Production of powder targets for neutron-induced cross section measurements

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This presentation

- Introduction
- Press equipment and method
- Press parameters and material
- Characterization
- Results
- Summary

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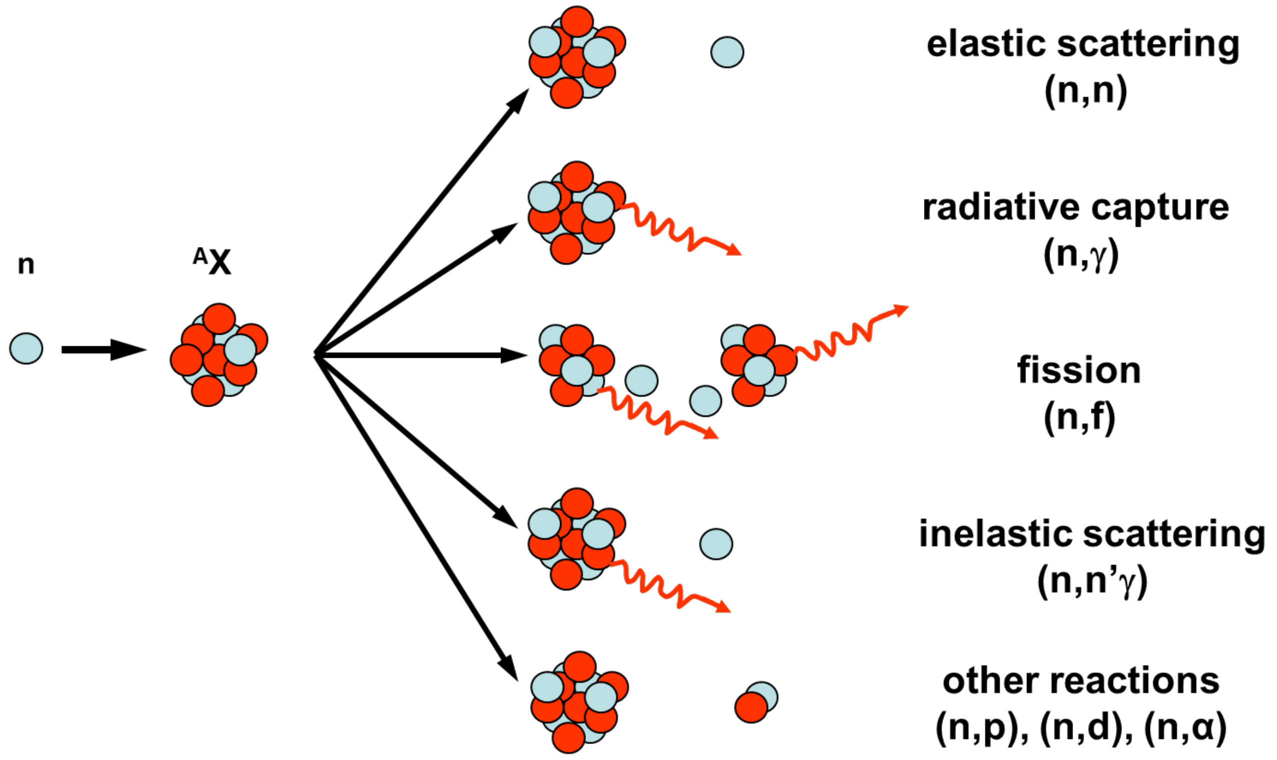
Introduction

The JRC is the largest provider of neutron data in the European Union. The data result from measurements at its two large-scale particle accelerators, and is used for nuclear safety and security in nuclear energy and for nuclear science applications.



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Targets for neutron cross-section measurements

Thin layers on thin substrates

- by molecular plating (U, Pu, Np, Am in nuclear controlled area)
- by physical vapour deposition (^{235}U and ^{238}U in nuclear controlled area; ^{6}LiF, ^{10}B, $C_{57}H_{110}O_6$, metallic Li deposits outside nuclear controlled area)

Samples with a wide range of thicknesses

- by rolling and punching metal discs (in- and outside nuclear controlled area)
- by pressing powders (in- and outside nuclear controlled area)
- by dissolving and diluting solutions (in- and outside nuclear controlled area)

This presentation

Preparation of

- ^{94}Mo, ^{95}Mo, ^{96}Mo targets for capture and transmission measurements to determine the neutron-induced cross-sections of ^{96}Mo
- ^{239}Pu target to measure ^{239}Pu(n,xn γ) cross sections
- Mass: 2 g
- Thickness: maximum 1 mm
- Mechanical stable
- Good strength
- Self-supporting

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This presentation

Available material: ^{94}Mo, ^{95}Mo, ^{96}Mo and ^{239}Pu **only as a powder**

Preparation method: uniaxial pressing

To avoid addition of impurities and enable simple recovery of limited and valuable material

- no adding fluids (e.g. solvent)
- no additives (e.g. binder)
- no lubricants
- no sintering

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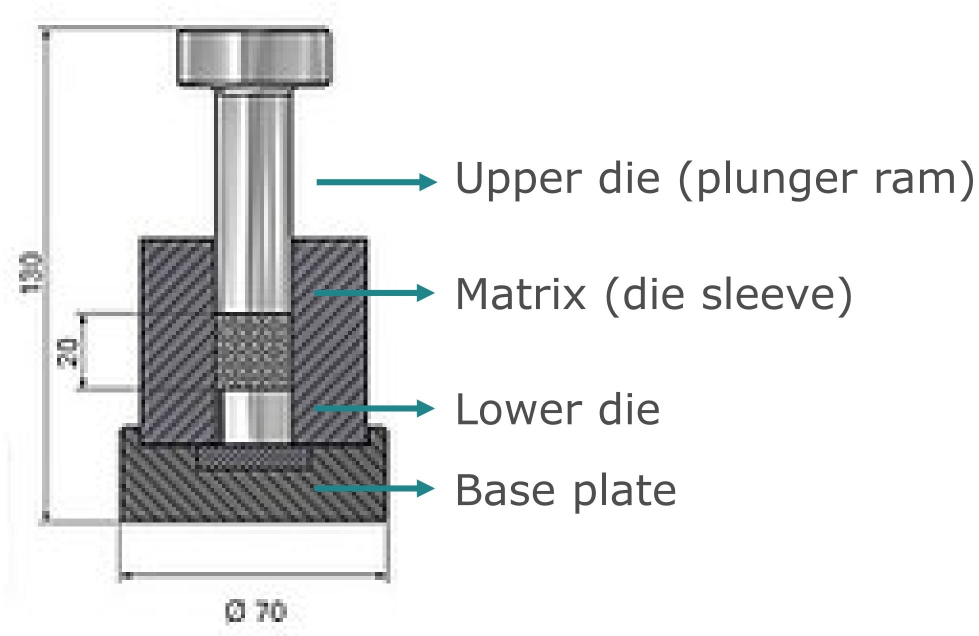
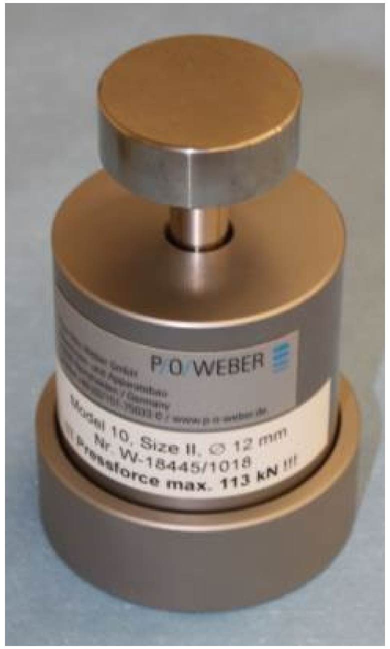
Press equipment





Ejector ring

Press tool, made of hardened steel and the corresponding lower and upper die.

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Press equipment

- Hydraulic lab press
- External manual hand pump
- Press load display

Specac press
15-t pressure load

Retsch press
25-t pressure load

P/O/Weber manual press, Model PW 30-BOX, enclosed in a glove box

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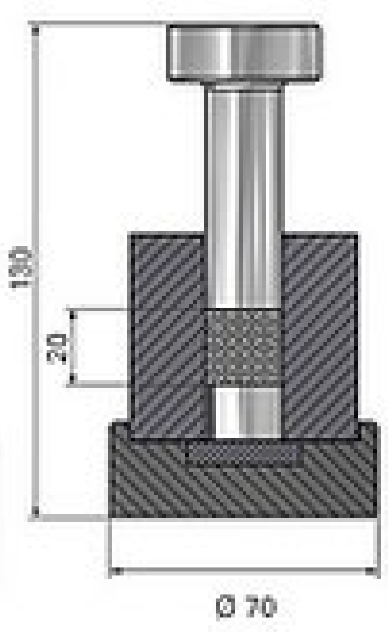
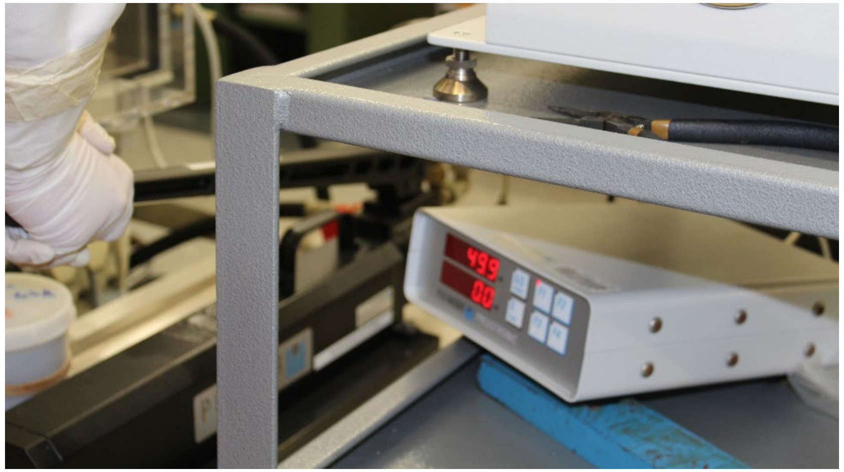
Uniaxial pressing

1. Die filling
2. Compaction
3. Ejection





André Moens turning the spindle down of the P/O/Weber manual 2-Column Laboratory Press, Model PW 30-BOX in a glove box and outside the glove box pumping the oil to raise the piston and reaching the designated press force shown on the gauge.

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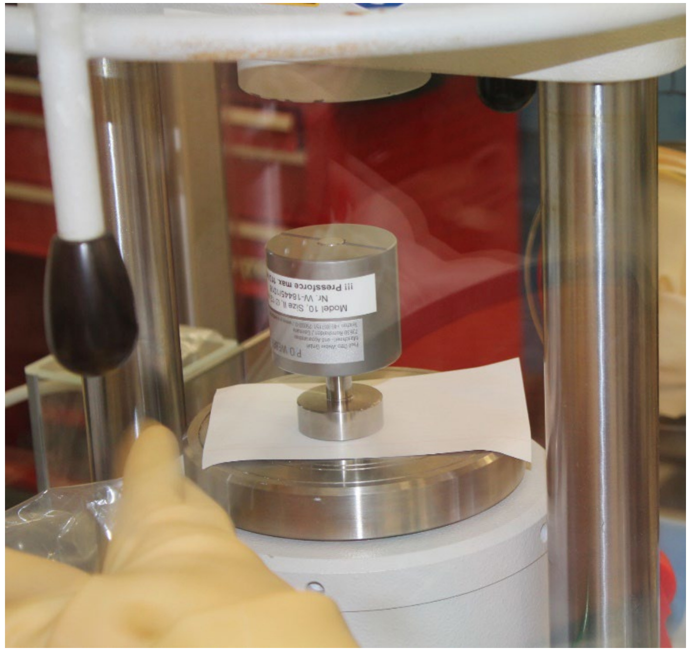
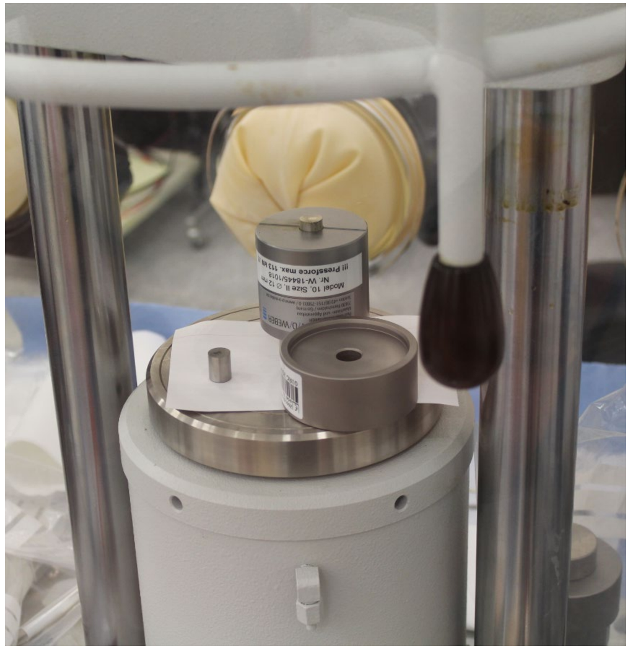
Uniaxial pressing

Release of the pellet with the ejector ring on top of the upside-down press tool.





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Simulation tests for Mo

Molybdenum

- **Molybdenum** pellet
  - Mass 2 g
  - Thickness as low as possible (max 1 mm)
  - Mechanical stable, good strength

Die diameter	Pressure Force	Powder	Powder mass	Pellet mass	Pellet thickness	Effective density	Strength
mm	kN		g	g	mm	%TD	
30	350-380	^{nat}Mo (<5 µm)	2.00		0.6		not good
30	300-390	^{nat}Mo (350 µm)	2.00		0.5		not good
20	160	^{nat}Mo (<5 µm)	2.02	2.00	0.9	70	good
20	160	^{nat}Mo (350 µm)	1.99	1.99	0.9	69	good

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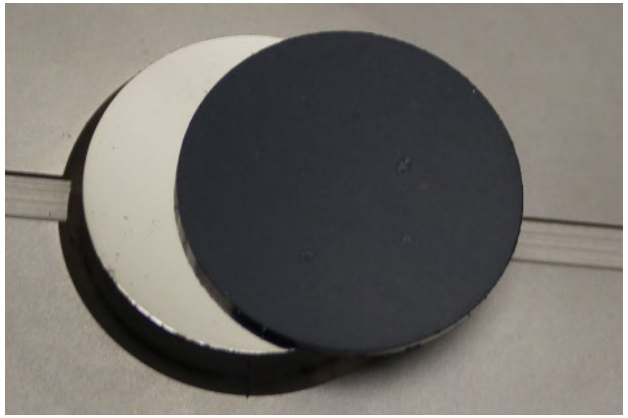
^{94}Mo, ^{95}Mo, ^{96}Mo pellets

Die diameter	Pressure Force	Powder	Powder mass	Pellet mass	Pellet thickness	Effective density	Strength
mm	kN		g	g	mm	%TD	
20	160	^{94}Mo	1.95	1.95	1.0	59	good
20	160	^{95}Mo	1.98	1.98	1.1	54	good
20	160	^{96}Mo	1.92	1.92	1.1	56	good



2 g ^{95}Mo
Diameter 20 mm
Thickness 1.1 mm

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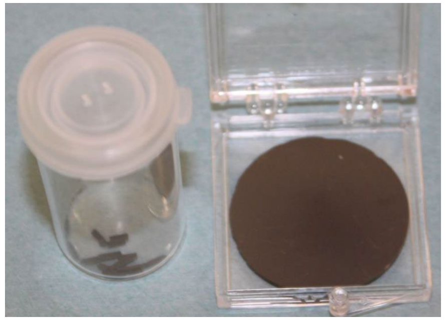
Simulation tests for $^{239}PuO_2$

- Pellet
  - Mass 2 g
  - Thickness as low as possible (max 1 mm)
  - Mechanical stable, good strength



2 g UO_2
Diameter 30 mm
Thickness 0.7 mm

Die diameter	Pressure Force	Powder	Powder mass	Pellet mass	Pellet thickness	Effective density	Strength
mm	kN		g	g	mm	%TD	
30	200	$^{nat}UO_2$	2.07		0.7		not good
12	60	CeO_2	0.46	0.46	1.0	55	good
12	60	CeO_2	0.55	0.55	1.1	59	good
12	53-60	$^{nat}UO_2$	0.66	0.65	1.1	46	good
12	62	$^{nat}UO_2$	0.54	0.54	0.9	48	good
12	62.5	$^{nat}UO_2$	0.47	0.47	0.8	47	good
12	62.5	$^{nat}UO_2$	0.41	0.41	0.7	46	good

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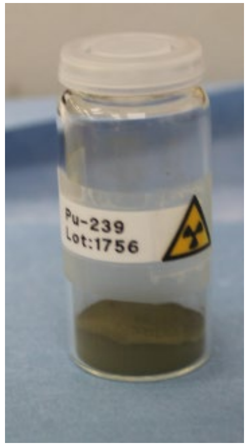
$^{239}PuO_2$ material

$^{239}PuO_2$ Batch 716
99.97% ^{239}Pu



$^{239}PuO_2$ Batch 1756
99.90% ^{239}Pu

$^{239}PuO_2$ Purified Batch 1756(p)
99.90% ^{239}Pu

- $^{239}PuO_2$ Batch 716 (ORNL batch nº Pu-239-277A) (purchased in 1979, history unknown)
- $^{239}PuO_2$ Batch 1756 (ORNL batch nº Pu-239-277BR) (purchased in 1989, history unknown)
- $^{239}PuO_2$ Batch 1756(p) is Batch 1756 after purification for americium at SCK CEN in Belgium in 2021 by peroxide precipitation, re-dissolution of the plutonium peroxide in nitric acid, oxalate precipitation and calcination at 735 °C

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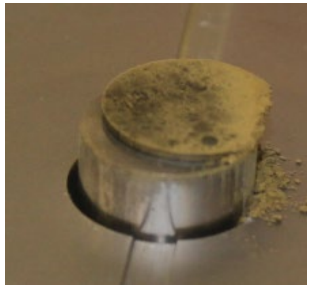
$^{239}PuO_2$ pellets: Pu batch 716

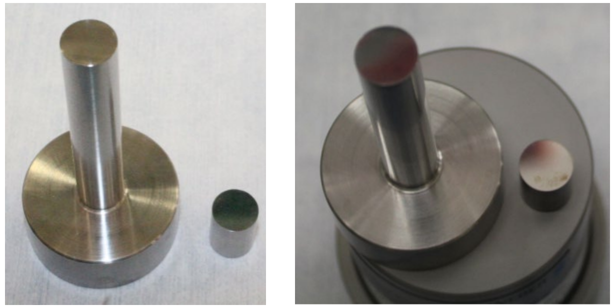
Die diameter	Pressure Force	Powder	Powder mass	Pellet mass	Pellet thickness	Effective density	Strength
mm	kN		g	g	mm	%TD	
12	60	$^{239}PuO_2$ no 716	0.50	0.50	0.75	48	good (only the 1st)

$^{239}PuO_2$ batch 716
99.97% ^{239}Pu

1st pellet



2nd pellet



Press tools before and after

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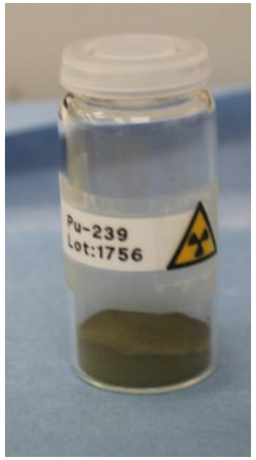
$^{239}PuO_2$ pellets: Pu batch 1756

Die diameter	Pressure Force	Powder	Powder mass	Pellet mass	Pellet thickness	Effective density	Strength
mm	kN		g	g	mm	%TD	
12	60	$^{239}PuO_2$ no 1756	0.50	0.50	0.8	48	good



$^{239}PuO_2$ batch 1756
99.90% ^{239}Pu

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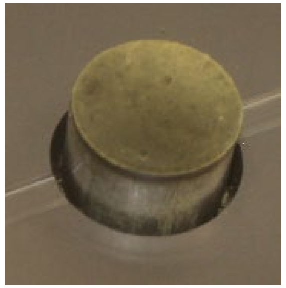
$^{239}PuO_2$ pellets: Pu batch 1756(p)

Die diameter	Pressure Force	Powder	Powder mass	Pellet mass	Pellet thickness	Effective density	Strength
mm	kN		g	g	mm	%TD	
12	60	$^{239}PuO_2$ no 1756p	0.50	0.50	0.8	48	good (only the 1st)

$^{239}PuO_2$ batch 1756(p) purified for Am



1st pellet

2nd pellet

Purification done at SCK CEN in Belgium in 2021 by peroxide precipitation, re-dissolution of the plutonium peroxide in nitric acid, oxalate precipitation and calcination at 735 °C

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$^{239}PuO_2$ pellets: summary

Die diameter	Pressure Force	Powder	Powder mass	Pellet mass	Pellet thickness	Effective density	Strength
mm	kN		g	g	mm	%TD	
12	60	$^{239}PuO_2$ no 716	0.50	0.50	0.75	48	good (only the 1st)
12	60	$^{239}PuO_2$ no 1756	0.50	0.50	0.8	48	good
12	60	$^{239}PuO_2$ no 1756p	0.50	0.50	0.8	48	good (only the 1st)

The bad plutonium pellets produced from the first batch n° 716, the heterogeneity of the colour and the corrosion of the press tools generated some questions related to the presence of impurities in the material.

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Characterization of Pu batch 716

ICP-MS

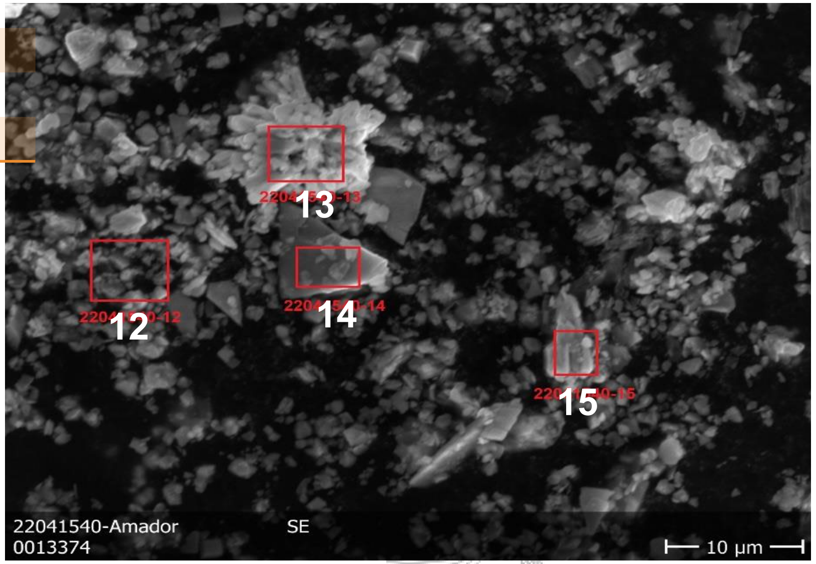
V	U	Ce	Np	Th	Ta
µg/g solid material					
12400	3340	20	15	9	5

Isotope	ppm	Wt%
U-233	2200	67
U-235	1100	32
U-238	41	1.2
U-236	1.1	< 0.05
U-234	0.4	< 0.05

SEM-EDX

position	12	13	14	15
	Wt%	Wt%	Wt%	Wt%
C	2.93	4.06	13.36	6.00
O	14.37	13.77	10.58	12.66
Si	1.49	0.65	0.58	1.06
Cl	2.04	1.69	1.10	1.68
Ca	1.48	1.82	1.34	1.54
Fe	0.55	0.23	0.44	0.33
U	2.92	2.81	1.95	2.28
Pu	74.23	74.96	70.65	74.46

Position	1-2-3-4-5-6-7	8-9	12-13-14-15
Shape	Pellet	Particules	Powder
C	x	x	x
O	x	x	x
Si	x	x	x
Cl		x	x
Ca		x	x
Fe/Ti			x
V		x	
U	x	x	x
Pu	x	x	x



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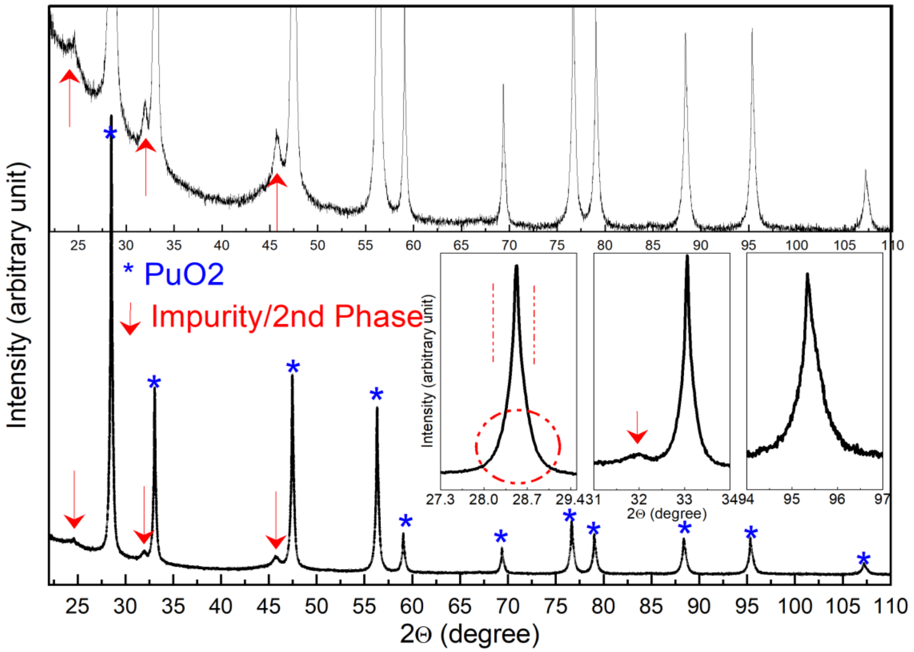
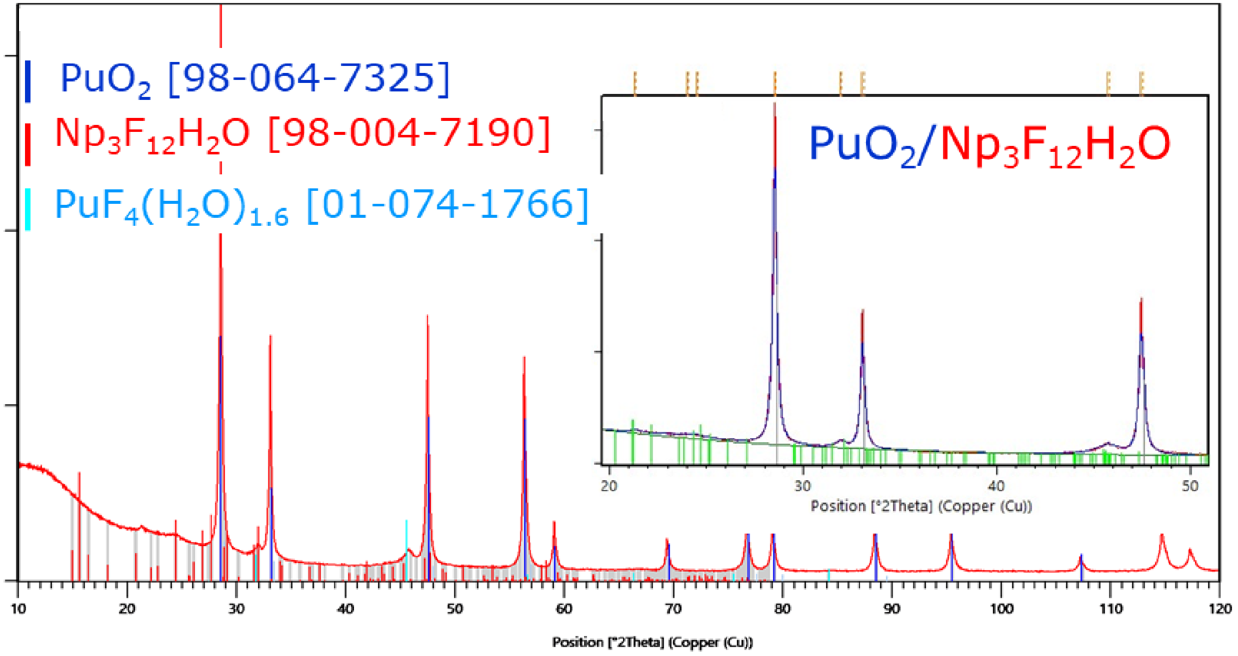
Characterization of Pu batch 716

XRD analysis





Inset: peak fitting with PuO_2 and $Np_3F_{12}H_20$ phases

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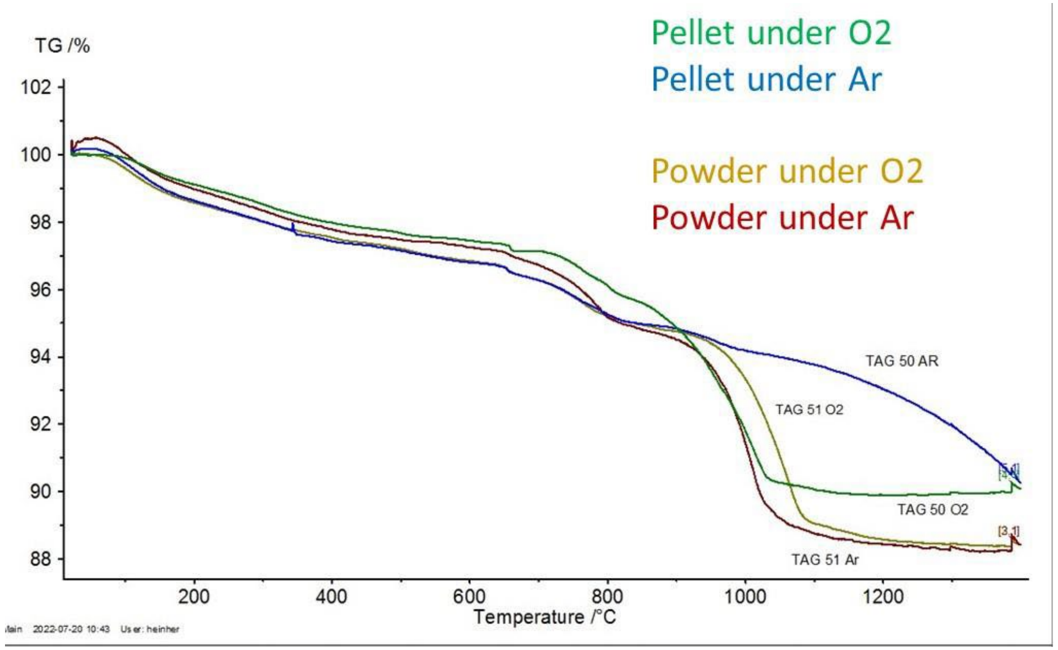
Characterization of Pu batch 716

DTA-GTA in O_2 and Ar



Important weight loss of 11 wt% as a result of the DTA-GTA in O_2 and Ar

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Summary

Pressure force and diameter were chosen to produce mechanical stable green pellets with a mass of 2 g and a thickness as thin as possible and maximum 1 mm by uniaxial pressing the powder with a hydraulic press without using fluids, binders, lubricants to avoid addition of impurities and enable simple recovery of limited and valuable material.

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Summary

Die diameter	Pressure Force	Powder	Powder mass	Pellet mass	Pellet thickness	Effective density	Strength
mm	kN		g	g	mm	%TD	
20	160	^{94}Mo	1.95	1.95	1.0	59	good
20	160	^{95}Mo	1.98	1.98	1.1	54	good
20	160	^{96}Mo	1.92	1.92	1.1	56	good
12	60	$^{239}PuO_2$ no 716	0.50	0.50	0.75	48	good (only the 1st)
12	60	$^{239}PuO_2$ no 1756	0.50	0.50	0.8	48	good
12	60	$^{239}PuO_2$ no 1756p	0.50	0.50	0.8	48	good (only the 1st)

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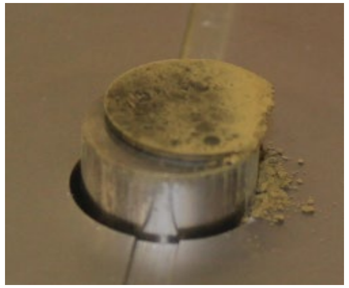
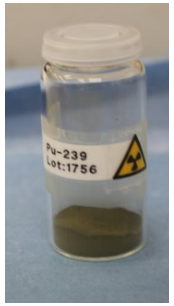
$^{239}PuO_2$ pellets: summary

Batch 716

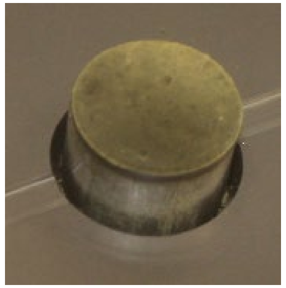


Analysed for impurities:
C, CO_2 and Cl



Batch 1756

Purified 1756 (p)



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Summary

- The analyses of the PuO_2 powder of batch 716 by ICP-MS, SEM-EDX, XRD and DTA-GTA showed beside the expected Pu, O and actinide elements from the radioactive decay of Pu, the important presence of V, C, and Cl. Carbon could be linked to the purification process of plutonium dioxide via thermal decomposition of hydrated plutonium(IV) oxalate.
- We can only assume that the presence of C, CO2, and Cl in the purified Pu material, in combination with water absorbed from the environment, caused corrosion of the steel press tools. This could result in end-capping by friction of the Pu material with the inner wall of the die sleeve and/or that volatile impurities caused high porosity and cracks in the Pu pellet.

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Summary

- Powder contamination, crystalline order and morphology of grains can influence the pressing process.
- This depend on
  - powder production process
  - storage container
  - purification process
  - process environment, etc.

  which is not always known!

My thanks go to

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