NRC «Kurchatov Institute» - IREA

NRC «Kurchatov Institute» Laboratory of luminescent and detector materials

Processing of scintillation ceramics based on complex oxides with garnet structure

Karpyuk P.V.[1,2], Dosovitskiy G.A.[1,2], Kuznetsova D.E.[1,2], Gordienko E.V.[1,2], Dosovitskiy A.E.[3], Korzhik M.V.[4]

[1] National research center “Kurchatov institute” - IREA, 107076 Moscow, Bogorodskiy val str. 3, Russia;
[2] National research center “Kurchatov institute”, 123182, Academik Kurchatov sqr. 1, Moscow, Russia;
[3] NeoChem JSC; [4] Institute for Nuclear Problems of Belarusian State University

2018

Polycrystalline form of scintillating materials in comparison to single crystal

Advantages

- composition variation
- different geometrical forms
- potentially lower cost

Disadvantages

- obtaining process with several dissimilar stages
- fully transparent ceramics - expensive

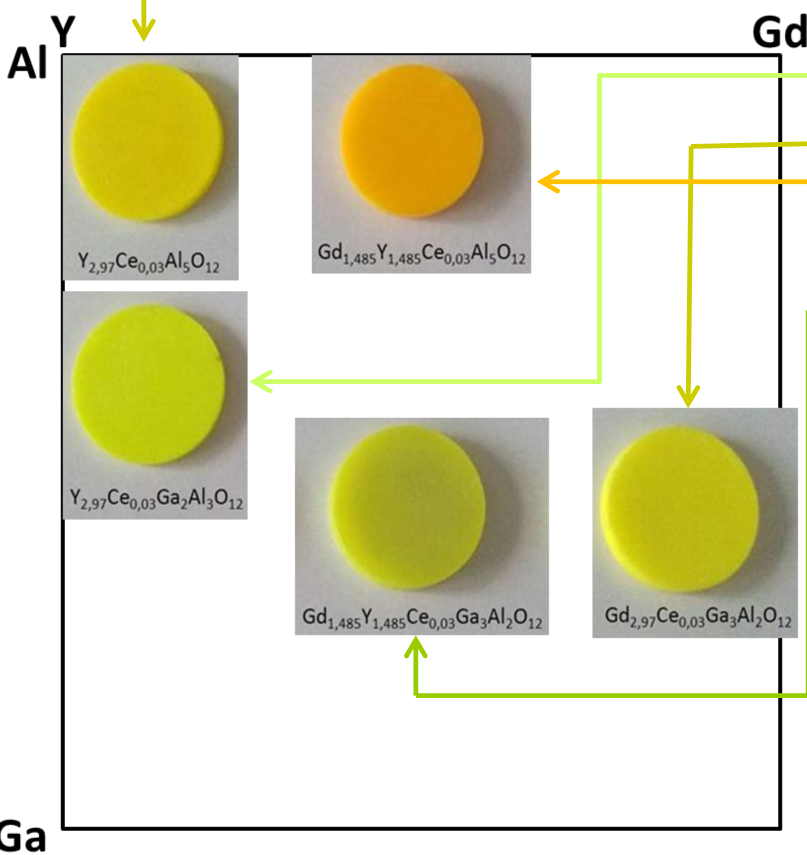
Main goal is obtaining of highly translucent ceramics with good scintillation properties (high light yield and short scintillation decay time).

Garnet phosphors family

General composition:

$Gd_{3-x-y}Y_xCe_yAl_{5-z}Ga_zO_{12}$

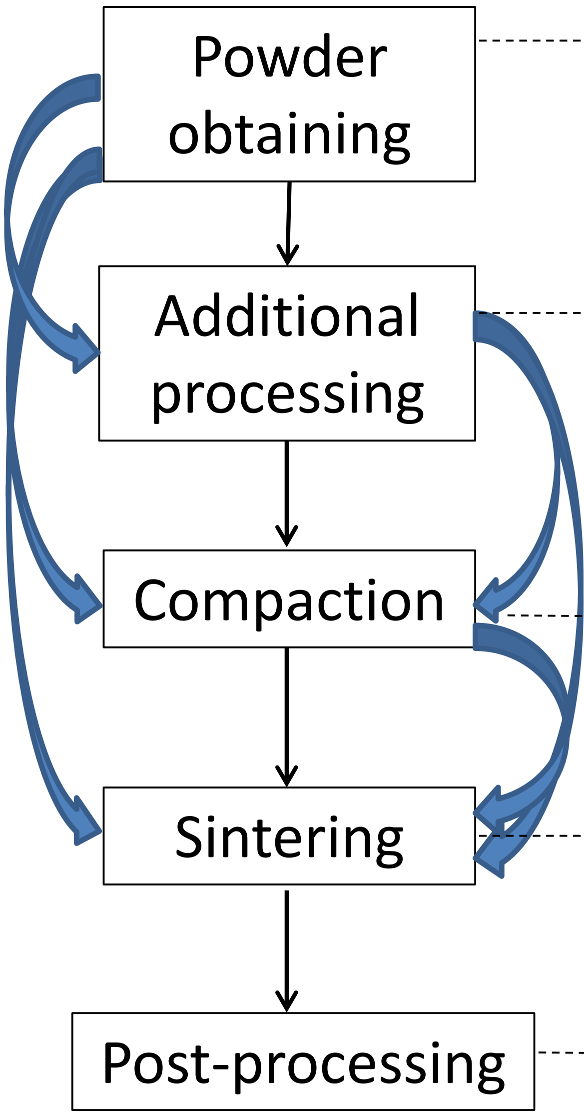
$Gd_{3-x-y}Lu_xCe_yAl_{5-z}Ga_zO_{12}$



Composition	Light yield, photons/MeV	Decay time, ns
YAG:Ce	23000	90 + slow
YAGG:Ce	24000	100
GGAG:Ce	56000	13 + slow
GYAGG:Ce	45000	14 + slow
GYAG:Ce	14000	90
LuAG:Ce	13000	39 +slow
LuAGG:Ce	20000	60
LuGAGG:Ce	30600	67 +slow

M.Moszyńskia Properties of the YAG:Ce scintillator / 1994.
Sidletskiy O. et al. Engineering of bulk and fiber-shaped YAGG: Ce scintillator crystals //2017.
Kamada K. et al. Growth and scintillation properties of 3 in. diameter Ce doped Gd3Ga3Al2O12 scintillation single crystal 2016.
Cherepy Comparative gamma spectroscopy with SrI2(Eu), GYGAG(Ce) and Bi-loaded plastic scintillators, 2010.
Kamada K. et al. Composition engineering in cerium-doped (Lu, Gd) 3 (Ga, Al) 5O12 single-crystal scintillators // 2011.

Process of ceramics obtaining



Synthesis of initial substance with preset composition,
Heat treatment → Phase formation

Fractioning, homogenizing, mechanical treatment → Particle size averaging

Powder formation into a "green body"

Obtaining of dense polycrystalline structure

Polishing to improve optical properties of sample surface, additional annealing

Powder obtaining

Required properties:
- Nano-sized primary particles
- Regular particle shape
- Composition homogeneity

Methods:
- Co-precipitation method
- Sol-gel method
- Pyrolysis
- Mixing of individual powders

	Nano-sized particles	Regular shape of particles	Composition homogeneity	Scaling possibility
Co-precipitation	+	+	+	+
Sol-gel	+	+	+	-
Pyrolysis	+	+	+	-
Mixing	+/-	+/-	-	+

Powder obtaining

Complex solution of Gd, Y, Ce, Ga, Al nitrates

↓

Precipitation

↓

Separation of precipitate

↓

Calcination

General composition:
$Gd_{3-x-y}Y_xCe_yAl_{5-z}Ga_zO_{12}$

YAG:Ce (Y, Ce, Al)

YAGG:Ce (Y, Ce, Al, Ga)

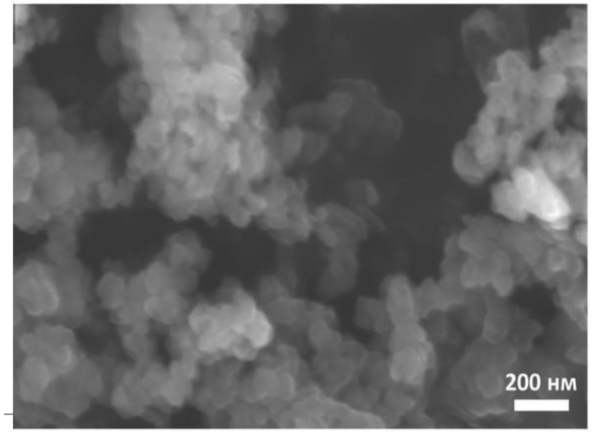
GYAG:Ce (Gd, Y, Ce, Al)

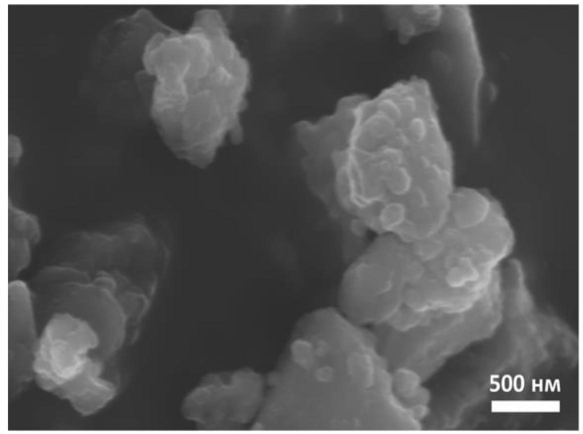
GYAGG:Ce (Gd, Y, Ce, Al, Ga)

GAGG:Ce (Gd, Ce, Al, Ga)

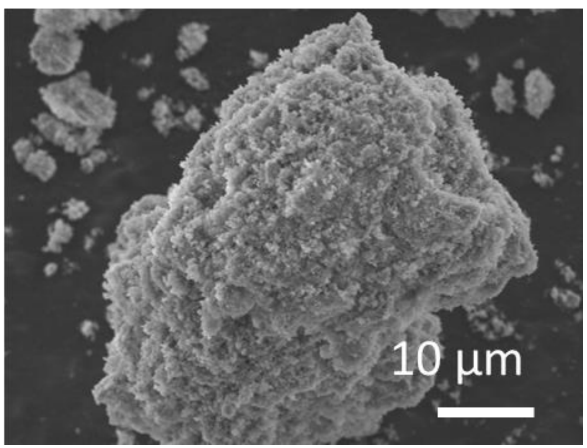
Powder obtaining

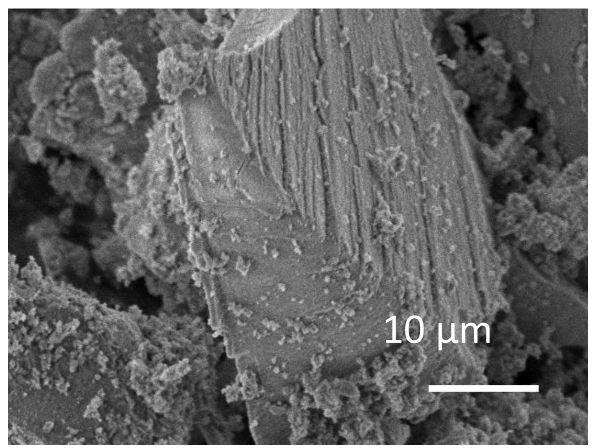
NH_4HCO_3 precipitant $NH_3 \cdot H_2O$ precipitant





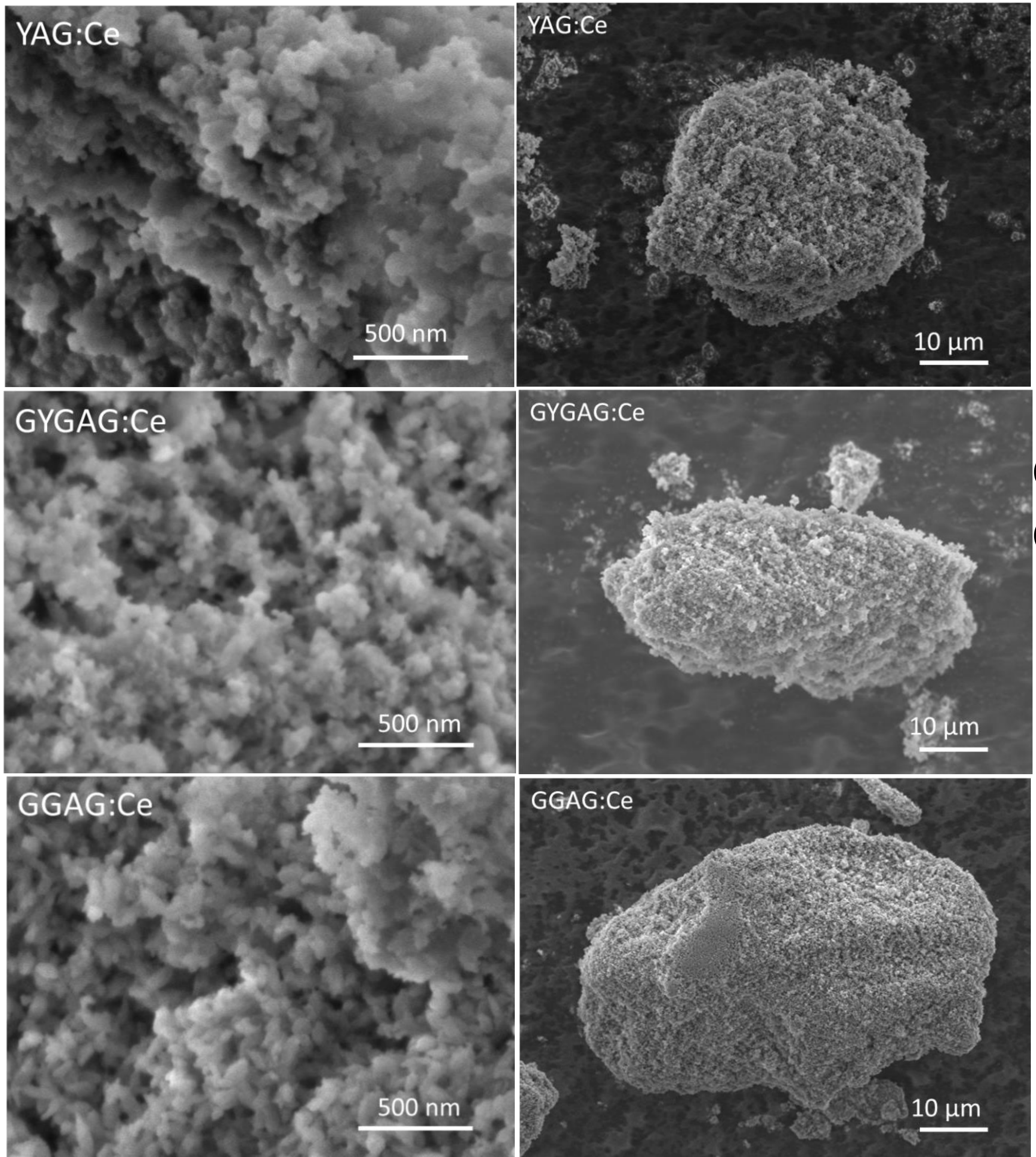
Primary particle size ~50 nm (after drying 100 °C)





Secondary particle size (after aggregation) ~50-100 µm

Powder obtaining



YAG:Ce (100 °C)
$Y_{2,97}Ce_{0,03}Al_5O_{12}$

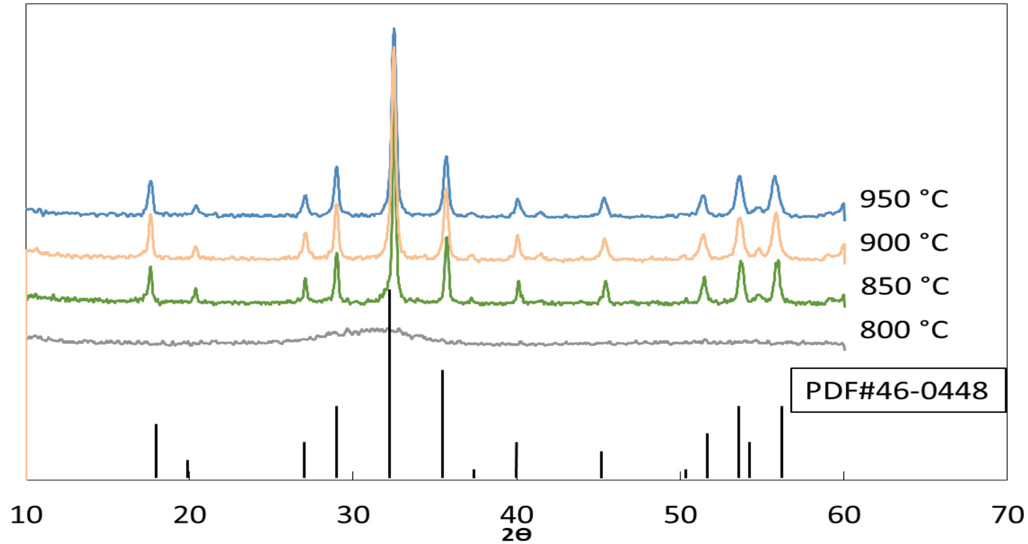
GYAGG:Ce (100 °C)
$Gd_{1,485}Y_{1,485}Ce_{0,03}Al_2Ga_3O_{12}$

GGAG:Ce (100 °C)
$Gd_{2,97}Ce_{0,03}Al_2Ga_3O_{12}$

Powder obtaining

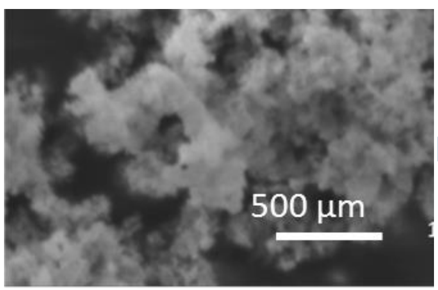
Heat treatment (calcination)

XRD- diagrams for GGAG:Ce after calcination at different temperatures

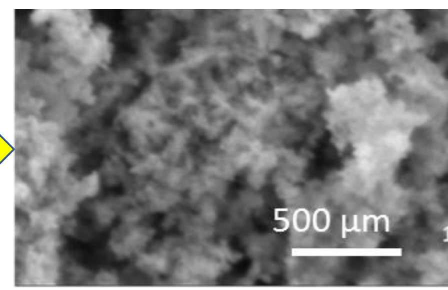


GGAG:Ce calcination

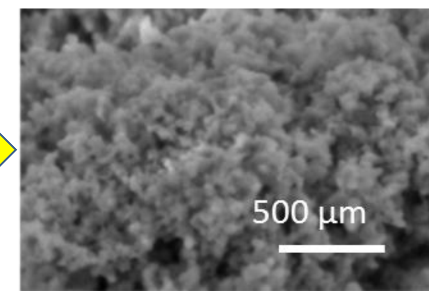
800 °C



850 °C

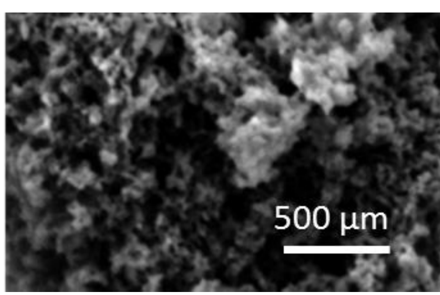


900 °C

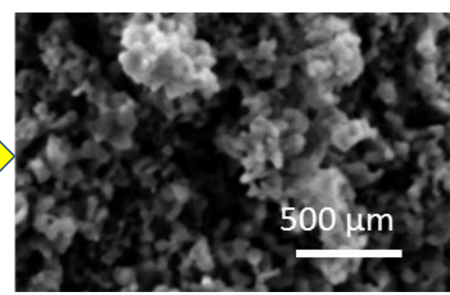


GYAGG:Ce calcination

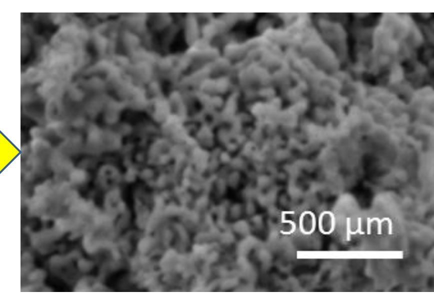
900 °C



950 °C



1000 °C

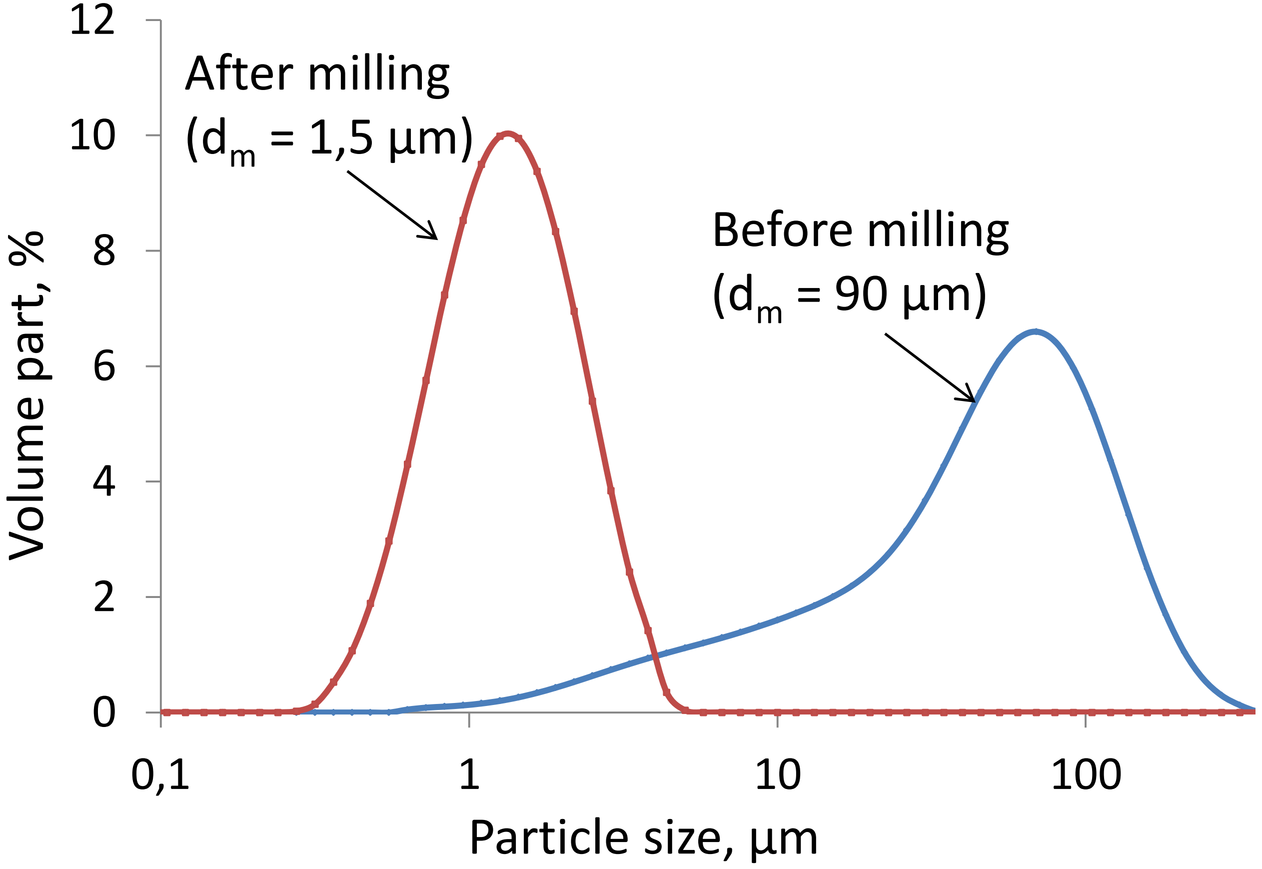


Additional processing

Mechanical treatment (milling)

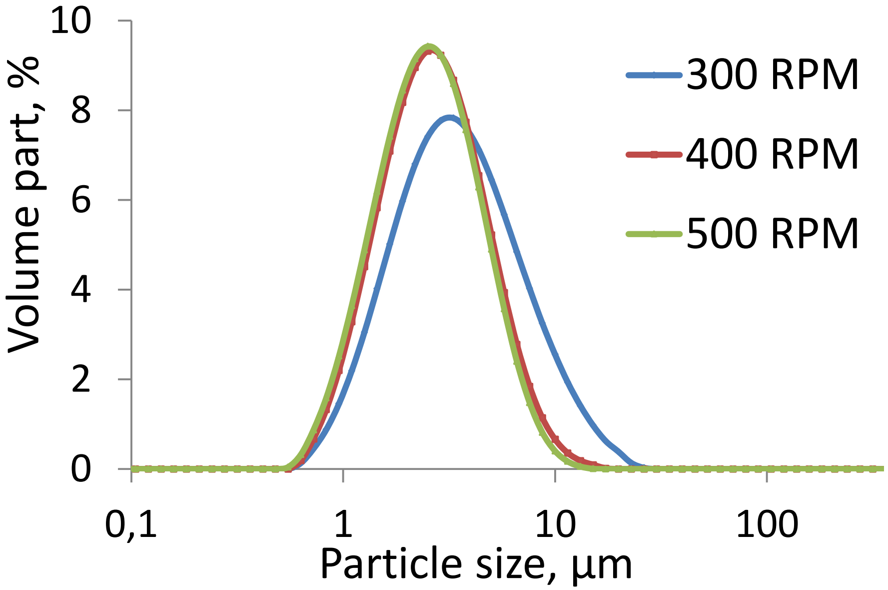
Milling was performed in planetary mill

Particle size distribution was determined by the laser diffraction method

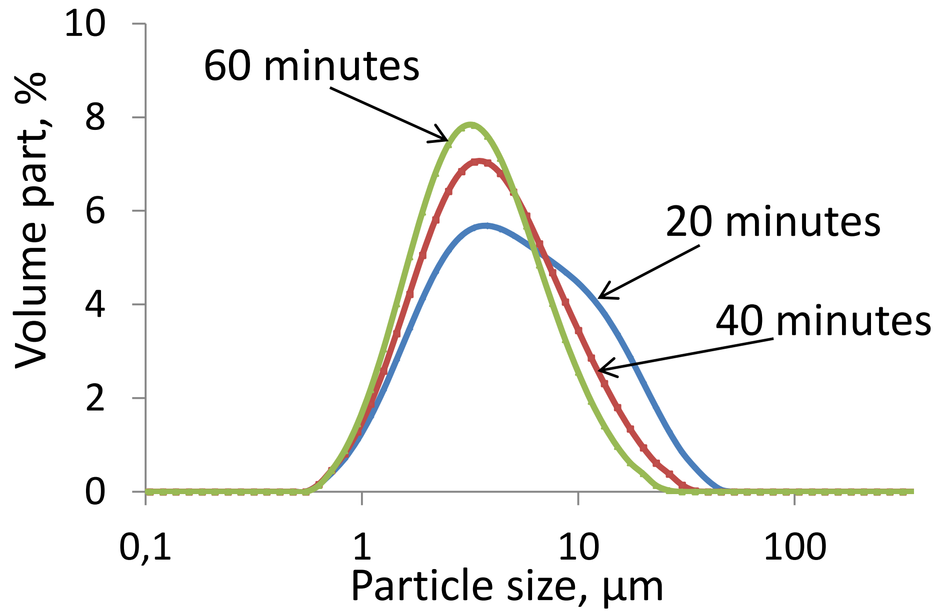


Milling: influence of process features

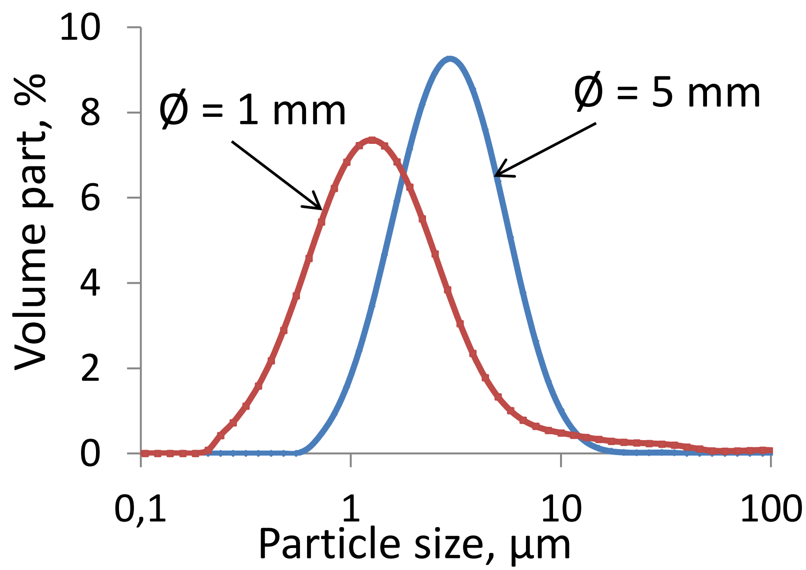
Milling rate



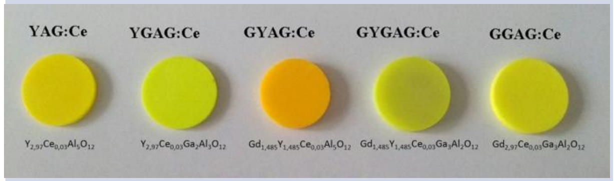
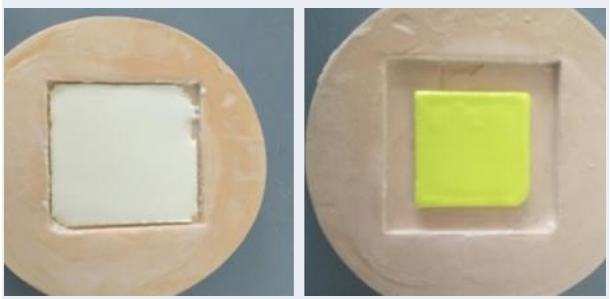
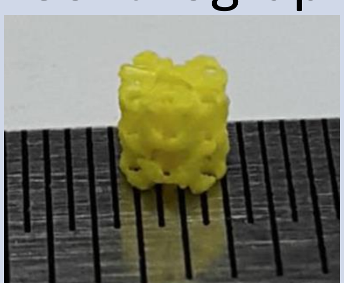
Milling time



Diameter of milling bodies



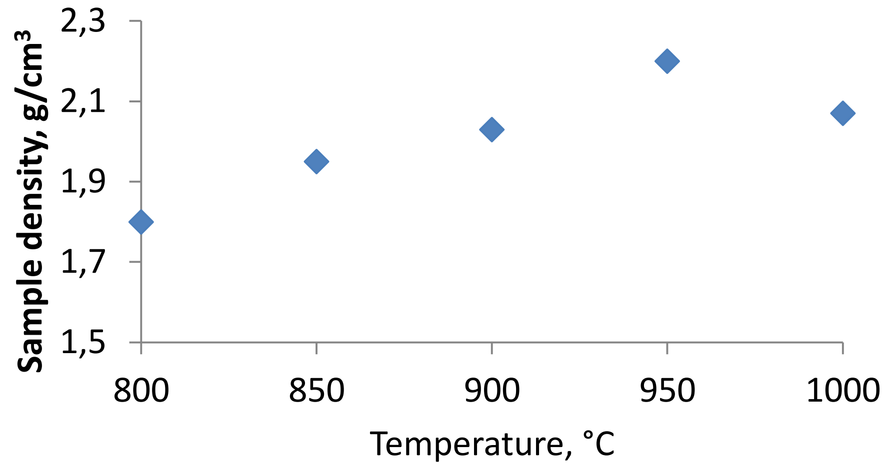
Compaction methods

<table>
<tr><th>Method</th><th>Features</th></tr>
<tr><td>Uniaxial pressing

</td><td>+ fast
+ cheap and widespread equipment
- internal stresses
- form of “green body” depends on geometry of pressing matrix</td></tr>
<tr><td>Slip-casting
</td><td>+ absence of internal stresses
+ various forms of obtained “green body”
- requires suspensions with large volume of solid phase
- potential admixtures from material of casting form</td></tr>
<tr><td>Stereolithography
</td><td>+ the most complex form of obtained “green body”
- slow rate of compaction
- requires special equipment</td></tr>
</table>

Powder synthesis & processing → Compaction

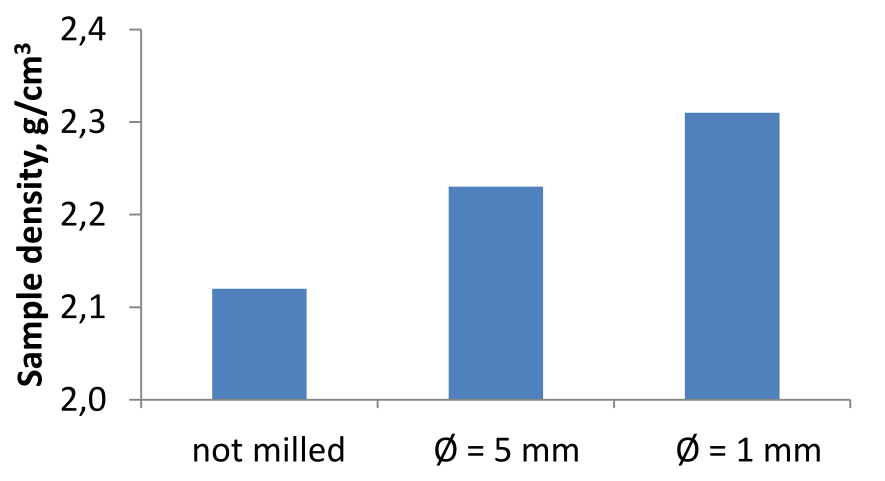
Initial powder of GYAGG- composition after calcination was milled by 1 mm Al_2O_3 milling bodies

Calcination temperature, °C	Sample density, g/cm^3
800	1,80
850	1,95
900	2,03
950	2,20
1000	2,07



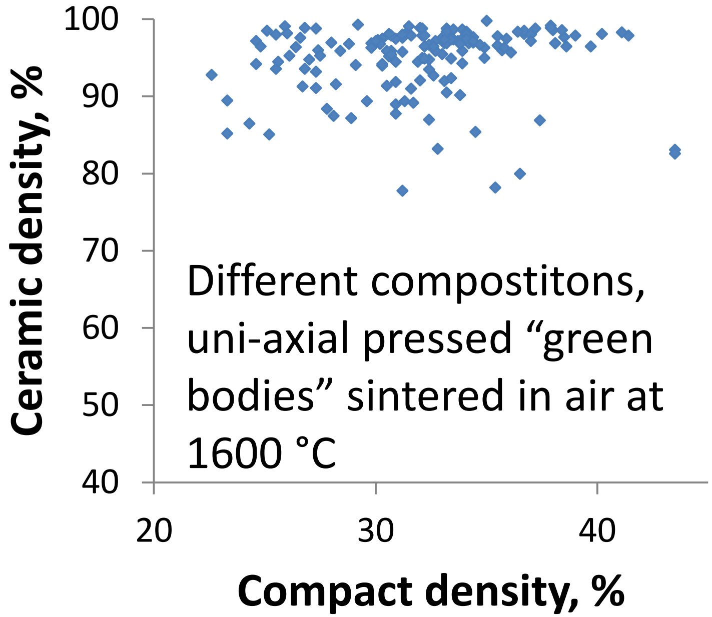
Initial powder of GYAGG- composition was calcined at 850 °C

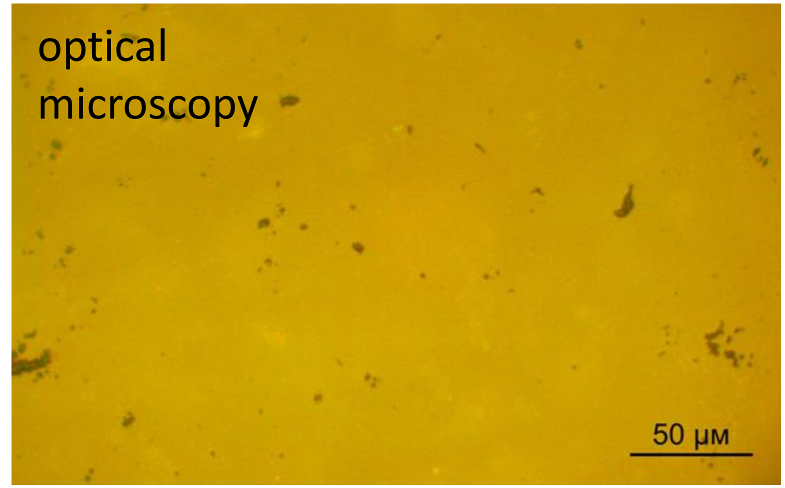
Milling	ρ, g/cm^3
-	2,12
Planetary mill, Al_2O_3 milling bodies Ø = 5 mm	2,23
Planetary mill, Al_2O_3 milling bodies Ø = 1mm	2,31



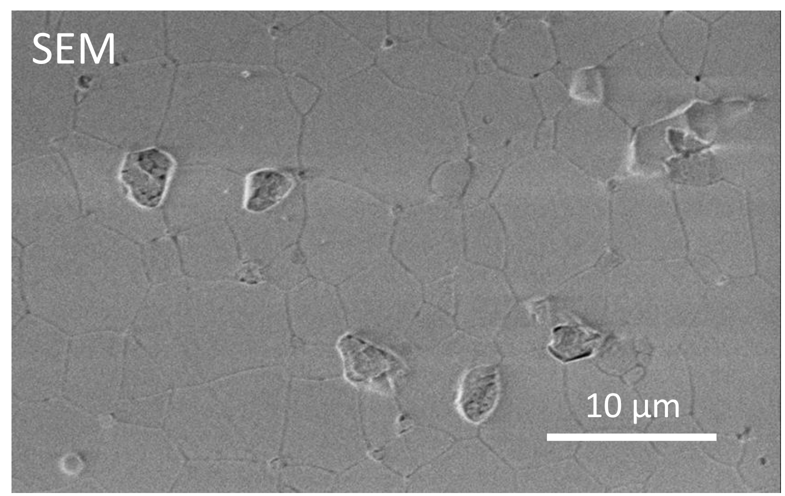
Compaction → Sintering

Sintering in **air**





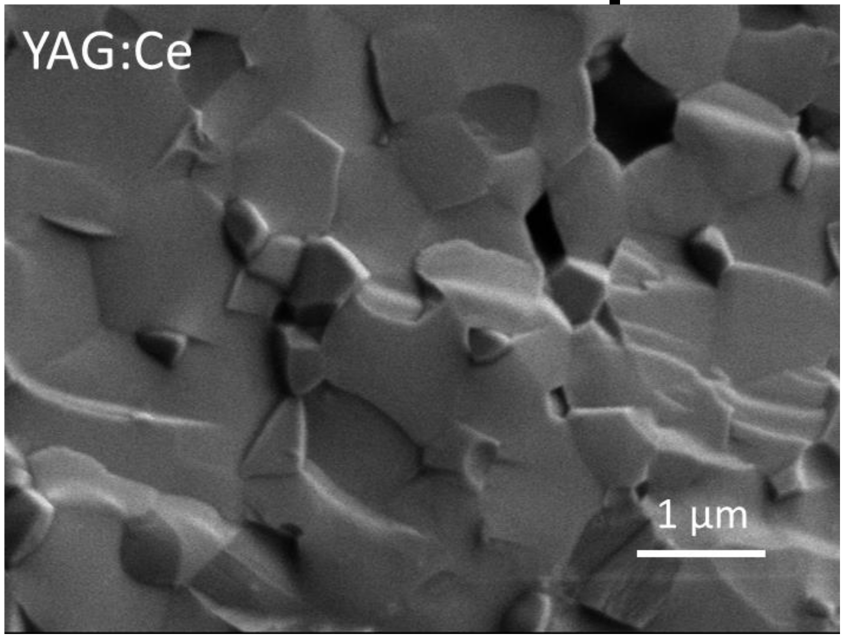
GYAGG:Ce, ~ 1,0% of pores

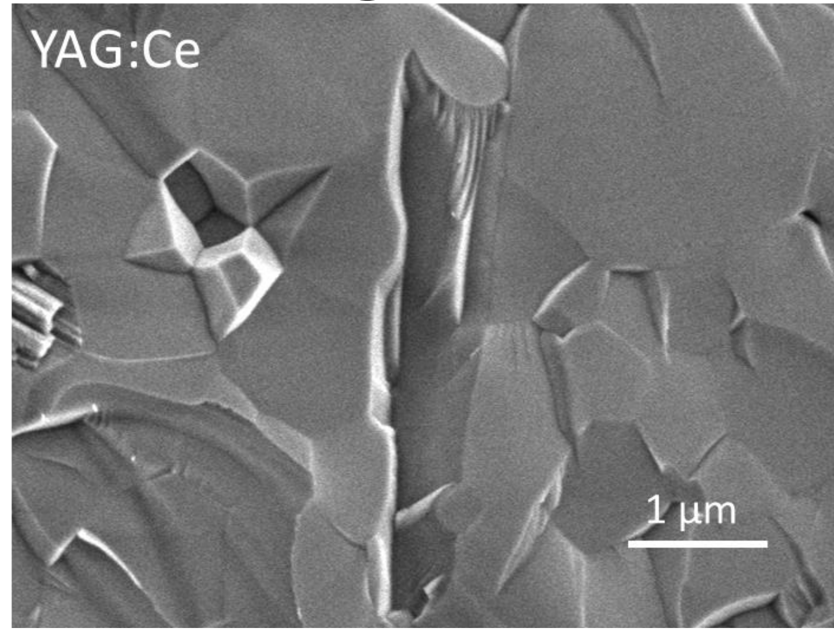


Precipitant	Compact density*, %	Ceramic density*, %
NH_4HCO_3	25 – 30	97 – 99
NH_3H_2O	35 – 45	93 – 95

*initial composition YAG:Ce (theoretical density – 4,55 g/cm^3)

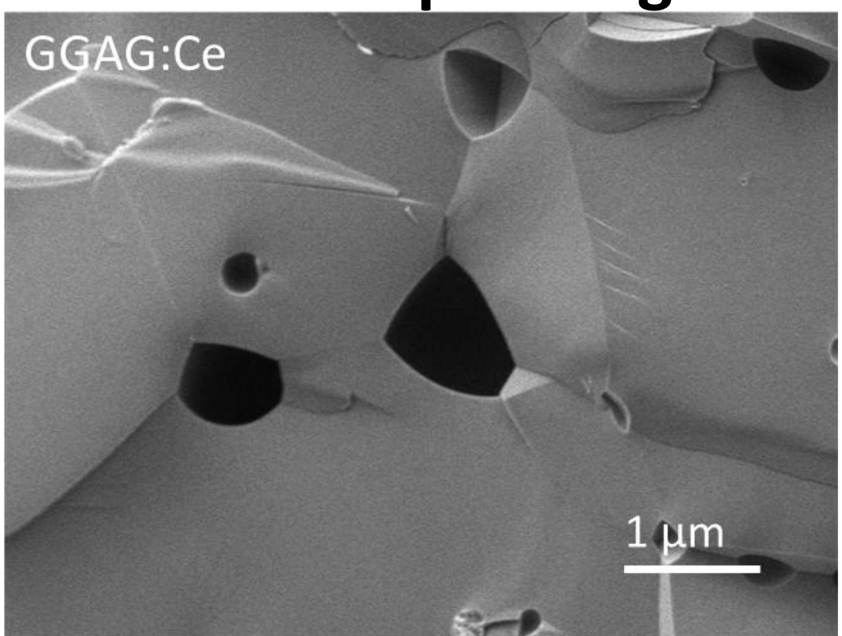
Compaction → Sintering

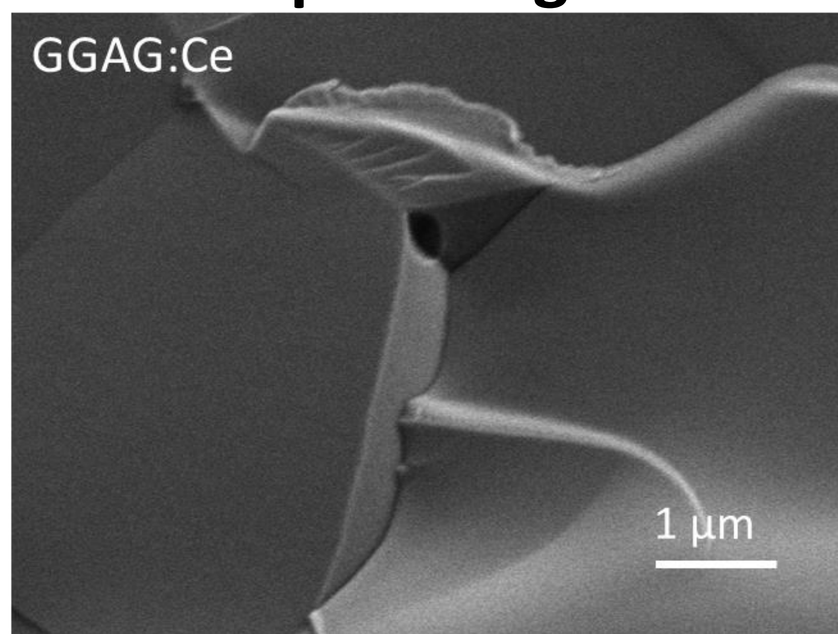




Uniaxial pressing

Slip-casting

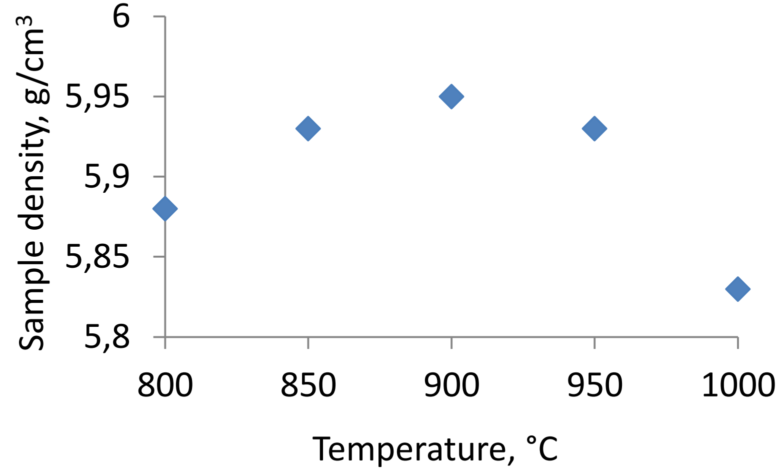




Powder synthesis & processing → Sintering

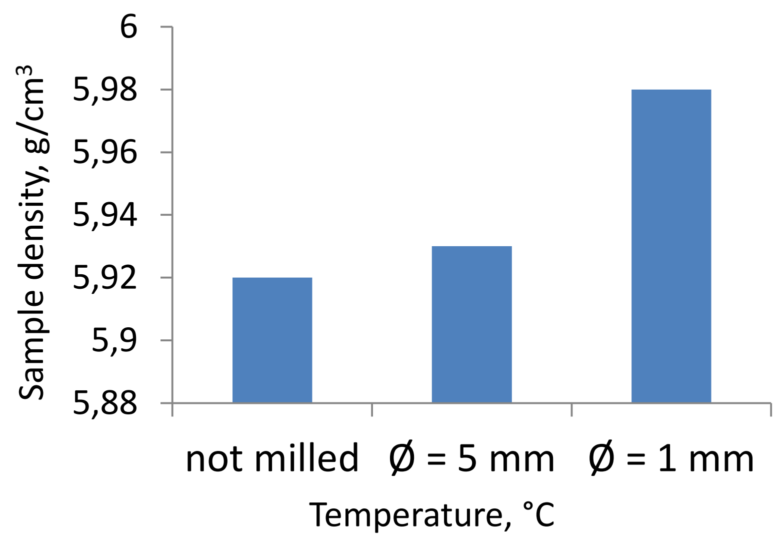
Initial powder of GYAGG- composition after calcination was milled by 1 mm Al_2O_3 milling bodies

Calcination temperature, °C	Ceramic density, g/cm³	% from theoretical (6,05 g/cm³)
800	5,88	97,2
850	5,93	98,0
900	5,95	98,3
950	5,93	98,0
1000	5,83	96,4



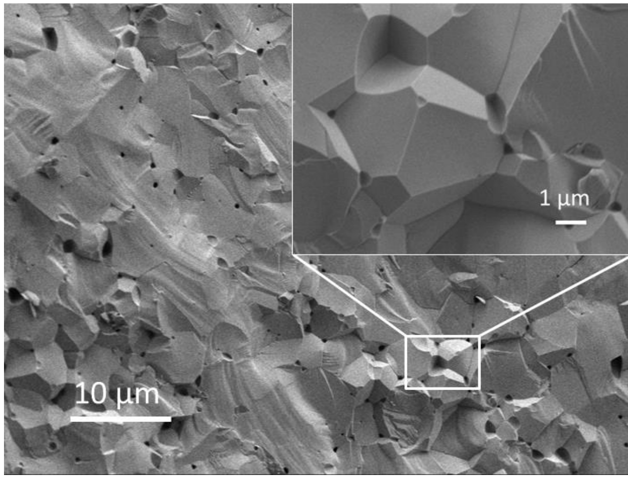
Initial powder of GYAGG- composition was calcined at 850 °C

Milling	Ceramic density, g/cm³	% from theoretical (6,05 g/cm³)
-	5,92	97,9
Planetary mill, Al_2O_3 milling bodies Ø = 5 mm	5,93	98,0
Planetary mill, Al_2O_3 milling bodies Ø = 1 mm	5,98	98,9

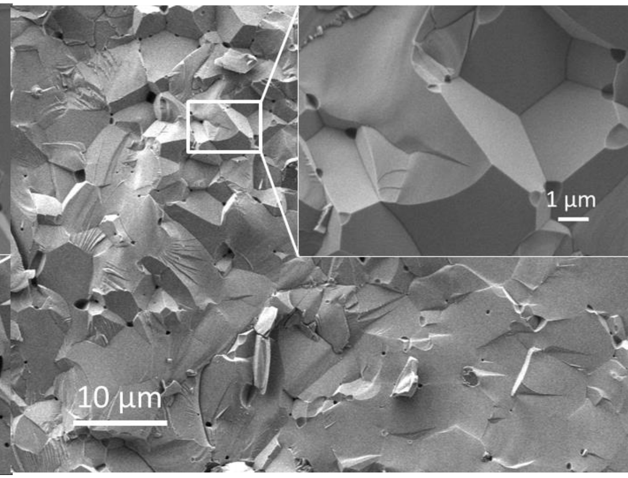


Powder synthesis & processing → Sintering

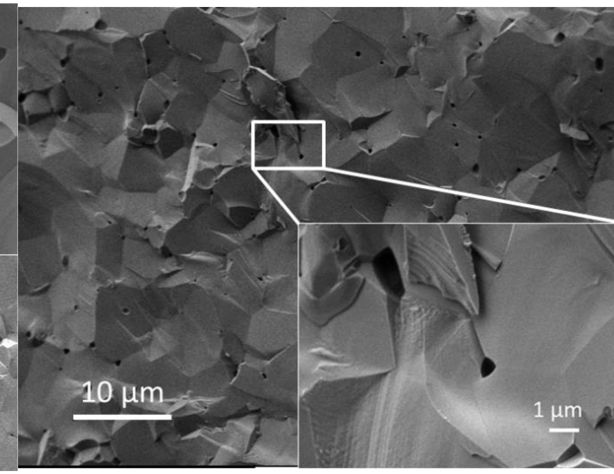
Initial powder of GYAGG:Ce composition was calcined at different temperatures, pressed and sintered in air atmosphere



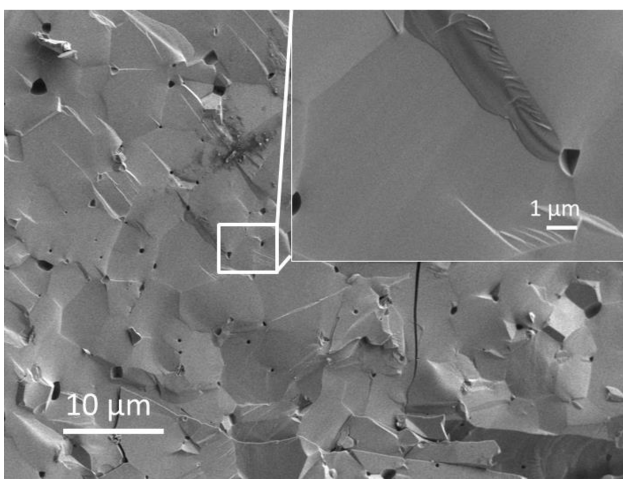
t° = 800 °C



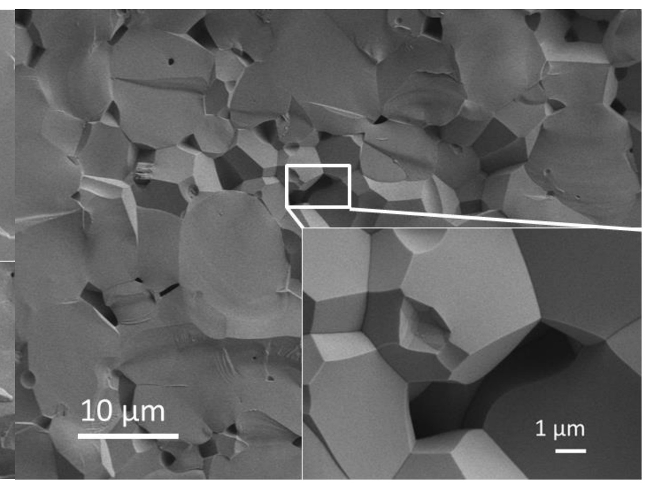
t° = 850 °C



t° = 900 °C



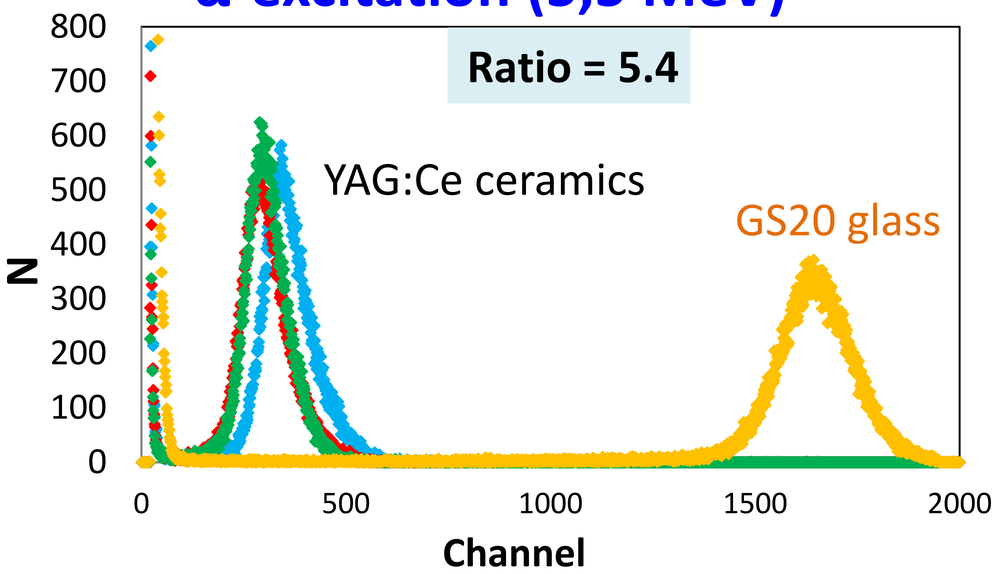
t° = 950 °C

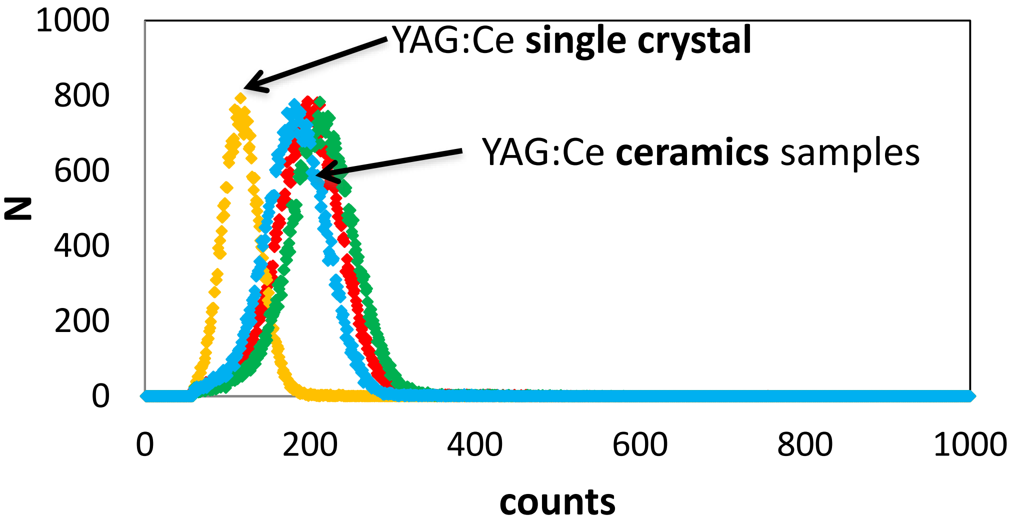


t° = 1000 °C

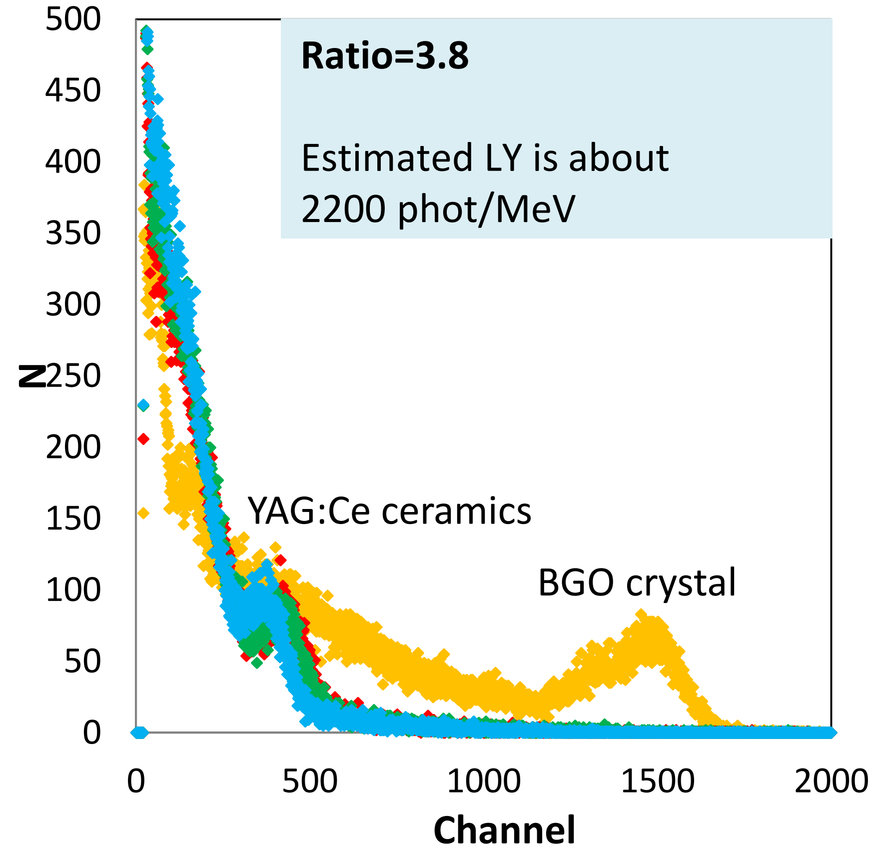
Scintillation properties of YAG:Ce in comparison to other scintillators

α-excitation (5,5 MeV)



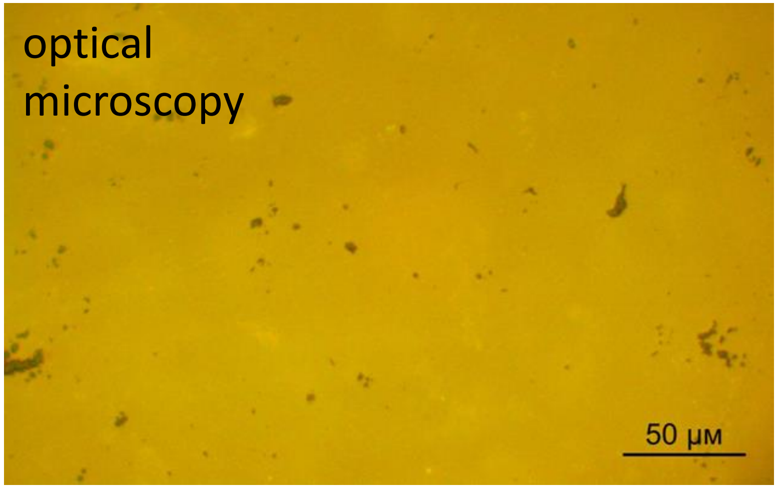


γ-excitation (662 keV), transmission



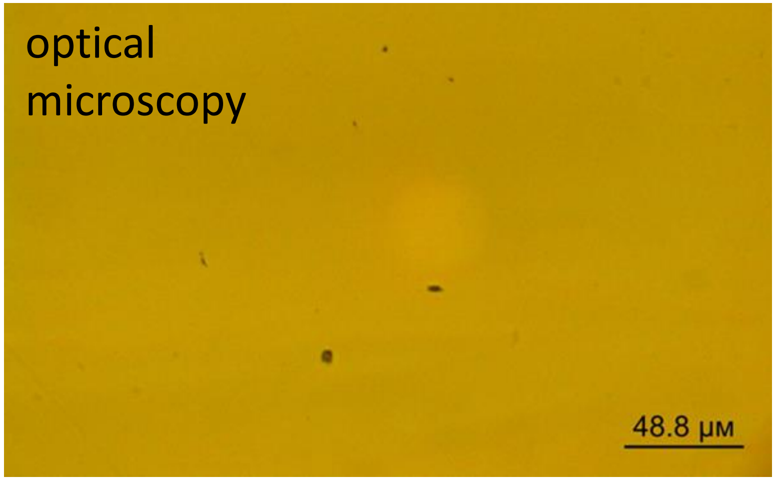
Sintering

Sintering in **air**

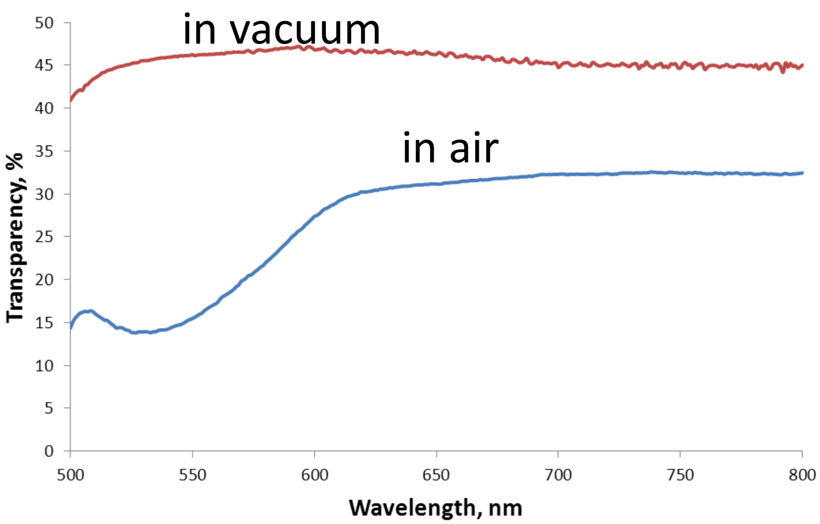


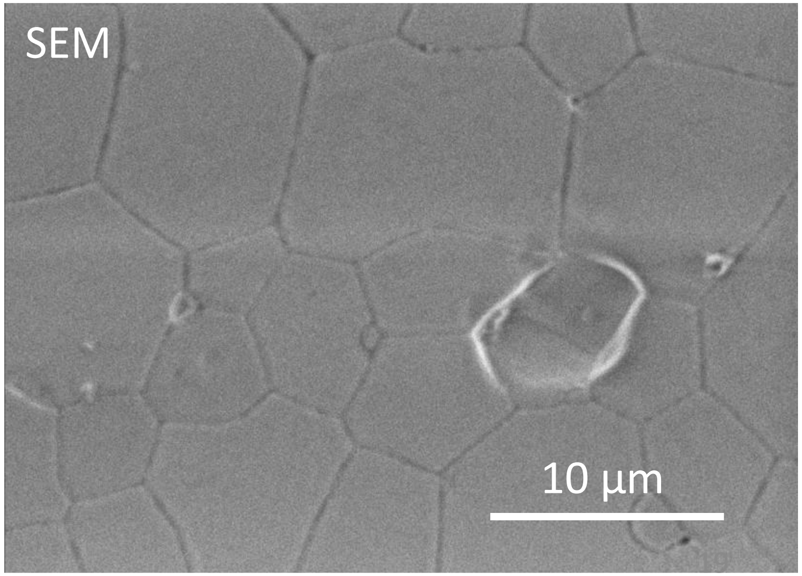
GYAGG:Ce, ~ 1,00% of pores
~ 30% of transparency

Sintering in **vacuum** ($p < 10^{-4}$ atm.)



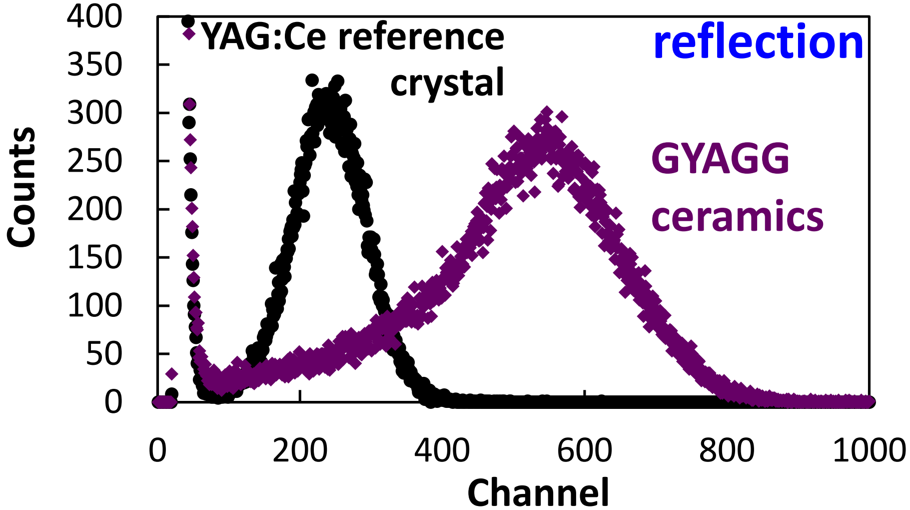
GYAGG:Ce, ~ 0,02% of pores
~ 45% of transparency

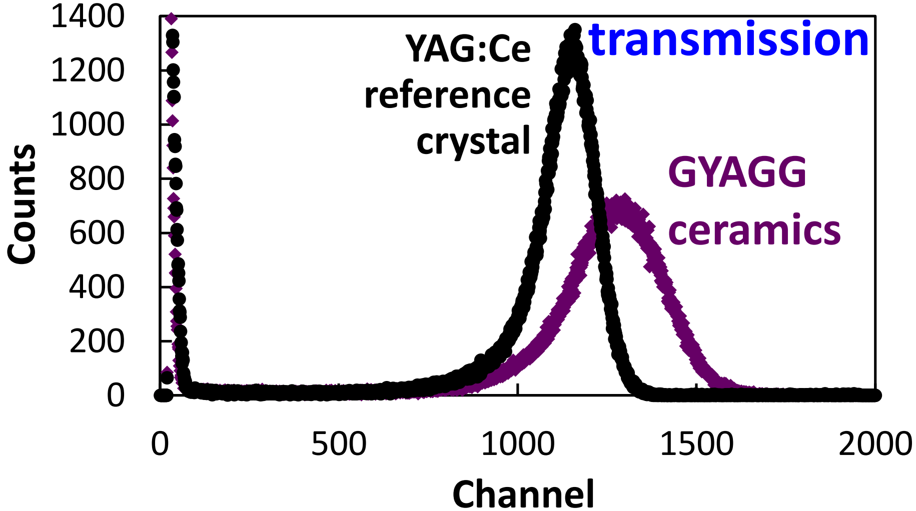




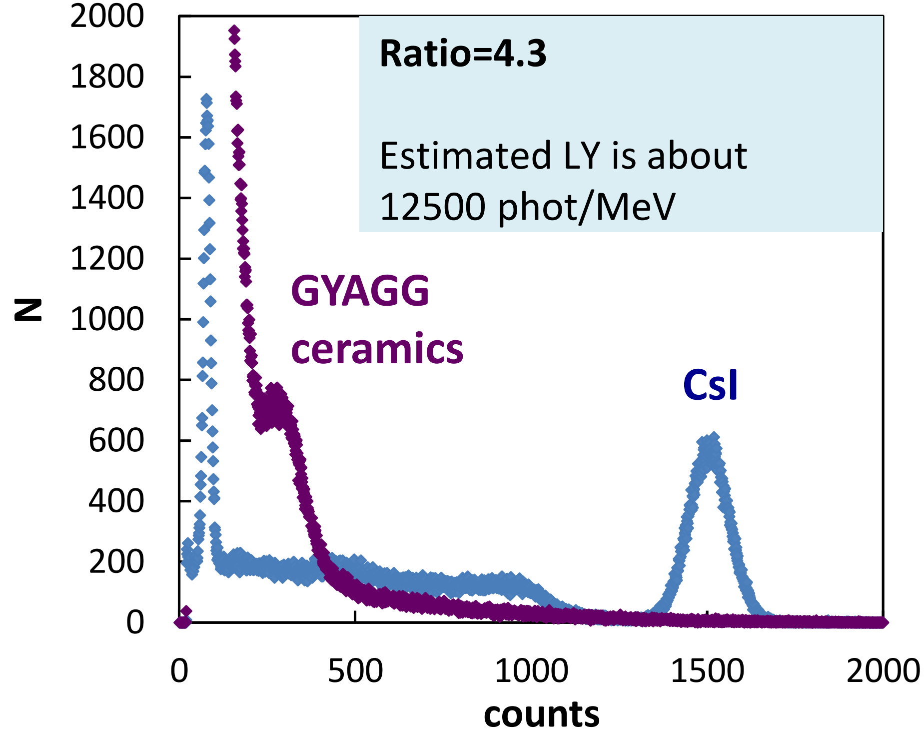
Measurements of translucent GYAGG ceramics in transmission (normal) geometry

α-excitation (5,5 MeV)



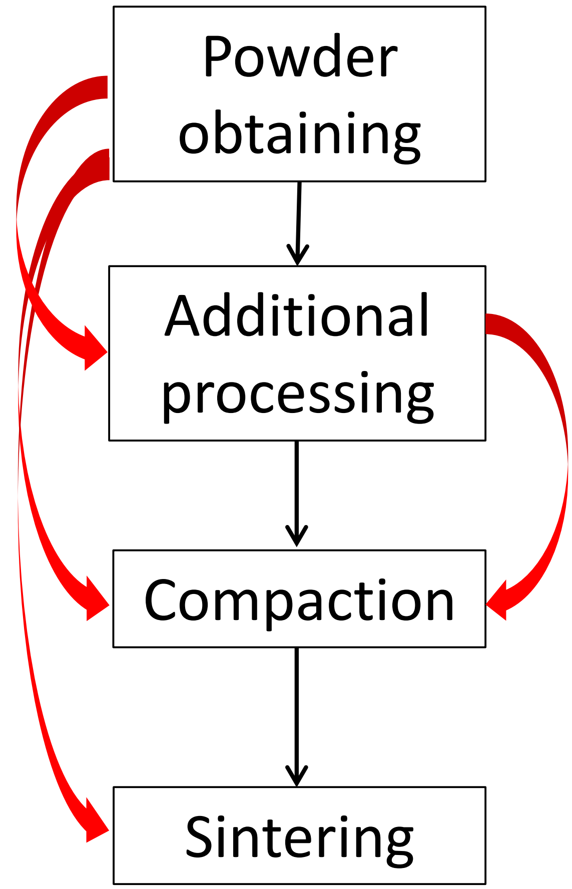


γ-excitation (662 keV), transmission

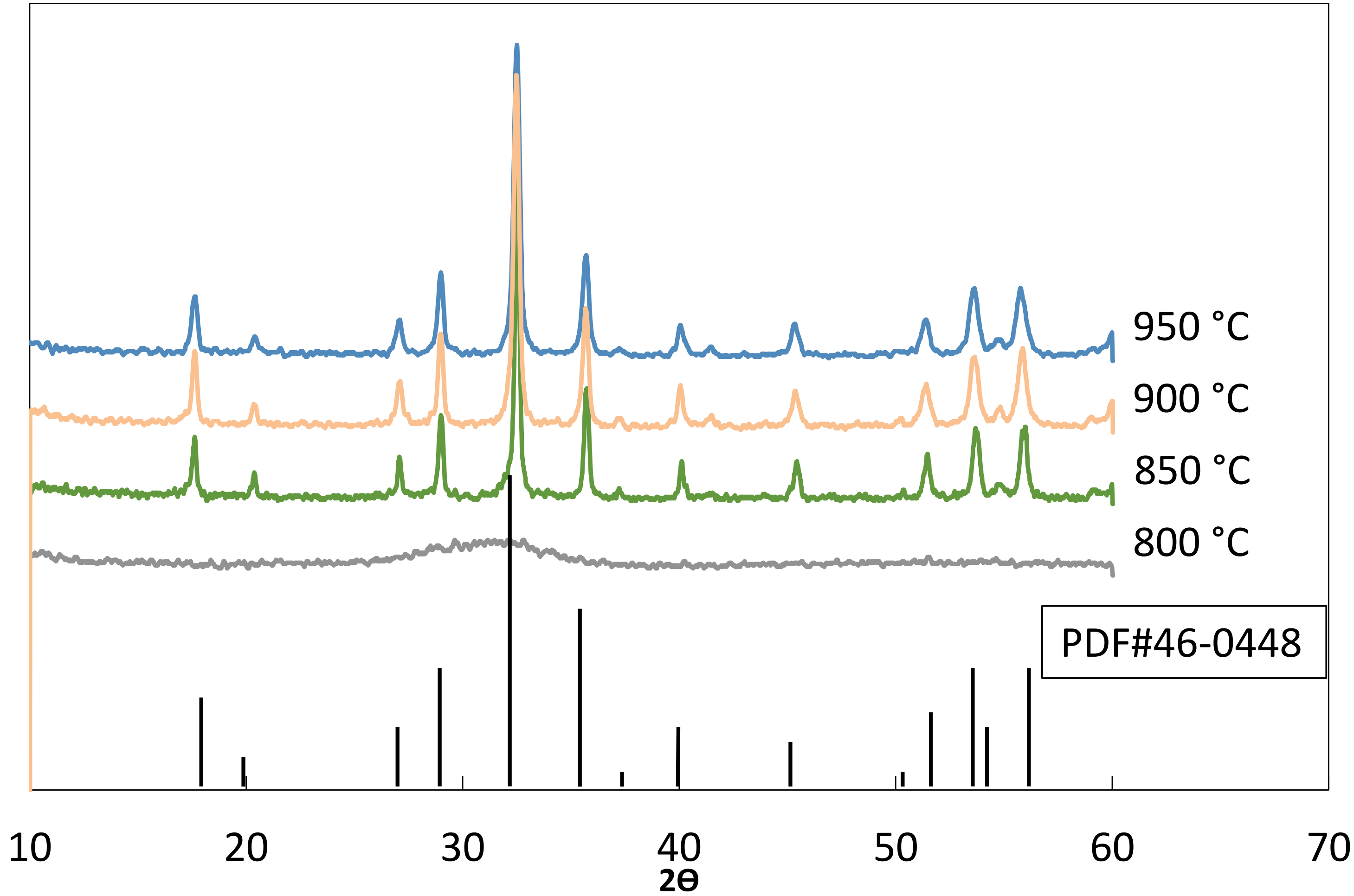


Conclusions

- Nature of precipitant could influence on microstructure of initial powders and density of final “green bodies” and ceramics
- Additional processing of obtained powders also influences on ceramic density
- Conditions of sintering are important for density and transparency of ceramics: sintering in vacuum could improve these parametrs
- Ceramic samples have better scintillation properties, compared to single crystal
- Composition variations could greatly improve light yield of obtained samples



Thank you for your attention!


950 °C
900 °C
850 °C
800 °C
PDF#46-0448
10
20
30
40
50
60
70
2Θ