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# Neutron diffraction measurements of residual stress distribution in large zirconia based refractory bricks produced by electro-fusion and casting

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## ABSTRACT

Electro-fusion and casting is used to produce large refractory bricks (~250 kg) containing a high amount of ZrO<sub>2</sub>. These bricks are used in glass-making furnaces where good mechanical performance is required at very high temperatures (>1500 °C). During the manufacturing procedure, they develop large residual stresses as a result of the cooling conditions and structural phase transformations they underwent. This leads to stress concentration and crack formation at different length scales. In order to characterize these phenomena, a 'multi-scale' analysis approach is under development, where different internal strain measurement methods are combined. In this approach we benefit from different gauge volumes provided by various diffraction methods, ranging from a few hundred nanometres to a few tens of millimetres. In the present paper, the results of neutron diffraction measurements on large ZrO<sub>2</sub> blocks are given. These results show the level of internal strains at the millimetre scale, based on (311) reflection of the monoclinic ZrO<sub>2</sub>. Overall, a range of 0.025% tensile to 0.1% compressive strain was observed. Clear strain gradients were also visible, as larger values in the interior of the block were encountered.

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## 1. Introduction

Refractory materials containing high amounts of ZrO<sub>2</sub> are widely used as flux blocks for glass-making furnaces owing to the resistance of ZrO<sub>2</sub> to the corrosive effects of molten glass [1,2]. The size of these blocks is several hundreds of millimetres. In order to fully benefit from the corrosion resistance, a lower degree of open porosity is required. Therefore, the large ZrO<sub>2</sub> blocks are produced by electro-fusion followed by casting. However, during the manufacturing process, thermal gradients, successive structural phase transitions (SPTs), and anisotropic thermal contraction of zirconia induce the development of significant amounts of internal residual stresses [3,4].

Under ambient pressure, pure zirconia undergoes three SPT during cooling from its melt [5]. It solidifies into a cubic crystal structure (c-ZrO<sub>2</sub>, space group *Fm* $\bar{3}$ *m*) at about 2700 °C; upon

cooling to 2300 °C it transforms to tetragonal (t-ZrO<sub>2</sub>, space group *P*<sub>4</sub><sub>2</sub>/*nmc*), and finally it becomes monoclinic (m-ZrO<sub>2</sub>, space group *P*<sub>2</sub><sub>1</sub>/*c*) around 1170 °C. This last SPT is a first order transition and is of martensitic type. Moreover, it induces a large volumetric expansion which, in the absence of any stresses, is typically close to 4.5%. Therefore the tetragonal–monoclinic SPT alone creates large amounts of strain in the material and induces cracking, which finally leads to breaking the pure bulk zirconia to pieces. In the case of refractory blocks containing a large volume fraction of zirconia, an amorphous matrix phase is used between the ZrO<sub>2</sub> crystals and acts as a medium for partial stress relief [1].

Because of the stress effect, the actual temperatures of the SPTs between tetragonal to monoclinic phases (both forward and backward transformations) are usually very different from that predicted for a perfect free single crystal. The transition of a single crystal from one state to the other occurs when the sign of the free energy balance between the two phases changes. This depends mainly on the temperature, the size of the considered crystal, and the energy associated with the interfacial strain. In a zirconia block containing a very large number of zirconia crystals (having various sizes and crystal orientations), the tetragonal–monoclinic SPTs

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occur thus in a large temperature range, and a hysteresis effect is observed both during the heating and cooling processes of the sample. Similar to what is observed in steels, this martensitic transition generally occurs at a lower temperature during the cooling than during the heating, and in some cases, residual tetragonal zirconia can be observed at room temperature.

Substantial efforts on the numerical modelling of these stresses have been ongoing for two decades [3,4,6]. Nevertheless, since the causes for the internal stresses are numerous, with each of them involving different but co-dependent phenomena, these models do not yet account for all deformation mechanisms and therefore are not yet predictive. In order to rigorously analyse the stress build-up within these  $\text{ZrO}_2$  blocks, an experimental multi-scale approach is being developed in which we combine neutrons, X-rays and electron diffraction. The global scale, including the contribution of a large number of crystals, is explored by both room temperature neutron diffraction and in-situ high-temperature X-ray diffraction experiments. The strain state is also probed at the very local scale through Laue microdiffraction and electron back scatter diffraction (EBSD). Thus the used diffraction volumes range from a few hundreds of nanometres to a few tens of millimetres. These different diffraction gauge volume sizes enable averaging over different scales and thus facilitate the analysis of the effects of different causes of stress on the system.

In this paper, we discuss the results of the first part of this analysis: the residual strains at millimetre scale as measured by *neutron diffraction*. Two different blocks of high zirconia content were prepared by Saint-Gobain CREE. These two blocks differ mainly in the composition of the amorphous matrix phase around the zirconia crystals.

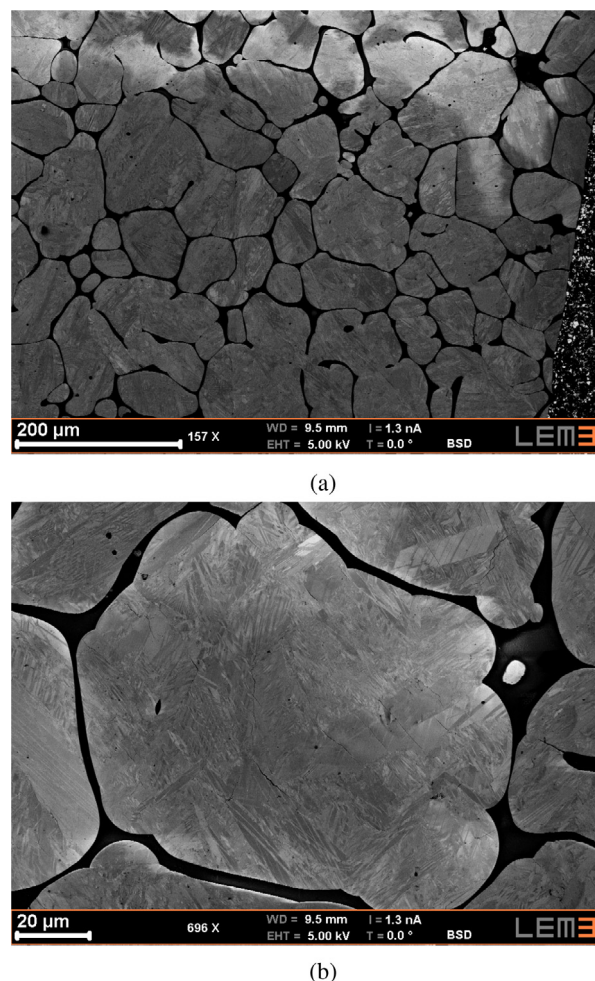
Neutron diffraction is a widely used technique for the measurement of the residual stress of crystalline matter on larger scales. Even though hard X-rays from high brilliance synchrotron sources are emerging as strong alternatives [7], neutrons still perform better at high penetration. A good review for the comparison of the two methods is given in [8]. Since neutron beams are less affected by intensity loss at greater depths than are X-ray beams, neutron diffraction is generally the method of choice when it comes to large engineering pieces. There is a vast literature on strain measurements made by neutron diffraction on large industrial metallic pieces [9–12]. However, to the best of our knowledge, there seems to be a clear lack of residual stress–strain studies with diffraction methods on ceramic materials produced at this scale. The only neutron measurements on refractory blocks with such a high zirconia content was done for the purpose of texture analysis [13]. Therefore the significance of the present paper is also that it is among the first in the field of residual strain analysis of an industrial size ceramic refractory material.

## 2. Experimental

### 2.1. Sample preparation and microstructure

Two blocks of different compositions were prepared and cast by the company Saint-Gobain CREE. These compositions are exactly the same as those reported in [13]. They therefore both contain 94 wt.%  $\text{ZrO}_2$ . The remaining content (6 wt.%) is alkalisilicate type glass for one block (labelled ZS hereafter) and borosilicate glass for the other (ZB hereafter).

Fig. 1a shows the typical microstructure of these materials exemplified by a sample of ZB composition which was cut and polished for SEM imaging after casting. The micrograph reveals large zirconia areas (in grey) formed during the solidification process [14], having sizes of a few hundred micrometres. Each of these areas was initially (i.e. at high temperature) a single crystal with a cubic



**Fig. 1.** Microstructure of the ZB sample.  $\text{ZrO}_2$  'domains' (in grey shades) in a glassy matrix (in black) (a), and a close-up of a single domain, showing the needle- and plate-like monoclinic  $\text{ZrO}_2$  crystallites (b). The image is taken from a fused-cast sample with the same composition as the measured block sample.

structure [15]. These areas are surrounded by the amorphous silicate phases (in black). As seen in Fig. 1b, they contain a large number of monoclinic  $\text{ZrO}_2$  crystals which are arranged in specific orientations with respect to each other. The sizes of these crystals range from 50  $\mu\text{m}$  down to a few hundreds of nanometres. The EBSD results on these materials [14] confirm a very well defined local texture within the mentioned areas of a few hundred micrometres. This is a result of orientation relationships established by the successive SPTs [15]. The texture at the macroscopic scale is, on the other hand, found to be random [13].

### 2.2. Neutron diffraction

Using neutron diffraction is the best option when one wants to scan very thick specimens up to a few centimetres depth. The neutron diffraction measurements were done at the SALSA beamline of Institute Laue-Langevin (ILL) in Grenoble, France. the  $\text{ZrO}_2$  blocks were measured in their entirety, i.e. without slicing or sampling. The only treatments made to the blocks were removing the riser (masselotte) and machining the surface to decrease the surface roughness.

The final dimensions were  $437 \times 240 \times 305 \text{ mm}^3$  for the block of composition ZB and  $500 \times 102 \times 688 \text{ mm}^3$  for the block of composition ZS along the x-, y- and z-directions of the reference system in Fig. 2. A schematic view of the ZB block is also given in

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