

CORROSION & DECOMMISSIONING

*R&D studies at CEA “corrosion service” in relation
with light water reactor accidents & decommissioning*

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First Anniversary of the Center for Fundamental Research
on Nuclear Decommissioning (CFReND)

Tohoku University, Sendai, Japan

December 1st, 2017

Introduction / background

Thermodynamics (corium)

ATTILHA setup

Database

Material behavior & irradiation

Experimental facilities

Carbon & Stainless steels

Long term issues

Atmospheric corrosion

Nuclear facility life cycle

- Siting of nuclear facility
- Design of nuclear facility**
- Construction & commissioning**
- Operation, management & maintenance**
- Severe accident management**
- Decommissioning of nuclear installations**
- Nuclear waste management**



Bold type: corrosion issues

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Decommissioning of nuclear reactors

- 110 commercial power reactors, 48 prototypes and over 250 research reactors have been retired from operation.
- About 17 of these had full decommissioning.
- 12 reactors closed after an accident/incident, including 8 (partial) core melts:
 - TMI (USA-1979)
 - Chernobyl (Ukraine – 1986)
 - Fukushima Daiichi (Japan, 2011)
- Corrosion issues**
 - During the pre-dismantling period when dismantling is not immediate
 - Specific corrosion issues after core meltdown
 - Nuclear waste interim & geological storages

Importance of corrosion during decommissioning

- Structural materials have been chosen for reactor standard operation, investigated for their behavior during accidents, but not for decommissioning conditions while they still have functions (mechanical, tightness,...)
- Long term corrosion issues (deferred dismantling)
- After core meltdown:
 - knowledge of the corium interactions, fuel debris and fission products (physico-chemical status) & behavior
 - corrosion & radiations
 - behavior of key components after accident conditions

⇒ Development of fundamental & technical knowledges

Subjects related to decommissioning investigated at the CEA “Corrosion Service”

Thermodynamics (corium)

Data acquisition

Database

Material behavior & irradiation

Experimentations *and radiolysis calculations*

Carbon steels & Stainless steels

Long term issues

Atmospheric corrosion / *Microbial Influenced corrosion*

Corrosion modelling & simulation

HIGH TEMPERATURE THERMODYNAMIC TOOLS FOR THE INVESTIGATION OF CORIUM BEHAVIOR

Experimental setup

Thermodynamic modeling

FROM RESEARCH TO INDUSTRY

cea den In-vessel & Ex-vessel Core Melts

Issue: core melt configurations

In-vessel corium Interactions:

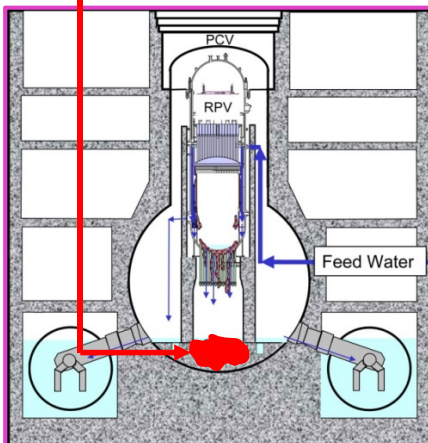
MOx + Zircaloy + stainless steel + Inconel + B₄C + Fission Products

“Prototypic” in-vessel corium system: **U-Zr-Fe-O**

The **Molten Corium Concrete Interaction** starts:

- The components of the concrete (CaO, SiO₂, Al₂O₃, MgO, H₂O, CO₂) are added to the already complex in-vessel system (U-Zr-O-Fe)
- As a first approximation the **Fe-U-Zr-Al-Ca-Si-O** system is representative of an ex-vessel corium

Fukushima



A better thermodynamic description of the in & ex-vessel corium sub-systems is needed to improve the thermal and thermo-hydraulics codes accuracy

Need of exp. data at very high temperatures
1500°C ≤ T ≤ 3200°C

Development of a specific exp. setup

Advanced Temperature and Thermodynamics Investigation by a Laser Heating Approach



ATTILHA: Development of a setup for high solid/liquid transitions

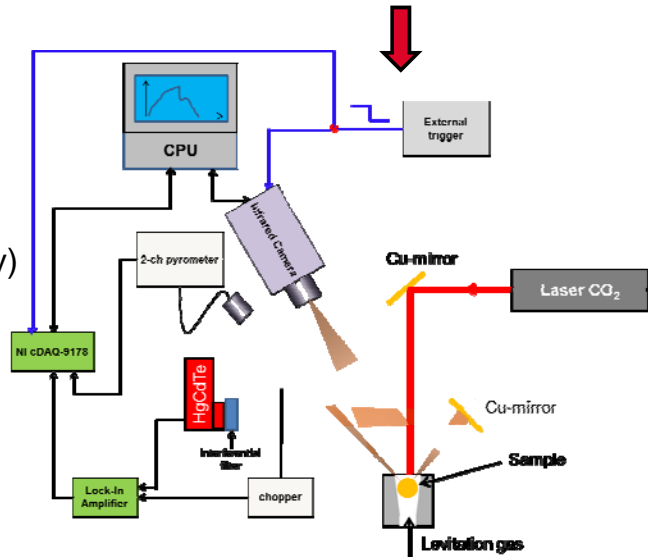
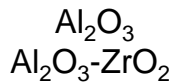
ATTILHA configuration: Contactless → Aerodynamic levitation

Acquisition of data on corium systems

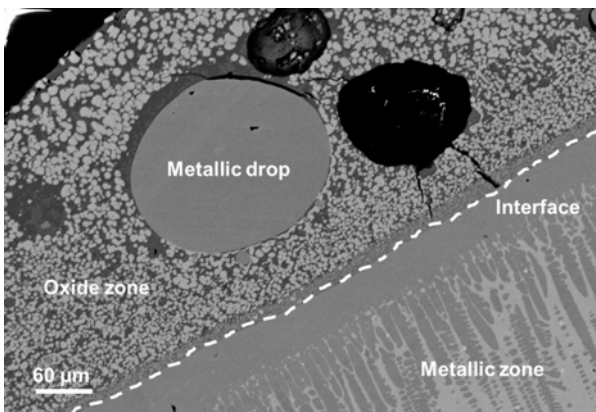
- Phase diagram data (liquidus, solidus)
- Thermo-radiative properties (IR emissivity)



All the instruments are synchronized
Validation on transitions in oxide systems



Miscibility gap in the Fe-Zr-O system



New result

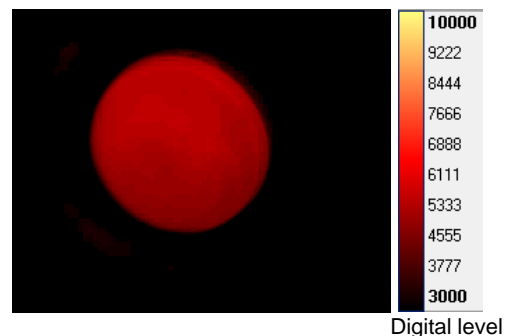
Starting composition: $\text{Fe}_{0.85}\text{Zr}_{0.15}$

Levitation gas: He

Composition moved into the ternary Fe-Zr-O system

tie-line: $\text{Fe}_{0.97}\text{O}_{0.03}$ – $\text{Fe}_{0.05}\text{Zr}_{0.32}\text{O}_{0.63}$

Infrared camera footage



Real speed 200 Hz
Video player 12.5 Hz

Observation of dynamic phenomena:

→ **Formation of 2 liquids in-situ**

Estimation of the emissivity ratio between the two liquids

→ $\epsilon_{\text{oxide}} \sim 2\epsilon_{\text{metal}}$

Development of Thermodynamic Databases using Calphad

OECD/NEA TAF-ID Project

AIM: To develop a **thermodynamic database** as a **computational tool** to perform thermodynamic calculations on advanced fuel materials using the Calphad method for **Phase diagrams & Thermodynamic properties**

CONTEXT: Fuel materials for Generation 2,3 & 4 reactors

APPLICATIONS

Fuel behaviour at high temperature under normal and off-normal conditions

Fuel fabrication & Fuel / cladding chemical interaction at high temperature

2013-2017 (version 1) / 2017-20xx (version 2) – coordinated by OCDE/NEA

TCOFF Project

AIM: The TCOFF project represents a suitable framework for exchange of technical information on topics related to thermodynamic databases available for modelling the fuel/core degradation process, which include thermodynamic functions/models, experimental data, calculation tools, calculation methods for quasi-equilibrium systems.

CONTEXT: The decommissioning of the Fukushima-Daiichi Nuclear power plants

APPLICATIONS

Task Force on “Thermodynamic characterisation of corium and fuel” (TF-1)

Task Force on “Thermodynamic characterisation of fission products” (TF-2)

CEA and JAEA experts have been collaborating for many years on thermodynamic studies for a better knowledge of the high temperature behavior of materials during severe accident events

OECD Projects:

- TAF-ID (Chair C. Guéneau – CEA)
- TCOFF (Chair M. Kurata – JAEA)

Submission of a common paper on the thermodynamic modeling of the U-Zr-O system in Journal of Nuclear Materials (Accepted):

Contribution to the thermodynamic description of the corium - the U-Zr-O system

A. Quaini, C. Guéneau, S. Gossé, N. Dupin, B. Sundman, E. Brackx, R. Domenger, M. Kurata, F. Hodaj

References :

Experimental contribution to the corium thermodynamic modelling – the U-Zr-Al-Ca-Si-O system

A. Quaini¹, C. Guéneau¹, S. Gossé¹, T. Alpettaz¹, E. Brackx², R. Domenger², A. Chocard², F. Hodaj^{3,4}



High temperature investigation of the solid/liquid transition in the PuO₂-UO₂-ZrO₂ system

A. Quaini¹, C. Guéneau^{1,2}, S. Gossé¹, B. Sundman³, D. Manara⁴, A.L. Smith⁵, D. Bottomley⁶, P. Lajarge¹, M. Ernstberger⁷, F. Hodaj^{1,4,8}

MATERIAL BEHAVIOR & IRRADIATION

- EXPERIMENTATION

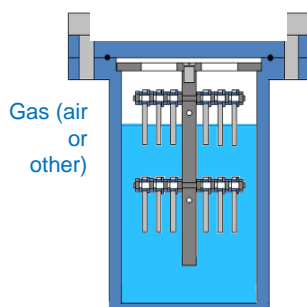
- CARBON & STAINLESS STEELS



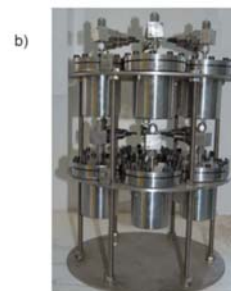
CORROSION & IRRADIATION

Experimental facility: CASIMIR

Gamma irradiation performed in the POSEIDON pond, at CEA-Saclay



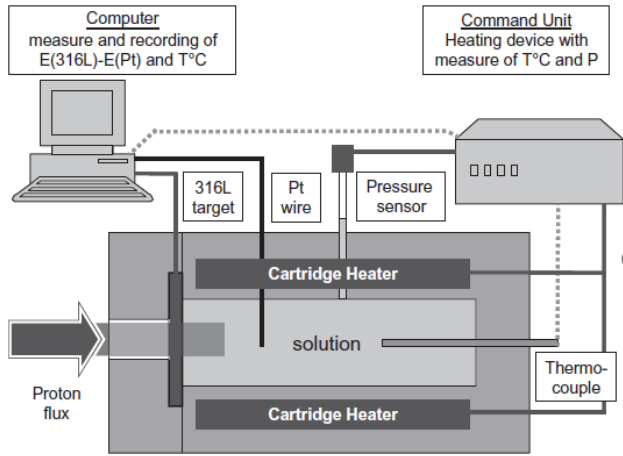
Water chemistry



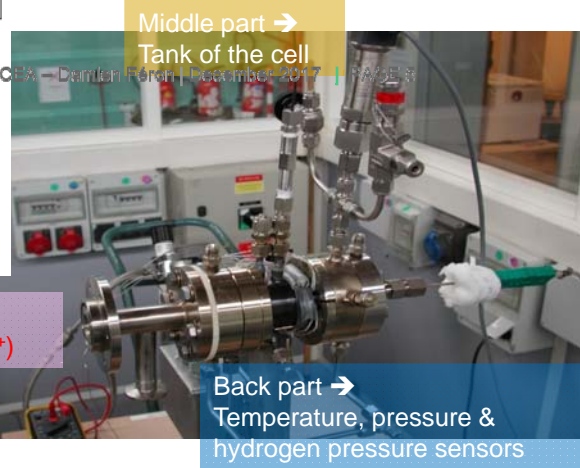
S. Perrin & al., LTC2016, Toronto, 2016



High temperature and high pressure electrochemical cell



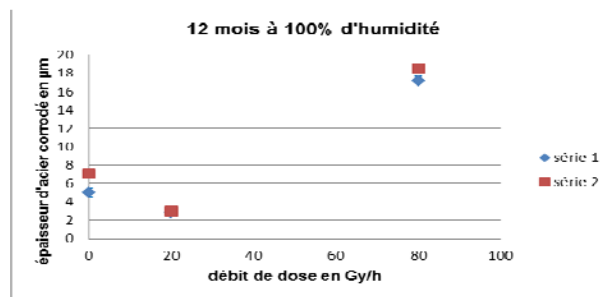
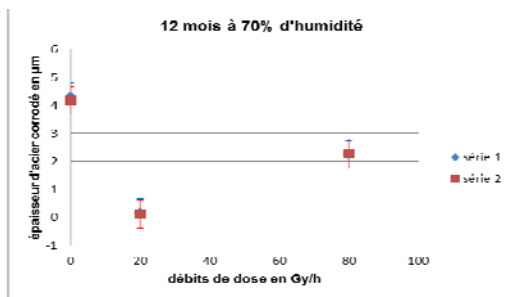
Temperature → 300 (°C)
 Pressure → 90 (bar)
 Hydrogen pressure → 400 (mbar)
 Free potential → E_{316L} vs. Platinum (V)



Brevet n° 08 56970
 du 15.10.2008, CEA (DEN & DSM)-LSI

ATMOSPHERIC CORROSION OF CARBON STEEL

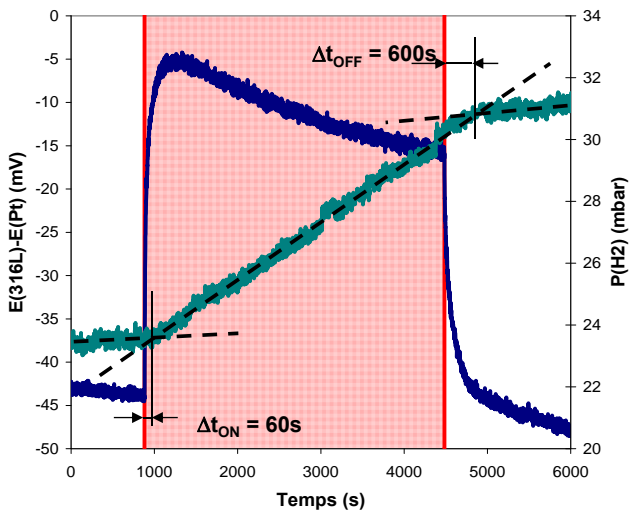
Corrosion of carbon steel (A37) in a closed humid atmosphere
 Influence of the humidity at temperature 80°C,
 and of gamma irradiation (20 & 80 Gy.h⁻¹)



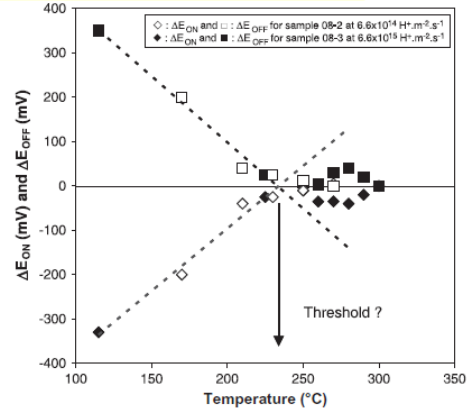
S. Perrin & al., LTC2016, Toronto, 2016

Generalized corrosion, but not uniform
 Lower corrosion rates at 70% RH (relative humidity) than at 100% RH, with or without irradiation
 Highest corrosion rates at 100% HR and 80 Gy.h⁻¹

Effects of radiolysis on the electrochemical behaviour of stainless steels



Potential and hydrogen partial pressure evolution at 300°C, proton flux = $6.6 \times 10^{11} \text{ H}^+ \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$



- PWR water chemistry (300°C) /316L
- in situ measurement of the radiolytic production of H_2
- Importance of the temperature

B. Muzeau & al., JNM 419 (2011) 241–247

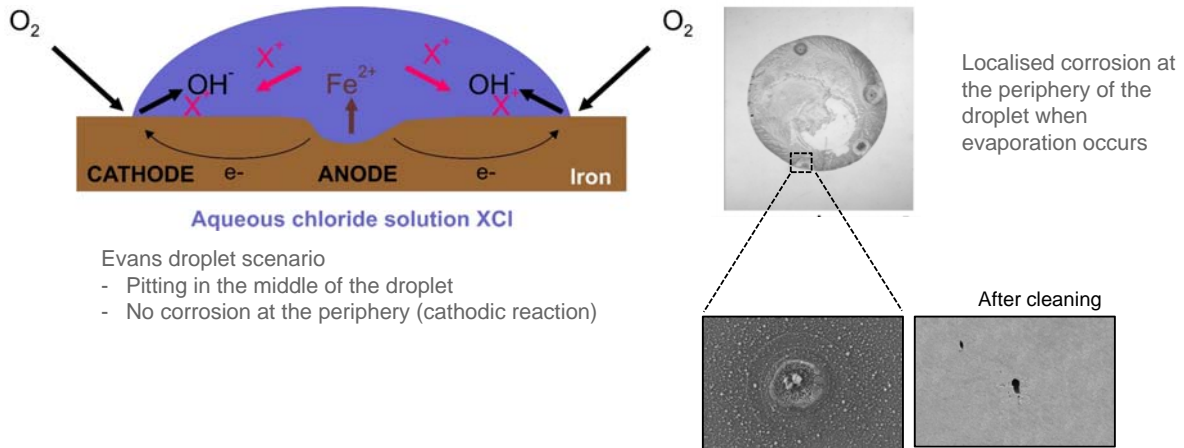
LONG TERM ISSUES

- ATMOSPHERIC CORROSION
- *Microbial corrosion*
- *Passivation & depassivation*
- *Corrosion modelling & simulation*

Phenomena: outdoor and indoor atmospheric corrosion, including closed atmospheres.

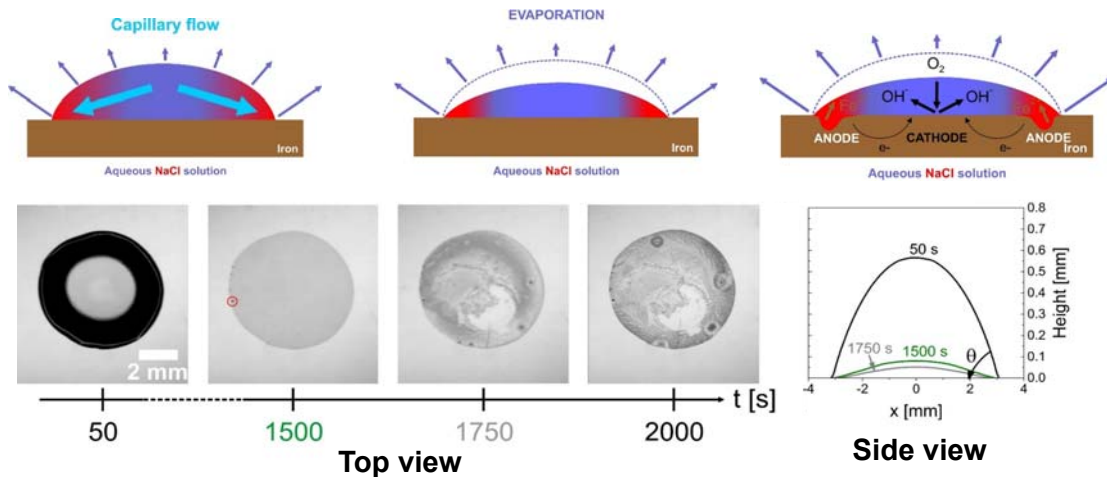
Applications: deferred dismantling, interim storage, first period of geological disposal.

Observations: shape of localised corrosions not in accordance with the well-known “Evans” droplet.



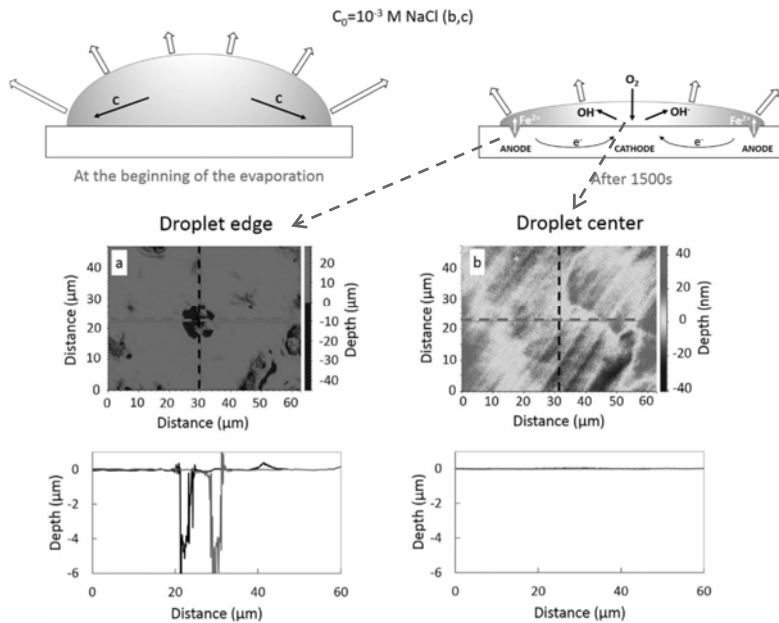
Corrosion under Evaporating Salty Sessile Droplets

Low salt concentration: $c_0 = 10^{-3}M$ NaCl



Evaporation of pinned salty sessile droplets causes **peripheral salt enrichment**
Local chloride enrichment promotes the **initiation of corrosion**

Strong correlation between evaporation process and localization of corrosion phenomena



Schematic diagram of the processes

Experimental observations on iron

Soulié, Lequien & al., Materials and Corrosion, Sept. 2017, Vol. 68 Issue 9, p927-93

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CONCLUSION

Decommissioning and corrosion

Fundamental knowledge needed in several areas

Thermodynamics

Effects of irradiation (low & high doses)

Corrosion phenomena even at low temperatures

Modeling and simulation

Prediction of phases and compounds in melt core

Long term prediction & coupling of multi-corrosion issues

Exchanges of information & collaborations

CEA - Tohoku U/CFReND - JAEA/CLADS cooperation
for safe decommissioning of 1F

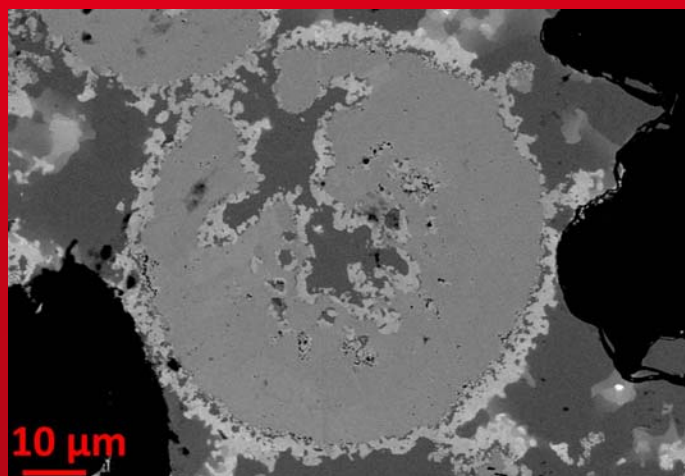
Fukushima Research Conference on
Corrosion Prediction and Mitigation for Key Components of Fukushima Daiichi NPS

Nov. 27-29, 2017
Tomioka-cho, Fukushima



◆ Long-term prediction of carbon steel corrosion
◆ Radiolysis effect on corrosion
◆ Re-passivation of localized corrosion
◆ Microbial influenced corrosion
◆ Reinforced concrete corrosion

Conference chair: Damien Feron (CEA)
Vice-chairs: Y. Watanabe (Tohoku)
M. Yamamoto (JAEA)



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