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PS-15.02.02 MASS TRANSPORT EFFECT WITHIN THE SOLID-LIQUID INTERFACE BOUNDARY LAYERS IN SOLUTION CRYSTAL GROWTH. By Yu Xiling*, Liu Youchen and Yue Xuefeng, Institute of Crystal Materials, Shandong University, Jinan 2510100, P.R. China

In kinetics theories of high temperature solution growth, it is considered that the mass transport within the solid-liquid interface boundary layer is a diffusion process only, and that the concentration gradient distribution in this layer is homogeneous. There is, however, a lack of experimental evidence of this fact.

Using holographic phase-contrast interferometric microphotography, we have carried out real-time recording of the dynamic processes which take place during the crystal growth of KTP from high-temperature solutions and of KDP from low-temperature solutions. The relation between the boundary layer thickness at different places on the same crystal face and the depth of the crystal soaked in the solution, the relation between the boundary layer thickness and supersaturation, the concentration distribution and the refractive index gradient distribution within the boundary layers have all been investigated in detail. The experiments demonstrate that the mass transport process within the solid-liquid interface boundary layers is the result of the coupled effects of diffusion and convection, no matter whether it is high-temperature or low-temperature solution growth. The distribution of the concentration gradient in the layers are exponential functions of the position under the free convection state.

PS-15.02.03 INVESTIGATION ON GROWTH AND SOME PROPERTIES OF POTASSIUM TANTALATE CRYSTALS. By Meng Xianlin*, Sun Youxuan, Zhu Li, Wang Ruihua, Wei Jingqian, Wang Jiyang and Lu Yaogang. Institute of Crystal Materials, Shandong University, Jinan 250100, China.

Potassium tantalate ($KTaO_3$, KT) single crystals with a stable cubic structure have a wide range of applications in LASER beam modulators, digital light deflectors and semiconducting devices. These crystals are also a promising substrate material because they exhibit no phase transition in a temperature range from 0° K to the melting point and have good lattice matching. Previously, KT crystals have been grown either by the slow-cooling method (D. Rytz, *J. Cryst. Growth*, **59**, 1982, 468), or a variation of the Kyropoulos Technique (W.A. Bonner *et al.*, *Am. Ceram. Soc. Bull.*, **44**, 1965, 9) but the crystal growth rate was very slow (0.03 to 0.14 mm/h). In our present wotk, high quality KT single crystals (36*35*16 mm^3 in size and 128 g in weight) have been grown by using the Czochralski technique and the growth rate is 10 times faster than that in the above mentioned two methods. The transmission spectrum of as-grown KT crystals was determined and the result indicates that the crystal is transparent in the band range from 0.4 to 2.6 μm. The morphology of the crystals were studied. It is observed that the growth morphology is mainly defined by {100} and {110} faces. The lattice parameter, a, and the density of the crystal, ρ, were determined: a = 3.988 Å, ρ = 7.020 * 10^3 Kgm^{-3}. The value of the density of KT crystals grown by the Czochralski technique is the closest to the theoretical value (7.018 * 10^3 Kgm^{-3}). This indicates that the crystals grown by the Czochralski technique have a better crystalline perfection than those grown by other methods. The dielectric constant, ε, of KT has been measured. We have found that, at lower temperatures (- 200 to 0°C), ε decreases rapidly with increasing temperature, whereas the rate of decrease becomes slower at higher temperatures (0 to 400 °C).

PS-15.02.04 CRYSTAL MORPHOLOGY OF YIG GROWN IN HIGH TEMPERATURE SOLUTIONS. H. M. Park, K. I. Seo and S. J. Chung*, Department of Inorganic Materials Engineering, College of Engineering, Seoul National University, San 56-1, Shinlim-Dong, Kwanak-Gu, Seoul, 151-742, Korea

Single crystals of YIG were grown in high temperature solutions of various contents of $BaO-xB_2O_3-(0.62-x)BaF_2$ ($0.21 \leq x \leq 0.62$) by top seeded solution growth. To investigate the effects of growth conditions on the anisotropic growth rate, slow cooling method as well as temperature gradient method were used and the viscosities of high temperature solutions were measured. By the slow cooling method, the solutions were slowly cooled at various cooling rates. By the temperature gradient method, nutrient of stoichiometric YIG composition was added to the solution at different constant temperatures. The grown crystals were up to 2.5 cm in size, and the crystal morphology was a combination of {211} and {220}. The relative development of the two faces depends on the growth rate in the direction perpendicular to each face. The relative growth rate can be calculated from the ratio of widths of the two faces. In the solution of increased BaF_2 contents and higher viscosity at higher temperatures or slower cooling rate, face {220} is dominantly developed, while in reversed conditions face {211} is dorminant. The morphology of grown crystal can be easily estimated if the growth temperature, cooling rate and the solution composition are known. X-ray topographic investigation and TEM observation on the grown crystals were carried out.

PS-15.02.05 CRYSTAL GROWTH OF $NdAl_3(BO_3)_4$ IN HIGH TEMPERATURE SOLUTIONS AND MORPHOLOGICAL INVESTIGATIONS. By S. T. Jung*, D. Y. Choi, S. J. Chung, Department of Inorganic Materials Engineering, Seoul National University, Seoul, 151-742, Korea.

$NdAl_3(BO_3)_4$ single crystals were grown from high temperature solutions by slow cooling method and top seeded solution growth technique. Fluorides such as BaF_2 or KF were gradually added to the flux of $BaO-B_2O_3$ or modified K_2O-MoO_3 respectively. As the amount of fluoride increased, increase of growth velocity, change of morphology, and contraction of the primary crystallization region in phase diagram were observed. When using modified K_2O-MoO_3 flux and the molar ratio of KF/K_2O was over 0.7, NAB rarely formed. The crystals grown using 50 cc Pt crucible were up to 10x10x20 mm^3 in size.

The lattice parameters and space group of grown crystal confirmed those already reported(E.L. Belokoneva and T.I. Timchenko, Sov. Phys. Crystallogr., 1983, 28(6), 658-661) by X-ray diffraction. The space group is C2/c. The results of energy dispersive X-ray analysis showed that Ba ion was not included in the crystals grown from

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$BaO-B_2O_3$ flux, whereas Mo ions were detected 1 - 3 at% in the crystals grown from a modified K_2O-MoO_3 flux. The orientation of the seed crystal affected crystal growth and morphology. In the case of seeding in the <10$\bar{1}$> direction, twins with the twin law of 3-fold rotation axis were usually formed. Untwinned single crystals could be grown by seeding perpendicular to the <10$\bar{1}$> direction. The crystal faces {010}, {111}, {11$\bar{1}$}, {001}, {021}, {102} were well-developed in the untwinned crystals. The changes of morphology according to the growth conditions were investigated. Optical and electron microscopic observation of the striation and crystal defects were carried out.

PS-15.02.06 ATTEMPTS TO GROW DELTA-CRYSTALS FROM BINARY SYSTEMS BY ZONE-MELTING. By W. Uebach, H. Bradaczek and G. Hildebrandt, Institut für Kristallographie, Freie Universität Berlin, Germany.

Delta-crystals, i.e. bulk crystals with an intrinsic continuous change in lattice spacing of at least 0.1% per centimeter, would open a new field in X-ray and neutron optics. A crystal grown from two substances which form the same lattice and have perfect miscibility will in general have lateral as well as axial changes in lattice parameters when grown by directional solidification of the melt. Almost all crystal growers try to avoid this well-known effect, we, however, tried to enhance it by subsequent zone melting and zone crystallization with the system lead chloride and bromide. For this purpose we constructed a programme controlled furnace made of fifty independent heater elements allowing for a wide range of temperature regimes. Temperatures around 500 °C could be stabilized within less than 1°C. A fine grain carbon crucible, about twelve centimeters high and protected by a silica tube was put in vertical position. A two centimeter melting zone was run from bottom to top without any external movement. All possibilities for preheating and annealing were given.
The main difficulty was that the crucibles used to crack when the crystal tip at the bottom was heated for a second time. It was observed in these cases that in the first zone run the salts had obviously reacted with the crucible walls and thus did not give way for the thermal expansion of the crystal tip in the second run. Furthermore the compositional change from one crystal end to the other was not by far as high as expected from model calculations. These calculations, however, were made under the assumption that in every stage of the growth the melt zone was completely homogenious. A control of this assumption seems impossible.
Until now our best results stem from a directional solidification experiment with the system potassium and rubidium chloride showing a lattice parameter difference of 0.4% over a distance of four centimeters. The quality of this single crystal was encouraging, the 'delta', however, could have been as high as 4% - theoretically.
New attempts will be made with the well-known and widely investigated system indium and gallium antimonide.

PS-15.02.07 CLUSTER FORMATION IN THE $LiNbO_3$ MELT BEFORE SOLIDIFICATION by P. Andonov*, CNRS-LMMM, Bellevue, Meudon Cedex, France; S. Kimura and T. Sawada, NIRIM, Tsukuba-shi, Ibaraki, 305 Japan

Structural analysis of the $LiNbO_3$ melt has been carried out by means of the Small Angle X-ray Scattering (SAXS) using the synchrotron radiation in the range of momentum transfer 0.020 $Å^{-1} \leq k \leq 1.400\ Å^{-1}$. Formation and evolution of the scattering particles were studied from 1673 K to 1513 K including a large undercooling domain. Their radius of gyration, size, shape and distance have been determined. Microclusters are present in all the T-domain and their number is sufficient to give an interference effect. By lowering the temperature, their size increases and macroclusters appear from 1550 K onwards. Previously, it was verified, in the same T-domain, that octahedrally coordinated niobium atoms exist in the melt as a fundamental local structure. So cluster models have been built using octahedral NbO_6 molecules. Single molecules could represent the smallest particles at high temperature. Dimers, chains of at most three or four NbO_6 octahedra and blocks of 2x2x1 NbO_6 octahedra could appear below 1670 K and subsist up to the solidification with also blocks having 3x2x1 NbO_6 octahedra and regular or deformed small aggregates constituted by two layers of four corner-shared niobium oxide molecules bonded by a lithium atom. Macroclustering due to a regrouping of microclusters explain the rapid increase of the viscosity. The numbers and the volumes of different particles were obtained from viscosity data and SAXS measurements. This cluster analysis using both determinations, is a very effective tool to study the nucleation and the clustering in the domain of the pre crystallization.

PS-15.02.08 CRYSTAL GROWTH OF $KTiOPO_4$ AND X-RAY TOPOGRAPHIC OBSERVATION. By J. H. Kim and S. J. Chung, Department of Inorganic Materials Engineering, Seoul National University, Seoul, 151-742, Korea.

Single crystals of potassium titanyl phosphate($KTiOPO_4$:KTP) were grown by top-seeding and slow cooling technique using $3K_2WO_4 \cdot P_2O_4$ as flux. Seeding was in the <010>, <001> and <011> direction, respectively, and the seed crystals were placed on the surface of the solutions for better observation and control of the growth process. Cooling rates were slower than 0.05℃/h at the beginning of growth and gradually increased up to 0.2℃/h. Inclusion-free crystals up to $30x40x40mm^3$ in size could be obtained by 3 weeks growing time. To obtain large crystals from a given crucible size, seeding in the <010> direction seems to be most suitable.

Grown crystals seeded in the <010> direction exhibit a slightly different morphology from that reported by L.K. Cheng, J.D. Bierlein and A.A. Ballman, J. Crystal Growth, 110, 1991, 697. The cleavage face of (200) is less developed and in some cases the (110) face appeared. The variation in crystal form arises from seed locations and orientations.

The structural quality of the slice cut from $KTiOPO_4$ crystal was studied by X-ray topography and electron microscopy. Dislocations, growth sector boundaries, inclusions, and growth striations were observed.