Positron-positronium trapping model in application to polycrystalline $BaGa_2O_4:Eu^{3+}$ ceramics

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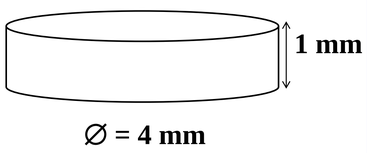
Introduction

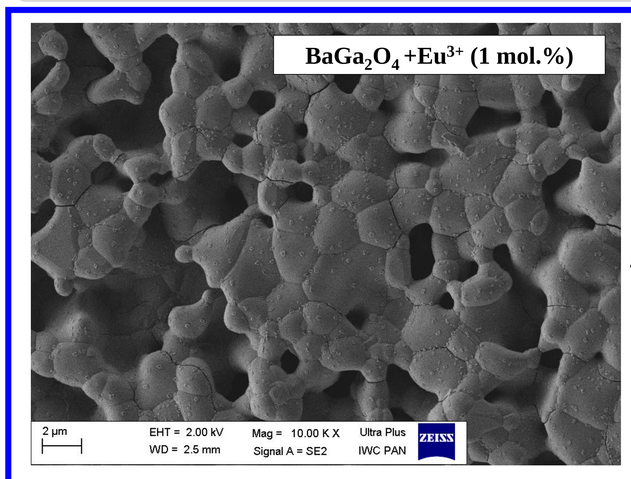
The $BaGa_2O_4$ ceramics doped with Eu^{3+} ions (1,3 and 4 mol%) synthesis were obtained by solid-phase sintering. The phase composition and microstructural features of ceramics were investigated using X-ray diffraction and scanning electron microscopy in comparison with energy dispersive methods. It is shown that undoped and Eu^{3+} -doped $BaGa_2O_4$ ceramics are characterized by a developed structure of grains, grain boundaries and pores. Additional phases are mainly localized near grain boundaries creating of additional defects. The evolution of defect-related free volumes in the $BaGa_2O_4$ ceramics due to the increase of the content of Eu^{3+} ions has been studied using positron annihilation lifetime spectroscopy technique. It is established that the increase in the number of Eu^{3+} ions in the basic $BaGa_2O_4$ matrix leads to the agglomeration of free-volume defects with their subsequent fragmentation. The presence of Eu^{3+} ions result in the expansion of nanosized pores and increase in their number with their future fragmentation.

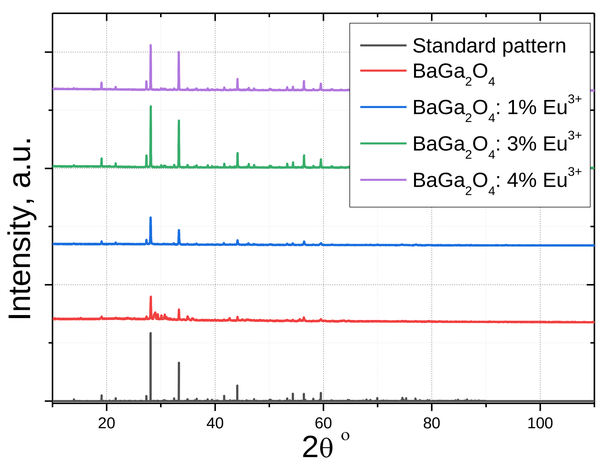
SAMPLE PREPARATION

The polycrystalline $BaGa_2O_4$ samples were prepared by solid-state reaction method. As starting materials $BaCO_3$ and Ga_2O_3 with purity 99.99% were used. Powders of stoichiometric composition with 0, 1, 3 and 4 mol.% of Eu_2O_3 (99.99%) were mixed in an agate mortar for 6 h with further pressing in a steel mold. Obtained pellets were annealed at 1200 ºC for 12 h in air. After that, the annealing of ceramic samples was carried out at 1300 ºC for 4h. The obtained polycrystalline ceramic samples were 4 mm in diameter and 1 mm in thickness.

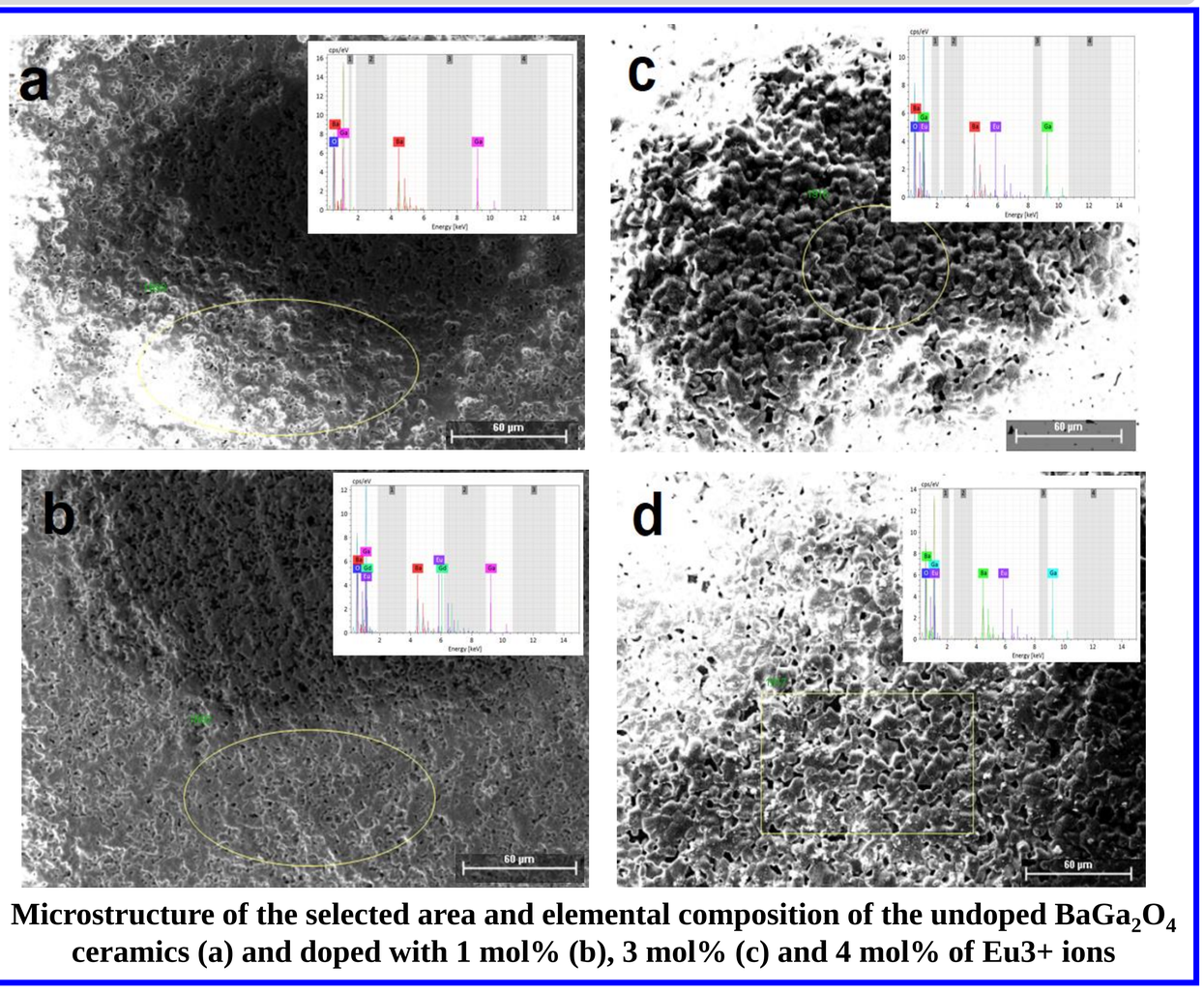


MICROSTRUCTURE and XRD DATA OF CERAMICS



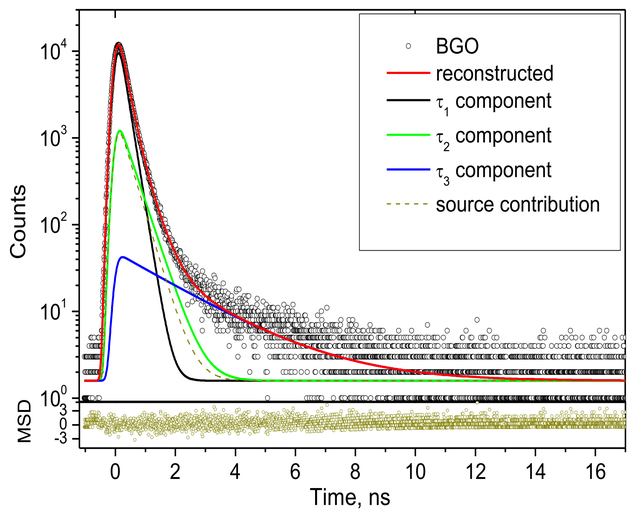


SEM and EDX results



Microstructure of the selected area and elemental composition of the undoped $BaGa_2O_4$ ceramics (a) and doped with 1 mol% (b), 3 mol% (c) and 4 mol% of Eu3+ ions

RESULTS: PAL characteristics



Sample	τ_1, ns	I_1, %	τ_2, ns	I_2, %	τ_3, ns	I_3, %
$BaGa_2O_3$	0.200	83.3	0.424	14.9	2.196	1.8
$BaGa_2O_3$+1 mol% Eu^{3+}	0.206	85.1	0.450	13.2	2.289	1.7
$BaGa_2O_3$+3 mol% Eu^{3+}	0.212	89.9	0.550	7.9	2.390	2.2
$BaGa_2O_3$+4 mol% Eu^{3+}	0.201	83.3	0.411	14.4	2.157	2.4

Sample	$\tau_{av.}$, ns	τ_b, ns	κ_d, ns^{-1}	τ_2 -τ_b, ns	τ_2/τ_b	R_3, nm
$BaGa_2O_3$	0.234	0.218	0.40	0.21	1.95	0.306
$BaGa_2O_3$+1 mol% Eu^{3+}	0.239	0.222	0.35	0.23	2.02	0.314
$BaGa_2O_3$+3 mol% Eu^{3+}	0.240	0.223	0.23	0.33	2.46	0.322
$BaGa_2O_3$+4 mol% Eu^{3+}	0.232	0.218	0.37	0.19	1.89	0.302

$\tau_{av.} = \frac{\tau_1 I_1 + \tau_2 I_2}{I_1 + I_2}$	**Mean positron lifetime:** reflects cumulative defect environment prevailing in sample
Lifetime τ_b: associated with the positron trapping in defect-free bulk	$\tau_b = \frac{I_1 + I_2}{\frac{I_1}{\tau_1} + \frac{I_2}{\tau_2}}$
$\kappa_d = \frac{I_2}{I_1}\left(\frac{1}{\tau_b} - \frac{1}{\tau_2}\right)$	**Positron trapping rate in defects**
τ_2- τ_b	Size measure of extended defects where positrons are trapped
Represents the nature of defects	τ_2/τ_b

Diagram explaining the evolution of defect-related voids in $BaGa_2O_4$ ceramics caused by Eu^{3+} doping

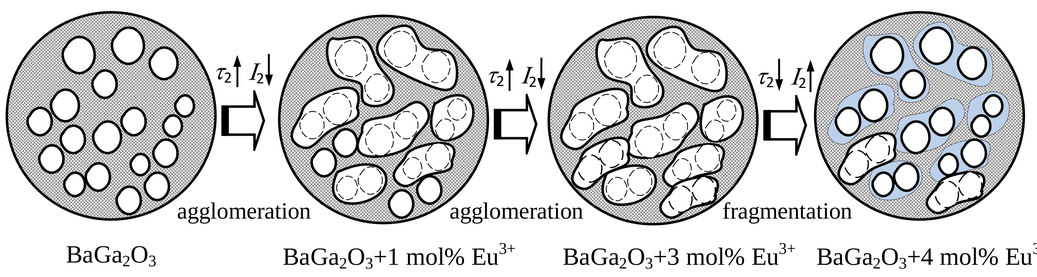
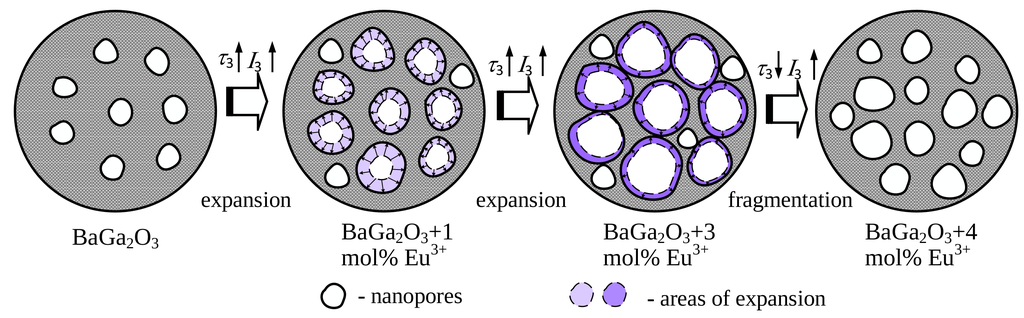


Diagram explaining the evolution of nanopores in $BaGa_2O_4$ ceramics caused by Eu^{3+} doping



Conclusion

The structural features and evolution of free-volume defects in the $BaGa_2O_4$ ceramics obtained by solid-phase synthesis from the initial $BaCO_3$ and Ga_2O_3 components with the addition of different amount of Eu_2O_3 content (1, 3 and 4 mol%) were investigated The structural features of ceramics were studied using XRD as well as SEM with EDX elemental analysis. It is established that in according to the quantitative analysis of the elemental composition, samples of the undoped $BaGa_2O_4$ ceramics have the largest deviations from the stoichiometric composition, they are three-phases. Such processes are apparently caused by the evaporation of the constituent synthesis powders during the annealing process at high temperatures. The detected signs correlate with the XRD data. Additional phases in ceramics are mainly localized near the grain boundaries and create defective centers for positron capture studied by PAL spectroscopy. Analyzing the second component of PAL spectra for the undoped and Eu^{3+}-doped $BaGa_2O_4$ ceramics, it was shown that an increase of Eu^{3+} content from 1 to 3 mol% leads to agglomeration of free-volume defects near grain boundaries of ceramics. At the same time, nanopores in ceramics expand and their number increases. Further increase in the content of Eu^{3+} ions are accompanied with fragmentation of both free-volume defects and nanopores.