$Zn_xMg_{1-x}WO_4$ - A new crystal scintillator

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Overview

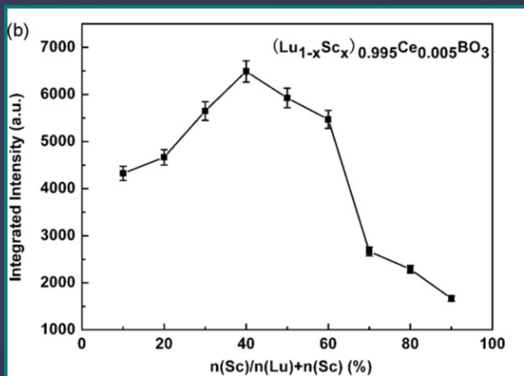
- Motivation
- The solid solutions preparation
- Properties of the new scintillator
- Summary

Motivation

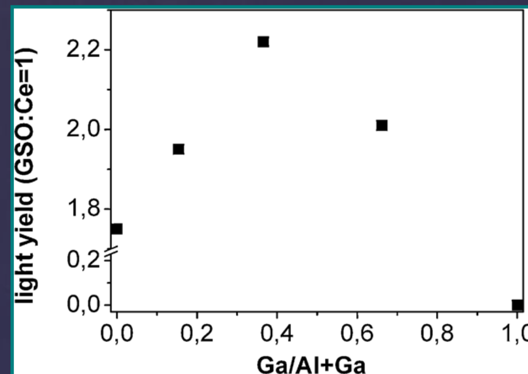
Requirements of 2β and Dark Matter searches to scintillator detectors

- a high light output at milli-Kelvin temperature;
- an extremely low level of radioactive contamination
- presence of specific nuclei (2β decay)
(for example $^{64,70}Zn$, ^{180}W, ^{186}W, ^{100}Mo)
- a variety of elements (heavy & light) in scintillation targets (Dark matter)
- diamagnetic properties (for bolometers)

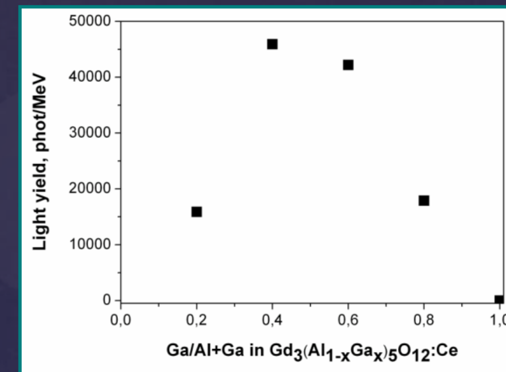
!Scintillation light output can be enhanced for the solid solutions of inorganic crystals



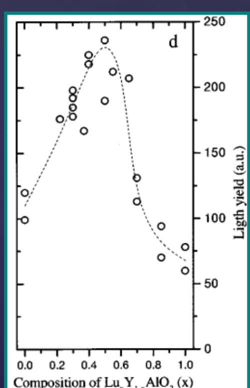
$(Lu,Sc)BO_3:Ce^{3+}$



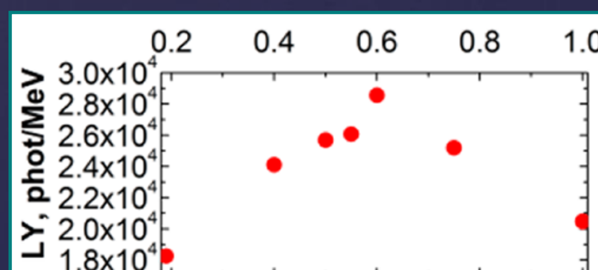
$Y_3(Al,Ga)_5O_{12}:Ce^{3+}$



$Gd_3(Al,Ga)_5O_{12}:Ce^{3+}$



$(Lu,Y)AlO_3:Ce^{3+}$



$(Lu,Gd)_2SiO_5:Ce^{3+}$

Belsky et al., IEEE TNS 48 (2001) 1095

Sidletskiy et al., Cryst. Gr. Des. 12 (2012) 4411

Wu et al., J. All. Comp. *509 (2011) 366*

Kamada et al., Cryst. Gr. Des. 11 (2011) 4484

Sidletskiy et al., MRB 47 (2012) 3249

! $ZnWO_4$ is one of the best candidate as a detector for cryogenic 2β decay and Dark Matter experiments

Large volume $ZnWO_4$ crystal scintillators up to ∅50 ×130 mm

Activity (mBq/kg)

	$ZnWO_4$(1)	$CaWO_4$(2)
^{232}Th	0.0015	0.7
^{235}U	≤ 0.003	1.6
^{238}U	0.002	300
Total α	0.18	400
^{40}K	≤ 0.4	
^{65}Zn	0.5	
^{90}Sr-^{90}Y	≤ 0.4	

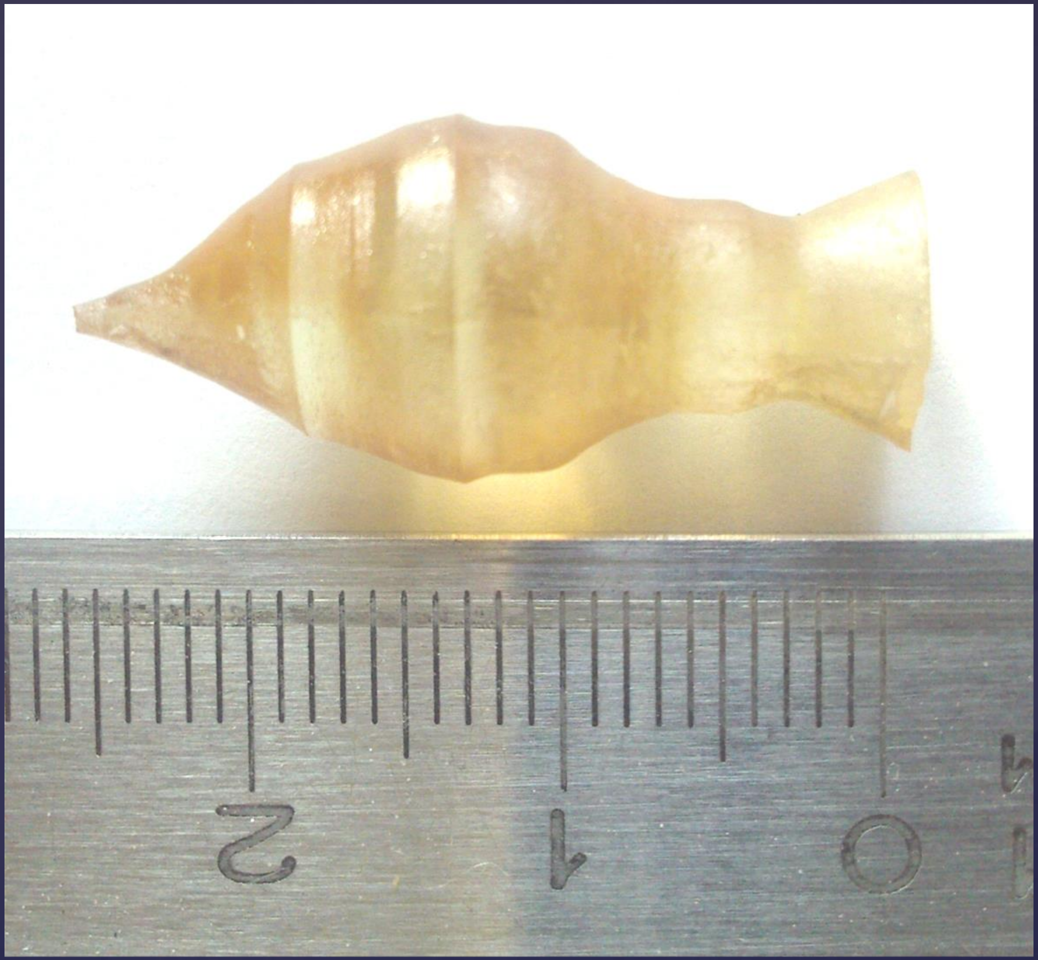
1) P. Belli, et al., NIM A 626-627 (2011) 31

2)NIMA 538 (2004) 657; APP 23 (2005) 249

The relative light output of $ZnWO_4$ at 10 K is 110-115 % that of $CaWO_4$

L. L. Nagornaya et al, "Growth of $ZnWO_4$ crystal scintillators for high sensitivity 2β experiments " IEEE Trans. Nucl. Sci. 55 (2008) 1469

$MgWO_4$ crystals for Dark Matter search



Cryogenic dark matter experi - ments require a variety of scintillation targets to verify the nature of a detected signal

The main difficulty of crystal growth from the melt is the presence in $MgWO_4$ of a phase transition below the melting point. Magnesium tungstate crystals of a few cm^3 volume were grown for the first time by pulling the seed from the melted flux solution

Radioactive contamination of the $MgWO_4$

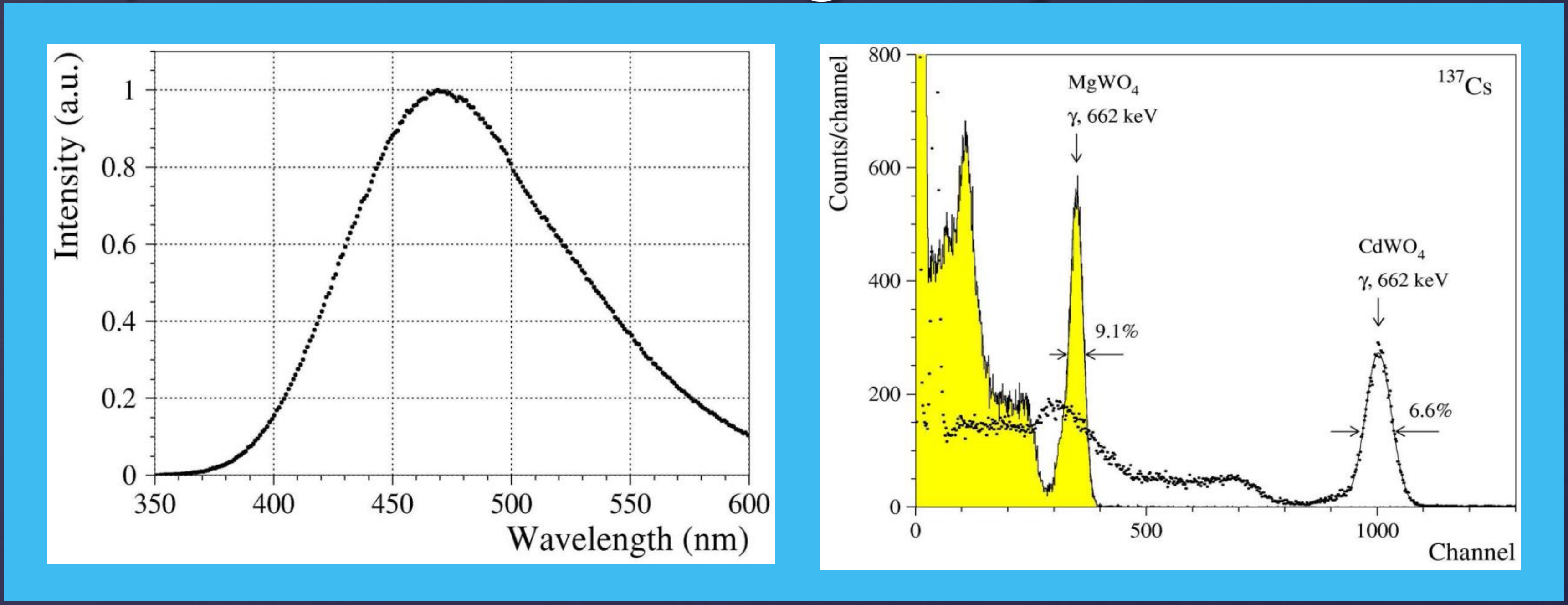
Radioactive contamination of the $MgWO_4$ crystal. Upper limits are given for $k = 1.645$ (or 90% C.L.).

Chain	Source	Activity ($Bq\ kg^{-1}$)
^{232}Th	^{232}Th	<0.28
	^{228}Th	<0.05
^{238}U	^{226}Ra	<0.05
	^{210}Pb	<2.4
	^{210}Po	5.7 ± 0.4
	^{40}K	<1.6
	$^{90}Sr-^{90}Y$	<1.5
	^{147}Sm	<0.54

F.A.Danevich et al,Nuclear Instruments and Methods in Physics Research A608(2009)107–115

$MgWO_4$ crystals are grown from a flux prepared from sodium tungstate, which caused contamination with radioactive impurities

XRL and Energy spectra of 662 keV ^{137}Cs γ-rays measured for $MgWO_4$



The crystal shows intense luminescence under X-ray excitation. The broad emission band exhibits maximum at 475 nm.

The relative photoelectron yield found to be 35% that of $CdWO_4$

The energy resolution of a small sample magnesium tungstate crystal scintillator giving 9.1 % for the 662 keV γ-quanta of Cs

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$ZnWO_4$ → monoclinic structure = $MgWO_4$ → monoclinic structure

→ ionic radius Zn^{2+} - 0.209 nm ≈ → ionic radius Mg^{2+} - 0.210 nm

continuous series of solid solutions of zinc and magnesium tungstate with different ratios of heavy zinc / light magnesium

The solid solutions preparation

Preparation of the charge

Raw material

ZnO - 99.995%

MgO -99.95%

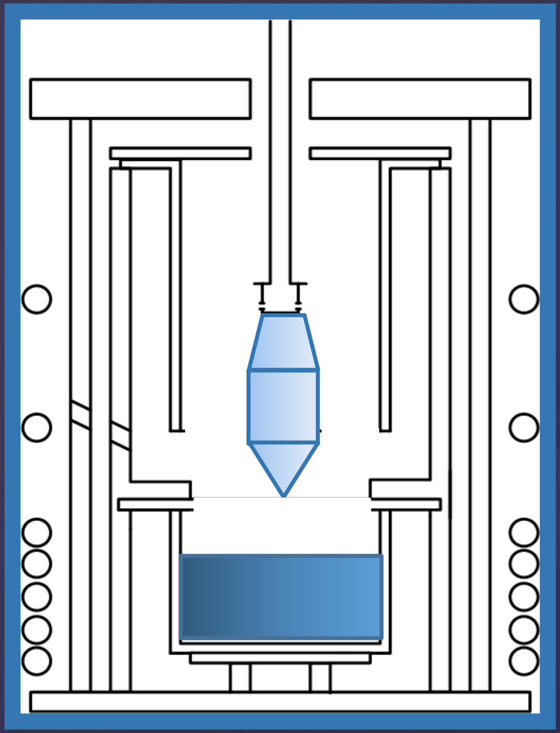
WO_3 - 99.995%

Solid phase synthesis

$$xZnO + (1-x)MgO + WO_3 \xrightarrow{900\ ^{\circ}C} \mathbf{Zn_xMg_{1-x}WO_4}$$

x = 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 0.95

Crystals growth



The single crystals of $Zn_xMg_{1-X}WO_4$ (x = 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 0.95) were grown by the Czochralski method

Growth Conditions

gradient $\Delta T_z \leq$ 10-20 deg/cm

pulling speed ν = 1.2-1.4 mm/hr

weight gain dm/dt = 2.0-2.5 g/hr

Analysis of the crystals composition

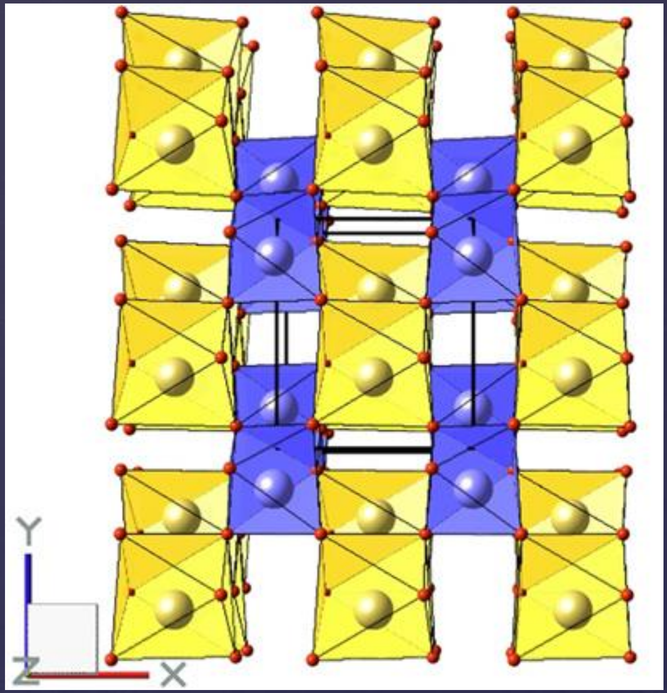
The intermediate values of x for all $Zn_xMg_{1-X}WO_4$ crystals were defined using the Scanning Electron Microscope with x-ray microanalysis. All values coincided with the ratio in the starting oxides

Impurities in the crystals

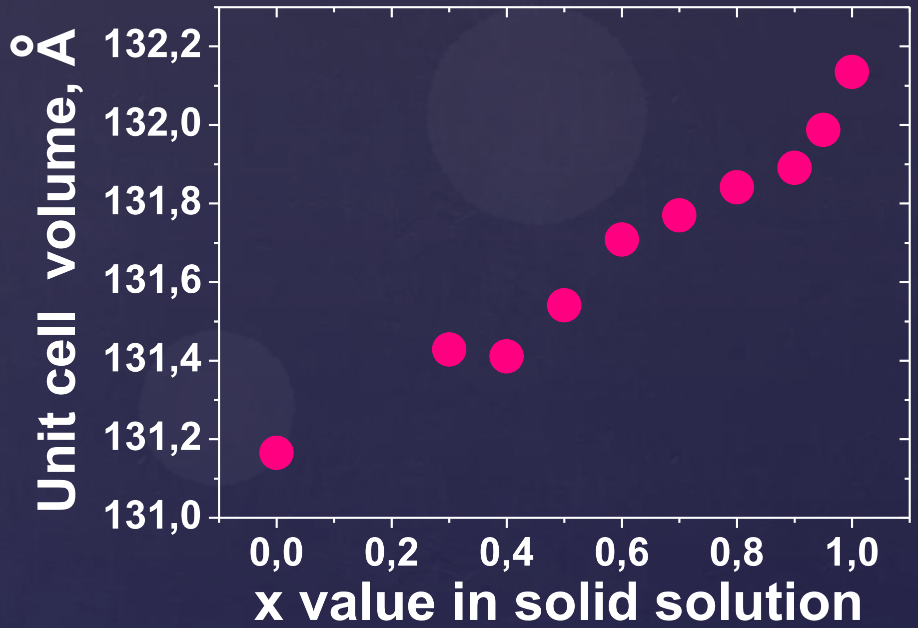
Sample	Element content, wt.%										
	Fe	Si	Mg	Cr	Sn	Ni	Mo	Pb	Al	Cu	Mn
$ZnWO_4$	0.0002	0.0005	0.001	0.0002	<0.0002	0.0001	0.001	0.0001	0.0002	0.00005	<0.0001
$Zn_{0.9}Mg_{0.1}WO_4$	0.0002	0.0005	-	0.00025	<0.0002	0.0001	0.0005	0.001	0.0002	0.0001	0.0003
$Zn_{0.8}Mg_{0.2}WO_4$	0.0002	0.0005	-	0.0002	<0.0002	0.0001	0.001	0.0001	0.00035	0.00005	0.0003
$Zn_{0.7}Mg_{0.3}WO_4$	0.00015	0.001	-	0.0002	<0.0002	<0.0001	0.001	0.001	0.0005	0.00005	0.0003
$Zn_{0.6}Mg_{0.4}WO_4$	0.00008	0.0005	-	0.0002	<0.0002	<0.0001	0.0005	0.0001	0.0002	0.00005	0.0003
$Zn_{0.5}Mg_{0.5}WO_4$	0.0001	0.0005	-	0.00025	<0.0002	<0.0001	0.0005	0.0001	0.0002	0.00005	0.00025
$Zn_{0.4}Mg_{0.6}WO_4$	0.0001	<0.001	-	<0.0002	<0.0002	<0.0002	<0.0002	0.00009	0.0002	<0.00005	<0.00005
$Zn_{0.3}Mg_{0.7}WO_4$	0.0001	<0.001	-	<0.0002	<0.0002	<0.0002	<0.0002	0.00008	0.0001	<0.00005	<0.00005
$MgWO_4$	0.001	<0.001	-	<0.0002	<0.0002	<0.0002	<0.0002	0.00006	<0.0001	<0.00005	<0.00005

Properties of the new scintillator

$Zn_xMg_{1-x}WO_4$ crystal structure

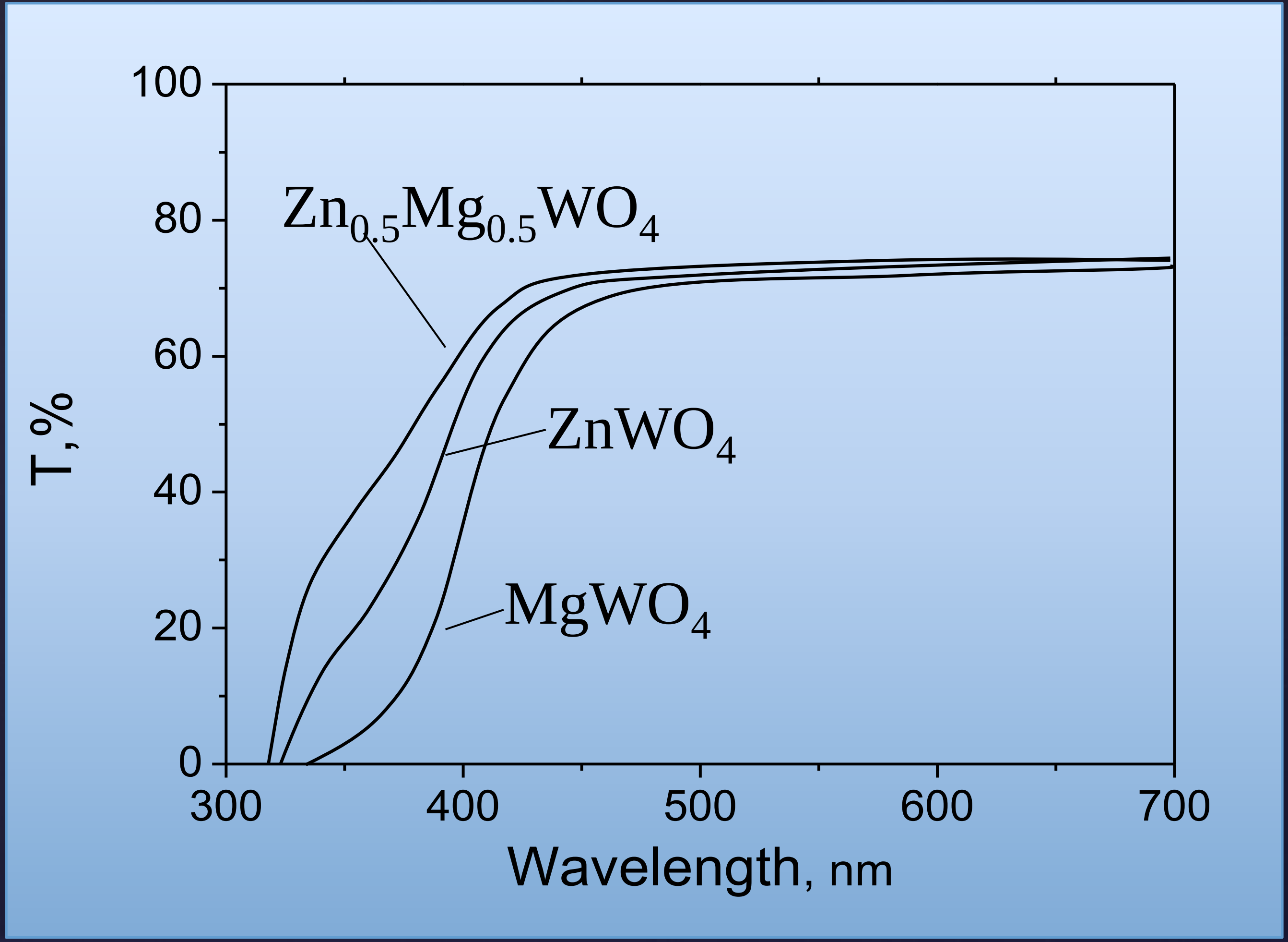


$Zn_xMg_{1-x}WO_4$ unit cell volume

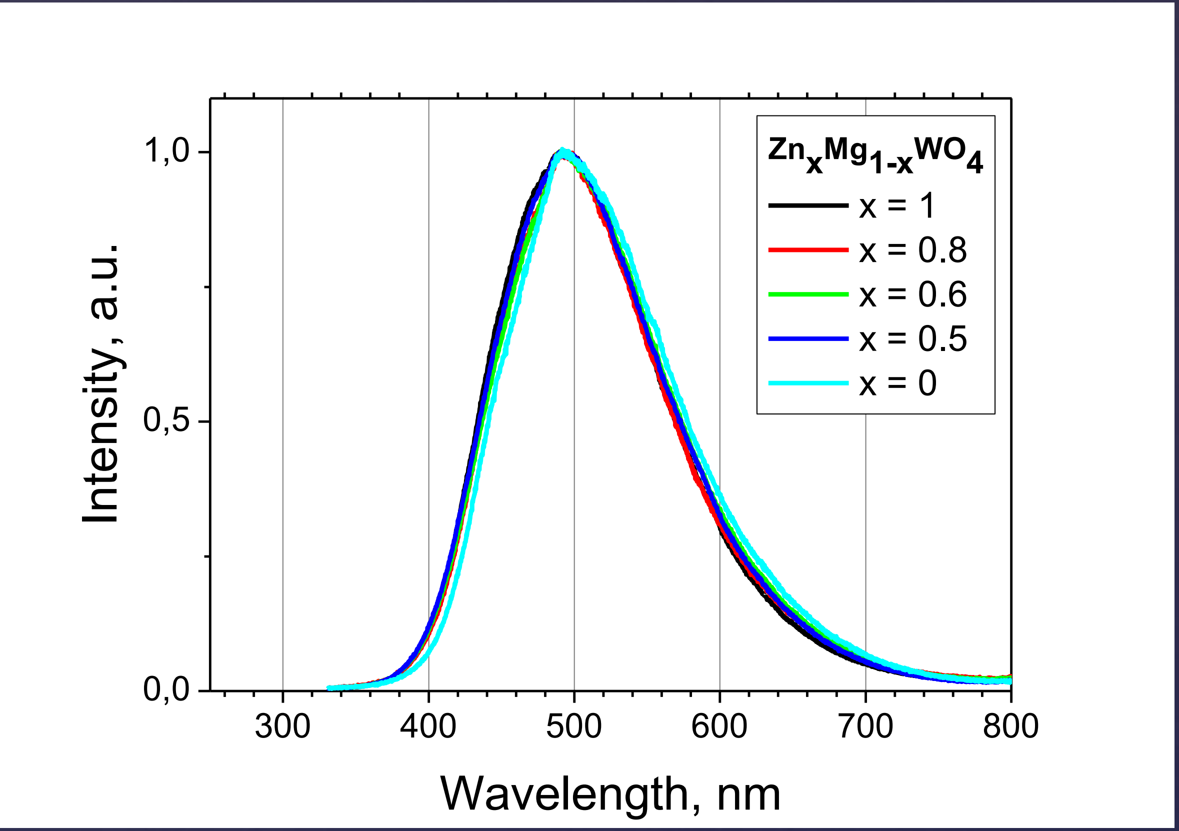


All samples of the solid solutions were wolframite structure (monoclinic)

Transmission spectra of tungstate crystals



Luminescence spectra of $Zn_xMg_{1-x}WO_4$



Normalized emission spectra under X-ray excitation, T = 300 K.

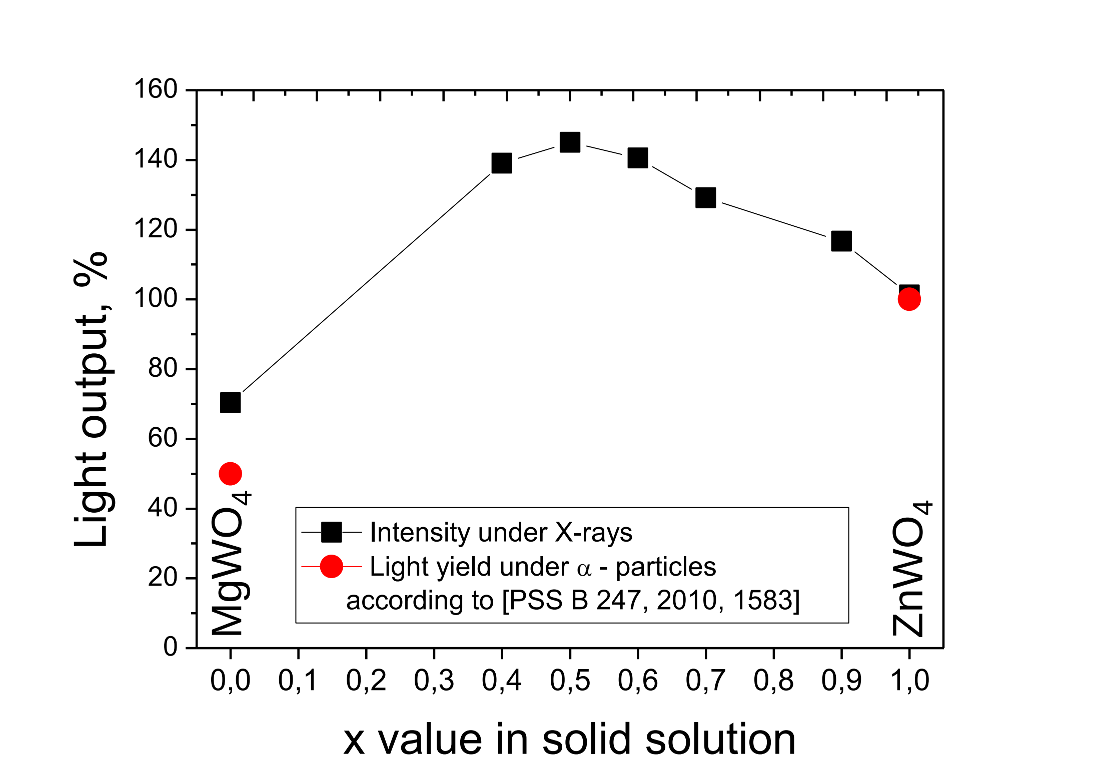
A single emission band is observed that is due to the emission of exciton, self-trapped on WO_6 complex.

The characteristics of emission center are similar for all of the studied solid solutions.

Decay times, E_{ex} = 4.1 eV

x	τ, mks
1	26
0.9	27
0.8	29
0.7	25
0.6	32
0.5	33
0	39

Light output of $Zn_xMg_{1-x}WO_4$



The effect of enhancement of light output is observed for intermediate values of x!

Summary

- Scintillation single crystals of $Zn_xMg_{1-x}WO_4$ solid solutions were obtained for the first time
- An increase of light output has been detected in $Zn_xMg_{1-x}WO_4$ solid solutions for intermediate values of x
- The first study of the properties of this scintillator shows its promise. Further studies of these scintillators are needed

Thanks a lot for your attentions !!!