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Joining of ultra-high temperature ceramics

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Joining of ultra-high temperature carbides

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Outline

- Introduction
- Aim of the work
- Starting materials
- Joint assembly
- Microstructure after joining
- Bonding mechanism
- Nanoindentation
- Conclusions & perspectives

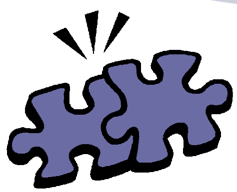
Introduction

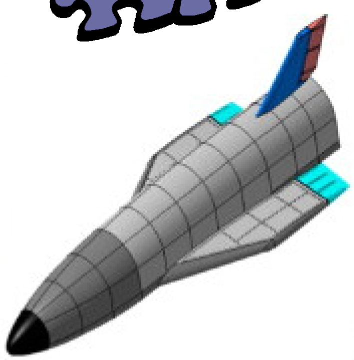
Materials for high temperature applications:

Borides and Carbides of Group IV, V transition metals

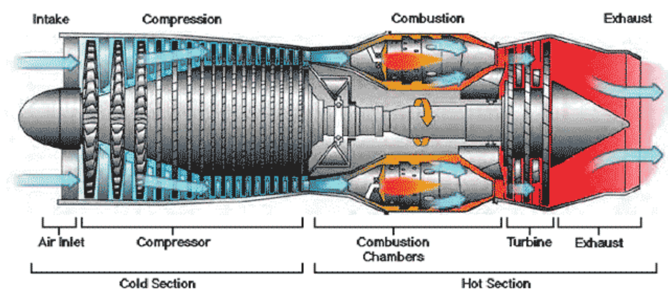
- High melting point
- High strength
- High hardness and stiffness
- Relatively good oxidation

- Low fracture toughness
- High density
- Processing often imposes simple shapes
- Difficult machining





How to succesfully **integrate UHTCs** into hypersonic structures/turbine engines without losing the high temperature properties?



Ceramic bonding

Solid state bonding
- ☺ high precision
- ☹ only simple shapes, flat components, high P, high T

Brazing and Active metal brazing
- ☺ pressureless, complicated shapes, moderate bonding T
- ☹ high vacuum and clean bonding atmosphere, limited excercise T

Liquid phase bonding
- ☺ pressureless, complicated shapes,
- ☹ possible degradation of the constituents, limited excercise T

Transient liquid phase bonding
- ☺ pressureless, complicated shapes, high precision, exercise T> bonding T
- ☹ brittle reaction products, cte mismatch

Aim of the work

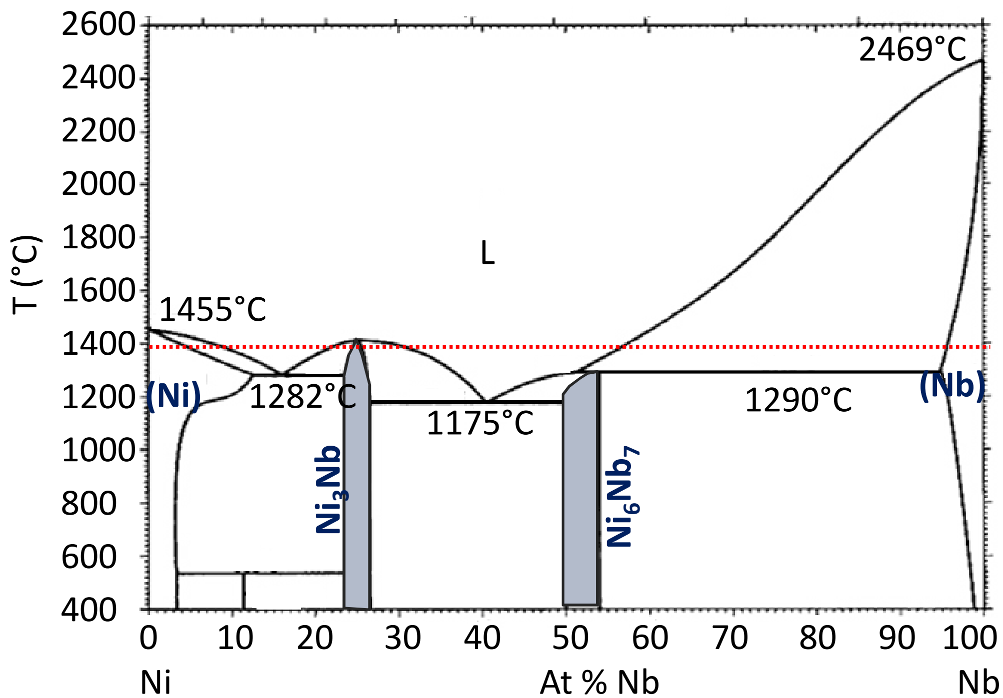
- Explore the TLP bonding method for joining UHTCs
- Basic study of the reactions between Nb-Ni and TMC
- Potential for strong refractory bonds

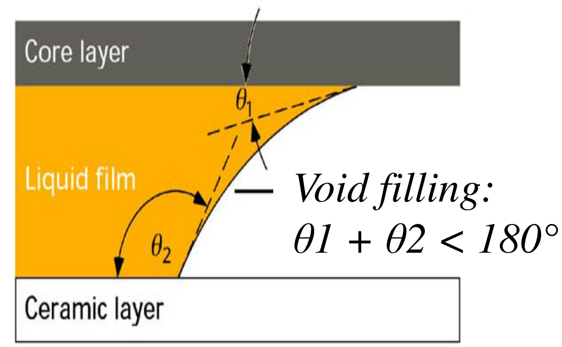
Nb-Ni transient liquid phase

A/B/A interlayer → Liquid formation a T<T_B → Liquid absorption/ reactive penetration

Ni-Nb E_1:1175°C
E_2:1290°C

Ni-based alloys show adequate *wetting* behavior towards UHTCs

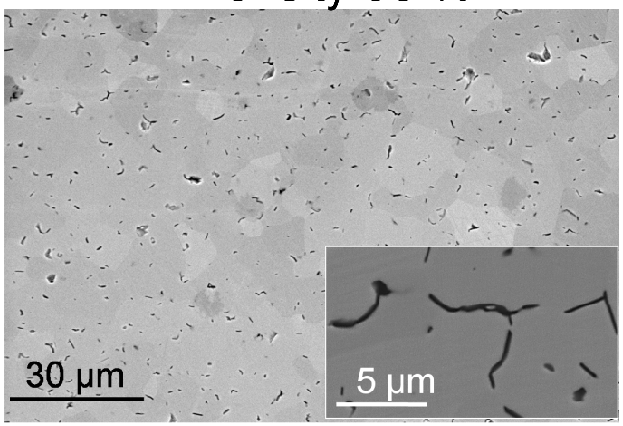




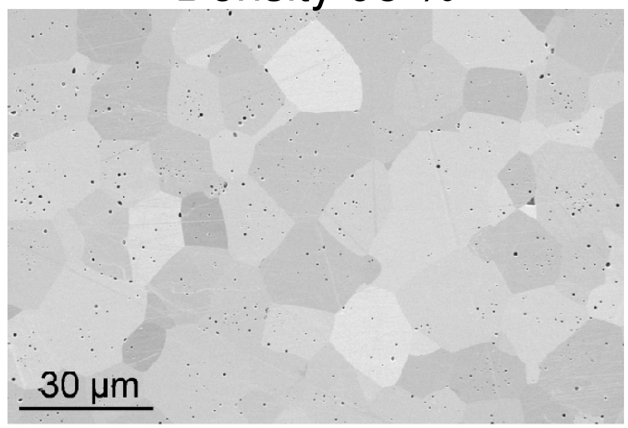
Starting materials

HP: 1900°C, 60 min, 30 MPa

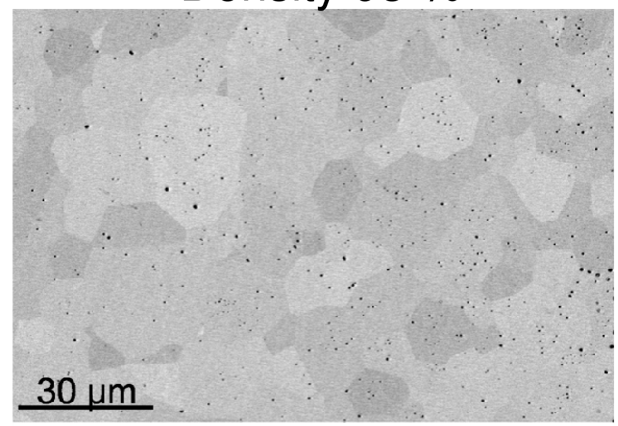
ZrC
Density 98 %



HfC
Density 98 %



TaC
Density 93 %

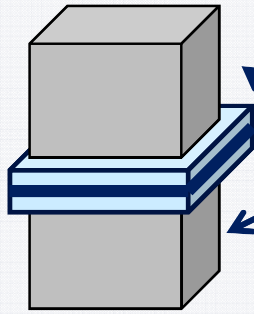


- Uniform microstructures
- Rounded or faceted relatively equiaxed grains
- Mean grain dimensions ~10 µm.
- Occasional oxide phases
- ZrC and HfC: <2 vol % of fine, closed porosity (pore size 0.3-1 µm)
- TaC residual porosity ≈5 vol %.
- ~8% of graphite in ZrC (≈1.5 wt% of free C in the starting powder)

Joint assembly

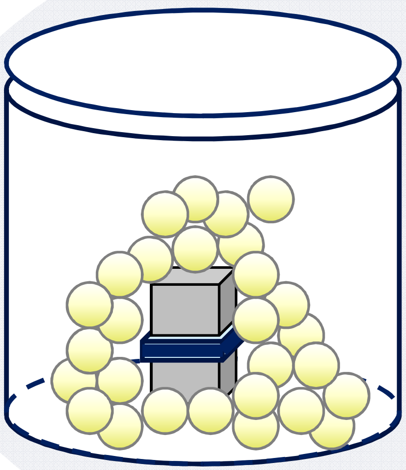
1

Ni/Nb/Ni
2/125/2 µm
platelet



Ceramic with
polished surface

2

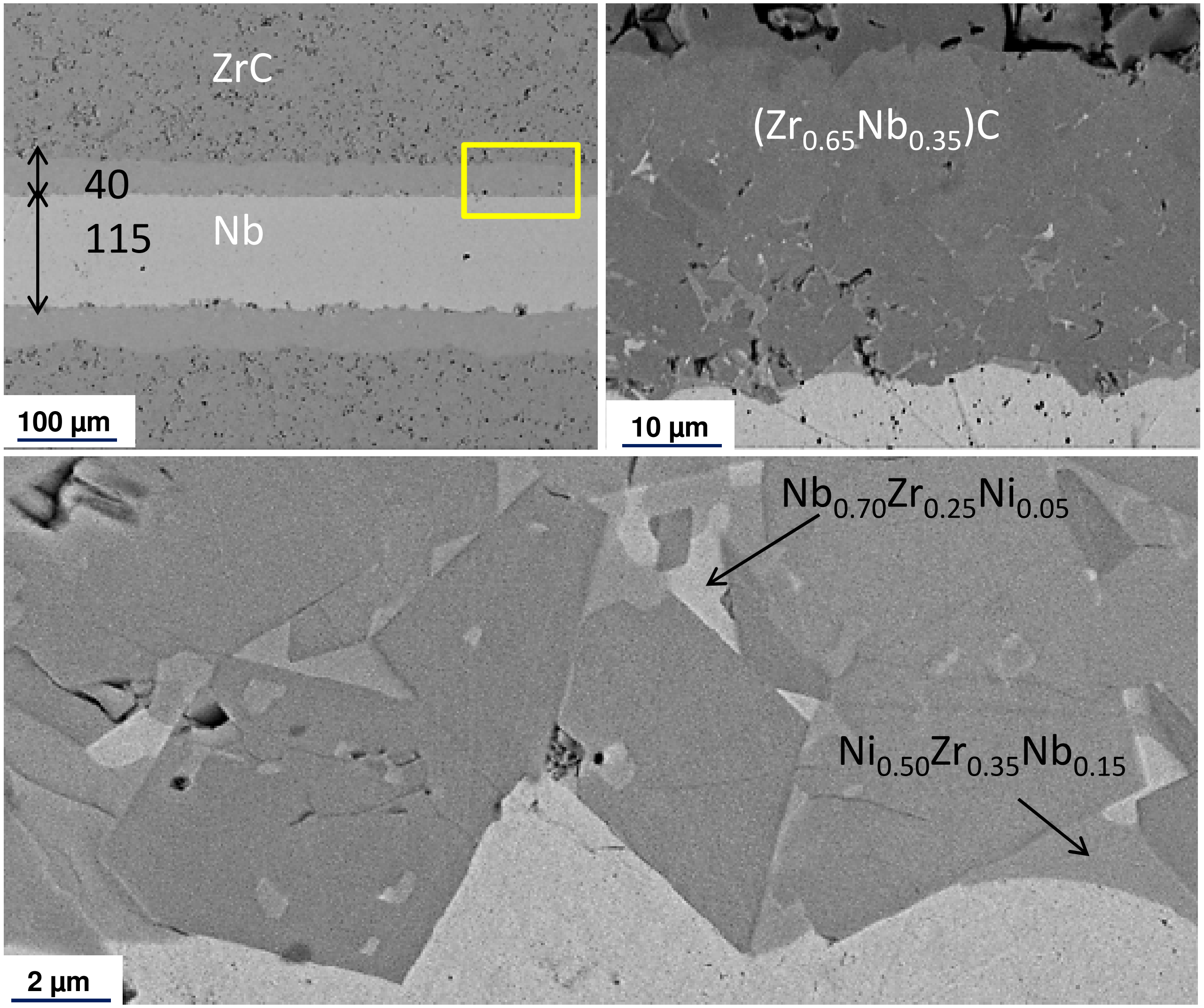


Al_2O_3 milling
media (<1.3 kPa)
& Al_2O_3 crucible

3

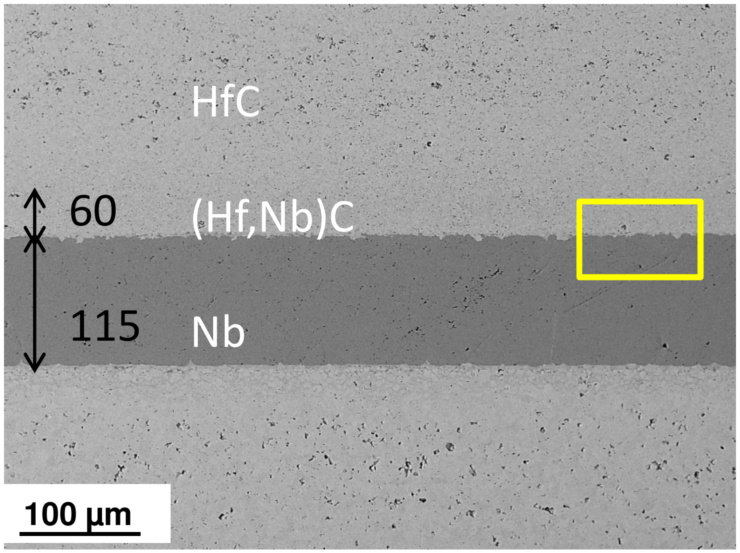
1400°C, 30', 10^{-4} Pa
4°C/min cooling,

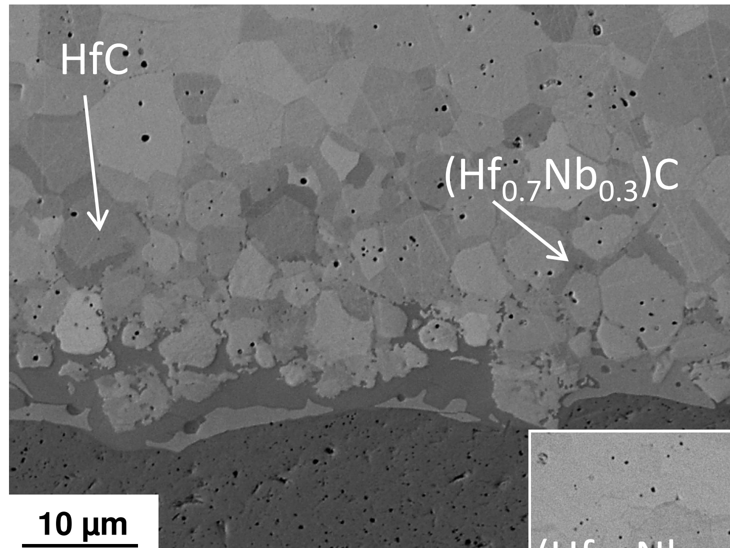
Monolithic ZrC



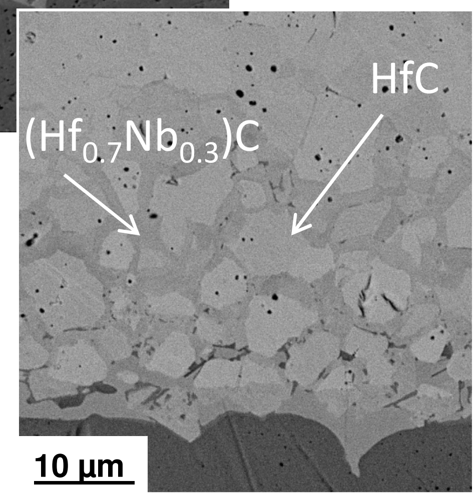
ZrC
(Zr,Nb)C
Nb-Zr-Ni
Ni-Zr-Nb

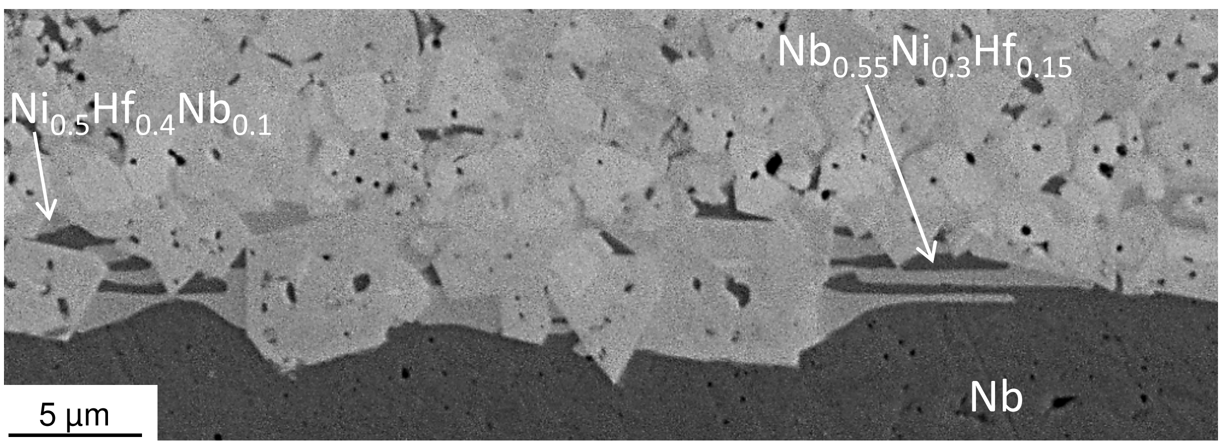
Monolithic HfC



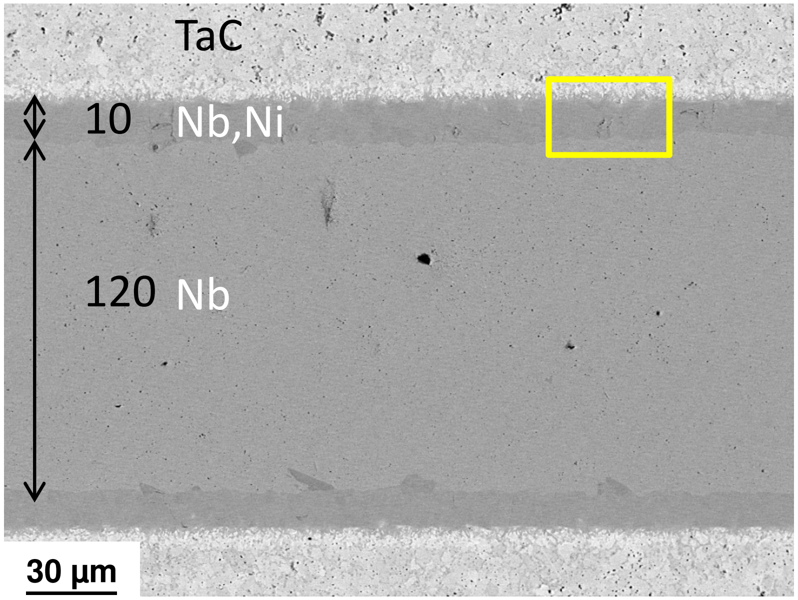


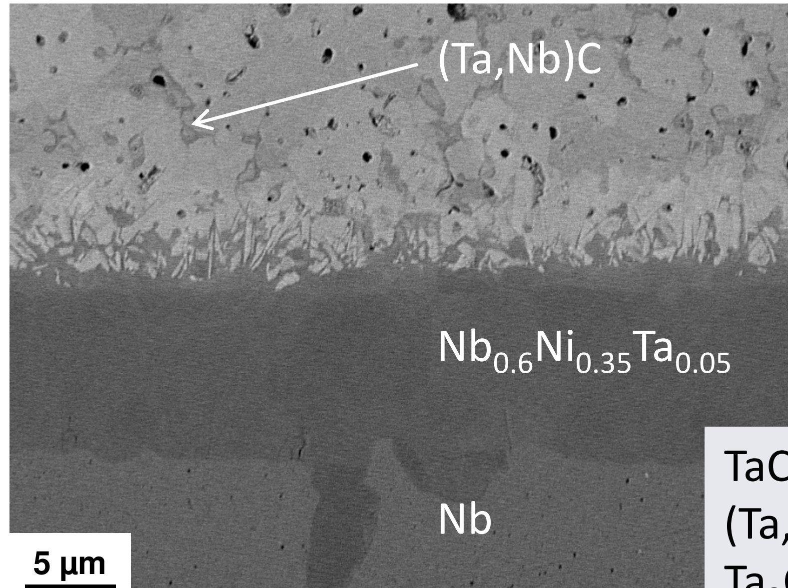
HfC
(Hf,Nb)C
Nb-Ni-Hf
Ni-Hf-Nb



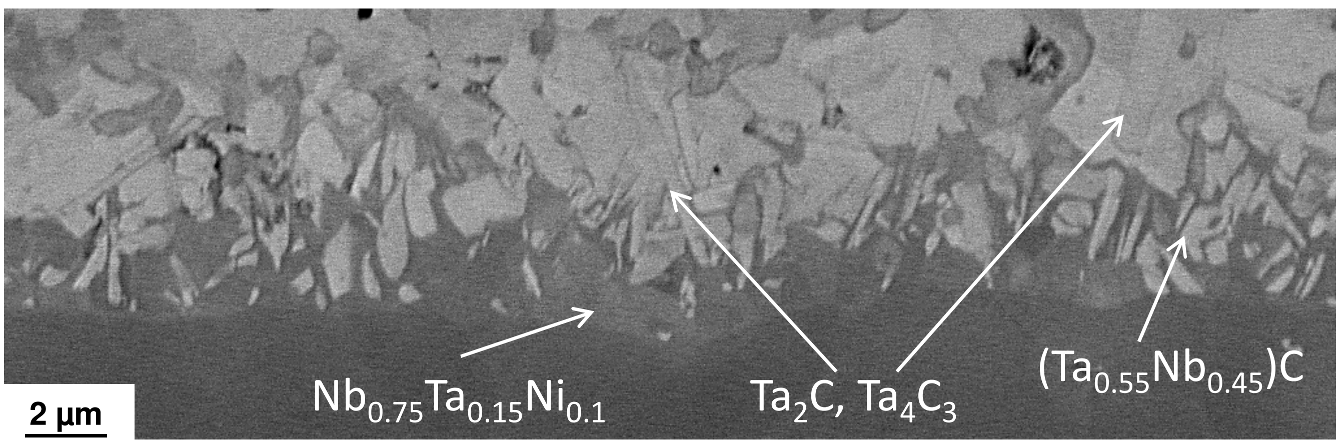


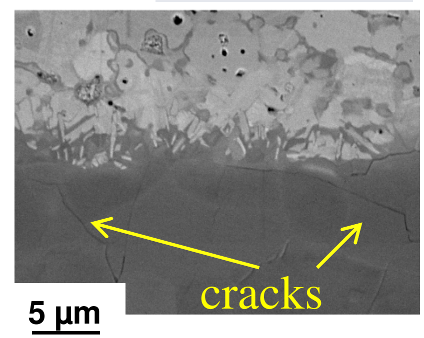
Monolithic TaC





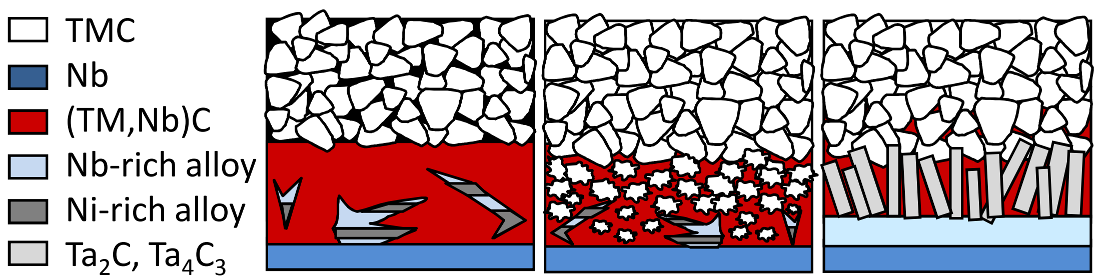
TaC
(Ta,Nb)C
Ta_2C, Ta_4C_3
Nb-Ta-Ni
Nb-Ni-Ta





Bonding mechanism

System	**ZrC**	**HfC**	**TaC**
New carbides	(Zr,Nb)C	(Hf,Nb)C	(Ta,Nb)C Ta_2C, Ta_4C_3
Metal alloys	NbZrNi NiZrNb	NbNiHf NiHfNb	NbTaNi NbNiTa

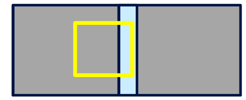


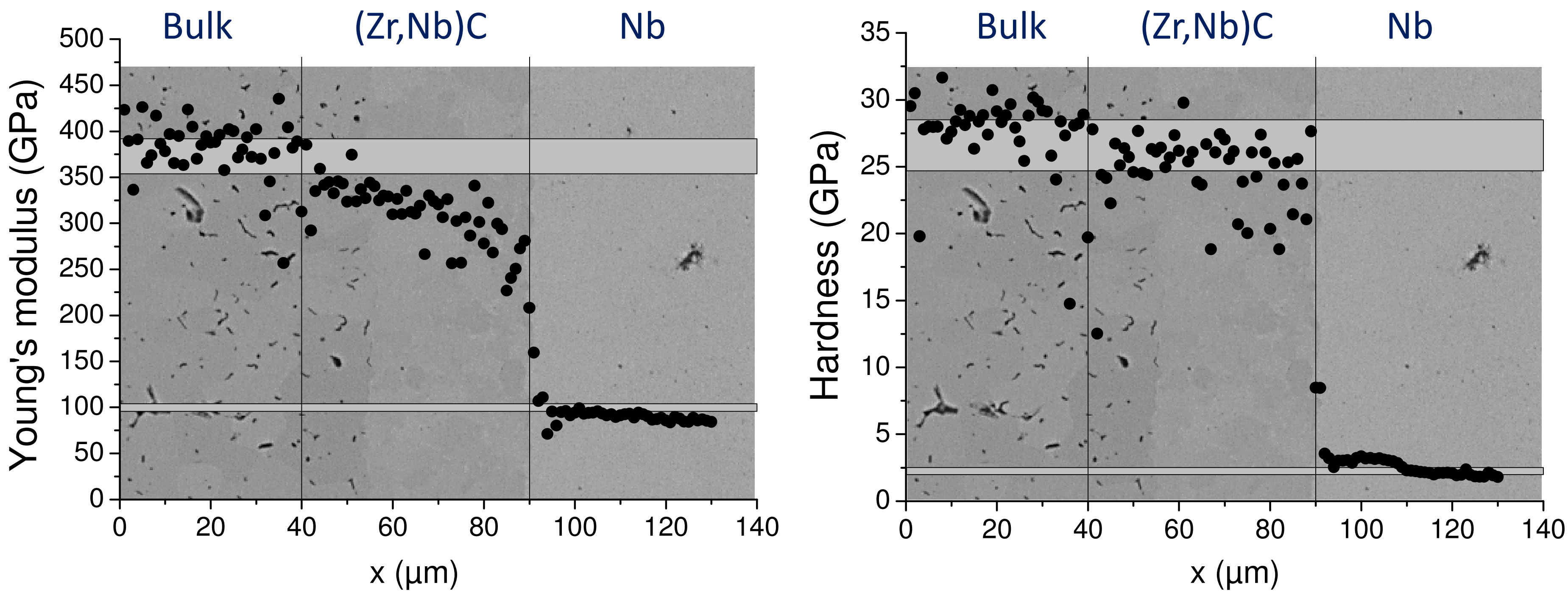
GENERAL BEHAVIOR:

- Nb-Ni eutectic→ 1176°C, 60 at%Ni
- Infiltration in the ceramic by capillary forces
- TMC dissolution in the liquid and precipitation of (TM,Nb)C
- Residuals of Ni_3Nb and Ni_6Nb_7 containing traces of the TM

- **ZrC**: homogeneous (Zr,Nb)C due to C impurities → Zr is highly soluble in Ni-Nb alloy and excess of C helps the carbo-reduction
- **HfC:** jagged HfC grains in (Hf,Nb)C → HfC is the least soluble in Nb-Ni alloy
- **TaC:** formation of Ta-rich carbides → TaC easily loses C giving rise to the Ta-rich carbides

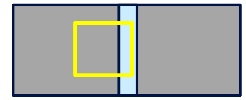
Nanoindentation on ZrC



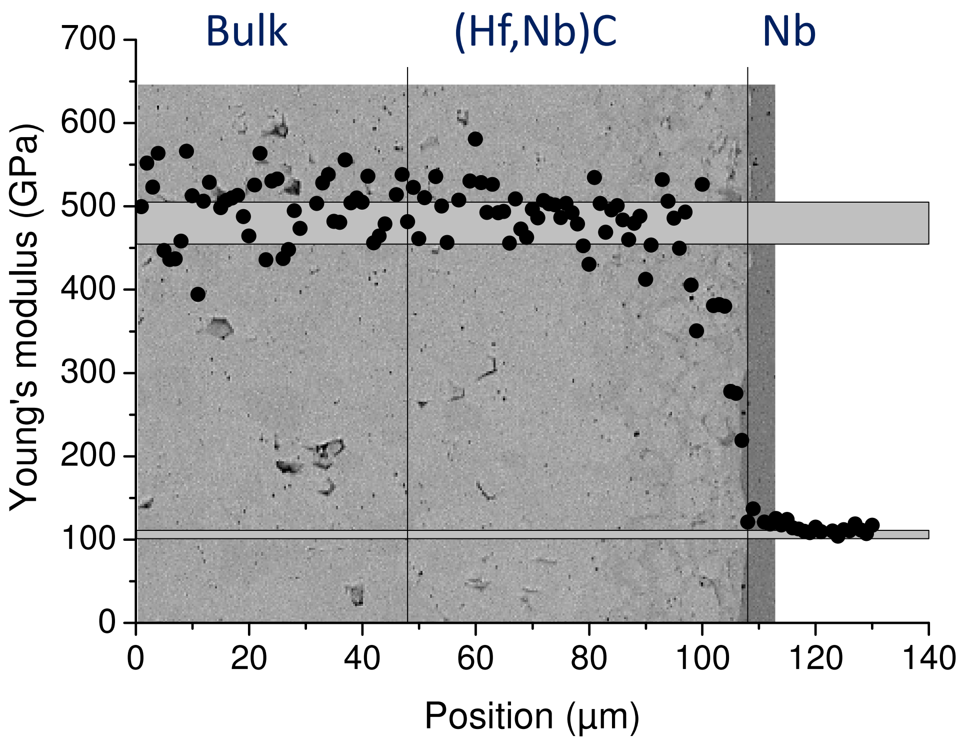


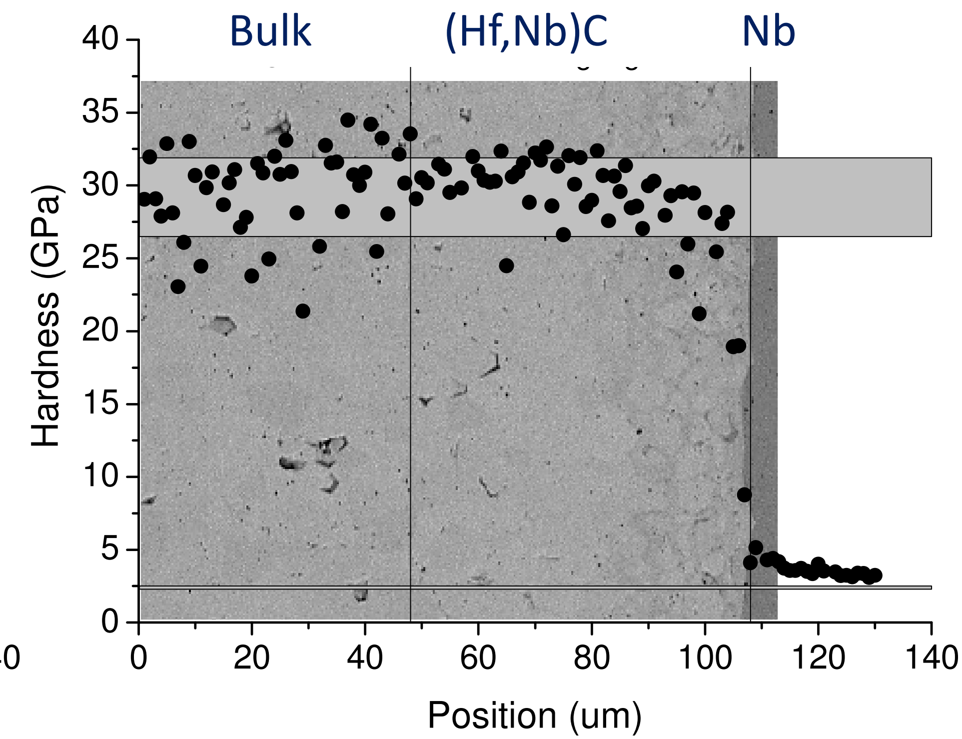
- Young's modulus slightly decreases in the (Zr,Nb)C region
- Almost no variation for hardness (H_{NbC}=18GPa)

Nanoindentation on HfC



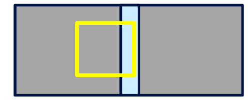
5 mN

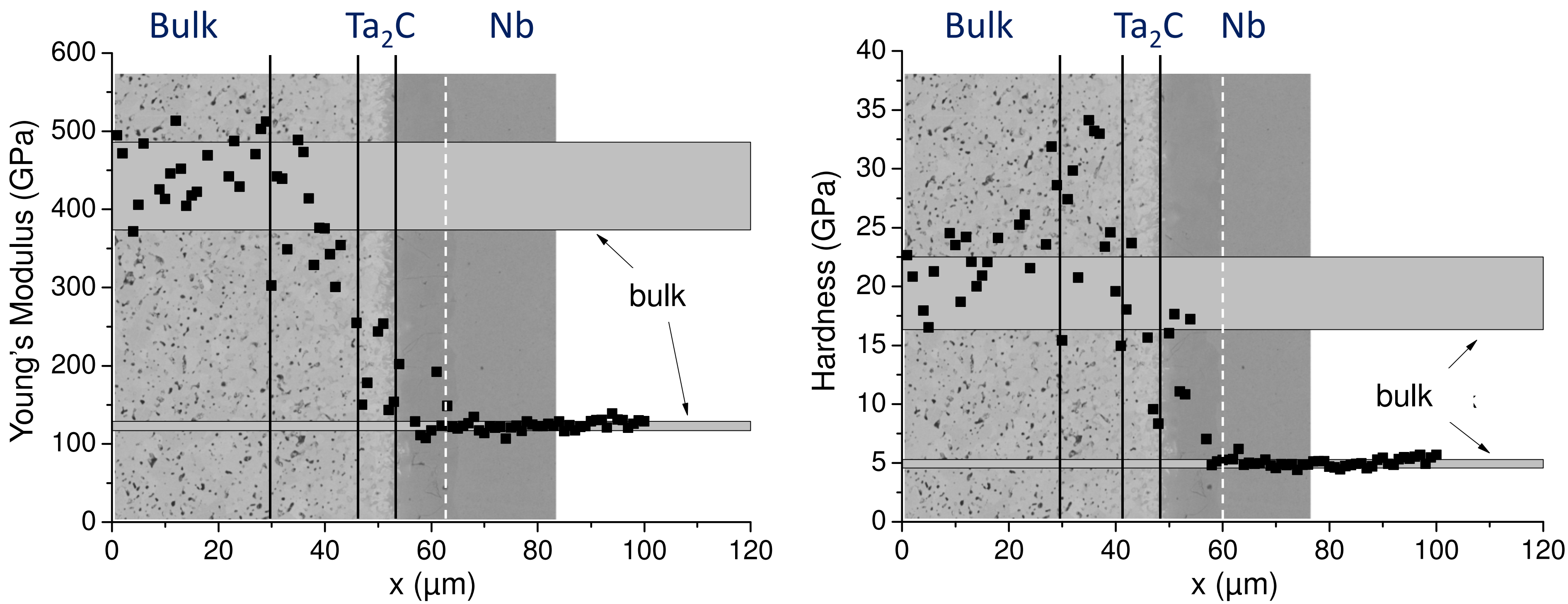




- Young’s modulus and hardness are basically not affected by the presence of (Hf,Nb)C

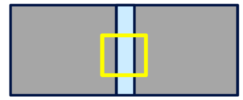
Nanoindentation on TaC



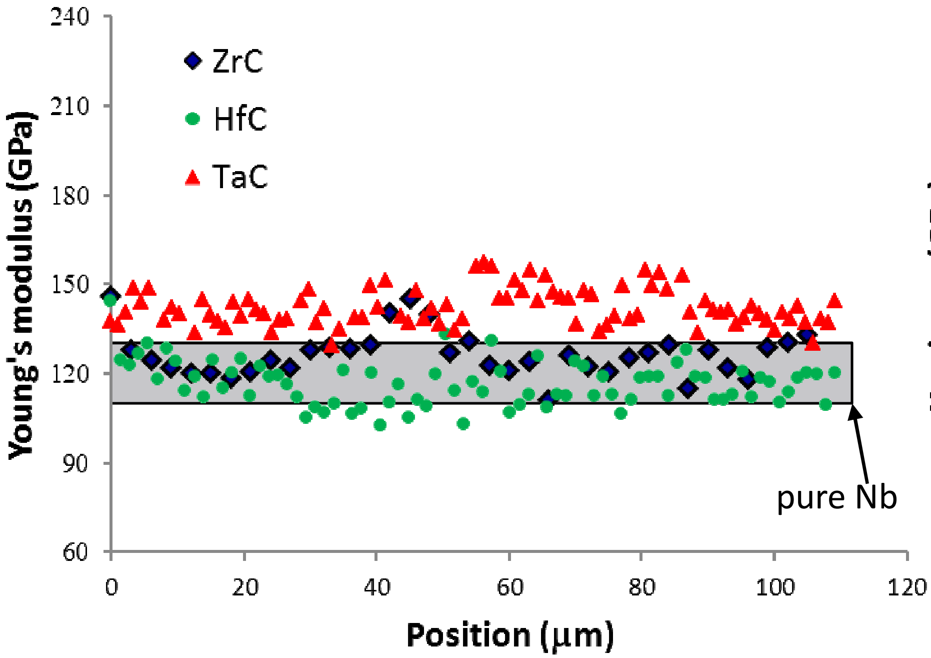


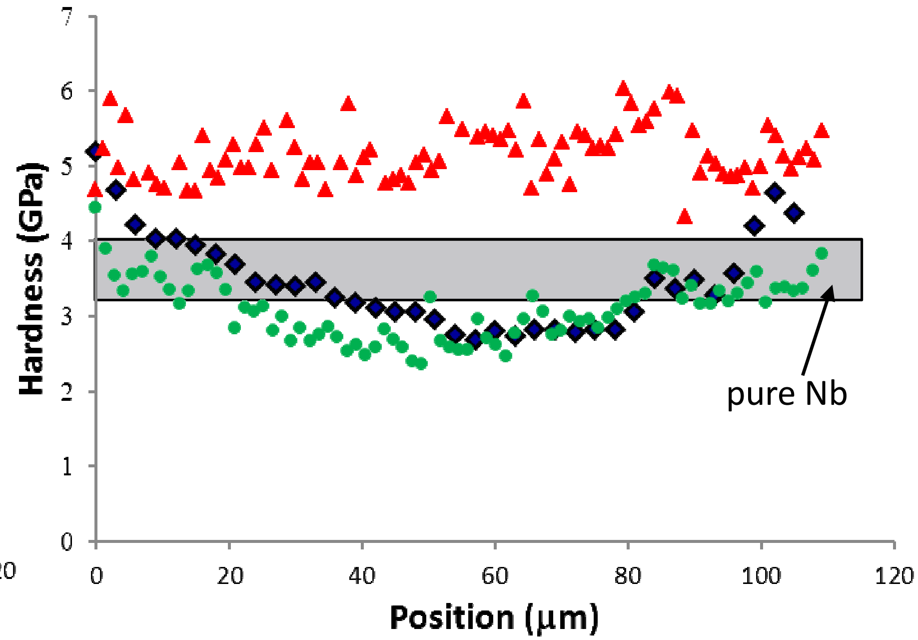
- Young's modulus decreases in the Ta-rich carbide region
- Hardness increases moving toward the joint owing to pores closure, but decreases in the Ta-rich carbide region (H_{Ta2C}=18GPa)

Nb in the joints



5 mN





- Nb is stiffer in TaC > ZrC = HfC; straight trend
- Nb is harder in TaC > ZrC > HfC; U-shaped trend owing to C enrichment
- Nb is hardest in TaC owing to C release from Ta-rich carbides formation

Conclusions & future perspectives

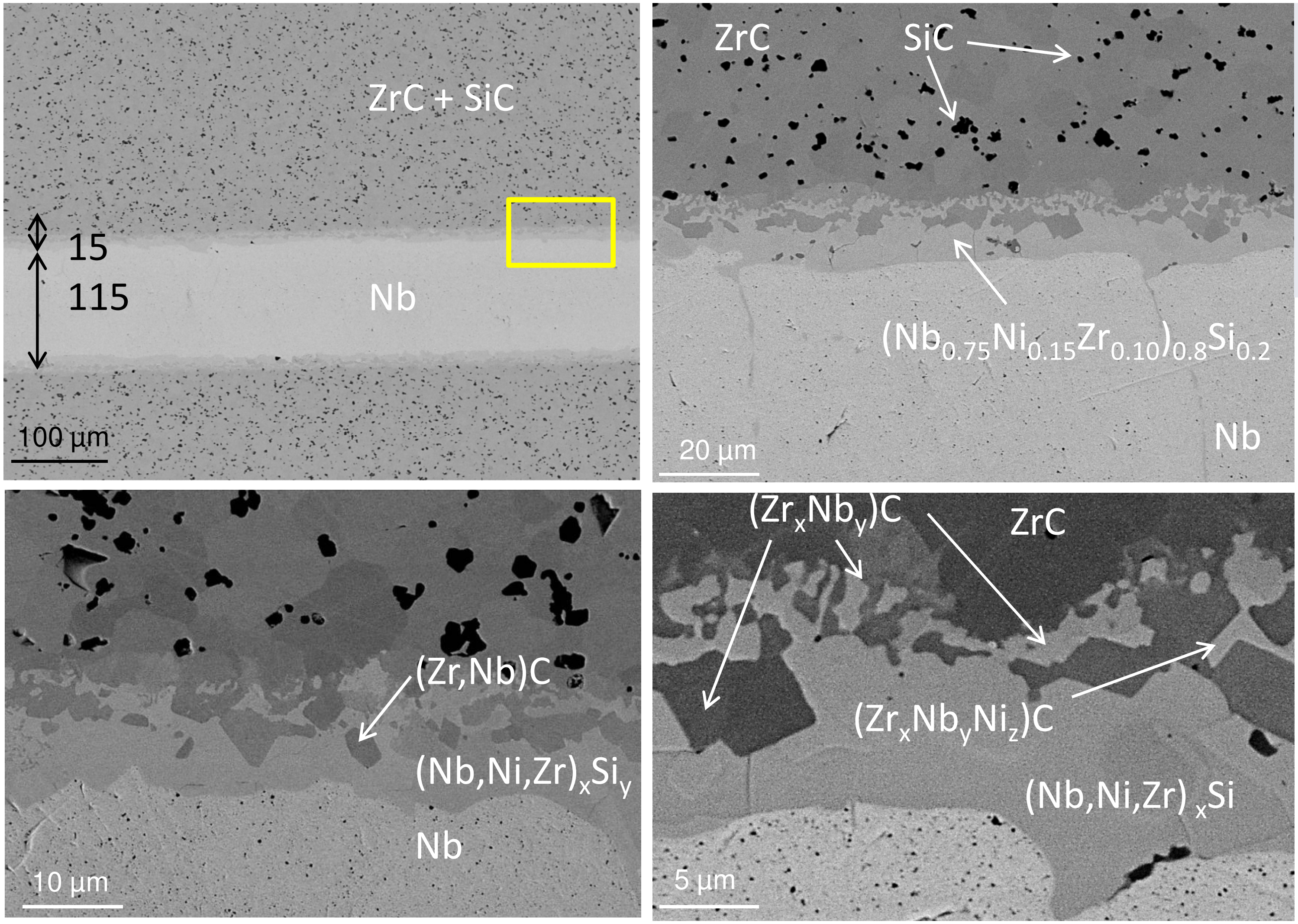
- Successful joining of **monolithic TM carbides** through transient liquid phase bonding at 1400°C with P<1.3 kPa.
- Good **compatibility** between TMC and Nb-Ni alloy, good cte match (~7.2 $10^{-6}K^{-1}$)
- **Nb-Ni melt** dissolves the carbides and forms (M,Nb)C. 2 residual metal alloys, Nb-rich and Ni-rich, are found close to the joint with traces of the TM
- ❖ **ZrC** displays a homogeneous reaction zone, thanks to C in the microstructure
- ❖ **HfC** is the least soluble in the Nb-Ni alloy
- ❖ **TaC** loses C evolving into Ta-rich carbides (Ta_2C, Ta_4C_3) → small cracks
- **Nanoindentation** showed no or little E&H difference from bulk and reaction zone in HfC, for ZrC and TaC E&H variations were observed.

IN PROGRESS…

- Preparation of specimen for 3-pt **strength** measurements.
- **$MoSi_2$ is detrimental** as it originates Nb,Ni-silicides forming cracks upon cooling.
- **SiC** present as secondary phase seems stable → promising for most UHTCs!

ZrC+15 $MoSi_2$*

*Most of $MoSi_2$ converted to SiC



ZrC+SiC
(Zr,Nb)C
Ta_2C
$(Nb,Ni,Zr)_xSi_y$
(Nb,Ni)C

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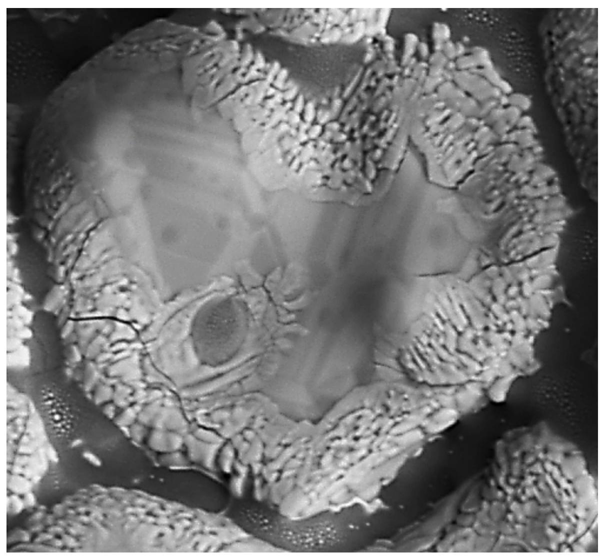

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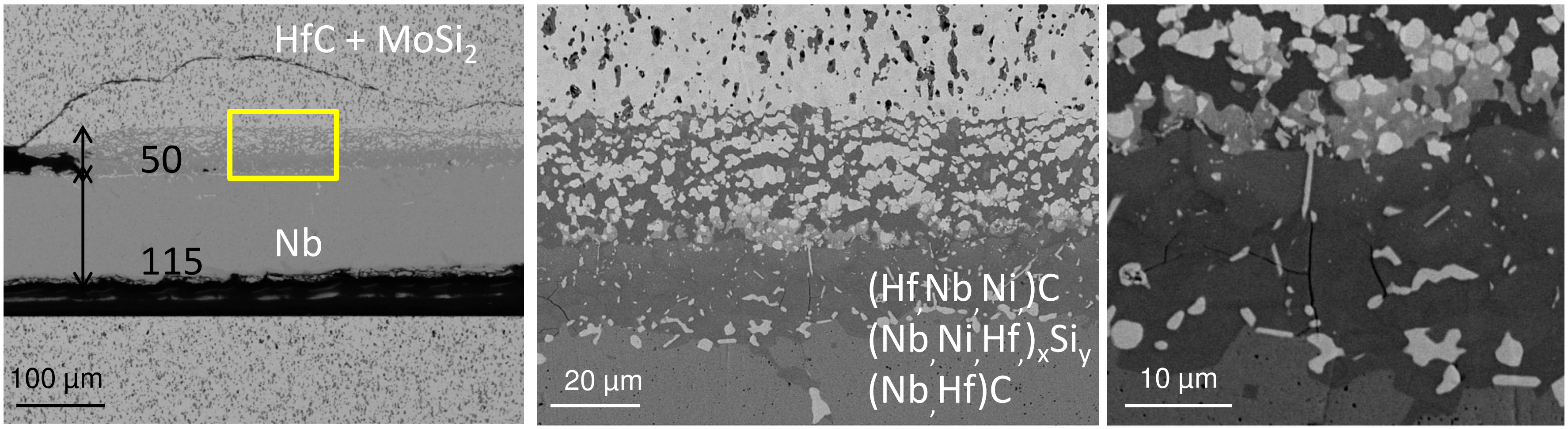
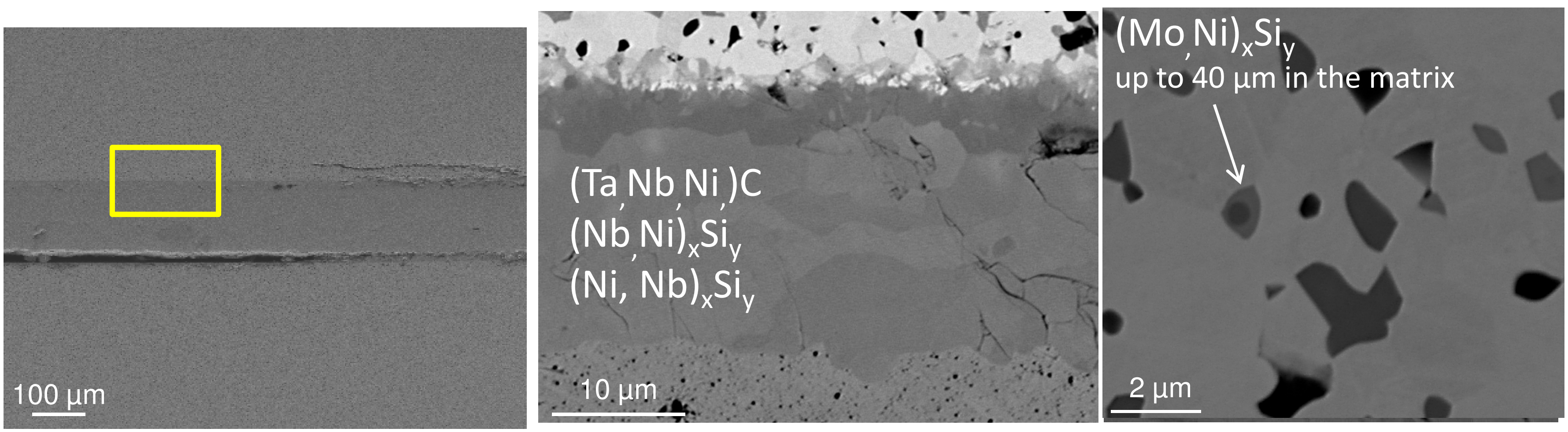
HfC/TaC + 15 $MoSi_2$

$HfC+15v\ MoSi_2$



$TaC+15v\ MoSi_2$



Starting matrices