Overview of ATF research and ongoing experiments at the Halden reactor project

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Explosion of reactor building at Fukushima nuclear power plant

\[ \text{Zr} + 2 \text{H}_2\text{O} \rightarrow \text{ZrO}_2 + 2 \text{H}_2 \]
Fig. 1. General overview of coolant–limited accident progression inside an LWR core.

Definition on accident tolerant fuel (ATF)

ATF concepts aim to *delay* the onset of high temperature oxidation as well as ballooning and burst to reduce the burden on reactor safety systems and *increase the coping time* for the reactor operators. In the Late phase, the confinement of fission products is also desirable.
Fukushima accident triggered a lot of research into Accident Tolerant Fuel

Alternative cladding materials are being investigated

Requirements:

- At least as good as standard fuel under normal operating conditions
- Low corrosion, low hydrogen generation
- Low neutron cross section
- Good retention of fission gases (especially Tritium)
- High melting temperature
- Sufficient strength at high temperature
- Reasonable cost
- Keep hydrogen penetration (from dissolved hydrogen in PWR coolant) low because otherwise water will be created within the fuel rod (combination with oxygen), leading to an increase of the inner rod pressure (fission gas + vapor pressure (100 bar))
Working groups

1. EERA, JPNM
3. IAEA Coordinated Research Project on Accident Tolerant Fuel Concepts for LWRs (ACTOF), see also IAEA-TECDOC-1797 (978-92-0-105216-2)
4. “Collaboration for Advanced Research on Accident Tolerant Fuel” (CARAT) network which is complementary to the Westinghouse-led (DOE supported) ATF program
<table>
<thead>
<tr>
<th>Organization Name</th>
<th>Location/Institution</th>
<th>Country/Institution</th>
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<tbody>
<tr>
<td>Argonne National Laboratory</td>
<td>University of Illinois</td>
<td>USA</td>
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<tr>
<td>Ceramic Tubular Products</td>
<td>ANSTO (Australia)</td>
<td>Australia</td>
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<td>Idaho National Laboratory</td>
<td>Uppsala University (Sweden)</td>
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<td>Los Alamos National Laboratory</td>
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<td>Oak Ridge National Laboratory</td>
<td>University of Virginia</td>
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<tr>
<td>University of Pretoria (South Africa)</td>
<td>CNNC,CGN, SNPTC, CAE, NPIC (China)</td>
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<td>University of Manchester (UK)</td>
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<td>Imperial College (London, UK)</td>
<td>Halden project (Norway, OECD) (Norway)</td>
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<td>AREVA</td>
<td>France</td>
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<td>Edison Welding Institute</td>
<td>USA</td>
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<td>Chalmers University (Sweden)</td>
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<td>KIT (Germany)</td>
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Different Accident Tolerant Fuel concepts

1. Different cladding materials
2. Modified fuel

Cladding material is most important.
Modified fuel important to reduce fission gas release once the cladding has been damaged.
Various concepts:

1. Coated Zircaloy claddings – different types
2. Molybdenum (alloy) cladding, coated with Zr or FeCrAl
3. FeCrAl (solid tube) – different variants
4. SiC-SiC
5. MAX phase (example Ti$_3$SiC$_2$)
6. Various types of layered claddings example Zr+SiC+some coating
1. Coated zircaloy claddings

<table>
<thead>
<tr>
<th>Type of coating</th>
<th>Institute/Company</th>
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<tbody>
<tr>
<td>Nitride coatings; CrN, TiAlN, CrAlN, TiN</td>
<td>IFE/Halden, KIT, …</td>
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<tr>
<td>Cr</td>
<td>CEA, Areva, EDF, KAERI, …</td>
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<tr>
<td>Si</td>
<td>KAERI</td>
</tr>
<tr>
<td>FeCrAl</td>
<td>KIT, ORNL, University of Illinois, …</td>
</tr>
<tr>
<td>SiC</td>
<td>KIT, …</td>
</tr>
<tr>
<td>MAX phase (Ti2AlC, Cr2AlC, Ti3AlC2, ..)</td>
<td>SCK.CEN, KIT, University of Tennessee, …</td>
</tr>
<tr>
<td>Carbide based; ZrC, TiC, TaC, NbC, ..</td>
<td>KIT, …</td>
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<tr>
<td>Oxides; Al2O3, SiO2</td>
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</tr>
<tr>
<td>ODS treated Zircaloy (no coating) ; Y2O3</td>
<td>KAERI, …</td>
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</table>

+ many more ….
CrN coating

- Initially proposed at Halden (R. Van Nieuwenhove) in 2011 (HWR-1028)
- CrN coating (2-4 micron) applied by a commercially available process (PVD) at relatively low temperature (<300°C)
- First in-pile testing in the Halden reactor on small samples in BWR and PWR conditions (in 2013)
- First in-pile testing on fuel rods in the Halden reactor (in 2014) (EHPG, Røros 2014)
Results and characteristics of CrN coatings

- The coatings are very uniform and free of cracks
- The coatings are very hard (developed to improve drills)
- Very good adhesion to substrate (zircaloy, AISI 316L, Inconel 600). No spalling off (even at high deformation)
- The coatings can be stretched by 1.5-2 % before narrow cracks appear.
- Excellent corrosion resistance due to protective chromium oxide layer (BWR, PWR, CANDU, supercritical water)
- Tubes up to 4 meter long can be coated (process is available)
- The coatings are cheap
- The coatings reduce hydrogen/tritium diffusion
- Survive irradiation in BWR, PWR, CANDU, supercritical water
1. Coated zircaloy claddings

A. CrN coating

B. Cr-coatings

Pursued by KAERI and AREVA/EDF/CEA, KIT (Germany)

Newly developed coatings
Present status: Out-pile tests only (very good performance)
Coating length presently limited: Order 20 cm
Flexible; can follow ballooning
Good resistance to high temperature steam testing (1200 C)
2. Mo claddings

EPRI, ...

Mo Alloy Strength Maintains to ~>1500°C
Mo-alloys (TZM, or Rhenium alloy), ODS-Mo

EPRI, Areva, Los Alamos National Laboratory

Tubes need to be thin walled because of higher neutron absorption (alternatively; fuel with higher enrichment)

Thin-walled (0.2 – 0.25 mm) Mo tubes coated with FeCrAl
Length: 1.5 meter tubes
Good oxidation resistance in high temperature steam

Open issues:
• Radiation embrittlement
• Corrosion under irradiation
3. FeCrAl

First developed by Hans von Kantzow (Sweden)
AB Kanthal was founded in 1931
Composition: Iron, chromium (20-30 %) and aluminium (4-7.5 %)
Used for heating wires (protective aluminium oxide)
Melting temperature up to 1500 ºC.
High temperature strength, good oxidation resistance.
Hydrogen/tritium diffusion through FeCrAl is rather large and could poses a problem*. The Aluminium oxide formed on the inside of the fuel cladding could however significantly reduce the outflux of tritium. Alternatively, an extra coating could be considered.

This needs further experimental investigation under realistic irradiation conditions.

The tubes need to be made thin to reduce neutron absorption. Alternatively, the enrichment has to be increased, leading to a 15-25 % increase in fuel cost.

4. SiC-SiC

- Generally seen as the most promising ATF material
- Largest international effort (US, France, Japan, Rep. Of Korea, P.R. of China, Russia, Sweden)
- Most challenging material
- Longest development time
- Experiments with un-fuelled SiC tubes have already been performed in the Halden reactor.
<table>
<thead>
<tr>
<th>Property</th>
<th>Performance</th>
</tr>
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</table>
| Thermal conductivity of composite | 3-5 W/m K (after irradiation)  
Fairly low!                      |
| Swelling                    | Up to 2 % (vol) and saturation after 1 dpa                                  |
| Strength                    | Good stability under irradiation                                            |
| Neutron cross section       | 25 % lower than zircaloy                                                   |
| Corrosion resistance        | Air: Very good  
High temperature steam: Good resistance  
Water at high temperature and pressure:  
Low resistance  
  ▪ Low pH dependence (K. Terrani, Oak-Ridge)  
  ▪ Corrosion increases with oxygen content  
  ▪ SiO₂ is not protective  
  ▪ Enhanced corrosion by irradiation |
| Joining of SiC              | Not yet demonstrated under relevant conditions                              |
Planned test IFA-796 (PWR) in the Halden reactor (Joint Halden Program) in 2017

<table>
<thead>
<tr>
<th>rod 1</th>
<th>rod 2</th>
<th>rod 3</th>
<th>rod 4</th>
<th>rod 5</th>
<th>rod 6</th>
</tr>
</thead>
<tbody>
<tr>
<td>CEA</td>
<td>KAERI</td>
<td>Westinghouse ORNL</td>
<td>ORNL FeCrAl-1</td>
<td>HIP-Mo</td>
<td>Zr</td>
</tr>
<tr>
<td>Zr 8 μm Cr</td>
<td>Zr 50 μm CrAl</td>
<td>WEC Zr Cr coating</td>
<td>ORNL FeCrAl-1</td>
<td>HIP-Mo</td>
<td>Zr</td>
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<tr>
<td>M5 15 μm Cr</td>
<td>Zr 50 μm CrAl</td>
<td>ORNL FeCrAl-2</td>
<td>HIP-Mo</td>
<td>Zr</td>
<td></td>
</tr>
<tr>
<td>M5 8 μm Cr</td>
<td>Zr 100 μm FeCrAl</td>
<td>WEC Zr Cr coating</td>
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IFE test with CrN coating unfortunately removed. Planned irradiation duration: 4-5 years
ATF fuel

Fuels with high thermal conductivity (with lower fuel temperature, less fission gas release and less stored heat)

1. $\text{UO}_2$-SiC composite
2. $\text{U}_3\text{Si}_2$
3. UN, and UN+$\text{U}_3\text{Si}_2$ (to reduce reaction with steam)
4. $\text{UO}_2$ + diamond
5. $\text{UO}_2$ + metal (such as Zr or Mo)
6. Micro-encapsulated fuel pellets for better fission gas retention
7. $\text{UO}_2$ + graphene (new proposal 2015; R. Van Nieuwenhove)
Graphene

- Strength 100 x strength of SS
- Very high thermal conductivity (5300 W/mK) – Effect of irradiation unknown
First ever production of UO$_2$ pellets with graphene at IFE

Need to reduce open porosity (which will increase thermal conductivity by maybe 25 % for 2 wt% graphene)
Conclusions (on claddings)

**Claddings**

<table>
<thead>
<tr>
<th>Type</th>
<th>Time to Deployment (Yr)</th>
<th>Critical issues</th>
</tr>
</thead>
<tbody>
<tr>
<td>CrN commercial PVD coating (Halden)</td>
<td>2</td>
<td>-</td>
</tr>
<tr>
<td>Other coatings, such as Cr (non-commercial)</td>
<td>5-7</td>
<td>? (irradiation not yet performed)</td>
</tr>
<tr>
<td>Steel alloy claddings</td>
<td>7-10</td>
<td>Hydrogen and tritium diffusion Fretting (thin)</td>
</tr>
<tr>
<td>Molybdenum</td>
<td>10-15</td>
<td>Embrittlement Corrosion (needs coating)</td>
</tr>
<tr>
<td>SiC (various concepts)</td>
<td>15-25</td>
<td>Corrosion, low thermal conductivity Hydrogen and tritium diffusion Brittle (?) Endcap brazing</td>
</tr>
</tbody>
</table>
Conclusions (on fuels)

Many variants under investigation with focus on increased thermal conductivity and improved fission gas retention.

New development at Halden on UO$_2$ fuel with graphene addition
Any Questions...
Just Ask!
SiC-SiC: Composition and fabrication methods

SiC fiber in a SiC matrix (SiC-SiC)

Not a new material: > 30 year in use (aerospace, fossile fuel, fusion research, Generation IV reactor research). Fibers Invented in 1970.

Ceramic fiber-reinforced ceramic matrix composites are usually abbreviated as CFRC or CMC

Different types of SiC fibers are commercially available; Tyrano-SA, Hi-Nicalon-S, Cef-NITE,..

The filaments have typically a diameter in the range 7-14 micrometer
The filaments can be used up to 1000-1400 C.
Typical density: 3 g/cm³
Fiber tow: about 800-1000 filaments in a bundle
Coating of the fiber: 50 – 200 nm using CVI. This interphase has a function to arrest and/or deflect the matrix microcracks.
A densification process (filling the open space within the fiber structure is needed).
This is also called «matrix formation»
The infiltration is done using SiC nanopowder and additives (Al₂O₃, Y₂O₃)
SiC Fiber Composites for Nuclear Application

- SiC/SiC fiber composite have been in development for about three decades, primarily in support of aerospace and fossil energy applications. Over the past twenty years fusion, and now fission programs are developing these materials.

Fiber pull out mechanism

**EPRI/INL/DOE Joint Workshop on Accident Tolerant Fuel - 2014**
February 27-28, 2014
San Antonio Westin Hotel, Texas

"Materials resistant to extreme conditions for future energy systems" - 12-14 June 2017, Kyiv – Ukraine
5. MAX phase

They exhibit unique deformation characterized by basal slip, a combination of kink and shear band deformation, and delaminations of individual grains

- Easy to machine (regular tool steels)
- Electrically conductive
- Produced by Kanthal (Sweden)
  - Maxthal 312 (Ti₃SiC₂); max temp 1000 C
  - Maxthal 211 (Ti₂AlC); max temp 1400 C, good oxidation resistance due to Al2O3 and TiO2 formation
  - Thermal conductivity 32-40 W/mK

Corrosion and irradiation studies are needed under relevant conditions to allow conclusions for usage as ATF cladding material