Nanoengineered Gd₃Al₂Ga₃O₁₂ scintillation materials with disordered garnet structure for novel detectors of ionizing radiation

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Main requirements to scintillators

- high stopping power
- high scintillation yield
- high energy resolution
- minimal level of afterglow
- multifunctionality

Garnet structure and its possibilities





C.N.	8	6	4
Y	1.019	0,9	-
Gd	1.053	0.938	-
AI	-	0.535	0.39
Ga	-	0,62	0.47

Y = Lu, Gd, Ca, Mg, Li, Zr, Hf

Al = Ga, Sc, In, Si, Ge, Fe, Cr, Mg.....

n + ¹⁵⁵Gd → ¹⁵⁶Gd + γ (8.5 MeV) n + ¹⁵⁷Gd → ¹⁵⁸Gd + γ (7.9 MeV). cross section of the neutron capture: ¹⁵⁵Gd - 61000 barn, ¹⁵⁷Gd 254,000 barn

Mixed Garnets scintillators properties

Material	Density, g/cm3	LY	Emission	Decay
Gd3Al3Ga2O12 :Ce		46000		~80 ns
Y3Al5O12:Ce (YAG)	4.6	16000	550	100
YGd2Ga3Al2O1 2:Ce		44000		50(90%); 120 (10%)

Enchance the ability for recombination of geminate pairs due to the local micro-nonuniformity related with modification of local structure of mixed garnets;

Diminish the thermo-activation energy of deep traps due to the shift of the bottom of the conduction band leads to covering some shallow traps located below of this band.

Influence of the synthesis approach on afterglow

Synthesis methods	Particle size	Homogeneity	Morphology	Synthesis temperatures (°C)	Afterglow time
Solid-solid reaction	Micrometer-scale	Bad	Bad	>1000	Long
Sol-gel method	Nano to micrometer-scale	Medium	Bad	≤1000	Medium
Combustion method	Nano to micrometer-scale	Medium	Medium	>1000	Short
Hydrothermal method	Nano to micrometer-scale	Good	Medium	<1000	Short
Co-precipitation	Nano to micrometer-scale	Good	Medium	>1000	Short
Template method	Nano to micrometer-scale	Good	Good	<1000	Short
Electrospinning	Nano to micrometer-scale	Medium	Medium	>1000	Short
Laser technique	Nano to micrometer-scale	Medium	Medium	none	Medium
Electron beam irradiation	Nano to micrometer-scale	Medium	Medium	none	Medium

• Table gives a comparison of synthesis methods in terms of particle size, required temperature, homogeneity, morphology, and afterglow time.

The development of approach for suppression of afterglow in GGAG crystals is crutial task!

We used 2 approaches: Co precipitation (CP) and Solid-state (SSS)

Sample	Raw material	Co-doping
SSR1	SSS	-
SSR2	SSS	Mg, 10ppm
SSR3	SSS	Mg, 50ppm
CP1	СР	-
CP2	СР	Mg, 50ppm

JSM-7800F Schottky FE-SEM

Extreme-high resolution imaging and full analytical capabilities

Specifications:

Mag: 25 to 1,000,000X kV: 10V to 30kV SEI: 1.0 nm (15kV) 0.8 nm (15kV, GB) 1.5 nm (1kV) 1.2 nm (1kV, GB) 3.0 nm (0.1kV, GB) BEI: 1.5 nm (3kV) STEM: 0.8 nm (30kV), 0.6 nm attainable Analytical: 3.0 nm (15kV, 5nA, WD=10mm)

Standard features:

Through the lens detector with energy filter In-chamber E/T SE detector Aperture angle control lens (ACL) Easy to use, remote-enabled GUI

Optional items:

Short WD retractable BE detector LV attachment (up to 300 Pa), LV BSE, LV SE GBSH – GB up to 5kV

Typical accessories: EDS, WDS, CL, EBSD, STEM, PCD





Line Scan – Magnification 25000X











Mapping - Magnification 50000 X







Element	Weight %	Atomic %	Net Int.	Error %	Kratio	Z	R	Α	F
GaL	26.65	14.82	2705.2	7.90	0.26	0.94	1.02	0.93	1.00
AIK	11.07	15.90	1704.1	2.51	0.06	1.17	0.90	0.42	1.00
CeL	0.83	0.23	32.1	35.67	0.01	0.79	1.13	1.03	1.07
GdL	36.69	9.05	930.8	4.40	0.33	0.75	1.12	1.03	1.05
Oxygen	24.76	60.00	0	0.00	0.00	0.00	0.00	0.00	0.00



Lsec: 21.9 0 Cnts 0.000 keV Det: Octane Pro Det

Line Scan analysis



Element	Weight 1	L Atomic Ti	NetInterv	Error %	K-ratio
OK	15.9	51.3	543.9	7.6	7.60246
Gal.	25.7	19.1	1300.8	13	1.9112
AK	6.5	12.6	435.3	8.2	8.22768
65.	31.8	17	629.6	14	3.76808





Elemen	t Weight 1	Atomic A	% Net Inter	. Error 1	6 K-ratio	
OK	16.1	51.4	662.5	7.4	7.42228	
GAL	26.3	19.1	1020.2	17	1.74292	
AIK	6.8	13	351.7	7.8	7.76984	
GdL	50.9	16.5	477.8	2.9	2.92237	

TSL measurements for SSR samples



<u>Co-doped sample shows TSL intensity at 5 times less</u>



Intensity, a.u.

LY measurements



Sample	Raw material	Co-doping	LY
SSR1	SSS	-	42000
SSR2	SSS	Mg, 10ppm	41000
SSR3	SSS	Mg, 50ppm	32000
CP1	СР	-	59000
CP2	СР	Mg, 50ppm	44000



Conclusions

CP samples show higher LY values that most probably related with lower concentration of intrinsic defects due to the lesser Ga evaporation at crystals growth in comparison with SSR samples;

The possibility of using linear EDX scanning for investigation samples inhomogeneity is shown;

The technology for co-doped samples should be improved for increase the LY;

Co-doping decrease TSL intensity only and does not impact on the traps distribution.