Questions ouvertes en solidification

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&

Commission thématique coulée solidification SF2M - GDR SAM

Outline of the presentation

1. Industrial motivations for solidification studies
   (with a special emphasis on the casting of aluminium alloys in Constellium)

2. Main topics with emphasis on unsolved problems and associated R&D

3. Perspectives and draft for a vision/ roadmap and a few words on GDR SAM

Industrial motivations

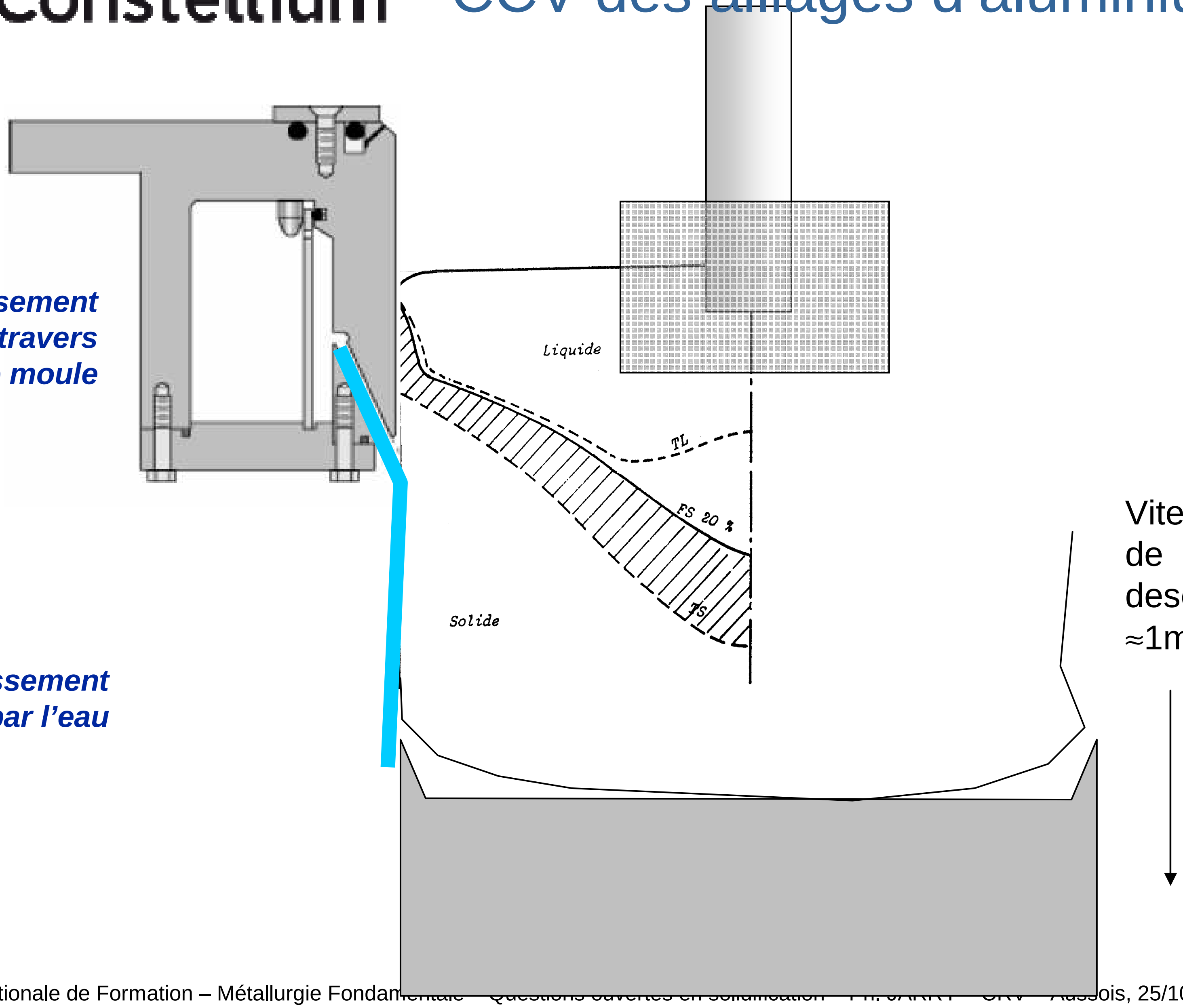
Industrial needs as expressed during Nancy september 2012 symposium of the « Commission thématique coulée » / GDR

- Industrial representatives (Industeel, Ascometal, Aubert & Duval, Areva, Timet) all agreed that the main solidification topics of industrial interest were:
  - Macrosegregation
  - Porosity
  - Hot cracking
  - Skin defects
- be it for static ingot casting, Vacuum Arc Remelting or ElectroSlag Remelting processes

- Constellium was the only one to add to this list the topic of solidification microstructure heredity (not only solidification *defect* heredity).

➢ *Is this a question of priority or an absence of needs?*

CCV des alliages d'aluminium



Refroidissement indirect à travers le moule

Refroidissement direct par l'eau

Vitesse de descente ≈1mm/s

Produits de coulée typiques:

plaque de laminage

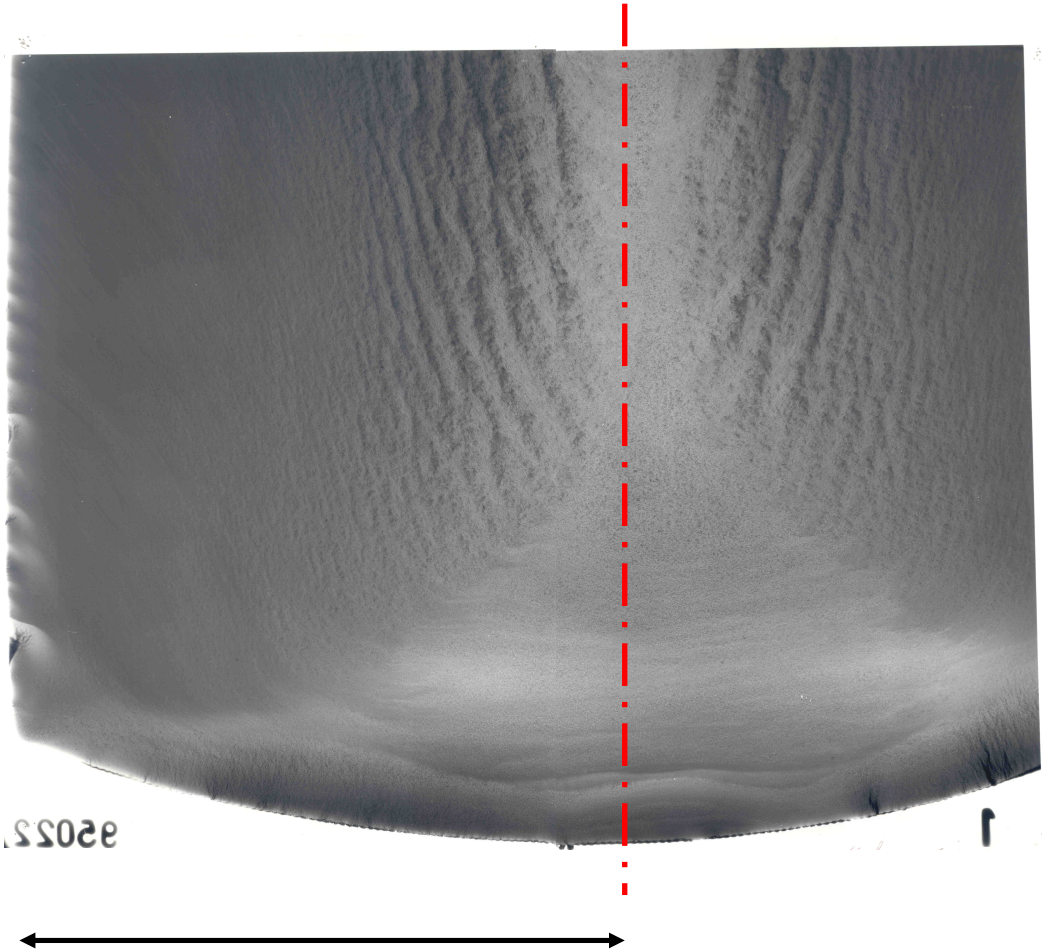
Principales applications:

Tôles aéronautiques

Tôles pour l’automobile

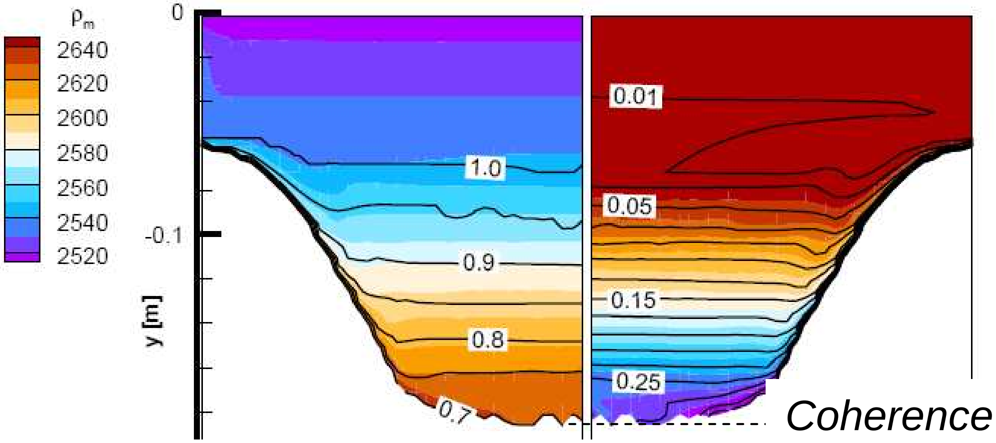
Bandes pour le boîtage

Macro- and meso-segregations

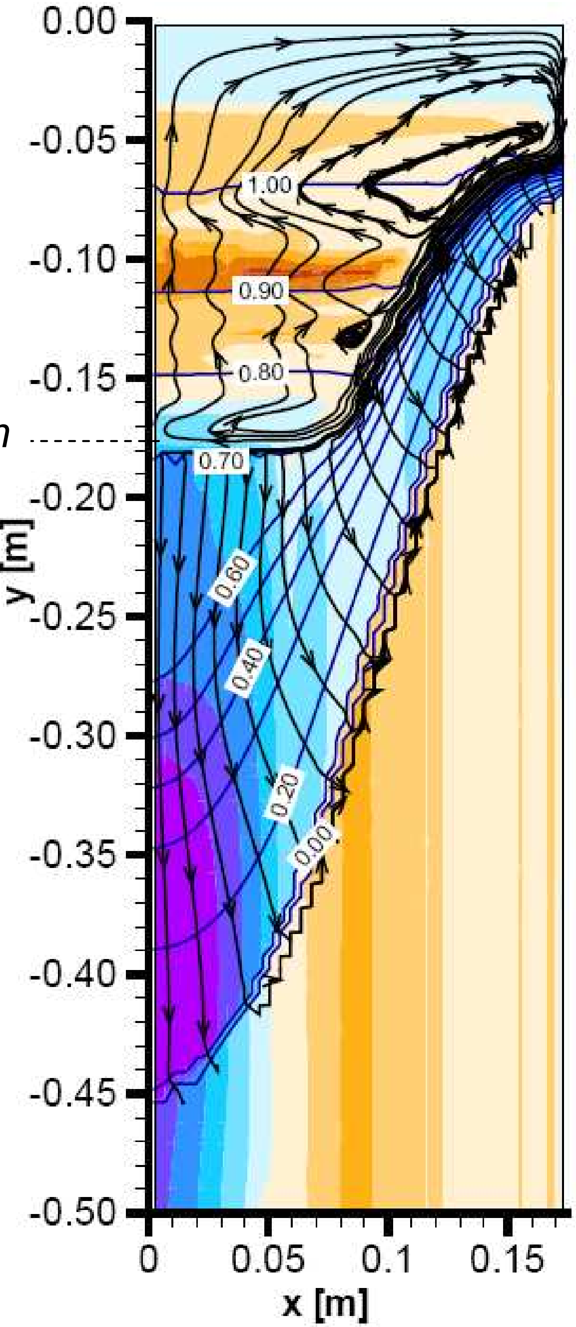


≈25cm

Radiographie X d’une tranche verticale de pied de plaque en alliage AlZnMgCu



Coherence isotherm



Modelling of macrosegregation with SOLID software, accounting for shrinkage driven flow, thermosolutal flow and grain transportation.

(Combeau-Založnik)

Both grain transportation to the sump bottom and shrinkage driven flow account for the negative centerline segregation.

Thermosolutal flow coupled with grain transportation account for mesosegegration.

Focus on:
Solidification studies driven by end properties

Mainly damage tolerance or formability properties

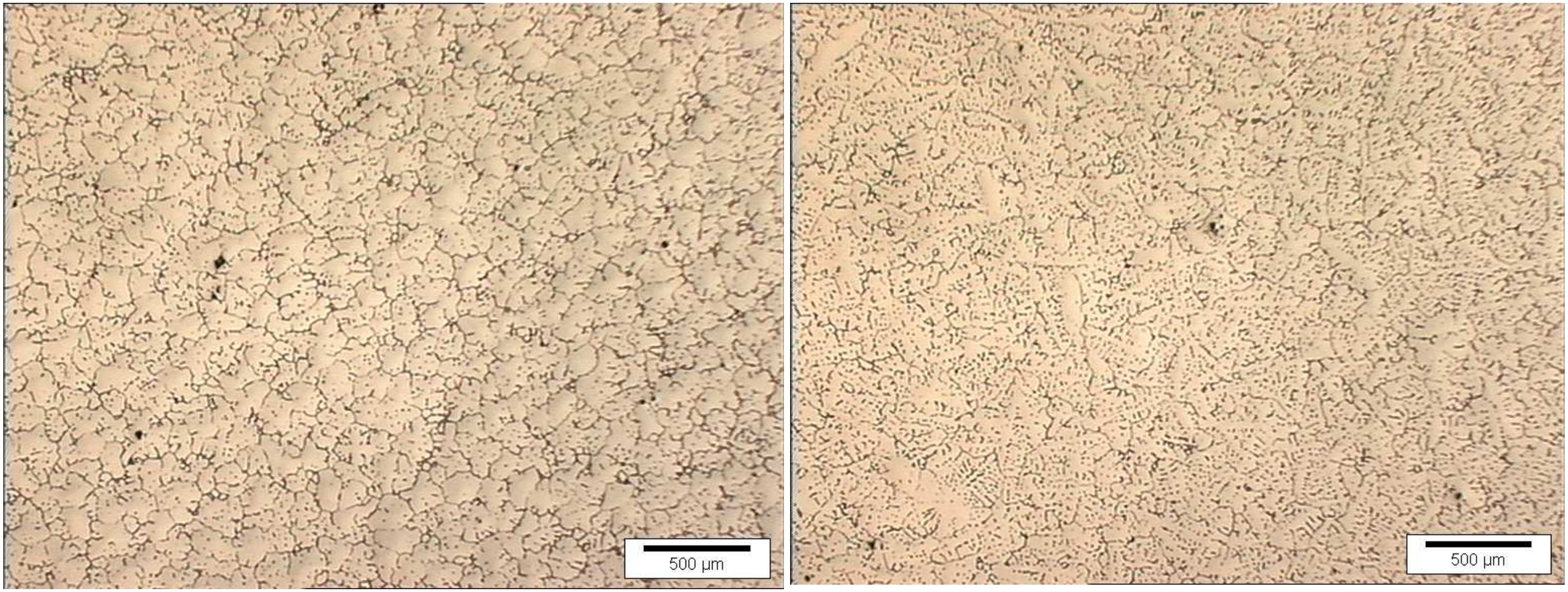
Heredity of microsegregation

Through:

1. Constituent phases (AlFe intermetallics) size and spatial distribution
2. Slow diffusing solutes microsegregation distribution (e.g. Zr in Al)

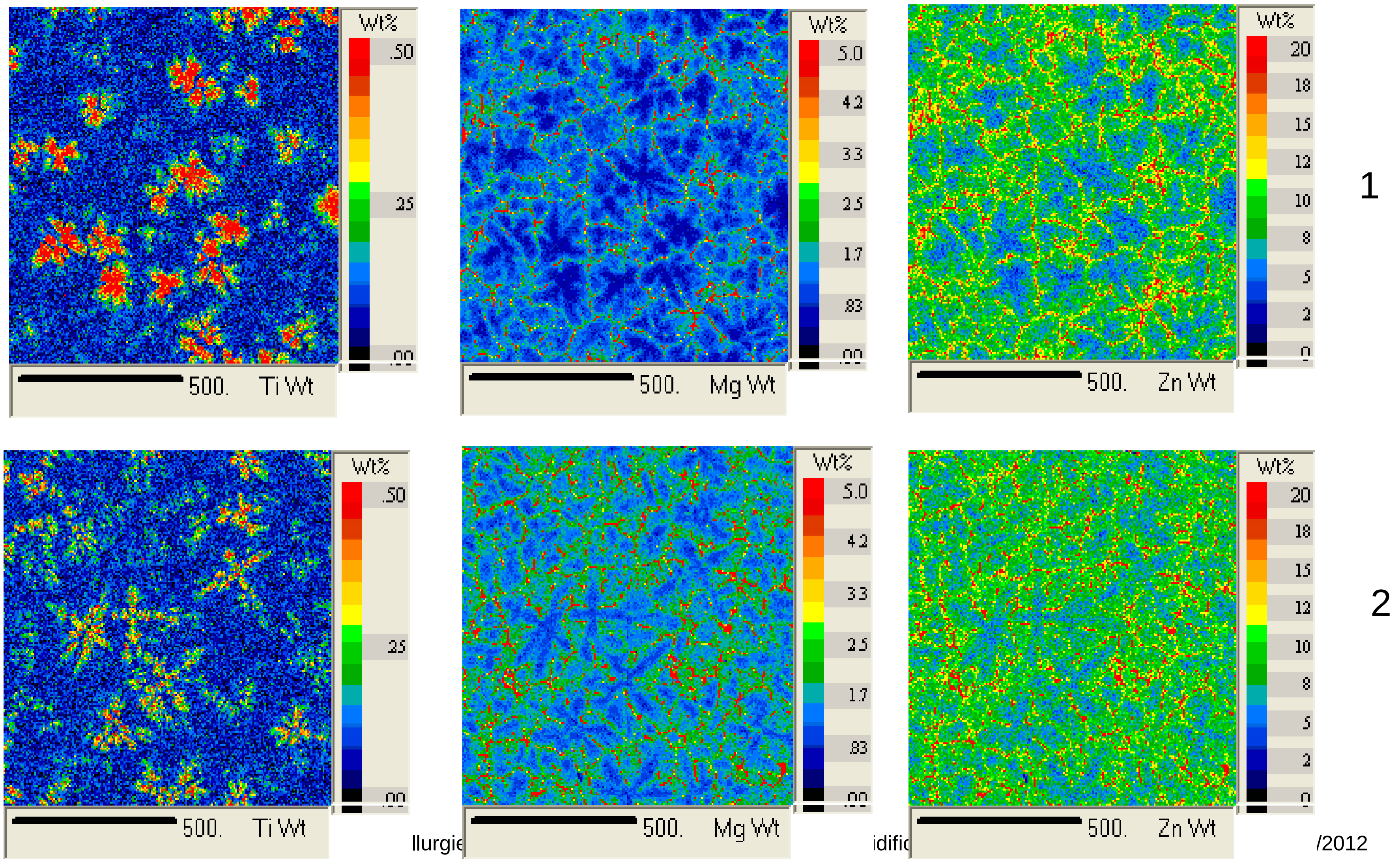
- Can we govern the solidification morphology so as to obtain a uniformly and finely distributed microsegregation (both in solid solution and in precipitation) ?

Lab study on 7xxx alloy

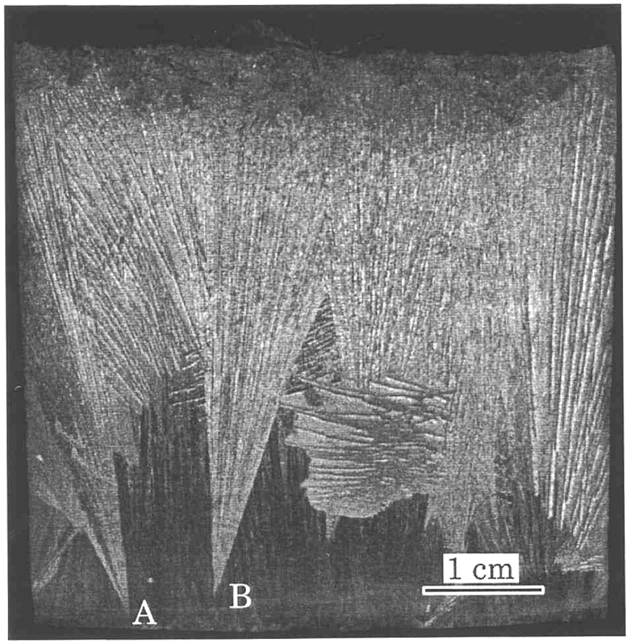


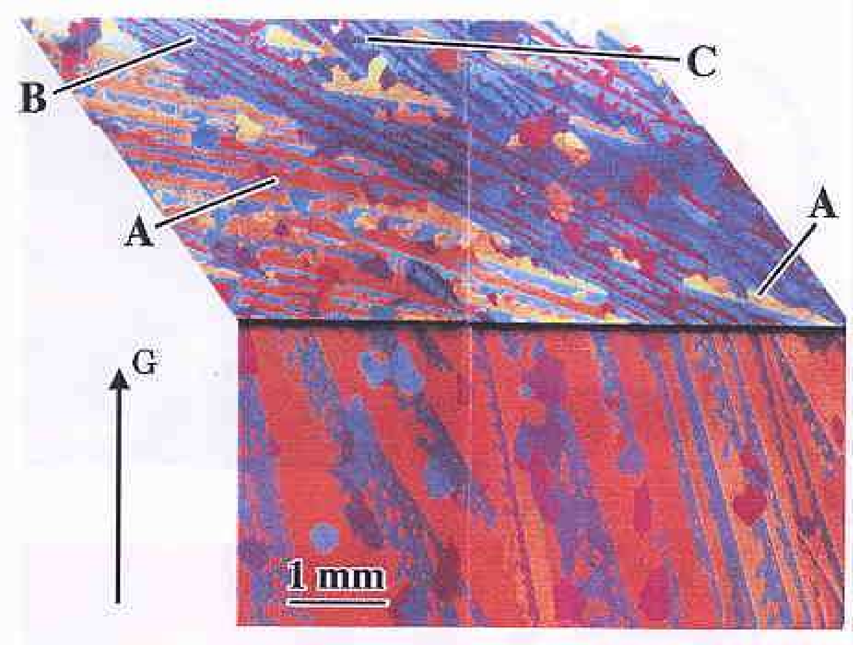
Same alloy, same dT/dt, varying inoculation and superheat in the laboratory

Influence of nucleation parameters on microsegregation distribution



Case study of an « exotic » solidification microstructure: twinned grains in aluminium alloys

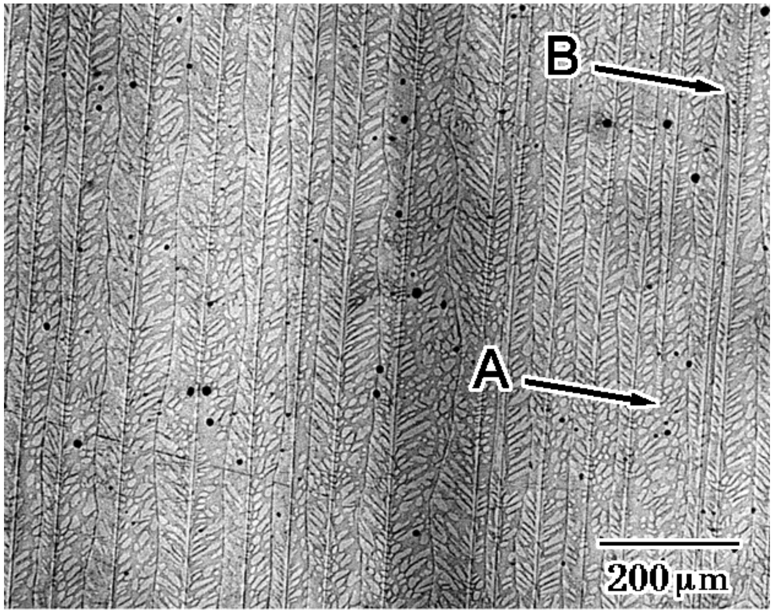




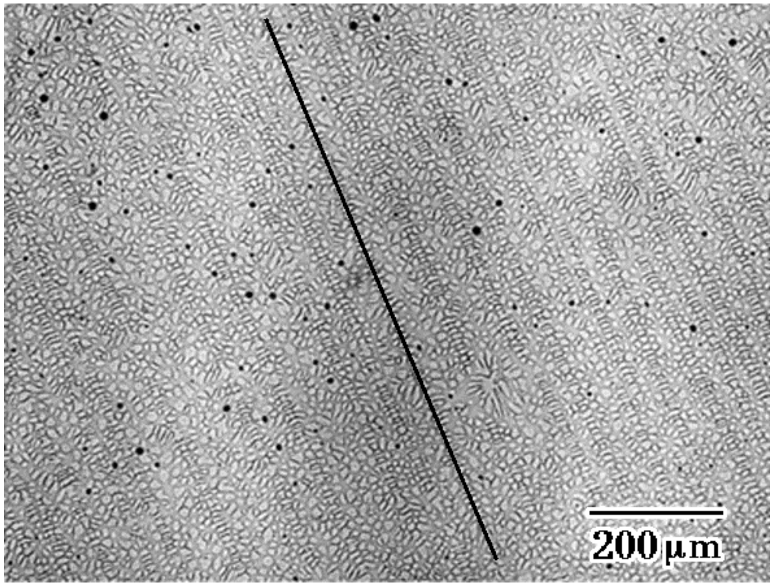
<u>left:</u> section parallel to the thermal gradient of an AlCuMg small ingot obtained by 1D solidification under high gradient, showing the feathery structure, globally parallel to the gradient, but with angle openings corresponding to cumulated little disorientations - the horizontal line structures correspond to the section of a feathery grain the twin planes of which are almost parallel to the section plane -

<u>right:</u> 3D reconstruction of a feathery structure (after anodic oxidation and observation under polarised light) : the plane of the lamellae contains the thermal gradient vector, but a cross-section shows random orientations.[Henry, 1999]

The feathery structure favours microsegregation scale reduction



a/ section parallel to the twin planes: typical space between the trunks: 50 to 100µm



Alloy Al 0.11%Cu

b/ section perpendicular to the twin planes (of which one is highlighted in black): typical space between the trunks: 10µm

3 microsegregation scales in feathery structures:

- **2 spaces between the primary trunks (10 and 100µm)**
- **space between the secondary arms = sdas**

↓

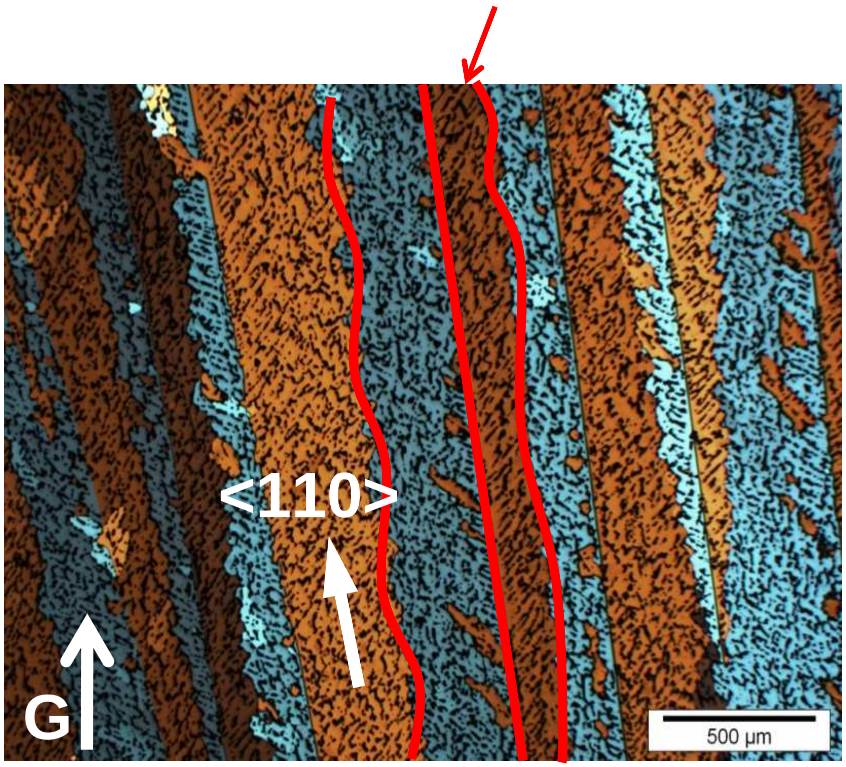
High steric constraint on the residual liquid

Very fine µsegregation

Source: S. Henry's thesis, Lausanne, 1999

Twinned Dendrite Growth in Al Alloys

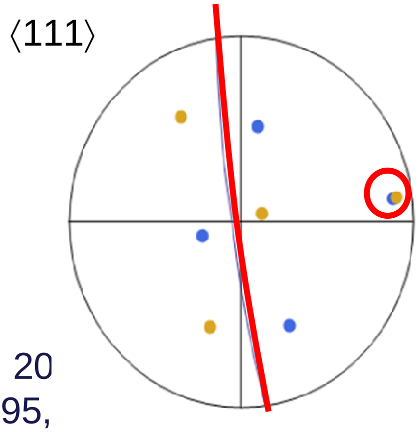
Feathery grains



Al-12wt%Zn

Twinned dendrite are observed in various Al alloys under conditions of

- High cooling rate (dT/dt > 5 K/s)
- High thermal gradient (G > 80 K/cm)
- Strong convection
- Poor or no inoculation



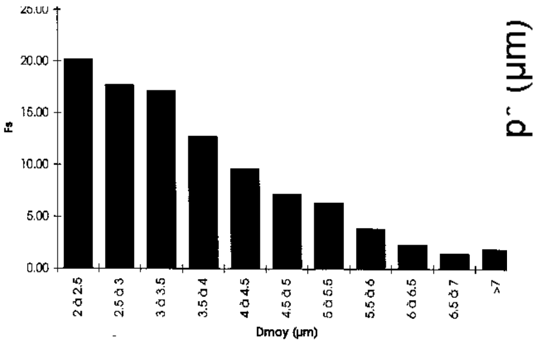
Electron BackScattered Diffraction Analysis (EBSD)

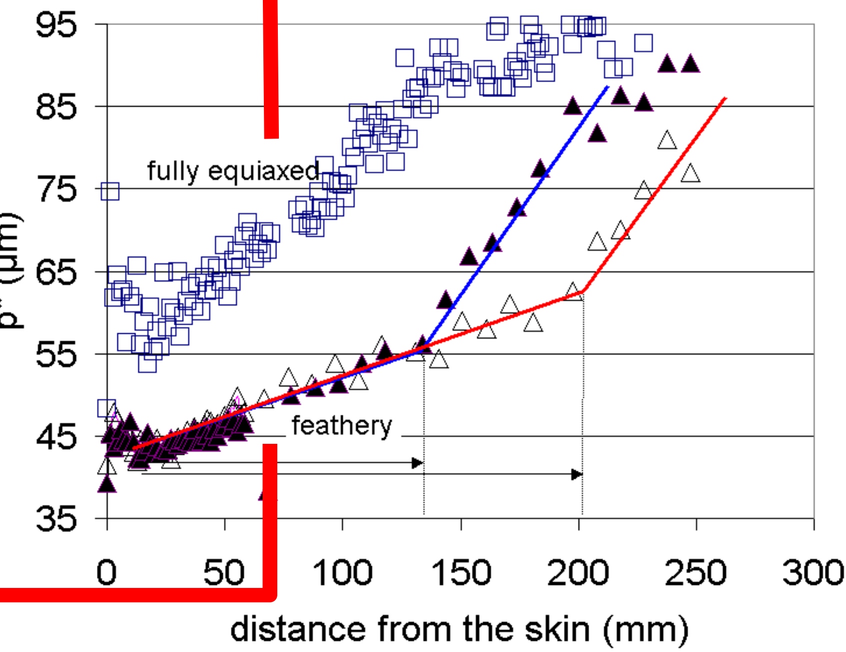
● **Untwinned**

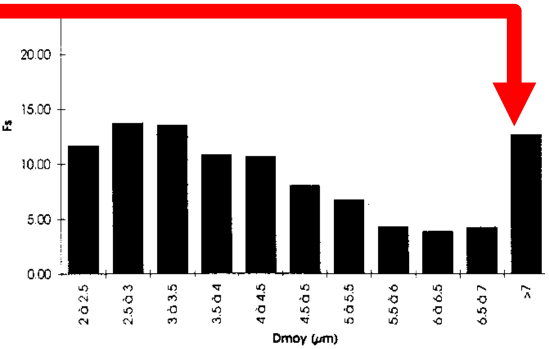
● **Twinned**

M.A. Salgado-Ordorica, et al., Acta Mater., 59, 5085, 20
S. Henry, T.Minghetti, M.Rappaz, Acta Mater., 46, 2495, 1998

Particle size histogram at final rolling thickness







Consequences: crack occurrence in 5182 can ends during rivet forming has a dependency upon the as-cast microsegregation.

Altered formability

Lessons from the the twinned grains microsegregation pattern

Dendrite morphology governed by ?

Solid liquid interfacial energy *anisotropy*

Solidification conditions promoting dendritic growth (nucleation undercooling, gradient, convection…)

$$V \propto \frac{D_\ell \Omega}{\Gamma_{s\ell}} = \frac{D_\ell}{\Gamma_{s\ell} Q} \Delta T$$

$$\begin{cases} \Omega = \dfrac{C_0 - C_L^*}{C_S^* - C_L^*} = \dfrac{\Delta T}{Q} \\ Q = m_L (k-1) C_0 \end{cases}$$

Equiaxed growth velocity

Tip velocity:

Slow diffusing solutes have a higher supersaturation than fast diffusing solutes => *What is the limiting step? and does that influence growth morphology?*

Ways of addressing the problem of growth morphology control:

1. Globular – dendritic transition criterium
2. Playing with anisotropy in phase-field simulation of dendrites
3. Experimental study of chemical influence on dendrite growth direction and morphology
4. Input of liquid structure studies (both experimental and by ab initio calculations

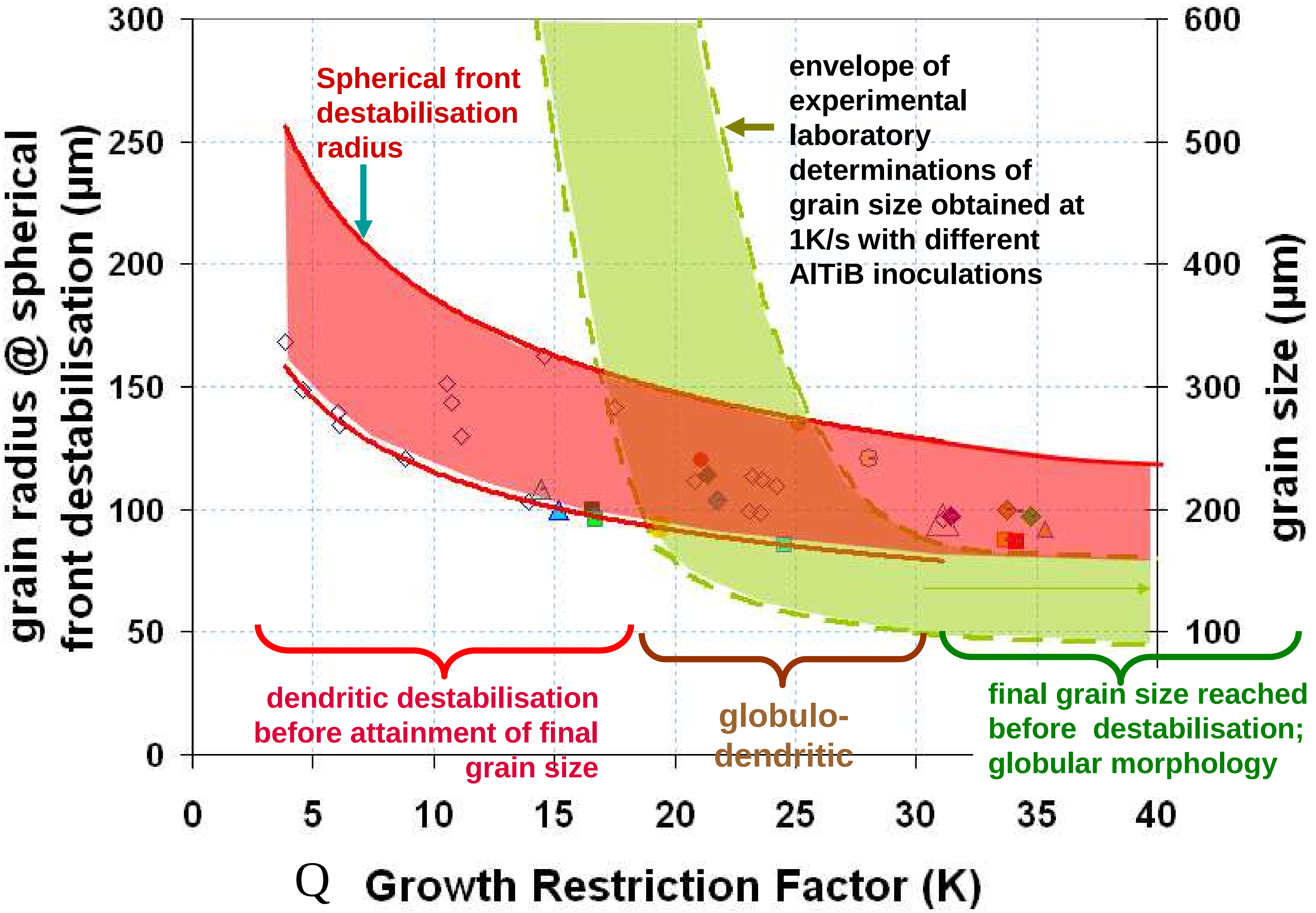
1- Dendritic vs globular growth

$$R_{g-d} = A(\varepsilon)\left[\Gamma_{s\ell}\,\frac{D_\ell}{Q}\,\frac{L_f}{c_p\dot{T}}\right]^{1/3}$$

Anisotropie de γ_{sl}

Diepers & Karma, 2004

Dantzig & Rappaz 2009



Spherical front destabilisation radius

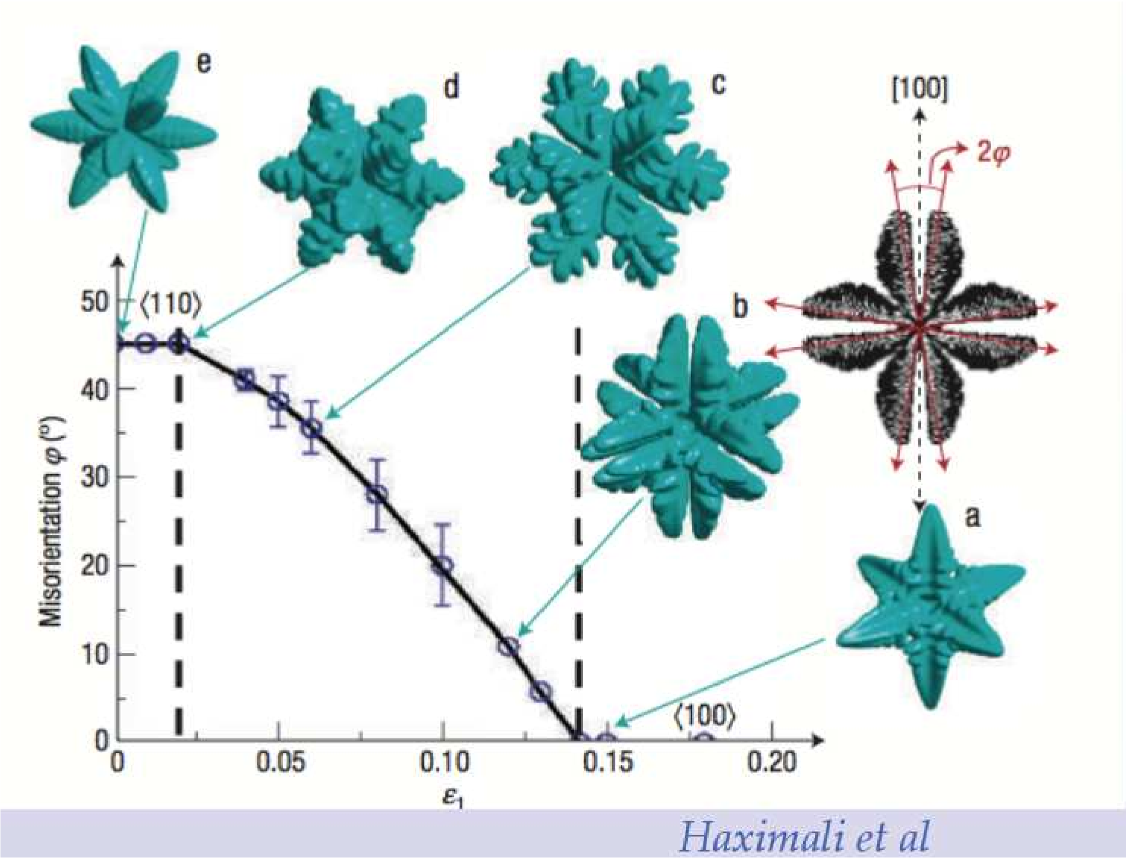
envelope of experimental laboratory determinations of grain size obtained at 1K/s with different AlTiB inoculations

dendritic destabilisation before attainment of final grain size

globulo-dendritic

final grain size reached before destabilisation; globular morphology

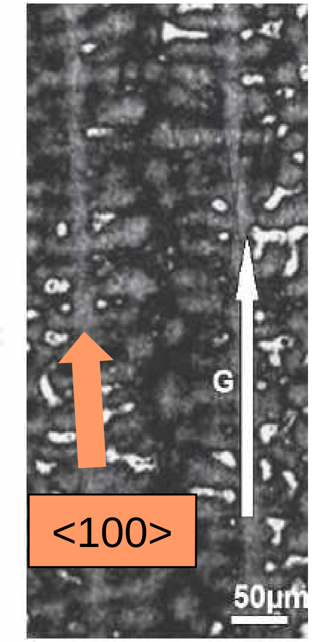
2- Liquid-solid interfacial energy anisotropy



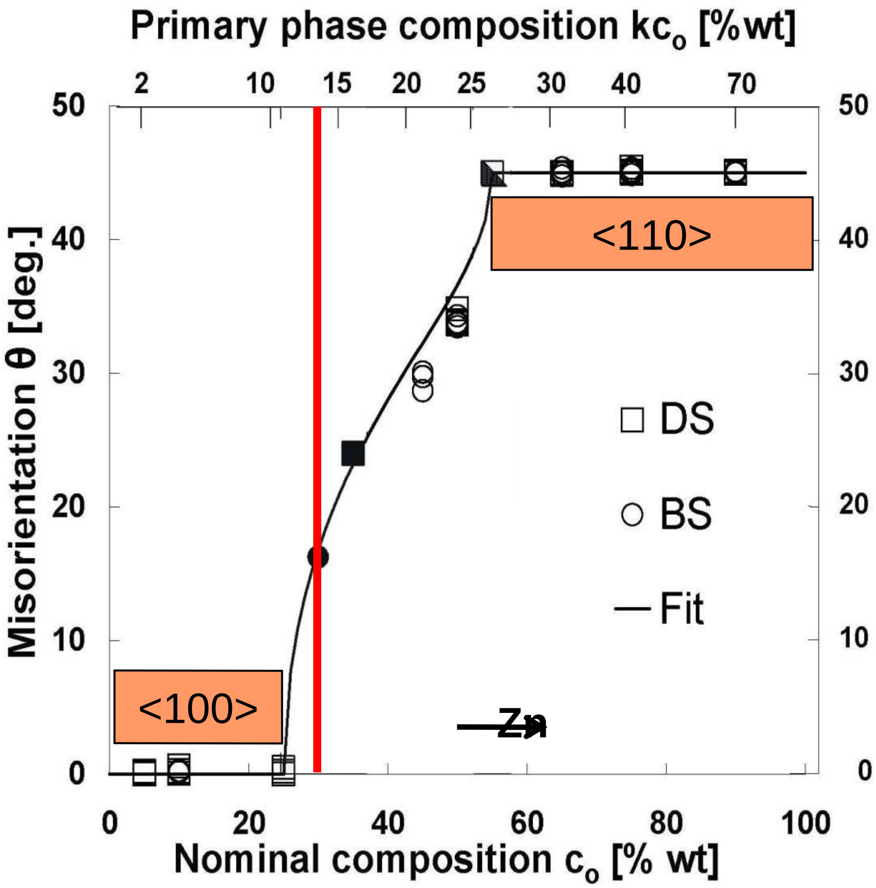
Haximali et al

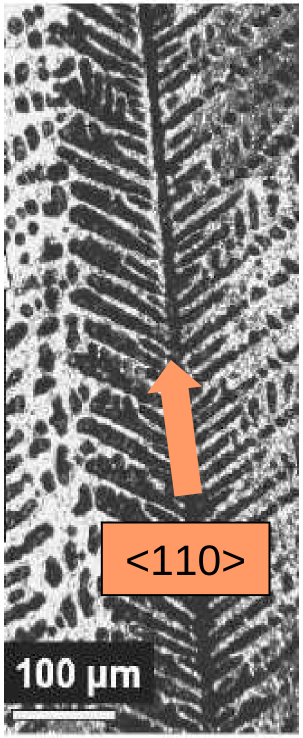
- Which growth morphologies will provide the finest microsegregation distribution? => need for quantitative assessment of 3D virtual morphologies to guide / inspire trials and choice criteria.

3- Growth Directions of Al-Zn Dendrites



Al-10%Zn





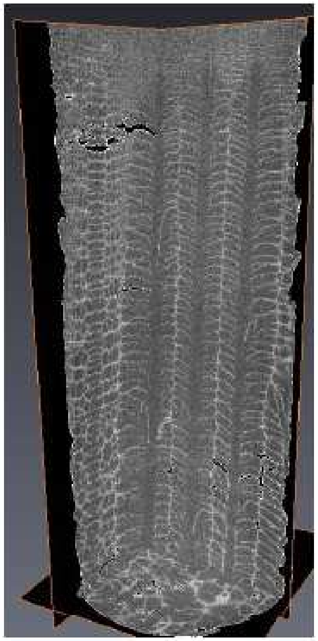
Al-90%Zn

Variability of Al growth direction: existence of a Dendrite Orientation Transition

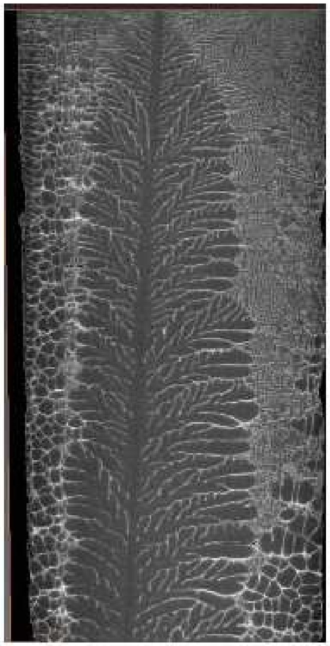
F. Gonzales, M. Rappaz, *Metal. Mater. Trans. A.*, **2006**, *37A*, 2797.

Growth Directions in Al-Zn-Cr

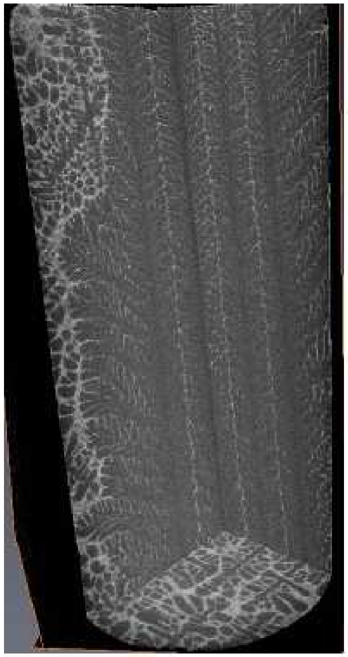
Kurtuldu,
Rappaz,
2011



Al-10% Zn
<100> trunk



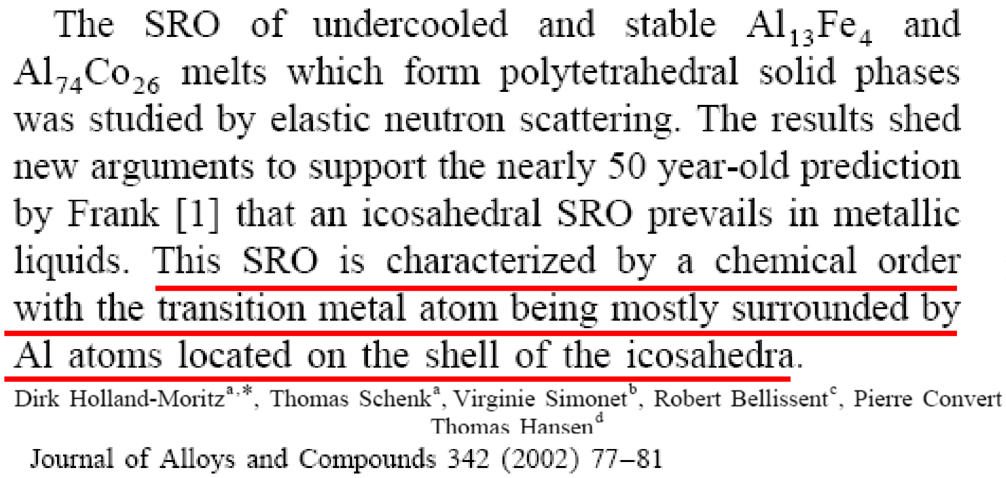
Al-10%Zn-200ppmCr
<100> trunk



Al-10%Zn-1000ppmCr
<210> trunk

- Does the growth direction influence in some way
  - the topology of the solute redistribution?
  - the maturation processes?

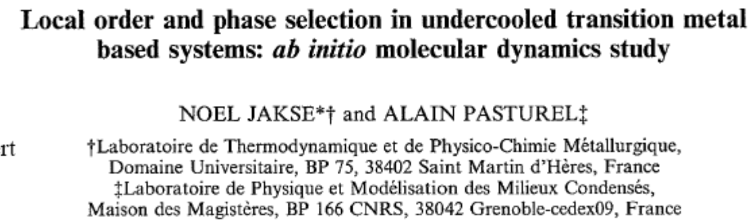
4- Influence of liquid Al-TM structure on TM atoms mobility



The SRO of undercooled and stable $Al_{13}Fe_4$ and $Al_{74}Co_{26}$ melts which form polytetrahedral solid phases was studied by elastic neutron scattering. The results shed new arguments to support the nearly 50 year-old prediction by Frank [1] that an icosahedral SRO prevails in metallic liquids. This SRO is characterized by a chemical order with the transition metal atom being mostly surrounded by Al atoms located on the shell of the icosahedra.

Dirk Holland-Moritz[a,*], Thomas Schenk[a], Virginie Simonet[b], Robert Bellissent[c], Pierre Convert
Thomas Hansen[d]

Journal of Alloys and Compounds 342 (2002) 77–81



Local order and phase selection in undercooled transition metal based systems: *ab initio* molecular dynamics study

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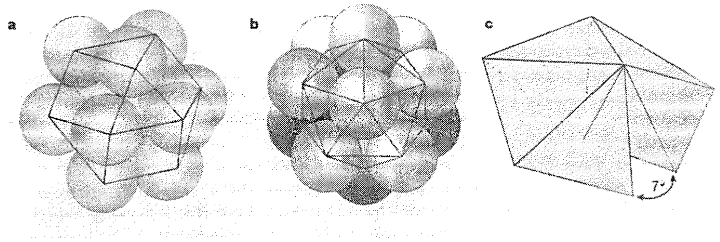
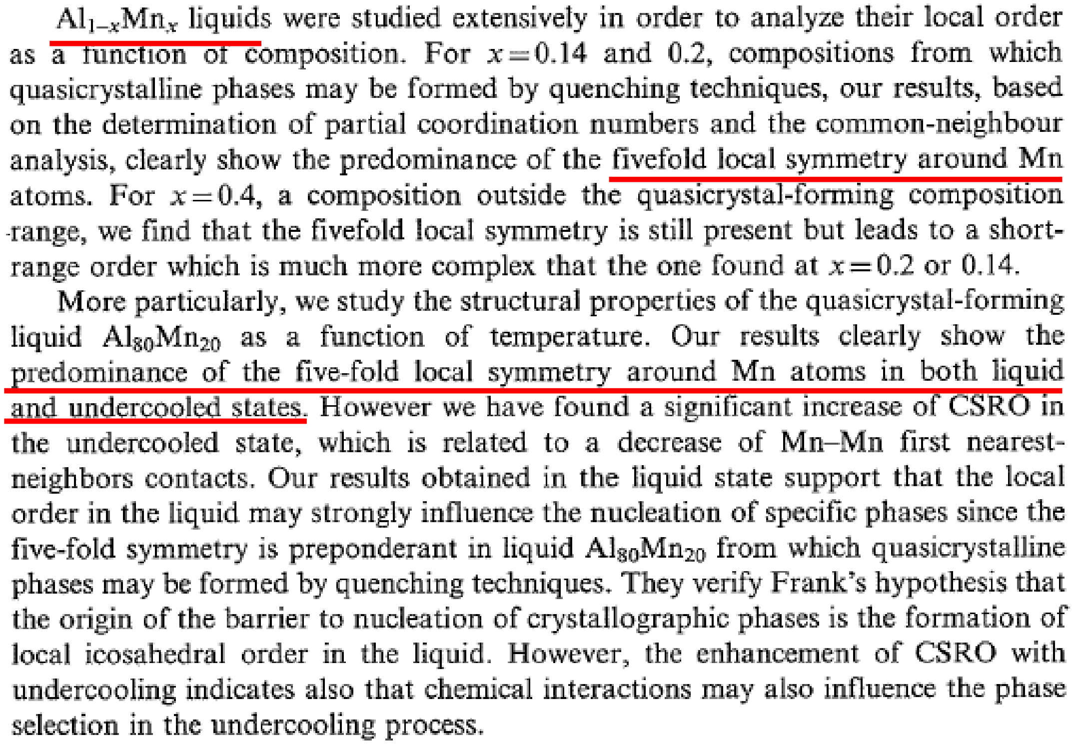


Briques élémentaires d'empilements atomiques ; 12 atomes entourant un atome central peuvent former : a) un arrangement cuboctaédrique comme dans les cristaux cubiques face centrée ; b) un arrangement icosaédrique, comme ceux trouvés dans les liquides. Un anneau de 5 tétraèdres réguliers partageant une même arête laisse un jour de 7° ; ce jour est très labile ; l'agitation thermique du liquide, sa fluidité et la diffusivité atomique élevées peuvent être qualitativement interprétées par la redistribution de ces jours. [Spaepen, 2000]



$Al_{1-x}Mn_x$ liquids were studied extensively in order to analyze their local order as a function of composition. For $x=0.14$ and 0.2, compositions from which quasicrystalline phases may be formed by quenching techniques, our results, based on the determination of partial coordination numbers and the common-neighbour analysis, clearly show the predominance of the fivefold local symmetry around Mn atoms. For $x=0.4$, a composition outside the quasicrystal-forming composition range, we find that the fivefold local symmetry is still present but leads to a short-range order which is much more complex that the one found at $x=0.2$ or 0.14.

More particularly, we study the structural properties of the quasicrystal-forming liquid $Al_{80}Mn_{20}$ as a function of temperature. Our results clearly show the predominance of the five-fold local symmetry around Mn atoms in both liquid and undercooled states. However we have found a significant increase of CSRO in the undercooled state, which is related to a decrease of Mn–Mn first nearest-neighbors contacts. Our results obtained in the liquid state support that the local order in the liquid may strongly influence the nucleation of specific phases since the five-fold symmetry is preponderant in liquid $Al_{80}Mn_{20}$ from which quasicrystalline phases may be formed by quenching techniques. They verify Frank's hypothesis that the origin of the barrier to nucleation of crystallographic phases is the formation of local icosahedral order in the liquid. However, the enhancement of CSRO with undercooling indicates also that chemical interactions may also influence the phase selection in the undercooling process.

Phase Transitions,
Vol. 80, Nos. 4–5, April–May 2007, 369–384

Conclusions and Perspectives (1)

- *ab initio* calculations of liquid alloys structure should help and guide us to:
  - Revisit the fundamentals of nucleation of primary alpha and intermetallic phases (precursor phases in liquid structure)
  - What actually controls the minimum achievable grains size?
  - Assess the mobility of species in liquid alloys, essential for understanding and predicting solidification morphology.

- The process counterpart of these fundamental studies should be to explore ways of *creating precursors of surfaces within the liquid* / wetting present inclusions by means of energy fields
  - Ultrasonication
  - High turbulence (vorticity surfaces are surface precursors in a one phase liquid)

  The input of scientists working on sonication or turbulence would be beneficial to the solidification community => who in CNRS or University?

Conclusions and Perspectives (2)

- Develop studies of chemical control of solidification growth morphology / interfacial energy anisotropy, both for dendrites and eutectics.

➢ *Is it possible to determine anisotropy coefficients from X-ray tomography on unidirectionally grown dendrites and comparison with Phase-Field calculations?*

  – Aluminium based alloys: Al has a very low liq sol interfacial energy anisotropy => very sensitive to minute additions. On-going studies.

  – Open question for Fe-based alloys (Fe-Mn, Fe-Nb): is there any hope to influence growth morphology and microsegregation distribution ?

(This is addressed in topic n°1 of GDR SAM)

Conclusions and perspectives (3)

Other identified topics of GDR SAM
(Solidification des Alliages Métalliques)

- Macrosegregation modelling with complete coupling between heat flow, thermosolutal convection and transport of growing grains. *Probably more input from the Fluid Mechanics community would be interesting (KH instabilities, Holmboe waves, etc…)*

- Porosity modelling / interaction with macrosegregation

- Hot Cracking modelling (*interesting granular approaches by Rappaz or Gourlay*)