Experimental report

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Proposal: 1-01-149 **Council:** 4/2015

Title: Measurement of rheology of solidifying aluminum alloys using in situ neutron diffraction during casting

Research area: Materials

This proposal is a new proposal

Main proposer: Jean-Marie DREZET

Experimental team: Nicolas CHOBAUT
Jean-Marie DREZET

Local contacts: Thilo PIRLING

Samples: Aluminium

Instrument	Requested days	Allocated days	From	To
SALSA	4	4	05/09/2016	12/09/2016

Abstract:

Stress build up in solidifying metallic alloys is of high importance as it impacts the integrity of the final casting through at least three aspects, associated distortions, formation of stress related defects such as solidification cracking and/or cold cracking and finally residual stresses that are detrimental for further material processing. The generation of stresses during casting is due to the conjunction of thermal gradients (usually high for productivity reasons and for obtaining fine grains) that impose thermally induced deformations on a material that has low yield strength close to its solidus temperature. In all castings, the alloy plastifies at high temperatures thus leading to the formation of more or less intense residual stresses. Macroscopic tensile strains and stresses are transmitted throughout the solid plus liquid mixture once grains or grain clusters have sufficiently coalesced. This transition called rigidity or mechanical coherency has been measured using in situ neutron diffraction casting. The goal of the present proposal is to determine the rheology of the as cast alloy using the same in situ casting experiments.

Experimental report on the measurements carried out at SALSA on Sept 2016

Measurement of rheology of solidifying aluminium alloys using in situ neutron diffraction during casting (exp. No. 1-01-149)

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Dr Jean-Marie Drezet, IMX, Ecole Polytechnique Fédérale de Lausanne

Coalescence corresponds to the formation of solid bridges between solidifying metallic 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 4th-9th Sept. 2016 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 Sept. 2016, kinetic mode was used for the diffraction peak acquisitions together with a large detector. With this mode, each neutron impacting the detector is considered as an event and both time and coordinates of this neutron are saved. The 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 10 = 160 mm^3 with open vertical slit. The neutrons had a wavelength of 1.72 Angstroms.

Casting procedure

Twenty four in situ castings were performed using binary Al-Cu alloys (4.43 wt. pct.), Al-Zn alloys (5 wt. pct.) and four industrial alloys AA2100, AA5182, AA6063 and AA7440. 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, 0.4% in weight) 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 one or 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 all over 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 synchronised using a 5V signal. The experimental setup, mould design and castings obtained at SALSA are presented in figure 1. Each sample weighted around 240 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. Centre: top view of dog bone shape mold with alumina foils in place. Right, 3 AA6063 samples, 2 dog bone samples and one 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.1 centre. 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. With a higher grain density, neutron statistics gets better. The results for the AlZn5 and AA6063 alloys cast in both free to contract and dog bone configurations are presented below. They are typical of the results obtained 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. When one inlet was used, temperature could not be measured at the hot spot (centre of the mould): in that case, temperature was extrapolated along the mould length to get the thermal history at the hot spot. A typical temperature evolution during casting is presented in fig. 2. The liquidus temperatures are used to calculate the Zn content. Although AlZn5 wt pct were cast, the Zn content at the hot spot is much higher owing to macrosegregation during solidification. Simultaneously neutron diffraction data was recorded using the event mode. To synchronise the thermal and ND data sets, a ND signal yielding 5 V during ND data acquisition was recorded with Netdaq. Fig. 2 centre shows two castings: one with one inlet that did not crack and the second one with two inlets where a hot tear appeared at the centre of the casting in the middle between the two inlets.

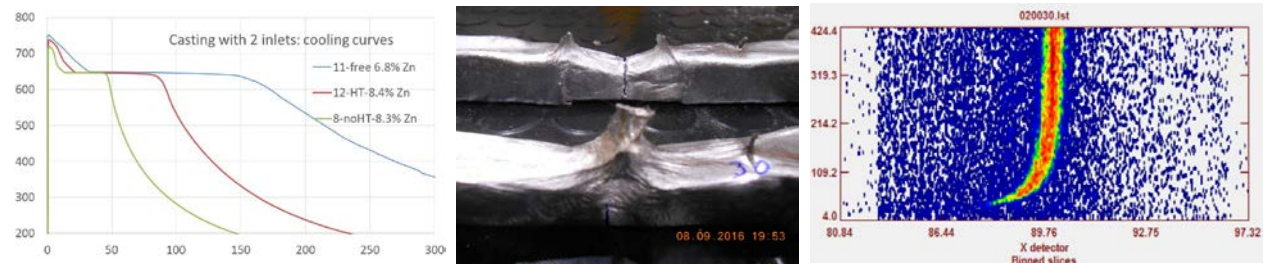

Fig.2: Left, temperature evolution during casting for 3 Al-Zn5% samples. Centre, AlZn5 cast with two inlets showing a hot tear and cast with one inlet showing no crack. Right, ND signal versus time (or temperature) with averaging over 9 periods.

The neutron diffraction data averaged over 9 periods using the dedicated routine called movav (sump and average m ND data sets in order to get better definition of the ND peaks) are presented in fig. 2 right. 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 three components: a constrained thermal and solutal contraction and an elastic strain associated with the development of a longitudinal stress.

Determination of the thermal strain, elastic strain and longitudinal stress
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. 1). The temperature and peak position evolution is presented in fig.3 for the AA6063 alloy. Using the associated peak shift, one can calculate the thermal strain of this alloy as there is no stress generation in this situation. A polynomial fit is used for the thermal component of the strain. 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. Results are presented in fig.3 right and left for two cracked castings. At temperatures higher than 400°C, the two signals associated with the cracked samples yield lattice parameters that are higher than the stress free samples. This is the signature of the development of straining of the solidifying alloy. The difference in straining is converted in elastic component and then in longitudinal stress using an elastic modulus of 1 GPa. Notice that this last quantity is highly questionable owing to the difficulty to measure it.

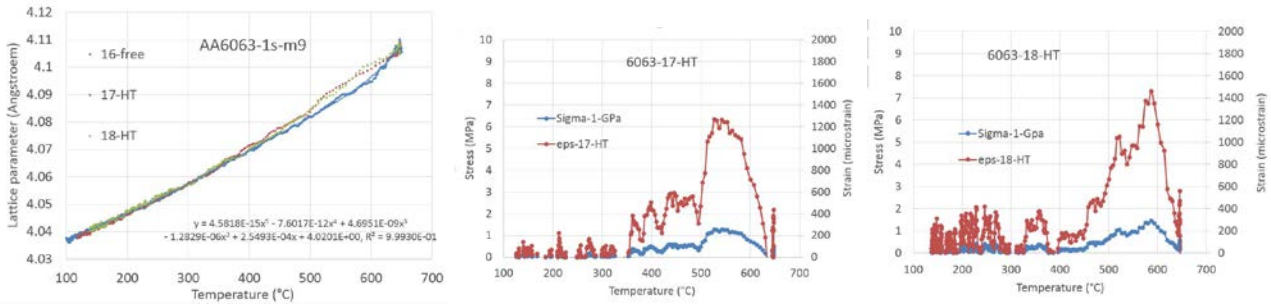

Fig. 3: left, lattice parameter during casting in a free to contract configuration and during constrained casting. Centre and right, elastic strain and longitudinal stress as a function of temperature for the two cracked AA6063 samples.

Comparison of the fraction of solid with the peak height and peak integral
During solidification, the volume fraction of solid increases from 0 (fully liquid) to 100 % (fully solid) over a temperature range that depends on the solute content. For the Al-Zn alloys, one can see in fig. 4 that both the peak height and the peak integral increase during solidification.

Thermo-solutal and shrinkage components of the lattice parameter
During solidification, the mean solute content in the solidified grains increase in both Al-Cu and Al-Zn alloys as they both have a partition coefficient that is lower than unity. In addition, the lattice parameter rapidly decreases owing to

solidification shrinkage. This rapid decrease is visible in Al-Zn alloys in fig. 4 by the initial large decrease in the lattice parameter or associated large increase in the peak angle. This transition takes place close to the liquidus temperature of the alloy when early grains grow rapidly. Using micro-segregation models such as the Scheil model for the calculation of the fraction of solid and mean concentration of the grains, the next step is to fit the lattice parameter in the solidification range taking into account a thermal, solutal and shrinkage components:

$$a = a_0\left[1 + \alpha_T(T\text{-}Tliq) + \alpha_s(C_s\text{-}kC_0) + f_s\Delta a\right]$$

Fig. 4 shows the determination of the 3 coefficients for the Al-Zn alloy: the shrinkage coefficient is 2.2% linear i.e. 6.6% in volume, typical value for Al alloys.

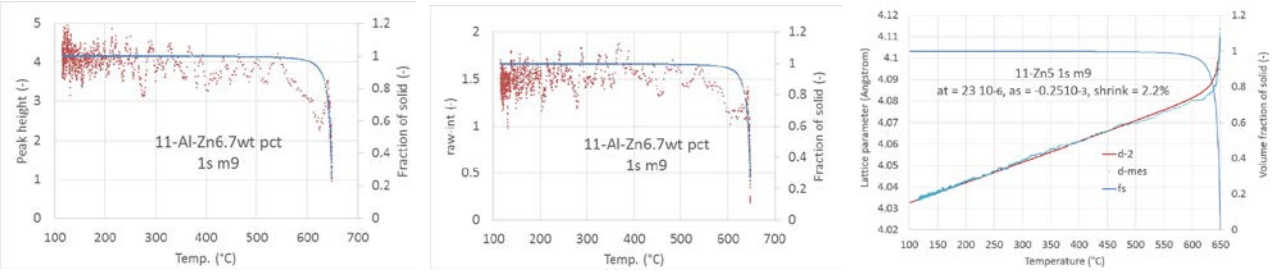

Fig. 4: left and centre, peak height and peak integral versus volume fraction of solid for the AlZn alloy cast in the free to contract configuration. Right, determination of the thermal, solutal and shrinkage coefficients.

Conclusions

Using a large a large detector and the event mode where each neutron position is recorded helped us to acquire a lot of information in solidifying aluminium alloys. Although further analysis of the data is required, preliminary results show that it is possible to:

- determine the solutal, thermal and shrinkage coefficients of the lattice parameter in solidifying Al-Cu and Al-Zn alloys. To the author's knowledge, this is the first time the rapid decrease of the lattice parameter during solidification is measured in metallic alloys.
- determine the coalescence and rigidity temperature of solidifying alloys,
- and evaluate the maximum strain, i.e. the ductility, a mushy alloy can sustain without initiating hot tears.

Stresses can be assessed as well although the elastic modulus is not known at such high temperatures. Stresses remain very low when a hot tear initiates and their values oscillate as found in the past using X-ray in situ castings. The structure accumulates stresses before a hot tear forms and relaxes those stresses by propagating a hot tear in the mushy alloy. The major difficulty resides in the fact that solidification is rather fast and that stresses remain particularly low.

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

J.-M. Drezet, B. Mireux, Z. Szaraz and T. Pirling: Determination of Coherency and Rigidity Temperatures in Al-Cu Alloys Using In Situ Neutron Diffraction During Casting, JOM (TMS) Volume 66, Issue 8 (2014), Page 1425-1430.

J.-M. Drezet, B. Mireux and G. Kurtuldu: In situ X-ray Diffraction during Casting: Study of Hot Tearing in Al-Zn alloys, Advanced Materials Research Vols 1120-1121 (2015) pp 1134-1141, Trans Tech Publications, Switzerland, doi:10.4028/www.scientific.net/AMR.1120-1121.1134.

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

The present Salsa data are to be referred as DOI (10.5291/ILL-DATA.1-01-149)