EXAMINATION AND EVALUATION OF DEBRIS FROM THREE-MILE ISLAND UNIT-2 (TMI-2)

presented by P. D. W. Bottomley (JAEA Invited Researcher)

*formerly Sector Head, European Commission DG JRC-Karlsruhe*  
*Directorate G - Nuclear Safety & Security*  
*Unit G.III.8 - Waste Management*  
*PO Box 2340, Hermann-von-Helmholtz Pl. 1, 76125 Karlsruhe, Germany*  
*dboksb3@gmail.com*
Outline of Presentation

• Introduction to JRC nuclear work programme
• TMI-2 Accident & Sample Investigation Project
• Other fuel debris samples
  • a) Phébus pf bundle PIE
  • b) Chernobyl samples
• Conclusions
The European Commission’s science and knowledge service

Joint Research Centre

JRC Karlsruhe Site = Unit G Nuclear Safety & Security

Unit G.III Nuclear decommissioning = Ka site + units in Ispra (HoD VVR)

Unit G.III.8 Waste Management ex-Hot Cells (HoU JS)

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The mission of JRC-Karlsruhe is to provide the scientific foundation for the protection of the European citizen against risks associated with the handling and storage of highly radioactive material.

JRC-Karlsruhe’s prime objectives are
• to serve as a reference centre for basic actinide research,
• to contribute to an effective safety and safeguards system for the nuclear fuel cycle, and
• to study technological and medical applications of transuranium elements.
Hot Cell facilities at JRC-Karlsruhe

- 24 hot cells (licensed capacity $10^6$ Ci = $3.7 \times 10^{16}$ Bq)
- ~160 kg of irradiated fuel (80 LWR fuel rods) and 3.5 kg Pu
- shielded SEM, OM, EPMA, SIMS, XRD
- infrastructure: supporting workshop incl. manipulators maintenance
- 3 hot labs for characterization of non-irradiated materials
Nuclear fuel studies at JRC-Karlsruhe

Topics
- thermal transport
- thermodynamics (specific heat, vapor pressure, melting point)
- matter transport ($f_g$ behavior, actinides, $f_p$)
- radiation damage: mechanisms and effects
- rim structure formation, evolution
- property changes
- microstructure characterization
- corrosion, creep

Tools
- density
- optical, electron microscopy, XRD
- EPMA, SIMS, TEM-SEM
- laserflash, POLARIS
- high T laser-heating (melting, vaporization, conductivity, high-P)
- Knudsen cell, hot cell annealing
- fuel rod (profilometry, radiography, spectroscopy, puncture)
- cladding behaviour (creep)
- (hot) indentation
- chemical analysis
- leaching, electrochemistry
- reprocessing/partitioning studies

multidisciplinary approach

analytical/modeling tools
TMI-unit 2 (TMI-2) underwent a prolonged Loss of Coolant Accident (LOCA) on March 28th 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.

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, Argonne National Lab, FZK-Karlsruhe, JRC-ITU Karlsruhe, 
PSI Würenlingen, Studsvik, CE A Saclay, AEA Windscale

as well as JAERI Japan.

The principal aims of the examination were to establish:

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

* from Zircaloy steam oxidation
TMI-2 Examination: Positions of ITU samples

Damaged core of the TMI-unit 2 reactor in its end-state

- Fuel rod remnant: C7 3-35
- Debris samples: H8 7.2-7.9
- Upper crust: D8-P2,3
- Lower crust: N5-P1-E O7-P4

1) molten core (yellow), 2) fused crust/agglomerate (orange), 3) loose debris above (red) – location of powder & loose fuel remnants.

Green labels show samples received by JRC-Karlsruhe under the OECD TMI-2 sample analysis project.

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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 ZrO$_2$ oxide layer present with $\alpha$-Zr(O) containing layers beneath.

Thin external oxide layers indicate only slight exposure to increased temperatures. (~800°C)
G12-P9-E Fracture Surface -SEM
a) & b) show dense (U-rich) phases (white) and lighter (Zr-rich) oxide phases (dark)

c) Many fine metallic Ag precipitates on the surface.

Fe-rich oxide phases also present.

Core bore rock was principally oxidic
Core bore rock G12-P9-B
Back-scattered image SEM image showing 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
G12-P10-A fracture surface a) secondary & b) backscattered images. (2760x)

Note a) fine eutectic lamella approx. 0.5µm wide (to estimate cooling rate); b) small spheres are Ag spheres from Ag-In-Cd absorber

G12-P2-E (Mag. ~900x) 2-phase corium, lighter U-rich oxide & darker, Zr-rich oxide +Fe inclusion. Matrix composition was approx. equimolar (U,Zr)O₂
Agglomerate crust O7-P4-EA
Fe-Ni nodules with pure Fe metallic core (lighter centres) surrounded by Fe-Ni oxide crust (darker zones) due to preferential oxidation (BEI-1000x)
SEM image of an agglomerate sample from O7-P4-E zone of the lower crust.

- a Fe-Ni nodule is seen with a Ni-rich oxide layer on the outside
- example of preferential oxidation of the Ni (intermediate oxygen levels)

Backscattered electron SEM image of an upper crust (or agglomerate) sample D8 P3-A.

Elements of fuel, cladding & structural material are seen.
There are wide grain boundaries (containing eg. Zr & Fe) in U-rich grains. This shows gradual material interactions indication of lower temperatures than core.
D8-P3-A Agglomerate sample (190x) interference micro showing metallic and oxidic zones, both with secondary phases.

Indicates incomplete interactions - lower temperatures or lower durations at a temperature

2 phase metallic / oxidic zone

2 phase metallic zone

Oxidic zone with some secondary precipitates
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.

A metallic-dominated phase from a control rod $\rho \sim 6.7 \text{ g/cm}^3$ (compared to steel $\rho = 7.93 \text{ g/cm}^3$) implies it is steel dominated.
H8 7-3 from above the upper crust - BEI of a debris sample
- Note nodule with a 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.
Macroscopic photo of various debris pieces (approx. 5x)

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

White pieces are UO₂

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

Long grey piece is zircaloy-UO₂ mixture

Note variety of debris particles: unchanged UO₂ fuel particles & cladding, oxidised cladding fused cladding, steel-cladding interactions. Particles have different origins and histories, and have suffered different temperatures.
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.
Conditions during accident

1) Max. Temperature
- Edge of reactor \( T < 800 \, ^\circ \text{C} \)
- Agglomerate \( T \sim 1500 \, ^\circ \text{C} \) (stainless st. mp)
- fully molten core \( T = 2000-2500 \, ^\circ \text{C} \)
  (some pure \( \text{UO}_2 \) seen \( T=2850 \, ^\circ \text{C} \)?)

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

Phases formed
Core - a \( \text{UO}_2 \) fuel & Zry cladding melt that oxidised in steam to form H2 and:
  an \( \text{U,Zr} \)-containing oxide with a \( \text{U} \)-rich phase, a \( \text{Zr} \)-rich phase &
  smaller amounts of \( \text{Fe,Ni,Cr} \) oxides & Ag nodules

Agglomerate - mixed metallic and ceramic phases
  from fuel/cladding/structure interactions (often incomplete)
  eg. \( (\text{U,Zr})_2 \text{O}_2 \) phases, \( (\text{Fe,Ni}) \)-Zr-U oxides, \( \text{Ni-Fe-Sn} \) metal,
  \( \text{Ni,Fe} \) partially oxidised nodules, & some Ag metal nodules
1) Core bore rock were fully oxidic U,ZrO$_2$ with Fe-rich (with Cr, Ni) phases.

2) Al$_2$O$_3$ also seen from burnable (Gd$_2$O$_3$) poison rods.

3) Slightly superstoichiometric

4) Oxygen potential during the accident were estimated at -150kJ/mol ($p_{H_2}/p_{H_2O} = 1$) at 2000°C to -510kJ/mol O$_2$ ($p_{H_2}/p_{H_2O} = 10^6$) for 1200°C temperature. Suggests high H$_2$ presence could be possible at times.

5) Core centre probably cooled down in 4-50 hours (estd. from various lamellae thickness in structure).

6) Agglomerates were mixed cladding/fuel/structural materials debris in partially oxidic & metallic form (Fe-Ni-Sn metal, (Fe,Ni)-Zr-U oxides, partially oxidised Ni,Fe nodules, metallic Ag nodules)
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.
Comparative Bundle Post irradiation Examination (PIE)

X-ray tomographic scans produced by IRSN directly after testing:
- a) FPT0 (trace (8day) irradiated fuel),
- b) FPT1 (2yr irradiated fuel) & steam flow
- c) FPT2 (low steam flow- reducing conditions).

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.

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

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

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Lava samples from Chernobyl reactor unit 2 at ITU

Part of a collaboration of JRC-Ka with Khlopin Institute, St. Petersburg (P. Pöml/B. Burakov)

Sample of brown lava and SEM micrograph of crystal structures found in this lava sample. The highly dense (white) phases are U-rich fuel particles in a poly-phase matrix.
Lava samples from Chernobyl reactor unit 2 at ITU

- X-ray image mapping shows the layered compositions during slow cooling to form a U-rich centre and Zr & Si-rich outer edge.
- This can be used to determine its temperature as it solidified. This data can be used to validate lava flow models and to understand the conditions of the accident.

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Conclusions

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

- Techniques include simple density & porosity measurements for preliminary characterization/sorting of samples, detailed compositional analysis (by SEM/TEM/EDS/WDX) and crystallographic determination (XRD)

- Assessing the compounds metals/oxides formed has enabled the possible interactions, and mechanisms of degradation to be proposed; the conditions of formation (eg. temperatures reached; O₂ potential/steam content); also estimates of H₂ formation can be made.
Conclusions (contd.)/ Outlook

• Later research (eg. Phébus PF project & Chernobyl but also elsewhere) has improved bundle degradation & PF release knowledge.
  - Reproducible corium pool geometry was demonstrated
  - Zry cladding interactions with reactor steels etc. (giving low-melting mixtures) that attack fuel & cause relocation at low temps.(~1200°C) was confirmed.
  - Detailed geometry effects (eg BWR/VVER design) & other materials (eg B₄C moderator) still need research.

• X-ray tomographic techniques have been very accurate and are powerful non-destructive techniques (other NDT techniques eg. γ-spectroscopy continue to advance)

• Modelling and simulation techniques are also a very important to understand reactor accidents and materials behaviour modelling.

• Current research is more oriented on understanding fission product behaviour, Corium interactions with reactor vessel and its retention & Corium interactions with concrete containment.