Overview on nuclear corrosion - Corrosion in the nuclear cycle

D. Feron

OVERVIEW ON NUCLEAR CORROSION

CORROSION IN THE NUCLEAR CYCLE

Nuclear corrosion sumer school | NuCoSS-15, Bled, Slovenia | July 5-10, 2015

Damien Féron, CEA/DEN/DPC/SCCME

JULY 2015

NUCLEAR CORROSION IN THE NUCLEAR CYCLE

Content: illustrations of corrosion processes in the nuclear cycle but far from the "traditional" water corrosion phenomena

Background

Reactors

- Liquid metal
- Gas

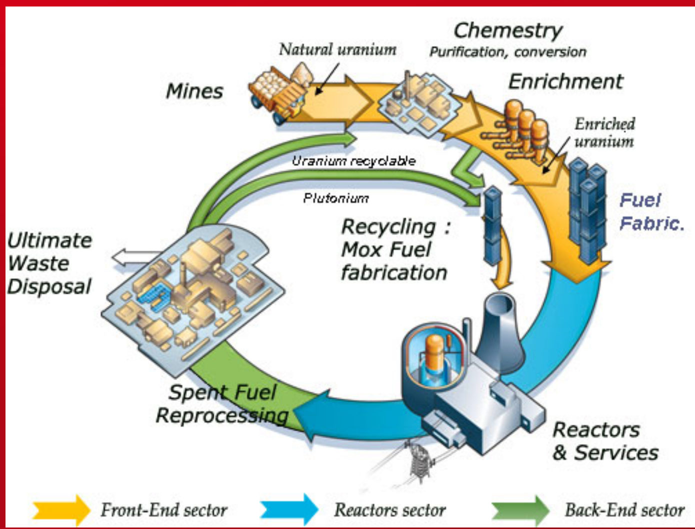
Reprocessing plants

- Nitric acid

Geological disposal

- Long term

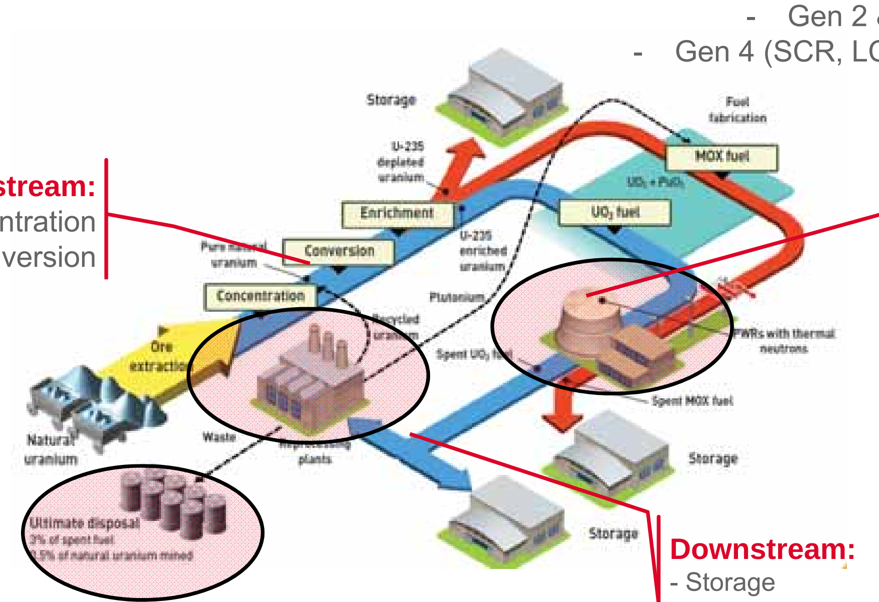
BACKGROUND



NUCLEAR CYCLE & CORROSION

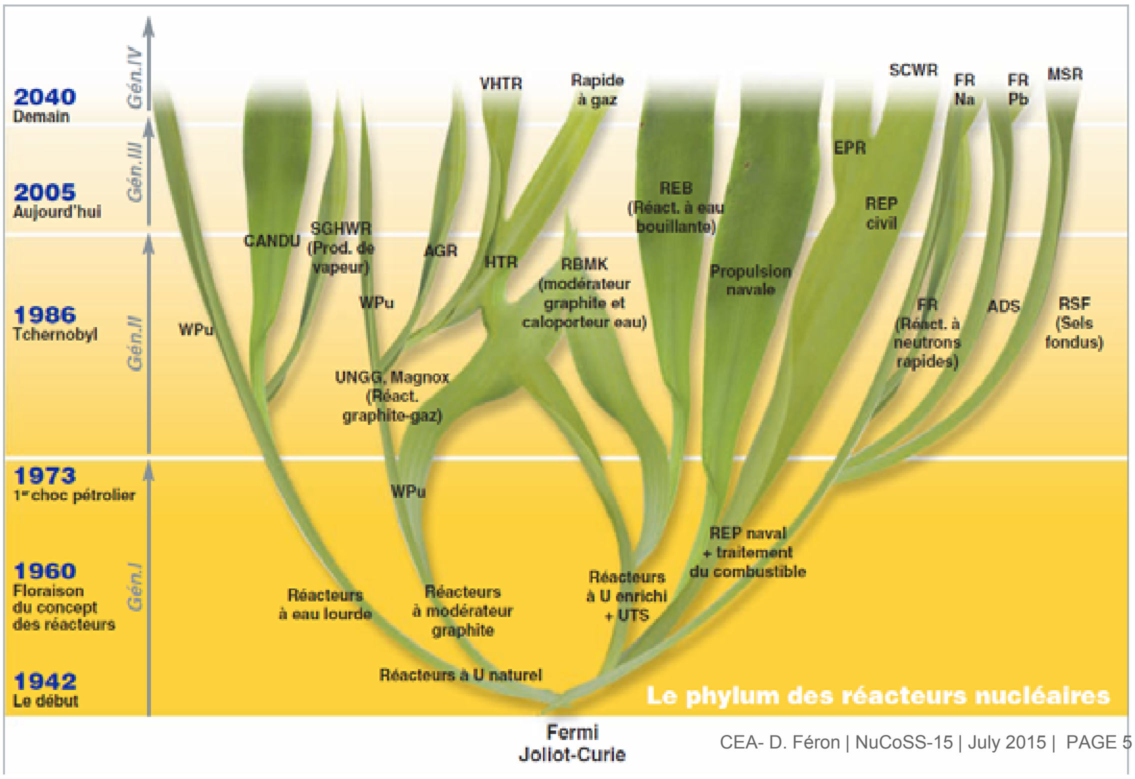
Nuclear reactors
- Gen 2 & 3 (PWRs)
- Gen 4 (SCR, LCR, GCR…)

Upstream:
- Concentration
- Conversion

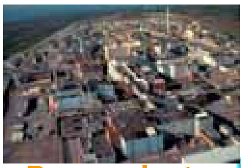


Downstream:
- Storage
- Reprocessing (nitric acid & glass)
- Disposals

DIVERSITY OF NUCLEAR POWER PLANTS (NPP)



NUCLEAR CORROSION FROM YESTERDAY TO TOMORROW



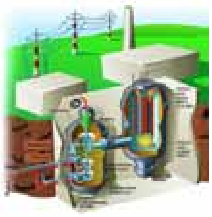
Reprocessing

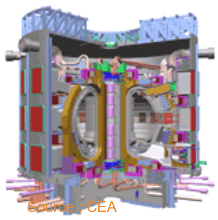
Generation IV and Fusion

New facilities

Power plants

First facilities





1950 1970 1990 2010 2030 2050 2070 2090

Material selection

Nuclear
Waste
disposal

Failures investigations
Performance increases

Life time (Power & other plants, …)
Long term prediction (HLNW disposal)
High temperatures (innovative materials)

NUCLEAR CORROSION
NUCLEAR PLANT OPERATING TIME

Safety improvements

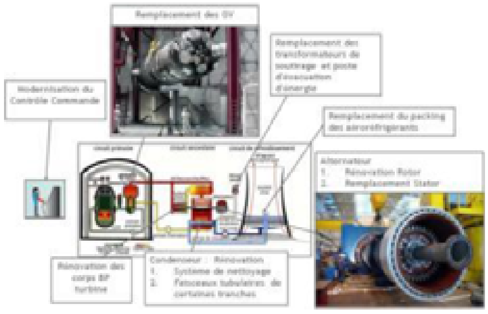
- **Ten-year inspection revaluation**
  - Safety inspection every ten years
  - One file per reactor submitted by EDF
- **40-year key stage: enhance the safety with the objectives of new reactors**
  - REDUCE the consequences of a core meltdown
  - REDUCE the probability of a core meltdown occurring
  - Meet WENRA* requirements as closely as possible
- **Coexistence of GEN 2 and GEN 3 (EPR)**

Confirmation of objectives and acceleration after Fukushima

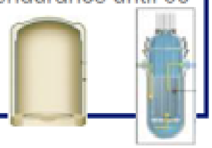
*Western European Nuclear Regulators Association

Ageing management

- **Replaceable components: Define the best compromise between routine maintenance/preventive replacement**



- **Non-replaceable components**
  - Vessel: Demonstration of endurance until 60 years
  - Containment: Repairs

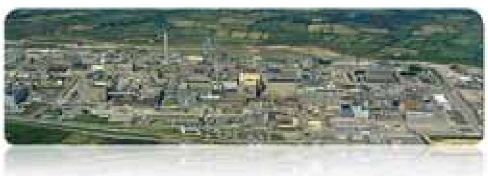


From C. Ancelin, Fontevraud 8, September 2014

ENVIRONMENTS IN THE NUCLEAR CYCLE

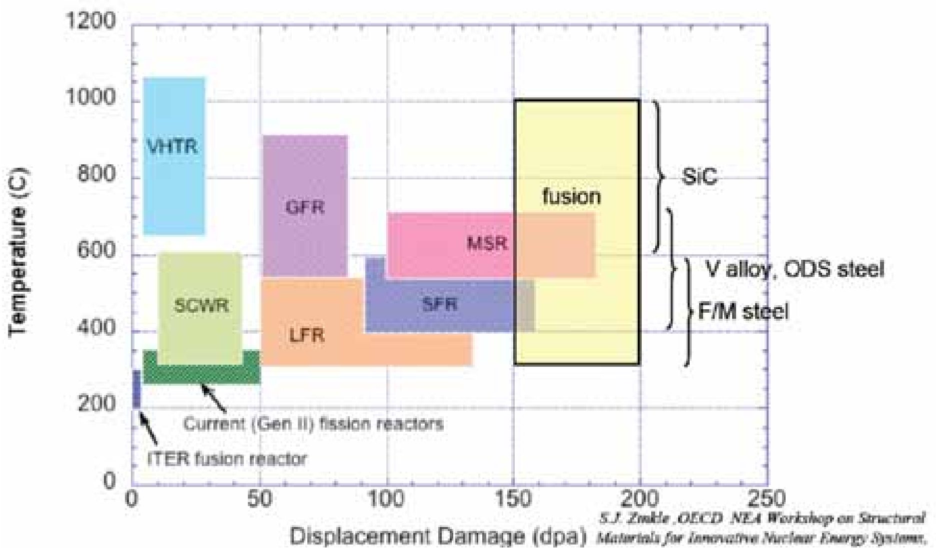
- **Upstream**
  - Sulfuric acid and fluorine environments
- **Fission power plants**
  - Light water reactors
  - Heavy water reactors
  - Supercritical water reactors
  - Liquid metal reactors (Na, Pb & Pb-Bi)
  - Gas cooled reactors (helium)
  - Molten salt reactors
- **Fusion**
  - Liquid metal (Li, Pb-Li)
  - Gas
  - Water
- **Downstream**
  - Nitric acid (reprocessing plants)
  - Liquid glass (high radioactive nuclear waste)
  - Atmospheric corrosion (interim storage)
  - Geological environments (final nuclear waste disposals)

GEN IV International Forum



EVOLUTION OF REACTOR ENVIRONMENTS

Comparison of Gen II – Gen IV and fusion environments of structural materials



S.J. Zinkle, OECD NEA Workshop on Structural Materials for Innovative Nuclear Energy Systems.

Three main evolutions

- **Increase of temperatures from 350°C to >1000°C**
- **Increase of dose from 10 dpa to 200 dpa**
- **Increase operating time from 40 years to ….**

MATERIALS IN THE NUCLEAR CYCLE

Non specific and specific materials

Materials with "standard" properties

- **functions:**
  Structural materials
  Containment barriers
  Confinement matrix
- **types:**
  Metallic alloys
  oxides (ceramics, glass…)
  graphite
  concrete
  polymers

Materials with nuclear properties

- **functions:**
  Fuel
  Neutron absorbing materials
  Neutron non- absorbing materials
- **types:**
  ceramics (UO_2, B_4C)
  Metallic alloys (AIC, Zircaloy)

from bibliography I

MATERIALS IN THE NUCLEAR CYCLE

Example of specific and non-specific materials with Zr alloys (« common » and « nuclear » materials)

Alloy	Zr	Zr 702*	Zircaloy 2	Zircaloy 4	Zr- 1 Nb	Zr- 2.5 Nb
Sn %			1.2-1.7	1.2-1.7		
Nb %					1	2.4-2.8
Fe %		0.20% max (Fe+Cr)	0.07-0.20	0.18-0.24		
Cr %			0.05-0.15	0.07-0.13		
Ni%			0.03-0.08			
O ppm		0.16% max	1200-1400	1200-1400	1200-1400	1200-1400
Co ppm	20		20	20	20	20
Hf ppm	100	4.5% max	100	100	100	100
U ppm	3.5		3.5	3.5	3.5	3.5
Application	Reprocessing plants	Reprocessing plants	LWR	LWR	LWR	LWR

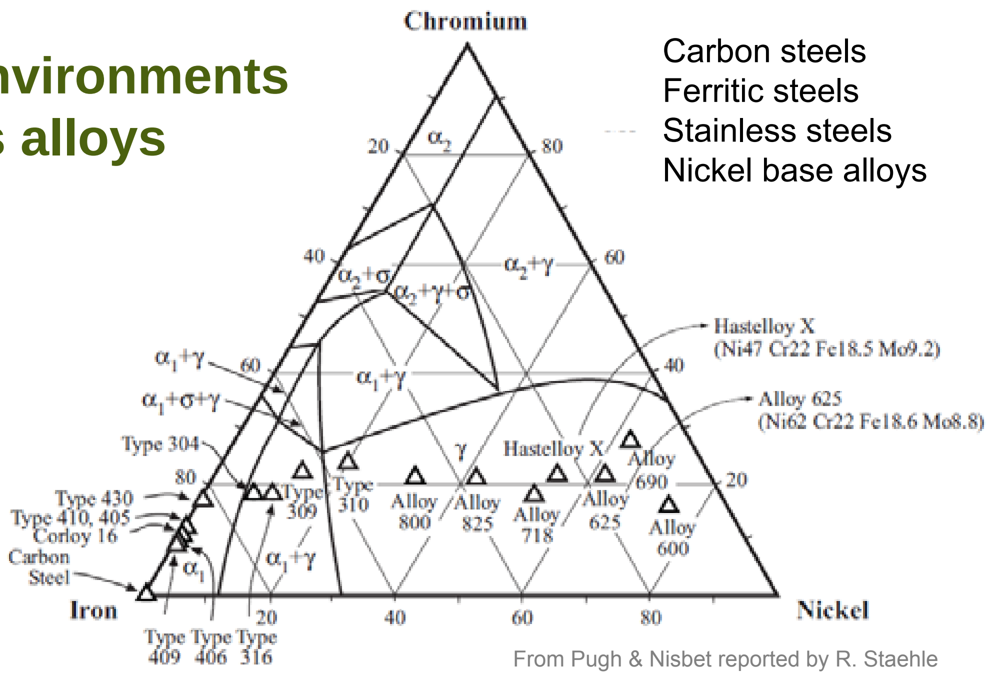
from bibliography I

Zr alloys without Hafnium have been developed specifically for cladding application

MAIN STRUCTURAL ALLOYS OF THE NUCLEAR CYCLE

Various environments
Numerous alloys

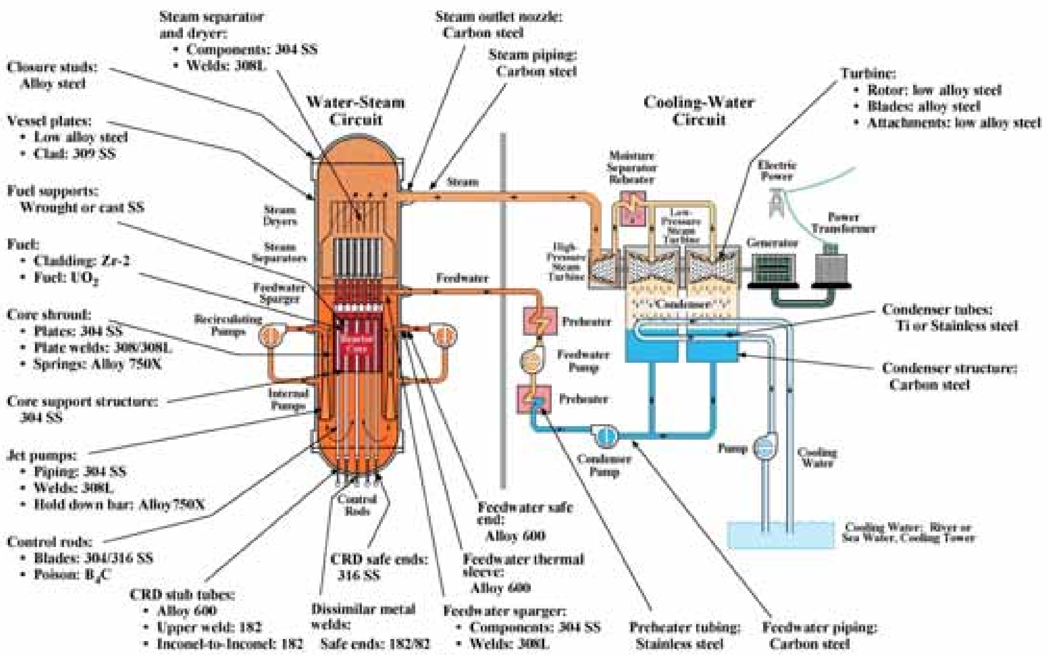
Carbon steels
Ferritic steels
Stainless steels
Nickel base alloys



From Pugh & Nisbet reported by R. Staehle

MAIN MATERIALS IN A NPP

BWR Components and Materials



From R. Staehle

MATERIALS/ENVIRONMENTS IN NPP

Reactor type	BWR	PWR	SFR	GFR	LFR	VHTR	SCWR	MSR
Coolant temperature	Water 288°C	Water 280-330°C	Sodium 370-550°C	Herlium 450-850°C	Lead or lead alloys 550-800°C	He 400-950°C	Water 280-550°C 24MPa	Molten salts (fluorines) 500-720°C
Cladding	Zirconium alloys	Zirconium alloys	Austenitis stainless steels / F/M ODS	SiC-SiC / Refractory metals	F/M steels ODS steels	Graphite SiC	Ni alloys and F/M steels	Graphite and carbon base materials
Structure	Stainless steels	Stainless steels (SG: Ni alloys)	316LN F/M steels	Refractory metals/ Ni alloys	High Si F/M steels	Ceramics ODS Ni alloys	Ni alloys and F/M steels	Graphite, carbon base materials, Hastelloy N
Dose (cladding)	10dpa (2-4dpa/an)	10dpa (2-4dpa/an)	200 dpa*	60/90dpa	150 dpa	7/25 dpa		

Cladding and structural materials used for present reactors and planned for innovative NPPs

From Nuclear corrosion science and engineering, 2012

MAIN CHALLENGES & ISSUES

Source : EDF

- Long term
  - nuclear facilities (60 years)
  - interim storage (100-300 years)
  - geological disposal (thousands of years)

Source : ANDRA

- High temperatures
  - Gen 4
  - Fusion

- Safety & liability
  - Confidence
  - Evolution

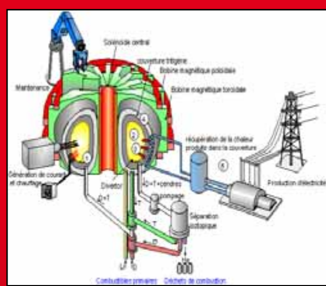
Major challenges linked with corrosion behaviour

Rapsodie

Phenix

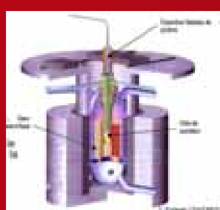
Superphenix

LIQUID METAL

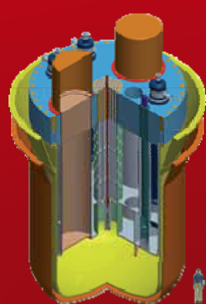


Fusion facilities

Astrid



ADS



Myrrha

Background Liquid metals

- **Liquid metals** with high thermal conductivity, high boiling point and adequately high specific heat are used to enhance **heat transfer.**
- The coolant of a **fast neutron reactor** should possess low neutron absorption cross section and be a poor moderator for achieving high neutron economy and for sustaining the hard neutron spectrum. **Liquid sodium** has been the coolant of choice for the fast neutron reactors.
- **Liquid lithium** in its pure form or as low melting **Pb-17Li** eutectic is considered as tritium breeder coolant in **fusion reactor** systems.
- **Liquid Pb and lead-bismuth** eutectic (LBE) alloy are the candidate coolants for ADS.

BACKGROUND LIQUID METALS

Thermo-physical properties of liquid metal coolants

Property	Coolant					
	Sodium	Lithium	Pb-Li	Pb	LBE	H_2O
Melting Point (K)	371	453.5	507	600.5	398	273
Boiling Point (K)	1156	1620	-	2018	1901	373
Density Kg/m^3 at 773K	845	487	9486	10520	10150	0.99 (at 323K)
Thermal conductivity at 773K (W/(K.m)	68.8	53.5	17	17.1	14.2	0.67 (at 323K)
Heat capacity kJ/(kg.K) at 773K	1.269	4.212	0.187	0.150	0.146	1.339 (at 323K)
Vapour pressure(Pa) at 773K	2 at 573K	0.08	0.002	0.002	0.0024	101325 Pa at 373K
Electrical Resistivity, Ω.m at 800K	28.28* 10^{-8}	38.2* 10^{-8}	135.2* 10^{-8}	103.6* 10^{-8}	125.8* 10^{-8}	182 * 10^{3} at 298K

from bibliography I

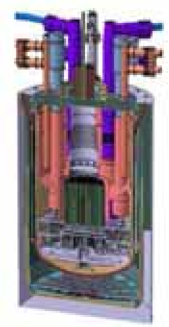
Sodium cooled reactors
“Fast breeder reactors” in the world (2013)

Reactor (Country)	Th. Power (elect.) MW	First start	Shutdown	Number of operating years
EBR-I (États-Unis) (NaK)	1,4 (0,2)	1951	1963	12
BR-5/BR-10 (Russie)	8 (0)	1958	2002	44
DFR (Royaume-Uni) (NaK)	60 (15)	1959	1977	18
EBR-II (États-Unis)	62,5 (20)	1961	1991	30
EFFBR (États-Unis)	200 (61)	1963	1972	9
RAPSODIE (France)	40 (0)	1967	1983	16
BOR-60 (Russie)	55 (12)	1968		44
SEFOR (États-Unis)	20 (0)	1969	1972	3
BN-350 (Kazakhstan)	750 (130)	1972	1999	27
PHÉNIX (France)	563 (250)	1973	2009	36
PFR (Royaume-Uni)	650 (250)	1974	1994	20
JOYO (Japon)	50-75/100 (0)	1977		35
KNK-II (Allemagne)	58 (20)	1977	1991	14
FFTF (États-Unis)	400 (0)	1980	1993	13
BN-600 (Russie)	1 470 (600)	1980		32
SUPERPHÉNIX (France)	3 000 (1240)	1985	1997	12
FBTR (Inde)	40 (13)	1985		27
MONJU (Japon)	714 (280)	1994		18
CEFR (Chine)	65 (25)	2010		2
PFBR (Inde)	1 250 (500)	En construction		
BN-800 (Russie)	2 100 (880)	En construction		
Total				418

from AIEA

LIQUID METAL & NUCLEAR REACTORS

Liquid metals

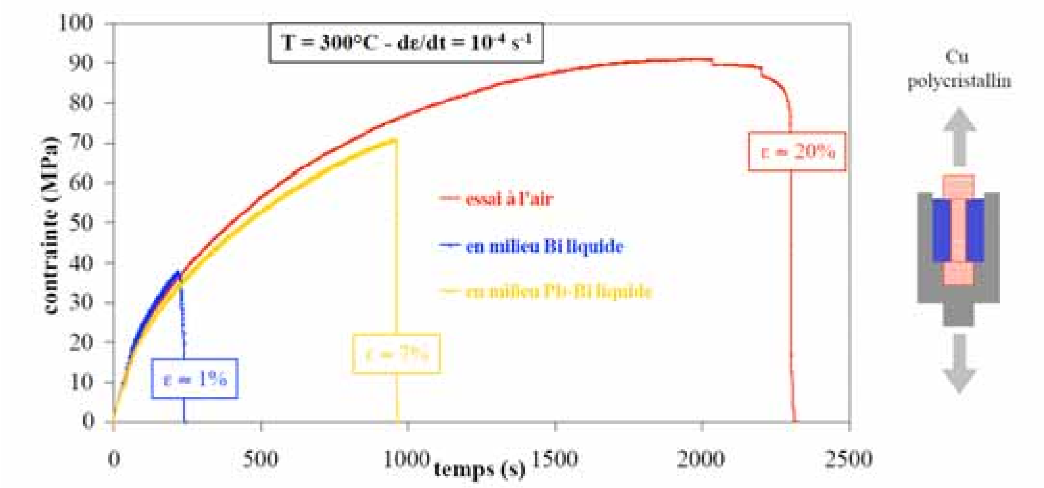


- **Sodium - SFR**
- Lead – LFR, Hybrid R. (secondary circuits of SFR)
- Lead-bismuth - LFR
- Lithium
- Lead-lithium

} Fusion

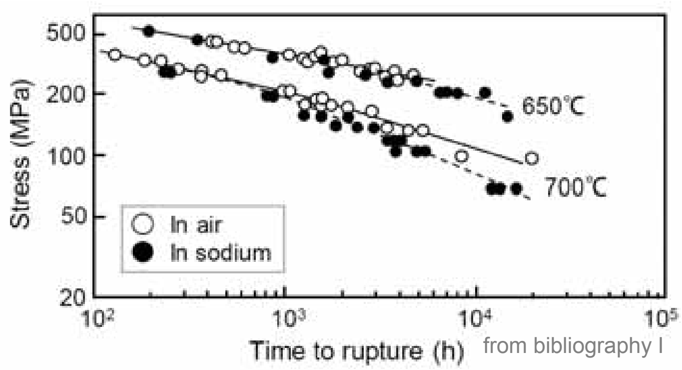
Corrosion/compatibility solid/liquid metals

- **Liquid metal embrittlement** « process resulting in a decrease of the toughness or ductility »
- General corrosion: Reaction/ Dissolution / Oxidation…
- Transfer processes (non-isothermal systems)



Dramatic decrease of the mechanical properties (solid copper in liquid bismuth a liquid Pb-Bi at 300°C)

liquid sodium



On 316L, same behaviour in pure sodium than in air (constant load testing)

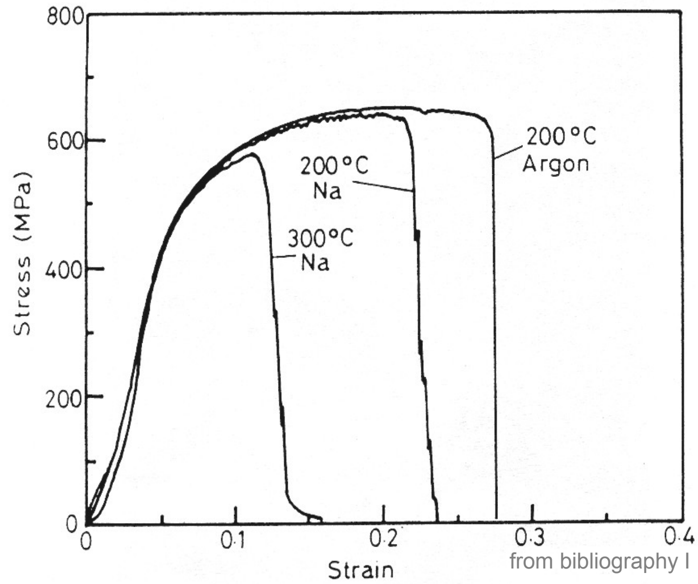
Liquid metal embritllement (LME) is often affecting metals and alloys stressed and in contact with molten metals: a major issue for components exposed to some liquid metals.

In pure liquid sodium (with low impurity content), it is generally assumed that there are **no deleterious effects (or no major effects) on rupture behaviour for ferritic and austenitic steels under constant load test conditions.**

Liquid metal embrittlement

liquid sodium

For environmental purities typical of sodium-cooled reactors, embrittlement has been reported only for low alloyed steels under severe loading conditions during slow strain tests : not relevant in normal SFR operating conditions



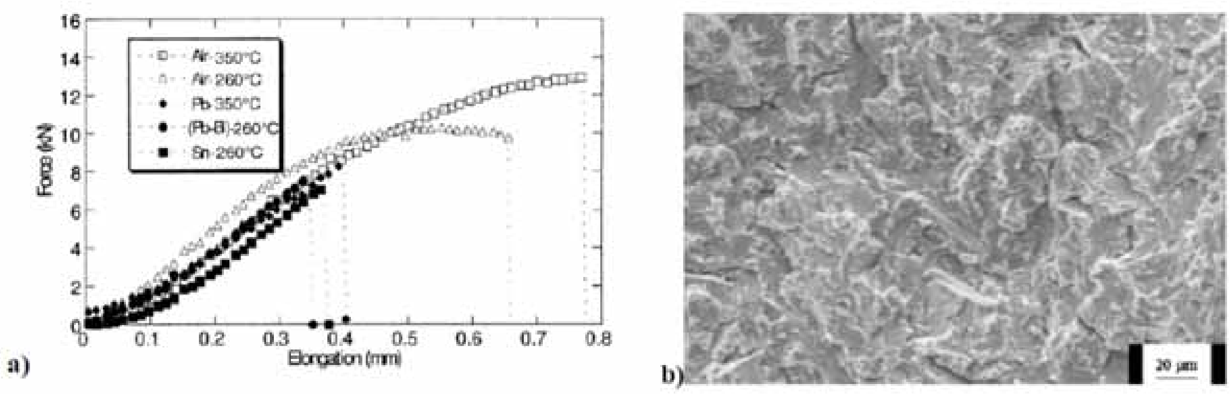
from bibliography I

Stress-strain curves for low alloyed steel in sodium and in argon at 200°C and 300°C, at 10^{-6} s^{-1}

Liquid metal embrittlement

FUSION reactors (systems)

IN LEAD, LEAD BISMUTH (AND TIN), A LOSS OF DUCTILITY IS OBSERVED ON STEELS (T-91)



from bibliography I

GENERAL CORROSION IN LIQUID METALS

- **Intermetallic compounds** formation linked to phase diagrams between the solid metal and the liquid metal

  ➔ Thermodynamics

- **Oxidation** of the steel elements (Fe, Cr, Ni, Mn, …) of the surface in contact with the liquid metal, if the oxygen content is high enough, then dissolution of the oxides and transfer of corrosion products (reaction with other dissolved species)

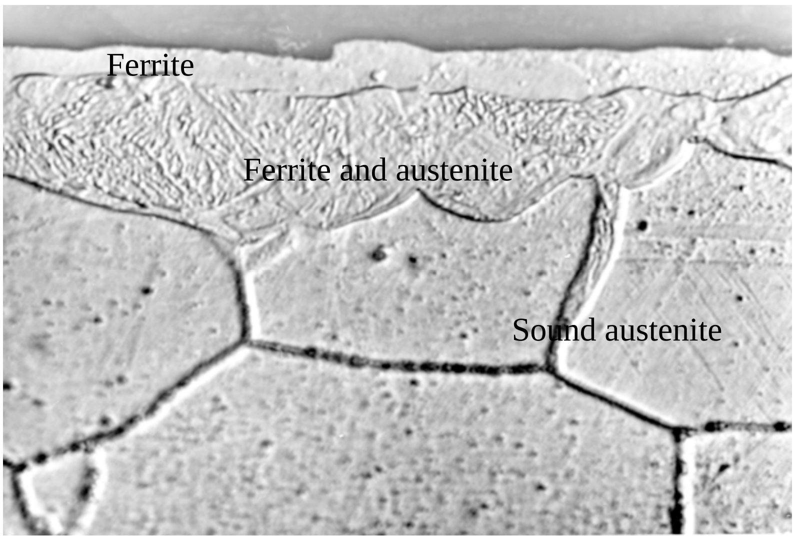
  ➔ Oxygen concentration (concentration of impurities)

- **Reaction with other impurities** (C, N….)

  ➔ Activities in solid and liquid metals

- **Dissolution** of the steel elements (Fe, Cr, Ni, Mn, …) of the surface in contact with the liquid metal, then transfer and a deposition on the reactor structures

General corrosion in liquid metal "dissolution"



from bibliography VIII

General corrosion : formation of a ferrite film at the surface (ferrite/ ferrite and austenite / Sound austenite)

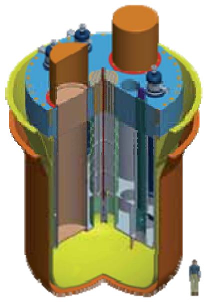
SUMMARY

✓Dissolution at low oxygen content: dissolution in Li, Na and Pb with low oxygen

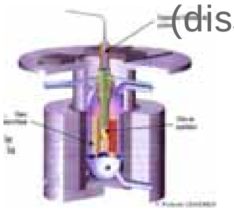
✓Oxidation may lead to a more or less protective layer.

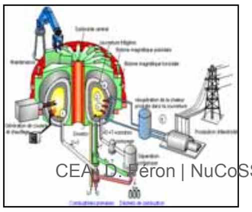
✓Ni solubilities are generally high and Ni alloys are not suitable.

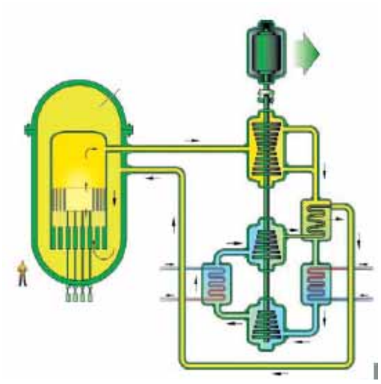
✓Liquid metal embrittlement: main issue for lead and lead eutectics.



✓Importance of the transfer processes between hot and cold parts of a circuit (dissolution/deposition).

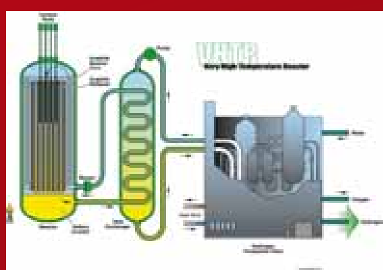






GAS (HELIUM) CORROSION

HELIUM COOLED REACTORS & FUSION



CORROSION IN GAS AT HIGH TEMPERATURE

Background

Chemical or electrochemical reaction between two phases (heterogeneous reaction)

Solide1 + Gas → Solide2

High temperature oxydation by oxygen

$M \rightarrow M^{z+} + ze^-$

$(z/4)\ O_2 + ze^- \rightarrow z/2\ O^{2-}$

$M + (z/4)O_2 \rightarrow MO_{z/2}$

- Water vapor

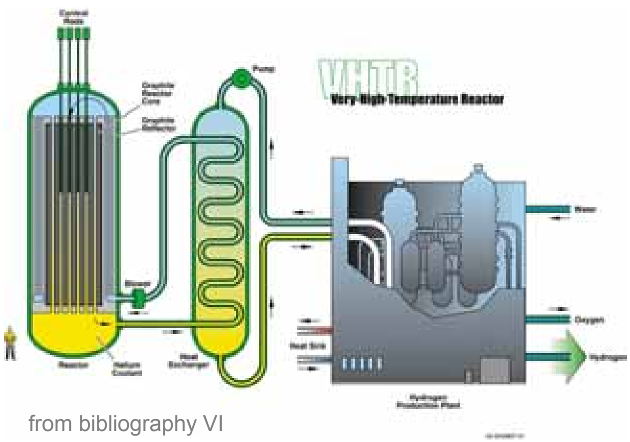
$$\nu_M M + \nu_O H_2O = M_{\nu_M}O_{\nu_O} + \nu_O H_2$$

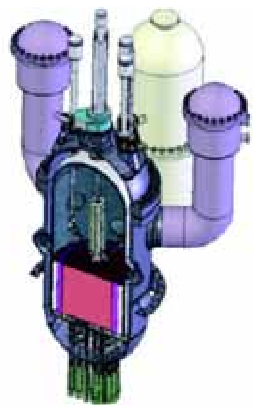
- Carbonic gas

$$\nu_M M + \frac{\nu_O}{2} CO_2 = M_{\nu_M}O_{\nu_O} + \frac{\nu_O}{2} CO$$

ILLUSTRATION

BEHAVIOR OF CHROMIA-FORMING ALLOYS IN HELIUM AT HIGH TEMPERATURE (VHTR)



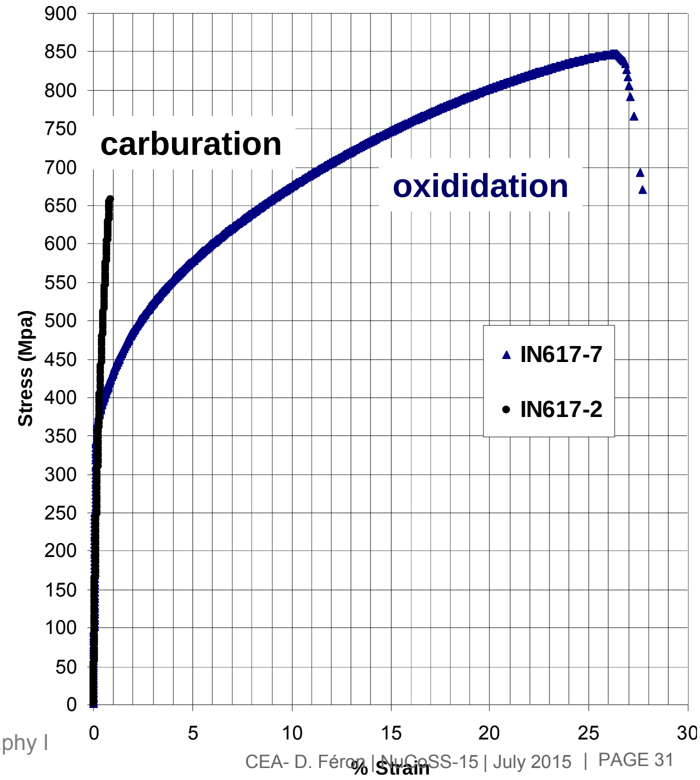


from bibliography VI

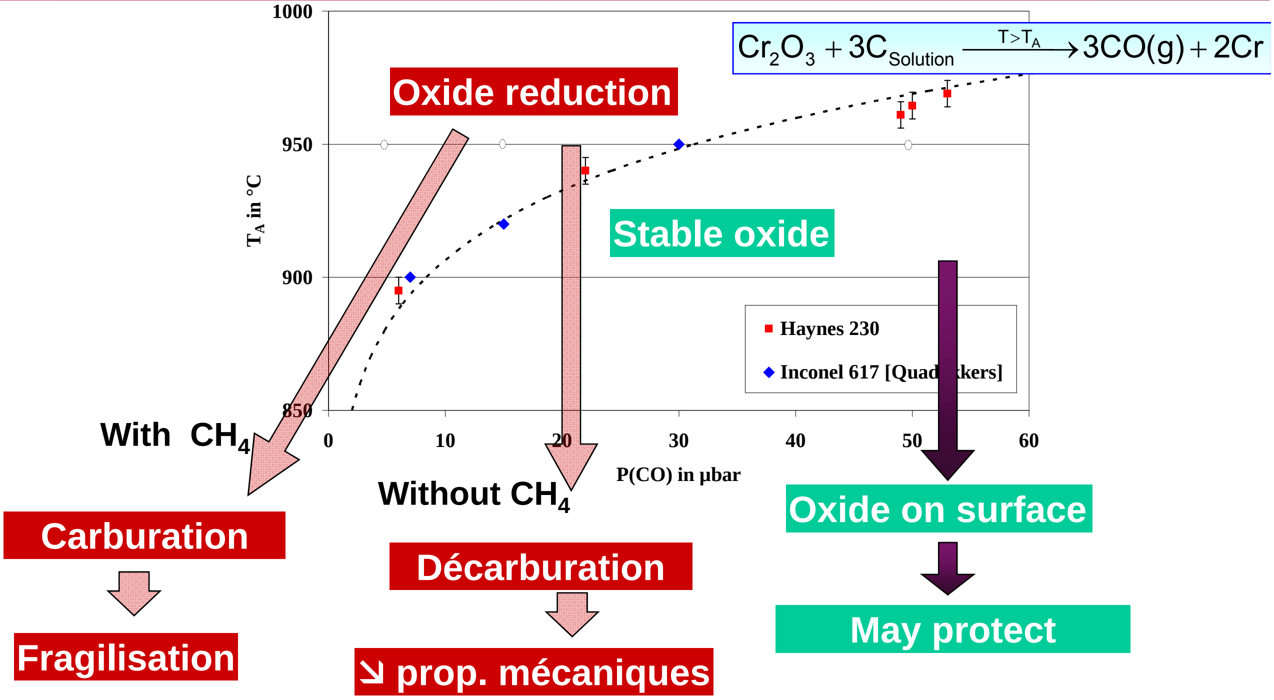
Effect of the carburation on mechanical properties

Carburized Alloy 617 specimens exhibit nil ductility at room temperature

Oxidized Alloy 617 exhibits 26% reduction in area at room temperature



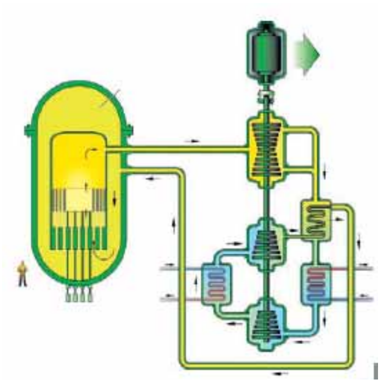
from bibliography I



from bibliography IV & VI

! Importance of the gas chemistry!
(control of impurity levels)

CORROSION IN HELIUM AT HIGH TEMPERATURES (OVER 800°C)



- IMPORTANCE OF THE IMPURITIES

-CHEMICAL CONTROL IS NEEDED

- IMPURITIES ARE NEEDED BUT AT THE RIGHT CONCENTRATIONS

- EVOLUTION OF THE CHEMISTRY WITH TEMPERATURE…..

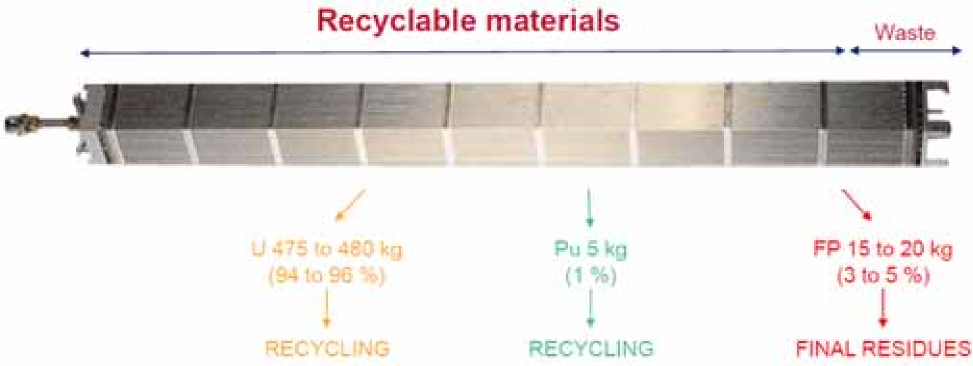
REPROCESSING PLANTS

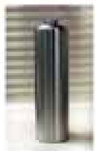
NITRIC ACID CORROSION

WHY REPROCESSING SPENT FUEL ?

Composition of the LWR spent fuel assembly (FA) after irradiation

➔ **1 LWR fuel assembly: 500 kg uranium before irradiation in the reactor**

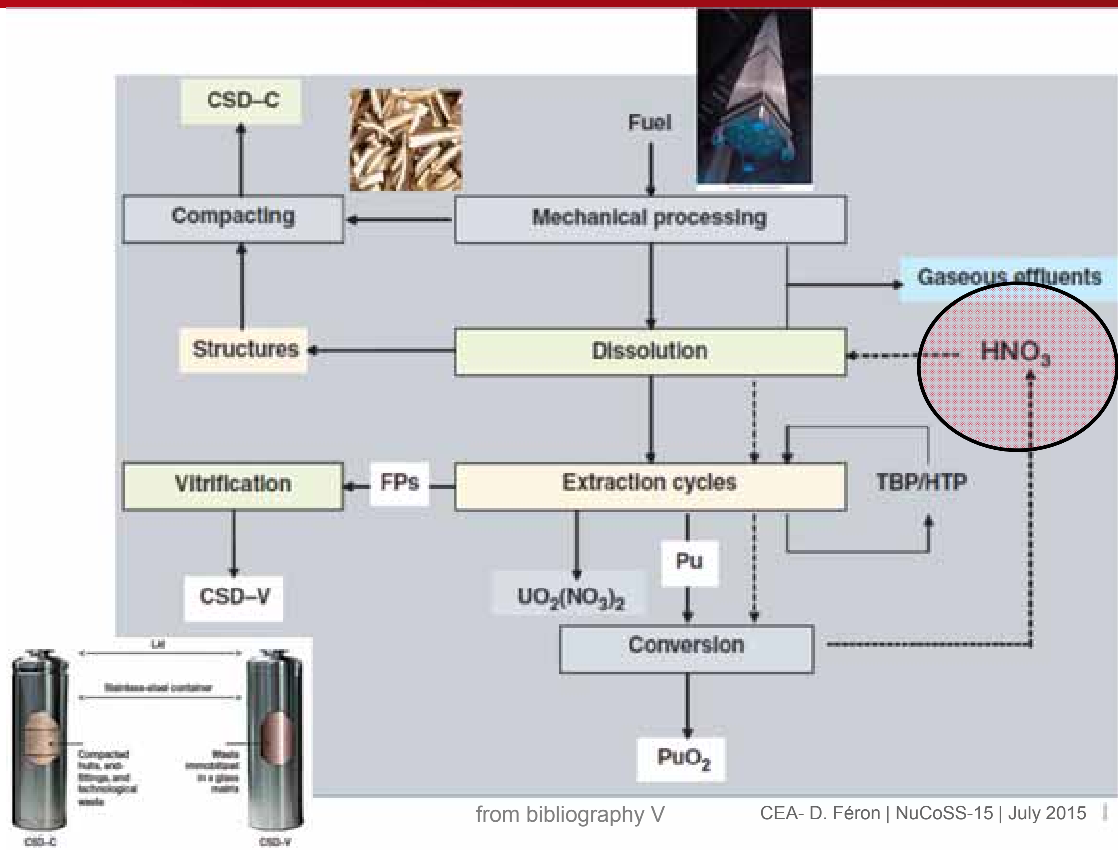




Recycling U & Pu

Very stable chemical form (glass) for hight radioactive elements

HOW SPENT FUEL REPROCESSING IS DONE?



from bibliography V

SPECIFICITIES OF NITRIC ACID MEDIA

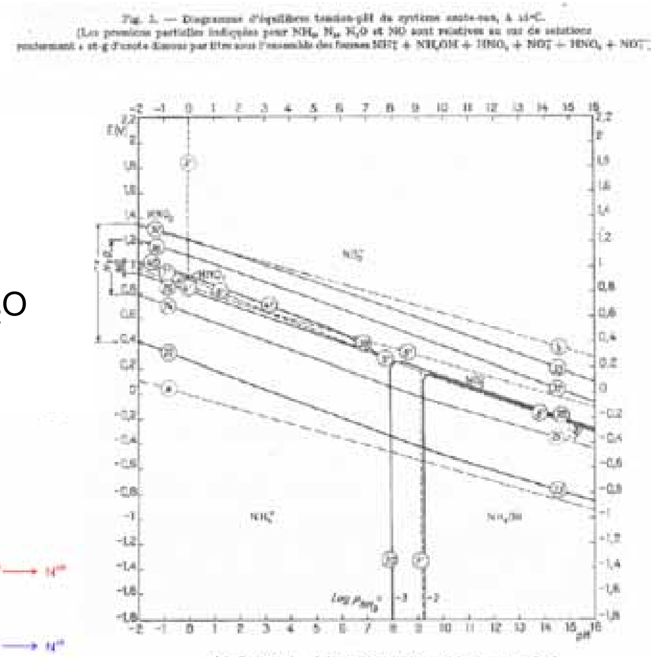
Nitric media

Acid

$HNO_3 \leftrightarrow NO_3^- + H^+$

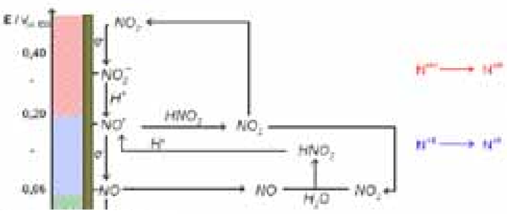
Oxidant

$HNO_3 + 2e^- + 2H^+ \leftrightarrow HNO_2 + H_2O$ (simplified)



M. Pourbaix, Atlas d'équilibres chimiques, 1963

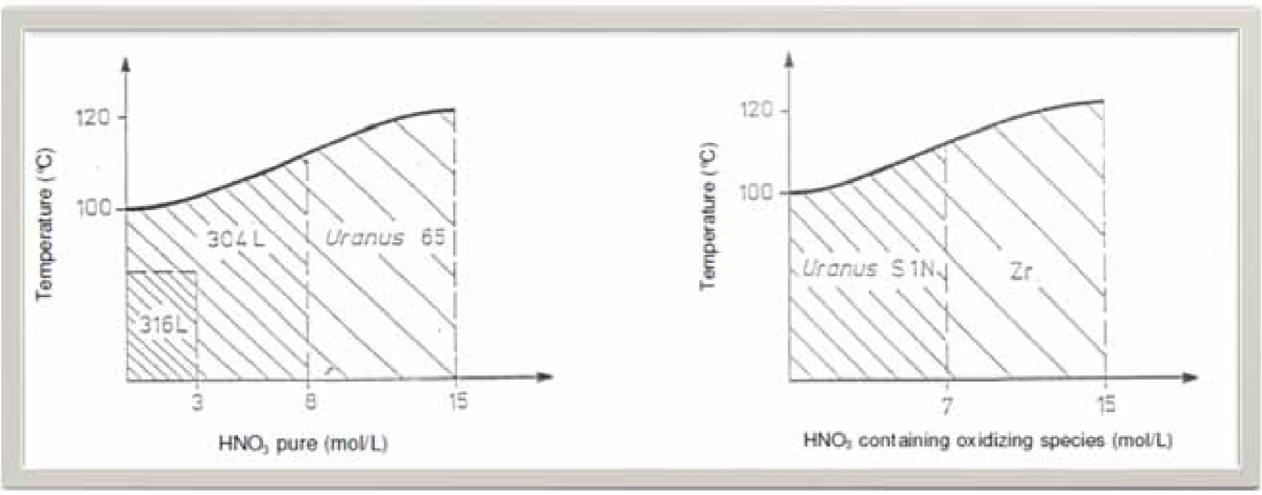
Cathodic reactions



autocatalysis

MATERIAL SELECTION IN REPROCESSING PLANTS

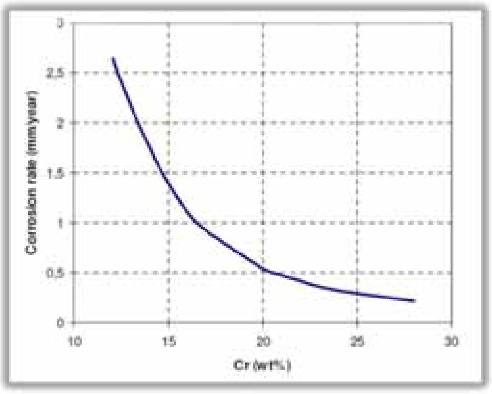
Typical limits of the use of austenitic stainless steels in nitric acid



P. Fauvet & al., JNL, 375, 2008, 52-64
P. Fauvet & al., EUROCORR 2006

AUSTENITIC STAINLESS STEELS USED IN SPENT FUEL REPROCESSING PLANTS

Grades AFNOR	Grades Commercial	C	Cr	Ni	Si	S	P	Mo	Mn	N	Nb
Z2 CN 18.10	AISI 304L	≤ 0.03	18	10	≤ 1.0	≤ 0.02	≤ 0.03	-	-	-	-
Z2 CN 25.20	URANUS 65 AISI 310	≤ 0.015	24-26	19-22	≤ 0.25	≤ 0.005	≤ 0.025	≤ 0.5	≤ 2	-	addition
Z2 CND 17.13	AISI 316L	≤ 0.03	17	13	≤ 1.0	≤ 0.02	≤ 0.03	2.5-3	-	-	-
Z1 CNS 17.15	URANUS S1N	≤ 0.015	16.5-18.5	13.5-15	3.8-4.5	≤ 0.005	≤ 0.025	≤ 0.5	≤ 2	≤ 0.035	addition

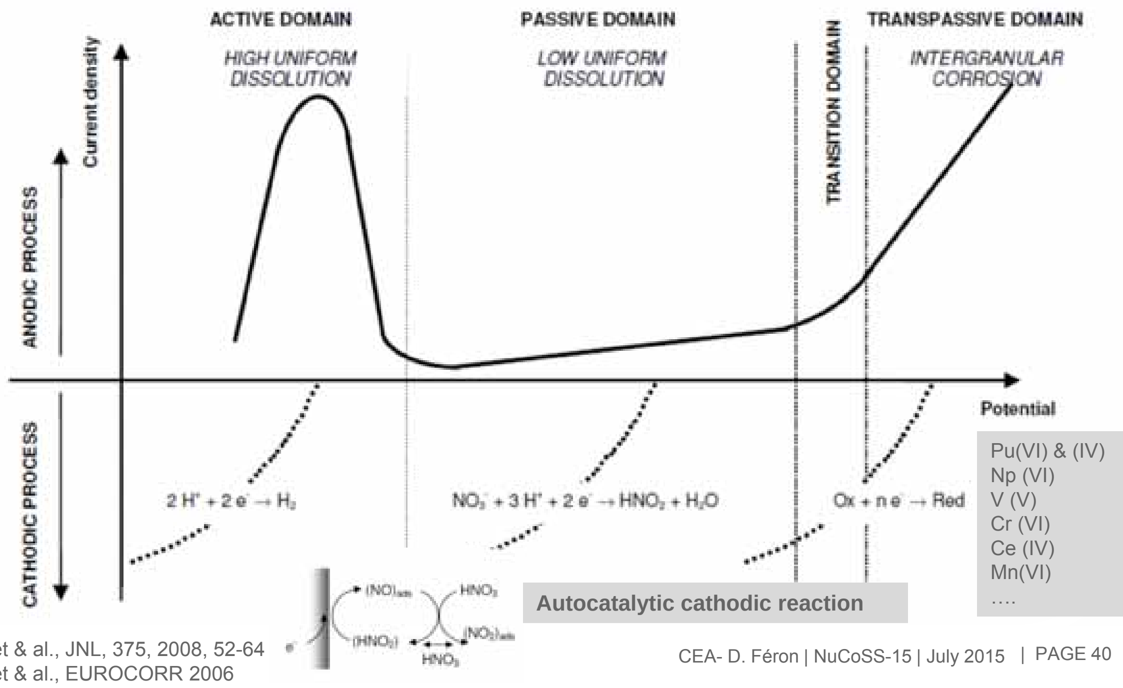


Corrosion rate as function of the chromium content of austenitic stainless steels in boiling 65% nitric acid

P. Fauvet & al., JNL, 375, 2008, 52-64
P. Fauvet & al;, EUROCORR 2006

STAINLESS STEELS IN REPROCESSING PLANTS

Electrochemical behavior of stainless steels in nitric acid



P. Fauvet & al., JNL, 375, 2008, 52-64
P. Fauvet & al., EUROCORR 2006

GALVANIC CORROSION IN REPROCESSING PLANTS

Metallic elements (from the cladding for instance) will be in contact with structural alloys in nitric acid

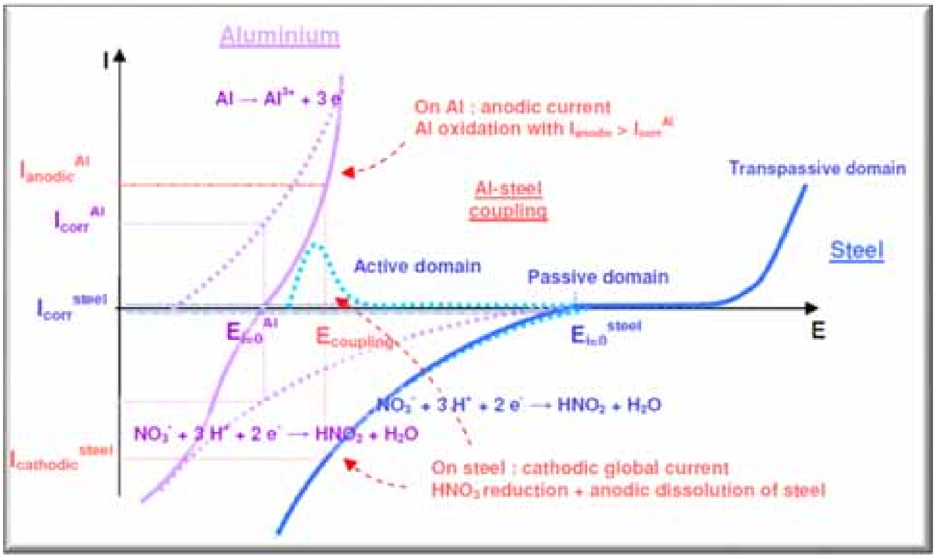
Metallic elements less noble than passive 304L & 316L	Metallic elements more noble than passive 304L & 316L
Al Zr Ta Ti	Zr (passive) Pt Au Graphite (Platinoïds: Pd, Rh, Ru)

- ✓ Cathodic reaction mainly on the more noble material
- ✓ Anodic reaction increases on the less noble material
- ➢ But it does not mean that the corrosion rate of SS decreases when SS is more noble in nitric acid …

P. Fauvet & al., JNL, 375, 2008, 52-64
P. Fauvet & al., EUROCORR 2006

GALVANIC CORROSION IN REPROCESSING PLANTS

Coupling between aluminium and stainless steels in nitric acid



Coupling ⇨ increase of the anodic reactions on Al and on SS, so **increase of the corrosion rates on both materials** (due to the SS active peak)

P. Fauvet & al., JNL, 375, 2008, 52-64
P. Fauvet & al., EUROCORR 2006

USE OF ZIRCONIUM ALLOY IN REPROCESSING PLANT

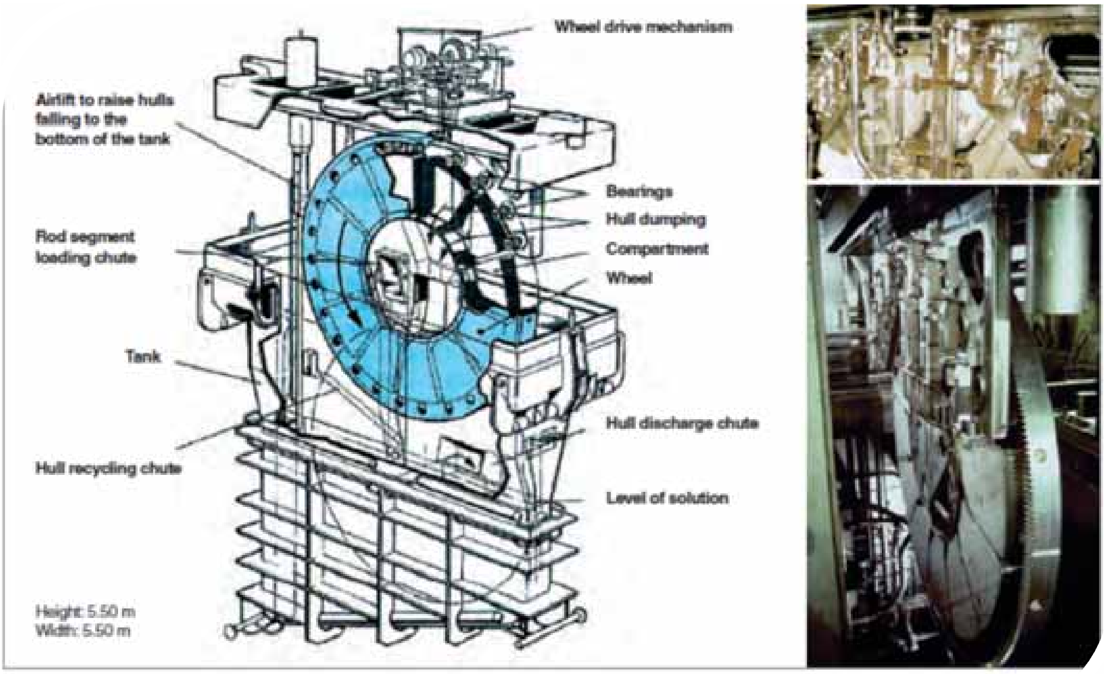


Fig. 35: Schematic, and photographs, of rotating-wheel dissolvers used in the UP3 and UP2-800 plants, at La Hague.

from bibliography V

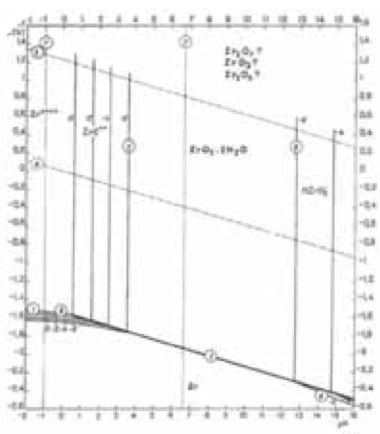
SOME FUNDAMENTALS ABOUT ZIRCONIUM CORROSION

Zirconium behavior

- Oxidation

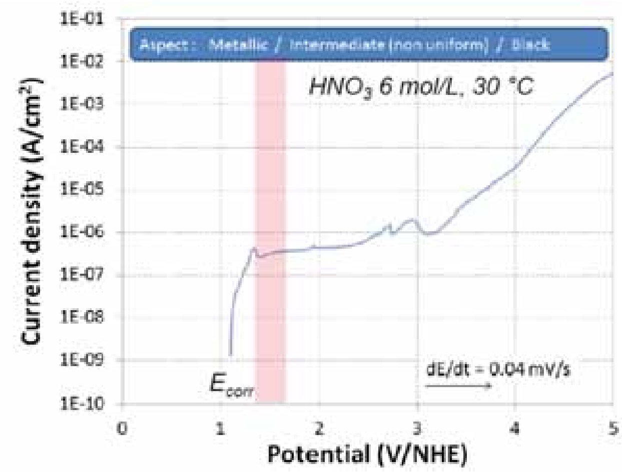
  $Zr \leftrightarrow Zr^{4+} + 4e^-$

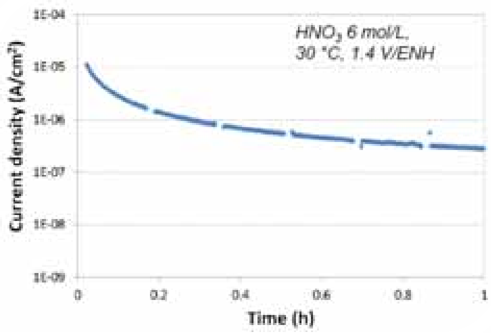
- In acid media, the oxide is not stable thermodynamically
- At low pH, domain of active corrosion following traditionnal use of Poubaix diagrams
- But, kinetics of formation and of dissolution of the oxide….



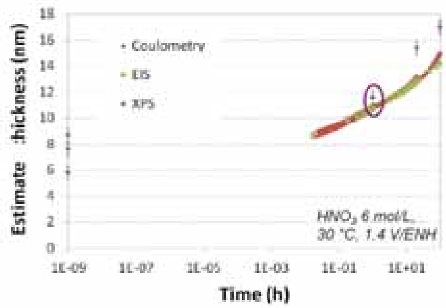
M. Pourbaix, Atlas d'équilibres chimiques, 1963

ZIRCONIUM ALLOY IN PURE NITRIC ACID



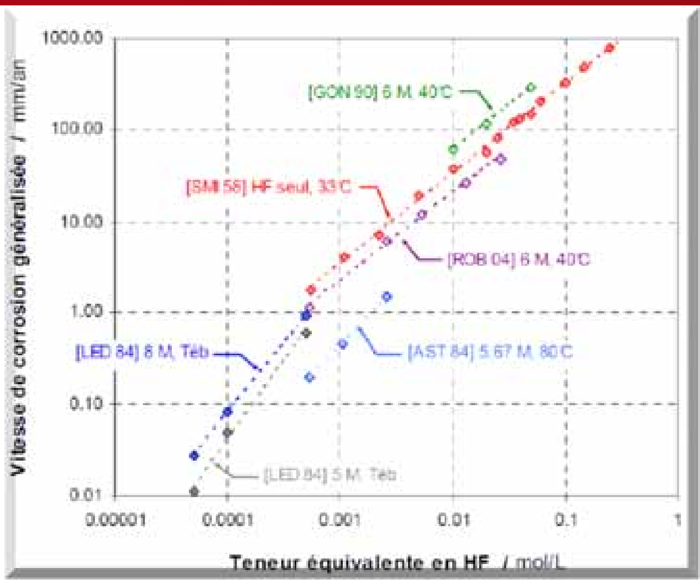


B. Gwinner & al., 64th ISE meeting, 2013
P. Lagoutharis & al., EUROCORR 2013



Passive behavior of Zr in pure nitric at very low pH

ZIRCONIUM BEHAVIOR IN NITRIC ACID WITH FLUORINE

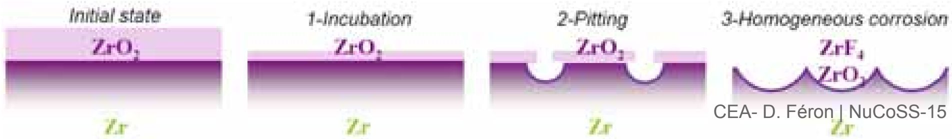


Increase of the corrosion rate with fluorine

formation of a soluble complex $(ZrF_n)^{(4-n)}$

B. Gwinner & al., 64th ISE meeting, 2013

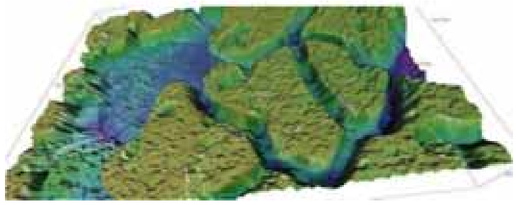
P. Lagoutharis & al., EUROCORR 2013



REPROCESSING PLANTS / NITRIC ACID CORROSION

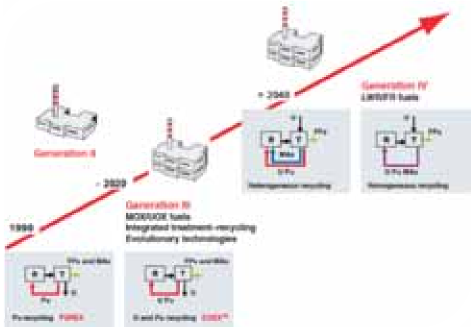
Nitric acid media

- Very different from water
- Autocatalytic HNO_3 reduction
- Oxidising species
- Galvanic coupling



Main alloys

- Austentitic stainless steels
  (304L, Uranus 65, 316L, Uranus S1N,…)
- Zirconium
- Titanium



UNDERGROUND REPOSITIRY

HOW TO PREDICT CORROSION FOR MILLENNIA ?

How to predict corrosion over millennia ?

Tests & Analyses

- *data for conception*
- *Identification of possible mechanisms*

Modeling & Simulation

- *Validation of mechanisms*
- *Long term extrapolation*

Analogues

- *Validation of time scale*
- *Mechanisms*

"Integrated" experiments

- *Coupling*
- *Interactions*
- *In-situ*

- **Communication**
- **Technology**
- **Science**

Illustration of the links between modelling and analogues

MODELLING & ARTEFACTS

Corrosion modeling of an iron based alloy in passive conditions

Arcorr experiments

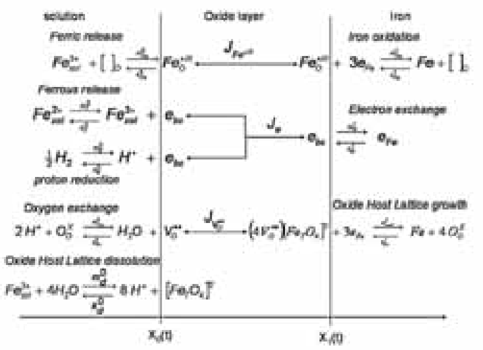


Fig. 3. Flow chart for electrons, octahedral cations and oxygen vacancies. Kröger–Vink notation is used. $[\,]_0$ corresponds to a free octahedral site.

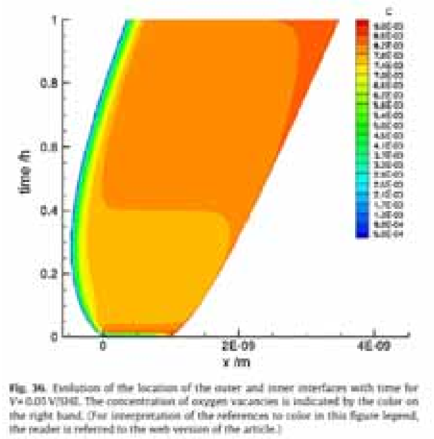
In situ experiments (Mont Terri and Bure underground laboratories)

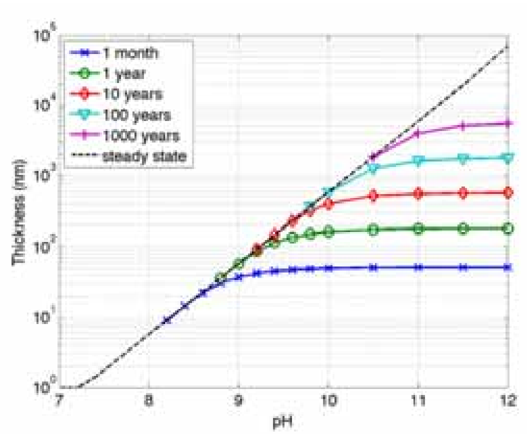
C. Bataillon & al. , Elect. Acta 55 (2010) 4451-4467

- **Corrosion modeling based on Fick and Poisson equations with moving interfaces (Diffusion Poisson Coupling Model - DPCM)**
- **Electrochemical experiments for data acquisition and verification**
- **Coupling with geochemical models**
- **Clay and concrete**

MODELLING & ARTEFACTS

Corrosion modeling of an iron based alloy in passive conditions
Example of obtained results



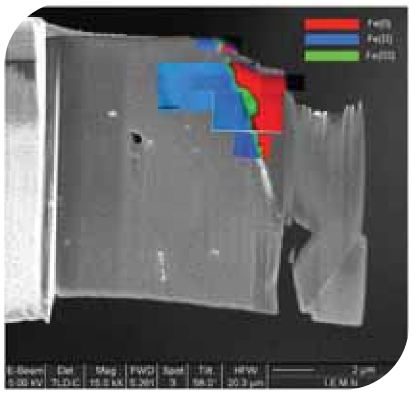


C. Bataillon & al. , Elect. Acta 55 (2010) 4451-4467

C. Bataillon & al. , J. Comput. Physics 231 (2012) 6213-6231

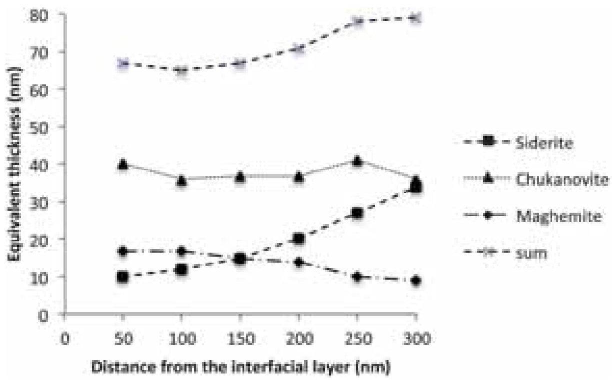
This model is under implantation in the nuclear waste simulations codes

MODELLING & ARTEFACTS



From A. Michelin & al., JAAS, 2012

Equivalent thicknesses of the different phases obtained from the fit of the extracted spectra.



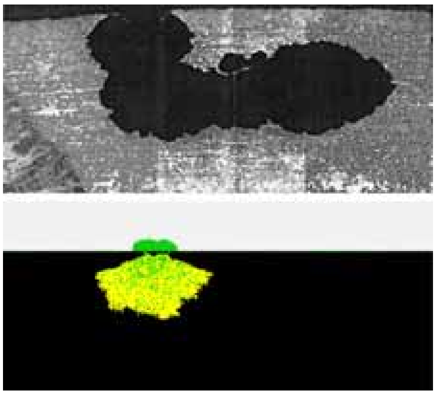
Support for modelling

- Corrosion products of a 450 year old archaeological iron nail in anoxic environment were investigated at the nanometer level using STXM
- **Interfacial layer of about 100 nm at the interface metal/oxide**
- Support the hypothesis of a nanolayer controlling the corrosion process (Point Defect Model and associated models)

MODELLING & ARTEFACTS

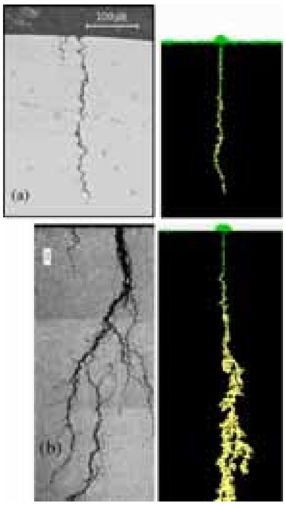
Use of cellular automata to model corrosion phenomena

Same description / difference: statistic of occurring events



Localised corrosion

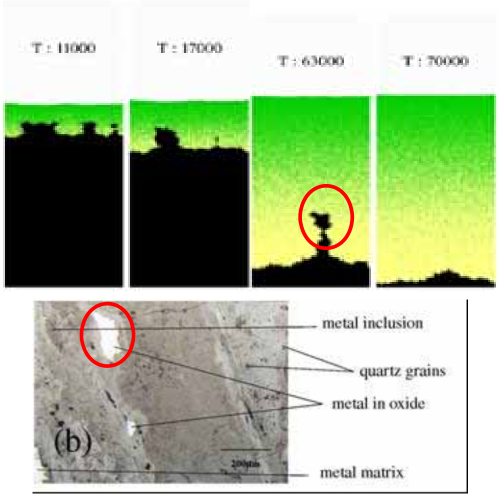
λ = 0.3 and ε = 0.005



Stress corrosion cracking

(a) λ = 0.99 and ε = 0.01,

(b) λ = 0.978 and ε = 0.001.

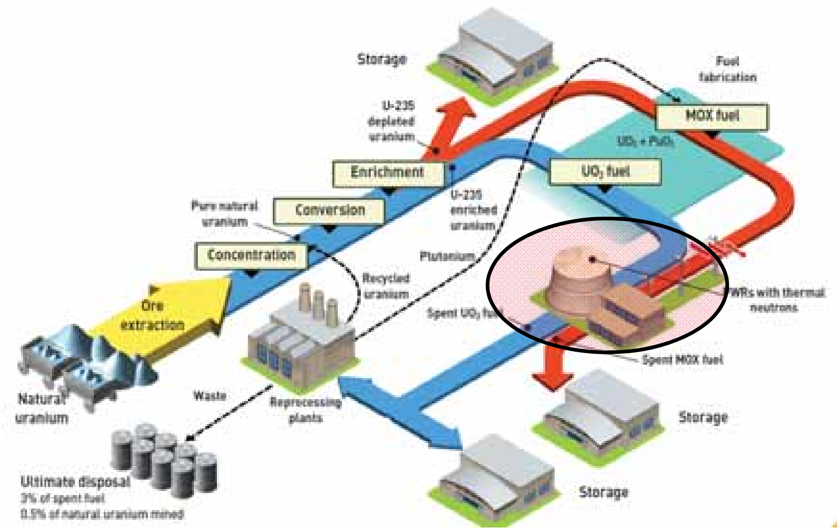


Evolution of the interface morphology (metal inclusions)

λ = 0.70 and ε = 0.30

D. di Caprio & al., Corrosion Science 53 (2011) 418–425

NUCLEAR CYCLE & CORROSION



Large number of environments, of materials, of sollicitations

Focus now on LWRs

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Have a nice nuclear corrosion summer school

NuCoSS - 15

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