MEASUREMENT OF DENDRITE GROWTH ON Al-Ni ALLOYS IN REDUCED GRAVITY

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ABSTRACT

It is well known that the growth kinetics in metallic melts controls microstructure evolution. If the melt is cooled below its equilibrium melting temperature prior to solidification, the state of a metastable undercooled melt is created. An undercooled melt possesses an enhanced free energy that enables the liquid to choose solidification pathways into various metastable solids of properties being different to their stable counterparts [1]. A very efficient method to undercool a metallic liquid is the application of containerless processing a liquid drop such that heterogeneous crystal nucleation on container walls is completely avoided [2]. Electromagnetic levitation is a power-full technique to produce a freely suspended drop without any contact to a solid or liquid medium with the extra benefit that it is accessible for direct observation of solidification far from equilibrium by proper diagnostic means. Under terrestrial conditions, strong electromagnetic fields are needed to compensate the gravitational force. That, in turn, causes forced convection inside the liquid drop and influences mass and heat transport, and consequently, crystal growth in undercooled melts.

If the reduced gravity environment is utilized the forces to compensate residual accelerations are several orders of magnitude smaller than the levitation force on Earth. In the present paper we report on results obtained in the Earth laboratory, during parabolic flight missions and during TEXUS 44 flight in 2008 using the TEMPUS facility for containerless processing of metals in space. The results are discussed within dendrite growth theory and give evidence for strong effects of gravitational driven effects in the solidification dynamics.

1. EXPERIMENTAL

Electromagnetic levitation in combination with a high speed video camera (Photron VKT) was applied with a frame rate of 50 000 pictures per second to measure the rapid dendrite growth velocity as a function of undercooling for Al-Ni alloys in terrestrial experiments and parabolic flight campaigns as well. Details of electromagnetic levitation technique for containerless undercooling and solidification experiments are given elsewhere [3]. Measurements of rapid dendrite growth by high speed camera technique are described in Ref. [4].

The TEMPUS facility was developed by the GERMAN AEROSPACE CENTER – AGENCY (DLR), constructed by ASTRIUM and successfully tested during three NASA spacelab missions under real spaceconditions [5]. Presently, an advanced multiuser facility for Electro-Magnetic Levitation (EML) is developed in a common effort by DLR and ESA for accommodation on board the ISS.

Undercooling creates a driving force for crystal growth in melts. Growth is very much governed by heat and mass transport. The TEMPUS facility was used during TEXUS 44 mission to measure the dendrite growth velocity of $Al_{68.5}Ni_{31.5}$ alloy. 180 seconds of µg-time was available for the present experiment. This time was efficiently used succeeding in three undercooling and solidification cycles. During the first cycle metal-oxides were still present on the surface of the sample, which limited the undercooling to a few degrees, $\Delta T_0 \approx 5$K. Subsequently, the sample was heated by more than 200 K above the liquidus temperature of this alloy, T_L= 1768K. As a consequence, the metal oxides disappeared and the sample undercooled during the subsequent solidification cycles to ΔT_1=185K and ΔT_2=228K.

2. SHARP INTERFACE THEORY OF DENDRITE GROWTH

The sharp interface theory basing upon the work of Lipton, Trivedi, Kurz [6] is extended by taking into account fluid flow motion [7]. The total undercooling ΔT as measured in the experiment is the sum of several contributions:

$$\Delta T = \Delta T_t + \Delta T_c + \Delta T_r + \Delta T_k \qquad (1)$$

with ΔT_t the thermal undercooling, ΔT_c the constitutional undercooling, ΔT_r the curvature undercooling and ΔT_k the kinetic undercooling. The thermal undercooling $\Delta T_t = T_i - T_\infty$ (T_i: interface and T_∞ the temperature of the undercooled melt) is obtained from the solution of the thermal transport equation yielding the temperature at the tip of a dendrite as:

$$T_i = T_\infty + \Delta T_{hyp} Pe_t \cdot \exp(Pe_t + Pe_{tf}) \cdot \int_1^\infty q^{-1} \exp\left[-qPe_t + (\ln q - q) Pe_{tf}\right] dq \qquad (2)$$

$\Delta T_{hyp}=\Delta H_f/C_p$ (ΔH_f: heat of fusion, C_p: specific heat of the melt) is the hypercooling, $Pe_t=(V\cdot R)/2a$ the thermal Peclet number with R the radius of curvature at the tip of a dendrite and a the thermal diffusivity. The effect of fluid flow on dendrite growth is taken into account by introducing a thermal Peclet number for fluid flow, $Pe_{ff}=(U_o\cdot R)/2a$ with U_o the fluid flow velocity in the melt. Since U_o cannot be measured in electromagnetically levitated sample, we estimate its value by an energy balance for the energetics of the magnetic field inside the levitation coil, the gravitational field and the viscous dissipation as:

$$U_o = \left[\frac{2}{\rho}\left(\rho g R_o + \frac{B_o^2(1-exp(-2R_o/\delta}{8\pi} + \frac{\rho\eta^2}{2\delta^2}\right)\right]^{1/2} \qquad (3)$$

with g the gravity acceleration, ρ the mass density, η the viscosity of the melt, δ the skin depth, R_o the radius of the sample, and B_o the average of the magnetic field. (C_i-C_o) with C_i the concentration at the interface and C_o the nominal composition.

The constitutional undercooling $\Delta T_c=m_e(C_i-C_o)$ with m_e the slope of the liquidus line. ΔT_c is obtained from the solution of the mass transport equation yielding the concentration at the tip of a dendrite, C_i, as

$$C_i = C_o + (1-k_e) C_o Pe_c \cdot \exp(Pe_c + Pe_{fc}) \cdot \int_1^\infty q^{-1} \cdot \exp\left[-qPe_c + (\ln q - q) Pe_{fc}\right] dq \qquad (4)$$

with $Pe_c=(VR)/2D$ the chemical Peclet number. The effect of fluid flow on the mass redistribution in front of the solid-liquid interface is taken into account by introducing the chemical Peclet number of fluid flow $Pe_{cf}=(U_oR)/2D$.

The curvature undercooling $\Delta T_r=T_l-T_i$ is given by the Gibbs-Thomson equation as

$$\Delta T_r = 2\Gamma\left(1 - 15\cdot\varepsilon_c \cos 4\theta\right)/R \qquad (5)$$

where $\Gamma=\sigma/\Delta S_f$ (σ: the interfacial energy, ΔS_f: the entropy of fusion) is the capillary constant (Gibbs-Thomson coefficient), R is the radius of curvature at the tip of a dendrite, ε_c is the parameter of anisotropy of the interface energy, and θ is the angle between the normal to the interface and the direction of growth along the growth-axis. The kinetic undercooling, is inferred from rate theory

$$\Delta T_k = V/\mu_K, \quad \mu_K = \mu_{Ko}\left(1 - \varepsilon_K \cos 4\theta\right) \qquad (6)$$

where μ_K is the kinetic coefficient for growth of the dendrite tip, ε_K is the parameter of anisotropy for the growth kinetics.

Eq. 1 gives a relation between undercooling ΔT and the product of $V\cdot R$ in terms of the Peclet numbers. For a solution of Eq. (1) one needs a second equation for the radius of curvature of the dendrite tip. From solvability theory one gets an expression for the tip radius of a pure thermal dendrite without constitutional effects

$$R = \frac{\Gamma}{\sigma^* \Delta T_{hyp} Pe_t};$$

with σ^* the stability parameter. (7)

$$\sigma^* = \sigma_o \cdot \varepsilon_c^{7/4}\left[1 + \chi(\mathrm{Re})\frac{U_o\Gamma}{a\Delta T_{hyp}}\right]$$

where σ_o is a constant. $Re=U_oR/\eta$ is the Reynolds number. The function $\chi(Re)$ is taken from Ref. [8]. For calculations of the stability parameter σ^* we choose the results of phase-field modeling [9] with $\sigma_o\varepsilon_c^{7/4}/\sigma^* = 1.675$ for the 3D upstream fluid flow imposed on the scale of a freely growing dendrite. Thus, for a congruently melting alloy as $Al_{50}Ni_{50}$ with constitutional undercooling, $\Delta T_c=0$, the velocity V and the tip radius R of the dendrite can be calculated from Eqs (1), (2) and (7) as a function of the initial undercooling ΔT.

For non-congruently melting alloys with $\Delta T_c \neq 0$, the stability analysis has to take into account constitutional effects leading to

$$\frac{2d_o a}{VR^2} = \sigma_o \cdot \varepsilon_c^{7/4}\left[\frac{1}{2} + \frac{a}{D}\cdot\frac{\Delta_c k_e}{\left[1-(1-k_e)Iv(Pe_c)\right]\cdot\Delta T_{hyp}}\right] \qquad (8)$$

with d_o the capillary constant, and $\Delta_e = m_eC_o(k_e-1)/k_e$ is the equilibrium interval of solidification with k_e the equilibrium partition coefficient. σ_o is a constant and ε_c is the anisotropy parameter.

3. RESULTS AND DISCUSSION

3.1 Experiments in the Earth laboratory and during parabolic flight campaigns

The dendrite growth velocity was measured as a function of undercooling for two Al-Ni alloysone of equiatomic composition $Al_{50}Ni_{50}$ and anotherone of $Ni_{60}Al_{40}$, respectively. Fig. 1 shows the phase diagram of Al-Ni. The compositions of the alloys investigated are marked by solid vertical lines.

The equiatomic alloy $Al_{50}Ni_{50}$ is a high melting intermetallic phase of body-centered cubic (bcc) structure with a type B2 superlattice, which melts congruently. The $Ni_{60}Al_{40}$ alloy is also primarily crystallizing in B2 superlattice structure of β-phase, however, melts incongruently. This means, constitutional effects and chemical segregation effects have to be taken into account, which are missing in case of the equiatomic alloy. The Raney-type alloy of $Al_{68.5}Ni_{31.5}$ forms by a peritectic reaction from the liquid. Peritectic reactions involve liquid (L) and a primarily formed solid α to form a solid phase γ according to L + α → γ. It requires diffusion in the solid of α-phase. Therefore, growth of γ-phase is expected to be very sluggish.

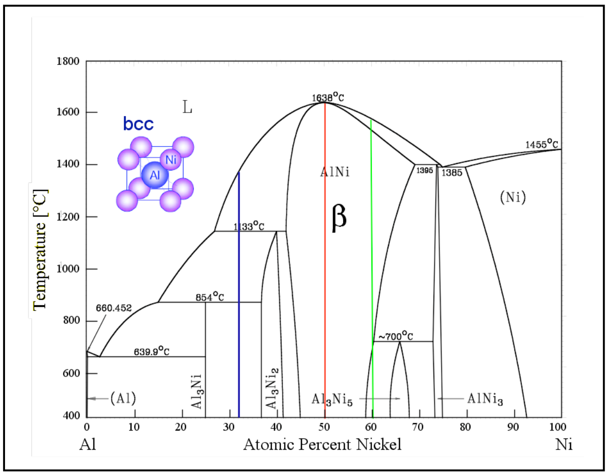

Figure 1: Phase diagram of Al-Ni. The compositions of the alloys investigated in the present work are marked by vertical lines.

Fig. 2 gives the results of measurements of the growth velocity V as a function of undercooling ΔT for the $Al_{50}Ni_{50}$ alloy [10]. The experimental results of levitation experiments using the TEMPUS facility during parabolic flight (squares and triangles) are well described by sharp interface theory without taking into account fluid flow by forced convection (dashed line). Also the data obtained in 1g (full circles) are in agreement with theory without convection in the velocity range $V > U_o$ (U_o: fluid flow velocity). This finding confirms that fluid flow motion is of influence to dendrite growth dynamics if the growth velocity is smaller or comparable with the fluid flow velocity. At smaller velocities $V < U_o$ systematic deviations occur between terrestrial data (full circles) and predictions of sharp interface theory if convection is neglected. These deviations are attributed to an enhancement of the growth velocity due to forced convection in electromagnetically levitated samples. The terrestrial data are well described by extended sharp interface theory assuming a maximum fluid flow velocity of $U_o \approx 1.2$ m/s (solid line). Even though a bit larger this value is comparable to the estimations of magneto-hydrodynamic simulations [11].

Fig. 3 exhibits results of equivalent measurements of dendrite growth velocity on the non-congruently melting alloy $Ni_{60}Al_{40}$. At small undercoolings the same behaviour of $V(\Delta T)$ is found as for $Al_{50}Ni_{50}$ alloy.

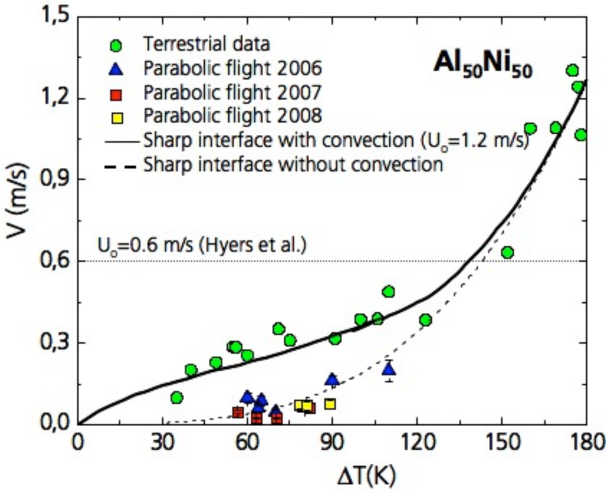

Figure 2: Dendrite growth velocity V as a function of undercooling ΔT for $Al_{50}Ni_{50}$ alloy. The experimental data are represented by the symbols and the lines give the predictions of sharp interface theory. The fluid flow velocity U_o in electromagnetic levitation is inferred from magneto-hydrodynamic simulations [11].

Also for the $Ni_{60}Al_{40}$ alloy sharp interface theory is successfully applied to describe the results obtained in reduced gravity (dashed line). The parabolic flight mission in 2006 offered to record video pictures during recalescence only with a frequency of 200 Hz. Therefore the data points (triangles) are associated with a large scatter while the data of the experiments during the parabolic flight mission in 2007 were taken by the high speed camera at a frequency of 50000 Hz. Consequently the squares show a much smaller uncertainty.

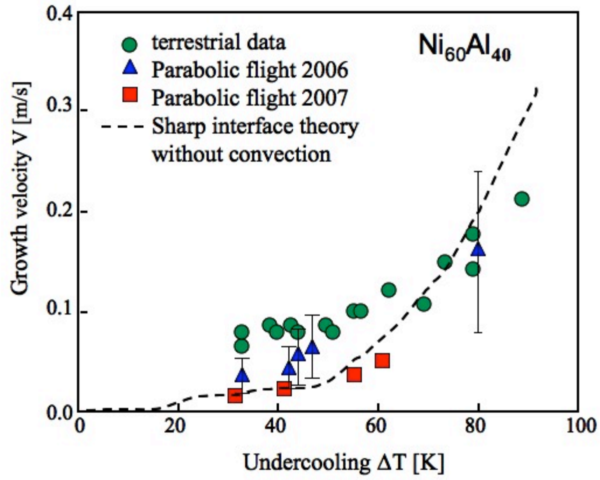

Figure 3: Dendrite growth velocity V as a function of undercooling ΔT for $Al_{40}Ni_{60}$ alloy. The closed circles give results of terrestrial experiments while he squares and triangles reproduce data from two parabolic flight missions using the TEMPUS facility on board.

The $Ni_{i60}Al_{40}$ alloy could be undercooled in the parabolic mission 2006 to about 156 K. One data point for the velocity was taken at this undercooling (not shown). The velocity measured is about by a factor of three smaller than the corresponding result measured in the Earth laboratory. But because of the low measuring frequency the error of this data point is relatively large. Nevertheless, it is still outside the confidence interval. In order to support this finding additional experiments on this alloy in reduced gravity are needed.

3.2 Experiments on TEXUS 44 flight

A Raney type $Al_{68.5}Ni_{31.5}$ alloy was investigated during the second phase of an experiment using the TEMPUS facility during TEXUS 44 flight. Fig. 4 shows the temperature-time-profile of three undercooling and solidification cycles.

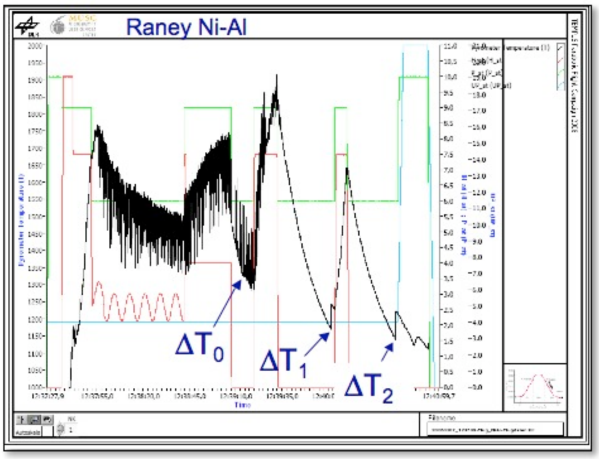

Figure 4: Temperature-time-profile measured during TEXUS 44 flight for three undercooling and solidification cycles of Raney type $Al_{68.5}Ni_{31.5}$ alloy.

During the first cycle a very noisy signal was recorded by the pyrometer. This was caused by the fact that solid metal-oxides were swimming on the liquid sample. Such metal oxides are very stable and possess a much higher surface emissivity than a pure metal. The rotating sample was moving through the observation window of the pyrometer with alternatively changing surface quality with views of metal oxide and pure metal. Since metal oxides act as heterogeneous nucleation of high catalytic potency the undercooling of the first solidification cycle was limited to $\Delta T = 5K$ only. In the subsequent heating period the sample was heated up to about 250 K above its melting temperature. This overheating procedure led to a rapid removal of the metal oxides from the sample surface. As a direct consequence the noise of the temperature signal disappeared and the liquid was undercooled to a large undercooling of $\Delta T = 186$ K. Eventually, a third cycle could be realized within the available experiment time and a further increase of the undercooling to $\Delta T = 214$ K was achieved. The TEMPUS TEXUS module was equipped with a video camera of small measuring frequency of 200 Hz, the determination of the growth velocity during the recalescence profile is associated with a large uncertainty of about 15% to 25%. Fig. 5 shows the growth velocity V as a function of undercooling of Raney type Al-Ni alloy as measured on ground (closed circles) and the data points as retrieved from the TEXUS experiment (triangles).

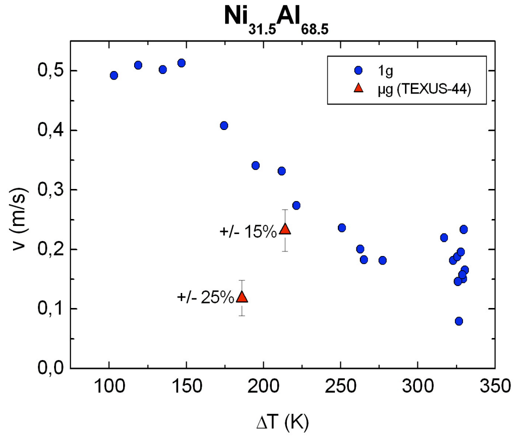

Figure 5: Dendrite growth velocity V s a function of undercooling, measured for Raney type Al-Ni alloy on ground (closed circles) and during TEXUS 44 flight mission (triangles).

Despite the relatively large uncertainty of the data points obtained during the TEXUS mission two significant and important differences of the experiments under normal gravity and in microgravity are observed. First, the growth velocity measured in microgravity is essentially smaller than the data taken under 1g condition. Second, the temperature dependence of the $V(\Delta T)$ relation differs very much. In reduced gravity, it is apparent that the growth velocity increases with increasing undercooling. Such behaviour is observed so far for all investigated metals, alloys and even semiconductors despite the fact that the materials differ very much in their physical properties [12]. An increase of the growth velocity with increasing undercooling is easily understood since the undercooling scales with the driving force for crystallization and, hence, should lead to an increase of the velocity of growing dendrites.

In so far, the decrease of the velocity with l undercooling as measured on ground on a variety of Al-rich Al-Ni alloys is very anomalous and unique. By taking into account the results of the TEXUS 44 flight this anomalous behaviour of dendrite growth dynamics of Al-rich Al-Ni alloys is obviously associated with gravity dependent mechanism in crystal growth in undercooled melts. Investigations of the physical origin on this anomalous behaviour of $V(\Delta T)$ is currently in progress within a project that was recently approved by the German Research Foundation (DFG) [13].

4. SUMMARY AND CONCLUSIONS

We have presented results of measurements of dendrite growth velocity as a function of undercooling for three differently concentrated Al-Ni alloys as the congruently melting intermetallic phase of B2 structure of equiatomic composition $Ni_{50}Al_{50}$, the non-congruently melting $Ni_{60}Al_{40}$ alloy and the Raney type Al-rich $Al_{68.5}Ni_{31.5}$ alloy. All alloys were undercooled essentially by electromagnetic levitation technique in the Earth laboratory and the dendrite growth velocity was measured with high accuracy by applying a high speed video camera with maximum picture frequency of 50000 pictures per second. In addition, all alloys were investigated by equivalent levitation experiments in reduced gravity making use of various parabolic flight missions and the TEXUS 44 mission in 2008. At growth velocities being smaller or comparable as the fluid flow velocity in the liquid a pronounced effect was found confirming that forced fluid flow leads to an enhancement of the heat and mass exchange at the solidification front with the consequence that also the growth velocity is enlarged by forced convection. This finding is very important since the growth dynamics controls the microstructure evolution. The microstructure itself is decisive for the physical and chemical properties of the as solidified material. Thus, fluid flow motion may be an effective process parameter in future casting processes of foundry industry to improve production chains of metallic materials.

An anomalous $V(\Delta T)$ relation is observed for Al-rich Al-Ni alloys such that the dendrite growth velocity is decreased with increasing undercooling i.e. with increasing driving force for crystallization. The results of the TEXUS 44 – TEMPUS mission, however, suggest a normalous growth behaviour of Raney type $Al_{68.5}$ $Ni_{31.5}$ alloy. The comparison of the findings of equivalent terrestrial and microgravity experiments suggests that the anomalous growth behaviour of Al-rich Al-Ni alloys may be caused by gravity related processes during non-equilibrium solidification of undercooled melts. Investigations to clarify this surprising growth behaviour are presently in progress. They certainly need further complementary experiments in the reduced gravity environment in the future.

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