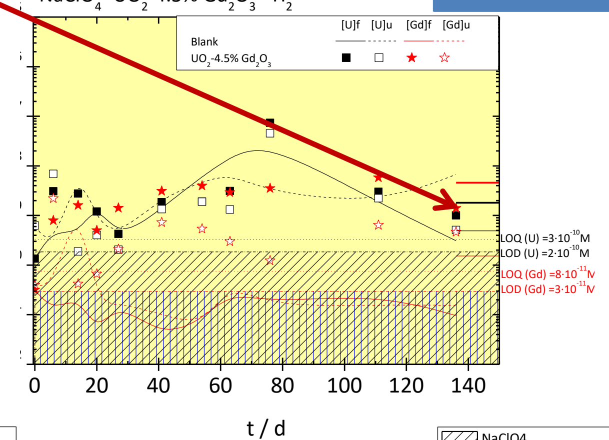
[Cr] = 10^{-9} M

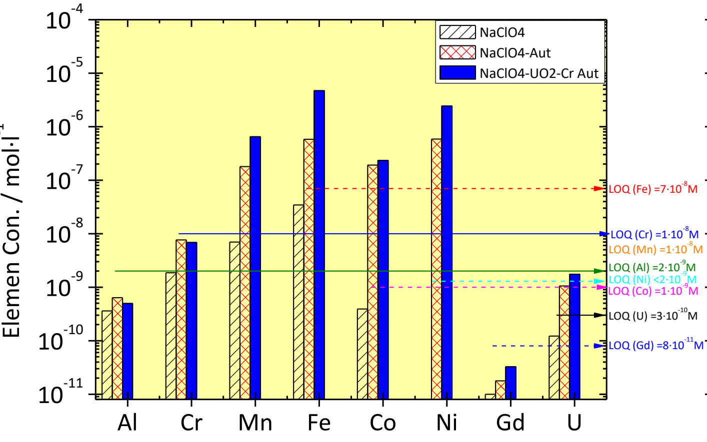


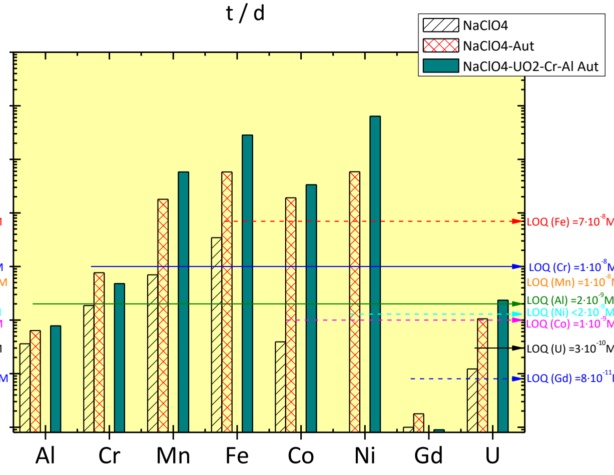
[Cr] = 10^{-9} M

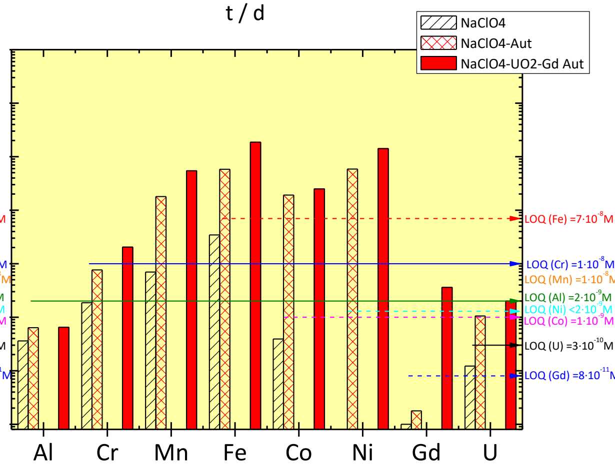


[Gd] = 10^{-9} M





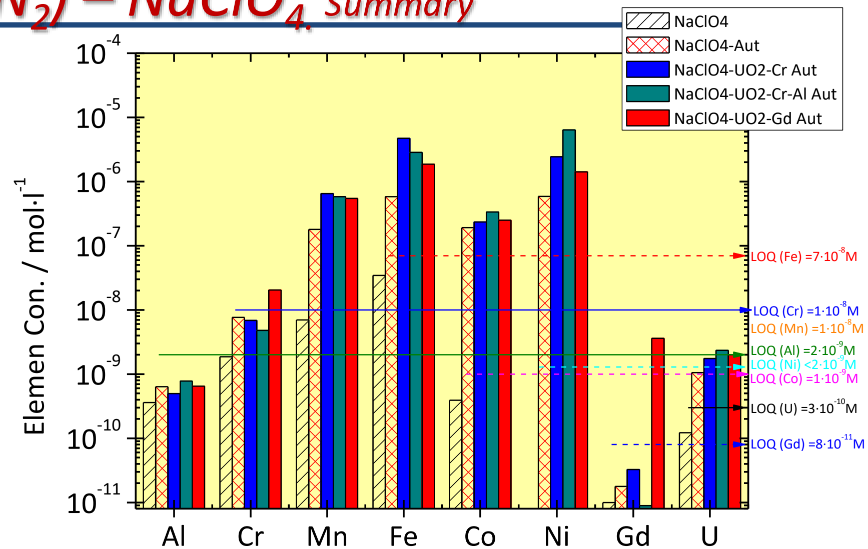




4. Dissolution tests with doped UO_2: Batch Autoclaves (7- 8 bar 4.7%H_2-N_2) – $NaClO_4$ Summary

- **<u>UO2-doped + $NaClO_4$ 0.02M + H_2</u>**
  - $P(H_2)$ = 0.37 atm; Eh = -0.02V; pH = 6.8
  - Δ(Sgeom/V) = 0.6 ➔ 0.7 m^{-1}

Indistinguishable **U concentrations** than those found in the leachant



PC	[U]f	[U]u	[Cr]f	[Cr]u	[Al]f	[Al]u	[Gd]f	[Gd]u	[Fe]f	[Fe]u
Blank	$(1\pm1)\cdot10^{-9}$	$(4\pm5)\cdot10^{-9}$	$(8\pm1)\cdot10^{-9}$	$(1\pm1)\cdot10^{-8}$	$(6\pm2)\cdot10^{-10}$	$(8\pm3)\cdot10^{-10}$	$(2\pm2)\cdot10^{-11}$	$(1.57\pm0.03)\cdot10^{-11}$	$(6\pm3)\cdot10^{-7}$	$(10\pm8)\cdot10^{-7}$
UO_2-0.06%Cr_2O_3	$(1.7\pm0.6)\cdot10^{-9}$	$(1\pm2)\cdot10^{-8}$	$(7\pm3)\cdot10^{-9}$	$(9\pm1)\cdot10^{-9}$	$(4.9\pm0.9)\cdot10^{-10}$	$(8\pm5)\cdot10^{-10}$	$(3\pm8)\cdot10^{-11}$	$(5\pm3)\cdot10^{-11}$	$(4.7\pm0.4)\cdot10^{-6}$	$(5\pm1)\cdot10^{-6}$
UO_2-4.5%Gd_2O_3	$(2\pm2)\cdot10^{-9}$	$(1\pm2)\cdot10^{-9}$					$(4\pm4)\cdot10^{-9}$	$(6\pm2)\cdot10^{-10}$	$(1.9\pm0.8)\cdot10^{-6}$	$(1.5\pm0.5)\cdot10^{-6}$
UO_2-0.05%Cr_2O_3-0.02%Al_2O_3	$(2\pm1)\cdot10^{-9}$	$(6\pm5)\cdot10^{-9}$	$(4.8\pm0.4)\cdot10^{-9}$	$(7\pm3)\cdot10^{-9}$	$(8\pm6)\cdot10^{-10}$	$(1\pm1)\cdot10^{-9}$	$(0.9\pm2)\cdot10^{-11}$	$(3\pm4)\cdot10^{-11}$	$(3\pm2)\cdot10^{-6}$	$(3\pm2)\cdot10^{-6}$

Literature

Carbol et al.➔ UO_2-10%^{233}U + 16, 1.6 y 0.16 bar H_2 + (0.01M NaCl - 2mM HCO_3^-) + pH = 8.4 ± 0.2*
Eh = -0.35 V➔ [U] ~ 10^{-12} - 10^{-11} M

LWR-UO_2 SNF 50.4 GWd·$(tHM)^{-1}$ in YCW (H_2 overpressure) **
Eh = -0.74 V➔ [U] = $1.5\cdot10^{-9}$ M

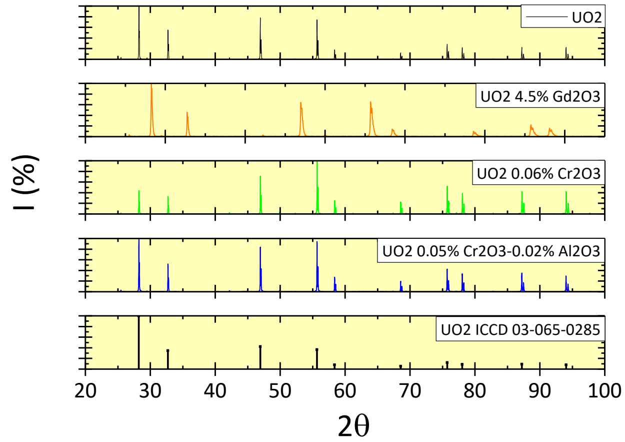
PWR 43 MWd/kg U + 4.9 atm H_2-0.03%CO_2 + (0.01M NaCl + 2mM $NaHCO_3$) + 70ºC ***
Eh = ??? ➔ [U] = 10^{-9} M

* Carbol et al. SKB TR-05-09 (2005)
** Loida, A. et al. (2012) MRS 1475, 119 – 124
*** Spahiu et al. (2004) Radiochimica Acta

4. Dissolution tests with doped UO_2: Batch Autoclaves (7- 8 bar 4.7%H_2-N_2) – $NaClO_4$. Pre and post characterization

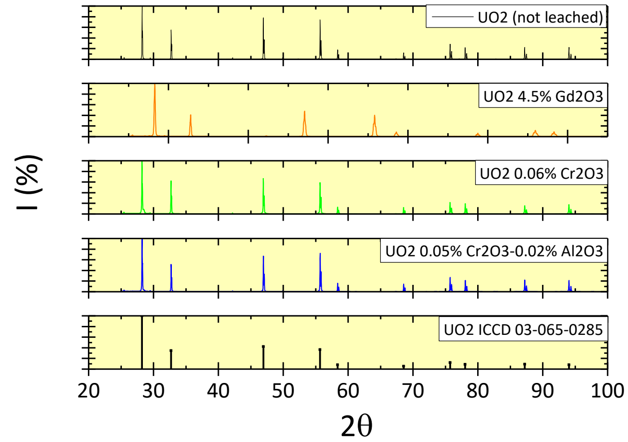
PRE-LEACHING

XRD (pre-leaching)



POST-LEACHING

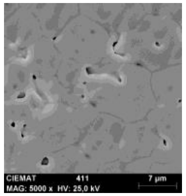
XRD (post-leaching)

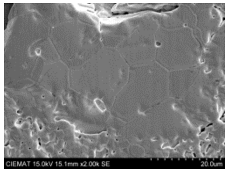


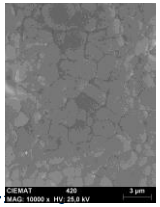
UO_2 0.06% Cr_2O_3

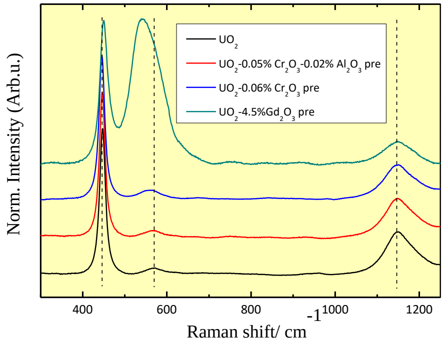
UO_2 0.05%Cr_2O_3 0.02%Al_2O_3

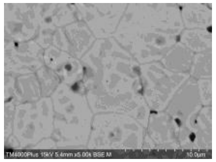
UO_2 4.5%Gd_2O_3

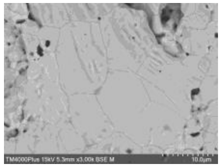


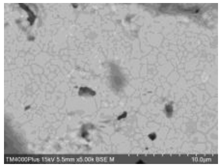


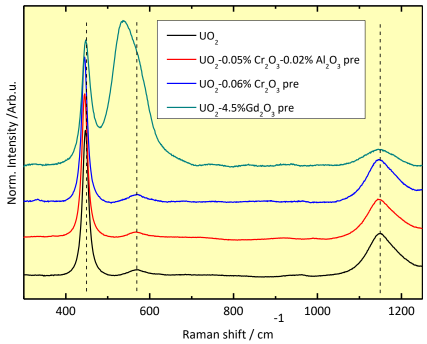






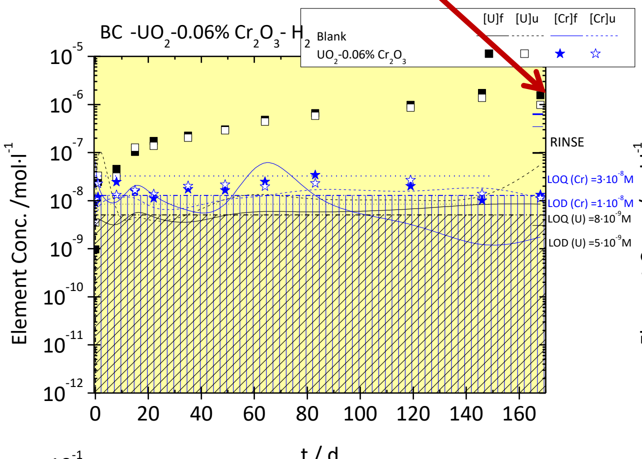


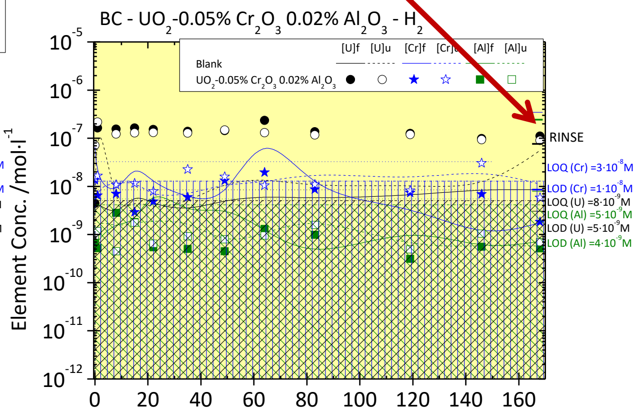


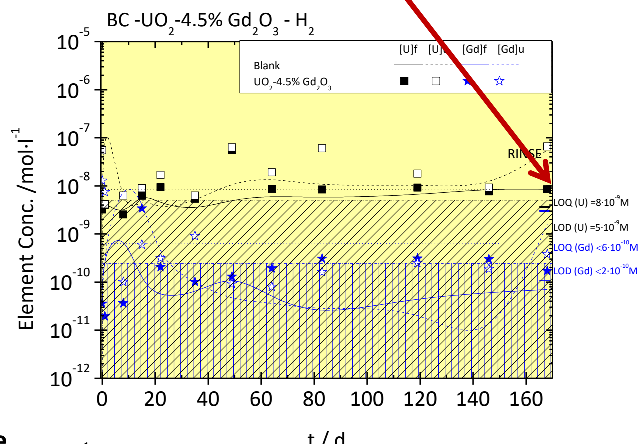


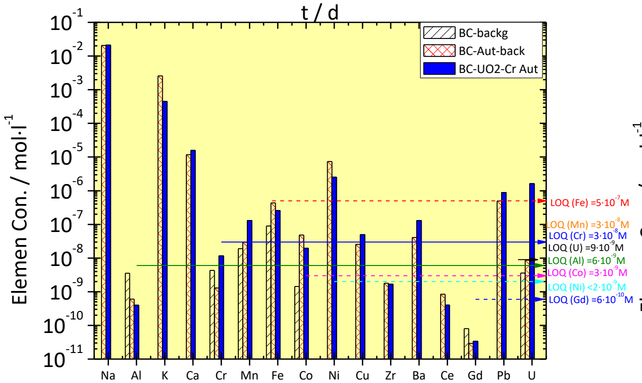
4. Dissolution tests with doped UO_2: Batch Autoclaves (7- 8 bar 4.7%H_2-N_2) – BC (Pt wire)

UO_2-0.06%Cr_2O_3	UO_2- 0.05% Cr_2O_3 - 0.02%Al_2O_3	UO_2-4.5%Gd_2O_3
[U] = 10^{-6} M	[U] = 10^{-7} M	[U] ≈ 10^{-8} M
[Cr] = 10^{-8} M	[Cr] = 10^{-8} M	[Gd] = 10^{-10} M

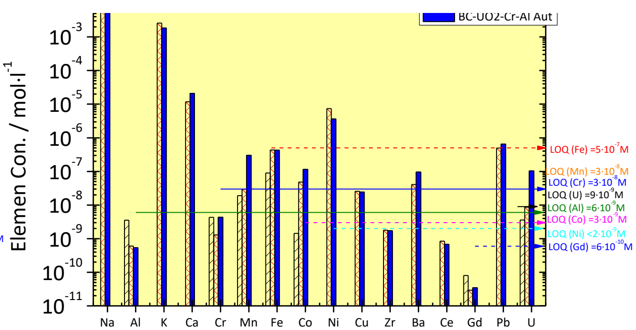


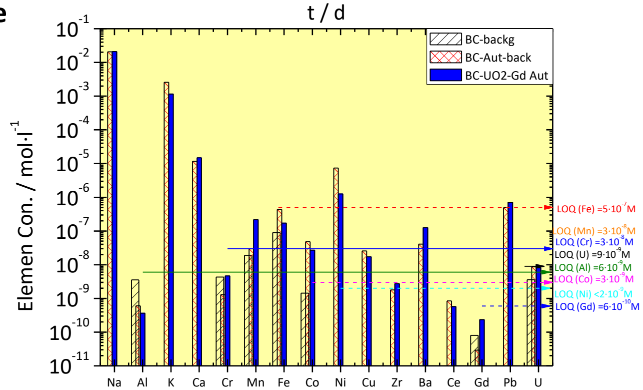






Increased element release compared to Perchlorate
➔ Bicarbonate effect

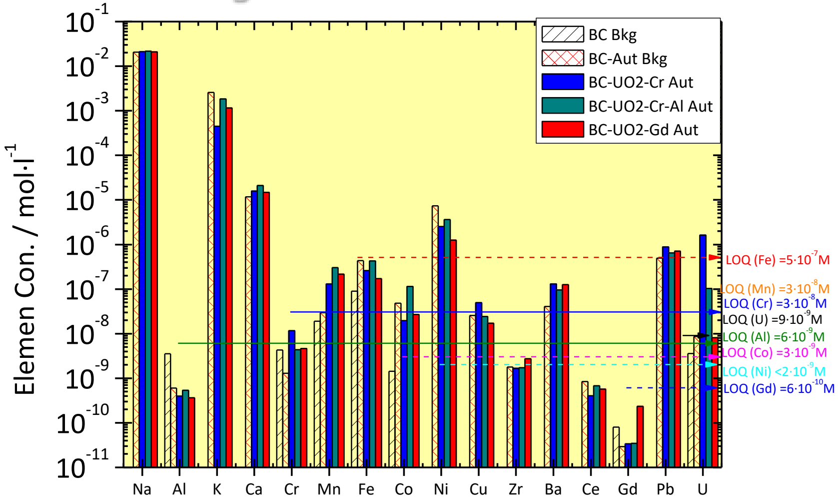




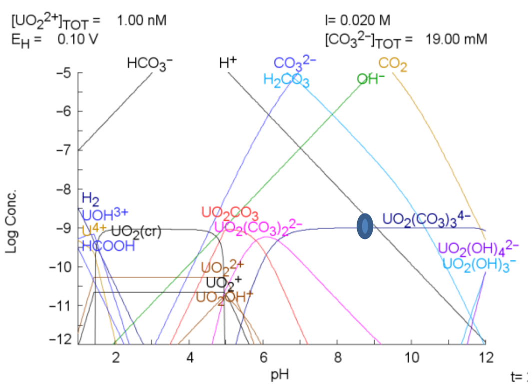
4. Dissolution tests with doped UO_2: Batch Autoclaves (7- 8 bar 4.7%H_2-N_2) – $NaHCO_3$ 0.019M

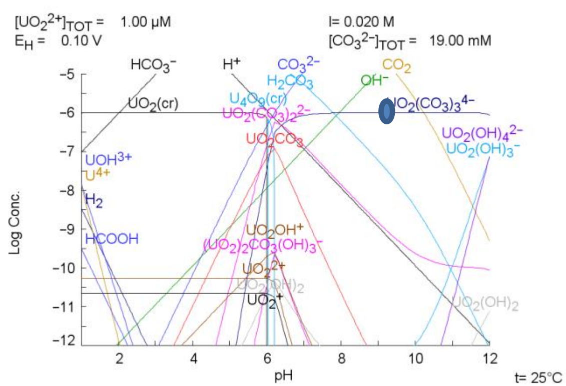
- ***UO2-doped + $NaHCO_3$ 0.02M + H_2***
  - $P(H_2)$ = 0.37 atm; Eh = 0.10±0.03V; pH = 9.23±0.04
  - Δ(Sgeom/V) = 0.6 ➔ 0.7 m^{-1}

Differences in **U concentrations** with doped UO_2 ➔ UO_2-Cr and UO_2-Cr/Al



BC	[U]f	[U]u	[Cr]f	[Cr]u	[Al]f	[Al]u	[Gd]f	[Gd]u	[Fe]f	[Fe]u
Blank	$(8.6\pm0.3)\cdot10^{-9}$	$(3\pm5)\cdot10^{-8}$	$(1.3\pm0.9)\cdot10^{-9}$	$(2\pm1)\cdot10^{-8}$	$(6\pm2)\cdot10^{-10}$	$(5.9\pm0.2)\cdot10^{-10}$	$(3\pm8)\cdot10^{-11}$	$(0.08\pm2)\cdot10^{-9}$	$(4\pm6)\cdot10^{-7}$	$(9\pm3)\cdot10^{-7}$
UO_2-0.06%Cr_2O_3	$(2\pm0.2)\cdot10^{-6}$	$(1\pm0.4)\cdot10^{-6}$	$(1\pm0.3)\cdot10^{-8}$	$(1\pm0.3)\cdot10^{-8}$	$(4\pm2)\cdot10^{-10}$	$(5\pm2)\cdot10^{-10}$	$(3\pm8)\cdot10^{-11}$	$(5\pm4)\cdot10^{-11}$	$(3\pm3)\cdot10^{-7}$	$(3\pm2)\cdot10^{-7}$
UO_2-4.5%Gd_2O_3	$(8\pm0.8)\cdot10^{-9}$	$(4\pm6)\cdot10^{-8}$					$(2\pm1)\cdot10^{-10}$	$(3\pm2)\cdot10^{-10}$	$(2\pm1)\cdot10^{-7}$	$(7\pm5)\cdot10^{-7}$
UO_2-0.05%Cr_2O_3-0.02%Al_2O_3	$(1.0\pm0.1)\cdot10^{-7}$	$(9\pm0.3)\cdot10^{-8}$	$(4\pm0.5)\cdot10^{-9}$	$(2\pm2)\cdot10^{-8}$	$(5.4\pm0.4)\cdot10^{-10}$	$(9\pm3)\cdot10^{-10}$	$(3\pm4)\cdot10^{-11}$	$(0.7\pm1.2)\cdot10^{-10}$	$(4\pm1)\cdot10^{-7}$	$(2\pm2)\cdot10^{-6}$





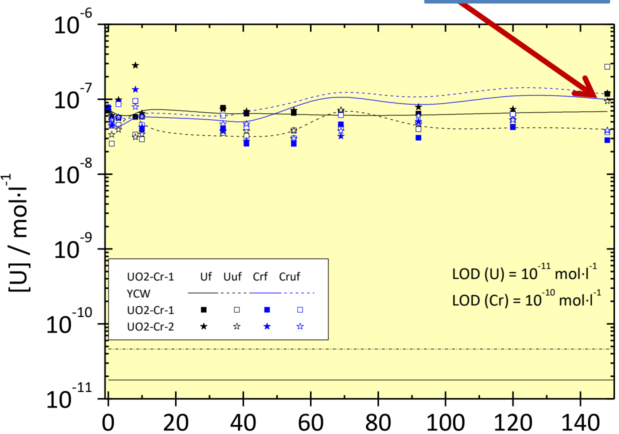
Post-leached samples are awaiting surface analysis

4. Dissolution tests with doped UO_2:
Static anoxic test-Batch (Ar atmosphere) - YCW

UO_2-0.06%Cr_2O_3

[U] ≈ 10^{-7} M

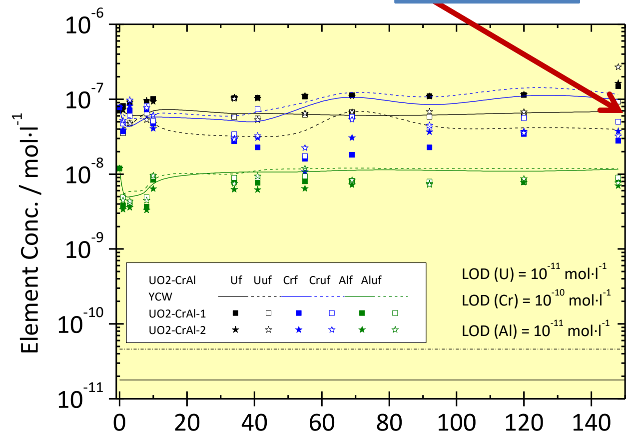
[Cr] = 10^{-8} M



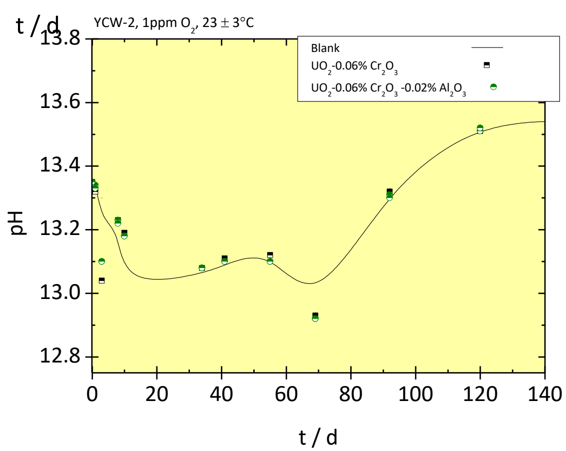
UO_2- 0.05% Cr_2O_3 - 0.02%Al2O3

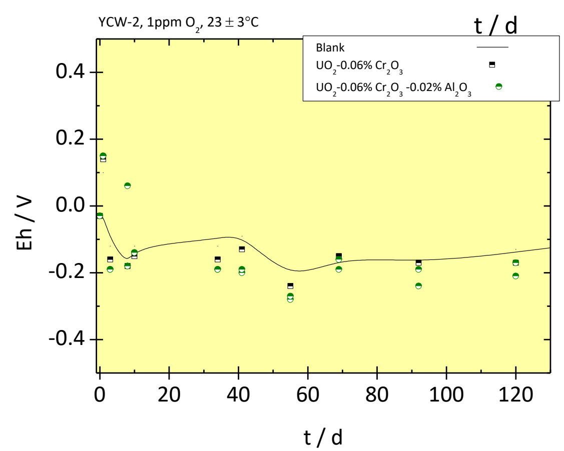
[U] = 10^{-7} M

[Cr] = 10^{-8} M



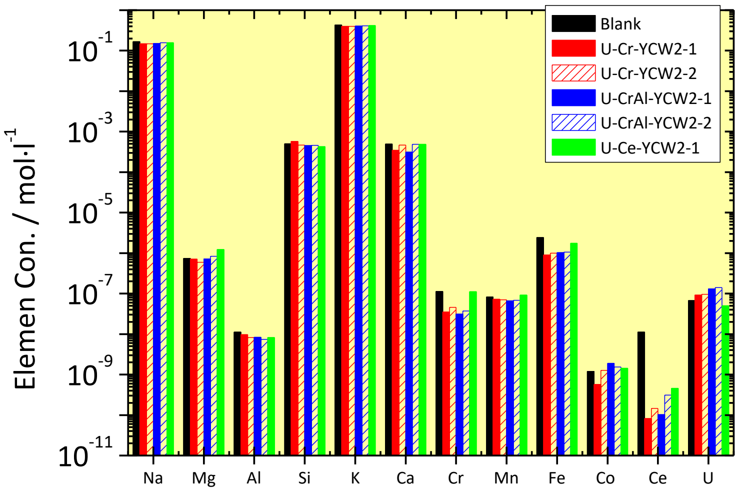
Detailed analysis: Identical **U concentrations** than the leachant





4. Dissolution tests with doped UO_2: Static anoxic test-Batch (Ar atmosphere) - YCW

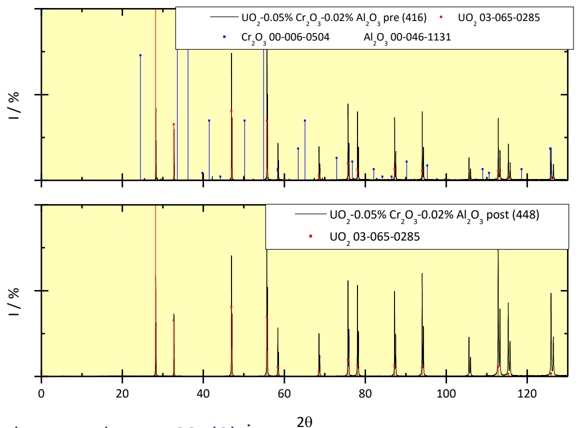
- ***UO2-doped + YCW-2 + Anoxic***
  - $P(H_2)$ = - atm; Eh = -0.17±0.03V; pH = 13.32±0.04
  - Δ(Sgeom/V) = 0.6 ➔ 0.7 m^{-1}



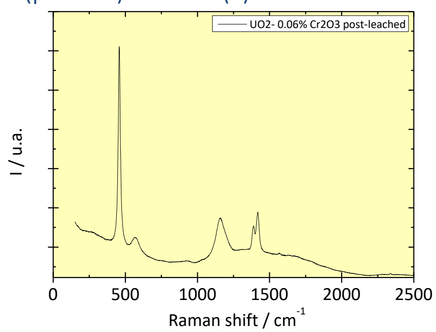
BC	[U]f	[U]u	[Cr]f	[Cr]u	[Al]f	[Al]u	[Fe]f	[Fe]u	[Ce]f	[Ce]u
Blank	6.39E-08	5.46E-08	1.05E-07	1.30E-07	1.13E-08	1.20E-08	1.75E-06	1.97E-06	1.40E-08	1.19E-09
UO_2-0.06%Cr_2O_3	8.20E-08	8.40E-08	4.00E-08	4.97E-08			1.02E-06	1.01E-06		
UO_2-0.05%Cr_2O_3-0.02%Al_2O_3	1.23E-07	9.40E-08	3.05E-08	4.66E-08	7.73E-09	8.07E-09	1.04E-06	1.14E-06		

4. Dissolution tests with doped UO_2: Static anoxic test- Batch (Ar atmosphere)

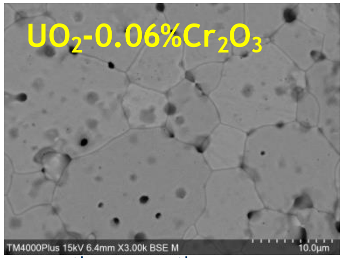
UO_2-0.06%Cr_2O_3



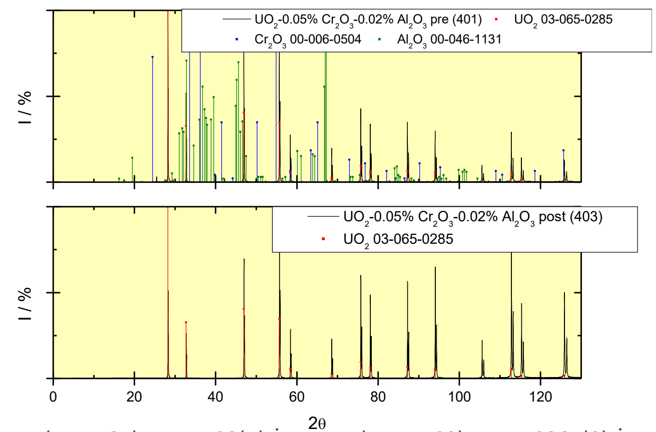
a (pre–411) = 5.47027(3) Å



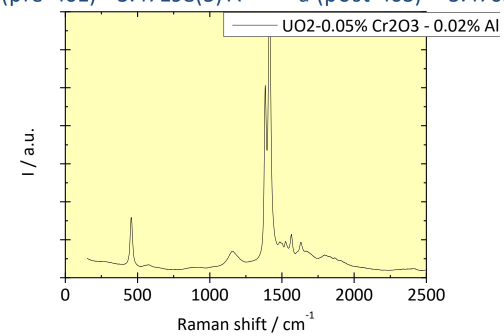
No secondary phases



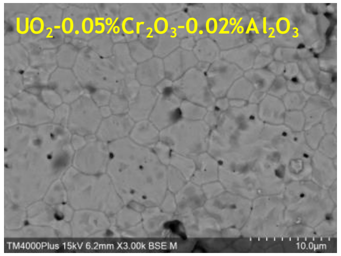
UO_2-0.05% Cr_2O_3-0.02%Al_2O_3



a (pre-401) = 5.47198(5) Å a (post-403) = 5.470961(9) Å

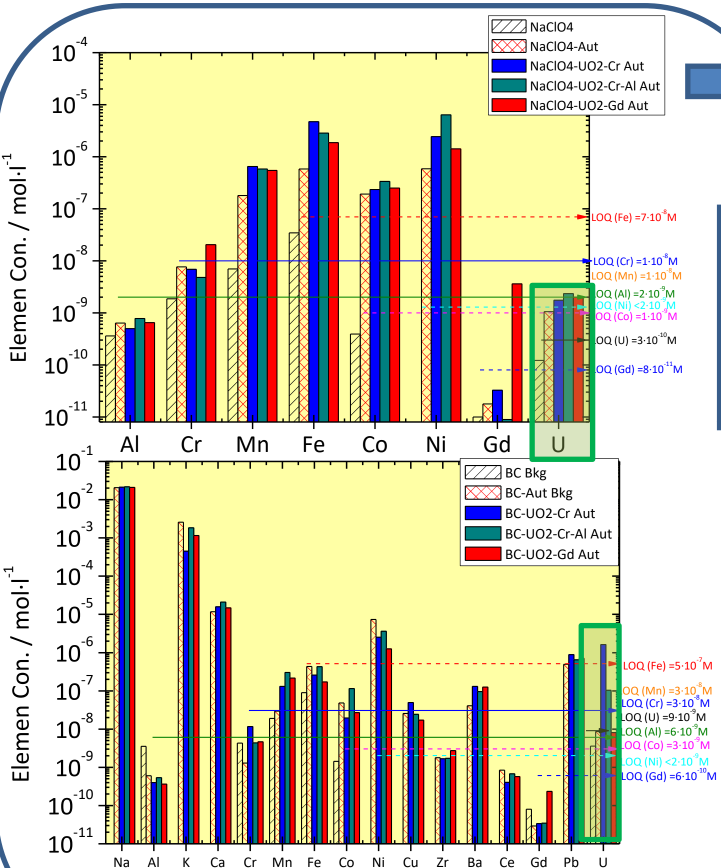


No secondary phases

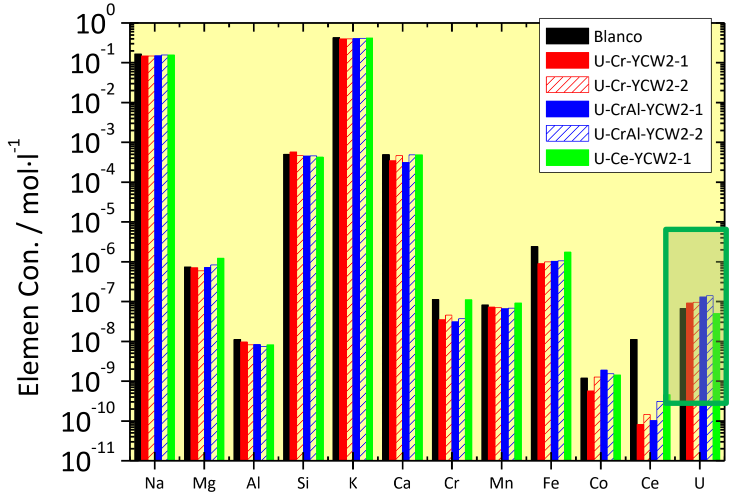


5. Conclusions

- Starting UO2 ➔ ground and <u>purified :</u> Starting with purified powders (ADU) provides improvements in grain size and microhardness, due to relieve the residual stresses (“*memory effect*”)
- Trivalent dopant are embedded in fluorite UO_2 crystalline structure and no significant differences between cold press and wet coating



- H_2(= 0.37 atm) with doped UO_2 samples in
  - in $NaClO_4$ 0.02M similar U concentration (10^{-9}M) = Background
  - in HCO_3- 19mM, 1-3 orders of magnitude higher in U conc.
- Anoxic Static leaching in YCW ➔ ≈ 10^{-7}M of U released = Background
- No secondary phases



- *Thanks to SCK-CEN (T. Mennecart) for his help in YCW preparation*
- *Thanks to all partners involved in High Level Waste Unit - CIEMAT*

Fabrication: Fernández, S.,
Purification (PUREX): Nuñez, A.
Raman spectroscopy: Bonales, L.J., Milena-Pérez, A.
XRD: Nebot, L.,
Operative work, glove box and ICP-MS: Durán, S., Anta, L., Serrano, L.

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