Progress in Am transmutation targets from EFTTRA

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The EFTTRA Collaboration: 15 years from 1992 to 2006

- Goal: transmutation of long-lived FP and MA
- Tc-99 and I-129 irradiations performed and reported
- Inert matrices selection and irradiations in Phenix and HFR
- Pre-selection of MgAl$_2$O$_3$ for Am targets transmutation
- Irradiations (Am) in HFR: T4 and T4bis (2 burn-up levels)
- Results not conclusive > Selection of Zirconia (once-through scenario) or MgO, Mo (multiple recycling scenario)
- Preparation of HELIOS irradiation in HFR
- Further developments
EFTTRA: a 15-years successful collaboration

- Agreement on experimental programmes, seminars, publications
- Financing by the partners (case by case)
- Financial (partial) support through EC Framework Programmes:
  - FP5 (EFTTRA T4)
  - FP6 (EUROTRANS IP: HELIOS, BODEX)
EFTTRA T4 and T4bis Objective

- MgAl$_2$O$_3$ spinel as IMF for *once-through transmutation*
- Development of Am spinel target fabrication
- First irradiation testing of Am spinel
- In-pile behaviour, transmutation demonstration
- Burn-up effect (two irradiation times)
EFTTRA T4 and T4bis main conclusions

- Am spinel fabrication demonstrated, but improvements implemented (matrix powder infiltration instead of green pellet infiltration)
- Irradiation successful (up to 650 days): no failure, good pellet structure
- Very high gas production (mainly He), low release (low T°)
- Very large pellet swelling (gas bubbles)
- Am-Spinel chemical instability: formation of AmAlO₃
EFTTRA-T4(bis) pellets: green pellet infiltration

Inhomogeneous Am distribution in spinel

HELIOS: sol-gel beads infiltration

Solid-solution formation in the sintered pellet
EFTTRA T4 and T4bis main conclusions

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- Irradiation successful (up to 650 days): no failure, good pellet structure
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- Very large pellet swelling (gas bubbles)
- Swelling also due to n-damage
- Am-Spinel chemical instability: formation of AmAlO3
The two maxima are due to formation and fission of Am-242m and Pu-239, respectively. The EFTTRA T4 irradiation showed a similar power profile, but it did not reach the second maximum due to the shorter irradiation time.
As-fabricated T4 fuel

Irradiated T4: 17% porosity

AmAlO₃

Irradiated T4bis: 25% porosity
T4bis Electron Probe Micro-Analysis

Composition of the 50% actinides remaining in the fuel

Pu: 71%
Cm: 19%
Am: 10%
EFTTRA T4bis Profilometry and X-ray image

Cross section for investigation

Axial position (mm)

Outer cladding diameter (mm)
Profilometry and optical micrograph of an MgAl2O4 single crystal <110> orientated irradiated with iodine ions of 72 MeV energy. The respective fluences are 1015 for a) and 1017 ions/cm² for b). The optical micrograph shows clearly the irradiated area popping out from the original crystal surface.
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- Irradiation successful (up to 600 days): no failure, good pellet structure
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X-ray diffraction of EFTTRA-T4 fuel

EFTTRA-T4 1996
spinel +
Am phase similar
to AmAlO$_3$

EFTTRA-T4 2006
spinel (crystalline!)
no Am phase

AmAlO$_3$ (Keller 1965)
EXAFS measurement

- annealing at 1600°C in Ar/H\textsubscript{2} (as for production in 1996)
- about 60 mg Am-spinel powder in a steel shielded Plexiglass cuvette
- measurement at INE Beamline at the Ångströmquelle Karlsruhe, ANKA

EXAFS provides information of local atomic structure:
- interatomic distance
- coordination number
- type of atom in coordination shells
- disorder
Am L$_{\text{III}}$-edge EXAFS of aged and annealed EFTTRA-T4

10 years aged EFTTRA-T4 fuel shows no peaks corresponding to a periodic structure.

Am L$_3$-edge EXAFS of annealed EFTTRA-T4 fuel can be fitted with AmAlO$_3$ structure.

$\Rightarrow$ AmAlO$_3$ is predominant in EFTTRA-T4 fuel.
General conclusion related to the use of spinel as IMF

- Conclusion based on T4+T4bis + other experiments in HFR and Phenix
- At low temperature, He retention leading to swelling
- At high temperature He release, but chemical instability
- Study started of a more stable IMF (once-through): Y-stabilised Zirconia
- Additional investigation of MgO and Mo for recycle strategies
- Need of irradiation experiments: HELIOS in HFR
EUROTRANS IP (FP6) Irradiation Tests

- **FUTURIX**: irradiation test in Phénix of TRU-fuels under EFIT relevant conditions

- **BODEX**: irradiation test in HFR to study the helium buildup and release from inert matrixes for IMF’s

- **HELIOS**: irradiation test in HFR to study the in-pile behaviour vs temperature of U-free Am targets
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- **Fuel**
- **Control rod**
- **Be reflector**
- **Irradiation positions**

**HELIOS**
<table>
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<tr>
<th>Pin Nr</th>
<th>Composition</th>
<th>Microstructure</th>
<th>As-fabricated density [g/cm³]</th>
<th>Fuel Manufacturer</th>
<th>Fuel Type</th>
<th>Remarks</th>
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<tr>
<td>1</td>
<td>$\text{Am}_2\text{Zr}_2\text{O}_7+\text{MgO}$</td>
<td>10-50 $\mu$m</td>
<td>0.7</td>
<td>0</td>
<td>CEA</td>
<td>Cer Cer</td>
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<td>$(\text{Am},\text{Zr},\text{Y})\text{O}_2$</td>
<td>Solid solution</td>
<td>0.7</td>
<td>0</td>
<td>JRC-ITU</td>
<td>Instrumented with Central TC</td>
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<td>$(\text{Am},\text{Pu},\text{Zr},\text{Y})\text{O}_2$</td>
<td>Solid solution</td>
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<td>$(\text{Zr},\text{Am},\text{Y})\text{O}_2+\text{Mo}$</td>
<td>60-120 $\mu$m max 30 vol%</td>
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<td>JRC-ITU</td>
<td>Cer Met</td>
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<td>5</td>
<td>$(\text{Pu},\text{Am})\text{O}_2+\text{Mo}$</td>
<td>40-150 $\mu$m max 30 vol%</td>
<td>0.3</td>
<td>1.2</td>
<td>JRC-ITU</td>
<td>Cer Met</td>
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Power calculations

- $\text{Pu}_{\text{fis}} 0.08 \text{ g/cm}^3 + ^{241}\text{Am} 0.7 \text{ g/cm}^3$
- $^{241}\text{Am} 0.7 \text{ g/cm}^3$
- Fission of $^{242}\text{Am}$
- Fission of $^{239}\text{Pu}$

Irradiation Time (Days)

Joint Research Centre

NEA P&T Exchange Meeting, Nimes, September 2006
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<td>Am₂Zr₂O₇ + MgO</td>
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<td>240</td>
<td>56</td>
<td>741</td>
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<td>(Am,Zr,Y)O₂</td>
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<td>0.7</td>
<td>170</td>
<td>40</td>
<td>811</td>
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<td>(Pu,Am,Zr,Y)O₂</td>
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<td>(Am,Zr,Y)O₂ + Mo</td>
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<td>0.7</td>
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<td>58</td>
<td>687</td>
<td>300</td>
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<td>(Pu,Am)O₂ + Mo</td>
<td>1.2</td>
<td>0.3</td>
<td>1600</td>
<td>370</td>
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➢ Onset of He release: ≈ about 550 °C
➢ Complete He release: ≈ about 1350 °C
(Am,Zr,Y)O$_2$ pre-fabrication tests

Sintered density > 90\%TD
HELIOS Status

- HELIOS irradiation shall start in 2007 in the HFR and last for about 1 year (i.e. 10 HFR cycles)
- Fuel production and hardware procurement & manufacturing underway
- Predicted temperatures in the fuels are acceptable (slightly lower than aimed)
- Destructive PIE’s results shall be available in 2009
Future EFTTRA plans

- Complete HELIOS experiment within EUROTRANS IP
- Integrate the results within other experiments: BODEX
- Demonstrate the high Am transmutation capability of Zirconia IMF with a 5-years irradiation in HFR: HELIOSbis (not decided – funding dependent)
- Looking at the feasibility of Cm transmutation in once-through mode