Scripta Materialia 109 (2015) 84–88

Contents lists available at ScienceDirect

Scripta Materialia

journal homepage: www.elsevier.com/locate/scriptamat

Separating macrostresses from microstresses in Al_2O_3–15 vol%SiC particulate reinforced composites

Ji Zou [a,*], Yuan Zhong [a], Jun-Zhan Zhang [a,c], Magnus Ekelund [b], Zhi-Jian Shen [a,*]

[a] Department of Materials and Environmental Chemistry, Arrhenius Laboratory, Stockholm University, S-106 91 Stockholm, Sweden
[b] AB Sandvik Coromant, Lerkrogsvägen 19, 126 80 Stockholm, Sweden
[c] College of Materials and Mineral Resources, Xi'an University of Architecture and Technology, 13 Yanta Road, Xi'an 710055, PR China

ARTICLE INFO

Article history:
Received 16 June 2015
Revised 19 July 2015
Accepted 20 July 2015
Available online 25 July 2015

Keywords:
Particulate reinforced composites
Residual stresses
Spark plasma sintering
Raman spectroscopy
Argon ion beam polishing

ABSTRACT

Macrostresses and microstresses coexist in ceramics, but are difficult to distinguish. In this work, these two types of residual stresses were separated by comparing the stress levels on the mechanically and argon beam polished surfaces of Al_2O_3–15 vol%SiC particulate reinforced composites. Stresses were measured using the shift of a Raman peak of β-SiC, the shift of a luminescence line of α-Al_2O_3 and a self-consistent approach. The critical grain size of SiC, over which will result in the crack formation in Al_2O_3 matrix, was calculated to be 13.5–26.4 μm. New microcracking did not form in Al_2O_3 grains during progressive mechanical polishing.

Residual stresses occur in the manufactured or sintered ceramics due to the inherent properties of the phases or the processing methods employed. These residuals are separated into two types depending on their effective areas, i.e. macrostresses and microstresses. The former normally develop over relatively large scales, much bigger than the grain size in the specimen. In contrast, microstresses vary among individual grains or even within in a single grain. For ceramic particulate composites, macrostresses generally develop near the outer surface of the ceramics, and can form due to surface finishing processes such as cutting, grinding or polishing that produce stress differences between the surface and interior [1]. Microstresses generally accumulate inside ceramic bodies during processing as a result of a mismatch in the coefficients of thermal expansions and/or elastic properties between the ceramic matrix and isolated reinforcing particles [2].

In early 1990s, high strength Al_2O_3–SiC particulate (SiC_p) reinforced composites nanocomposites (AS_p) were developed and commercialized as cutting tools for casting iron and nickel based alloys [3,4]. Addition of 5 vol% nano-sized SiC particles improved the bending strength from ~350 MPa for monolithic Al_2O_3 ceramics to ~1 GPa for composites and as high as 1.5 GPa for annealed composites [3,5]. One explanation for the high strength is the residual compressive stress. The stress was considered to be generated during the diamond grinding of Al_2O_3–SiC_p composites. An extra heat treatment could help the crack healing on the tensile surface of the specimen, but only partially relieve the compressive stress in the ground layers. The crack propagation was inhibited by the compressive stress in the matrix. Accordingly, the strength of annealed Al_2O_3–SiC_p composites was improved [3,5]. Another mechanism has been proposed simply relies on the experimental observation: the incorporation of nano SiC grains make pulling out of Al_2O_3 grain more difficult. Therefore, the defect density on the ground surface of AS_p was reduced and its surface finishing quality was enhanced [6,7]. The extraordinary high strength of Al_2O_3–SiC_p nanocomposites is still a controversial topic, but clearly separating macro- and microstresses could provide insight into the strengthening mechanism [7–9]. One method to determine the intrinsic microstress value is polishing machined surfaces with progressively finer media. Nevertheless, stresses measured on free surfaces are normally different than those measured for bulk ceramics, which could be due to relaxation of the microstresses or incomplete relaxation of macrostresses during polishing [10].

Ion beam milling provides an opportunity to produce surfaces that are nearly free of artifacts and distortions produced by conventional cutting and grinding processes [11]. Therefore, ion beam milling may be a method to minimize residual surface stresses without annealing. The aim of this study is to measure residual stresses on the surfaces of AS_p prepared by argon ion beam

* Corresponding authors at: School of Metallurgy and Materials, University of Birmingham, B15 2TT Birmingham, UK (J. Zou).
E-mail addresses: zouji1983@gmail.com (J. Zou), shen@mmk.su.se (Z.-J. Shen).

http://dx.doi.org/10.1016/j.scriptamat.2015.07.026
1359-6462/

(ion-beam cross section polishing, CP) and conventional mechanical polishing (MP) to determine the criterion for its microcracking.

Granules of Al_2O_3 containing 15 vol%SiC_p produced by freeze granulation were provided by Sandvik Coromant. The starting β-SiC and α-Al_2O_3 powders had average particle size of ~100 nm. Compacts were densified by spark plasma sintering (Dr. Sinter 2050, SPS Syntex Inc., Kawasaki, Japan) under vacuum (~10 Pa). During sintering, temperature was monitored by a pyrometer focusing on a hole in the die body near the outer edge of the cylindrical samples. For sintering, granules were poured into a graphite die with an inner diameter of 20 mm and then a uniaxial pressure of 75 MPa was maintained during the cycle. Powders were first heated to 600 °C in 3 min and then the temperature was continuously increased to 1450 °C at 100 °C/min. After 5 min at 1450 °C, the SPS power was shut off and the furnace cooled naturally, which was an average rate of ~300 °C/min from 1450 °C to 600 °C.

For MP, specimens were polished with successively finer diamond abrasives ranging in diameter from 15 μm to 100 nm. Another batch was prepared by argon CP (SM9000, JEOL, Japan), which is shown schematically in Fig. 1a. The ion beam impinged on the edge of a polished surface, producing a semicircular flat surface on the cross section as described elsewhere [11]. For the present study, AS_p was polished using an accelerating voltage of 5 kV for 12 h. By repeating the aforementioned procedures, several regions with maximum depth of ~160 μm were obtained by CP.

The Raman shifts for SiC and the spectroscopic measurements of Al_2O_3 phases were conducted by a micro confocal Raman spectrometer (HR 800, LabRAM, France) equipped with an air cooled Nd:YAG laser (532 nm/50 mW). Different filters were adopted to reduce the incident intensity and the laser spot size on the polished surface. All the patterns were acquired from at least ten random areas on polished surfaces. For a specified area, the measurement was repeated at least ten times. Raman peak positions and intensities (Fig. 2a) were determined by fitting these peaks to a Lorentzian function. For the fluorescence spectra, deconvolution (Fig. 2b) was completed using a mixed Lorentzian (R1 and R2) and Gaussian function (R1′ and R2′) [12]. The exposure times for Raman and fluorescence measurements were 20 s and 5 s, respectively. A higher laser power and longer exposure time were needed for collecting Raman spectra to produce an acceptable signal to noise ratio. Different exposure times were examined for a laser power of 0.5 W to assess thermal effects on peak positions, but none were noted for exposure times up to 5 min. The apparatus was calibrated using the 520.7 cm^{-1} line from a silicon wafer before each measurement.

A typical area after argon ion beam polishing is shown in Fig. 1b. Benefiting from a gentle flow of ionized argon gas, most of the individual Al_2O_3 and SiC grains could be directly recognized, without any etching process. They were distinguished by the differences in channeling or charge contrast. As we can see from Fig. 1c, no apparent pores were detected in AS_p. Density measurement also confirmed a high relative density (98.5%) was reached. In Fig. 1d, SiC grain segregations were obvious, however, they are well dispersed in the Al_2O_3 matrix (Fig. 1c), with a maximum agglomeration dimension of 1 μm. The average size of Al_2O_3 and primary SiC grains in AS_p is around 250 nm and 150 nm, respectively.

Natural concentration of Cr^{3+}, as impurity, always exists within α-Al_2O_3 lattice. When the Al_2O_3 containing ceramics were irritated by a laser, apart from seven Raman active phonon modes ($2A_{1g} + 5E_g$) of α-Al_2O_3 (Fig. 2a), the lines from Cr^{3+} fluorescence (R1 and R2 peaks, Fig. 2b) could be clearly identified as well. Assuming an unstressed material showing a band centered at ν_0 (cm^{-1}), its peak shift will occur when it is subject to a level of stresses. The full relationship between stress level and peak shift could be described by a Taylor's expansion, this regulation works in both Raman and fluorescence peaks. If a higher order term in Taylor's expansion is neglected, simple equations to get residual stress values could be obtained by inputting the peak shift data. This hypothesis is valid since the higher derivatives in Taylor's expansion are always much smaller than the first two [13]. According to this principle, the relationship between Raman peak shift (α-Al_2O_3 [14] and β-SiC [15], ω in cm^{-1}) or fluorescence

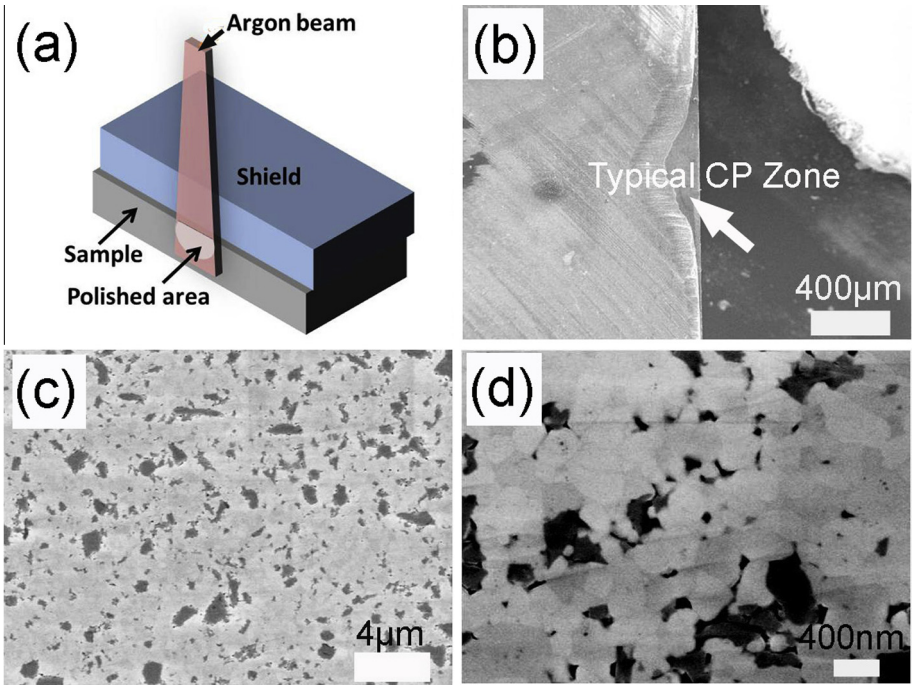

Fig. 1. Schematic process for a typical ion-beam cross section polishing (CP) (a) and SEM images of cross section polished Al_2O_3–15 vol%SiC_p composites. (b) A typical argon beam polishing zone; (c) and (d) are the detail microstructures in AS_p with different magnifications. Note that the grain size of Al_2O_3 (gray color) and SiC (black color) could be directly observed after CP without any etching (d). All the SEM images were collected by secondary electrons under an accelerating voltage of 5 kV and a working distance of 9 mm.