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Joining of UHTC Boride Composites using Metallic Interlayer

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Kyushu University 2011
100th Anniversary

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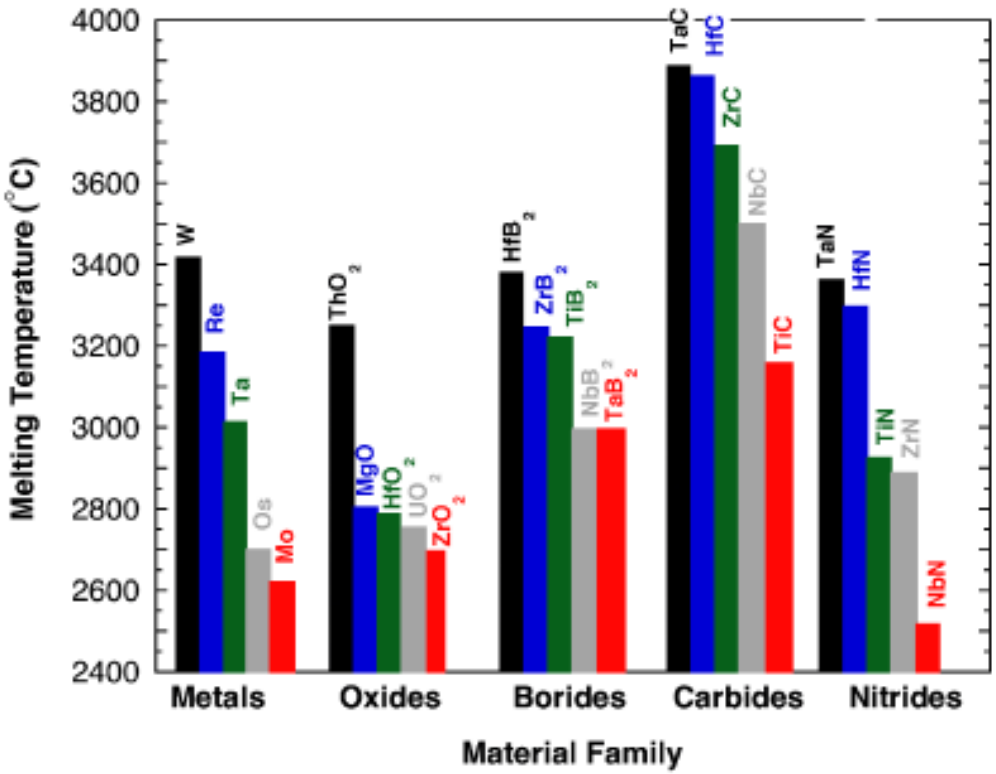
Background - Ultra High Temperature Ceramics



Several carbides, borides and nitrides of the group IV and V metals are considered UHTCs based on melting temperatures in excess of 3000 ℃ and other excellent properties.

William G. Fahrenholtz et al. : J. Am. Ceram. Soc., 90 [5] 1347–1364 (2007)

Objective

The fabrication of large complex-shape parts will be very challenging, and for many applications, the practical use of these materials in complex structures will require effective methods of joining.

Without joining methods, the full promise of these materials will not be realized.

Outline

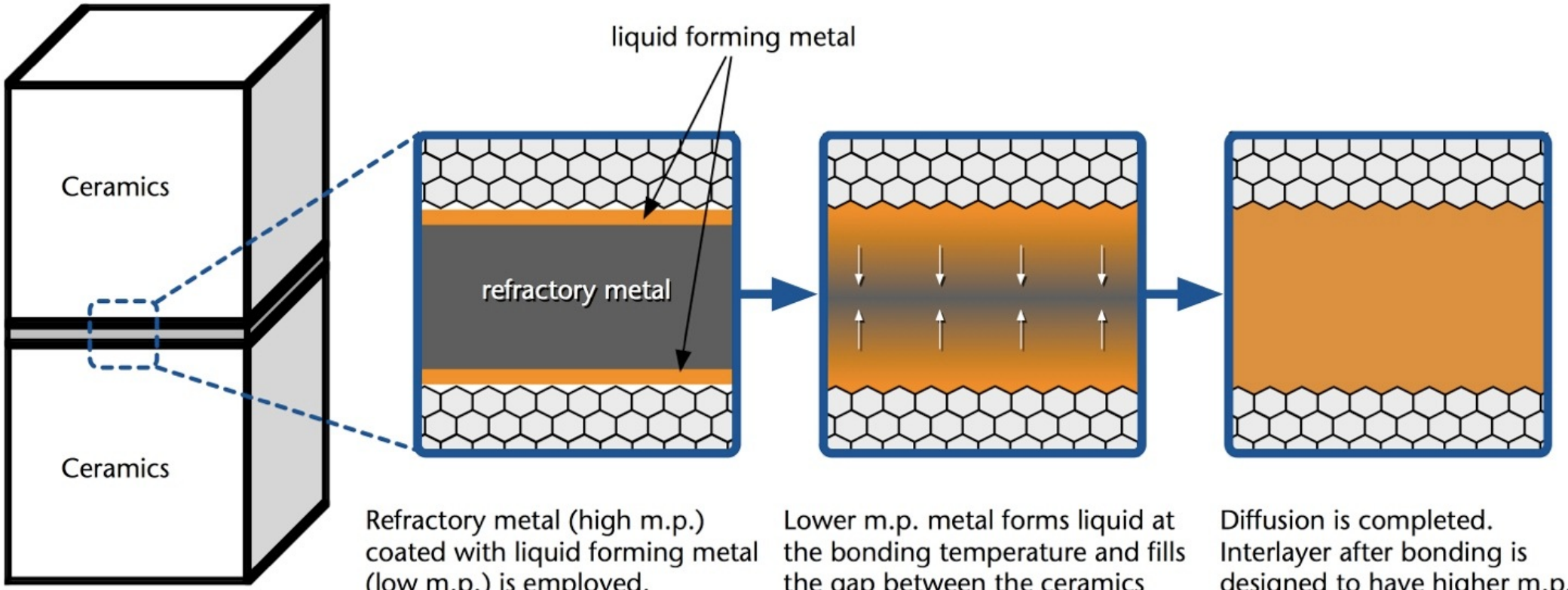
1. Transient Liquid Phase (TLP) Bonding of HfB_2 Composite

2. Joining of ZrB_2 Composite using Reactive Metal

Transient Liquid Phase (TLP) Bonding of HfB_2 Composite

Journal of Materials Science 47 [24] pp.8454-8463 (2012)
Journal of Materials Science 49 [2] pp.654-664 (2014)

TLP Bonding of Ceramics

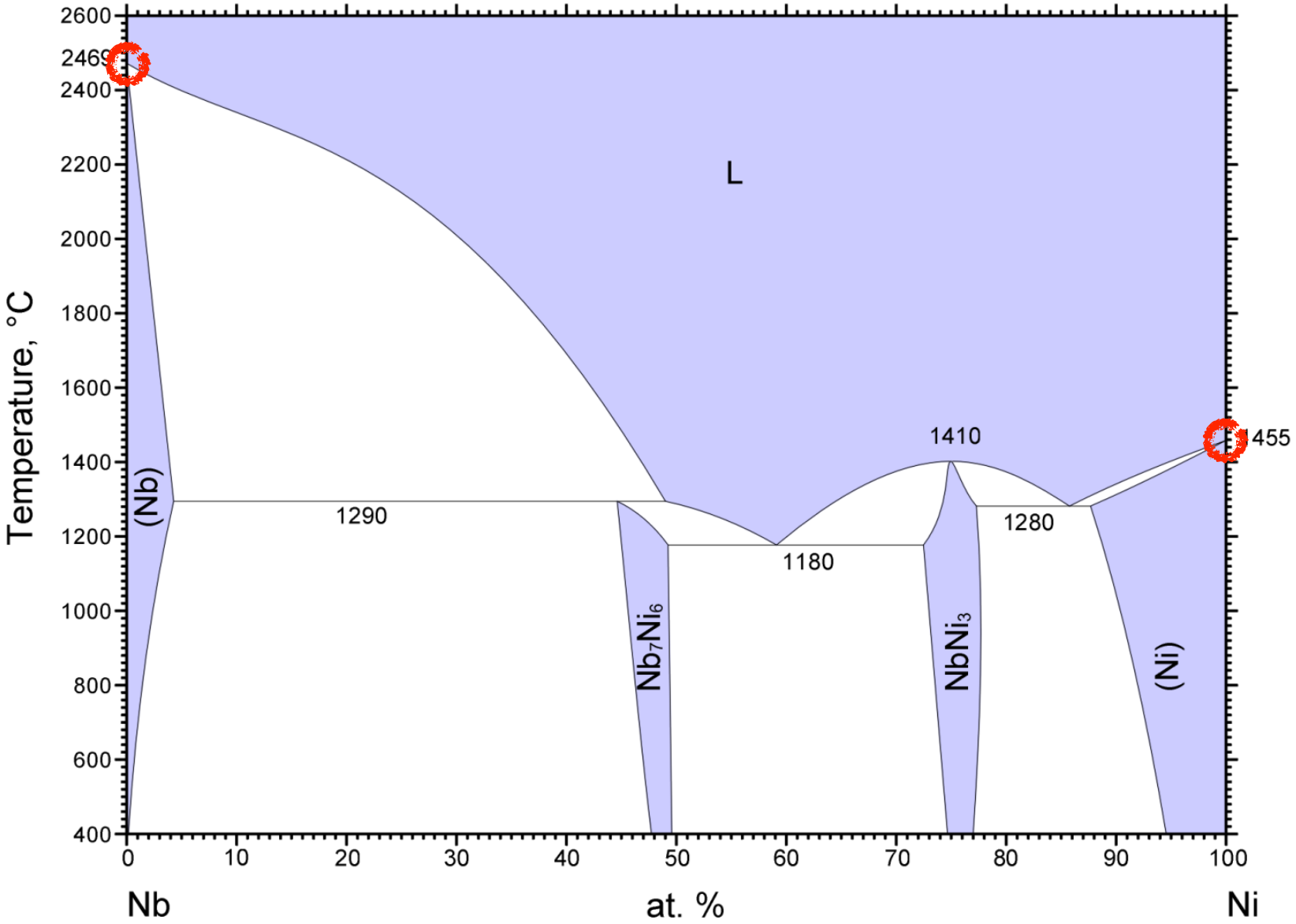


Refractory metal (high m.p.) coated with liquid forming metal (low m.p.) is employed.

Lower m.p. metal forms liquid at the bonding temperature and fills the gap between the ceramics and the inserted layer. The liquid forming metal rapidly diffuses into the core-layer, resulting in solidification.

Diffusion is completed. Interlayer after bonding is designed to have higher m.p. than the bonding temperature.

The Phase Diagram Nb-Ni System



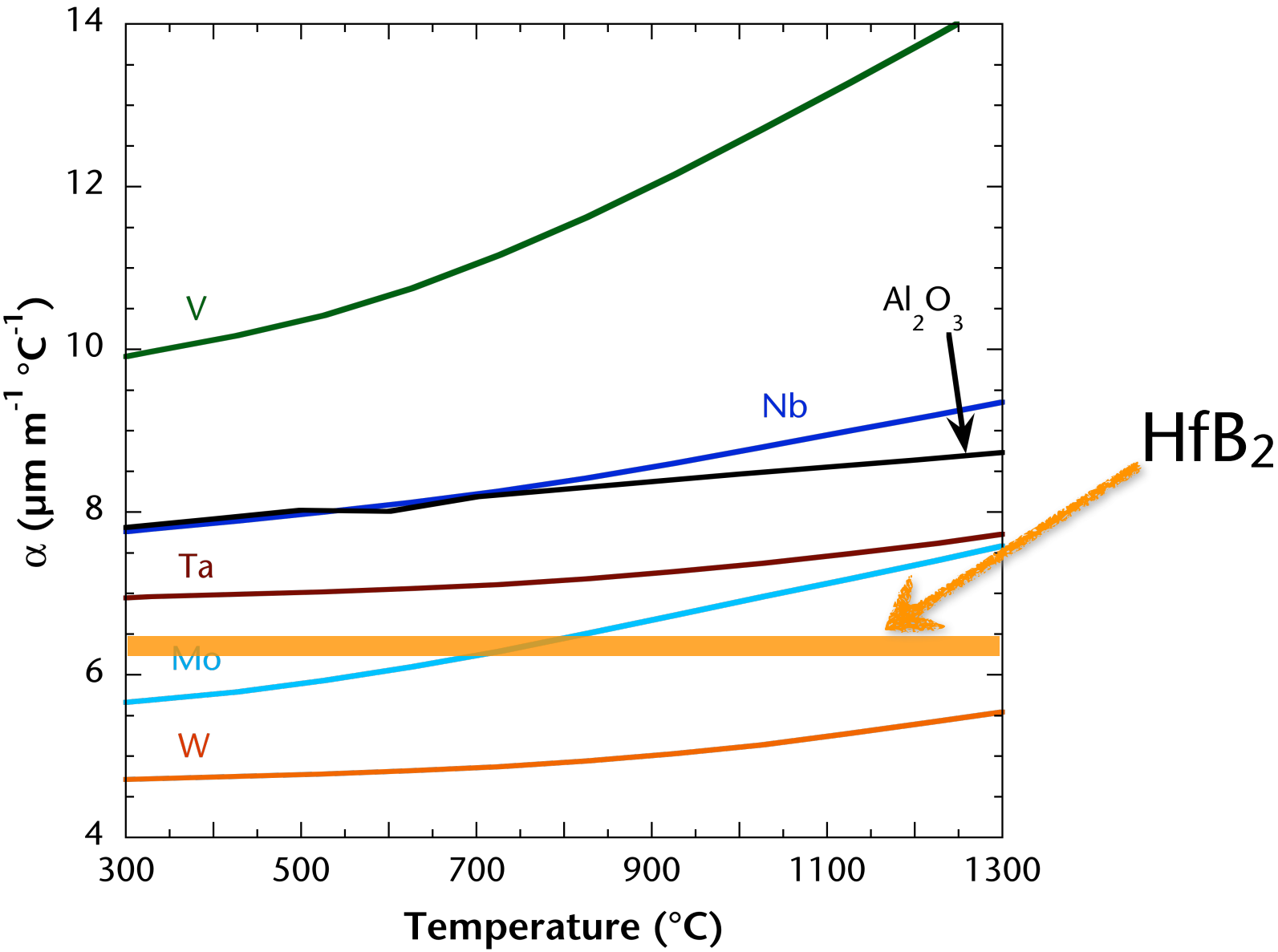
Qualifications for TLP Interlayer

1. CTE Matching

2. Liquid Phase

3. Wettability

CTE Matching



Qualifications for TLP Interlayer

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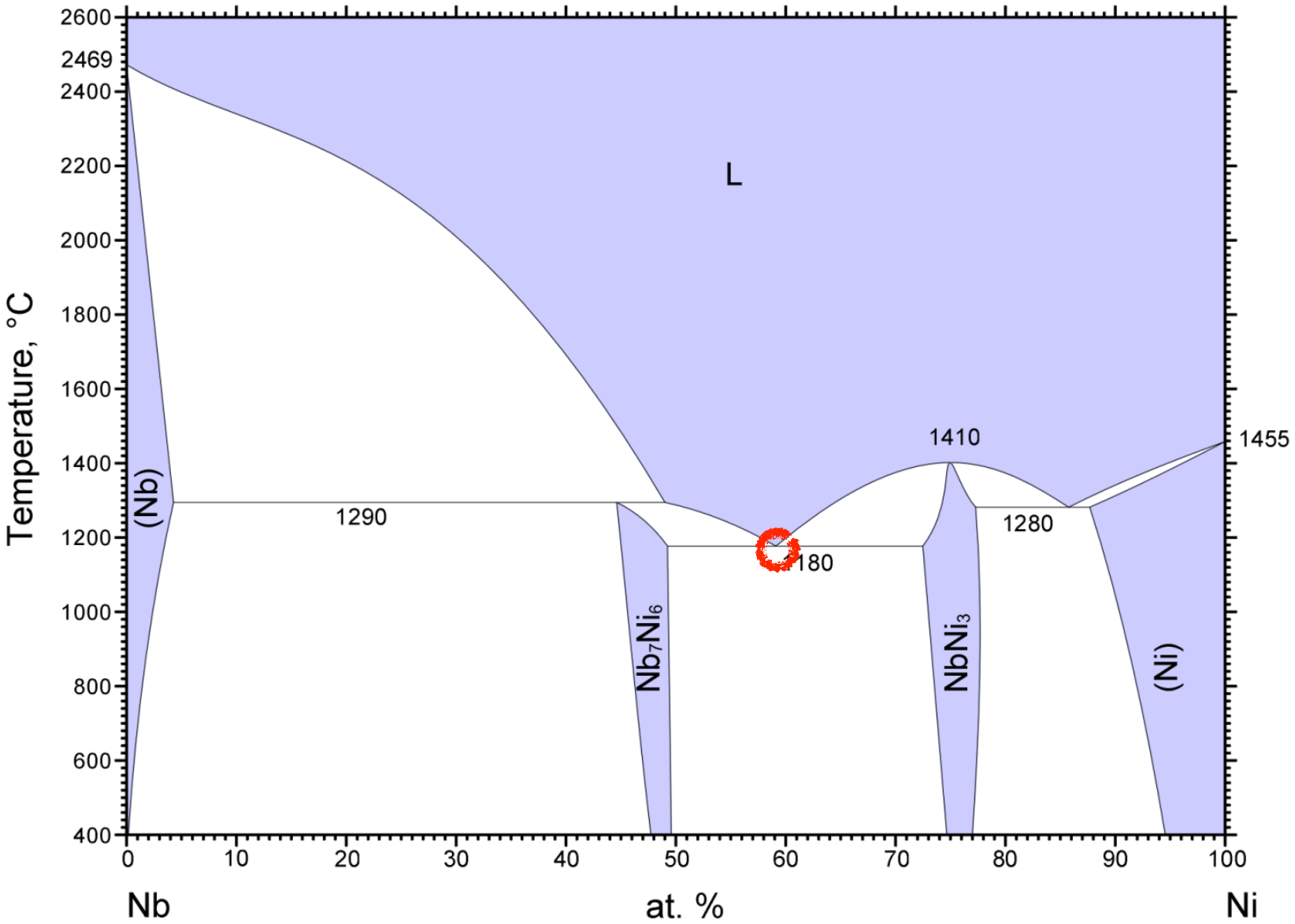
The Phase Diagram Nb-Ni System



Qualifications for TLP Interlayer

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2. Liquid Phase ✓
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Experimental Procedure of HfB_2 Composite Processing

Powder mixing

HfB_2 (-325 mesh, 99.9%),
+10vol%$MoSi_2$ (-325 μm, 99.9%)
ZrO_2 balls+PE pot for 24 h

Molding

23 mmΦ
300 MPa CIP

Sintering

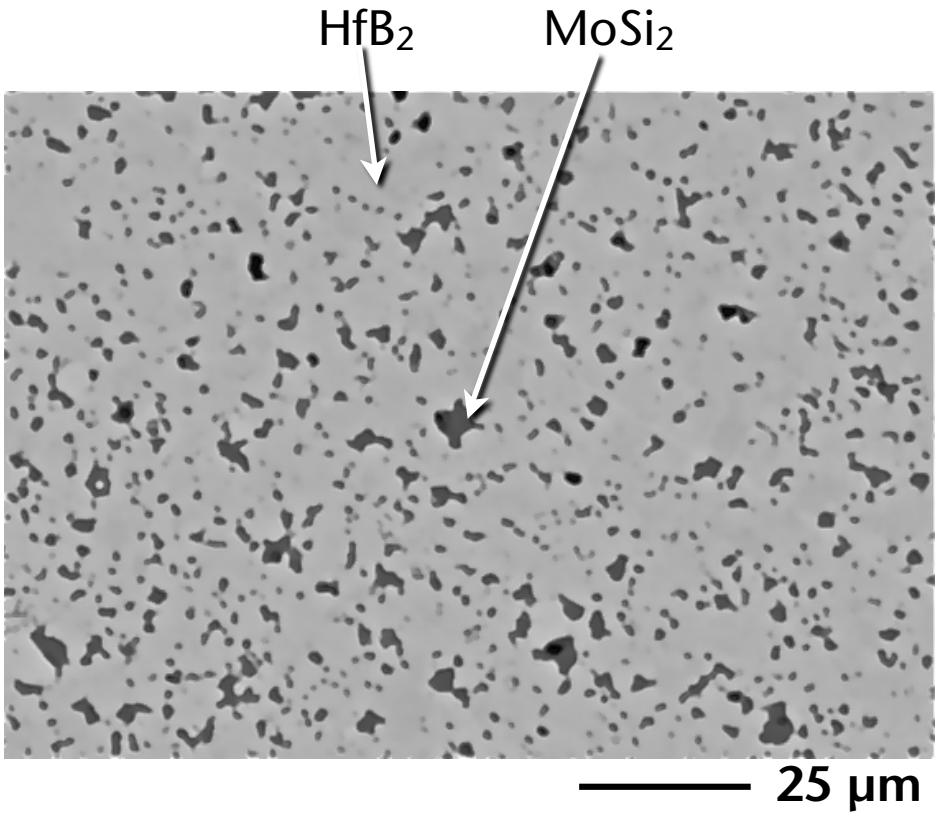
1950 ℃ for 60 min
Under 0.1 MPa Ar

Polishing

with diamond slurries

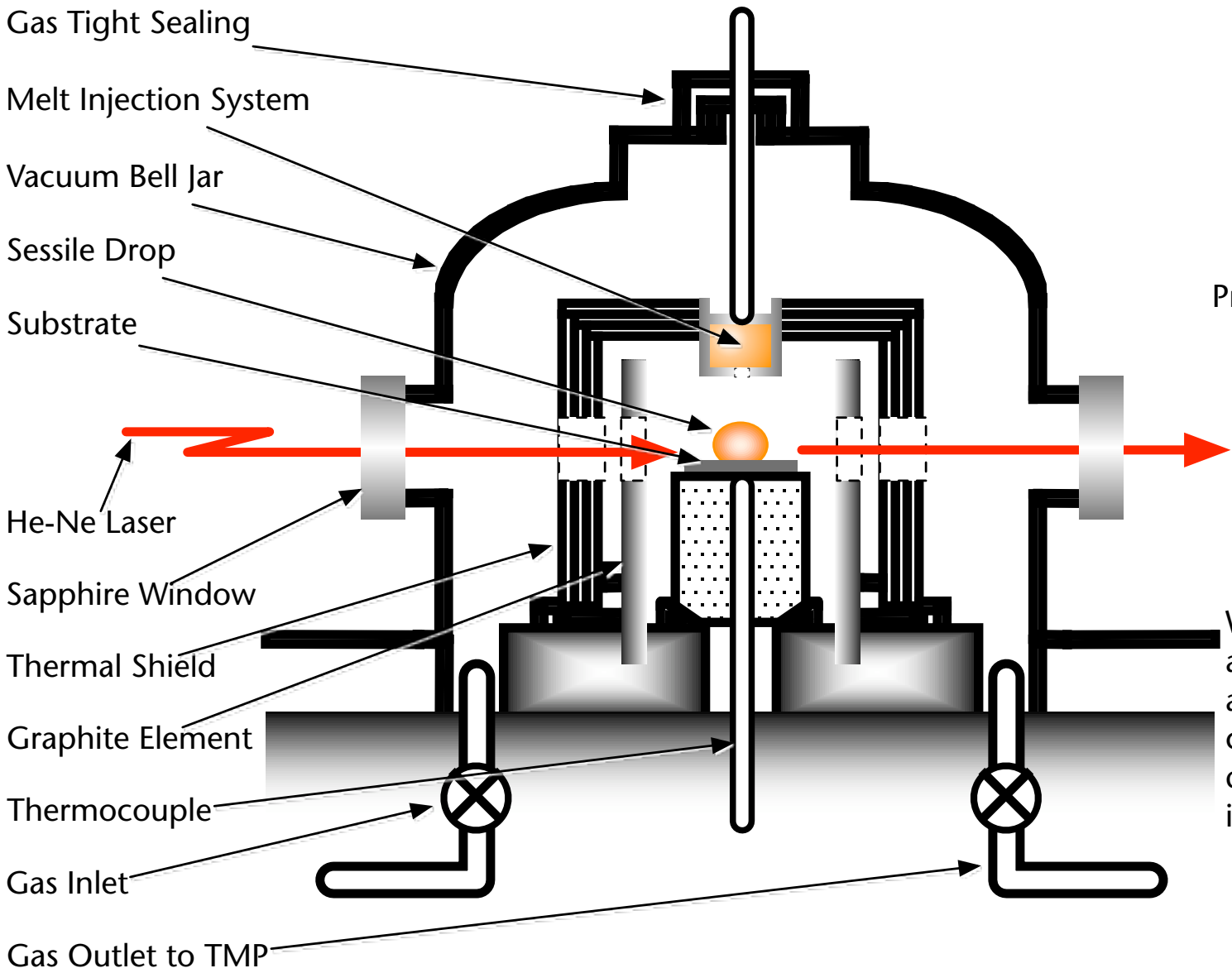
Wetting test

BSE image of HfB_2-10vol%$MoSi_2$ Polished Section

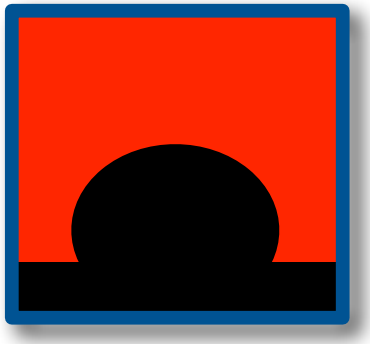


98% relative density

Schematic of Wetting Furnace



Projection Image on CCD



Wettability (contact angle) at elevated temperature is accurately evaluated by curve-fitting method on digitalized projection image of melt.

Experimental Procedure of Wetting Test

Heating at 15 K/min

under 10^{-3} Pa vacuum

Taking pictures

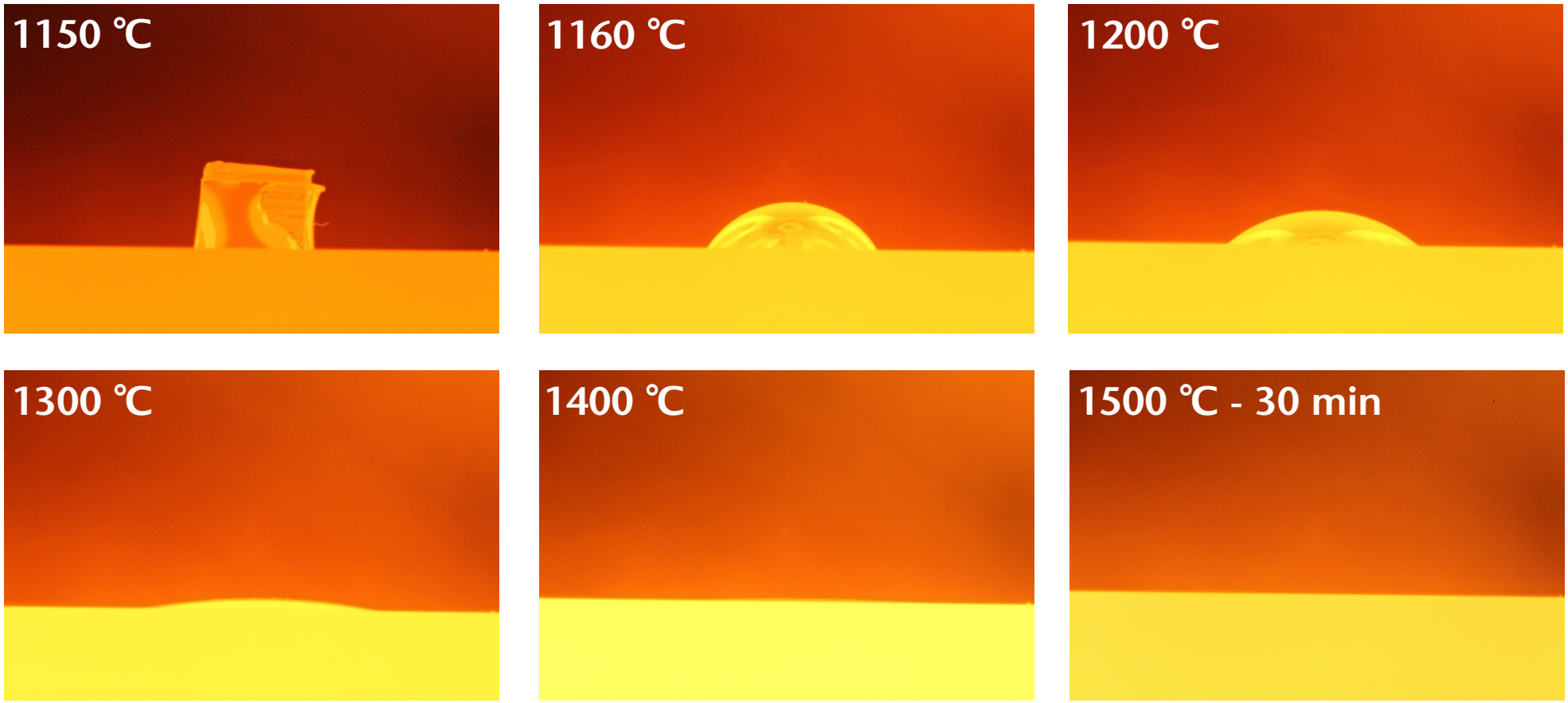
20 ℃ interval on heating

Holding at 1500 ℃

Taking pictures

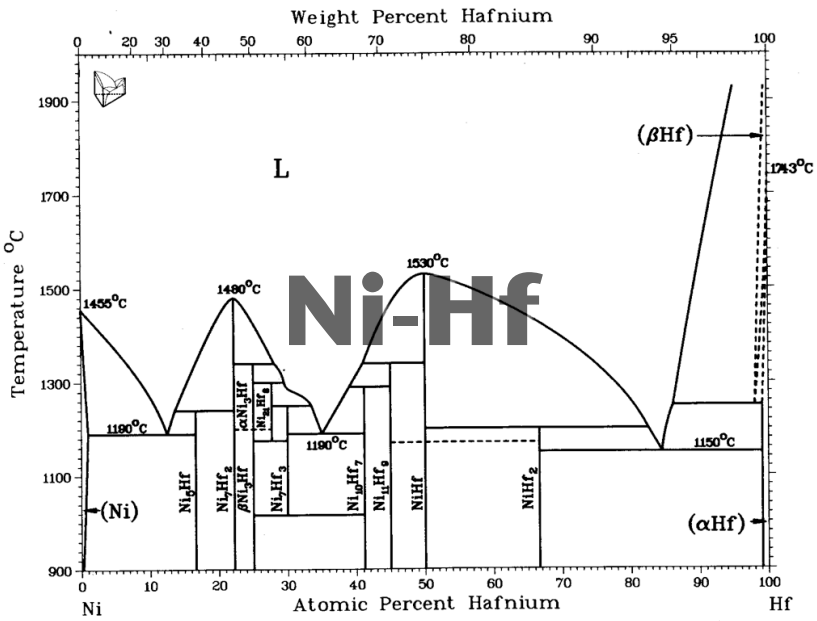
60 sec interval on holding

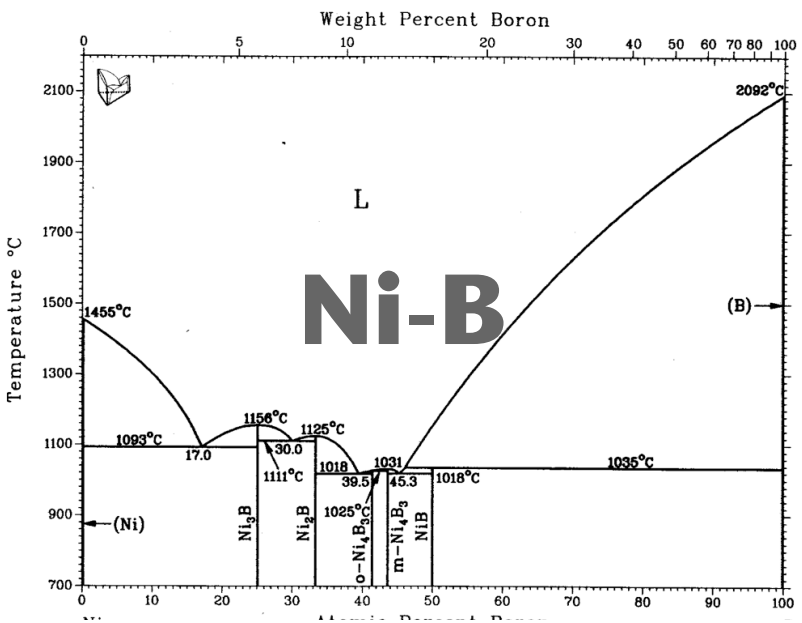
HfB_2-10vol%$MoSi_2$ / pure Ni

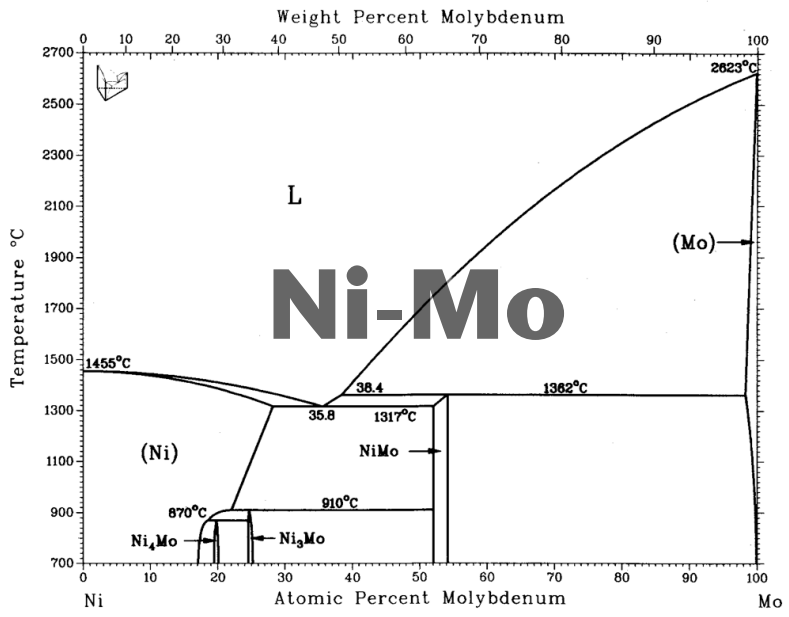


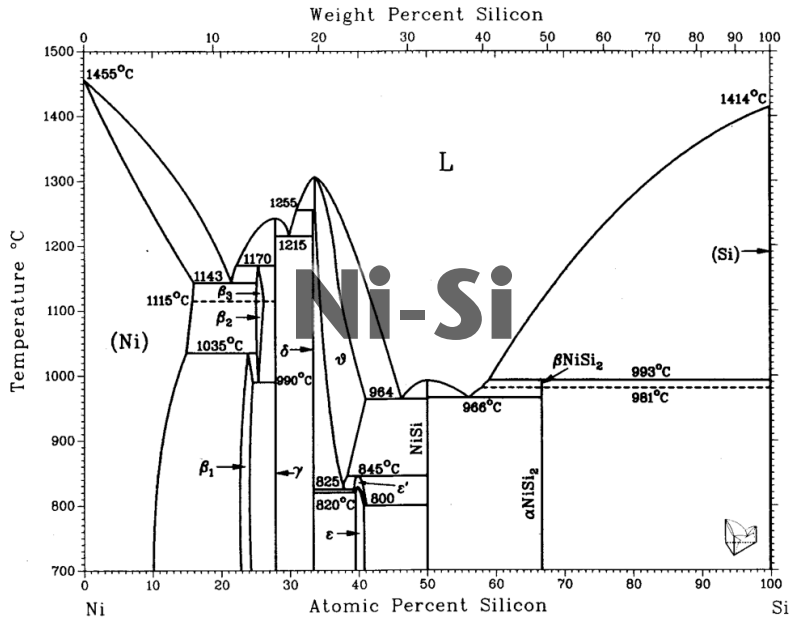
Ni started melting at 1150 °C !

Phase Diagrams of Ni based Binary

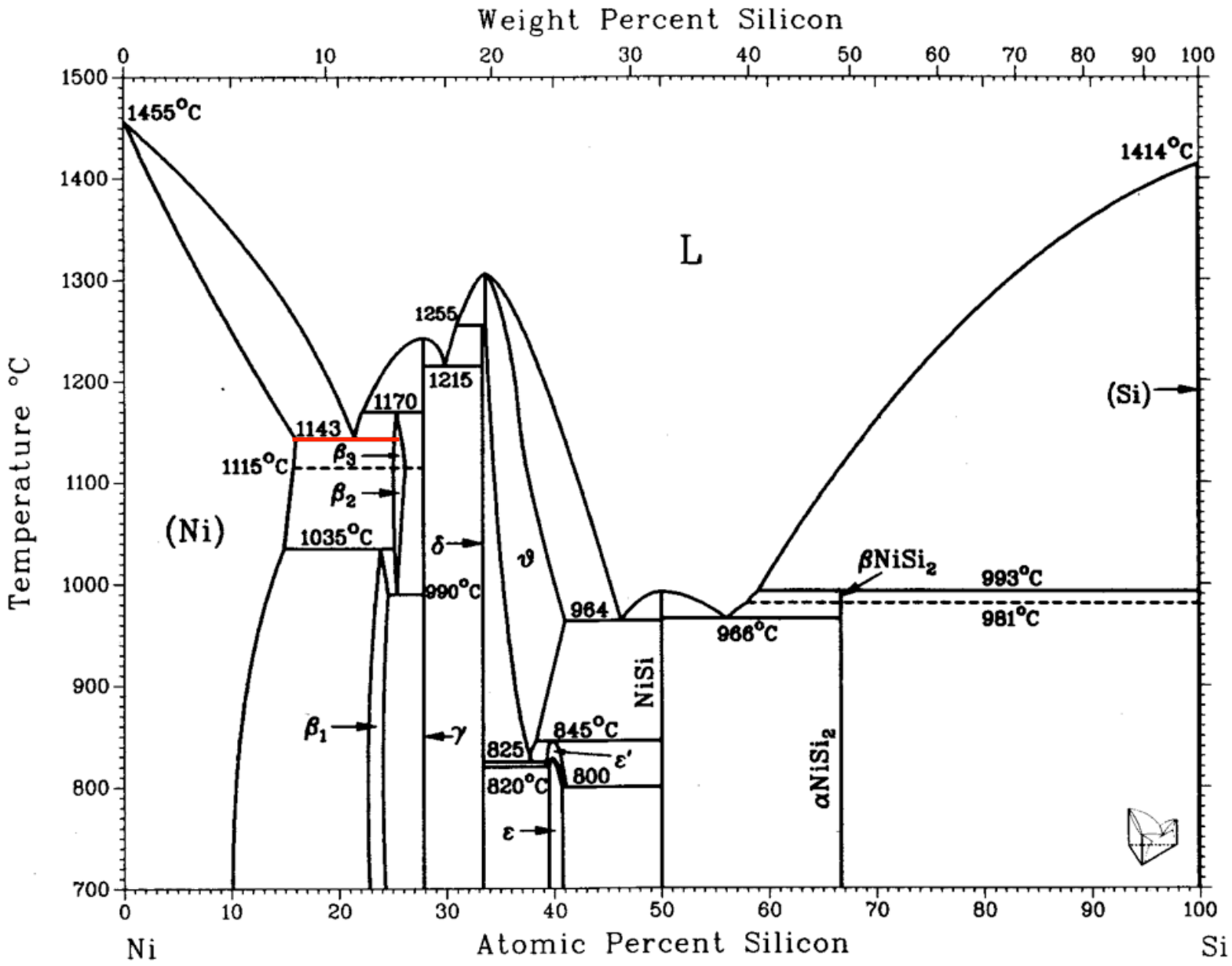




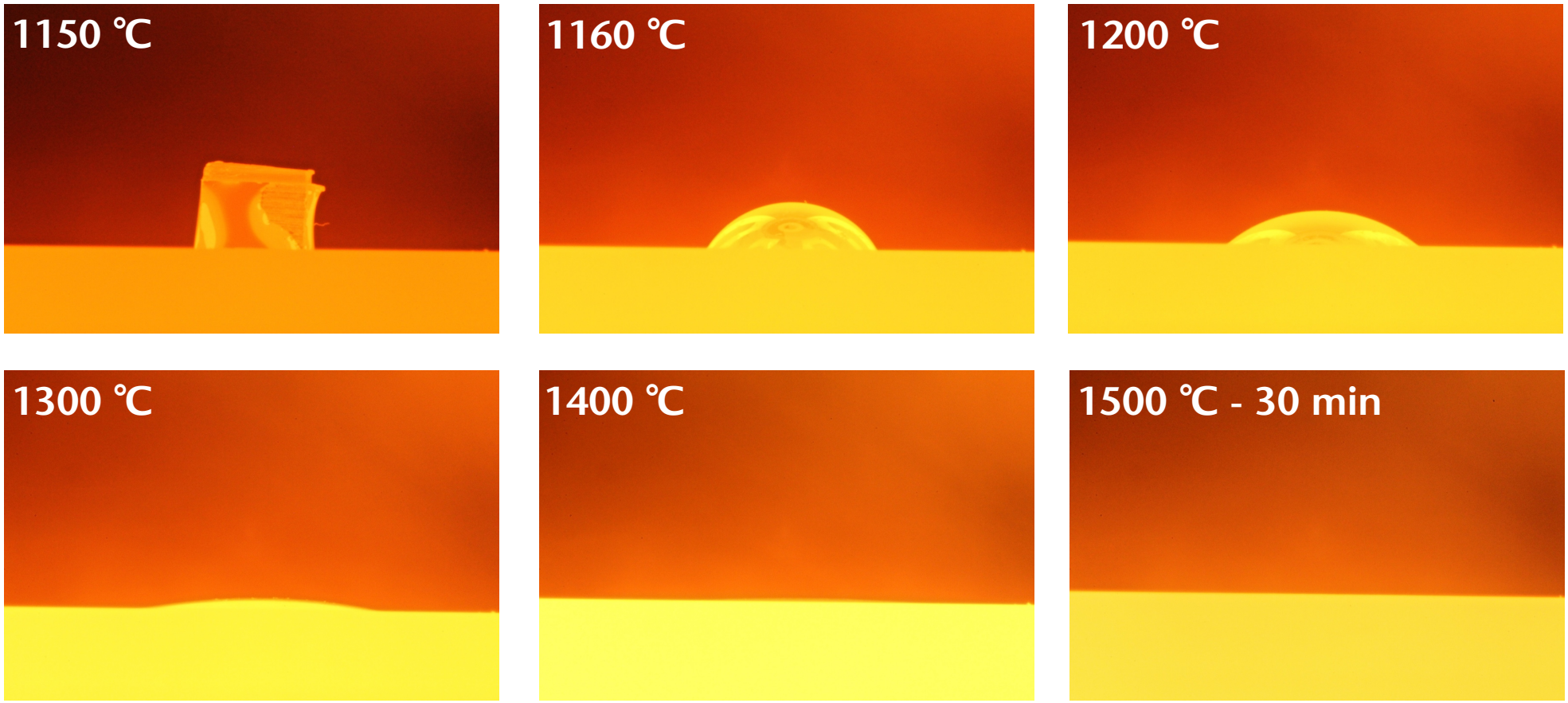




The Phase Diagrams of Ni-Si System

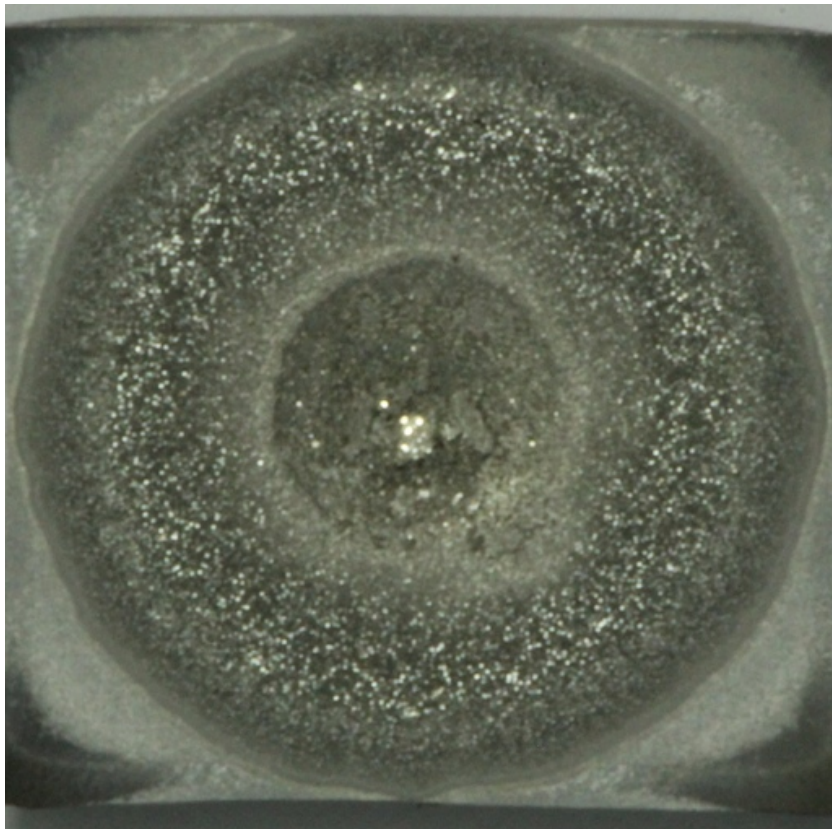


HfB_2-10vol%$MoSi_2$ / pure Ni

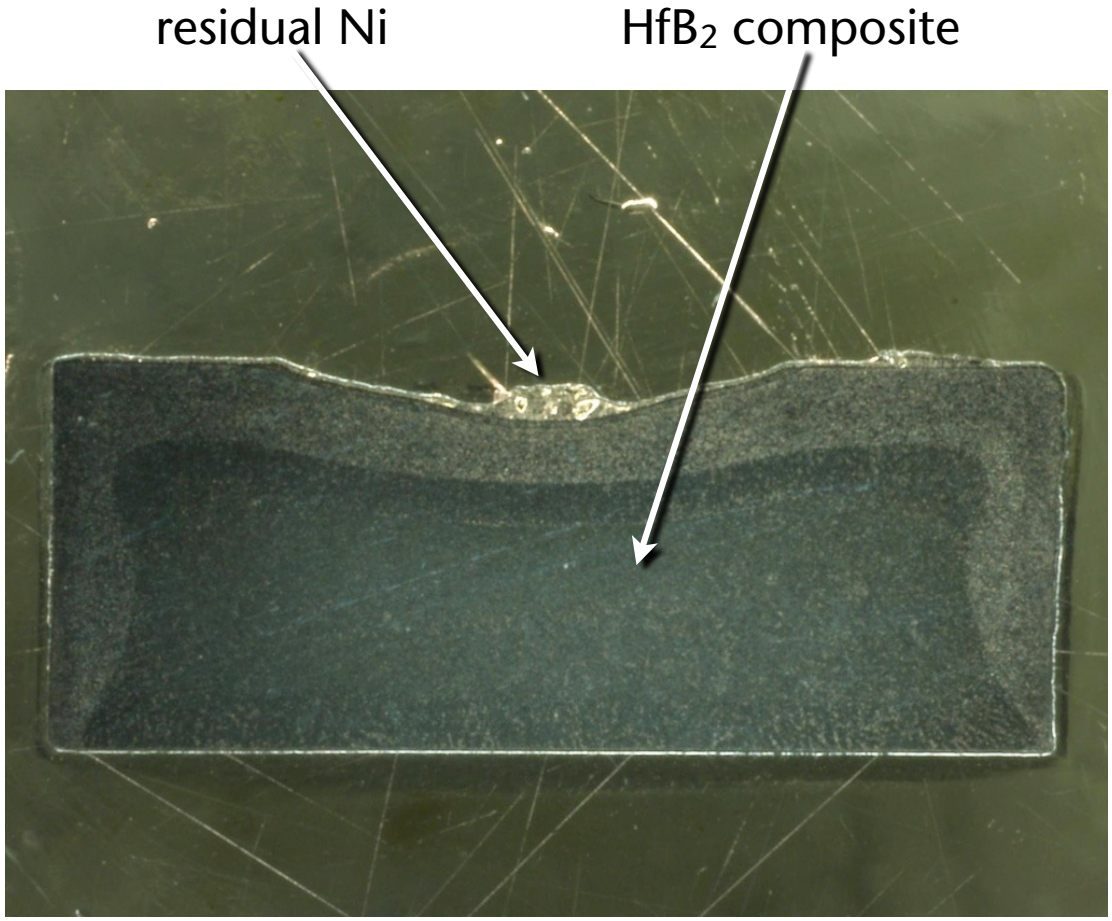


The wettability of HfB_2 composite is very good.

OM images, HfB_2-10vol%$MoSi_2$ / pure Ni

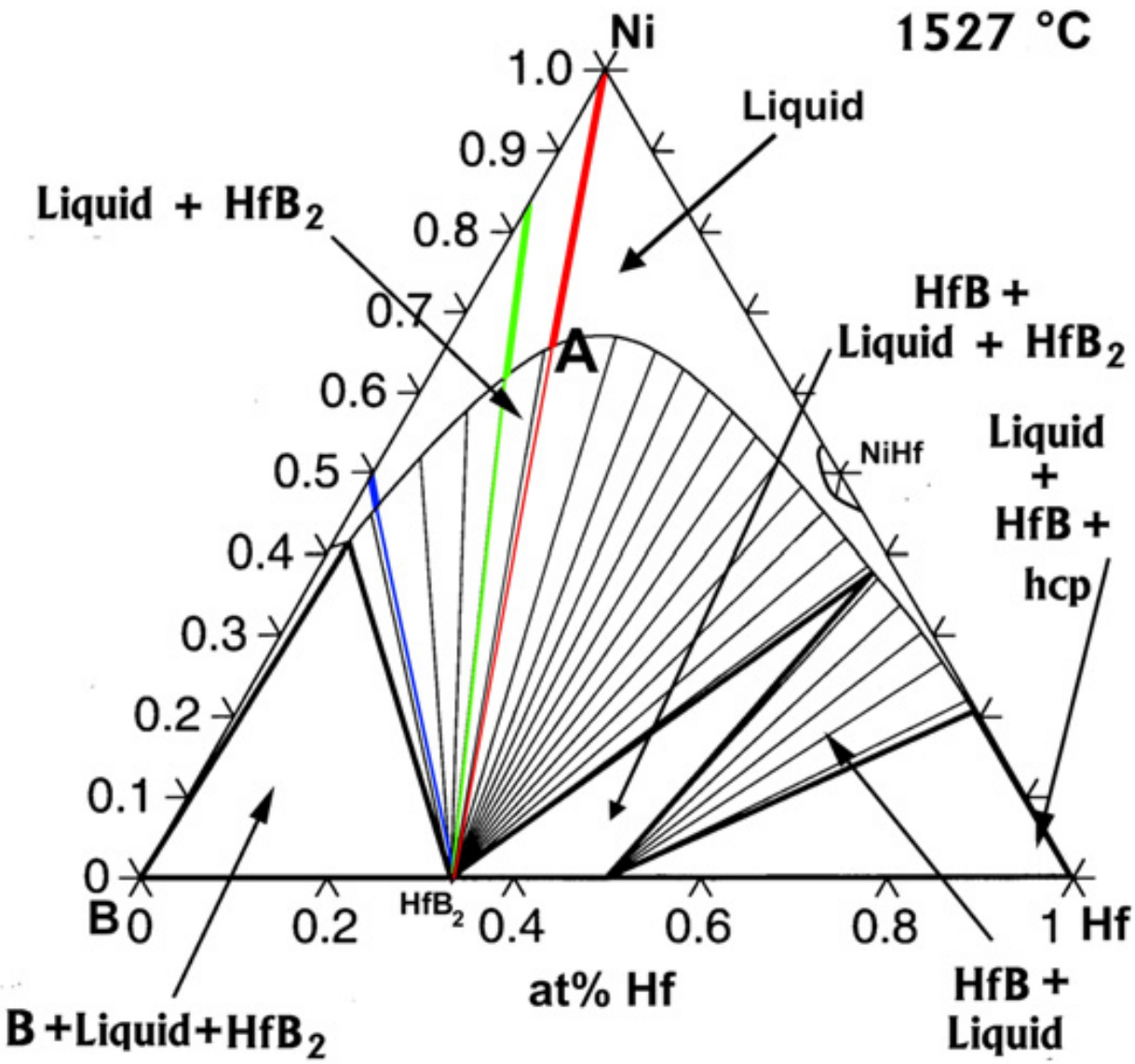


Top view



Cross-sectional view

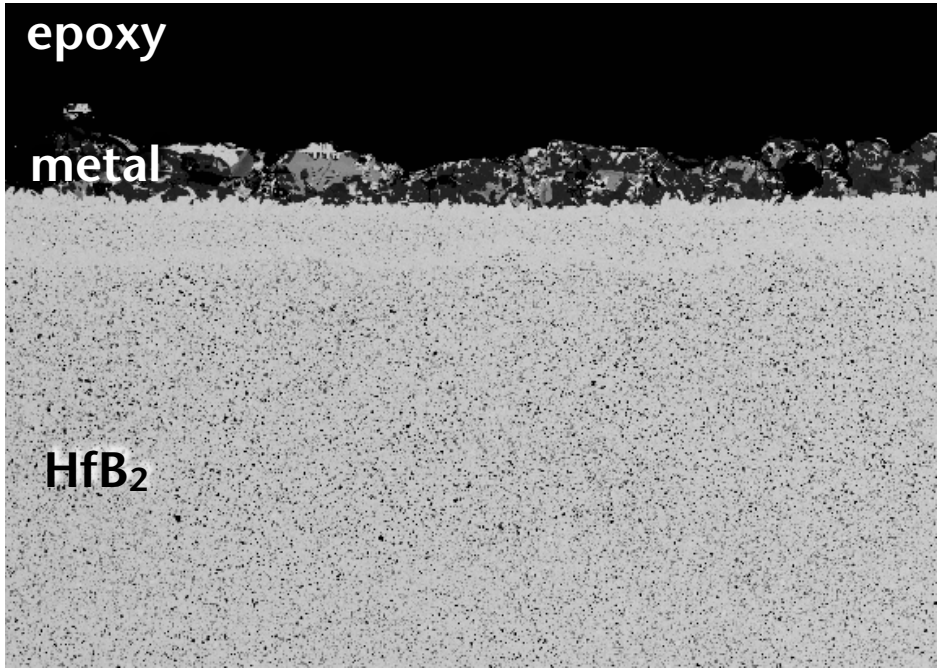
Solubility of HfB_2 into Ni Liquid

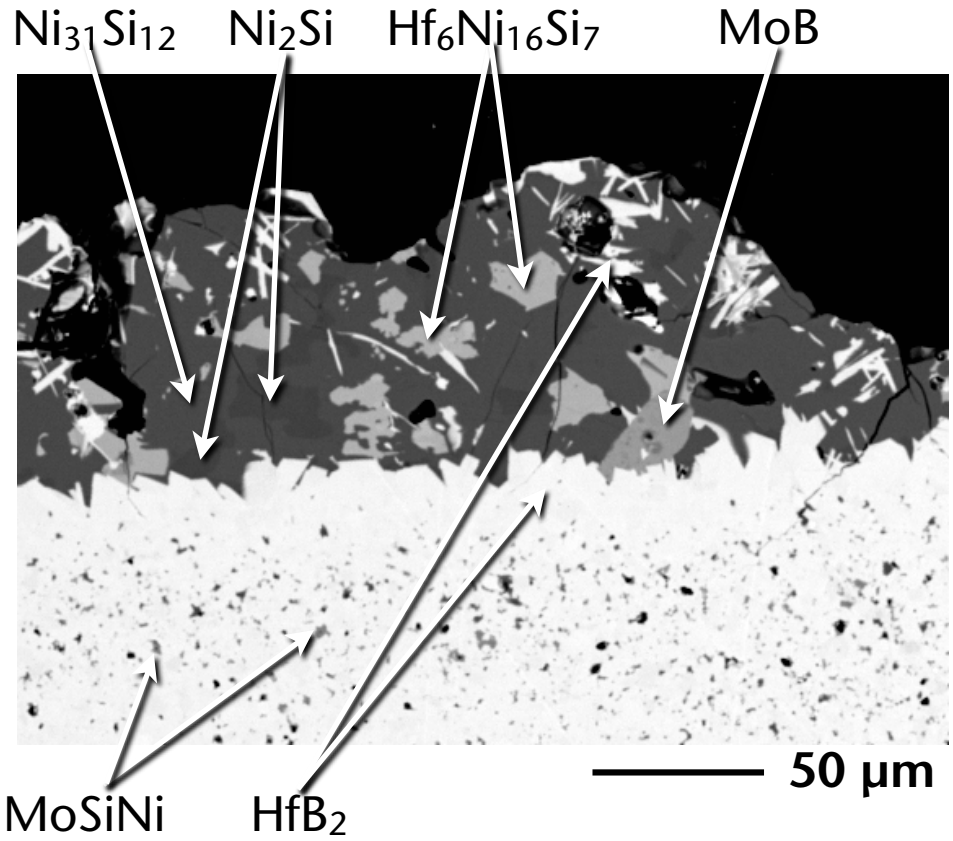


Large solubility at h.t.

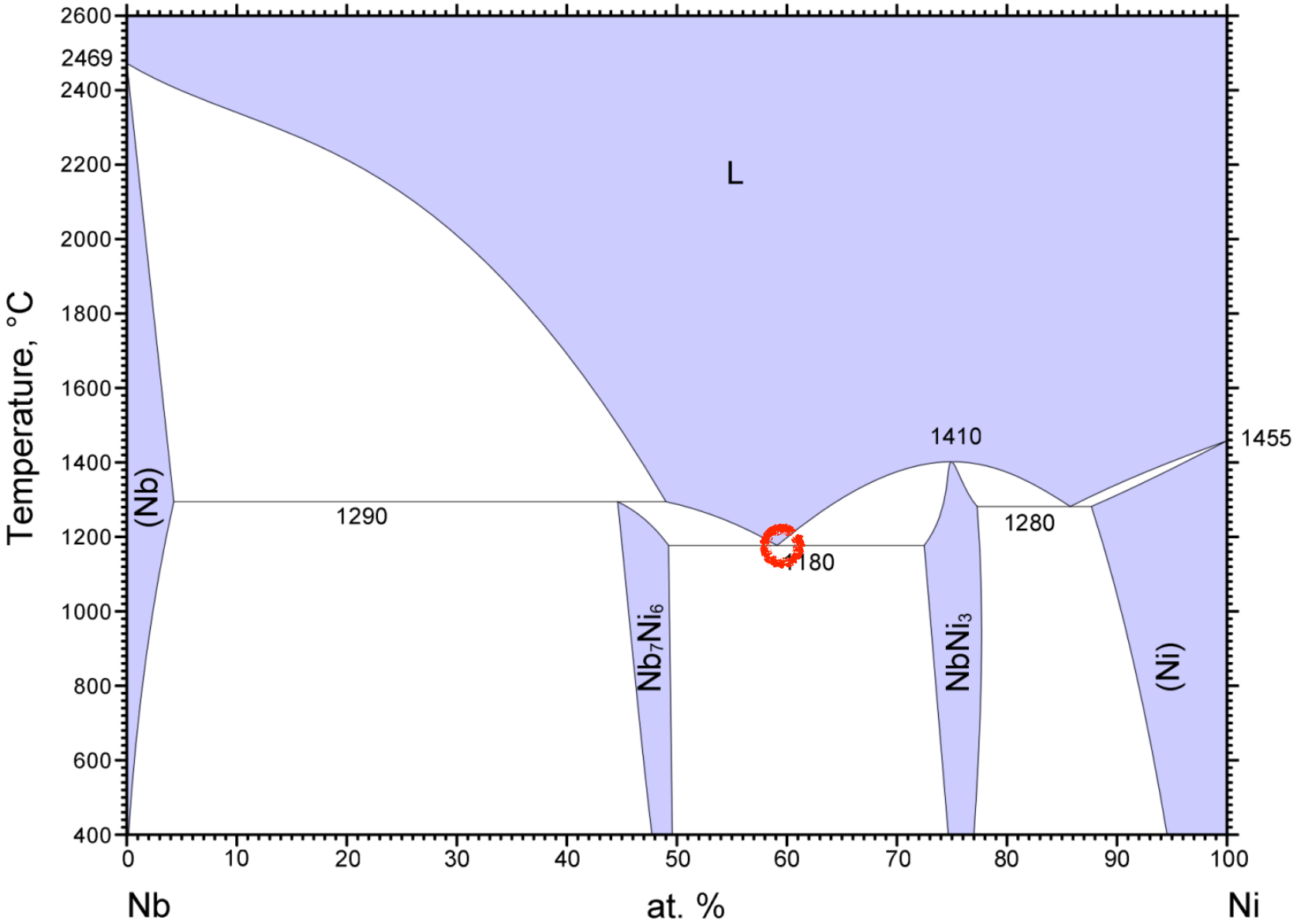
A. Passerone et al. : CALPHAD, 34 (2010) 6–14

BSE images, HfB_2-10vol%$MoSi_2$ / pure Ni

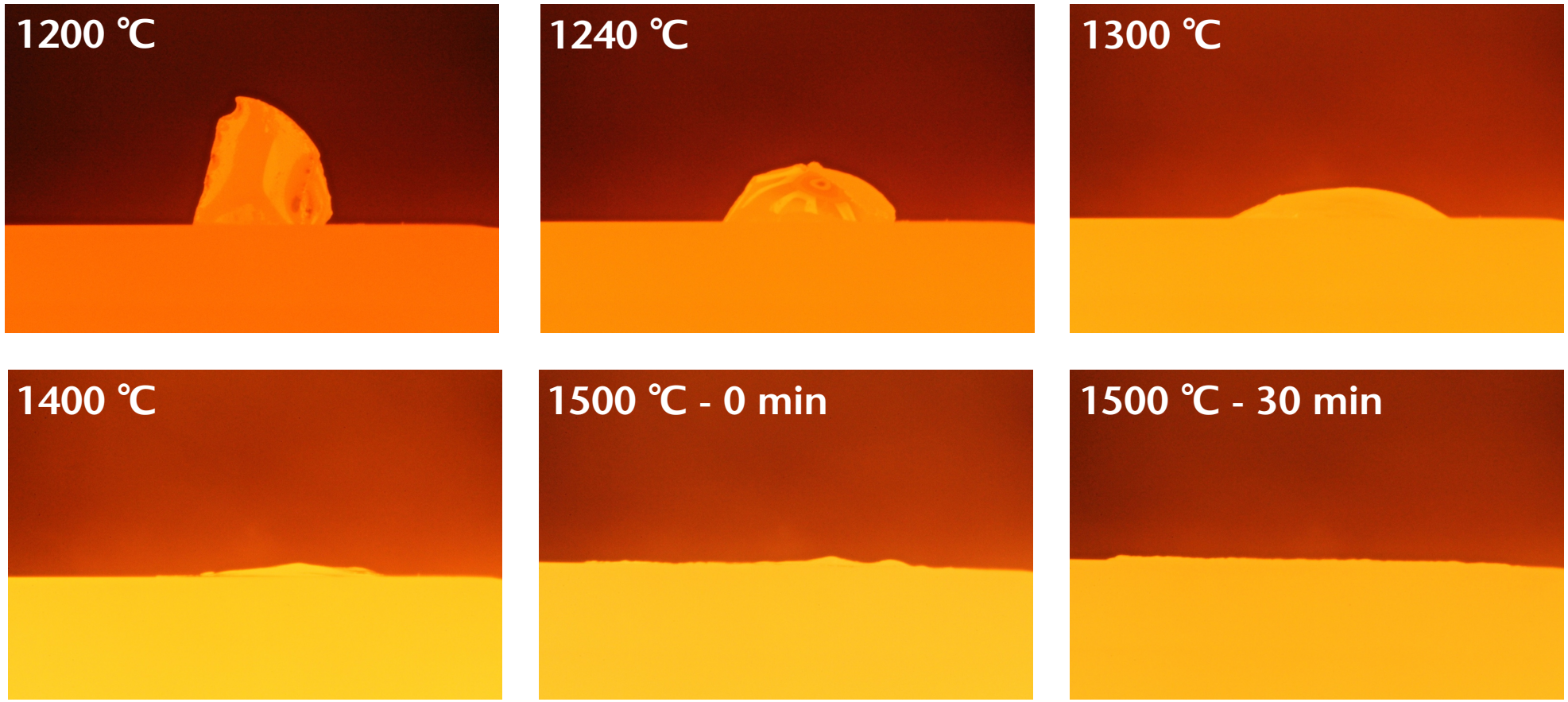




The Phase Diagram Nb-Ni System

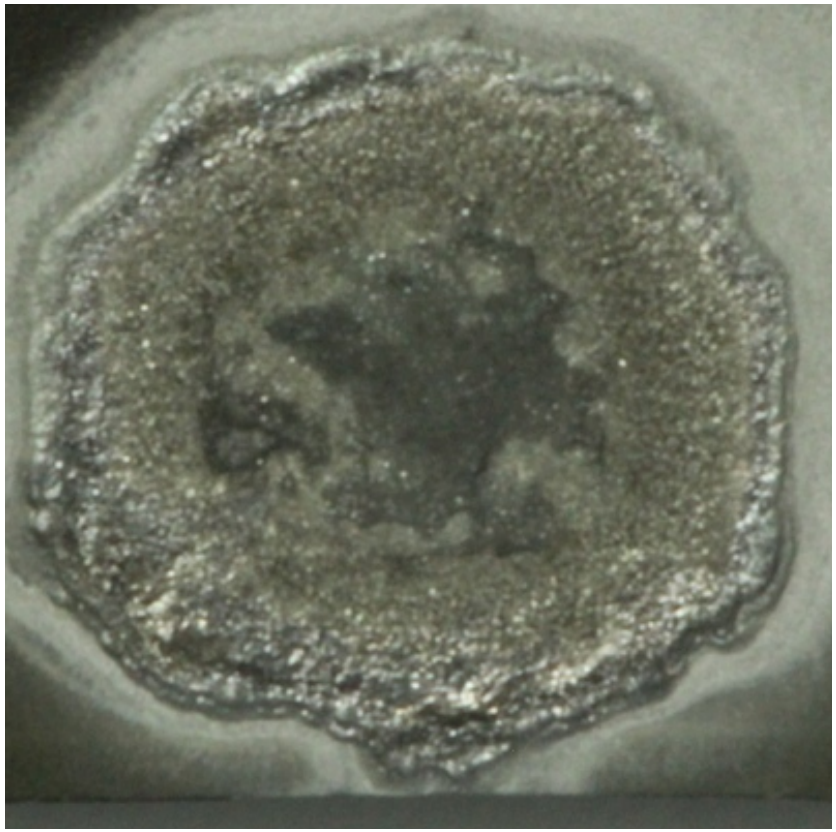


HfB_2-10vol%$MoSi_2$ / Ni-40at%Nb

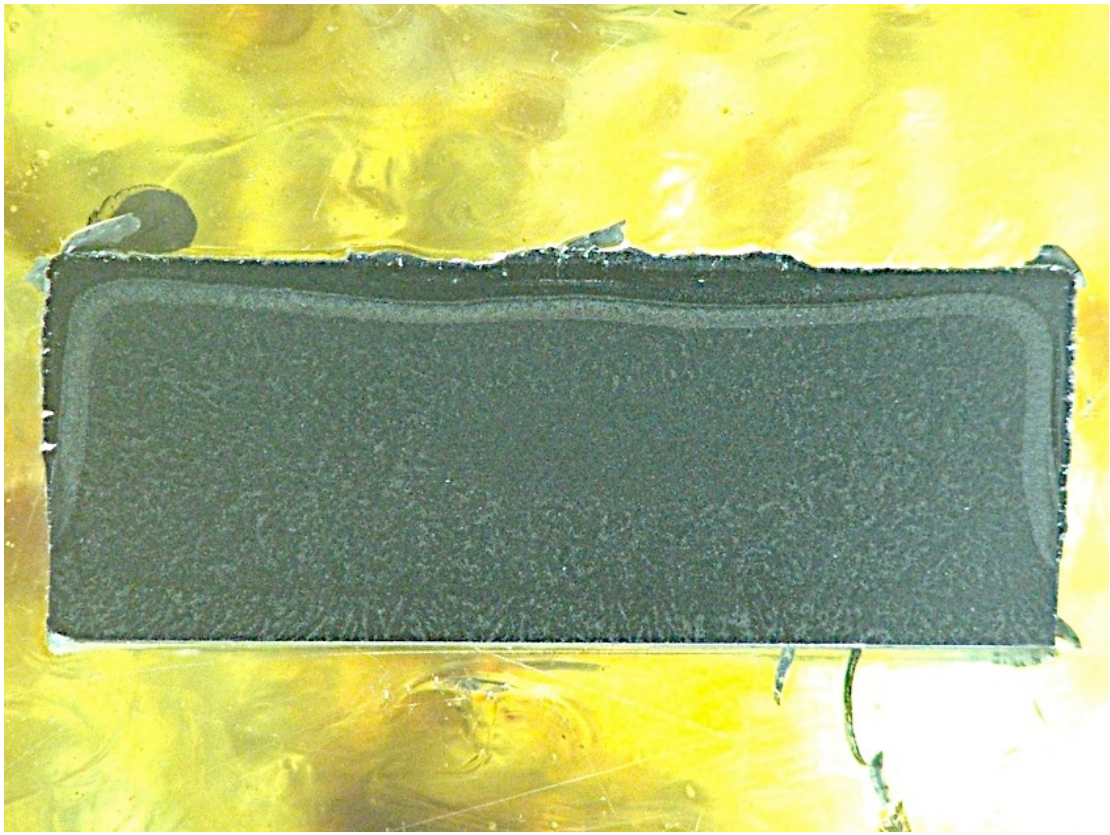


The wettability of HfB_2 composite is very good.

OM images, HfB_2-10vol%$MoSi_2$ / Ni-40at%Nb

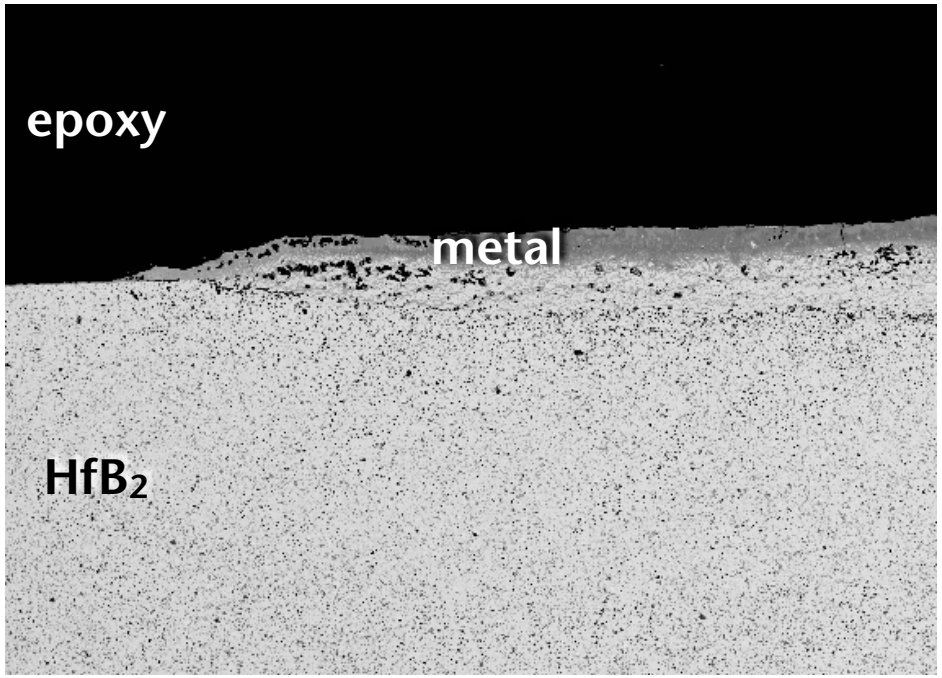


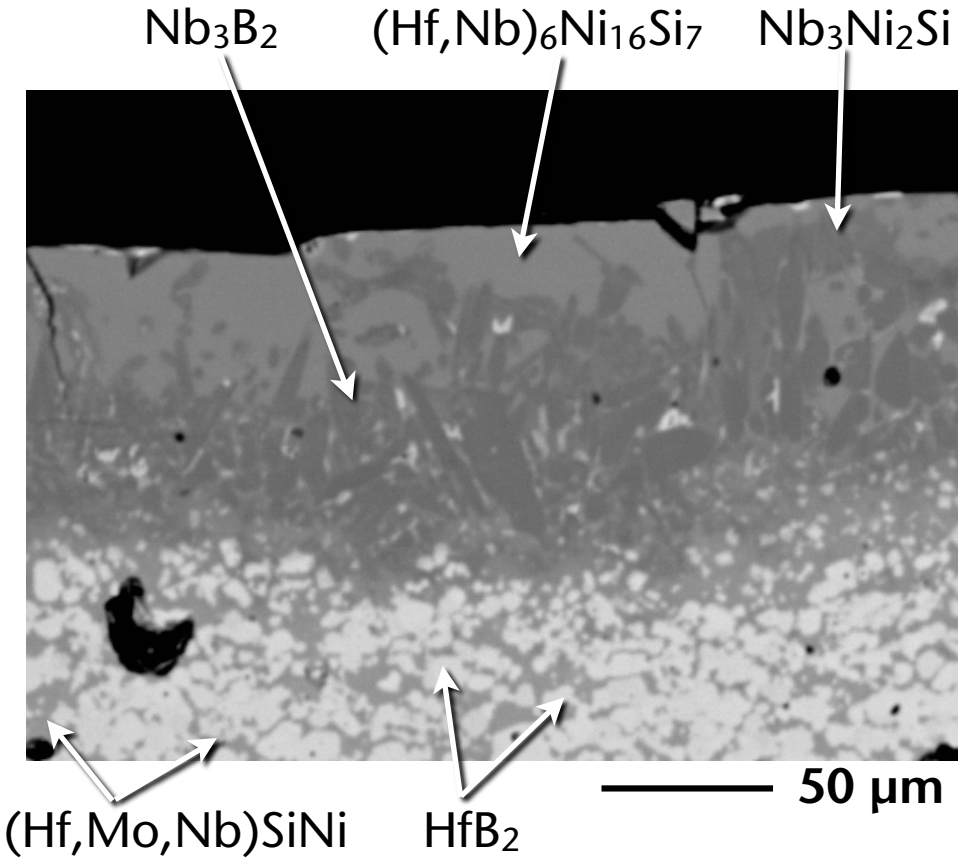
Top view



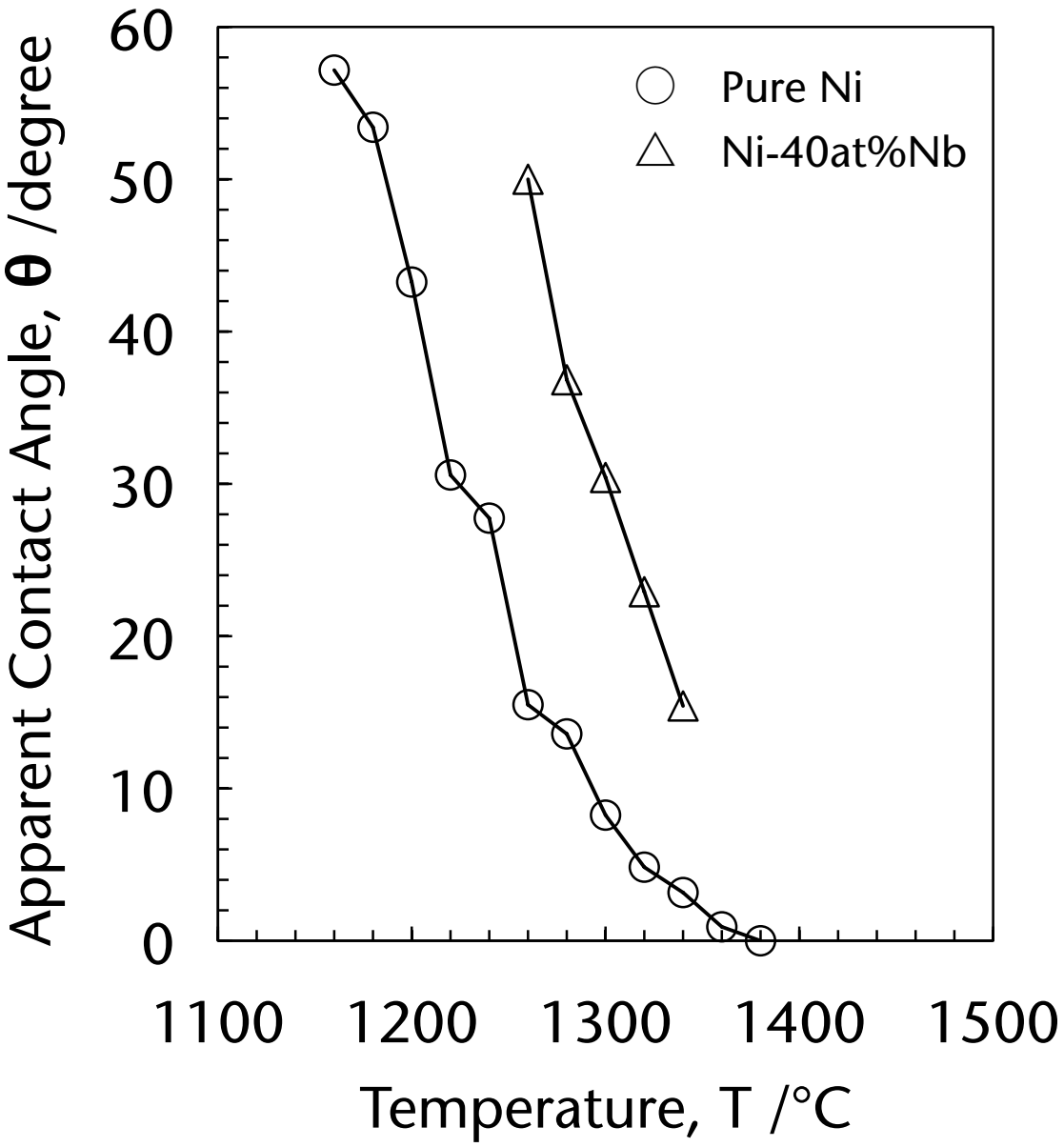
Cross-sectional view

BSE images, HfB_2-10vol%$MoSi_2$ / Ni-40at%Nb





Apparent Contact Angle of Ni-Nb Alloy



Good wettability
against Ni-Nb alloy

J. Mater. Sci. 2012, 47, 8454-8463

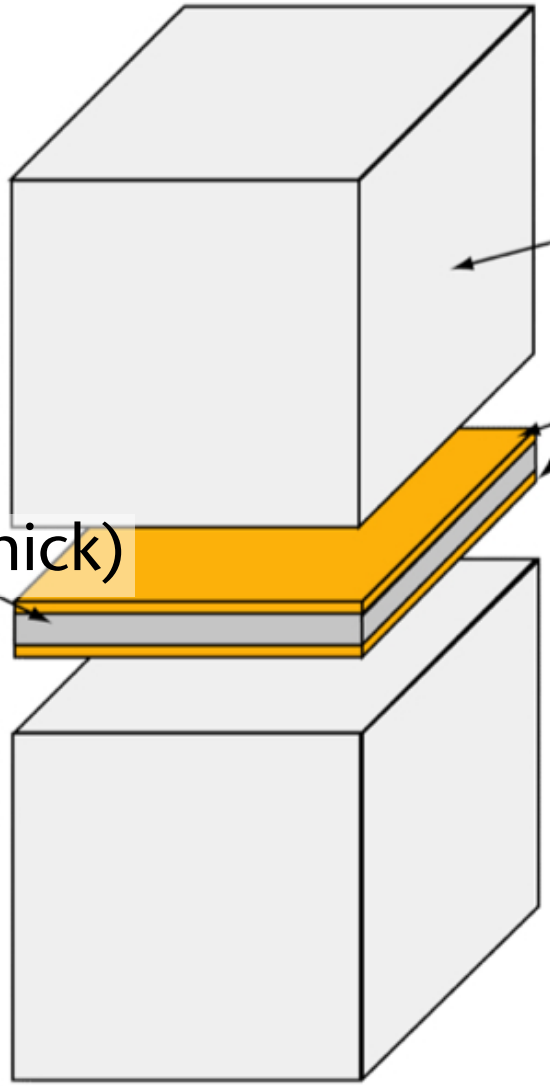
Qualifications for TLP Interlayer

1. CTE Matching ✓

2. Liquid Phase ✓

3. Wettability ✓

TLP Bonding of HfB_2 Composite using Ni-Nb Interlayer



TLP Bonding of HfB_2 Composite using Ni-Nb Interlayer

Heating at 15 K/min

Holding

1500 ℃ for 30 min
8.5 MPa Hot-press

Cooling

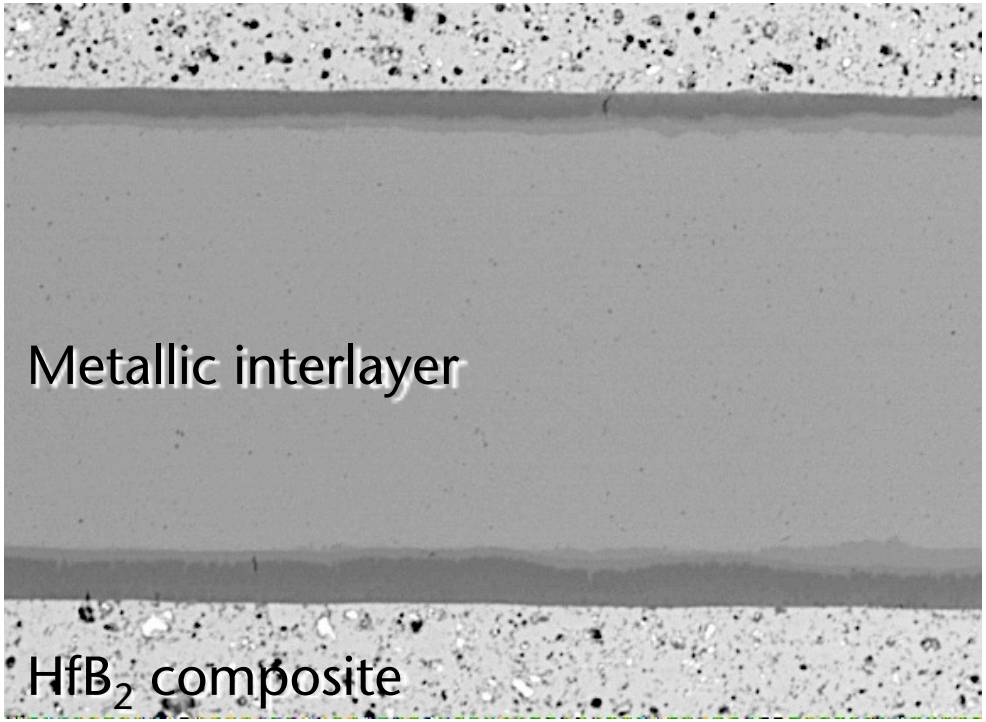
by cutting off the power

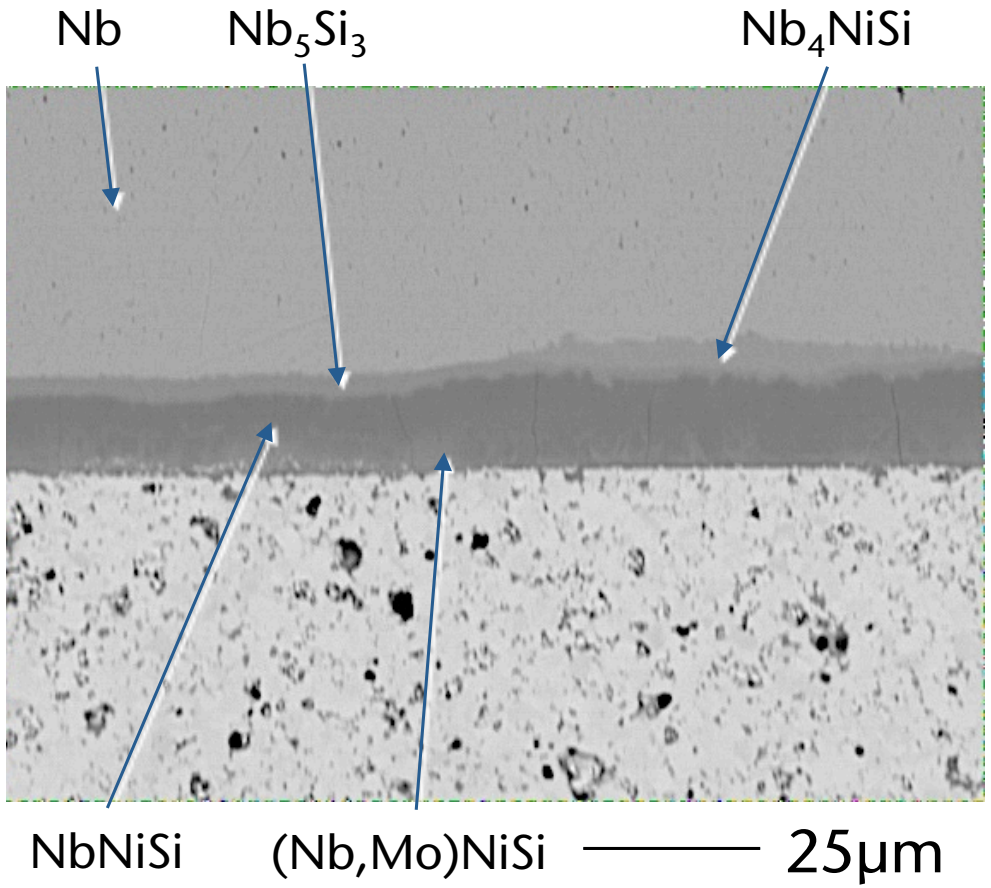
Microstructural Analysis
Mechanical Test

Appearance of TLP Joint

Well bonded!

BSE images of the interface, after 1500 °C for 30 min





Silicides were formed at the interface.
TLPB was not completed.

4-Point Bending Strength of TLP Joints at r.t. and at h.t.

EDM Machining

5 beams for r.t.
3 beams for h.t.

Polishing + Beveling

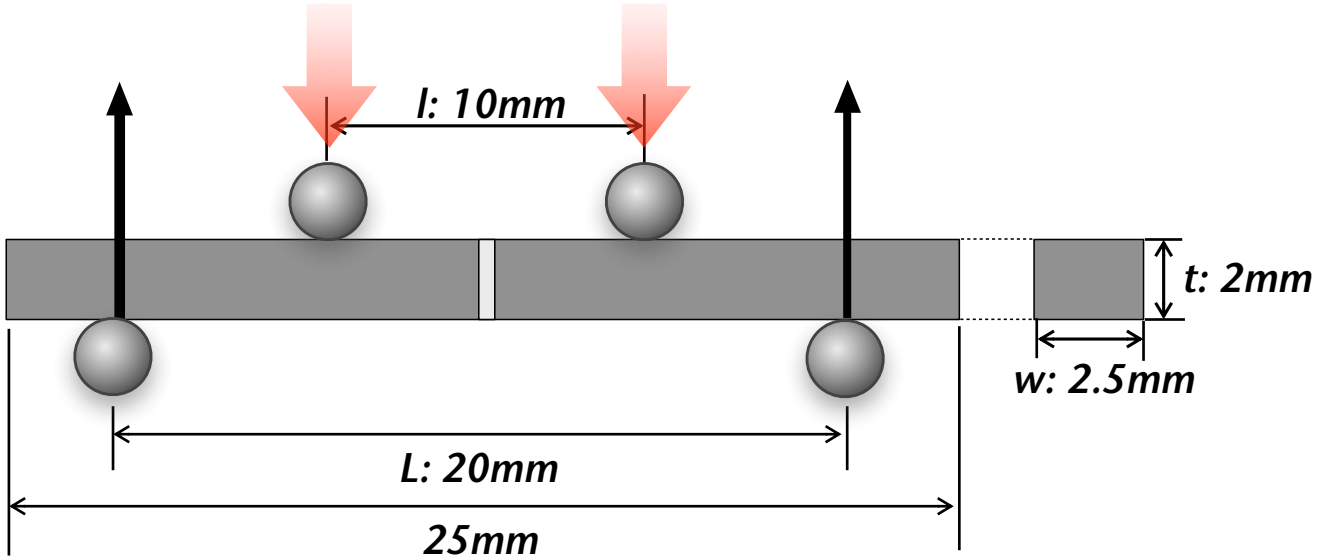
finishing with c-SiO_2

Testing at r.t.

1.0 mm/min

Testing at h.t.

1.0 mm/min
at 1000 °C, Air
10 K/min



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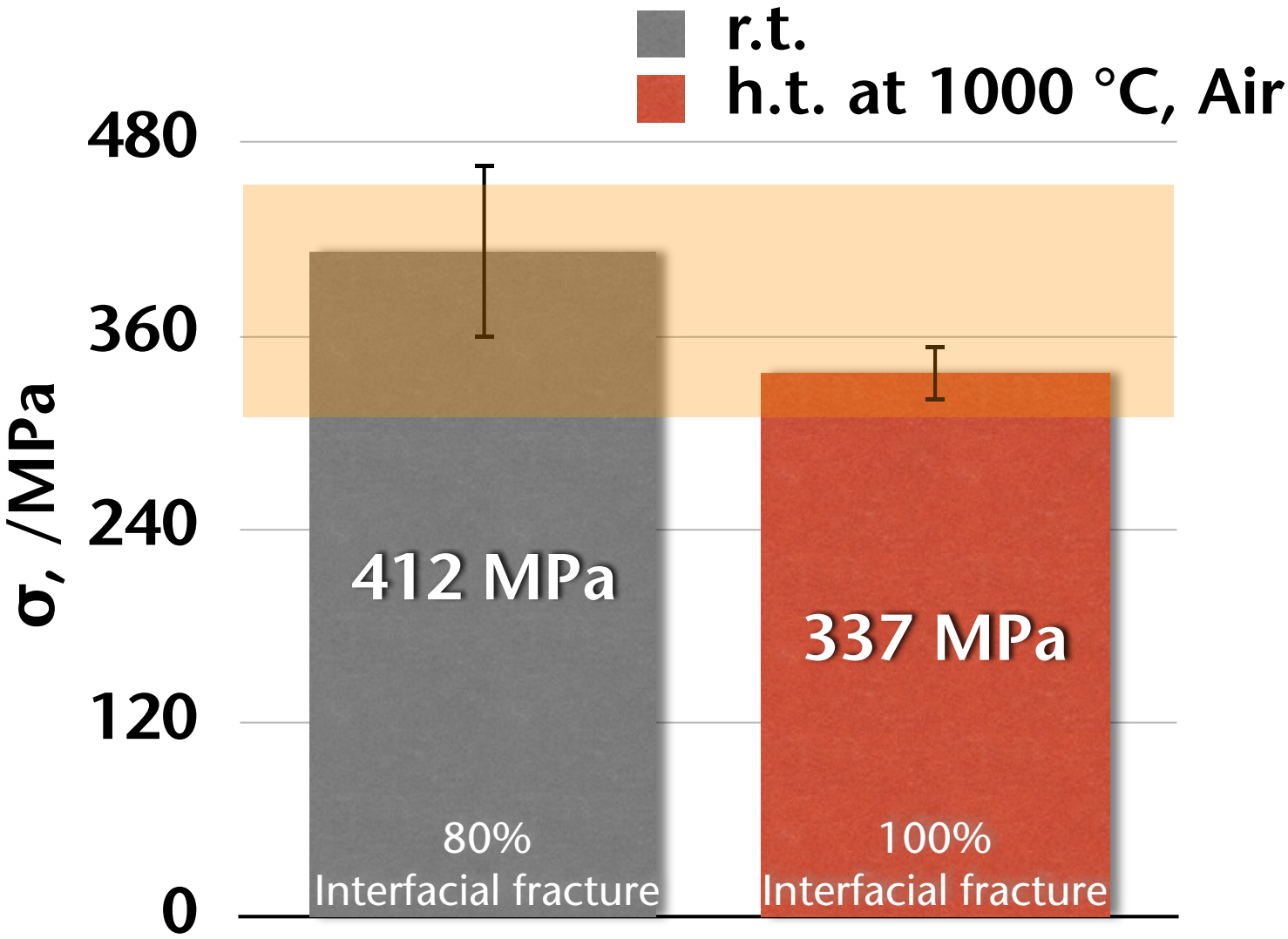


Experimental Procedure of "Drip" Wetting Test

Heating at 15 K/min

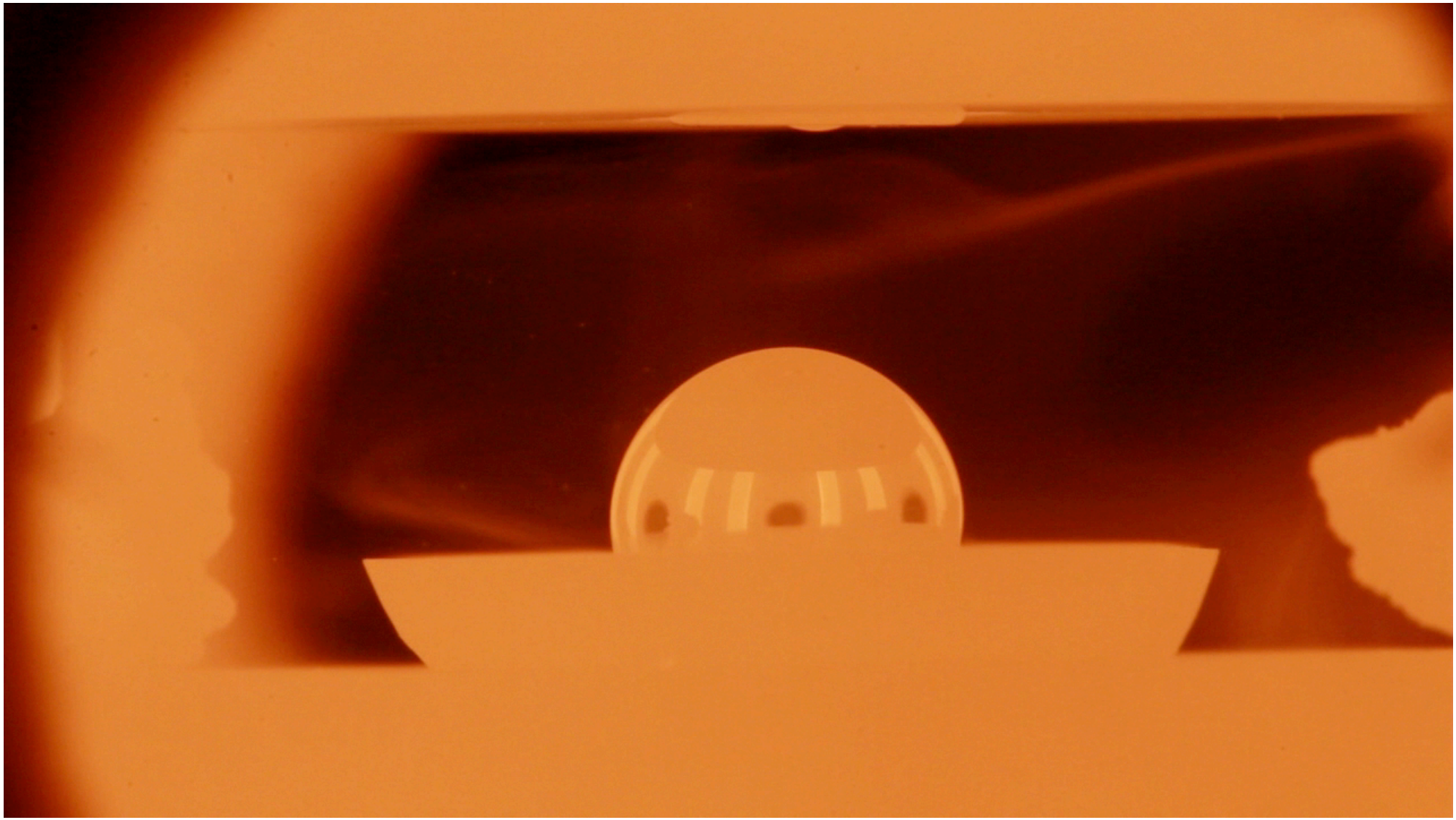
under 10^{-3} Pa vacuum

Dripping Ni melt at 1500 ℃

onto UHTC substrate

Movie shooting

A Typical Movie of “Drip” Wetting Test at 1500 °C



Joining of ZrB_2 Composite using Reactive Metal

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+10vol%$MoSi_2$ (-325 μm, 99.9%)
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Molding

23 mmΦ
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Sintering

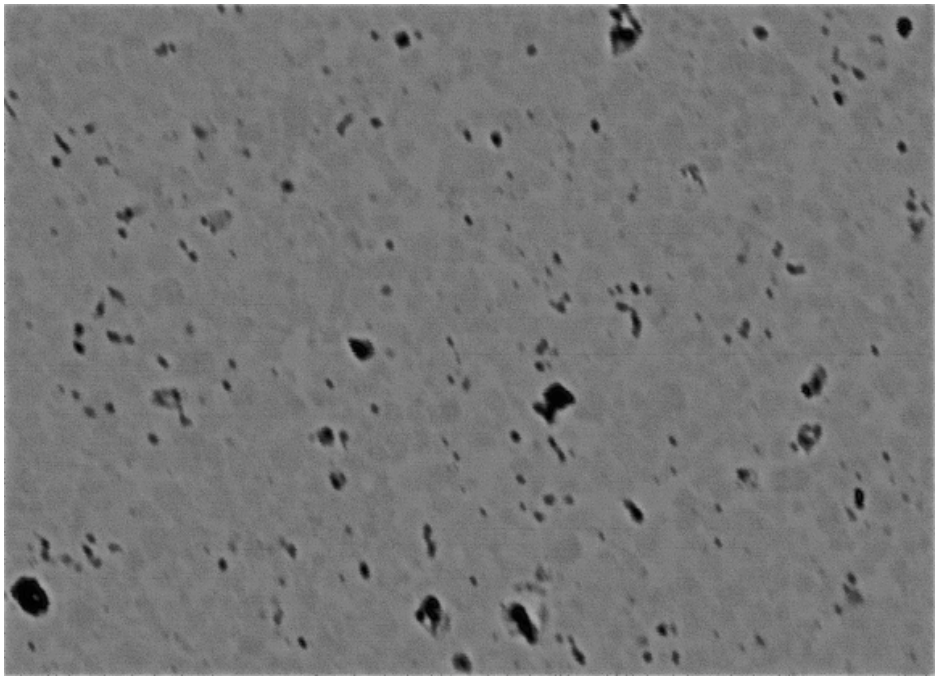
2000 ℃ for 60 min
Under 0.1 MPa Ar

Polishing

with diamond slurries

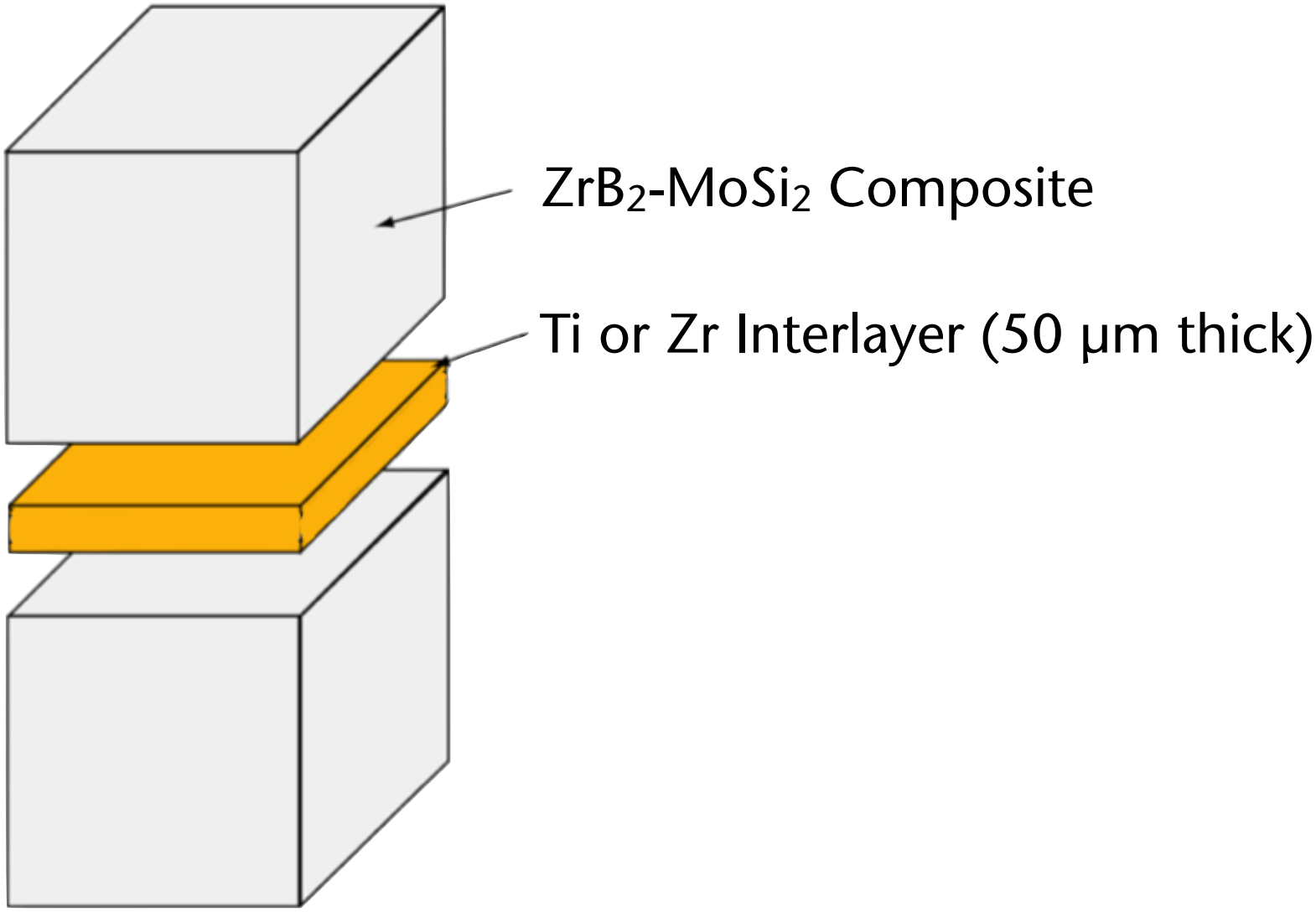
Joining

BSE image of ZrB_2-10vol%$MoSi_2$ Polished Section



98% relative density

Joining of ZrB_2 Composite using Ti or Zr Interlayer



Joining of ZrB_2 Composite using Ti or Zr Interlayer

Heating at 15 K/min

Holding

1500 ℃ for 30 min
8.5 MPa Hot-press

Cooling

by cutting off the power

Microstructural Analysis
Mechanical Test

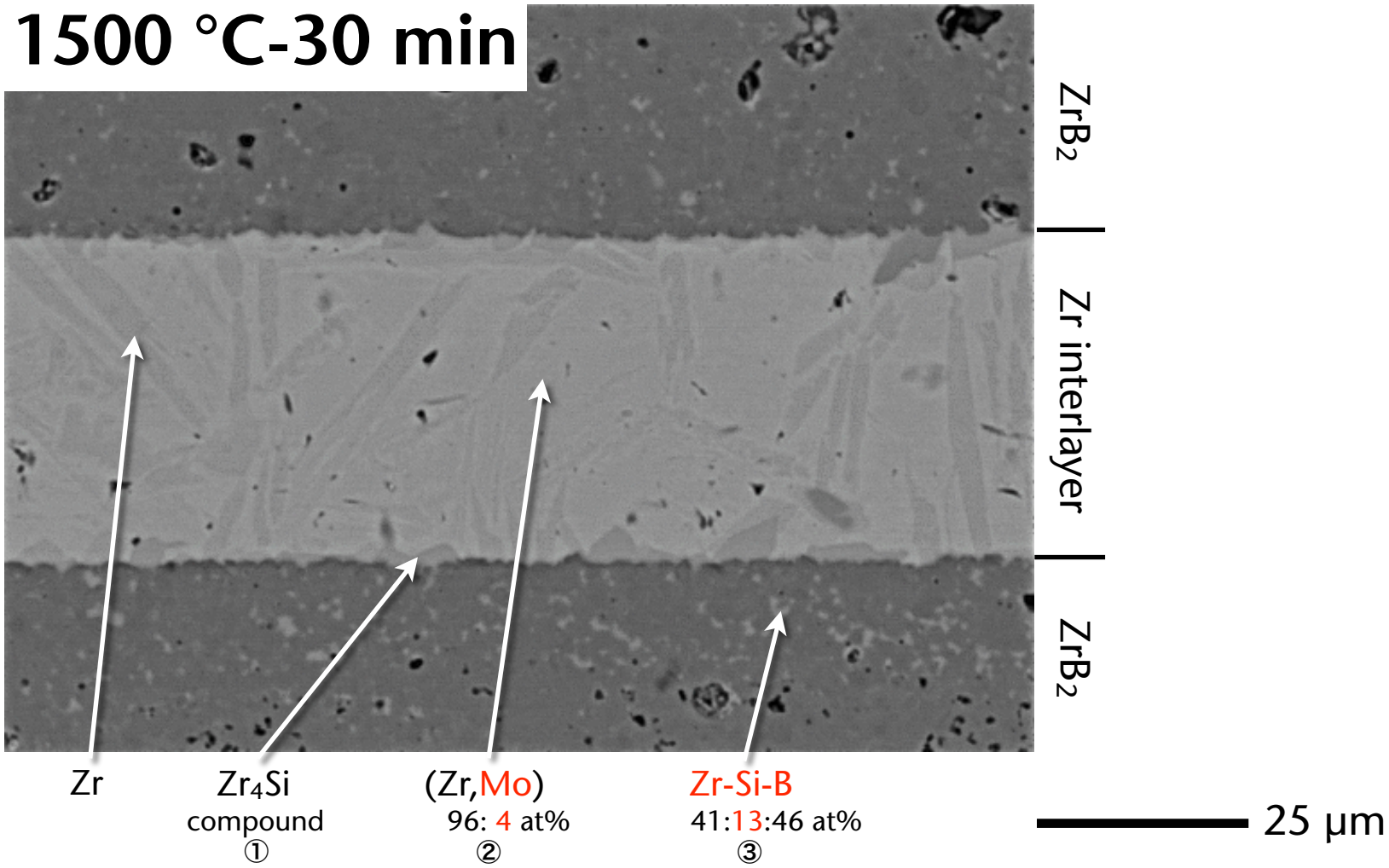
Appearance of Joints after 1500 °C for 30 min

Ti Interlayer

Zr Interlayer

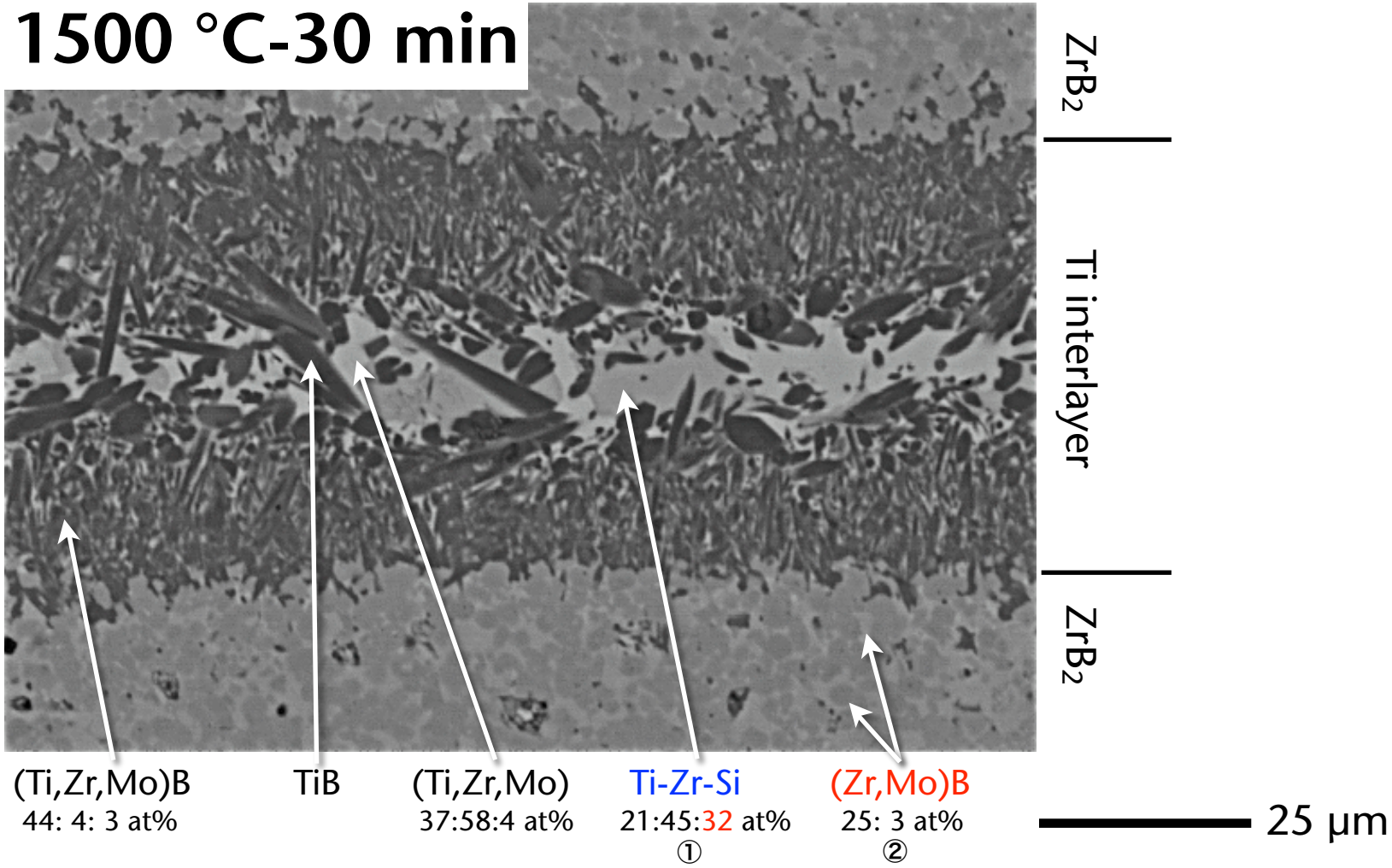
Well bonded!

BSE image, Bonded Interface of $ZrB_2/Zr/ZrB_2$



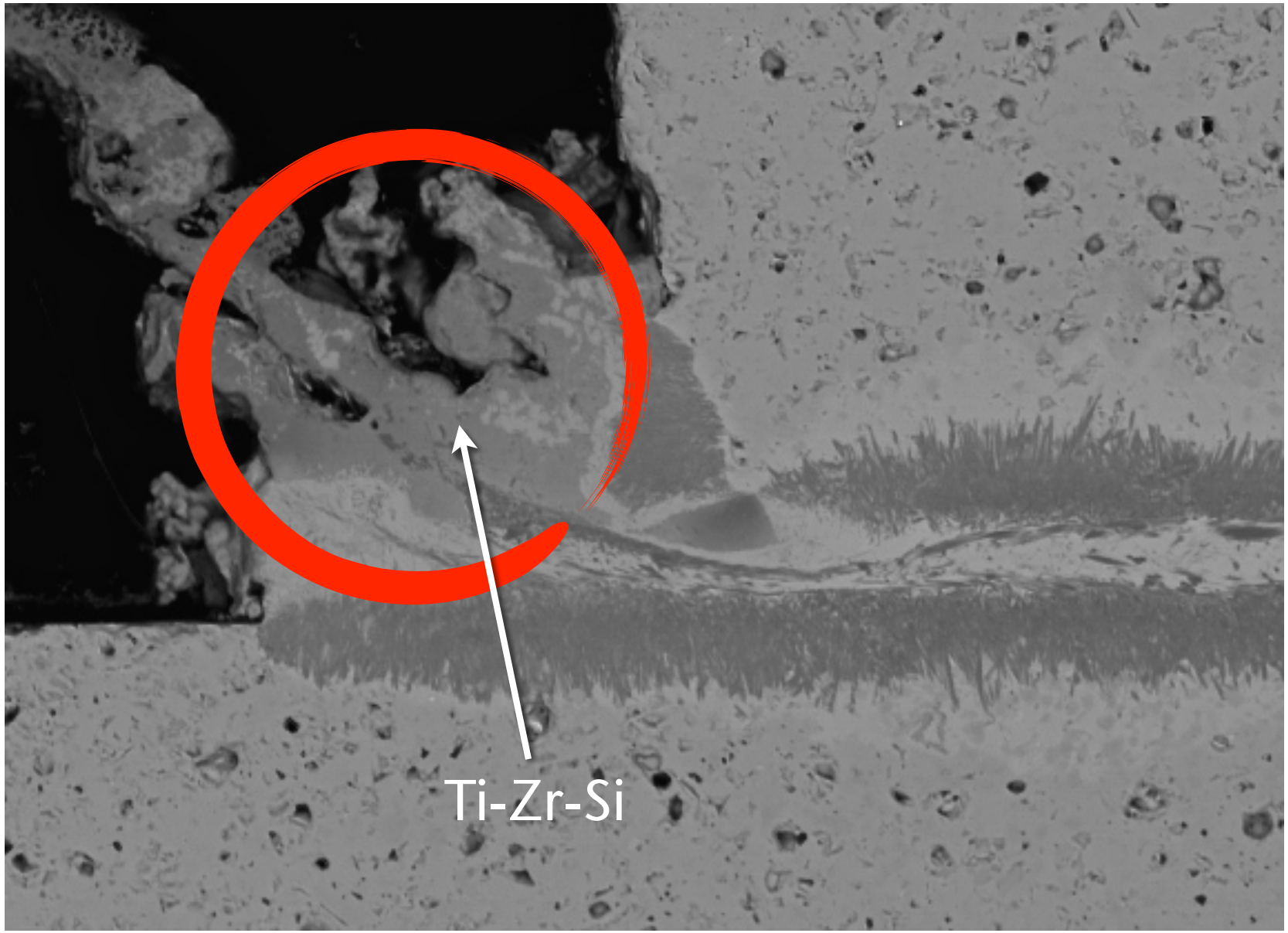
Interfacial reaction was found to be modest in solid state.

BSE image, Bonded Interface of ZrB_2/Ti/ZrB_2



Interfacial reaction was found to be very intensive.

Squeezed Liquid at Ti/ZrB_2 Interface



4-Point Bending Strength of Joints at r.t. and at h.t.

EDM Machining

5 beams for r.t.
3 beams for h.t.

Polishing + Beveling

finishing with c-SiO_2

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1.0 mm/min

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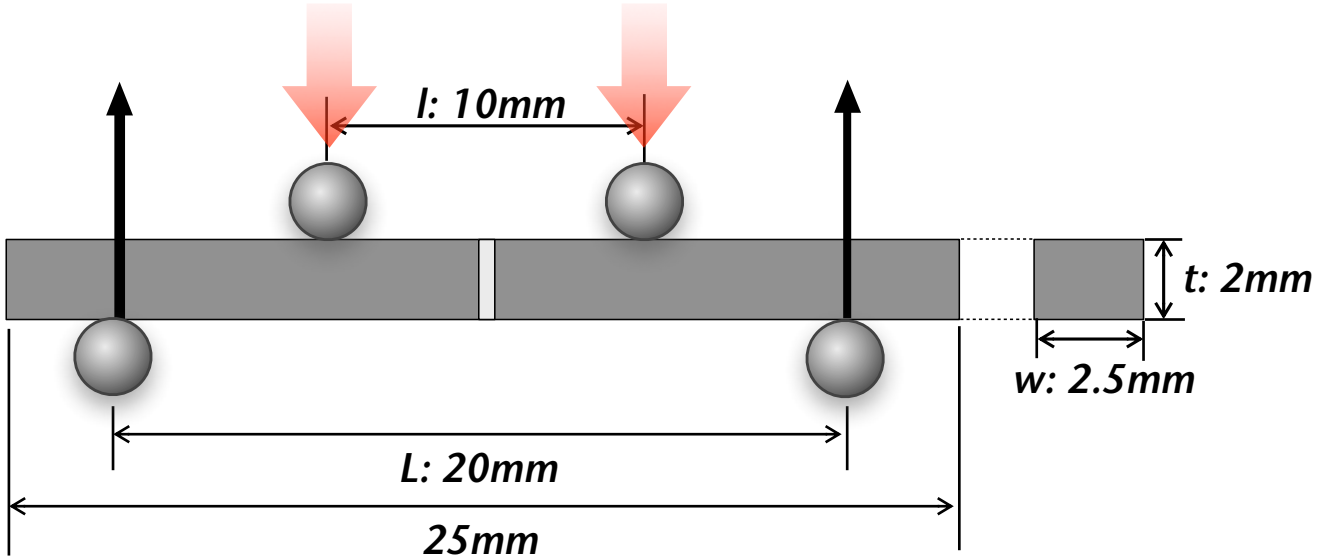
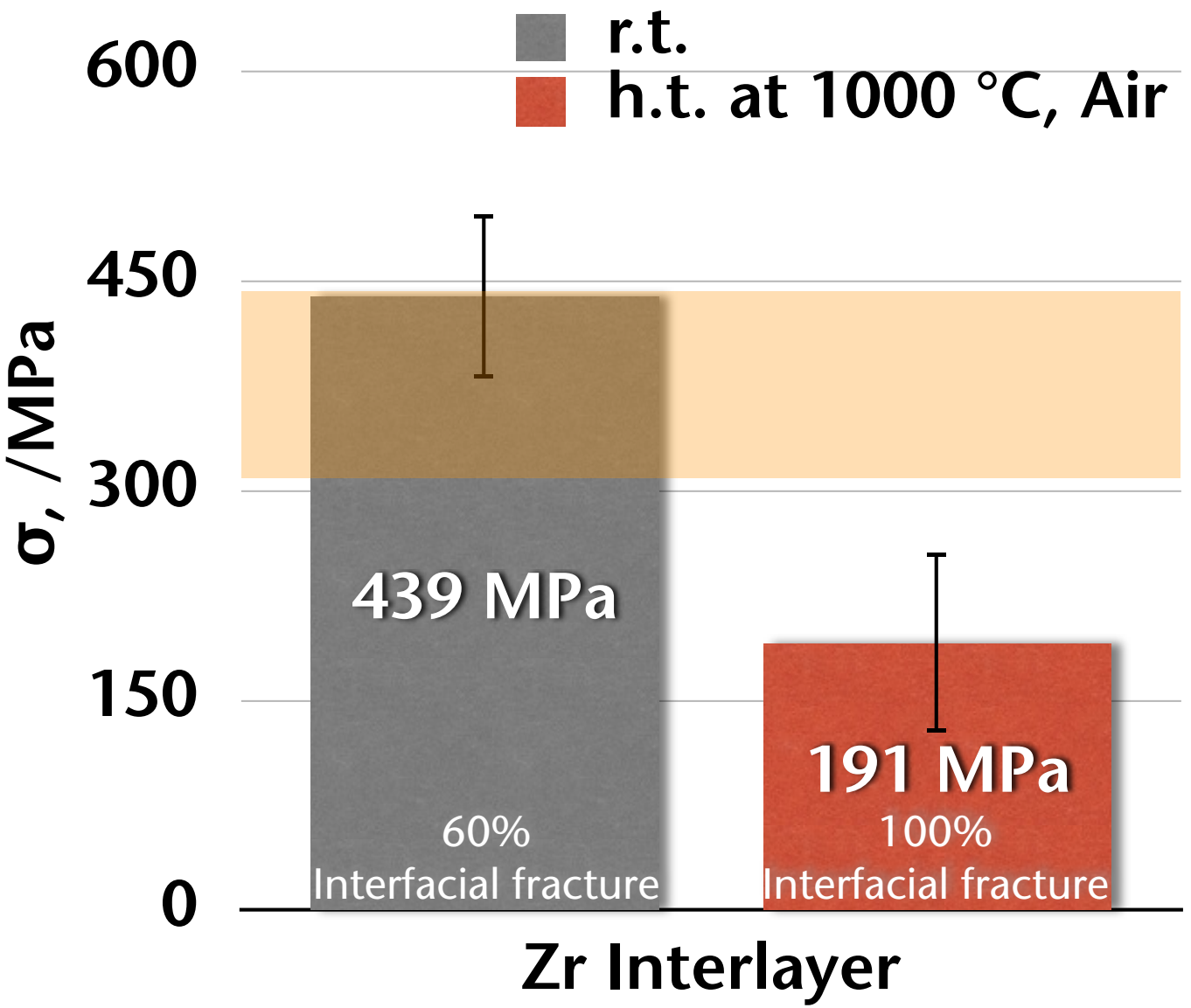
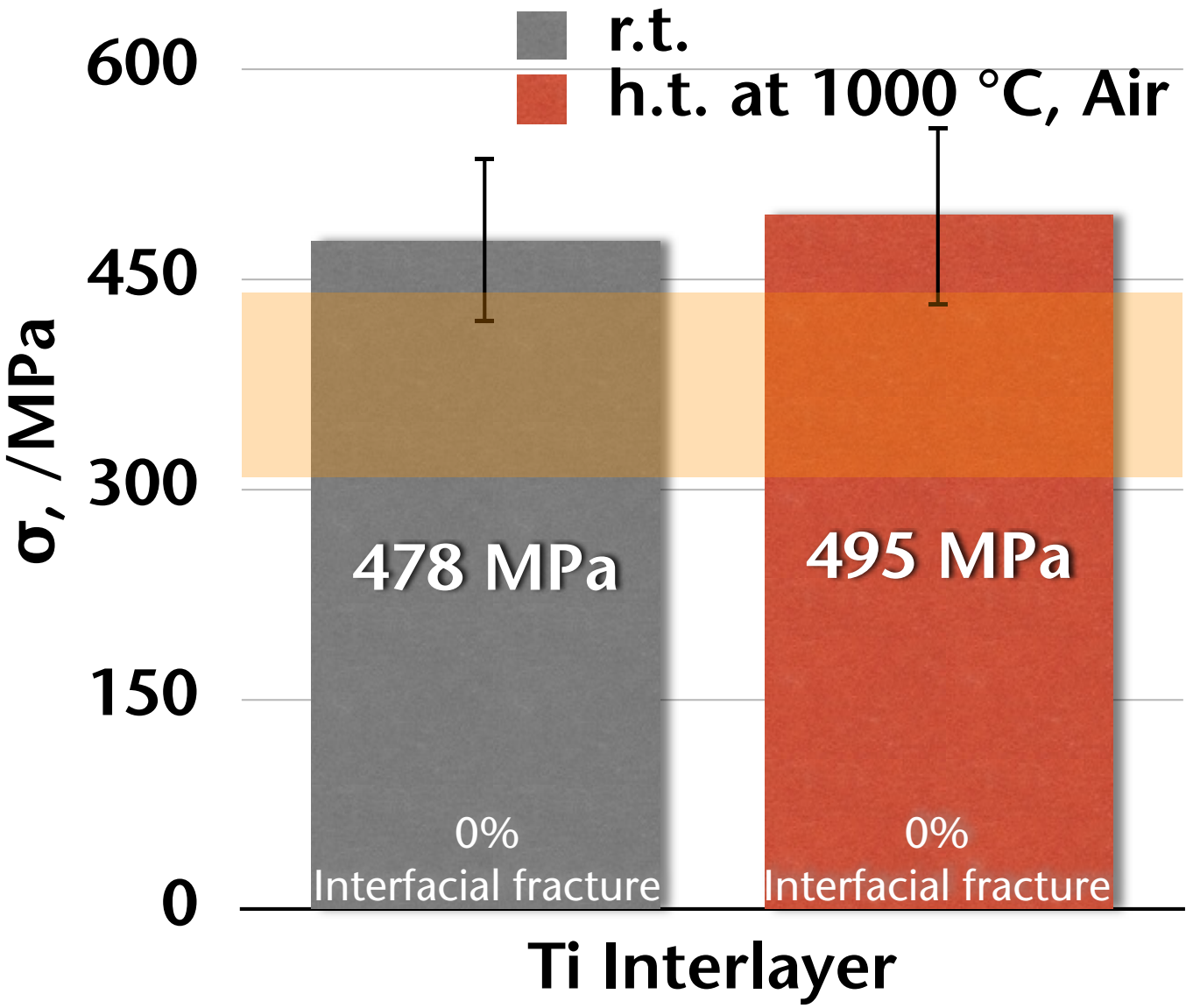


4-Point Bending Strength of Joints



4-Point Bending Strength of Joints with Ti Interlayer



Conclusions

1. The result of the TLP bonding for HfB_2 composite using the Ni/Nb/Ni interlayer at 1500 °C has shown that the interlayer and composite were well-bonded. The layers of intermetallic compounds related to Si were found in the interfacial region, suggesting detrimental interfacial reaction with $MoSi_2$.

2. The result of the joining for ZrB_2 composite using the Ti or Zr interlayer at 1500 °C has revealed that both the interlayers and composite were well-bonded. Especially, the joint with the Ti interlayer exhibited remarkable bending strength even at high temperature.

Acknowledgements

Consiglio Nazionale delle Ricerche
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Dr. Laura Silverstroni, Dr. Diletta Sciti,
and Stefano Guicciardi, CNR-ISTEC, Italy

Prof. Andreas M. Glaeser, UC Berkeley, USA

Acknowledgements

Japan Society for the Promotion of Science