New Transparent MeliliteCeramic from Glass Crystallization

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Our recent work has proved the possibility to obtain new transparent polycrystalline ceramics by complete and congruent crystallization from their parent glass. This process combines the advantages of glass (shaping ability and large scale production) with the intrinsic ceramic properties. Moreover, crystallization from glass enables fully dense ceramics with thin grain boundaries to be obtained. The presence of residual porosity (acting as scattering centers), which is usually the major drawback preventingtransparency in polycrystalline ceramics, is here avoided.

Starting from this idea, we have first elaborated $BaAl_4O_7$ [1][2] transparent polycrystalline ceramics. This new materials exhibits two orthorhombic polymorphs both transparent in the visible and near infrared ranges, with a grain size distribution from 1 to 5 µm. Doped with Eu^{2+} [3], $BaAl_4O_7$ shows efficient scintillating properties, and the transparency can be enhanced through the crystallization of a second phase[4]. Following this primary work, other transparent polycrystalline ceramics were synthesized using the same concept. For example $Sr_3Al_2O_6$ [5] which is transparent up to 6 µm and the large scale $Sr_{1+x/2}Al_{2+x}Si_{2-x}O_8$ [6] ceramics showing 90% transmittance in the visible and near infrared ranges.

The main objective of the present new study is to elaboratetransparent melilite ceramics. The melilite structure is particularly flexible and can be substituted by numerous cations[7]. Our innovative crystallization from glass synthesis process enables the elaboration of new melilite compositions ($SrTmGa_3O_7$ and $SrYbGa_3O_7$) which cannot be obtained from classic solid state method. Surprisingly, the resulting structure of some$Sr\mathit{Ln}Ga_3O_7$ (with *Ln* = Ho, Er, Tb and Tm) materials synthesizedbycrystallization from glass is different from the traditional melilite phase. Actually, their structure shows an original superstructure with a tripling of the "a" parameter compared to the classic melilite structure. This superstructure can beexplained by a local orderingof the *Ln*/Sr ions as demonstrated by both synchrotron powder diffraction and high resolution STEM. Finally we present the possibility to synthesize transparent polycrystalline melilite ceramic exhibiting visible light emission from up-conversion mechanism in the $SrYb_{1-x}Er_xGa_3O_7$ system.

[1] M. Allix, Highly Transparent $BaAl_4O_7$ Polycrystalline Ceramic Obtained by Full Crystallization from Glass, Advanced Materials,2012, 24 5570

[2] M. Allix, Aluminate transparent glasses, glass-ceramics and ceramics, International patent, WO2013079707, (2013).

[3] G. Patton, Light yield sensitization by X-ray irradiation in $BaAl_4O_7$: Eu^{2+} ceramic scintillator obtained by full crystallization from glass, *Phys. Chem. Chem. Phys.,*2014, 16, 24824

[4] M. Boyer, Enhanced Transparency through Second Phase Crystallization in $BaAl_4O_7$ Scintillating Ceramics, *Cryst. Growth Des.*, 2016, 16 (1), pp 386–395

[5] S. Alahraché, Perfectly transparent $Sr_3Al_2O_6$ polycrystalline ceramic elaborated from glass crystallization, *Chem. Mater*. 2013, 25, 4017

[6] K. Al Saghir; Transparency through Structural Disorder: A New Concept for Innovative Transparent Ceramics. *Chem. Mater.*2015, *27*, 508–514

[7] M. Boyer, Transparent Polycrystalline Sr*RE*Ga_3O_7Melilite Ceramics: Potential Phosphors for Tuneable Solid State Lighting. *J. Mater. Chem. C*2016.