Surface Characteristics of three Accident Tolerant Fuel Cladding Materials and their Potential Impact on Critical Heat Flux in LWRs

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Nuclear Energy

- **Carbon Free Energy**

- Worldwide Nuclear power reactors → 440
- U.S. Nuclear power reactors → 94
- Research and test reactors in U.S. → 30
- Nuclear Energy supply nation’s 20% electricity need

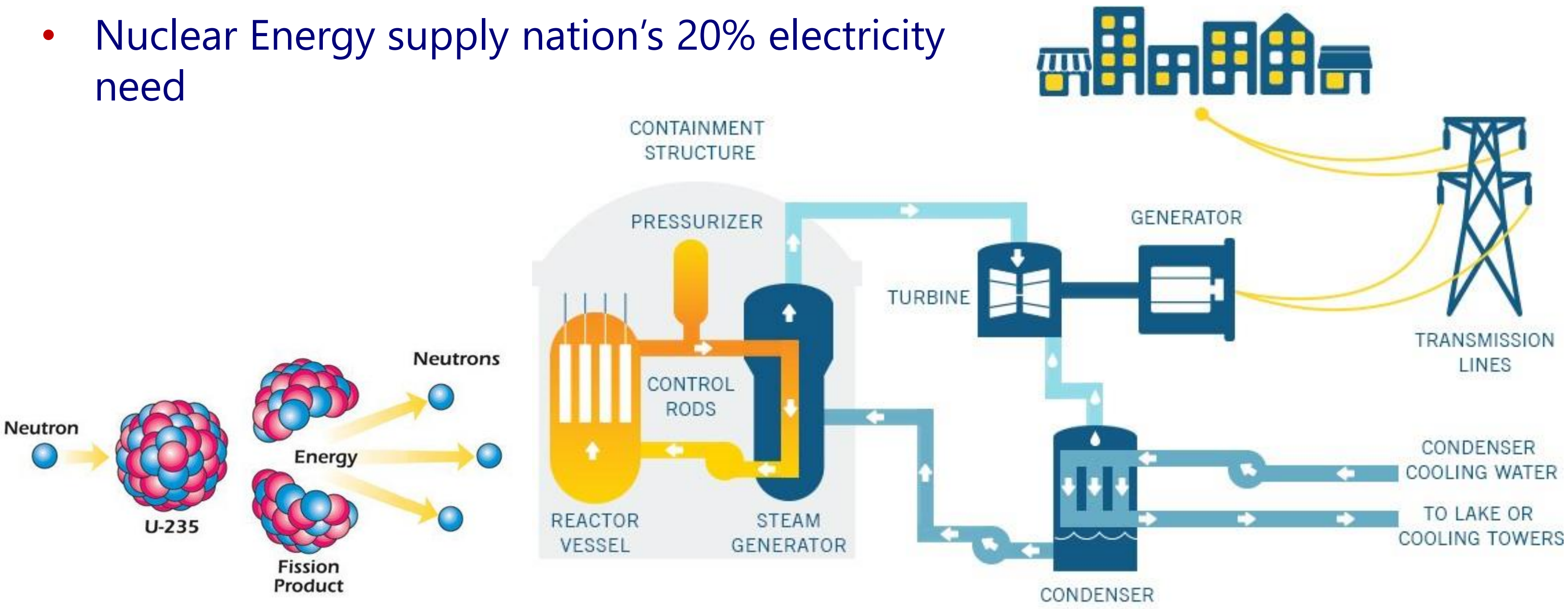


Fig. 1 Basic concept of Nuclear Power Generation (Pressurized Water Reactor (PWR))

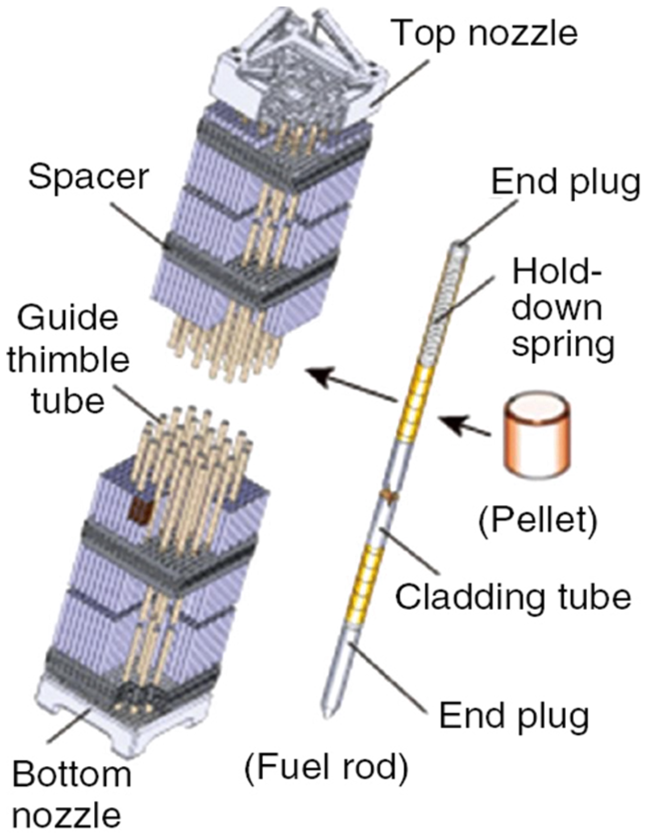
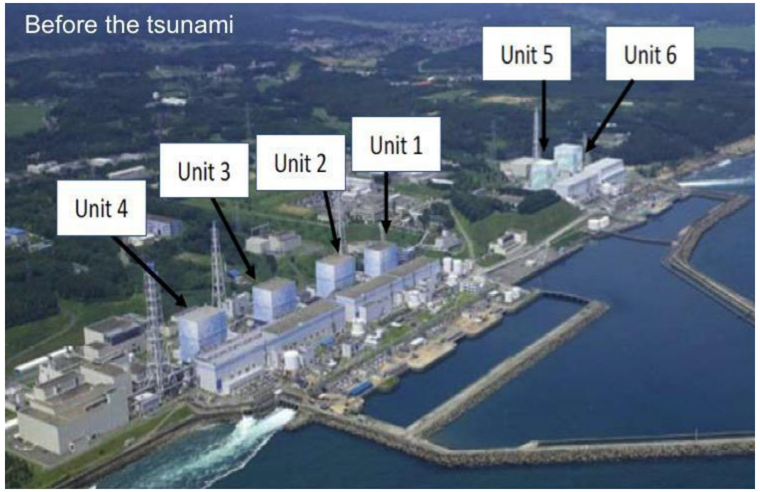


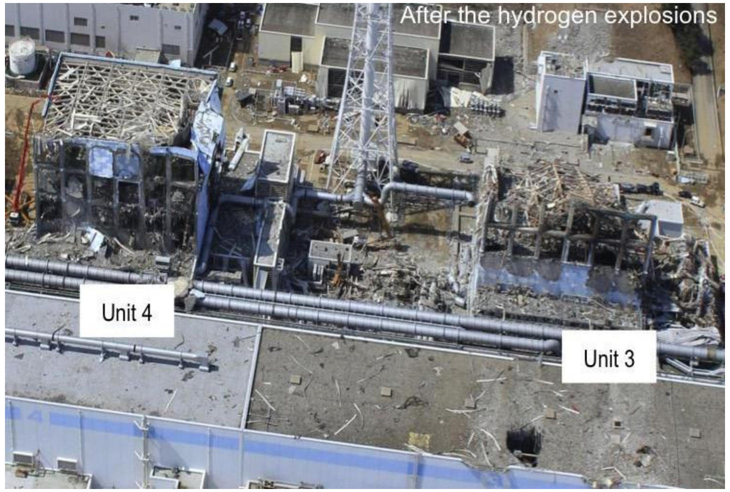
Fig. 2 Schematic view of a PWR fuel assembly *(Tang2017 et. al.)*

Rich, Alex K., and Tom Warhol. "Nuclear Power: An Overview." Points of View Reference Center. N.p., 1 Mar. 2016. Web. 24 Oct. 2016.

Challenges

○ **Fukushima nuclear reactor accident**





Fukushima Daiichi nuclear accident on March 11th, 2011 (Tohoku earthquake and tsunami)

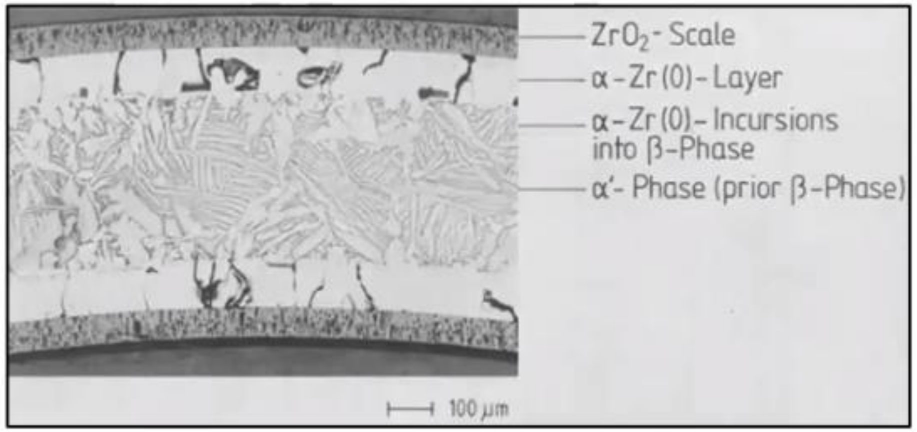
- Disabled the emergency power generators
- Insufficient cooling → three nuclear reactor meltdown
- Core temperatures > 1200 °C
- **Hydrogen-air explosions**
- The release of radioactive material

The Fukushima Daiichi Accident Report by the Director General (2015)

Fuel oxidation

$$Zr + 2H_2O \rightarrow ZrO_2 + 2H_2$$

$$\Delta H = -586\ kJ/mol$$

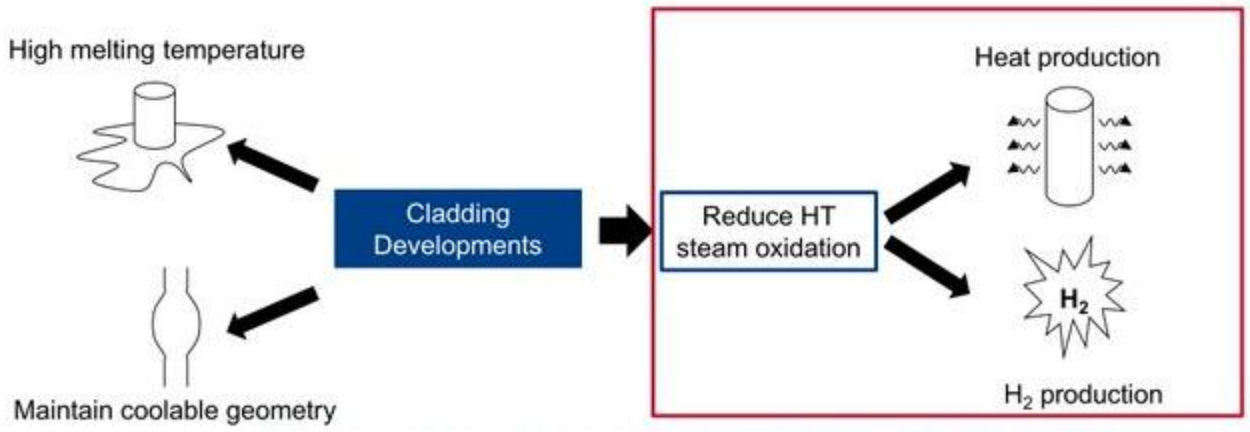


Cross section of Zircaloy-4 after steam oxidation at 1400 °C for 2 min (Leistikow et. al. (1985))

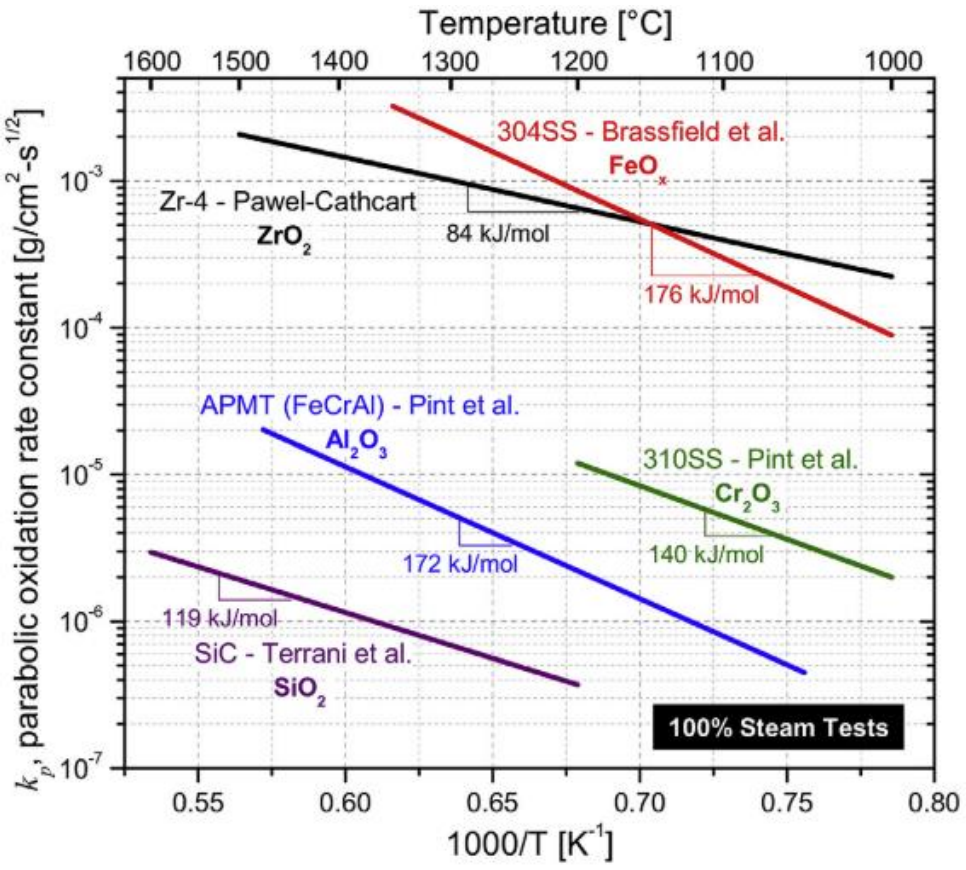
Accident Tolerant Fuels

After Fukushima, US Congress mandated DOE to develop "**melt-proof**" nuclear fuel technology.

According to DOE, "the fuels with enhanced accident tolerance are those that, in comparison with the standard UO_2 – Zr system, can **tolerate loss of active cooling in the core for a considerably longer time period** while maintaining or improving the fuel performance during normal operations".



Focus on cladding development: limiting the high temperature steam oxidation



Parabolic oxidation rate for various cladding materials and their resulting oxide in steam as a function of temperature *(Pint2015 et. al., Terrani2014 et. al.)*

Concepts of ATF

ATF Claddings

Surface modification | **Replacement**

Concept 1
Zr alloys
Surface coating

Concept 2
Alloys

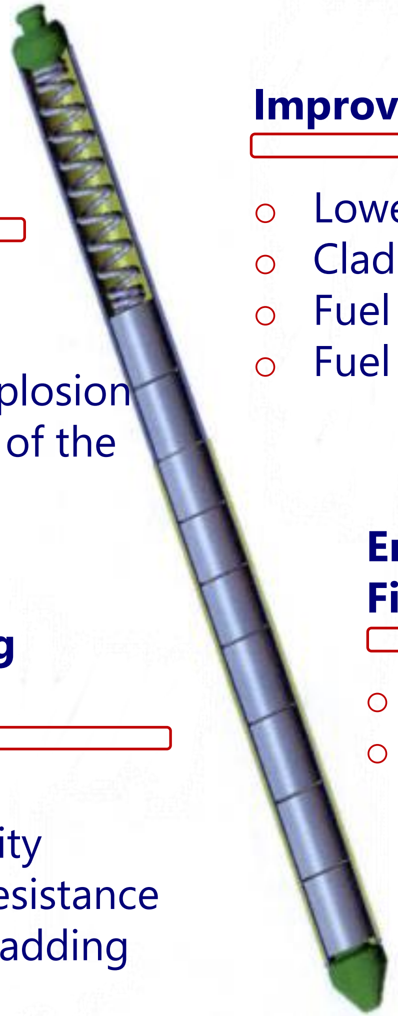
Surface modification	Replacement
FeCrAl	APMT
Cr	C26M
Mo	310SS
SiC	

Improved Clad Reaction Kinetics with Steam

- Heat of Oxidation
- Oxidation rate
- Hydrogen bubble and explosion
- Hydrogen embrittlement of the clad

Improved Cladding Properties

- Clad fracture
- Geometric stability
- Thermal shock resistance
- Melting of the cladding



Improved Fuel Properties

- Lower operating temperatures
- Clad internal oxidation
- Fuel relocation
- Fuel melting

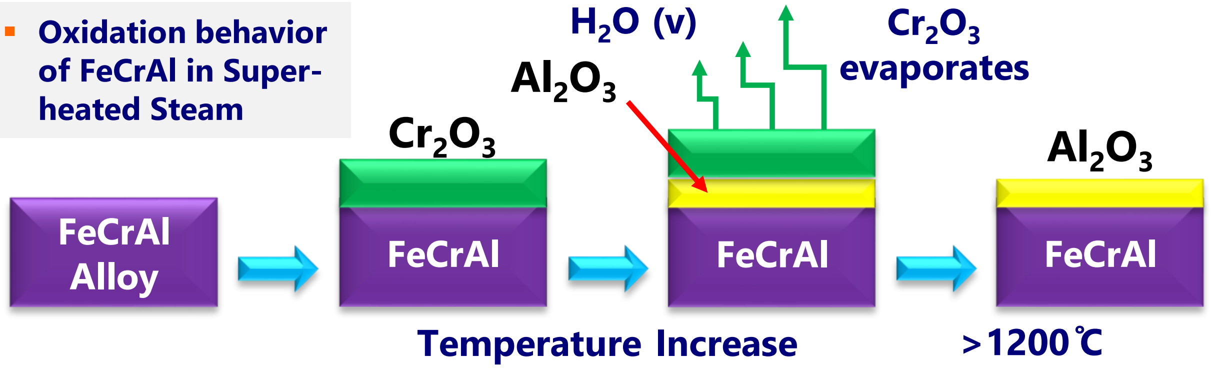
Enhanced Retention of Fission Products

- Gaseous fission products
- Solid/liquid fission products

Areva's Enhanced Accident Tolerant Fuel Program (2017)

Current Research: FeCrAl – An ATF

- Oxidation behavior of FeCrAl in Super-heated Steam



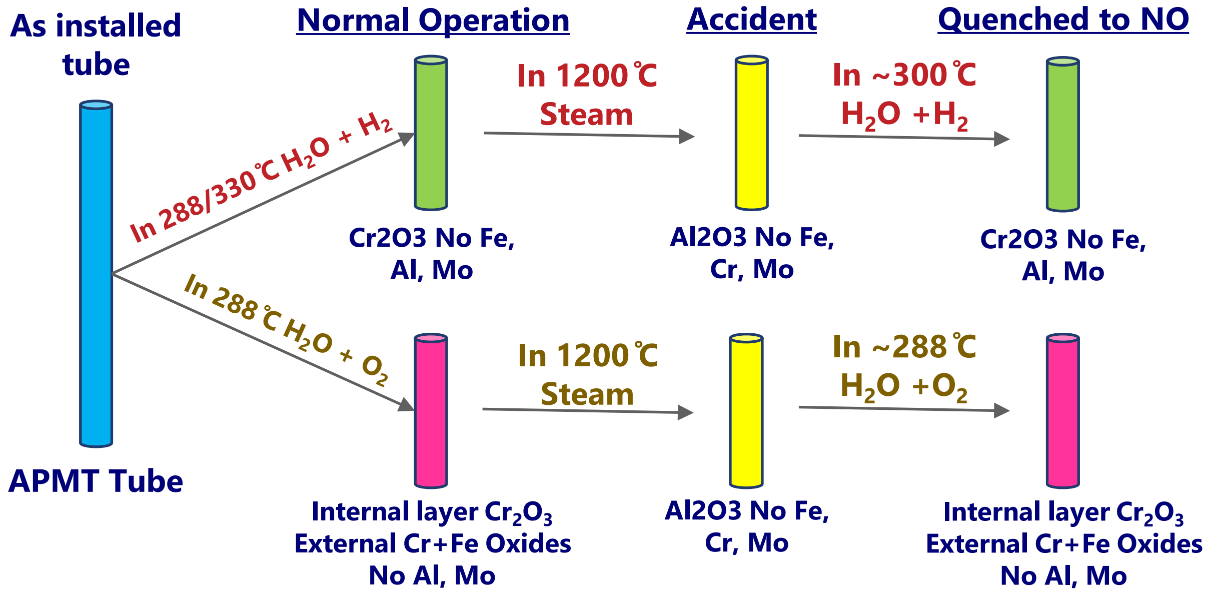


Fig. 1 Schematic representation of how Al and Cr in FeCrAl react to normal operation and accident conditions

Rebak et al. – 2017 – Iron-chrome-aluminum alloy cladding for increasing safety in nuclear power plant

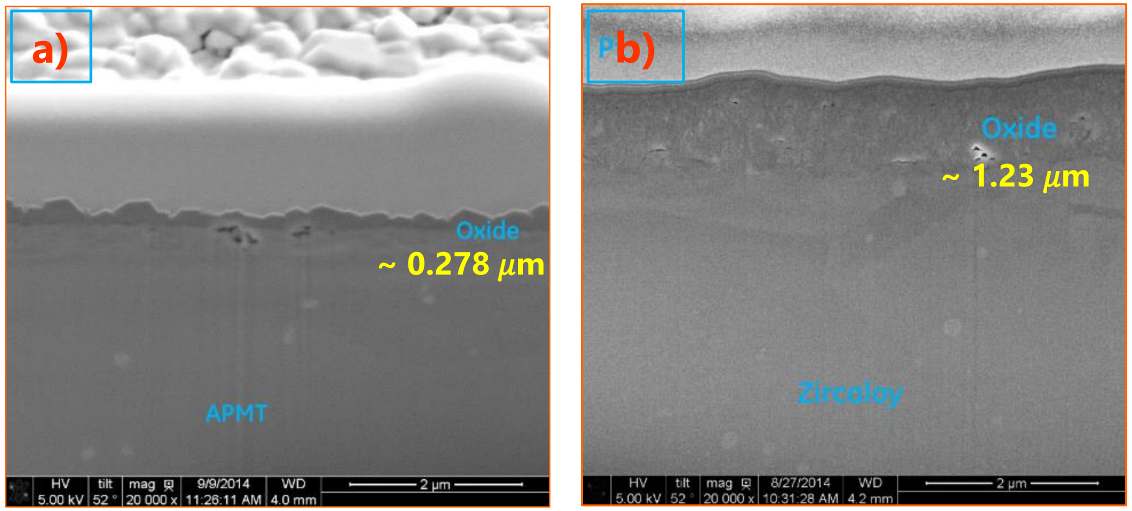


Fig. 2 Cross sections of coupons exposed to BWR simulated conditions for one year (288℃ + 2 ppm O_2)

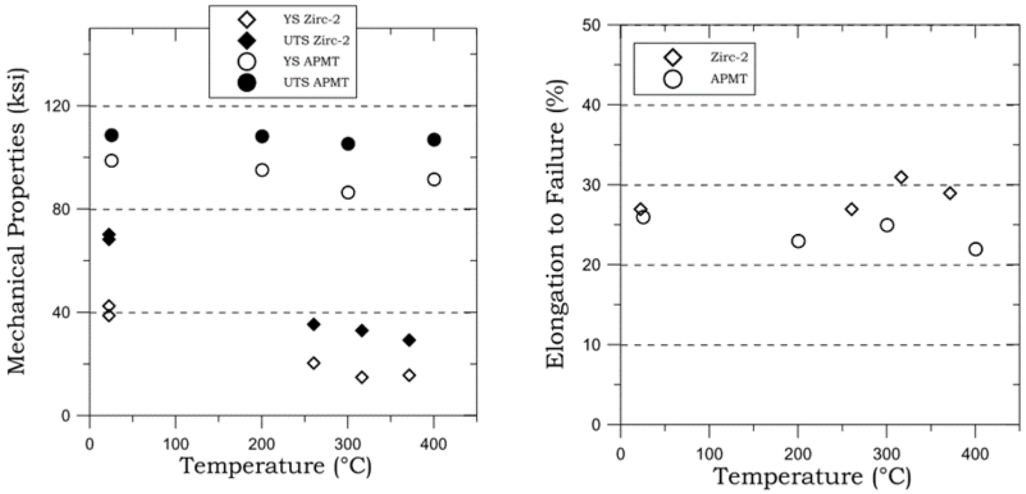


Fig. 3 Mechanical properties of Zircaloy-2 and APMT without irradiation

Rebak et al. – 2016 – FeCrAl Alloys for Accident Tolerant Fuel Cladding in Light Water Reactors

Current Research: Cr-coated Zircaloy Cladding

Westinghouse

Coating materials: Cr

Fabrication method: Cold spraying

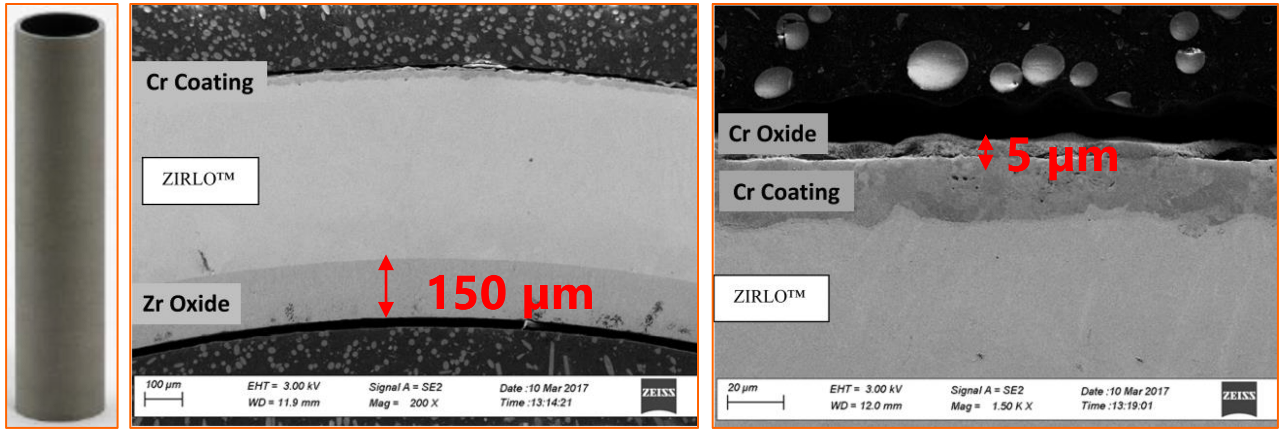


Fig. 1 Cross section microstructures after ultra-high temperature oxidation in 1300℃ steam for 20 min

No spalling of the coating was observed:
Fig. 2 Cr-coated samples after tensile testing: a) 1% strain, b) 10% strain and c) test to failure

Shah et al. – 2017 – Development of Surface Coatings for Enhanced Accident Tolerant Fuel (ATF)

Framatome

Coating materials: Cr

Fabrication method: Physical Vapor Deposition

Fig. 3 Zircaloy clad with/without Cr-coating after LOCA in 1200℃ steam for 6000 s

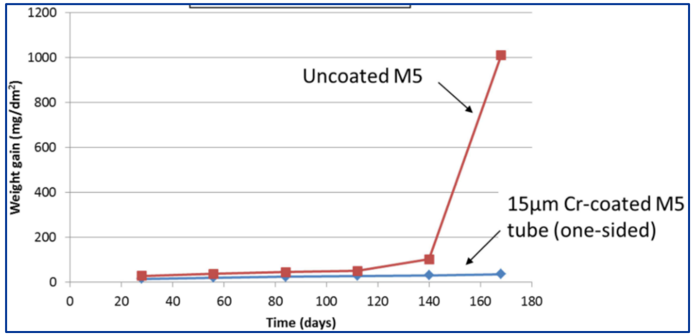


Fig. 4 Corrosion of Cr-coated M5 exposed to 360℃ PWR water

Equipment of PVD

Bischoff et al. – 2018 – AREVA NP's enhanced accident-tolerant fuel developments focus on Cr-coated M5 cladding

Motivation of this work

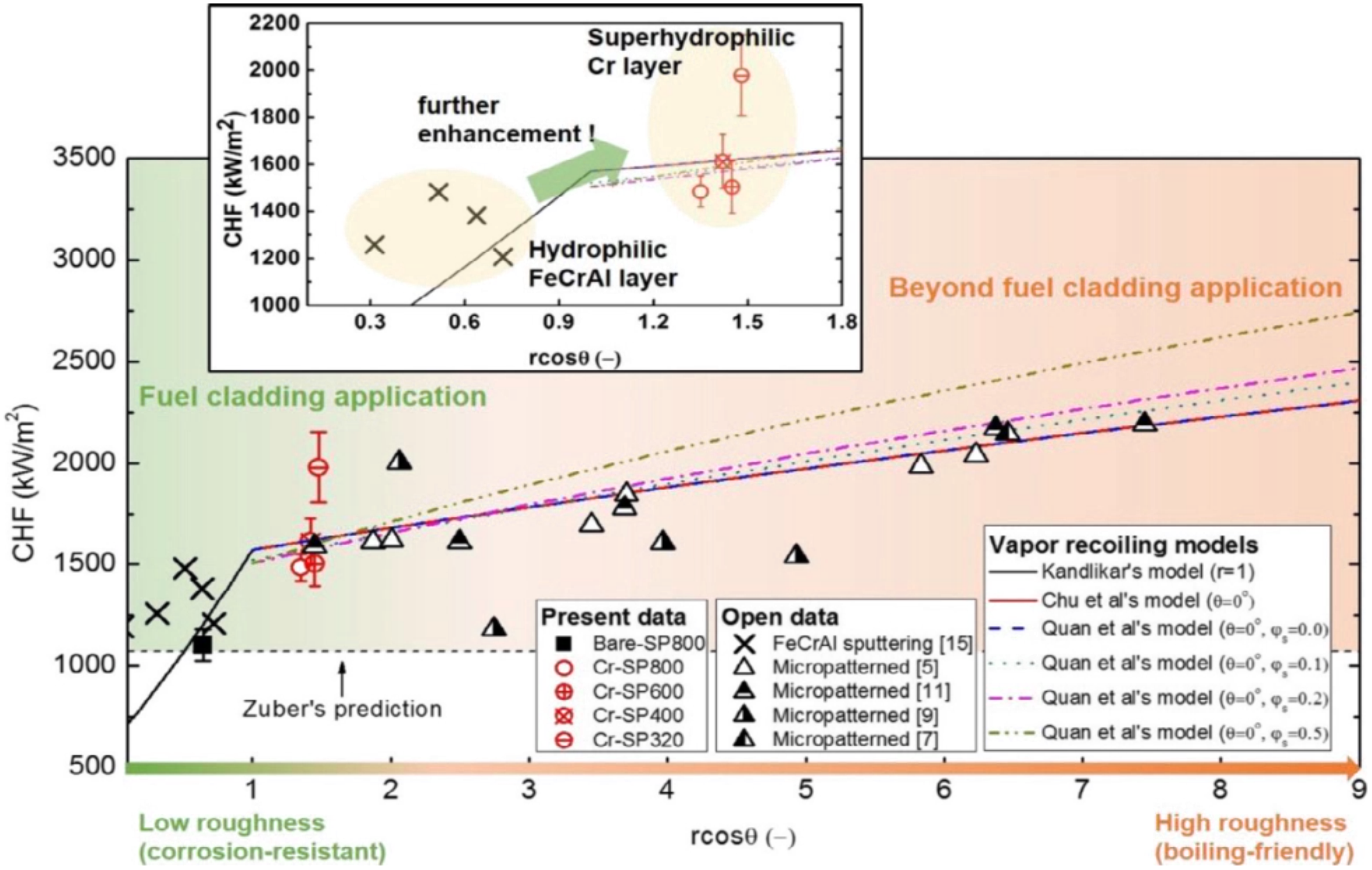


Figure 1: Comparison of Critical Heat Flux (CHF) data in term of roughness factor r and contact angle θ. (r is surface area ratio of geometric area to projected area) ***(Son et al.(2017)).***

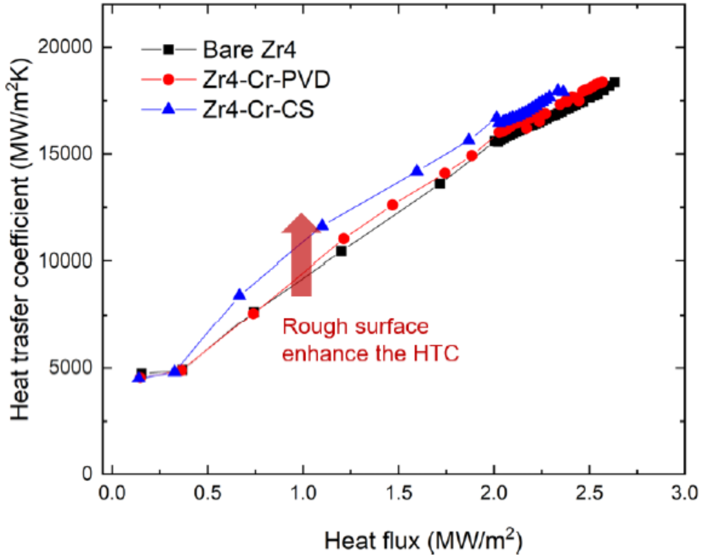


Figure 2: Comparison of average heat transfer coefficient (HTC) of the bare Zr4, Zr4-Cr-PVD and Zr4-Cr-CS rod, respectively *(Lee et al.(2020)).*

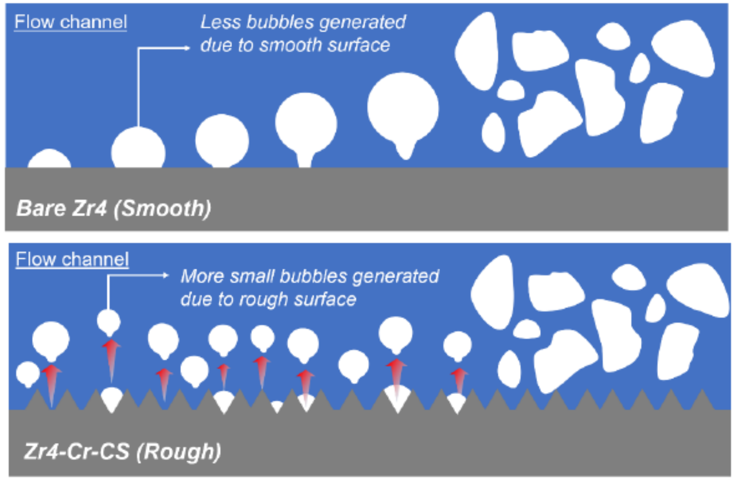
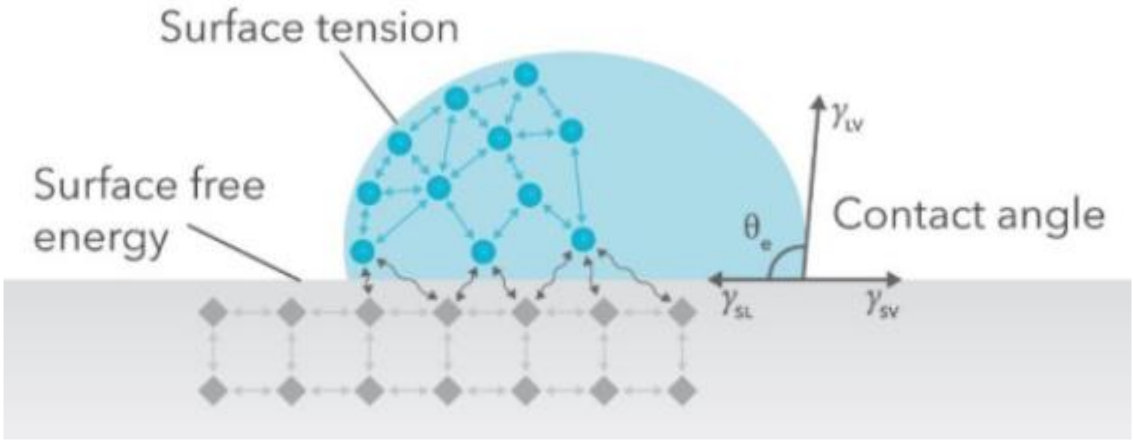
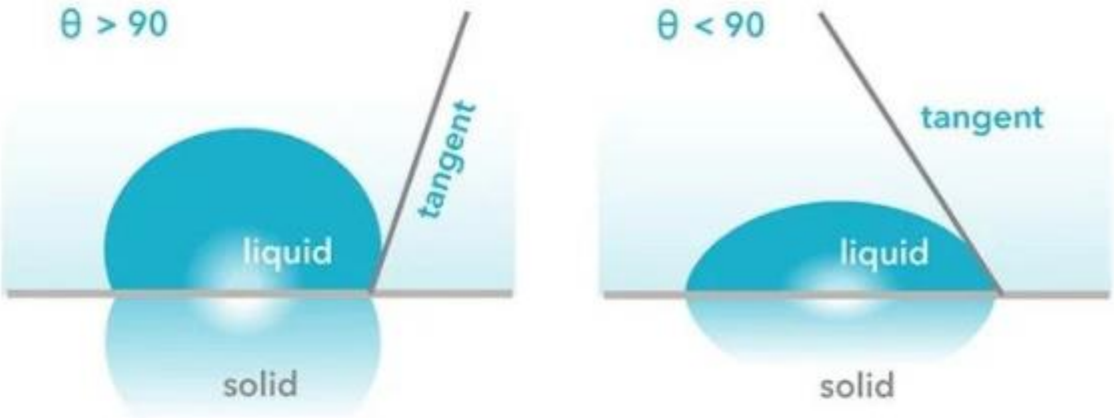


Figure 3: Schematic diagram for the higher HTC on the Zr4-Cr-CS rough surface compared to the bare Zr4 smooth surface *(Lee et al.(2020)).*

Wetting



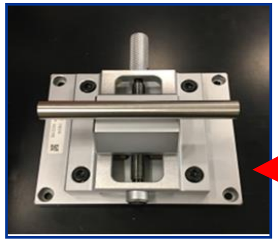


hydrophobic

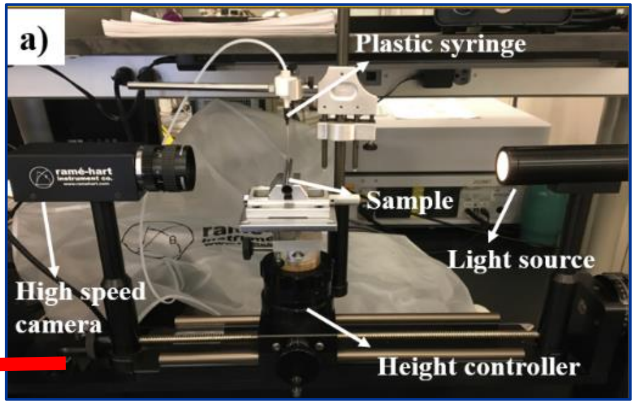
hydrophilic

Young's law

$$\gamma_{LV}\cos\theta = \gamma_{SV} - \gamma_{SL}$$



Sample stage

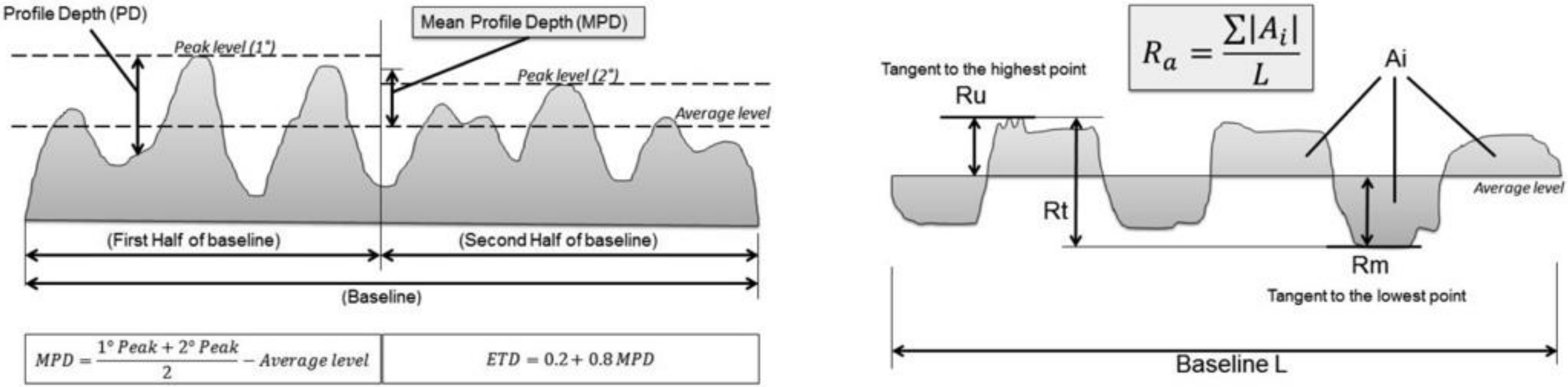


Rame-hart Contact Angle Goniometer
To measure wettability of surface

- **Hydrophilic**: Droplet fills the roughness asperities and wets entire solid surface
- **Hydrophobic**: Droplet sits raised up on the roughness asperities, cause air pockets underneath the droplet

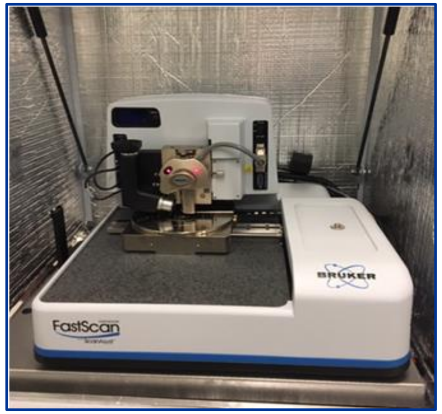
Kock-Yee Law & Hong Zhao et. al. 2016

Roughness



Understanding of the surface roughness parameters ***(Bitelli et. al.(2012))***

Mitutoyo SJ-410 profilometer



Bruker Dimension Icon Atomic Force Microscope

- **Ra:** The arithmetic average roughness
- **Rz:** The average of the five highest peaks and the five deepest valleys
- **RSm:** The arithmetic mean value of the width of profile elements

CHF test

Figure 1: Atmospheric Pressure Loop at University of Wisconsin – Thermal Hydraulic Lab

Table I. Testing parameters used for the flow boiling CHF test

Testing parameter	Value
Cladding tube OD (mm)	9.5 (Zr-based alloys) and 10.26 (FeCrAl alloys)
Cladding wall thickness (mm)	0.51 (Zr-based alloys) and 0.4 (FeCrAl alloys)
Working fluid	Water
Nominal heat flux (MW/m^2)	0 - 3
Inlet temperature (°C)	24
Inlet pressure (kPa)	115
Nominal mass flux (kg/m^2s)	750
Heated length (mm)	457.2
Hydraulic diameter (mm)	10.50 (Zr-based alloys) and 9.74 (FeCrAl alloys)

Experimental CHF values

Sample	CHF value (MW/m^2)
AR-Zr4	2.63
600G-Zr4-Cr-PVD	2.58
AR-Zr4-Cr-CS	2.31
APMT	2.54
C26M	2.26

(R. V. Umretiya, et. Al. J. of Nuc. Mat.. 541 (2020) 152420)

Surface Characteristics before and after CHF test: Cr-Zr4

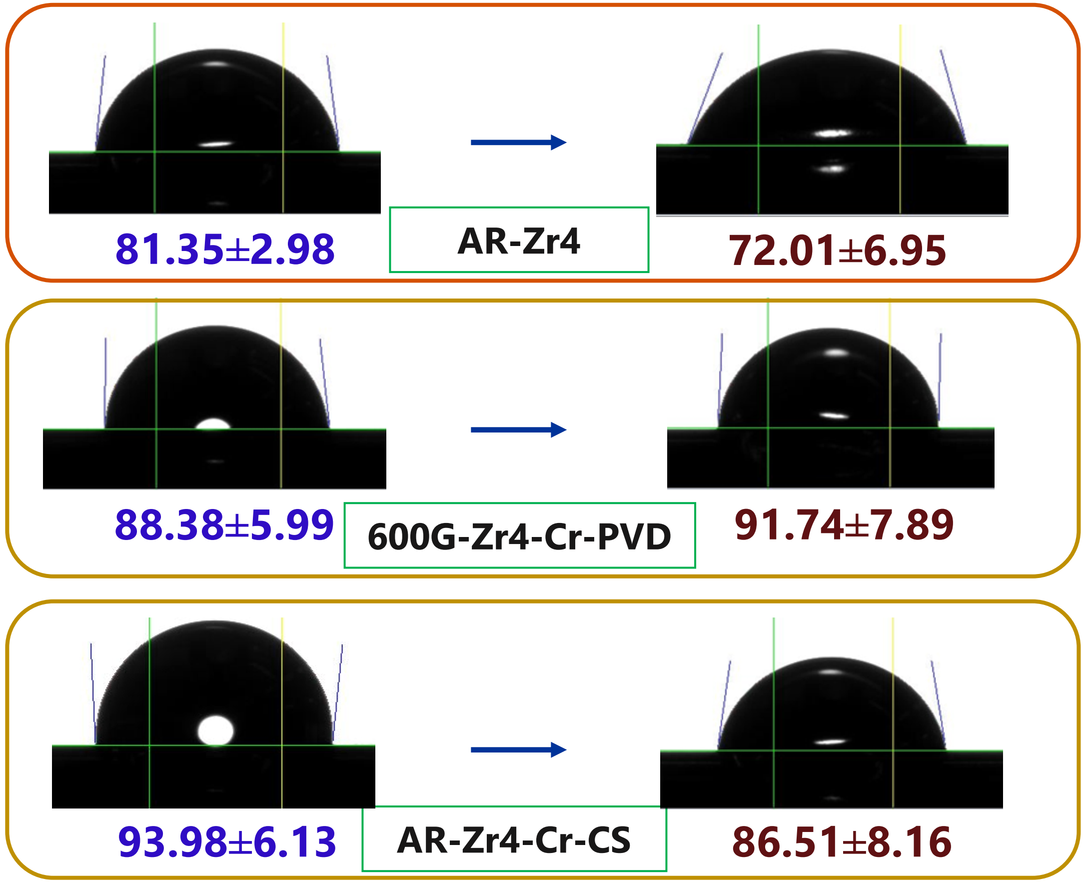


Figure 1: Comparison of Static Contact angle for all samples

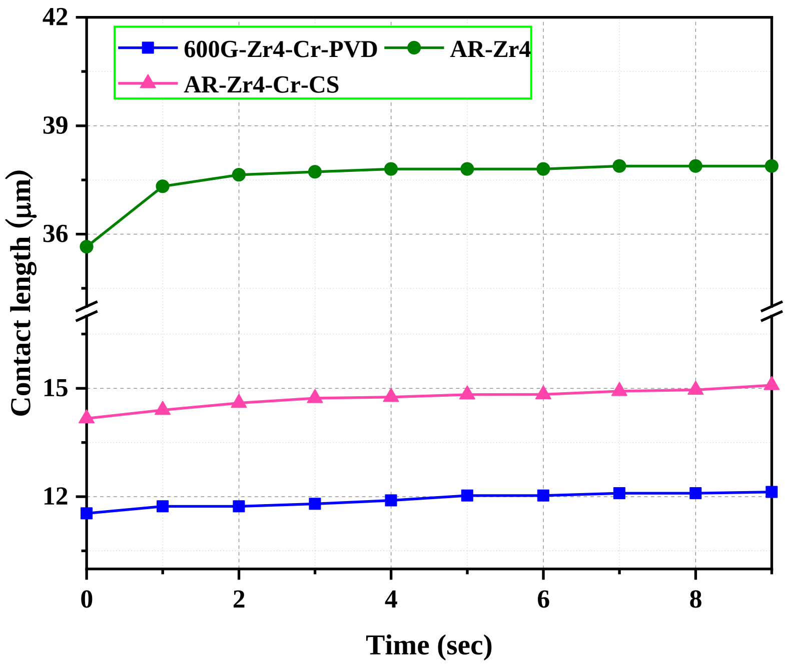


Figure 2: Droplet spreading after CHF test

- Post-CHF samples showed improved wettability for substrate Zr-4 and Cold Spray coated samples however wettability for PVD coating was slightly decreased
- Roughness for all samples was slightly increased. These surface parameters could lead to better thermal-hydraulic performance in reactor

Roughness data measured using Contact Profilometer (Mitutoyo SJ-410)

Sample	Ra (μm)	Rq (μm)
AR-Zr4	0.43	0.54
600G-Zr4-Cr-PVD	0.47	0.61
AR-Zr4-Cr-CS	1.04	1.33

After CHF

Sample	Ra (μm)	Rq (μm)
AR-Zr4	0.56	0.79
600G-Zr4-Cr-PVD	0.51	0.66
AR-Zr4-Cr-CS	1.11	1.50

(R. V. Umretiya, et. Al. J. of Nuc. Mat.. 541 (2020) 152420)

Surface Characteristics before and after CHF test : FeCrAl

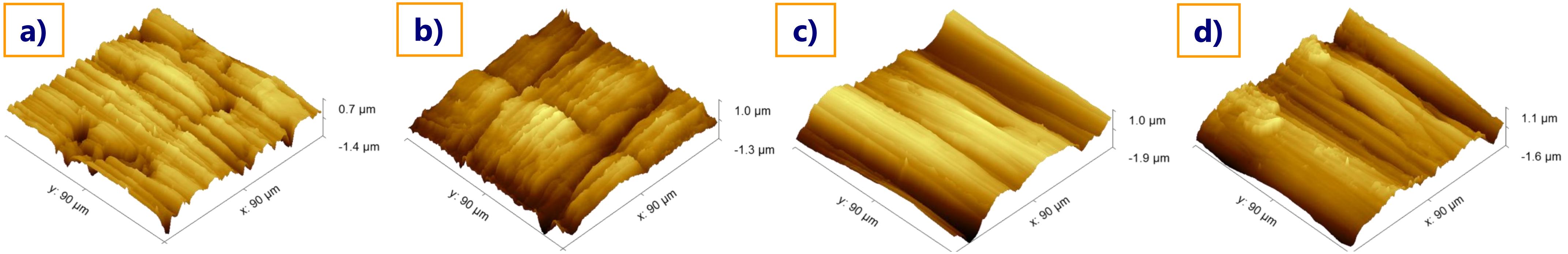


Figure 1: AFM topography : a) APMT_Pre-CHF, b) APMT_Post-CHF, c) C26M_Pre-CHF and d) C26M_Post-CHF

Roughness data measured using Contact Profilometer (Mitutoyo SJ-410)

Sample	Contact profilometry measurements (μm)			AFM measurements (nm)	
	Ra	Rz	RSm	Ra	r
APMT_Pre-CHF	**0.40±0.09**	3.89±0.99	70.11±7.24	205	1.02
APMT_Post-CHF	**0.47±0.08**	4.68±1.03	76.86±8.22	243	1.03
C26M_Pre-CHF	**0.80±0.12**	8.75±1.65	130.27±22.73	454	1.02
C26M_Post-CHF	**0.90±0.10**	9.62±1.83	138.86±22.92	550	1.04

- Post-CHF samples showed significant increase in wettability (given by decrease in contact angle in Fig. 2) for both APMT and C26M
- Roughness for both FeCrAl samples was slightly increased.

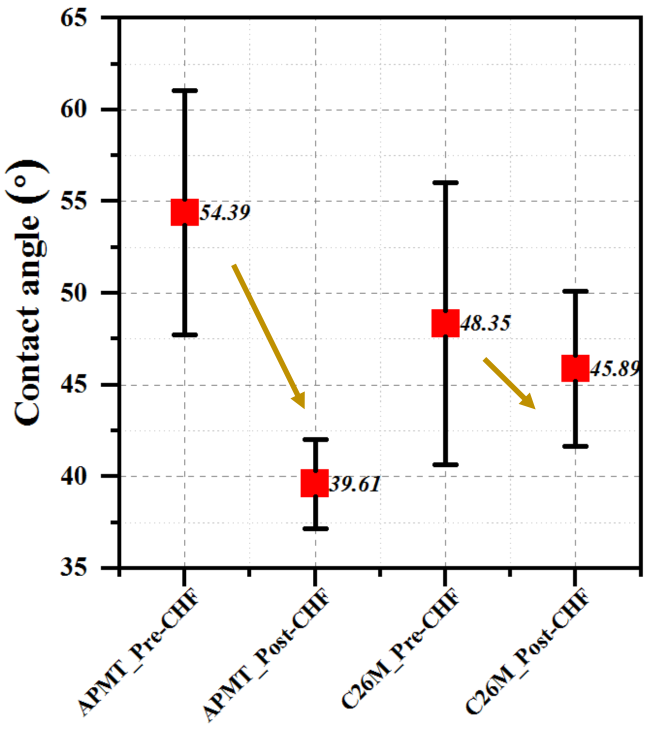


Figure 2: Comparison of static contact angle (left) and droplet spreading (right) for FeCrAl alloys before and after CHF test

(R. V. Umretiya, et. Al. J. of Nuc. Mat.. 541 (2020) 152420)

Experimental Results: CHF Test

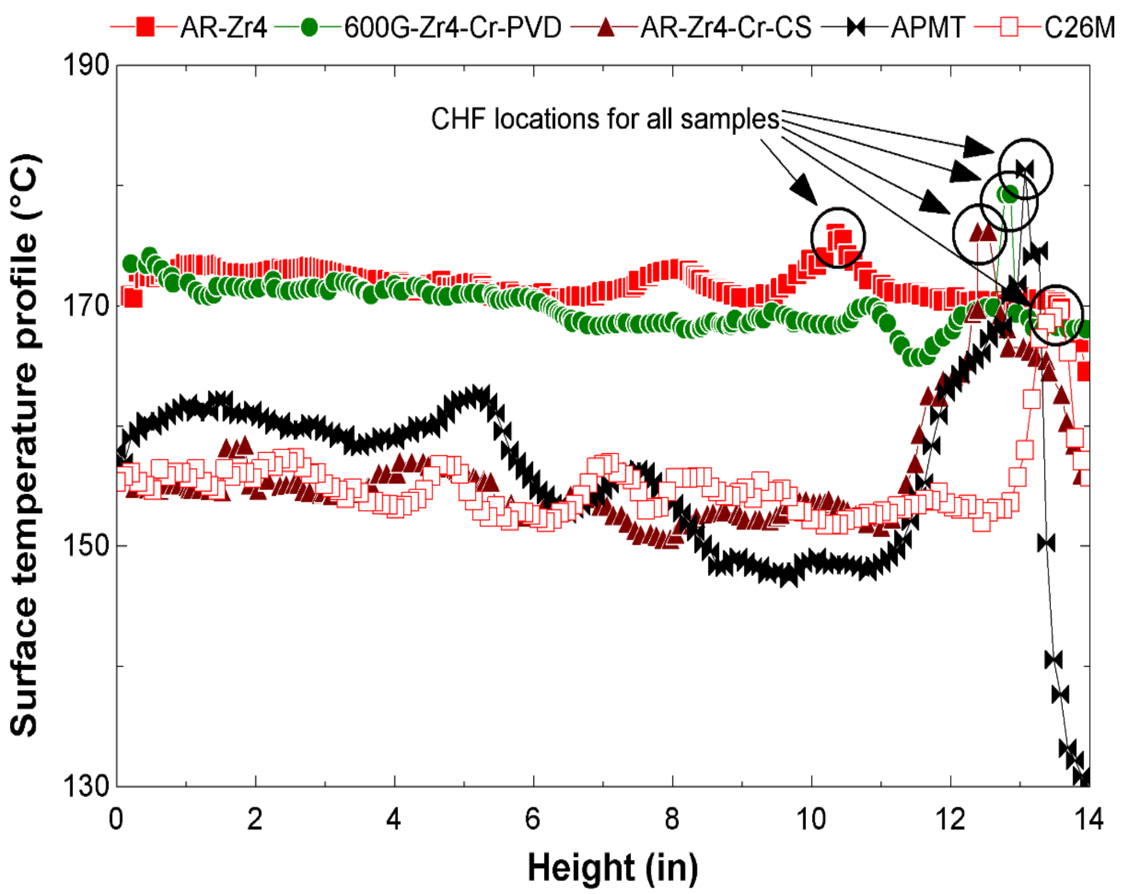


Figure 1. Comparison of experimental tube outer surface temperature for Zircaloy-4, both types of Cr-coatings, and FeCrAl alloys (all heat flux values are in MW/m²).

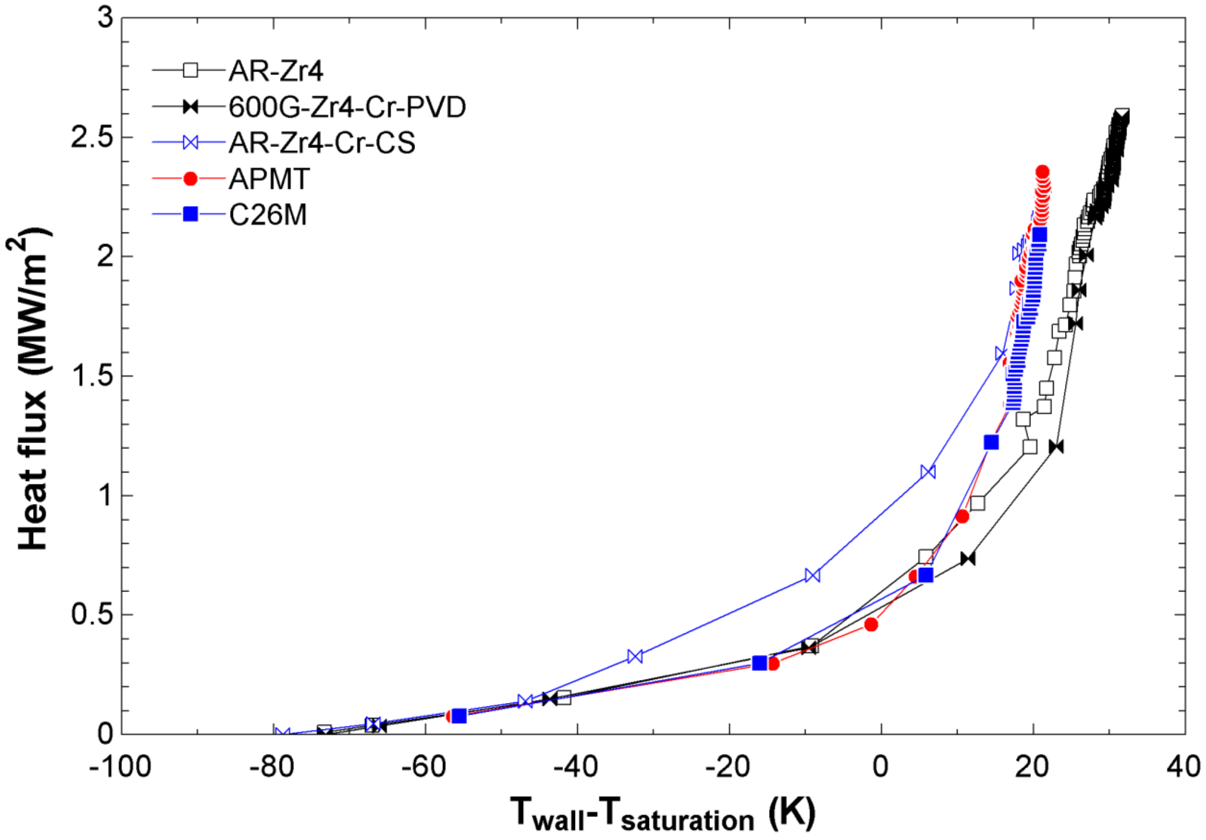
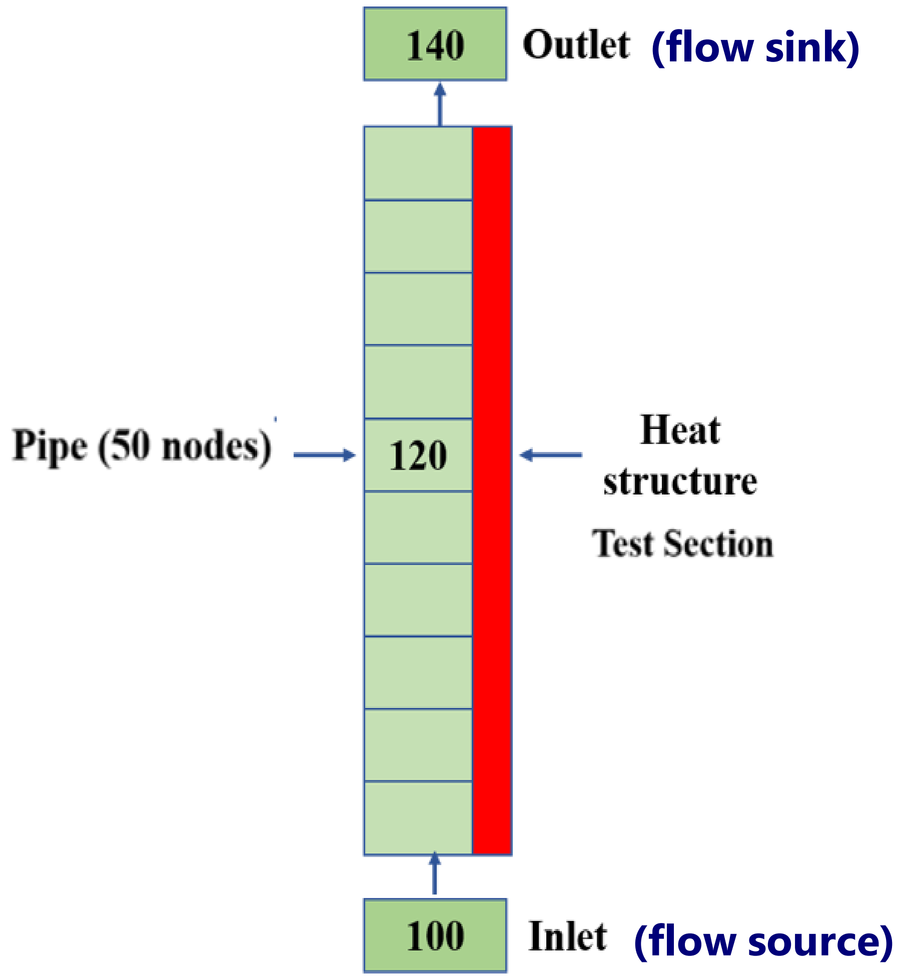


Figure 2. Boiling curves for the tested samples obtained based on the thermocouple's data.

Simulated Results: CHF Test



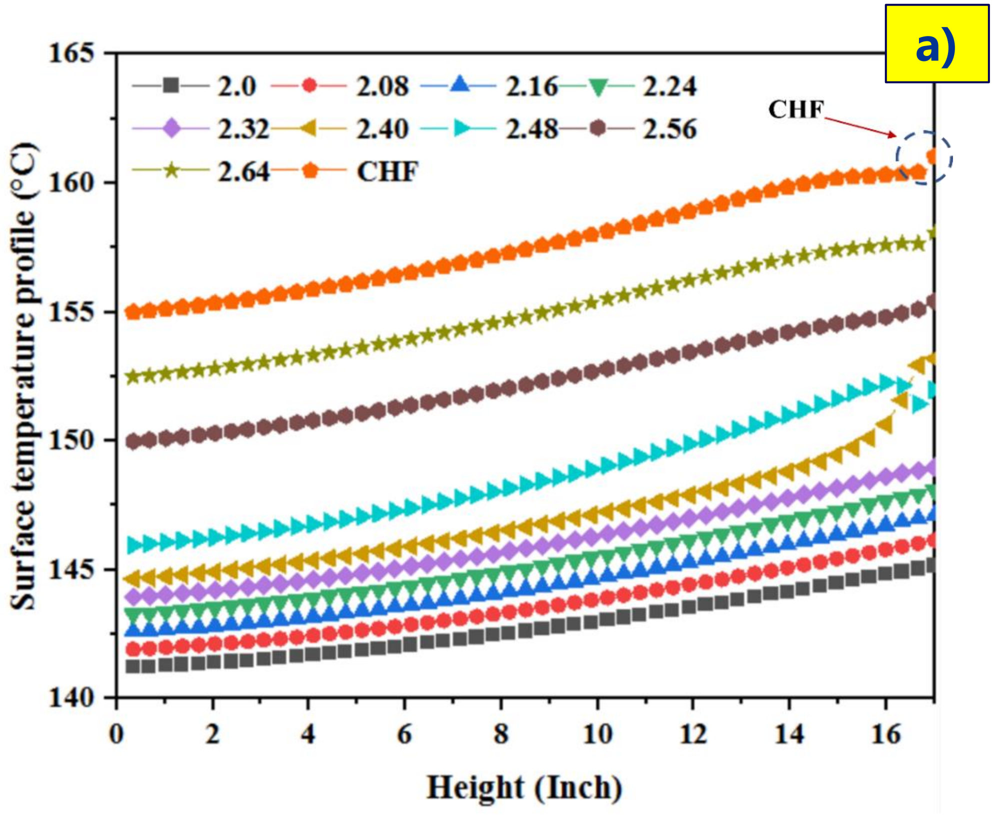
Nodalization diagram of RELAP5-3D model

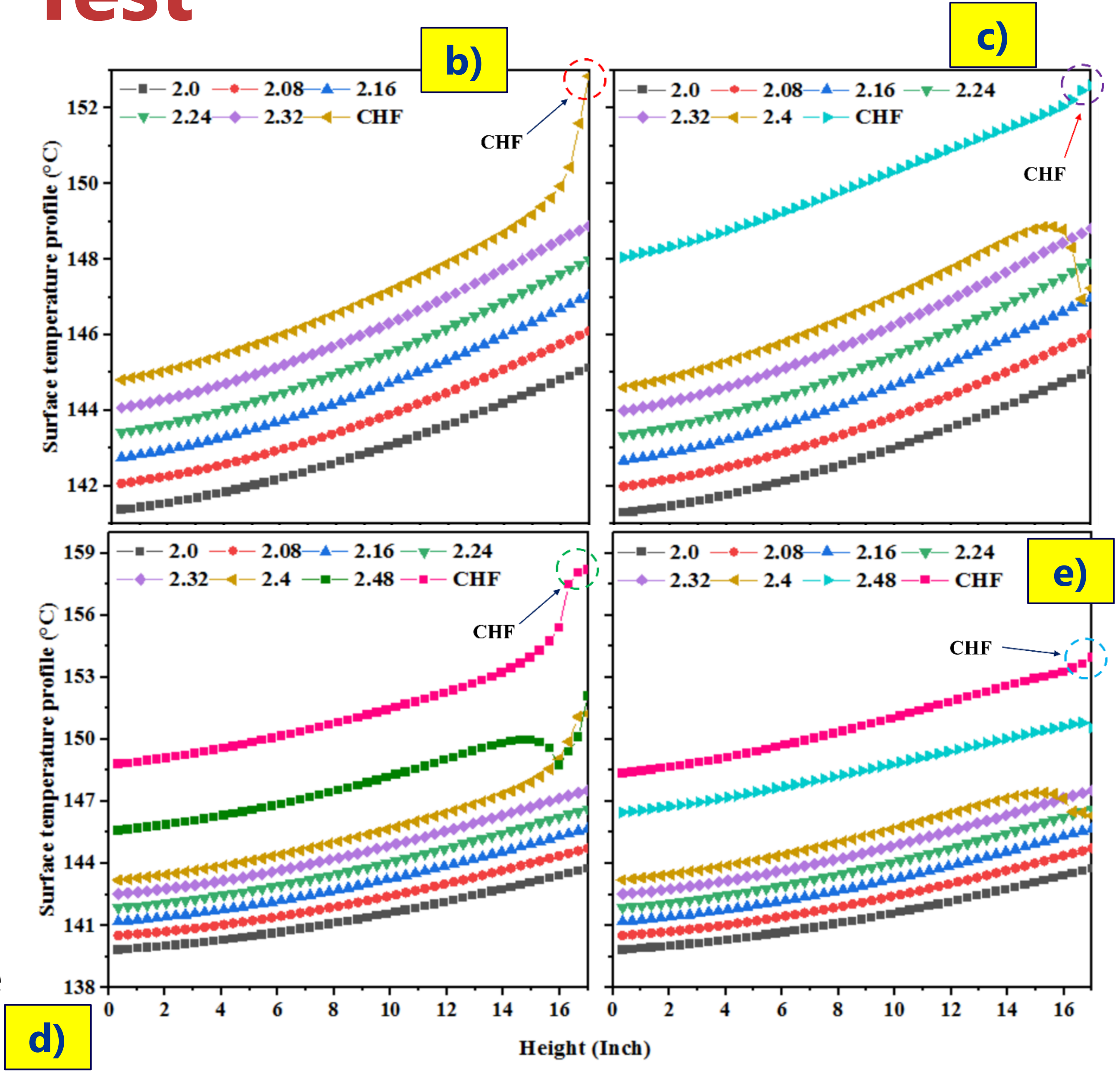
Summary of experimental and simulated CHF and PCT data for all samples

Material	CHF (MW/m^2)			PCT (°C)		
	Experimental	RELAP5-3D	Error (%)	Experimental	RELAP5-3D	Error (%)
AR-Zr4	2.60	2.72	4.62	175.86	161.05	-8.42
600G-Zr4-Cr-PVD	2.57	2.40	-6.61	179.35	152.85	-14.78
AR-Zr4-Cr-CS	2.28	2.48	8.77	176.25	152.6	-13.42
APMT	2.54	2.56	0.79	212	158.20	-25.38
C26M	2.26	2.56	13.27	212	153.95	-27.38

- A minimum timestep of 1×10^{-8} s was used for the dynamic simulations.
- The Groeneveld look-up table was used to predict CHF in the RELAP5-3D model.
- To better simulate the flow boiling CHF experiments, the Risk Analysis and Virtual Environment (RAVEN) was coupled with RELAP5-3D.

Simulated Results: CHF Test





Comparison of simulated tube outer surface temperature at different heat flux for (a) substrate Zircaloy-4; Cr-coated Zircaloy-4: PVD (600G-Zr4-Cr-PVD) (b) and Cold Spray (AR-Zr4-Cr-CS) (c); FeCrAl alloys: APMT (c) and C26M (d) (all heat flux values are in MW/m^2)

Summary and F~~ea~~uture work

- The surface properties of all samples were assessed in terms of surface topography, roughness characteristics, and wettability measurements
- CHF experiments using these cladding materials were conducted at atmospheric pressure within a flow boiling facility
- The RELAP5-3D simulation results of CHF and PCT were within reasonable error with experimental data
- Future work will include a transient heat flux model in RELAP5-3D which can also incorporate the effect of surface characteristics in CHF and HTC using existing correlations in the literature
- Furthermore, sensitivity analysis will be performed to investigate the impact of HTC, and material thermal properties on CHF and PCT

Thanks!! Questions?

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