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Determination of residual stress distributions in polycrystalline alumina using fluorescence microscopy

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ABSTRACT

Maps of residual stress distributions arising from anisotropic thermal expansion effects in a polycrystalline alumina are generated using fluorescence microscopy. The shifts of both the R1 and R2 ruby fluorescence lines of Cr in alumina are used to create maps with sub-μm resolution of either the local mean and shear stresses or local crystallographic *a*- and *c*-stresses in the material, with approximately ±1 MPa stress resolution. The use of single crystal control materials and explicit correction for temperature and composition effects on line shifts enabled determination of the absolute values and distributions of values of stresses. Temperature correction is shown to be critical in absolute stress determination. Experimental determinations of average stress parameters in the mapped structure are consistent with assumed equilibrium conditions and with integrated large-area measurements. Average crystallographic stresses of order hundreds of MPa are determined with characteristic distribution widths of tens of MPa. The stress distributions reflect contributions from individual clusters of stress in the structure; the cluster size is somewhat larger than the grain size. An example application of the use of stress maps is shown in the calculation of stress-intensity factors for fracture in the residual stress field.

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1. Introduction

Single crystal alumina (Al_2O_3, corundum) has a trigonal crystal structure that exhibits anisotropic thermal expansion: The coefficient of thermal expansion (CTE) is greater along the crystallographic *c*-axis than in the basal plane that contains the three *a*-axes perpendicular to the *c*-axis. [1]. As a consequence, on cooling an unconstrained corundum single crystal the *c*-axis contracts more than the *a*-axes (contraction is isotropic in the basal plane), leading to an anisotropic stress-free strain state. On cooling *polycrystalline* Al_2O_3 from a high temperature, however, the mutual constraint of neighboring unaligned grains impedes the free CTE strains for an individual grain and residual stress fields are developed in the polycrystalline microstructure in reaction to the constraint [2]. Considerations of mechanical equilibrium for a polycrystal in which the grains are randomly oriented show that the mean normal stress in the crystallographic *c*-direction is tensile, $\overline{\sigma}_c>0$, and that in the *a*-direction is compressive, $\overline{\sigma}_a<0$ [3]. Mechanical equilibrium requires the mean spherical stress in the microstructure be zero, $\overline{\sigma}_M = 0$, such that the mean normal stresses in the crystallographic frame have the relationship $\overline{\sigma}_c = -2\overline{\sigma}_a$. Estimates for the magnitudes of these stresses on cooling polycrystalline Al_2O_3 from typical sintering temperatures are approximately 100 MPa, although the value depends on grain size, grain shape, the presence of other phases, and cooling rate [2,4–7].

The nature of the microstructurally-driven residual stress field significantly affects the mechanical properties, especially fracture, of polycrystalline Al_2O_3. At the smallest length scale, residual tensile stresses can be relieved in the structure on cooling through the formation of spontaneous "microcracks"; these are typically localized to fracture of a single grain boundary facet or grain, and occur most frequently in large-grained materials [7–10]. Such microcracks form preferential locations for material removal under erosion or wear conditions and form strength-limiting flaws in structural applications. At an intermediate scale, sharp surface contacts generate localized plastic deformation zones in the material with attendant tensile stress fields that initiate and stabilize cracks, typically a few grain diameters in length [11]. The microstructural residual stress field can significantly impede or assist in the formation of such contact flaws, leading to significant strength degradation and potential material removal [9,12]. At the largest length scale, long cracks traversing many grains in polycrystalline ceramics are often observed to have microstructural elements behind the crack tip acting to restrain crack opening and thus impede crack propagation [13,14]. Such elements are particularly pervasive in Al_2O_3 and typically consist of either ligamentary bridges of a few grains formed by discontinuous crack propagation or frictional interlocks at a single grain boundary facet perpendicular to the predominant crack propagation direction [15]. In both cases, the local microstructural residual stress field is critical to the formation and subsequent deformation of the

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restraining elements [15,16]. Such elements lead to significant increases in the toughness of the material with increases in crack length [9,12,15].

As a consequence of the above effects, knowledge of the microstructurally-driven residual stress field is critical in materials selection and design considerations for polycrystalline Al_2O_3. If the magnitude of the residual stress field is too large, the material will be prone to microcracking and hence would not be a good choice for applications in which resistance to small cracks is important, *e.g.*, a wear-resistant coating. If the magnitude of the residual stress field is too small, the material will not exhibit much microstructural toughening and hence would not be a good choice for applications in which resistance to large cracks is important, *e.g.*, a hermetic feed-through. If the distribution of the residual stress field is too heterogeneous, the material will exhibit variable crack initiation response at sharp contacts and hence would not be a good choice for applications in which strength predictability is important, *e.g.*, a radar window. Hence, in order to optimize fabrication methods for polycrystalline Al_2O_3 so as to generate microstructures appropriate for specific applications, and to predict how a given microstructure will perform, a method is required that can measure and map polycrystalline Al_2O_3 residual stress distributions.

A method with sufficient spatial and stress resolution for mapping microstructure-related stress distributions in Al_2O_3 is fluorescence microscopy. The method is based on the shift in the fluorescence bands associated with Cr, a ubiquitous substitutional impurity for Al in Al_2O_3 [17]. The crystal field arising from the octahedral arrangement of O ions surrounding the Cr ions in Al_2O_3 leads to two closely-separated R1 and R2 bands that fluoresce at a wavelength of approximately 694 nm. It is this fluorescence that gives ruby (Al_2O_3:Cr) its characteristic red color. Applications of stress, changes in temperature, and different Cr compositions all distort the octahedra and associated crystal field and lead to shifts in the energies of the R1 and R2 fluorescence peaks. Early application of this phenomenon was the incorporation of calibrated ruby chips into diamond anvil cells for use as pressure gauges during high-pressure experiments [18]. Subsequent application was the measurement and mapping of stress in single-crystal and polycrystalline Al_2O_3, although works considering two-dimensional (2-D) mapping of microstructural residual stresses are few. A recent work [19] significantly extended spatial and stress resolution for 2-D Al_2O_3 stress mapping, in addition to introducing new stress-mapping methodologies. In that work, stress in a series of Cr-doped polycrystalline Al_2O_3 materials was mapped with sub-micrometer spatial resolution and about 10 MPa stress resolution. The work used the intensities and shifts of the R1 peak to define grain boundaries in the microstructures and generate σ_c stress maps, respectively. Scales for the stress maps were determined from the R1 peak-shift distributions and assumed Gaussian stress distributions. The work [19] provides a brief but comprehensive review of application of Cr fluorescence shift to stress measurement, with an emphasis on 2-D mapping.

Here, stress mapping in polycrystalline Al_2O_3 is further extended with attention focused on stress distribution determination. The determination includes four new and important experimental and analytical features: First, the determination is accomplished by using information from *both the R1 and R2* peak shifts, significantly increasing the precision of the stress determination. Second, stress determination includes explicit *correction for temperature* changes during mapping, significantly increasing the accuracy of the stress determination—in fact, it will be shown that temperature correction is critical to obtaining absolute estimates of stress. Third, *no particular form for the stress distributions* will be assumed. Fourth, the maps will *divide the stress in two ways*, providing both local spherical and shear, as well as *a*- and *c*-direction, stress distributions. As before [19], stress determination will involve a correction for the effect of Cr composition on peak shift. In addition to generating information regarding residual stress distributions, the measurements will enable an experimental test of the equalities given above for the equilibrium mean stress values: $\overline{\sigma}_M = 0$ and $\overline{\sigma}_c = -2\overline{\sigma}_a$. The next section develops the piezospectroscopic analysis required to convert R1 and R2 fluorescence peak shifts into stress maps and distributions, including comparisons with previously used large-area scans. Also included is a brief description of the fracture mechanics analysis employed in an example application of stress mapping in predicting the behavior of microcracks. This is followed by a description of the experimental methods, which are similar to those of the previous work. The results are presented first as maps and histograms of fluorescence intensity and shifts, and then as 2-D stress maps and histograms of stress distributions. Discussion centers on comparison of the results obtained here with those obtained in prior fluorescence-based residual stress studies of Al_2O_3, the likely next steps in advancing the quantification of residual stress mapping using fluorescence, and comparison of the fluorescence mapping technique with techniques used to assess residual stress in other material systems.

2. Analysis

2.1. Stress optical analysis for mapping

The energies, ν, (in wavenumbers, cm^{-1}) of the R1 and R2 fluorescence lines in Al_2O_3:Cr are given by [20–23]

$$\nu^{(1)} = \nu_0^{(1)} + \Delta\nu^{(1)} + \Delta\nu_T^{(1)} + \Delta\nu_C^{(1)} \tag{1a}$$

$$\nu^{(2)} = \nu_0^{(2)} + \Delta\nu^{(2)} + \Delta\nu_T^{(2)} + \Delta\nu_C^{(2)}, \tag{1b}$$

where the superscripts $^{(1)}$ and $^{(2)}$ here and throughout indicate parameters associated with the R1 and R2 lines, respectively. The ν_0 values are the energies or line positions of a reference material in the unstressed state at a reference temperature (here taken to be 298.8 K) with negligible Cr composition (here taken to be undoped sapphire). Fig. 1 shows example spectra taken from single (pixel) locations on a sapphire reference and a polycrystalline Al_2O_3:Cr material. The ν_0 values of the R1 and R2 lines of the sapphire are indicated, as are the total shifts, $\Delta\nu + \Delta\nu_T + \Delta\nu_C$, for the polycrystalline material. The shifts in line positions associated with changes in temperature and composition, $\Delta\nu_T$ and $\Delta\nu_C$ respectively, will be considered below. Here attention is first focused on the shift associated with stress, $\Delta\nu$. In hyperspectral fluorescence mapping, a spectrum of fluorescence intensity as a function of wavenumber, $I(\nu)$, similar to that in Fig. 1, is obtained at every point in a map. The spectra are then analyzed to obtain $\Delta\nu$ at every point in the map, such that stress maps can be generated.

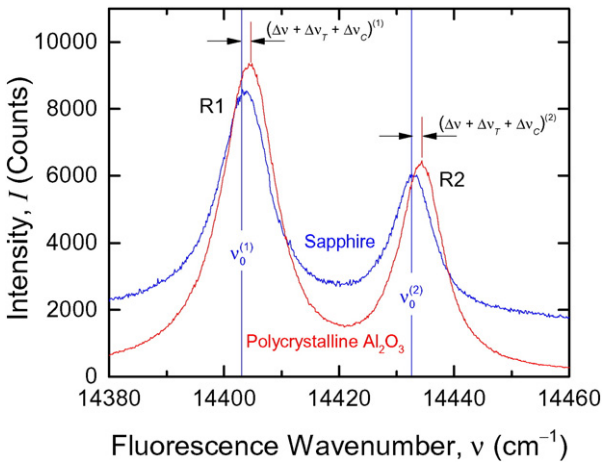

Fig. 1. Single location fluorescence spectra of single-crystal sapphire and a polycrystalline Al_2O_3 material, showing the R1 and R2 "ruby" lines and the shift of these lines in the polycrystalline material due to residual stress, temperature, and compositional changes. For easy comparison the polycrystalline Al_2O_3 spectrum has been reduced to 75% intensity.