Structural properties of AlN crystals grown by physical vapor transport

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Recent major developments in GaN semiconductor technology have driven the interest towards aluminum nitride (AlN) and gallium nitride (GaN) bulk crystals, which are considered ideal substrates for III-nitride epitaxy. Up to now the potential of these materials in substrate applications has been disadvantaged by the lack of sufficiently large and perfect single crystals. In this context, preparation of AlN crystals by physical vapor transport (PVT) seems to be very promising. By sublimating AlN powder at about 2300°C and subsequent condensation of vapor species on the crucible lid we have achieved preparation of dense, polycrystalline material containing single-crystalline grains with up to 50 mm^3 in volume. Furthermore, self-nucleation of separate single crystals on crucible walls lead to freestanding single crystals with volumes of up to 100 mm^3. The grown material is single-phase wurtzite AlN containing about 100 ppm wt of oxygen; transition metals are present only in the sub-ppm range.

High structural quality of AlN crystals is considered a major prerequisite for substrate use. Thus we investigated the structural properties of the obtained material in order to compare structural properties of freestanding single crystals (grown at low temperature gradient, almost absent stress, but zonar structures) vs. bulky material (grown at higher temperature gradient by unidirectional crystallization, higher stress).

Freestanding single crystals demonstrate typical habit with mostly smooth facets and almost no macroscopic defects. Grains in the polycrystalline boule can vary significantly in their structural quality, and some of them show voids and Al inclusions as identified e.g. by EDX, which indicate that AlN decomposition at the growth interface is an important issue. Accordingly, bulk AlN phonon modes' FWHM taken by Raman spectroscopy, the position of X-ray diffraction peaks, and the reflectivity of the reststrahlenband in FTIR can vary significantly from one grain to another. However, best values for the FWHM of the (004) X-ray diffraction peak being below .025°, and phonon modes' FWHM of 5 1/cm prove that growth of high-quality AlN crystals is possible also at high temperature gradient, i.e. high growth rate.

Utilizing various X-ray techniques we found that polycrystalline boules are textured in c-axis direction. Single grains deviate from this orientation by 10.6° in average. Furthermore, etching in eutectic KOH/NaOH melt was used to identify the crystal polarity. In the case of freestanding single crystals, the mirror-like (0001)Al surface as well as adjacent rhombohedron facets are resistant to the etchant, whereas the more defective (0001)N surface is chemically very active. It shows up that the vast majority of grains in a polycrystalline boule are aligned along the (0001)Al direction, i.e. the growth interface is normally formed by Al-facets and adjacent rhombohedron faces. Those grains which are aligned along (0001)N direction, however, are defective, thus leading to the different appearance and structural quality of the grains in polycrystalline boules.