Hot tear formation and coalescence observations in organic alloys

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Abstract

Hot tear formation has been observed during the solidification of a succinonitrile–acetone (SCN-acetone) alloy by pulling the columnar dendrites in the transverse direction with a stick. It is observed that the cracking of the mushy zone (hot tears) always occurs at grain boundaries. At low volume fraction of solid, the opening can be compensated for by leanersolute 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 or air bubbles induced by solidification shrinkage. Moreover, coalescence/bridging of dendrite arms has been carefully observed and the temperature at which this occurs has been measured. This allowed us to determine the corresponding solid fraction. It is observed that coalescence between columnar dendrites inside a grain (intragranular coalescence) occurs at a higher temperature/lower solid fraction than coalescence of dendrites. Located across a grain boundary (intergranular coalescence), these results bring a new light on the formation of hot tears in metallic alloys.
I. Introduction

Hot tears are cracks which initiate during solidification, i.e. at non-zero solid fraction. They represent a major defect commonly encountered during the casting of large freezing range alloys and can lead to catastrophic cracking of the cast parts. In Figure 1a, the example of a cracked rolling sheet ingot of very large dimensions is shown. Here, two hot tears which were initiated during the start-up phase degenerated in two long cracks all along the ingot. In extrusion billets, hot tears are located in the centre of the cast part, as shown in Figure 1b. The consequences of this phenomenon lead to a considerable loss in productivity, which can even reach 10% for some very sensitive alloys. The study of hot tear formation has gained a new interest during the last few years, in particular with the derivation of predictive criteria [1,3,5,6] which can be implemented in FEM models of casting.

Figure 1. a) Hot tears in an aluminium slab that led to a complete cracking of the ingot. b) Typical hot tear in extrusion billets [2].

Hot tears originate from a lack of liquid feeding of the mushy zone [1], especially at the end of solidification and more precisely, as highlighted by Campbell, when grains start to impinge and finally touch one another, but are still surrounded by a continuous liquid film [3].

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 is the alloy to hot tearing. Clyne and Davies [4] defined a criterion in which the time interval spent by the mushy zone in a vulnerable stage 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. Rappaz et al. [1] extended Feurer's approach in order to take into account the feeding associated with both solidification shrinkage and tensile deformation of the solidifying material. 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, \textit{in situ} observations of hot tear formation are rare. Nevertheless, transparent organic alloys offer an interesting alternative to observe the formation of hot cracks. Succinonitrile with acetone (SCN-acetone alloy) was selected by Farup et al. [7] 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 of the present work is to visualise \textit{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. In section III, observations of hot tear formation are described. In section IV, observation of coalescence between dendrites and between grains is detailed.

II. Set-up description

The set-up used to induce hot tears in SCN-acetone is essentially the same as that used by Farup et al. [7]. As depicted in Figure 2, the succinonitrile-acetone alloy contained in a thin glass cell is solidified in a constant temperature gradient (Bridgman type experiment), by moving it away from a heat source with an electrical motor at a constant velocity (typically 10 $\mu$m/s). In Farup’s experiments, a “puller” was used to manually pull apart the growing dendrites in the transverse direction, whereas now this is achieved by a supplementary electrical motor (better control of the applied strain rate). Fuchsin was added to the transparent alloy in small quantity (typically 0.05 wt\%) so as to enhance the contrast between the different phases of the system (liquid, solid, air). Temperature was measured every second by placing a type-K thermocouple, 50 $\mu$m in diameter, in the cell. This measurement was coupled with video recording of the growing dendrites in order to deduce the temperature at which dendrites coalesce or bridge.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{Schematics of the experimental device used to observe \textit{in-situ} the formation of hot tears in succinonitrile-acetone alloys.}
\end{figure}
III. Observation of hot tear formation in SCN-acetone

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 [7], 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 3, except maybe some enhanced segregation.

![Figure 3. Picture of a healed hot crack.](image)

**Figure 3. Picture of a healed hot crack.**

**Clip 1** shows how, by using a stick to induce a tensile opening in the organic alloy, deformation concentrates at the grain boundary. The dotted lines indicate the position of grain boundaries. In this case, the opening takes place between grains 2 and 3 (i.e., closest to the stick). Moreover, the solid fraction at this point of solidification is still low enough to allow liquid metal to feed the opening owing to the relatively high permeability of the mush. This leads to the formation of a “healed” crack avoiding any formation of hot tears.

![Clip 1. “Healed” hot crack (Double-click on the picture to start the movie).](image)

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It can be observed in Clip 1 that the tensile deformation induced by the stick is compensated by some liquid flow. The direction of this flow is revealed by the movement of some equiaxed grains that have nucleated in the solute-enriched liquid and which are moving to the right. During the opening, some dendritic arms grow in the liquid spacing between the two grains, as shown in Figure 4.

![Figure 4](image)

**Figure 4.** Picture of an opening fed by some solute-enriched liquid.

2) When the "stick" is pulled at a sufficiently high solid fraction, i.e., deep in the mushy zone where the permeability is low, some liquid cannot feed the opening from the tip of the dendrites and an intergranular crack appears, as shown in Figure 5 and Clip 2. 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 air 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 5](image)

**Figure 5.** Picture of two hot cracks growing in intergranular liquid film regions.
The pressure in the intergranular liquid associated with both the solidification shrinkage and the tensile deformation increases owing to the inability of the liquid to feed the opening and reaches the cavitation pressure at which an air bubble appears. One can clearly see in Clip 2 air bubbles propagating into the opening thus creating a hot crack.

**Clip 2.** Hot tear formation by propagation of air bubbles in the opening (Double-click on the picture to start the movie).

**Clip 3** shows more precisely the flow of air bubbles in the opening created by the pulling of the “stick” in the SCN-acetone alloy. It explains clearly how the entrapped porosity contributes to the formation of a hot tear. Moreover, it highlights the unstable equilibrium due to the associated curvature depression between the opening of the crack, the tip radius of the bubble and the movement of the bubble.

**Clip 3.** Hot tear growing in intergranular liquid regions (Double-click on the picture to start the movie).
IV. Observation of coalescence in SCN-acetone

*Figure 6a* shows a typical cooling curve obtained with the thermocouple inserted in the cell. 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 changes at 1100 s).

By associating temperature measurements with video observations of the solidifying transparent alloy, the liquidus temperature, \( T_{\text{liq}} \), has been measured (see *Figure 6b*). In the same manner, the coalescence temperature of dendrites across a grain boundary (intergranular coalescence temperature), \( T_{\text{cg}} \), and within a grain (intragranular coalescence temperature), \( T_{\text{cd}} \), have been evaluated, as shown in *Figures 7 and 8*, respectively. Coalescence or bridging of dendrite arms is established when the liquid film between two adjacent dendrites or two grains is no more continuous (see *Figures 7b and 8b*). Intergranular and intragranular coalescence does not occur simultaneously: *Picture 7* was taken about 400 s after *Picture 8*. These "coalescence temperatures" are highly dependent on the experimental conditions and on the disorientation of the grains. For this reason, the values of \( T_{\text{cd}} \) and \( T_{\text{cg}} \) reported in *Figure 6a*, are mean values estimated from several measurements. \( T_{\text{cd}} \) is around 43.8°C whereas \( T_{\text{cg}} \) is much lower, 27.6°C. Indeed, for dendrites belonging to the same grain, no grain boundary energy has to be overcome to establish bridges.

![Figure 6. 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 TC (determination of the liquidus temperature of the alloy, \( T_{\text{liq}} \).](image)

Using the back-diffusion model of Clyne and Kurz [8] and the thermophysical properties of Glicksman et al. [9,10], Grasso et al. [11] estimated the solid fraction at intergranular and intragranular coalescence: \( f_s(T_{\text{cg}}) = 0.99 \) and \( f_s(T_{\text{cd}}) = 0.95 \).
Figure 7. Pictures used to determine the temperature of coalescence a) of dendrites across a grain boundary ($T_{CG}$); b) magnification showing spots where coalescence is complete (B) and where some liquid films are still present (A). The upper part of the picture shows some completely solidified region, where only segregation of Fuchsin can be observed.

Figure 8. Pictures used to determine the temperature of coalescence a) of dendrites belonging to the same grain ($T_{CD}$); b) magnification showing spots where coalescence is complete (B) and where some liquid films are still present (A).
V. 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. Coalescence of dendrite arms has also been observed by catching the very moment when liquid films are no longer continuous. 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, around 99%. 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. Indeed, coalescence is a key parameter for the understanding of the mechanical property of the mush and of the hot tearing phenomenon. Theoretical foundation of coalescence within a grain and at grain boundary is also being established [12].

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