International Spring School on Forefront Alloys and Advanced Materials for Extreme Conditions

15 – 17 May 2017

Sardinia, Italy

Brazing of SiC materials for space applications

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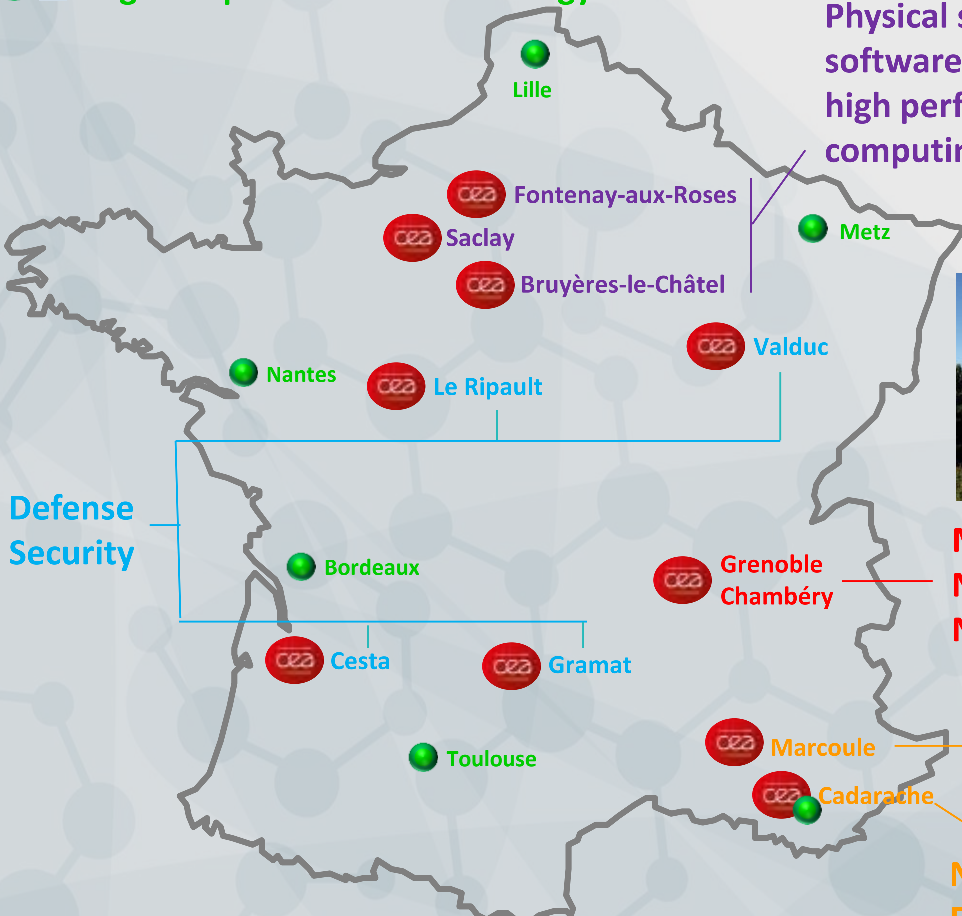
- Examples of applications
- SiC / metal brazing
- Conclusions

CEA & LITEN

CEA : 10 Research centers in France

6 Regional platforms of technology transfer



Physical sciences, software technologies high performance computing, biomedicine

Defense Security

Micro-Nanotechnologies
Nanobiotechnologies
New technologies for energy

Nuclear :
Nuclear fuel cycle
and waste management

Nuclear :
Fusion, fission, Bioenergies, solar

Conception and Assembling Laboratory

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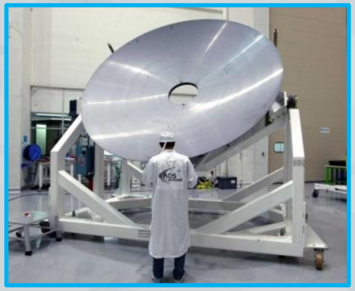
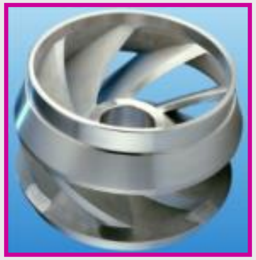
Brazing



HIP

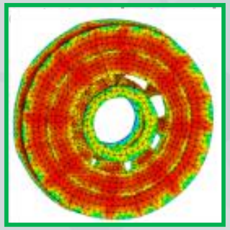
Diffusion bonding

Powder metallurgy



Simulation

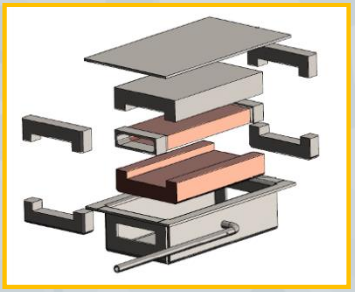
Processes (HIP, brazing)
Sizing
Behaviour law



Ansys
Abaqus
Cast3M

Conception

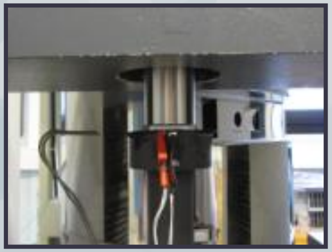
Heat exchanger / reactors
Complex components



Solidworks
Catia

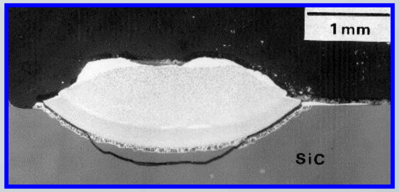
Mechanical testing

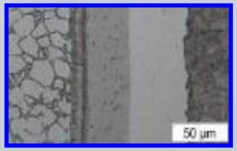
Tensile, torsion, creep
toughness, fatigue
Test @ HT 1400° C
air/vacuum/He/H_2
H2 embrittlement



Materials

Metallography
Physico-Chemistry
Reactivity
Spreading kinetic





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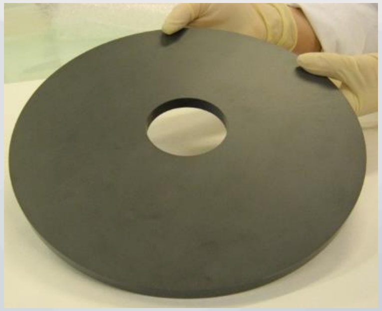
Interest of SiC for extreme conditions

▪ The Silicon Carbide SiC is a high performance ceramic :

- Use at 1450°C under air, 1800°C in inert atmosphere* (no melting of SiC)
- Low density ρ (3,15*)
- Low Coefficient of Thermal Expansion CTE
  ($2{,}2\ 10^{-6}\ K^{-1}$ at 20°C, $4{,}8\ 10^{-6}\ K^{-1}$ at 500°C, $6\ 10^{-6}\ K^{-1}$ at 1000°C *)
- High thermal conductivity (180 W/m.K at 20°C, 66 W/m.K at 500°C *)
- High thermal stability (thermal conductivity / CTE)
- High strength (bending strength 400MPa at 20°C, tensile strength 210 MPa at 20°C*)
- High stiffness E/ρ (E young modulus 420 GPa from 20 to 1000°C*)
- high thermal shock resistance, high heat resistance
- High corrosion resistance, hardness, wear resistance
- Low nuclear activation, semiconductor.



⇩

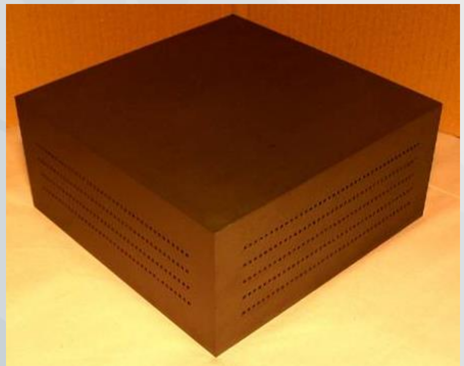
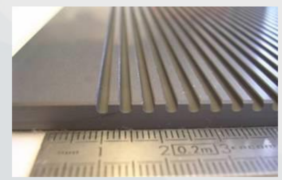
- Sintered SiC is interesting for applications in the field of energy
- Sintered SiC is used in space applications (mirror for telescopes, structural parts)

*Reference : Boostec® SiC (group Mersen)

Manufacturing SiC components

- **How to manufacture large and / or complex shape SiC components ?**

⇨ **Joining of simple shape and small SiC elements**

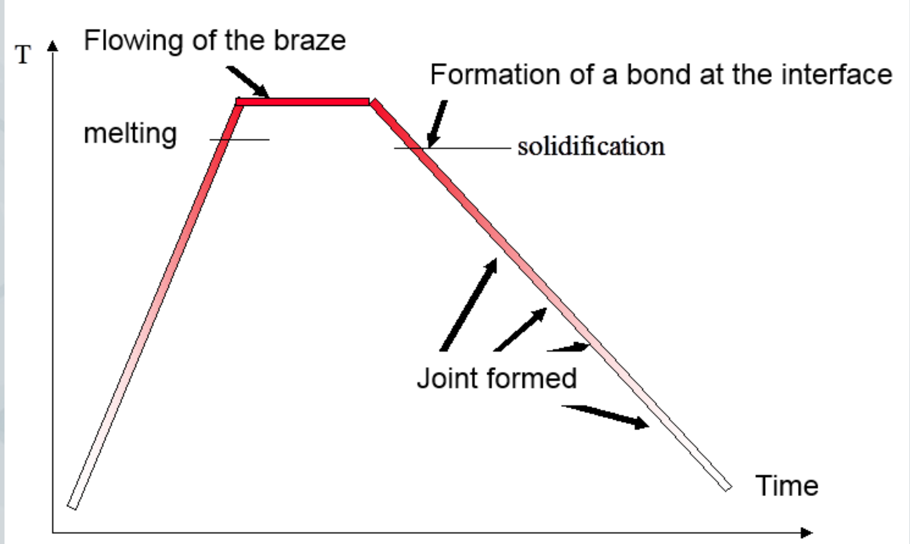
SiC plates with channels joined by brazing to form a compact SiC heat exchanger (CEA).

- **Joining methods :**
  - Mechanical attachment (use of rivets and screws, holes in the ceramic) not well suited to SiC
  - Adhesive bonding by using polymeric materials : working temperature range limited and sensitive to environment
  - Diffusion bonding : needs a pressure and an excellent surface contact
  - Welding : involves melting of pieces edges
  - **Brazing** : melting of a filler metal without melting of the materials to be joined

Fundamental parameters of brazing

- Brazing : method for joining ceramics and metals using a liquid metal or alloy as bonding material (the filler alloy or braze) without melting of the substrates. The liquid braze flows into the gap between the susbtrates and then solidifies during cooling to form a permanent bond.

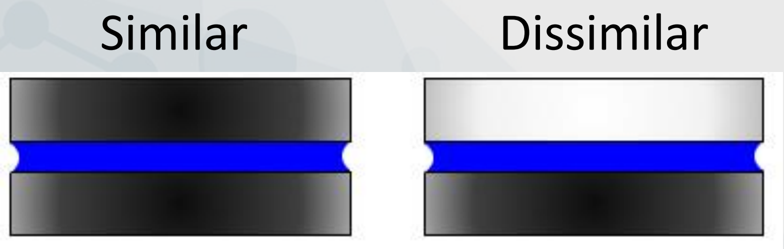
- $T_{braze\ melting} < T_{brazing} < T_{substrate\ melting}$



Fundamentals parameters of brazing

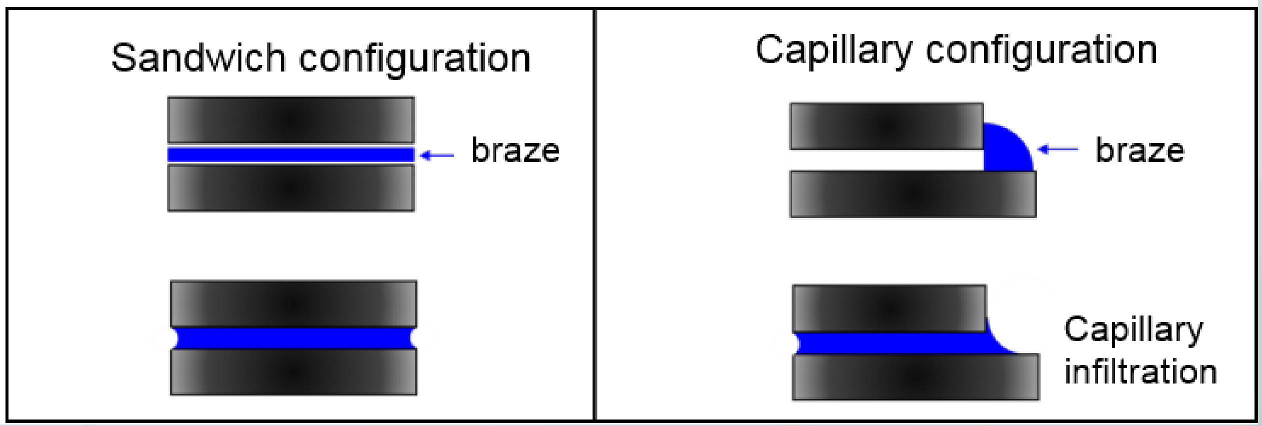
- Different types of joints

Compact SiC heat exchanger



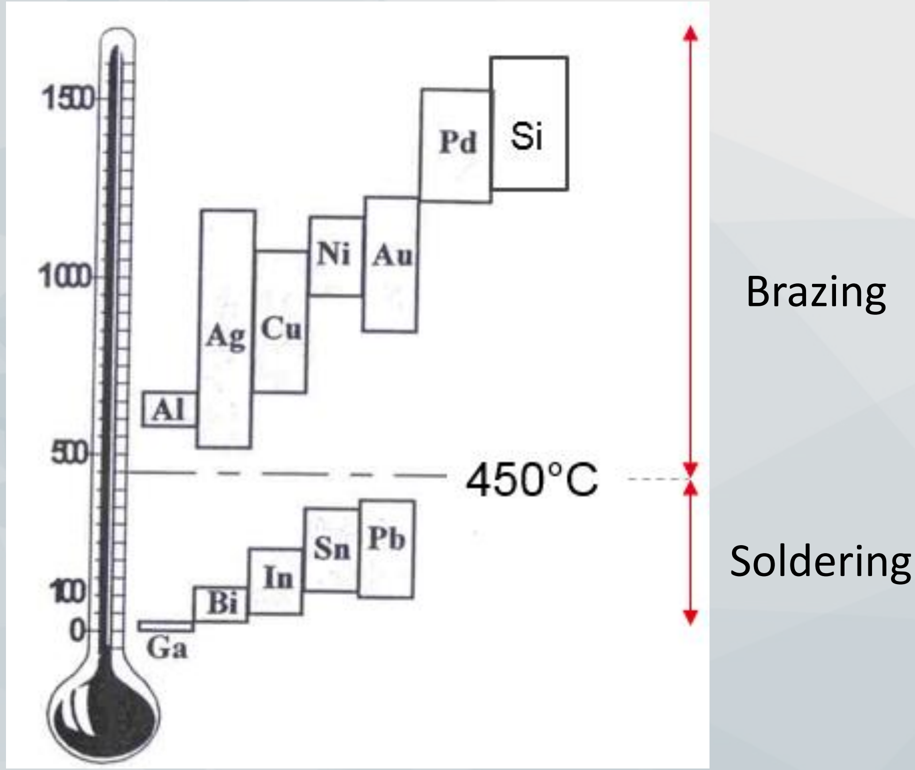
Brazing CFC / Cu

- Brazing configurations : « sandwich » or capillary



Fundamentals of brazing

- Brazing and soldering :
  - Soldering : $T_{melting}$ braze < 450°C
  - Brazing : $T_{melting}$ braze > 450°C

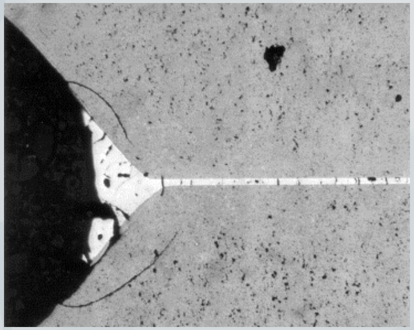


Fundamentals parameters of brazing

- Chemical and physical compatibility
  - Good wetting of the surfaces to be brazed (a good flowing over the surfaces)
  - Formation of a strong braze / substrate interface

- Thermomechanical compatibility :
  - Low stresses in the region of the joint

SiC/Inco – $T_{brasage}$ 800°C



Cracks around the fillets

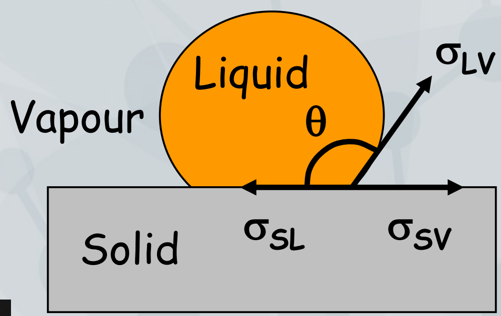
Fundamental parameters of brazing

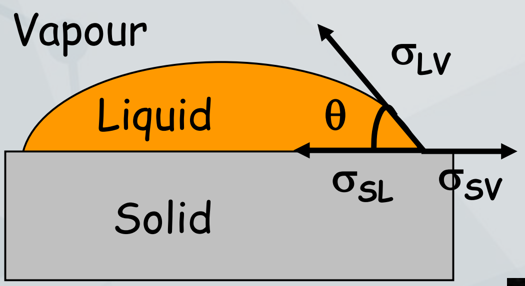
- Wetting and contact angle θ :

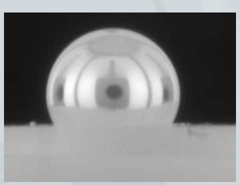
Equilibrium contact angle θ at a solid / liquid / vapour junction (triple line)

depends on the three interfacial energies σ_{SV} , σ_{SL} , σ_{LV} :

$$\cos\theta = \frac{\sigma_{SV} - \sigma_{SL}}{\sigma_{LV}}$$ (Young equation)

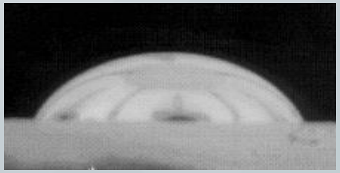






$\theta > 90°$
No wetting

$\theta < 90°$
Wetting



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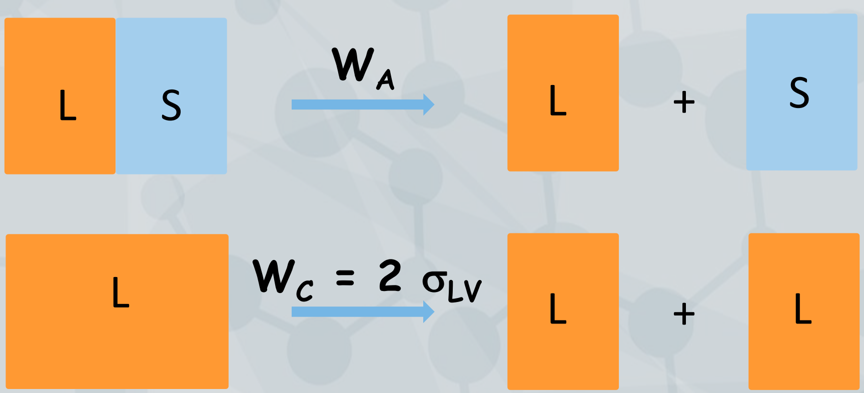
Fundamental parameters of brazing

- Work of adhesion Wa : $W_A = \sigma_{LV} + \sigma_{SV} - \sigma_{SL}$
  energy to separate reversibly a unit area of a solide (S) / liquid (L) interface and create two new surfaces (solid / vapour and liquid / vapour).
- Wetting results from a competition between liquid /solid adhesion forces that promote a good wetting and cohesion forces in the liquid that minimise the formation of the liquid /solid interface.



W_c work of cohesion

$$\cos\theta = \frac{W_A}{\sigma_{LV}} - 1$$

ADHESION (W_A)

COHESION (σ_{LV})

Fundamental parameters of brazing

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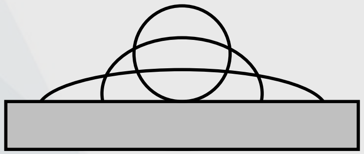
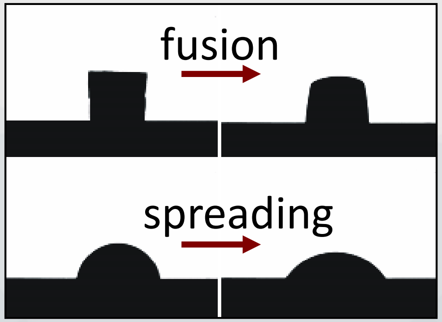
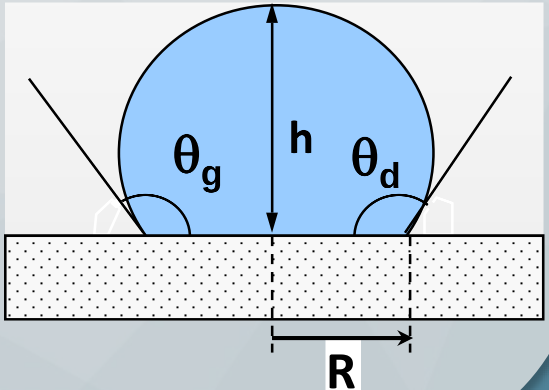
Sessile drop method :



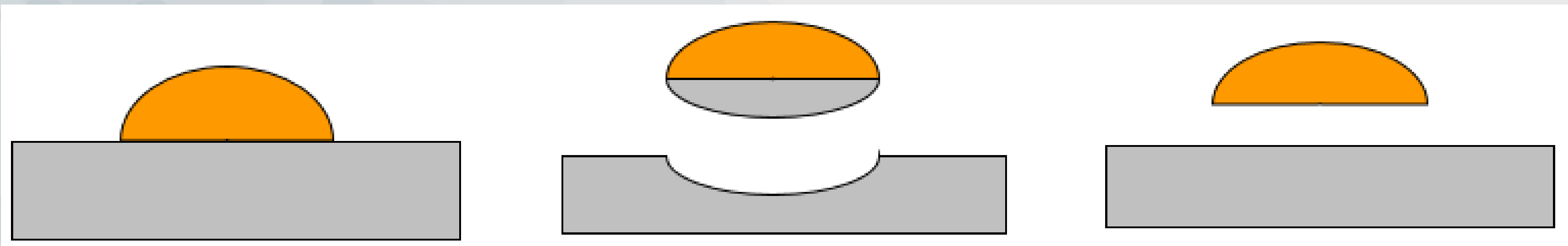
- Small piece of solid braze (max : 100 mg) is placed on a substrate and then heated above its melting point
- The spreading of the drop over the substrate is observed *in situ* through a window in the furnace and the drop profile is recorded with a CCD camera.
- Information derived from the drop profile
  - Contact angles θ ($\theta = f(t, T)$)
  - Radius of the substrat contact area R
  - Height of the drop h





Fundamental parameters of brazing

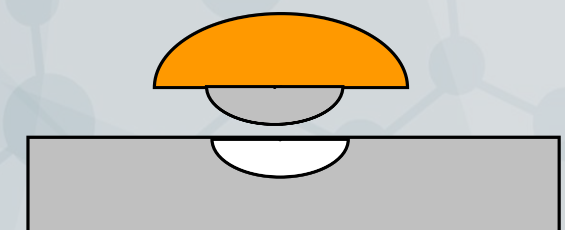
- Mechanical behaviour after solidification : adherence drop /substrate



No failure
→ strong interface

Cohesive failure
→ strong interface

Adhesive failure
→ weak interface



Mixte failure
→ heterogeneous interface

Fundamental parameters of brazing

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- NON-REACTIVE WETTING / REACTIVE WETTING

Non reactive system

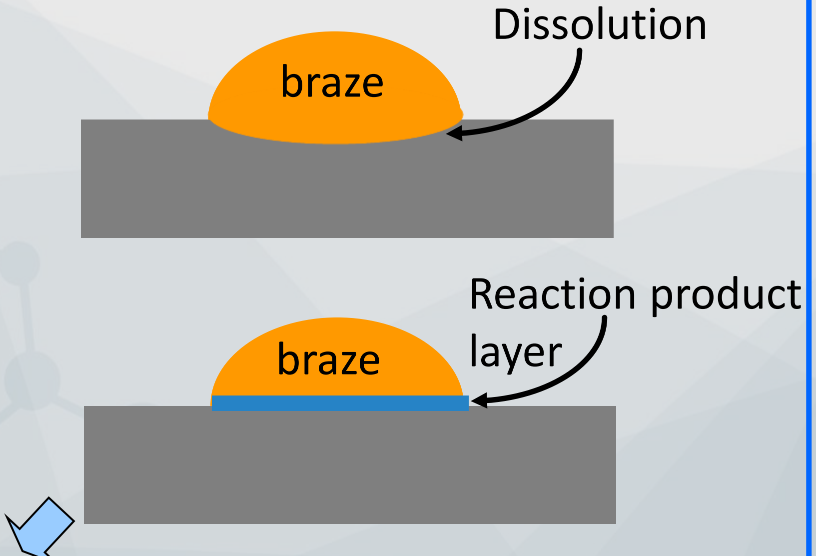
No reaction product, no dissolution of the solid substrate in the liquid braze. The interfacial energies are constant during the experiment, the spreading is controlled by the viscosity.

Liquid metals :
low viscosity η ~ 1 mPa.s
spreading time 10^{-1} to 10^{-2} s (v : 1m/s)

Glasses :
high viscosity η > 10 Pa.s
spreading time > 10^{2} s

Reactive system

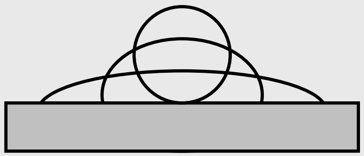
- Dissolution of the solid in the liquid braze
- Formation of a reaction product



Wetting depends on the new compound formed at the interface : less or more wettable than the substrate.

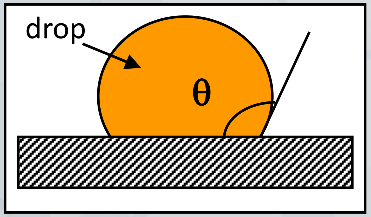
Fundamental parameters of brazing

- **Key parameters controlling the wetting :**
  - Temperature : θ ↗ when T ↘
  - Time t (kinetic)
  - Atmosphere in the furnace : vacuum or gas, vacuum level, PO_2, pressure, nature of the gaz (Ar, He, H_2), metallic or graphite furnace
  - Roughness of the solid surfaces
  - Impurities at the solid surfaces
  - Composition of the braze : concentration of the active element for example
  - Interfacial reactivity



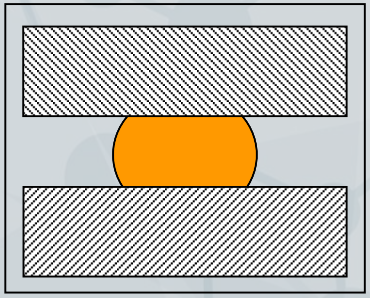
Fundamental parameters of brazing

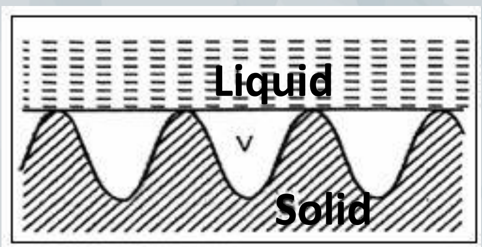
Wetting and brazing



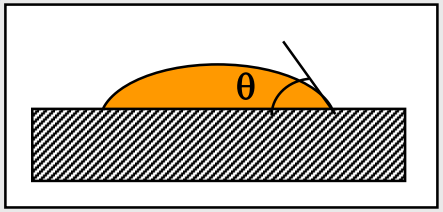
θ > 90°

Non-wetting system



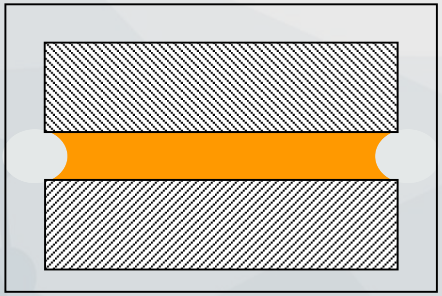


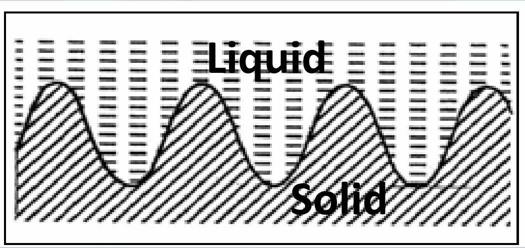
Low effective contact



θ < 90°

Wetting system



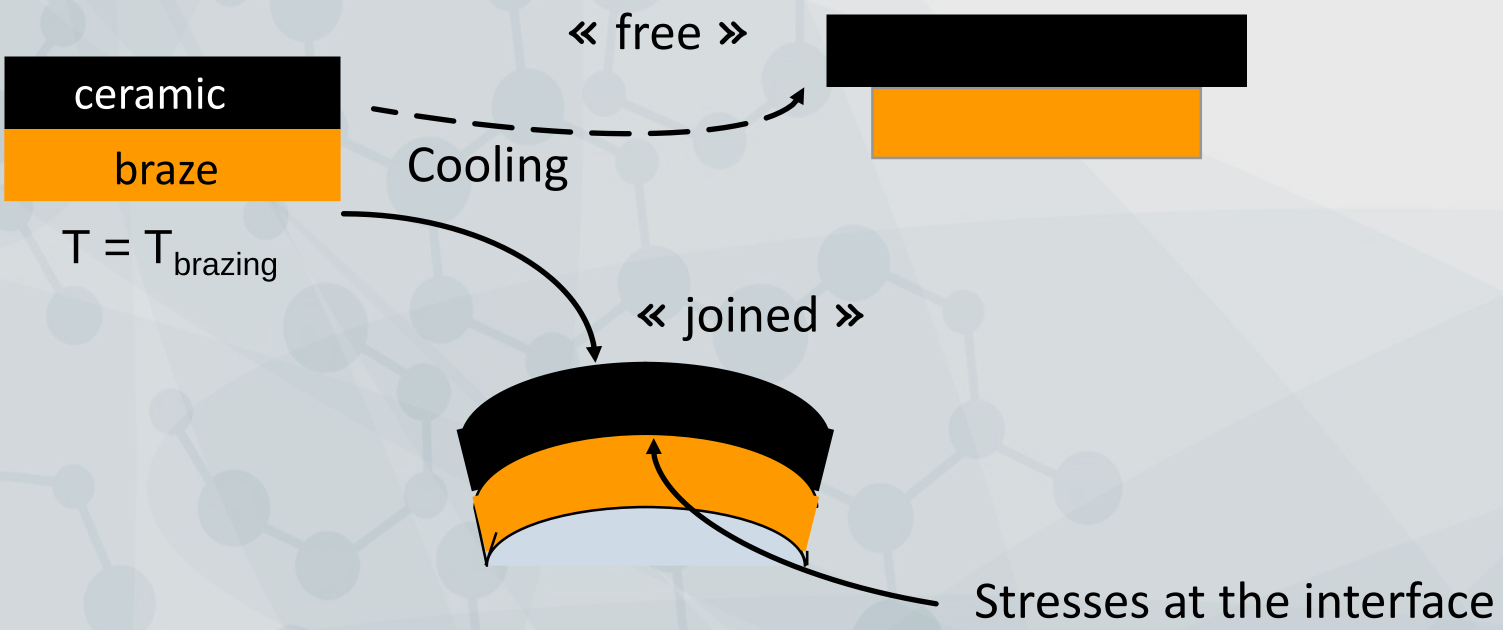


High effective contact

Brazing is possible θ << 90° + Capillary configuration is possible

Fundamental parameters of brazing

- Thermomechanical compatibility : low stresses in the region of the joint ⇨ low mismatch between the thermal expansion coefficients of the braze and the ceramic substrate.



Brazing of SiC components

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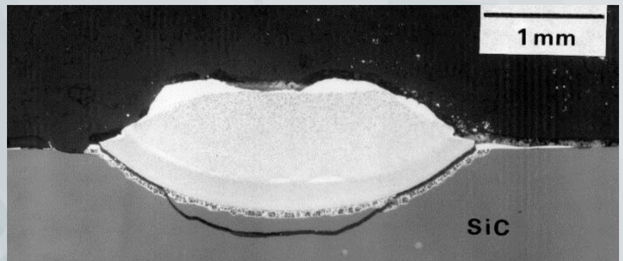
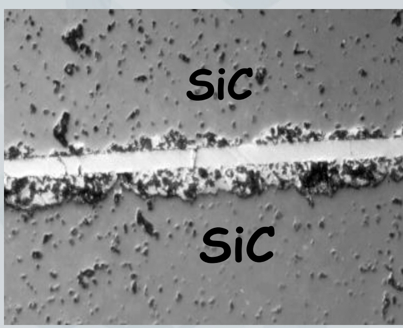
- The main problem of the brazing of SiC : **a strong reactivity with metals**

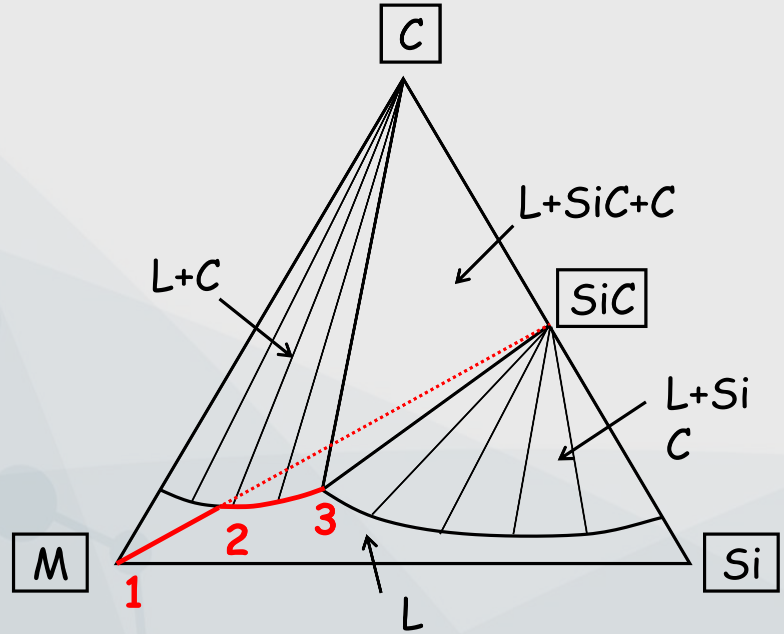


Decomposition of SiC by pure Ni
(sessile drop 15 min / 1350°C)

Joining configuration



Strong reactivity of SiC with the refractory commercial brazes (based on Ni, Co, Pd or Cu…) ⇨ dissolution of SiC, formation of brittle compounds



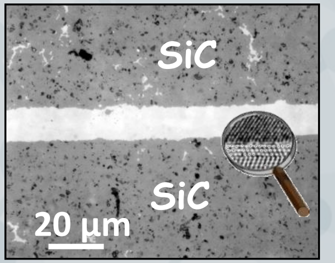
Decomposition of SiC :

- Dissolution of C and Si in M **(1 to 2)**
- Precipitation of graphite **(2)**
- Equilibrium : L + SiC + C **(3)**

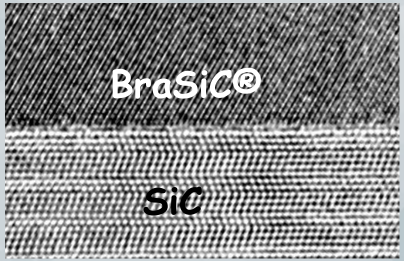
SiC/SiC Brazing : the BraSiC® process

- CEA developed the **BraSiC® process** : **a non reactive** brazing process for joining SiC based materials under vacuum or inert atmosphere (sintered SiC and composites SiC/SiC, C/SiC)

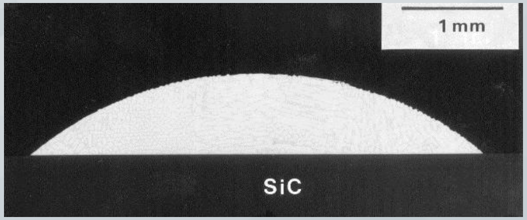
- **More than 20 BraSiC® filler alloys based on Si** developed for a large range of applications (800°C < melting point of the braze < 2000°C)
- CEA has **25 years of experience** on this process : basic research background in collaboration with N. Eustathopoulos at the SIMAP laboratory of Grenoble, R&D for industrial applications (financial support : CNES, Europe, ANR, SAFRAN, Airbus…), technology transfer for space applications.
- **Properties :**
  - No reactivity at the atomic scale. The interface is sharp.
  - Good wetting of SiC $\theta < 60°$.
  - Strong adherence of the SiC / BraSiC® alloy interface.



Optical microscopy



High resolution Transmission Electron Microscopy



Good wetting of SiC

SiC/SiC Brazing : the BraSiC® process

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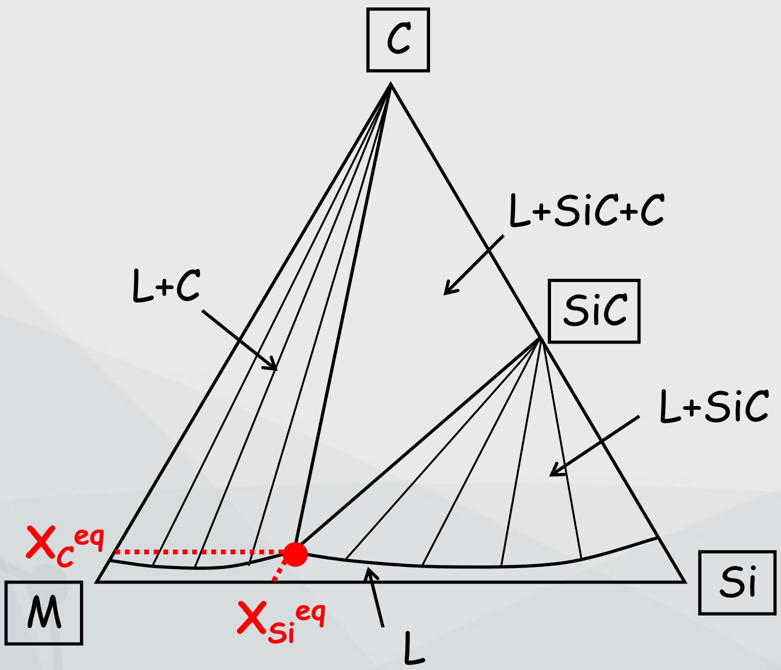
- Non reactive brazing alloys based on Si

For example in the system M-Si-C :

No reactivity for : $X_{Si} > X_{Si}^{eq}$

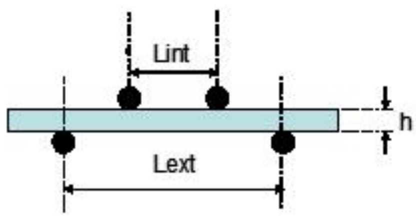


M	T(°C)	X_{Si}^{eq}	Réf.
Ag	1200	0.004	Rado, PhD 97
Cu	1150	0.14	Rado, PhD 97
Ni	1360	0.37	Rado et al., Acta Mater. 99
Co	1500	0.41	Gasse, PhD 96
Fe	1360	0.27	Kalogeropoulos et al., Acta Metall. 95

N. Eustathopoulos
PhD advisor

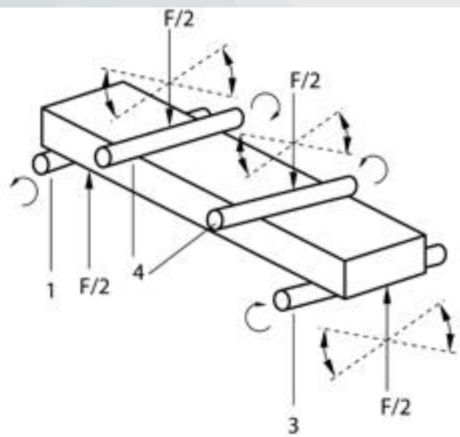
SiC/SiC Brazing : the BraSiC® process

- Mechanical characterisation of the joints
  - Determination of fracture strength by four point bending tests

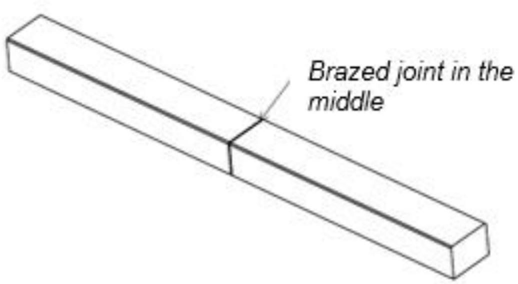


$$\sigma = \frac{3F(L_{EXT} - L_{INT})}{2ah^2}$$

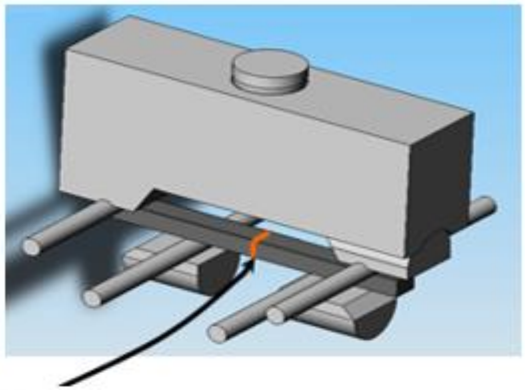
F maximum load (N),
L_{EXT} outer span (mm),
L_{INT} inner span (mm)
a specimen width (mm)
h specimen thickness (mm)



Standard : NF EN 843-1 and ASTM C1211-02



Specimen dimensions : 3 x 4 x 46 (mm³)



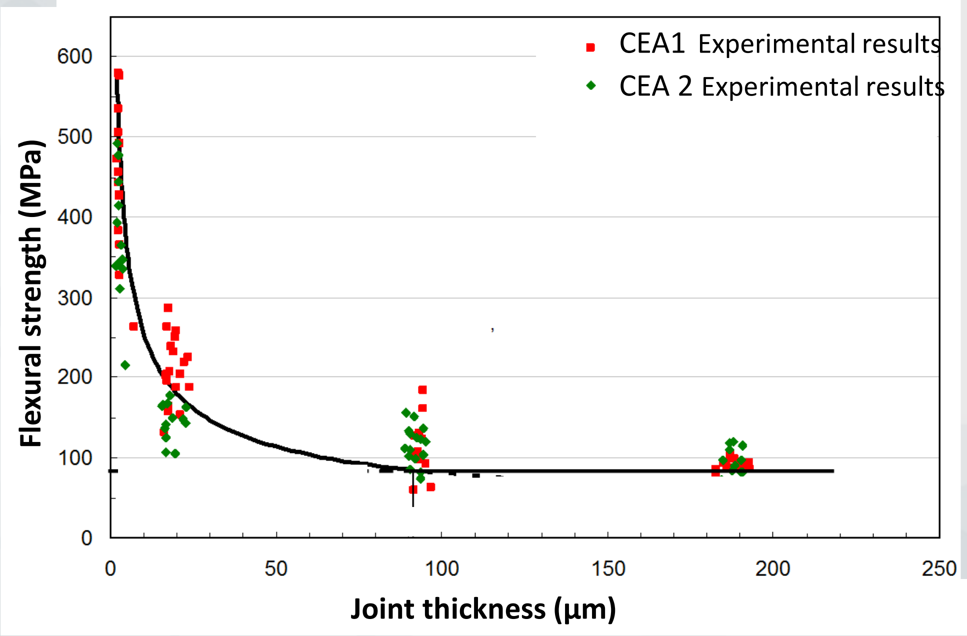
Specimen

Four point jig in alumina

SiC/SiC Brazing : the BraSiC® process

- Mechanical behaviour (2 BraSiC® alloys CEA 1 and CEA2)
  - Flexural strength

Statistical behaviour of the flexural strength depending of the joint thickness (Le Minh Nguyen PhD 2011) .



  - Young modulus : E_{CEA1}=164±8.4 GPa measured by nano indentation (E_{SiC}=420 GPa)
  - Fracture toughness : K_{1C} CEA1= 0.69±0.13 (MPa.$m^{1/2}$)
    K_{1C} SiC = 2.5±0.4 (MPa.$m^{1/2}$)

Examples of applications

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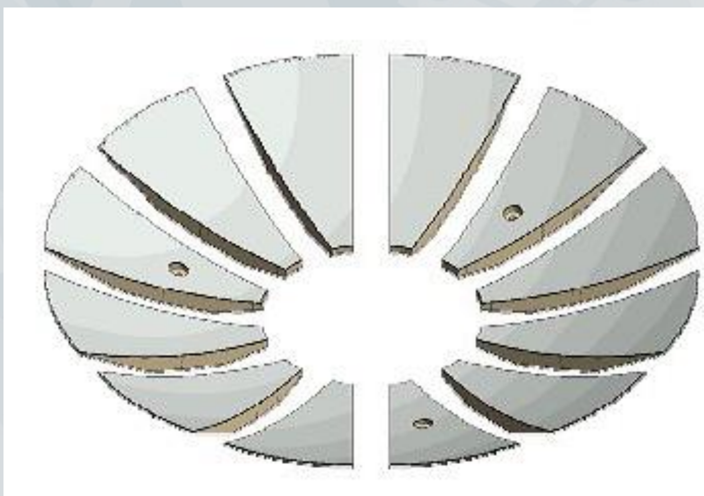
- Technology transfer of the BraSiC® process to the Airbus Defense & Space (EADS Astrium) for the fabrication of SiC components for the telescopes
- Telescope Herschel : manufacturing of the largest space reflector in the world (ϕ 3,5 m) by brazing 12 SiC petals.



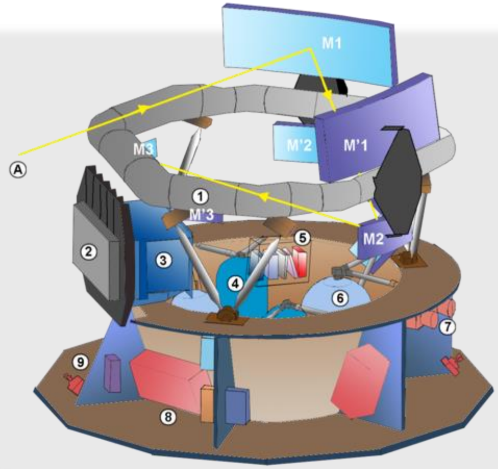
Brazing 12 petals to manufacture a single structure

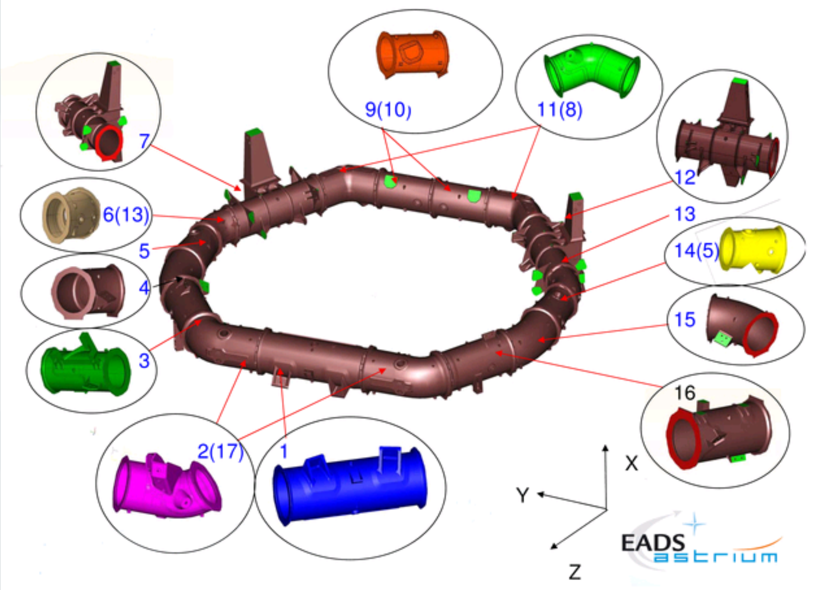
After brazing

Manufacturing of the Herschel mirror by Airbus Defense & Space and Boostec.

Examples of applications

- Telescope GAIA : brazing the torus
  ϕ 3 m (19 SiC elements)



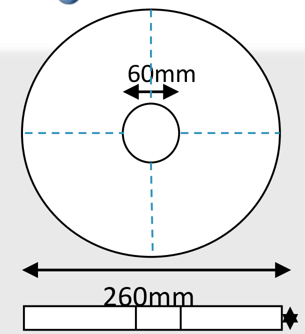


Schematic diagram of the GAIA torus

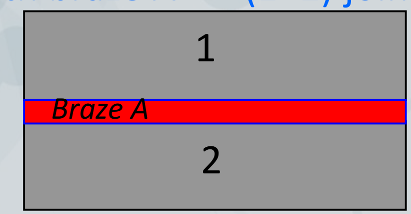
Torus brazed by Airbus Defense & Space and Boostec

Examples of applications

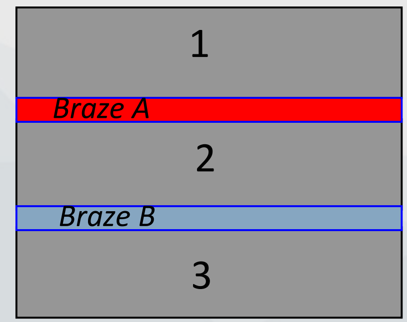
- The **multi-staged brazing** (R&D project at CEA with CNES financial support) : manufacturing and test of a mock-up constituted of 4 quarters of disc

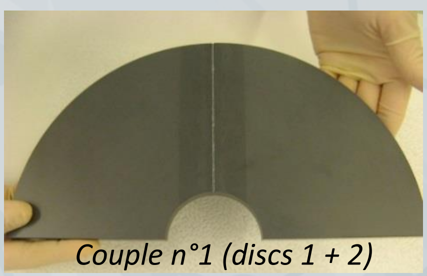


Stage A : Brazing (1) + (2) at 1440°C with braze A ⇨ (1+2) joined

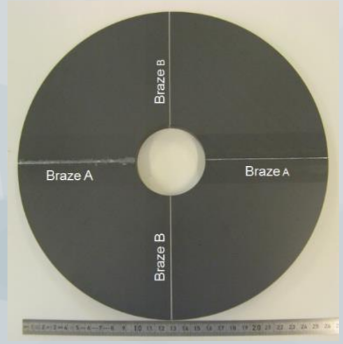


Stage B : Brazing (1+2) + (3) at 1250°C with braze B ⇨ (1+2+3) joined

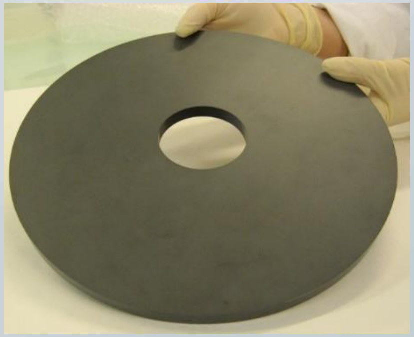



Couple n°1 (discs 1 + 2)


Couple n°2 (discs 3 + 4)



Couples n°1 and n°2 brazed

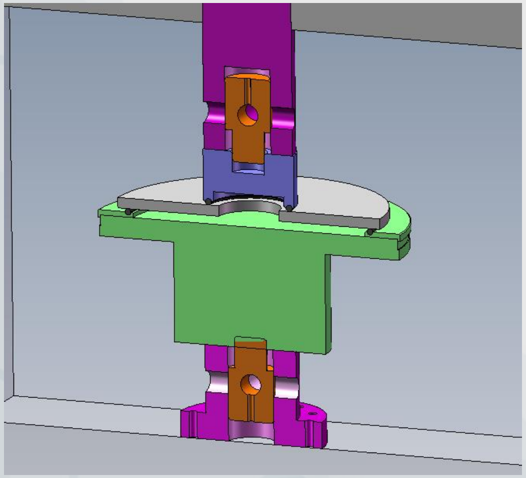

Mock-up brazed

International Spring School on Forefront Alloys and Advanced Materials for Extreme Conditions, Sardinia, Italy, 15 to 17 May 2017

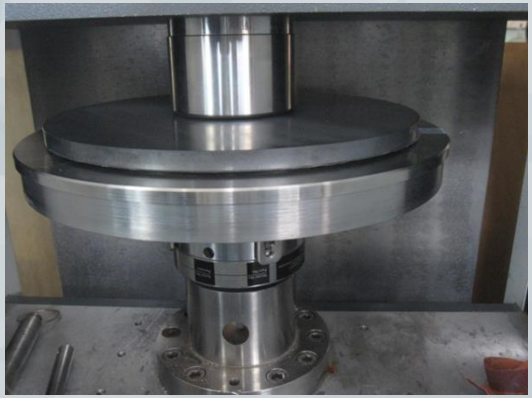
Examples of applications

- Mechanical test of the mock-up until failure

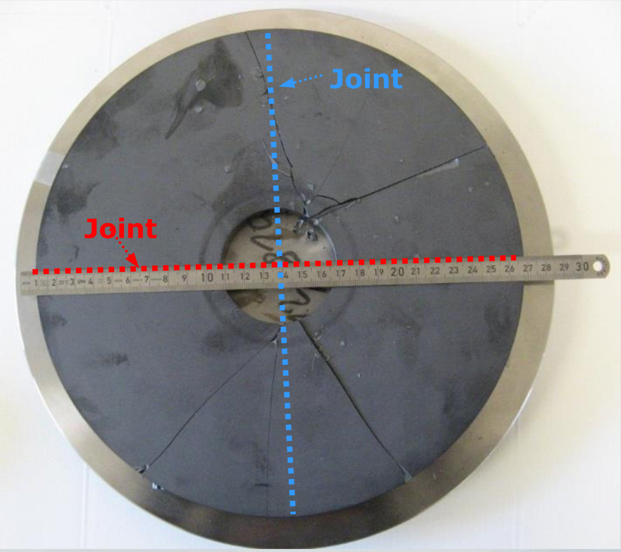
Electromechanical Compression machine (20 kN)



Cut-view of the set-up for axisymmetric flexural test mounted on the compression machine



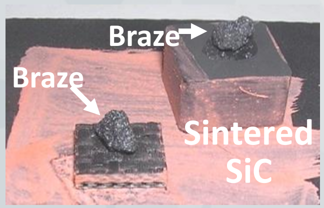
Set-up and mirror during mechanical test



Mirror after failure : cracks are not localized in the brazed joints.

Examples of applications

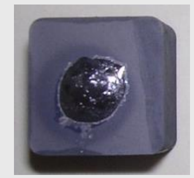
- Brazing of sintered SiC with SiC/SiC composites (nuclear application) : the difficulty is the infiltration of the liquid braze in the composite



Composite

Melting of the braze

Composite

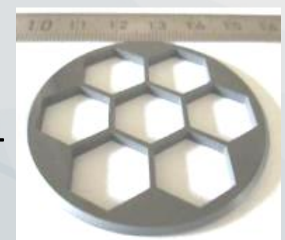


Sintered SiC

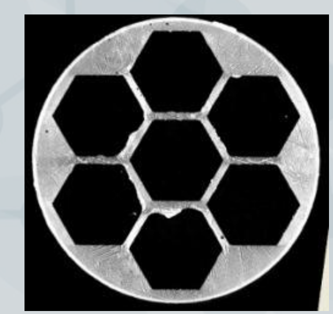
The composite is covered with a seal coat (coating) to suppress the infiltration

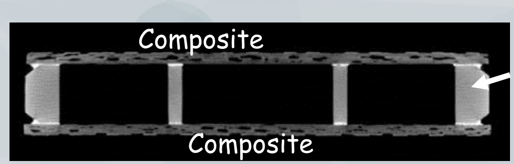
SiC / SiC composite disc

+



Sintered SiC honeycomb



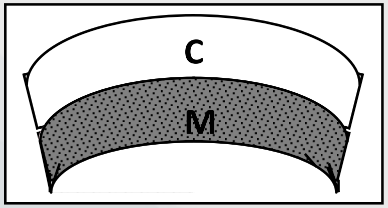


SiC

Controls by X-ray tomography (braze in white)

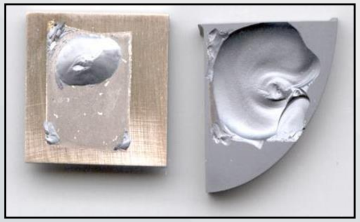
SiC / metal brazing

- **SiC / metal brazing difficulties :**
  - Strong reactivity of SiC with metals
  - Mechanical incompatibility : the major problem
    - CTE SiC = 4 10^{-6} K^{-1}
    - CTE most metals > 10^{-5} K^{-1}



High stress level in the assembly

⇨ Failure in the SiC is possible during cooling, especially from high temperature



SiC/Inco – $T_{brazing}$ 800°C

- **Optimisation of the joint to minimise the stress and limit the reactivity :**
  - Design : SiC in compression
  - Use of interlayers to limit residual stress (compliant, elastic)
  - Use of a chemical barrier to limit reactivity

Conclusions

- The BraSiC® process is an interesting solution for the similar brazing of SiC :
  - A great knowledge of the process and properties of the joints
  - A process conducted until the stage of technology transfer
  - Extension to composites

- The brazing of SiC with metals :
  - The mechanical incompatibility is the major problem
  - A great effort is necessary to develop SiC / metal brazing, especially for high temperature applications

Thank you!

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