# ACCIDENT TOLERANT FUEL DEVELOPMENT

Dr. Michael Rushton on behalf of Dr. Simon Middleburgh Nuclear Futures Institute, Bangor University



BANGOR UNIVERSITY

### **REASON FOR ABSENCE**



### OVERVIEW

- Why develop accident tolerant fuels?
- Key aims for accident tolerant fuel
- Examples of claddings being developed
- Examples of fuels being developed
- Licensing new fuels



# WHY DEVELOP ATF?

- The nuclear industry has strived to improve safety since its inception.
- Severe accidents are defined by the envelope that the system's materials can operate within.
- Accidents such as Chernobyl, TMI, and now Fukushima spur on advances in technology and improve working practices.
- Some operators are demanding ATF products.







### WHAT HAPPENED AT FUKUSHIMA?

- Station blackout caused cooling of the pressure vessel to be disrupted and temperatures inside the core to rise.
- Zirconium melts at 1855 °C but loss of mechanical integrity happens at 875 °C (Zr a  $\rightarrow \beta$  phase transformation) causing fuel ballooning
- This limits cooling further aiding a runaway reaction. The water reaction proceeds at 1200 °C.
- Fuel pellets melt at ~2850 °C allowing significant flow of fuel through the crippled reactor.

Highly exothermic reaction:  $Zr + 2H_2O \rightarrow ZrO_2 + 2H_2$ Lots of heat Lots of pressure



### AIMS FOR ACCIDENT TOLERANT FUEL

- Major aims:
  - Prevent similar run-away reaction between steam and Zr in water reactors.
  - Maintain a coolable geometry in all accident scenarios.
- Other aims:
  - Reduce the overall fuel cycle cost.
  - Lower the fuel failure rate due to fuel degradation mechanisms (e.g. fretting and hydrogen pickup).
  - Improve operational versatility of fuel operation.





a) Debris-induced wear

b) Grid-to-rod fretting wear

c) Excessive cladding oxidation

"Self-sufficient nuclear fuel technology development and applications" Kim et al. Nuclear Engineering and Design 249, P. 287-296

# ANATOMY OF A NUCLEAR FUEL ASSEMBLY

#### Boiling Water Reactor



Major components:

- Fuel pellet (normally UO<sub>2</sub>, sometimes MOX)
- Cladding (Zr-based)
- Grid spacers (Ni-based in BWR, Zr-based in PWR).
- Tie rods and water rods (Zrbased).
- Channel box (Zr-based BWR only).
- Bottom filter (Steel-based)
- Top/bottom tie plates (Steel-based)

#### Pressurised Water Reactor



### TECHNOLOGIES AND TIME-SCALES



### PREVENTING THE STEAM REACTION

- Coatings for Zr cladding
  - Cr-metal
  - Alternative alloy
  - Ceramic-based
- Alternative cladding material
  - Iron-based
  - SiC-SiC cladding
  - Mometal

All considered in terms of corrosion, dissolution and structural strength/stability.



"Accident tolerant fuels for LWRs: A perspective" Zinkle et al. Journal of Nuclear Materials 448 P. 374-379

## CLADDING COATINGS

- Range of deposition methods have been explored.
  - Cold-spray
  - Atomic layer deposition
  - Pulsed laser deposition (PLD)
  - Chemical vapour deposition (CVD)
- Scalability and uniformity have been engineering challenges.
- Chemical/mechanical interaction between coating and substrate an issue.



Cr cold spray





**CrN PVD** 

### BENEFITS

- No need for complete rod material re-design (mechanical/creep properties of Zr are excellent).
- No significant change from current manufacturing routes.
- Benefits in normal operation in terms of fuel failures.
- Often coupled with a significant reduction in H-pickup.

### CHALLENGES

- Coatings tend to spall off (some are better than others).
  When this happens – oxidation can be worse. Metals are better than ceramics here.
- Coatings tend to chemically interact with the Zr-alloys. Some promote lower melting points or phase transformations (e.g. Cr).
- Some coating methods are slow and expensive (Cold spray better than vapour methods).

The majority of fuel vendors are considering Cr coatings. CrN also promising. Commercial products very likely.

### IRON BASED CLADDING

- Steel based and FeCrAl alloys are being considered due to their significantly lower corrosion rate in high temperature steam.
- Mechanical properties are excellent.
- Biggest issue is the neutronic penalty compared to Zr-alloys.
  - Fuel would need to be enriched beyond 5 wt.% U-235 the industry standard and hard upper limit in the USA (~6.5%).
  - Some high density fuels may over-come this issue.
- Also potential negative chemical interactions between fuels and cladding.



"Advanced oxidation-resistant iron-based alloys for LWR fuel cladding" Terrani et al. <u>Journal of Nuclear Materials</u> <u>448.</u> P. 420-435

### SILICON CARBIDE COMPOSITES



"Accident Tolerant Fuel Analysis" INL/EXT-14-33200

### SILICON CARBIDE COMPOSITES



### BENEFITS

- Extremely high melting/sublimation point.
- High stiffness/modulus @ high T.
- Low water reaction rate at extended temperatures.



# CHALLENGES

- Manufacturability.
- Cost.
- Sealing end-plugs.
- Hermeticity.
- · Ceramic nature of failure.
- Unsuitable for use in tensile regimes (rod internal pressure).
- Low thermal conductivity when irradiated.

Intrinsic Material Issue

• Potential negative pellet chemical interactions.

"In situ observation of mechanical damage within a SiC-SiC ceramic matrix composite" Saucedo-Mora et al. Journal of Nuclear Materials 481, P. 13-23

### SIC IS ALSO BEING CONSIDERED AS CHANNEL BOX MATERIAL FOR BWRS



Issues similar for cladding but not in contact with fuel – so a little easier.

Radiation induced swelling the largest problem (could prevent control blade movement).

Reduced amount of Zr in core by ~30% by volume.

### FAILED/UNLIKELY DESIGNS



Mo alloy variants found to be excessively expensive and poor under accident and normal operating conditions. Not under active development.

- Molybdenum claddings were championed early on but were found to be unsuitable.
- Looking as though <u>most</u> ceramic coatings are not suitable for light water reactor operation.
- Steels unlikely to be used in the USA due to the strict limits on fuel enrichment at present.

### IMPROVING FUEL CYCLE COSTS

- All major fuel vendors have advanced pellets that improve fuel behaviour and fuel cycle costs.
- Offsets cost of more robust cladding and some offer additional safety characteristics.
- Range from doped UO<sub>2</sub> pellets that have minor improvements to fuel cycle cost but good reactions with coolants.
- To significantly enhanced fuel cycle cost pellets such as uranium mononitride – with slight drawbacks in coolant interactions.



# INCREASED ENRICHMENT UO<sub>2</sub>

#### Pros

- No significant variability in terms of fuel performance and accident behaviour.
- UO<sub>2</sub> is fantastic in terms of melting point and coolant dissolution.
- Very stable with increasing burnup (accommodation of fission products is high).
- Manufacture routes very mature.

#### Cons

- UO<sub>2</sub> has a poor thermal conductivity meaning centre-line temperatures are hot.
- Low U-density.
- Licensing beyond 5 wt.% a significant regulatory challenge in some markets.



Pellets waiting for rod loading



Pellets after sintering

### DOPED FUELS

Doped pellets used to improve density and some in-reactor behaviour.

- A common dopant is Cr (both Westinghouse and Framatome/Areva have Cr-pellet designs).
- Westinghouse have operated ADOPT for >10 years in BWR market.
  - Improvements to pellet cladding mechanical interactions.
  - Transient fission gas release rates.
  - Dissolution rates into coolant.
  - Manufacturing slightly more complicated, but not too far from standard UO2.
- Other doped fuels include alumina-silicate dopants which significantly improve pellet-cladding mechanical interactions but appear to be difficult to manufacture.

### DOPED FUELS

#### Microcell- 2-10 vol.%



Metallic grain boundaries provide a compliant material with larger fission product accommodation and high thermal conductivity.

Large additions mean that fuel is displaced and manufacturing routes are complex.

#### Cr<sub>2</sub>O<sub>3</sub> additions – 500-2000 ppm



Larger grains – more compliant material with larger fission product accommodation

Small additions mean that density improvements outweigh dopant amounts

#### Alumino-silicate – 2000-5000 ppm



Larger grains – more compliant material with larger fission product accommodation.

However, displaces a significant amount of uranium and sintering of fuel is very difficult.

# HIGH DENSITY FUELS

- Three major high density fuels being considered:
  - $U_3Si_2$  (~20% more dense than  $UO_2$ )
  - UN (~40% more dense than  $UO_2$ )
  - U-alloy (~40% more dense than  $UO_2$ )
- All have significantly higher U density compared to UO<sub>2</sub>.
- The highest (after additions and porosity is considered is UN with ~40% additional U atoms per cm<sup>3</sup>.



#### U<sub>3</sub>Si<sub>2</sub> pellets manufactured at INL



U-Mo alloy spheres coated in Al

### HIGH DENSITY FUELS

#### **Benefits**

- Density
- All have significantly higher thermal conductivities compared to  $UO_2$  (cooler centre-line temperatures).

#### Drawbacks

- All have poor reactions with water. Oxidize to uranium oxides in water.
- All require significant new manufacture routes and factories.
- Melting point of U<sub>3</sub>Si<sub>2</sub> and U-alloys low and likely to melt in an accident (such as a reactivity initiated accident or loss-of-coolant scenario. The power to melt value is a key metric here.
- UN requires N isotope enrichment to N-15 to prevent neutron poisoning effect of N-14 (costs are currently falling but still an order of magnitude too high).

U<sub>3</sub>Si<sub>2</sub> has been leading (including test reactor time) but melting point and manufacturing issues seem to be fundamental drawbacks. UN now being considered more intensively.

# COMPOSITE FUELS

Composite fuels have been considered to attempt to gain benefits of multiple fuel systems. Two major classes:

- Those that include  $UO_2$  for oxidation resistance.
  - ZrB<sub>2</sub>-UO<sub>2</sub> encapsulated additive to provide burnable absorber capability – some with increase in U-235 enrichment.
  - UN-UO<sub>2</sub> composite increasing the density of the fuel whilst maintaining a corrosion rate largely similar to UO<sub>2</sub>.
- Non-oxide concepts
  - UN-U<sub>3</sub>Si<sub>2</sub>



#### $U_3Si_2 - UN$ composite

"Fabrication and thermophysical property characterization of  $UN/U_3Si_2$  composite fuel forms" J.T. White et al. Journal of Nuclear Materials 495, P. 463-474

# UO<sub>2</sub> – UN COMPOSITE FUEL



#### **Advantages**

- Improves U-density
- Improves thermal conductivity

#### **Disadvantages**

- Requires UN manufacture routes in addition to UO<sub>2</sub> routes.
- Requires N-15 enrichment.
- Reaction in water worse than  $UO_2$ .

"UO<sub>2</sub>–UN composites with enhanced uranium density and thermal conductivity" J.H. Yang et al. Journal of Nuclear Materials 465, P. 509-515

# **BORIDE-CONTAINING UO<sub>2</sub>**

- Borides have been used as coatings on UO<sub>2</sub> to act as a burnable absorber. Westinghouse's Integral Fuel Burnable Absorber (IFBA) is a good example.
  Borides have been used as coatings too reactive at the beginning of life (need to lower enrichment)
- By including them within the fuel bulk, clear improvements to thermal conductivity and burnable absorber behaviour can be made.
- Issues are mainly related to manufacturability of the fuel concept. Similar to UO<sub>2</sub>-UN composites.

"Fuel with advanced burnable absorbers design for the IRIS reactor core: Combined Erbia and IFBA" F. Franceschini et al. Annals of Nuclear Energy 36, P. 1201-1207



IFBA allows more U-235 but still has a significant reactivity peak. Better if absorber was inside pellet (more self-shielding effects).

# EXPERIMENTAL VERIFICATION AND LICENSING

The fuel system must be licensed for operation in commercial reactors.

Typically done in stages and historically has taken ~20 years for small iterations on fuel design (e.g. Cr-additions).

Requirement for ATF has made the industry innovate. Still require major steps to be taken:

- Test pellets ( $U_3Si_2$ , UN and some doped fuels are in this stage now).
- Lead test rods in commercial reactors (U<sub>3</sub>Si<sub>2</sub> planned, Cr-coating testing underway).
- Lead test assemblies in commercial reactors
- Re-load quantities (fully licensed) Cr-additive fuel is in this stage for Westinghouse and the fuel company formerly known as Areva.



In Europe this is a major issue: Halden test reactor closure has been announced.



ATR

Halden



## FUEL PERFORMANCE AND DESIGN

Testing is used to provide data to show that it is safe to operate the new nuclear fuels in commercial reactors.

This is done by combining all of the post-irradiation examination (PIE) data and on-line measurement data into a multiphysics code called a fuel performance code.

Mechanistic modelling is being used to accelerate the licensing and reduce the number of highly expensive test-reactor experiments.



Ensures fuel is safe to operate. No melting. No radioactive release. No problems.

"Mechanistic materials modeling for nuclear fuel performance" M. Tonks et al. Annals of Nuclear Energy 105, P. 11-24

### CONCLUSIONS

- Accident tolerant fuels are being developed to reduce the risks associated with a significant reactor incident.
- Cladding development provides the majority of the accident tolerance – but at an economic cost.
- New fuels are being developed to offset this cost and further improve safety/performance of the fuel system.
- In the near term: Cr-coated Zr cladding coupled with  $Cr_2O_3$  doped fuel will be commercially available.
- Following this: more advanced materials are being targeted with a significant licensing effort required (UN and composite fuels leading the novel fuel types).

### THE SILVER LINING



This is the first time in 50 years that we have put so much effort into new nuclear materials for commercial power reactors. Exciting times.