Solubility Limits of La and Y in Aluminum Oxynitride (AlON) at 1870°C

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Aluminum oxynitride (AlON) is a polycrystalline ceramic material with potential use in applications requiring high strength combined with optical transparency [1-3]. Due to its cubic spinel structure, polycrystalline AlON has isotropic optical and thermal properties, making it a candidate material to replace single crystal forms of oxides currently in use for optical applications

In order to achieve optical transparency full density is required, and as a result the sintering process for AlON usually includes elevated temperatures combined with pressure and/or long sintering durations. To overcome this difficulty and for controlling microstructural evolution, dopants are often introduced. The solubility of these elements is very low (assumed to be in the order of tens of ppm), which results in their enrichment to grain boundaries even at very low doping levels. Until recently, the precise measurement of solubility limits in ceramics has not been performed directly [4].

Since there is no data in literature regarding the solubility limits in AlON, we chose to develop the measurement method on Al_2O_3 and then apply it to AlON. Since Coble's work in the 1960's, the influence of Mg doping on alumina has been extensively studied. It is one of the major dopants for alumina, used to reduce the grain boundary mobility and growth rate during sintering [5,6]. La, Y and Mg are common dopant for AlON used to control the microstructure evolution [7,8].

The samples were prepared by ball milling the starting materials for 24 hours. Dopants were introduced as nitrates with a concentration well *above* the solubility limit. Green bodies were prepared by pressure filtration, and sintering was conducted at 1600ºC and 1870ºC for Al_2O_3 and AlON, respectively. X-Ray diffraction and energy dispersive spectroscopy measurements were performed to ensure the presence of secondary phases which indicates that the matrix grains are saturated (at the solubility limits).

The solubility of Mg was measured using a scanning electron microscope (SEM) combined with wavelength dispersive spectroscopy (WDS). The WDS measurements were conducted on polished samples, saturated with Mg, and rapidly cooled from the sintering temperature (1600°C). Prior to the measurements, the WDS working conditions were optimized in order to improve the detection limit and measurement precision. Since no etching was performed following mechanical polishing (to prevent changes in the near-surface chemistry), it was not possible to identify grain boundaries in the SEM confidently. To overcome this difficulty, a large number of measurements (>300) were performed at random locations on the cross section of the sample. The purpose was to obtain a distribution of the Mg concentration that can be divided into three main regions (see Figure 1). The first region (marked as A in Figure 1)

corresponds to the Mg concentration within the alumina grains, which is the *solubility limit* if secondary spinel particles exist in the equilibrated microstructure. Region C in Figure 1 corresponds to the Mg concentration of the spinel particles that reside within the interaction volume, and Mg from the alumina grain. Region B is an intermediate region, because of overlap of the electron probe (and interaction volume) between regions A and C, and because of the possible contribution of Mg segregated at grain boundaries. Figure 2 presents SEM fractographs of La-doped and Y-doped AlON samples which underwent sintering at 1870°C for 24 hours and were rapidly cooled. The secondary phase particles, whose presence confirms that the AlON grains are at the solubility limit, are brighter due to their higher BSE contrast. WDS analysis, based on the approach used to measure the solubility limit of Mg in Al_2O_3, resulted in solubility limits of La and Y in AlON at 1870°C to be 498±82 and 1775±128 ppm, respectively.

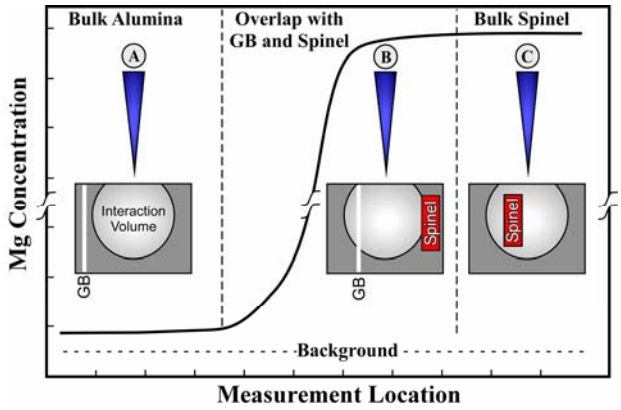

Figure 1: Schematic drawing of the Mg concentration distribution versus the measurement location.

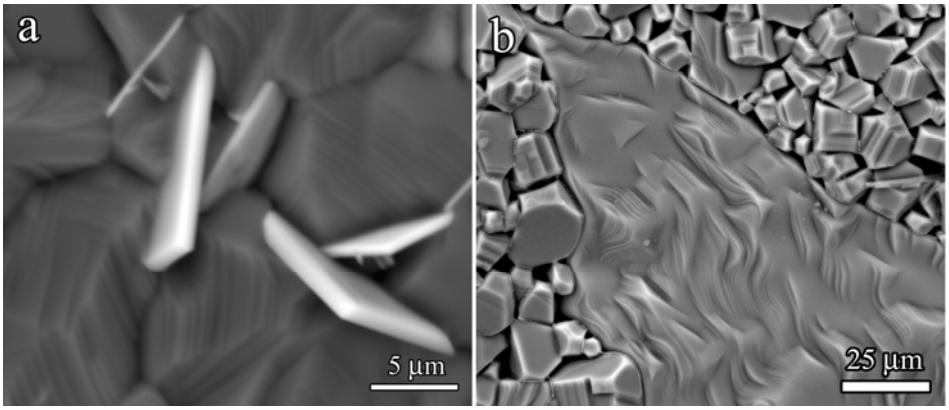

Figure 2: Backscattered electron (BSE) SEM fractographs of (a) La-doped AlON and (b) Y-doped AlON samples which underwent sintering at 1870°C for 24 hours and were rapidly cooled.

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