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Temperature Dependence of the Luminescence output of $CdWO_4$ Crystal. Comparison with CaF_2:Eu

George Saatsakis[a], Dionysios Linardatos[b], Konstantinos Ninos[c], IoannisValais[b], Nektarios Kalyvas[b], Athanasios Bakas[c], Ioannis Kandarakis[b], George Fountos[b], George Panayiotakis[a] and Christos Michail[b,*]

[a]*University of Patras, School of Medicine, Department of Medical Physics, GR-15310, Rion, Greece*
[b]*University of West Attica, Department of Biomedical Sciences, Athens 12210, Greece*
[c]*University of West Attica, Radiation Physics, Department of Biomedical Engineering, Materials Technology and Biomedical Imaging Laboratory, Athens 12210, Greece*

Abstract

Scintillators are radiation converters applied in medical imaging detectors, in applications at harsh environments, including in geophysical detectors for deep geology boreholes, non-destructive testing (NDT) in gas and oil facilities, space, marine exploration, etc. In this study the luminescence efficiency dependence of single-crystal scintillators was examined with increasing temperature. Cadmium tungstate ($CdWO_4$) was examined against calcium fluoride doped with europium (CaF_2:Eu). The dimensions of the single crystals' samples were 10x10x10 mm^3 and were irradiated using X-ray radiographic exposures (90 kVp, 63mAs) to measure the light output with temperature (22 to 128 °C). The luminescence efficiency was found in both cases maximum at the lowest examined temperature (23.06 efficiency units-E.U for $CdWO_4$ and 22.01 E.U. for CaF_2:Eu, at 22 °C-environmental). With increasing temperature, the luminescence efficiency constantly decreased for both crystals due to thermal quenching (5.32 efficiency units for $CdWO_4$ and 4.43 for CaF_2:Eu, at 128 °C). In the mid-range (50-80 °C) $CdWO_4$ shows increased differences compared to CaF_2:Eu. $CdWO_4$ has a higher density (7.9 g/cm^3) and luminescence signal than CaF_2:Eu (3.18 g/cm^3), thus it is suitable, besides medical imaging, also for operation in harsh environments.

Keywords: Radiation detectors; Single Crystals; $CdWO_4$, CaF_2:Eu, Temperature dependence

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* Corresponding author. Tel.: +30-210-538-5387.
E-mail address: cmichail@uniwa.gr

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1. Introduction

Scintillators are radiation converters (Kandarakis 2016, Salomoni et al. 2018), applied in, medical imaging (radiography, computed tomography, positron emission tomography, mammography), at harsh environments, including detectors in geophysical research for deep geology boreholes, non-destructive testing (NDT) in gas and oil facilities, space, marine exploration, high energy physics, homeland security, etc. (Kytyr et al. 2010, Mares et al. 2012, Michail et al. 2016a, Michail et al. 2018a, Hu et al. 2019, Martini et al. 2019, Mykhaylyk et al. 2019). Thousands of different crystal scintillators have been produced depending upon the specific application. Some conventional crystal scintillators are sodium-iodide, Gadolinium-Oxyorthosilicate, Bismuth-Germinate-Oxide, Lutetium-Oxyorthosilicate, Yttrium-Orthoaluminate-Perovskite (Melcher et al. 1991, Van Eijk 2002, Michail et al. 2016b, Karpetas et al. 2017). In harsh environmental applications, scintillators are subject to extreme conditions of pressure, temperature, or radiation dose rates, resulting in variations in the luminescence output (Melcher et al. 1991, Bulatovic et al. 2013, Rothkirch et al. 2013, Bisong et al. 2019, Lebedev et al. 2019, Patri et al. 2019, Saxena et al. 2019). Due to these limitations, the detectors that will be used should have properties like adequate light output under elevated temperature, chemical stability, suitable mechanical properties, and energy resolution suitable for such conditions (Yang et al. 2014).

Cadmium tungstate ($CdWO_4$) is one of the most widely applied scintillators for various applications (Ziluei et al. 2017). $CdWO_4$ is a very dense scintillating material (ρ= 7.9 g/cm^3), with a very short radiation length of 10.6 mm (Galashov et al. 2014, Ruiz-Fuertes et al. 2017, Ziluei et al. 2017, Michail et al. 2020). The light yield of $CdWO_4$ can be found ranging across a wide range of values upon crystal manufacturer (Table 1) (Lecoq 2017, Ziluei et al. 2017, Michail et al. 2020). Some of the $CdWO_4$ advantages may be that it is a low-cost material, non-hygroscopic, and can tolerate high rates of radiation (van Eijk 2002, Lecoq 2016, Ziluei et al. 2017, Eritenko et al. 2020). As disadvantages may be considered the difficulty of fabricating in large samples and, of course, the existence of cadmium an element of considerable toxicity (van Eijk 2002, Lecoq 2016).

On the other hand, calcium fluoride doped with europium (CaF_2:Eu) can be easily found in nature and can be manufactured in large quantities at low cost. CaF_2:Eu has been used as a single crystal, in medical physics and spectroscopic applications, charged particle detection, in the quest for dark-matter, in radiation detectors for low energies, solar cell application, homeland security, etc. (Knoll 2000, Chen 2008, Mikhailik and Kraus 2010b, Lecoq et al. 2017, Dujardin et al. 2018, Fan et al. 2018, Yanagida 2018). CaF_2:Eu offers excellent operational characteristics, and it has good properties, for particle or low-energy radiation detectors. Furthermore, it is an excellent choice for vacuum applications since it has shallow vapor pressure. The energy resolution of CaF_2:Eu do not degrade noticeably with temperature.

Table 1. Comparison of $CdWO_4$ and CaF_2:Eu single-crystal's intrinsic and mechanical properties (Advatech 2020, Michail et al 2019, Michail et al. 2020).

		Crystal material	
		$CdWO_4$	CaF_2:Eu
Properties Mechanical	Units	Value	
Density	g/cm^3	7.9	3.18
Atomic Number (Effective)		61-66	16.5
Melting Point	ºK	1325	1360
Linear Expansion Coeff.	C^{-1}	$10.2x10^{-6}$	19.5×10^{-6}
Thermal Conductivity	$Wm^{-1}K^{-1}$	4.69(@300K)	9.7
Hardness	Mho	4-4.5	4
Hygroscopic	-	Όν	No

Both crystals have high melting points at 1325°C ($CdWO_4$), and 1360°C (CaF_2:Eu) and are robust to mechanical and thermal shocks, which is essential property for extreme environmental applications (Wang et al. 2018). Thus, investigation of these crystals for harsh environmental applications could be of interest.

Table 2. Comparison of $CdWO_4$ and CaF_2:Eu single-crystal's optical properties (Advatech 2020, Michail et al. 2019, Michail et al. 2020).

Crystal properties	$CdWO_4$	CaF_2:Eu
Emission maximum (nm)	490	435
Emission wavelength range (nm)	380-800	395-525
Decay-time (ns)	5000	950
Light yield (photons per MeV)	6200-28000	13000-30000
Photoelectron-yield (percent of NaI:Tl)	30-50	50
Radiation length (cm)	1.06	3.05
Refractive-index	2.2-2.3 (@max nm)	1.47 (@435nm)

In this article the luminescence output of $CdWO_4$ is compared with that of CaF_2:Eu crystals with increasing temperature, upon X-ray irradiation, for applications of detectors in harsh environments (temperature or radiation flux) (Rutherford et al. 2016, Saatsakis et al. 2020). The luminescence efficiency was measured under a typical X-ray excitation (Koukou et al. 2015).

2. Materials and Methods

$CdWO_4$ and CaF_2:Eu crystals of equal dimensions (10x10x10mm) and polished surfaces (Advatech 2020) were examined. An X-ray unit (BMI General Medical Merate tube) with rotating anode (Tungsten) was used to irradiate the crystals using 90 kVp and 63mAs, to measure the light photon intensity dependence with temperature (22 to 128 °C). The X-ray beam was filtered with an external aluminium filter (20 mm), simulating the beam quality alternation by a typical human chest (Michail et al. 2018b). The crystal sample was heated up to 128°, using a Perel 3700-9 2000W heating gun. The temperature on the crystal surface was monitored using an Extech RH101 infrared digital thermometer (0.1% accuracy).

2.1. Luminescence Output

The light flux emitted by the crystal samples upon X-ray irradiation was measured using the following methodology. The crystal was placed at the upper port of the integrating sphere (Oriel 70451). In the output port of the sphere a photomultiplier (PMT) (EMI 9798B) was coupled (Fig. 1). The photomultiplier's photocathode (extended S-20) signal was fed to a Sub-Femtoamp electrometer (Keithley, 6430) (Saatsakis et al. 2019). This set-up was used to measure electrometer's electric current in order to estimate the light flux (Table 3):

$$\dot{\Psi}_{\Lambda} = \frac{I_{elec}}{\tau_0 (s_{PC} a_s)} \cdot \frac{1}{A_{sc}} \tag{2.1}$$

Table 3. Light flux calculation parameters.

	Light flux calculation parameters
I_{elec}	Electrometer's current (pA)
s_{PC}	Photocathode's maximum photosensitivity (pA/W)
a_s	Spectral matching factor between the emitted light from the crystal and the sensitivity of the extended S-20 photocathode
A_{sc}	Irradiated crystal surface
τ_0	Integrating sphere's throughput (15.6)

Then the light output over the x-ray exposure rate of the crystals was calculated as (Michail et al. 2019):

$$\eta_A = \dot{\Psi}_\lambda / \dot{X} \tag{2.2}$$

Where $\dot{X}$ is the exposure rate incident on the crystal. Efficiency-units (EU) are expressed in $\mu W \times m^{-2} / (mR \times s^{-1})$. The S.I. equivalent is $\mu W \times m^{-2} / (mGy \times s^{-1})$, where mGy is the corresponding air Kerma.

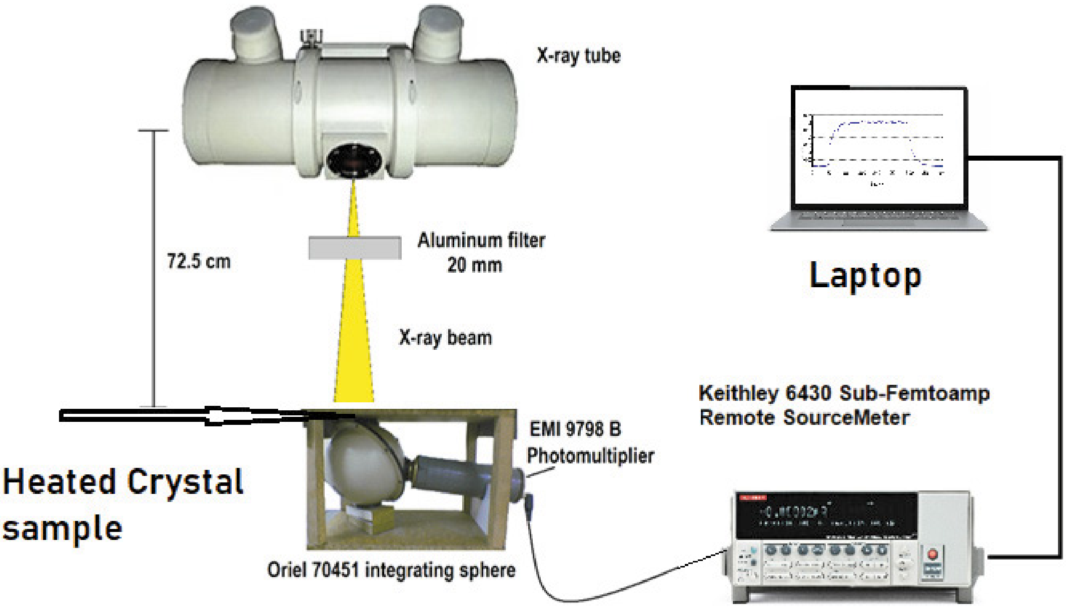

Fig. 1. Luminescence efficiency apparatus.

3. Results and Discussion

Figure 2 shows luminescence output results for the examined crystal samples at a temperature range from 22 to 128 °C. This is an indicative temperature range that can be found in logging detectors in which crystals are subjected to temperatures in the range from minus 0 °C to more than 200 °C (Melcher et al. 1991). Exposure of the scintillator to excessive heating or X-ray flux can result in crystal cracking (Pokluda et al. 2015, Kastengren 2019).

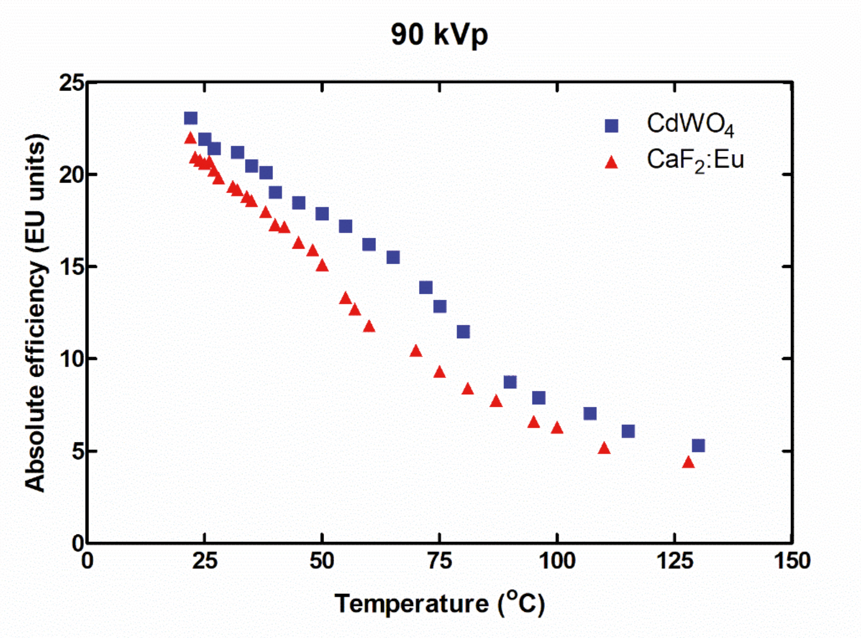

Fig. 2. Comparison between the luminescence efficiency of the examined single crystals in the temperature range from 22 to 128° C.

Figure 2 shows that in the examined crystal samples, the luminescence efficiency is temperature-dependent (Kastengren 2019). With increasing temperature, the luminescence efficiency constantly decreased for both crystals due to thermal quenching (5.32 efficiency units for $CdWO_4$ and 4.43 for CaF_2:Eu, at 128 °C). When the temperature increases, the light output decreases, since it is affected by radiation-less transitions whose probability increase with temperature, along with possible lattice-defects or impurities that will reduce the signal (Melcher et al. 1991). The latter is crucial when large crystal sizes should be prepared. The luminescence signal was found in both crystals maximum at the lowest examined temperature (23.06 E.U. for $CdWO_4$ and 22.01 E.U. for CaF_2:Eu at 22 °C). In the mid-range (50-80 °C) $CdWO_4$ shows increased differences compared to CaF_2:Eu.

4. Conclusion

In this research, the effect of temperature on the luminescence output of $CdWO_4$ and CaF_2:Eu single crystals, for applications in harsh environments, was examined. The luminescence output values of both crystals decrease down to 77-79% when the crystal surfaces were heated to the maximum operating temperature. Both crystals showed almost similar behavior upon temperature, with $CdWO_4$ performing slightly better due to its intrinsic properties, such as the higher density (7.9 g/cm^3 for $CdWO_4$, 3.18 g/cm^3 for CaF_2:Eu) and the light output of this crystal. These findings could render $CdWO_4$ preferable for applications, besides medical imaging, in applications of crystals at harsh environments.

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