



Densification mechanisms of metallic systems by SPS at the microscopic scale

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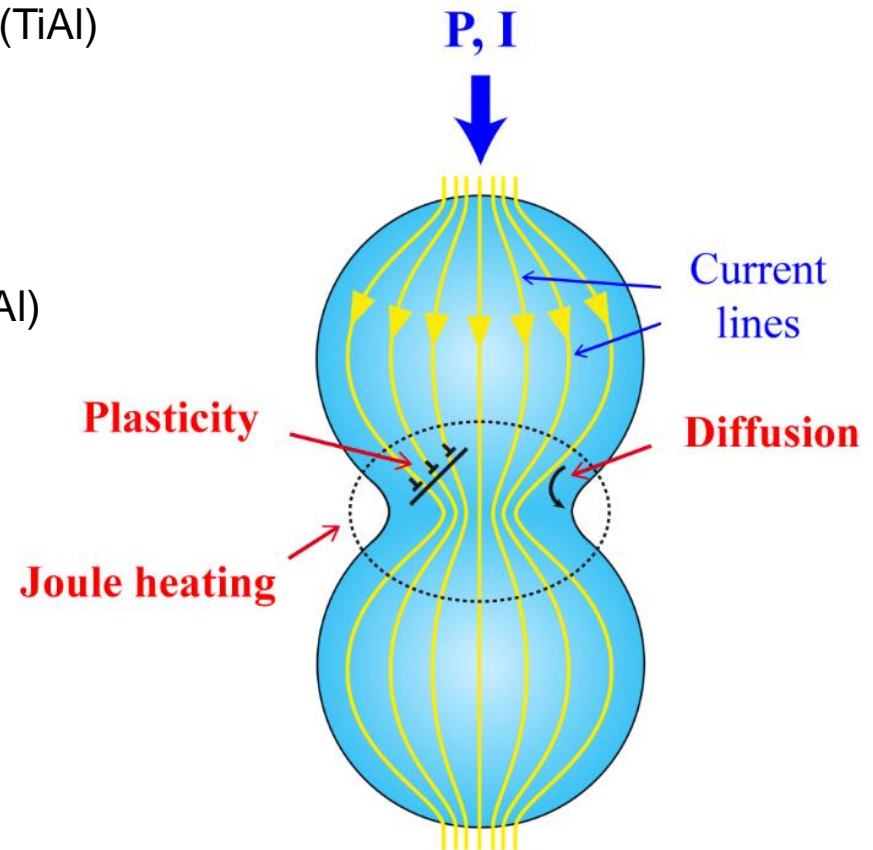
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Outline

Joule overheating? → predictive FEM modelling (TiAl)

Electroplasticity ? → FIB liftouts at the necks (TiAl)

Electromigration ? → SPS vs. HP
→ Ag-Zn diffusion exp.



Conclusions : microscopic densification mechanisms

FEM modelling

Conditions

- $r / R = 0,04$
- $E = 15 \text{ V/m}$ applied at $t=0$ (exp. value)
- No heat loss by radiation
- No contact resistances

Local equations solved

$$\nabla \cdot (\nabla V) = 0$$

Electricity eq.

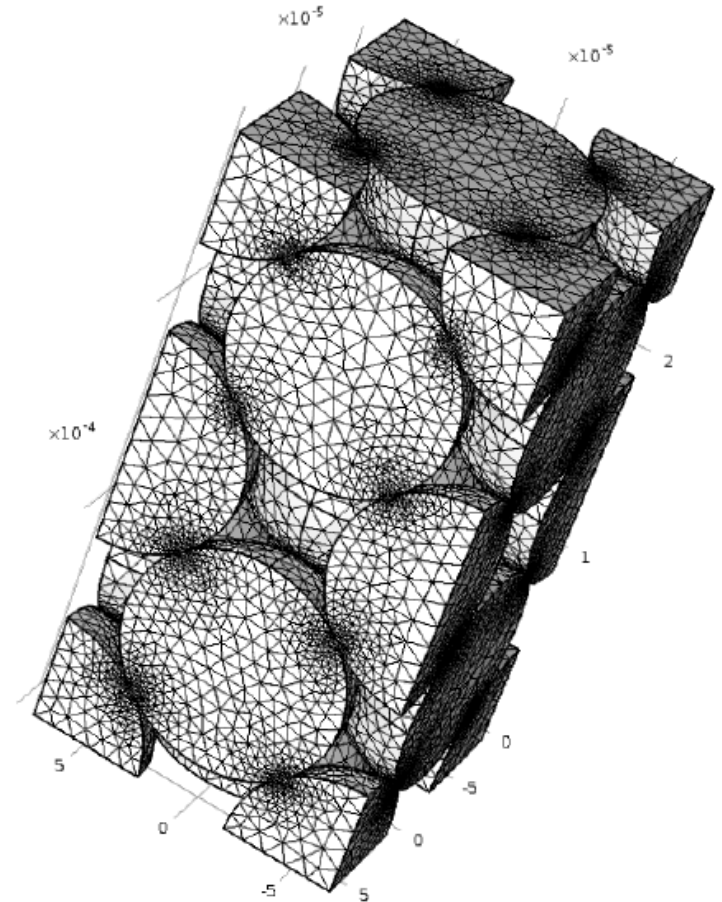
$$\nabla \cdot (-\lambda \nabla T) + \rho c_p \left(\frac{\partial T}{\partial t} \right) = \sigma (\nabla V)^2$$

Heat eq.

V : tension (V), T : temperature (K)

Temperature-dependent physical constants
of TiAl at 1000°C :

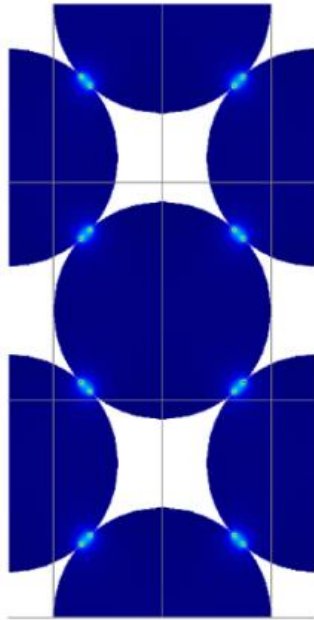
λ : thermal conductivity	$23 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$
σ : electrical conductivity	$8,1 \times 10^5 \cdot \Omega^{-1}\cdot\text{m}^{-1}$
c_p : thermal capacity	$693 \text{ J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$
ρ : density	$3900 \text{ kg}\cdot\text{m}^{-3}$



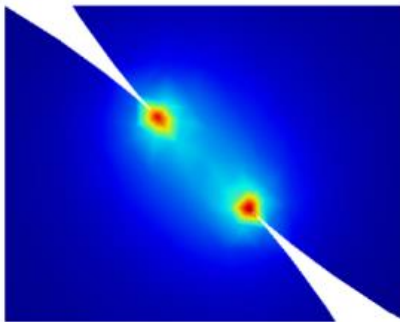
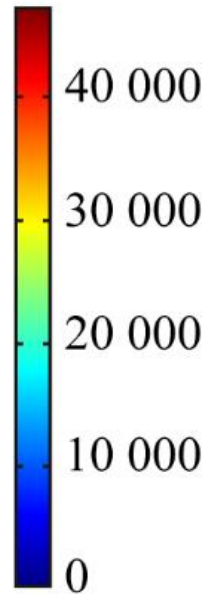
FEM modelling

Current density map

Average
current
 500 A/cm^2



Current
density
 A/cm^2

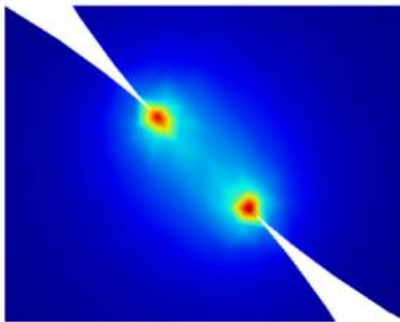
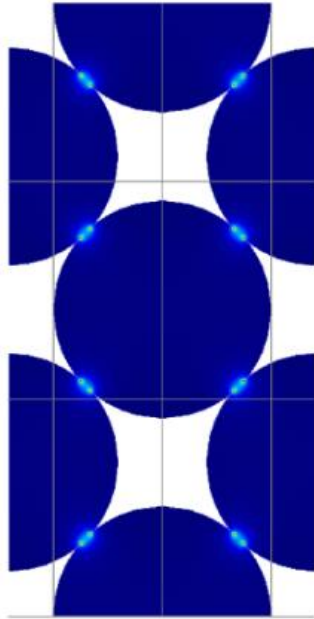


Local current $\approx 100 \times$ average current

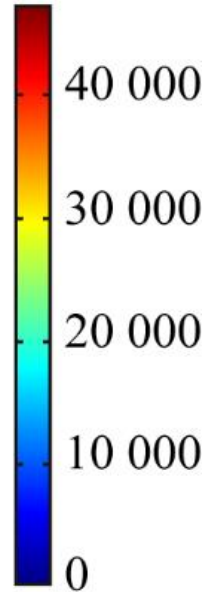
FEM modelling

Current density map

Average current
 500 A/cm^2

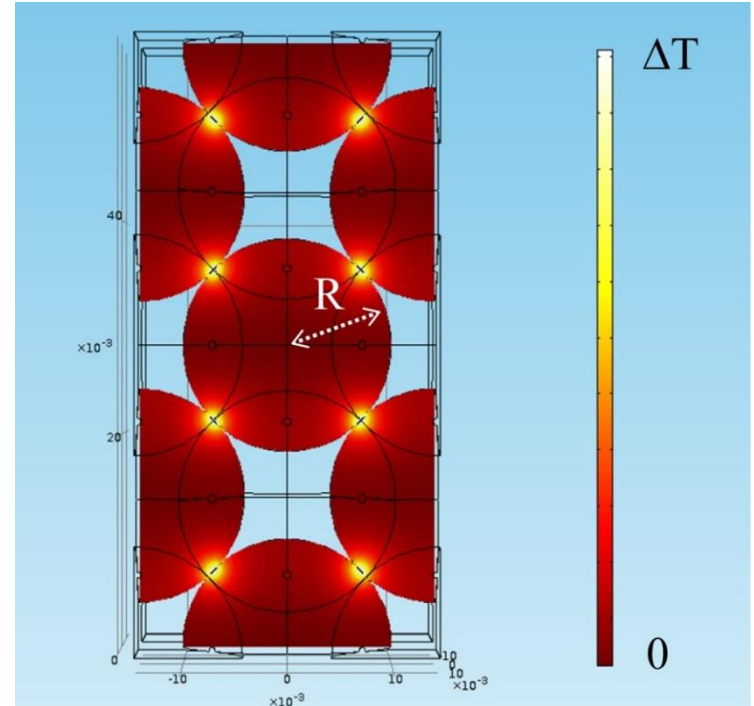


Current density
 A/cm^2



Local current $\approx 100 \times$ average current

Temperature map



$R = 1 \text{ cm} \rightarrow \Delta T = 100^\circ\text{C}$

$R = 100 \mu\text{m} \rightarrow \Delta T = 0,1^\circ\text{C}$

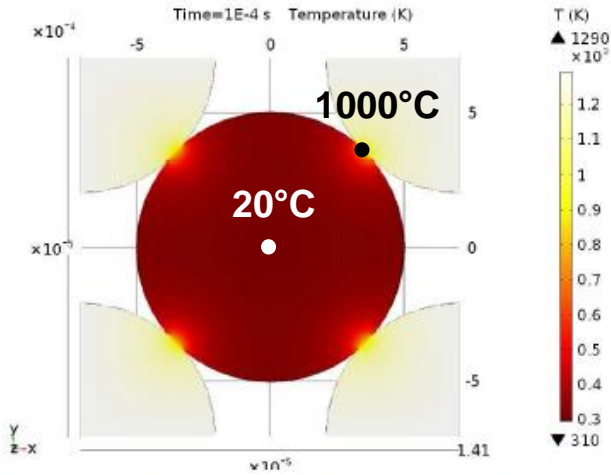


Very weak overheating
at the necks

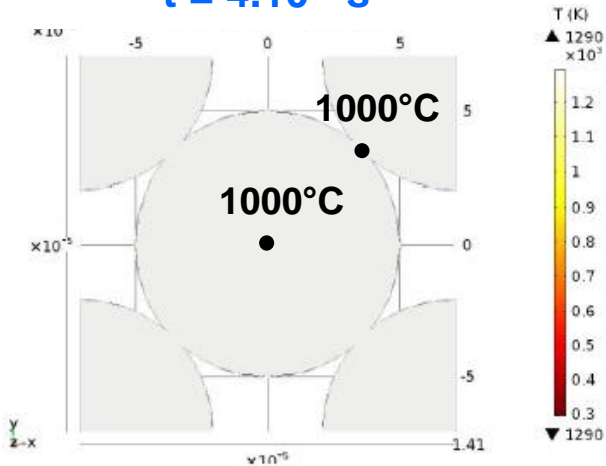
FEM modelling

Diffusion of heat generated at hot spots

$t = 0$

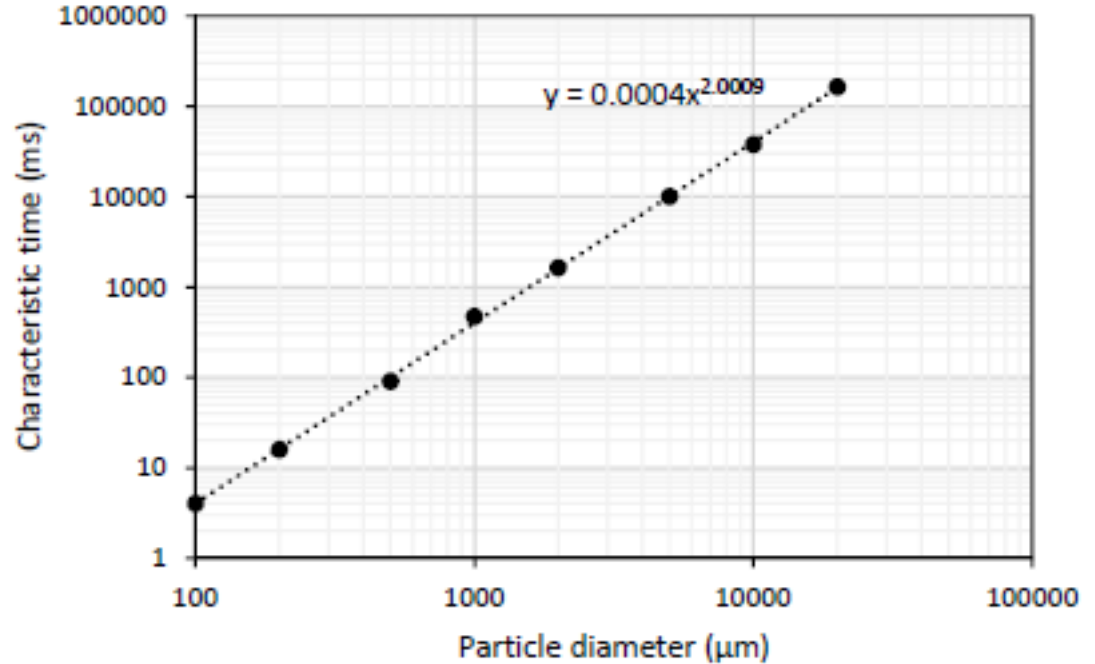


$t = 4 \cdot 10^{-3} \text{ s}$



Particles 100 μm
in diameter $\rightarrow t \approx 4 \text{ ms}$

Time for homogenize temperature in the particles (at $\pm 1^\circ\text{C}$):



Parabolic law: heat transfer by diffusion

Limited overheating at the necks: fast heat diffusion at the particle scale (100 μm)

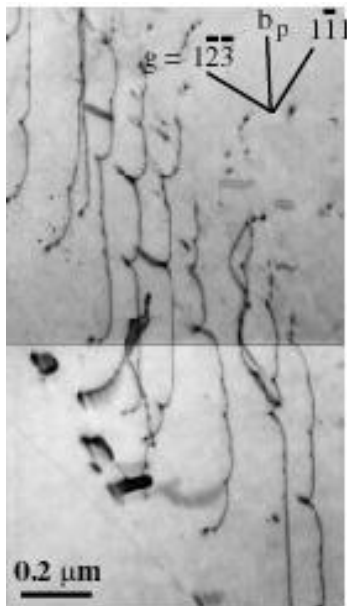


Other phenomena induced by high currents at the necks:
electroplasticity? electromigration?

Electroplasticity at the necks

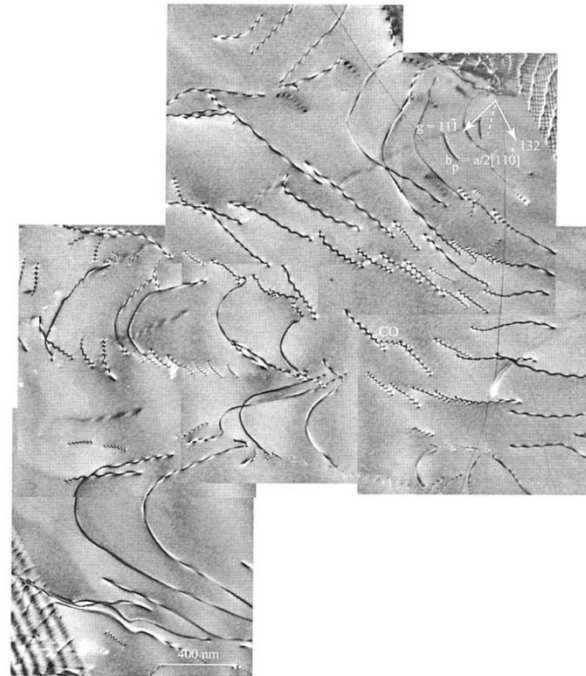
Electroplasticity: force exerted by high currents on the dislocations, considered as impenetrable cylinders

Plasticity mechanisms in TiAl



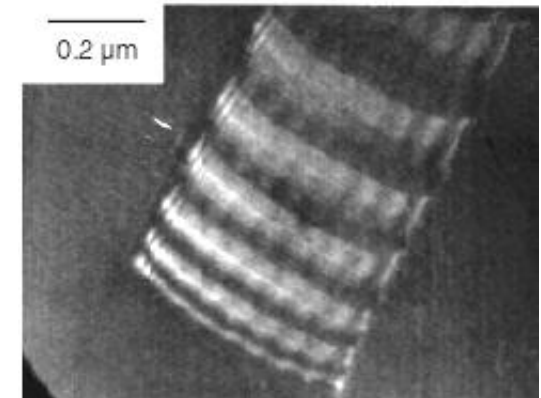
Screw dislocations
anchored on pinning points

Glide



Curved dislocations

Glide + climb

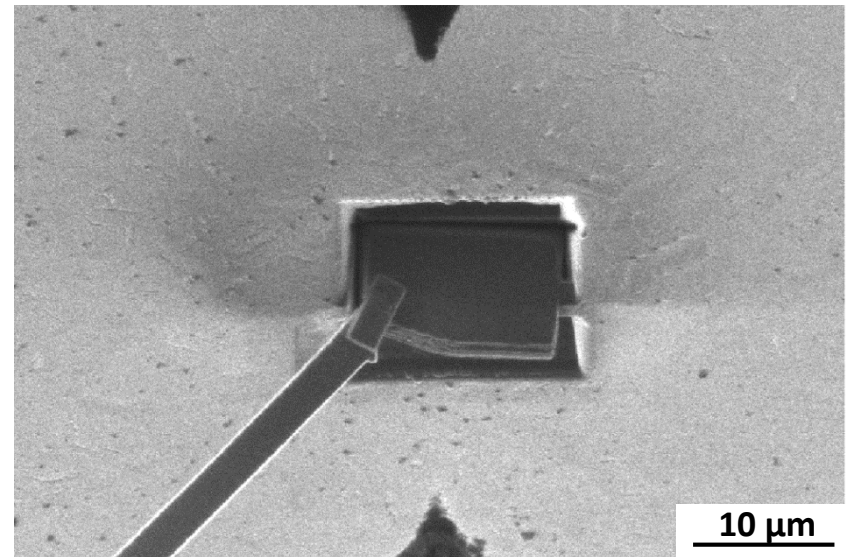
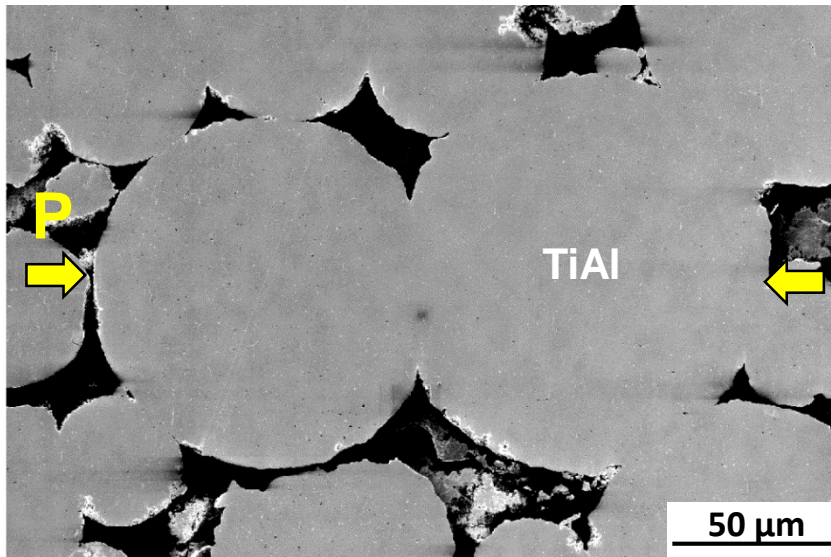


Twinning

High current \rightarrow change of mechanism? Ex: activation of cross-slip, of mixed climb,...

Electroplasticity at the necks

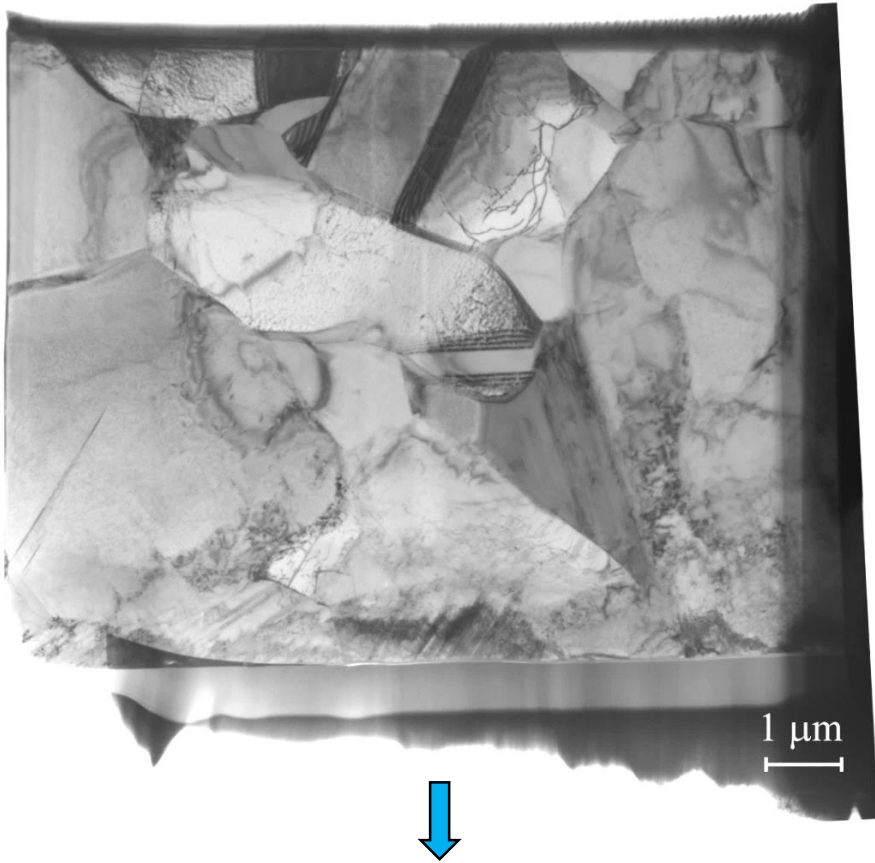
Identification of the plasticity mechanisms: TEM thin foil lift-out by FIB



Electroplasticity at the necks

TEM images of FIB thin foils

900°C – Center

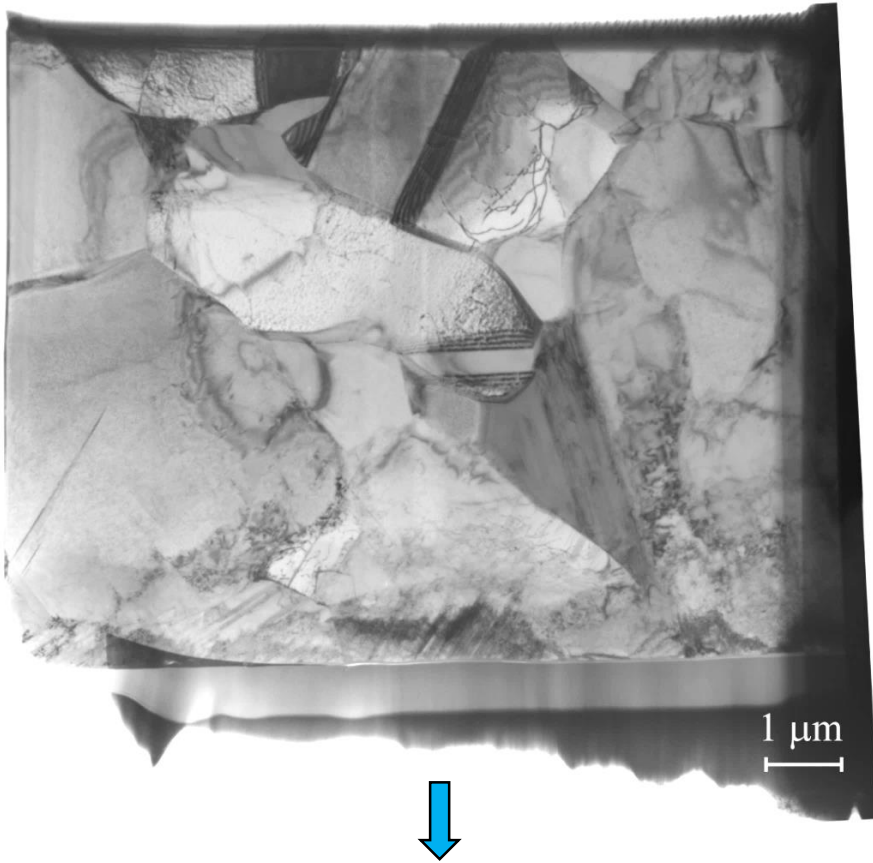


Microstructure identical to the initial state

Electroplasticity at the necks

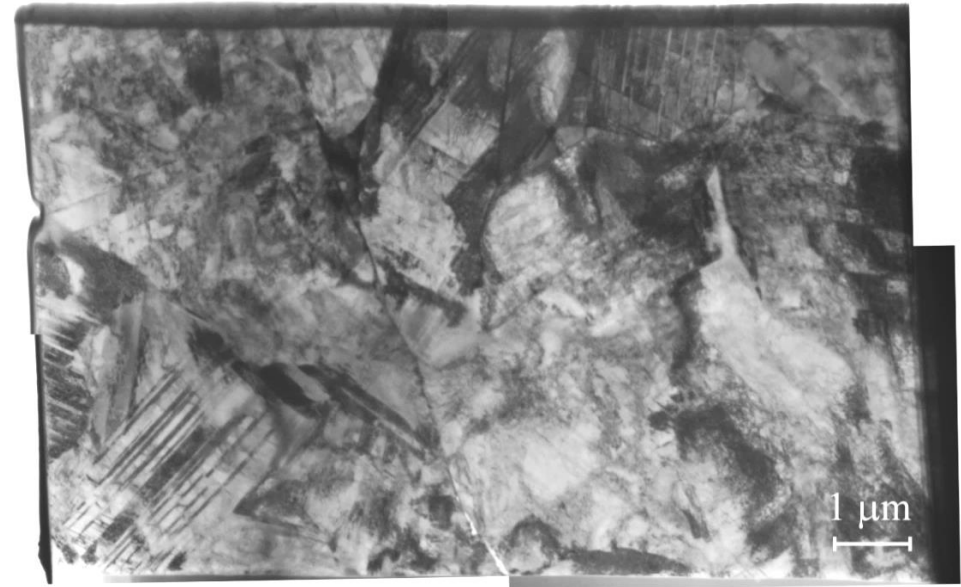
TEM images of FIB thin foils

900°C – Center



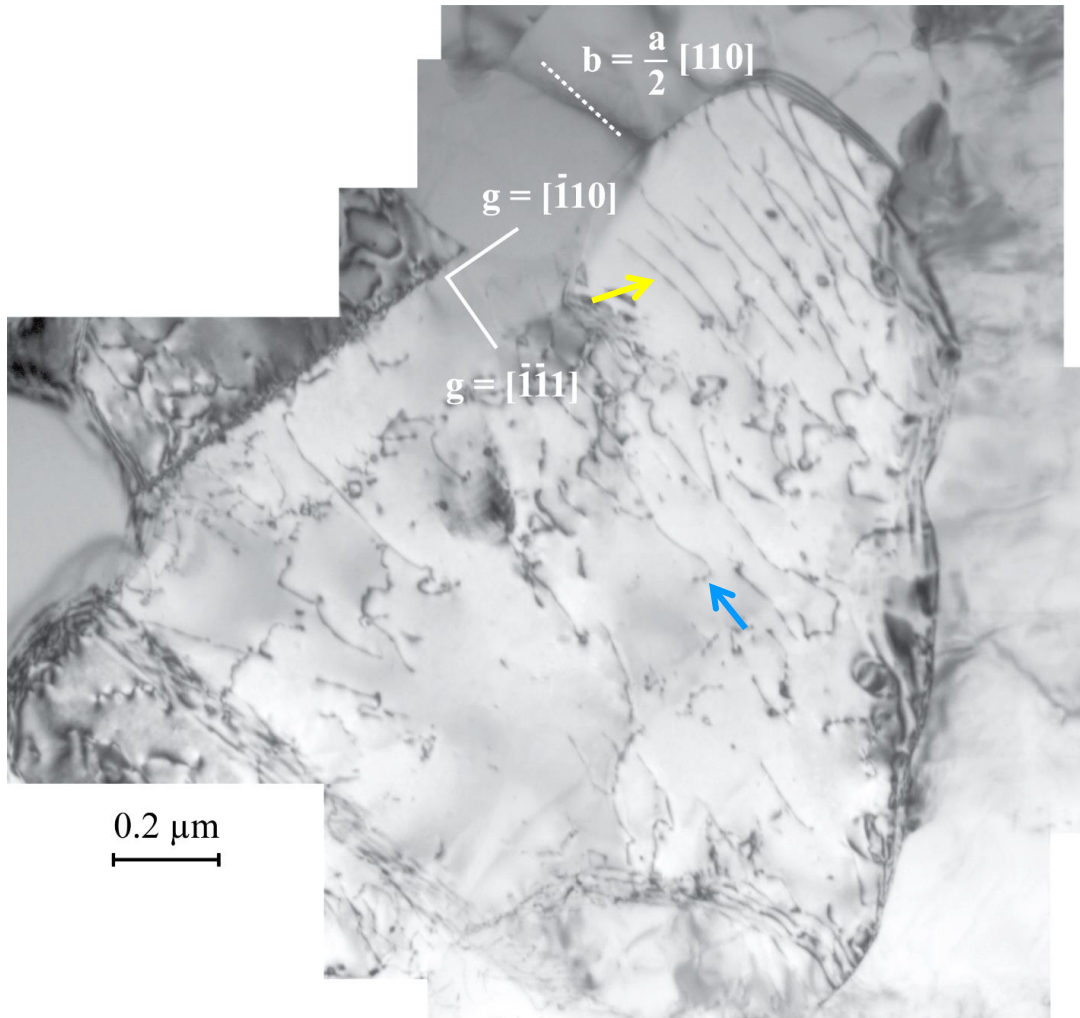
Microstructure identical to the initial state

900°C – Neck



Intense plastic deformation

Electroplasticity at the necks



Screw dislocations → Glide

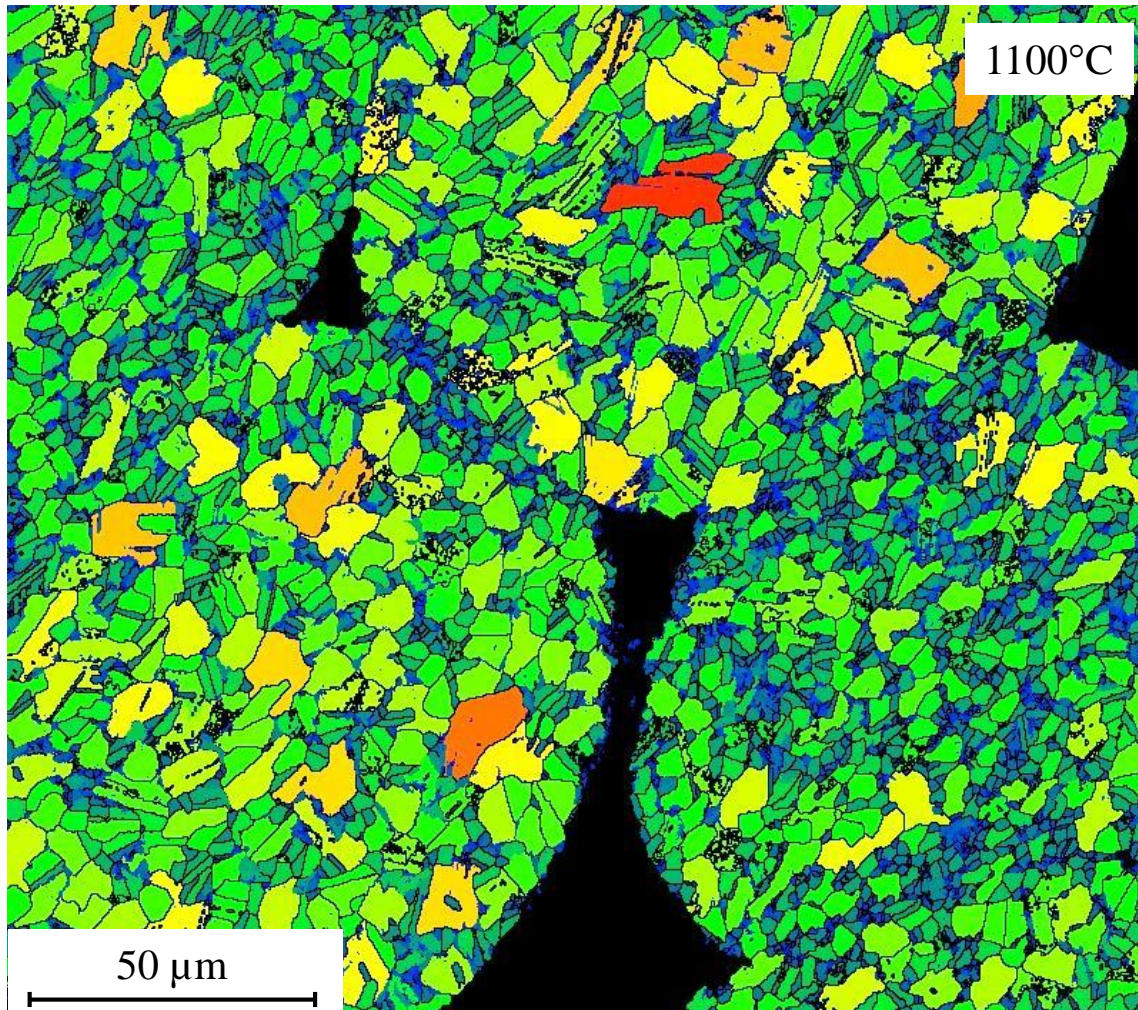
Helices → Climb



Mechanism:
Climb-controlled glide

Electroplasticity at the necks

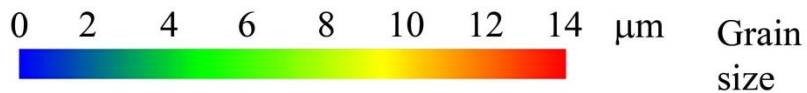
EBSD grain size map



Small grains at the necks



Recrystallization



Electroplasticity at the necks

- Intense plastic deformation by twinning and climb-controlled glide
- Relaxation of deformation by recovery-recrystallization

Densification by classical metallurgical phenomena

No change of mechanism



Higher kinetics induced by current?

SPS-HP comparisons

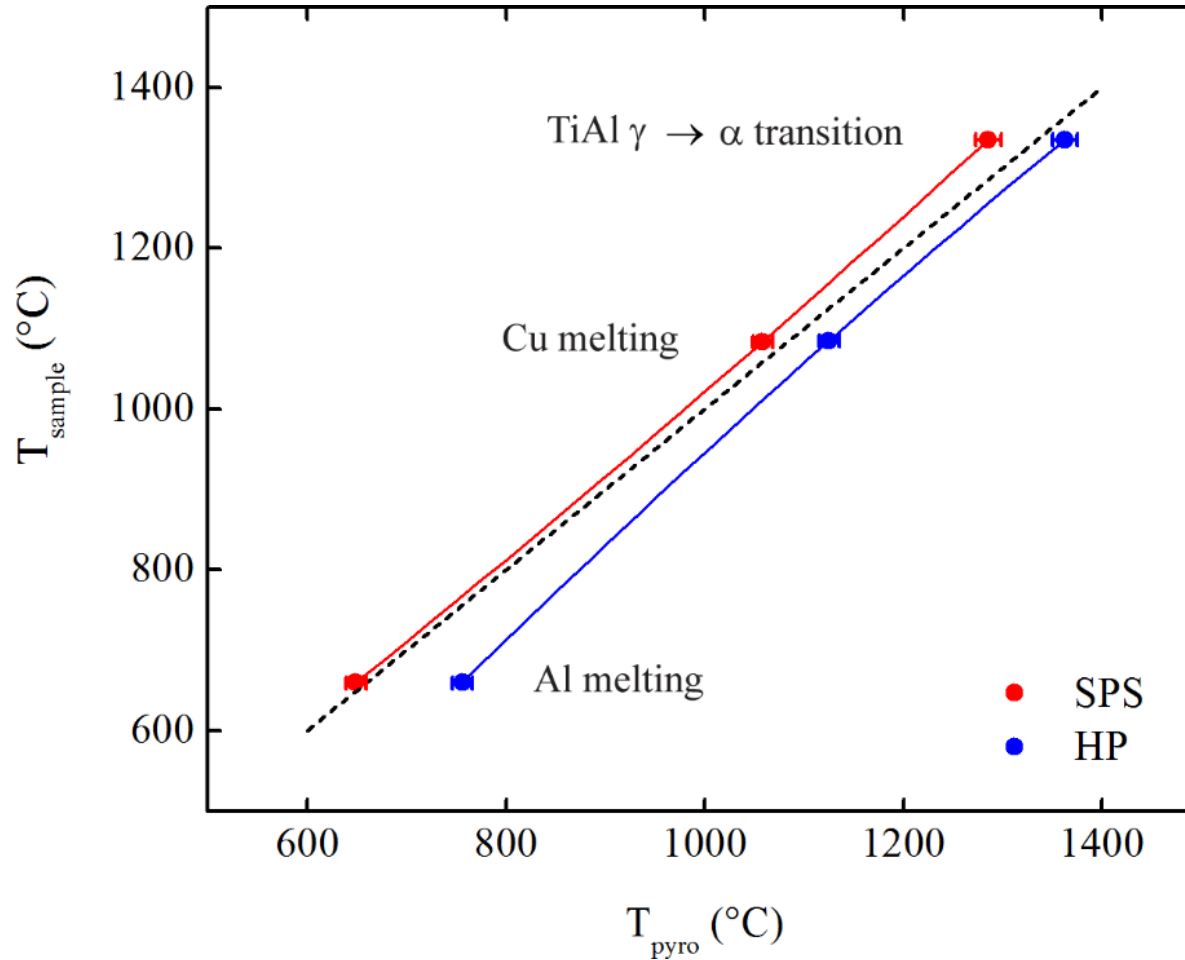
Aim: evaluate the role of the current on densification kinetics



Necessity of identical cycles and sample temperatures

SPS-HP comparisons

Sample temperature calibration

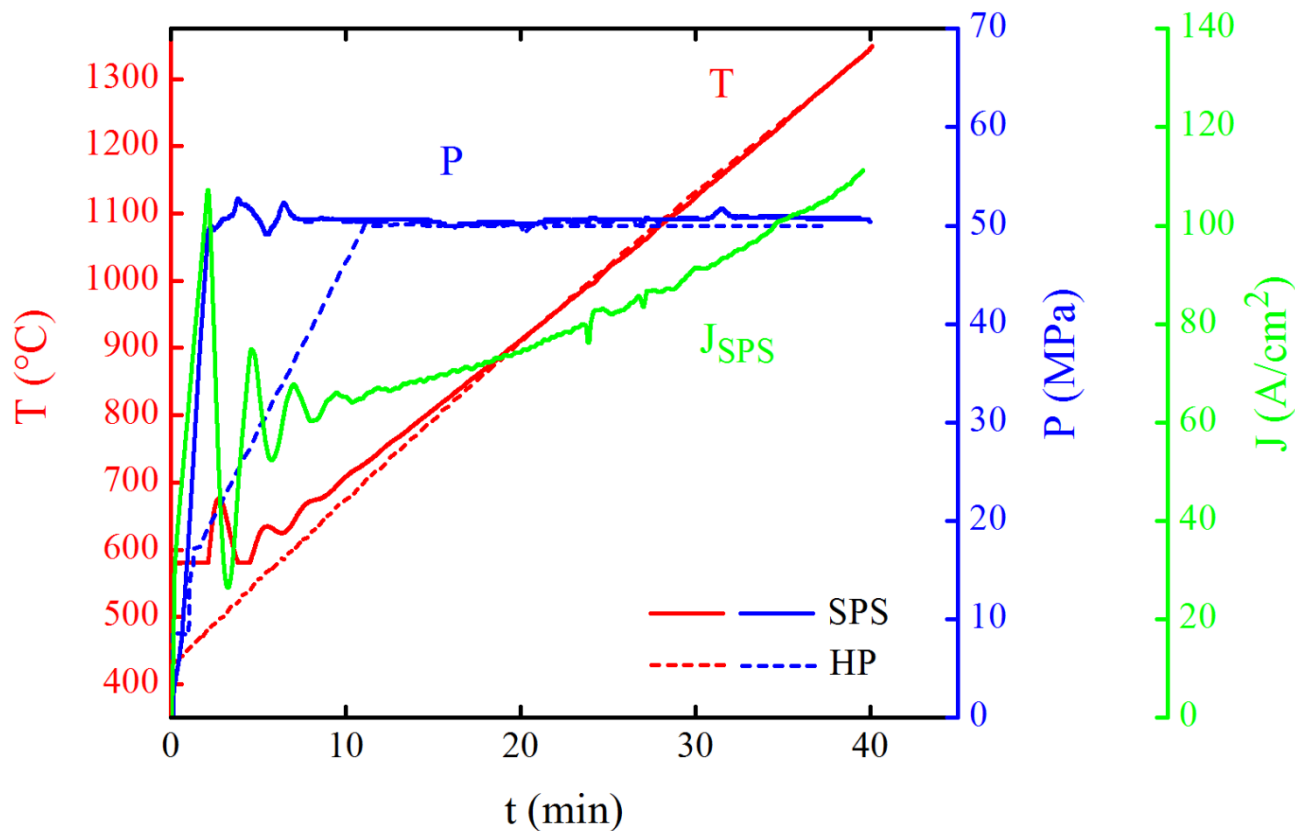


Accuracy of sample temperature: ± 20 °C

SPS-HP comparisons

Pressure - temperature cycles

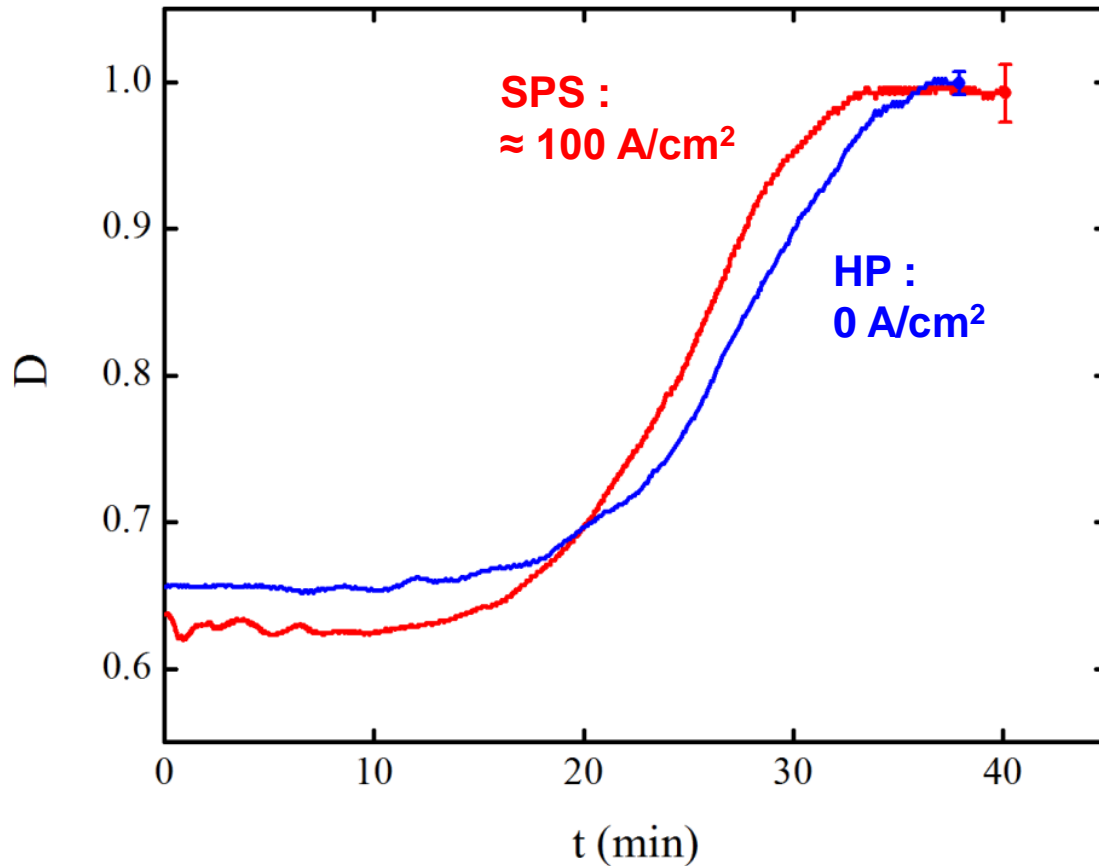
50 Mpa – 20°C/min



J ≈ 100 A/cm²

SPS-HP comparisons

Densification cycles



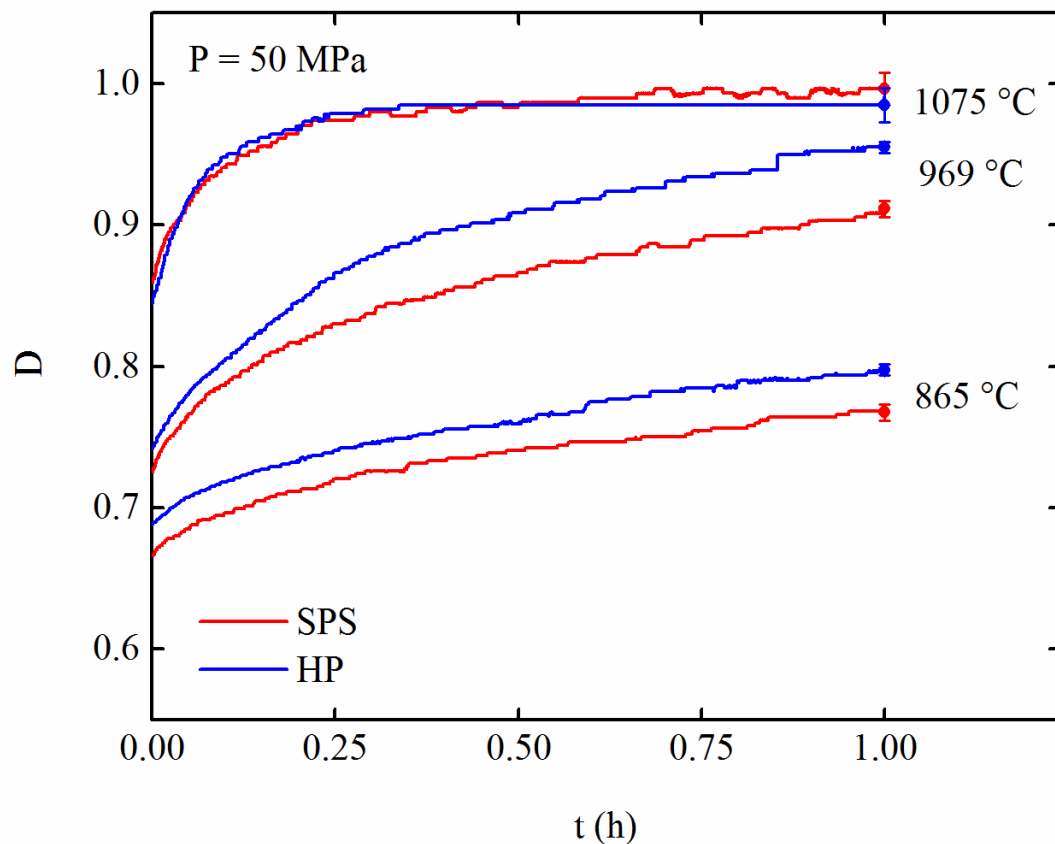
Close densification kinetics



No influence of current

SPS-HP comparisons

Isothermal densification curves



	n	Q (kJ/mol)
SPS	1,9 ± 0,3	308 ± 20
HP	1,5 ± 0,3	276 ± 40



TiAl constitutive relation:

$$\dot{\epsilon} = A_0 \exp\left(-\frac{Q}{RT}\right) \sigma^n$$

$$\dot{\epsilon} = G_1(D) \dot{D}$$

Von Misès stress in the powder particles

$$\sigma = G_2(D) P$$

$$\ln \dot{D} = f(\ln P) \rightarrow \mathbf{n}$$

$$\ln \dot{D} = f\left(\frac{1}{T}\right) \rightarrow \mathbf{Q}$$

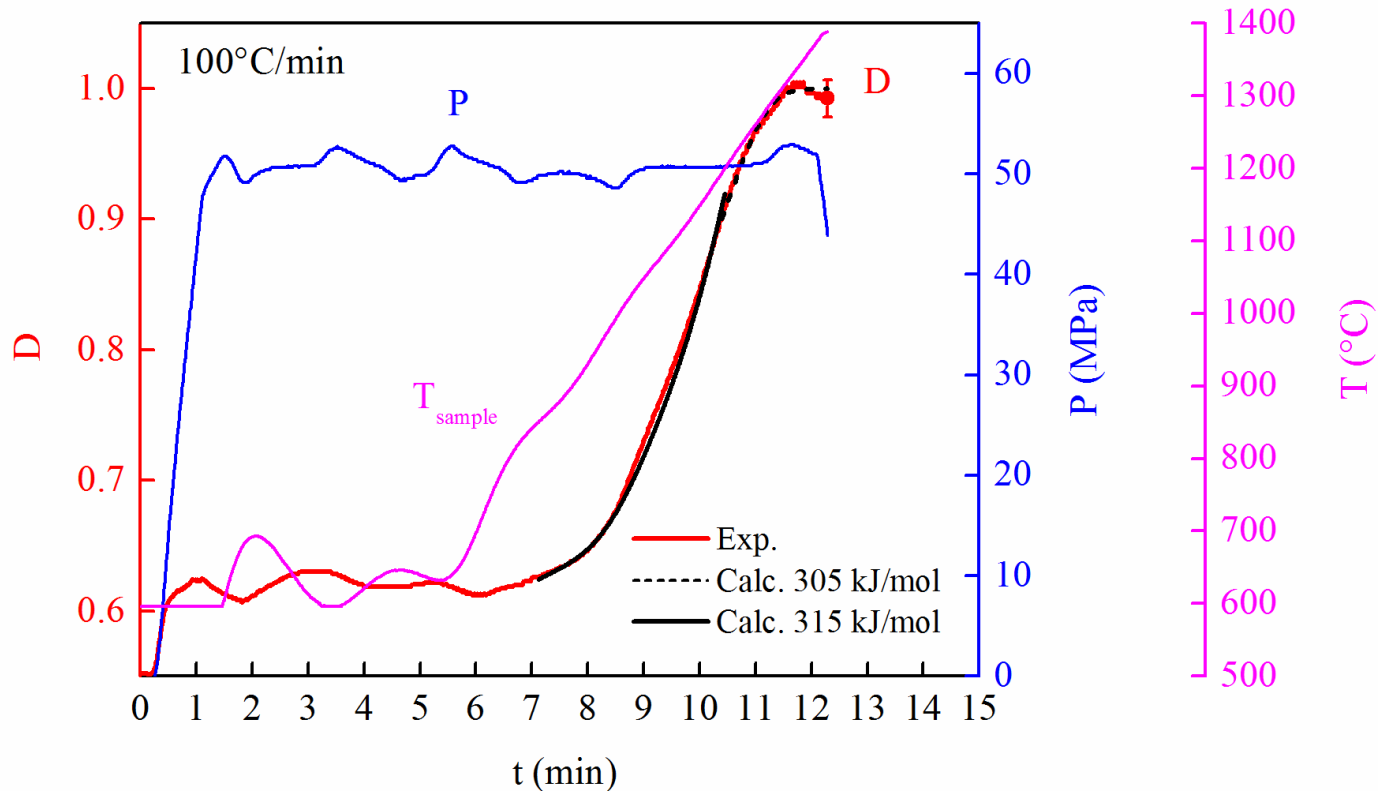
Typical values for bulk TiAl

Calculation of the densification cycle

Ashby, Arzt et Helle (1983, 1985)

$$D < 0,9 : \dot{D} = 5,3(D^2 D_0)^{1/3} \frac{1}{\sqrt{3}} \left(\frac{D - D_0}{1 - D_0} \right)^{1/2} \dot{\epsilon}$$

$$D > 0,9 : \dot{D} = \frac{3}{2} \times \frac{D(1 - D)}{[1 - (1 - D)^{1/n}]^n} \dot{\epsilon}$$



$n = 1,9$

$Q = 305 - 315 \text{ kJ/mol}$ (exp : 308 kJ/mol)



Good reproduction of the anisothermal densification kinetics

Densification kinetics – Summary

- Close densification kinetics by SPS ($\approx 100 \text{ A/cm}^2$) and by HP (0 A/cm^2)
- Activation energy for densification ($\approx 300 \text{ kJ/mol}$) close to that of bulk self diffusion of Al in TiAl (360 kJ/mol , Herzig 1999)
- Anisothermal densification kinetics (100°C/min) well reproduced analytically by Ashby et al's modèle (1983, 1985)



No modification of kinetics by currents $\approx 100 \text{ A/cm}^2$

→ **Effect of stronger currents?**

Electromigration in Ag-Zn

Electromigration :

Perturbation of the electron flow by the diffusing atoms when they are in the activated state →
momentum transfer



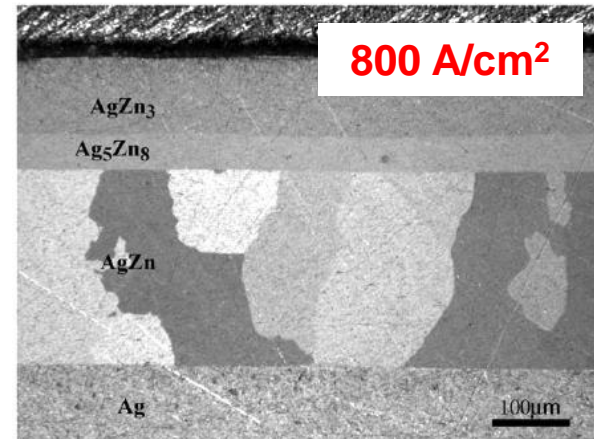
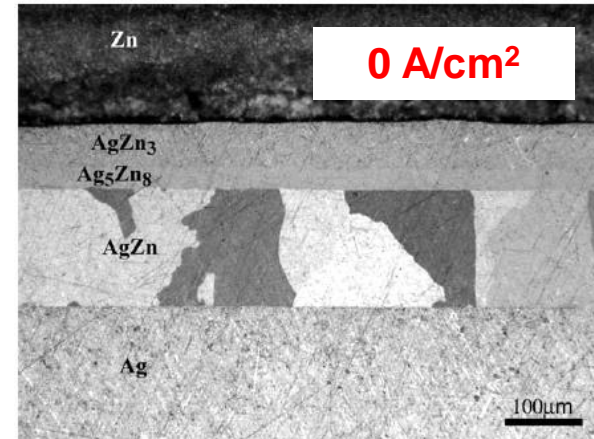
Modification of 2nd Fick's law:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - \frac{D}{RT} Fz^* \rho J \frac{\partial c}{\partial x}$$



Drift with time of the diffusion profile

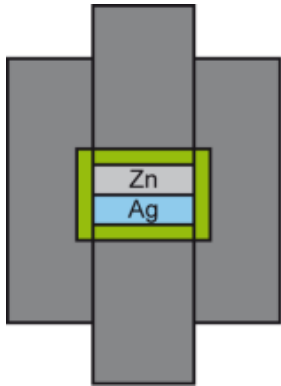
350°C – 5 h



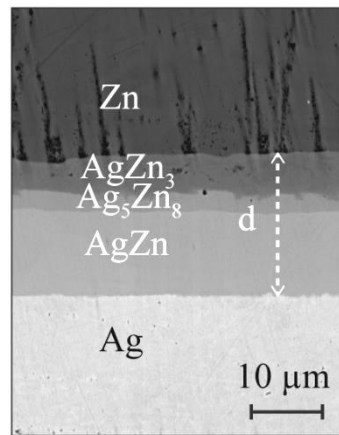
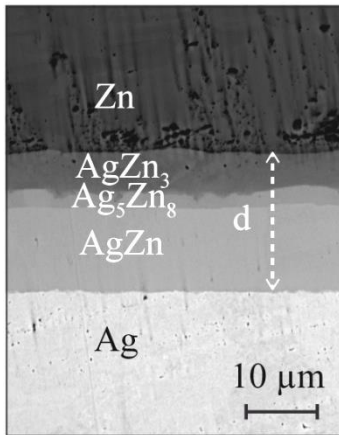
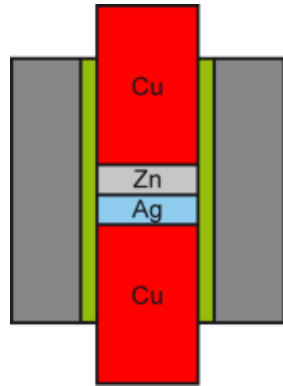
Friedman et al, Intermetallics 12 (2004) 589

Electromigration in Ag-Zn

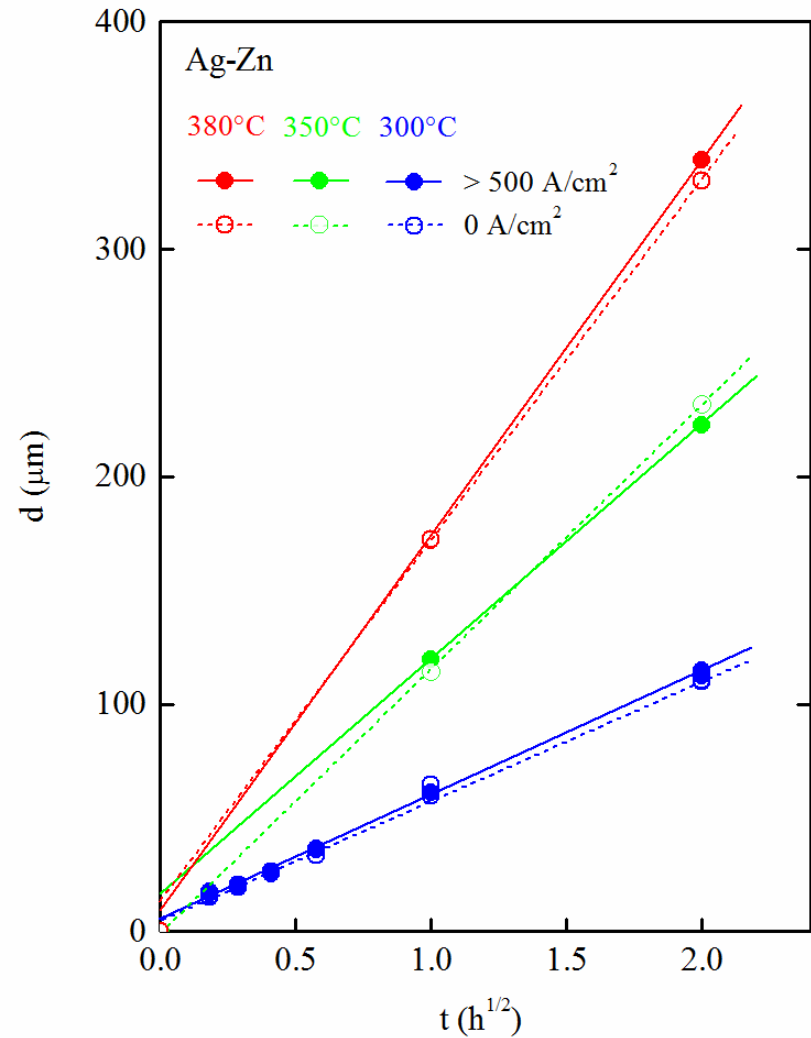
0 A/cm²



> 500 A/cm²



300°C – 2 minutes



Parabolic kinetics

No electromigration

Conclusions

- **Mechanisms controlling densification:**

Plastic deformation by dislocations followed by recovery/recrystallization

Kinetics controlled by Al bulk diffusion

- **Effect of current?**

- Negligible Joule overheating

- Current $\approx 100 \text{ A/cm}^2$: no acceleration of densification

- Current $> 500 \text{ A/cm}^2$: no electromigration in Ag-Zn

SPS conventional conditions



Densification by classical metallurgical mechanisms, without acceleration by the current