Coated ATF 성능평가 및 설계기준 개발

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사고저항성 핵연료: 단기 및 장기 전략

한국원자력학회 핵연료 및 원자력재료 연구부회

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서울대학교
SEOUL NATIONAL UNIVERSITY

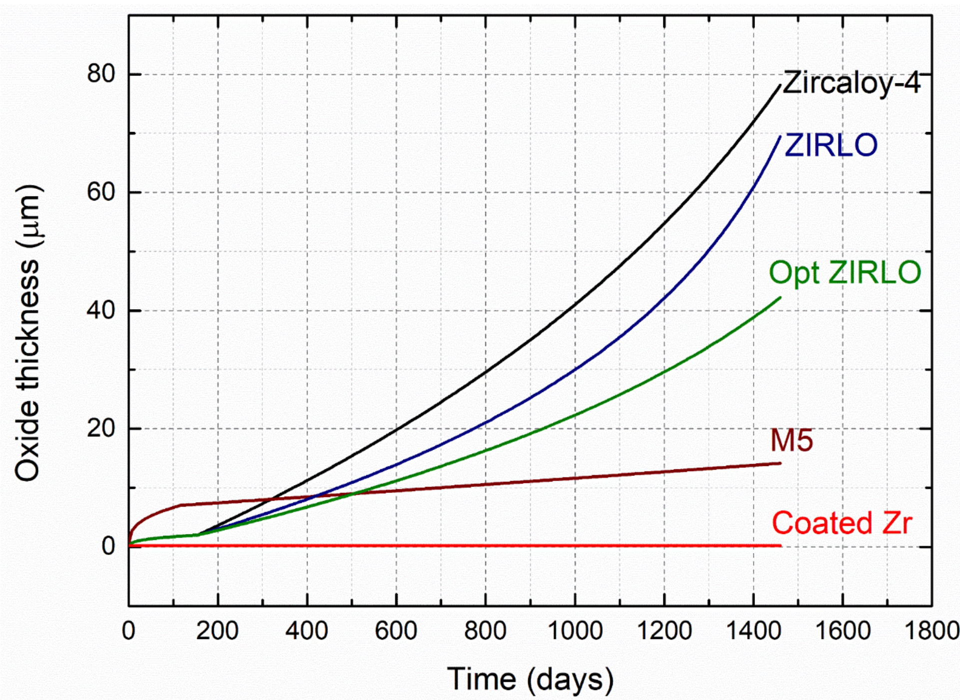
목차

Chapter 1

Coated ATF 성능평가 및 설계기준 개발

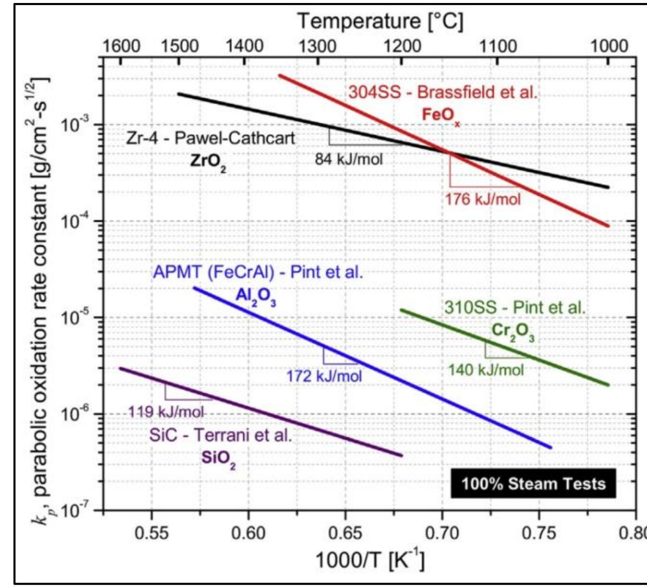
코팅 피복관의 기술적 의의

정상운전 내산화성



Zr 기반
피복관
개발 방향

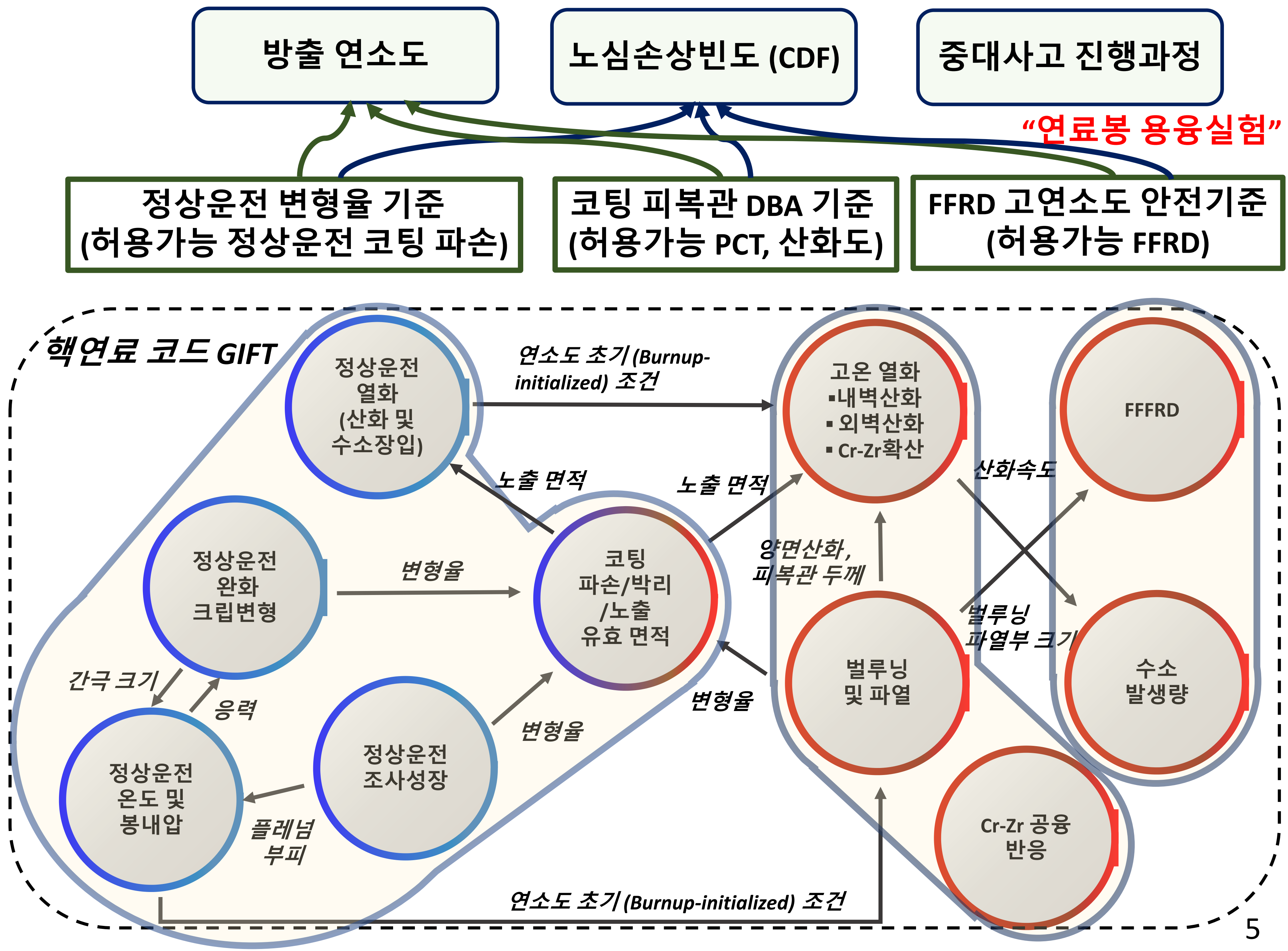
사고상황 (고온증기) 내산화성



기존 피복관 개발의 연장선

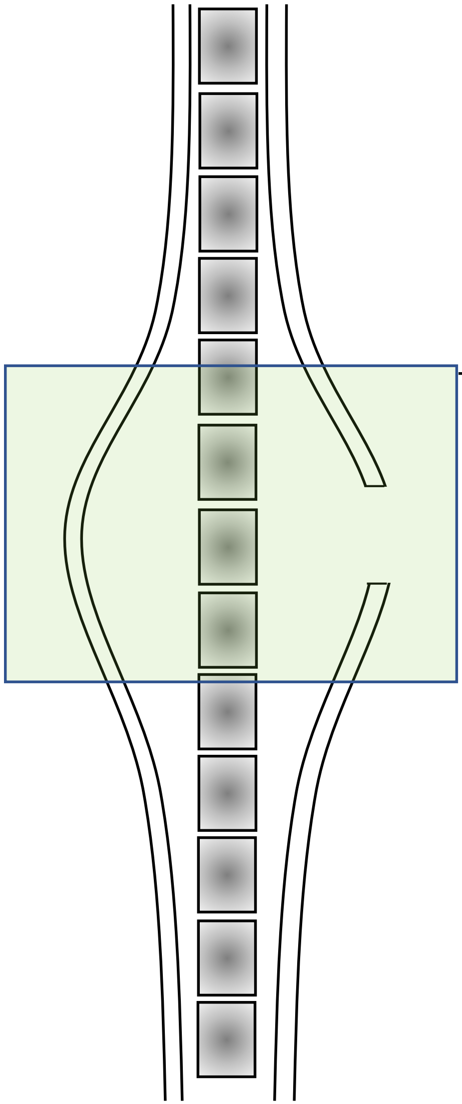
새로운 (경험해보지 못한) 성능

- **경제성 :** 연소도 증진에 따른 경제성 향상 여부
- **안전성:** 노심손상빈도 (CDF) 영향과 중대사고진행 완화 여부
- **지속가능성:** 연소도 증진에 따른 사용후핵연료 배출량 감소
  열화 현상 완화에 따른 사용후핵연료 관리비용 감소



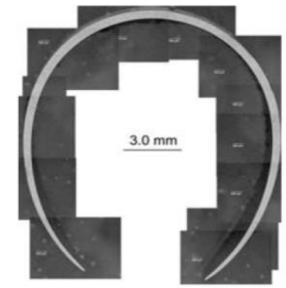
방출 연소도
노심손상빈도 (CDF)
중대사고 진행과정
"연료봉 용융실험"
정상운전 변형율 기준
(허용가능 정상운전 코팅 파손)
코팅 피복관 DBA 기준
(허용가능 PCT, 산화도)
FFRD 고연소도 안전기준
(허용가능 FFRD)
핵연료 코드 GIFT
정상운전 열화 (산화 및 수소장입)
연소도 초기(Burnup-initialized) 조건
고온 열화
▪내벽산화
▪ 외벽산화
▪ Cr-Zr확산
FFFRD
노출 면적
노출 면적
산화속도
정상운전 완화 크립변형
변형율
코팅 파손/박리 /노출 유효 면적
양면산화, 피복관 두께
벌루닝 파열부 크기
간극 크기
응력
정상운전 온도 및 봉내압
정상운전 조사성장
변형율
변형율
벌루닝 및 파열
수소 발생량
플레넘 부피
Cr-Zr 공융 반응
연소도 초기(Burnup-initialized) 조건

Post-Quench ductility near the burst hole: a key DBA limit



Exacerbated embrittlement near the burst region

① Double side oxidation



② Thinning of the wall

$$ECR_{double-sided} = 87.8 * \frac{\Delta W_{CP}}{\delta_{avg,deformed}}$$

NUREG 2119, U.S NRC

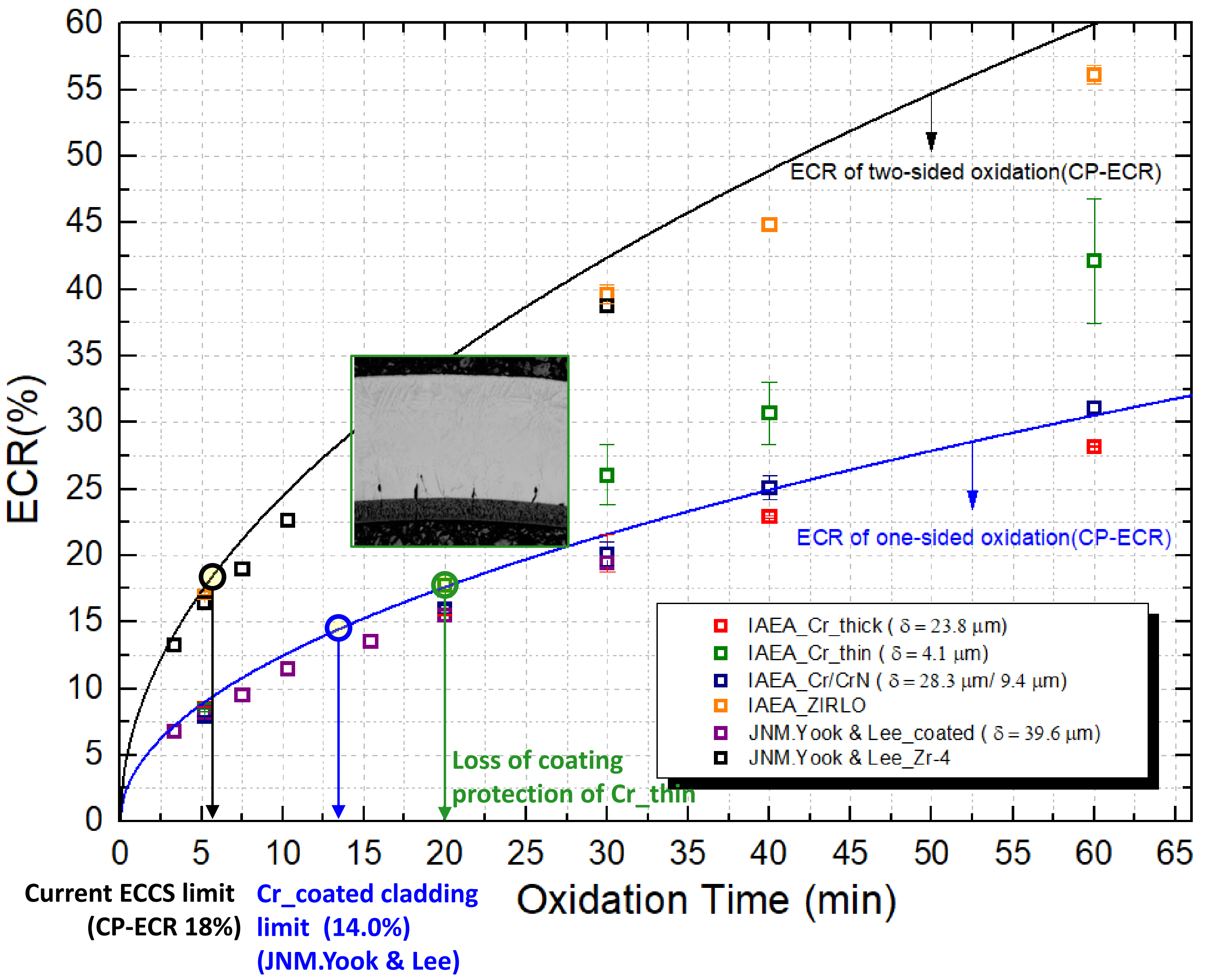
Therefore, the Office of Nuclear Regulatory Research recommendation for how to treat the ballooned region is that the time-at-temperature limit developed based on ring-compression data to limit oxidation is applied uniformly to the entire rod, with the provisions for the balloon outlined in the existing rule to use the average wall thickness in the rupture region to calculate the CP-ECR.

ATF-ISG-2020-01, U.S NRC

Differences in oxidation kinetics between zirconium-based cladding and chromium-coated cladding change the relationship between oxygen diffusion and oxide growth. This issue is further complicated within the burst region where cladding inner diameter oxidation is based on zirconium alloy kinetics and cladding outer diameter oxidation would be based on chromium coating kinetics. Hence, the applicability of the 17-percent equivalent cladding reacted analytical limit and, more generally, the use of maximum local oxidation as a surrogate SAFDL for cladding embrittlement is questionable.

The NRC staff should ensure that, if the applicant elects to ignore the potential benefits expected with chromium coatings and continues to use the existing 10 CFR 50.46 analytical limits, then supporting evidence has been provided to demonstrate residual ductility of the coated cladding up to these analytical limits.

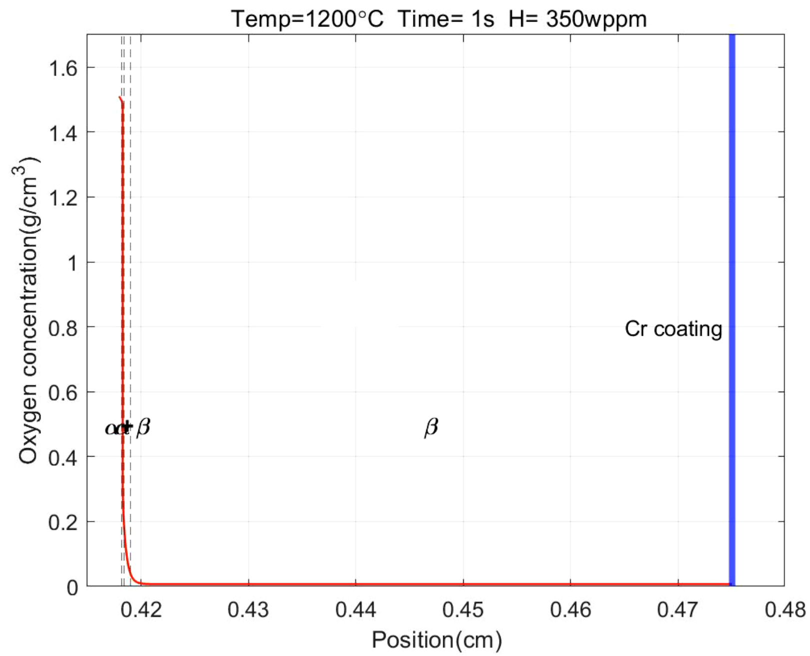
Coated cladding: effectively single side oxidation for DBA consideration

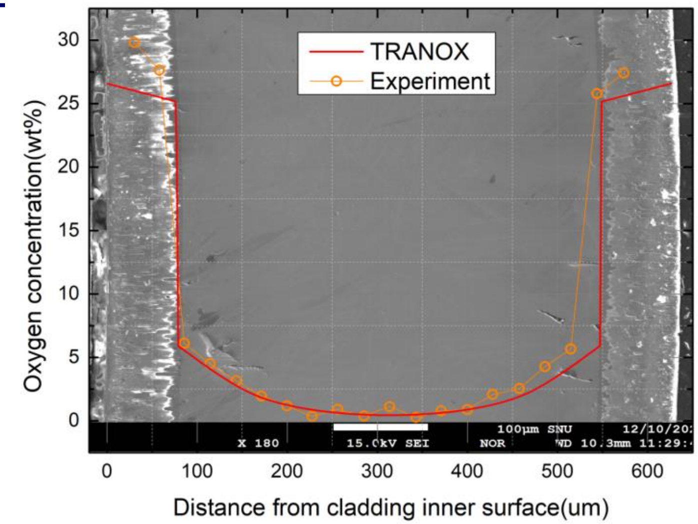


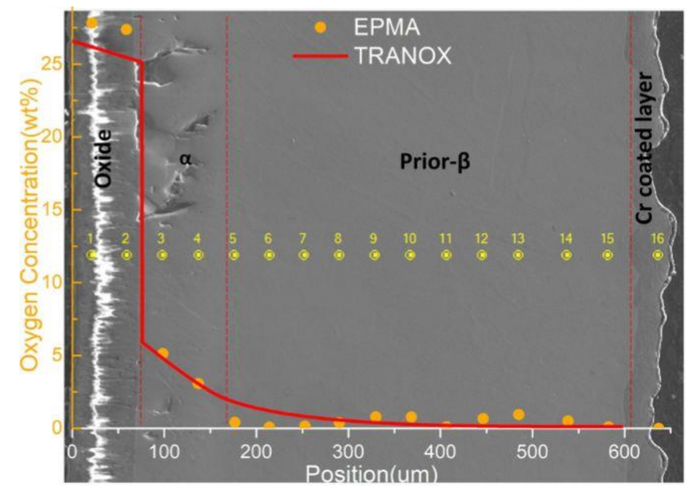
TRANOX : Mechanistic model of Zircaloy oxidation

- **TRANOX Capability**

- **Oxygen distribution, ECR, alpha and beta phase thickness**
- **Transient temperature**
- **Presence of hydrogen (high burnup)**
- **Pre-transient oxide layer (high burnup)**
- **Coated Cladding**





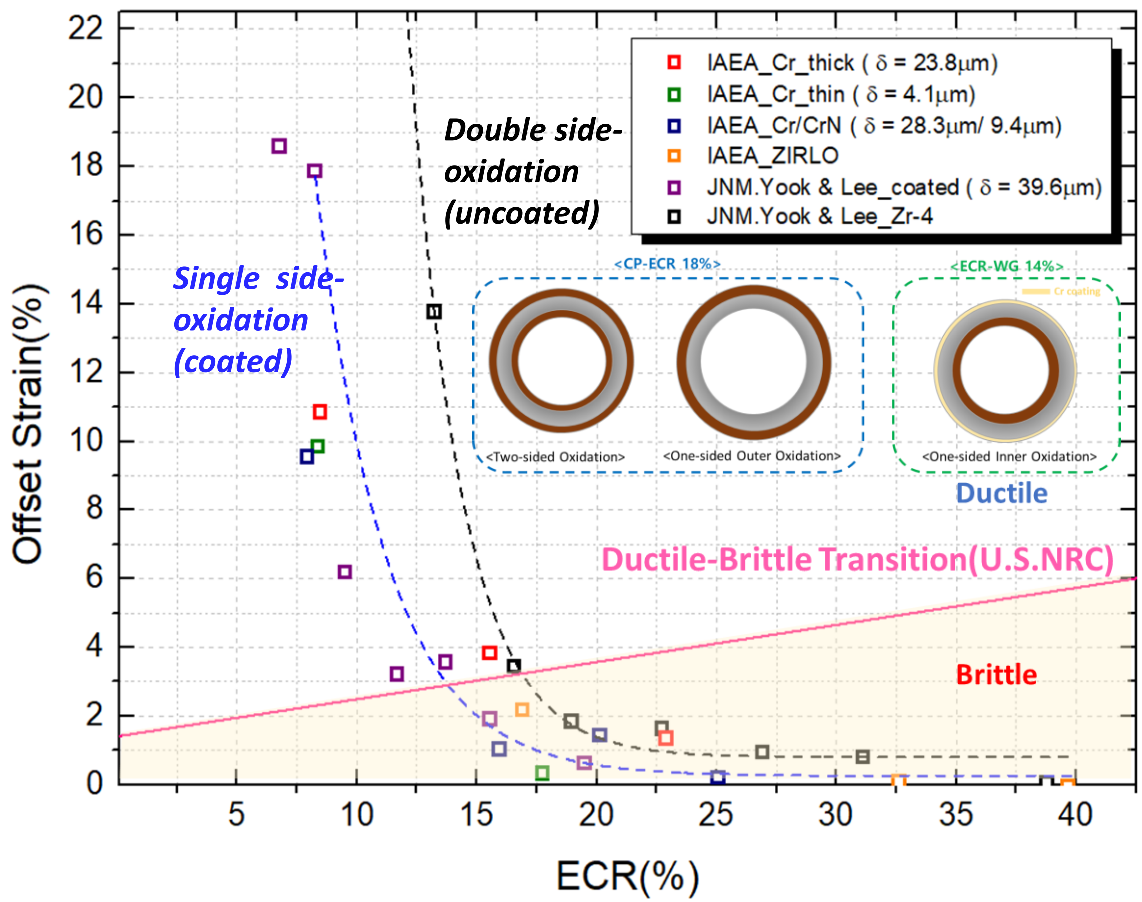


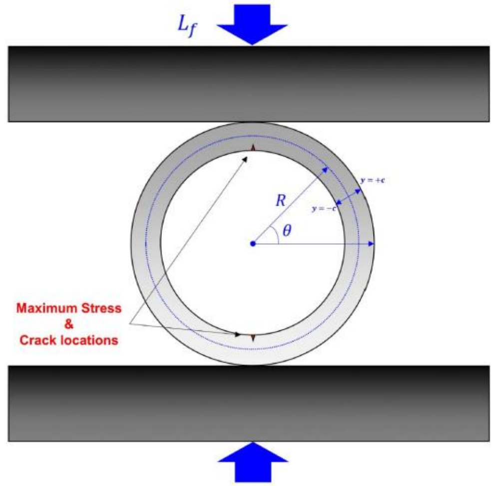
[1] 2021. D. Kim et al., TRANOX: Model for Non-Isothermal Steam Oxidation of Zircaloy Cladding. *Journal of Nuclear Materials* , 556, 153153.

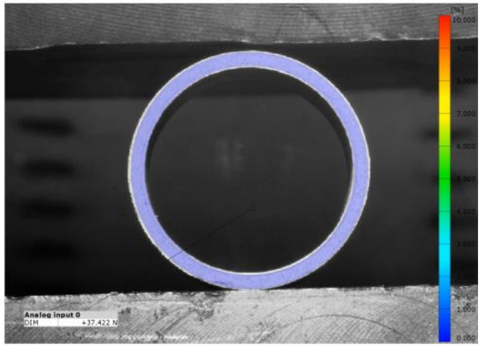
[2] 2022. H. Yook, et al.,. Post-LOCA Ductility of Cr-coated cladding and its embrittlement limit. *Journal of Nuclear Materials*, 558, 153354.

[3] 2022. D. Kim et al., Study of high burnup effect on steam oxidation of Zircaloy and its regulatory implications via the development of pre-transient oxide model of TRANOX. *Journal of Nuclear Materials*, *in press*.

Result: Ring Compression Tests



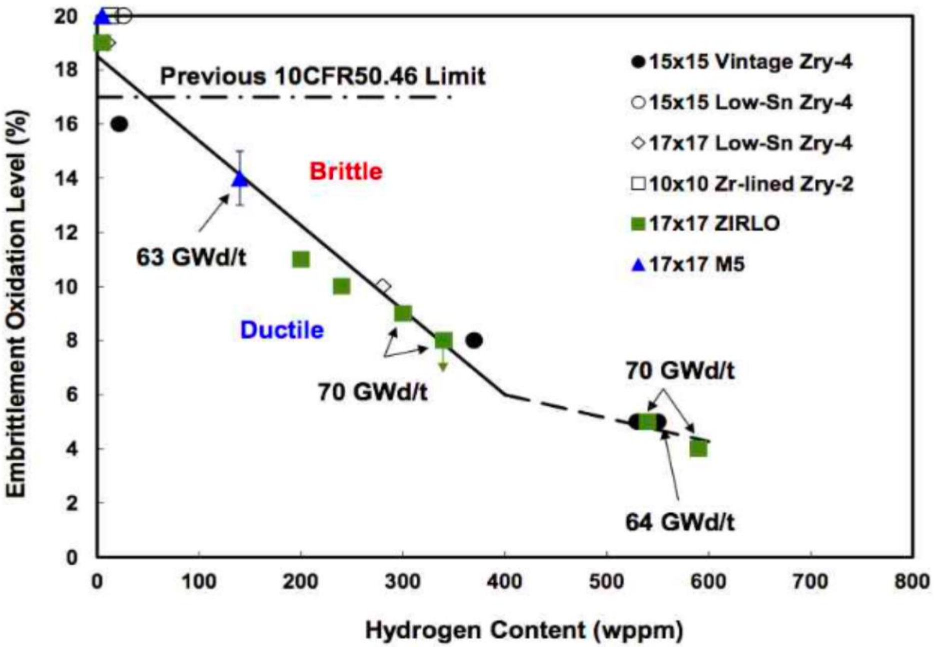




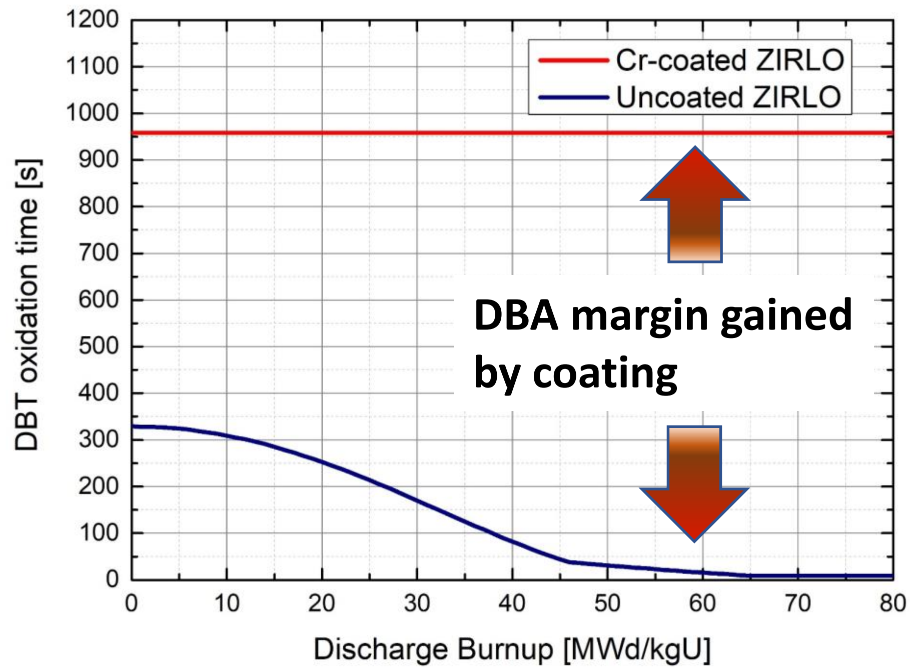
Key deliverables:

- Ductility limit is reached by inner side oxidation. Hence it is insensitive to coating manufacturing process.
- Nature of RCT biasedly lowers the ECR limit for coated cladding: the lower ECR limit for coated cladding is not due to the ductility carrying phase (prior- β). Even with thicker prior- β phase, coated cladding undergoes premature ductile to brittle transition under RCT. This is primarily because it has thicker brittle phases (ZrO_2 + α-Zr(O)) at the inner surface compared to uncoated cladding for the same ECR

Greater benefit for higher burnup



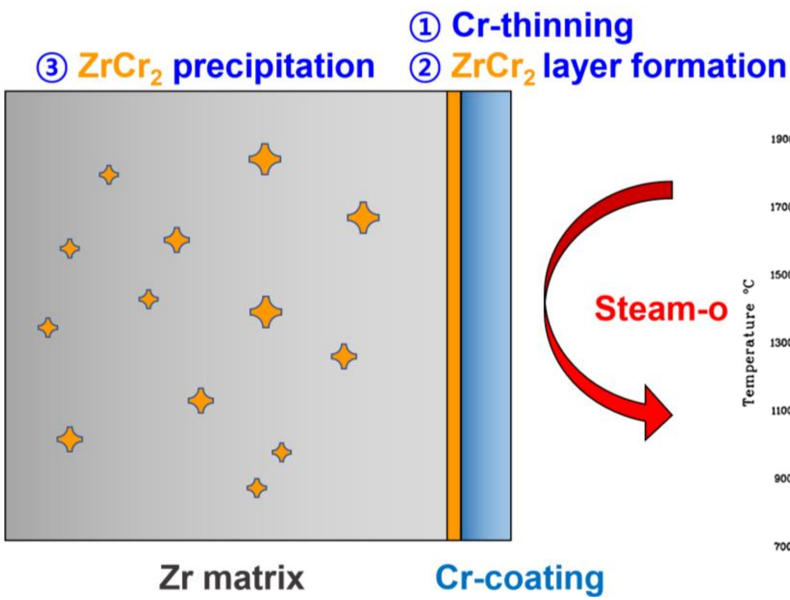
<Decreasing allowable ECR limit with $Bu_d\ (H)$>, U.S. NRC

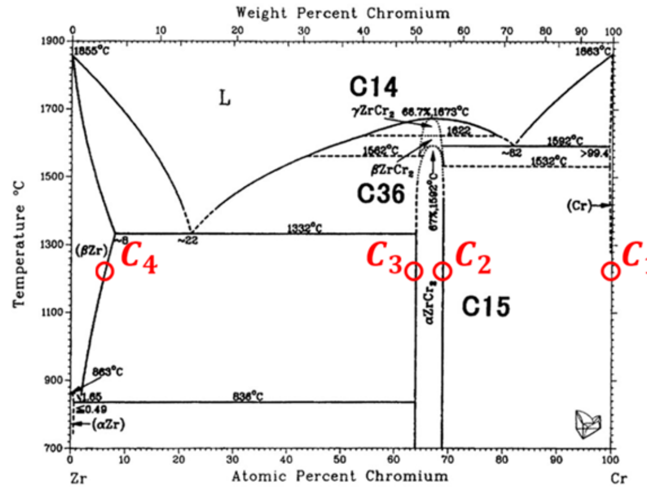


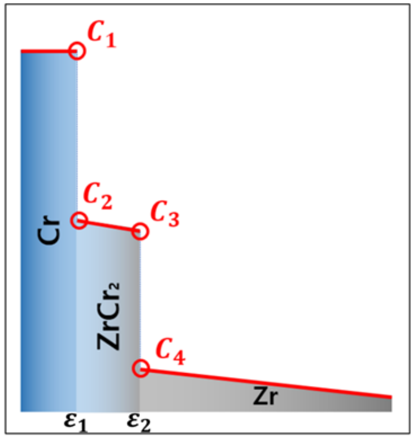
<Time required to reach ductile to brittle transition (DBT) by exposure to 1200 ℃ steam, Cr-coated ECR Limit: 14%
Uncoated ECR Limit: decreasing with Bu_d>

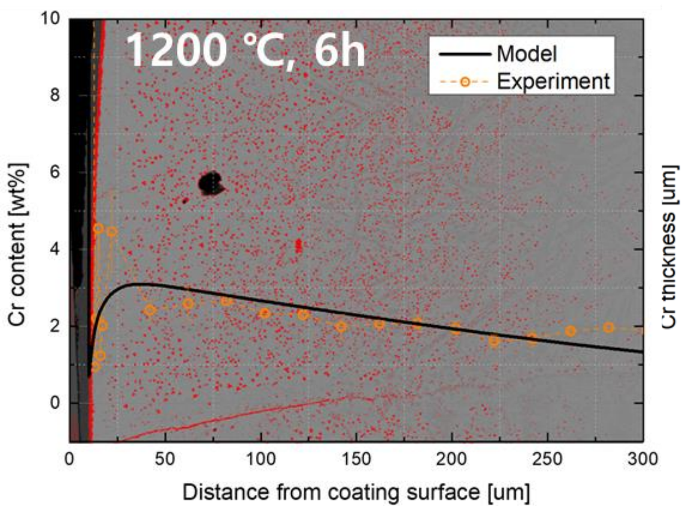
- **Dramatic retention of embrittlement margin with burnup :**
  Applying the draft rule of the U.S NRC, burnup extension is practically impossible. Successful coated cladding, however, is expected to retain embrittlement margin at high burnup, providing a basis for future burnup extension.

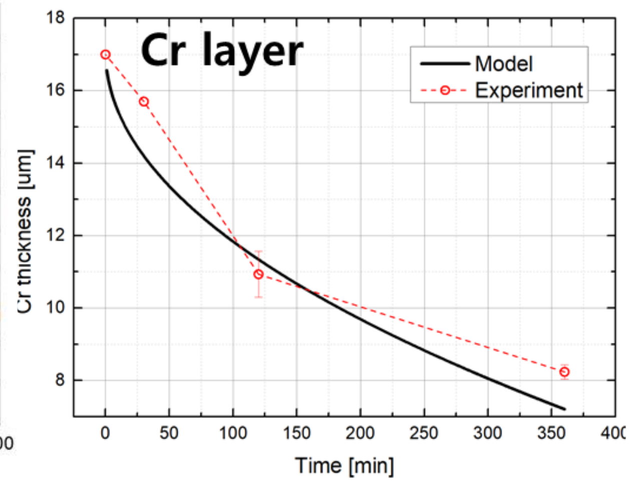
Extra cladding embrittlement mechanism via Cr-Zr diffusion

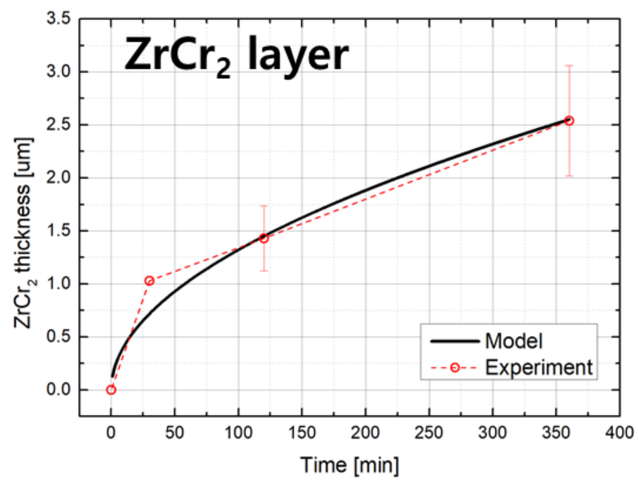






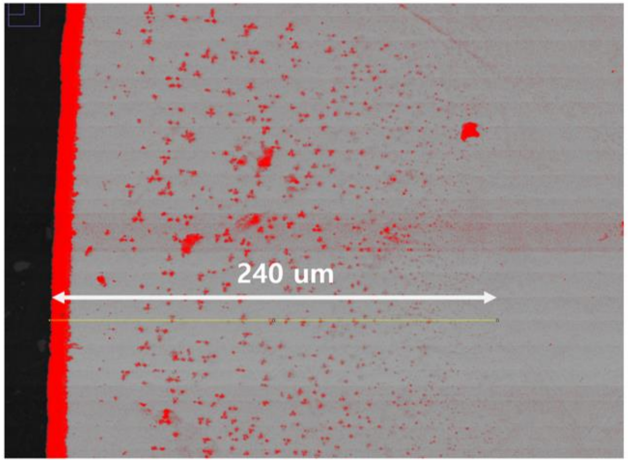


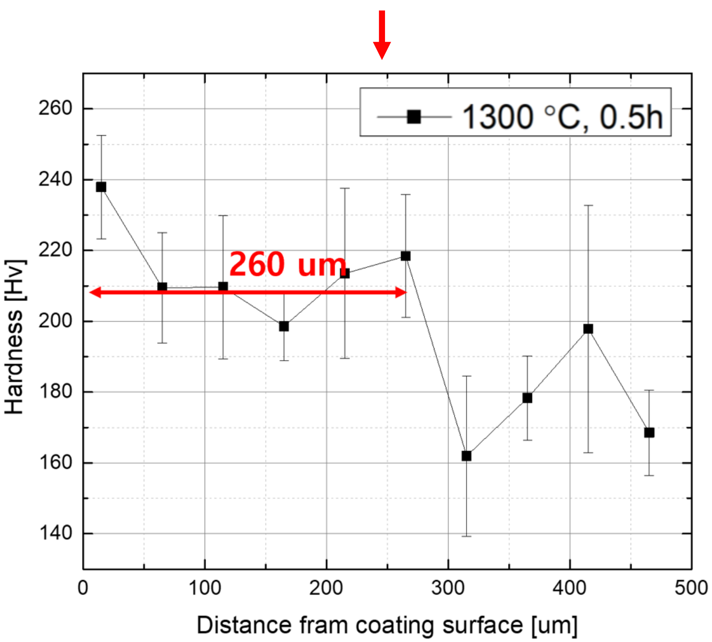




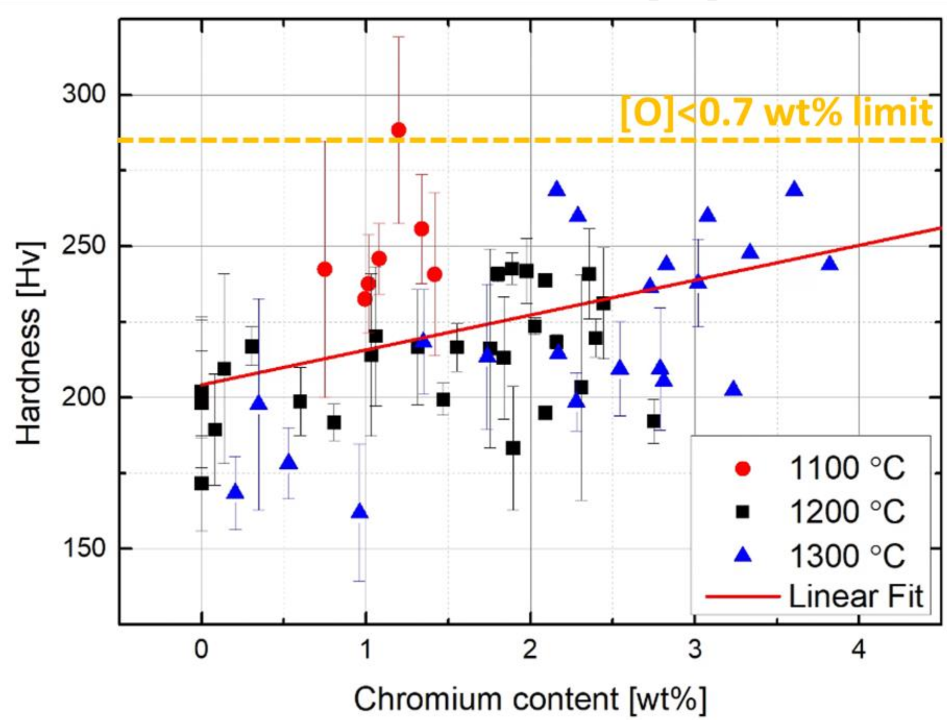
<Cr-Zr diffusion experiments and TRANOX modeling>

Sign of cladding embrittlement





$$Hardness = 11.535 * [Cr] + 205$$



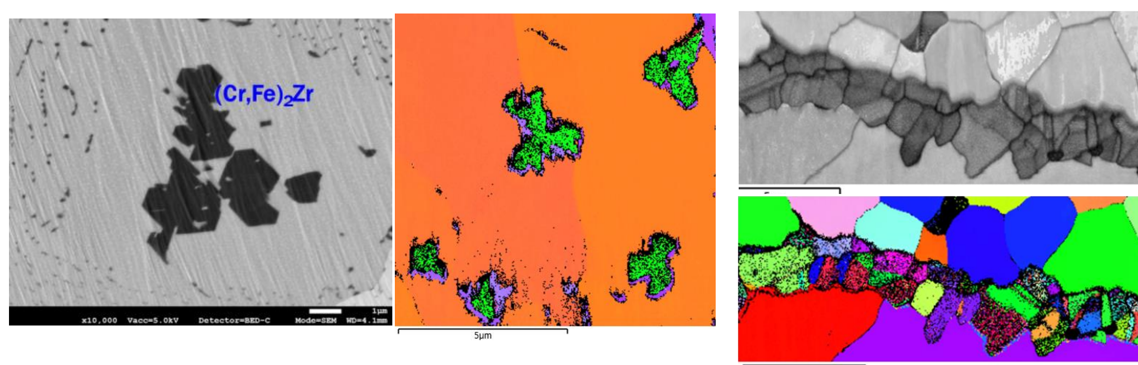
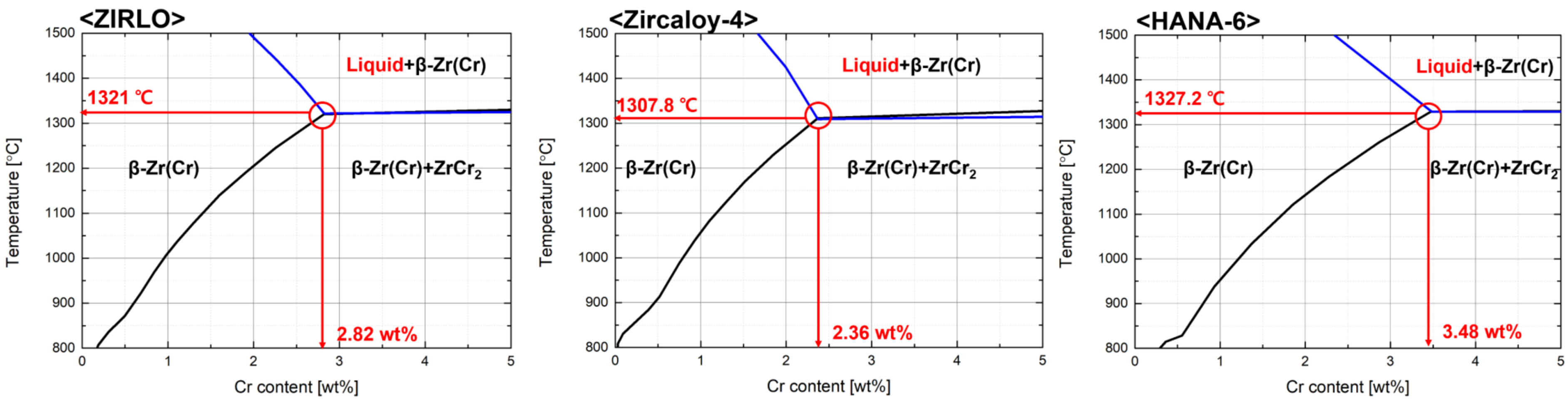


Fig. Relation between hardness and precipitation

Eutectic reaction of Zr-alloys with Cr

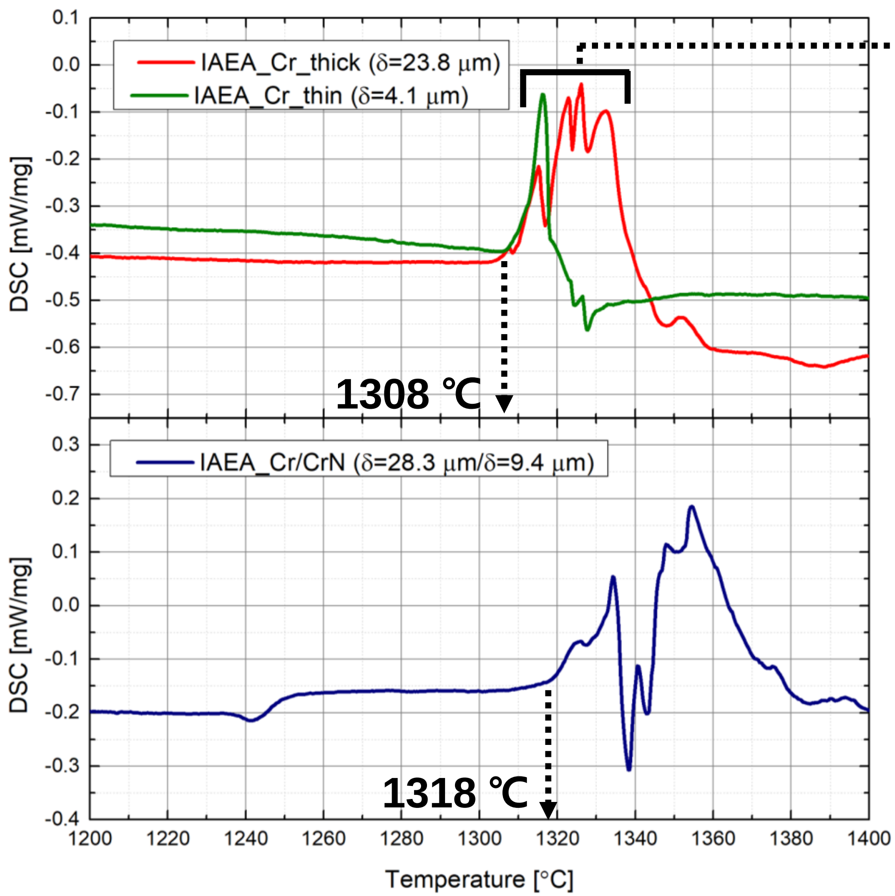


< Part of Zr-Cr phase diagram of Zr-alloys obtained by Thermo-Calc (TCZR1 database: Cr, Fe, H, Nb, Ni, O, Sn, Zr) >

< Table. Predicted eutectic temperature and Cr content of Zr-alloys obtained by Thermo-Calc >

	ZIRLO	Zircaloy-4	HANA-6
Composition [wt%]	Zr(bal)-0.5Sn-0.11Fe-Cr-0.11O-0.98Nb	Zr(bal)-1.45Sn-0.21Fe-Cr-0.12O	Zr(bal)-Cr-1.1Nb
Predicted eutectic temperature [℃]	1321.9	1307.8	1327.2
Predicted eutectic Cr content [wt%]	2.83	2.36	3.48

DSC results

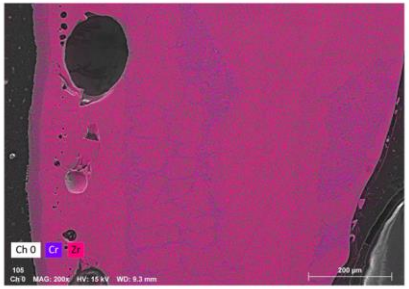


<DSC Curve of coated ZIRLO >

Larger peak area for thick coating

- Target temperature : **1400 ℃**
- Heating rate : **20 ℃/min**

<Surface change after eutectic reaction (Cr/CrN coated ZIRLO)>



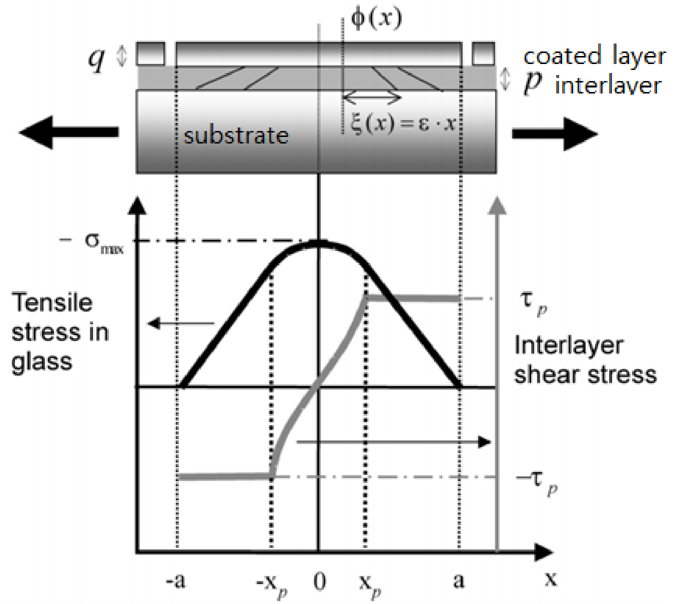
< Table. Eutectic temperature and peak area of coated ZIRLO >

	Theoretical	Cr_thick	Cr_thin	Cr/CrN
Eutectic temperature [℃]	1321.9	1308.4	1308	1318
Eutectic peak area [J/g]	-	26.31	7.59	26.04

Coated Cladding Crack Analysis: Shear-lag model

Shear-lag model

- Shear-lag model was first presented by O. Volkersen in 1938 and is one of the most basic concepts in load transfer between two adherends physically connected through an adhesive.
- The shear-lag model is used to explain the stress relationship between the coating layer and the substrate layer.
- The crack density of the coated cladding can be calculated from seven major parameters.



<Shear-lag model used in coated cladding stress analysis>

$$\rho = \frac{1}{L} = \frac{\tau_b}{2Ed_1}\left[Acosh\left(arsinh\left(\frac{B}{-A}\right)\right) + Barsinh\left(\frac{B}{-A}\right) + \epsilon + \frac{\sigma_r}{E}\right]^{-1}$$

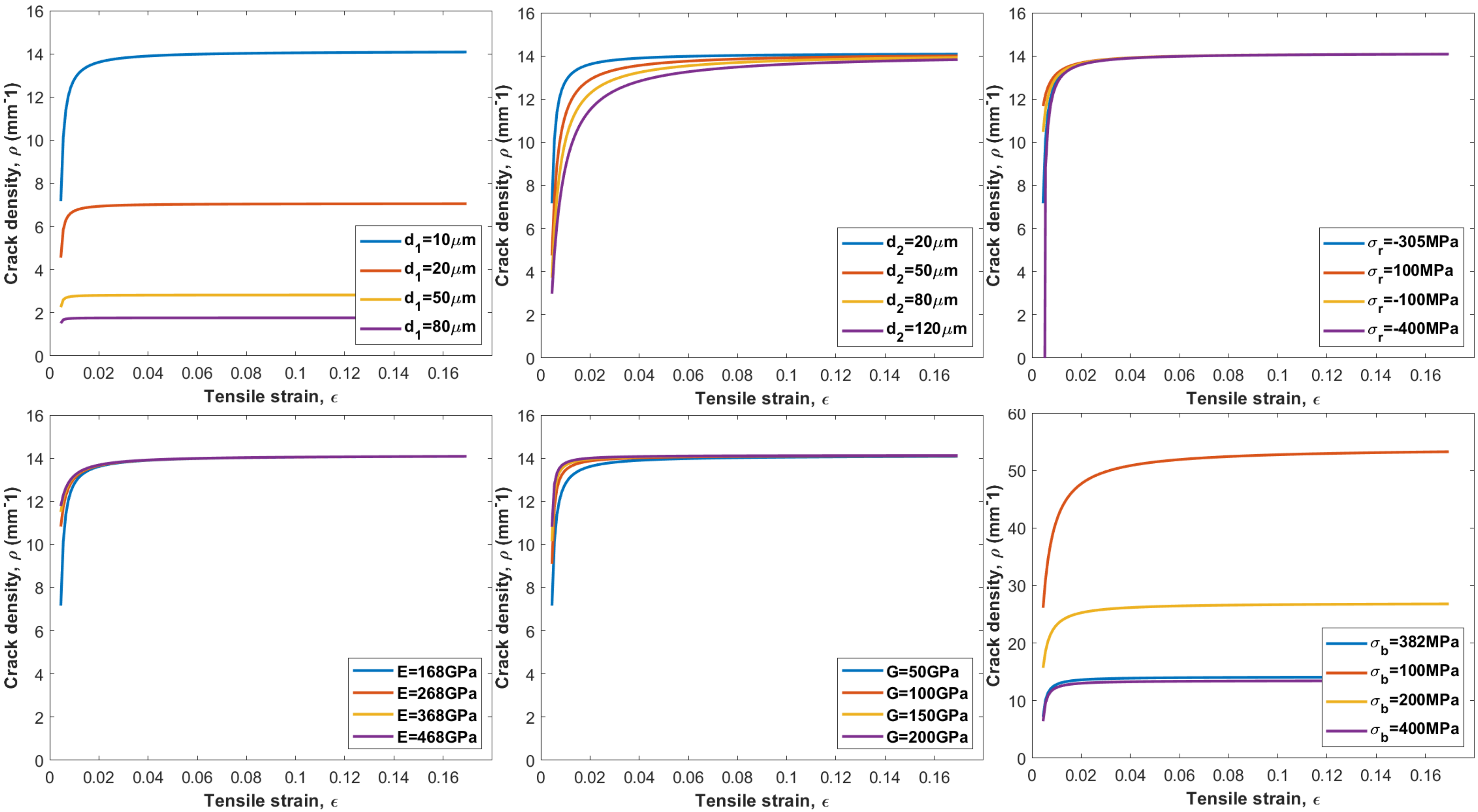
$$A = \frac{\sigma_b - \sigma_r}{E} - \epsilon,\ B = \tau_b\sqrt{\frac{d_2}{GEd_1}}$$

$\rho^* = \frac{\tau_b}{2d_1\sigma_b}$, ρ^*: saturation crack density

<Table. Major parameters and measurement methods>

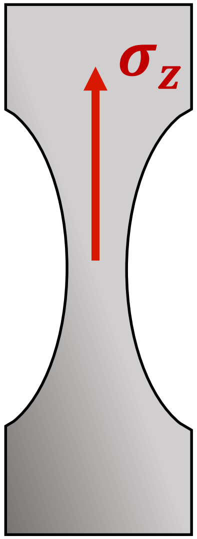
Parameters	Unit	Measurement Method
d_1	**μm**	**Microscopy**
d_2	μm	Theoretical evaluation
E	GPa	Nanoindentation
G	GPa	Tensile test
σ_r	MPa	XRD
σ_b	MPa	In-situ tensile test
τ_b	MPa	In-situ tensile test

Coated Cladding Crack Analysis: Preliminary results



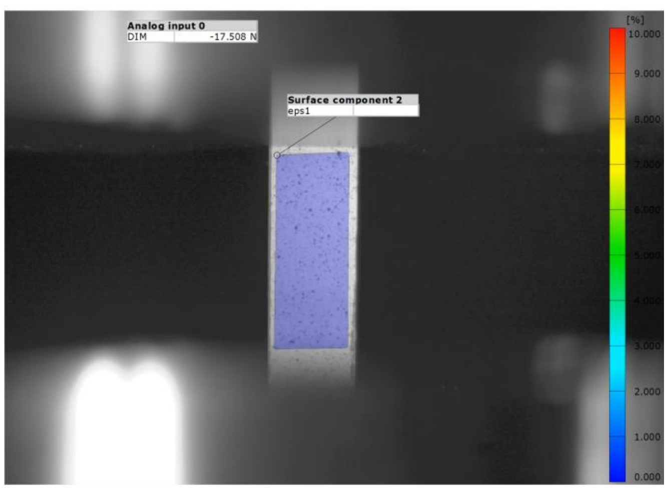
d_1: Coated layer thickness, d_2: Interlayer thickness, E: Elastic modulus, G: Shear modulus, σ_r: Residual stress in the as-received coating, σ_b: Fracture strength of coating

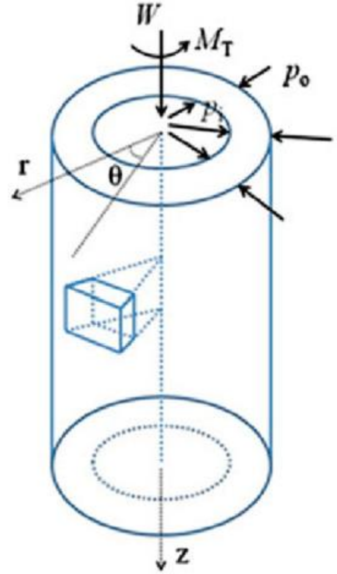
Experiments: Cr-coated cladding crack test (DIC video)



① 1D shear-lag model:

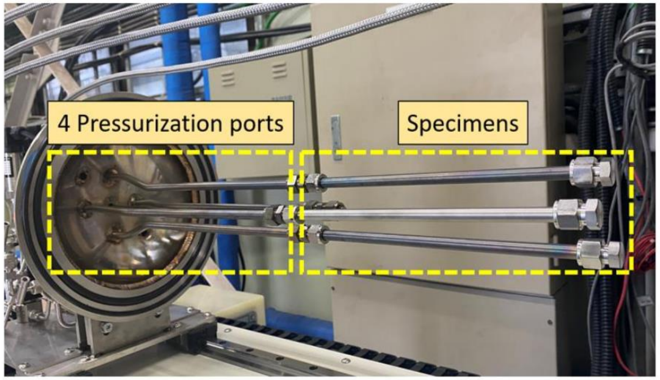
Experimentally obtaining Cr-specific parameters

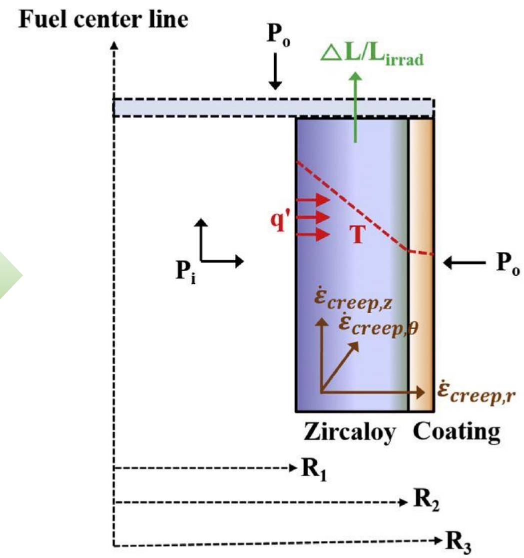




② Expanding to 3D stress states of Cr-coated cladding

Experimentally obtaining Cr-specific parameters

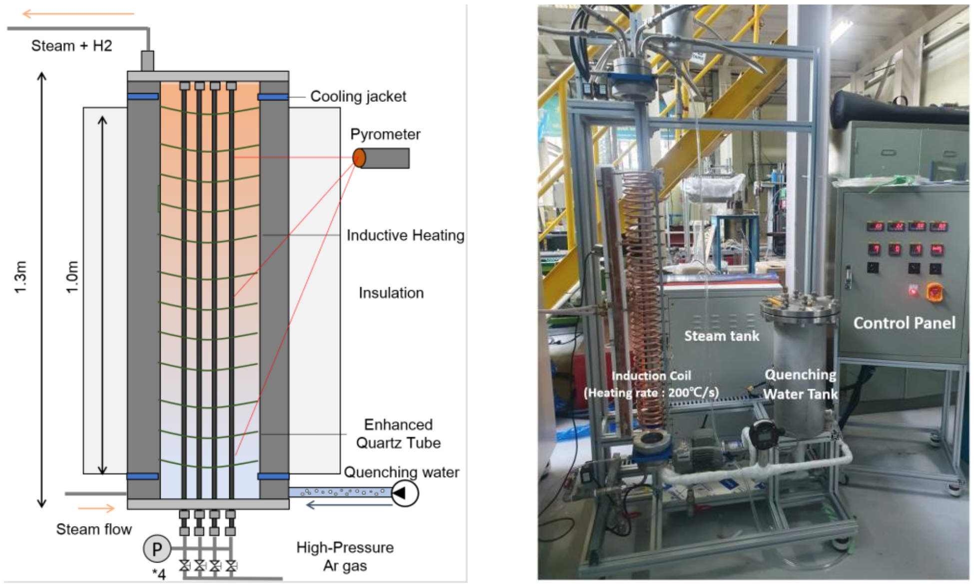




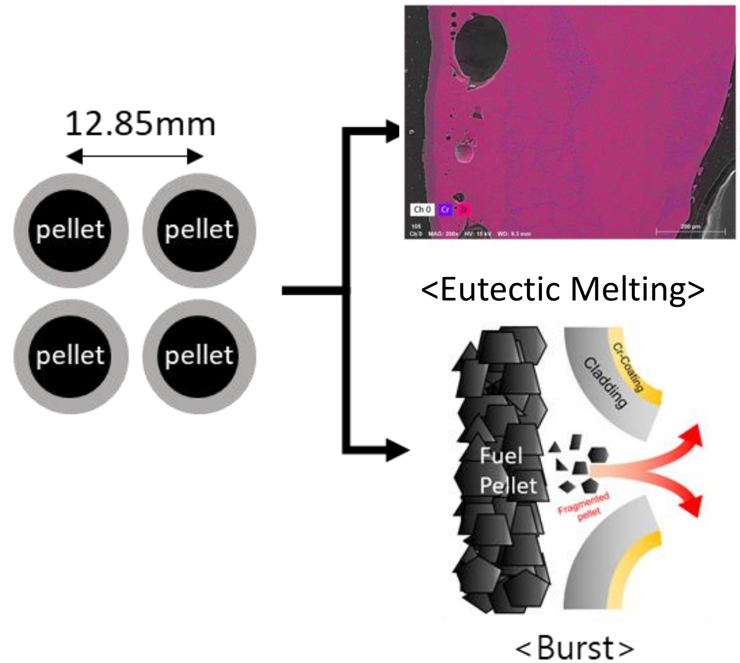
③ Implementing in the fuel performance code

Integral LOCA Experiment

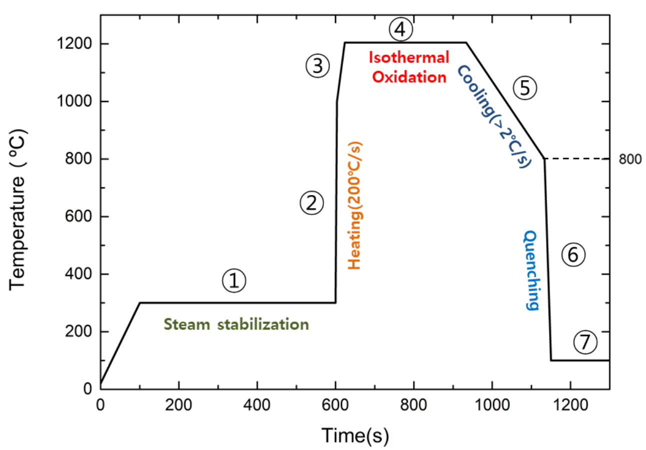
Integral LOCA Facility



<Schematic and figure of facility>



<Integral LOCA tests enabling Fuel rod burst and Melting >

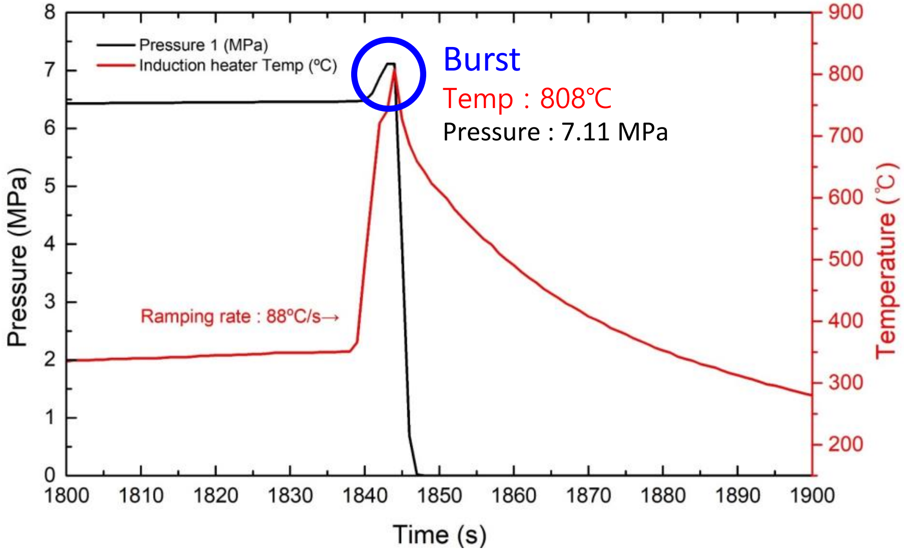


<LOCA experiment procedure>

- Integral LOCA Experiment of test section 1m
- High heating rate(~200℃/s) and maximum temperature through induction heating
- FFRD simulation through Al_2O_3 or ZrO_2 pellet insertion
- 4 rod experiment simulating subchannel of reactor

Cr-coated Cladding Integral LOCA Experiment

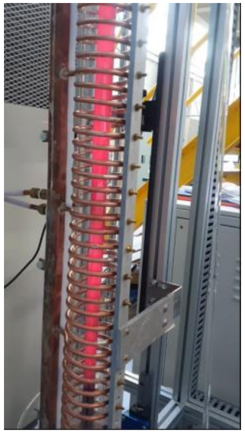
Cladding Ballooning & Burst Experiment



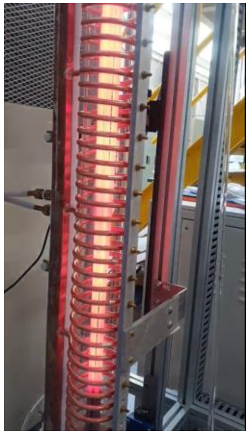
<Temperature and pressure change in experiment>

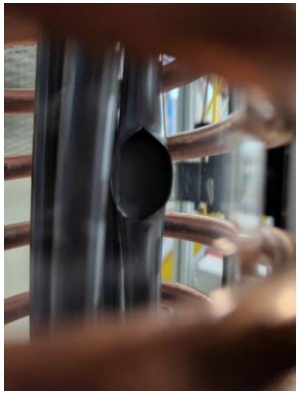
1840s

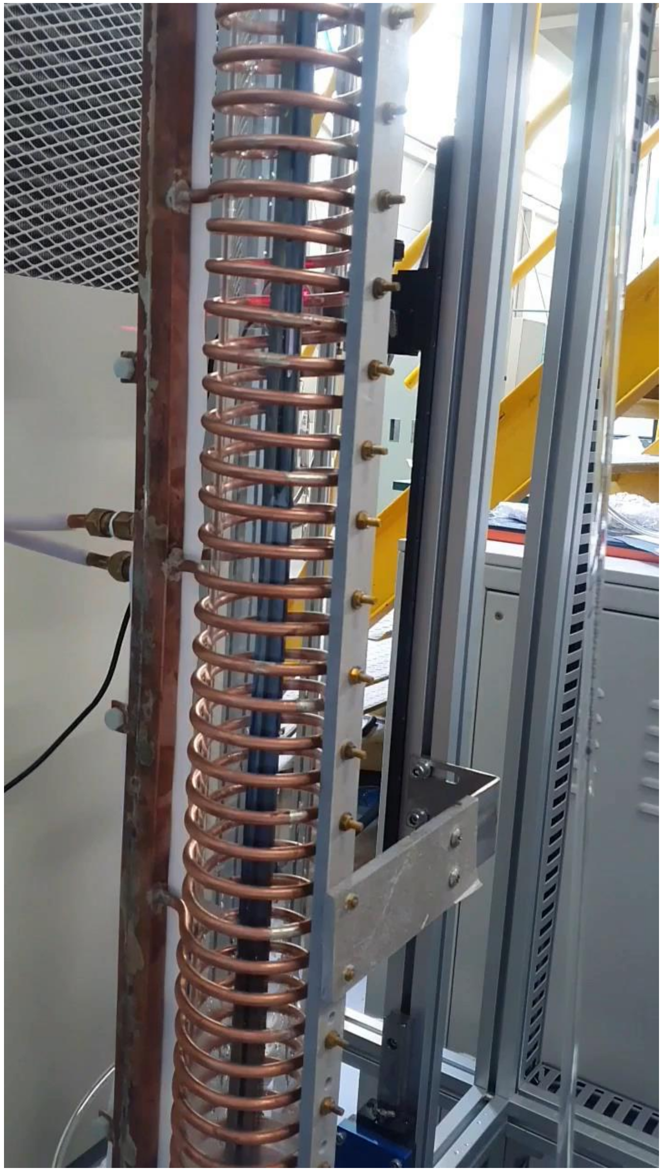
1842s



1844s







<Temperature and structure change of cladding> <Bursted cladding>

GIFT 코드 목표 성능

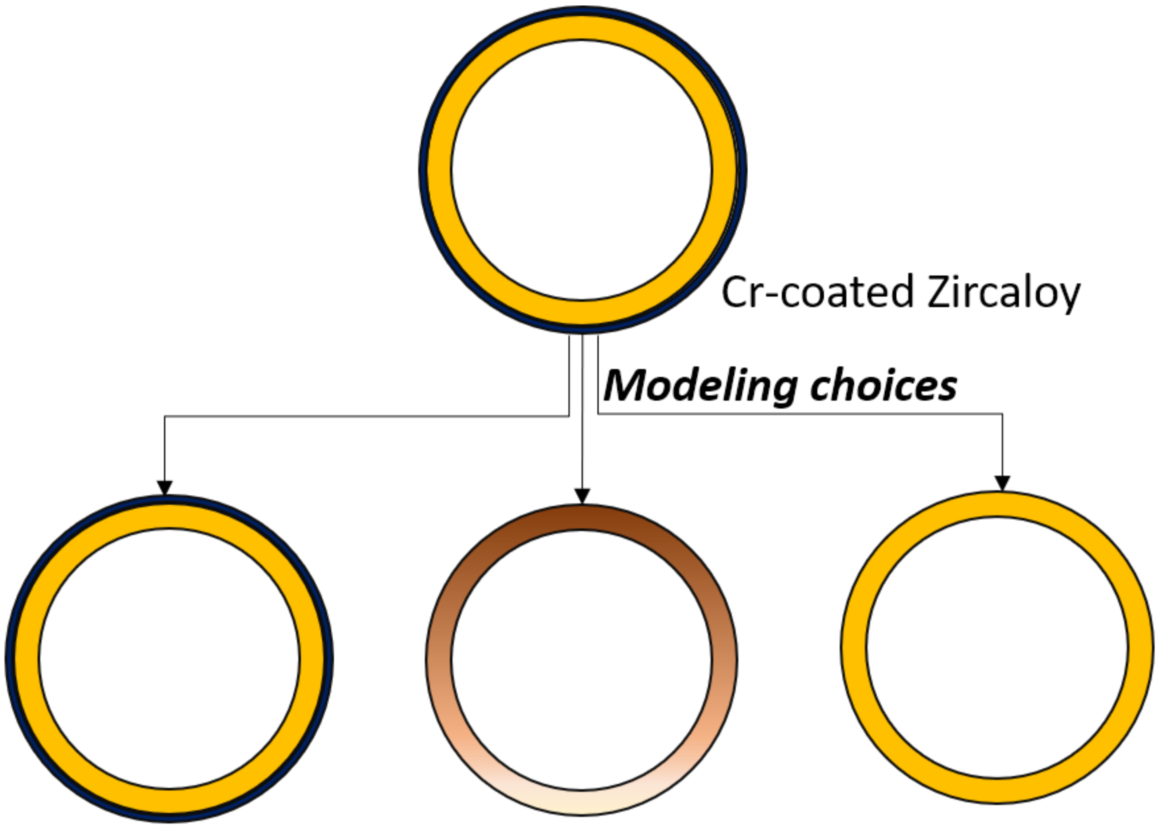
GIFT 코드개발 (written in C++)

고심도 ATF 모사를 위한 추가 모델 성능 정리

(흰색) : 현행 체계 모델 적용가능 (녹색) : ATF 특화 고심도 모델 필요 부분

		FRAPCON-4.0/FRAPTRAN-2.0	ATF 모사를 위한 추가 모델 성능	관련 평가인자
	소결체 거동 모델	UO_2 소결체	UO_2 소결체	-
①	피복관 구조해석 모델 충실도	1D 모델 (Thin-wall approximation)	2차원 축대칭 모델 도입 필요	코팅 박리/균열
②	다층 피복관 구조 해석 모델	반영 안됨	모체와 코팅막 크립 및 소성 변형 모사 필요	코팅 박리/균열
	소결체-피복관 기계적 상호작용 (PCMI)	Rigid Pellet Model	Rigid Pellet Model	-
③	축방향 피복관 조사성장 구조해석	반영 안됨	반영 필요	코팅 박리/균열
④	피복관 벌루닝 및 파열	ATF 코팅영향 미반영	ATF 코팅영향 반영필요	대변형 관련
⑤	산화모델	실험상관식 기반 단순모델. 코팅 미반영.	고심도 해석모델. Zr-Cr-O 거동모델 개발 필요	사고후 잔류연성
⑥	건식저장 사용후 핵연료 모델	제한적	모사 필요	범용
⑦	수소 확산 모델	반영 안됨	모사 필요	범용
⑧	가돌리니아 삽입 모델	반영 안됨	i-SMR 적용 관점에서 모사 필요	범용

GIFT 피복관 구조해석 모델 현안



Brute-force multi-layer model

- can predict coating stress
- no longer valid once coating partially fails

Composite model

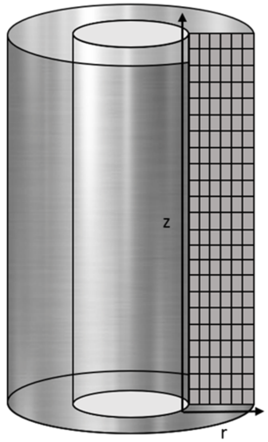
- can efficiently approximate integral behavior of dual layers
- No calculation for coating structural integrity

Bare-Zircaloy Approximation

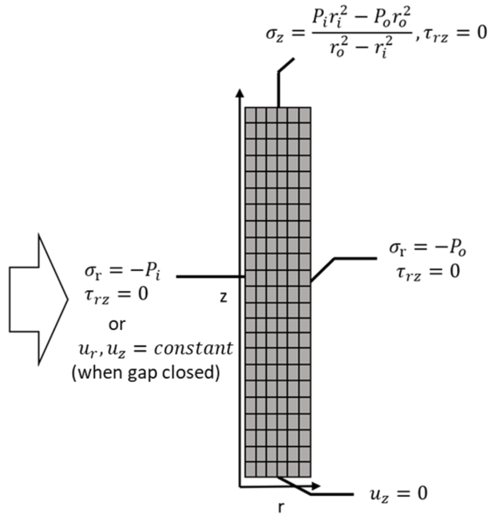
- good when coating layer is thin
- No calculation for coating structural integrity

GIFT 피복관 구조해석 모델

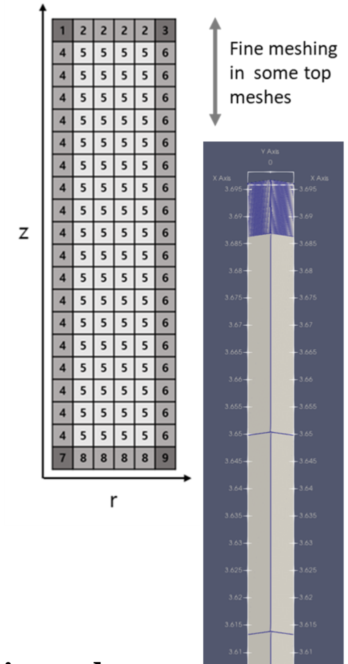
2D Axis-symmetric full-length cladding mechanical model



2D (axis-symmetric) full length fuel rod



Boundary conditions



Governing equations and numerical solver, $A\vec{x} = \vec{b}$

Equilibrium Equation

$$\frac{\partial \sigma_{rr}}{\partial r} + \frac{\partial \tau_{rz}}{\partial z} + \frac{\sigma_{rr} - \sigma_{\theta\theta}}{r} = 0 \quad (1)$$

$$\frac{\partial \sigma_{zz}}{\partial z} + \frac{\partial \tau_{rz}}{\partial r} + \frac{\tau_{zr}}{r} = 0 \quad (2)$$

Constitutive Equation

$$\epsilon_{rr} = \frac{\sigma_{rr}}{E_{rr}} - \frac{V_{\theta r}\sigma_{\theta\theta}}{E_{\theta\theta}} - \frac{V_{zr}\sigma_{zz}}{E_{zz}} + \alpha_{rr}\Delta T + \epsilon_r^p + \epsilon_r^c + S_{rr} \quad (3)$$

$$\epsilon_{\theta\theta} = \frac{\sigma_{\theta\theta}}{E_{\theta\theta}} - \frac{V_{z\theta}\sigma_{zz}}{E_{zz}} - \frac{V_{r\theta}\sigma_{rr}}{E_{rr}} + \alpha_{\theta\theta}\Delta T + \epsilon_\theta^p + \epsilon_\theta^c + S_{\theta\theta} \quad (4)$$

$$\epsilon_{zz} = \frac{\sigma_{zz}}{E_{zz}} - \frac{V_{rz}\sigma_{rr}}{E_{rr}} - \frac{V_{\theta z}\sigma_{\theta\theta}}{E_{\theta\theta}} + \alpha_{zz}\Delta T + \epsilon_z^p + \epsilon_z^c + \frac{\Delta L}{L} + S_{zz} \quad (5)$$

$$\gamma_{rz} = \frac{\tau_{rz}}{G} \quad (6)$$

Deformation(kinematic) Relations

$$\epsilon_{rr} = \frac{\partial u_r}{\partial r} \quad (7)$$

$$\epsilon_{\theta\theta} = \frac{u_r}{r} \quad (8)$$

$$\epsilon_{zz} = \frac{\partial u_z}{\partial z} \quad (9)$$

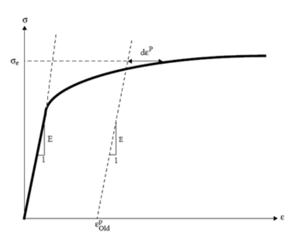
$$\gamma_{rz} = \frac{\partial u_r}{\partial z} + \frac{\partial u_z}{\partial r} \quad (10)$$

10 Unknowns

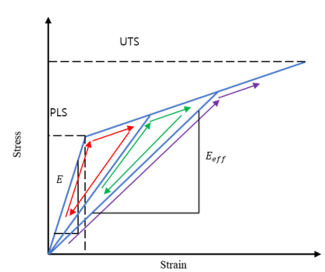
$$\sigma_{rr}, \sigma_{\theta\theta}, \sigma_{zz}, \tau_{rz}, \epsilon_{rr}, \epsilon_{\theta\theta}, \epsilon_{zz}, \gamma_{rz}, u_r, u_z$$

$$c_{11}\frac{\partial^2 u_r}{\partial r^2} - c_{11}\alpha_{rr}\frac{\partial \Delta T}{\partial r} - c_{11}\frac{\partial S_{rr}}{\partial r} + c_{12}\frac{\partial}{\partial r}\left(\frac{u_r}{r}\right) - c_{12}\alpha_{\theta\theta}\frac{\partial \Delta T}{\partial r} - c_{12}\frac{\partial S_{\theta\theta}}{\partial r} + c_{13}\frac{\partial}{\partial r}\left(\frac{\partial u_z}{\partial z}\right) - c_{13}\alpha_{zz}\frac{\partial \Delta T}{\partial r}$$
$$- c_{13}\frac{\partial S_{zz}}{\partial r} + c_{44}\left(\frac{\partial^2 u_r}{\partial z^2} + \frac{\partial^2 u_z}{\partial r \partial z}\right) + \frac{c_{11} - c_{21}}{r}\left[\frac{\partial u_r}{\partial r} - (\alpha_{rr}\Delta T + S_{rr})\right] + \frac{c_{12} - c_{22}}{r}\left[\frac{u_r}{r} - (\alpha_{\theta\theta}\Delta T + S_{\theta\theta})\right]$$
$$+ \frac{c_{13} - c_{23}}{r}\left[\frac{\partial u_z}{\partial z} - (\alpha_{zz}\Delta T + S_{zz})\right] = 0$$

The displacement formulation of governing equations



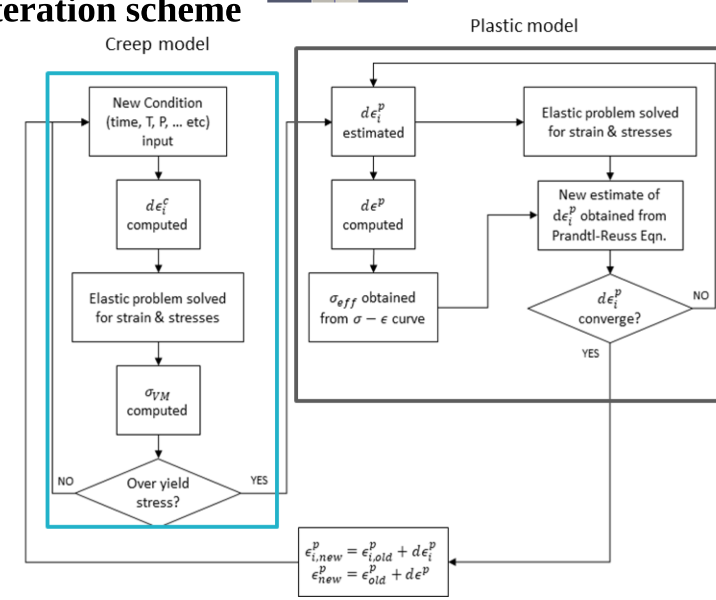
Prandtl-Reuss flow rule for zirconium-based alloy



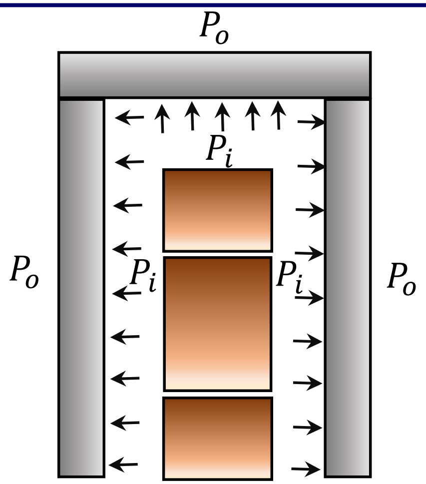
Pseudo-ductile behavior for ceramic composites

Material-dependent plasticity model

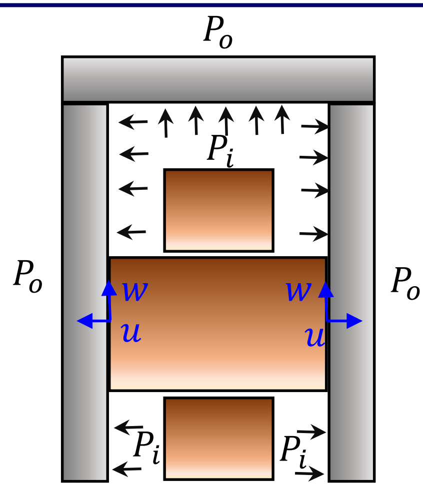
Creep and plastic deformation iteration scheme



GIFT 피복관 구조해석 모델 – closed gap cladding behavior



Open gap



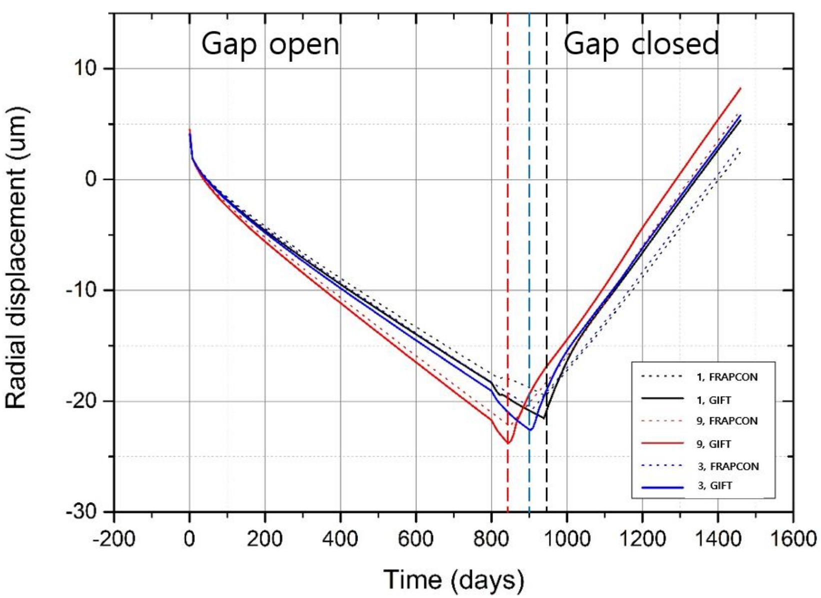
closed gap

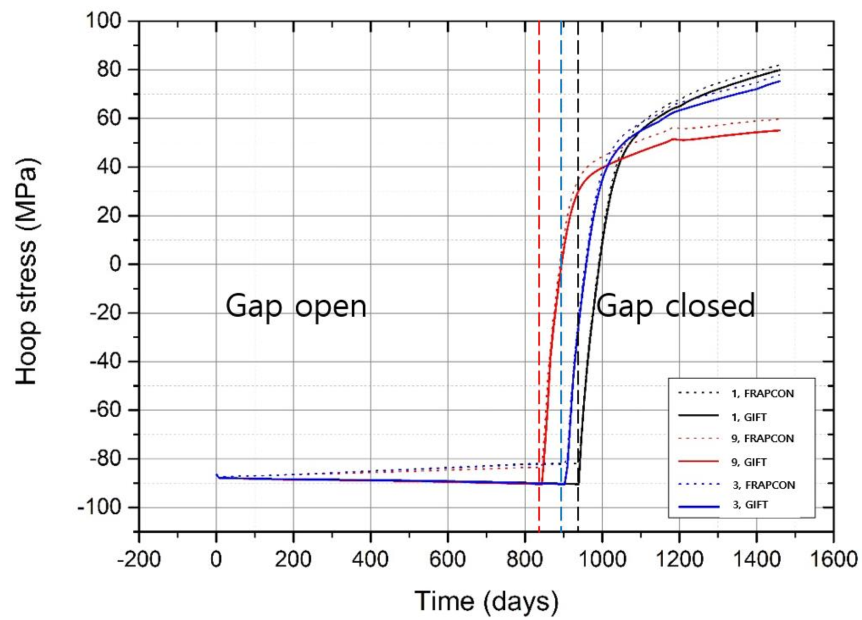
$$A\vec{x} = \vec{b}$$

Discretization scheme & inter-node interaction

Boundary conditions

Matrix and B.C conditioning for every different pellet-cladding interaction state in the fuel rod



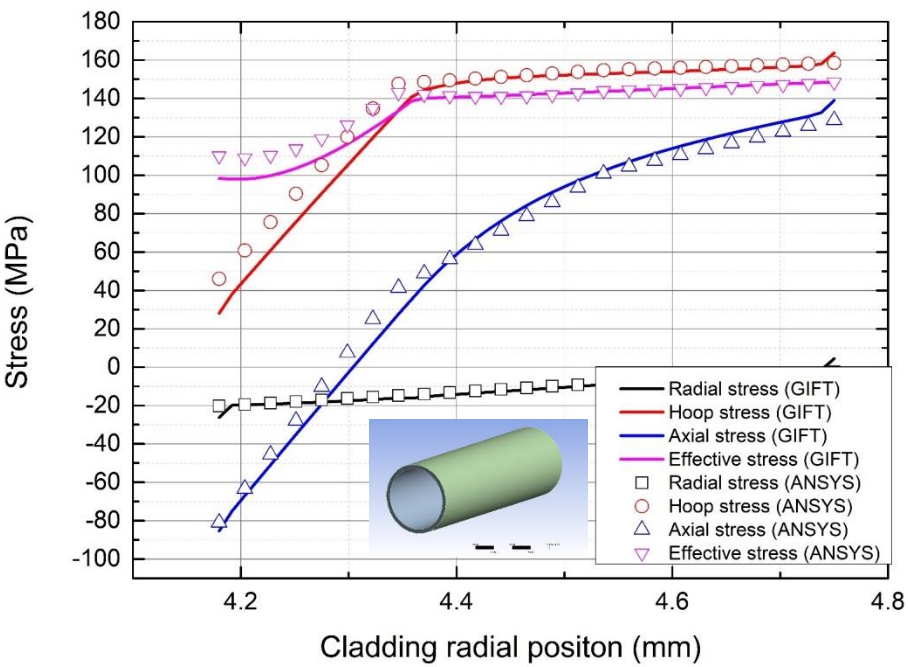


<Cladding performance comparison with FRAPCON>

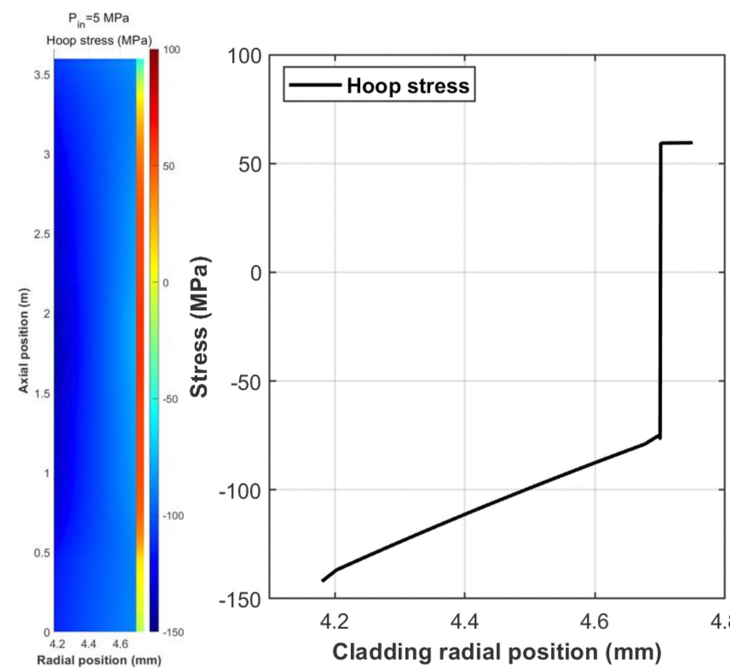
GIFT 피복관 구조해석 모델 – 소성변형 독립검증

○ 코팅피복관 해석 모델 적용

- 모체와 코팅층에 서로 다른 물성치 및 Stress-Strain curve를 도입
- ANSYS 와 소성변형 구조해석 비교 수행, 높은 일치도 확인
- 소성 및 조사성장을 고려한 Cr코팅 피복관 구조해석 계산 성공적 구현



〈ANSYS와의 소성이 고려된 구조해석 결과 비교 검증〉

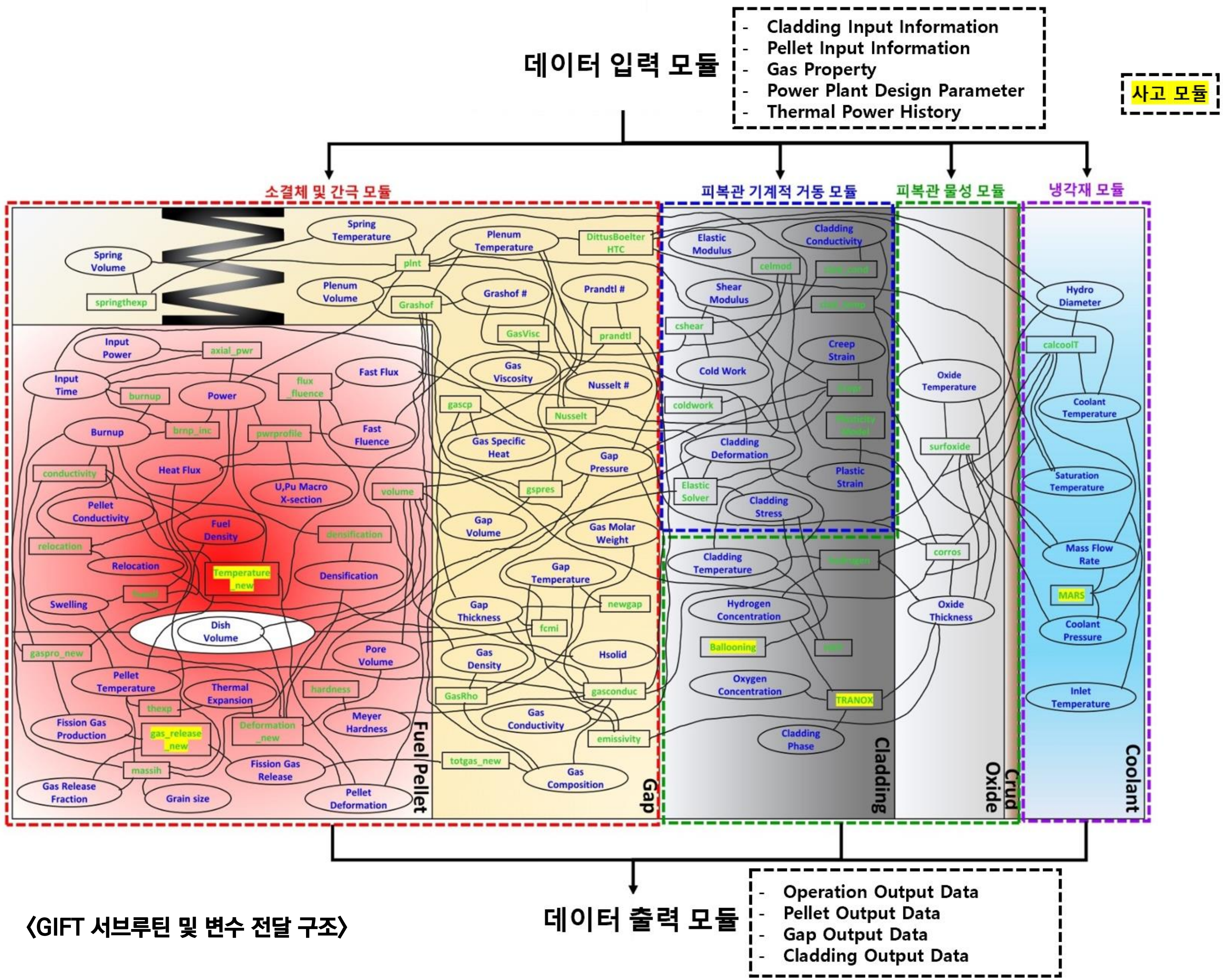


〈Cr 코팅 피복관 구조해석 예시결과〉

Simulation conditions

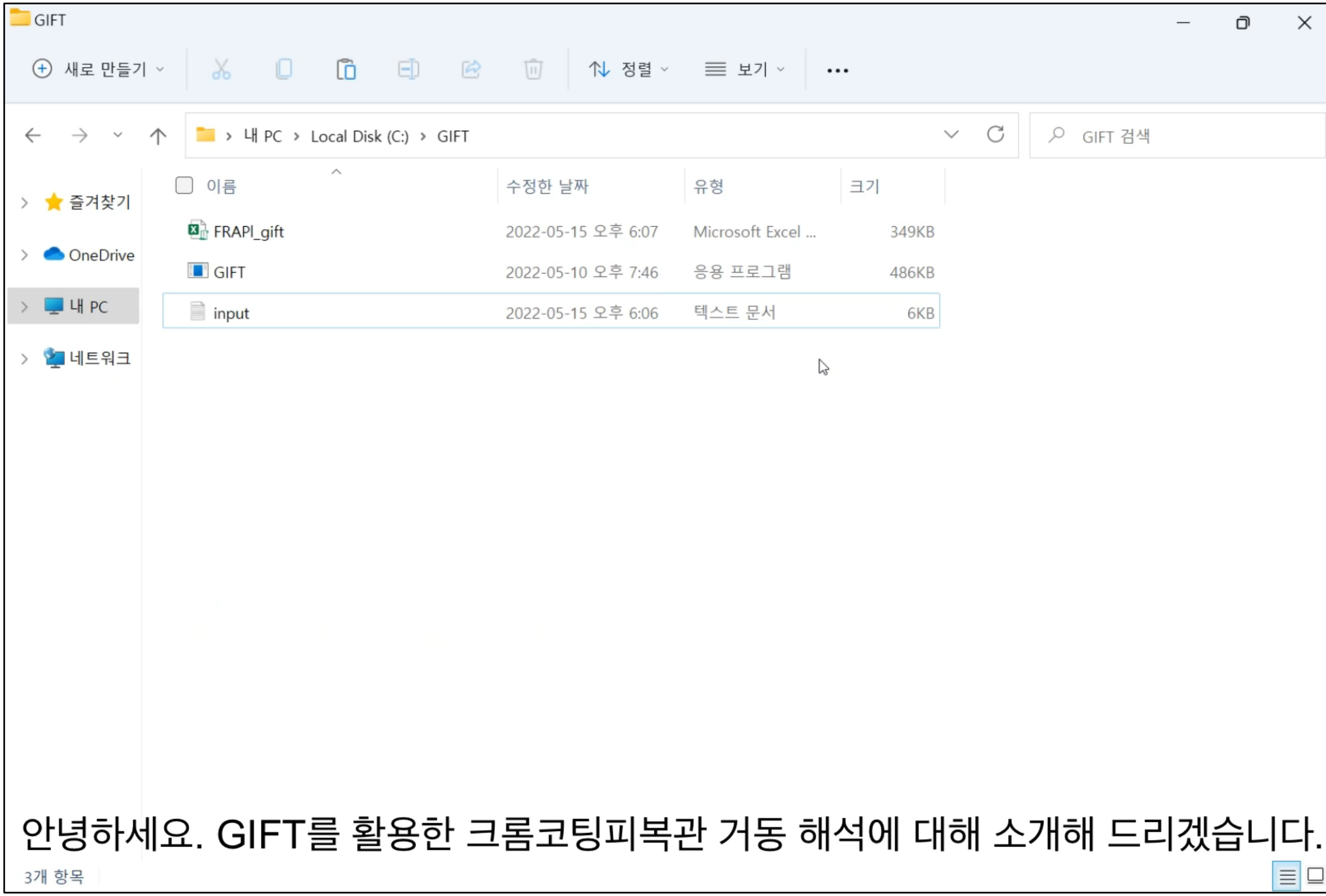
* PZR pressure	= 15.5 MPa	* LHGR	= 17.8 kW/m
* Hot leg temp	= 598.15 K	* Fast neutron fluence	= $10^{25}\ n/m^2$
* Cold leg temp	= 565.15 K		

서울대학교 개발 핵연료 코드 GIFT

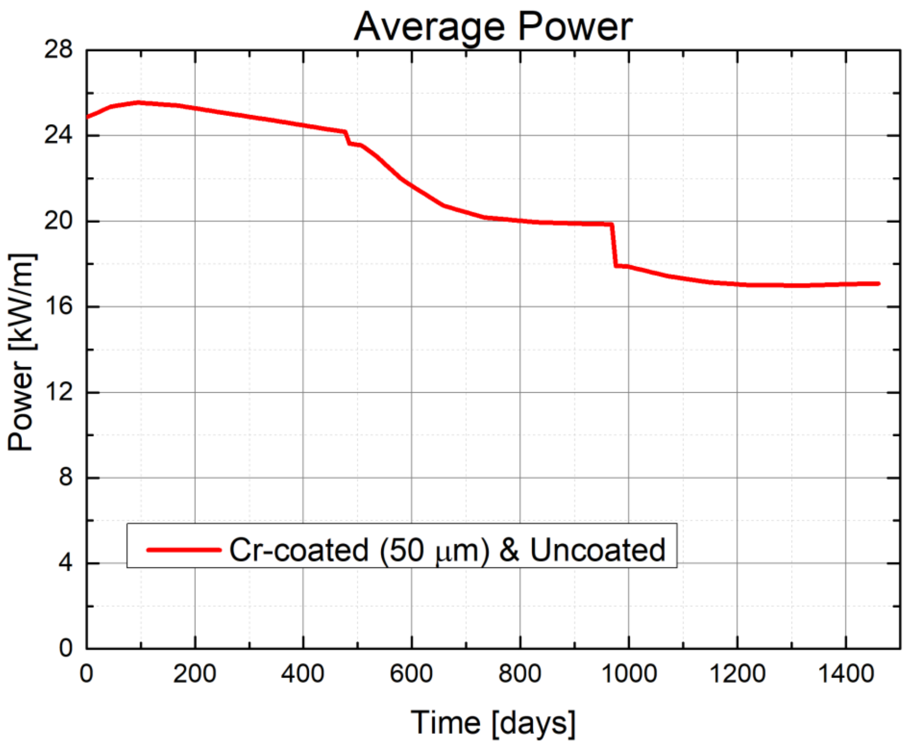


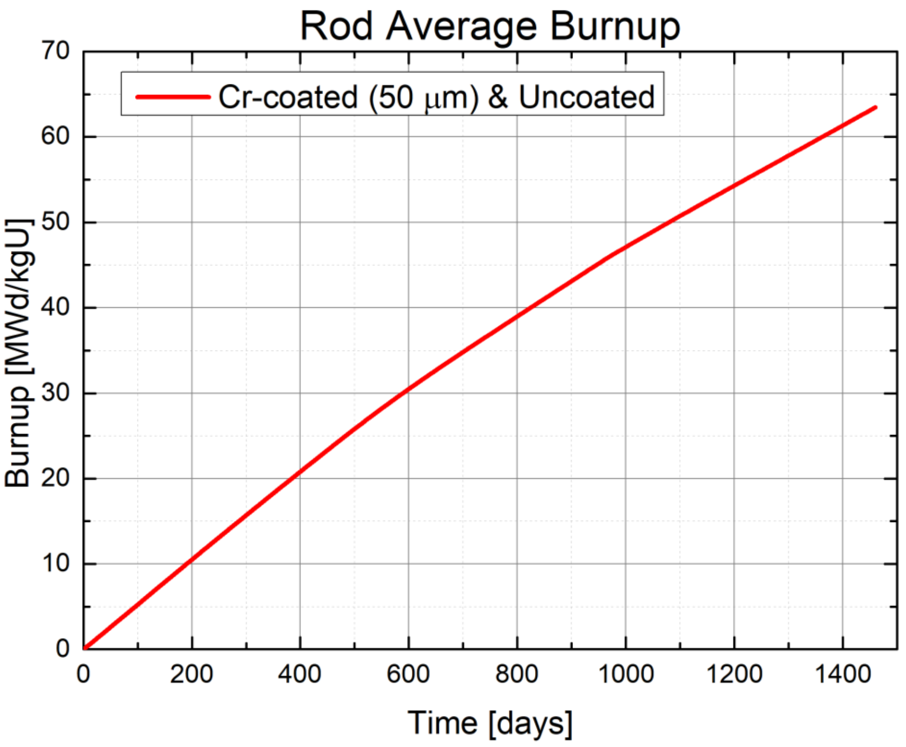
〈GIFT 서브루틴 및 변수 전달 구조〉

Introduction of GIFT code

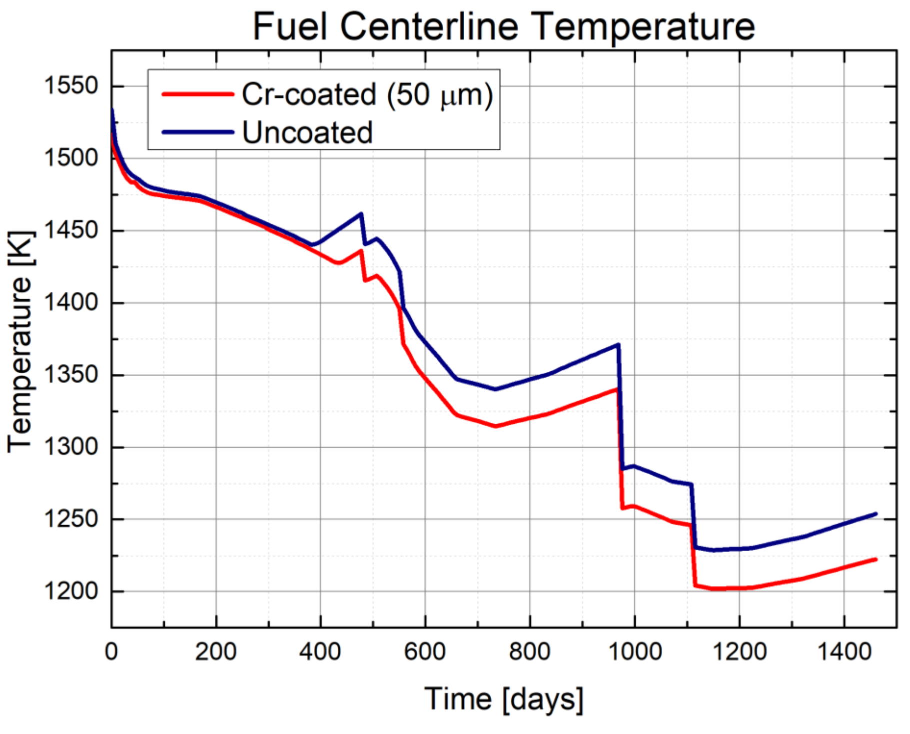


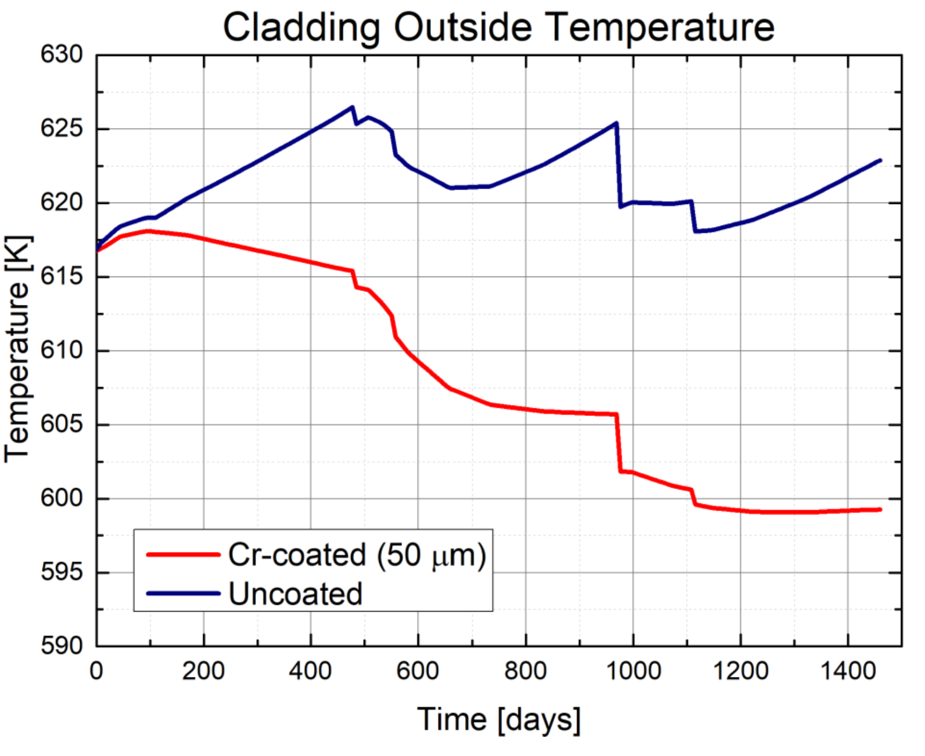
Power and Burnup



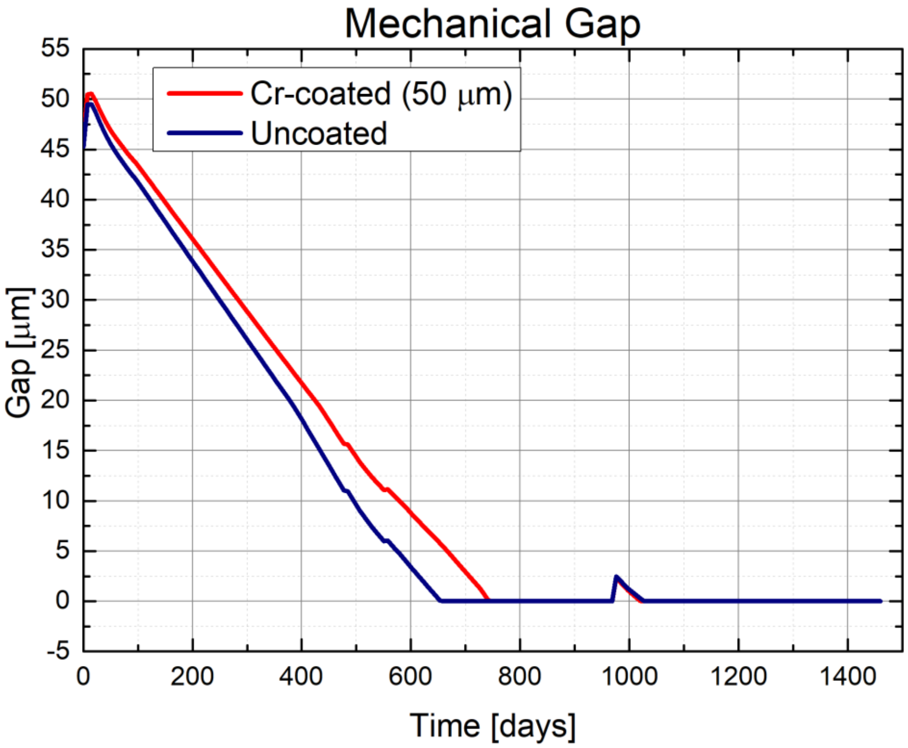


Fuel & Cladding temperature

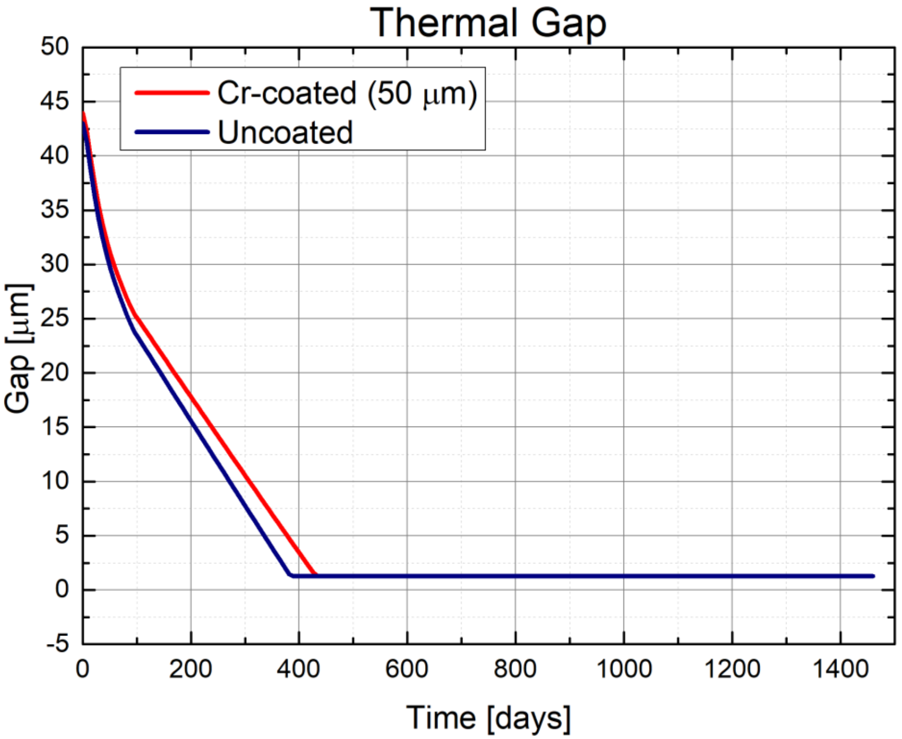




Gap size

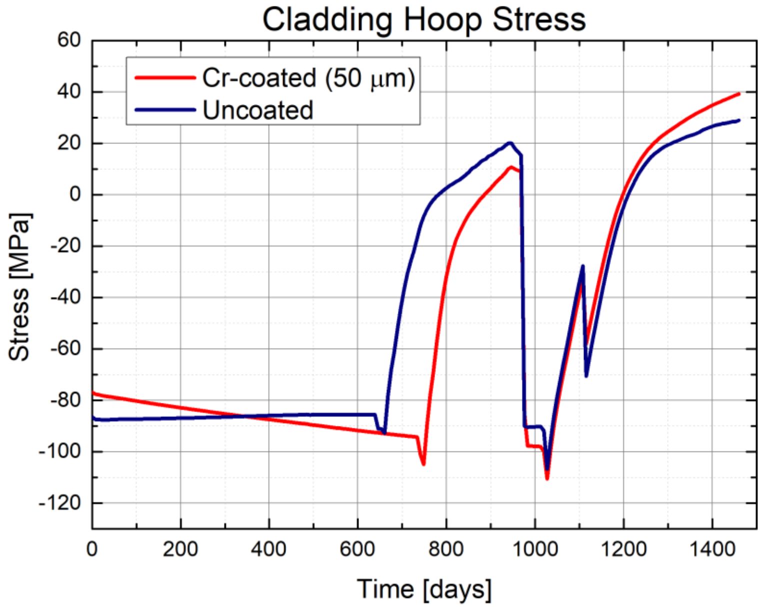


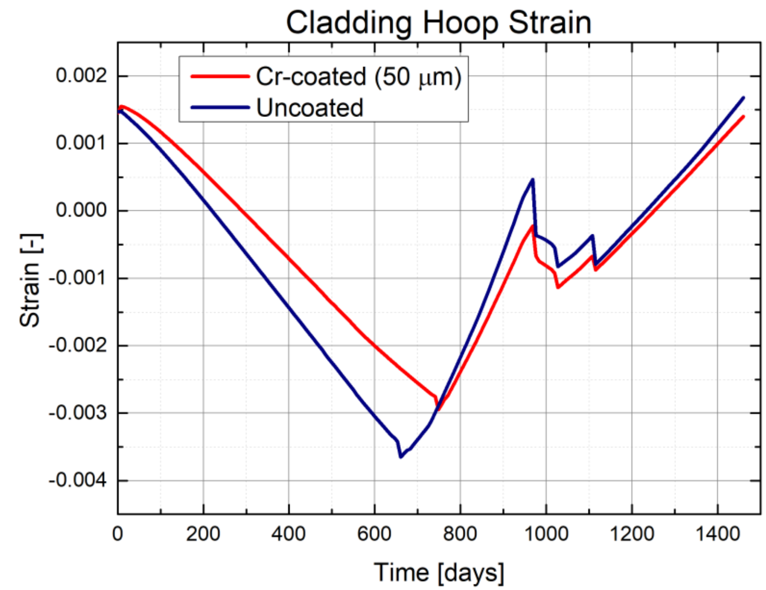
Mechanical Gap
Cr-coated (50 μm)
Uncoated
Gap [μm]
Time [days]
55
50
45
40
35
30
25
20
15
10
5
0
-5
0
200
400
600
800
1000
1200
1400

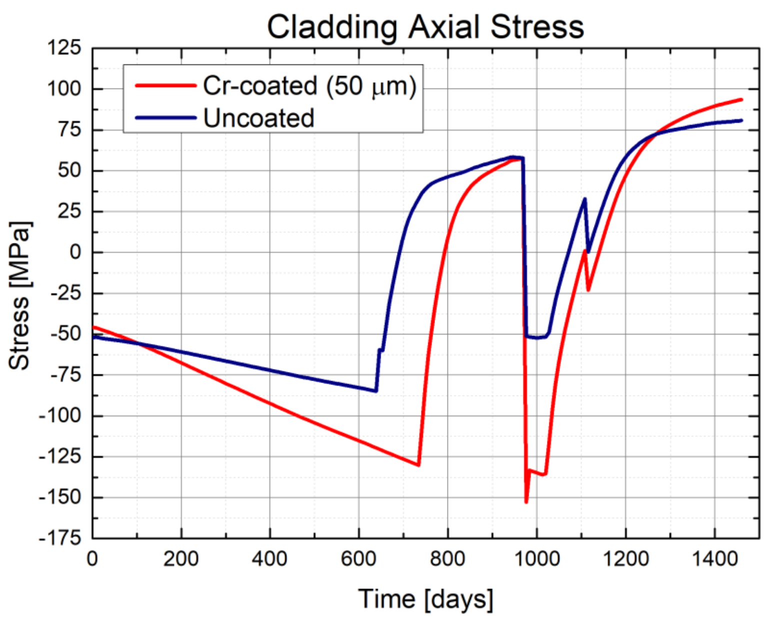


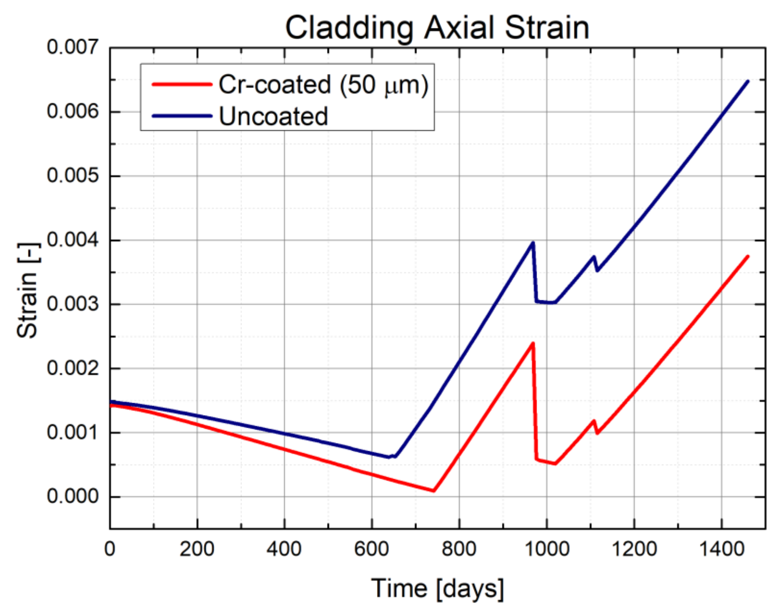
Thermal Gap
Cr-coated (50 μm)
Uncoated
Gap [μm]
Time [days]
50
45
40
35
30
25
20
15
10
5
0
-5
0
200
400
600
800
1000
1200
1400

Stress & Strain

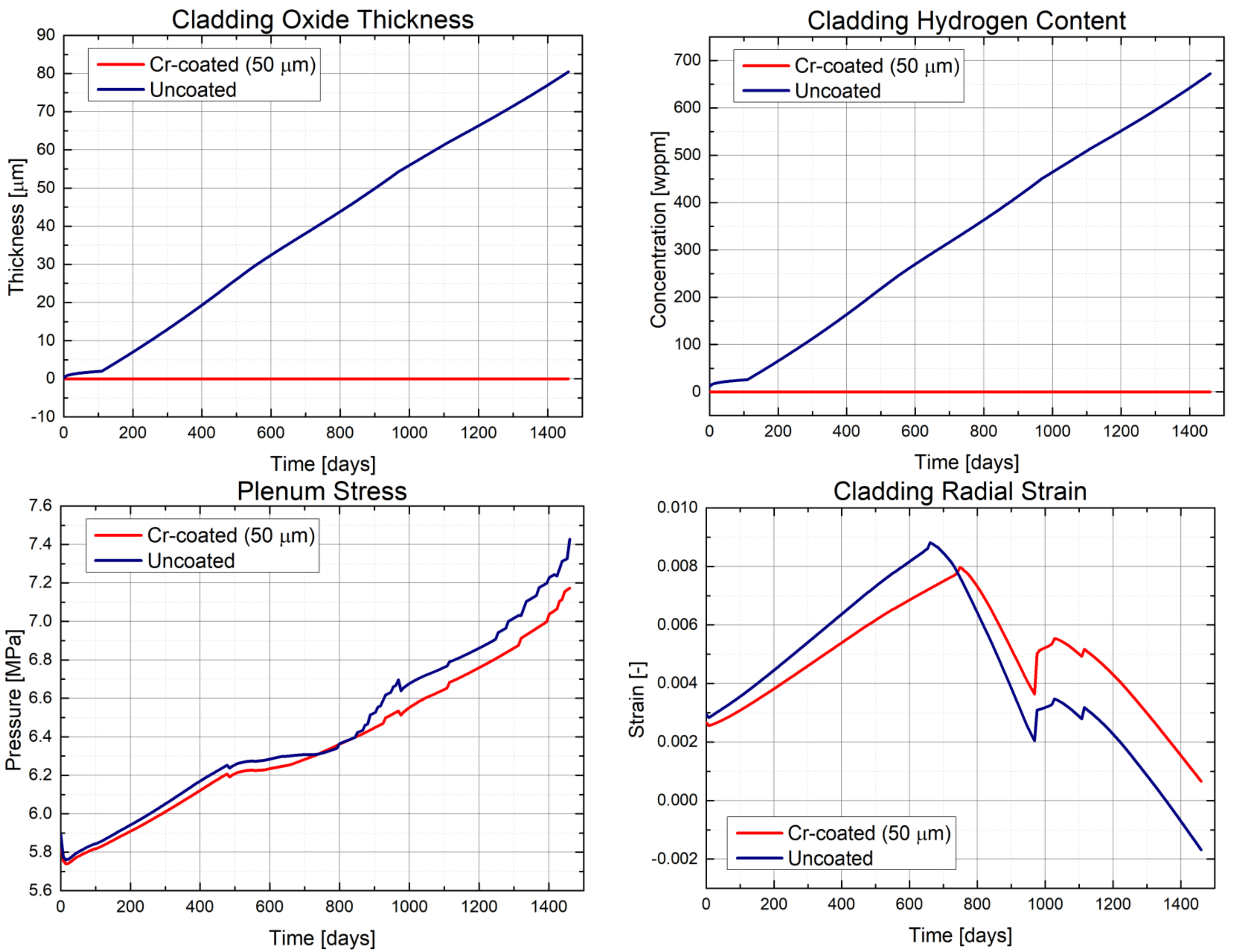




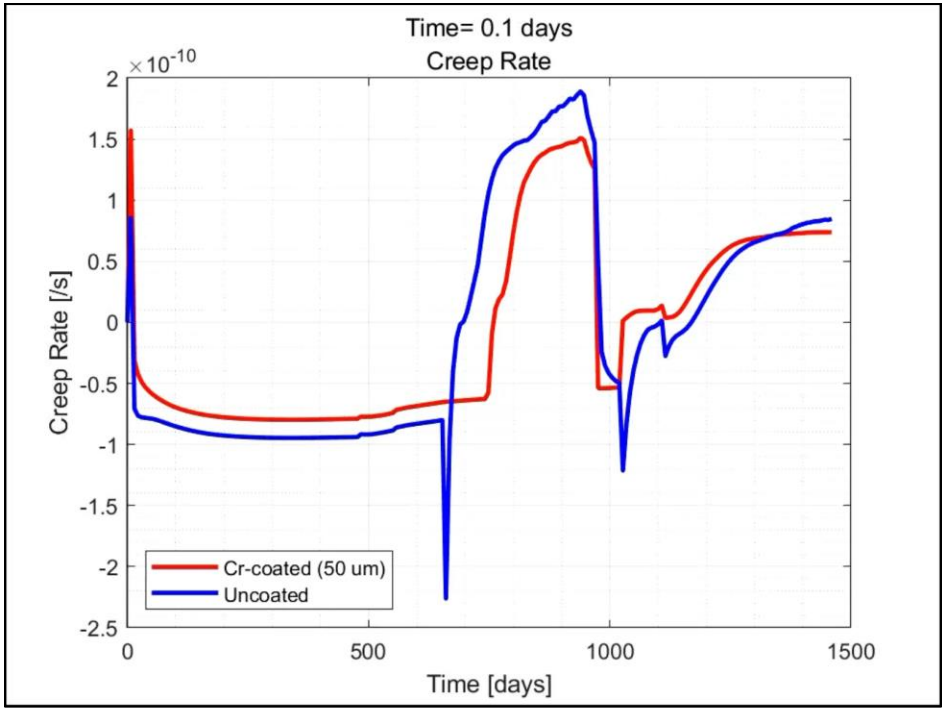




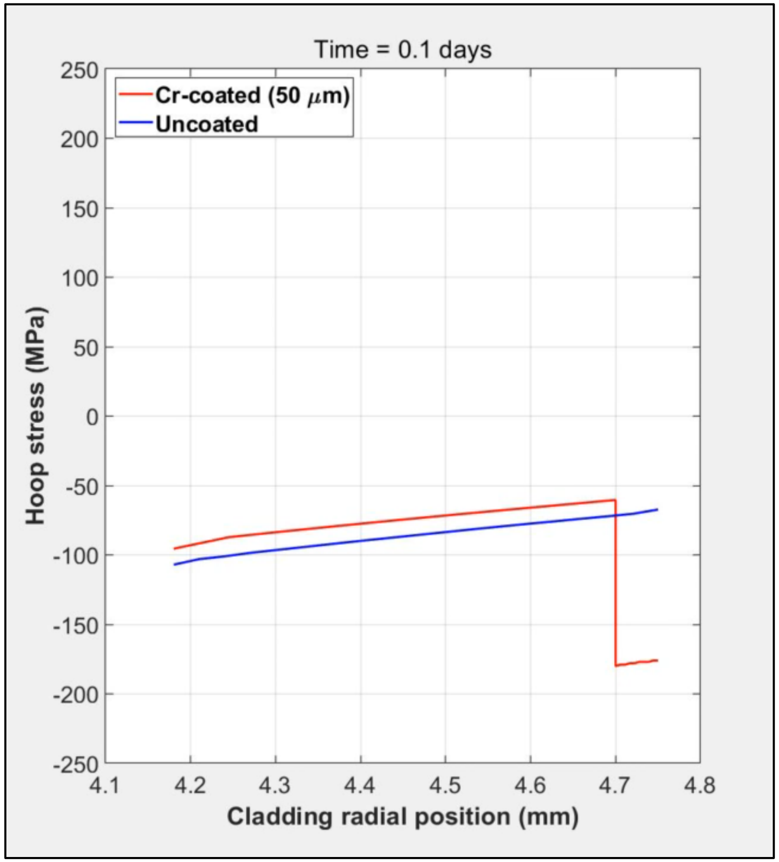
Mechanical integrity of fuel



Creep rate and Stress field



<Cladding Creep Rate>

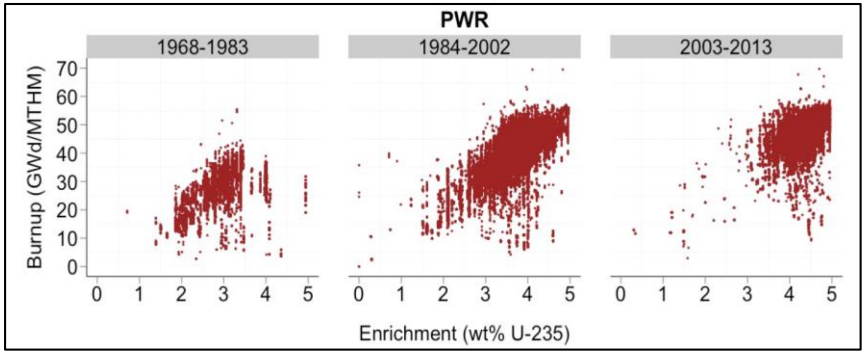


<Cladding Hoop Stress Distribution>

Chapter 3
결론 및 제언

결론 및 제언:

- **경제성 :**
  - ▶ 연소도 증진을 위해서는 5% 이상의 농축도 필수
  - ▶ 한미원자력협정에근거하여 5% 이상 농축 우라늄에 대한 공급을 준비할 필요 있음
  - ▶ FFRD 가 허용가능 연소도 결정에 중요할것으로 봄.



Burnup and initial enrichment for each PWR assemblies in the U.S (US Commercial Spent Nuclear Fuel Assembly Characteristics: 1968-2013, U.S NRC, NUREG/CR-7227)

- **안전성:**
  - ▶안전 여유도를 유지하며 연소도 증진 가능 → “경제적 안전여유도”
  - ▶ CDF 유의미한 변화는 없을것으로 예상.
  - ▶ 공융현상 매우 중요 (Eutectic).
  - ▶ 공융현상으로 인한 핵연료봉 손상/붕괴모드 실험 중요.
  - ▶ 중대사고 진행 완화에 미치는 영향을 평가하는것이 중요.
    → 핵연료봉 스케일 공융 실험 중요

결론 및 제언:

- **지속가능성:**
  ▶ 연소도 증가에 따른 사용후핵연료 배출량 감소

  ▶ 에너지 생산량 당 사용후 핵연료 관리비용 감소

  ▶ 우라늄 농축도 증가에 따른 습식저장 재임계 문제 (해결 가능)

  ▶ 건식저장 핵연료 특성변화:
  기존 LWR: 높은 수소화물, 낮은 봉내압 , 낮은 붕괴열
  ATF LWR: 낮은 수소화물, 높은 봉내압 , 높은 붕괴열

  ▶ 수소화물재배열, 지연된 수소화물 크래킹 등에 의한 건식저장 안전성 문제 대폭 완화 → 사용후핵연료 관리비용 감소 가능

  ▶ 높은 봉내압으로 인한 건식저장시 Creep 변형이 주요. 핵연료 봉내압으로 인한 Creep 변형은 Self-regulating 하기에 문제가 제한적

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Massachusetts Institute of Technology

KIMS Korea Institute of Materials Science 한국재료연구원

FNC (주)미래와도전
FNC Technology Co., Ltd.

Thank you for your attention!