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

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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

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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)

NDF Forum 01.07.2017

EURATOM Work Programme at the JRC (ie. Nuclear WP of JRC)

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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

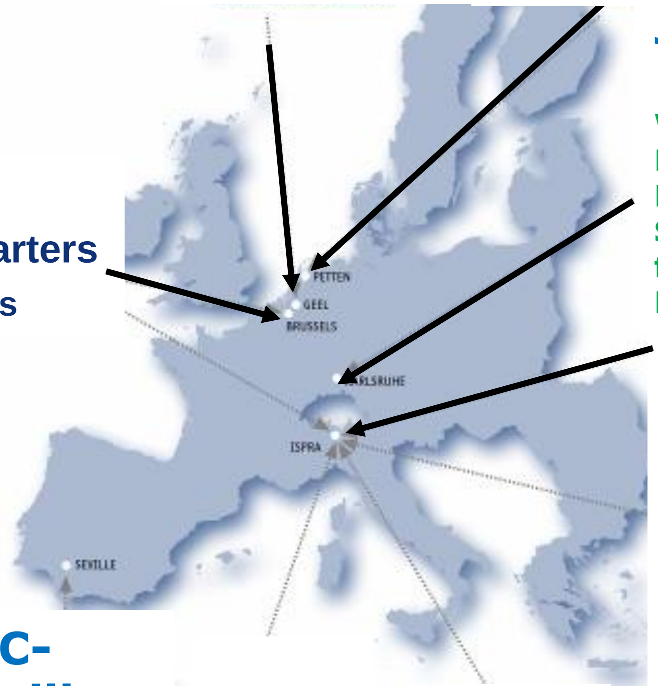
- *to serve as a reference centre for basic actinide research,*
- *to contribute to an effective safety and safeguards system for the nuclear fuel cycle, and*
- *to study technological and medical applications of transuranium elements.*

JRC-Geel
Standards for Nuclear Safety

JRC-Petten
Knowlege for Nucl. Safety, Nucl. Reactor Safety & Emergency Preparedness

JRC-Karlsruhe,
Waste Management Remediation, Nuclear Fuel Safety, Nuclear Safety, Safeguards & forensics, Advanced Nucl. Knowledge

JRC-Headquarters - Brussels



JRC- Ispra
Nucl. Sec. &Safeguards Nuc. Dcommissioning, Nucl. Safety-Ispra

JRC-Seville
Prospective studies

JRC Sites

Non–nuclear insitutes

1) **Health & Consumer Protection,**
2) **Protection & Security of the Citzen,**
3) **Institute for Environment & Sustainibility**

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Hot Cell facilities at JRC-Karlsruhe

- 24 hot cells (licensed capacity 10^6 Ci = 3.7 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

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Topics

thermal transport

thermodynamics (specific heat, vapor pressure, melting point)

matter transport (*fg* behavior, actinides, *fp*)

radiation damage: mechanisms and effects

rim structure formation, evolution

property changes
microstructure characterization

corrosion, creep

LWR,
advanced reactors
HTR

high burnup UO_2
MOX
MX
inert matrices

cladding

steady state
off-normal, accident
storage

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

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Three Mile Island (TMI-2)

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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,
CEA 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/H_2 production*) prevailed**

→ **hence what degradation reactions were likely**

*** from Zircaloy steam oxidation**

TMI-2 Examination: Positions of ITU samples

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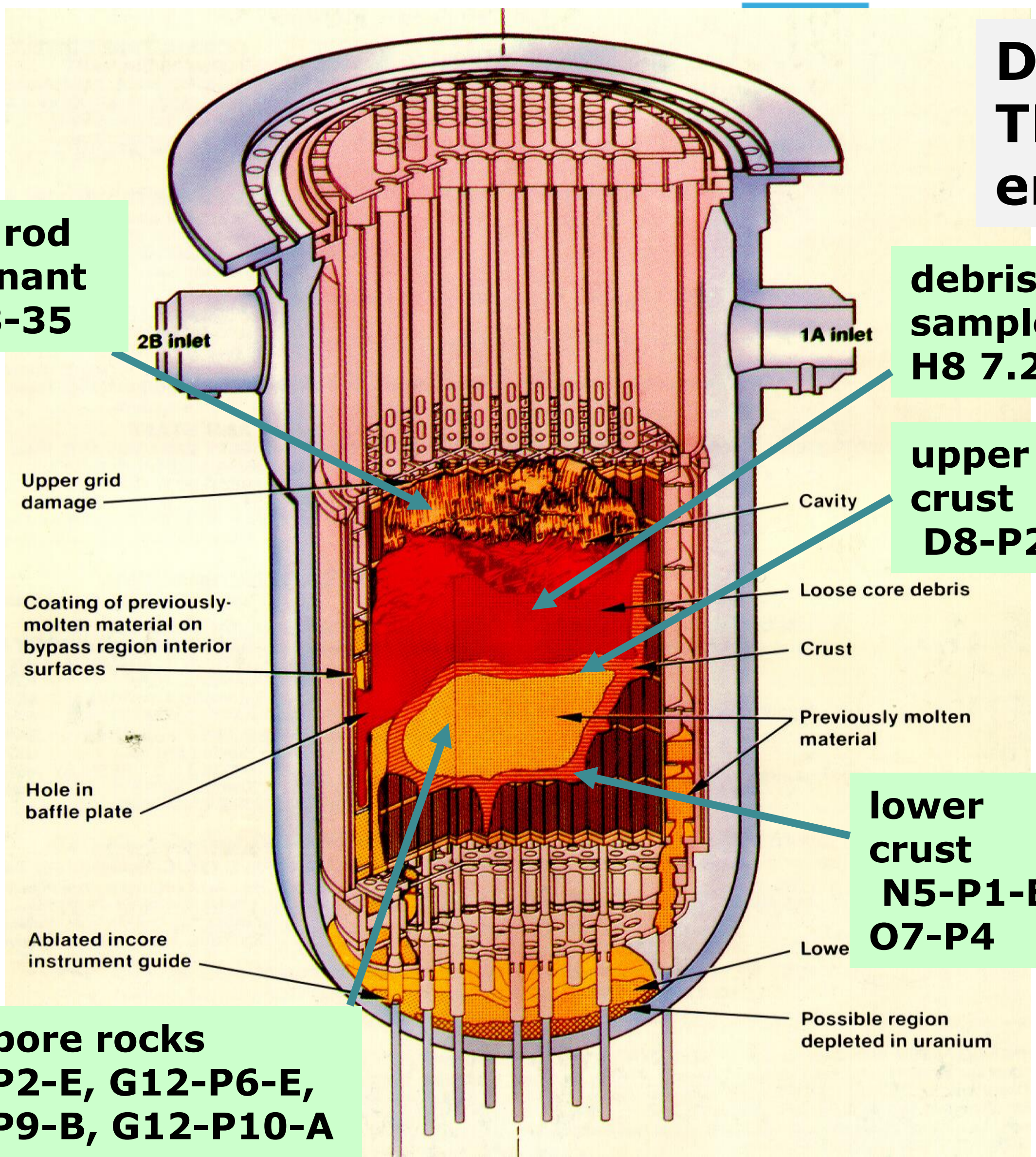

fuel rod remnant C7 3-35

debris samples H8 7.2-7.9

upper crust D8-P2,3

lower crust N5-P1-E O7-P4

core bore rocks G12-P2-E, G12-P6-E, G12-P9-B, G12-P10-A

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

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.

TMI-2 Examination: Debris lying on top of melted core

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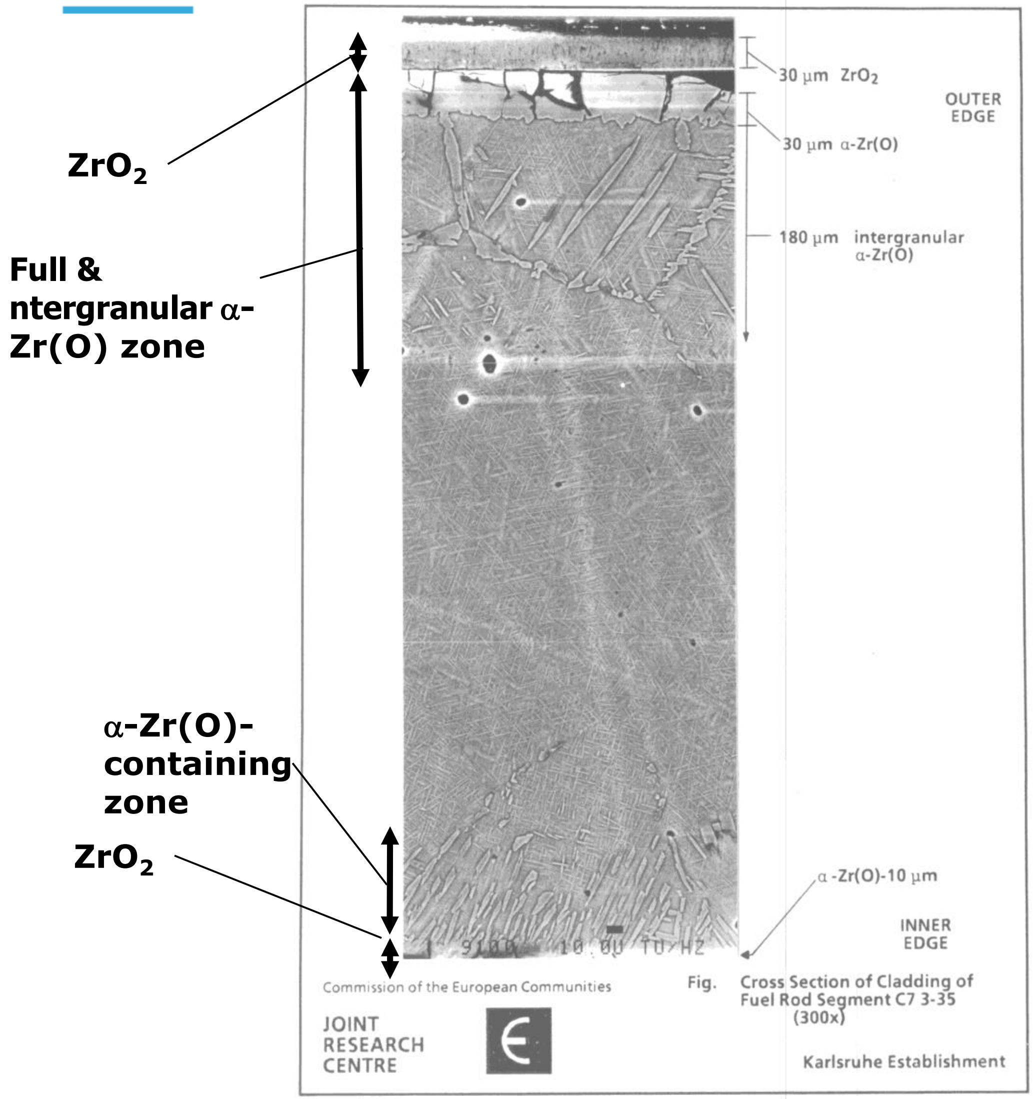
Cross-section of cladding of fuel rod remnant C7-3-35



C7-3-35 Fuel rod segment.
Note thin external ZrO_2 oxide layer present with α-Zr(O) containing layers beneath.

Thin external oxide layers indicate only slight exposure to increased temperatures. (~800C)

TMI-2 Examination: Core bore rock

Core bore rock was principally oxidic

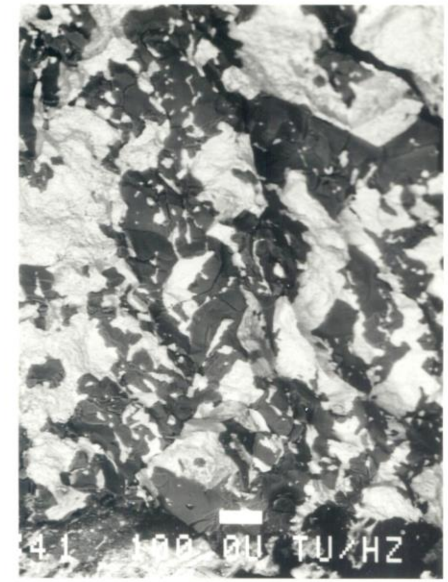
<u>G12-P9-E Fracture Surface -SEM</u>

a) & b) show dense (U-rich) phases (white) and lighter (Zr-rich) oxide phases (dark)

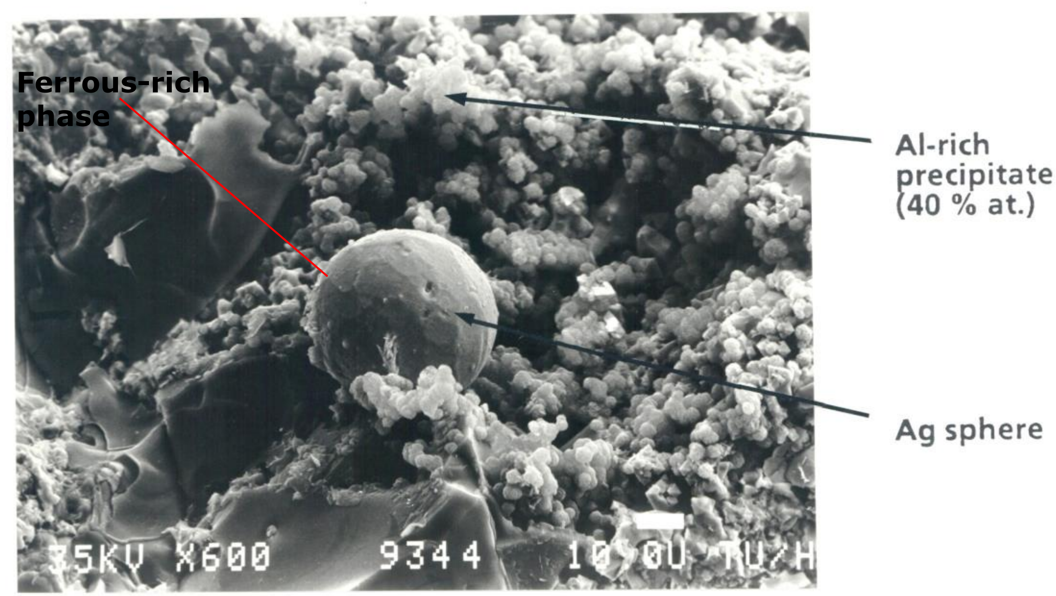
c) Many fine metallic Ag precipitates on the surface.

Fe-rich oxide phases also present.

a) Secondary Electron Image 60x mag



b) Back-scattered Electron Image 60x mag



c) Silver Sphere Precipitate 600x mag

TMI-2 Sample Examination
Core bore rock samples

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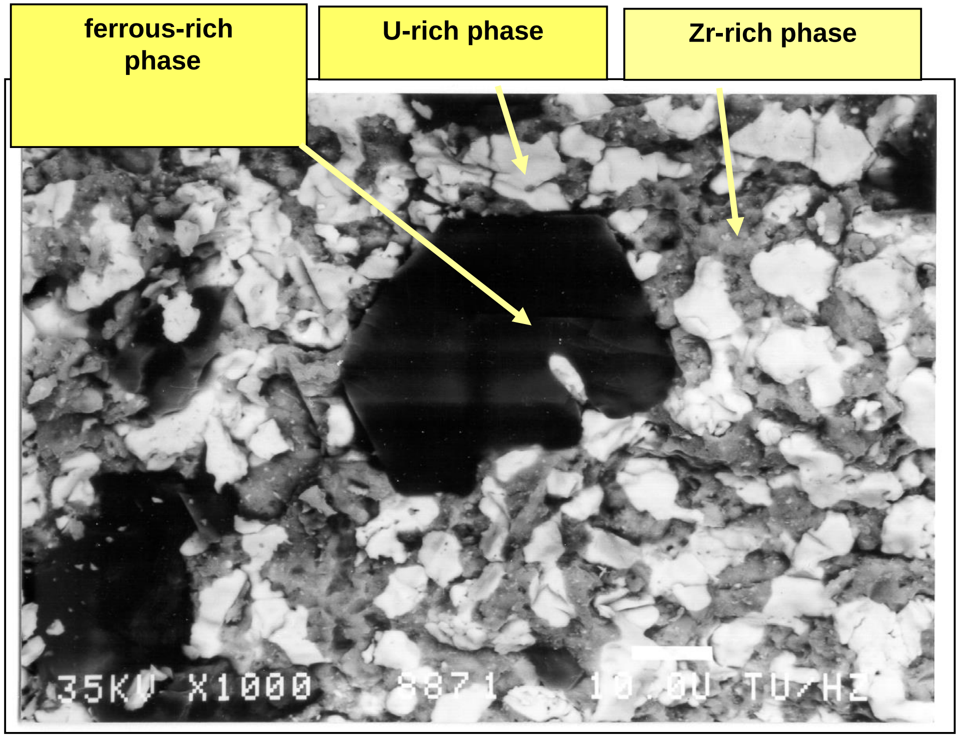
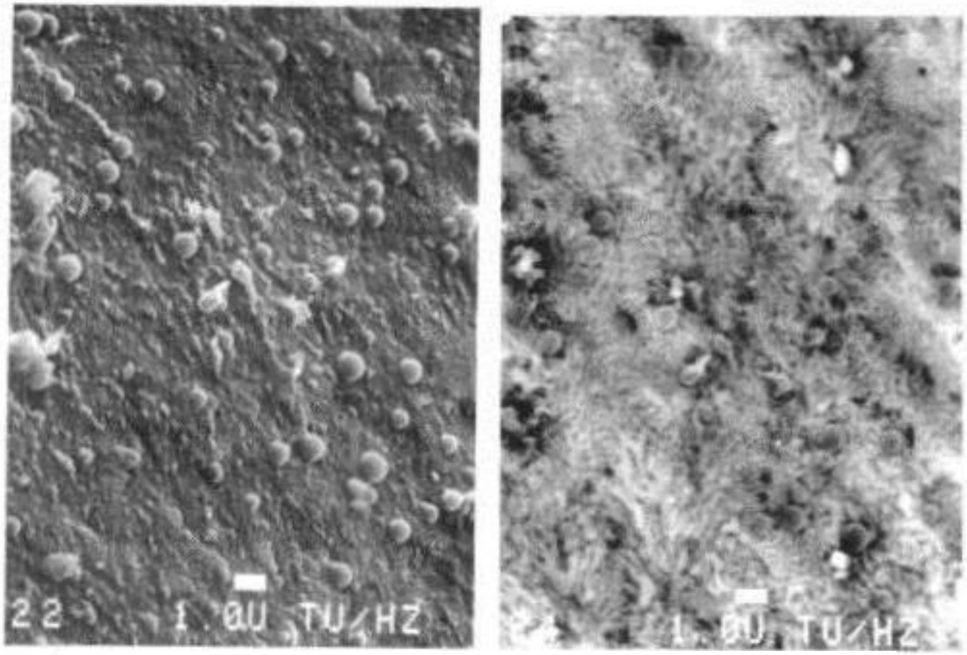
TMI-2 - example of Molten core – bored–out rock samples



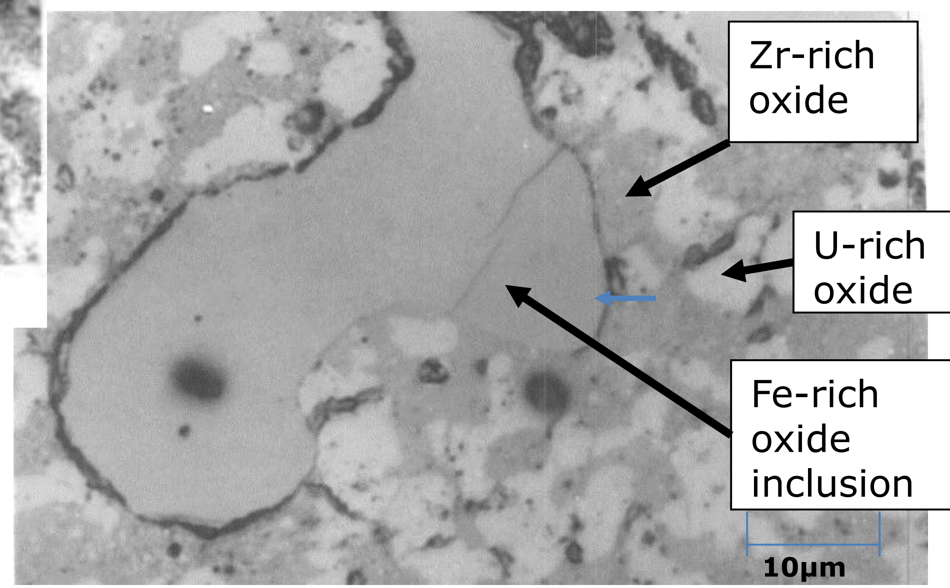
Core bore rock G12-P9-B
Back-scattered image SEM image showing 3 phases in the fully molten rock:
- **dense or white (U-rich) phases;**
- **less dense (grey) (Zr-rich)phases;**
- **light element (dark) Fe ,Ni, Cr-containing phases**

TMI -2 Examination Core Bore Sample Analysis



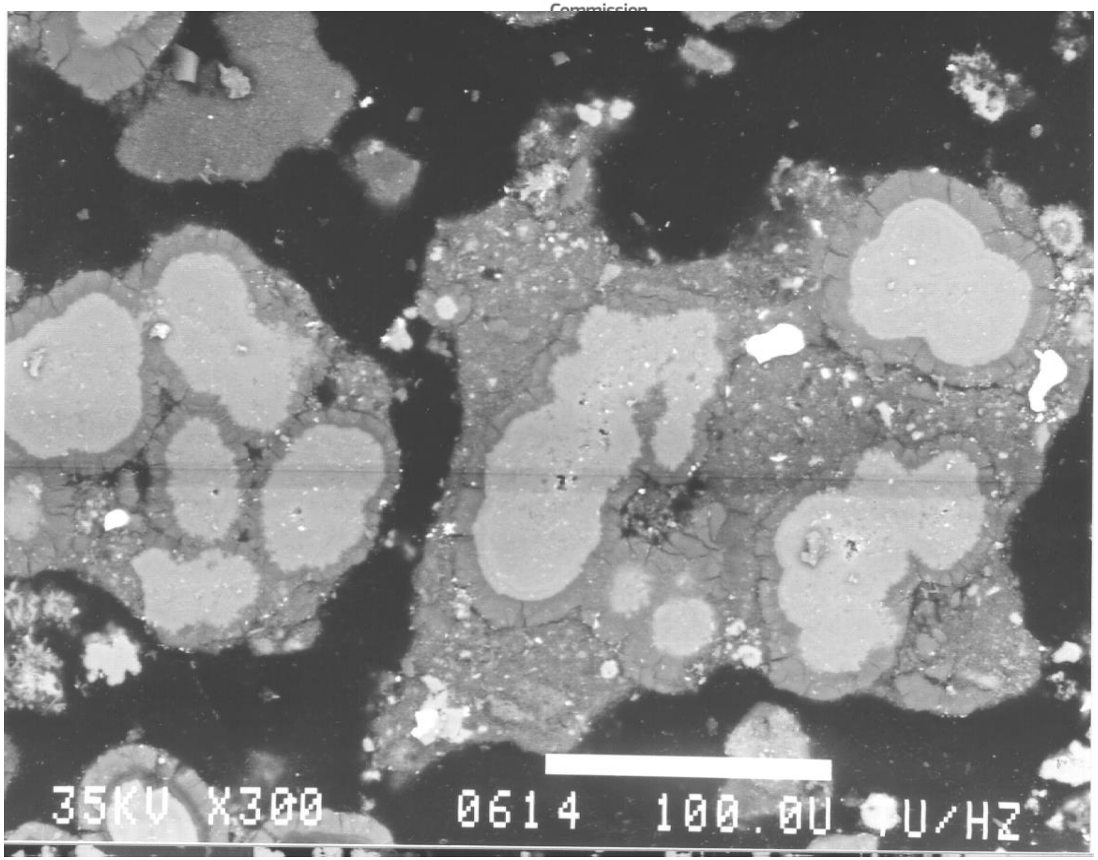
G12-P10-A fracture surface a) secondary & b) backscattered images. (2760x)

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 $(U,Zr)O_2$

TMI -2 Sample Examination: Agglomerates

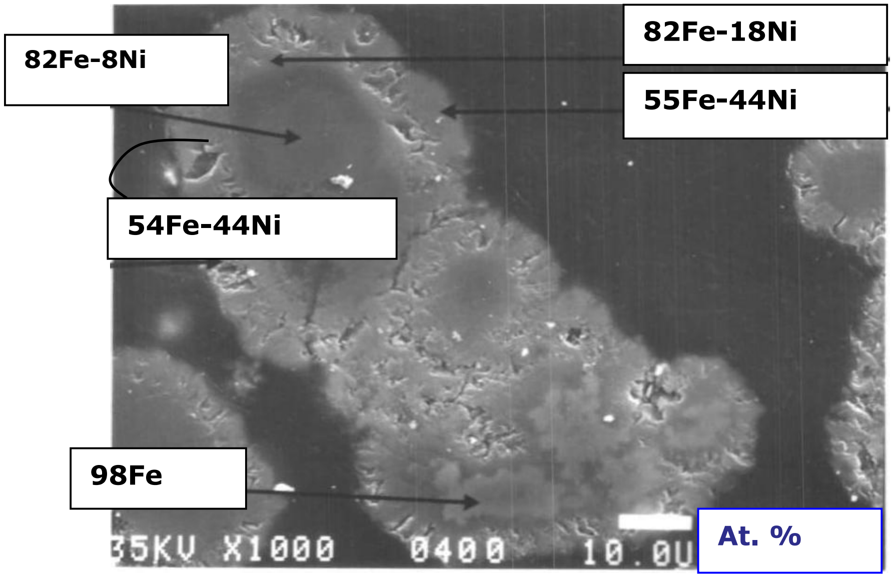


Agglomerate crust O7-P4-EA

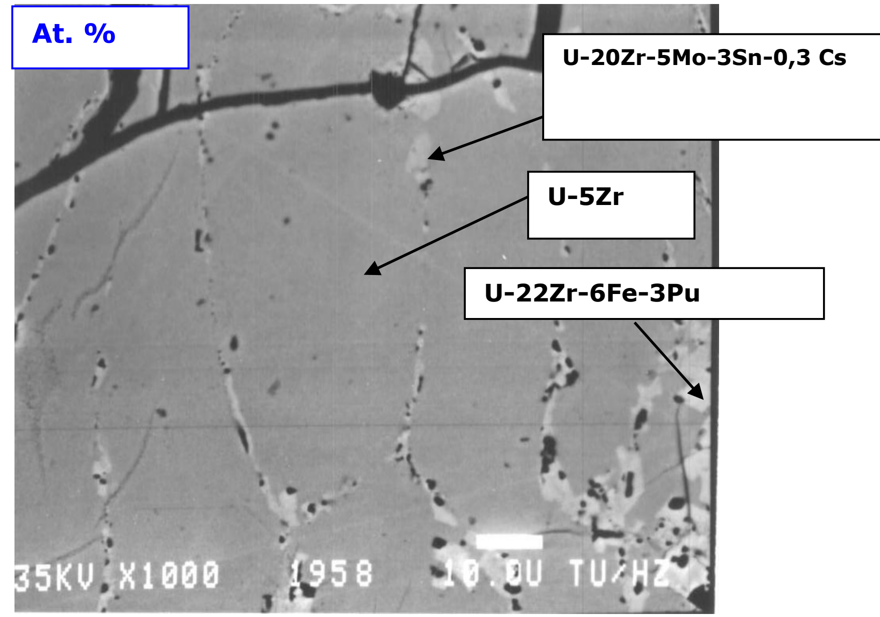
Fe-Ni nodules with pure Fe metallic core (lighter centres) surrounded by Fe-Ni oxide crust (darker zones) due to preferential oxidation (BEI-1000x)

TMI-2 Sample Examination Agglomerates

TMI-2 Crust Samples (Agglomerates)



SEM image of an agglomerate sample from O7-P4-E zone of the lower crust.



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

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

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

TMI-2 (Agglomerate material - interference macros)

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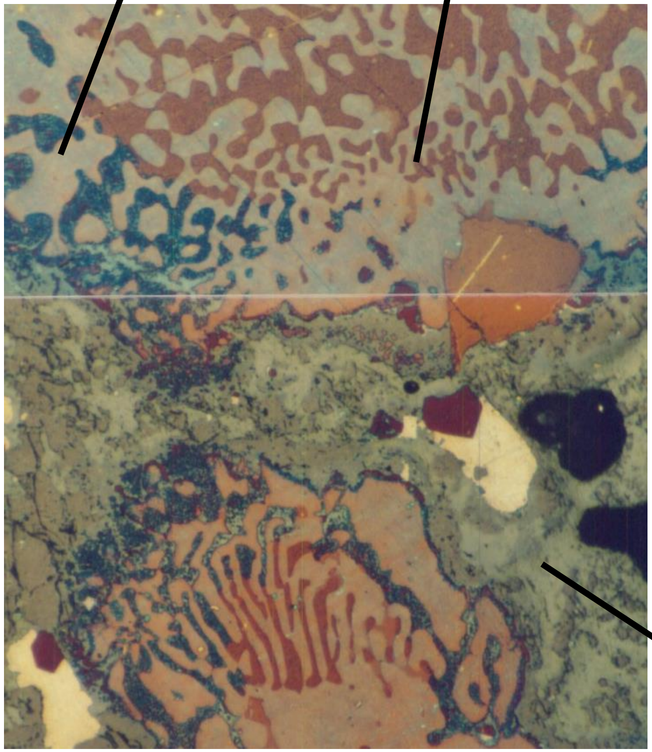
D8-P3-A Agglomerate sample (190x)
interference micro showing metallic and oxidic zones, both with secondary phases.

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



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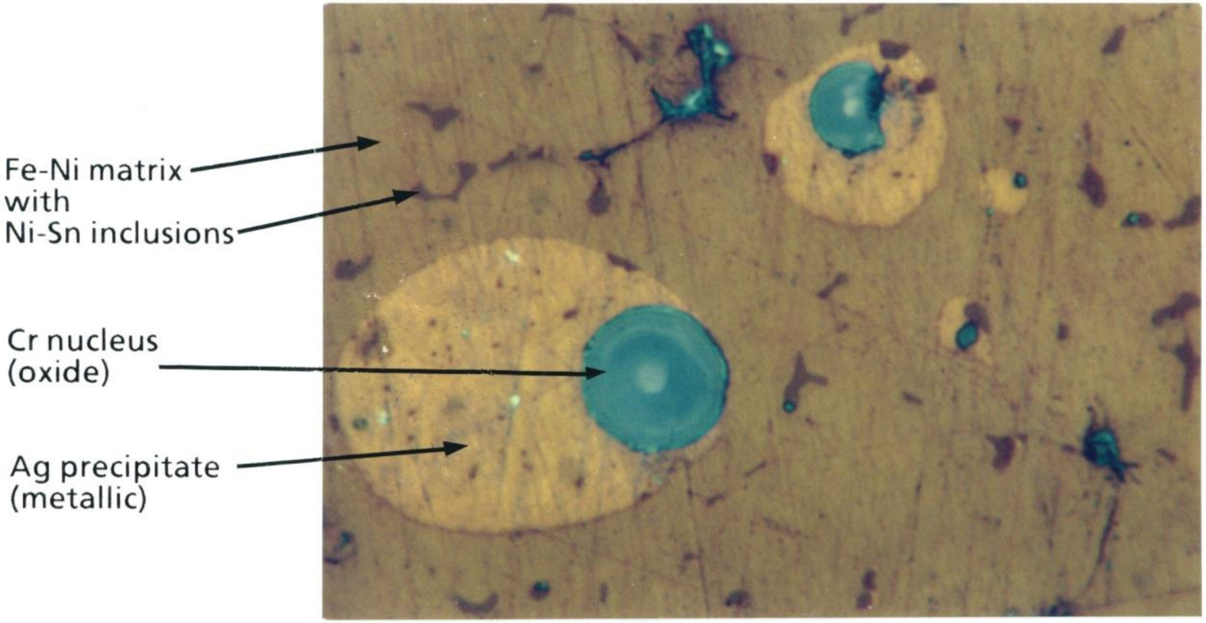
TMI-2 Agglomerate - metallic and oxidic phases

Example of partially oxidised material from agglomerate



b) Interference Micrograph (1033x)

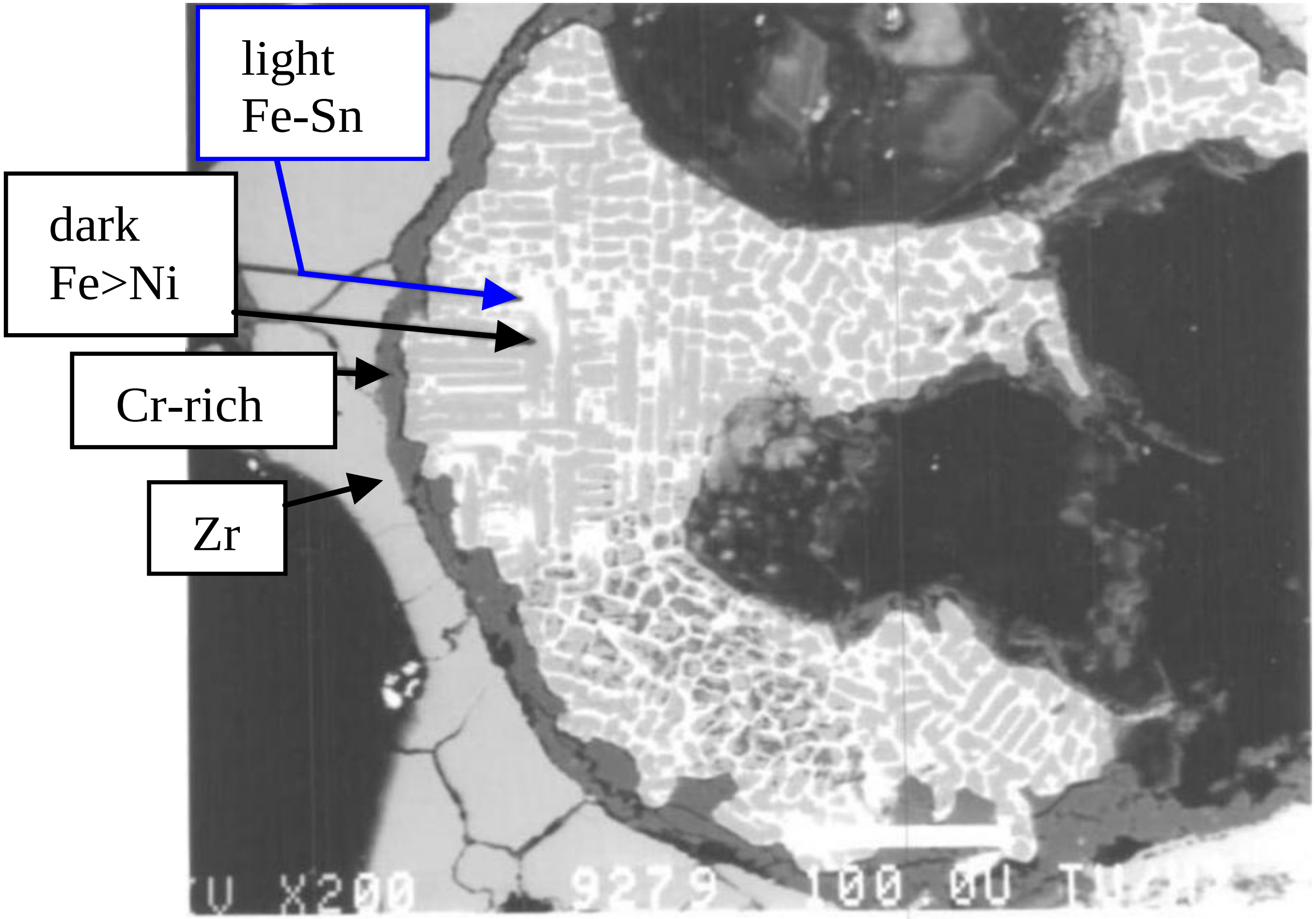
Agglomerate N5-P1-E (1033x).
(CuO vapour coating)
Stainless steel control rod cladding with Ag spheres that contain Cr oxide nuclei.

A metallic-dominated phase from a control rod
ρ ~6.7 g/cm^3 (compared to steel ρ= 7.93 g/cm^3) implies it is steel dominated.

TMI-2 Debris samples

H8 7-3 from above the upper crust <u>- BEI of a debris sample</u>

- **<u>Note nodule with a lamellar structure</u> 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 Analysis -Debris

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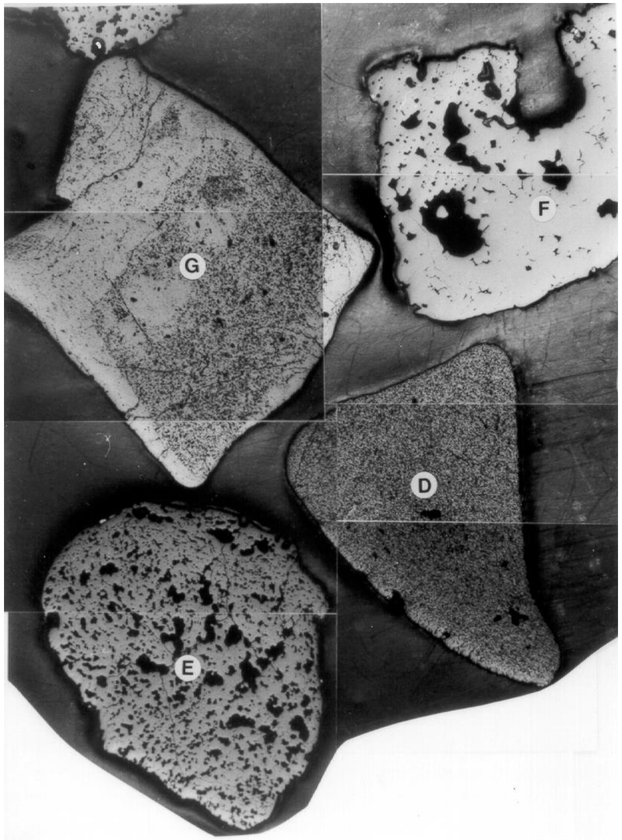
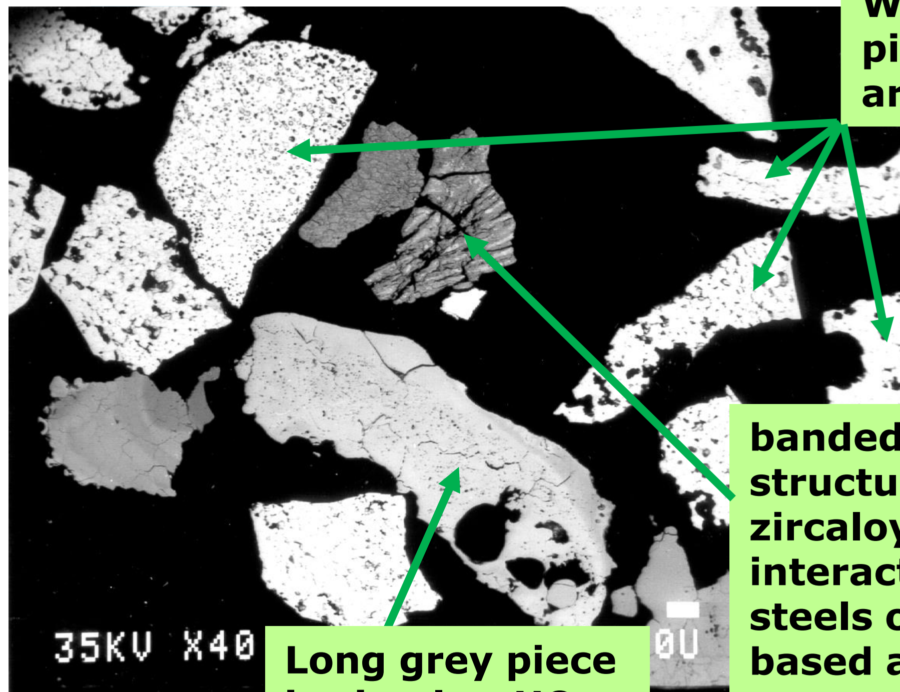


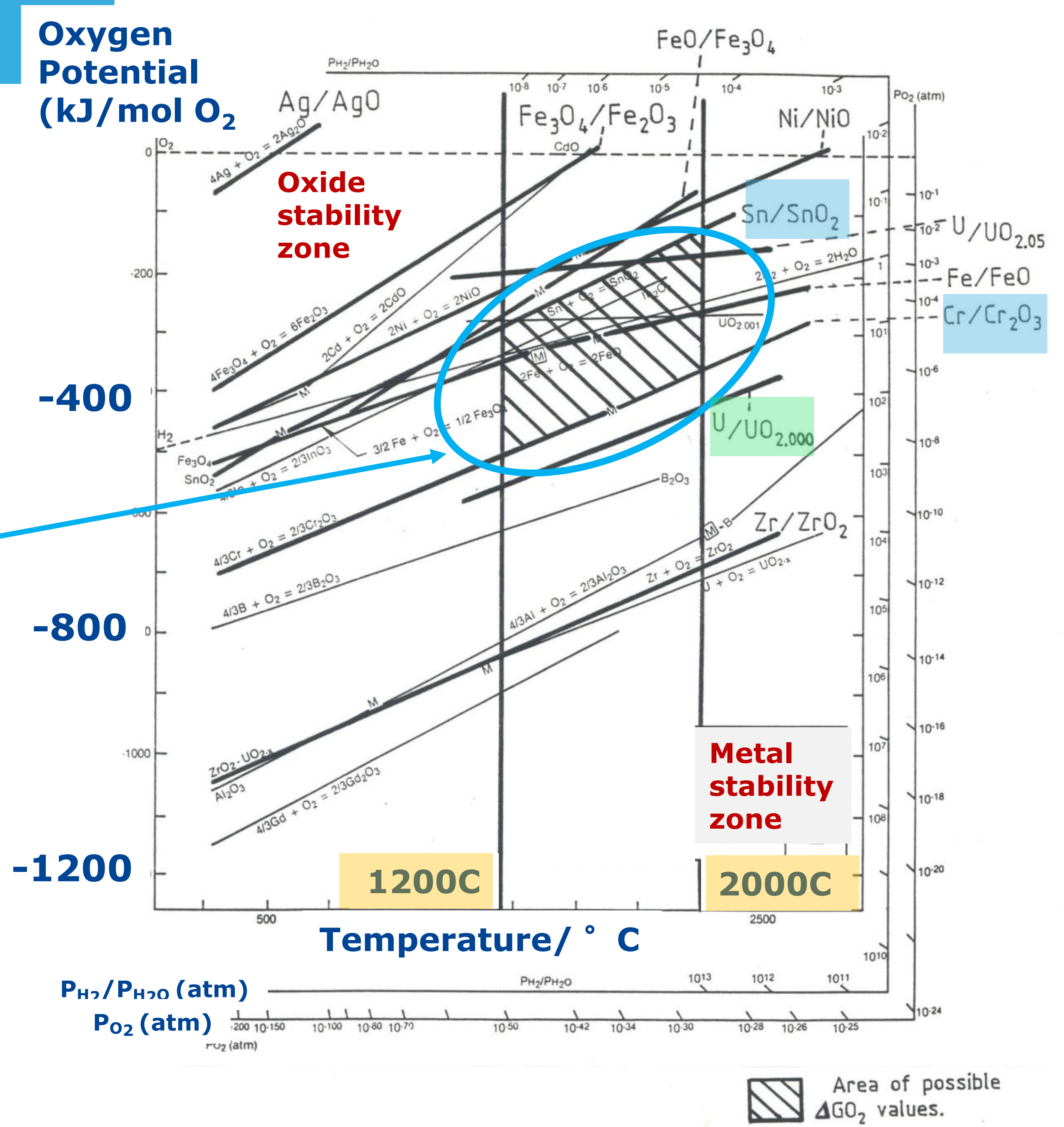
Macroscopic photo of various
debris pieces
(approx. 5x)



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

Note variety of debris particles: unchanged UO_2 fuel particles & cladding, oxidised cladding fused cladding, steel –cladding intractions. 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_{O2} or P_{H2}/P_{H2O}) in the reactor for the central rocks and surrounding agglomerate zones

TMI-2 Accident: Summary of conditions and temperatures experienced-1

Conditions during accident

1) Max. Temperature

- Edge of reactor T < 800° C
- Agglomerate T~1500° C (stainless st. mp)
- fully molten core T= 2000-2500C

(some pure UO_2 seen T=2850C?)

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 UO_2 fuel & Zry cladding melt that oxidised in steam to form H2 and: an U,Zr-containing oxide with a U-rich phase, a Zr-rich phase & smaller amounts of Fe,Ni,Cr oxides & Ag nodules

Agglomerate - mixed metallic and ceramic phases from fuel/cladding/structure interactions (often incomplete) eg. $(U,Zr)O_2$ phases, (Fe,Ni)-Zr-U oxides, Ni-Fe-Sn metal, Ni,Fe partially oxidised nodules, & some Ag metal nodules

TMI-2 Accident: Summary of conditions and temperatures experienced-2

1)Core bore rock were fully oxidic U,ZrO_2 with Fe-rich (with Cr, Ni) phases.

2)Al_2O_3 also seen from burnable (Gd_2O_3) poison rods.

3)Slightly superstoichiometric

4)Oxygen potential during the accident were estimated at -150kJ/mol (p_{H2}/p_{H2O} = 1) at 2000C to -510kJ/mol O_2 ($p_{H2}/p_{H2O} = 10^6$) for 1200C temperature. Suggests high H2 presence could be possible at times.

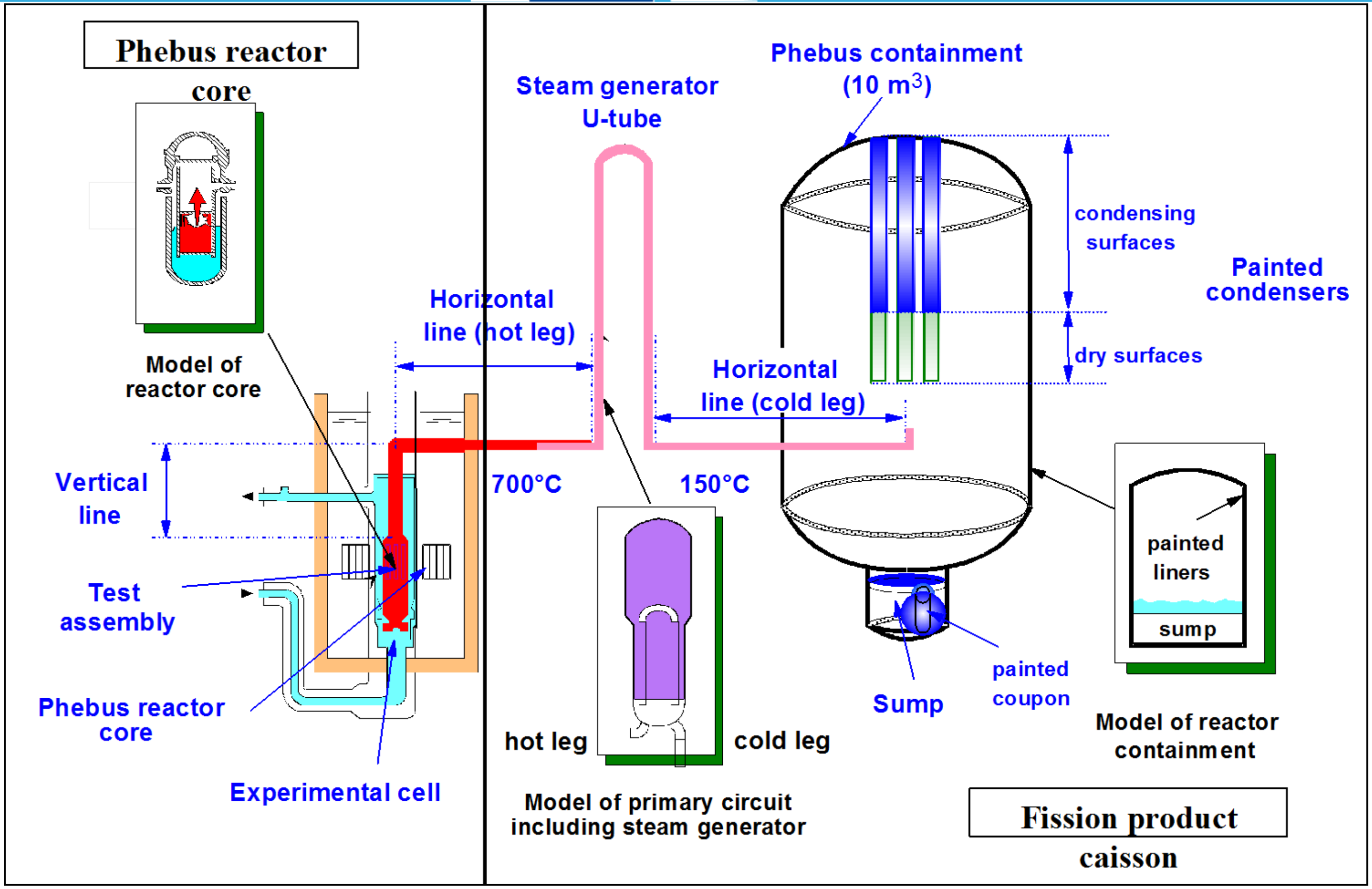
5) Core centre probably cooled down in 4-50 hours (estd. from various lamellae thickness in structure) .

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 project

lead by IRSN, Cadarache (with EC support) & participation of many EU national institutes)

Phébus PF Facility



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.

Phébus FP project

IRSN Cadarache Tomography

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

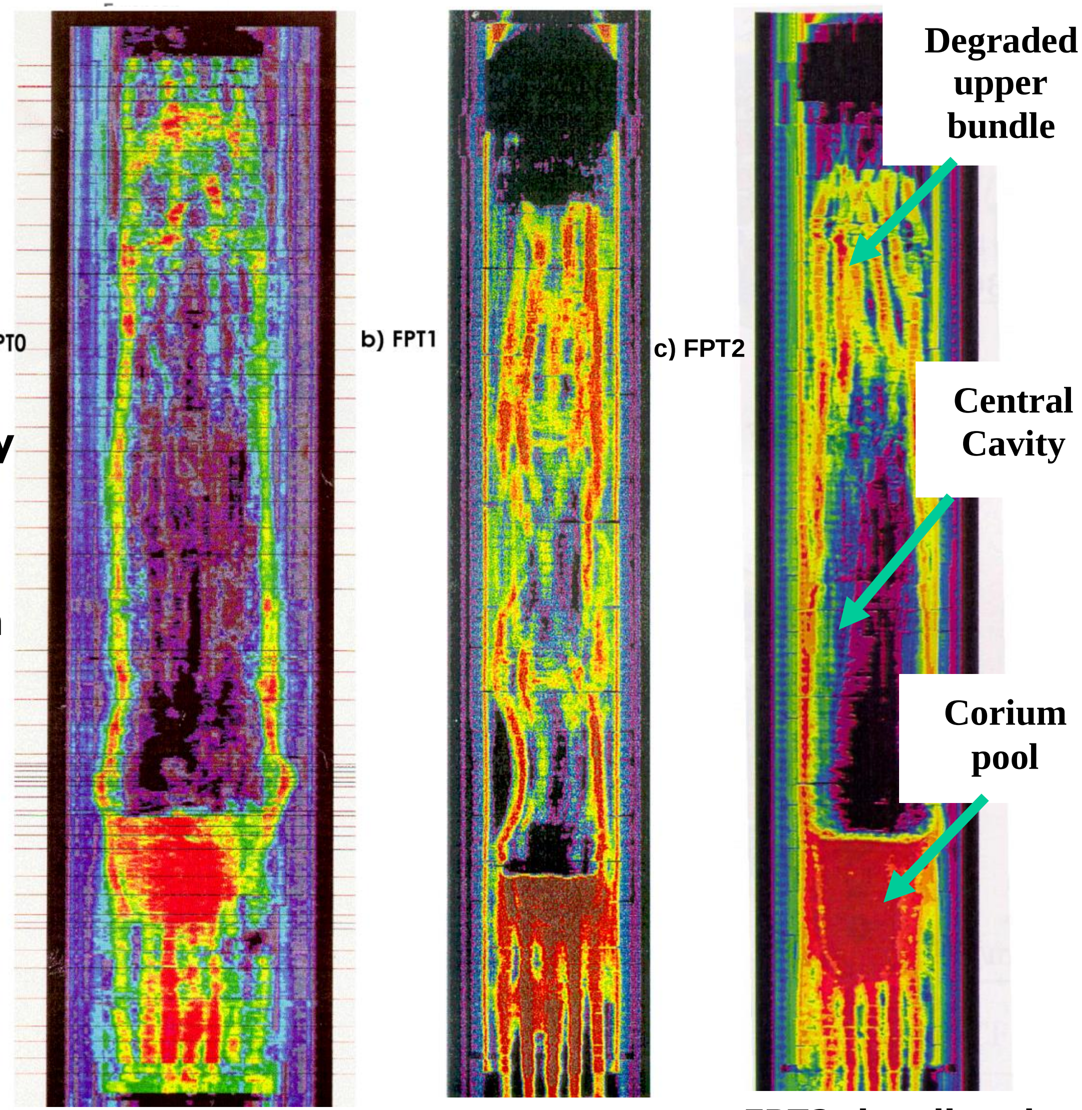
c) FPT2 (low steam flow- reducing conditions).

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 .



FPT0-trace irradiated fuel – full steam flow

FPT1- irradiated fuel –full steam flow

FPT2- irradiated fuel –low steam flow

Phébus FP project

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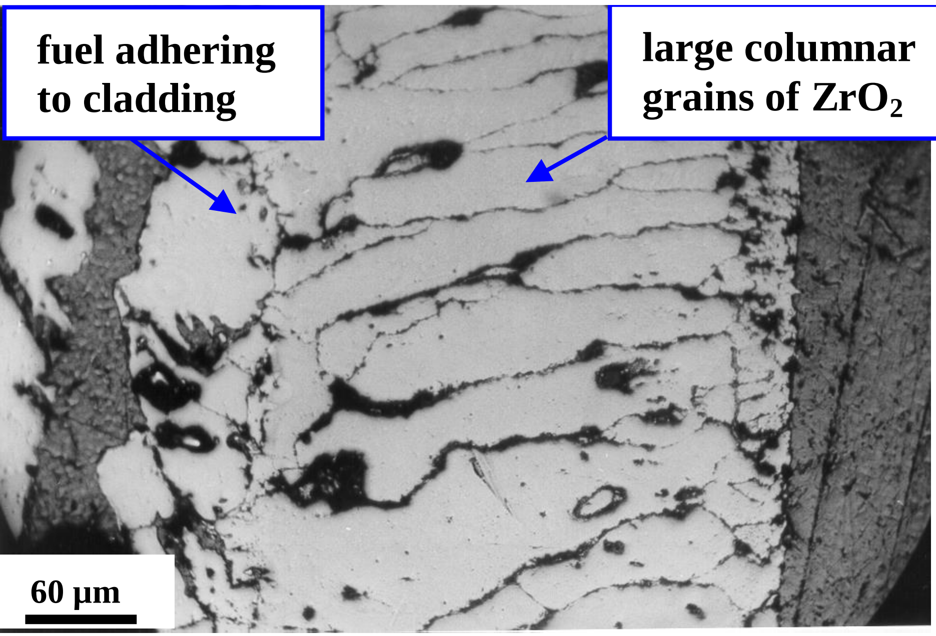
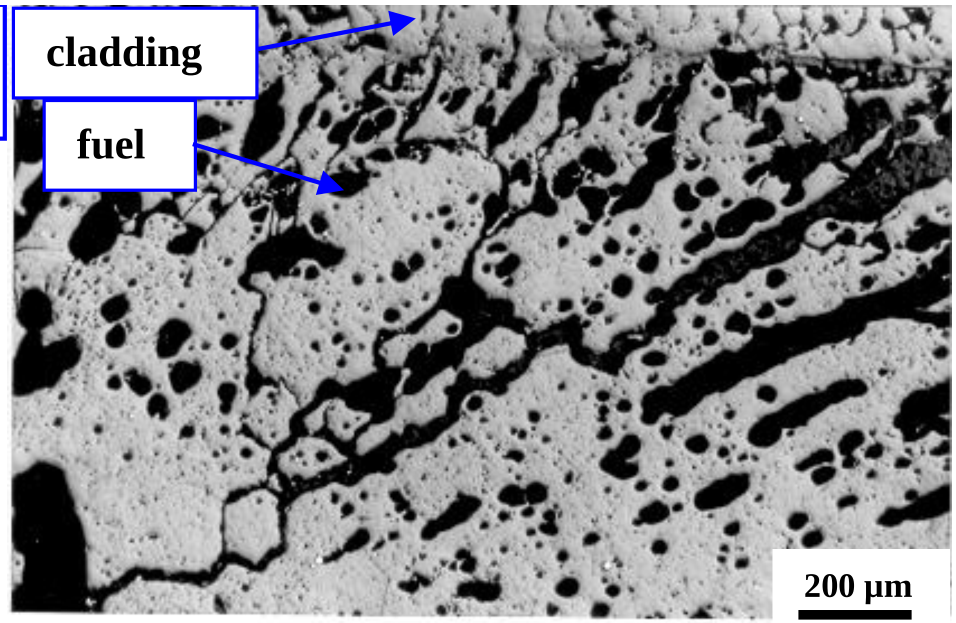
Upper Phébus bundle (FPT1)



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



Micrograph of degrading irradiated fuel at the +607mm 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

Phébus FP project

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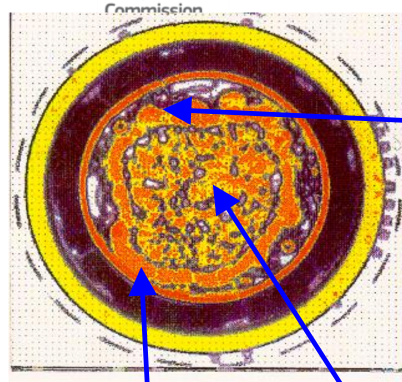
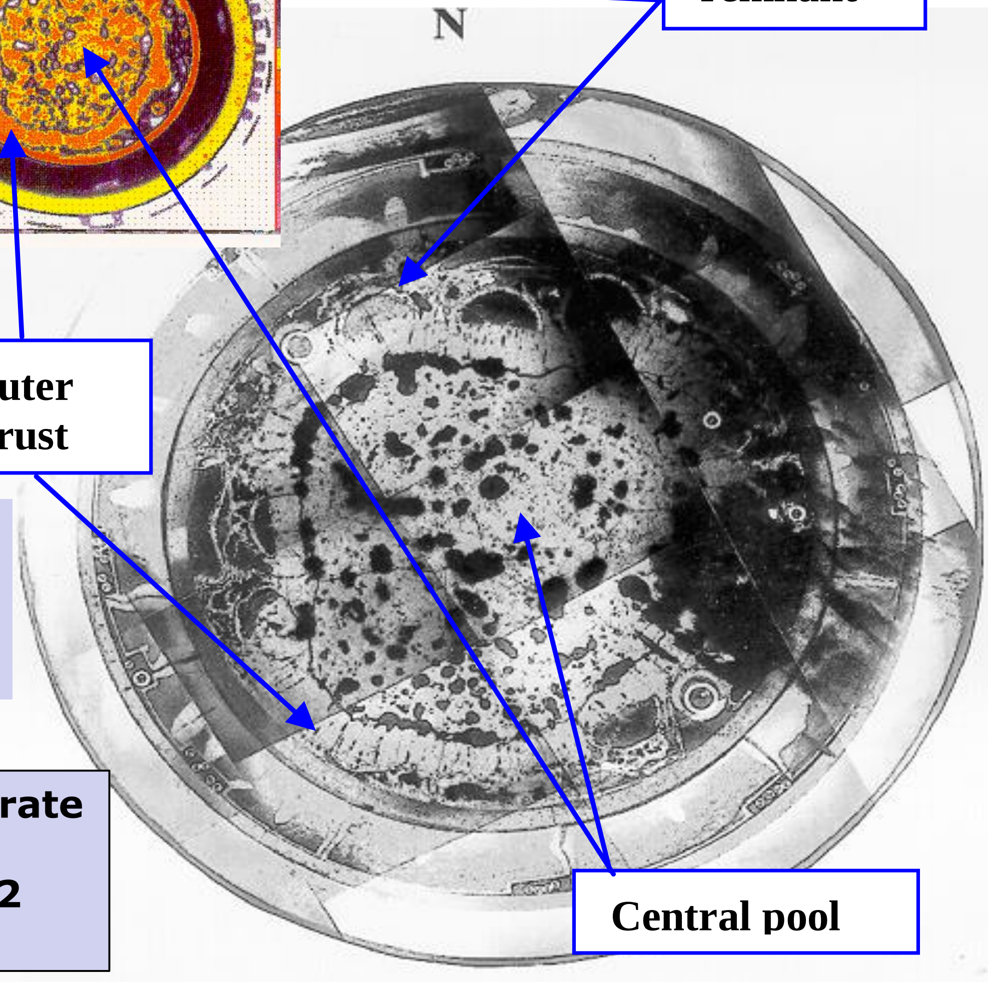
Corium Pool PIE

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





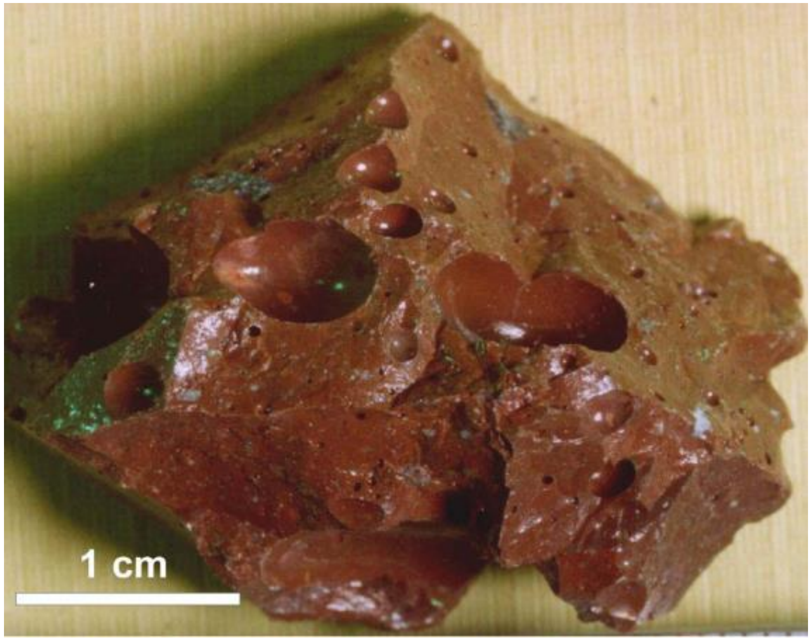
Corium pools compositions of FPT0, FPT1 & FPT2: U/Zr atomic ratio= 1.06 to 1.44

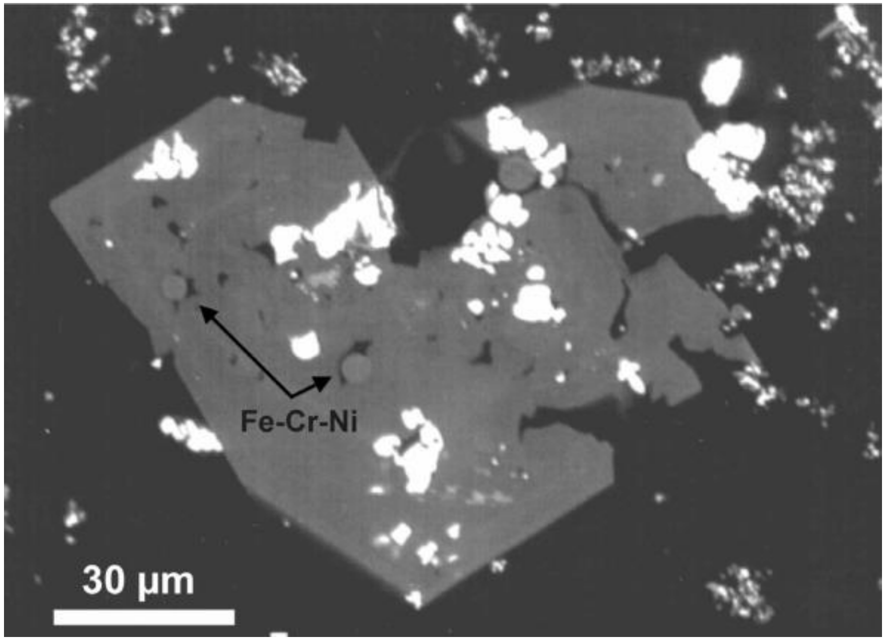
TMI-2 core (G12-P 9-B) U/Zr estd. at 1.18

- Non-destructive tomography is very accurate
- Corium pool composition reasonably consistent between Phébus tests and TMI-2 data

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)



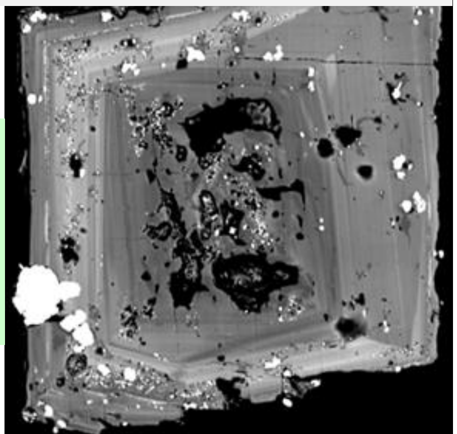


Sample of brown lava and SEM micrograph of crystal structures found in this lava sample. The highly dense (white) phases are U-rich fuel particles in a poly-phase matrix.

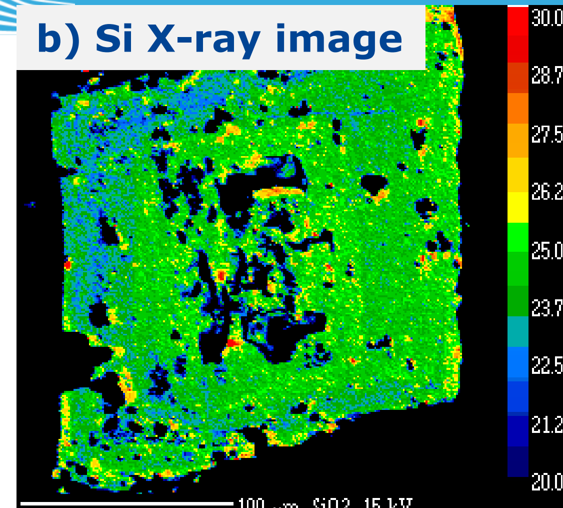
Lava samples from Chernobyl reactor unit 2 at ITU

Backscattered image of U zircon crystal with inclusions.

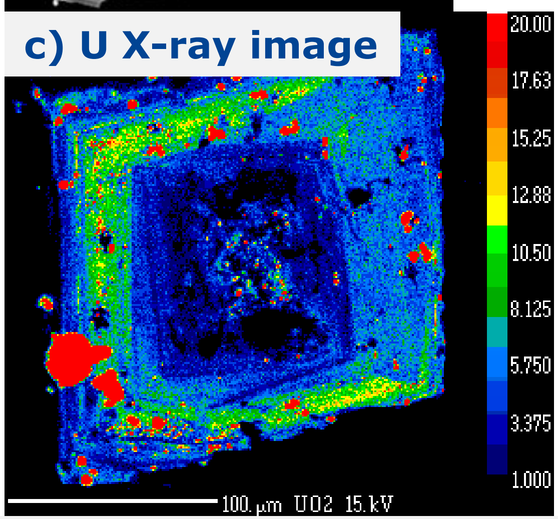
a) secondary electron image



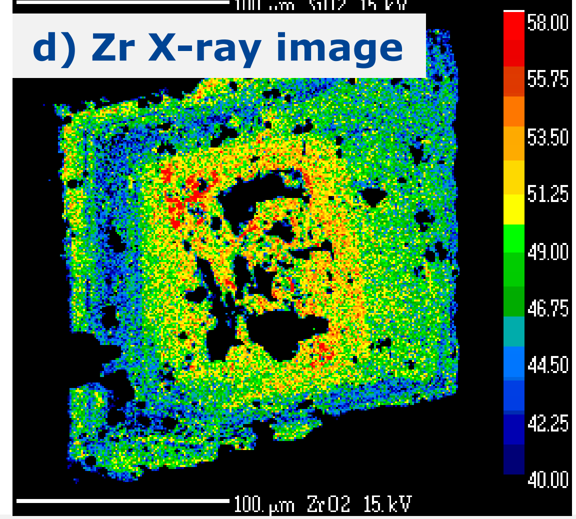
b) Si X-ray image



c) U X-ray image



d) Zr X-ray image



- **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**

Conclusions

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- **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; O_2 potential/steam content); also estimates of H_2 formation can be made.**

Conclusions (contd.)/ Outlook

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- **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.(~1200C) was confirmed.**
  - **Detailed geometry effects (eg BWR/VVER design) & other materials (eg B_4C 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.**