FeCrAl Oxidation Model Development on the ASTEC Code and Preliminary Validation Against QUENCH Experiments

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Content

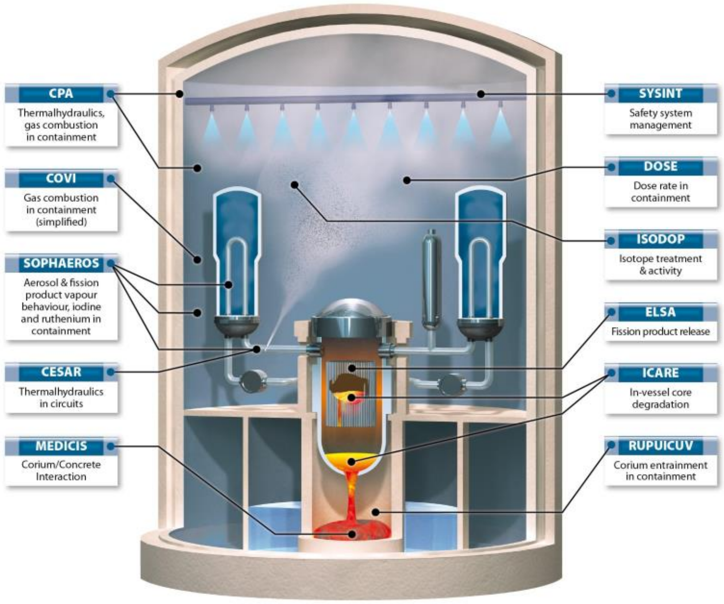
Motivation

- KIT strategy for severe accident (SA) analyses → continuous improvement of the codes to evaluate the progression and the radiological consequences of SA in current and innovative NPPs
- ATF claddings have the potential to mitigate the progress of hypothetical severe accident scenarios, e.g., delayed hydrogen onset timing, reducing the hydrogen production, compared with Zr cladding
- A KIT/INR and IRSN shared activity is going on to improve the ATF-related modelling capabilities (cladding) of the ASTEC* code
- Goals of the work
  - Improvement and implementation of the ASTEC modelling for predicting the hydrogen generation and mass gain by the oxidation of FeCrAl under steam atmosphere
  - ASTEC validation against the SETs and QUENCH-19 bundle test performed at the KIT QUENCH large-scale facility

*CHATELARD, P., et al., “Main modelling features of the ASTEC V2.1 major version”, Annals of Nuclear Energy 93 (2016) 83–93.

The Accident Source Term Evaluation Code (ASTEC)

- Developed by IRSN (IRSN all rights reserved, [2024])
- Simulating the entire Severe Accident (SA) sequence from the initiator up to the fission product release to the environment
- ASTEC models all the physical phenomena that occur during a core meltdown accident
- ICARE module describes the in-vessel degradation phenomena
- Chemistry model: Oxidation of cladding material by steam or O2



Accident Tolerant Fuels (ATF)

- ATF
  - Better performance
  - Near-Term
    - Coated claddings
    - Dopped pellets
    - FeCrAl cladding
      - H-T steam oxidation
      - Strength
      - H-T accident condition
      - Corrosion
      - No hydrides
  - Longer-Term
    - Uranium nitride pellets
    - Silicone carbide claddding
    - Extruded metallic fuel

B136Y3

- Fe
- 12.97 Cr
- 6.19 Al
- 0.03 Y
- <0.01 C

C26M2

- Fe
- 11.87 Cr
- 6.22 Al
- 1.98 Mo
- 0.03 Y
- <0.01 C
- 0.2 Si

ASTEC code improvement for FeCrAl: approach

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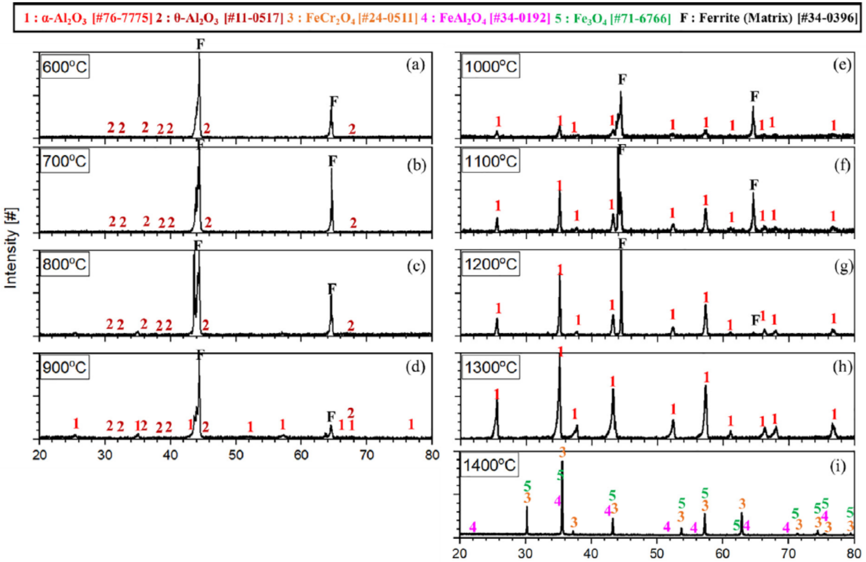
- Based on the experimental data from the QUENCH team…
- Simplification of the FeCrAl oxidation process in steam environment
- Implementation of the oxide kinetics physical models
- Implementation of the physical models for the mass gain and thickness oxide layer evaluations
- Use of the FeCrAl thermophysical properties, i.e. specific enthalpy, density, heat capacity

FeCrAl Oxidation Model

- Steam oxidation reaction

$$3\ FeCrAl_{(s)} + 4H_2O_{(g)} \rightarrow Fe_3O_{4\,(s)} + 3\ Cr_2O_{3\,(s)} + 4\ Al_2O_{3\,(s)} + 6\ H_{2\,(g)}$$



- Single oxide layer model

$$2\ Al_{(s)} + 3\ H_2O_{(g)} \rightarrow Al_2O_{3\,(s)} + 3H_{2\,(g)}$$

- Composition

80.8%Fe12.97%Cr6.19%Al

Fig. 1 Oxidation kinetics present in the FeCrAl alloy oxidation process from: Kim, C., Tang, C., Grosse, M., Maeng, Y., Jang, C., & Steinbrueck, M. (2022). Oxidation mechanism and kinetics of nuclear-grade FeCrAl alloys in the temperature range of 500–1500 C in steam. *Journal of Nuclear Materials*, *564*, 153696.

Oxidation kinetics

$873 < T < 1173$	$1173 \leq T \leq 1273$	$1273 < T < 1648$	$T \geq 1648$
Transient Alumina	Alumina	α-Alumina	FeO-Oxide

It evaluates the rate of aluminum oxide mass gain

Oxygen Mass Gain

- parabolic correlation
- different coefficients according oxidation kinetics

$$\frac{dm_{O_2}^2}{dt} = K_p \qquad (1)$$

$$K_p \left[\frac{kg^2}{m^4 \cdot s}\right] = A_{gain} e^{\left(-\frac{B_{gain}}{RT}\right)} \qquad (2)$$

- dm_{O_2}: mass gain
- K_p: rate constant for mass gain
- A_{gain}: pre-exponential constant
- B_{gain}: energy activation
- R: ideal gas constant
- T: temperature
- dt: time

Oxidation kinetics implementation

Transient Alumina

$$873 < T < 1173$$
$$A_{gain} = 5.37582 \cdot 10^{-3}$$
$$B_{gain} = 1.84730 \cdot 10^{5}$$

Alumina

$$1173 \leq T \leq 1273$$
$$A_{gain} = 4.69155 \cdot 10^{-12}$$
$$B_{gain} = 0.00000 \cdot 10^{0}$$

α-Alumina

$$1273 < T < 1648$$
$$A_{gain} = 5.01760 \cdot 10^{0}$$
$$B_{gain} = 2.87748 \cdot 10^{5}$$

FeO-Oxide

$$T \geq 1648$$
$$A_{gain} = 2.39940 \cdot 10^{8}$$
$$B_{gain} = 3.52514 \cdot 10^{5}$$

ASTEC equation

- $\frac{dm_O{}^2}{dt} = K_p \rightarrow m_O^{i+1} = \sqrt{\left\{\left(m_O^{i+1}\right)^2 + \left[A_{gain} e^{\left(\frac{-B_{gain}}{RT}\right)} \Delta t\right]\right\}}$

```
STRUCTURE PROPERTY NAME "SteamOxidation"
    STRU TEST X 1000. END

    HELP "Oxygen mass gain obey to the following law :"

    HELP "m_O   (t+dt) = S ((m_O   (t)/S)**(1/model) + AGAIN EXP(-BGAIN/(R.T)) * dt )**model"

    STRUCTURE MODEL NAME 'ATFKIT' LAW 'COEFF' VARIABLE 'T' VUNIT 'K' RUNLOW 0. RUNUPP 5000.
       SRG VALUE AGAIN 5.0176000D0 BGAIN 2.8774800D5  MODEL 0.5 TERM
    END

    STRUCTURE MODEL NAME 'ATF-KIT' LAW 'COEFF' VARIABLE 'T' VUNIT 'K' RUNLOW 0. RUNUPP 5000.
       SRG VALUE AGAIN 5.3758224D-3  BGAIN 1.8473000D5  MODEL 0.5 TERM
       X  1172.9K
       SRG VALUE AGAIN 4.6915560D-12 BGAIN 0.0000000D5  MODEL 0.5 TERM
       X  1273.K
       SRG VALUE AGAIN 5.0176000D0   BGAIN 2.8774800D5  MODEL 0.5 TERM
       X  1652.9K
       SRG VALUE AGAIN 2.3994010D8   BGAIN 3.5251400D5  MODEL 0.5 TERM
    END

 END
```

Fig. 2 FeCrAl oxidation kinetics implementation on ASTEC code

Thickness Oxide Layer

$$2\,Al + \frac{3}{2}O_2 \rightarrow Al_2O_3$$

$$m_{Al_2O_3} = m_{O_2}\frac{MM_{Al_2O_3}}{f_s MM_{O_2}} \quad (3)$$

$$\delta_{Al_2O_3} = \frac{m_{Al_2O_3}}{S}\left[\frac{1}{\rho_{Al_2O_3}}\right] \quad (4)$$

$$\delta_{Al_2O_3} = m_o\frac{MM_{Al_2O_3}}{f_s MM_{O_2}}\frac{1}{\rho_{Al_2O_3}} \quad (5)$$

Hydrogen Production

$$2\,Al_{(s)} + 3\,H_2O_{(g)} \rightarrow Al_2O_{3\,(s)} + 3H_{2\,(g)}$$

$$mH_2 = \frac{3m_{Al_2O_3}MMH_2}{PM_{Al_2O_3}} \quad (6)$$

Implementation

$$\frac{m_{O_2}^{i+1}}{S} = \sqrt{\left\{\left(m_{O_2}^{i+1}\right)^2 + \left[A_{gain}e^{\left(\frac{-B_{gain}}{RT}\right)}\Delta t\right]\right\}} \quad (7)$$

$$\frac{m_{Al_2O_3}^{i+1}}{S} = \frac{m_{O_2}^{i+1}}{S}\frac{2MM_{Al_2O_3}}{3MM_{O_2}} \quad (8)$$

$$\delta_{Al_2O_3}^{i+1} = \frac{m_{Al_2O_3}^{i+1}}{S}\frac{1}{\rho_{Al_2O_3}} \quad (9)$$

$$m_{Al_2O_3}^{i+1} = \frac{m_{Al_2O_3}^{i+1}}{S}(S) \quad (10)$$

$$m_{H_2}^{i+1} = \frac{3m_{Al_2O_3}^{i+1}\cdot MM_{H_2}}{PM_{Al_2O_3}} \quad (11)$$

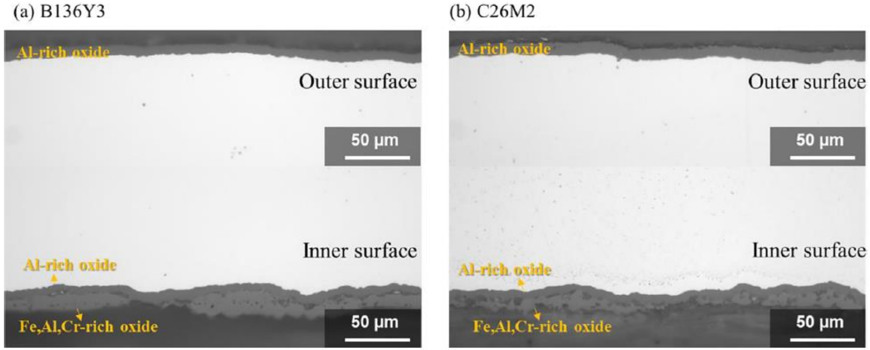


Fig. 3 Thickness oxide layer for FeCrAl alloys from*

*Kim, C., Tang, C., Grosse, M., Maeng, Y., Jang, C., & Steinbrueck, M. (2022). Oxidation mechanism and kinetics of nuclear-grade FeCrAl alloys in the temperature range of 500–1500 C in steam. *Journal of Nuclear Materials*, *564*, 153696.

Single effect test

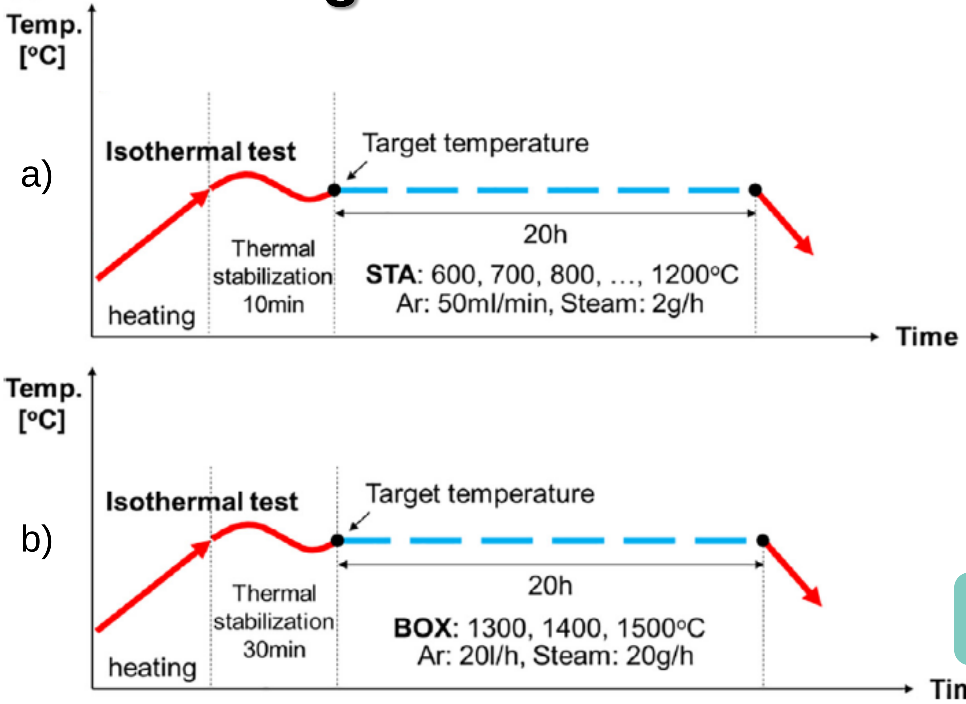
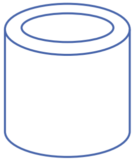
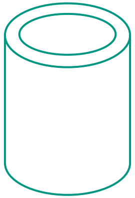


Fig. 4. The test schedules for the isothermal steam oxidation in (a) STA and (b) BOX facilities edited from*



873K-1473K
L: 10 mm



1573K-1773K
L: 20 mm

Fig.6 Sample before isothermal test from*

- Validation data
  - Weight gain $\left(\frac{mg}{cm^2}\right)$
  - Thickness oxide layer (m)
  - Hydrogen production (gr)

*Kim, C., Tang, C., Grosse, M., Maeng, Y., Jang, C., & Steinbrueck, M. (2022). Oxidation mechanism and kinetics of nuclear-grade FeCrAl alloys in the temperature range of 500–1500 C in steam. *Journal of Nuclear Materials*, *564*, 153696.

Mass gain

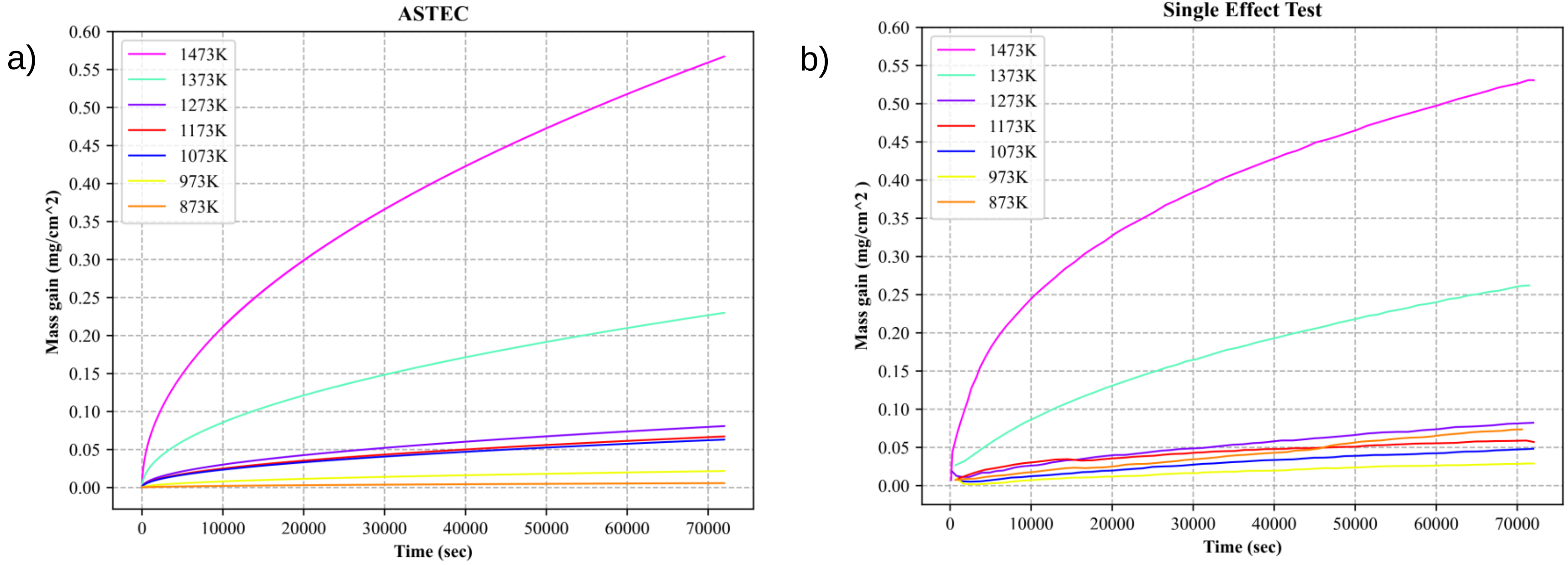


Fig. 5 Weight gains a) obtained from ASTEC b) from isothermal single effect test (extracted data from*)

*Kim, C., Tang, C., Grosse, M., Maeng, Y., Jang, C., & Steinbrueck, M. (2022). Oxidation mechanism and kinetics of nuclear-grade FeCrAl alloys in the temperature range of 500–1500 C in steam. *Journal of Nuclear Materials*, *564*, 153696.

ASTEC vs. Experiments Comparison

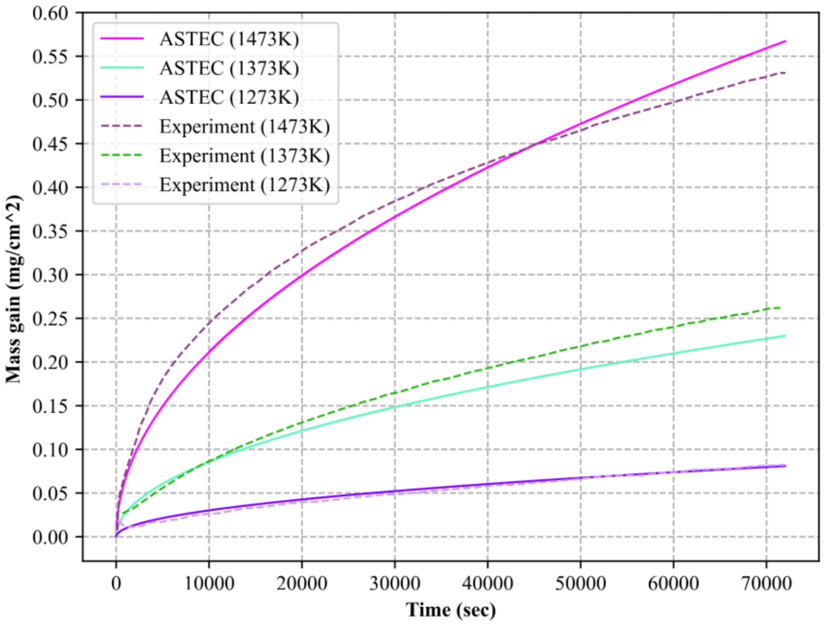


Fig. 6 Weight gains comparison Isothermal single effect test vs ASTEC

- The improvements allow ASTEC to reasonably well predict the experimental results.

Total Mass Gain 'ASTEC'

- 1373K

  $\Delta m = 0.567$
- 1473K

  $\Delta m = 0.229$
- 1573K

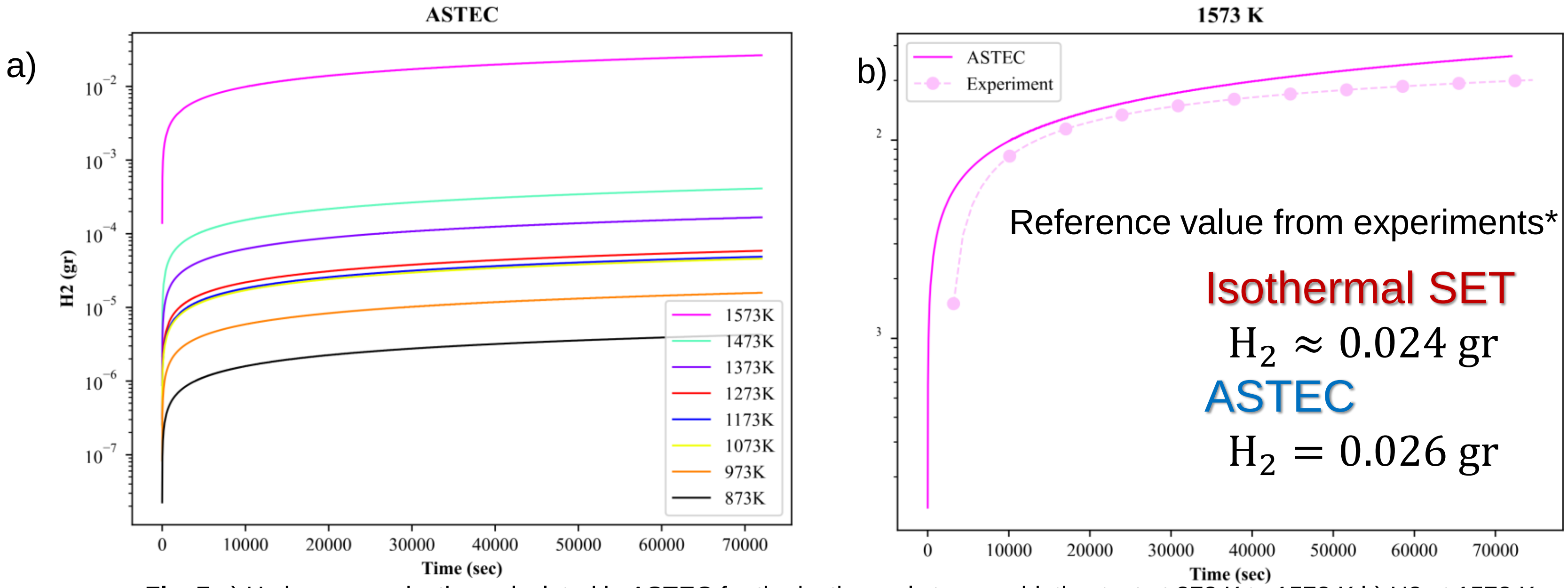
  $\Delta m = 0.080$

Thickness oxide layer

T (K)	Experiment Inner Surface (µm)	ASTEC Inner Surface (µm)	Difference Inner Surface (µm)	Experiment Outer Surface (µm)	ASTEC Outer Surface (µm)	Difference Outer Surface (µm
873	Nodular oxides	0.047	---	Too thin	0.047	---
973	Thin thickness	0.017	---	Thin thickness	0.175	---
1073	Thin thickness	0.509	---	Thin thickness	0.509	---
1173	Thin thickness	0.543	---	Thin thickness	0.543	---
1273	Thin thickness	0.654	---	Thin thickness	0.654	---
1373	~2.000	1.864	0.136	~1.500	1.864	0.364
1473	~4.000	4.605	0.605	~3.000	4.608	1.608
1573	~6.500	8.258	1.758	~6.500	8.258	1.758

- ASTEC results are in reasonable agreement with the experimental data

Hydrogen production



Reference value from experiments*

Isothermal SET

$H_2 \approx 0.024 \text{ gr}$

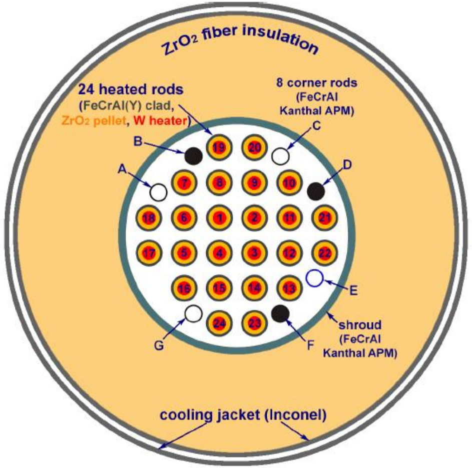
ASTEC

$H_2 = 0.026 \text{ gr}$

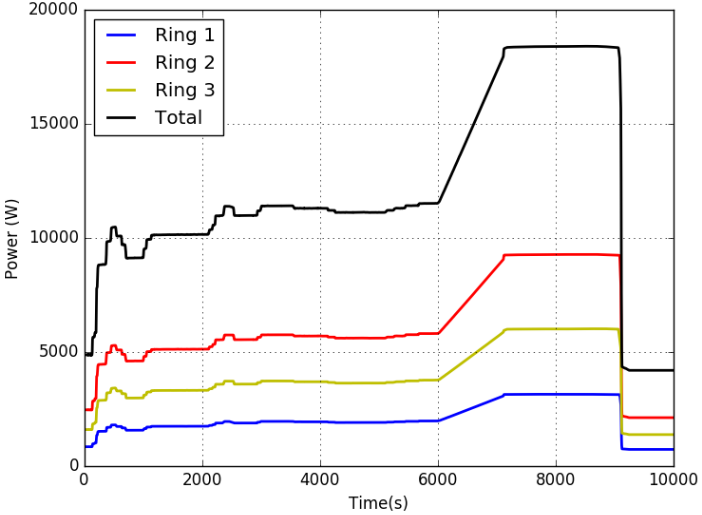
Fig. 7 a) Hydrogen production calculated in ASTEC for the isothermal steam oxidation test at 873 K to 1573 K b) H2 at 1573 K

*Kim, C., Tang, C., Grosse, M., Maeng, Y., Jang, C., & Steinbrueck, M. (2022). Oxidation mechanism and kinetics of nuclear-grade FeCrAl alloys in the temperature range of 500–1500 C in steam. *Journal of Nuclear Materials*, *564*, 153696.

QUENCH 19 test conduct

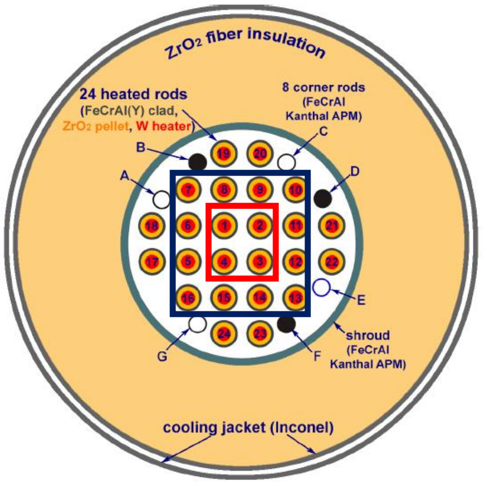


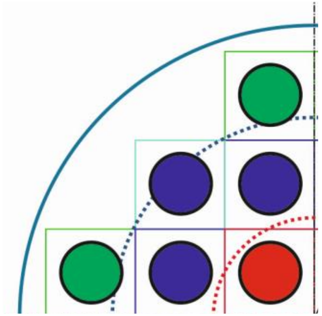
Phase 1	Heating up to ~600 °C (4 kW).
Phase 2	Power increase up to 11.5 kW (pre-oxidation).
Phase 3	Power increased up to 18.12 kW (5 W/s) (T_{pct}~1500 °C).
Phase 4	Phase 4: power reduced to 4.1 kW.

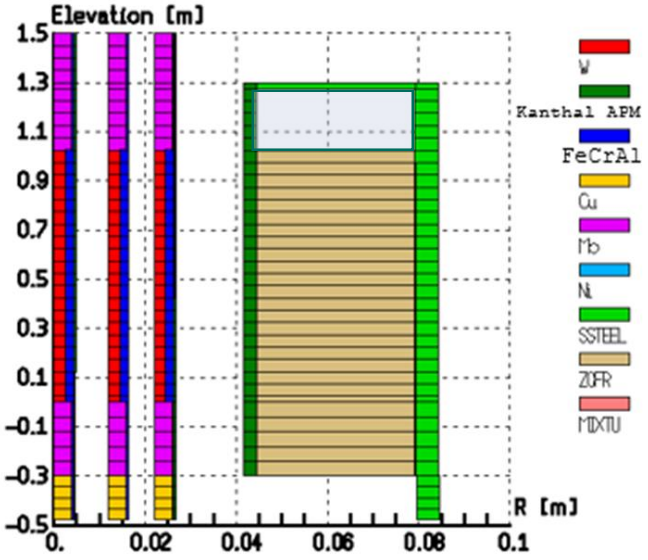


- ➢ Atmosphere of Ar (3.45 g/s) and superheated steam (3.6 g/s).
- ➢ Reflooding at ~9100 s
  - ➢ Fast initial injection of 4 L of water
  - ➢ Slow injection 48 ~ g/s of water

ASTEC model of QUENCH 19







Ch. 3, 8 rods, r_{ext}= 41.5 cm

Ch. 2, 12 rods, r_{ext}= 28.4 cm

Ch. 1, 4 rods, r_{ext}=14.2 cm

- Accidental presence of 4 l of water in the gap between the shroud and the cooling jacket modelled (J. Stuckert).

Cladding temperature

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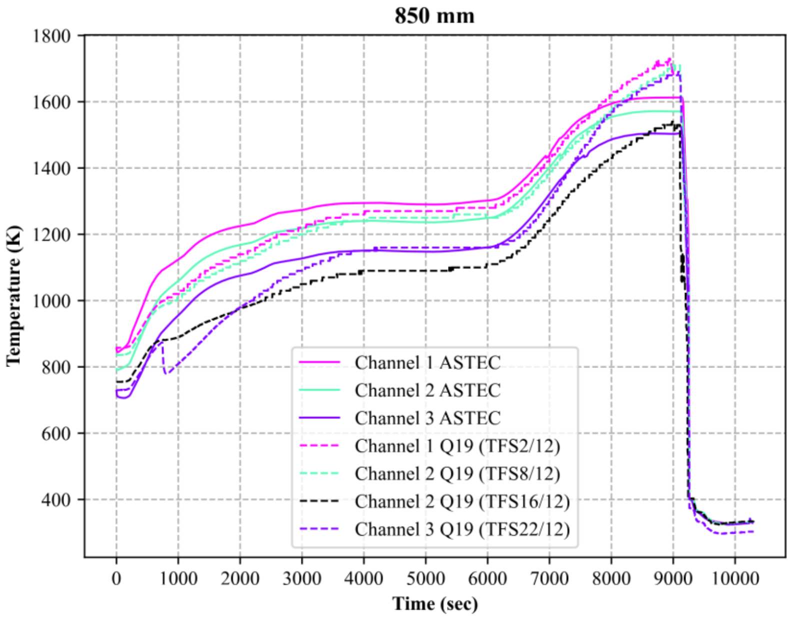
a)



b)

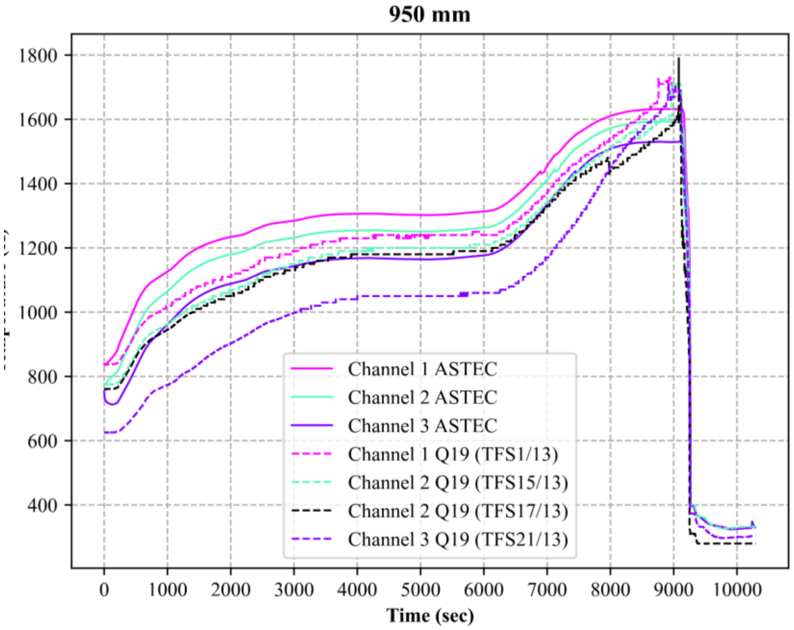


Fig. 8 Comparison Clad Temperature ASTEC vs bundle test a) 850 and b) 950 mm

- ➢ Higher temperatures from ASTEC
- ➢ Different behavior of temperature curves

Hydrogen production

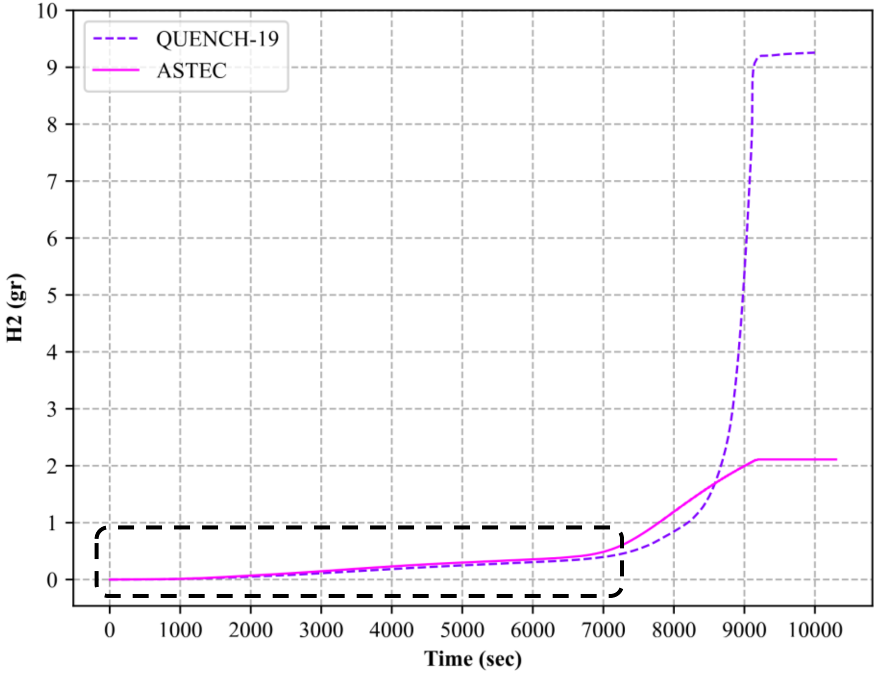


Fig. 9 Comparison total amount of hydrogen production from ASTEC vs bundle test QUENCH-19

- Experiment
  $H_2 = 9.3$ grams
- Cladding oxidation process
  $H_2 = 7.2$ grams
- ASTEC
  $H_2 = 2.1$ grams

Conclusion

- A KIT/INR and IRSN shared activity has been going on to improve the ASTEC modelling for FeCrAl cladding behavior in steam environment at low and moderate temperatures
- Modelling extensions have been implemented in ASTEC also based on the physical models resulting from the experimental investigations by the QUENCH team
- Validation of the improved ASTEC FeCrAl modelling has been performed
- QUENCH SETs:
  - ASTEC rather well predicts the experimental data with respect to Weight gain, Thickness oxide layer, and Hydrogen production in the experimental temperature range (873 K – 1573 K)
- QUENCH-19 bundle test
  - ASTEC well reproduces the experimental temperatures and hydrogen production as long as T<1500K
  - At very high T, important deviations appear in temperatures and H2 production, due to current model limitations:
    - Oxidation of Al only in the model overestimates Al_2O_3 layer and leads to a too strong stable protective role
    - Real Fe oxidation must be included in the modelling, with corresponding oxide layers

Outlook

- Application of the improved ASTEC modelling capabilities to postulated accidental scenarios in a generic SMR

- Application of ASTEC to the activities on the Cr-coated cladding materials in the frame of the OECD/NEA QUENCH-ATF project

Thank you for your attention