Evaluation of aluminide coatings as hardfacing material candidate in sodium fast reactor

F. Rouillard, J.-L. Courouau, B. Duprey, S. Mathieu, Y. Bouizi, M. Vilasi, G. Boissonnet, F. Pedraza, I. Proriol-Serre, J.-B. Vogt

This work was funded by NEEDS project (EDF-AREVA-CEA-CNRS)

EVALUATION OF ALUMINIDE COATINGS AS HARDFACING MATERIAL CANDIDATE IN SODIUM FAST REACTOR

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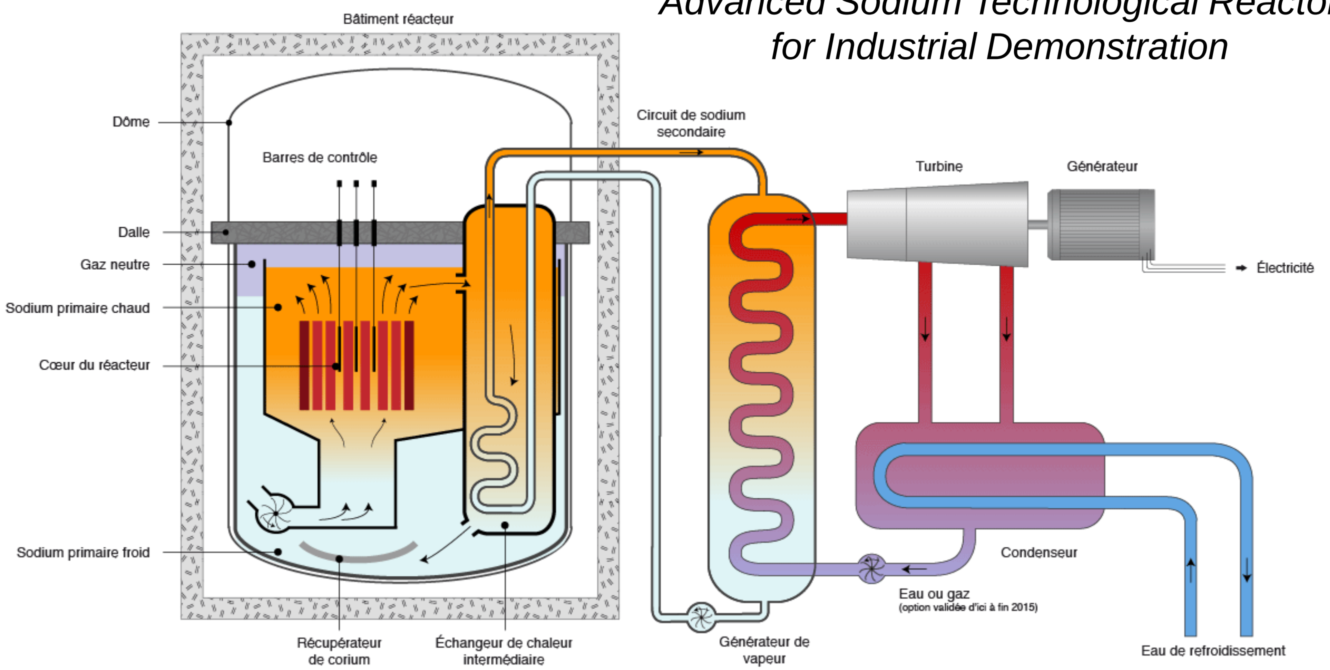
2- Jean Lamour Institute, University of Lorraine, Nancy, France
3- LASIE - University of La Rochelle, France
4- UMET - University of Lille, France

CONTEXT : NEW HARDFACING ALLOYS FOR TRIBOLOGICAL APPLICATIONS OF SFR

Sodium-cooled Fast Reactor ASTRID (600 MWe)

Advanced Sodium Technological Reactor for Industrial Demonstration

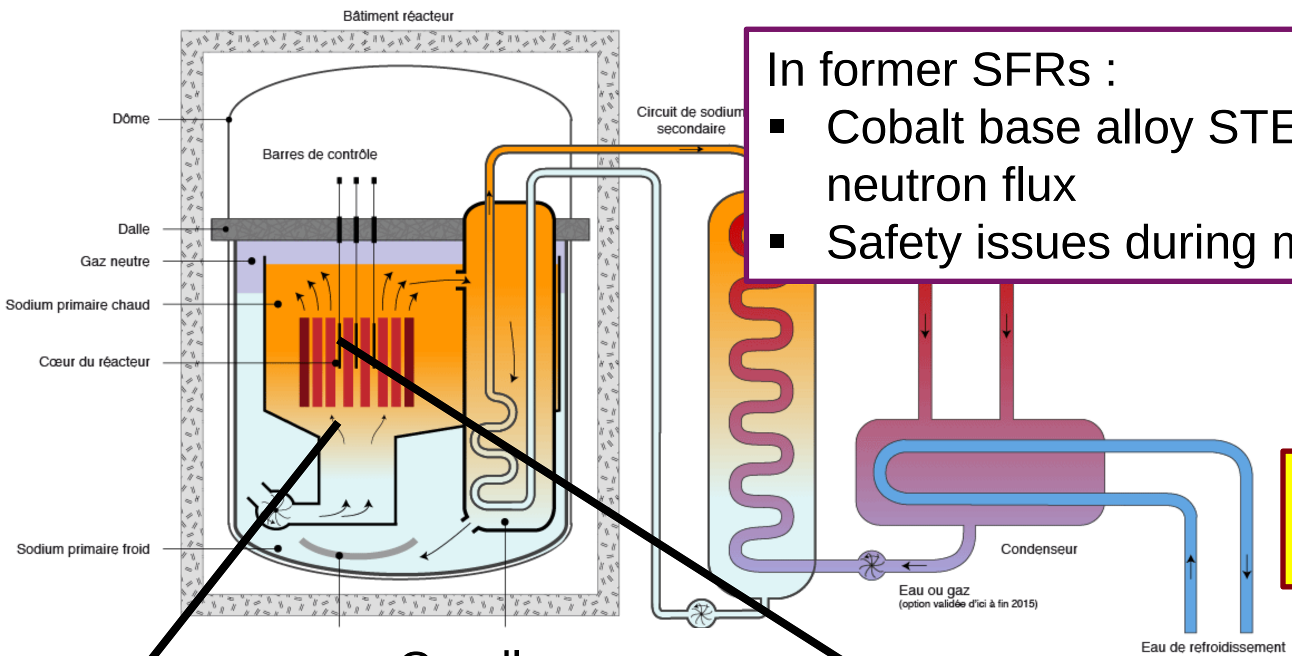


Main goals of ASTRID:
- Multi-recycling of plutonium (uranium ressource preservation)
- Minor actinide transmutation (nuclear waste reduction)
- Enhanced safety (as for Gen III reactors)

Many components need to move during :
- nominal operation
- or during maintenance operation

CONTEXT : NEW HARDFACING ALLOYS FOR TRIBOLOGICAL APPLICATIONS OF SFR

Sodium-cooled Fast Reactor ASTRID (600 MWe)



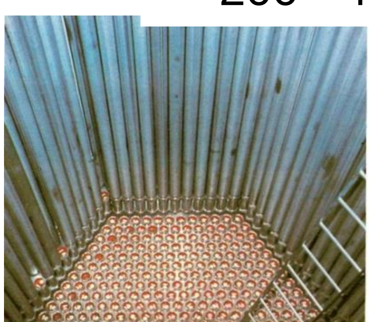
In former SFRs :

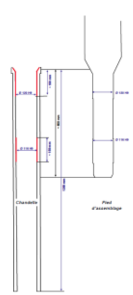
- Cobalt base alloy STELLITE but activation under neutron flux
- Safety issues during maintenance operation

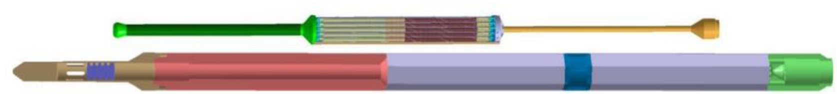
Research program on coating alternatives

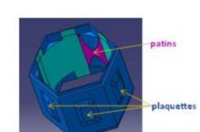
« Candles » :
200 - 400°C, 60 years

Core components : 400-500°C, 4 years









FROM RESEARCH TO INDUSTRY

EXPECTED PROPERTIES OF TRIBOLOGICAL MATERIAL

- ❑ Good friction coefficient
- ❑ Good wear resistance
- ❑ Good corrosion resistance in Na
- ❑ Good behaviour under irradiation
- ❑ Good mechanical behaviour : resistance to thermal and mechanical shocks
- ❑ Good adhesion

Fabrication process = robust and vendor = existing or expected in near future

CEA-AREVA-EDF program :

- ❑ Thick coatings (2 mm) : Nickel base alloys deposited by PTA or laser
- ❑ **Thin coatings (<100 µm) : Aluminides**

- Good compatibility in Na from results obtained during the « golden age » of SFRs in the 70's and 80's.
- Good tribological properties : good friction coefficient associated to the formation of stable surface $NaAlO_2$ or $Na_2Al_2O_4$ in Na

JOINT WORK IN COLLABORATION BETWEEN 4 LABS

Aluminides coatings synthesis by *pack-cementation* (IJL) and *slurry* (University of La Rochelle)

- Na compatiblity
- Tribological properties in Na

- Behaviour under irradiation (experiments conducted in Russian BOR60 reactor)

Mechanical testing on coated sample :
« Small Punch Test » in Na

JOINT WORK IN COLLABORATION BETWEEN 4 LABS

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3

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1) ALUMINIDE COATINGS SYNTHESIS

FROM RESEARCH TO INDUSTRY

STEEL GRADES AND COATING

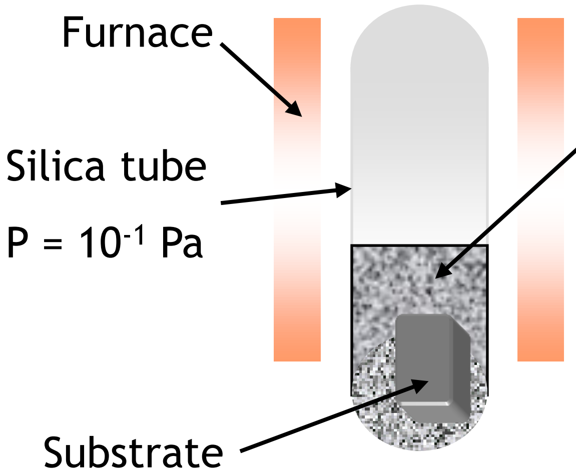
%weight	Fe	Cr	Ni	C	Mo	N	Aluminide coating process
316L(N)	Bal	17	13.2	0.04	2.3	0.06	Pack cementation
T91	Bal	9	0.2	0.1	1	/	Pack cementation + slurry

Both coating processes were realized without or with pre Ni plating treatment

Fe rich aluminides

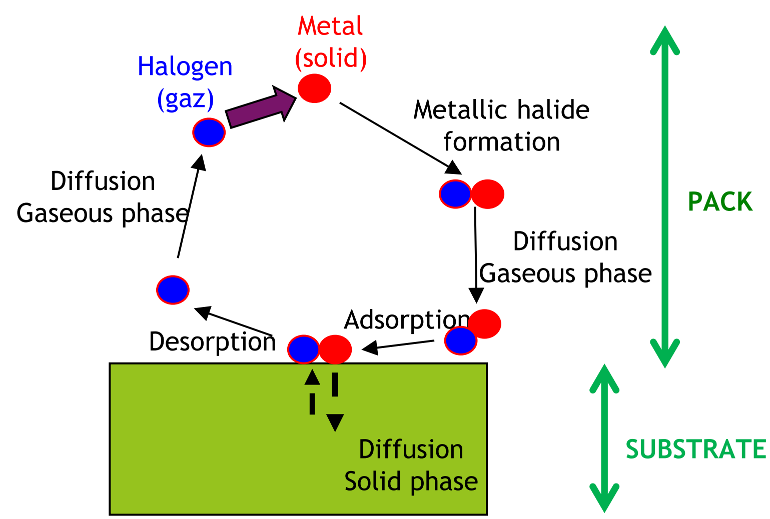
Ni rich aluminides

PACK-CEMENTATION – LOW AL ACTIVITY (PC)



"pack" composition :

- halide activator – $CrCl_3$ – 1 wt%
- inert powder (Al_2O_3) – 59 wt%
- Master alloy (Ni_2Al_3) – 40 wt%

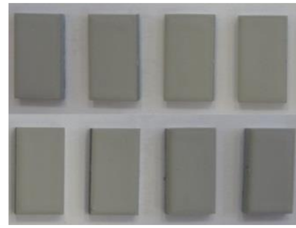


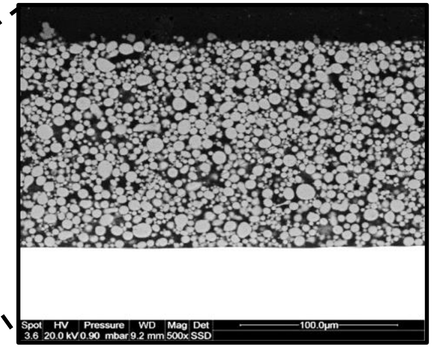
980°C – 4h in air
(muffle furnace)

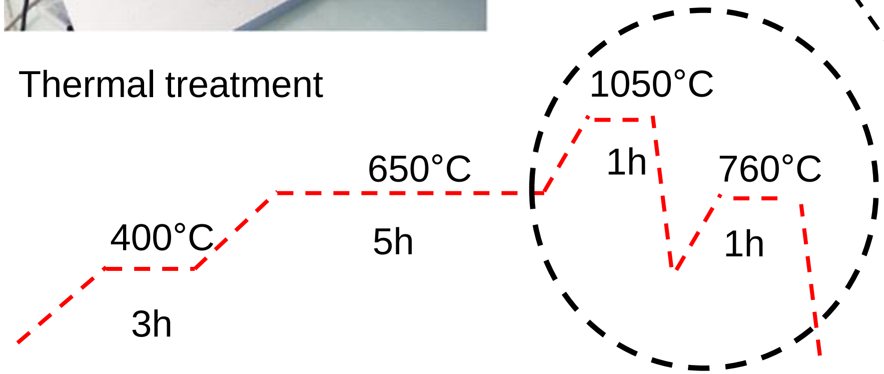
When pre nickel plated : 18 µm then vacuum annealed at 400°C before PC

FROM RESEARCH TO INDUSTRY cea den

SLURRY – HIGH AL ACTIVITY (SL)

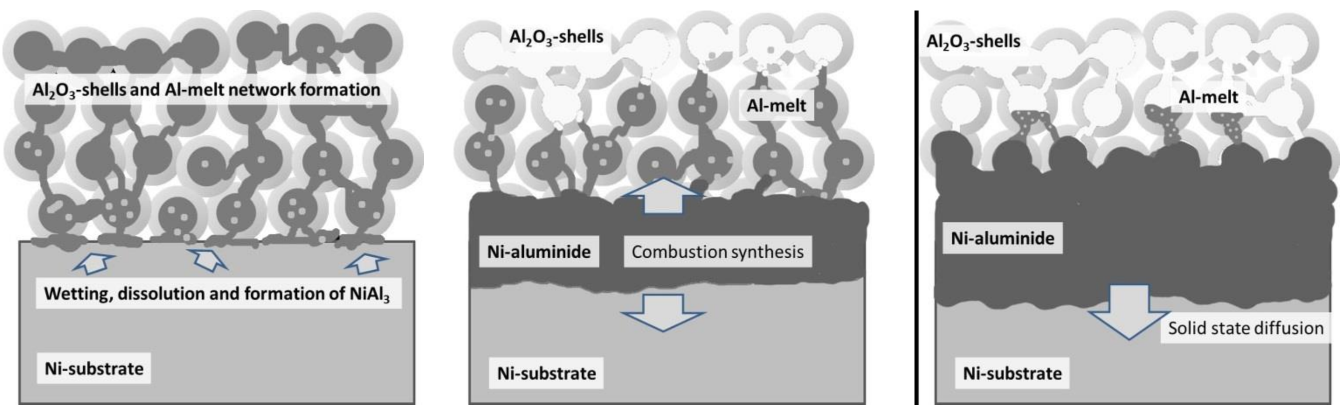






Final annealing step to recover the microstructure and the mechanical properties of T91 substrate

Wetting > Combustion synthesis > interdiffusion

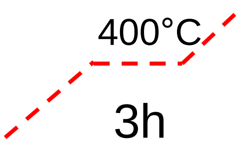


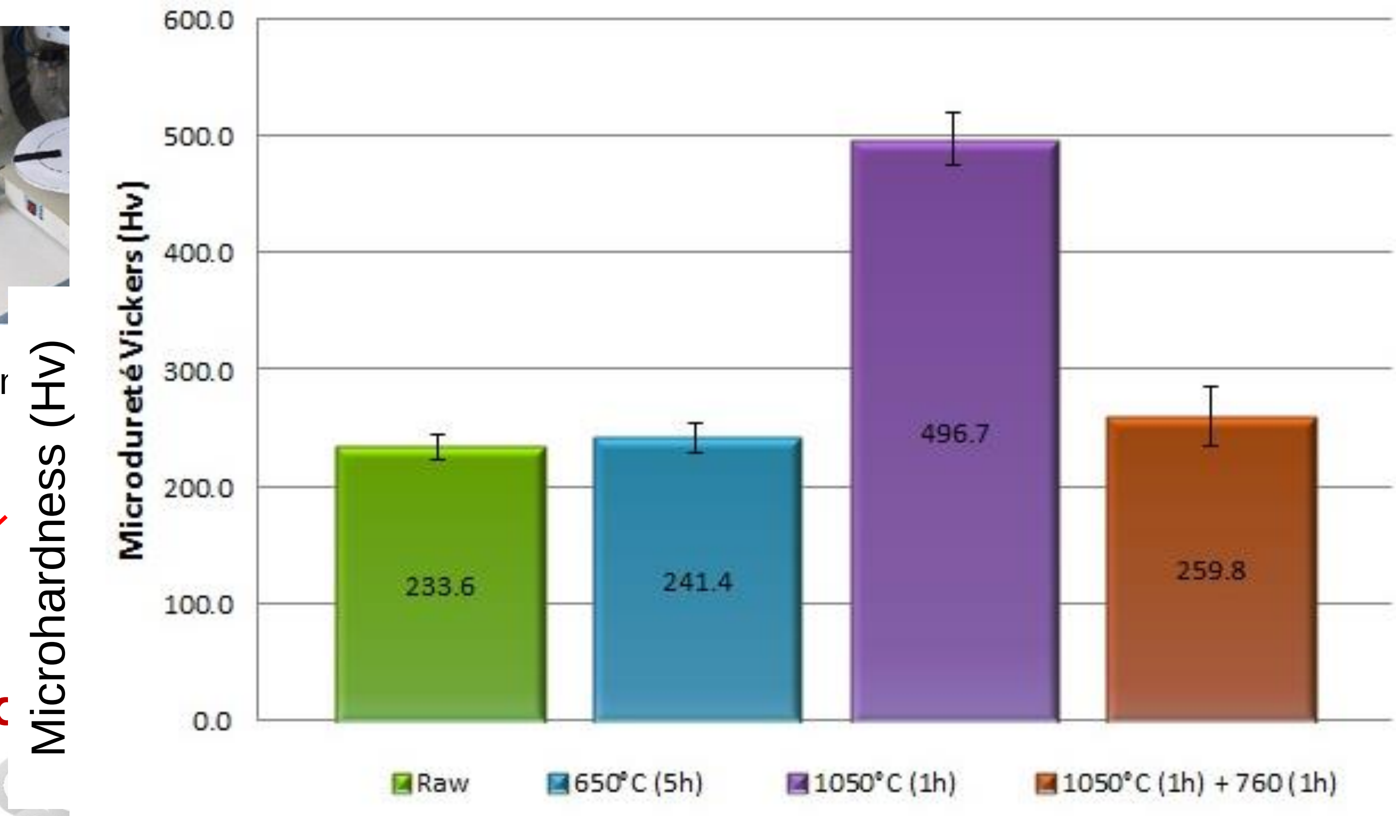
Ni & Fe substrates

* *M. Galetz et al. Intermetallics (2014)*

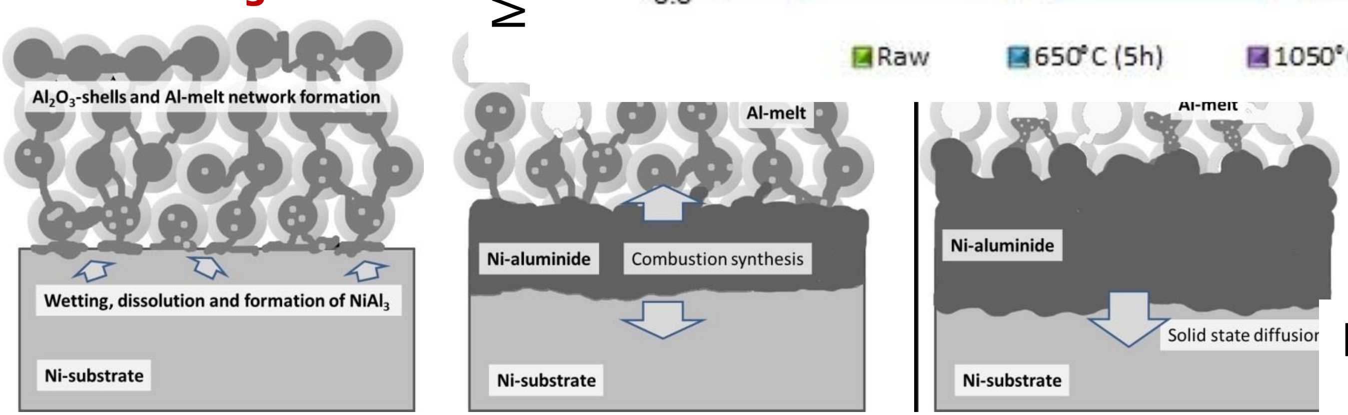
SLURRY – HIGH AL ACTIVITY (SL)

Thermal treatr





Wetting > C



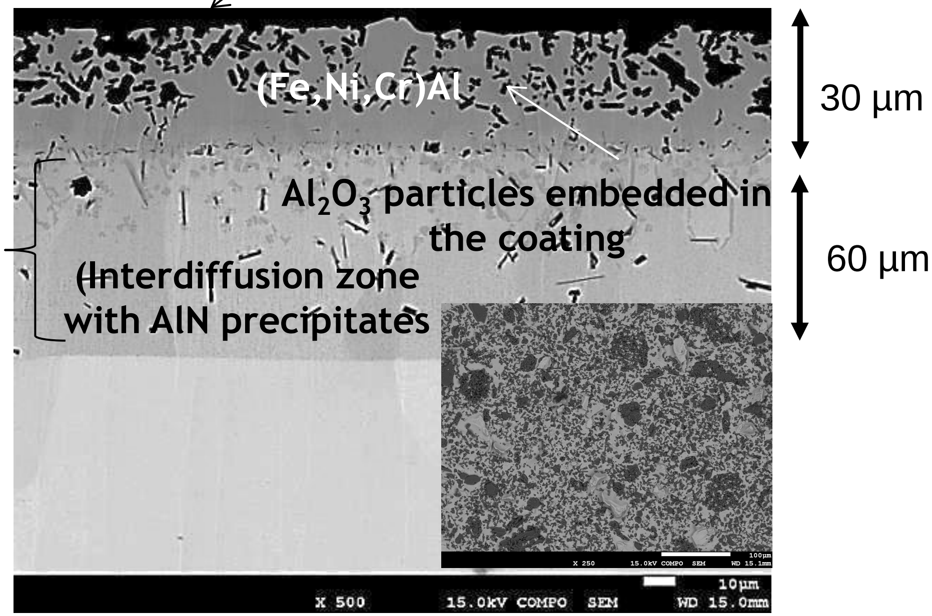
Ni & Fe substrates

Last step : grit-blasting of the top coat to remove the residue (bisque)

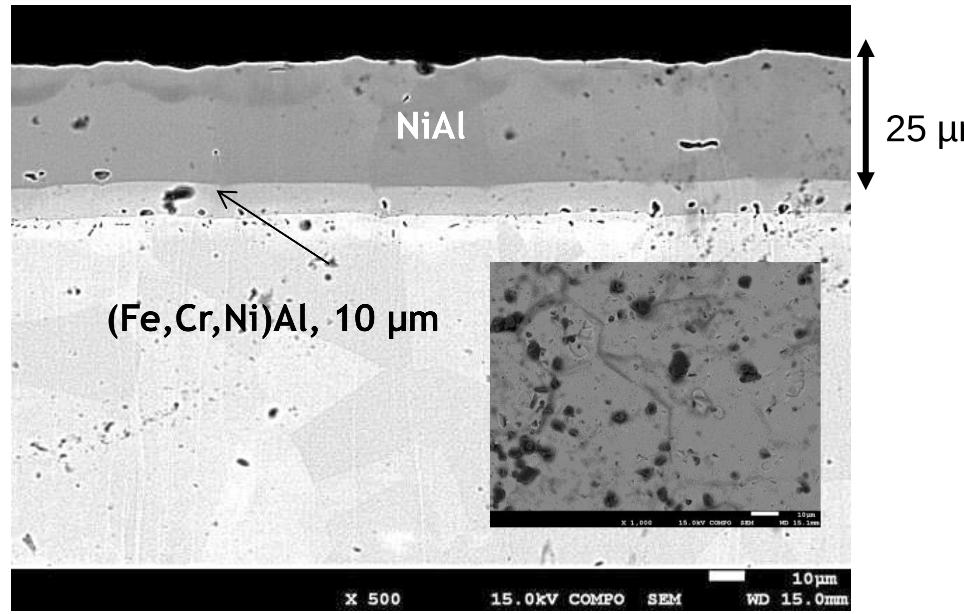
COATING OBTAINED BY PC ON 316L(N)

Without pre Ni plating

Al_2O_3 and Ni_2Al_3 particles stuck on the surface



With pre Ni plating

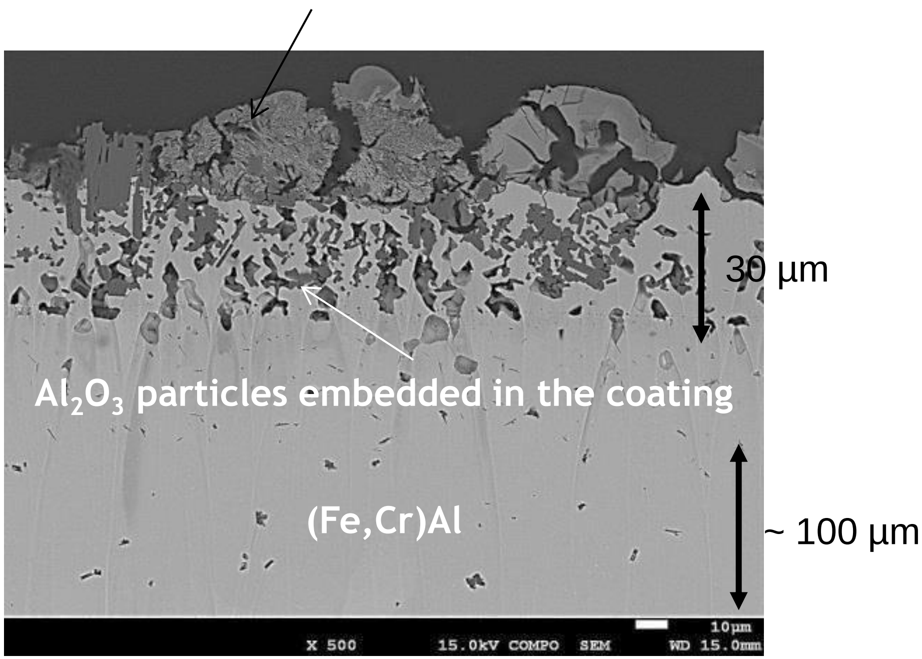


- **Composite coating** : B2-(Fe, Ni, Cr) Al with « hard » embedded Al_2O_3 particles
- Outwards aluminide growth

- NiAl
- Less particles stuck on the surface
- Lower outwards aluminide growth rate
- Thin interdiffusion zone

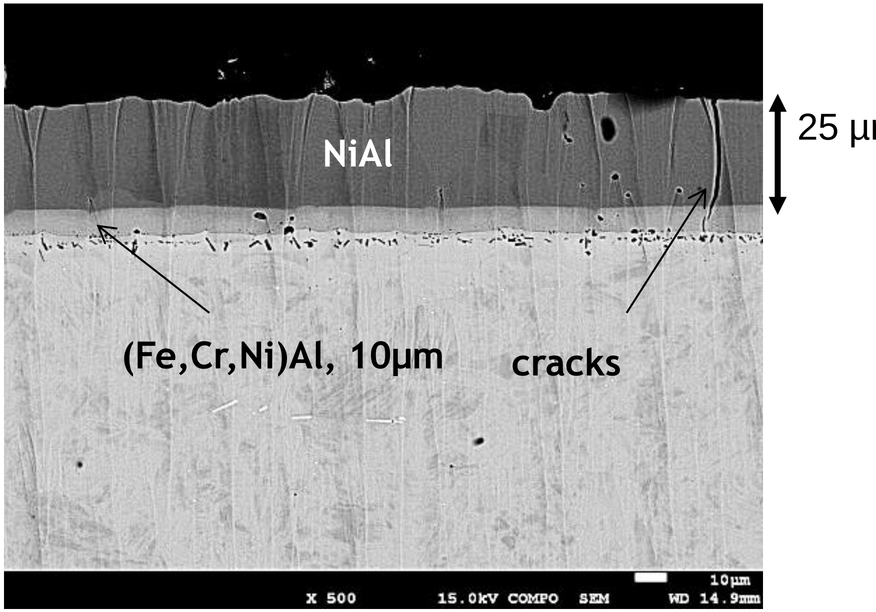
COATING OBTAINED BY PC ON T91

Without pre Ni plating



- **Composite coating** : B2-(Fe, Cr) Al with « hard » embedded Al_2O_3 particles
- Outwards aluminide growth
- Very large interdiffusion zone

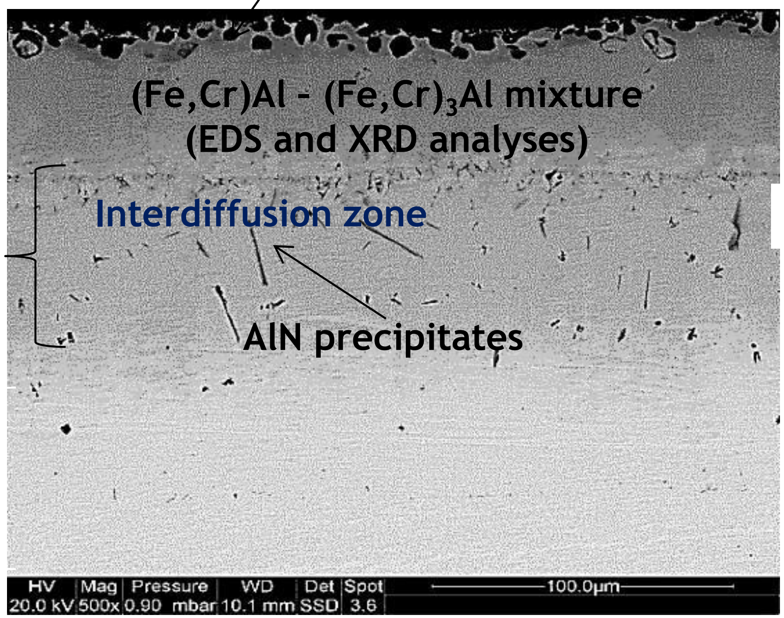
With pre Ni plating



- NiAl
- Less particle stuck on the surface
- Thin interdiffusion zone
- **Cracks = due to higher thermal expansion difference between T91 and NiAl ?**

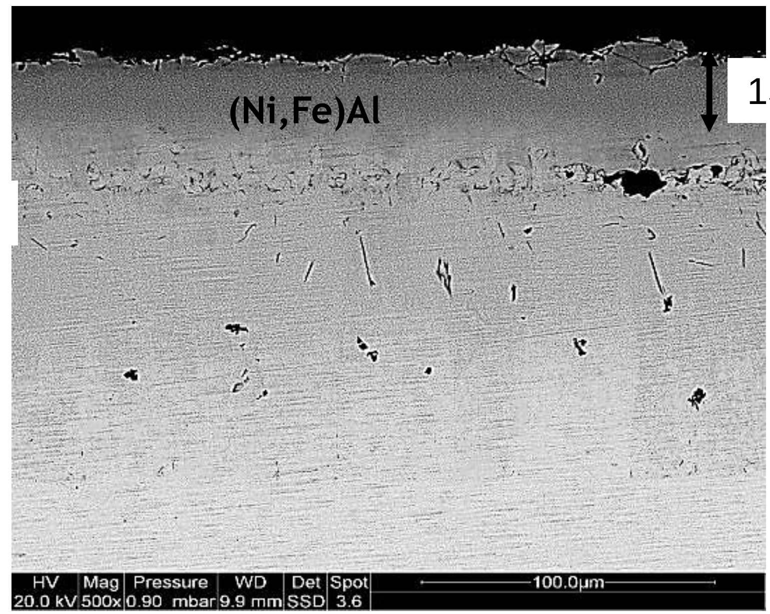
COATING OBTAINED BY SL ON T91

Without pre Ni plating



120 µm

With pre Ni plating



- B2-(Fe, Cr) Al with porosity at the surface
- Inwards aluminide growth
- Surface porosity

- NiAl
- Same thickness (preferential inwards Al diffusion)

2) COMPATIBILITY OF THE COATINGS IN NA

MAIN CORROSION PROCESSES IN HT NA

3 main corrosion processes

Dissolution
(governed by solubility)

MAIN CORROSION PROCESSES IN HT NA

3 main corrosion processes

Oxidation
(governed by oxygen content in Na)

Dissolution
(governed by solubility)

MAIN CORROSION PROCESSES IN HT NA

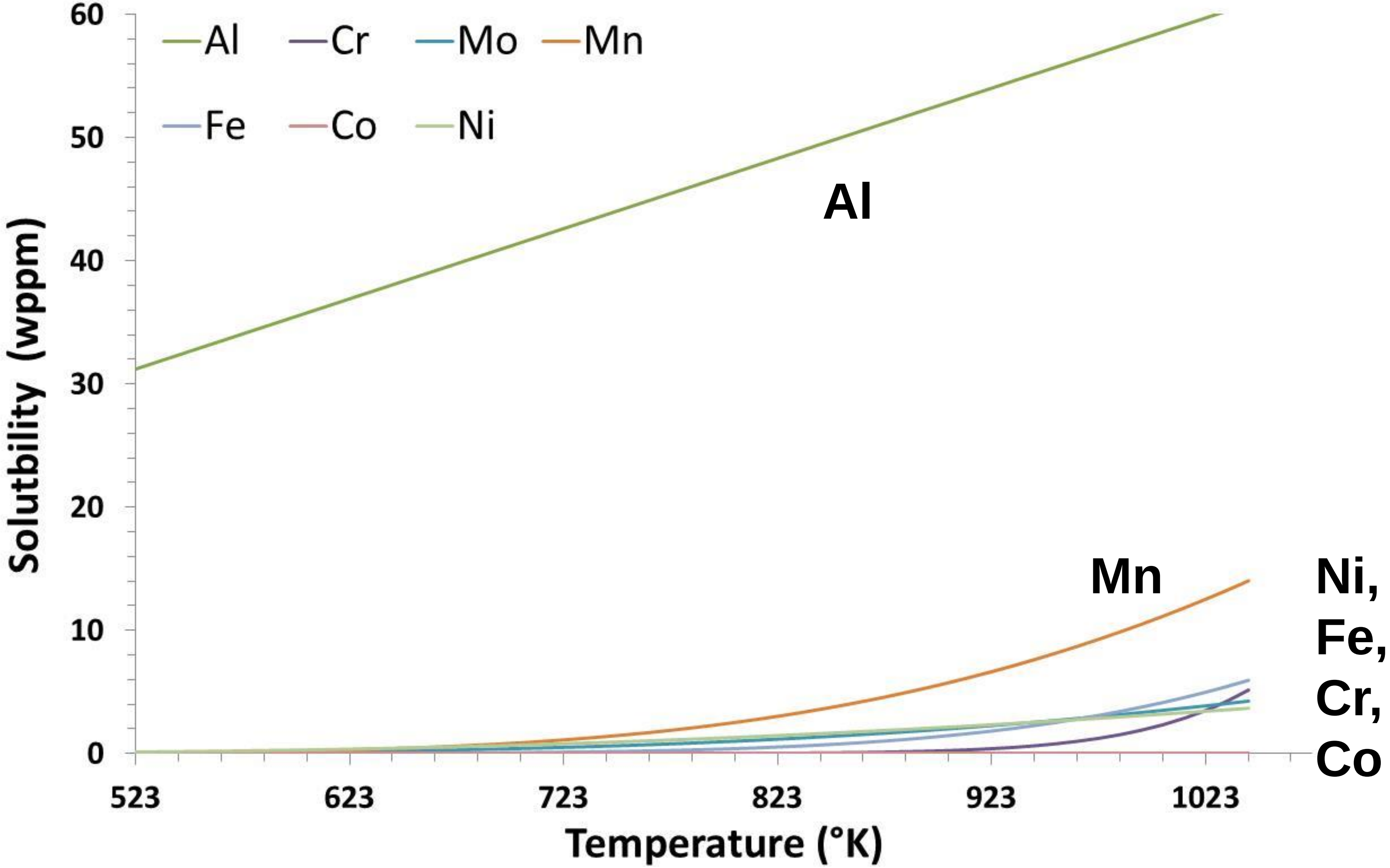
3 main corrosion processes

De- or carburisation
(governed by carbon content in Na)

Oxidation
(governed by oxygen content in Na)

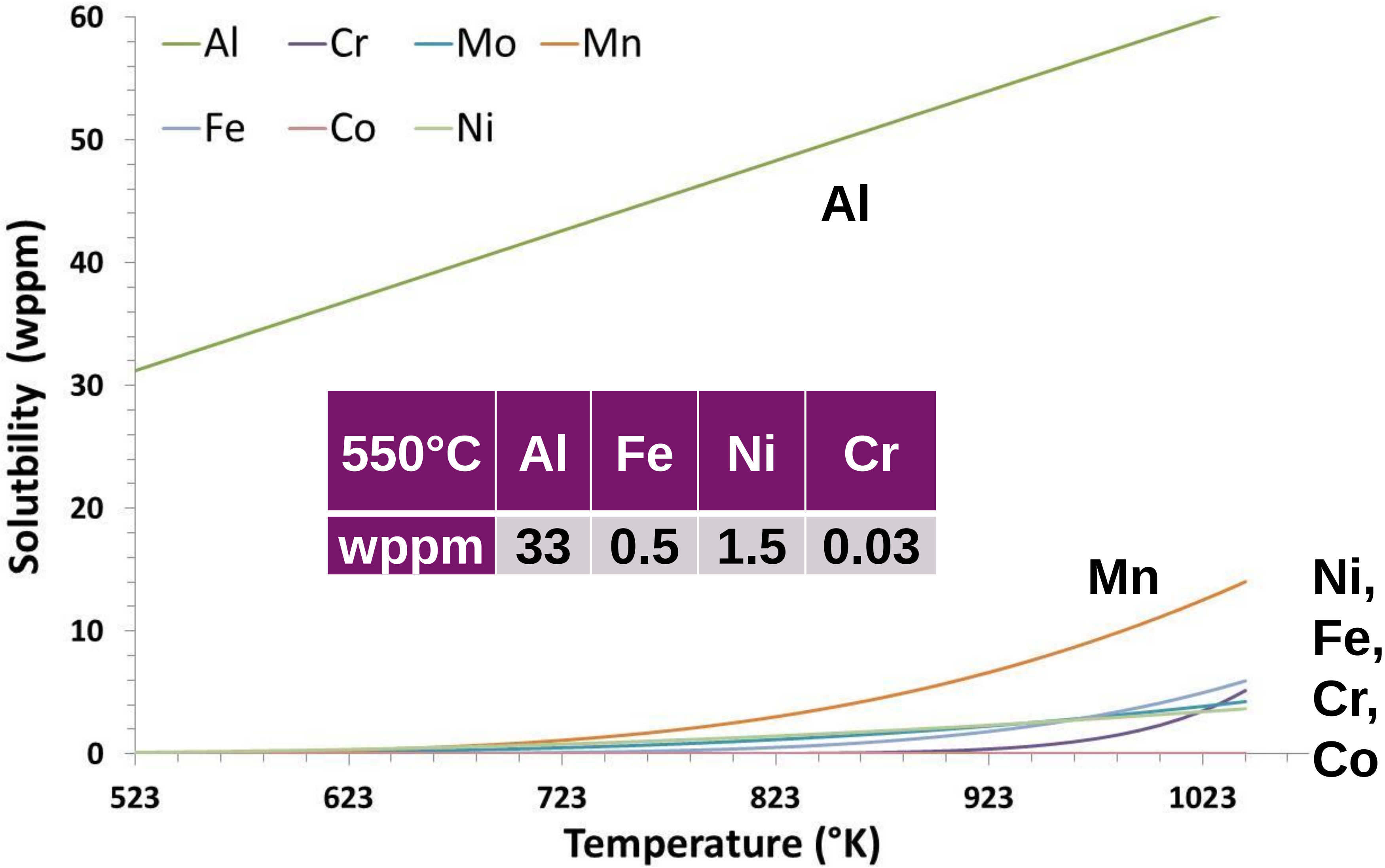
Dissolution
(governed by solubility)

DISSOLUTION IN NA



[Applied chemistry of the alkali metals, Borgstedt, 1987]

DISSOLUTION IN NA

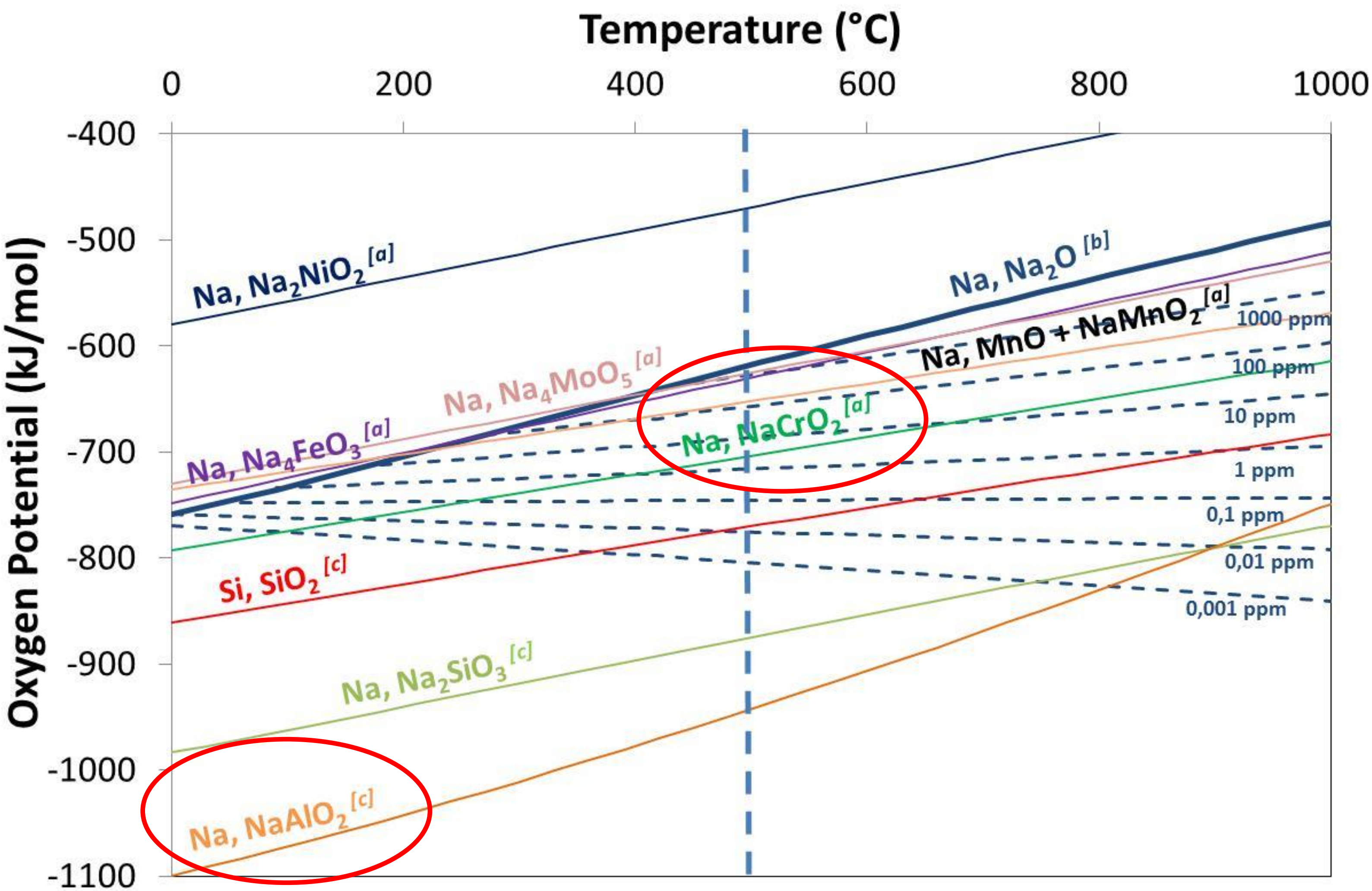


550°C	Al	Fe	Ni	Cr
wppm	33	0.5	1.5	0.03

[Applied chemistry of the alkali metals, Borgstedt, 1987]

OXIDATION IN NA

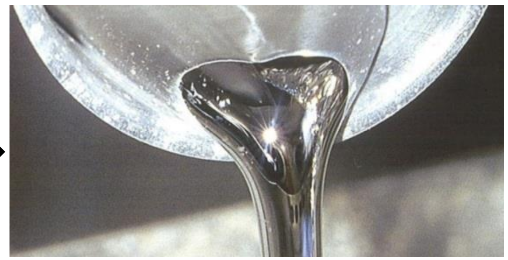
- All « usual » oxides are not stable in Na
- Formation of ternary oxides Na-M-O : Na_2O + oxide = Na-M-O

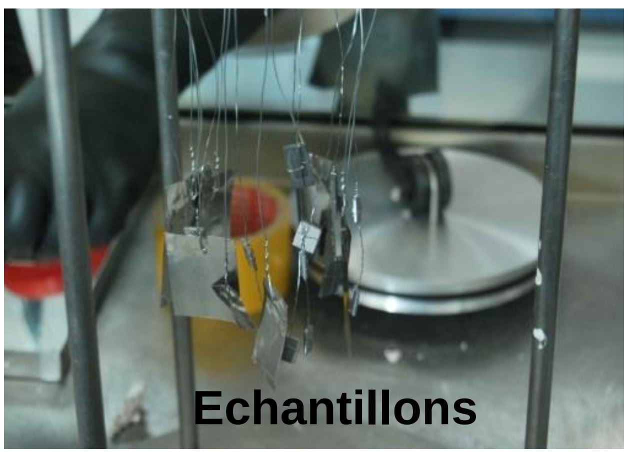


CORROSION TEST IN HT SODIUM

2.5 L static Na in Mo crucible
Temperature : **550 °C**
Exposure times : 850 h and 4250 h

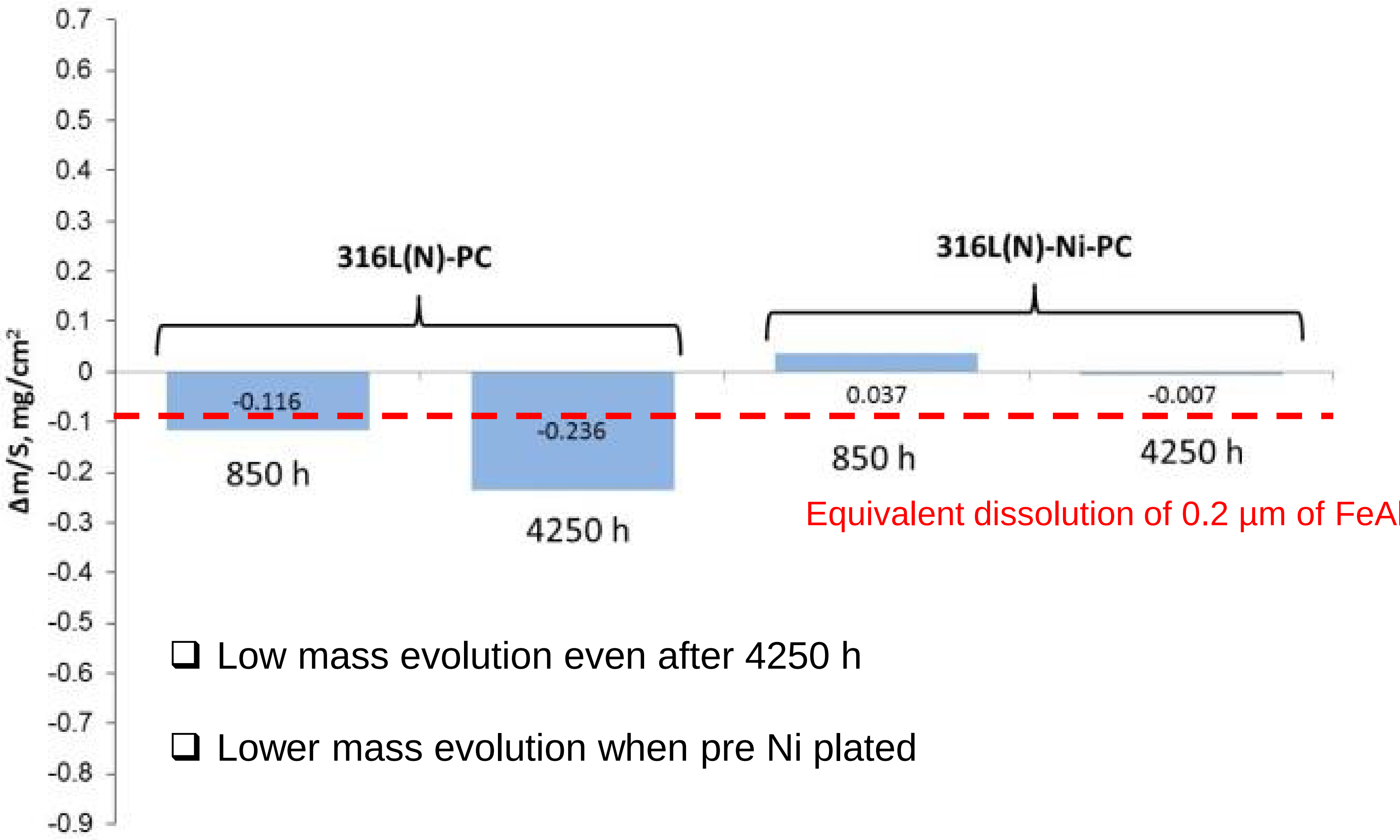
- Oxygen purified Na by Zr foil getter (600°C) before (3 days) and during sample exposure : **[O] < 5 ppm**
- Cr sample to detect any Al dissolution





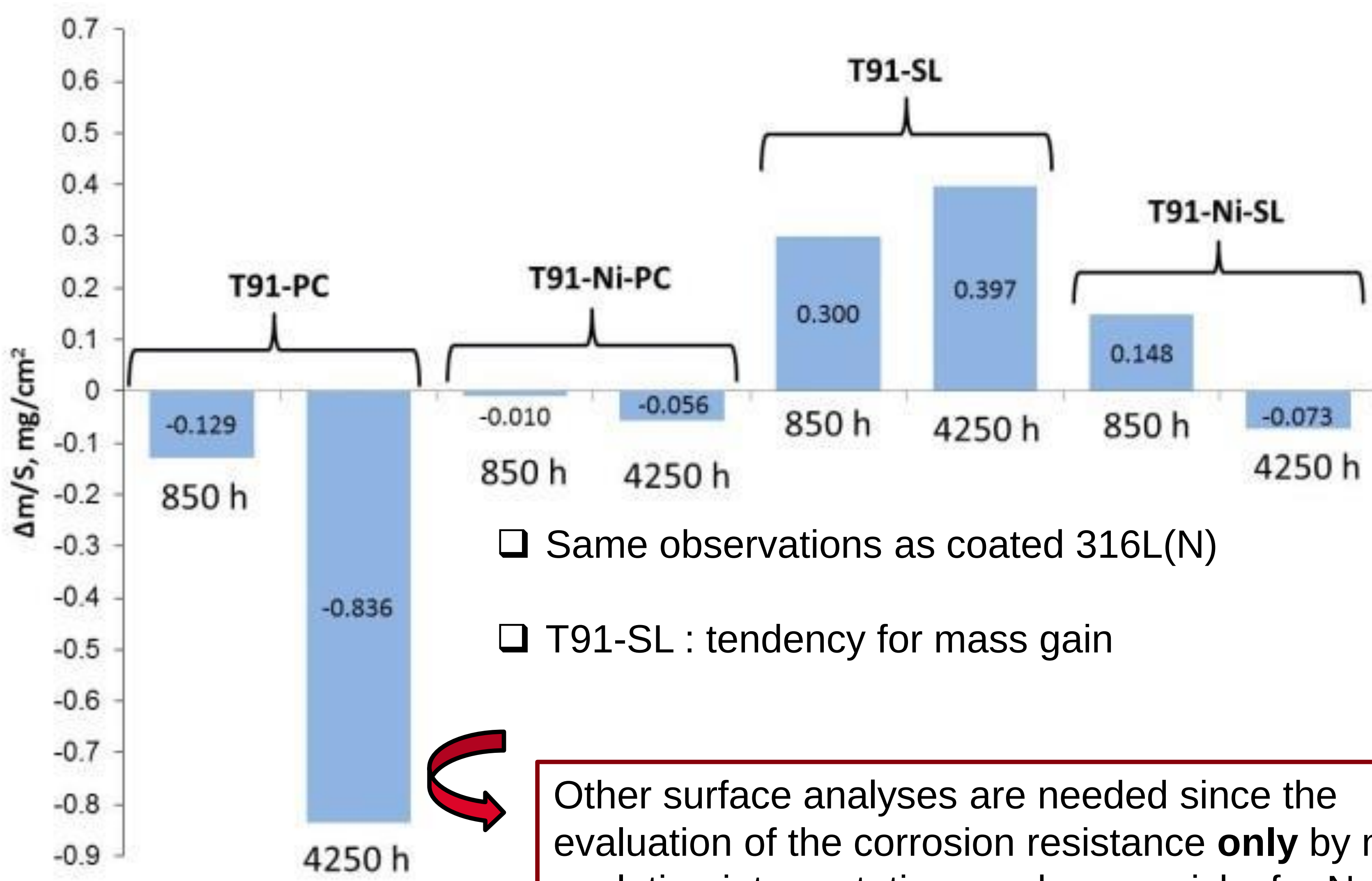
Specific washing procedure (ethanol+water) to remove residual surface sodium
Analyses : mass evolution, GDOES, SEM, Raman spectroscopy, XRD

PACK-CEMENTED 316L(N) SAMPLES



- ❑ Low mass evolution even after 4250 h
- ❑ Lower mass evolution when pre Ni plated

T91 SAMPLES COATED BY PC OR SL



- Same observations as coated 316L(N)
- T91-SL : tendency for mass gain

Other surface analyses are needed since the evaluation of the corrosion resistance **only** by mass evolution interpretation can be very risky for Na

MASS EVOLUTION INTERPRETATION

Mass evolution

+

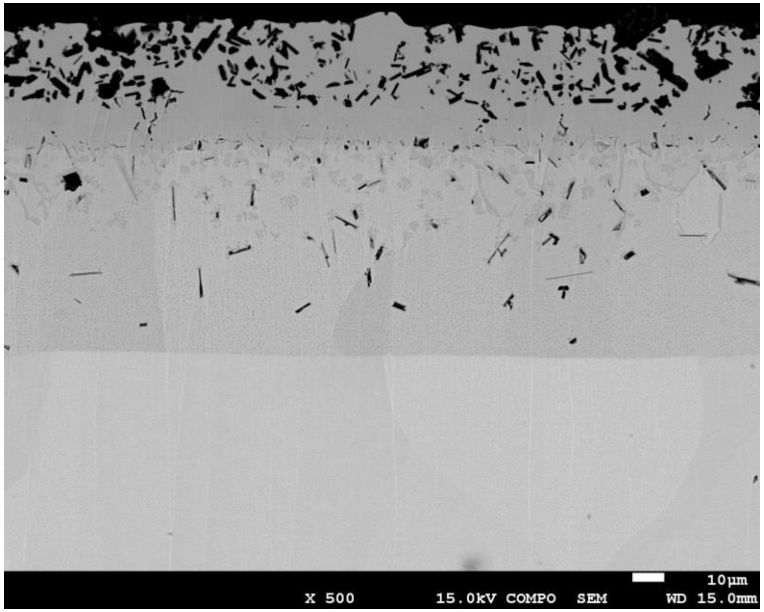
- Formation of ternary compounds (Na-Me-O/C) during exposure
- Formation of Na carbonates/hydroxide during washing procedure
- **Na penetration**

-

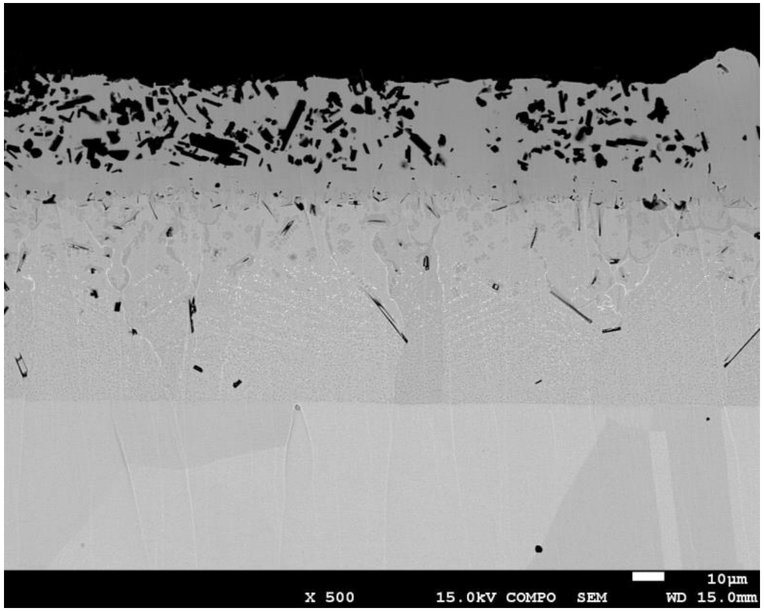
- Dissolution of metallic elements or ternary compounds during the exposure in Na
- Dissolution of metallic elements or ternary compounds during the washing procedure of samples
- Loss of cement particules stuck on the coating surface

SEM IMAGES OF 316L(N) –PC BEFORE AND AFTER EXPOSURE

316L(N) **before** exposure

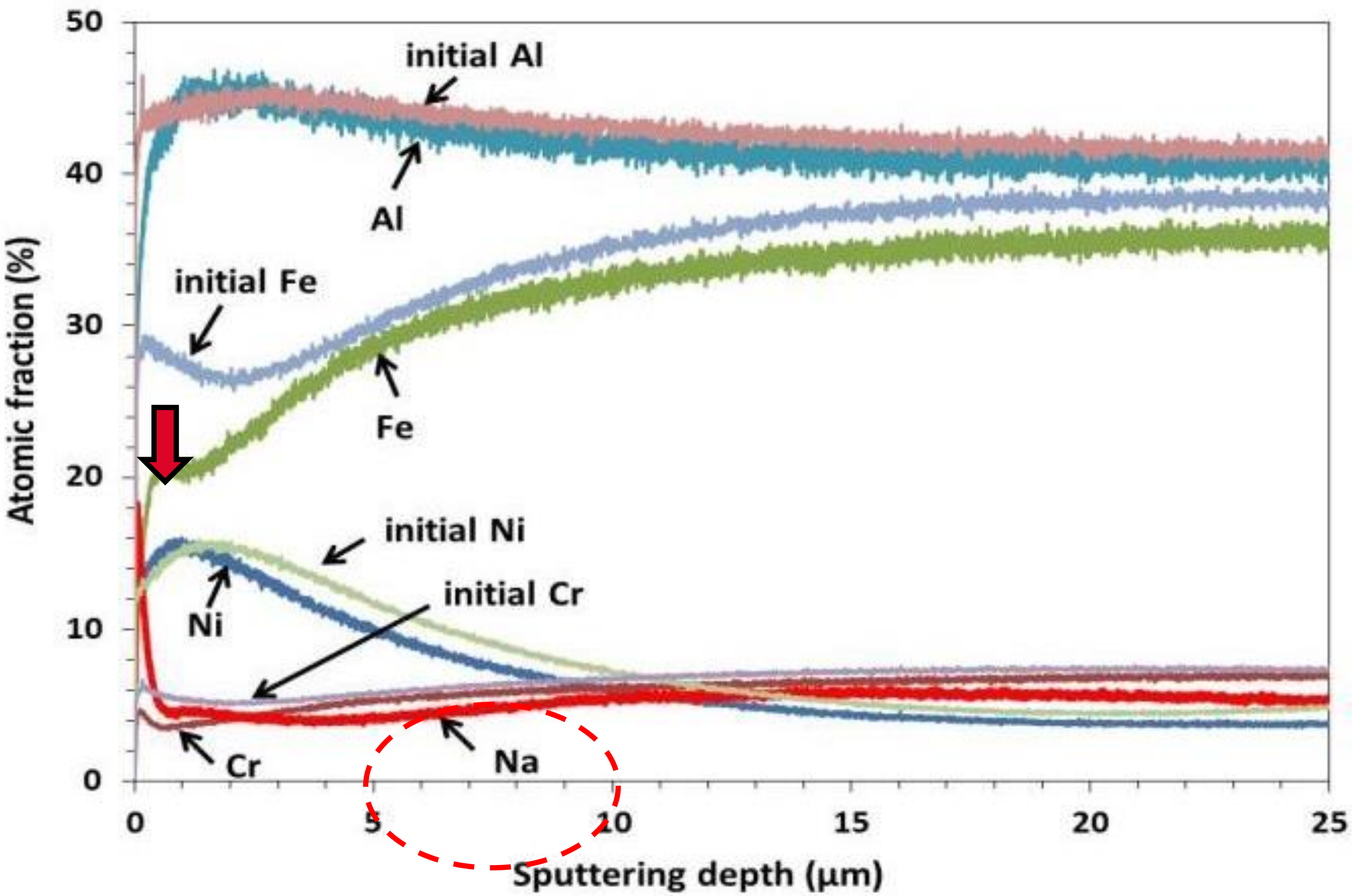


316L(N) **after** exposure in Na – 4250 h



- ❑ No clear degradation of the coating and the substrate-coating interface

GDOES PROFILES FOR 316L(N)-PC

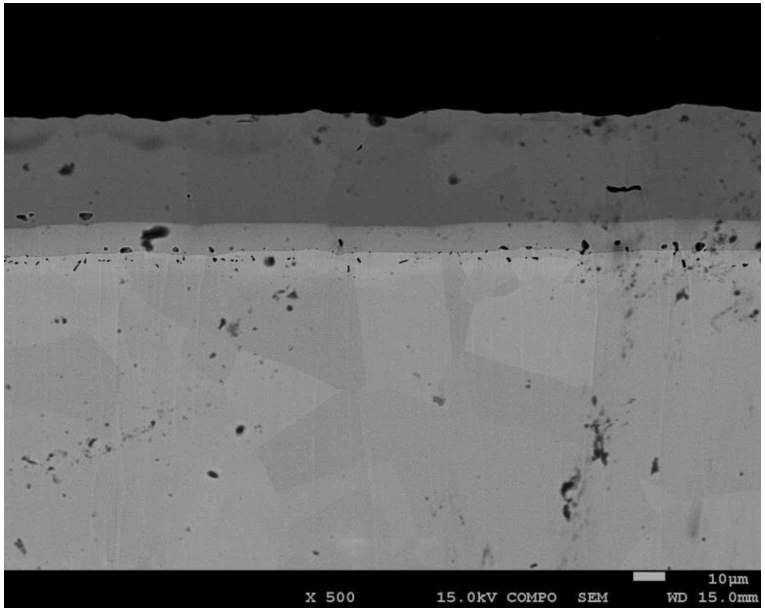


- ❑ No strong evolution of the surface coating composition as respect to its main elements : Al, Ni, Cr, Fe
- ❑ Deep penetration of Na > 25 µm in the Al_2O_3 particles enriched zone :

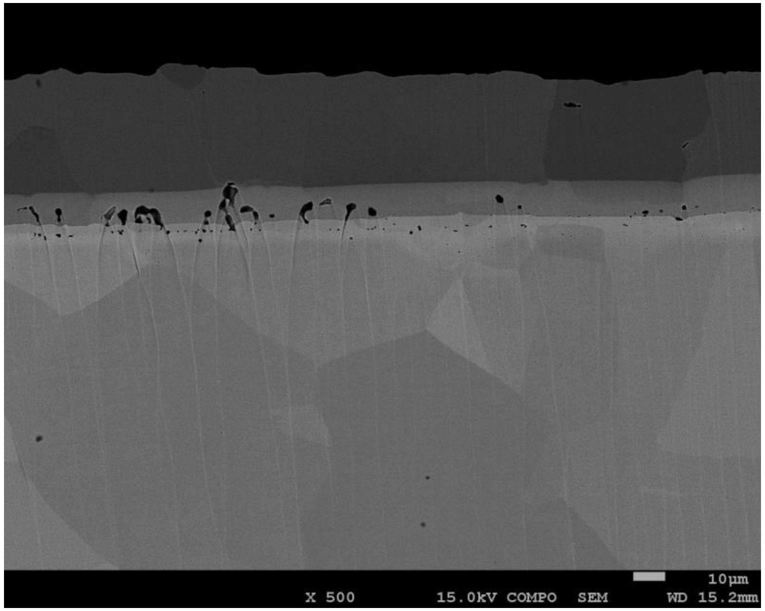
interfaces = high diffusion paths ?

SEM IMAGES OF 316L(N) - NI - PC BEFORE AND AFTER EXPOSURE

316L(N) **before** exposure

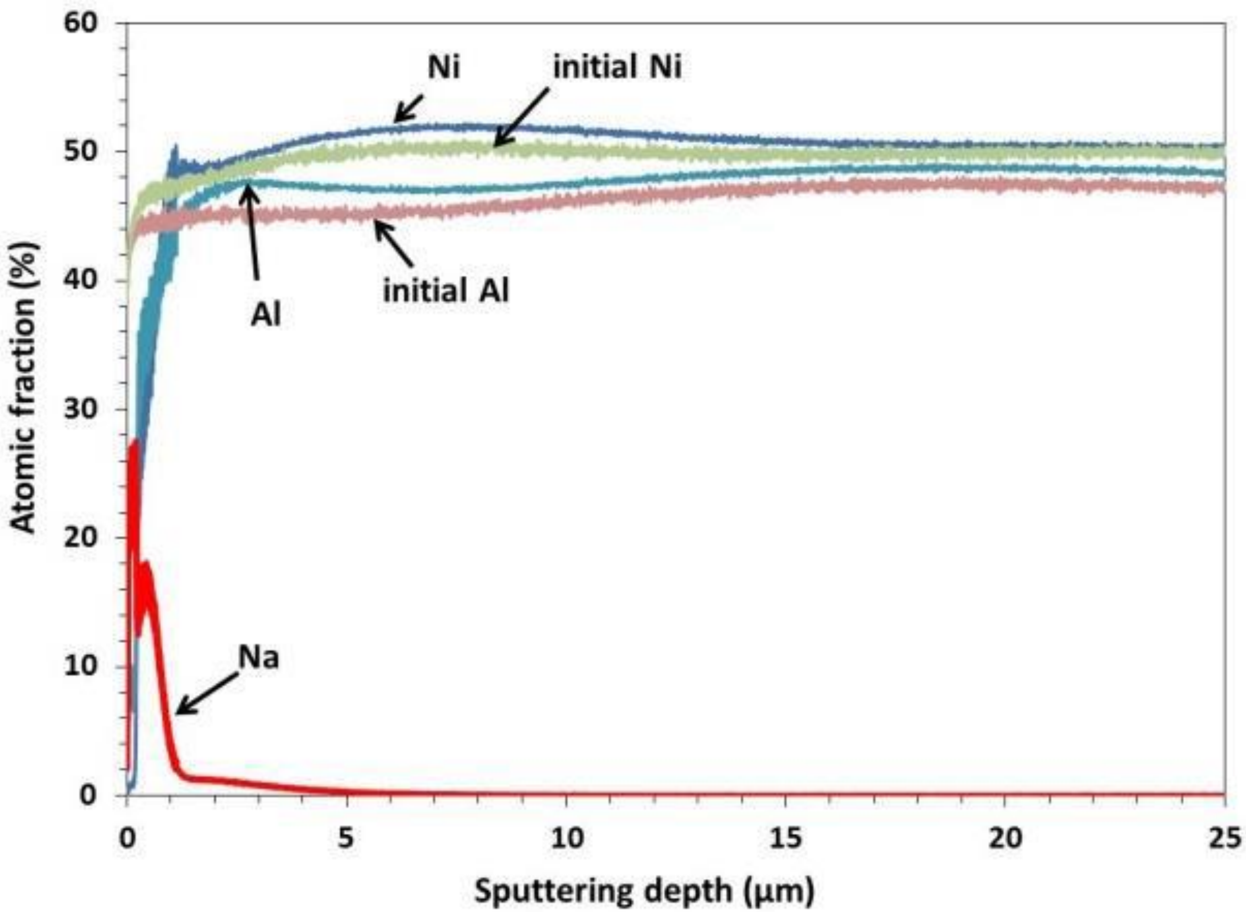


316L(N) **after** exposure in Na – 4250 h



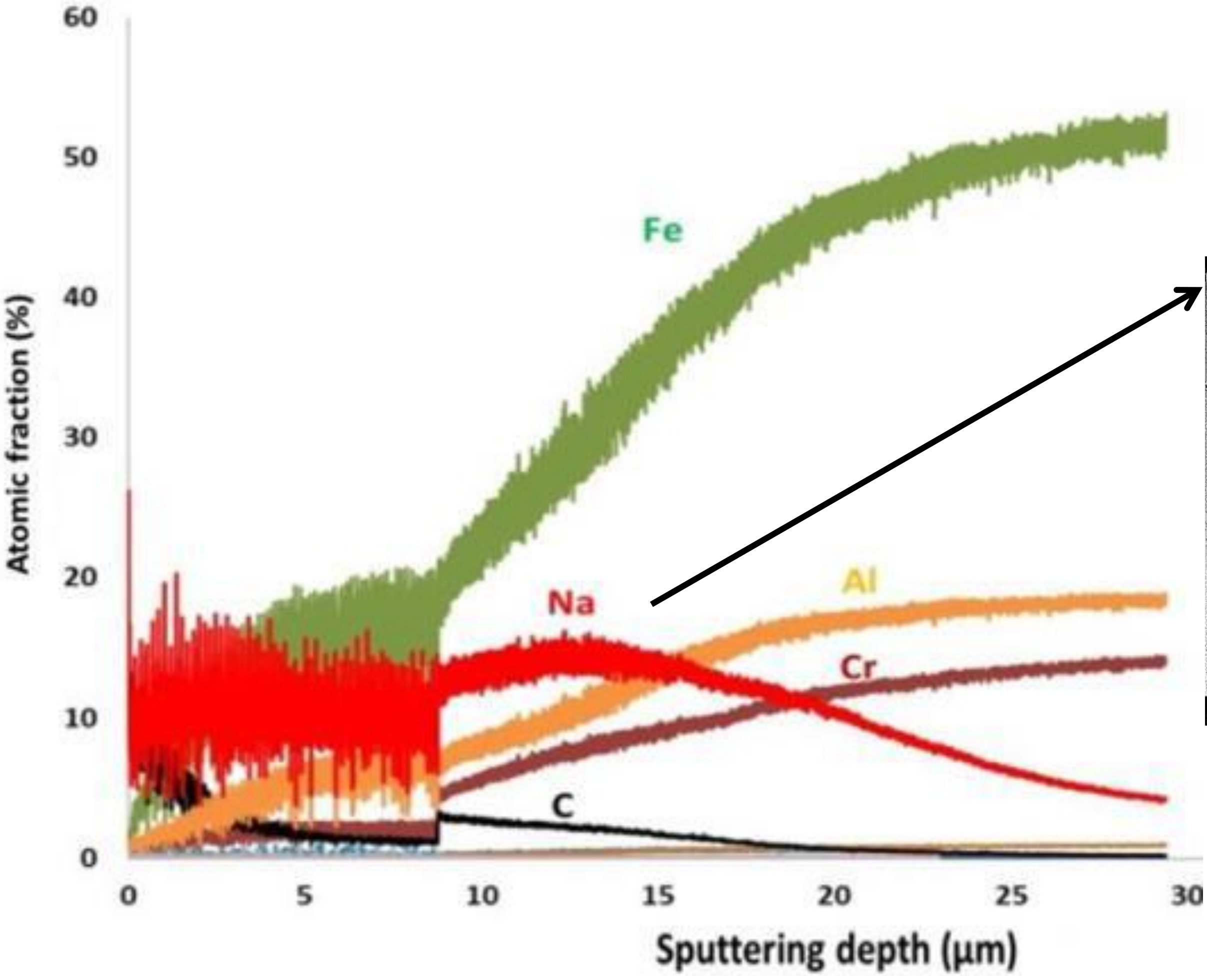
- ❑ No clear degradation of the coating surface
- ❑ Formation of voids in the Fe rich NiAl zone (Kirkendall effect)

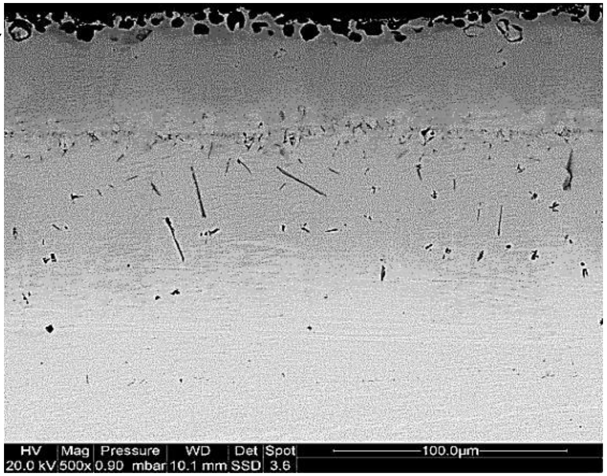
GDOES PROFILES FOR 316L(N)-NI-PC



- ❑ Same observations as for 316L(N) – PC : no clear evolution of concentrations
- ❑ **But lower penetration of Na** < 5 µm : dense coating

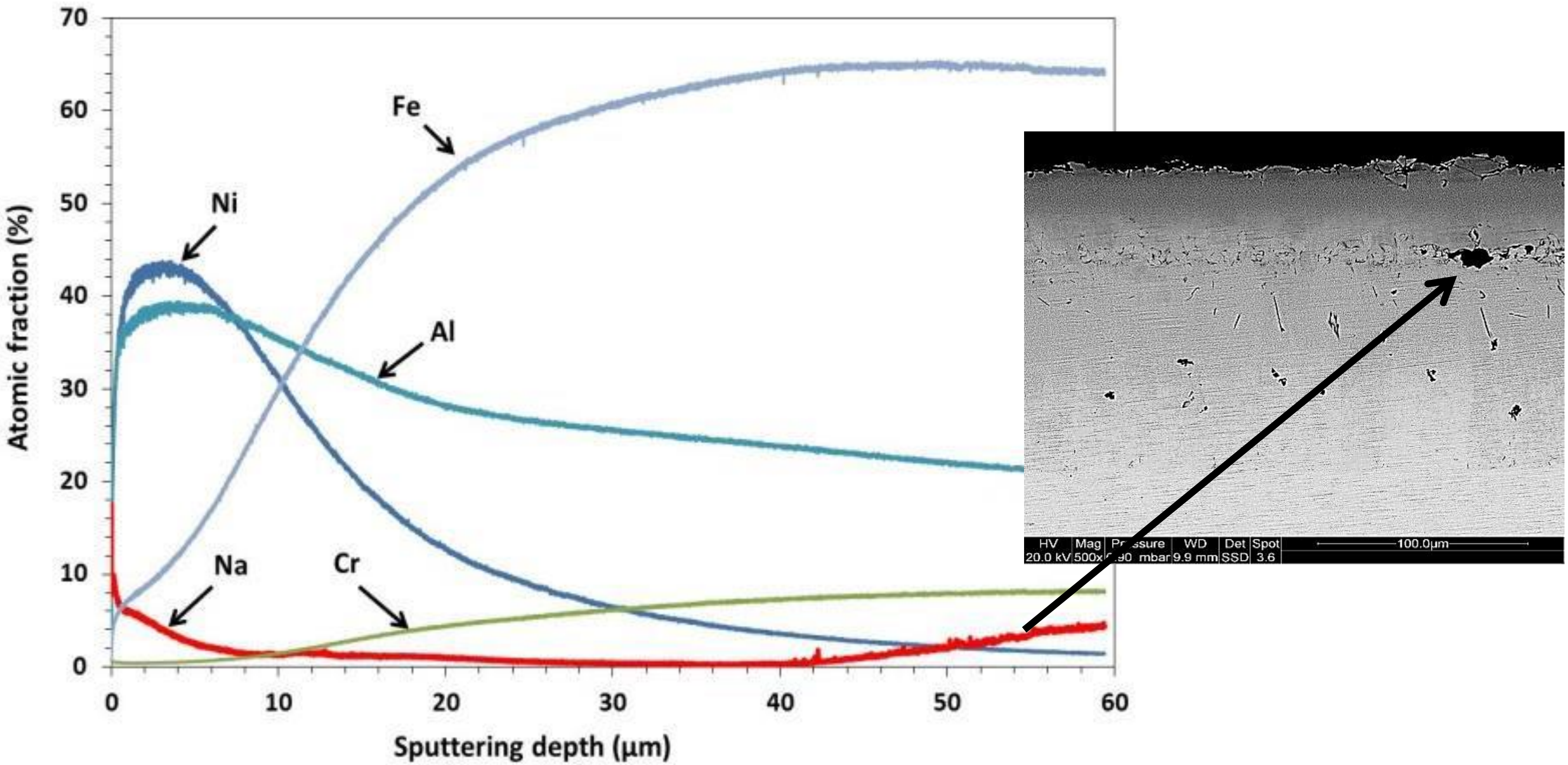
T91 COATED BY SLURRY





- ❑ No formation of ternary compounds could be detected (Raman, XRD, SEM-EDX). Only aluminides and Al_2O_3
- ❑ Na accumulation in the porous region of the coating then penetration

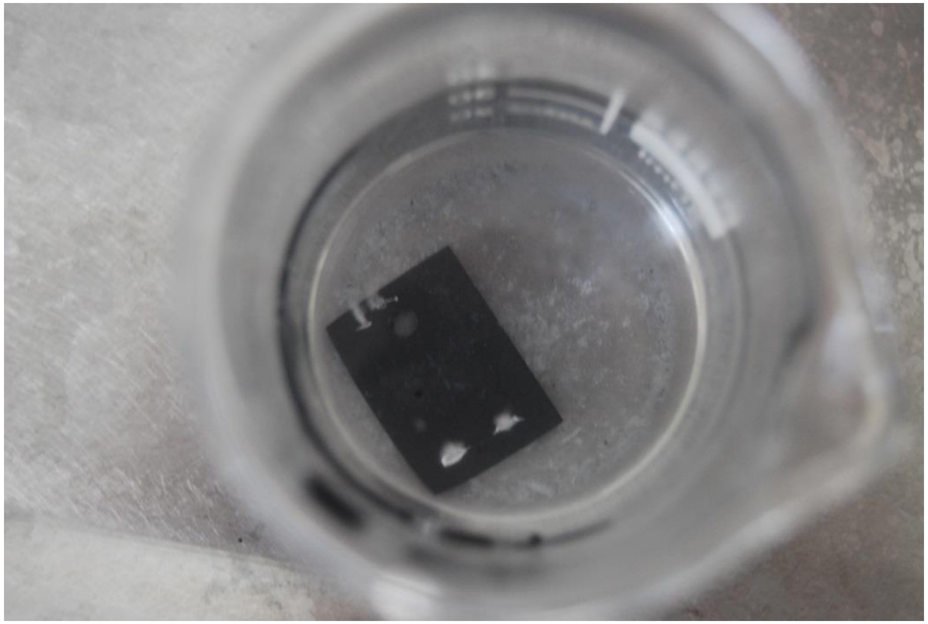
T91 – NI COATED BY SLURRY



Lower Na on the surface but penetration and accumulation detected deeper (accumulation in voids ?)

AL DISSOLUTION ?

Yes !



EDX analysis on Cr sample surface

3 wt% of Al

[from Al_2O_3, Fe-Ni aluminides]

CONCLUSIONS FROM EXPOSURE TEST

- No severe degradation of the aluminide coatings whatever the process used (PC or SL) : Aluminides are quite stable in HT Na

In good agreement with results from litterature :

- Maximum corrosion rate of NiAl on IN718 = 0.75 µm/year at 625°C *[Johnson et al., LIMET 1976]*
- No evolution of the concentration of Al, Fe, Cr and Ni in the aluminide deposited on 304, 316 and IN718 alloys after exposure in flowing Na at 400 °C for 3450 h completed by 350 h at 550 °C *[Depierre at al., Materials Science And Engineering* ***88****, 287 (1987)]*

- But still :
  - Slight Al dissolution
  - Na penetration (function of the density of the microstucture of aluminides, particles, cracks, …) :

CONCLUSIONS FROM EXPOSURE TEST

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  In good agreement with results from litterature :

  - Maximum corrosion rate of NiAl on IN718 = 0.75 µm/year at 625°C *[Johnson et al., J IMET 1976]*

  in

)]

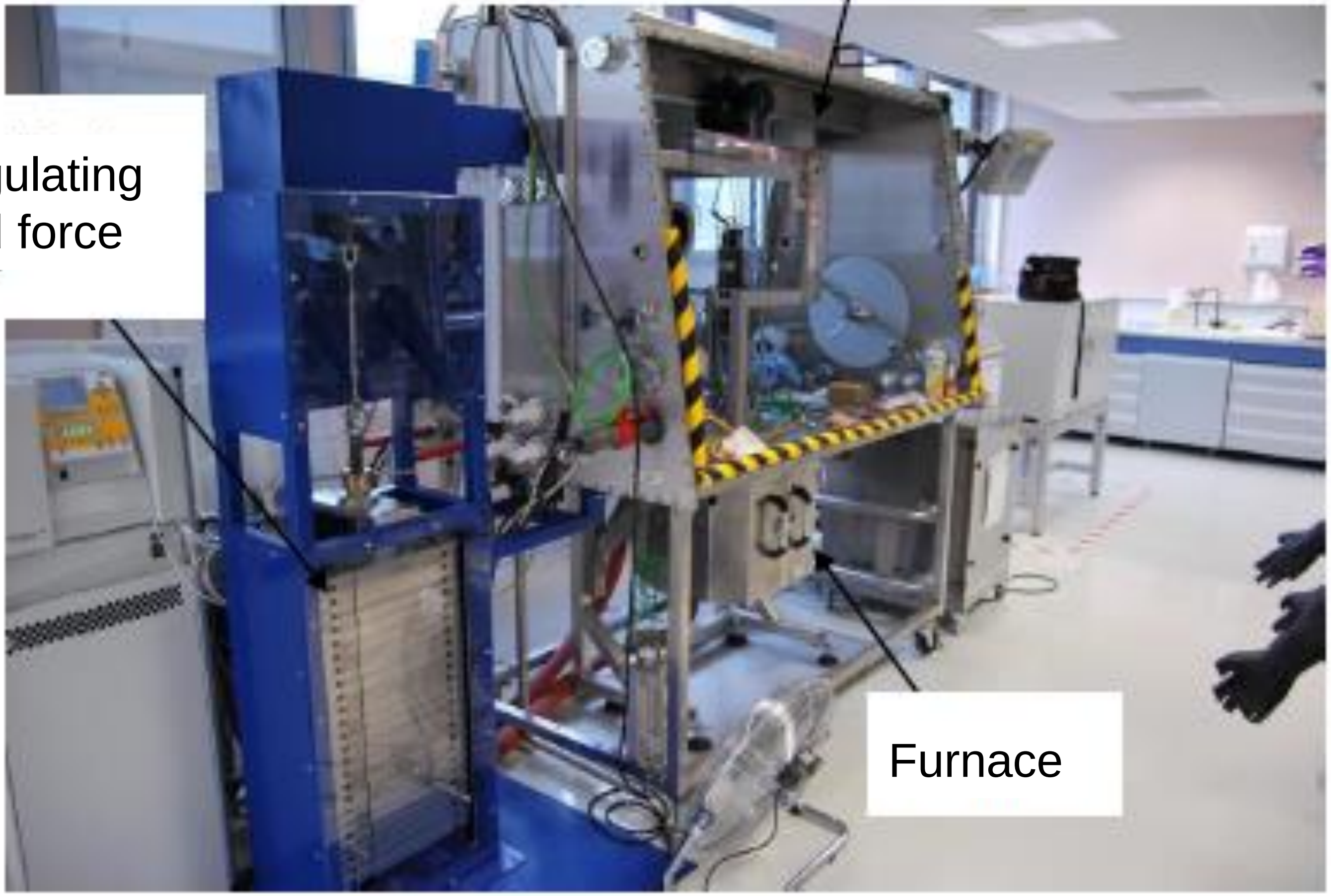
What about the tribological behaviour of aluminides in Na ?

- But still :
  - Slight Al dissolution
  - Na penetration (function of the density of the microstucture of aluminides, particles, cracks, …) : effect of tribological behaviour ?

SODIUM FACILITY FOR FRICTION-WEAR TESTS

Glove box

Weights regulating the applied force



Furnace

SODIUM FACILITY FOR FRICTION-WEAR TESTS

316L(N) pin

Sliding surface

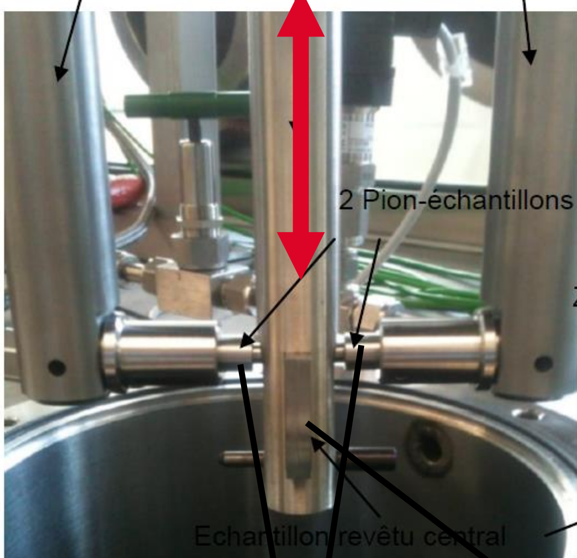
8 mm

16 mm

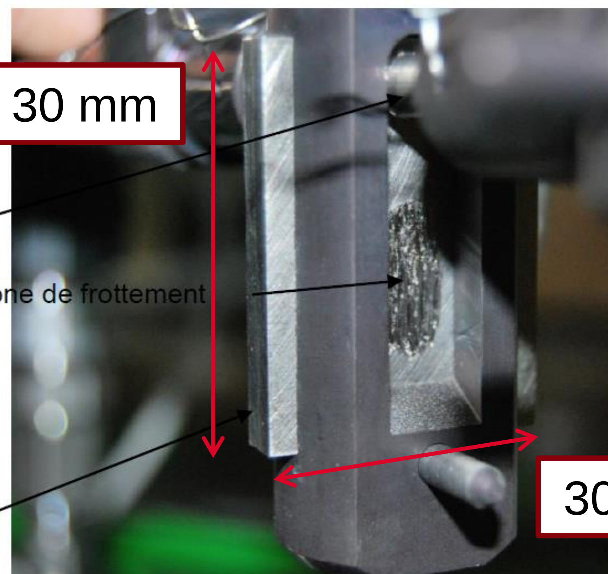
9 mm

Arms applying normal force F_n

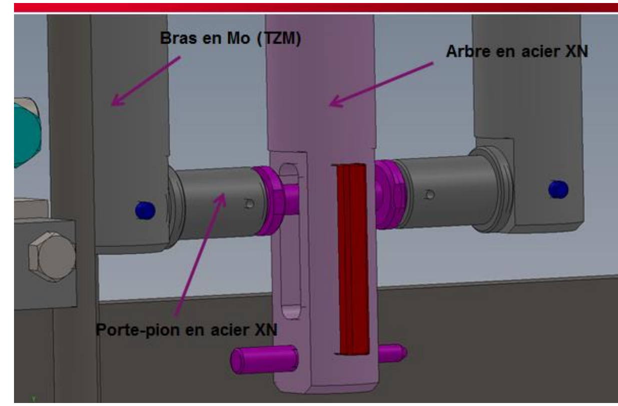
Oscillating translation movement



30 mm



30 mm



SolidWorks

316L(N) Pins

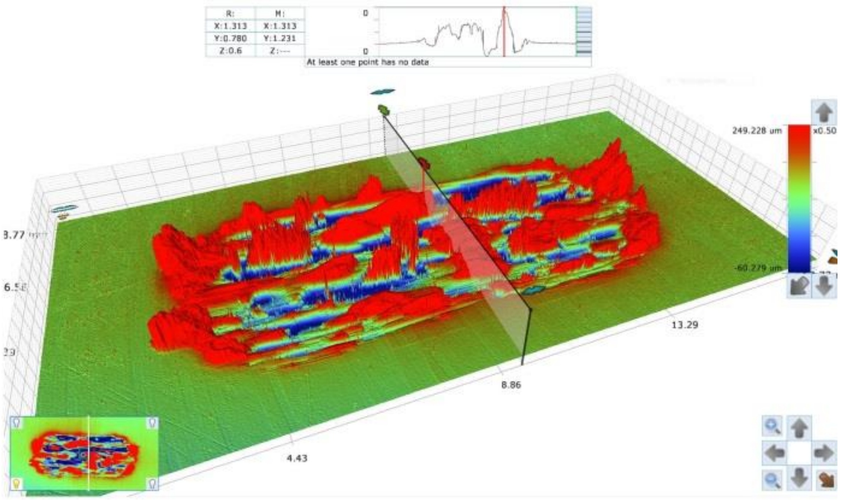
Coated sample

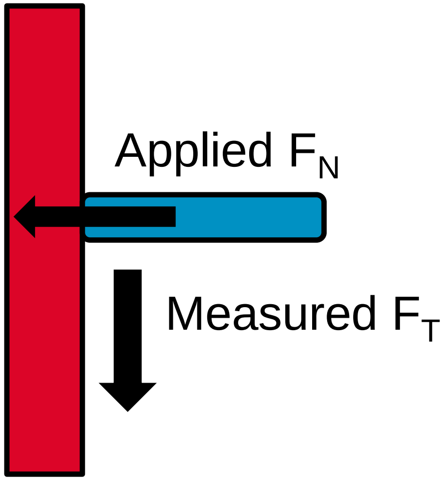
WHAT CAN WE MEASURE ?

- Transversal force F_T needed for the oscillating translation movement :

→ **FRICTION COEFFICIENT**

- Mass variation : pins and coated sample
- Mascroscopic images → **WEAR**
- Profilometric measurments : lost, gained and deplaced volume of material





$$f = F_T / F_N$$

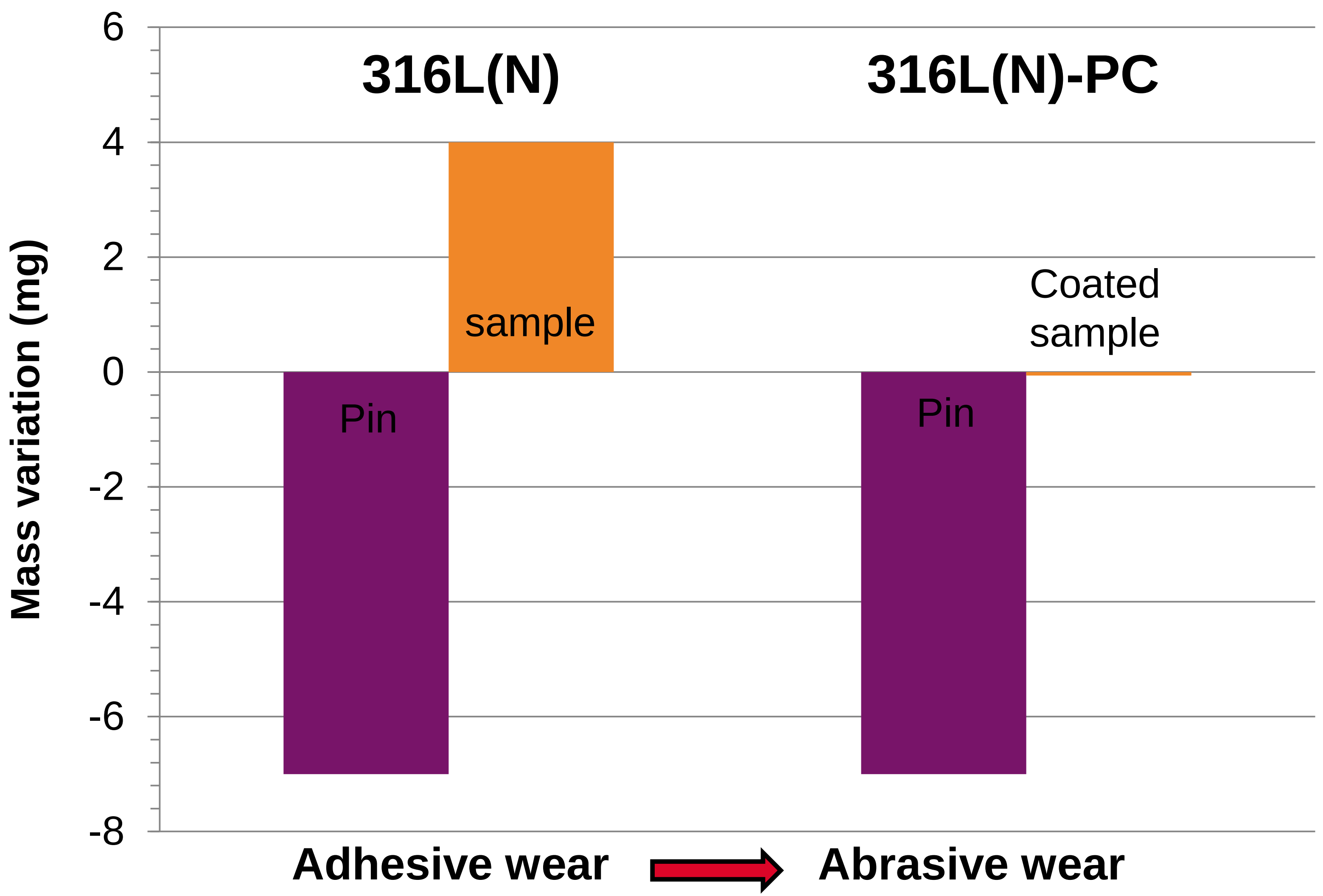
EXPERIMENTAL CONDITIONS

Na temperature	200°C
Translation rate	1 mm/s
Tracing length	10 mm
Total distance	4 m
Applied Stress	31 MPa

Pin material	316L(N)
Coated sample	316L(N) or 316L(N)-PC

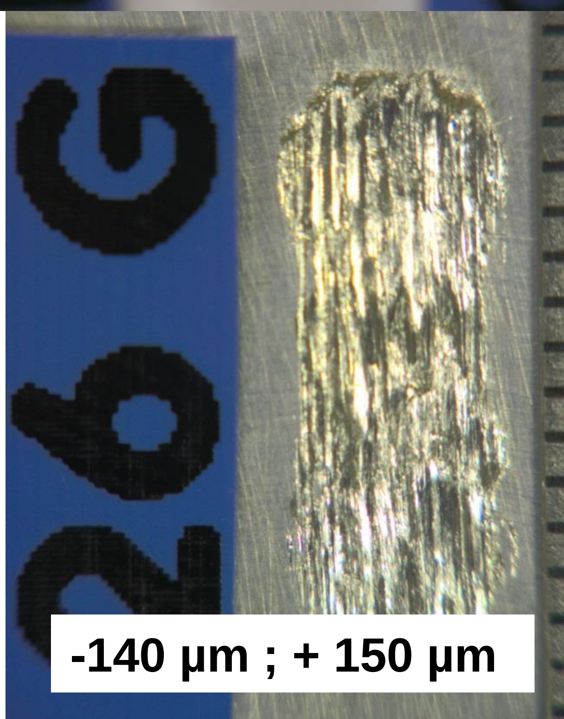
2 friction tests : with or without coatings

MASS EVOLUTION



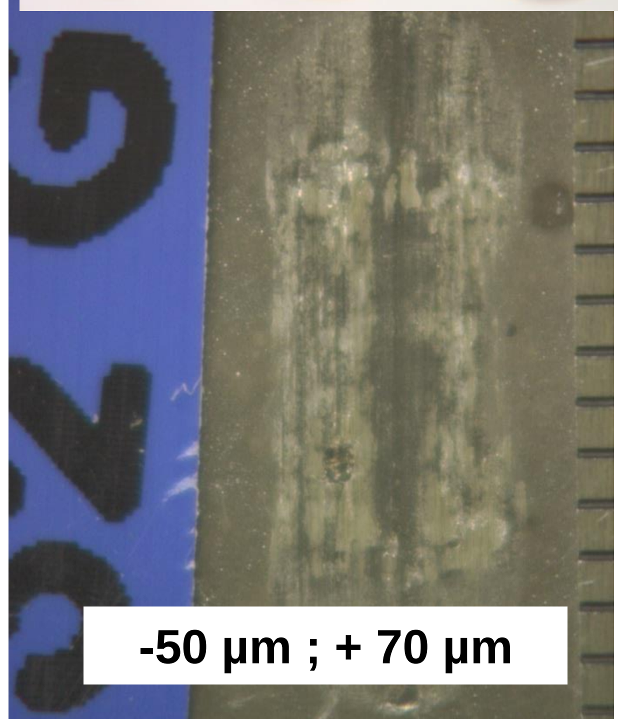
MACROSCOPIC IMAGES

316L(N) - 316L(N)



-140 µm ; + 150 µm

316L(N) - 316L(N) PC

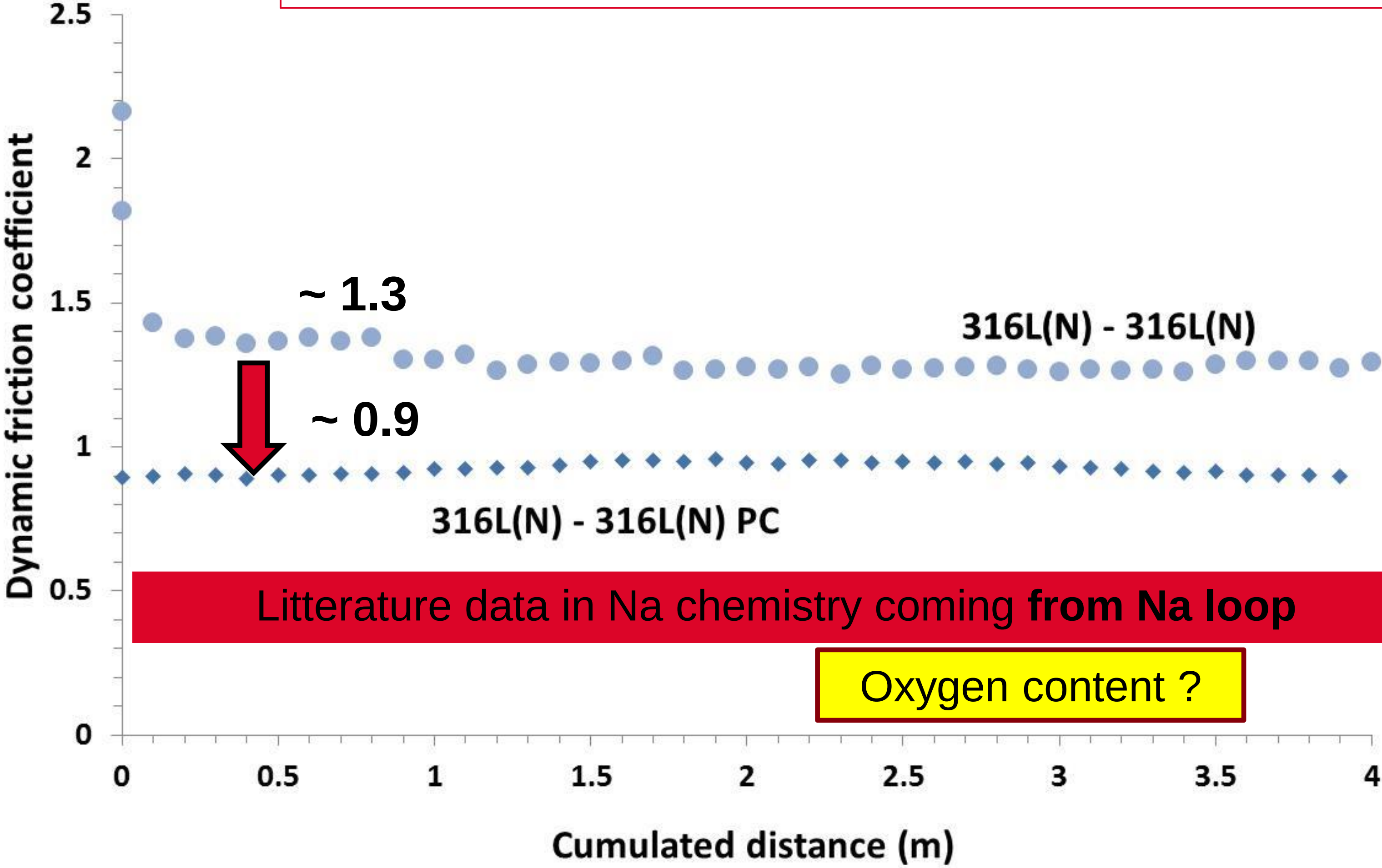


-50 µm ; + 70 µm

Lower wear for 316L(N) PC

FRICTION COEFFICIENT MEASUREMENTS

Lower friction coefficient for 316L(N)-PC



Litterature data in Na chemistry coming **from Na loop**

Oxygen content ?

CONCLUSIONS AND PROSPECTS

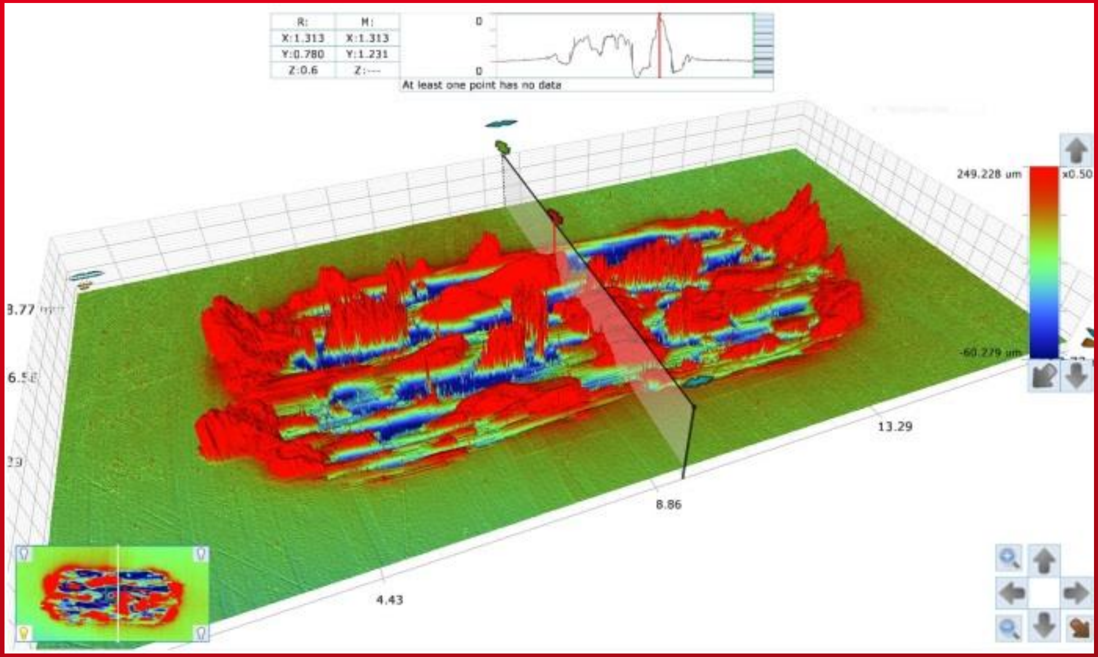
Conclusions

- ❑ Good compatibility of aluminides in Na (up to 550°C)
- ❑ Good wear resistance and lower friction coefficient than without coating

Prospects

- ❑ What are the influence of the aluminide coating on the wear/friction properties (nature, roughness) ?
- ❑ What is the influence of the [O] content in Na on the wear/friction properties ?
- ❑ Development of new coatings (additonnal elements) favoring the formation of lubricating surface compounds in Na (reactor chemistry)

Thank You for your attention



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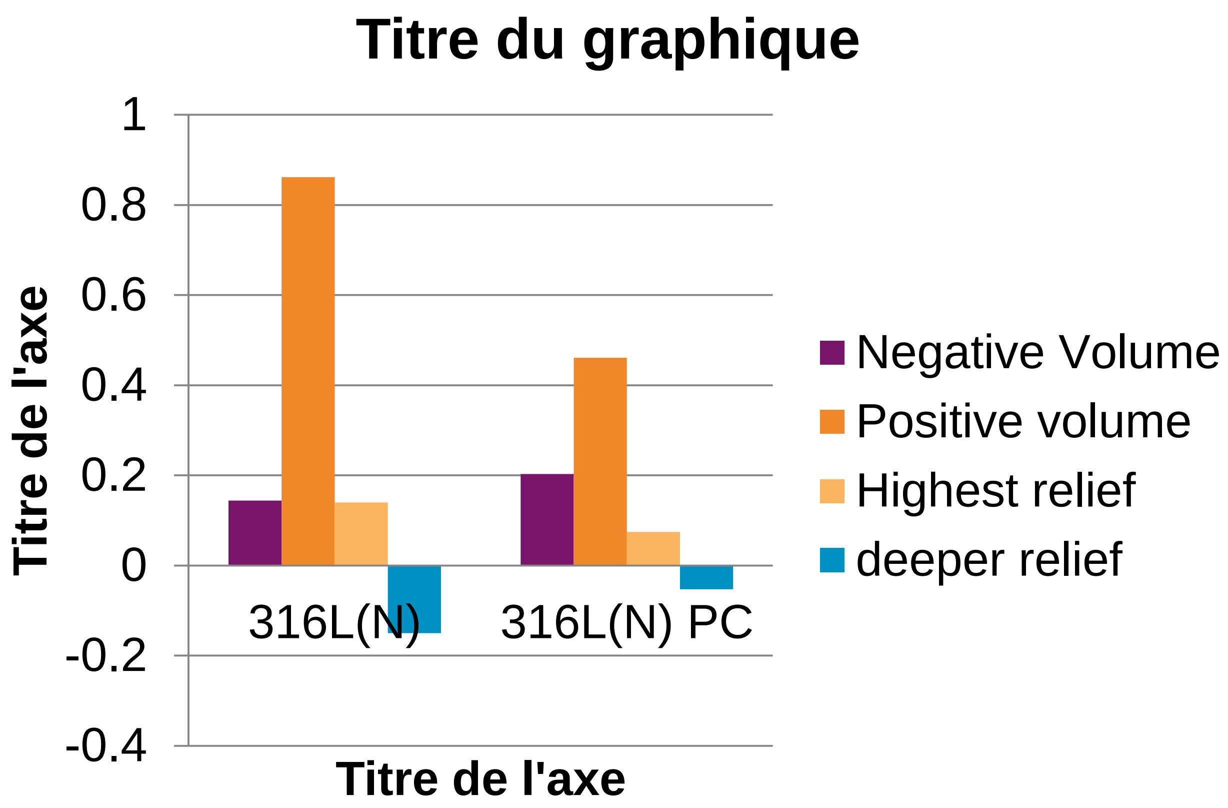
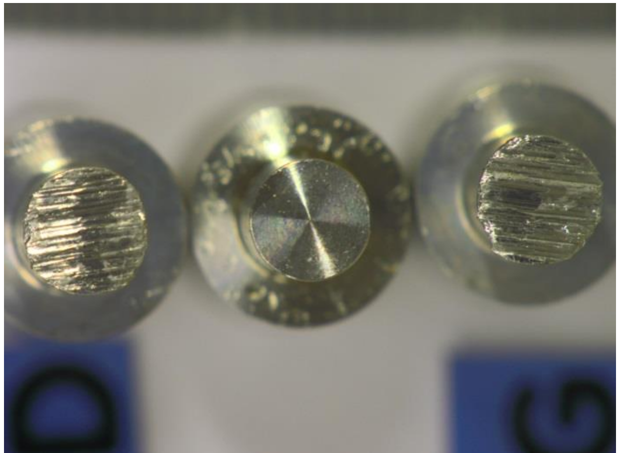
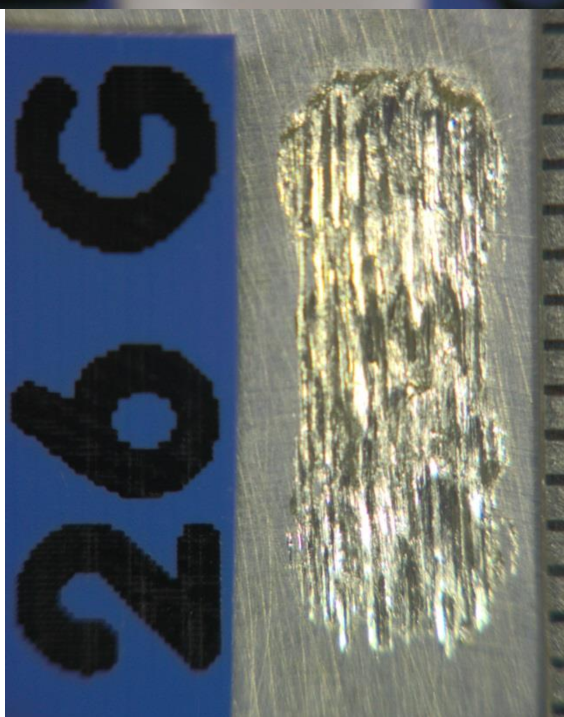

Titre du graphique
1
0.8
0.6
0.4
0.2
0
-0.2
-0.4
Titre de l'axe
316L(N)
316L(N) PC
Titre de l'axe
Negative Volume
Positive volume
Highest relief
deeper relief

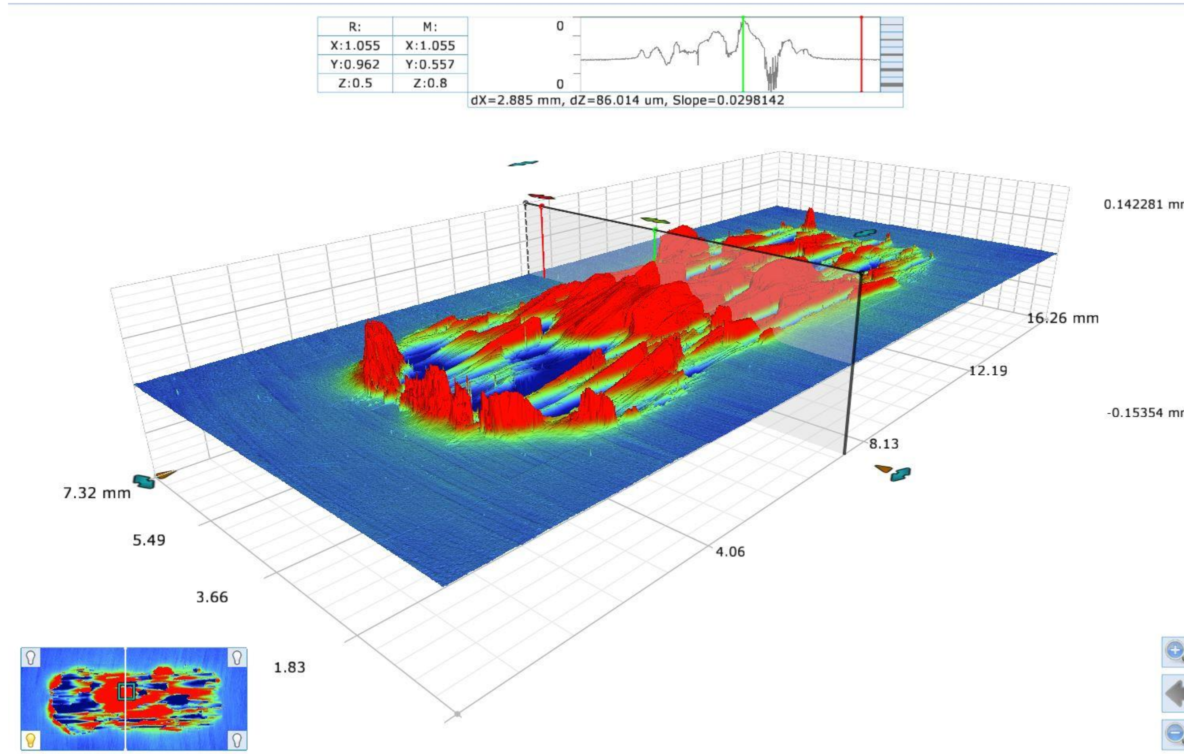
Test	Pin (mg)	Coated sample (mg)	Wear
316L(N) - 316L(N)	-7	+4	Adhesive wear
316L(N) - coated 316L(N) PC	-7	-0.06	Abrasive wear
316L(N) - coated 316L(N) Ni PC	-10	+4	Adhesive wear

316LN – 316LN 200°C



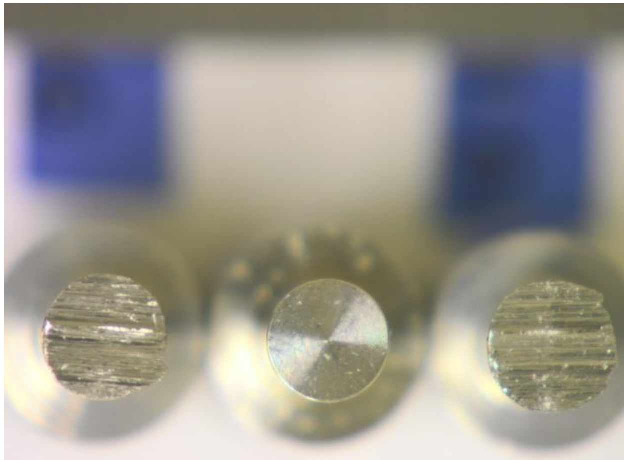


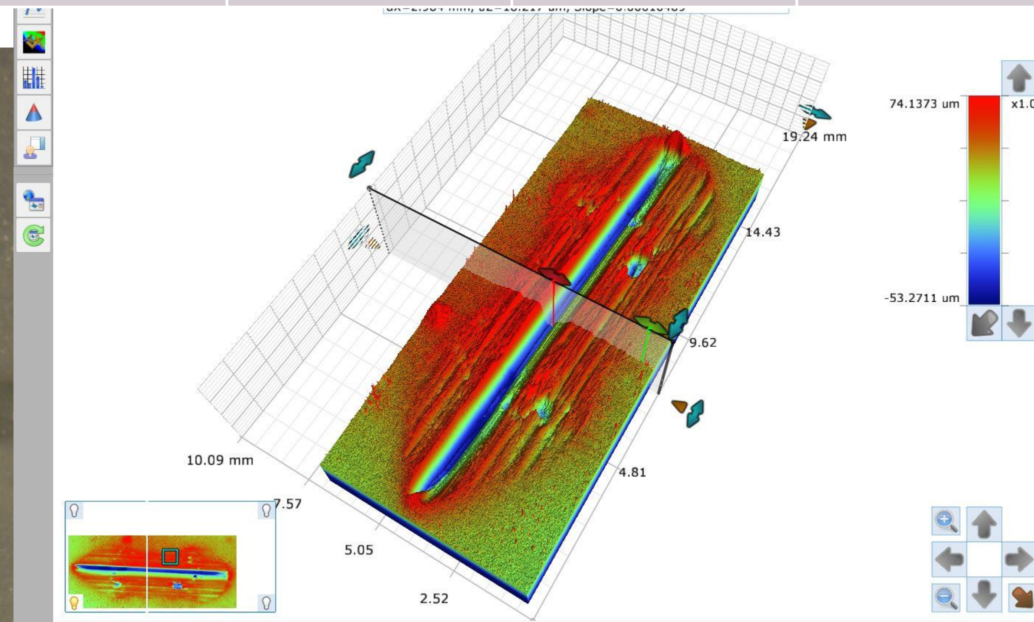
Volume + (mm3)	Volume -	Gained	Relief (µm)
0.862	0.144	+0.718	-150-+140



316LN – 316LN PC - 200°C AND 500°C

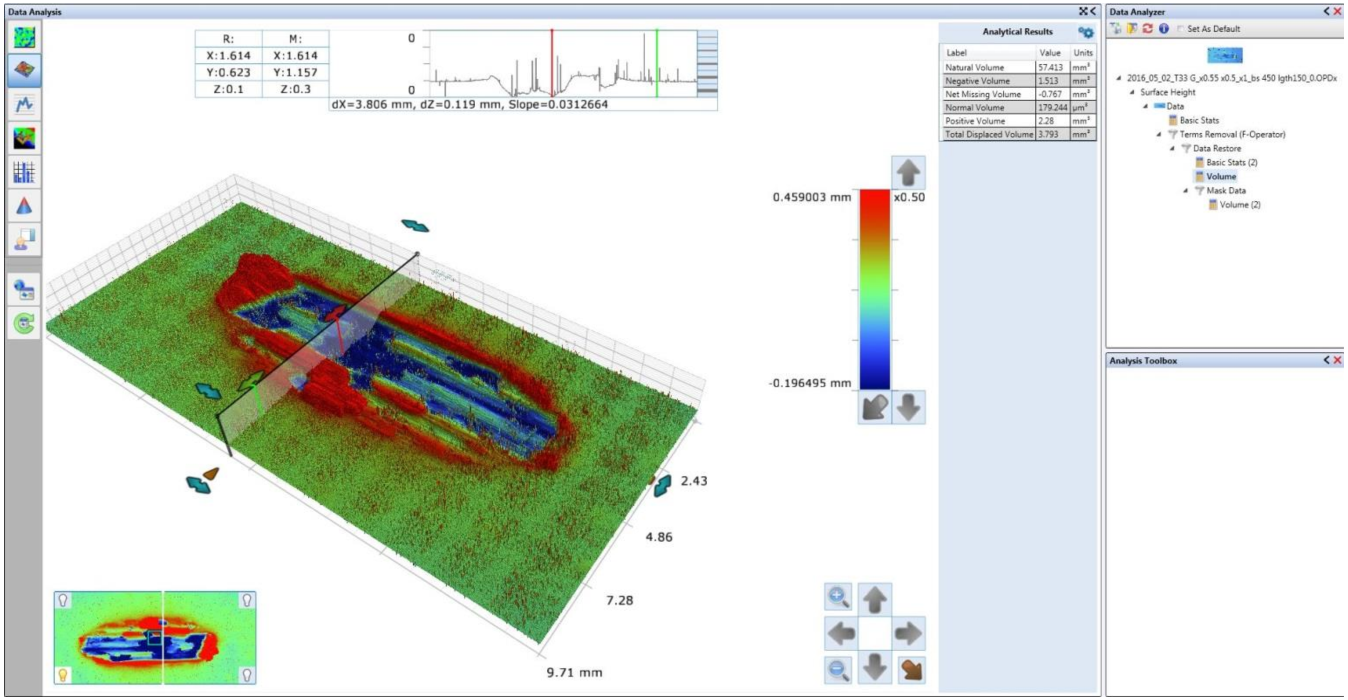
Volume + (mm3)	Volume -	Gained	Relief (µm
0.461	0.203	+0.258	-53-+74





316LN – 316LN NI PC - 200°C AND 500°C

Volume + (mm3)	Volume -	Gained	Relief (µm)
2.28	1.51	+0.77	-196-+460



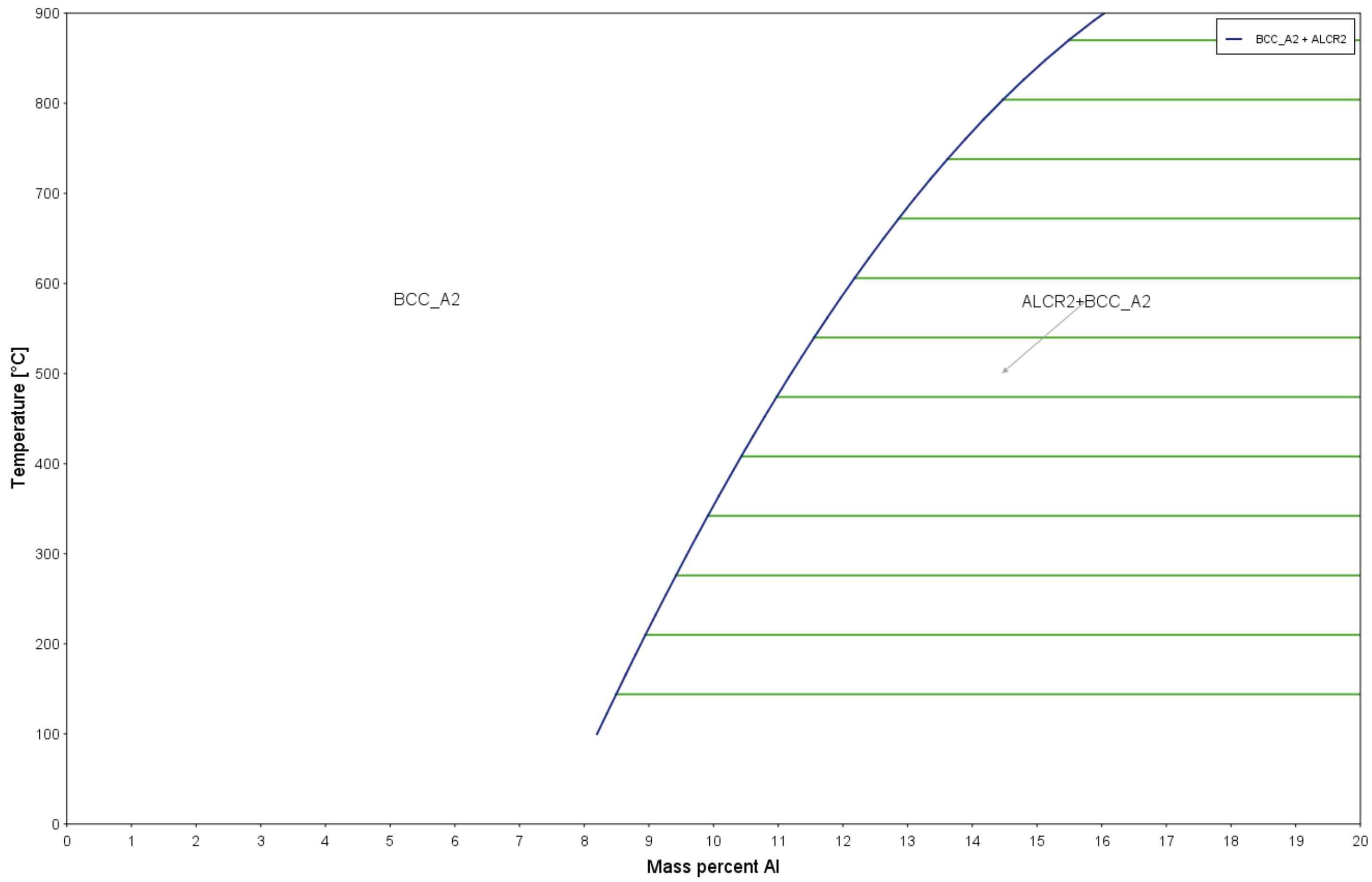
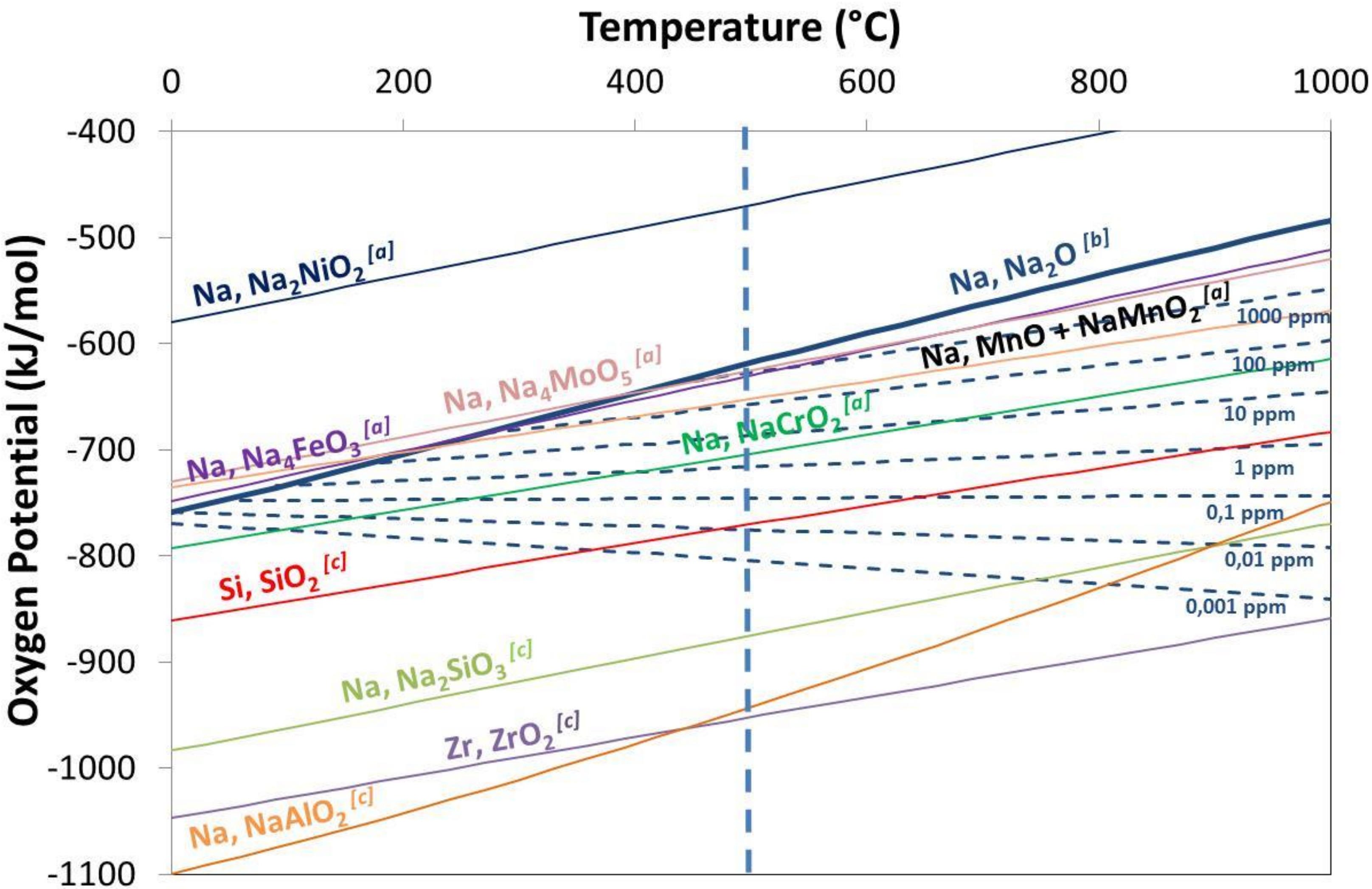
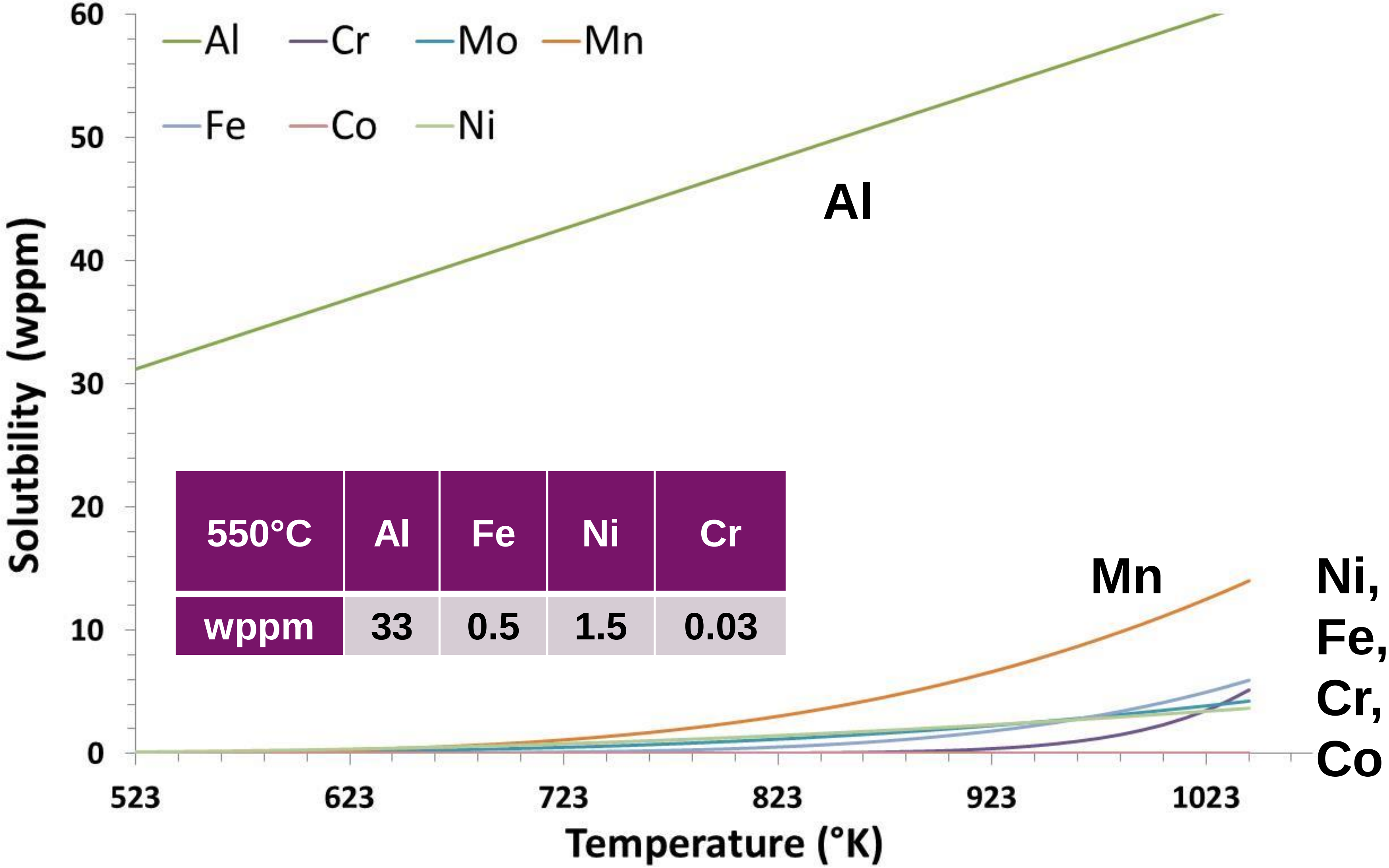


Diagramme thermos du binaire Cr-Al

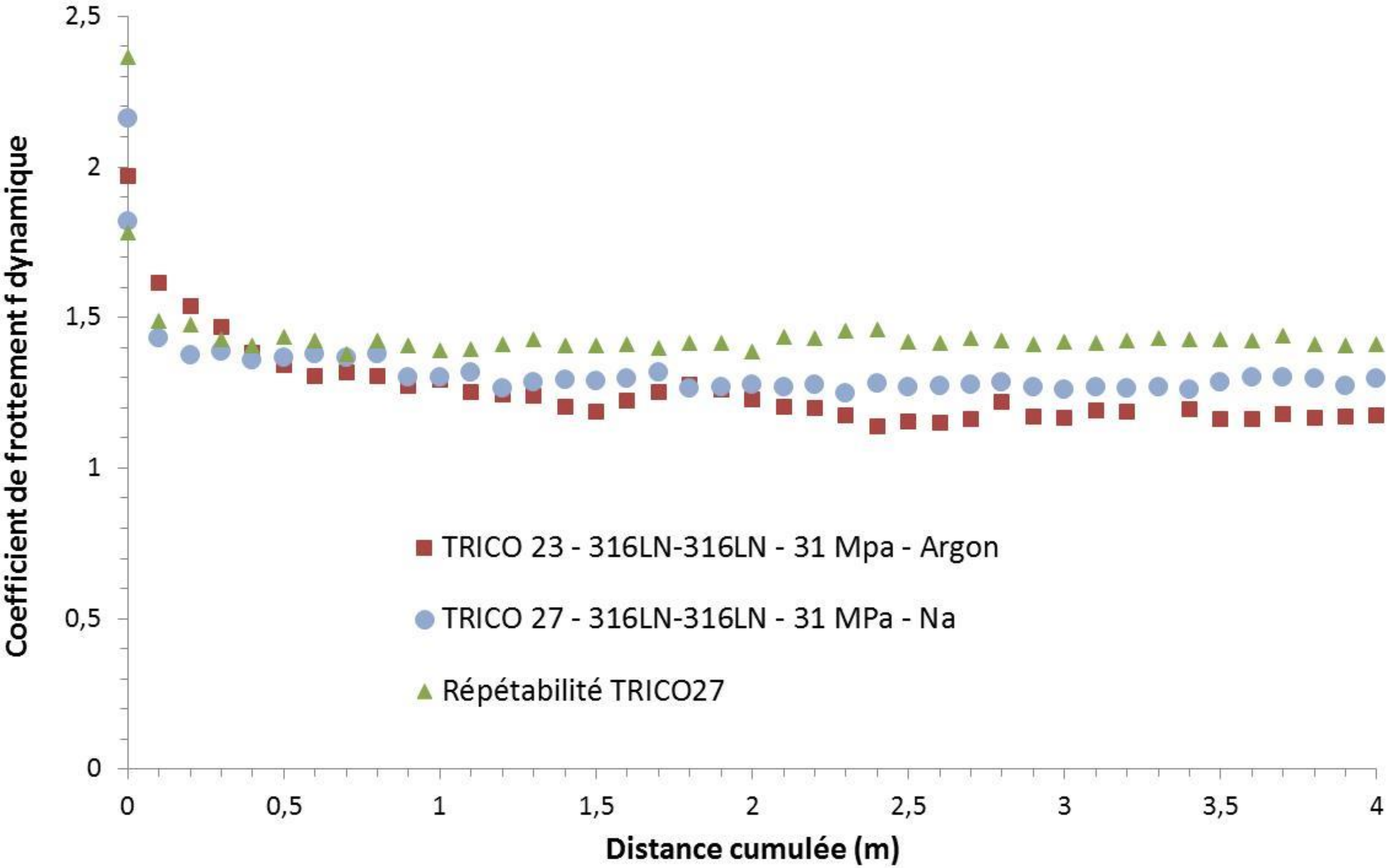
A 550°C, solubilité de Al = 11.6 wt% puis formation de $AlCr_2$


Temperature (°C)
0
200
400
600
800
1000
-400
-500
-600
-700
-800
-900
-1000
-1100
Oxygen Potential (kJ/mol)
Na, Na2NiO2 [a]
Na, Na2O [b]
Na, MnO + NaMnO2 [a]
Na, Na4MoO5 [a]
Na, Na4FeO3 [a]
Na, NaCrO2 [a]
Si, SiO2 [c]
Na, Na2SiO3 [c]
Zr, ZrO2 [c]
Na, NaAlO2 [c]
1000 ppm
100 ppm
10 ppm
1 ppm
0,1 ppm
0,01 ppm
0,001 ppm

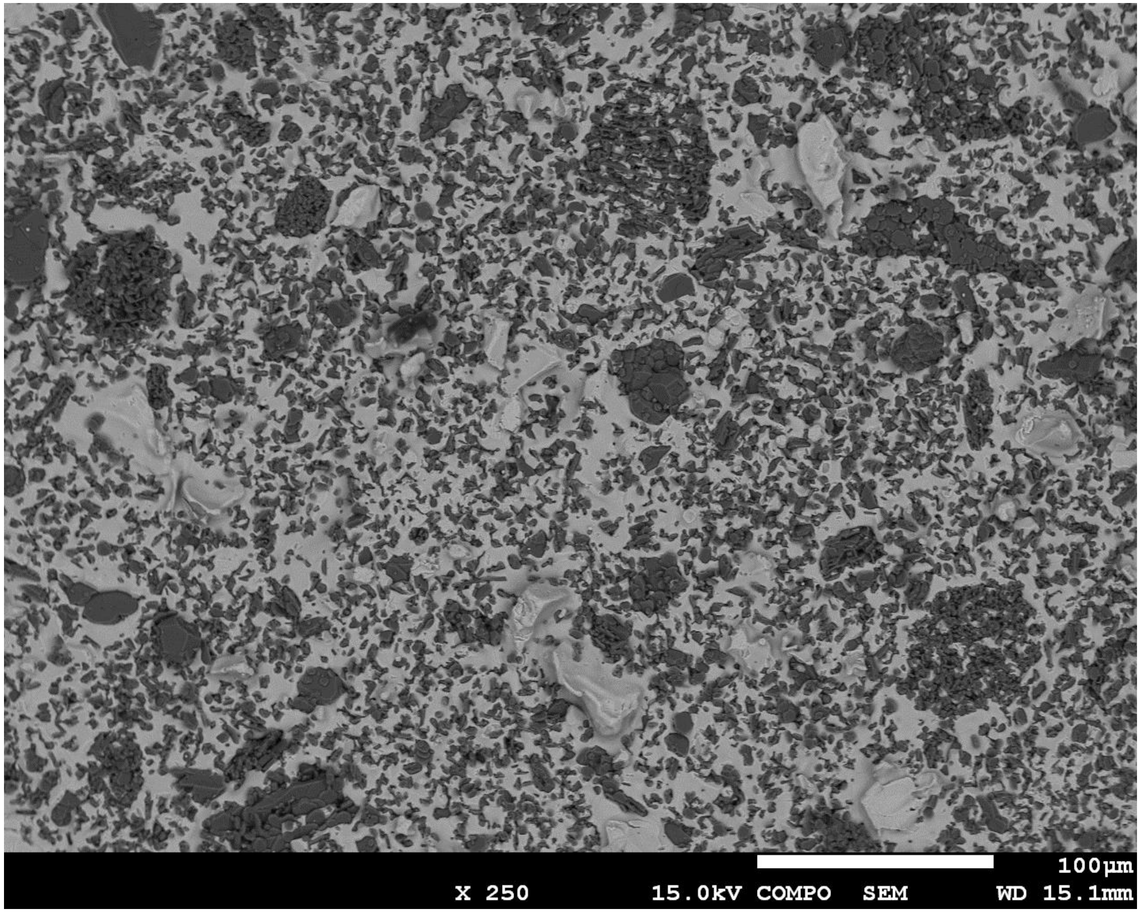


550°C	Al	Fe	Ni	Cr
wppm	33	0.5	1.5	0.03

[Applied chemistry of the alkali metals, Borgstedt, 1987]


Coefficient de frottement f dynamique
2,5
2
1,5
1
0,5
0
0
0,5
1
1,5
2
2,5
3
3,5
4
Distance cumulée (m)
TRICO 23 - 316LN-316LN - 31 Mpa - Argon
TRICO 27 - 316LN-316LN - 31 MPa - Na
Répétabilité TRICO27

316L(N) – as received - PC



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Thank You for your attention

More information :

www.portail-fluides-supercritiques.com

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