Experimental report

06/06/2016

Proposal: **1-02-171**

Council: 10/2014

Title: Heterogeneous stress field within large fused-cast zirconia bricks probed by neutron diffraction

Research area: Engineering

This proposal is a new proposal

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Samples: ZrO_2

Instrument	Requested days	Allocated days	From	To
SALSA	7	7	19/06/2015	26/06/2015

Abstract:

The manufacturing of high quality glasses and the need to increase the lifetime of furnaces used for glass fabrication, imply the development of new high zirconia refractory materials with good thermomechanical properties. This material is indeed used to make large bricks (~450×400×250 mm3, and 250kg) that are adjusted to build the inner wall of huge glass furnaces. During the fused-cast process used to elaborate these bricks, zirconia is undergoing two phase transitions (cubic → tetragonal → monoclinic) at ~2300°C and ~1000°C respectively, the latter being associated with a large increase of the lattice cell volume (~ 4%). A heterogeneous microstructure is observed in such bricks, and large internal stresses, which could be at the origin of defects and sometimes failures, are expected.
The aim of the present proposal is to perform diffraction experiments on a whole brick in order to investigate the effect of the elaboration process on the residual stresses distribution, at a scale rrepresentative of macroscopic gradients on a whole brick.

Aim

Refractory blocks containing high amount of ZrO_2 prepared by electro-fusion and casting develop high amount of residual stresses as a result of the cooling conditions and structural phase transformations (SPTs) they undergo. This leads to stress concentration and crack formation at different length scales (Cockcroft et al., 1994). In order to characterize these phenomena, a ``multi-scale" analysis approach is under development wherein different internal strain measurement methods are combined. In this framework, we have conducted strain measurement experiments at the SALSA beamline on ILL. Two blocks of different compositions were prepared and cast by our industrial partner. Compositions of these blocks are 94 wt% ZrO_2 and 6 wt% borosilicate glass for the block labelled ZB and 94 wt% ZrO_2 and 6 wt% alkalisilicate glass for the one labelled ZS .

Experiments

ZrO_2 blocks were measured in their entirety, i.e. without slicing or sampling. Final dimensions were 437x240x305 mm^3 for the block of composition ZB and 500x102x688 mm^3 for the block of composition ZS. Each one of the measured blocks, weighing approximately 250 kg., was mounted on the hexapod of beamline SALSA which serves as a goniometer for large industrial pieces.

Measurements were done on fixed 2θ ranges of 25-40° and 55-70°. The area of the horizontal cross-section of the diffraction volume was 4x4xsin(2θ) mm^2. The height of the gage was 20 mm. Wavelength was set to 1.644 Å during the entire experiment. Counting for neutrons per given position was done either until 50000 total neutron counts were received on the detector or for one hour, whichever condition was reached earlier.

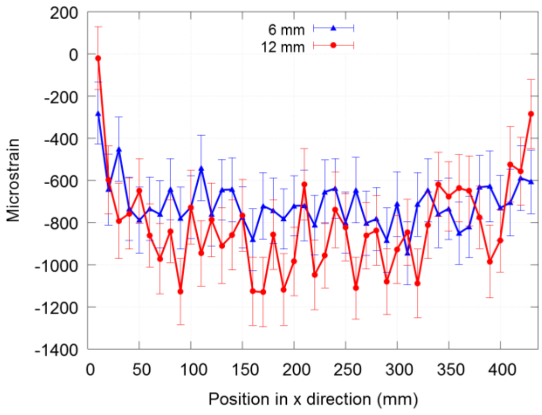
For the treatment of the measured diffraction images, LAMP software written by the ILL staff was used (Richard et al.). For radial integration of the 2D diffraction patterns, a software called DataGUInz (Klosek, 2014) was utilized. After integration, a correction is applied to the data to remove the possible effects of radial collimator foils on the diffracted beam intensity.

In order to obtain the exact maxima positions for the peaks, a Gaussian function is used: Fits were done for the peak around 2θ=61°, which contains information on the $(\bar{3}11)$ reflection. Other fits were made on the $(\bar{1}11)$ reflection for one of the blocks (ZS) to compare strain values based on different reflections.

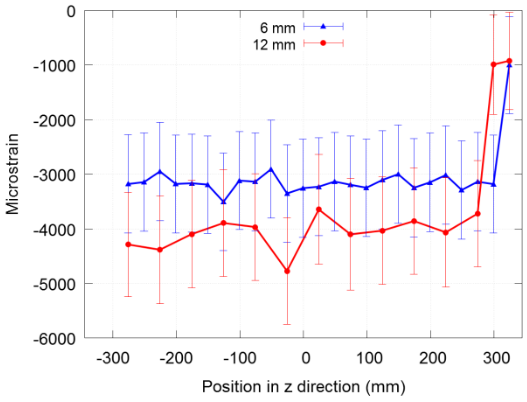
Reference peak positions were determined by measuring cubes of 10x10x10 mm^3 cut from the corners of the blocks. The cutting was done for the purpose of producing a sample which is free of internal stresses on scale of interest of this measurement. These reference samples yielded $\overline{2\theta}_0$=60.682(8)° for the $(\bar{3}11)$ peak of ZB composition. For the $(\bar{3}11)$ and $(\bar{1}11)$ peaks of the ZS composition, the results are $\overline{2\theta}_0$= 60.705(25)° and $\overline{2\theta}_0$=27.796(25)° respectively.

Results

In general, the measurements yield between 250 microstrain (=strain x 10^6) in the tensile direction and 1000 microstrain in the compressive directions for the block of composition ZB. The block of composition ZS shows a slightly smaller range of strain. There is a clear strain gradient from the surface to the centre across the large faces of the ZB block as shown in Fig.1a.



(a)



(b)

Figure 1 : Results of strain measurements for a scan made along (a) a horizontal line on the larger surface of the ZB block based on ($\bar{\mathbf{3}}\mathbf{11}$) reflection and (b) a vertical line (the casting direction) on the larger surface of the ZS block based on ($\bar{\mathbf{1}}\mathbf{11}$) reflection.

For the ZS block, measurements based on the $(\bar{1}11)$ reflection of the monoclinic crystal results in significantly higher amount of strain (~5 times larger). One of the reasons that could partially explain this observation is the anisotropic elastic modulus of ZrO_2.

Again for the ZS a line along the casting axis (z-direction) was measured for the $(\bar{1}11)$ crystal planes. As shown in Fig.1b, the strain-level is generally aligned with what was measured along the horizontal direction (x- and y-directions) for the same reflection. Nevertheless, the sharp increase of the compressive strain might be hinting towards a different gradient along this direction.

Based on our results, the effects of the cooling conditions and the martensitic phase transformations on the development of the internal stresses were found to be multifaceted. Thermal gradients forcing volumetric expansions due to phase transformations to occur at different times in different locations within the block should be regarded as a peculiar case of these materials. This is believed to be one of the most prominent causes of internal deformations at this scale within these blocks produced by casting. Our results at this point should be especially helpful for the mechanical modelling of this phenomenon.

References

Cockcroft, S. L., Brimacornbe, J. K., Walrod, D. G., & Myles, T. A. (1994). *J. Am. Ceram. Soc.* **77**, 1512–1521.

Klosek, V. (2014). Internal report of the Laboratoire Leon Brillouin Saclay (France).

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