Direct observation of hot tear formation in organic alloys

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April 3rd, 2001

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Abstract

Following the work of Farup et al. [1], hot tear formation has been observed during the solidification of a succinonitrile–acetone alloy by pulling the columnar dendrites in the transverse direction with a pulling stick. In order to get a better resolution and to observe the coalescence of dendrites deep in the mushy zone, a dye has been added to the alloy. Pulling of the dendrite arms has also been improved by connecting the pulling stick to a small electrical motor, thus allowing for a better control of the deformation rate. The opening of the mushy zone (hot tears) always occurs at grain boundaries. At low volume fraction of solid, the opening can be compensated for by leaner-solute interdendritic liquid (i.e., "healed" hot tears). At higher volume fraction of solid, hot tears directly nucleate in the interdendritic liquid or develop from pre-existing micropores induced by solidification shrinkage. The measurement of the temperature and solid fraction at which coalescence between two grains occurs, allows with the help of the Rappaz-Drezet-Gremaud (RDG) hot tearing criterion [2] to evaluate the cavitation depression of the succinonitrile–acetone alloy.

I. Introduction

Hot tearing, as well as porosity, is a major defect of cast parts, which is issued from a lack of feeding of the mushy zone. However, unlike porosity, which is induced by solidification shrinkage, hot tearing results from tensile stresses, which tend to open the mushy zone [2]. This problem starts to be particularly critical at the end of solidification and more precisely, as highlighted by Campbell, *"when grains grow to the point at which they collide with each other, but are still largely surrounded by residual liquid"*[3]. As can be imagined easily, those *"stresses are issued by differential thermal contraction upon cooling"* as specified by Rappaz et al. [2].

Nevertheless, most hot tearing criteria neglect the importance of thermomechanical aspects and simply consider the solidification interval of the alloy [3]: the larger the freezing range, the more susceptible the alloy will be to hot tearing. Clyne and Davies [4] defined a criterion in which the time interval spent by the mushy zone in the vulnerable region appears. As an alternative, Feurer [5] focused on the liquid present between the grains and argued that a hot tear will nucleate as a pore if the liquid is no longer able to fill the intergranular openings. Unfortunately, he only considered the contribution of solidification shrinkage. Rappaz et al [2] extended Feurer's approach in order to also take into account the feeding associated with tensile deformation of the solidified material in the direction transverse to the dendritic growth. Recently, Farup and Mo [6] formulated a two-phase model of a deforming, solidifying mushy zone where both interdendritic liquid flow and thermally induced deformation of the solid phase were taken into account.

Although the investigations on as-cracked surfaces flourish, *in situ* observations of hot-tear formation are rare because of the technical problems involved with metallic alloys. Due to these problems, the transparent organic model alloy of succinonitrile (SCN) with acetone was selected by Farup et al. [1] to induce hot tearing by mechanical pulling of the mushy zone during directional solidification. The same device has been used in the present investigation with two improvements : i) the use of a second solute element, a dye, which helps in distinguishing the liquid, the solid and the voids ; ii) the control of the pulling speed by an electrical motor. The main purpose is to visualise *in situ* hot tear formation during solidification, and in particular to study the nucleation of hot tears and the coalescence of grains.

Section II of the present work is dedicated to the description of the experimental set-up and to the observation of hot tears as well as grain coalescence. In section III, the RDG hot tearing criterion is briefly summarised and used for determination of the cavitation depression in SCN-aceton, with the help of the measured temperature at which coalescence of grains occurs.

II. Observation of hot tears in organic alloys

a) Set-up description

The set-up for inducing hot tearing in SCN-acetone is essentially the same as that used by Farup et al. [1]. As depicted in Figure 1, a cell containing the succinonitrile-acetone alloy is submitted to a constant temperature gradient by moving it away from a heat source with an electrical motor at a constant velocity (typically 10 µm/s). In Farup's experiments, a "puller" was used to manually pull apart the growing dendrites in the transverse direction, whereas now this was achieved by a supplementary electrical motor (better control of the applied deformation). Fuchsin was added to the transparent alloy in small quantity (typically 0.05wt%) so as to enhance the contrast between the different phases of the system (liquid, solid, air). Temperature was measured by placing a thermocouple in the cell. The temperature measurement was coupled with video recording of the growing dendrites in order to deduce the temperature at which dendrites coalesce or bridge. For dendrites belonging to the same grain (i.e., no grain boundary energy), this temperature, T_{cd}, is higher than that characterising bridging of dendrite arms across a grain boundary (T_{cg}).
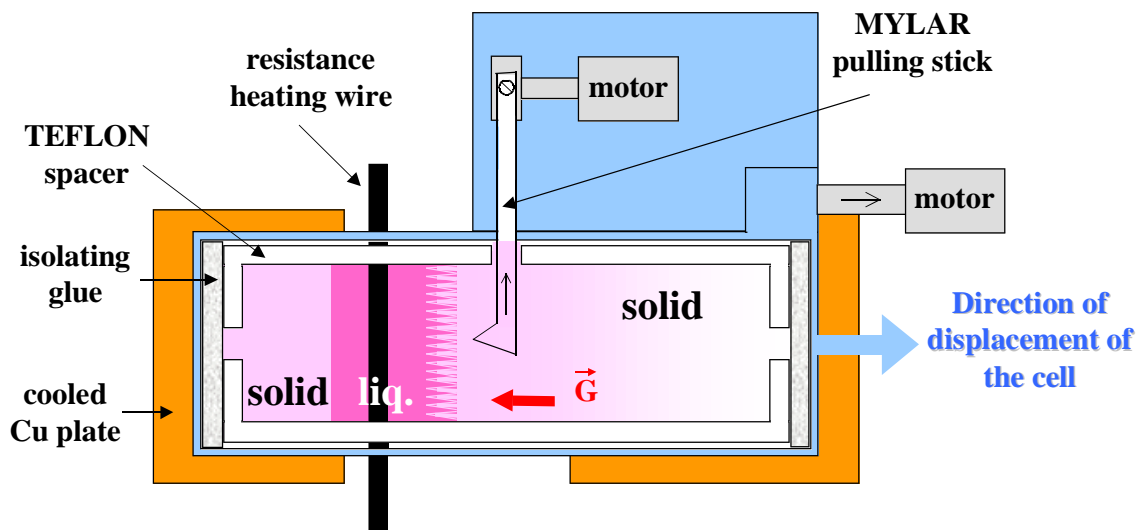

Figure 1.   Schematics of the experimental device used to observe in-situ the formation of hot tears in succinonitrile-acetone alloys.

b) Observation of hot tears

The observation of hot tears is performed on time by a colour video camera mounted on an optical microscope positioned on top of the set-up. As already reported in [1], it has been confirmed that hot tears always appear between two grains; the grain boundary being the last part of the system to solidify. Moreover, two situations can be distinguished from these observations :

1) When the "stick" is pulled at too high liquid fractions, liquid can feed the opening because of the high permeability of the mush. Therefore, the "crack" is healed by solute-enriched liquid and no defect results from this event as shown in Figure 2, except maybe some enhanced segregation.
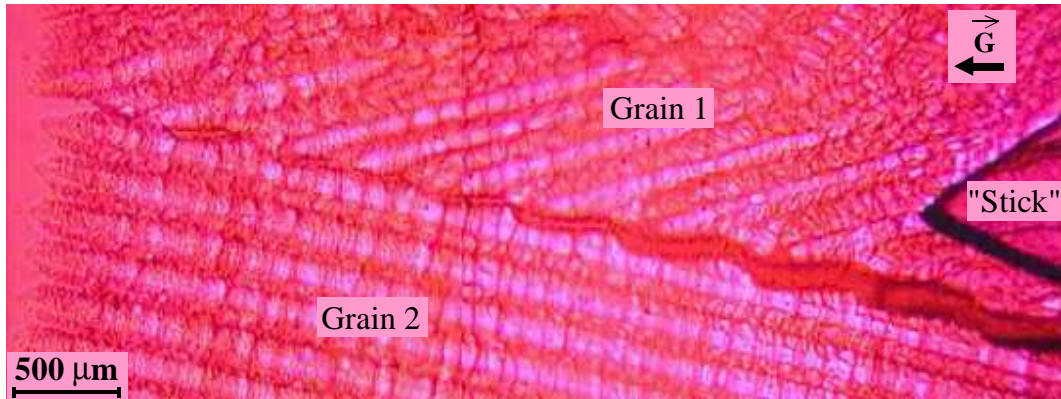

Figure 2.   Picture of a healed hot crack.

2) When the "stick" is pulled at a sufficiently high solid fraction (i.e., low permeability), the liquid cannot feed the opening from the tip of the dendrites and an intergranular crack appears, as shown in Figure 3. Moreover, it can be seen from the deformation of the dendrites during pulling that they are particularly ductile at this temperature. Please note that two cracks are growing in parallel in this figure. The bubble with the larger tip radius (top) is slightly ahead of the sharper one (bottom) as a result of the associated curvature depression (Laplace term).
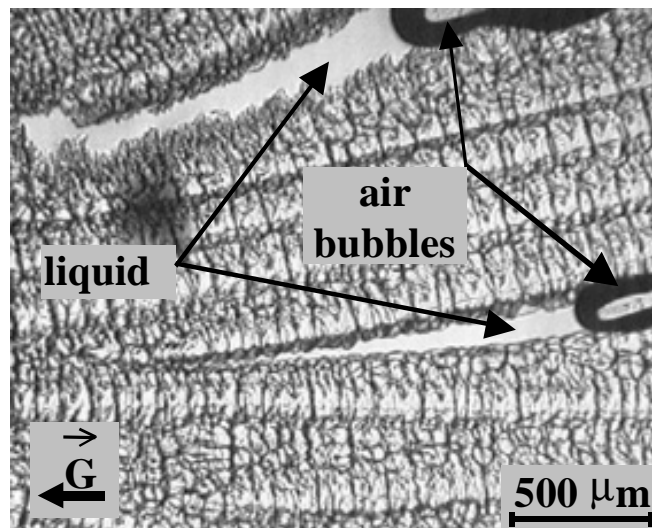

*Figure 3.   Picture of two hot cracks growing in intergranular liquid regions.*

c) Temperature measurements

A type-K thermocouple, 50 μm in diameter, has been inserted in the cell. Temperature was recorded every second. Figure 4a shows a typical cooling curve. The cooling rate is constant during the experiment (–0.03 K/s), until heating is switched off at lower temperature to speed up solidification (slope change at 1100s).

By coupling temperature measurements with video observations of the solidifying transparent alloy, the liquidus temperature, T_{liq}, has been measured (see Figure 4b). In the same manner, the coalescence temperature of dendrites across a grain boundary, T_{cg}, and within a grain, T_{cd}, have been evaluated as shown in Figures 5a and 5b, respectively. Coalescence or bridging of secondary dendrite arms is established when the liquid film between two adjacent dendrites or two grains is no more continuous. This "temperature" is highly dependent on the experimental conditions and on the disorientation of the grains. For this reason, the values of T_{cd} and T_{cg} given in Figure 4a, are mean values estimated from several measurements.
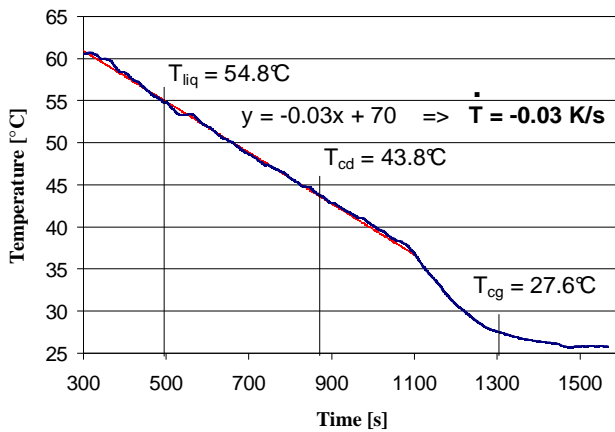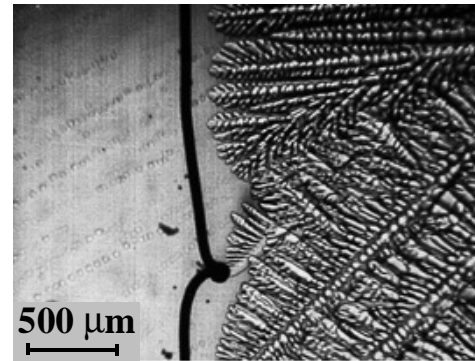
a)	b)

Figure 4.   a) Typical temperature history during the growth of columnar dendrites in the SCN-acetone system, b) micrograph of the dendrites when the tips reach the thermocouple (determination of T_{liq}).

As solidification proceeds under Bridgman conditions, the cooling rate is the product of the thermal gradient, G, and the speed of the isotherms, V_T, which is equal to the withdrawal speed. This allows to determine the value of the thermal gradient, 3000 K/m.

Although the strain field is highly non-uniform and is localised at grain boundaries, an average strain rate imposed to the alloy by the displacement of the stick was evaluated roughly as the ratio of the speed of the transverse motor to the width of the cell: about 4.10^{-5} s^{-1} for a pulling speed of 10 μm/s.
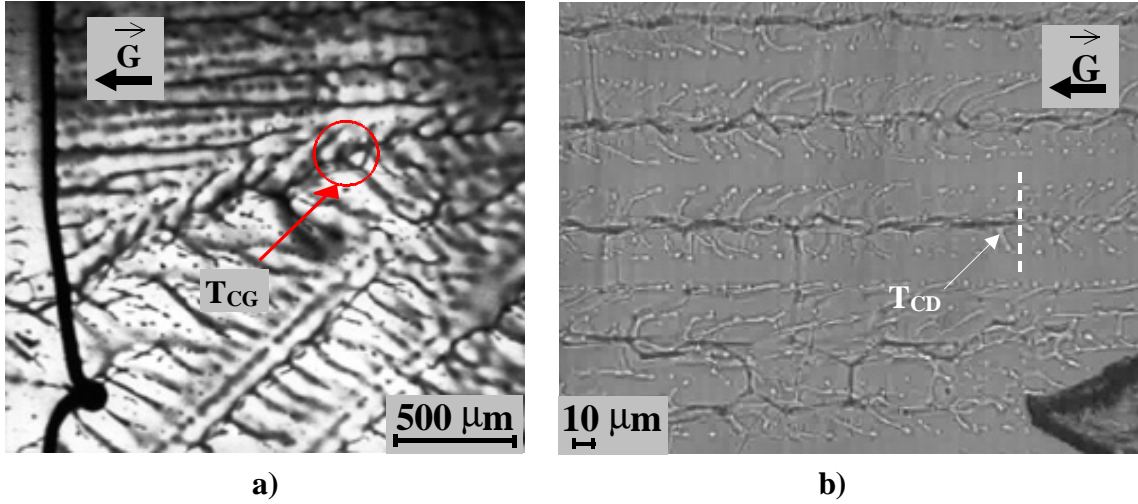
Figure 5. *Pictures used to determine the temperature of coalescence a) of dendrites across a grain boundary (T_{cg}) and b) of dendrites belonging to the same grain (T_{cd}).*

III. Application of RDG criterium to the SCN-acetone alloy

Before applying the hot tearing criterium recently derived by Rappaz, Drezet and Gremaud [2] to the organic alloy investigated here, it is briefly summarised. The determination of the coalescence temperature in inter- and intragranular liquid films is further detailed.

The experimental situation encountered with the organic alloy (previous section) is precisely that considered in the RDG approach. It is summarised in Figure 6. In this schematic diagram, the position, x_{liq}, of the dendrite tips is indicated together with the positions in the mushy zone where dendrites coalesce or bridge. For dendrites belonging to a single grain, this occurs at higher temperature (position x_{cd}) since there is in principle no grain boundary energy barrier. At the grain boundary, the dendrites coalesce at a position, x_{cg}, which must be a function of the disorientation. Below this position, the whole mush is coherent and thermal strains, ε, are transmitted to the partially coalesced dendrites. Although strains are localised, the RDG criterion considers a uniform mushy zone in the transverse direction. Due to solidification shrinkage and thermal strains, interdendritic liquid has to flow from the right and thus the pressure field in this medium might look like the profile shown at the bottom of Figure 6. The minimum pressure, p_{min}, is given by :

$$p_{min} = p_m - \Delta p_{sh} - \Delta p_c = p \qquad (1a)$$

or

$$\Delta p_{max} = \Delta p_{sh} + \Delta p_c = \Delta p = p - p \qquad (1b)$$

Δp_ε and Δp_{sh} are the pressure drop contributions associated with deformation and shrinkage, respectively (taken as positive values), p_m is the metallostatic pressure. If p_{min} is equal to a cavitation pressure, p_c, then a pore (a crack) will form at the roots of the dendrites.

In order to calculate these two contributions, a mass balance is performed at the scale of a small volume element (see Ref. [2] for details) in a reference frame attached to the isotherms and under steady state conditions. Assuming no porosity formation, the volume fraction of liquid, f_l, is equal to $(1- f_s)$ and the specific masses of the two phases, ρ_s and ρ_l, are assumed to be constant, but not equal; the solidification shrinkage factor β is given by (ρ_s/ρ_l-1). The velocity of the liquid is related to the pressure gradient in the liquid via the Darcy equation and the permeability of the mushy zone is given by the Carman-Kozeny approximation [2].
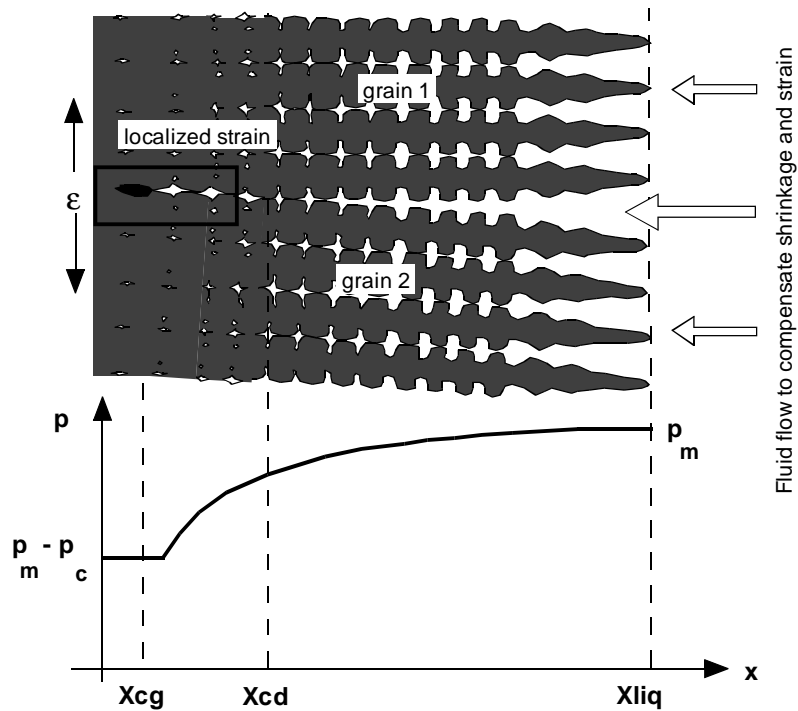

Figure 6.   *Schematic of the formation of a hot tear between two columnar grains as a result of a localised strain transmitted by the coherent dendrites below. The pressure drop in the intergranular liquid film is also indicated. x_{cg} , x_{cd} and x_{liq} stand for the positions of the isotherms of grain coalescence, dendritic coalescence and liquidus, respectively.*

Considering that the fluid moves along the thermal gradient only, i.e., along the x axis, whereas the solid deforms in the transverse direction under an uniform strain rate, the two contributions, Δp_{sh} and Δp_ε, are given by [2] :

$$\Delta p_{sh} + \Delta p_{mec} = \frac{180\mu}{G\lambda_2^2}\left[v_T\beta A + \frac{(1+\beta)B\dot{\varepsilon}}{G}\right] \quad \text{with}$$

$$A = \int_{T_c}^{T_{liq}} \frac{f_s^2 dT}{(1-f_s)^2} \quad \text{and} \quad B = \int_{T_c}^{T_{liq}} \frac{f_s^2 \int_{T_c}^{T} f_s \cdot dT}{(1-f_s)^3} dT \qquad (2)$$

T_c is the temperature at which coalescence (or bridging) of the dendrite arms occurs, that is T_{cg} for intergranular liquid films (see Figure 6) and T_{cd} for intragranular liquid films. μ is the viscosity of the liquid.

Equation 2 reveals that the shrinkage contribution is proportional to the shrinkage factor β and to the speed of the isotherms, v_T, whereas the mechanical contribution is proportional to the strain rate, epoint. Both contributions are inversely proportional to the square of the secondary dendrite arm spacing, λ_2. The hot tearing approach presented here is now applied to the organic alloy used in the experimental work in order to determine its cavitation depression.

Apart from the coalescence temperature, T_c, the two parameters A and B depend only on the solidification path of the alloy, i.e., on the relationship between the solid fraction, f_s, and the temperature, T. They can be calculated using a back-diffusion model such as that of Clyne and Kurz [7,8].

Although Fuchsin was added to the organic alloy, the binary system SCN-acetone was considered to calculate its solidification path [7,8], and thus :

$$f_s(T) = \frac{1}{1 - 2\alpha_s^* k}\left[1 - \left[\frac{T_m - T}{T_m - T_L}\right]^{\frac{1 - 2\alpha_s^* k}{k - 1}}\right] \qquad (3)$$

where k is the partition coefficient, T_m the melting point of pure SCN and is related to the Fourier number, α_s, via the relationship :

$$\alpha_s^* = \alpha_s[1 - \exp(-\alpha_s^{-1})] - 0.5\exp(-0.5\alpha_s^{-1}) \quad \text{with} \quad \alpha_s = D_s t_f/\lambda_2^2 \qquad (4)$$

D_s is the diffusion coefficient in the solid and t_f is the solidification time. The material properties of the binary system SCN-Acetone are given in Table 1. The diffusion coefficient in the solid was taken equal to that in the liquid divided by 10^3. The secondary dendrite arm spacing was evaluated from Figure 5a (around 80 µm). The solidification time is about 800 seconds (see Figure 4a). These values yield $\alpha_s = 0.1625$ and $= 0.1391$. Taking an initial composition of 0.5 wt. pct of acetone, the solid fraction versus temperature curve $f_s(T)$ can be calculated: it is shown in Figure 7. The predicted liquidus temperature is 56.68°C, whereas the measured tip temperature is 54.8°C; this difference can be explained by the undercooling of

the dendrite tips (2°C as calculated by Esaka [9] under such conditions) and maybe by the Fuchsin addition.

The values of T_{cg} and T_{cd} estimated in the first section are also reported in Figure 7 with an estimation of their error. From such measurements it is estimated that $f_s(T_{cg}) = 0.99$ whereas $f_s(T_{cd}) = 0.94$. Knowing the $f_s(T)$ relationship, the two integrals A and B appearing in Equation 2 can be computed up to different values of T_c (or corresponding solid fraction, f_c), as shown in Figure 8. The values of A and B highly depend on the lower limit of integration, T_c: indeed the two integrals tend to infinity when the solid fraction approaches 1. Reporting in Figure 8 the values of f_{cg} and f_{cd} previously determined, it clearly appears that A and B are much larger, at least one order of magnitude, at grain coalescence than at dendritic coalescence. In other words, the pressure drop contributions associated with deformation and shrinkage (see Equation 2) are much larger in the intergranular liquid films than in the intragranular films : this supports the fact that hot tears always appear at grain boundaries, even not considering strain localisation.

Table 1 : Relevant material parameters for the SCN–acetone system

Reference Melting point	T_m	58.08 ºC	[10]
Viscosity	μ	2.6 10^{-3} Pas	[11]
Slope of liquidus	m	-2.8 Kwt%$^{-1}$	[9]
Partition coefficient	k	0.1	[9]
Solidification shrinkage	β_s	0.047	[12]
Liquid diffusivity	D_l	1.3 10^{-9} m²/s	[9]
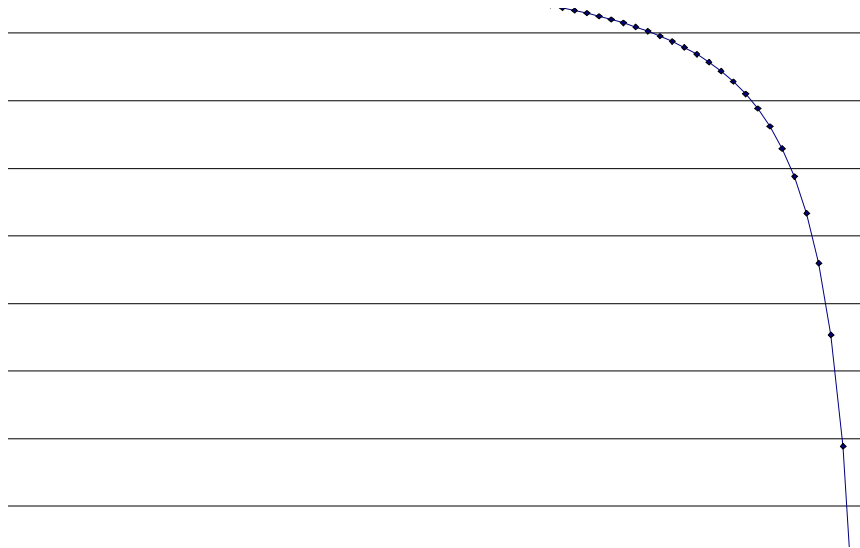
Figure 7.   Solid fraction versus temperature for SCN-0.5wt% acetone together with the indication of coalescence points of dendrite belonging to the same grain, T_{cd}, and localised at a grain boundary, T_{cg}.
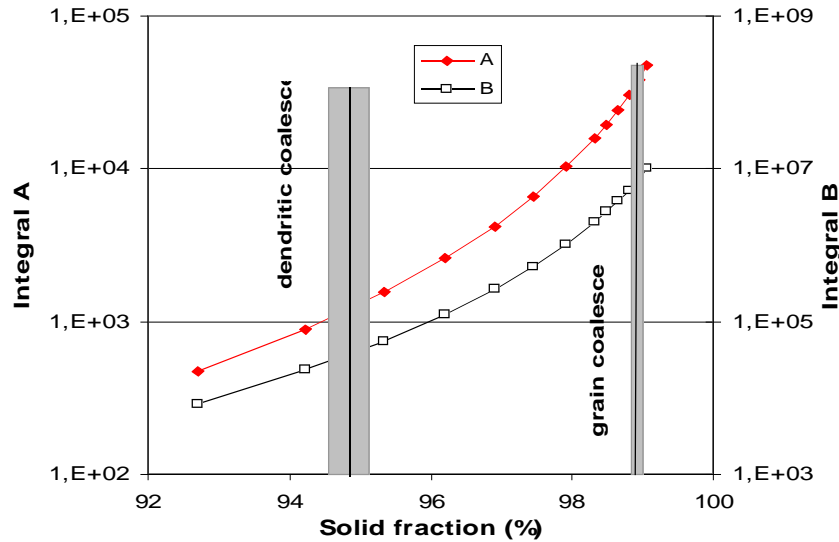
Figure 8.   Integrals A and B for SCN-0.5wt% acetone as a function of the solid fraction at coalescence, f_c.

Finally, using the computed values of A and B at grain coalescence, one can determine the cavitation depression of the organic alloy with the help of Equation 2 and using the average strain rate of 4.10^{-5} s^{-1}. This gives a value of 4 kPa for the cavitation depression. This is comparable with the value of 2 kPa found by Drezet and Rappaz [13,14] using thermomechanical computations of the ring mould tests of Al-4.5wt%Cu alloy. Nevertheless, the cavitation depression reaches 30 kPa if the strain rate increases to 4.10^{-4} s^{-1} owing to some localisation of deformation at grain boundaries.

IV. Conclusion

Hot tears have been observed during the solidification of a SCN–acetone alloy by pulling the columnar dendrites in the transverse direction with a pulling stick. At low volume fraction of solid, the opening can be compensated by interdendritic liquid (i.e., "healed" hot tears). At higher volume fraction of solid, hot tears directly nucleate in the interdendritic liquid at grain boundaries. Intragranular coalescence is found to occur at a solid fraction around 95% whereas intergranular coalescence takes place much deeper in the mushy zone at a higher solid fraction, 99%. The RDG hot tearing approach clearly shows that the intergranular liquid films are much more sensitive to hot cracking than the intragranular films. Further work should be carried out in order to determine more carefully when bridging of dendrites occur as a function of the grain boundary energy. Coalescence is a key parameter for the understanding of mechanical property of the mush at high volume fraction of solid and of strain localisation.

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