Le changement de phase
liquide ➔ solide

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Action Nationale de Formation « METALLURGIE FONDAMENTALE »
22-25 octobre 2012, Aussois, France
Alloys are produced by primary melting and transformed by secondary melting, thus involving the liquid-to-solid phase transformation.

Primary production of alloys in 2010

<table>
<thead>
<tr>
<th></th>
<th>Fe</th>
<th>Al</th>
<th>Cu</th>
<th>Zn</th>
<th>Pb</th>
<th>Ni</th>
<th>Sn</th>
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</thead>
<tbody>
<tr>
<td>World (Mtons)</td>
<td>1'518</td>
<td>40.4</td>
<td>14.6</td>
<td>12.9</td>
<td>8.8</td>
<td>1.4</td>
<td>0.3</td>
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<tr>
<td>France (Mtons)</td>
<td>15.8</td>
<td>0.421</td>
<td>0.428</td>
<td>0.163</td>
<td>0.088</td>
<td>0.107</td>
<td>-</td>
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<tr>
<td>France (people)</td>
<td>23’800</td>
<td>3’594</td>
<td>3’600</td>
<td>-</td>
<td>-</td>
<td>4’300</td>
<td>-</td>
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</tbody>
</table>

Solidification is part of metallurgy textbooks

### Laboratories/People in France

- GdR CNRS 3328 « Solidification des Alliages Métalliques » (COMBEAU)
  - « Formation des microstructures » (BILLIA, AKAMATSU, LACAZE, DALOZ)
  - « Dynamique de la zone pâteuse » (RAPPAZ, ZALOZNIK, SUERY)
  - « Procédés » (GANDIN, FAUTRELLE, DREZET, PEYRE, BIGOT, DUFFAR)

<table>
<thead>
<tr>
<th>Laboratory</th>
<th>People</th>
<th>Section CN</th>
<th>Laboratory</th>
<th>People</th>
<th>Section CN</th>
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<td>IJL, Nancy</td>
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<td>CDM, Evry</td>
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<td>14, 15</td>
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<td>IM2NP, Marseille</td>
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<td>5, 15</td>
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<td>INSP, Paris</td>
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<tr>
<td>IRPHE, Marseille</td>
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<td>LPMC, Palaiseau</td>
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<tr>
<td>LSMX, Lausanne, CH</td>
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<td></td>
<td>CEA-INES, Le Bourget-du-Lac</td>
<td>6</td>
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</table>

**Commission SF2M « Coulée et Solidification » (JARRY)**

[http://spinonline.free.fr/GDR](http://spinonline.free.fr/GDR)
Processes

- **Semi-finished products**
  - continuous, semi-continuous, ingots (casting)

- **Near-net-shape products**
  - expendable: sand, plaster, shell, plaster, investment, loast foam (casting)
  - non-expendable: centrifugal, semi-solid, die, permanent mold (casting)

- **Assembly**
  - welding (TIG, MIG, MAG, laser, …)
  - soldering, brazing
Defects

- Part integrity (cracks, misruns, …)
- Structure selection (columnar, equiaxed, dendritic, eutectic, …)
- Phase selection (nature and size)
- Crystallographic texture (fiber)
- Segregation (inverse, buoyancy forces, grain sedimentation, intergranular, interdendritic, freckles, …)
- Porosity (hot spot, shrinkage, gas)
- Residual stresses
- Hot tears
Structures

- Structures formed from a binary melt
  - dendritic, $l_1 \rightarrow \alpha + l_2$ (monovariant reaction),
  - peritectic, $l + \alpha \rightarrow \beta$ (invariant reaction),
  - eutectic, $l \rightarrow \alpha + \beta$ (invariant reaction),
  - congruent, $l \rightarrow \alpha$ (invariant reaction),
  - monotectic, $l_1 \rightarrow l_2 + \alpha$ (invariant reaction).

- Selection and fraction of structures and phases are influenced by
  - alloy composition (industrial = multicomponent),
  - departure from thermodynamic equilibrium (nucleation and growth undercooling),
  - diffusion in phases (with $D^s << D^l$),
  - phase flow (melt and solid),
  - heat flow (temperature gradient, isotherm velocity).
Segregations

- Segregations form due to
  - thermodynamic equilibrium between phases with different solubility of species, defined by the segregation coefficient $k = (X^s/X^l)_{T,p}$

- Main outputs of segregation studies
  - composition profile and average composition of phases,
  - fraction of structures and phases,
  - fraction of phases in each structure,
  - solidification path (above information as a function of temperature from $T_L$ to $T_S$)

- Simple examples of solidification paths, with common assumption a uniform liquid composition ($D^l >> 0$)
  - Lever Rule (LR): equal chemical potentials of all species in all phases ($D^s >> 0$)
  - Gulliver-Scheil (GS): same as LR at the s/l interface with $D^s = 0$
  - Partial Equilibrium (PE): mixture of GS for substitutionals and LR for interstitials
**Scales**

- Atom Probe Tomography
- Electron microscopy
- Optical microscopy
- Casting

**Observations**

- Molecular dynamics
- Phase field
- Cellular automaton
- Continuum mechanics

**Simulations**

**Introduction**
## Methods

<table>
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<tr>
<th>Scales</th>
<th>Indirect</th>
<th>Tracking</th>
<th>Direct</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Macroscopic</strong></td>
<td><strong>Volume averaging over a multiphase domain</strong></td>
<td><strong>Solidification paths</strong></td>
<td><strong>Tracking of domain boundaries</strong></td>
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<tr>
<td><strong>Grain</strong></td>
<td><strong>Volume averaging over each independent phase</strong></td>
<td><strong>Continuum mechanics</strong></td>
<td><strong>Solidification paths and interface kinetics</strong> for the prediction of grain size distribution</td>
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<tr>
<td><strong>Microscopic</strong></td>
<td></td>
<td></td>
<td><strong>Structure kinetics</strong> for the prediction of grain size distribution</td>
</tr>
</tbody>
</table>

- **Solidification paths and interface kinetics for the prediction of average phase fractions and compositions**
- **Diffuse interface for the prediction of microstructure**
Contents

- **Introduction**
  - Statistics, Processes & Defects
  - Structures & Segregations
  - Scales & Methodologies

- **Direct microscopic methodology**

- **Indirect microscopic methodology**
  - Observations: electromagnetic levitation and atomization
  - Kinetics modeling coupled with equilibrium calculations
  - Applications and current extensions

- **Indirect macroscopic methodology**
  - Phase field modeling of microstructures

- **Direct macroscopic methodology**
  - CAFE modeling
  - Application to ESRF observations
  - Application to directionally solidified grain structure
  - Application to a macrosegregation benchmark experiment

- **Perspectives**
Direct microscopic methodology

- The phase field method tracks to phase interfaces
- Applications to solidification include quantitative studies on structure dynamics (PLAPP, Ecole Polytechnique, Palaiseau)
- Most developed models consider multicomponent alloys and multiple phase transformations coupled with thermodynamic databases (Code MICRESS, ACCESS, Aachen, DE)
Limits of interface tracking

- Biggest phase field result for a network of dendrites in Al-Si?

Size is given by the number of grid point: 4096x1024x4096 (only few cubic millimeters!)

Parallel computation with up to 4’000 GPUs and 16’000 CPUs, reaching 2 petaFLOPS

GPU = Graphics Processing Unit, CPU = Central Processing Units, petaFLOPS = $10^{15}$ Floating point Operations Per Seconds

Indirect microscopic methodology

- **Collaborations**
  - D. TOURRET, MINES ParisTech, Sophia Antipolis
  - D. HERLACH, Th. VOLKMANN, D. TOURRET, DLR, Köln, DE
  - M. CALVO-DAHLBORG, U. DAHLBORG, C. M. BAO, University, Rouen
  - G. REINHART, University Paul Cézanne, Marseille,
  - G. N. ILES, ESRF/ILL, Grenoble

- **Funding**
  - European Community, Brussels, BE
  - European Space Agency, Noordwijk, NL
  - Bundesvereinigung Materialwissenschaft und Werkstofftechnik e.V., Berlin, DE
  - Deutsche Forschungsgemeinschaft, Bonn, DE
  - Bundesanstalt für Materialforschung und –prüfung, Berlin, DE

Observations

- Gas atomization of Al-Ni alloys

SEM image of a Ni-75at% Al 100 µm diameter droplet produced by gas atomization
Concurrent multiple phase transformations

- Constant and equal density in all phases
- Uniform T with heat exchange rate Q
- Zones & phases defined by boundaries & interfaces
  - (0): \( l^{(0)} \) \( s_1^{(1)} + l^{(1)} \)
  - (1): \( s_1^{(2)} + s_2^{(2)} + l^{(2)} \)
  - (3): \( s_1^{(3)} + s_2^{(3)} + s_3^{(3)} \)
- Microstructure propagation at zone boundaries with growth kinetics
  - \( s_1 \): dendritic
  - \( s_2 \): peritectic
  - \( s_3 \): eutectic
- Thermodynamic equilibrium at phase interfaces
- Unknowns: average composition and phase fractions
  - \( l^{(0)} \), \( l^{(1)} \), \( s_1^{(1)} \)
  - \( l^{(2)} \), \( s_1^{(2)} \), \( s_2^{(2)} \)
  - \( s_1^{(3)} \), \( s_2^{(3)} \), \( s_3^{(3)} \)
Modeling

Indirect micro

- **Average total mass**
  
  over phase \((\alpha)\):
  \[
  \frac{\partial}{\partial t} \left( g^\alpha \right) = \sum_{\beta (\beta \neq \alpha)} \left( S^{\alpha \beta} v_n^{\alpha \beta} \right)
  \]
  on interface \((\alpha \beta)\):
  \[
  v_n^{\alpha \beta} + v_n^{\beta \alpha} = 0
  \]

- **Average solute mass**
  
  over phase \((\alpha)\):
  \[
  g^\alpha \frac{\partial}{\partial t} \left( \langle w^{\alpha \alpha} \rangle \right) = \sum_{\beta (\beta \neq \alpha)} \left[ S^{\alpha \beta} \left( w^{\alpha \beta} - \langle w^{\alpha \alpha} \rangle \right) \left( v_n^{\alpha \beta} + \frac{D^\alpha}{I^{\alpha \beta}} \right) \right]
  \]
  on interface \((\alpha \beta)\):
  \[
  \left( w^{\alpha \beta} - w^{\beta \alpha} \right) v_n^{\alpha \beta} + \frac{D^\alpha}{I^{\alpha \beta}} \left( w^{\alpha \beta} - \langle w^{\alpha \alpha} \rangle \right) + \frac{D^\beta}{I^{\beta \alpha}} \left( w^{\beta \alpha} - \langle w^{\beta \beta} \rangle \right) = 0
  \]

- **Average energy balance**
  
  Global heat balance:
  \[
  \rho \frac{\partial \langle H \rangle}{\partial t} = \rho \sum_{\alpha} \left( \langle H^{\alpha \alpha} \rangle \frac{\partial g^\alpha}{\partial t} + g^\alpha \frac{\partial \langle H^{\alpha \alpha} \rangle}{\partial T} \frac{\partial T}{\partial t} + g^\alpha \frac{\partial \langle H^{\alpha \alpha} \rangle}{\partial \langle w^{\alpha \alpha} \rangle} \frac{\partial \langle w^{\alpha \alpha} \rangle}{\partial t} \right) = -h_c S^{\text{ext}} (T - T_{\text{ext}})
  \]

- **Additional relations and Unknowns**
  
  Geometry & Composition profiles assumptions:
  \[ S^{\alpha \beta}, I^{\alpha \beta}, S^{\text{ext}} \]
  Boundary conditions:
  \[ h_c, \text{ Text Data: } D^\alpha, \Delta T_n^{\alpha} \]
  Thermodynamic equilibrium:
  \[
  \langle H^{\alpha \alpha} \rangle, \frac{\partial \langle H^{\alpha \alpha} \rangle}{\partial T}, \frac{\partial \langle H^{\alpha \alpha} \rangle}{\partial \langle w^{\alpha \alpha} \rangle}
  \]
  Interface:
  Equilibrium:
  \[ w^{\alpha \beta} \]
  Boundary:
  Growth kinetics:
  Solute balance & Continuity:
  \[ v_n^{\alpha \beta} \]
  Partial Derivative Equations:
  \[ g^\alpha, \langle w^{\alpha \alpha} \rangle, T \]
Electromagnetic levitation

**Containerless processing**
- Controlled conditions, almost uniform temperature, observation of nucleation events

**Droplet: benchmark system for volume averaged models**
- **Alloy**: Ni - 75 at% Al
- **Thermodynamic database**: PBIN
- **System radius** \(d\): 3.82 \(\times\) 10\(^{-3}\) m
- **Dendrite arm spacing** \(d\): 36 \(\times\) 10\(^{-6}\) m
- **Diffusion coefficients**
  - Liquid: \(10^{-9}\) m\(^2\) s\(^{-1}\)
  - \(\text{Al}_3\text{Ni}_2, \text{Al}_3\text{Ni}\): \(5 \times 10^{-11}\) m\(^2\) s\(^{-1}\)
- **Gibbs-Thomson coefficients**
  - \(\text{Al}_3\text{Ni}_2\): \(2 \times 10^{-8}\) K m
  - \(\text{Al}_3\text{Ni}^M\): \(3.5 \times 10^{-8}\) K m
  - \(\text{Al}^M\): \(1.86 \times 10^{-7}\) K m
- **Nucleation undercooling**
  - \(\text{Al}_3\text{Ni}_2, \text{Al}_3\text{Ni}, \text{Al}\): 0 K, 150 K, 15 K
- **Heat transfer** \(d\): 213.8 W m\(^{-2}\) K\(^{-1}\)
- **External temperature** \(d\): 293 K

\(d\): *deduced from measurements*

**Regime I: Classical Fourier number analysis**

The peritectic transformation controls the phase fractions due to diffusion in the solid phases. It is favored for lower cooling rates (larger particles).

**Regime II: Al₃Ni primary growth**

Primary growth of Al₃Ni controls the phase fraction. It prevents the development of Al₃Ni₂.

**Regime III: Al₃Ni₂ and Al₃Ni growth competition**

Mixed regime where the peritectic reaction for Al₃Ni can catch-up with the dendritic reaction of Al₃Ni₂ and become primary when the cooling rate is increased.

**Evidence of growth competition between phases formed from the melt**

Current extensions: multicomponent alloys (AM1), multiple phase transformations (FeCCr)
Collaborations

- T. CAROZZANI, M. BELLET, H. DIGONNET, MINES ParisTech, Sophia Antipolis
- Y. FAUTRELLE, K. ZAIDAT, Institut National Polytechnique de Grenoble, Grenoble
- G. REINHART, N. MANGELINCK-NOËL, H. NGUYEN-THI, B. BILLIA, Aix-Marseille Université, Marseille
- J. BARUCHEL, ESRF, Grenoble

Funding

- European Space Agency, Noordwijk, NL
- Agence Nationale de la Recherche, Paris
Two-phase model assuming $\langle \rho \rangle^s = \langle \rho \rangle^l = \rho_0$ = constant and $\langle v \rangle^s = 0$

**Total mass conservation**

$\nabla \cdot \langle v \rangle = 0$

**Average flow velocity**

$\langle v \rangle = \langle v^l \rangle = g^l \langle v \rangle^l$

**Momentum conservation**

$$\rho_0 \frac{\partial \langle v^l \rangle}{\partial t} + \frac{\rho_0}{g^l} \nabla \cdot (\langle v^l \rangle \times \langle v^l \rangle) - \nabla \cdot (\mu^l \nabla \langle v^l \rangle) + g^l \nabla p^l - g^l \rho g + \frac{\mu^l}{K} g^l \langle v^l \rangle = 0$$

**Boussinesq approximation**

$$\rho = \rho_0 \left[ 1 - \beta_{th} (T - T_0) - \beta_w (w^l - w_0^l) \right]$$

**Carman-Kozeny permeability**

$$K = \left( g^{l3} \lambda_2 \right)/ \left[ 180 (1-g^l)^2 \right]$$

**Energy conservation**

$$\rho_0 \left( \frac{\partial \langle H \rangle}{\partial t} + \langle v \rangle \cdot \nabla \langle H \rangle^l \right) - \nabla \cdot (\kappa^l \nabla T) = 0$$

**Average enthalpy**

$$\langle H \rangle = g^s \langle H \rangle^s + g^l \langle H \rangle^l$$

**Solute mass conservation**

$$\frac{\partial \langle w \rangle}{\partial t} + \langle v \rangle \cdot \nabla \langle w \rangle^l - \nabla \cdot (D^l g^l \nabla \langle w \rangle^l) = 0$$

**Average composition**

$$\langle w \rangle = g^s \langle w \rangle^s + g^l \langle w \rangle^l$$

The FE method provides a solution for $\langle v \rangle, \langle H \rangle, \langle w \rangle, T$

G. Guillemot et al. 2007 Journal of Crystal Growth 303 58
Transport between scales

- A CA grid of square cells is superimposed on top of the FE mesh
- Topological links are defined between the FE nodes and the CA cells
- Quantity at cell $\xi$, $\xi_v$ (e.g., $\langle v \rangle$, $\langle H \rangle$, $\langle w \rangle$, $T$), is computed from quantity at nodes $n$, $\xi_n$, thanks to interpolation coefficients $c$

$$\xi_v = \sum_n c^n_v \xi_n$$

and oppositely,

$$\xi_n = \left( \sum_v c^n_v \xi_v \right) / \left( \sum_v c^n_v \right)$$

Carozzani et al. 2012 MSMSE 20 015010
Nucleation algorithm

- **State index of cell \( \nu \) wrt structure (\( j \)), \( l^{(j)}_{\nu} \)**
  - 0: no growing structure
  - 1: growing structure
  - 2: structure growth over

- **Structure nucleation in cell \( \nu \)**
  - Test for \( l^{(j)}_{\nu} = 0 \) prior to nucleation, change to \( l^{(j)}_{\nu} = 1 \) after nucleation
  - Test for \( \Delta T_{\nu} > \Delta T_{\text{nucl}}^{(j)}_{\nu} \), the activation undercooling of the nucleation site of structure (\( j \)) located in cell \( \nu \) (undercooling = temperature difference between the local liquidus temperature, \( T_L \), and the actual cell temperature, \( T_{\nu} \))
  - Initialization of a set of Euler angles \( (\phi_1, \Phi, \phi_2)^{(j)}_{\nu} \) defining the 6 perpendicular \(<100>\) directions
  - Growth center \( C^{(j)}_{\nu} \) coincides with cell center \( C_{\nu0} \)
  - Initialization of the lengths of the preferred \(<100>\) growth directions \( R^{(j)<100>}_{\nu} \) (for a \(<100>\) dendritic structure (\( j \)))
Growth algorithm

Structure growth in cell $\nu$

- Increase of lengths $R^{(j)<100>}_{\nu}$ by time integration of a phenomenological dendrite tip growth kinetics model that depends on local temperature, $T_{\nu}$, composition, $<w>_{\nu}$, and liquid velocity, $<v>_{\nu}$

- Calculation of volume associated with the growth shape of cell $\nu$, $V^{(j)}_{\nu \text{ capt}} = f(C^{(i)}_{\nu}, R^{(j)<100>}_{\nu})$, and the maximum volume required to capture all neighboring cells, $V^{(j)}_{\nu \text{ max}}$

- Calculation of the fraction of structure (j) in cell $\nu$, $g^{(j)}_{\nu} = \min \left[ \frac{(V^{(j)}_{\nu} - V^{(j)}_{\nu \text{ capt}})}{(V^{(j)}_{\nu \text{ max}} - V^{(j)}_{\nu \text{ capt}})}, 1 \right]$

Structure propagation to a neighboring cell $\mu$

- Test for $I^{(i)}_{\mu} = 0$ prior to capture, test for liquid in cell $\mu$, change to $I^{(i)}_{\mu} = 1$ after capture

- Test for the center of cell $\mu$ inside the growth shape associated with cell $\nu$

- Propagation of the grain $(\phi_1, \Phi, \phi_2)^{(j)}_{\mu} = (\phi_1, \Phi, \phi_2)^{(j)}_{\nu}$

- Calculation of $C^{(i)}_{\mu}$, $R^{(j)<100>}_{\mu}$, $V^{(j)}_{\mu \text{ capt}}$, $V^{(j)}_{\mu \text{ max}}$ and $g^{(j)}_{\mu}$
Nucleation kinetics

- Gaussian distribution of nucleation sites for a structure (j) as a function of the undercooling
  - Random selection of cells for assignment of nucleation sites
  - Random selection of Euler angles ($\phi_1, \Phi, \phi_2$) for each nucleation site

Growth kinetics

- Phenomenological growth kinetics model for the shape associated to the cells
  - Ivantsov solution and marginal stability (dendrite tip kinetics)
  - Extension to account for the fluid flow intensity and direction with respect to the growth directions (assumed <100> for cubic materials)

Mushy zone solidification

- Tabulation of thermodynamic properties for each phase in structure (j) assuming a given solidification path (phase volume fraction, phase composition and phase enthalpy as a function of temperature and the average composition)
ESRF directional set-up

Experimental set-up at the European Synchrotron Radiation Facility (ESRF, beam line ID19)

- Bridgman Furnace
- Vacuum chamber
- Monochromatic beam (~ 13.5 keV)
- Vacuum chamber
- FReLoN camera

Sample dimension: 37 mm x 6 mm x 200 µm

Soft crucible: graphite foils + Mo frame and clips

Field of view: 15 x 6 mm²
Pixel size: 7.46 µm
1 frame / 3 seconds

In situ real-time X-Ray imaging of phases in Al - 3.5 wt% Ni

Direct macro

In-situ imaging of solidification

- Directional solidification of Al - 3.5 wt% Ni

V = 1.5 µm/s

- **Dendritic grain**
  - envelope delimited by the dendrite tips
  - mixture of dendritic Al-rich solid \( s \) plus interdendritic liquid \( d \) or interdendritic eutectic
  - growth in extradendritic liquid phase \( l \), stopped by the extradendritic eutectic \( e \)

- **Segregation of elements at s/l interface**
  - average composition \( w_s, w_d, w_l \)
  - average fraction of phases \( g_s, g_d, g_l \)

- **Dendritic-to-eutectic transition (DET) 1.5 µm/s**

- **Columnar-to-equiaxed transition (CET) upon velocity jump at 15 µm/s**

- **Distribution of inter- and intra-granular eutectic?**
  - Role of dendritic grain structure on eutectic distribution?

Al - 3.5 wt% Ni,

\[ G = 20 \, \text{K/cm}, \, V = 1.5 \rightarrow 15 \, \text{µm/s} \]

X-Ray radiography
Measured parameters as inputs for simulations

- Characterization of the nucleation event for each grain: position $x_i$, $y_i$ + orientation $\theta_i$
- Measurement of the average dendrite arm spacing: 130 $\mu$m
- Measurement of time evolution of the size of the mushy zone

The interaction of the liquid flow with the structure is computed
The segregation pattern is linked with the grain structure and the eutectic fractions
The Columnar-to-Equiaxed Transition (CET) is reproduced
Inter- and intra-granular eutectics are partly distinguished
Dendritic-to-eutectic transition is qualitatively retrieved
To be improved…

- Direct tracking of the primary eutectic structure is missing to reach better comparison with in-situ observation.
- 3D effects can not be fully neglected.
- Assessment of nucleation undercooling and better measurements of the temperature fields would permit improved comparison.
- Measurements of the composition field and the liquid velocity fields are missing.
- Coupling with thermo-mechanics is required to give access to shrinkage and deformation.
- Sedimentation of the grains is not accounted for.
Parallel FE solver
- Coupled solutions for heat, solute and fluid flows
- 3D automatic remeshing

Parallel CA solver
- Dynamic allocation of CA grid based on FE mesh
- Nucleation and growth of dendritic and eutectic

Coupling between the CA and FE solvers
- Computation of mushy zone fraction from CA cells
- Conversion of average enthalpy at FE nodes

Coupling with thermodynamic
- Tabulated enthalpies as a function of temperature
- Tabulated solidification path (ThermoCalc+PBIN)

Competition between columnar and equiaxed structures

Carozzani et al. 2012 MSMSE 20 015010
- Retrieves well the occurrence of a recalescence recorded at 140 mm in the equiaxed region and the inflection recorded at 100 and 120 mm (validation of the coupling for heat flow together with the growth undercooling)
SnPb in rectangular cavity – Set-up

- Developed at Institut National Polytechnique (Grenoble, FR)

- Inspired from Hebditch and Hunt 1974 Metall. Trans. 5 1557
  - Left-hand-side (LHS) and right-hand-side (RHS) heat exchangers with independent control of the time evolution of the temperature
  - 100x60x10 mm$^3$ geometry
  - Sn - 3 wt% Pb alloy
  - $T_{\text{LHS}} - T_{\text{RHS}} = 40$ °C
  - Cooling rate = -0.03 °C/s
  - Thermocouples
    - 10 columns, 5 rows
  - Positions from LHS (mm)
    - L21  L24  L27  L30
    - 5    35   65   95
- Accurate measurements with control temperature at LHS and RHS boundaries
- Possibility to extract time evolution of temperature maps
SnPb solidification paths – Tab.

Phase diagram (PBIN)

Solidification paths (LR)

Temperature (°C) vs Pb composition (wt.%)

LIQ

FCC

Temperature (°C) vs Volume fraction of phases (-)

BCT

38.3 wt%

181.4°C

4.2 wt% 83 wt%
SnPb properties – Tab.

Phase composition

Phase enthalpy
SnPb in rectangular cavity – Flows

- CA grid: 200 µm (7’500’000 cells)
- FE mesh:
  - Initial: 1’200 µm (76’000 nodes)
  - Final: [780-1’200] µm
- Time step: 0.1 s
- Cluster: 64 processors
- CPU time: 4 days

Nucleation sites
- RHS: \(n=5 \cdot 10^4 \text{ m}^{-2}, \Delta T_a=1.5 \, ^\circ\text{C}, \Delta T_\sigma=0.5 \, ^\circ\text{C}\)
- Volume: \(n=10^7 \text{ m}^{-3}, \Delta T_a=5 \, ^\circ\text{C}, \Delta T_\sigma=0.5 \, ^\circ\text{C}\)

Isothermal surfaces every 2°C
Flow (max arrow 2.5 cm s\(^{-1}\))
(Time acceleration x100)

T. Carozzani et al. 2012
Metallurgical and Materials Transactions in press.
SnPb in rectangular cavity – Segreg. Direct macro

Radiography

3D FE simulation

3D CAFE simulated segregation map
SnPb in rectangular cavity – Grains

Casting surface

CAFE V_I
n = 10^8 m^{-3}
\Delta T_a = 3.5 ^\circ C
\Delta T_\sigma = 0.5 ^\circ C

CAFE V_II
n = 10^7 m^{-3}
\Delta T_a = 5 ^\circ C
\Delta T_\sigma = 0.5 ^\circ C
SnPb in rectangular cavity

Temperature [°C]

Time [s]

Solid fraction [-]

Direct macro
SnPb in rectangular cavity

Direct macro

Temperature [°C] vs Time [s]

CAFE V_II

Measured

Solid fraction [-]

SnPb in rectangular cavity

Direct macro
SnPb in rectangular cavity

Direct macro

Improvement of nucleation kinetics required. Fragmentation?
Main scientific problems

- Nucleation laws not well characterized
- Fragmentation of mushy zone known as a source of crystals, but not understood
- Upscaling: modeling of a unique single grain with a direct microscopic method (e.g., phase field) for improving macroscopic models (e.g., CAFE)
- Coupling with deformation of the mushy zone during the formation of the solidification structure required (hot tear, shrinkage porosity, macrosegregation)
- Links with subsequent thermomechanical processing steps while inheriting from the solidification structure (e.g., PFZ formed upon homogenization heat treatment)
- Measurement of missing properties (e.g., interfacial energy and its anisotropy, mobilities)

Direct simulation of structures

- Need for more dedicated experiments with well controlled boundary conditions, with in-situ and post-mortem characterizations, to be compared with more integrated modeling with multiple scale physics and thermodynamic properties.