Status of testing of commercially available PVD coatings at the Halden Reactor Project

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CONTENT

Coating testing at IFE/Halden

- Introduction
- Small samples in BWR, PWR conditions (in-pile testing!)
- Small samples in Candu reactor conditions (out-of-pile)
- **Coated fuel rods (in-pile testing!)**
- Small samples in supercritical water

1. Coating testing at Halden - Introduction

- Work was initiated in **2009** in Halden in order to improve instruments and to provide protective coatings for fuel and materials Gen IV (liquid lead, supercritical water)
- Methodology: Use existing industrial coatings!
- Presentation at Enlarged Halden Program Group Meeting in Sandefjord, **2011**
  - "Coatings and plasma treatment for nuclear applications", R. Van Nieuwenhove, J. Balak and B. C. Oberländer, HWR-1028, December 2011
- Coatings can improve significantly the performance of fuel claddings in present day reactors (see next slide)
- After the Fukushima accident (11 March 2011), a lot of research on coatings started in many countries for the development of accident tolerant fuel (ATF).

Importance of coatings for normal operation in present-day reactors

- More demanding conditions
  - Longer cycles, higher burn-up
  - Higher initial Lithium hydroxide (LiOH)
  - More corrosive environment (temperature, chemical)
- Reduction of failures
  - Avoid fuel failures induced by fretting
  - Reduction of hydrogen uptake (and embrittlement)

Different approaches

- Evolutionary
  - Modified Zr-based alloys (Opt. ZIRLO, M5, MDA, NDA, HANA, etc.)
  - Surface modifications (such as ODS treatment) and coatings
- Revolutionary
  - SiC, Fe-Cr-Al, metal-ceramic hybrid cladding, Mo-based alloys (Accident Tolerant Fuel, Gen IV)

Selected coatings for testing (Halden)

Method: PVD (most uniform, free of pores, cheap)

Coating materials:

- TiAlN
- CrN
- AlCrN
- ZrO_2

Substrate:

- AISI 316L
- Inconel 600
- Zircaloy

Coating company used: Oerlikon Balzers

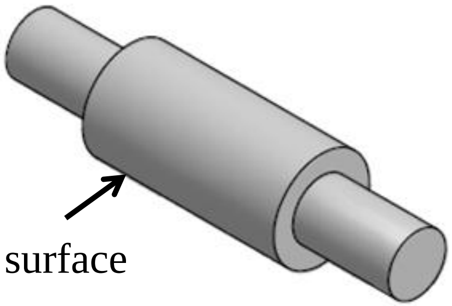
- 100 coating centres in 34 countries

BWR, PWR conditions (in-pile testing!)

Testing of various coatings (on small samples) has been performed in the Halden reactor

R. Van Nieuwenhove, IFA-774: The first in-pile test with coated fuel rods Enlarged Halden Program Group Meeting in Røros, 2014

In-pile tests (BWR and PWR)


Coated surface

Diameter : 5 mm
Length coated part: 10 mm
Coating thickness: 1-2 µm (PVD)

<table>
<tr><th>IFA</th><th>Experiment</th><th>Loop</th><th>Total duration Exposure (FPD)</th><th>Coatings/samples</th></tr>
<tr><td>731</td><td>In-core cladding corrosion (2012)</td><td>PWR</td><td>126</td><td><table><tr><th>Sample</th><th>Base material</th><th>Coating</th></tr><tr><td>1</td><td>Zr-4</td><td>TiAlN</td></tr><tr><td>2</td><td>Inconel 600</td><td>TiAlN</td></tr><tr><td>3</td><td>Inconel 600</td><td>ZrO_2</td></tr><tr><td>4</td><td>Inconel 600</td><td>CrN</td></tr></table></td></tr>
<tr><td>733.1</td><td>Crack initiation (2011-2012)</td><td>BWR</td><td>287</td><td><table><tr><th>Sample</th><th>Base material</th><th>Coating</th></tr><tr><td>1</td><td>Inconel 600</td><td>TiAlN</td></tr><tr><td>2</td><td>Inconel 600</td><td>CrN</td></tr></table></td></tr>
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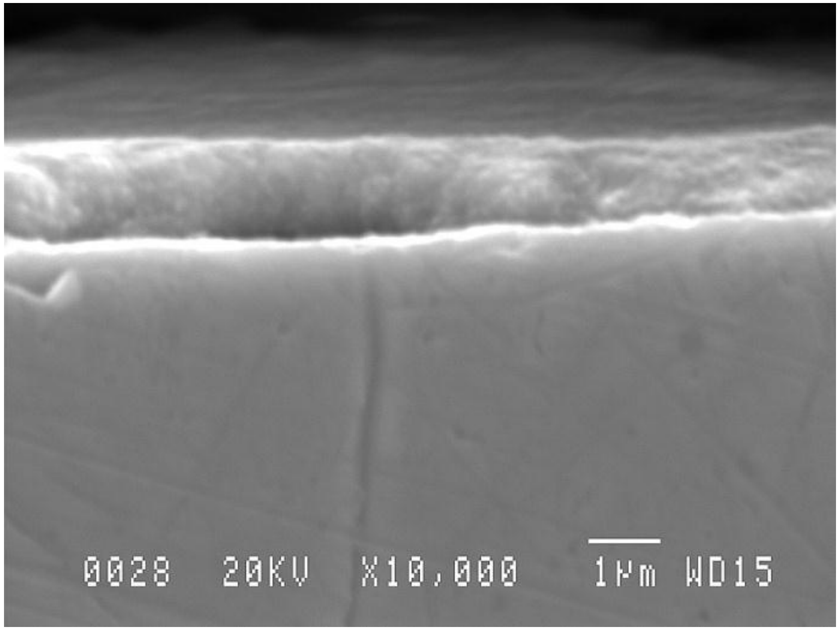
RESULTS

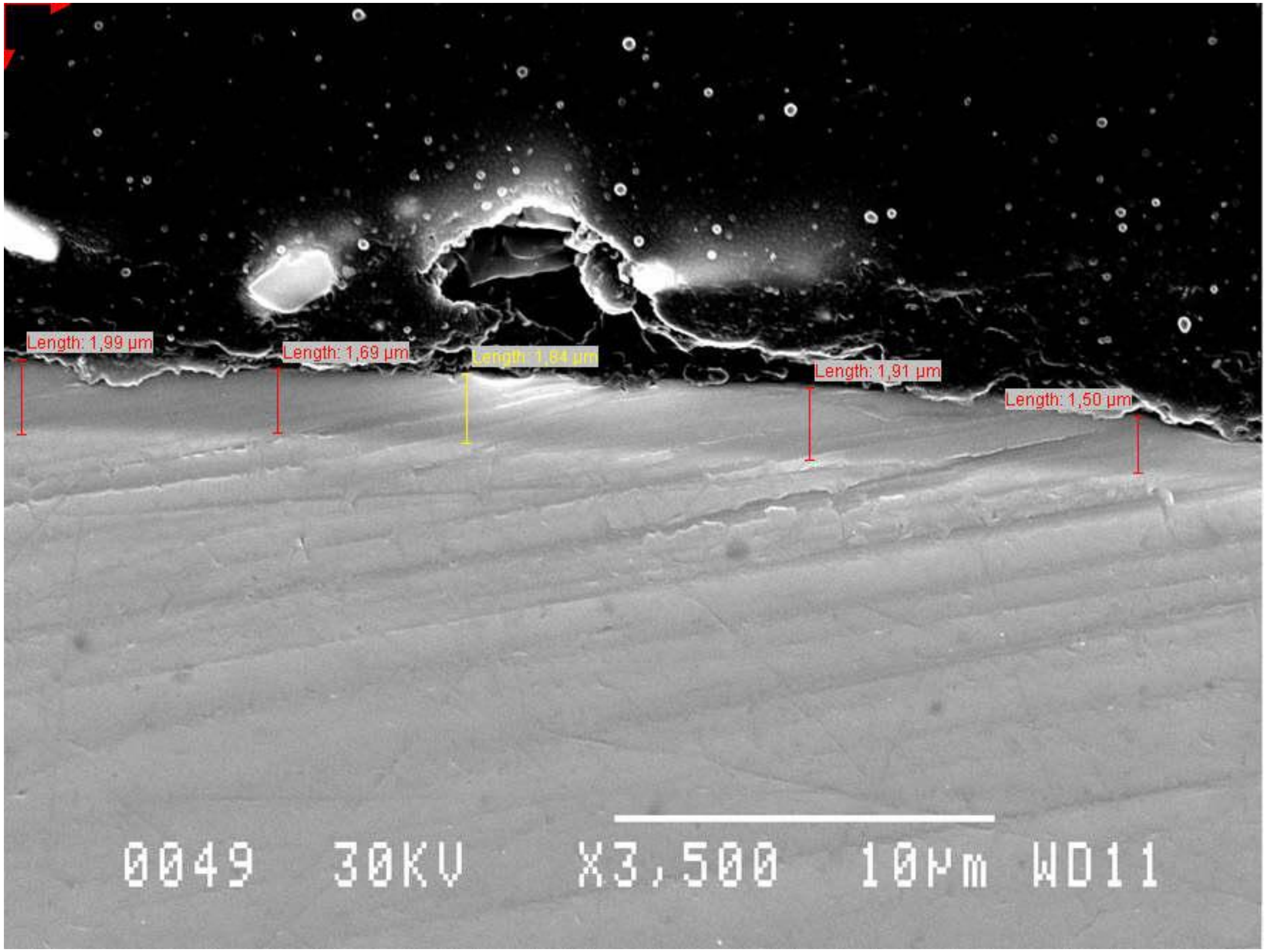
Condition	Thermal fluence	Full power days	TiAlN	CrN Base: Inconel 600	ZrO_2 Base: Inconel 600
BWR	4.5 10^{20} n/cm^2	287	OK (Inconel 600)	OK	--
PWR	2.1 10^{20} n/cm^2	126	OK for Zr-4 Disappeared on Inconel 600	OK	Disappeared



IFA-733 (BWR)
CrN coating after irradiation

- Protective chromium oxide
- layer formed

IFA-731 (PWR)
CrN coating after irradiation



Testing of small coated samples in CANDU conditions (out pile)

Tests performed in collaboration with AECL Canada

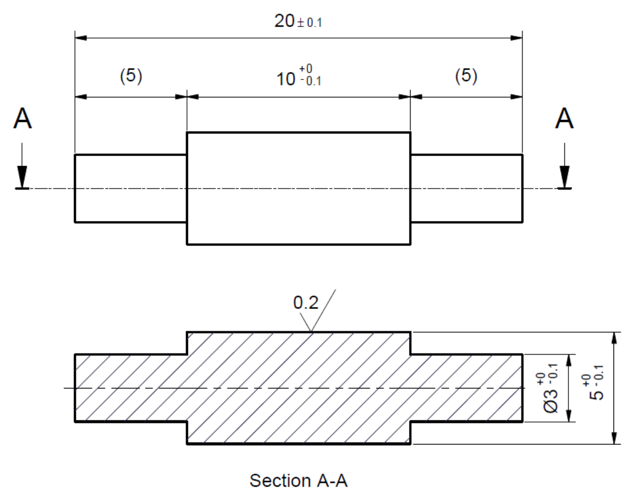
R. Van Nieuwenhove, K. Daub (AECL), H. Nordin (AECL) Investigation of the Impact of Coatings on Corrosion and Hydrogen Uptake of Nuclear Components, NUMAT Conference, 27-30 October, 2014, Clearwater Beach, Florida
(paper available upon request)

Very good corrosion protection obtained by CrN in CANDU conditions (120 days, 350 C, 165 bar, heavy water)

Coatings & Materials Studied

- Zr-4 coated
    - CrN
    - TiAlN
    - AlCrN

- SS-316L coated
    - CrN
    - AlCrN
    - TiAlN + AlCrN

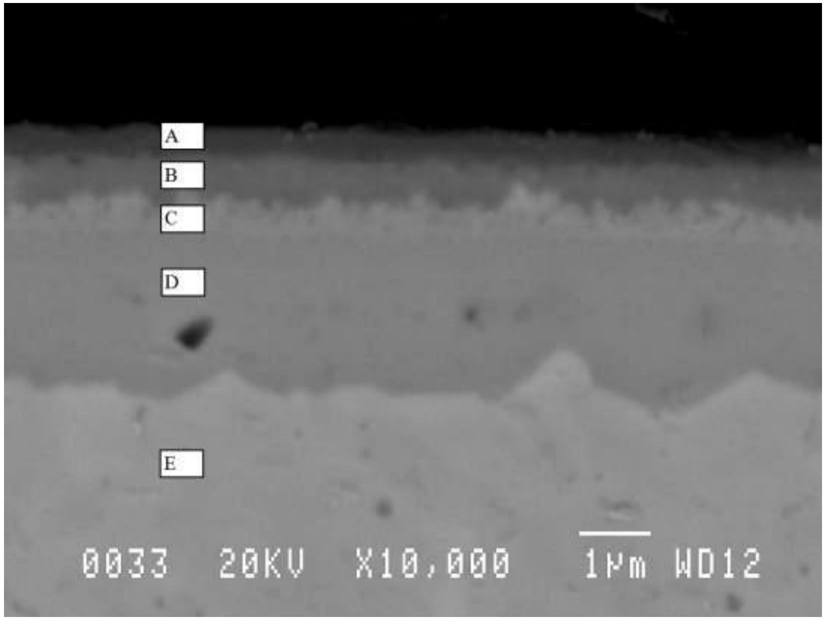


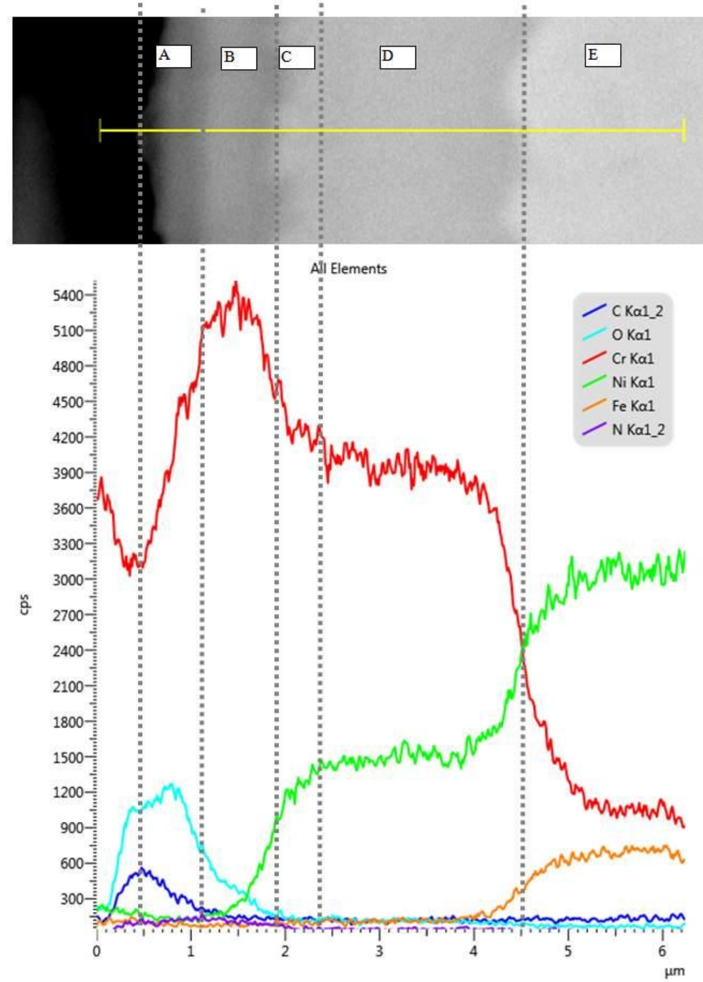
Testing of coatings in supercritical water

Testing performed in a supercritical water loop at VTT, Finland

R. Van Nieuwenhove, Juraj Balak (IFE), A. Toivonen (VTT), Sami Pentillä (VTT), Ulla Ehrnsten (VTT), Investigation of coatings, applied by PVD, for the corrosion protection of materials in supercritical water, ISSCWR-6, Shenzhen, China, March, 2013

Paper can be obtained on request (rudivn@hrp.no)



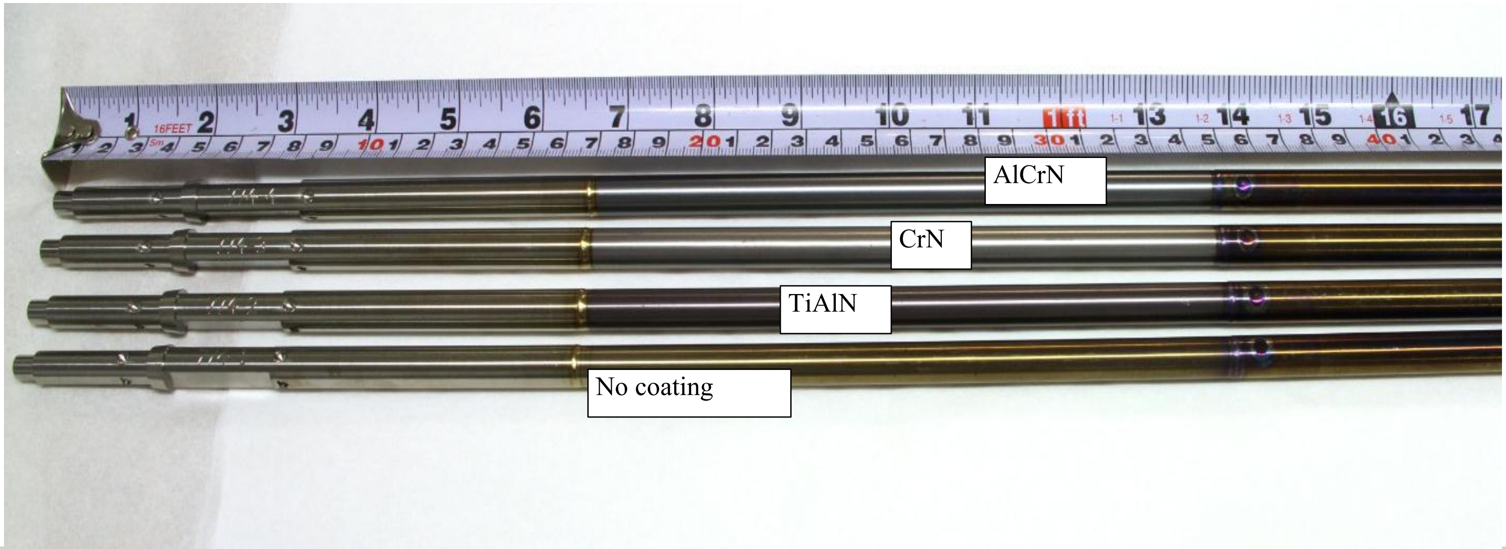


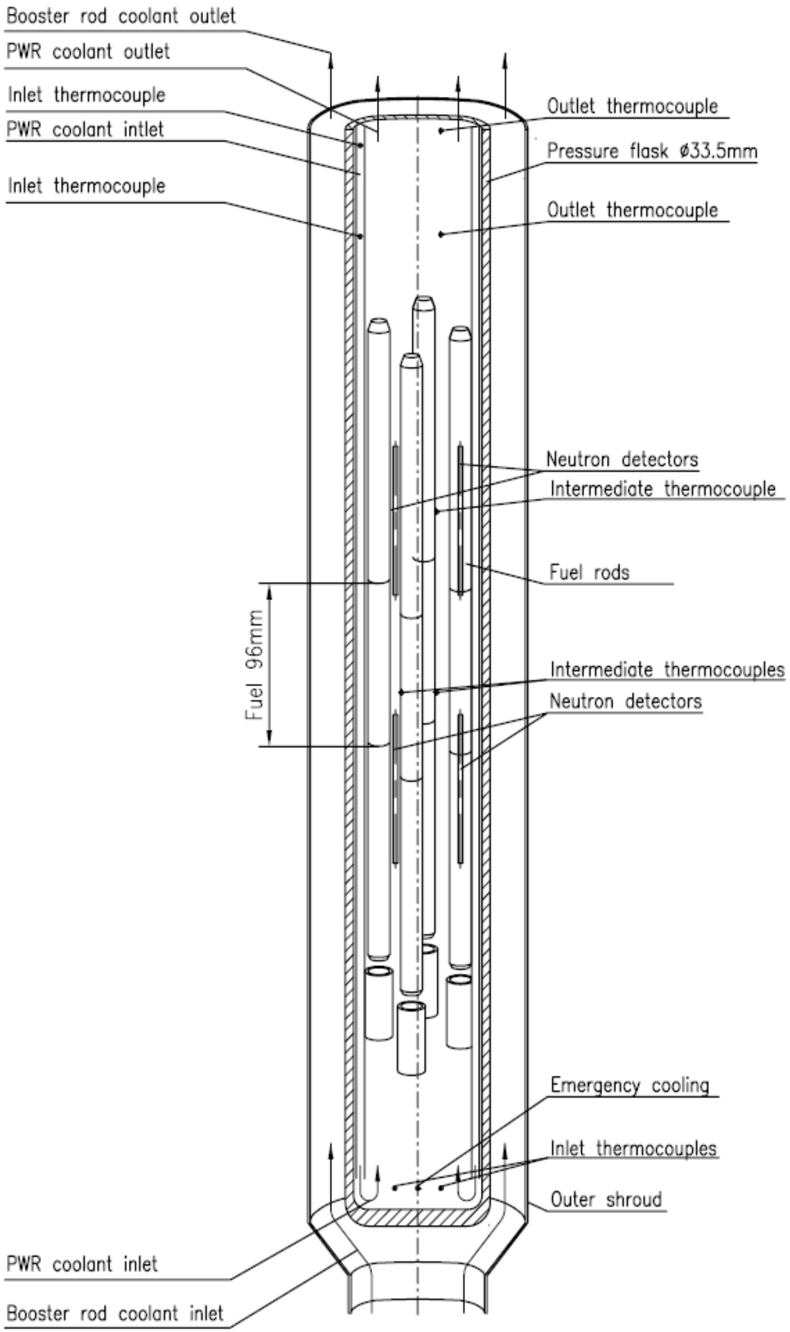
SEM analysis of CrN coating (Inconel 600) after SCW exposure at 650 ºC

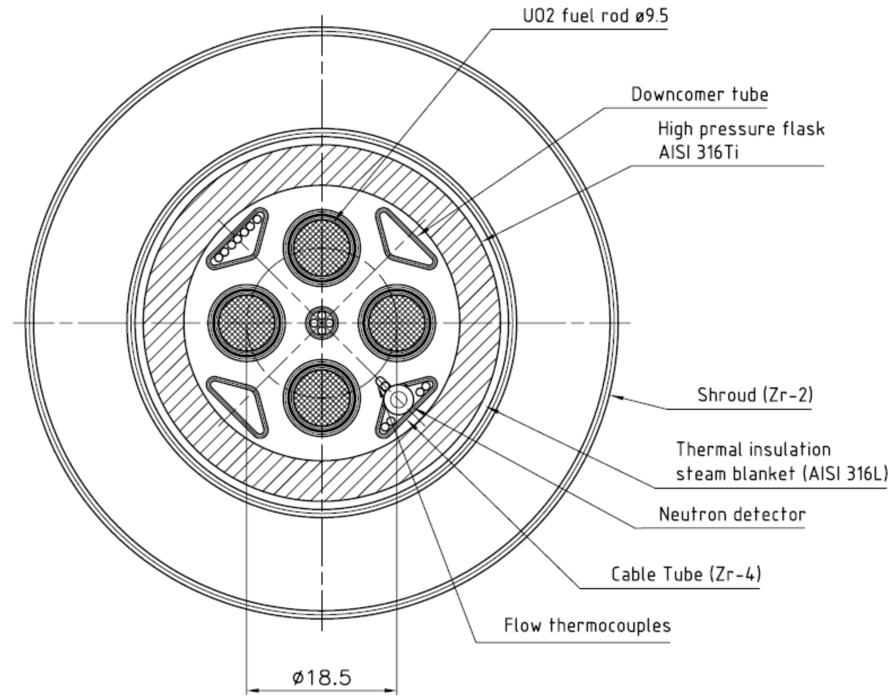
Coating layer intact (after 1000 h)

In-pile testing of coatings on fuel rods in PWR conditions

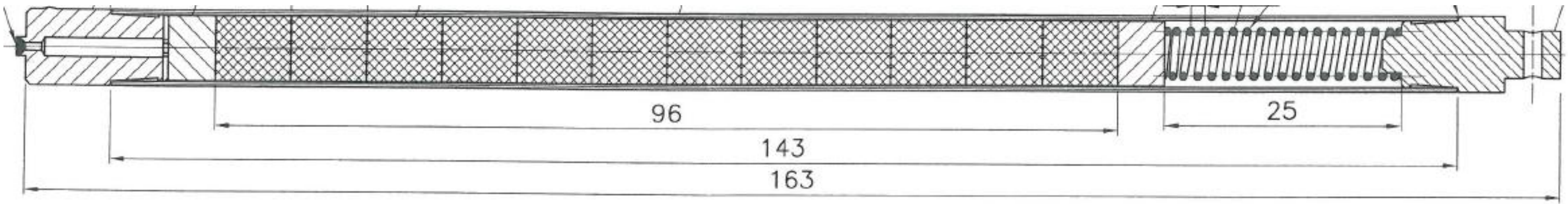
- First experiment (in the world) with coated **fuel rods**
- PWR conditions (2 reactor cycles) at 320 ºC
- Three rods with coatings: TiAlN, CrN, **AlCrN** (PVD)
- One reference rod
- Irradiation started on 15.03.2014, ended 11.2014 (143 days)
- Fast fluence 1.89 E20, thermal fluence 4.1E20 (n/cm^2)




Booster rod coolant outlet
PWR coolant outlet
Inlet thermocouple
PWR coolant intlet
Inlet thermocouple
Outlet thermocouple
Pressure flask ø33.5mm
Outlet thermocouple
Neutron detectors
Intermediate thermocouple
Fuel rods
Fuel 96mm
Intermediate thermocouples
Neutron detectors
Emergency cooling
Inlet thermocouples
Outer shroud
PWR coolant inlet
Booster rod coolant inlet


UO2 fuel rod ø9.5
Downcomer tube
High pressure flask
AISI 316Ti
Shroud (Zr-2)
Thermal insulation
steam blanket (AISI 316L)
Neutron detector
Cable Tube (Zr-4)
Flow thermocouples
ø18.5

Fuel rod characteristics



Double cladding: In view to ease PIE examination of the coated cladding

- Outer, coated cladding (Zr-4): 9.5/8.36 mm
- Inner cladding, not coated (Inconel 600): 8 / 7 mm
- Pellet outer diameter: 6.83 mm
- Fuel rod enrichment : 5 %
- Fill gas : He
- Designed to reach a linear heat rate of 20 kW/m

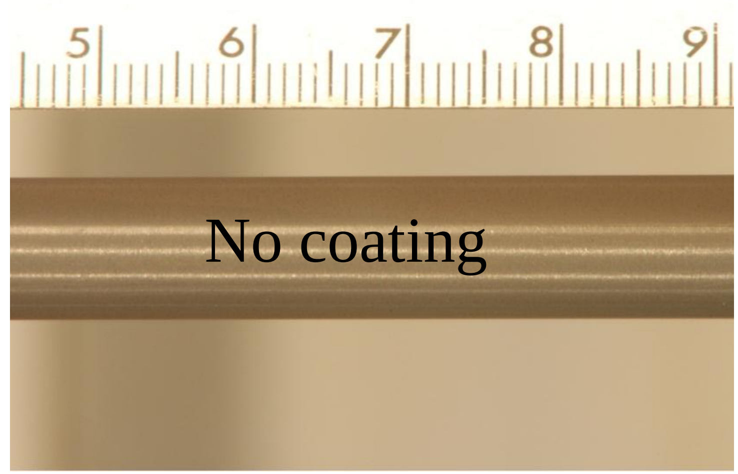
Coating evaluation methodology

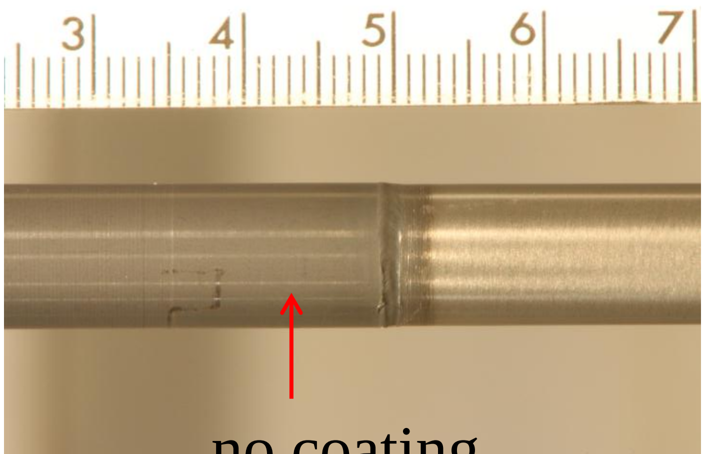
1. Pre-characterization of the coatings
   - SEM of cross section (coating thickness)
   - Hardness measurements
2. After irradiation, the outer cladding was retrieved and cut into sections
   - SEM, hardness measurements
   - Check what remains of the coating

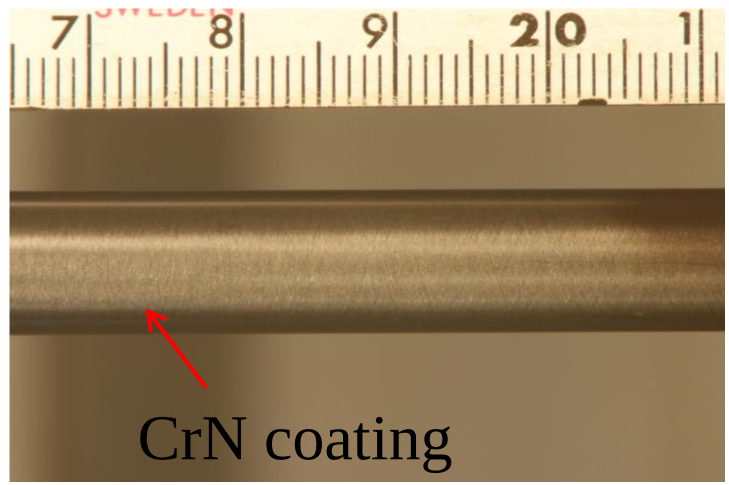
R. Van Nieuwenhove, IFA-774: The first in-pile test with coated fuel rods Enlarged Halden Program Group Meeting in Røros, 2014

From visual inspection, the CrN seems to survive very well!

Further SEM examinations
Will be performed in 2015







Important Note:

Coating of full-scale fuel rods (> 4 m) is possible today

PVD-Arc machine with 24 arc evaporators.
The coatings can be applied with a good coating uniformity.
The coating thickness difference over the whole length is lower than 1µm at a layer thickness of about 4µm.

Deformation limits of some PVD coatings
(based on tests at Halden)

- CrN : between 1.5 % and 2 %
- AlCrN: larger than 2.5 %
- TiAlN: between 1.5 % and 2 %

When cracks occur, the cracks are narrow without spallation of the coating occurs

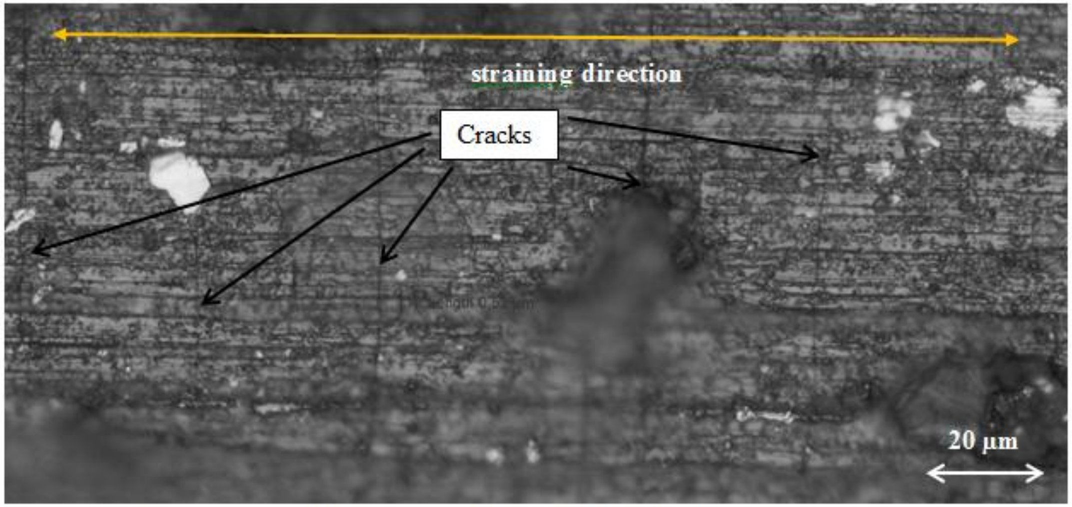


Figure 8: CrN coating on AISI 316L, at 300 °C and 2 % strain (ID 12).

Further work

- Investigate effect of defect/scratch in coating (out-pile and in-pile test in preparation)
- ATF fuel testing within the Halden Project
- Perform LOCA test on coated fuel rods
  - Out-pile
  - In-pile (Halden reactor)

Conclusions

- Industrial PVD coatings have been tested under various conditions
  - ✓ Corrosion protection (CrN) for PWR, BWR, CANDU
  - ✓ Perfect corrosion protection for supercritical water (CrN)
  - ✓ The coatings are irradiation resistant: First in-pile testing on samples and fuel rods in the Halden reactor
  - ✓ Additional benefits: Reduction of hydrogen absorption, reduced hydrogen generation in accident conditions, avoid fretting
  - ✓ The coatings have sufficient flexibility