

## Automatic Crystal Size Determination in the Micrometer Range from Spotty X-Ray Diffraction Rings of Powder Samples

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Crystal sizes can be calculated from peak intensities of spotty diffraction rings produced by a polycrystalline sample. Such patterns are collected using a small X-ray beam and an area detector. Peak intensities can be automatically measured using specially designed software. Crystal sizes can be determined from peak intensities after calibration using samples of the same material whose sizes are already known. This technique is independent of the aggregation state of the material. Also, crystal sizes of different mineral phases present in a sample can be analyzed independently. The present paper analyzes the potential of this methodology as applied to crystal size characterization of crystalline powder materials. Graded SiC and  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> abrasive powders were selected as test material for this study. A high correlation was found between parameters determined by X-ray diffraction and crystal size determined by means of optical microscopy and laser diffraction. The crystal sizes determined ranges from 3 to 80  $\mu$ m, a much larger range than that obtained by conventional X-ray line-broadening analyses. Additionally, upper and lower size ranges of applicability of this technique can be extended further by using different collimator diameters. The estimated error in crystal size measurements was within 5%.

### I. Introduction

CERAMICS properties (i.e., mechanical, magnetic, superconducting) are considerably dependent on their crystal size and orientation.<sup>1,2</sup> Thus, grain size characterization and selection is one of the first steps of ceramic preparation. There are many analytical techniques available for grain size characterization (i.e., optical microscopy, sieving, sedimentation, light diffraction).<sup>3</sup> The range of applicability and the time for analyses vary considerably among different techniques. This paper revisits and modifies a classical but unexploited methodology,<sup>4–6</sup> which, based on X-ray diffraction (XRD), provides valuable information regarding crystal sizes and that is independent of sample aggregation state. It can be applied to powder or fully dense ceramics. Furthermore, unlike other techniques, it does not require highly dispersed samples for analyses.

The diffraction pattern of a powder sample is composed of concentric Debye–Scherrer rings. These rings result from the superposition of a few spots from each illuminated crystal that is oriented in the Bragg condition. When the size of the X-ray beam is much greater than the crystal size (several orders of magnitude), a diffraction pattern constituted by continuous rings is formed. When the beam size is of the same or a few orders greater than the crystal size, fewer crystals are illuminated and spotty diffraction rings form. Crystal size estimation has

been performed in the past by manually counting the number of spots or by measuring spot dimensions in a Debye ring collected using photographic films.<sup>4–8</sup> However, this method has not been extensively used as its accuracy was insufficient and it required extreme care in sample preparation.<sup>9,10</sup> Now, the advent of recent technology (X-ray diffractometers equipped with CCD area detectors) has enabled the collection of much more precise information of the diffraction pattern than was previously obtainable using photographic films.<sup>11</sup> A new method derived from the former one is proposed here, which uses the intensity of reflection peaks in a Debye–Scherrer ring instead of the number of reflection spots in the ring to determine the size of the crystals. Note that the intensity of individual reflections in a ring can vary between several orders of magnitude (from 0 to 200 000 counts), while the number of reflection spots in a ring vary only within a few orders of magnitude (from 0 to 200). The intensity of reflections is therefore a much more sensitive parameter than the number of reflection spots. Thus, using this new methodology, crystal size can be automatically and precisely determined. In a sister paper, this methodology was also successfully applied to determine the crystal sizes in fully dense ceramics.<sup>12</sup> Now, it is applied to powder samples. Specifically, a set of abrasive samples (SiC and  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>) of known sizes has been used to determine the range of applicability and the precision of this technique. Sample size information determined from XRD has been compared with data obtained by optical microscopy and light diffraction techniques. It is thus demonstrated that the XRD methodology is independent of sample aggregation state and that it can be applied either to powder or fully dense ceramics.

### II. Materials and Methods

#### (1) Powder Samples

Factory-graded brown-alumina (B-Al<sub>2</sub>O<sub>3</sub>), micro-alumina ( $\mu$ -Al<sub>2</sub>O<sub>3</sub>), and silicon carbide (SiC) abrasive powders, with ASTM grit sizes ranging from 120 to 1200 numbers (100  $\mu$ m down to 3  $\mu$ m), were provided by K. C. Abrasive Company (Kansas City, KS).

#### (2) Crystal Size Characterization

(A) *Optical Microscopy and Light Diffraction:* Crystal size distribution of powder samples with sizes ranging from 3 to 40  $\mu$ m was determined by light diffraction (LD) using a Multi-sizer 3 (Beckman Coulter Inc., Fullerton, CA). Crystal sizes of coarser samples were determined using optical microscopy (SZ, Olympus, Tokyo, Japan) previously by dispersing a small amount of powder over a glass slide. Size measurements were performed by image analysis using ImageTool software (National Institutes of Health, Bethesda, MD). Crystal diameter was determined by averaging lateral size of crystals in pictures taken at three different locations within each sample. Abrasive crystals studied are equiaxial and their volumes can be calculated as that of a sphere  $\frac{4}{3}\pi(D/2)^3$ , where  $D$  is the lateral size of crystals.

(B) *X-Ray Diffraction:* Powder samples were consolidated with polyester resin. Approximately, 1 g of powder was

V. A. Hackley—contributing editor

Manuscript No. 20767. Received July 13, 2005; approved January 30, 2006.

This study was funded by Grant REN2003-07375, Research Group RNM 179 of Junta de Andalucía, and the Programa Ramon y Cajal (Spain).

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