

SCK•CEN

-

ThO₂ for LWR applications

Historic notes and on-going research



DINSDAG 17 SEPTEMBER 2013

zoeken op v

DDS DE DAGELIJKSE STANDAARD

HOME NIEUWS SPORT
DE AARDE GLOBAL WARMING

DOSSIER
KERNENERGIE

Alternatieve
vuur"

Jonathan Willeman - 08/01/13

© belga. De kerncentrale van Doel

Terwijl veel Europese
tijdperk vaarwel zegg
ontwikkeling van een
thorium, door voorste
energie doorbraak sin

Dat is misschien wat
overdreven, zegt Geoff
kernenergie-expert van
universiteit van Cambri

Thorium: alternatief voor windmolens?

f Delen

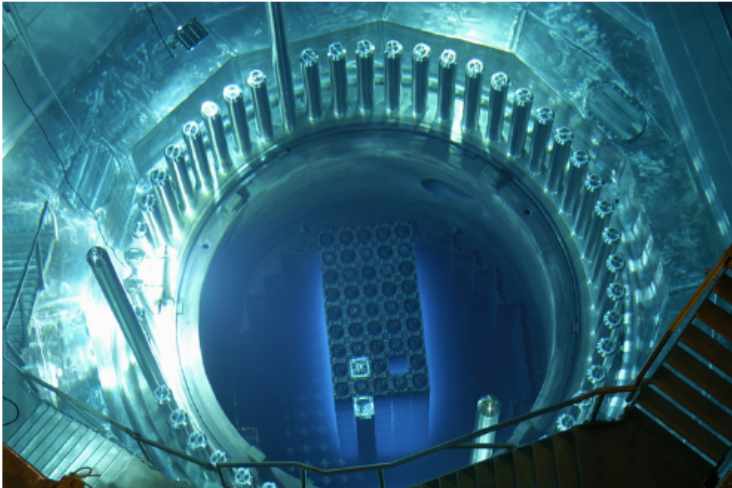


Foto: Thorium reactor

Door Hans Labohm 22 oktober 2014

& MEDIA VKS

OPMERKELIJK

Life Women
Business | You

van thorium

IEER OVER
ernenergie Energie Japan Wetenschap

nuclear
alternative to

Reactorblok kerncentral
afgesloten na brand

Kerncentrale Californië
gesloten

Belgische kernreactoren

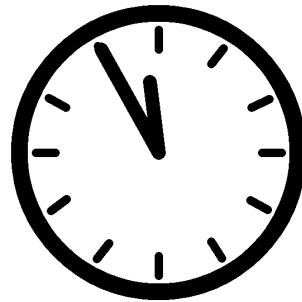
Geen haarscheurtjes in
Borssele

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**Did thorium really remain overlooked for decades?
Is thorium the wonder solution for our energy supply?**

Let's go back into time ...

2017



1953

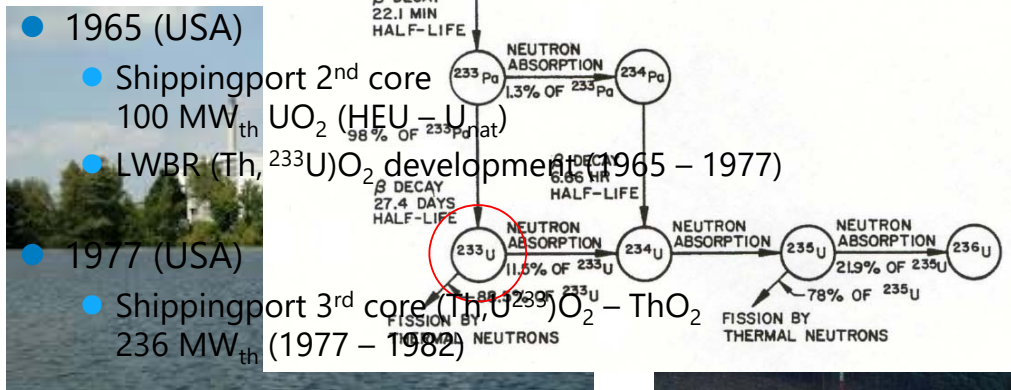
Prelude (1953 – 1958)

- **1953**
 - 1st successful ascent of Mount Everest
 - "Atoms for Peace" (D. Eisenhower, 8/12/1953): Shippingport NPP
- **1957**
 - 1st satellite (Sputnik, USSR)
 - 1st criticality of Shippingport NPP (1957 – 1964) 60 MW_{th} U-Zr & UO₂
- **1958**
 - 1st Pizza Hut (Kansas city)
 - World expo in Brussels
 - SCK•CEN and BELGONUCLEAIRE started



Mixed oxide breeder fuels (1960 – 1980)

- 1960 (USA)
 - Feasibility studies on Th-²³³U LWBR cycle
- 1963 (Belgium)
 - (U,Pu)O₂ LWR MOX in BR-3
 - (U,Pu)O₂ FBR MOX in BR-2 (& DFR, Rapsodie, Phenix)



Breeder programs (1980 – 2000)

- 1982 Stop of the LWBR program in the Shippingport reactor
- 1989 Stop of the Fast Breeder program in Belgium
- 1997 Stop of Superphénix in France



Thorium: miracle solution ?

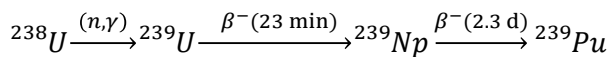
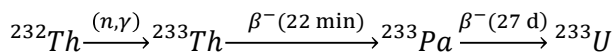
Some facts and figures



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U-Pu versus Th-U breeding



^{232}Th is a fertile isotope for the fissile isotope ^{233}U much in the same way as ^{238}U for ^{239}Pu

^{233}U outperforms ^{235}U and ^{239}Pu in thermal and epithermal spectra in terms of eta yield (n emitted per n absorbed) & has an acceptable delayed neutron fraction

	^{233}U	^{235}U	^{239}Pu
ν	2.48	2.43	2.87
$(\sigma_f/\sigma_t)_{\text{th}}$	0.096	0.171	0.504
$(\sigma_f/\sigma_t)_{\text{epi}}$	0.182	0.489	0.618
η_{th}	2.26	2.08	1.91
η_{epi}	2.10	1.63	1.77
β	0.0031	0.0069	0.0026

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[illegible]

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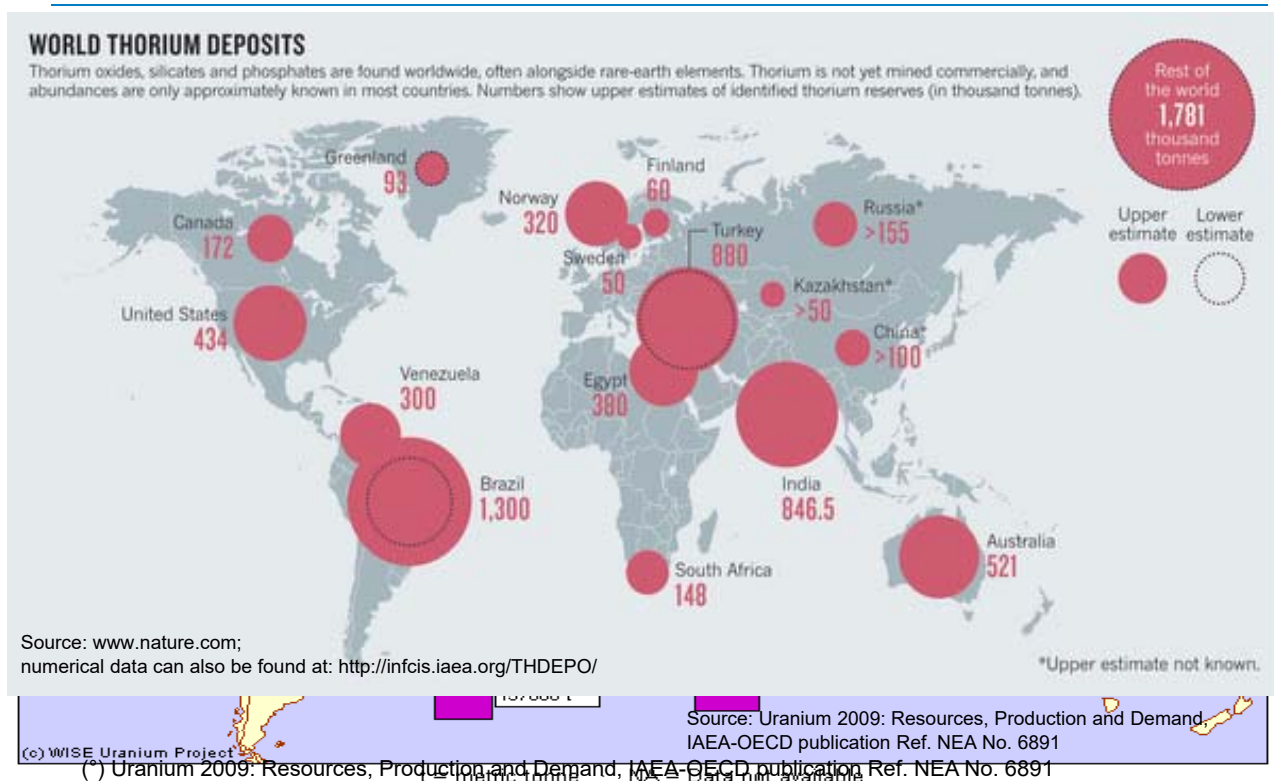
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- Acceptable « neutronics and physics » behavior
 - (+) reduced breeding of long living minor actinides
 - (+) breeding is possible in thermal and epithermal spectrum
 - (-) seed fissile isotope needed (^{235}U , ^{239}Pu or ^{241}Pu)
 - (-) smaller delayed neutron fraction in ^{233}U fission than ^{235}U fission
 - (-) "long" decay time of ^{233}Pa (27 days) compared to ^{239}Np (2.2 days) : reactivity increase after shutdown & ^{234}U production
 - (+ & -) production of ^{232}U (hard gamma emitter)
- Reactor feedback exists for various reactor types:
 - **HTR** (Peach Bottom & Fort St. Vrain, USA; AVR & THTR, Germany; Dragon, UK)
 - **HWR** (CIRUS, India)
 - **PWR** (Shippingport, USA)
 - **MSBR** (ORNL, USA)
- **Most feedback exists for Th- ^{235}U , lesser for Th- ^{233}U or Th-Pu**

Possible strategies for thorium utilization

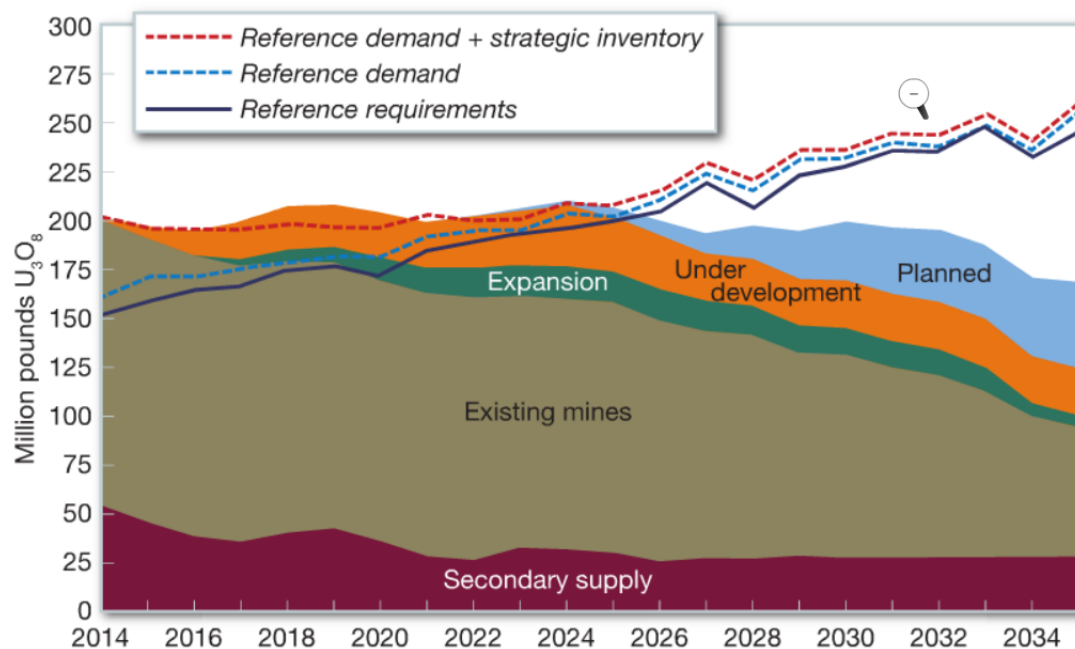
- Natural resource concerns (economic, geopolitical, security & flexibility of supply)
- Optimal use of already separated fissile material (reprocessed uranium, civil and military Pu stockpiles)
- Reduced production of long lived Minor Actinides & reduction of existing Pu inventories

Thorium: an alternative for uranium?

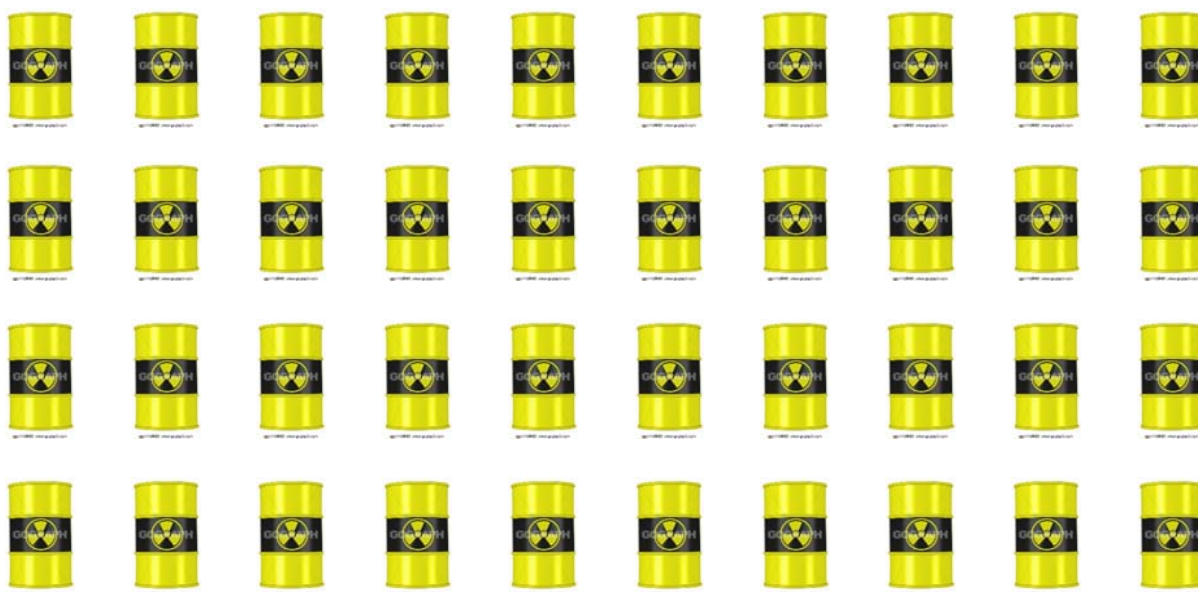


Uranium mining: ~40 000 tons/yr

Figure 3. World uranium projected supply and requirements through 2035



Uranium mining

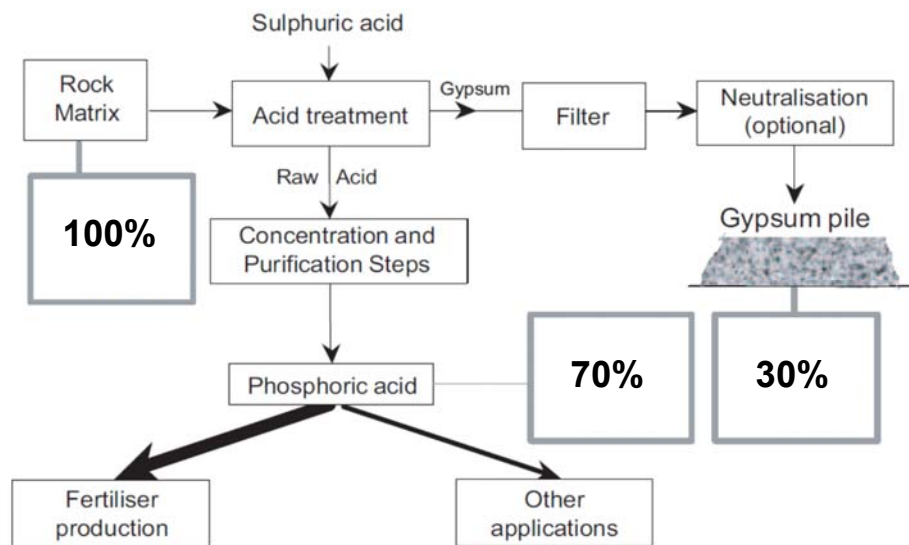


Uranium ~ 40 000 tons/year
Thorium: 100 tons/year



Unintended Th mining

- Phosphate mining: 200 000 000 tons/year



- Th-content 100 ppm → 20 000 tons/year

... Where does it land ?



Other unintended Th mining ...

- Ilmenite (TiO_2)



- Bastnazite, monazite*, ... (REE)



(*) Th is separated from monazite

... We can do better !

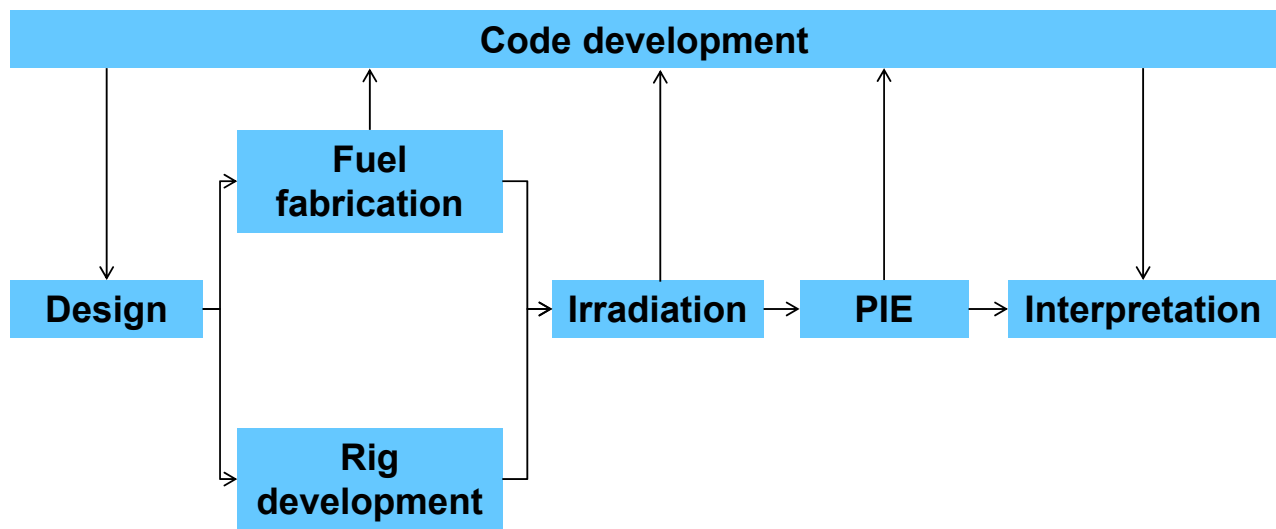
(Th,Pu)O₂ fuel experience at SCK•CEN

- Observations (1999)
 - A thorium-based fuel cycle implemented in the current fleet of reactors needs to go through a Th-²³⁵U or Th-Pu phase
 - SCK•CEN stood at the cradle of U-MOX since the early 1960's and has been leading, together with Belgonucleaire, the research, development and licensing of MOX fuel for LWR use
- Decision (1999) **to embark on a (Th,Pu)O₂ study for LWR application**
 - In 1999, SCK•CEN and ITU launched a research program to compare in-reactor performance of (Th,Pu)O₂ with UO₂ and (U,Pu)O₂
 - Since 1999, ThO₂ fuel has continuously been studied at SCK•CEN: fabrication, in-reactor performance, safety studies, spent fuel behavior (final disposal context), post-irradiation examinations, ...

(Th,Pu)O₂ fuel experience at SCK•CEN

- **European context:**
 - Thorium-based MOX was studied during the last decade in a pre-competitive context to reduce the nuclear waste burden
 - 5th FWP : Thorium Cycle (2000-2006) (*) coordinated by SCK•CEN
 - 5th FWP : OMICO* (2001-2007)
 - 5th FWP : Red Impact (2004-2007)
 - 6th FWP : LWR-DEPUTY* (2006-2011)

(Th,Pu)O₂ fuel experience at SCK•CEN



Successive (Th,Pu)O₂ R&D programs

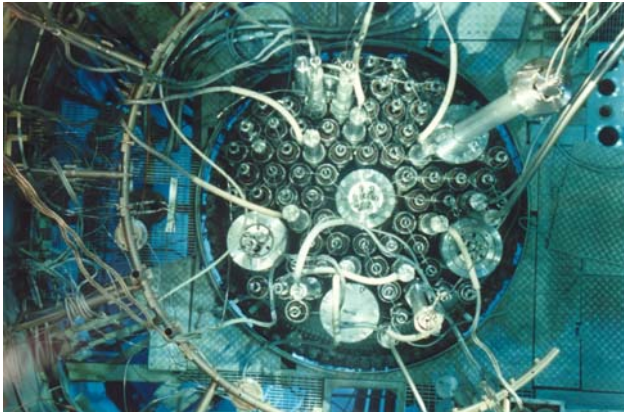
- OMICO (2001-2007)
 - Design
 - Fabrication
 - Instrumented irradiation (BOL)
 - Code development
- LWR-DEPUTY (2006-2011)
 - Post-irradiation examinations
 - Radiochemical analysis & reference datasets
 - Core assessments
- THOMOX (2012-today)
 - Irradiation to industry-relevant burnup
 - Code development

OMICO (2001-2007)

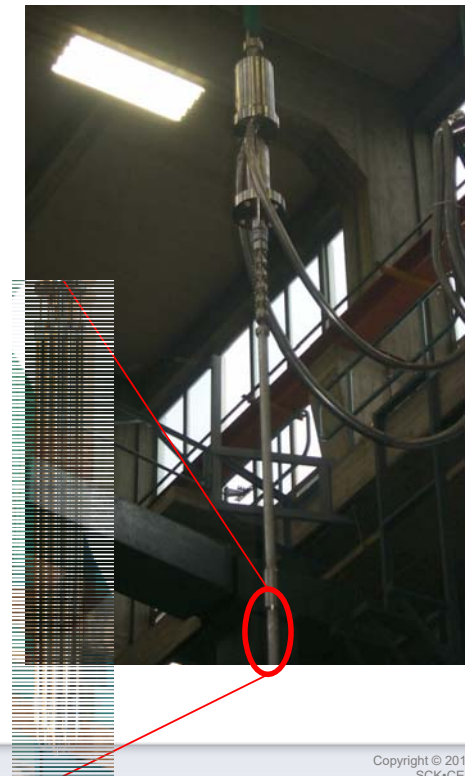
Instrumented pin assembly at ITU



Assembly & irradiation: TCH & BR-2

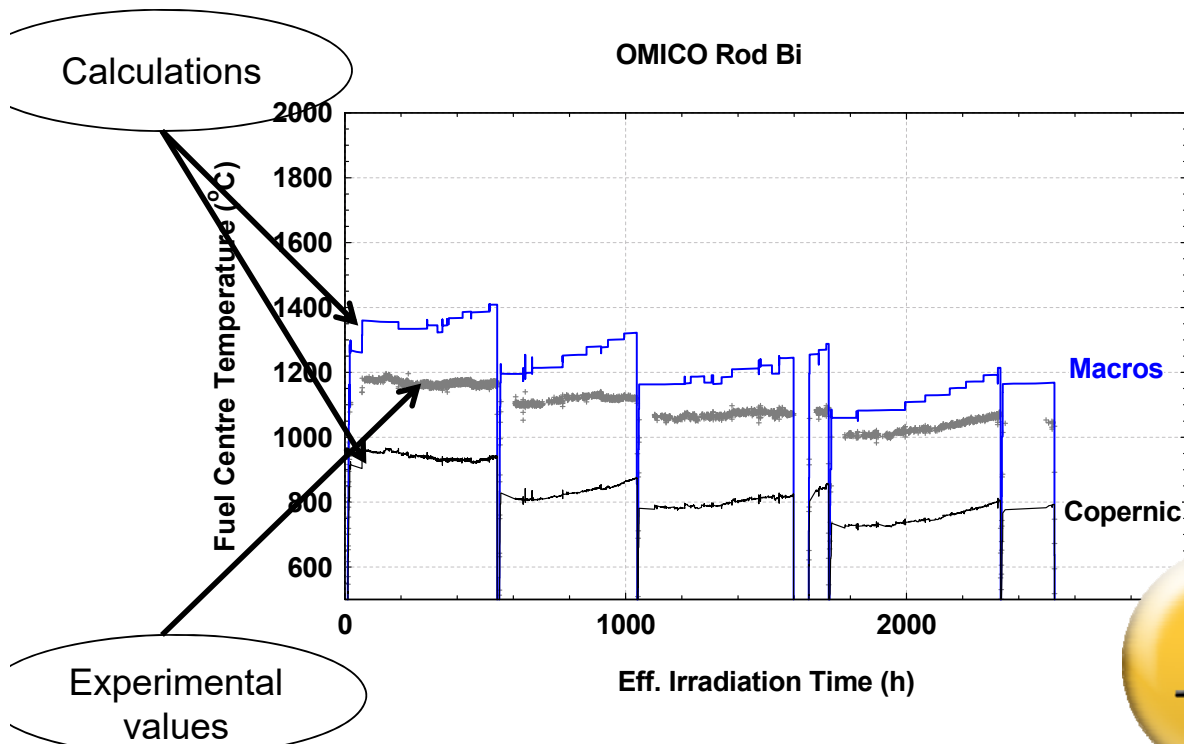


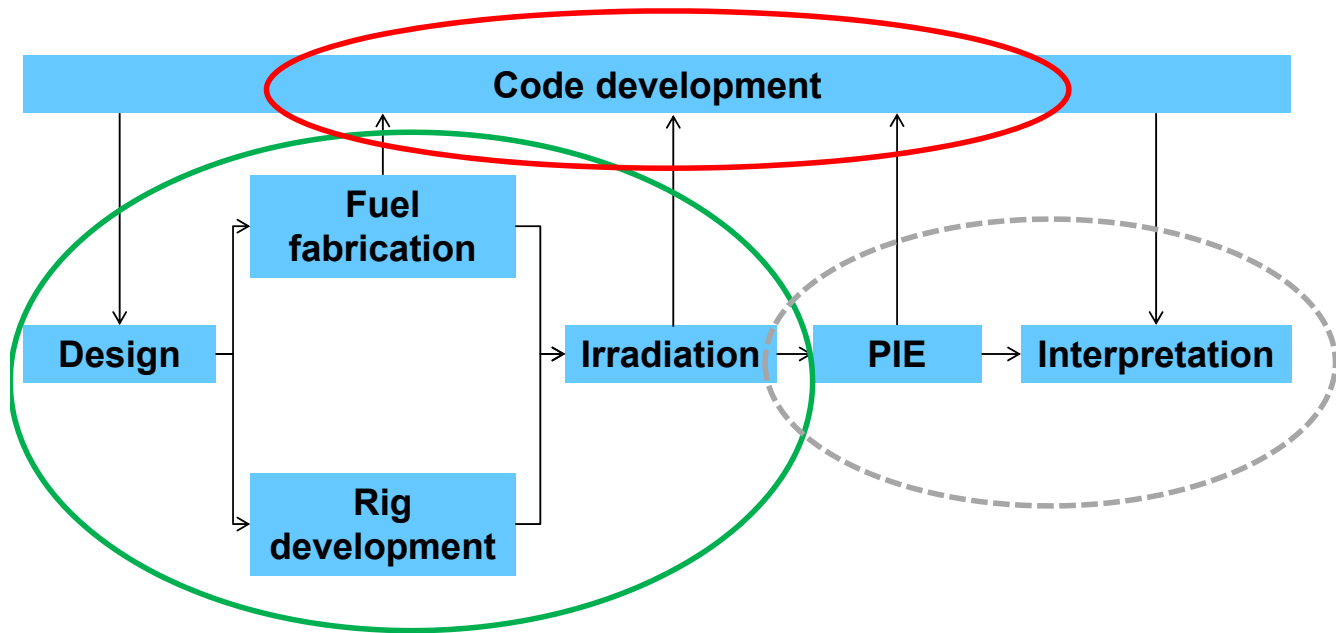
OMICO ready to be transferred to BR-2
(29-10-2004)



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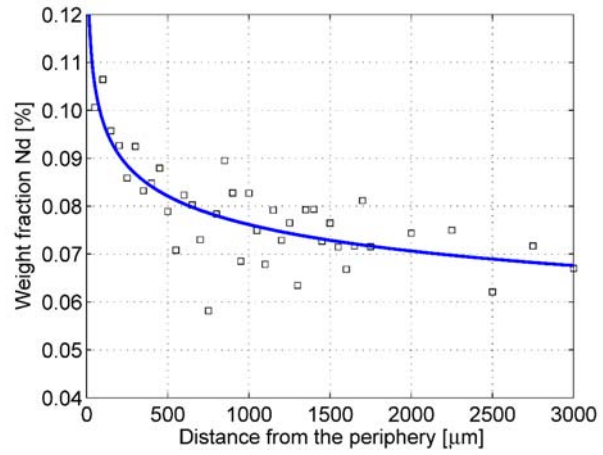
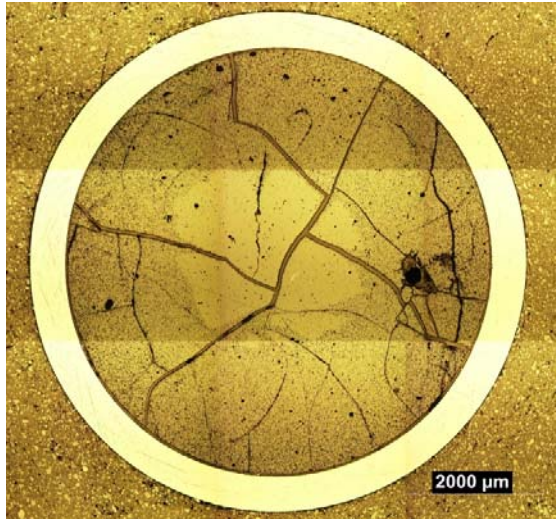
In-reactor performance of $(\text{Th,Pu})\text{O}_2$ in BR2 Model predictions (2007) versus experiment: SCK·CEN & AREVA



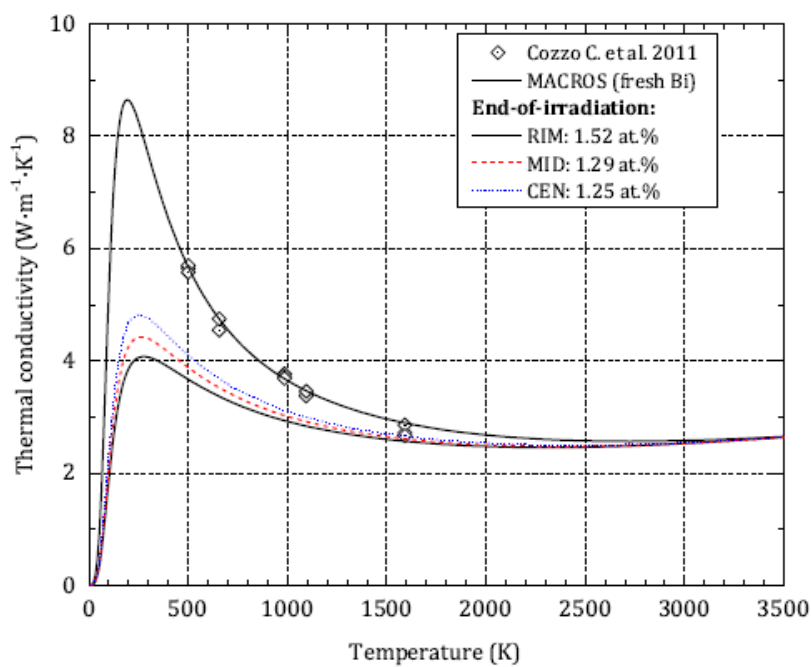


LWR-DEPUTY (2006-2011)

Post-irradiation examinations of (Th,Pu)O₂

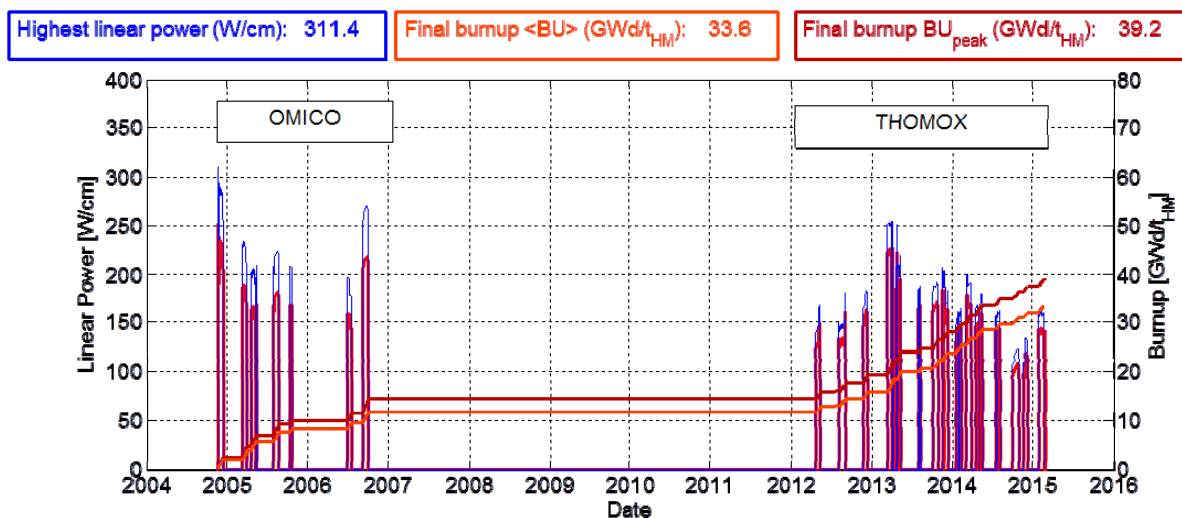


(Th,Pu)O₂: Thermal conductivity measurements & model



THOMOX (2012-today)

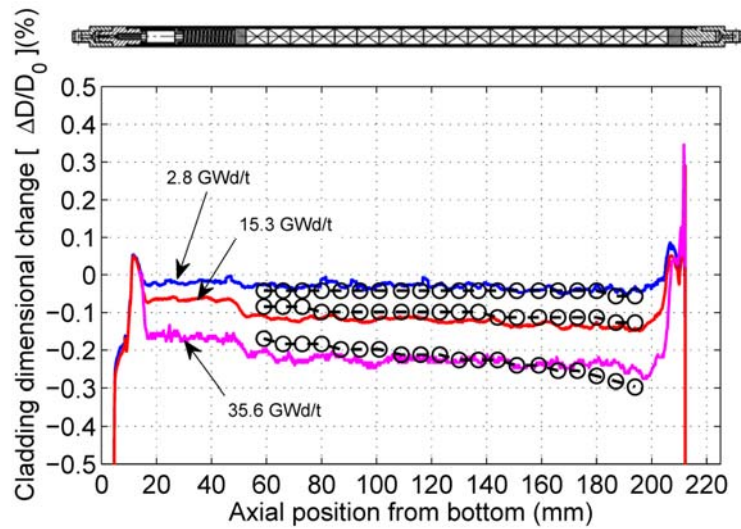
In-reactor performance of (Th,Pu)O₂ in BR2 Irradiations 2004-2015



Non-instrumented (Th,Pu)O₂ irradiations
Objective: reach ca. 40 GWd/t_{HM} (industrially-relevant burnup)

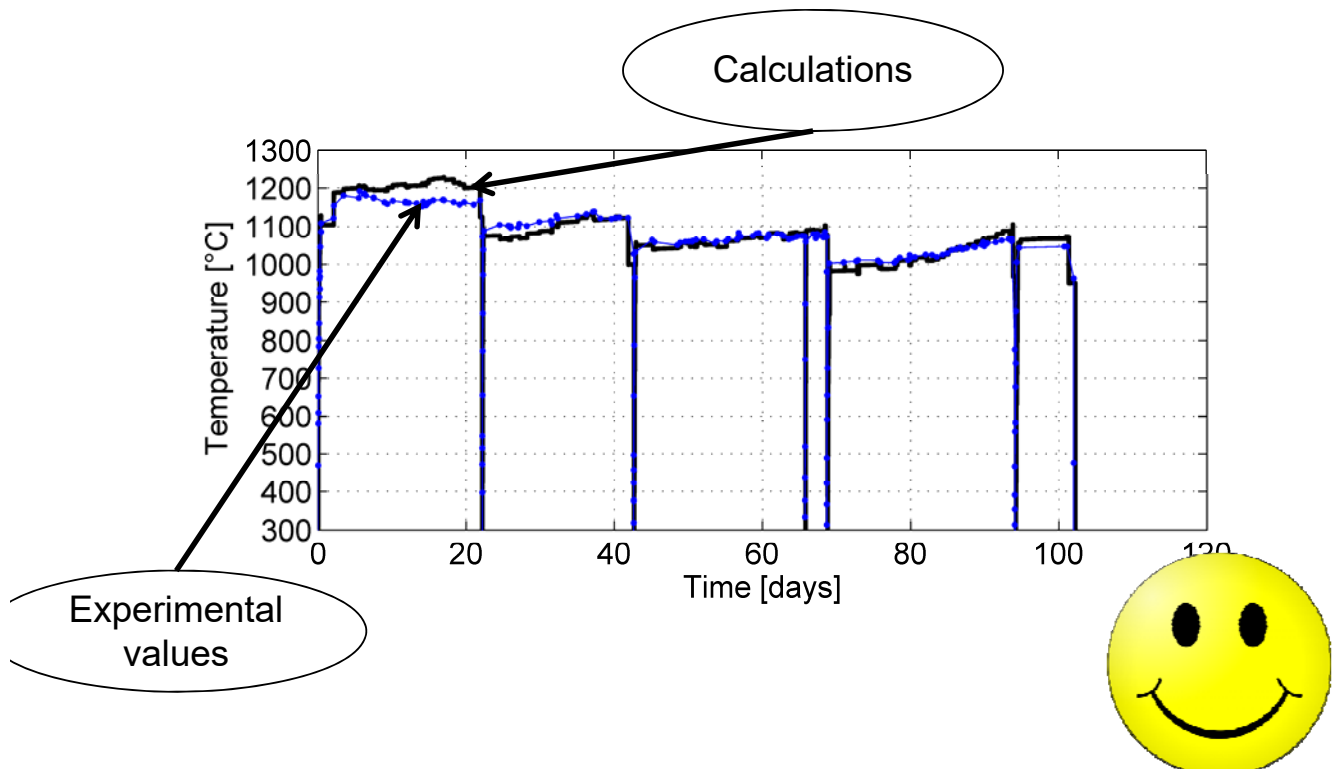
In-reactor performance of (Th,Pu)O₂ in BR2

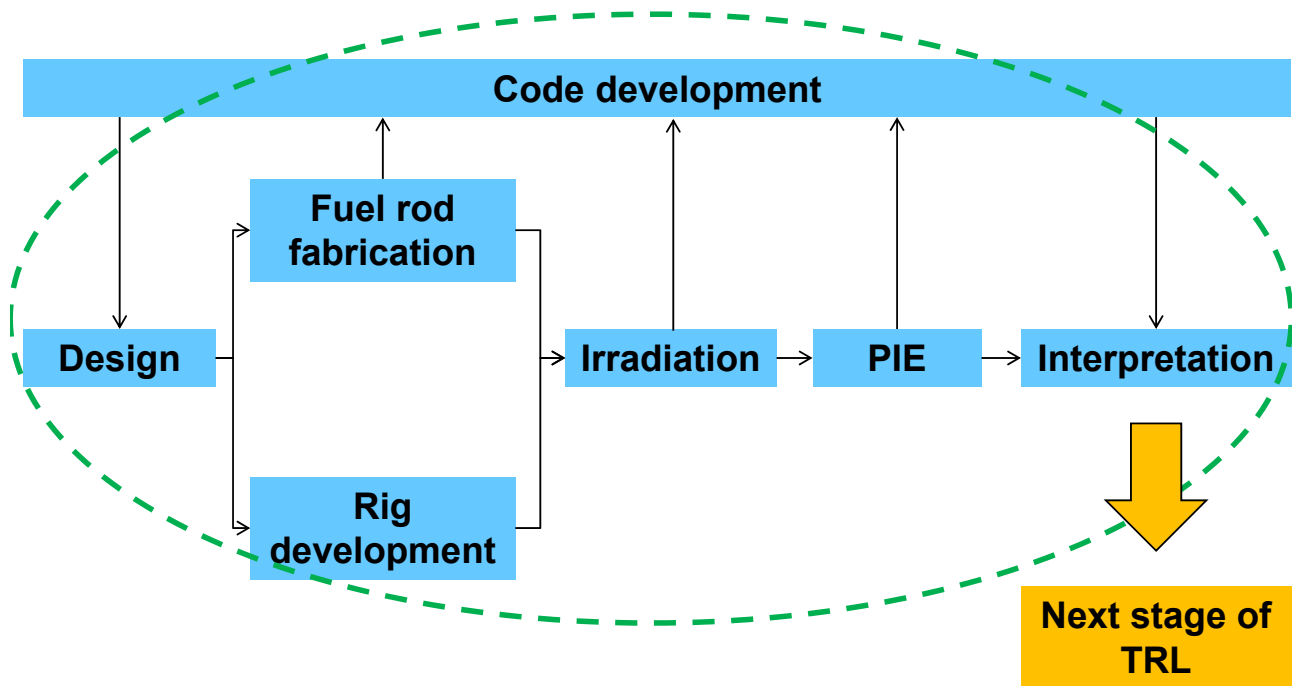
Post-irradiation examinations



In-reactor performance of (Th,Pu)O₂ in BR2

Model predictions (2015) versus experiment





Thorium and thorium-plutonium fuel R&D Where do we stand today?

Table 1
Proposed application of technical readiness levels to reactor fuel development and qualification

TRL	TRL Function	Generic Definition	Fuel Development-Specific Definition	Fuel Dev. Phase
1	Technology Selection	Technology concepts and/or applications formulated	Technical review leading to identified technology options	1
2	Technology Down-Selection	Analytical and experimental validation of critical parameters and/or proof of concept	Fuel candidates selected from options, based on selection criteria	2
3	Final Process Definition	Component and/or bench-scale validation in a relevant environment	Calculation analysis and lab-scale experimentation and characterization addressing feasibility, including: fabrication process development, property measurement, and ex-pile tests	3
4	Final Process Definition	Component and/or bench-scale validation in a relevant environment	Establish proof of concept. Fabrication of fuel elements in representative conditions. Parameters and features established	4
5	Final Process Definition	Component and/or bench-scale validation in a relevant environment	Performance phenomena identified with irradiation testing of prototype fuel elements in representative conditions (e.g., neutron dose rates, fuel and cladding temperatures, cladding damage rates) is performed and assessed	5
6	Final Process Definition	Component and/or bench-scale validation in a relevant environment	Prototypic rod/compact and assembly element irradiation in representative conditions. Repetitive compositions. Design parameters investigated	6
7	Final Process Definition	Component and/or bench-scale validation in a relevant environment	System prototype demonstration in representative conditions. Repetitive compositions. Design parameters investigated	7
8	Final Process Definition	Component and/or bench-scale validation in a relevant environment	Actual system completed and qualified through test and demonstration	8
9	Final Process Definition	Component and/or bench-scale validation in a relevant environment	Actual system proven through successful mission operations	9

- Performance qualification of (Th,Pu)O₂ has been achieved

- Open issues

- Elaborating production routes that can be implemented industrially
- Demonstrating the in-reactor performance in a power plant
- Fuel back-end solutions should be further quantified

- Today, interest in Th-Pu is rapidly growing worldwide. SCK•CEN continues its R&D:

- Implementation of a new fuel synthesis laboratory
- PhD & master thesis works on fabrication and solid state research
- Collaboration with industry on valorisation of thorium stockpiles

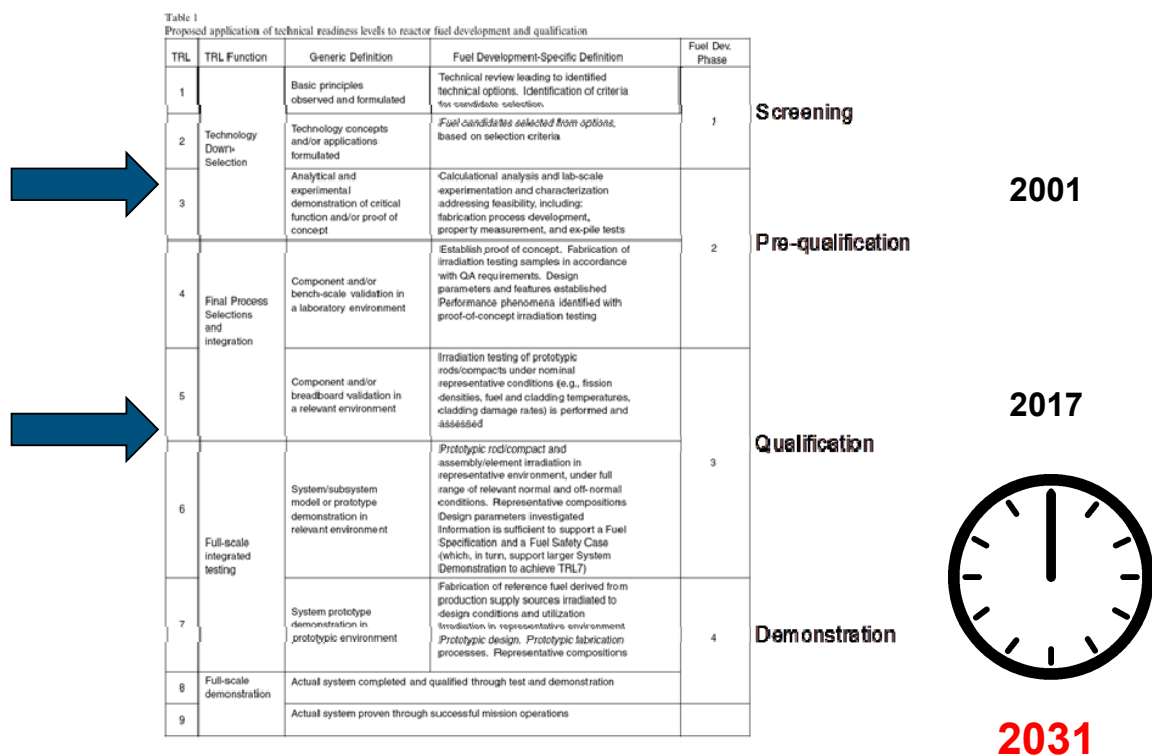


Solid state research and fabrication aspects of ThO_2

- *"UO₂ and ThO₂ doped nuclear fuel"* (A. Baena, 2011-2016 PhD thesis)
 - Solid state study on the fluorite structure effects upon doping with different lanthanides
 - Associated to her thesis: 3 master theses + 1 internship:
 - *"Advanced manufacturing routes for ThO₂ doped with lanthanides"* (M. Pasniewsky)
 - *"Synthesis of ThO₂ doped with Gd via wet chemical routes"* (P. Zsabka)
 - *"Recycling of ThO₂ scrap from nuclear fuel pellets fabrication"* (A. Schneider, L. Gijsemans)
- *"Improving the sintering behaviour of ThO₂"* (T. Wangle, 2015-2019 PhD thesis, industrial sponsor: Solvay)
 - Converting thorium nitrate into sinterable oxide powders
 - Associated master thesis / internships:
 - *"Effect of pH, T and concentration on the oxalic precipitation of Th(IV) from nitric acid media"* (M. Beliš)
 - *"Recycling of ThO₂ scrap from nuclear fuel pellets fabrication"* (R. Lommelen)

- The next steps:
 - **Elaborating production routes that can be implemented industrially**
 - We're working on it ...
 - **Demonstrate in-reactor performance**
 - Qualification of neutronics codes: benchmarking and uncertainty analysis
 - Assembly design (neutronic) for given core management
 - AOO, DBA analysis
 - Perform lead irradiation tests (power reactor: normal operation, test reactor: off-normal conditions)
 - ... still a lot to do ...

Thorium and thorium-plutonium fuel R&D Where do we stand today and where do we go?



Thorium is not a miracle solution, but it surely is a viable option ...



Open literature publications

- [1] M. Verwerft, S.E. Lemehov, M. Wéber, L. Vermeeren, V. Kuzminov, V. Sobolev, Y. Parthoens, B. Vos, S. Van den Berghe, H. Segura, P. Blanpain, J. Somers, G. Toury, J. McGinley, A. Schubert, P. Van Uffelen, D. Haas, *Oxide Fuels: Microstructure and Composition Variations (OMICO) - Final report*, in: SCK•CEN, Mol, report EUR 23104 (2007).
- [2] T. Cardinaels, M. Verwerft, D. Haas, J. Somers, G. Toury, *Innovative MOX fuels: advanced (U,Pu)O₂ and (Th,Pu)O₂ MOX*, Proceedings of 2010 LWR Fuel Performance/TopFuel/WRFP, Orlando, Florida, USA, 2010.
- [3] M. Verwerft, M. Gysemans, A. De Bremaecker, O. Schitthelm, R. Nabbi, A. Rineiski, B. Vezzoni, V. Romanello, S. Mittag, S. Kliem, J. McGinley, S. Van Winckel, D. Papaioannou, A. Worrall, C. Grove, D.F. da Cruz, P.R. Hania, F.C. Klaassen, N. Kivel, R. Restani, M. Martin, P. Darilek, J. Breza, C. Strmensky, *LWR-DEPUTY - Light Water Reactor fuels for Deep Burning of Pu in Thermal Systems Final Report (FI6W-036421)*, in: SCK•CEN, Mol, report ER-200 (2011).
- [4] D. Haas, M. Hugon, M. Verwerft, *Overview of European experience with thorium fuels*, in: J.P. Revol, M. Bourquin, Y. Kadi, E. Lillestol, J.P. de Mestral, K. Samec (Eds.), *Thorium Energy for the World*, Springer International Publishing, Switzerland, 2016.
- [5] B. Boer, S. Lemehov, M. Wéber, Y. Parthoens, M. Gysemans, J. McGinley, J. Somers, M. Verwerft, *Irradiation performance of (Th,Pu)O₂ fuel under Pressurized Water Reactor conditions*, J. Nucl. Mater. 471 (2016) 97-109.
- [6] A. Baena, T. Cardinaels, B. Vos, K. Binnemans, M. Verwerft, *Synthesis of UO₂ and ThO₂ doped with Gd₂O₃*, J. Nucl. Mater. 461(1) (2015) 271-281.
- [7] A. Baena, T. Cardinaels, K. Govers, J. Pakarinen, K. Binnemans, M. Verwerft, *Lattice contraction and lattice deformation of UO₂ and ThO₂ doped with Gd₂O₃*, J. Nucl. Mater. 467 (2015) 135-143.
- [8] A. Baena, T. Cardinaels, J. Vleugels, K. Binnemans, M. Verwerft, *Activated sintering of ThO₂ with Al₂O₃ under reducing and oxidizing conditions*, J. Nucl. Mater. 470 (2016) 34-43.
- [9] A. Baena, T. Cardinaels, J. Van Eyken, J.L. Puzzolante, K. Binnemans, M. Verwerft, *Effect of sintering atmosphere on the hardness of ThO₂*, J. Nucl. Mater. 477 (2016) 222-227.
- [10] R. Bès, J. Pakarinen, A. Baena, S.D. Conradson, M. Verwerft, F. Tuomisto, *Charge compensation mechanisms in U_{1-x}Gd_xO₂ and Th_{1-x}Gd_xO_{2-x/2} studied by X-ray Absorption Spectroscopy*, J. Nucl. Mater. 489 (2017) 9-21.
- [11] P. Zsabka, G. Leinders, A. Baena, T. Cardinaels, K. Binnemans, M. Verwerft, *Synthesis of gadolinium-doped thorium dioxide via a wet chemical route: Limitations of the co-precipitation method*, J. Nucl. Mater. 489 (2017) 211-221.
- [12] T. Wangle, V. Tyrpekl, S. Cagno, D. T., O. Larcher, T. Cardinaels, J. Vleugels, M. Verwerft, *The effect of precipitation and calcination parameters on oxalate derived ThO₂ pellets*, J. Nucl. Mater. (Submitted).
- [13] S. Cagno, L. Gysemans, V. Tyrpekl, T. Cardinaels, M. Verwerft, K. Binnemans, *Use of triflic acid in the recycling of thorium from nuclear fuel production scrap*, Journal of Sustainable Metallurgy (Accepted for publication).
- [14] V. Tyrpekl, M. Beliš, T. Wangle, J. Vleugels, M. Verwerft, *Alterations of thorium oxalate morphology by changing elementary precipitation conditions*, J. Nucl. Mater. (Submitted).
- [15] A. Baena, T. Cardinaels, J. Vleugels, K. Binnemans, M. Verwerft, *Sintering of thorium oxide comprising materials*, in: W.I.P. Organization (Ed.) 2017.

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