Experimental report

10/06/2016

Proposal: **1-01-137** **Council:** 4/2014

Title: In situ diffraction during casting: determination of rigidity point in grain refined Al-Cu alloys

Research area: Materials

This proposal is a continuation of 1-01-134

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Samples: Al-Cu alloys

Instrument	Requested days	Allocated days	From	To
SALSA	4	4	25/07/2015	29/07/2015

Abstract:

Rigidity (or mechanical coherency) temperature is a major input parameter in simulation and optimization of casting and welding processes, as it dictates the temperature below which thermal straining starts. With respect to hot tearing, this temperature is also a key parameter. In metallic alloys, it falls in the solidification interval at a high solid volume fraction. In situ neutron diffraction measurements had been performed by the authors in 2013 at ILL on the instrument SALSA (Grenoble) to study mechanical coherency and onset of stress generation in an Al-13wt.% Cu alloy. The feasibility of such a technique was proven by using the 3 times larger detector and 4 mm focus collimator's at SALSA. With the present proposal, we aim to cast different Cu contents (0.5 wt. pct to 4 wt. pct, maximum susceptibility to hot tearing is 1 wt. pct) in both cracking and non cracking conditions to determine the optimum composition to avoid hot tearing. In the absence of hot tearing, the “free to contract” configuration will allow us to determine the CTE of the alloy up to room temperature. Then the stress generation within the alloy will allow us to determine the as-cast rheology of the alloy.

Experimental report on the measurements carried out at SALSA on July 2015

In situ diffraction during casting: determination of rigidity point in grain refined Al-Cu alloys (exp. No. 1-01-137)

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Coalescence is an important transition in solidification of metallic alloys. It corresponds to the formation of solid bridges between grains. It starts at the coherency point when the grains touch each other but are unable to sustain any mechanical loads and ends up at the rigidity point when the structure is able to sustain tensile strains and stresses. In other words, rigidity temperature (T_{rig}) is reached when the solid phase is sufficiently percolated to transmit tensile and shear strains. This temperature is important as it determines the very instant macroscopic stresses start to build up owing to thermally induced deformations. It is also an important parameter in the hot tearing (solidification cracking) resistance of some alloys.

The present document reports the experimental campaigns which aimed at determining the rigidity temperature of aluminium alloys during their solidification using in situ neutron diffraction. The measurements were carried out at SALSA beamline (ILL, Grenoble) on 25^{th}-29^{th} July 2015 with the assistance of Thilo Pirling. The principles of such measurements are fully detailed in the proposal associated with the present report. During the measurements in July 2015, faster diffraction peak acquisitions (time resolution of 0.5 s and 0.2 s) were possible owing to the use of a larger detector collecting more neutrons in less time. This detector was an ion chamber type, 2D position sensitive. The detection was ensured by 2 mm spaced wires. The active area was 260 x 260 mm^2 with a 1 mm resolution due to interpolation. This detector had 256 x 256 channels, giving a 0.044 deg/channel angular resolution. The sample to detector distance was 1243 mm and the gage volume was 4 x 4 x 15 mm^3 with open vertical slit.

Casting procedure

Thirty one in situ castings were performed using pure Al, binary Al-Cu alloys (1 and 4.5 wt. pct.), Al-Zn alloys (5 wt. pct.) and four industrial alloys AA2100, AA5182, AA6063 and AA7449. The alloy was fully melted in a crucible equipped with electrical cartridges. About 30 min was necessary to melt the alloy. A 10 minute homogenization time was then observed. In some castings, Al-Ti-B2 master alloy (grain refiner, 4%) was added in the crucible and an additional 10 minute period was necessary to let the grain refiner melt entirely. A 5 cm thick plate made of Paris plaster played the role of both tundish with two metal distribution holes and support for the four thermocouples. The plate was carefully positioned on top of the mould using four guiding pins. Due to its thermal inertia, the mould made of 6 kg of copper and 5 kg of stainless steel was not cooled down by running water at its extremity. Some Promat alumina insulating foils 1mm in thickness were placed at the centre of the mould to localise the hot spot and thus the hot tear and to avoid leakage of the metal through the two neutron windows machined out in the mould. In some cases, they were also placed at the extremity of the mould to reduce the overall cooling and get longer solidification times.

The crucible was tilted automatically with the help of pressurized air to pour the alloy into the mould. The temperature and neutron diffraction data acquisition was started using the large detector to get a good time resolution. The experimental setup, mould design and castings obtained at SALSA are presented in figures 1 to 3. Each sample weighted around 260 g for the bone configuration and 120 g for the free to contract configuration. Each dog bone casting was 15 mm wide, 18 cm long and 2 cm high.

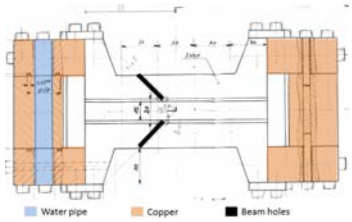

Fig.1: dog bone shape mould design (top view): two holes within the mould avoid neutron absorption by steel. Promat alumina foils prevent metal leakage through the holes.

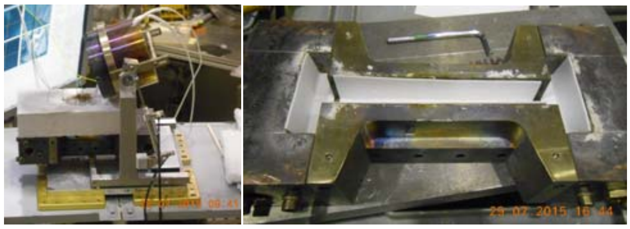

Fig.2: left, experimental setup after tilting the crucible; right, top view of dog bone shape mold with alumina foils in place.

Fig.3: left, cracked casting in the mould; right, five Al-Cu samples, 3 dog bone samples and two free to contract samples.

Hot tearing control

The study focused on the stress generation and possible cracking of the sample during its solidification (hot tearing). To control the occurrence of hot tearing, cooling rates were tuned by changing the coverage of the mould by the alumina foils as shown in fig.2 right. The free to contract configuration was used to obtain the solute and temperature dependent thermal contraction coefficient. This quantity was required to calculate the longitudinal stress development during solidification. The use of grain refiner was also tested. The results for the AA6063 and AlCu1

alloys cast in both free to contract and dog bone configurations are presented below. They are typical of the difficulties we faced during this campaign.

Data acquisition: temperature and neutron diffraction (ND)

As mentioned, a plate equipped with 4 type K thermocouples was placed over the mould. The temperature signals were recorded using the Netdaq program working on a PC and allowed us to calculate the thermal gradient during solidification along the axis of the mould. A typical temperature evolution during casting is presented in fig. 4a. The x position is the distance to the middle of the sample, i.e. to the hot spot where hot tears are expected to form. The casting temperature is around 740°C and the liquidus plateau is clearly visible. At each time a parabolic temperature profile can be fitted using the four measured temperatures.

Simultaneously neutron diffraction data was recorded using the kinetic acquisition mode using a frequency of 2 Hz first and then 5 Hz. Indeed it appeared that with neutron recorded during 0.2 s, a peak with enough count could be fitted with enough precision to find its angular position. A dead time of ca 8 s was required by Salsa to save the data before starting a new acquisition set. To synchronise the thermal and ND data sets, a ND signal yielding 5 V during ND data acquisition was recorded with Netdaq. Fig. 4b shows the ND signal together with the many dead times. This synchronisation was tedious: temperature signals should be recorded by the Salsa data acquisition system. Fig. 4b shows the AA6063 samples cast in both dog bone and free to contract configurations. A hot tear is visible at the very centre of the sample.

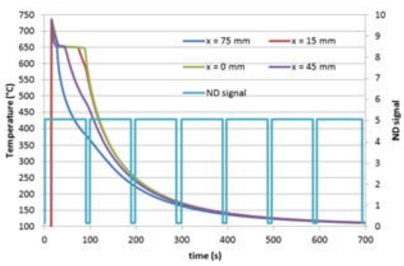

Fig.4a: Temperature profiles during casting the sample 20-AA6063.

Fig.4b: cracked sample 20-AA6063 and free to contract sample.

The raw neutron diffraction signal is presented in fig. 5a with a time resolution of 0.5 s. At high temperatures (above the liquidus of the alloy), no ND peak are detectable. When temperature decreases, a peak starts to appear and shifts to larger diffraction angle during cooling owing to the straining of the casting. This straining has two components: a constrained thermal contraction and an elastic strain associated with the development of a longitudinal stress. A dedicated routine called movav allowed us to sump and average m ND data sets in order to get better definition of the ND peaks. The use of this routine is illustrated in fig. 5 with different values of m, 3, 5 and 10. Indeed, the peak gets better defined but the evolution of its position gets smeared. A value of 5 appeared to be reasonable. Notice that the time resolution was increased to 1 s at lower temperatures thus yielding better peak signals.

Figure 6 presents the angular peak position as a function of the number of averaged periods, zero, three and five. The error on peak position greatly decreases as averaging is made over more periods. Nevertheless, the error remains quite high at the beginning of the experiment, i.e. at high temperatures when crystallites start to form and coalesce together.

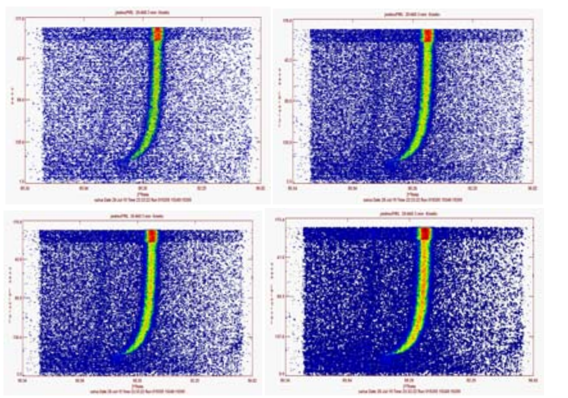

Fig.5: ND signal versus time (or temperature): upper left, raw data, upper right, averaged data over 3 periods, lower left, averaging over 5 periods and lower right, averaging over 10 periods (sample 20-AA6063).

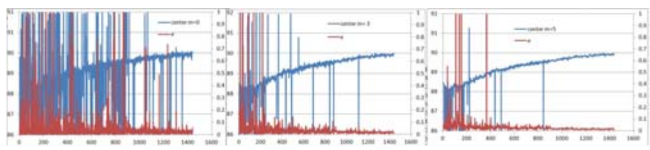

Fig.6: Peak angle position and associated error using raw data (left), averaging over 3 periods (middle) and averaging over 5 periods (right), sample 21-AA6063.

Figure 7 left shows the peak shift evolution during cooling sample 20-AA6063 using an averaging over 5 periods, i.e over 2.5 s. At the beginning, no diffraction peak is present owing to the fact that the alloy is still liquid. The error on the peak position is very high, above 10%. After a certain time, crystals have started to form and to diffract. Diffraction peaks can be fitted with a lower error, typically lower than 1 %. Unfortunately, the time when the very first peak appeared (coalescence temperature) and when they started to shift (rigidity temperature) is absent as ND data were not saved during that period. Using the temperature recorded at the hot spot of the casting, the peak shift can be presented as a function of temperature (cf. Fig. 7 right). No data are present between 650°C and 530°C because of the dead time required to store the ND data. Below 530°C peak shifts to higher angular position owing to the straining of the casting in the dog bone mould.

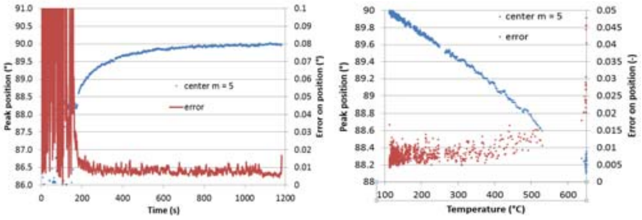

Fig.7: left, peak position versus time, right peak position versus temperature at the hot spot.

Determination of the thermal strain

To compute the evolution of the longitudinal stress that develops during casting in the dog bone configuration, the same alloy was cast in a free to contract configuration (cf. fig. 4b). The temperature and peak position evolution is presented in fig.8. Using the associated peak shift, one can calculate the thermal strain of this alloy as there is no stress generation in this situation. Fig. 9 presents the peak shift during casting and the derivation of both the thermal strain and the coefficient of thermal expansion (CTE) of the alloy. Over the range 130°C – 630°C a mean CTE of 32 10^{-6} /K is found (cf. fig. 9 left). As temperatures within the solidification interval are of interest, the mean CTE in the range 480°C – 640°C is slightly higher, 37 10^{-6} /K. All these values are common values for aluminium alloys.

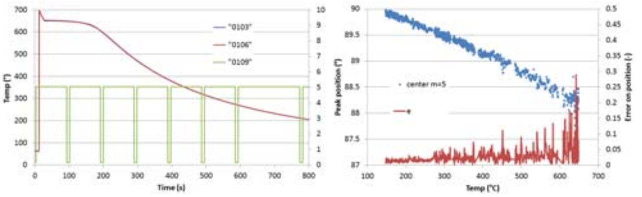

Fig. 8: temperature and peak position evolution during casting in a free to contract configuration.

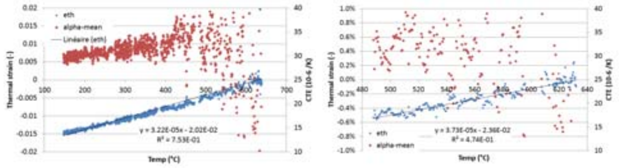

Fig. 9: thermal strain and coefficient of thermal expansion of the AA6063 alloy as a function of temperature (21-AA6063).

Stress generation during casting AA6063 (0.5 s periods)

By differentiating the peak shift of a given alloy cast under both the free to contract and dog bone configurations, one can calculate the stress generation within the sample. Let us defined d0 as the (311) lattice parameter at a reference temperature T0 higher than the rigidity temperature, Trig, where crystallites are sufficiently big so that a lattice parameter can be measured using diffraction. The rigidity temperature is defined as the temperature where grains are sufficiently coalesced to transmit tensile stresses. One of the gaol of the present study is to determine this temperature for different alloys. The stress generation during casting is computed by comparing the (311) lattice spacing in the free configuration, dfree, and in the dog bone configuration, ddog:

$$d = d0 \text{ at To with } \lambda = 2d0\sin\theta_0\ ,\ d_{free} = do\left(1+\alpha(T\text{-}T0)\right) \text{ and } \varepsilon_{free} = \frac{d_{free}\text{-}do}{do} \text{ at } T < To$$

$$\lambda = 2d_{dog}\sin\theta_{dog}\ ,\ \lambda = 2d_{free}\sin\theta_{free}\ ,\ \varepsilon_{dog} - \varepsilon_{free} = \varepsilon_{dog} - \varepsilon_{th} = \frac{\sigma}{E(T)} = \frac{d_{dog}\text{-}do}{do} - \frac{d_{free}\text{-}do}{do} = \frac{d_{dog}\text{-}d_{free}}{do}$$

$$\sigma(T) = E(T)\sin\theta_0\left(\frac{1}{\sin\theta_{dog}} - \frac{1}{\sin\theta_{free}}\right) \quad \text{with } \sin\theta = \sin\left(\frac{\pi}{180}\frac{centre}{2}\right)$$

The evolution of the peak position in both configurations and the associated longitudinal stress is presented in fig. 10 using a fixed Young's modulus of 35 GPa and an averaging over 5 periods, i.e. over 2.5 s. The calculated stress shows a rather erratic evolution but remains positive with low values. One has to notice that the sample cracked (cf. fig. 4b) and thus low stresses are expected.

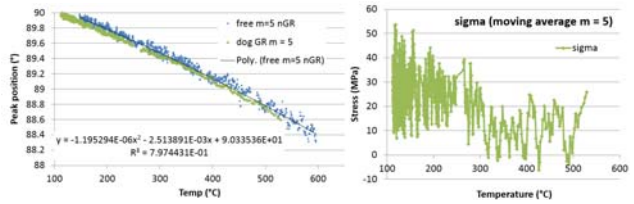

Fig. 10: left, peak evolution in free to contract and dog bone configurations. Right, stress generation in the AA6063 alloy sample. Notice that this sample cracked and thus stresses remain very low.

Stress oscillations might be explained by the dynamic nature of hot tearing where at the same time crystallites coalesce, transmit tensile stresses (stress build up) and cracks initiate and release those stresses. The averaging might also impact these results: this point is further studied in the next section.

Stress generation during casting Al-Cu1% (0.2 s periods)

Figure 11 shows the temperature and ND data recording for the Al-Cu 1 wt pct in both free to contract and dog bone configurations.

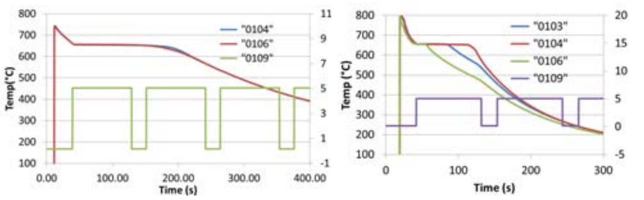

Fig. 11: left, temperature evolution in free to contract AlCu1 casting and right, in dog bone configuration (28-AlCu1 and 30-AlCu1).

Using the ND data recorded during the free to contract casting, a mean CTE of 35.5 10^{-6} /K is obtained as shown in fig. 12 left. This quantity is then used to calculate the stress build up during casting in the dog bone configuration. Results are presented in fig. 13 using a Young's modulus of 35 GPa and averaging over 3, 5 and 7 periods, i.e. over 0.6 s. 1s and 1.4 s, respectively. Averaging has an impact on the results: it decreases the error in the peak position but at the same time, it reduces the amplitude of the oscillations. In Fig. 14, the solid fraction curve is reported for the Al-Cu 1 wt. pct alloy together with an attempt to determine the rigidity temperature of the alloy. This one is defined as the temperature at the build-up of the first stress accumulation. Using m = 3, rigidity seems to occur between 630°C and 640°C, i.e. at solid volume fractions between 80% and 90 %.

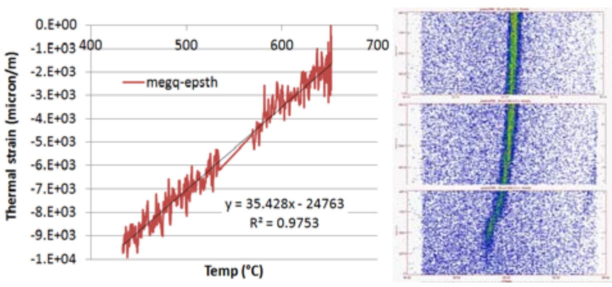

Fig. 12: left, determination of the value of CTE at high temperatures.

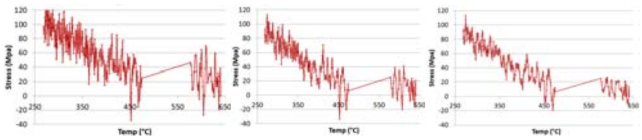

Fig. 13: impact of averaging over 3 (left), 5 (middle) and 7 (centre) periods on stress development in AlCu1 casting (30-AlCu1).

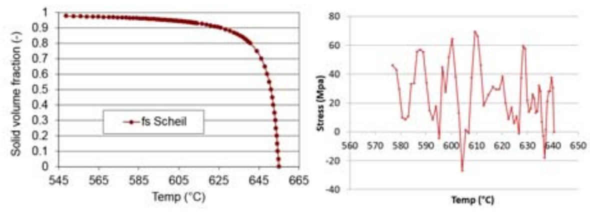

Fig. 14: solid fraction curve for the Al-Cu 1 wt. pct alloy and determination of the rigidity temperature of the alloy.

Conclusions

Neutron diffraction can theoretically be used to determine coherency and rigidity points, i.e. coalescence transition, in solidifying aluminium alloys. Although a large detector allowed us to obtain good time resolution, of the order of 0.2 s, it appears that we are still at the limit of detection of the very beginning of stress generation during casting. Stresses remain very low when a hot tear initiates and their values oscillate were also found using X-ray in situ castings. They are explained by the fact that the structure accumulates stresses before a hot tear forms and relaxes those stresses by propagating in the mushy alloy. For the next measurement campaign at Salsa (proposal 1-01-149), one should:

- use the kinetic count mode without dead time periods during ND data savings,
- record temperature signals on Salsa acquisition system to synchronize them with ND data,
- cast with much longer solidification times using thicker Promat insulating foils in both cracking and not cracking conditions.

With all this, one should be able to determine the stress development although we are seeking for very low stresses.

J.-M. Drezet, B. Mireux, Z. Szaraz and T. Pirling: *In situ* Neutron Diffraction during Casting: Determination of Rigidity Point in Grain Refined Al-Cu Alloys. *Materials* **2014**, *7*, 1165-1172, http://www.mdpi.com/1996-1944/7/2/1165

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J.-M. Drezet, B. Mireux, G. Kurtuldu, O. Magdysyuk and M. Drakopoulos, Measurement of Mechanical Coherency Temperature and Solid Volume Fraction in Al-Zn Alloys Using In Situ X-ray Diffraction During Casting, Metallurgical and Materials Transactions A: Volume 46, Issue 9 (2015), Page 4183-4190, doi: 10.1007/s11661-015-3041-y