Tb^{3+} Doped Silicoborate Glass Scintillator for High Resolution Synchrotron X-Rays Imaging Application

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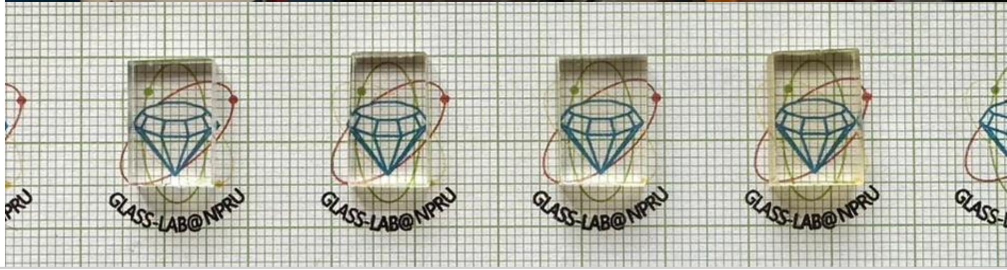


Research Background & Motivation

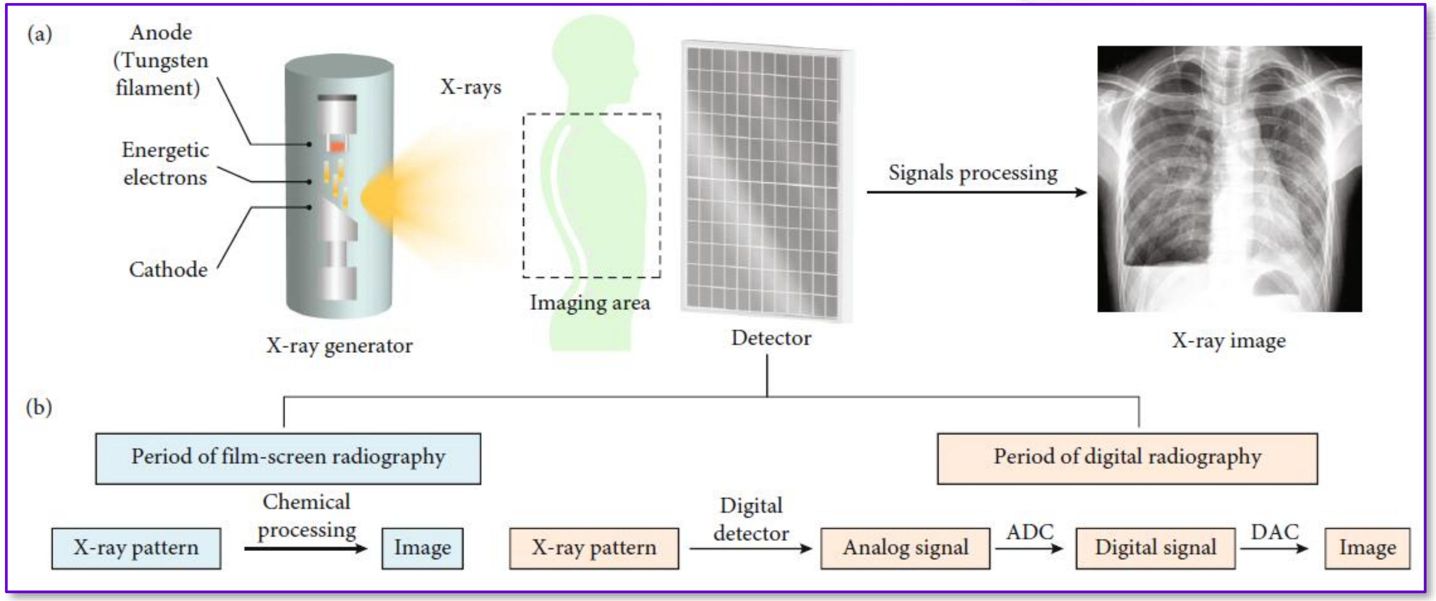


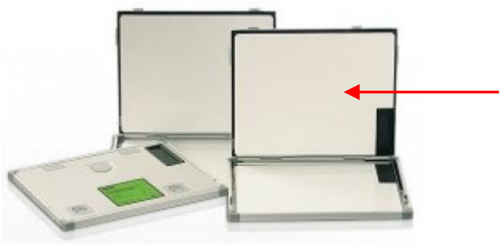
Figure 1: (a) Schematic illustration of an X-ray imaging system. **(b)** The development of X-ray radiography with the evolution of X-ray detectors.

Radiography is a medical imaging technique that uses X-rays to create images of the internal structures of the body.

- It is commonly used for diagnostic purposes to visualize bones, organs, and tissues.
- The X-ray machine emits a controlled amount of radiation, which passes through the body and is captured on a film or digital detector on the other side.

Research (2021), Article ID 9892152, https://doi.org/10.34133/2021/9892152

- **Intensifying screen**



Phosphor
- $CaWo_3$

Rare earth elements
- La_2O_2S
- LaBrO
- Gd_2O_2S

https://pjxray.com/green-400-speed-screens-and-cassettes-1417.html

- **Computed radiography (CR)**

Photostimulable phosphor (PSP) plate
- BaFBr: Eu^{2+}
- CsBr: Eu^{2+}

https://www.indiamart.com/proddetail/fuji-x-ray-cr-cassette-23371763791.html

- **Digital radiography (DR)**

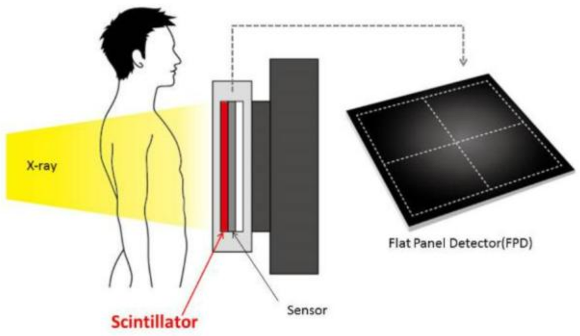
Material of scintillators
- CsI: Tl
- GOS: Tb (Terbium doped Gadolinium oxysulfide)

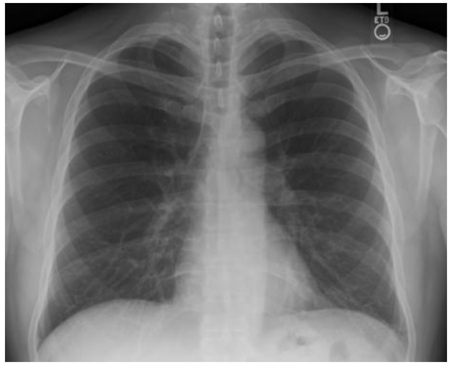
https://www.trademed.com/products/8577/Flat-Panel-Detector.html

Research Background & Motivation

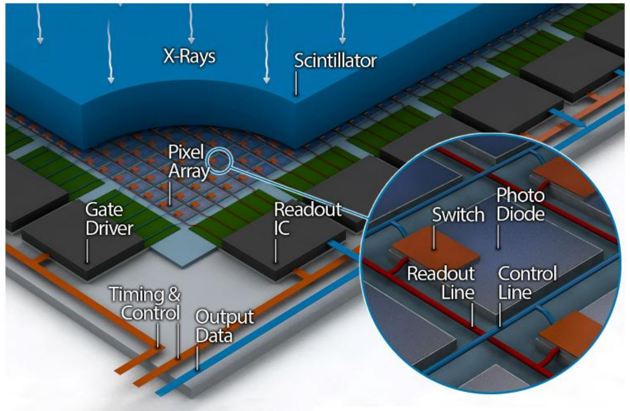
Detector in General Radiography

- **General Radiography**





- **Flat Panel Detector**



Scintillators – is a materials that spontaneously emit light at wavelengths within or near the visible region of the electromagnetic spectrum upon exposure to ionizing radiation

https://www.cappa.ie/photonic-applications/
https://ams.com/how-flat-panel-detectors-work

Types of scintillator

Types of scintillator	Advantages	Disadvantages
• Gas scintillator ^3He	Excellent γ/neutron discrimination	Expensive
• Single-crystal scintillators	High light yield	Difficult to grow
• Plastic scintillators	Easy to form	Low light output
• **Glass scintillators**	- Can be variety of size and shapes - **Doped with composition or lanthanide ions into the glass matrix; improve radiation absorption & tune emission wavelengths**	- Quite fragile and easily damaged - Light output weak; added activator to solve

J.D. Musgraves, et al. Handbook of glass 2019

Research Background & Motivation

- **Lanthanides in the periodic table with atomic number and weight**

58	59	60	61	62	63	64	65	66	67	68	69	70	71
Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
140.115	140.9076	144.24	(145)	150.36	151.965	157.25	158.9253	162.50	164.9303	167.26	168.9342	173.04	174.967

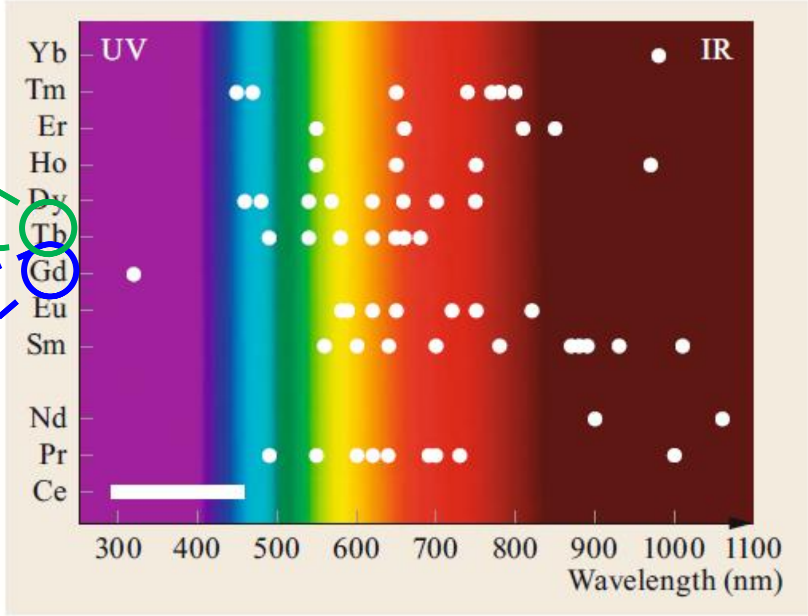
❖ Dopants, which serve as activators

create the energy gap in luminescence center resulting to..

- ✓ **Increase efficiency**
- ✓ **Control emission characteristics**
- ✓ **Enhance luminescence properties**

Good for green display device and suitable for the X-ray detecting material

Good interaction with radiation and high efficiency of energy transfer to Ln^{3+} ions.



Wavelengths of the main emissive transitions of the trivalent lanthanide (Ln^{3+}) coordination compounds in the near-UV to NIR range.

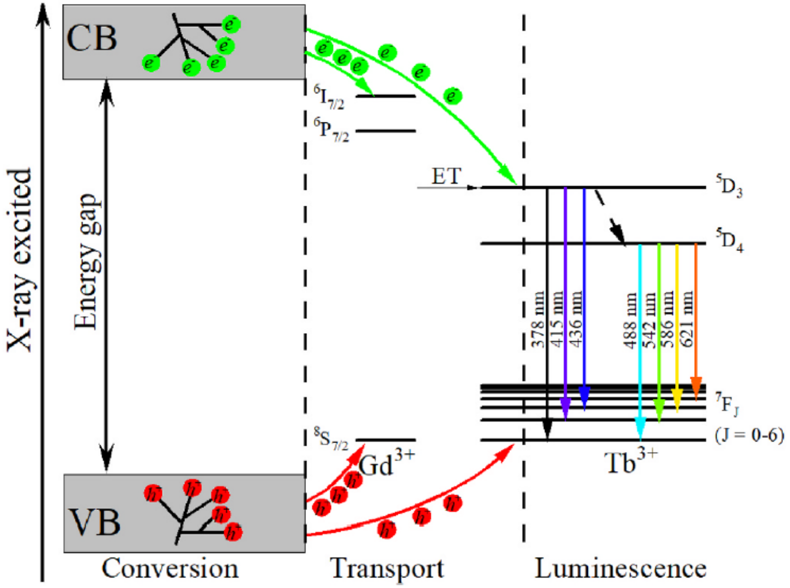


Fig. 6. Schematic diagram scintillating mechanism for ABSG:*x*Tb glasses, where VB denotes valance band and CB denotes conduction band.

References

J. David Musgraves, et al., Springer Handbook of Glass, 2019, https://doi.org/10.1007/978-3-319-93728-1

Palvi Gupta, et al., 2015, Mater. Res. Express, https://doi.org/ 10.1088/2053-1591/2/7/076202

WenJun Huang, et al., Ceramics International 48 (2022) 17178–17184, doi.org/10.1016/j.ceramint.2022.02.274

Objectives

- To develop Tb^{3+} doped silicoborate glass to be used as a glass scintillator.

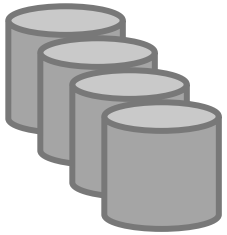
- To investigate the physical, optical, luminescence properties of Tb^{3+} doped silicoborate glass.

- To study the possibility of glass to be used as a scintillator, carried out based on the “Synchrotron radiation X-ray tomographic microscopy (SRXTM)” technique.

Methodology

❑ *Glass formula: $xTb_2O_3 : 40Na_2O : 7.5Gd_2O_3 : 5SiO_2 : (47.5\text{-}x)B_2O_3$ (x = 0, 1, 2 and 3 mol%)*

Weigh chemicals according to the formula



Alumina crucibles
15 g./batch

Melting in electrical furnace at **1400°C / 3 hrs.**

Pouring at room temperature

Quenching at room temperature

Annealing at **500°C / 3 hrs.**

Cutting and Polishing

Glass sample
Dimension:
$1 \times 1.5 \times 0.3$ cm^3

❑ *Investigation its property of…*

- **Physical and optical properties**
  - ➢ Density, Molar volume
  - ➢ Absorption spectra
- **Luminescence properties**
  - ➢ Photoluminescence (PL) → Excitation & Emission spectra, Decay time
  - ➢ X-ray induced luminescence
  - ➢ PLQY
- **X-ray imaging**

Research Results

Physical and optical property

❖ Density and molar volume

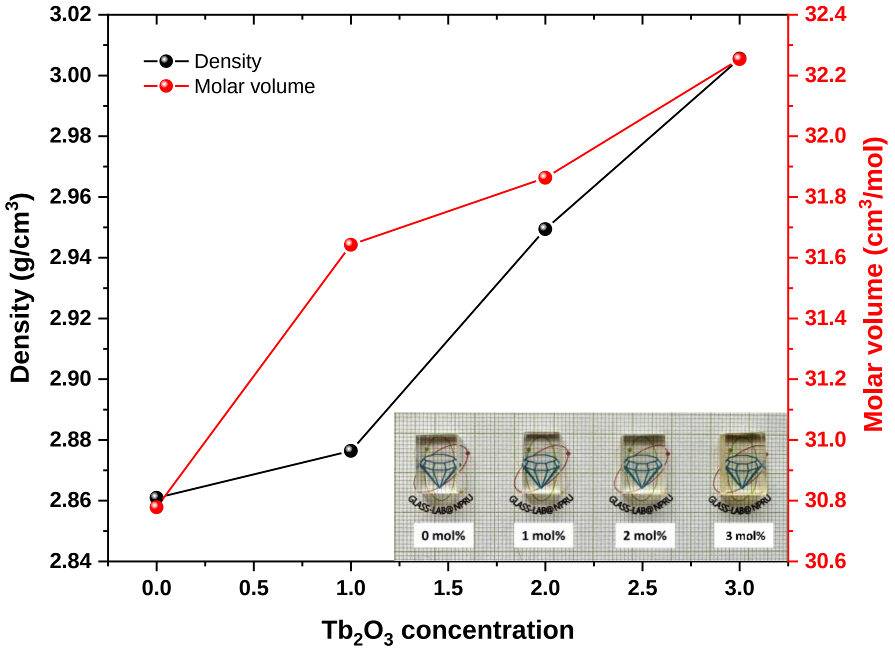


Fig.1. Density and Molar volume of xTb:7.5Gd glasses

❖ Absorption spectra measured by UV-Vis-NIR spectrometer

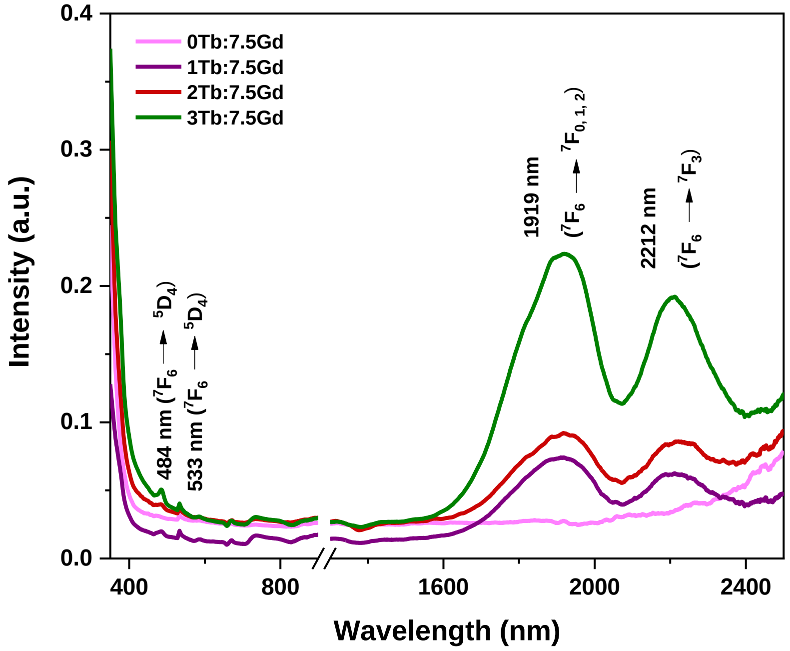


Fig.2. Vis-NIR absorption spectra of xTb:7.5Gd glasses

- **Density and molar volume** increases with Tb_2O_3 concentrations increasing; B_2O_3 components were replaced by Tb_2O_3.
- Tb^{3+} can absorption the light at Vis and NIR region. **Absorption spectra** of these wavelengths increased with increasing concentration of Tb_2O_3.

Materials Today: Proceedings 43 (2021) 2635–2640
Hugo R. Fernandes., et al., Materials 2018, 11, 2530; doi:10.3390/ma11122530
WenJun Huang, et al., Ceramics International 48 (2022) 17178–17184, doi.org/10.1016/j.ceramint.2022.02.274

Research Results

Luminescence property

- Photoluminescence spectra measured by Agilent Cary Eclipse fluorescence spectrometer

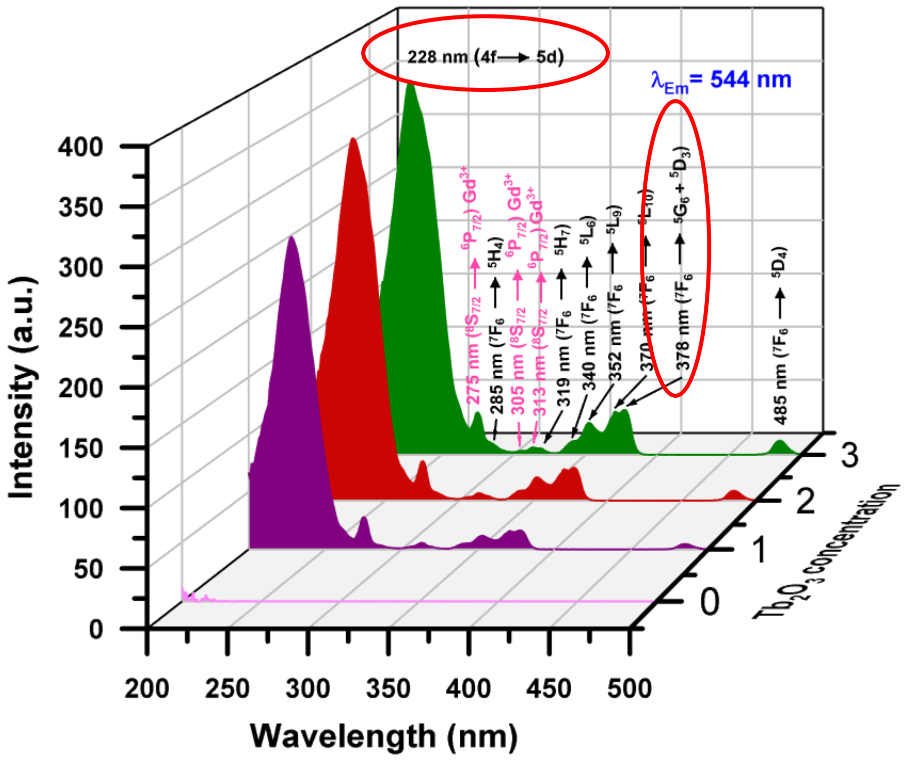


Fig.3. Excitation spectra of xTb:7.5Gd glasses under λ_{Em} = 544 nm

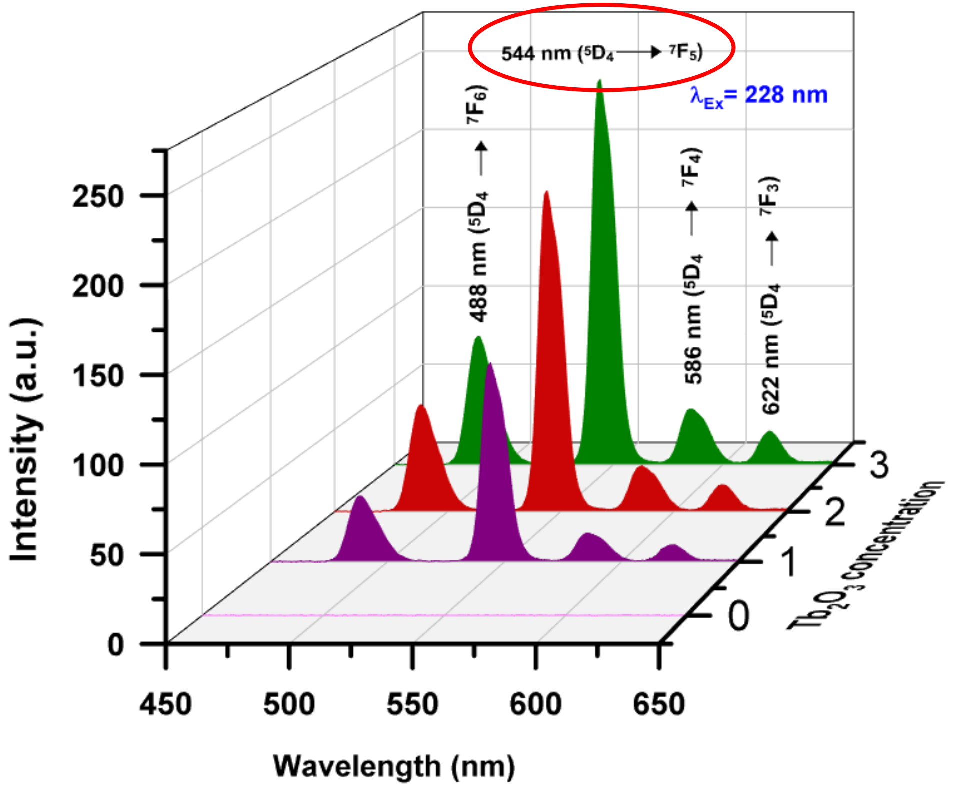


Fig.4. Emission spectra of xTb:7.5Gd glasses under λ_{Ex} = 228 nm

- The emission spectra under 228 nm excitation showed the strong green emission peaks at 544 nm corresponding to $^5D_4 \rightarrow {}^7F_5$ transition in 3.0 mol% of Tb^{3+}-doped glass.

Research Results

Luminescence property

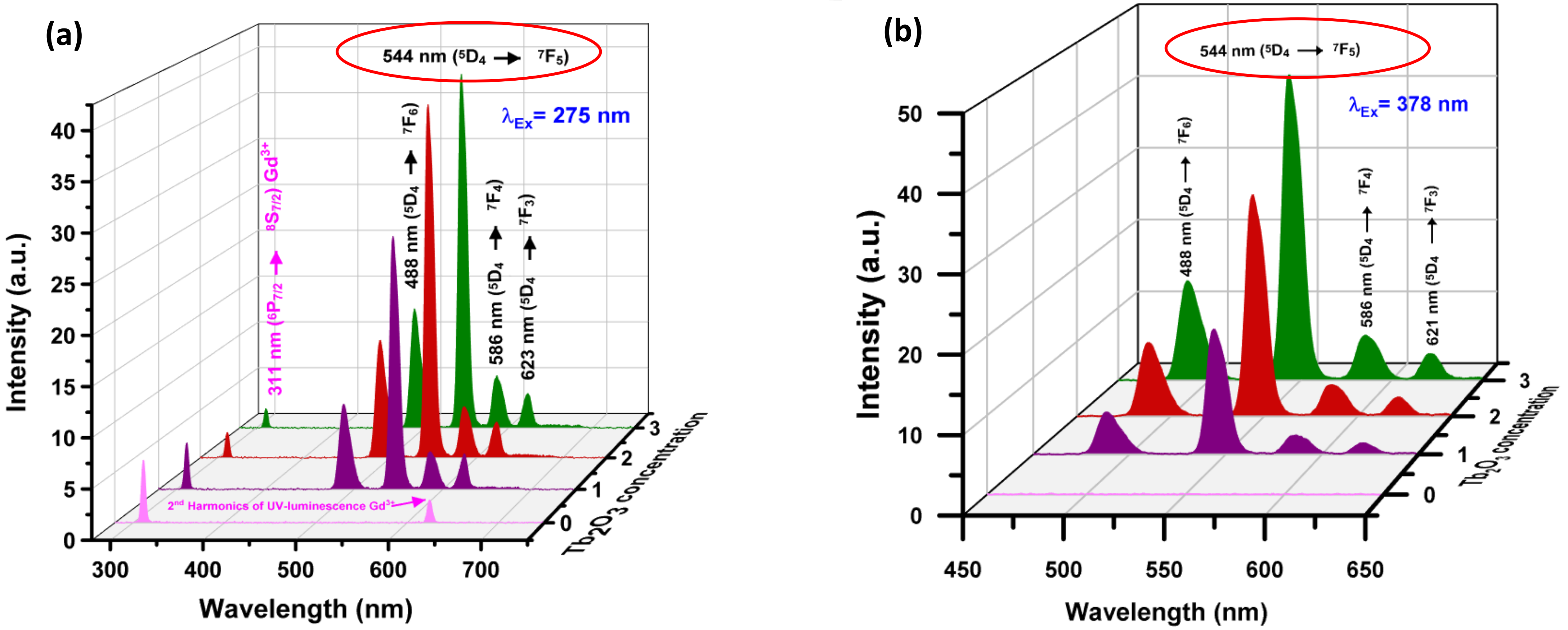


Fig.5. Emission spectra of xTb:7.5Gd glasses under λ_{Ex} = 275 nm (a) and 378 nm (b)

- The additional peak centering at 311 nm describes the Gd^{3+} transition emission under $^6P_{7/2} \rightarrow {}^8S_{7/2}$ while the additional peak at 623 nm is the half-harmonic of 275 nm.

Research Results

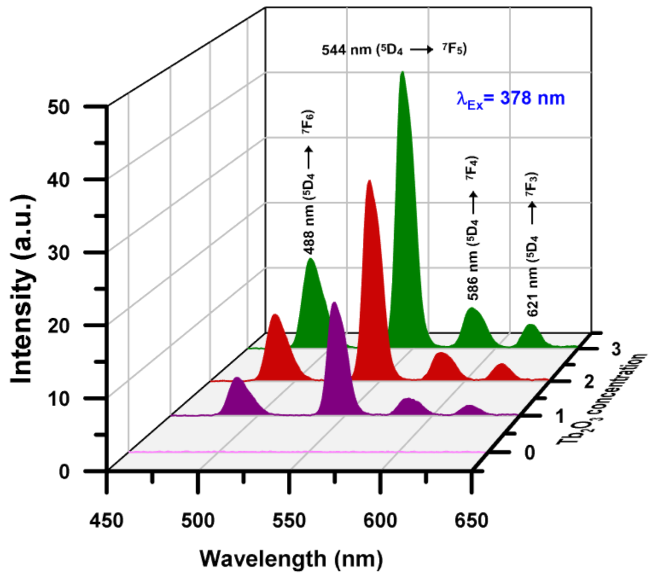
Experimental photoluminescence quantum yield (PLQY)

- Photoluminescence quantum yield was utilized by spectrofluorometer with an excitation source of a xenon lamp and K-integrating sphere

Table 1

The experimental photoluminescence quantum yield (PLQY) of xEu:7.5Gd glasses under λ_{Ex}= 378 nm at the wavelength of 400 – 650 nm.

Glass	Experimental PLQY (%)	Abs Error	Re Error
1Tb:7.5Gd	22.65	-	-
2Tb:7.5Gd	26.79	0.108	0.004
3Tb:7.5Gd	30.85	0.081	0.003



- This represents high-quality luminescence materials and indicates efficient light emission for a given amount of absorbed energy.

Research Results

X-ray excitation spectra

- X-ray induced luminescence spectra irradiated by X-ray, setting as 50 kV and 30 mA at NPRU, Thailand

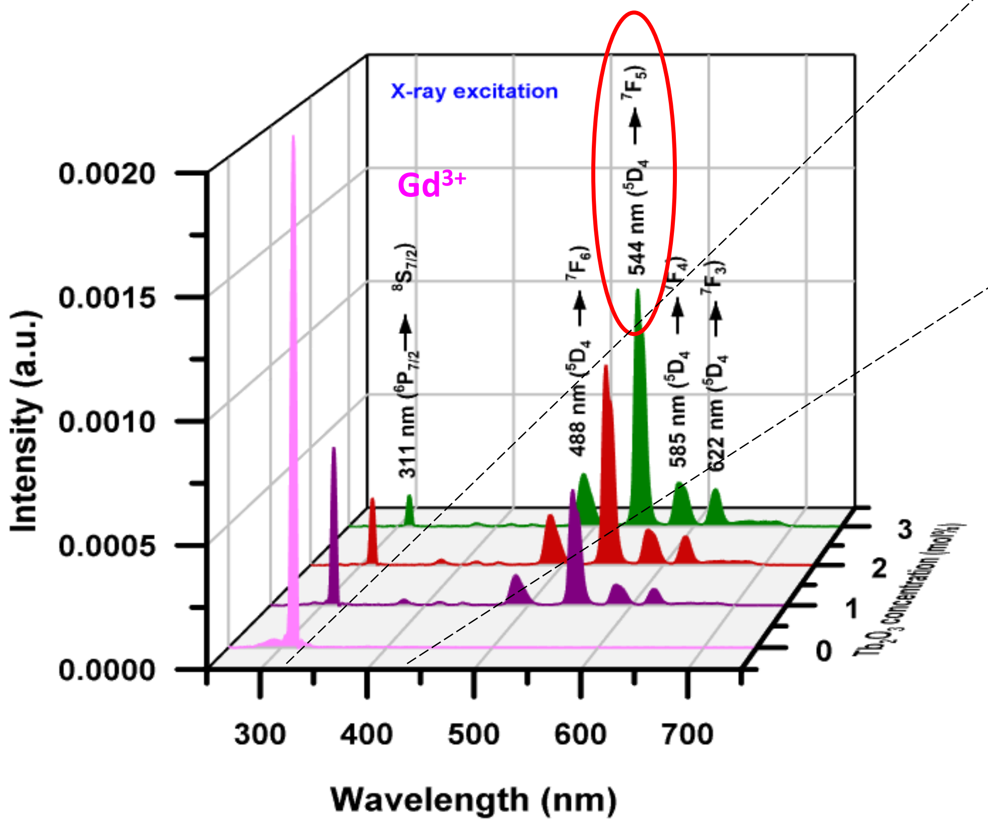
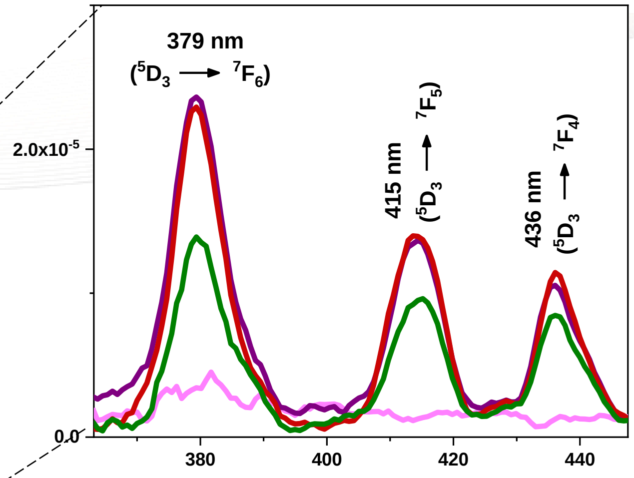
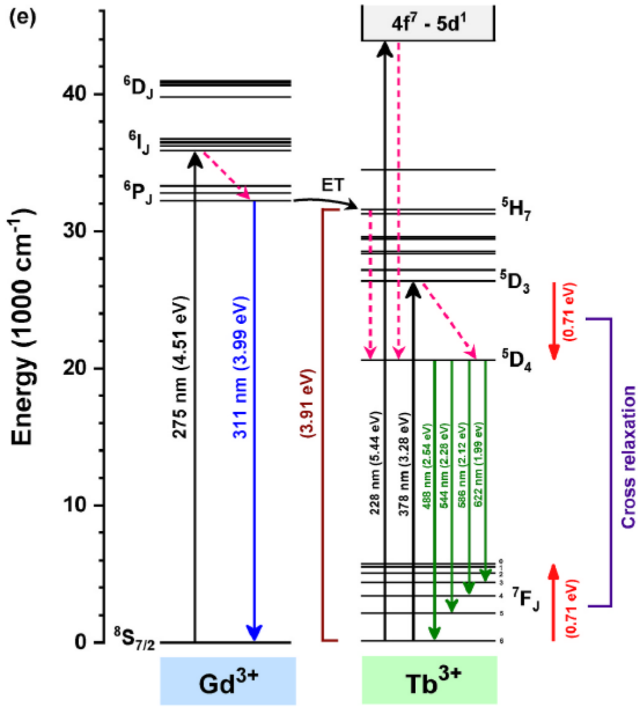


Fig.6. X-ray excitation spectra of xTb:7.5Gd glasses



- The emission intensity at $^5D_4$ increases with adding Tb_2O_3 concentrations while $^5D_3$ decreased



Research Results

X-ray excitation spectra and CIE chromaticity diagram

- Comparing the total integrated area of 3Tb:7.5Gd glass and BGO

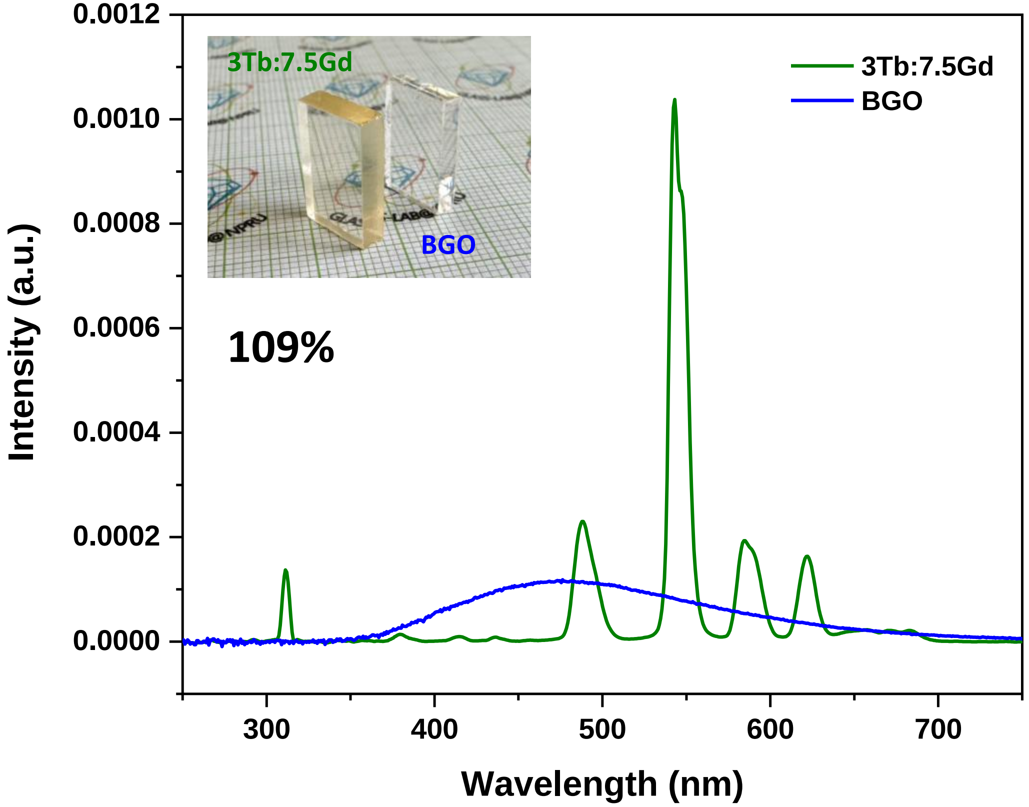


Fig.7. X-ray excitation spectra of 3Tb:7.5Gd compared with BGO

- The color of emitted light investigated by the International Commission on Illumination (CIE 1931), using emission data

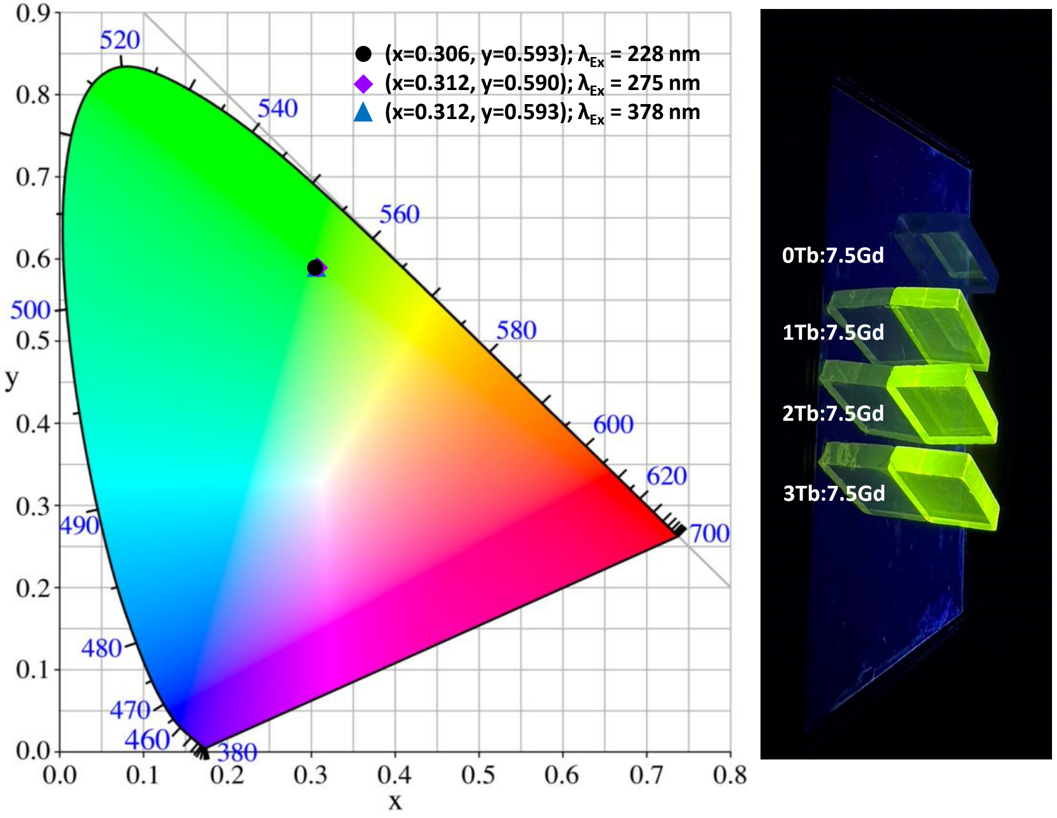


Fig.8. CIE chromaticity of xTb:7.5Gd glasses

Research results

Decay curves and Energy transfer efficiency

❖ Decay curves

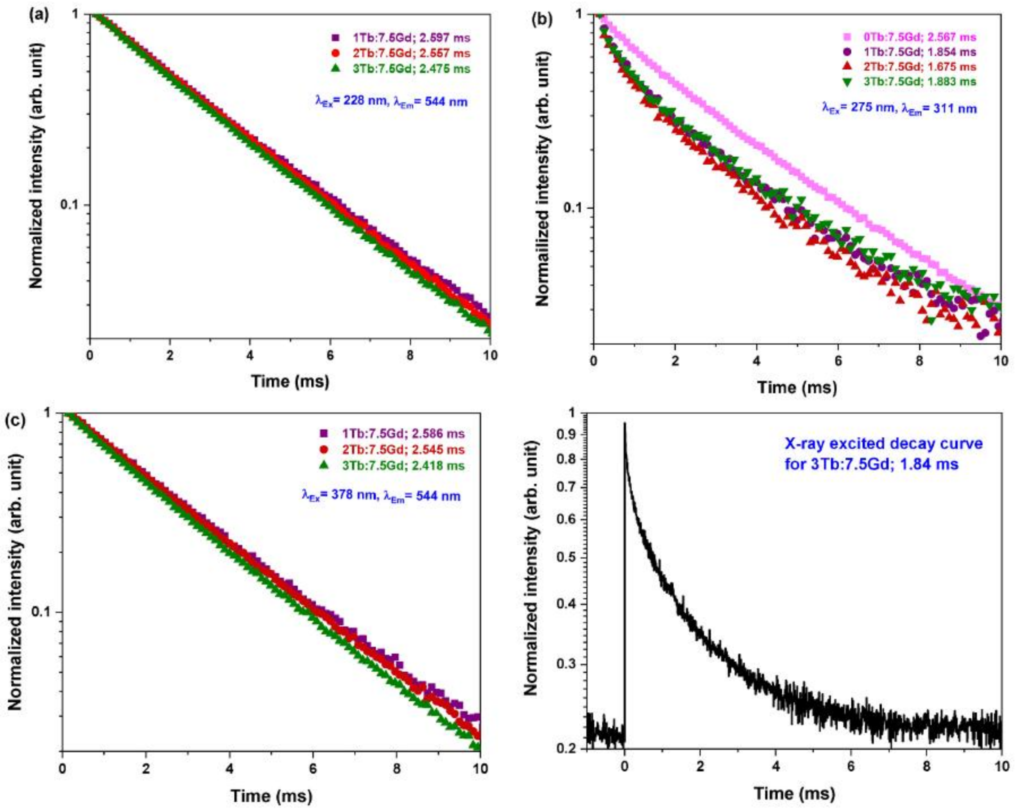


Fig.9. Decay time of the xTb:7.5Gd glasses, excitation at (a) 228 nm (b) 275 nm (c) 378 nm and (d) X-rays excitation.

❖ Energy transfer efficiency (η_{ET})

$$\eta_{ET} = 1 - \frac{\tau_{Gd-Tb}}{\tau_{Gd}}$$

τ_{Gd-Tb} = the decay time of the donor (Gd^{3+}) in the presence of the acceptor (Tb^{3+})

τ_{Gd} = the decay time of the donor in the absence of the acceptor

Table 2 The emission decay time (τ) under λ_{Ex}=275 nm, λ_{Em}=311 nm and energy transfer efficiency (η_{ET}) for xTb:7.5Gd glasses

Glass sample	τ (ms)	η_{ET} (%)
(τ_{Gd})		
0Tb:7.5Gd	2.567	-
(τ_{Gd-Tb})		
1Tb:7.5Gd	1.854	28
2Tb:7.5Gd	1.675	35
3Tb:7.5Gd	1.883	27

Research Results

X-ray imaging

;Experiment at BL1.2W: X-ray Imaging & X-ray Tomographic Microscopy (XTM), Synchrotron Light Research Center, Nakhon Ratchasima, Thailand

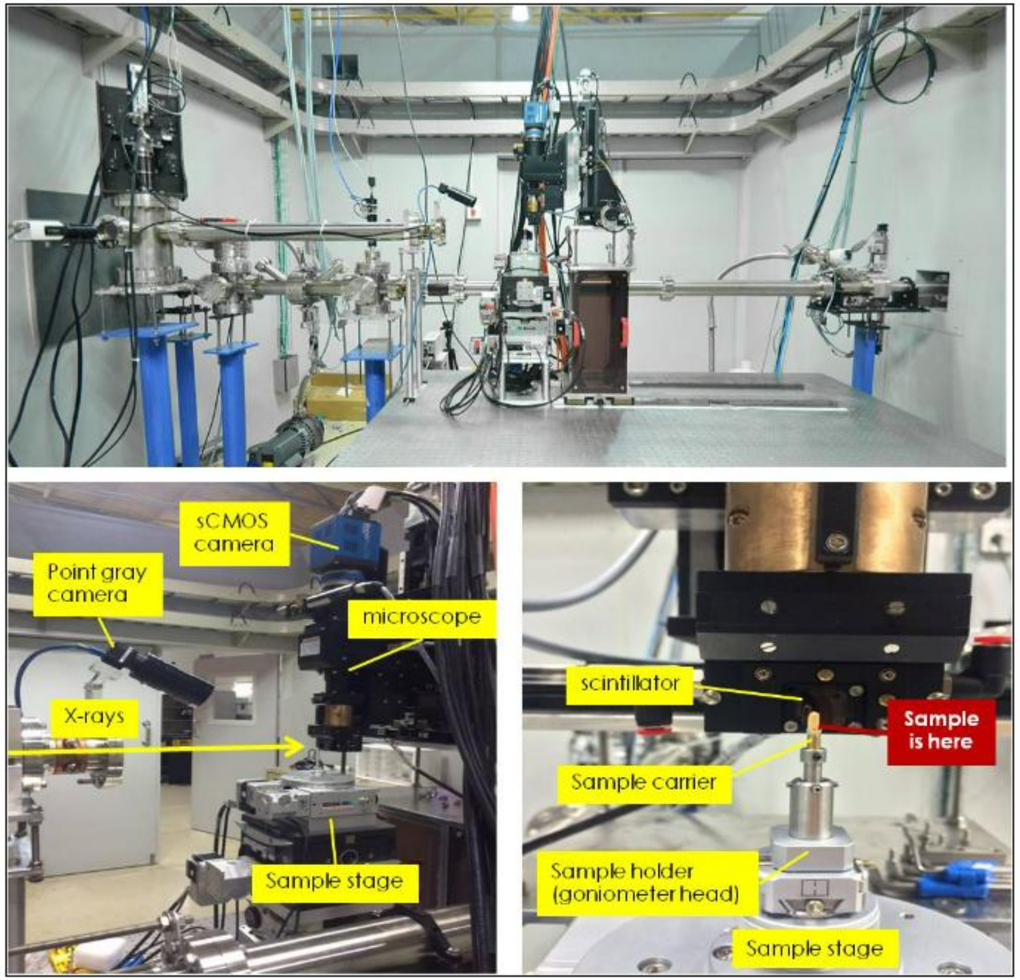


Figure 3 Experimental hutch of BL1.2W (commissioning mode).

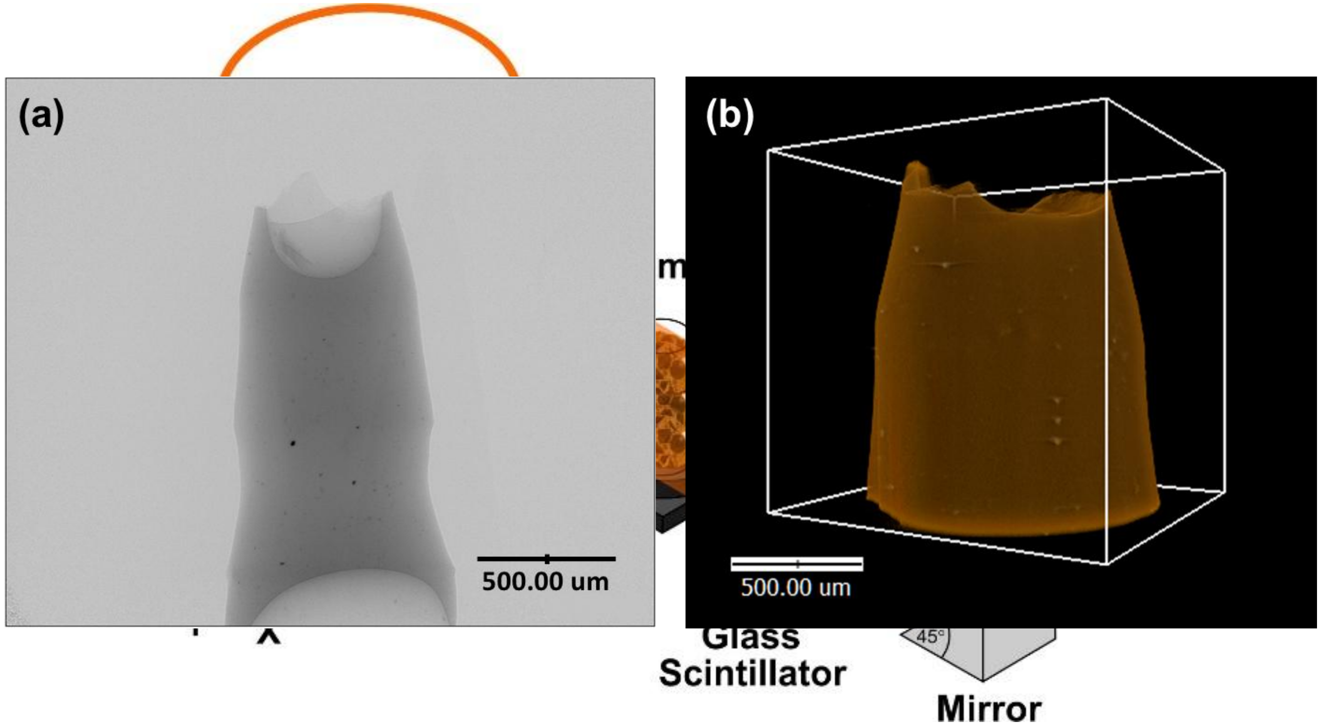


Fig.11. The 2D X-ray image (a) and the 3D reconstruction from 2D X-ray images of slime (b) by 3TbEu:7.5Gd glass as a scintillator.

Fig.10. The schematic diagram of the X-ray imaging process at BL1.2W, SLRI, Thailand

Research Results

X-ray imaging

- Spatial resolution can be expressed using spatial frequency, which describes the number of line pairs in a certain length (lp/mm).

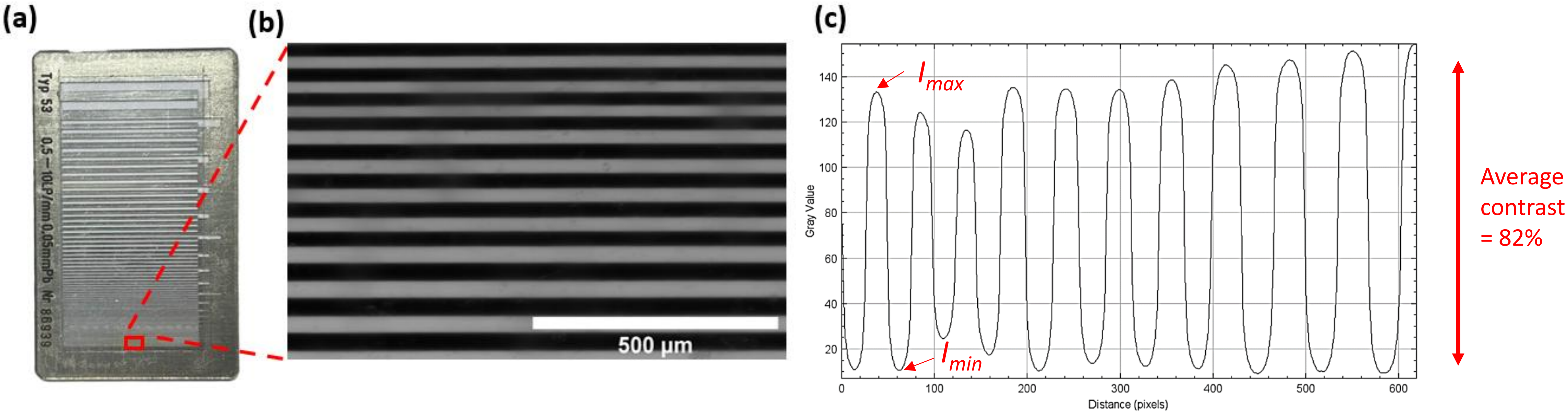


Fig.12. Line pair pattern (a) and the 2D X-ray image of line pair pattern at 10 lp/mm (b) and plot profile (c) of 3Tb:7.5Gd glass as a scintillator.

$$\% \text{ Contrast} = \left[\frac{I_{max}-I_{min}}{I_{max}+I_{min}}\right]$$

where I_{max} is the maximum intensity of gray value and $I_{\min}$ is the minimum intensity gray value

Yu Yu Kachurin et al 2021 J. Phys.: Conf. Ser. 2127 012021

http://qualitycontrol3.weebly.com/limiting-spatial-resolution.html

Summary

xTb:7.5Gd glass

Physical and Optical property

- Developed glass is heavy up to 3.01 g/cm^3.
- Glass performs the enhanced non-bridging oxygen with increment of Tb_2O_3 content.
- Tb^{3+} doped glass absorbs photons in Vis and NIR region.

Luminescence property

- PL and RL under all excitation makes the strongest emission of Tb^{3+} at 544 nm ($^5D_4 \rightarrow {}^7F_5$) in 3.0 mol% doped glass.
- PLQY of 3Tb:7.5Gd was 30.85%
- Photoluminescence decay curve of Eu^{3+} is in millisecond order.

X-ray imaging

- Special resolution of X-ray imaging is 10 lp/mm
- Possibility to be a glass scintillator, confirmed by X-ray imaging

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Thankyou

for your kind attention