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INTERFACE RESIDUAL STRESSES IN DENTAL ZIRCONIA USING LAUE MICRO-DIFFRACTION

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Abstract

Due to their aesthetic value and high compressive strength, dentists have recently employed ceramics for restoration materials. Among the ceramic materials, zirconia provides high toughness and crack resistant characteristics. Residual stresses develop in processing due to factors including grain anisotropy and thermal coefficient mismatch. In the present study, polychromatic X-ray (Laue) micro-diffraction provided grain orientation and residual stresses on a clinically relevant zirconia model ceramic disk. A 0.5 mm x 0.024 mm region on zirconia was examined on a 500 nm scale for residual stresses using a focused poly-chromatic synchrotron X-ray beam. Large stresses ranging from – to + 1GPa were observed at some grains. On average, the method suggests a relatively small compressive stress at the surface between 47 and 75 MPa depending on direction.

Introduction

Typical core restorative materials in modern dentistry include alumina, zirconia, or noble metal alloys that are veneered with porcelain. Zirconia provides high toughness and crack resistant characteristics. The phase transformation from tetragonal to monoclinic produces local volume expansion favoring crack closure and results in additional strength. However, during crown fabrication in dental laboratories, residual stresses develop due to factors including grain anisotropy, thermal coefficient mismatch, and the overall complex tooth geometry. Along with stress induced phase changes, grain-grain stresses are significant. In the present study, polychromatic X-ray (Laue) micro-diffraction provided grain orientation and residual stresses on clinically relevant disk shaped dental zirconia. The ceramic disk was fabricated considering dental laboratory thermal history and material properties typically utilized for dental restorations. The deviatoric strains developed within zirconia, as a result of grain anisotropy and the coefficient thermal expansion mismatch, were determined by highly focused polychromatic

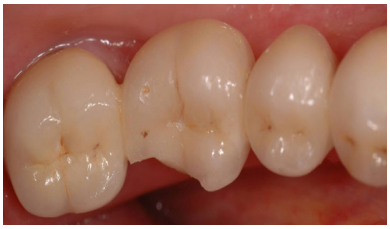


Figure 1 Fractured porcelain veneer layer off the top of a 4 unit Zirconia FPD [1]

synchrotron X-ray Laue diffraction at Beamline 12.3.2 of the Advanced Light Source. Laue diffraction spots obtained from the sample were analyzed by indexing and fitting the peaks from the tetragonal phase of zirconia.

The analysis revealed a histogram of the residual stresses, for each component of the deviatoric residual stress tensor. While the mean of the deviatoric stresses were typically, between -47 and -75 MPa (compressive as in Figure 6), a small fraction of localized compressive and tensile stresses reached 1 GPa. The study also resulted in an observation of the relation of peak fitting parameters. For example, the angular deviation parameter and the peak search box size showed an important influence on the accuracy of stress results.

Method

The current experiment measured the grain-to-grain residual stress within zirconia. Laue diffraction was utilized to observe the stress in individual grains. A polychromatic beam focused to a cross section of $0.5 \times 0.5\ \mu m^2$ at Beamline 12.3.2., of the Advanced Light Source, was made incident on the zirconia sample. Emerging diffracted X-rays from the sample, produce spots on the CCD oriented in 90° reflection geometry as shown in Figure 2.

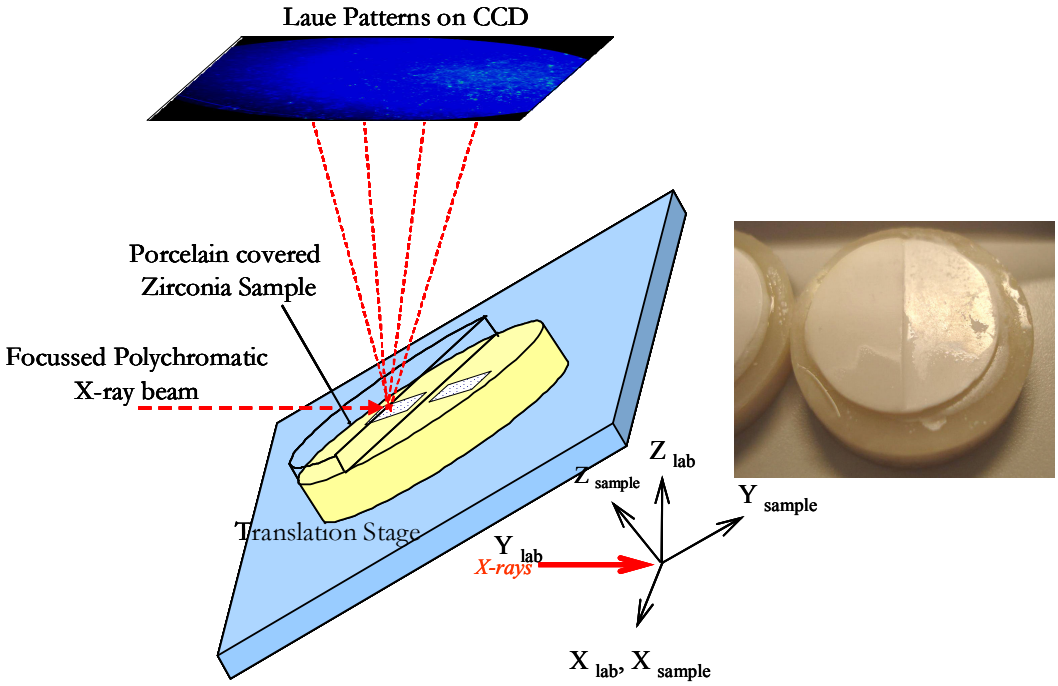


Figure 2 Schematic of the sample orientation in 90° reflection geometry. The sample consists of two semicircular zones where raster scans were obtained to study the effect of residual stresses. Only the zone without porcelain is discussed here.

Spots registered for every position of the beam on the sample were collected onto an X-ray CCD and analyzed using XMAS [2] (X-ray micro-diffraction analysis software). The X-ray beam, has an energy range of 6-22 keV. The measurements were carried out on the zirconia surface as well as the porcelain covered side of zirconia. After a beam spot is incident for a fixed interval on the sample, the control software translates the sample to a new location. Using an automated scan, the sample was rastered with micro-focused beam spot. TiO_2 marker dots were utilized to facilitate positioning of the sample. The sample re-alignment was done in successive steps using a focused laser beam aligned parallel to the X-ray beam. Results from the analysis of the data collected on the zirconia sample without porcelain are presented in this paper.

Sample Preparation

The samples consisted of a disk 1 mm thick made of sintered polycrystalline yttria stabilized zirconia. The disk was half coated with a porcelain layer 0.5 mm thick which is ground and polished. The porcelain layer and the zirconia disk were fabricated under typical dental laboratory thermal cycling for fabrication of all ceramic zirconia-porcelain systems. No further processing was required for Laue-XRD measurements.

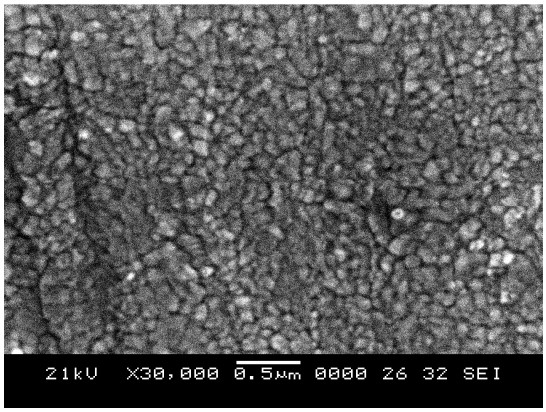


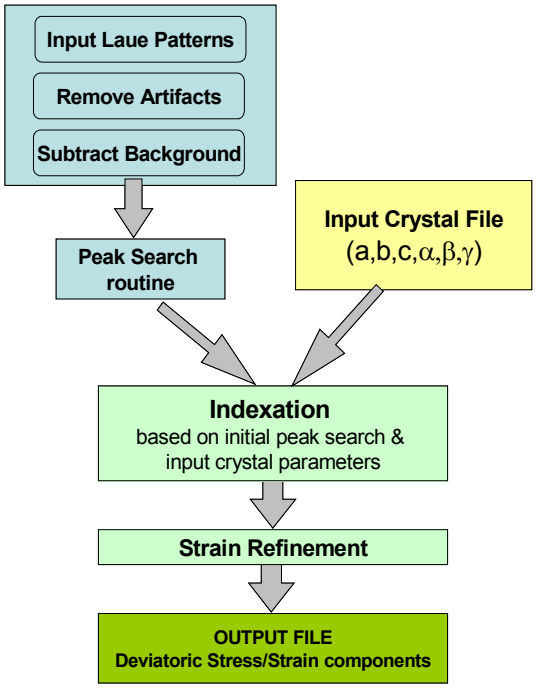
Figure 3 SEM image of the neat Zirconia disk surface at 30000X (bottom) magnification. The grain sizes are in the submicron range (0.2 µm).

For SEM analysis, the zirconia disks were polished using fine diamond slurry (0.1 micron diamond size). After the initial polishing process the samples were thermally etched at a temperature of 1100^{o}C for 20 minutes. Thermal etching reveals the grain boundaries, which are not well defined otherwise. SEM images showed that the grains were of submicron size (0.1 - 0.3 µm).

Indexation

Laue patterns collected for every spot on the sample were analyzed using the XMAS software. The sample surface was scanned with a 500 nm cross section beam over a rectangular grid of size 0.5 mm by 0.024 mm. Figure 4, lists the step-wise process of indexing an acquired Laue

pattern. The Laue pattern at every data point of the raster scan was input to the XMAS software. The raw image was in TIFF format and consisted of the CCD artifacts which were eliminated by a reference amorphous image subtraction followed by background subtraction. The background subtraction step also involved normalizing the intensities of the peaks. Input parameters to XMAS included the reference crystal lattice parameters, the energy range, sample to detector distance, detector tilt, and sample orientation parameters.



The crystal lattice parameters for the tetragonal zirconia phase were determined using powder diffraction of zirconia powder (using a Bruker D8 Diffractometer). Figure 4 illustrates the process of artifact removal and background subtraction. Red numbers indicate the indexed grains in Zirconia after an initial peak search based on the Lorentzian peak profile. Based on the initial parameters and reference crystal file, the software analyzes the peak positions indexes them and returns the orientations for the different grains. Following indexation, based on the deviation of the reflections from the expected position and the reference *d*-spacing, the strains are refined for each indexed grain in every image. The strain obtained from polychromatic diffraction (Laue diffraction) consists of the deviatoric components only (marked red in Eqn.).

$$\varepsilon_{ij} = \varepsilon = \begin{pmatrix} \varepsilon_{11} - \Delta/3 & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{12} & \varepsilon_{22} - \Delta/3 & \varepsilon_{23} \\ \varepsilon_{13} & \varepsilon_{23} & \varepsilon_{33} - \Delta/3 \end{pmatrix} + \begin{pmatrix} \Delta/3 & 0 & 0 \\ 0 & \Delta/3 & 0 \\ 0 & 0 & \Delta/3 \end{pmatrix}$$

$$\text{With } \Delta = \varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33}$$

Where, Δ is the hydrostatic strain.

A separate measurement using a monochromatic X-ray beam is required to determine the hydrostatic component. The output file of a particular analysis is contains a sequential list file consisting of the strain components, respective stress components, the resolved shear stress/strain values for every data point on the sample. Upon analyzing the sequential list, it was found that

every data point on the sample indexed 1 to 3 new grains. If the grain size was larger than the X-ray beam cross-section as well as the step size between exposures, then the same grain would index at neighboring locations, as in [3]. Here, the cross-sectional area of a grain was on the order of the area of the focused beam and the one micron step size was larger than the typical grain diameter (Figure 3). In addition, more diffracted intensity is available from the surface layer of grains where X-ray absorption is minimal. The deeper grains provide fewer, dimmer peaks making them more difficult to index.

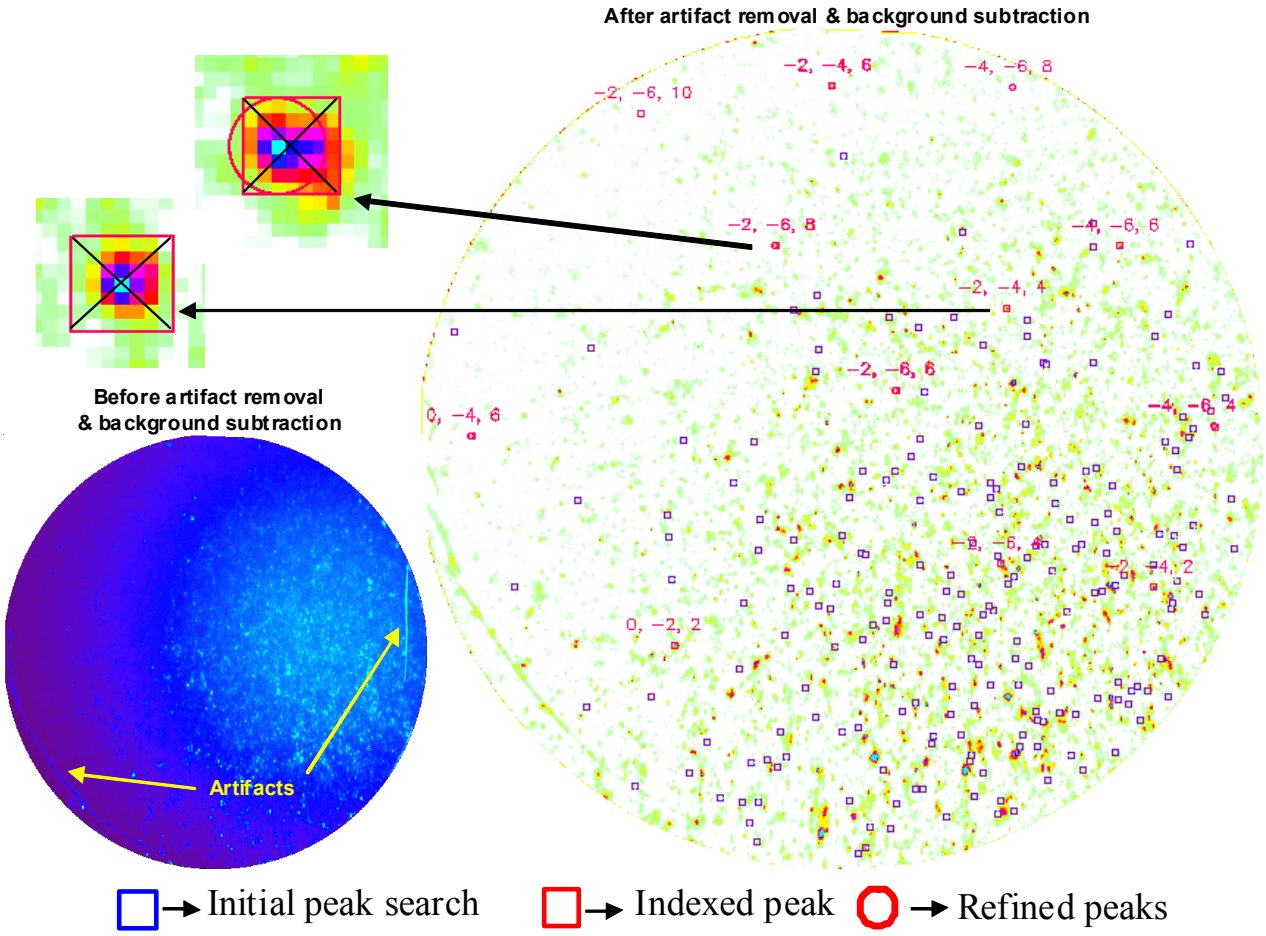


Figure 4 Indexed pattern of the tetragonal phase of zirconia. 10 peaks were utilized in determining the strain. The close up view of the indexed peak shows the peak search and fit. The symbol "□" indicates the brightest peaks found during the initial automated peak search. Based on the input reference crystal parameters, further peak search and fitting is carried out autonomously. Found peaks are marked by the symbol "□".

For any given indexation, a minimum of 6 peaks were chosen as a threshold. Based on the initial peaks and the found peaks, the entire pattern is indexed. As the number of peaks included in the strain refinement increases, so does the strain accuracy. Where here, strain accuracy is indicated

by observing physically relevant strains. Figure 4 illustrates an indexed pattern of the tetragonal zirconia phase. As seen in Figure 5, with less spots per indexation, the range of calculated strains increases.

Discussion

The analysis of Laue diffraction images leads to a sequential list of the refined strains. The distribution of the six deviatoric components of stress and strain, from the scanned area consisting of 6000 data points from a monolithic neat zirconia disk is presented in Figure 6. The output of the analysis consists of the strain components which are transformed to stress components using the stiffness tensor [4] for tetragonal zirconia. Results indicate an average stress between 47 and 75 MPa in compression depending on direction, along with smaller shear and tensile components.

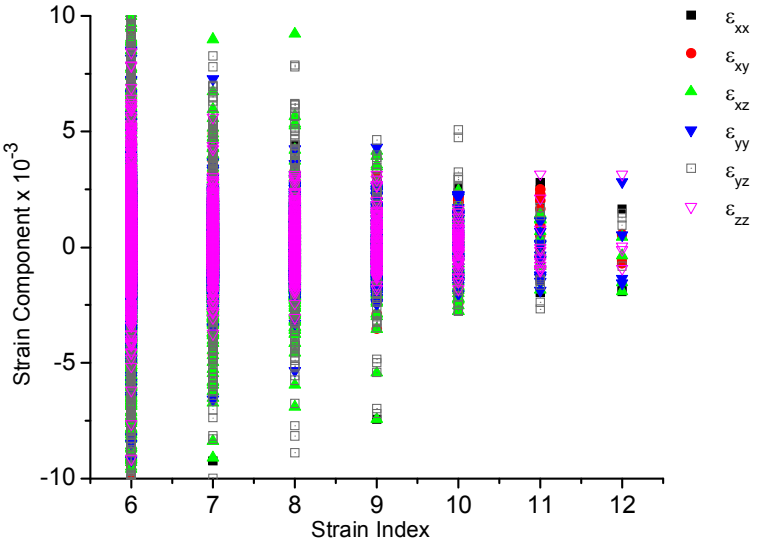


Figure 5 Illustration of Strain Magnitude and number of peaks. The accuracy of strains is low for less number of indexed peaks.

However, more than a few grains, between 9.7% (yz) and 0.2% (zz), reached stresses up to 1 GPa. Large compressive and tensile strains up to ±6000 με were observed. Such strains have been previously observed in graded alumina zirconia components, and zirconia disks.[5,6,7,8] The beam cross section, nominally 500 nm × 500 nm, provided 1-3 indexed grains. Typically only one grain per exposure had sufficient intensity for strain refinement.

Observed in the Laue pattern are a large number of spots, which (due to the small grain size) correspond to grains beneath the surface that give too few peaks to be indexed. It is possible

some monoclinic phase also contributes to the large number of peaks, but recent monochromatic measurements suggest less than 5% monoclinic phase is present. The present results were obtained by analyzing only the tetragonal phase for indexation due to the relatively brighter peaks and high symmetry. Indexation of the monoclinic phase would be more computationally intensive.

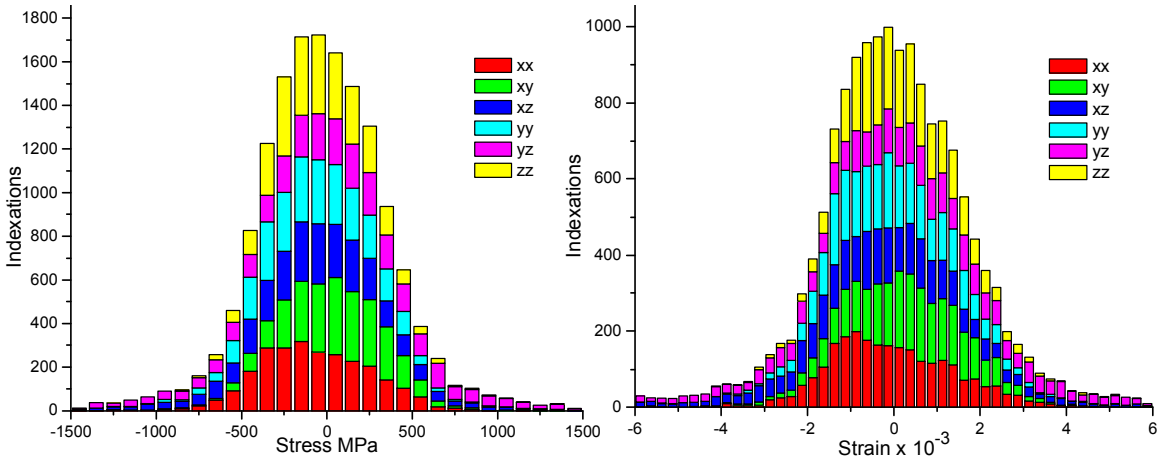


Figure 6 Left: Distribution of the stress components from the raster scanned data points. Maximum stresses of upto ±1 GPa were observed. Right: Distribution of the Strain components (100 MPa bins). Strains, up to 5000 με are observed (250 με bins).

Conclusion

Grain specific residual stresses were observed in many grains on a ceramic disk. These 2500 tetragonal zirconia grains typically ranged from 100 nm to 300 nm across. Laue patterns from a highly focused polychromatic X-ray beam were indexed and used to observe the deviatoric strains. On average, around 300 spots were observed per exposure for the zirconia sample. Typically, 1 to 3 grains could be indexed per exposure with an incident beam spot of 500 nm x 500 nm. Approximately 5% of the spots provided the highest 10% of intensities and belonged to the tetragonal phase. Large residual stresses exceeding – to + 1GPa were observed at some grains. Such high residual stress concentration may be detrimental to clinical performance where complex geometries along with an aggressive mechanical and chemical environment may synergistically contribute to clinical failures of zirconia all-ceramic dental crown systems.

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