Translucent Y₃Al₅O₁₂ ceramics : electron microscopy characterization

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TRANSLUCENT $Y_3Al_5O_{12}$ CERAMICS: ELECTRON MICROSCOPY CHARACTERIZATION

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TEM observations of ceramic YAG samples doped with SiO_2 or MgO are reported. In both cases a grain size of a few micrometers has been observed. Neither grain boundary phases nor second phases at triple junctions could be detected in both type of ceramics within the lattice resolution of the samples (better than 0.5 nm). Furthermore, aluminium rich inclusions occurred frequently.

1. INTRODUCTION

The most well-known technical ceramic is probably Al_2O_3 . This material can be sintered to translucency if the powder is doped with a slight amount of MgO (see e.g. ref. 1). The various opinions concerning the dopant behaviour presented in the literature seem to converge towards a $MgAl_2O_4$ second phase in the triple points (at high dopant levels) and to a slight enrichment of Mg at the grain boundaries, probably as nonstoichiometric $MgAl_2O_4$ at discrete spots (see ref. 2, for a recent review and also the introduction of ref. 3). Recently, also the sintering of $Y_3Al_5O_{12}$ (YAG) powder to translucency was reported⁴ using either SiO_2 or MgO as a dopant. For this ceramic, similar questions concerning the dopant behaviour could be raised. As a first step towards the solution of this problem, a transmission electron microscopic (TEM) study on the grain boundary structure was done.

2. EXPERIMENTAL

The materials were prepared using the wet-chemical route⁴. Various dopant levels for SiO_2 and MgO were used. Sintering was done in a tungsten vacuum furnace at about 0.001 Pa at temperatures between 1700 and 1800 °C. The microstructure of the various ceramics was revealed by scanning electron microscopy (SEM)

either using the as-fired surfaces or fracture surfaces. In both cases the surfaces were covered with a thin gold layer to prevent electrostatic charging during examination.

For observation in the transmission electron microscope (TEM) the samples must be transparent for electrons (typically 100 nm for 100 kV electrons). Discs, 3mm in diameter and 200 μm thick are obtained from bulk material. Conventional grinding and polishing techniques were used for the SiO_2 doped ceramics to reduce the thickness to about 30 μm . For the ceramic materials under study this lapping is rather time consuming (typically several weeks). Therefore, the newly introduced sphere-on-tape method⁵ was employed to thin the MgO doped YAG discs. Using an instrumentation recorder with half-inch lapping tape (0.3 μm Al_2O_3 particles) loaded on the reverse side with a ruby sphere (diameter 4 mm) about 100 μm material can be eroded from one side of the sample in less than about half a day. By inverting the sample a second hole can be made on the reverse side of the disc. A displacement probe continuously monitors the depth of the hole. Finally, the discs are ion-etched on both sides in a single-specimen, two-beam ion mill at a glancing angle of about 10°. In addition to the advantage of rapid thinning with the sphere-on-tape method, these type of samples are also easier to handle than the conventionally

FIGURE 1

As-fired surfaces of YAG (1200 wt ppm SiO_2) ceramics (fig. 1a) and YAG (500 wt ppm MgO) ceramics (fig. 1b) (bar = 10 μm).

lapped ones. The discs are 3 mm in diameter and 200 μm thick with in the middle an area of about 1 mm in diameter with a gradual change in thickness; after carbon coating the edge of the 50- μm -diameter hole can be studied by TEM.

Observations were made with a Philips EM400T transmission electron microscope combining double tilt specimen handling with a line resolu-

tion down to 0.2 nm. For elemental analysis an X-ray detector (EDAX) at a take-off angle of 20° was used.

3. CHARACTERIZATION

The microstructure of the YAG ceramics is shown in fig. 1. In these SEM photographs the as-fired surfaces of the sintered samples are shown. For both the SiO_2 and the MgO dopant a regular microstructure with an average grain size of a few micrometers is observed. A similar impression is obtained from TEM observations.

Fig. 2a shows a bright-field image of an SiO_2 doped ceramic. Boundaries between the grains are visible as almost straight dark lines. If the plane of a grain boundary is not precisely parallel to the incident electron beam, so-called thickness fringes are observed at the grain boundaries. In fig. 2b a corresponding dark-field image is shown. SEM/EDAX observations further showed that Al-rich inclusions are frequently present (fig. 3). These inclusions were also examined in the TEM (fig. 4); EDAX analysis showed that Y (and Si) are almost ab-

FIGURE 2

Bright-field (fig. 2a; bar = 2 μm) and dark-field (fig. 2b) image of polycrystalline YAG (1200 wt ppm SiO_2).

FIGURE 3

Scanning electron micrograph (fig. 3a; bar = 10 μm) and corresponding X-ray mapping image (Al K α ; fig. 3b) of YAG (1200 wt ppm SiO_2).

sent in the inclusion. Point-count analysis of several micrographs and X-ray diffraction of various samples revealed that on average about 0.5 vol% of inclusions is present but that in some cases values up to about 15 vol% were encountered. Since the specimens were prepared to be strictly stoichiometric, the appearance of

the inclusions is due to the sintering procedure.

In order to elucidate whether any intergranular phase is present, TEM diffraction observations were made. From fig. 2 it can be seen that the grains are directly bonded to each other and that an intergranular phase is not present. To verify this, the lattices of two neighbouring grains were resolved. An image is formed based upon the transmitted incident beam and a single diffracted beam of each grain giving rise to fine fringes, e.g. modulations with the lattice spacing (fig. 5). Part of the electron diffraction pattern for both grains is shown in the inset of fig. 5.

The lattice fringes in the upper grain of fig. 5 correspond to the 211 reflection (spacing 0.489 nm). The spacing in the lower grain is twice that of the 321 reflection (spacing 0.321 nm).

From fig. 5 it can be seen that the lattice fringes practically meet at the interface of both grains. In addition, no amorphous contribution can be detected either in the diffraction pattern or in dark field at the grain boundary. The thickness of any intergranular film can not

FIGURE 4

Dark-field image (bar = 200 nm) of an Al-rich inclusion in a grain of YAG (1200 wt ppm SiO_2). Note the strain-induced contrast contour around the inclusion.

FIGURE 5

Interface boundary (bar = 10 nm) between two YAG grains (2000 wt ppm SiO_2). In the inset, part of the electron diffraction pattern of both grains is depicted.

FIGURE 6

Edge-on grain boundary (bar = 5 nm) in YAG (2000 wt ppm SiO_2). In the inset, the zero-order $\bar{1}11$ Laue zone for the lower grain is depicted.

be larger than 0.5 nm, therefore.

Another grain boundary of YAG(SiO_2) is shown in fig. 6. Both grains display distances of 0.424 nm (220 reflection). The lower grain is lattice resolved; the corresponding zero-order $\bar{1}11$ Laue zone is shown in the inset of fig. 6. The diffraction condition consists apart from the directly transmitted beam, of the six surrounding, equidistant diffracted beams. Again, the grains appear to be directly bonded to each other without an amorphous phase in between.

Similar observations of the structure at grain boundaries at various locations in the MgO-doped ceramics were made. Fig. 7 shows a typical example of a bright- and dark-field image pair of a few YAG(MgO) grains. The misorientation between some grains is less than 1° and so-called low-angle boundaries are observed, especially in the dark-field situation. In fig. 8 a junction between three grains is lattice resolved. The two grains at the right side of the picture clearly display two interpenetrating lattice fringe images. Both grains show the 220

reflection (0.424 nm); in addition, the $\bar{4}22$ reflection (0.245 nm) is present in the lower grain. It is seen that over the interface between the upper and left grain, Moiré fringes directed almost perpendicular to the grain boundary are present.

From our microstructural observations we conclude that within the lattice resolution of the samples (less than 0.5 nm) neither grain boundary phases nor second phases at triple junctions are present in SiO_2 - and MgO-doped YAG ceramics. In contrast to the present results on YAG ceramics for grain boundaries in translucent polycrystalline MgO-doped Al_2O_3 studied by TEM, Clarke⁶ showed that Ca is detectable (1 wt%) at three-grain junctions, whereas Mg is not (detection limit 1 wt%). At high MgO dopant levels, however, Mg is also present in the second phase at the triple junctions^{2,3}. Using weak-beam, conventional strong-beam and electron diffraction techniques Carter et al.⁷ revealed periodicities in the grain boundaries of MgO-doped Al_2O_3 . The thickness of the boundary

FIGURE 7

Bright-field (fig. 7a; bar = 1 μm) and dark-field (fig. 7b) image of polycrystalline YAG (500 wt ppm MgO). Several low-angle grain boundaries are observed.

layer was estimated as 6 nm. A significant difference in grain boundary structure between the garnet and alumina ceramic is thus observed.

4. CONCLUSION

Electron microscopic observations of YAG ceramics doped with different amounts of SiO_2 or MgO revealed no grain boundary phases or second

FIGURE 8

Lattice and Moiré fringes in three YAG grains (500 wt ppm MgO; bar = 10 nm).

phases at triple junctions, at least within the lattice resolution of the samples (better than 0.5 nm).

On the other hand, aluminium rich inclusions have been observed frequently. The influence of these inclusions on the elastic, hardness and fracture behaviour of various YAG ceramics will be published elsewhere⁸.

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