Applications of luminescence dosimeters on FLASH-RT studies: literature review on materials and methods

Dr. Daniel Villani
PITZ Physics Seminar, 29.06.2023

HELMHOLTZ

Outline

1 Brief overview on luminescence dosimetry theory
2 Main samples used
3 Main readers used
4 Main uncertainty components
5 Successful experiences of UHDR/FLASH-RT studies measurements using TLDs/OSLDs

1. Brief overview on luminescence dosimetry theory

How does luminescence dosimetry work?

The luminescent materials for dosimetry are ionic crystals (insulators or semiconductors) that have energy levels in their crystalline structure. These energy levels can be described with “the banding model”.

The process of luminescence dosimetry consists of two main actions:

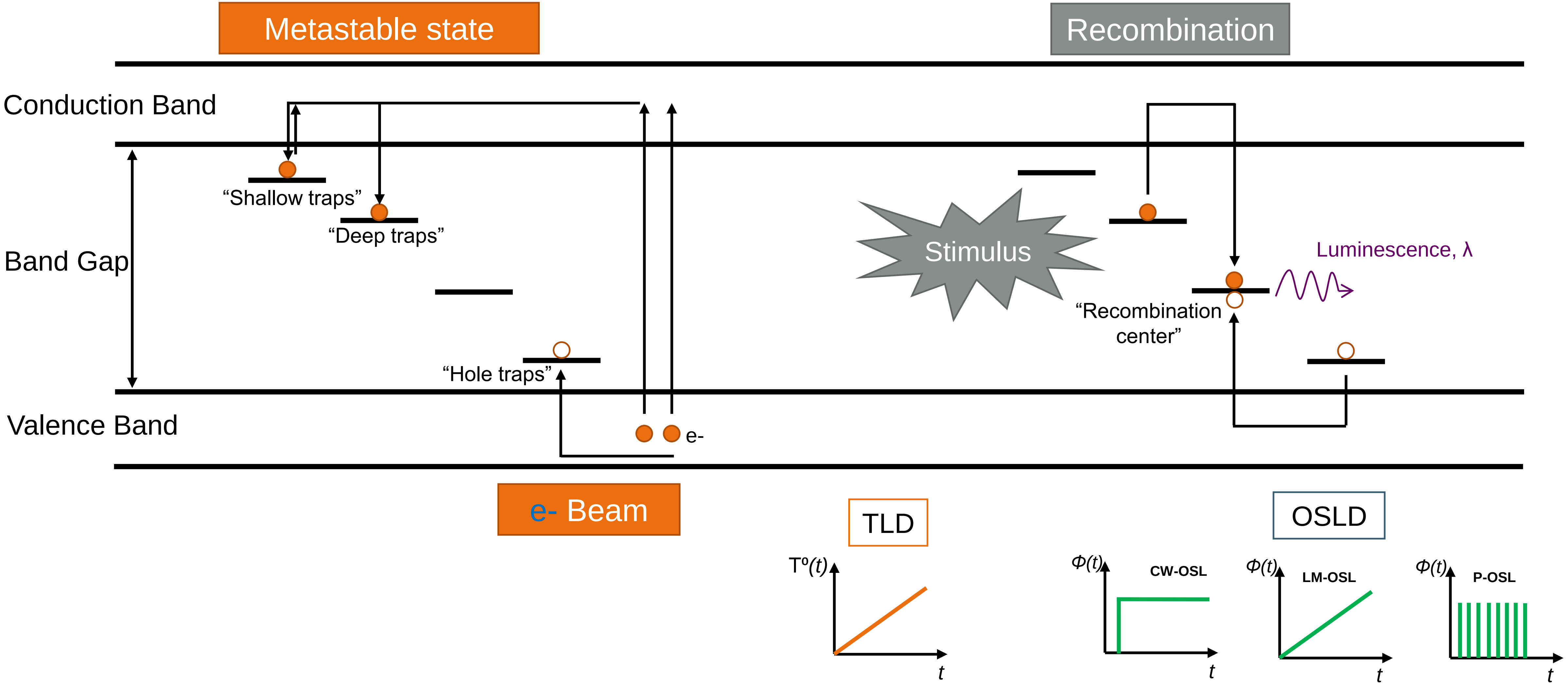
- The disturbance of the equilibrium of the material, taking it to a metastable state, and
- The relaxation of the system when stimulated, bringing it back to equilibrium.

Although the relaxation process can occur naturally, its probability at room temperature is usually very low.

[1] Yukihara, E.G., McKeever, S.W., Andersen, C.E., Bos, A.J., Bailiff, I.K., Yoshimura, E.M., Sawakuchi, G.O., Bossin, L. and Christensen, J.B., 2022. Luminescence dosimetry. *Nature Reviews Methods Primers*, *2*(1), p.26.

1. Brief overview on luminescence dosimetry theory

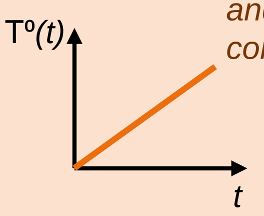
The phenomenon explained by the banding model



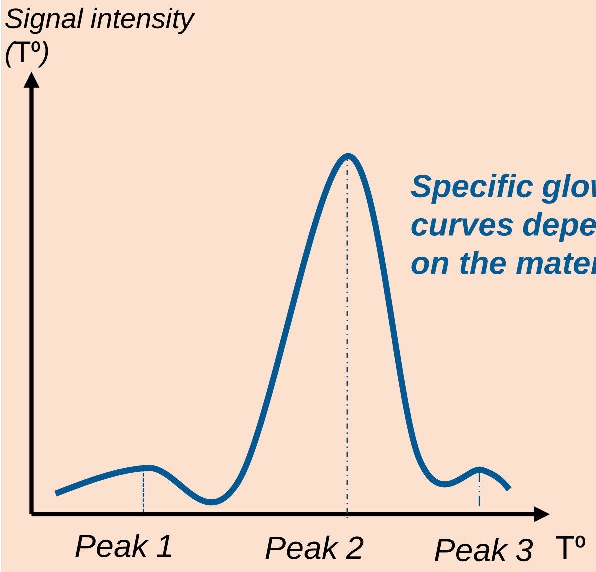
[1] Yukihara, E.G., McKeever, S.W., Andersen, C.E., Bos, A.J., Bailiff, I.K., Yoshimura, E.M., Sawakuchi, G.O., Bossin, L. and Christensen, J.B., 2022. Luminescence dosimetry. *Nature Reviews Methods Primers*, *2*(1), p.26.

1. Brief overview on luminescence dosimetry theory

TLD



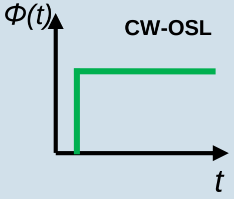
TTP: Heating rate, time and top temperature control

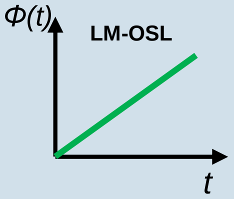


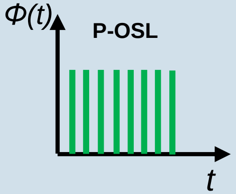
Different depths of traps

***Integrated area** above the curve*

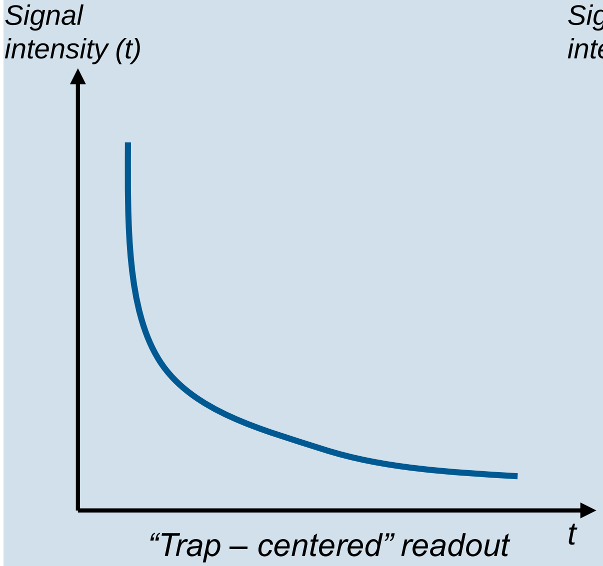
OSLD



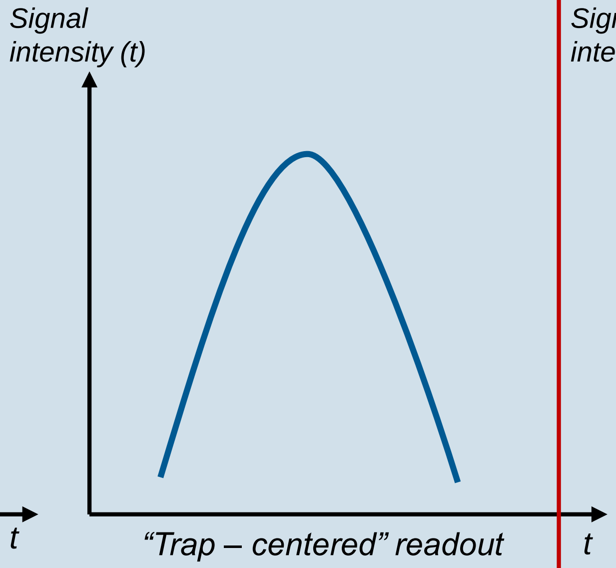




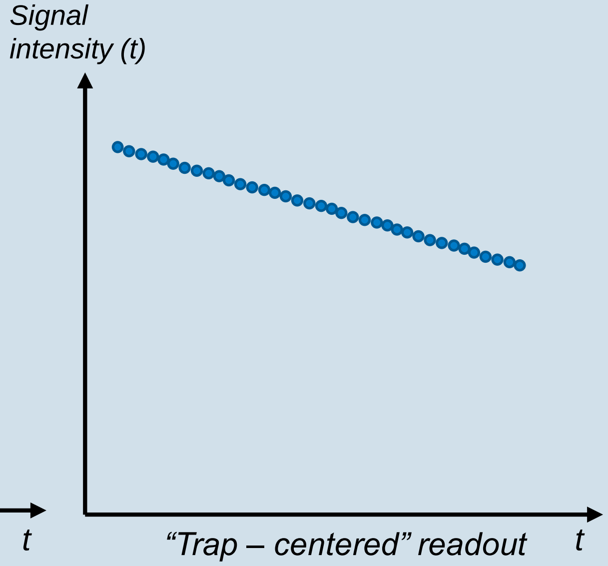
This method can be characterized for non-destructive readings (Landauer microStar readers)



*First point of measuremet or **integrated area** above the curve*



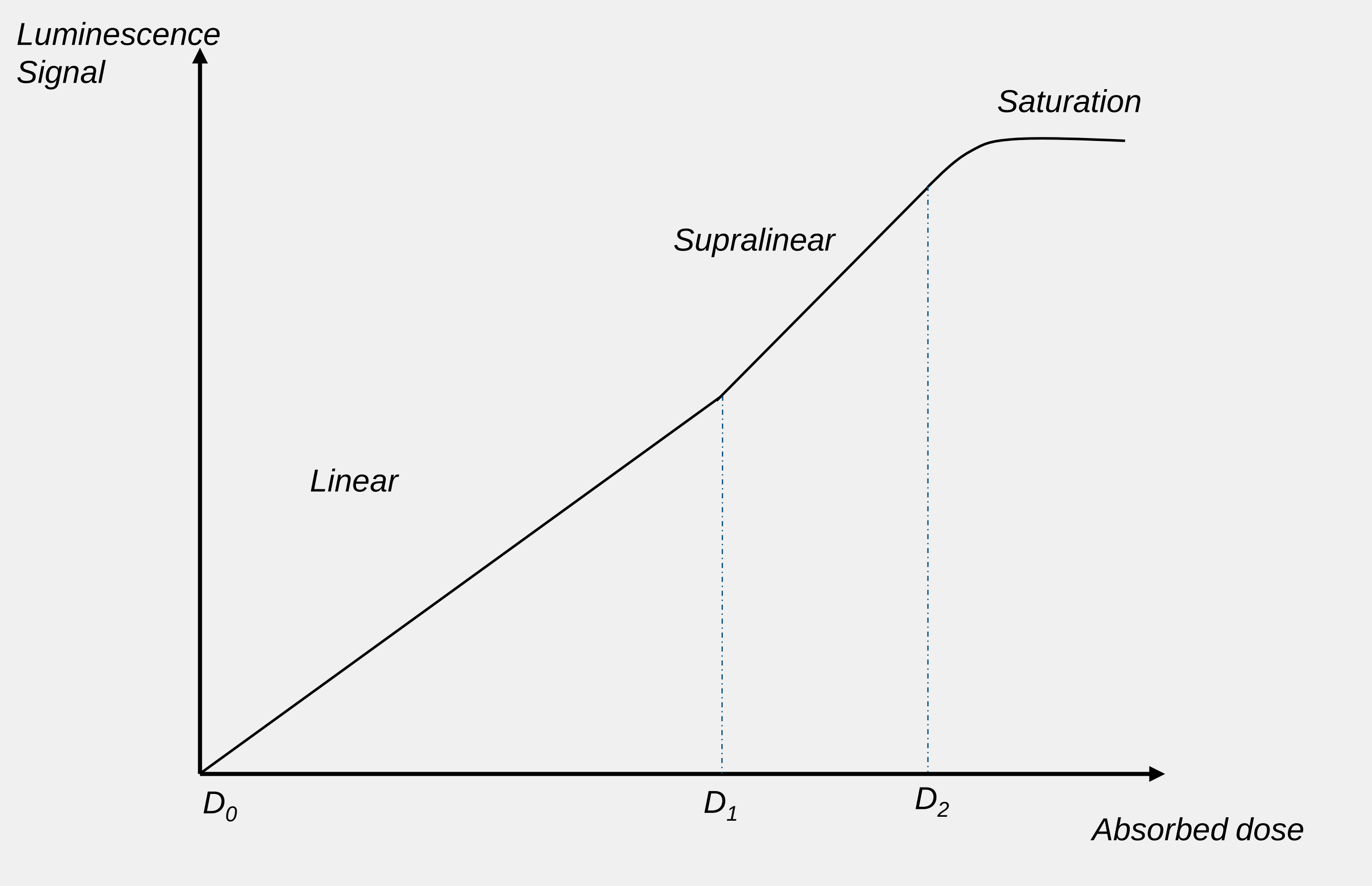
Integrated area above the curve



First point of measurement or average of n points

1. Brief overview on luminescence dosimetry theory

Linearity and Supralinearity



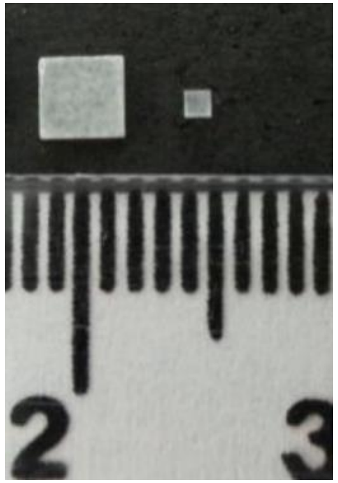
"The competition between different trapping and recombination centers during either irradiation or readout can cause the material to produce more signal that would be expected based on the response at low doses. This is called supralinear dose-response. An example of a competition process that could lead to supralinearity is the presence of a competing deep trapping center." [2]

[2] Yukihara, E.G. and McKeever, S.W., 2011. *Optically stimulated luminescence: fundamentals and applications*. John Wiley & Sons. p. 127.

2. Main samples used

TLDs[1]		
Material/Name	**Main characteristics**	
LiF:Mg,Ti (TLD-100, Harshaw Thermo Scientific)	Dimensions:	3.2 x 3.2 x 0.9 mm^3
	Batch uniformity:	± 15%
	Z_{eff} (host only):	8.2
	Density:	2.64 $g.cm^{-3}$
	Useful dose response range:	50 µGy to 1 kGy
	Linearity:	Linear < 1Gy, Supralinear 1Gy < D < 1kGy
	Repeatability:	± 2%
µLiF:Mg,Ti (microcubes TLD-100, Harshaw Thermo Scientific)	Dimensions:	1 x 1 x 1 mm^3
	Batch uniformity:	± 15%
	Z_{eff} (host only):	8.2
	Density:	2.64 $g.cm^{-3}$
	Useful dose response range:	50 µGy a 1 kGy
	Linearity:	Linear < 1Gy, Supralinear 1Gy < D < 1kGy
	Repeatability:	± 2%

Main emission ~410nm



[1] Yukihara, E.G., McKeever, S.W., Andersen, C.E., Bos, A.J., Bailiff, I.K., Yoshimura, E.M., Sawakuchi, G.O., Bossin, L. and Christensen, J.B., 2022. Luminescence dosimetry. *Nature Reviews Methods Primers*, *2*(1), p.26.

2. Main samples used

OSLDs[1]		
Material/Name	**Main characteristics**	
Al_2O_3:C (nanoDot, Landauer Inc.)	Dimensions:	5mm diameter x 0.2mm thickness
	Batch uniformity:	± 5 % for screened samples, ± 10 % for unscreened samples
	Z_{eff} (host only):	11.3
	Density:	3.96 g.cm^{-3}
	Useful dose response range:	5 µGy to 500Gy
	Linearity:	Linear < 5Gy, Supralinear 5Gy < D < 100Gy
	Repeatability:	± 2% or better
BeO (ezClip, Dosimetrics)	Dimensions:	4.7 x 4.7 x 0.5mm^3
	Batch uniformity:	NA
	Z_{eff} (host only):	7.2
	Density:	2.85 g.cm^{-3}
	Useful dose response range:	30 µGy to 10Gy
	Linearity:	Linear < 10Gy
	Repeatability:	± 2% or better

Main emission ~335nm

Main emission ~365nm

[1] Yukihara, E.G., McKeever, S.W., Andersen, C.E., Bos, A.J., Bailiff, I.K., Yoshimura, E.M., Sawakuchi, G.O., Bossin, L. and Christensen, J.B., 2022. Luminescence dosimetry. *Nature Reviews Methods Primers*, *2*(1), p.26.

3. Main readers used

TL only (commercial packages)
Thermo Scientific Harshaw 3500 (manual)
Thermo Scientific Harshaw 5500 (automated)
Mirion RE 2000A
OSL only (commercial packages)
Landauer microStar
Landauer microStar ii
Dosimetrics BEOSL reader
TL/OSL (research)
Freiberg Instruments Lexsyg Smart
DTU Risø TL/OSL

DTU Risø TL/OSL reader

Freiberg Instruments Lexsyg Smart TL/OSL


Harshaw 3500


Harshaw 5500


Mirion RE 2000A


Dosimetrics BEOSL


Landauer microStar ii


Landauer microStar

4. Main uncertainty components

Characteristics of the sample
Size
Shape
Mass
Density and Z_{eff} (Energy dependence)
Sensitivity (ammount of luminescent material within the sampe)
Reader stability
Dark counts and current
PMT sensitivity
Stimulation source (heat or light control)
Calibration
Irradiation (radioactive source or x-ray/electronic system)
Tracebility of the measurements
Annealing
Furnance/Oven temperature (TL)
Wavelenth and intensity of the light (OSL)

4. Main uncertainty components

In order to "reduce" the number of uncertainty components, one can use the "S/S_R method" proposed by Yukihara et al.[3] for both calibration of the TL/OSL system and dose readout

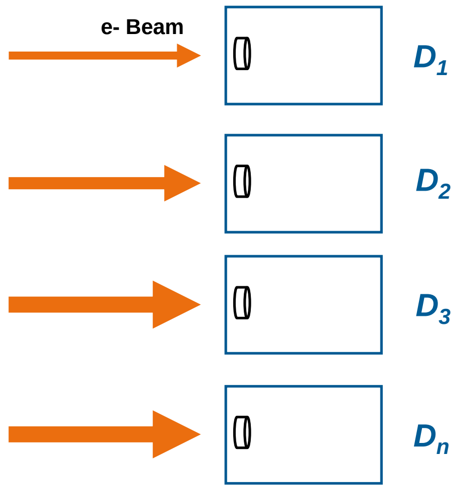
This method is very advantageous when considering doing FLASH-RT dosimetry due to the increase in accuracy on the measurement

[3] Yukihara EG, Mardirossian G, Mirzasadeghi M, Guduru S, Ahmad S. Evaluation of Al_2O_3:C optically stimulated luminescence (OSL) dosimeters for passive dosimetry of high-energy photon and electron beams in radiotherapy. Med Phys. 2008 Jan;35(1):260-9. doi: 10.1118/1.2816106.

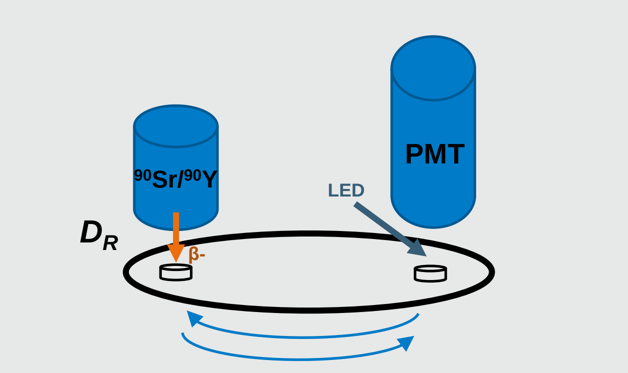
4. Main uncertainty components

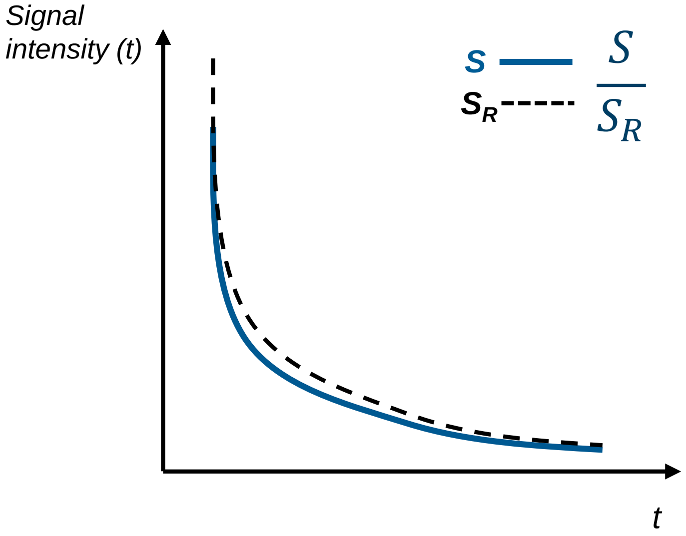
The S/S_R method

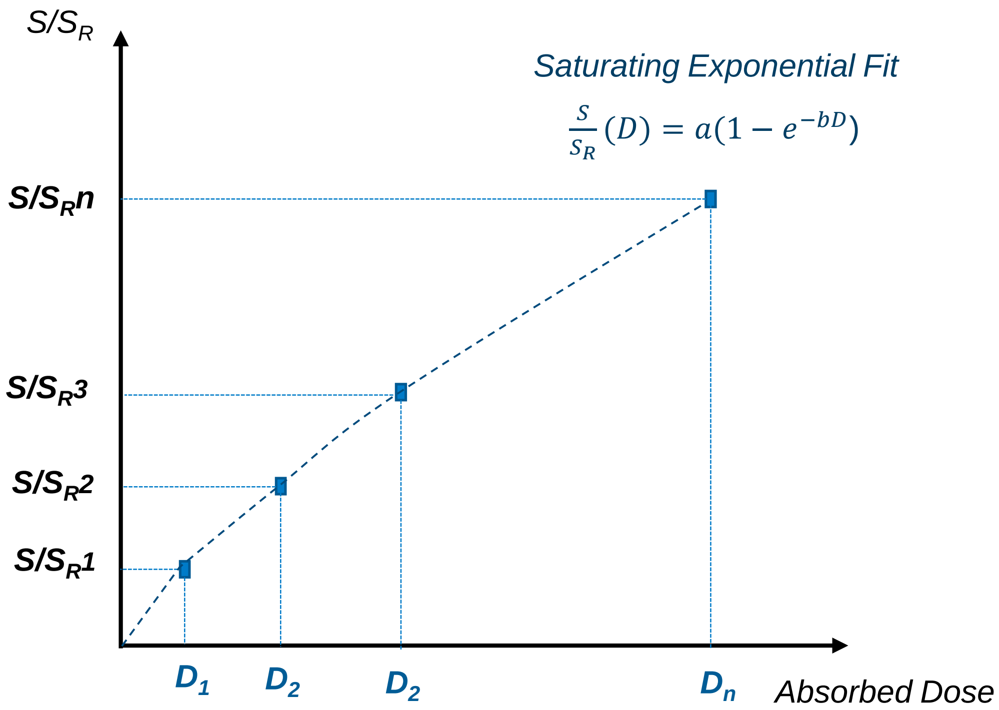
Building a calibration curve



Reader with built-in irradiation source







Dose measurements become relative and most of the uncertainty componentes are negligible

[3] Yukihara EG, Mardirossian G, Mirzasadeghi M, Guduru S, Ahmad S. Evaluation of Al_2O_3:C optically stimulated luminescence (OSL) dosimeters for passive dosimetry of high-energy photon and electron beams in radiotherapy. Med Phys. 2008 Jan;35(1):260-9. doi: 10.1118/1.2816106.

4. Main uncertainty components

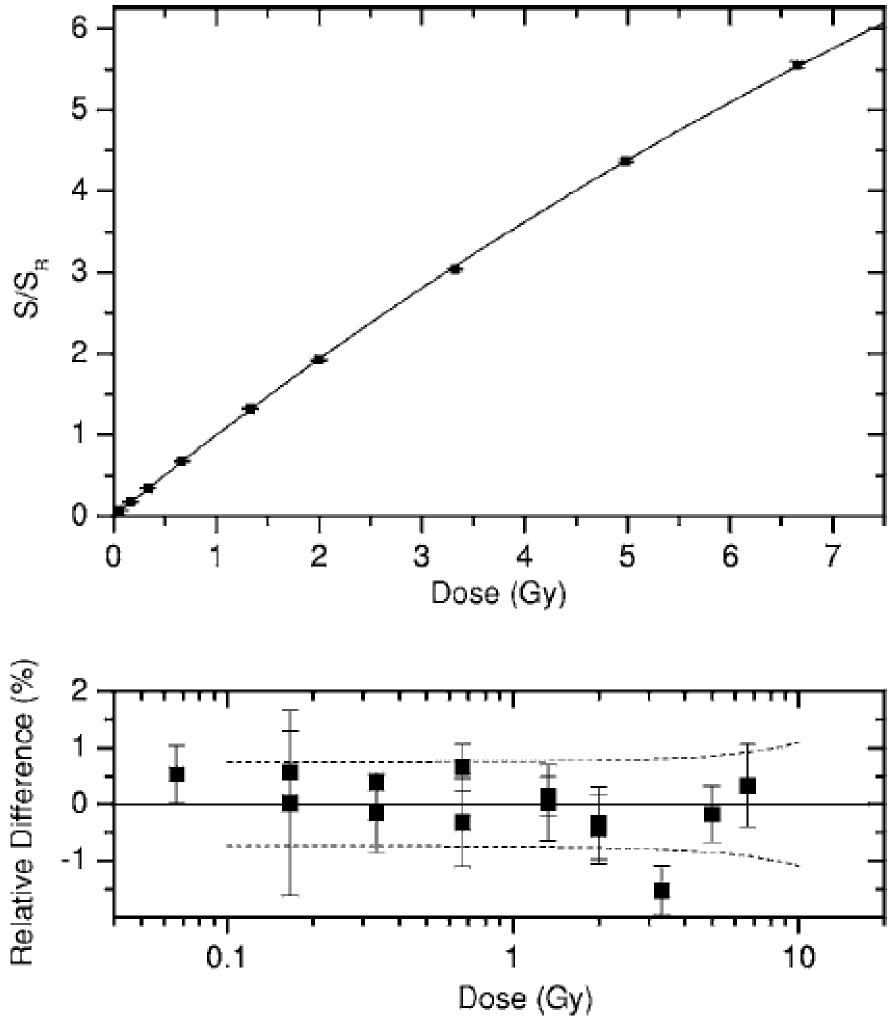


FIG. 3. (a) Ratio S/S_R between the OSL signal S after irradiation in the accelerator with the indicated dose, and the OSL signal S_R after subsequent irradiation with a reference dose. (b) Relative difference between the OSL data and the fitted saturating exponential [Eq. (1)] with parameters $a = 15.11 \pm 0.20$ and $b = (0.0686 \pm 0.0011)$ Gy^{-1}. The dashed lines are the relative standard deviation of the dose σ_D as a function of dose, calculated using Eq. (2) and the above fitted values for a and b, as well as a covariance of -2.12×10^{-4} Gy^{-1} obtained from the fitting procedure. The data points represent the mean value of five dosimeters and the error bars represent the experimental standard deviation. [Note that the error bars are smaller than the size of the symbols in (a).] Duplicate irradiations were carried out for some doses.

[3] Yukihara EG, Mardirossian G, Mirzasadeghi M, Guduru S, Ahmad S. Evaluation of Al_2O_3:C optically stimulated luminescence (OSL) dosimeters for passive dosimetry of high-energy photon and electron beams in radiotherapy. Med Phys. 2008 Jan;35(1):260-9. doi: 10.1118/1.2816106.

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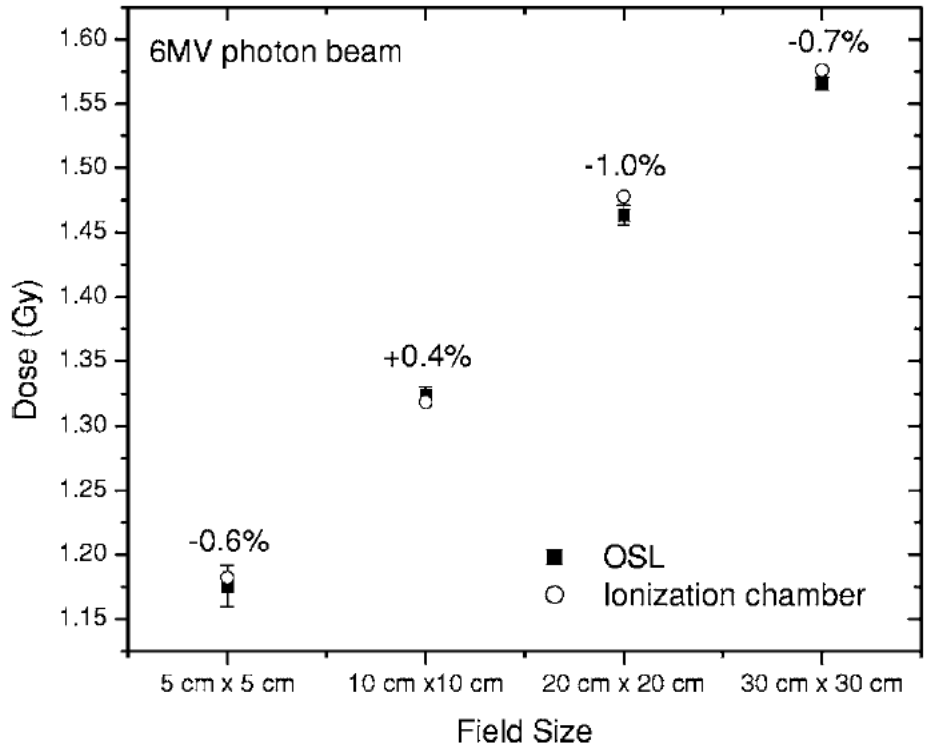
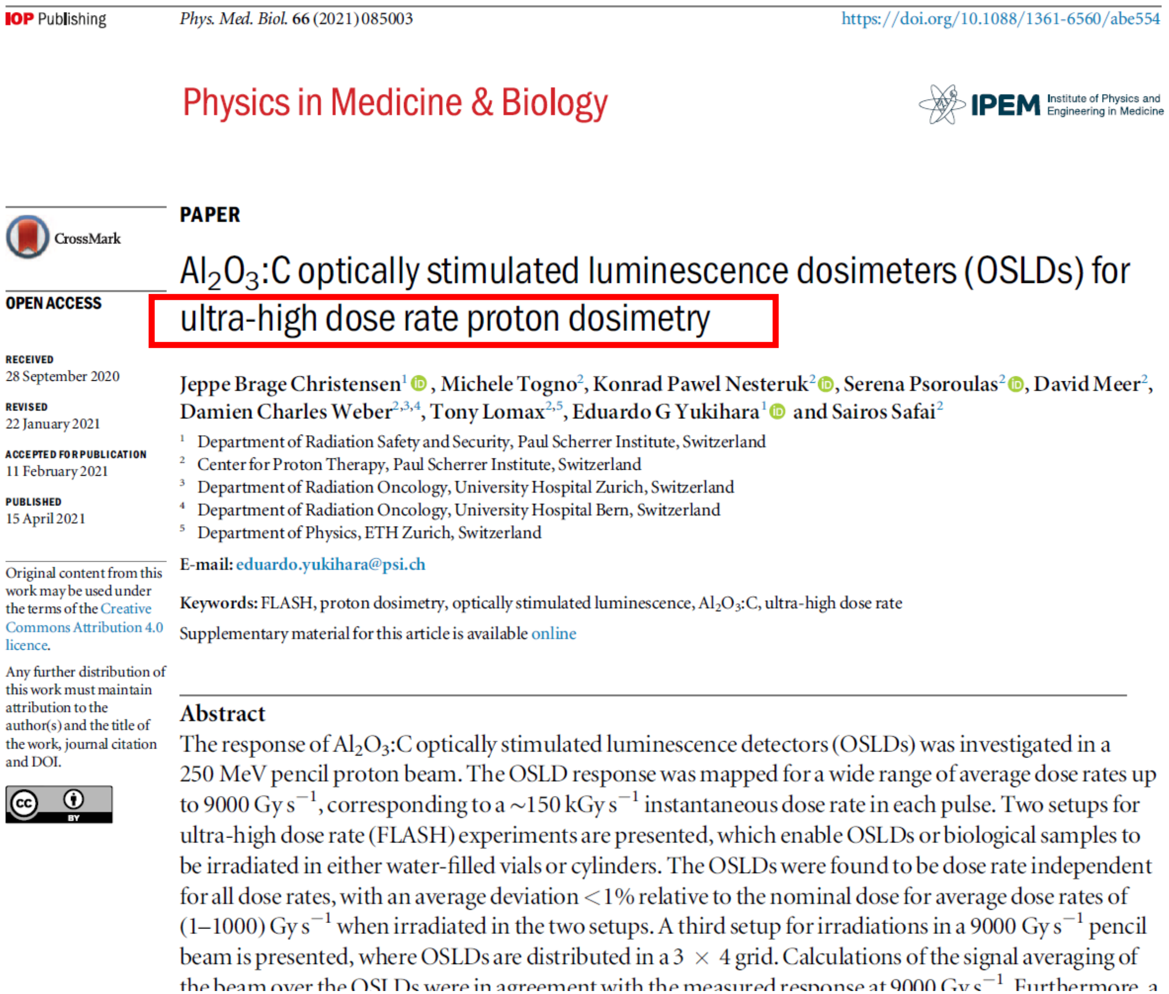


FIG. 9. Doses obtained using OSLDs and ionization chambers for different field sizes. In all cases the irradiations were carried out with 6 MV photon beam, 200 MU, and at 10 cm depth in water. The data points are the average of 5 OSLDs and the error bars represent the experimental standard deviation.

OSL measurements were comparable to ionization chamber measurements for conventional RT

[2] Yukihara EG, Mardirossian G, Mirzasadeghi M, Guduru S, Ahmad S. Evaluation of Al_2O_3:C optically stimulated luminescence (OSL) dosimeters for passive dosimetry of high-energy photon and electron beams in radiotherapy. Med Phys. 2008 Jan;35(1):260-9. doi: 10.1118/1.2816106.

5. Successful experiences of FLASH-RT measurements using TLDs/OSLDs



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PAPER

Al_2O_3:C optically stimulated luminescence dosimeters (OSLDs) for ultra-high dose rate proton dosimetry

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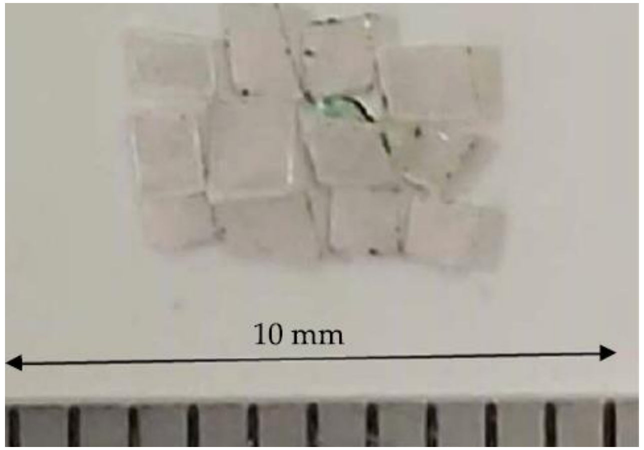
Keywords: FLASH, proton dosimetry, optically stimulated luminescence, Al_2O_3:C, ultra-high dose rate

Supplementary material for this article is available online

Abstract

The response of Al_2O_3:C optically stimulated luminescence detectors (OSLDs) was investigated in a 250 MeV pencil proton beam. The OSLD response was mapped for a wide range of average dose rates up to 9000 Gy s^{-1}, corresponding to a ~150 kGy s^{-1} instantaneous dose rate in each pulse. Two setups for ultra-high dose rate (FLASH) experiments are presented, which enable OSLDs or biological samples to be irradiated in either water-filled vials or cylinders. The OSLDs were found to be dose rate independent for all dose rates, with an average deviation <1% relative to the nominal dose for average dose rates of (1–1000) Gy s^{-1} when irradiated in the two setups. A third setup for irradiations in a 9000 Gy s^{-1} pencil beam is presented, where OSLDs are distributed in a 3 × 4 grid. Calculations of the signal averaging of the beam over the OSLDs were in agreement with the measured response at 9000 Gy s^{-1}. Furthermore, a



Grossly cut Al_2O_3:C samples

DTU Risø TL/OSL reader

[4] Christensen, J.B., Togno, M., Nesteruk, K.P., Psoroulas, S., Meer, D., Weber, D.C., Lomax, T., Yukihara, E.G. and Safai, S., 2021. Al_2O_3: C optically stimulated luminescence dosimeters (OSLDs) for ultra-high dose rate proton dosimetry. Physics in Medicine & Biology, 66(8), p.085003. https://doi.org/10.1088/1361-6560/abe554

5. Successful experiences of FLASH-RT measurements using TLDs/OSLDs

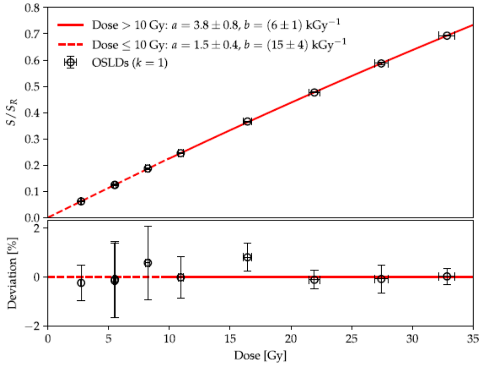


Figure 2. The ratio of the signal S from the experimental irradiation to the reference irradiation S_R as a function of the dose measured with an ionization chamber. Equation (1) is fitted separately to the data above and below 10 Gy. The lower figure shows the deviation of the data points to the fit. The 5.5 Gy irradiation was repeated to investigate the reproducibility.

S/S_R calibration

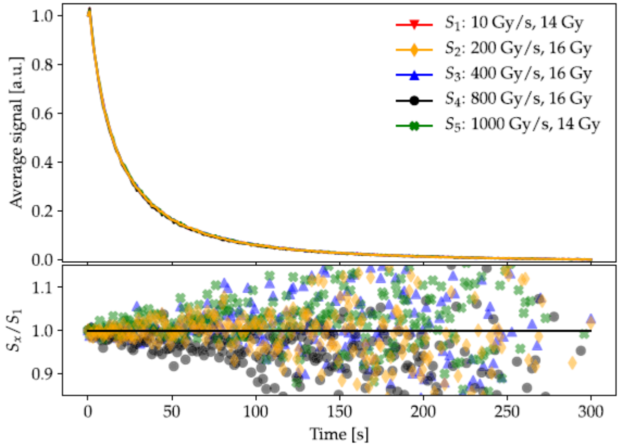
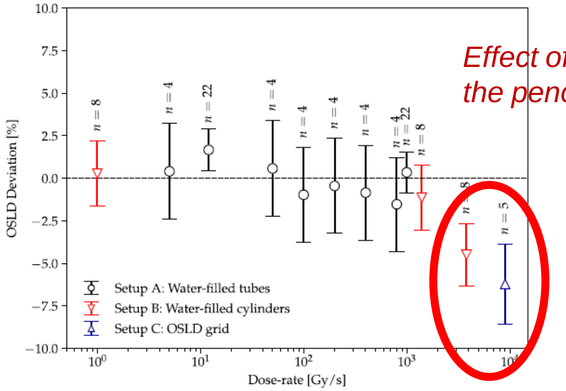


Figure 4. Each curve shows the average of four OSLD readout signals normalized to the entry, where the background signal has been subtracted. The lower scatter plot shows the ratio of each signal to the signal obtained from the irradiation with the lowest dose rate.

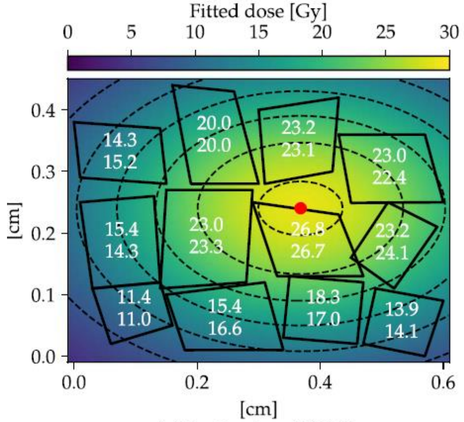
Different dose rates



Effect of averaging dose from the pencil beam

Figure 10. The deviation of the OSLD measured doses relative to the nominal dose as a function of the dose rate for the three methods A, B, and C. The number of aggregated data points for each dose rate is given above each marker. The decreasing response above 1000 Gy s^{-1} is due to signal averaging of the narrow pencil beam over the OSLDs.

Different dose rates at different setups



(b) Center dose: 29.5 Gy

2D measurements with sample array

[4] Christensen, J.B., Togno, M., Nesteruk, K.P., Psoroulas, S., Meer, D., Weber, D.C., Lomax, T., Yukihara, E.G. and Safai, S., 2021. Al_2O_3: C optically stimulated luminescence dosimeters (OSLDs) for ultra-high dose rate proton dosimetry. Physics in Medicine & Biology, 66(8), p.085003. https://doi.org/10.1088/1361-6560/abe554

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PAPER

Characterization of LiF:Mg,Ti thermoluminescence detectors in low-LET proton beams at ultra-high dose rates

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[2] Center for Proton Therapy, Paul Scherrer Institute, Villigen PSI, Switzerland
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Keywords: dose rate dependence, LiF:Mg,Ti, proton dosimetry, FLASH dosimetry, thermoluminescence detectors

Abstract

Objective. This work aims at characterizing LiF:Mg,Ti thermoluminescence detectors (TLDs) for dosimetry of a 250 MeV proton beam delivered at ultra-high dose rates (UHDR). Possible dose rate effects in LiF:Mg,Ti, as well as its usability for dosimetry of narrow proton beams are investigated. *Approach.* LiF:Mg,Ti (TLD-100^{TM} Microcubes, 1 mm × 1 mm × 1 mm) was packaged in matrices of 5 × 5 detectors. The center of each matrix was irradiated with single-spot low-LET (energy >244 MeV) proton beam in the (1–4500) Gy s^{-1} average dose rates range. A beam reconstruction procedure was applied to the detectors irradiated at the highest dose rate (Gaussian beam sigma <2 mm) to correct for volumetric averaging effects. Reference dosimetry was carried out with a diamond detector and radiochromic films. The delivered number of protons was measured by a Faraday cup, which was employed to normalize the detector responses. *Main results.* The lateral beam spread obtained from the beam reconstruction agreed with the one derived from the radiochromic film measurements. No dose rates effects were observed in LiF:Mg,Ti for the investigated dose rates within 3% ($k = 1$). On average, the dose response of the TLDs agreed with the reference detectors within their uncertainties. The largest deviation (−5%) was measured at 4500 Gy s^{-1}. *Significance.* The dose rate independence of LiF:Mg,Ti TLDs makes them suitable for dosimetry of UHDR proton beams. Additionally, the combination of a matrix of TLDs and the beam reconstruction can be applied to determine the beam profile of narrow proton beams.

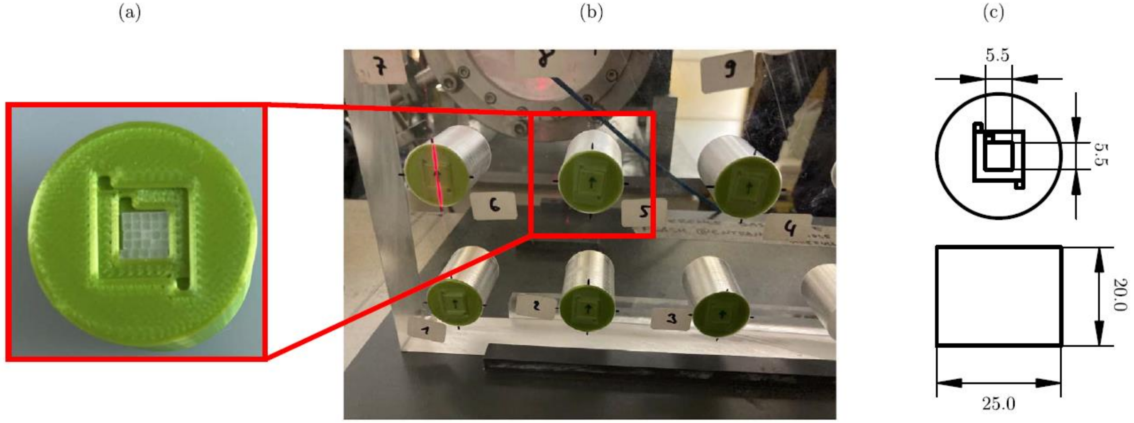

Figure 1. (a) 3D-printed cylinder containing the matrices of 5 × 5 TLD-100^{TM} Microcubes. The package lid is not shown in this figure. (b) The cylinders were inserted in a PMMA phantom for the irradiation and centred with the beam axis. (c) Technical drawing of the package, where the dimensions are expressed in millimeters.

Standard μLiF:Mg,Ti (microcubes TLD-100) samples

Freiberg Instruments Lexsyg Smart TL/OSL reader

[5] Motta, S., Christensen, J.B., Togno, M., Schäfer, R., Safai, S., Lomax, A.J. and Yukihara, E.G., 2023. Characterization of LiF: Mg,Ti thermoluminescence detectors in low-LET proton beams at ultra-high dose rates. Physics in Medicine & Biology, 68(4), p.045017. https://doi.org/10.1088/1361-6560/acb634

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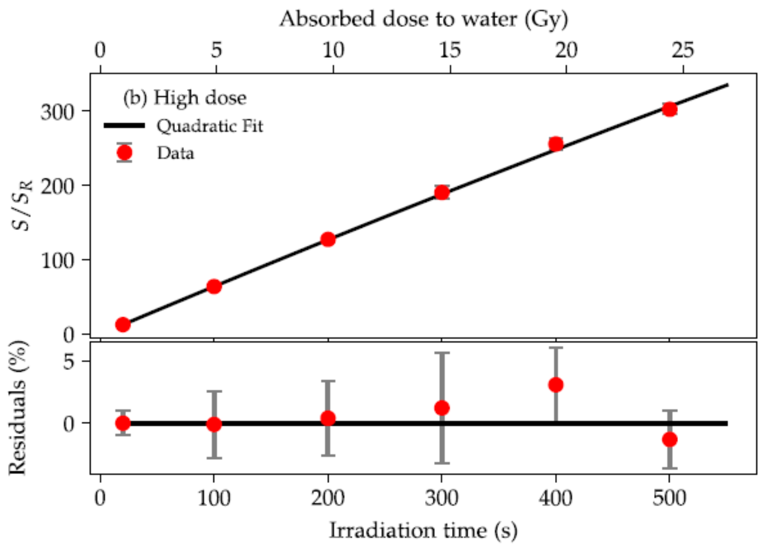

Figure 4. Calibration curves obtained by irradiating TLD-100™ Microcube detectors with the beta source in the Lexsyg Smart reader, for the (a) low dose and (b) high dose ranges. To convert the irradiation time into absorbed dose to water, a reference irradiation with a 230 MeV clinical proton beam was performed. Each point is the average of 5 samples and the error bars indicate the standard deviation of the data. The curves were fitted with quadratic functions, whose residual plots are reported in the lower figures.

S/S_R calibration

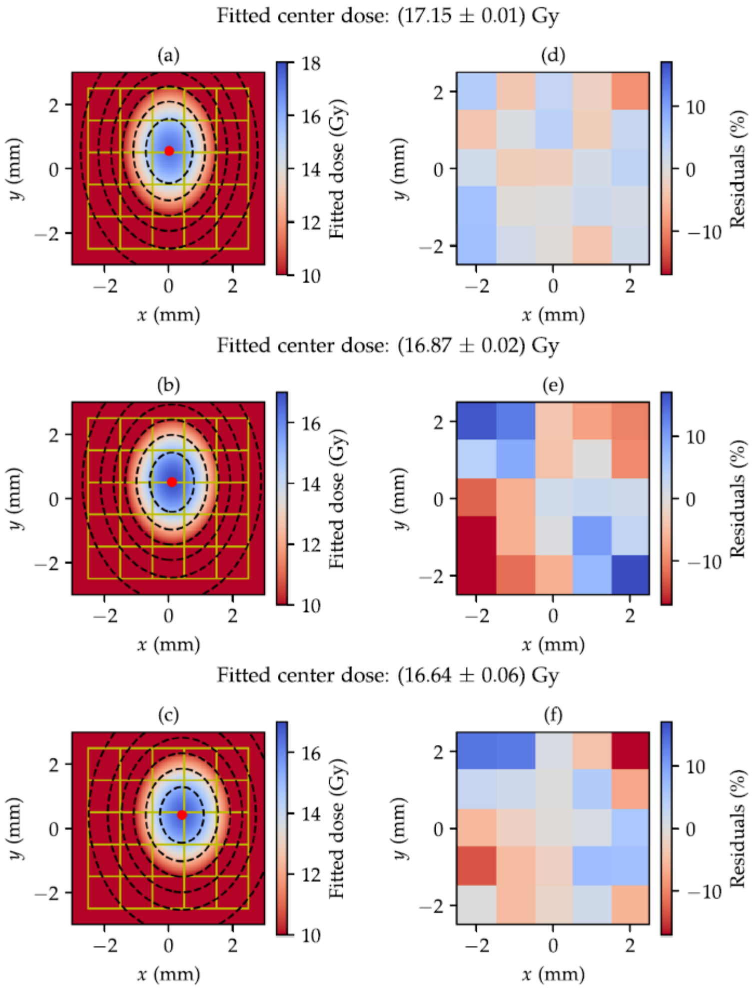

Figure 6. (a)–(c) show the dose maps obtained with the beam reconstruction procedure applied to the TLD-100™ Microcube matrices irradiated at ~4500 Gy s^{-1}. The yellow grids indicate the TLD matrices and the black dashed lines are the contour lines of the fitted Gaussian beam profile, whose center is marked by the red dot. The calculated beam central dose is also indicated for each irradiation. (d)–(f) represent the residual maps, where each square is a TLD.

2D measurements with sample array

[5] Motta, S., Christensen, J.B., Togno, M., Schäfer, R., Safai, S., Lomax, A.J. and Yukihara, E.G., 2023. Characterization of LiF: Mg,Ti thermoluminescence detectors in low-LET proton beams at ultra-high dose rates. Physics in Medicine & Biology, 68(4), p.045017. https://doi.org/10.1088/1361-6560/acb634

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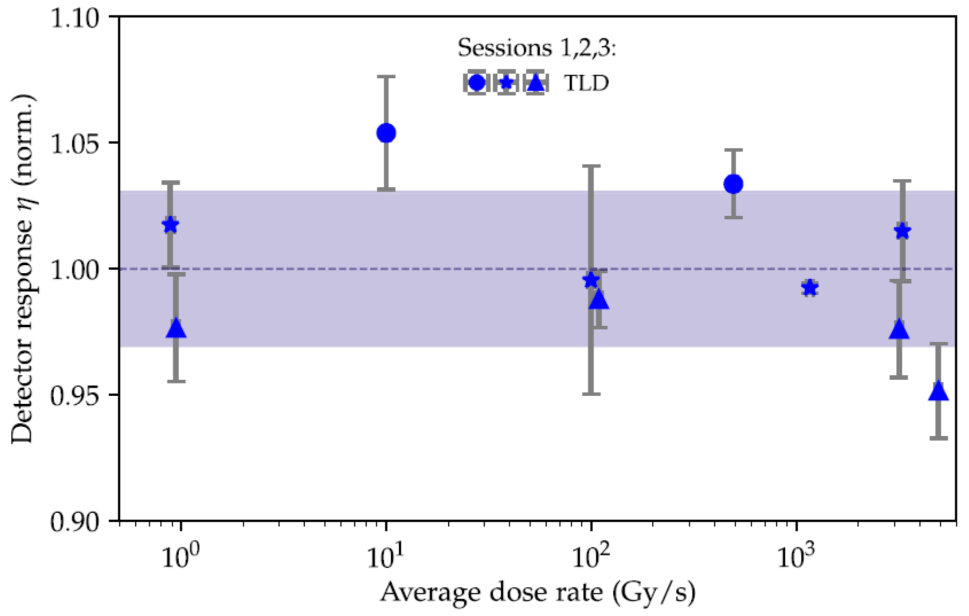

Figure 9. Response of TLD-100™ Microcubes as a function of the dose rate, normalized to the average value. The blue band shows one standard deviation around the mean. The order of the symbols follows the irradiation session numbering.

Different dose rates

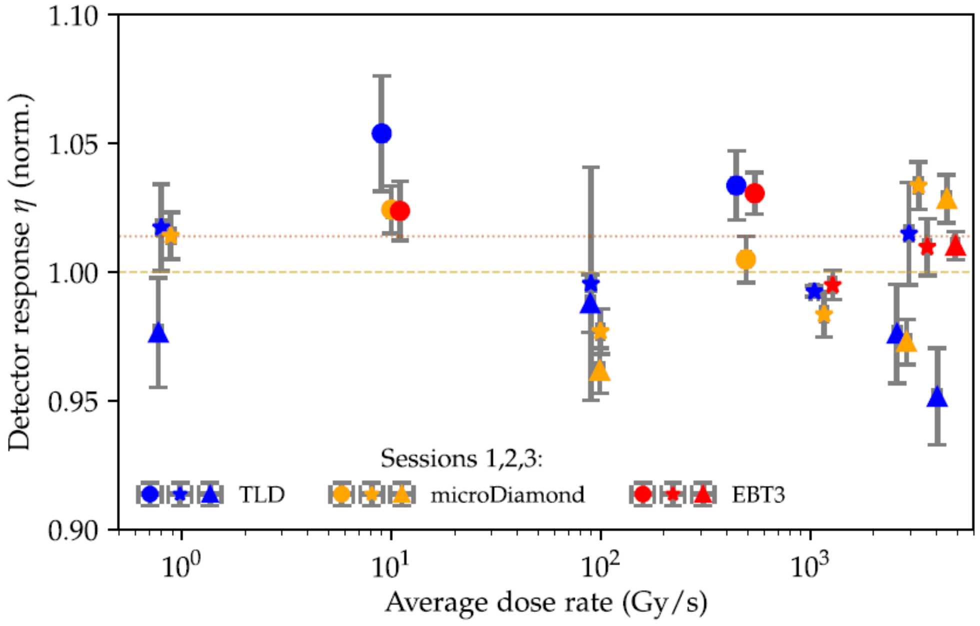

Figure 10. The TLD response compared to that of the reference detectors (microDiamond and EBT3 films) as a function of the dose rate. The data are normalized to the average response of TLD-100™ Microcubes. The dashed yellow line represents the average response of the microDiamond and the red dotted line the EBT3 films one. The order of the symbols follows the irradiation session numbering. The data relative to one dose rate in an irradiation session were offset (±10%) to help the reader distinguish the data and the uncertainties.

Different dose rates checked with different detectors

[5] Motta, S., Christensen, J.B., Togno, M., Schäfer, R., Safai, S., Lomax, A.J. and Yukihara, E.G., 2023. Characterization of LiF: Mg,Ti thermoluminescence detectors in low-LET proton beams at ultra-high dose rates. Physics in Medicine & Biology, 68(4), p.045017. https://doi.org/10.1088/1361-6560/acb634

5. Successful experiences of FLASH-RT measurements using TLDs/OSLDs

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Investigation of TL and OSL detectors in ultra-high dose rate electron beams

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[6] Motta, S. Christensen,. J. B. Frei, F. Peier, P. and Yukihara, E.G. 2023 Phys. Med. Biol. In Press https://doi.org/10.1088/1361-6560/acdfb2

5. Successful experiences of FLASH-RT measurements using TLDs/OSLDs

Table 1: Properties, preparation, and readout parameters for the TL and OSL materials employed in this study.

Material	Size (mm^3)	Annealing or bleaching	Readout technique	Reader parameters [d,e]	t_R (s)	Signal
LiF:Mg,Ti (TLD-100™) [a]	3.2 × 3.2 × 0.9	400 °C for 1 h + 100 °C for 2 h	TL	5 °C s^{-1}, 400 °C, IRSL/TL-410 nm [f]	5	Area peak 4+5
LiF:Mg,Cu,P (TLD-100H™) [a]	3.2 × 3.2 × 0.9	240 °C for 1 h	TL	1 °C s^{-1}, 240 °C, BSL/TL-365 nm (25 %) [g]	100	Area peak 4
CaF_2:Tm (TLD-300™) [a]	3.2 × 3.2 × 0.9	400 °C for 1 h	TL	5 °C s^{-1}, 400 °C, BSL/TL-365 nm (25 %) [g]	5	Area peak 3+5
Al_2O_3:C (Luxel™) [b]	~3 × 3 × 0.3	Green LED	CW-OSL	Green LED, 60 mW cm^{-2}, 300 s, GSL/TL-365 nm (OD2) [h]	300	Area OSL curve
BeO [c]	4.7 × 4.7 × 0.5	900 °C for 10 min	LM-OSL	Blue LED, 80 mW cm^{-2}, 240 s, BSL/TL-365 nm (25 %) [g]	100	Area OSL curve

Producer: [a] Thermo Fischer Scientific Inc. [b] Landauer Inc. [c] Materion Corp.
[d] TL: heating rate, maximum temperature, detection filter combination
[e] OSL: light stimulation, maximum irradiance, stimulation duration, detection filter
[f] Schott BG 39 (3 mm) + Semrock HC 414/46 (3.5 mm)
[g] Hoya U340 (2.5 mm) + Delta BP 365/50 EX (4.5 mm) + Schott NG 4 (1 mm)
[h] Schott BG 3 (2 mm) + Delta BP 365/50 EX (5 mm) + Thorlabs ND 20B (1 mm)

Luminescent materials used

2.2 Irradiations

2.2.1 Irradiations with the electron beam at METAS

The irradiations were carried out with a Scanditronix microtron accelerator (Uppsala, Sweden) at the Swiss Federal Institute of Metrology (METAS). A 15 MeV electron beam with 3 µs pulse duration was delivered in all irradiation sessions. The pulse repetition frequency was varied between 1 Hz and 25 Hz, as indicated in table 2.

Beam parameters

Table 2: Beam parameters for the four irradiation sessions. D_{pulse} and D_{total} indicate the dose per pulse and the total delivered dose, respectively, as derived from the integrating current transformer, calibrated in absorbed dose to water against a reference ionization chamber. The instantaneous dose rate $\dot{D}_{inst}$ is the ratio of the dose per pulse and the pulse duration (3 µs). $u(D_{total})$ is the uncertainty of the delivered dose and z_{ref} is the reference depth in water chosen for the irradiations.

# pulses	Frequency (Hz)	D_{pulse} (mGy)	D_{total} (Gy)	$u(D_{total})$ (%, $k=1$)	$\dot{D}_{inst}$ (kGy s^{-1})	z_{ref} (g cm^{-2})
224	25	3.6	0.8	1.1	1	3.30
7	1	125	0.9	1.5	42	3.29
2	1	586	1.2	1.2	195	3.50
1	1	970	1	2.9	324	3.51

[6] Motta, S. Christensen,. J. B. Frei, F. Peier, P. and Yukihara, E.G. 2023 Phys. Med. Biol. In Press https://doi.org/10.1088/1361-6560/acdfb2

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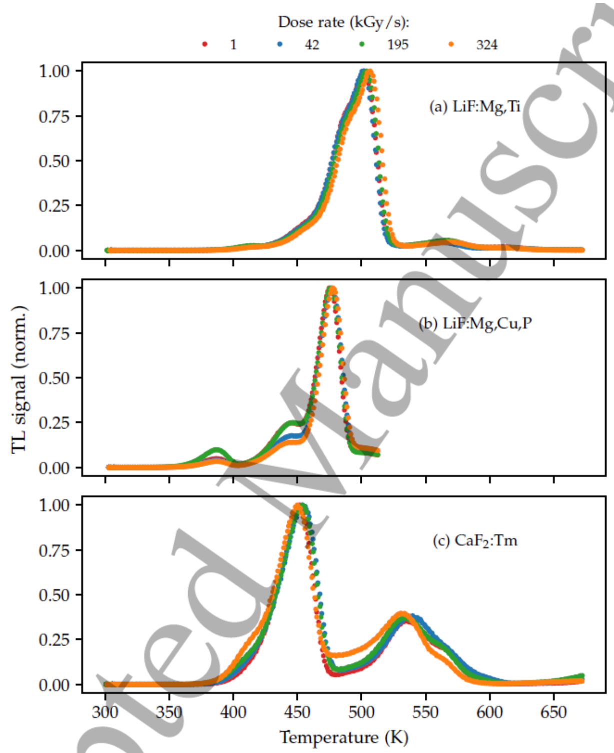

Figure 7: Examples of TL curves for each dose rate for the investigated materials: (a) LiF:Mg,Ti, (b) LiF:Mg,Cu,P, (c) CaF_2:Tm. The TL curves are normalized by the maximum intensity. No trend of the TL curves with the dose rate was observed.

Dose-rate evaluation for the TLDs used

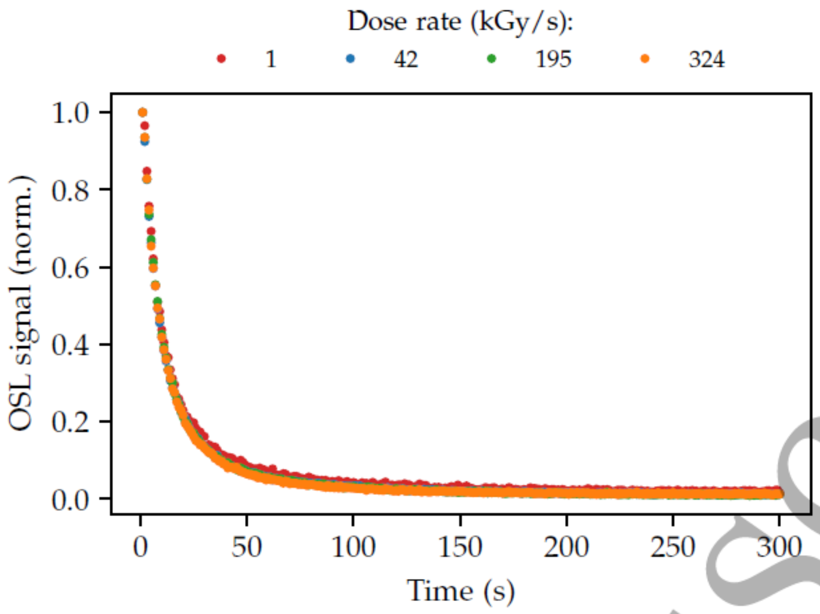

Figure 8: Example of an OSL curve from Al_2O_3:C for each dose rate. The OSL curves are normalized to the maximum intensity.

Dose-rate evaluation for the OLDs used

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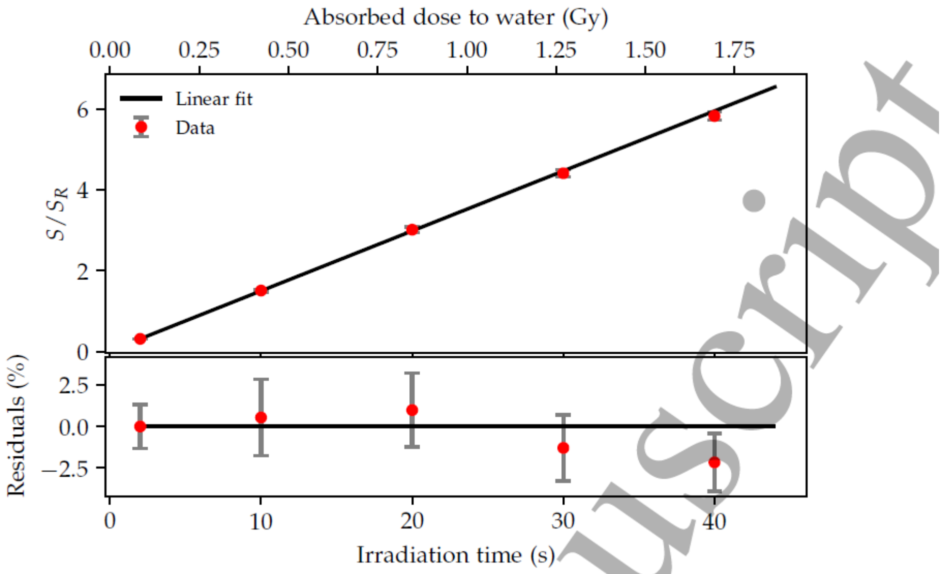

Figure 5: S/S_R calibration curve of LiF:Mg,Ti obtained with the beta source in the Lexsyg Smart reader. Each point denotes the average of five detectors and the error bars represent the standard deviation of the data. The data points were fitted with a linear function, whose residual plot is reported in the lower figure. The conversion from indicated value (irradiation time with the reader source) to absorbed dose to water was obtained by irradiating the detectors in the reference electron beam at 1 kGy s^{-1} (instantaneous dose rate).

S/S_R calibration

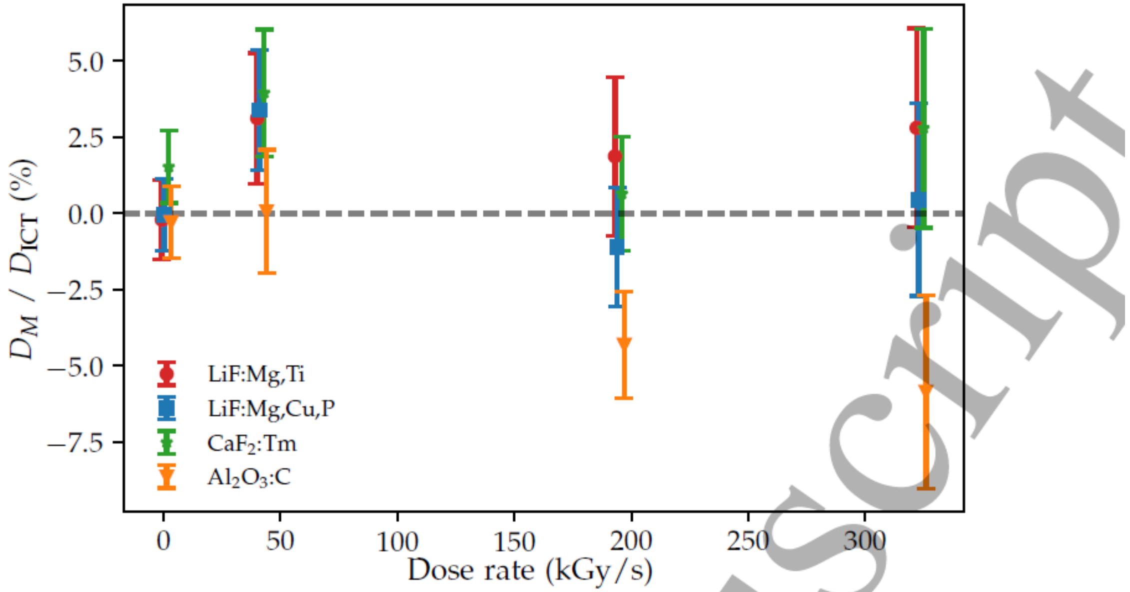

Figure 11: Deviation of the investigated TL and OSL materials relative to the dose estimated from the reference ICT. The TLD/OSLD measured dose D_M is calculated considering the central detectors around (0,0) and according to the formalism described in Section 2.3. The error bars represent the uncertainties ($k = 1$) of the ratios, which include the uncertainties of the dose measured by the TLDs/OSLDs and the uncertainty of the delivered dose (see table 3). A horizontal offset was introduced to help distinguish the data and the error bars.

Measurent comparison with reference ICT estimation

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Summary

- TLDs and OSLDs are suitable for doing FLASH-RT dosimetry measurements;
- They can provide point-measurements or 2D measurements with array of sensors within mm resolution;
- They've been proven to be dose rate independent for protons and electrons at UHDR regimes;
- "S/S_R method" of measurement is already standardized at PSI;
- Some care is needed regarding dose averaging effects on pencil beams;
- Reader with built-in beta source is a "must have" to obtain the most accurate measurement possible;
- We could do this type of measurement in our FLASH*Lab*@PITZ

Thank you