Hot cell investigation of irradiated fuel debris from the Three Mile Island unit 2 (TMI-2) reactor

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Outline of Presentation

• TMI-2 Accident & Sample Investigation Project
• TMI-2 samples and results
• Selected results from Phébus pf
• Conclusions
TMI-unit 2 (TMI-2) underwent a prolonged Loss of Coolant Accident (LOCA) on March 28\textsuperscript{th} 1979. Subsequently the examination of TMI-2 samples was organised as OECD-NEA, CSNI project under the initiative of the US Dept. of Energy (US-DOE) involving most major European national research institutes.
Three Mile Island (TMI-2)

INEL Idaho (as principal contractor) had extracted and examined the samples from TMI-2. It then shipped samples to N. American & European institutes:

- AECL Canada, PSI Würenlingen, Switzerland
- Argonne National Lab. US, Studsvik, Sweden
- FZK-Karlsruhe, Germany, CEA Saclay, France
- JRC-ITU Karlsruhe, EC, AEA Windscale, UK
- as well as JAERI Japan & KAERI, Korea (later transport).

Principal aims of the examination:

- what was corium (& other phases) composition
- what temperatures were reached
- what conditions (oxygen potentials/H₂ prodn*) prevailed
  hence what degradation reactions were likely

* from Zircaloy steam oxidation
TMI-2 Examination: Main zones of damaged reactor

TMI-unit 2 reactor in its end-state

Main Zones from which JRC-Karlsruhe & other institutes received samples:
1) molten core (yellow), 2) fused crust/agglomerate (orange), 3) loose debris above (red).
TMI-2 Examination: Debris lying on top of melted core

Cross-section of cladding of fuel rod remnant C7-3-35

C7-3-35 Fuel rod segment. Note thin external ZrO₂ oxide layer present with α-Zr(O) containing layers beneath.

Thin external oxide layers indicate only slight exposure to increased temperatures. (~800°C)
TMI-2 Examination: Core bore samples

Oxidic core bore rock is principally oxidic

G12-P9-E Fracture Surface -SEM
a) & b) show dense (U-rich) phases (white) and lighter (Zr-rich) oxide phases (dark)
TMI-2 Examination: Core bore samples

- Many fine metallic Ag precipitates on the surface.
- Fe-rich oxide phases also present.

Oxidic core bore rock apart from Ag nodules

G12-P9-E Fracture Surface -SEM

- Many fine metallic Ag precipitates on the surface.
- Fe-rich oxide phases also present.
Core bore rock G12-P9-B (BEI-SEM image)

3 phases in the fully molten rock:
- dense or white (U-rich) phases;
- less dense (grey) (Zr-rich) phases;
- light element (dark) Fe, Ni, Cr-containing phases
TMI -2 Examination: Core Bore samples

G12-P10-A fracture surface a) secondary & b) backscattered images. (2760x)

G12-P2-E (Mag. ~900x)
Corium of lighter U-rich oxide & darker, Zr-rich oxide +Fe inclusion.

Note 2–phase corium with
a) fine eutectic lamella ~0,5µm wide
b) small Ag spheres from Ag-In-Cd absorber
c) matrix composition approx. equimolar (U,Zr)O₂
Agglomerate crust O7-P4-EA (BEI-1000x) preferential oxidation of Fe-Ni nodules - pure Fe metallic core (lighter centres) surrounded by Fe-Ni oxide crust (darker zones). Indicates intermediate pO$_2$ levels
TMI-2 Sample Examination: Agglomerates

TMI-2 Upper Crust Sample (Agglomerates)

There are wide grain boundaries (containing eg. Zr & Fe) in U-rich grains. This shows gradual material interactions indication of lower temperatures than core.

Backscattered electron SEM image of an upper crust (or agglomerate) sample D8 P3-A.
TMI-2 sample examination (Agglomerate –upper crust)

D8-P3-A Agglomerate sample (190x)

Interference micro showing metallic and oxidic zones.

Incomplete interactions - lower temperatures or short time at temperature

2 phase metallic /oxidic zone

2 phase metallic zone

Oxidic zone with some secondary precipitates
TMI-2 sample examination: Agglomerate - metallic and oxidic phases

Example of partially oxidised material from agglomerate

Agglomerate N5-P1-E (1033x).
(CuO vapour coating)
Stainless steel control rod cladding with Ag spheres that contain Cr oxide nuclei.
H8 7-3 Debris from above the upper crust (BEI)
- Porous nodule has lamellar structure of Fe-Ni & Fe-Sn phases
- Surrounded by a Cr-rich layer.
- Nodule itself is surrounded by Zr.
- Fe-Fe-Ni & Fe-Sn phases have a lustrous metallic appearance.
TMI -2 Sample examination: - Debris

Macroscopic photo of various debris pieces (approx. 5x)

Debris H8-7-5-1 located on the upper agglomerate (40x)

Note variety of debris particles. Particles have different origins and histories, and have suffered different temperatures.

White pieces are UO₂

banded structure is zircaloy interacted with steels or Ni-based alloys

Long grey piece is zircaloy-UO₂ mixture
TMI-2 Ellingham Diagram of samples

Range of oxygen potential observed in core bore rocks & agglomerate zones

This gives an indication of the conditions (T and P_{O_2} or P_{H_2}/P_{H_2O}) in the reactor for the central rocks and surrounding agglomerate zones

UKAEA estimates from fuel remnants
Conditions during accident

1) Max. Temperature
   - Edge of reactor  \( T < 800 \degree C \)
   - Agglomerate  \( T \approx 1500 \degree C \) (stainless st. mp)
   - fully molten core  \( T = 2000-2500C \)
     (some pure UO\textsubscript{2} seen \( T=2850C ? \))

2) Cool-down
   - slow (2-54 h)
   - Agglomerate- more rapid & variable
   - Edge of core - transient rise in temp.; only slight degradation

3) Oxygen potential during the accident is estimated at \(-150k\text{kJ/mol} \) \( (p_{H_2}/p_{H_2O} = 1) \) at 2000C to \(-510k\text{kJ/mol} O_2 \) \( (p_{H_2}/p_{H_2O} = 10^6) \) for 1200C. Suggests high H\textsubscript{2} presence could be possible at times.
Phases formed
Core  - a $\text{UO}_2$ fuel & Zry cladding melt that oxidised in steam* and formed a U,Zr-containing oxide The core also contained small amounts of Fe,Ni,Cr oxides & Ag nodules.

Agglomerate - mixed metallic and ceramic phases from fuel/cladding/structure interactions (often incomplete) eg. $(\text{U,Zr})\text{O}_2$ phases, $(\text{Fe,Ni})$-Zr-U oxides, Ni-Fe-Sn metal, Ni,Fe partially oxidised nodules, & Ag metal nodules

* generating $\text{H}_2$
Phébus FP (fission product) with a driver core, test bundle and vertical line leading to the simulated primary circuit. The circuit has a horizontal line with a simulated steam generator before the airborne fission products pass into a containment tank & a sump to collect liquids.
Degradation results common to all 3 tests:

a) an heavily-oxidised upper bundle
b) melting of hot central zone & its relocation to produce:
c) a corium pool at quarter-height
Phébus FP project

Upper Phébus bundle (FPT1)

Micrograph of degrading irradiated fuel at the +607mm height of the FPT1 bundle - lenticular porosity due to fission gas bubble movement in overheated fuel.

Micrograph of fully oxidised cladding from a fuel rod at +607 mm height of FPT1 bundle.

Note heavily oxidised cladding and degraded fuel, in semi-liquified condition.
Corium Pool PIE

Comparison of the corium pool of FPT1 bundle at +228mm height (from bundle bottom) with the tomography for the same position.

Corium pools compositions of FPT0, FPT1 & FPT2: U/Zr atomic ratio = 1.06 to 1.44

TMI-2 core (G12-P 9-B) U/Zr estd. at 1.18

- Non-destructive tomography is very accurate
- Corium pool composition reasonably consistent between Phébus tests and TMI-2 data
- Note the outer crust around the central pool
Conclusions

• Examination of samples in TMI-2 investigation have enabled major advances in understanding of reactor degradation mechanisms.

• Assessing the compounds metals/oxides formed has enabled the mechanisms of degradation and conditions of formation to be proposed (eg. temperatures reached; O₂ potential/steam content/ H₂ vol. estimates).

• Techniques started with simple measurements (eg. density, porosity & g-spec) on all samples, detailed compositional analysis by SEM/TEM/EDS/WDX and then chemical analyses was performed on selected samples.

• X-ray ( & γ–spec.) tomographic techniques are now very accurate & powerful non-destructive techniques. They continue to advance.
Conclusions (contd.)/ Outlook

• Later research (eg. Phébus PF project) has improved bundle degradation mechanism & PF release knowledge.
  - Reproducible corium pool geometry was demonstrated
  - low-melting point interactions of Zry cladding with reactor steels that attack fuel & cause relocation at low temps.(~1200°C) was confirmed.
  - $\text{B}_4\text{C}$ moderator can interact with Zry and steel very rapidly > 1200°C to aid degradation effects.
  - $\text{B}_4\text{C}$ interacts with fission products (I,Cs) and affects their release to the exterior.

• Detailed geometry effects (eg. BWR/VVER lower head) still need research.

• Small scale (or inactive) tests are also important to investigate particular effects or mechanisms

• Modelling and simulation techniques are very important to understand materials behavior in all severe accidents.
Many thanks for your attention!

Selected publications/further references

- Metallurgical examination of bore samples from the three mile island unit 2 reactor core, PDW Bottomley & M. Coquerelle, Nuclear Technology, 1989 Aug; 87; 120-136.
- TMI-2 Examination results from the OECD-CSNI Program- Vols 1 & 2, D. Akers, G: Bart, P. Bottomley et. al., NEA/CSNI/ R(91) 9, April 1992 (EGG-OECD-9168)

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