Wetting and interfacial phenomena in relation to joining of advanced ceramics

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National Research Council of Italy

ICMATE
Institute of Condensed Matter Chemistry
and Technologies for Energy

Outline

- **Introduction**
  - Basics of wetting at high temperatures
  - Basics of ceramic joining by brazing
  - Methodologies: experimental complex for wetting studies
- **Examples of wetting and joining of SiC**
  - dissolutive wetting: SiC/Ni-Si
  - formation of new phases: SiC/Al-Ti
  - active elements: SiC/Sn-Ti
- **Conclusions**

Wetting and joining

Metal/Ceramic or Ceramic/Ceramic **joining** is necessary in order to:

- integrate ceramic components into metallic structures;
- enhance the individual characteristics of different materials;
- assemble ceramics in complex shapes whenever sintering is difficult.

Within this field, many processes are possible, for example:

- Mechanical joining
- Adhesive bonding
- Diffusion bonding (solid state processes)
- Soldering (<450°C)
- Brazing (>450°C)
- Transient Liquid Phase Bonding

Liquid state processes

Wetting and joining

A **liquid based process** (e.g. brazing, transient liquid phase bonding) is often preferable:

- to limit surface preparation,
- to avoid high pressures
- when the adjoining surfaces are not flat.

When designing new joining methods to be realized through **liquid media,** it is fundamental to understand:

✓ how well the brazing alloy wets the adjoining surfaces;

→Wetting tests (sessile drop)

✓ the chemical interactions between the liquid and the solid and how to control them;

→ Microanalysis and calculation of multi-component phase diagrams

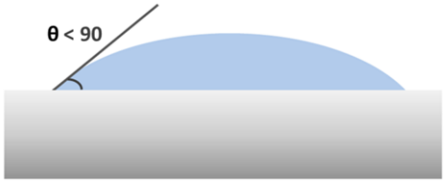
✓ the processing-microstructure-property relationship.

→ Joints manufacturing and characterization (e.g. mechanical, corrosion, ageing tests

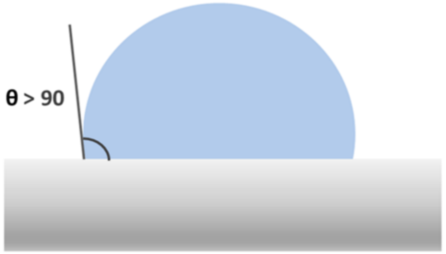
Wetting

Wettability is the tendency of a liquid to contact a solid surface and it is measured in terms of the **contact angle** between the liquid and the solid.

Sessile drop



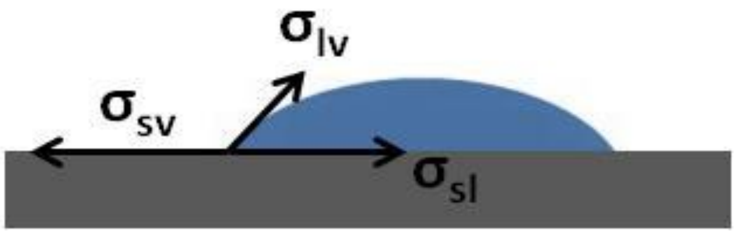
Wetting



Non wetting

Young's equation: equilibrium contact angle in relation with the interfacial tensions between the three phases:

$$\sigma_{sv} = \sigma_{LS} + \sigma_{LV} \cos \theta$$



Young-Duprè equation: work of adhesion between the liquid and the solid:

$$W_{ad} = \sigma_{LV}(1+\cos \theta)$$

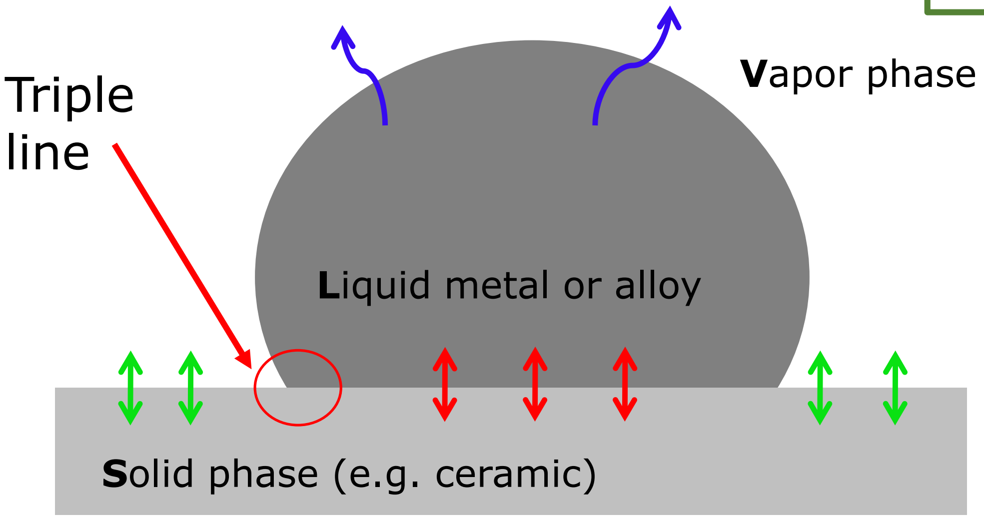
Gibbs adsorption (multicomponents):

$$d\sigma = - s dT - \Sigma_i \Gamma_i d\mu_i$$

Sessile drop

σ_{LV} : surface tension of the liquid
θ : contact angle

Sessile drop



Young's equation

$$\cos\theta = (\sigma_{sv} - \sigma_{LS}) / \sigma_{LV}$$

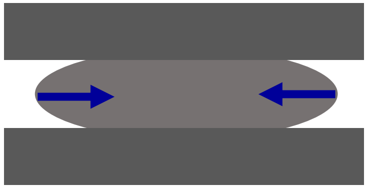
- **S/L interface**
Dissolution of the solid phase, infiltration of the liquid into the solid, formation of interfacial compounds, adsorption of active elements (e.g. Ti, Cr)

- **L/V interface**
Evaporation, adsorption of gaseous compounds (e.g. oxygen), oxidation

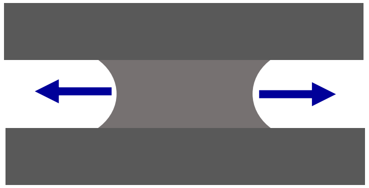
- **S/V interface**
Oxidation-deoxidation of the surface, selective evaporation (e.g. SiO)

Wetting and joining

A braze alloy relies on capillarity to distribute the liquid alloy along the adjoining interfaces

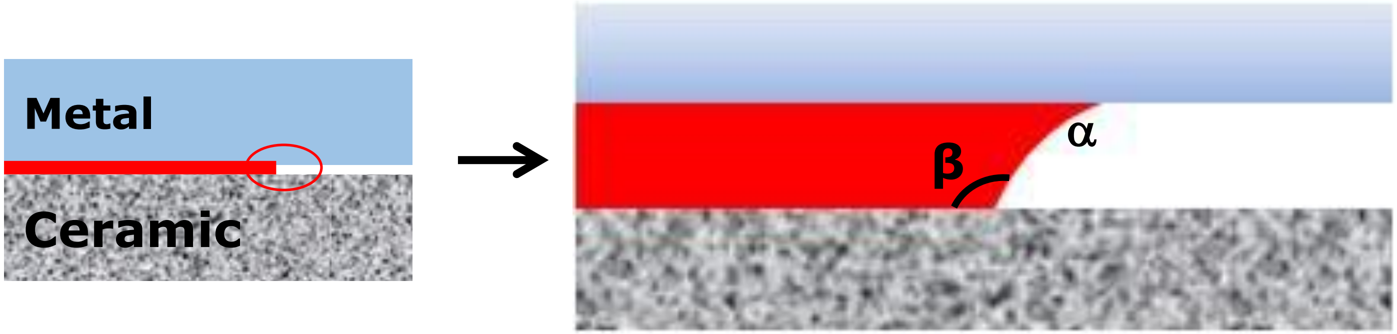


Non-wetting both substrates



Wetting both substrates

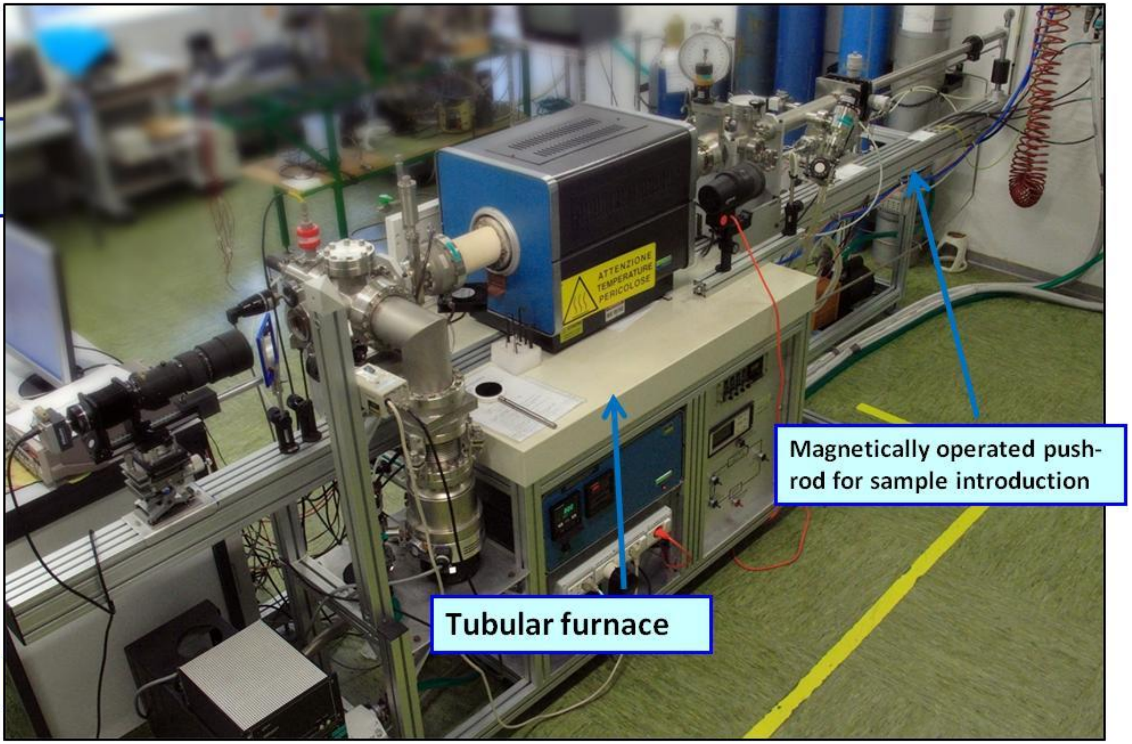
Conditions for spontaneous liquid infiltration



$\alpha + \beta < 180°$

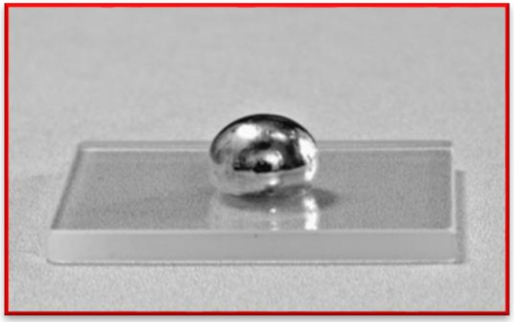
Experimental apparatus for sessile drop tests

ASTRAView software



Digital Camera

Example of sessile drop during test (θ<90°)



Example of sessile drop after cooling (θ>90°)

$T_{max} = 1600°C$

Working atmosphere: Vacuum: 10^{-4} Pa @1500°C or gas mixtures (e.g. Ar, Ar/H_2, O_2, CO/CO_2) with analysis of PO_2 at inlet and outlet.

Fast heating and cooling: samples introduced into the experimental chamber through an external manipulator.

Analysis of wetting kinetics and surface tension by software (AstraView).

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Nature of interfacial reactivity between SiC and liquid metals

Reactivity for pure metals	Metal feature	Ex.
None		Pb, Au, Ag, Sn.
Me + SiC → silicide + C	No stable carbide, stable silicides	Ni, Co, Fe, Cu
Me + SiC → Si + carbide	Stable carbides	V, Al, Nb
Me + SiC → silicide + carbide	Stable carbides, stable silicides	Zr, Hf, Cr, Ta, W, Ti, Mo

The reactivity of liquid metals with SiC must be controlled in order to select the nature of the reaction products and the thickness of the reaction zone.

Dissolution in metal/SiC systems

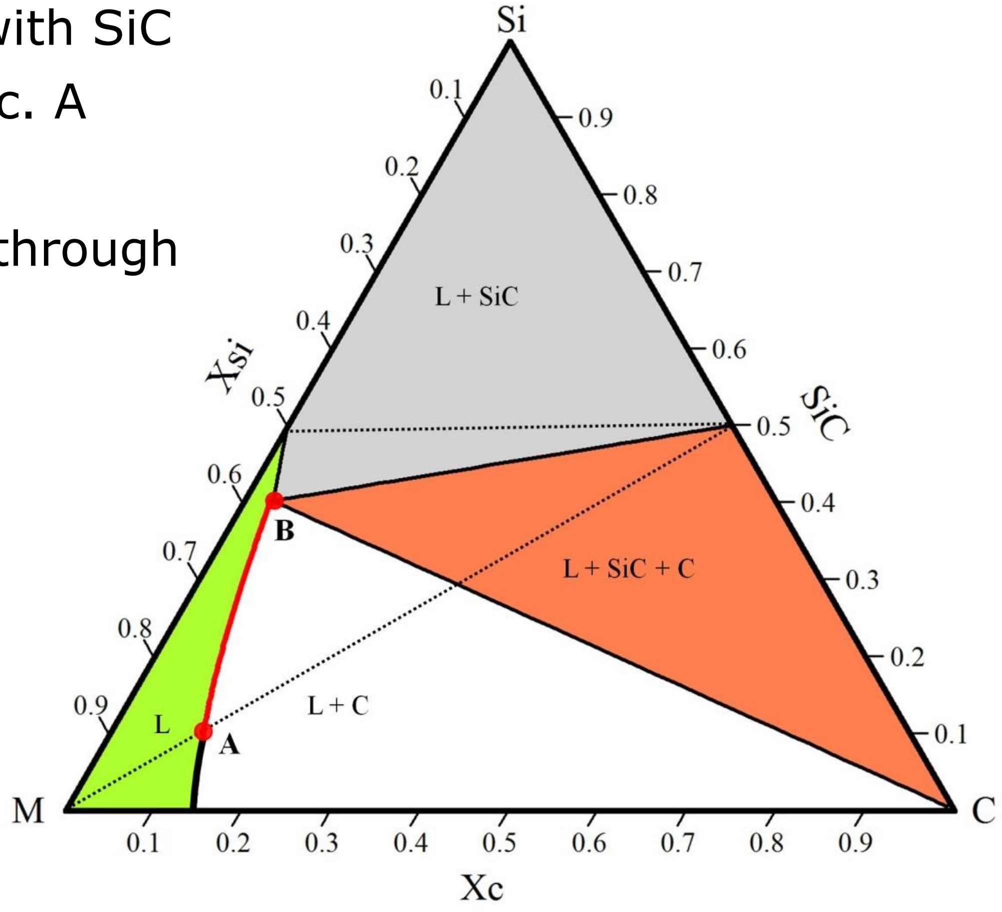
Schematic isothermal section of M–Si–C phase diagram
(M = metal which does not form stable carbide)

1 - M (e.g. Co, Ni) in contact with SiC

2 - Dissolution of SiC until conc. A

3 - Precipitation of C

4 - Dissolution continues to B through the red line



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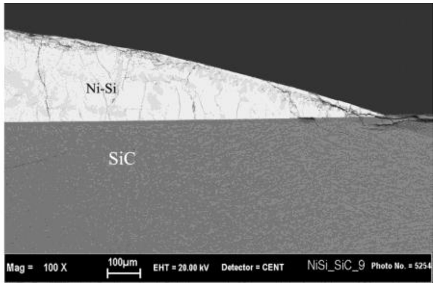
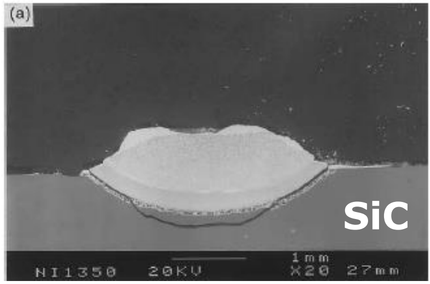
Ni-Si alloy:**
NON reactive



Pure Ni*:
reactive (dissolutive)



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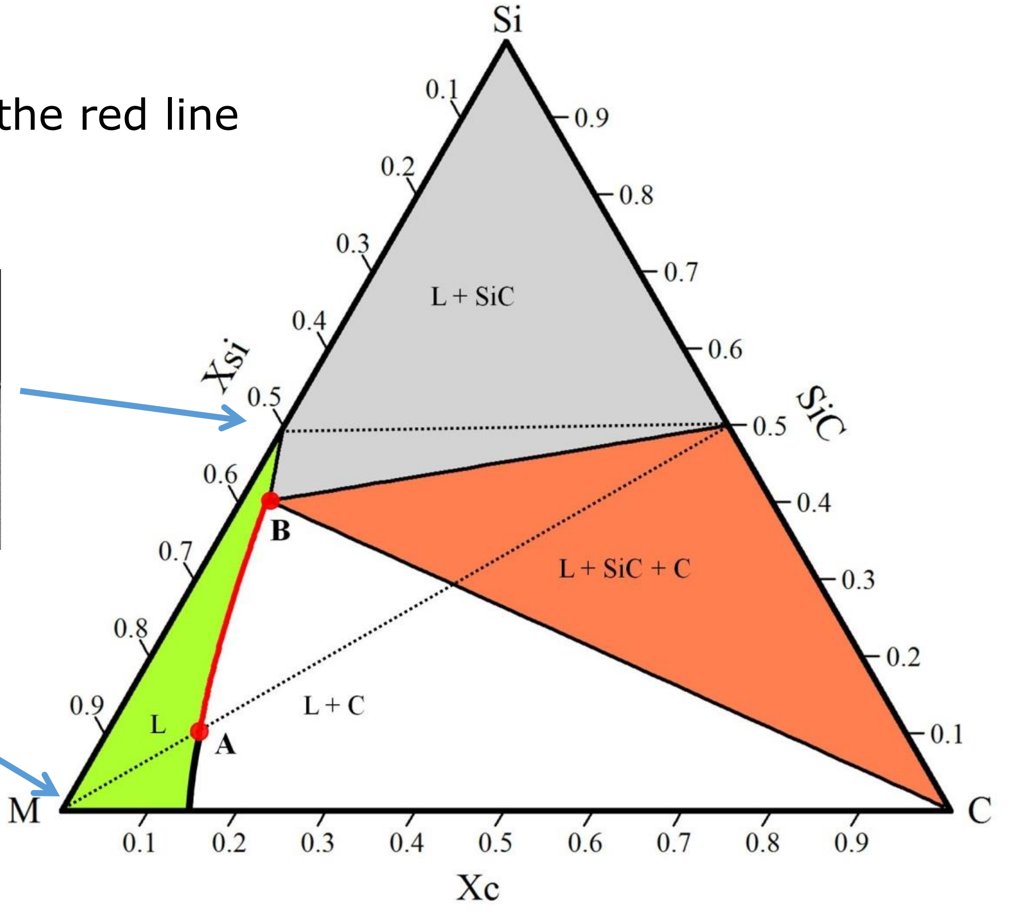
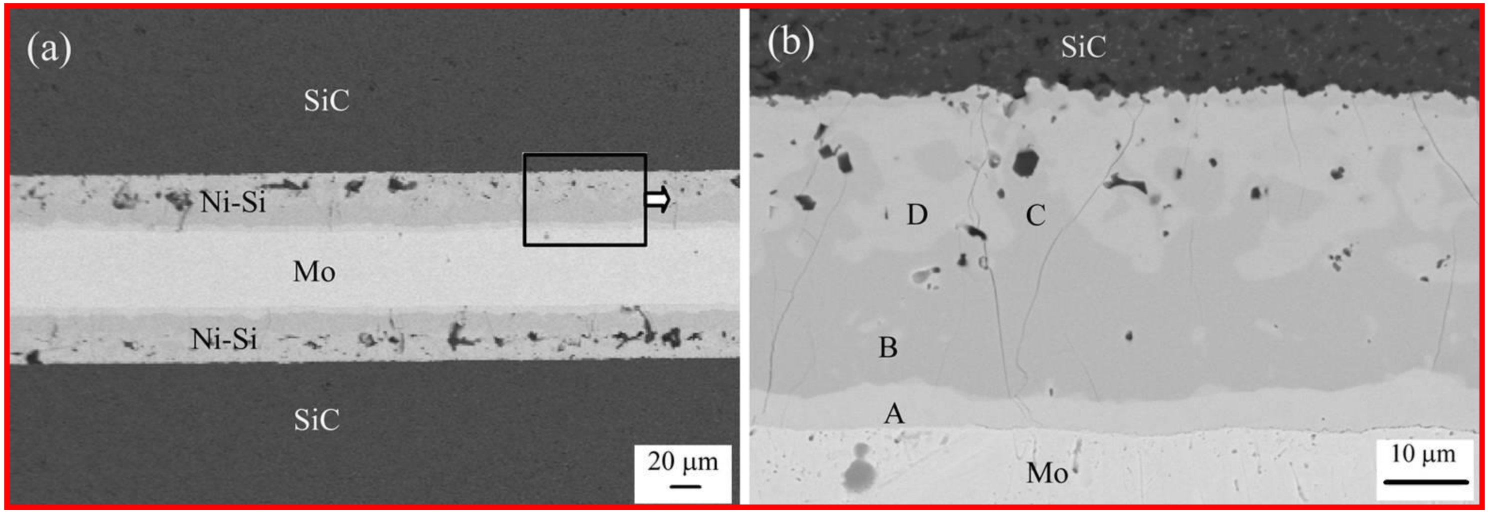
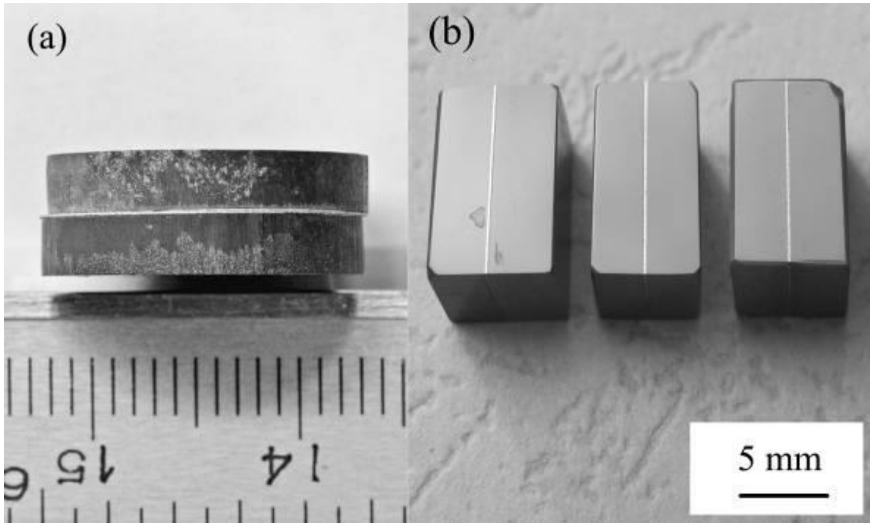


* Rado C., Kalogeropoulos S., Eustathopoulos N., Acta Materialia **47** (1999) 461-473
Liu G.W., Valenza F., Muolo M.L., Qiao G.J., Passerone A., J. of Materials Science **44 (2009) 5990-5997

SiC-SiC joining with Mo interlayer, 1350°C – 10 min





Liu G.W., Valenza F., Muolo M.L., Passerone A., SiC/SiC and SiC/Kovar joining by Ni-Si and Mo interlayers, J Mater Sci 45(16) (2010) 4299-4307

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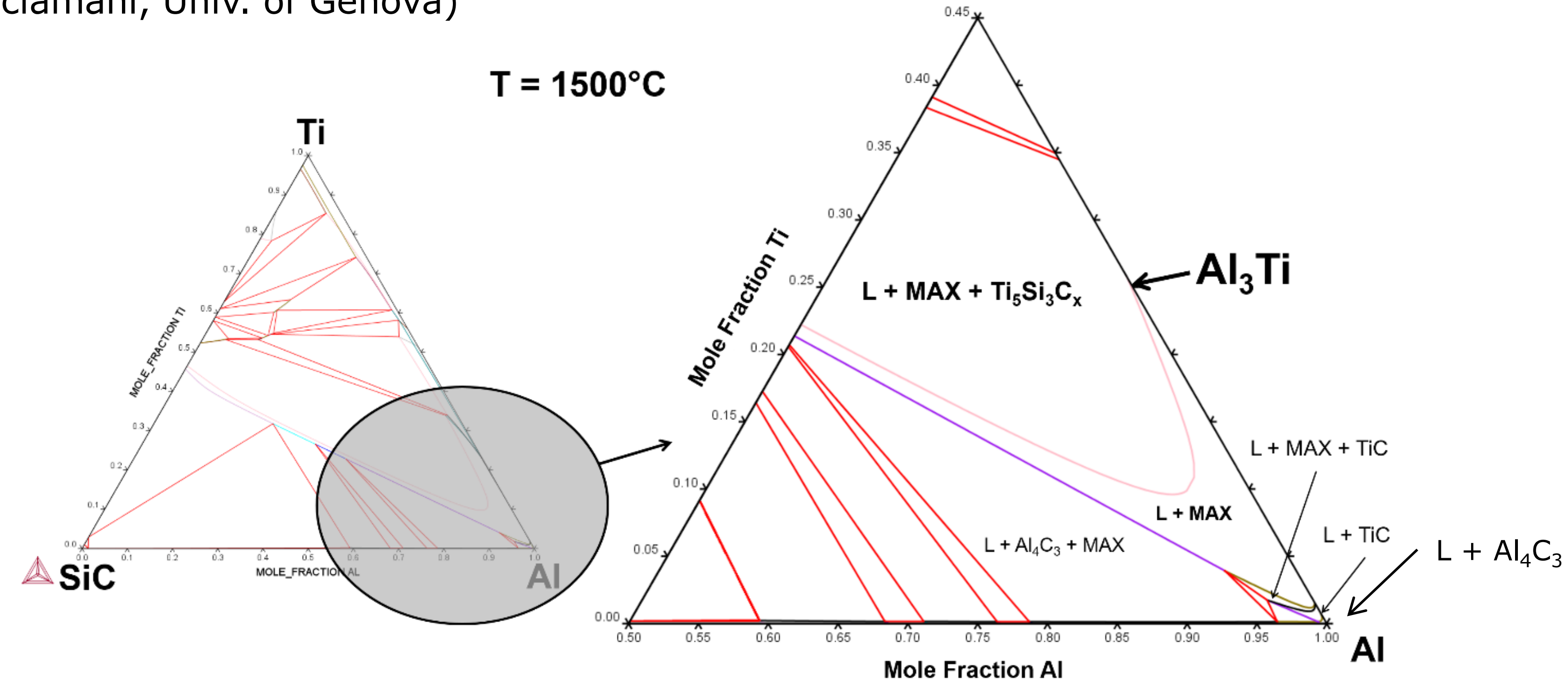
Wetting of SiC by Al + Ti

SiC-Al-Ti system

SiC-Al-Ti isothermal section – Al-rich corner

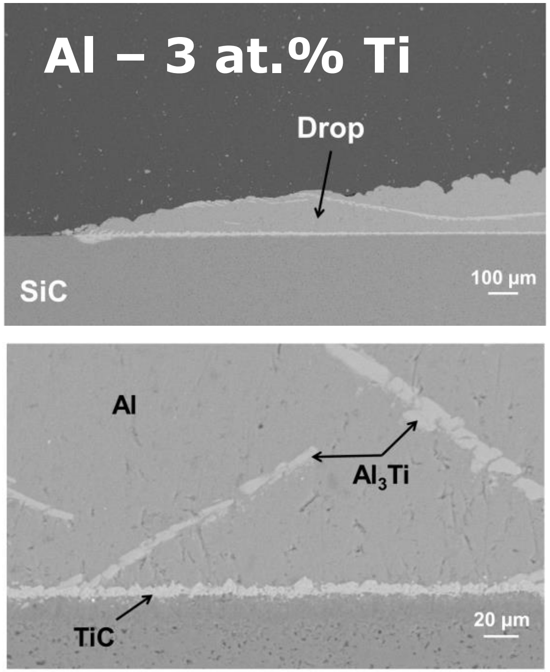
(determination of multicomponent phase diagam by CALPHAD, collaboration with prof. Cacciamani, Univ. of Genova)

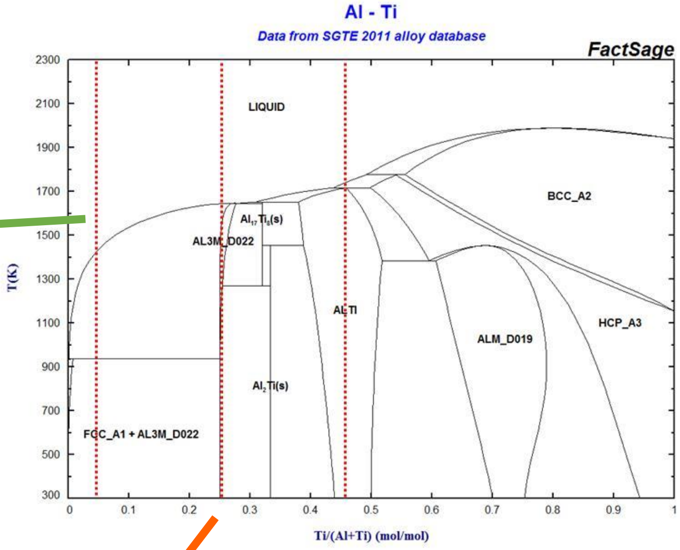


Main goal: to obtain SiC/SiC joints through the **in-situ formation of a $Ti_3Si(Al)C_2$ phase** starting from simple Al-Ti interlayers. Similarly to a **transient liquid phase bonding process**, the in-situ formation of the desired phase at the process temperature would allow to overcome the thermal stresses associated with the solidification by cooling of the interfacial phases over a broad temperature range.

S.Gambaro, F.Valenza, G.Cacciamani, M.L. Muolo, A.Passerone, F.Toche, R.Chiriac, O.Dezellus, High-Temperature-Reactivity evaluation of Al-Ti alloys in contact with SiC, submitted to Journal of Materials SCience

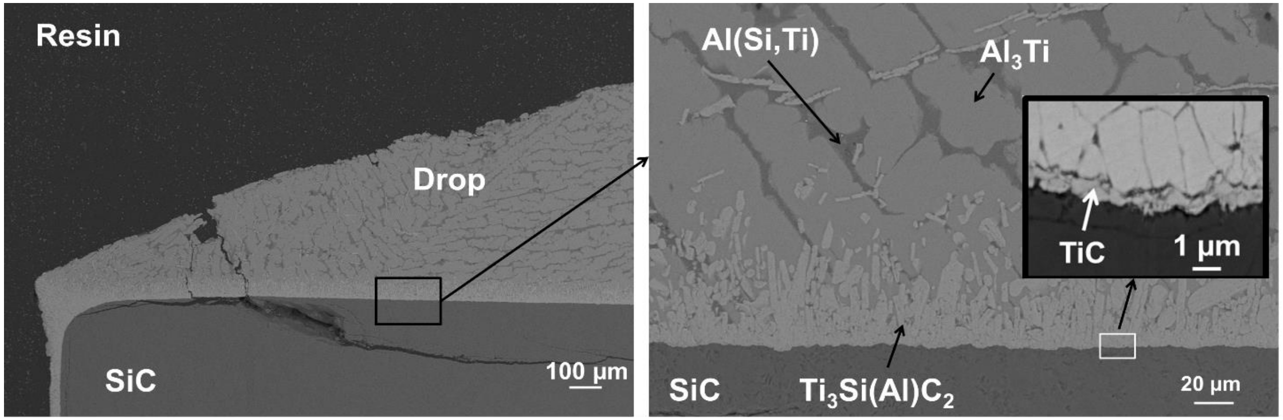
Wetting results: microstructures





T = 1500°C

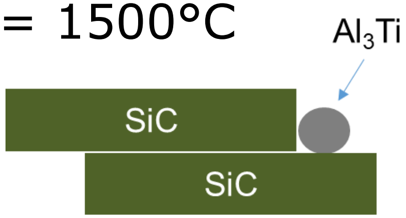
Al – 25 at.% Ti (Al_3Ti)



Valenza, F., Gambaro, S., Muolo, M.L., Salvo, M., Casalegno, V., *Wetting of SiC by Al-Ti alloys and joining by in-situ formation of interfacial $Ti_3Si(Al)C_2$*, Journal of the European Ceramic Society, 38 (2018) 3727-3734

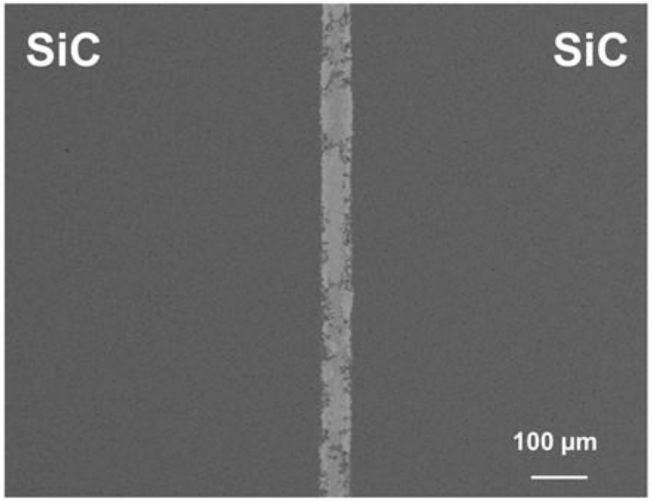
Joining of SiC by in-situ formation of $Ti_3Si(Al)C_2$ phase

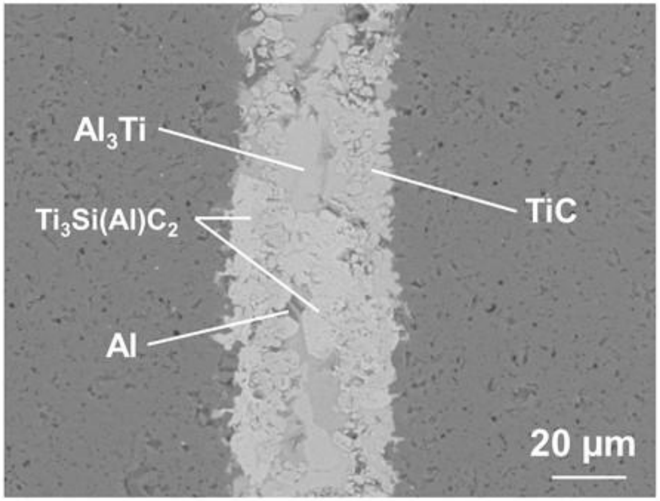
T = 1500°C



Infiltration of Al_3Ti

No cracks
Resulting phases:
$Ti_3Si(Al)C_2$, Al_3Ti, Al

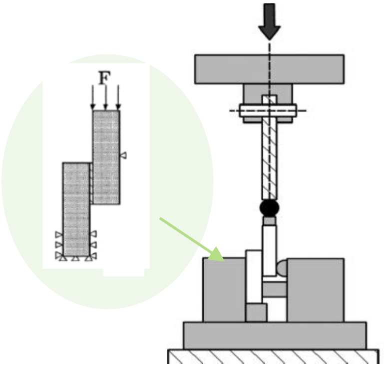




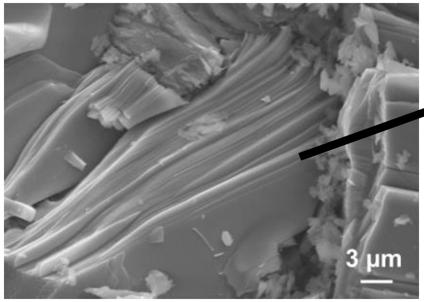
Mechanical tests

at Politecnico di Torino

τ = 296 MPa ± 20

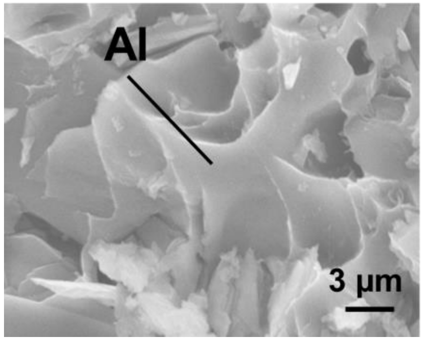


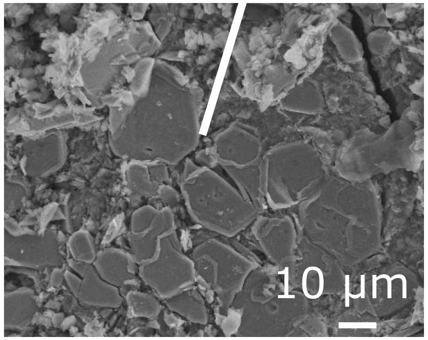
Fracture surfaces



Kink bands in $Ti_3Si(Al)C_2$

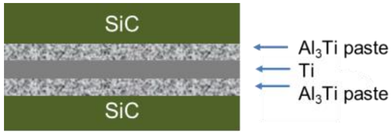
Transgranular fracture in TiC

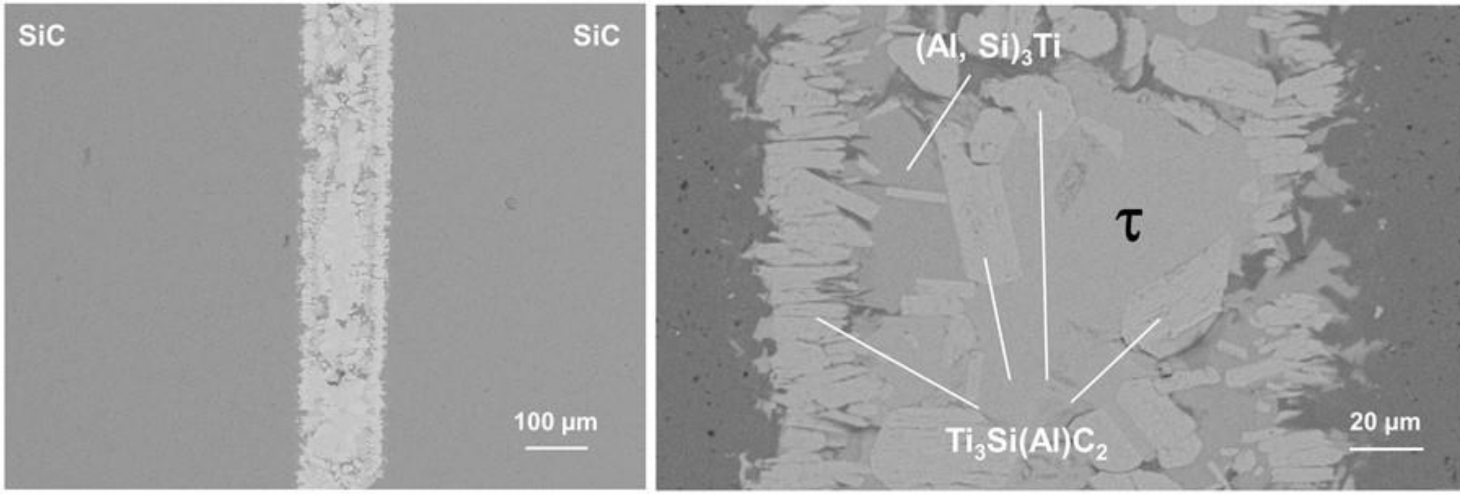




Joining of SiC by in-situ formation of $Ti_3Si(Al)C_2$ phase

Al_3Ti / Ti foil / Al_3Ti





No cracks

Thickness: ~ 50 µm

Resulting phases: $Ti_3Si(Al)C_2$, $(Al, Si)_3Ti$, τ phase in the Al-Si-Ti phase diagram (Al 13, Si 34, Ti 53 at.%)

τ = 85 MPa

Nature of interfacial reactivity between SiC and liquid metals

Reactivity for pure metals	Metal feature	Ex.
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Wetting of Sn-Ti alloys on SiC

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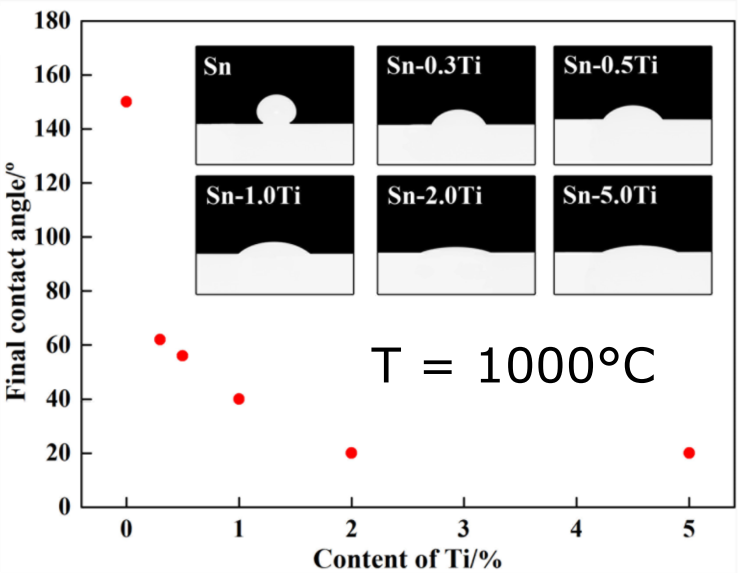

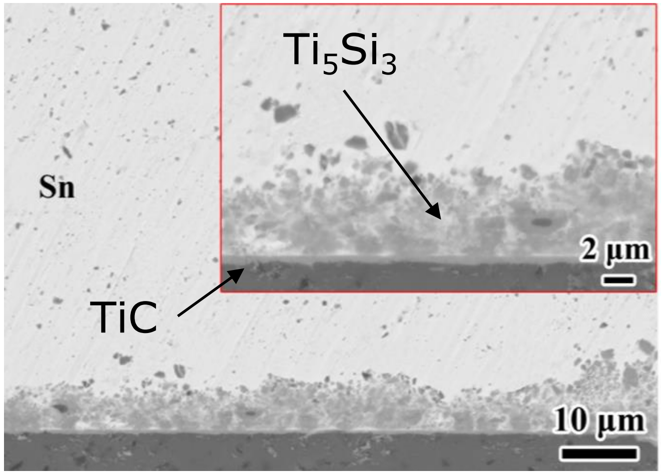

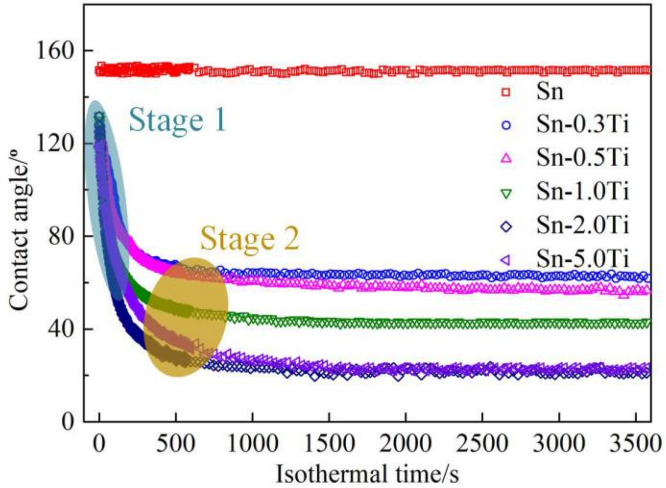

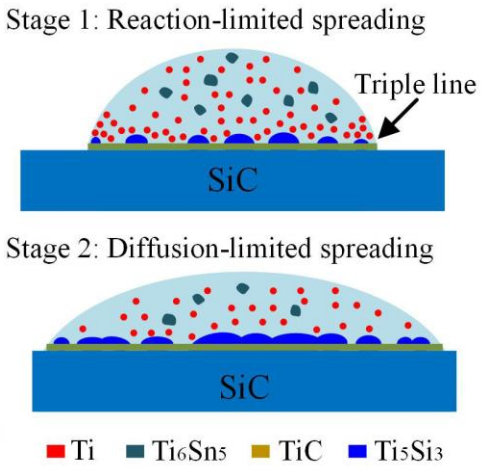

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Conclusions

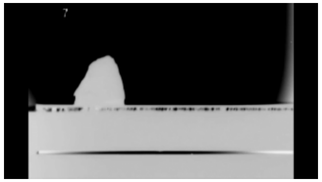
The study of metal-ceramic interfaces at high temperature is relevant in applications which involve the combination of these two class of materials (e.g. production of metal-ceramic joints, casting of special alloys etc.).

Joining processes for ceramics based on liquid phases (e.g. brazing) need the knowledge of liquid-solid interfacial phenomena.

In particular, it is fundamental to investigate:
- the wettability of liquid alloys on ceramics
- the evolution of interfaces
- the interfacial reactions and how to control them

Phase diagrams could assist the choice of the most proper braze compositions in order to promote wetting, to avoid deleterious reactions (e.g. dissolution) or to get new phases.

Active metals could be used to enhance the wetting behaviour of non-wetting alloys.



Acknowledgments

S. Gambaro, M.L. Muolo, A. Passerone, F. Valenza: CNR-ICMATE

Prof. G. Cacciamani (Univ. Genova): CALPHAD calculations

Wei Fu (visiting Ph.D student from Harbin Inst. of Technology): Sn-Ti/SiC work

Prof. GW Liu (Jiangsu Univ. – China): Ni-Si/SiC work

Prof. M. Ferraris, Prof. V. Casalegno, Prof. M. Salvo (Politecnico di Torino): mechanical testing

Fundings

The research leading to these results has received funding from:

European project ADMACOM (Advanced manufacturing routes for metal/composite components for aerospace, www.admacomproject.eu); EU-FP7 2007-2013, grant agreement 609188.

Italian Flagship Project, RITMARE (La ricerca italiana per il mare); Italian Government, MIUR.

INSURFCAST, «Innovative Surfaces for Superalloys casting processes», ERA-LEARN 2020 support action funded by EU-H2020, M-ERA.NET Joint Call 2016

Thank you
for your attention!