

THE LOW TEMPERATURE AEROSOL SYNTHESIS OF YAG:Ce³⁺ NANOSTRUCTURES: COMPARATIVE STUDY OF THE XRPD MICROSTRUCTURAL PARAMETERS

Lj. Kandić¹, K. Marinković¹, L. Mančić¹, G. del Rosario², O. Milošević¹

¹Institute of Technical Sciences of Serbian Academy of Sciences and Arts, K.Mihajlova 35/IV, 11000 Belgrade, Serbia

²University Rey Juan Carlos I, Tulipan s/n. Móstoles, Madrid 28933, Spain

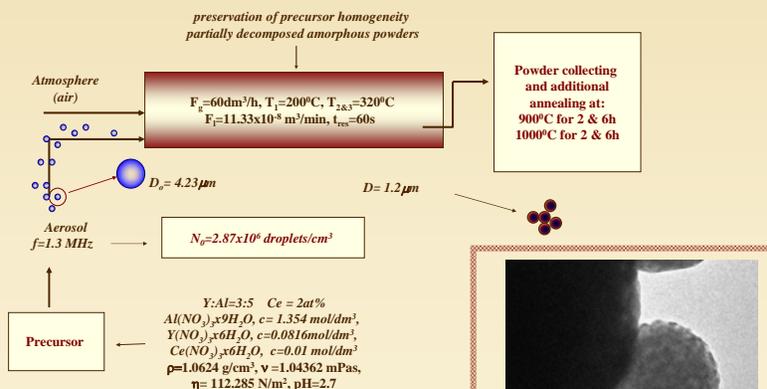
Abstract

Comparative analysis for the YAG:Ce³⁺ microstructural parameters is performed through Fullprof and Koalariet-XFit programs. Both programs are based on Rietveld method of structural refinement where fitting is done by convolution and corrections of XRD peak width for instrument broadening through either using the reference specimen (Fullprof) or the fundamental parameters approach (Koalariet-XFit).

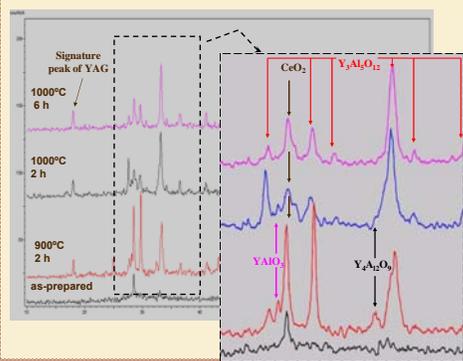
YAG:Ce³⁺ powder is obtained via low temperature aerosol route at 320°C followed with post annealing treatment at 900 and 1000°C. Besides targeting cubic garnet structure Y₃Al₅O₁₂ (YAG), the presence of either perovskite YAlO₃ (YAP) or monoclinic Y₄Al₂O₉ (YAM) phase is detected. Since garnet phase represents a very promising host phosphor material being doped with rare-earth ions, the most important criteria determining its applicability in various optical devices is the uniform distribution of the luminescent center in the host lattice as well as the cubic YAG phase crystallinity.

Comparison of data obtained for same the sample through usages of the above mentioned XRPD analyses gives insight of the validity of the refined parameters.

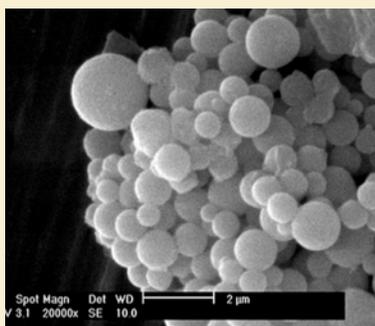
Low temperature aerosol decomposition



Phase development

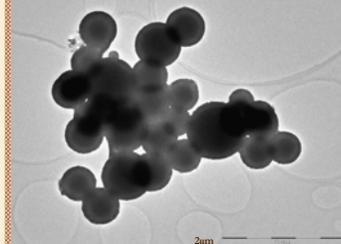


SEM (XL30ESEM Philips)

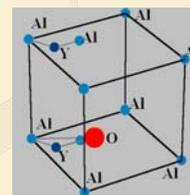


Spherically shaped and submicron sized as prepared particles
 $D_{\text{mean}} = 0.7 \mu\text{m}$

TEM (Tecnai 20 Philips)



Composite Inner particle structure



Structure refinement

Al-O: 1.93189 1.76415
 Y-O: 2.32885 Y-Al: 3.01420

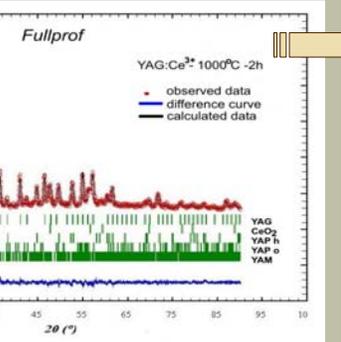
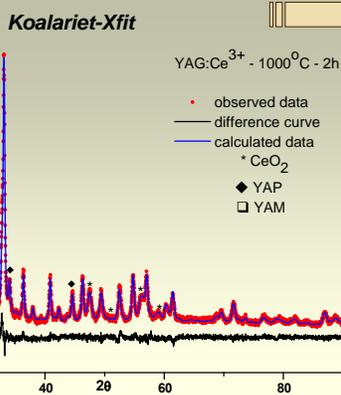
(XRPD data: step 0.02, time 15s)

Koalariet program (fundamental parameter approach): $\text{gof} = 0.914$ $\text{rwp} = 7.433$
 background: polynomial function of the 8th degree

YAG crystalline	Ia-3d cubic inter-crystalline	CeO ₂ crystalline	Fm-3m cubic inter-crystalline	YAP P6 3/mmc hexagonal
lp (Å): 12.0568_0.0001	12.0801_0.0001	lp: 5.4059_0.001	5.3933_0.0036	lp _{1,2} : 3.6847_0.001 lp ₃ : 10.5003_0.0015
wt%: 47.988_0.461	12.110_0.587	12.110_0.587	1.011_0.116	15.723_0.173
CS (Å): 647.25_38.53		126.56_15.52		460.47_22.39
strain: 0.485_0.024	1.309_0.181	0.0001_0.1793	1.07008_0.17978	0.499_0.039

Fullprof program: $X^2 = 0.803$ $\text{Rwp} = 15.3$ $R = 5.86$ $\text{Rexp} = 17.61$
 background: linear interpolation between 34 points

YAG crystalline	Ia-3d cubic inter-crystalline	CeO ₂ crystalline	Fm-3m cubic inter-crystalline	YAP P6 3/mmc hexagonal
lp (Å): 12.0454(7)		lp: 5.3955(3)		lp _{1,2} : 3.6742(3) lp ₃ : 10.4883(9)
wt%: 57.8(7)		12.3(2)		20.6(3)
CS (Å): 373.58(0.12)		95.48 (0.04)		236.43(0.25)
strain: 26.5226(0.0216)		25.3791(0.0052)		11.2298(0.0096)



Conclusion

Fundamental parameter approach in Koalariet uses physical based models to generate the line profile shape so it enables recognizing of crystalline and intercrystalline domains presence in YAG and CeO₂ and gives more accurate insight in nanocrystalline particle nature.

Standard approach in Fullprof uses analytical-function fit (Tompson-Cox-Hastings pseudo-Voigt function) resulting in more correlated crystalline size and strain parameters determination where intercrystalline domains could not be considered.

Both programs allow to obtain structural and compositional information for all crystalline phases present in analysed sample but in the case of low weight percentages they could not be modelled.