# Chapter X: Optically stimulated luminescence dosimeters

Optically stimulated luminescence dosimeters

- Again → luminescence process → now stimulation by light (UV, visible, IR)
- When optically stimulated luminescence is used in radiation dosimetry → OSL dosimeter
- Fast-expanding during last years
- Remark → when a default is **excited** by light → radiophotoluminescence (RPL) → end of the chapter
- References:
  - L. Bøtter-Jensen, S.W.S. McKeever, A.G. Wintle, *Optically stimulated luminescence dosimetry* (Elsevier Science) 2003
  - S.W.S. McKeever, Optically stimulated luminescence dosimetry, Nuclear Instruments and Methods in Physics Research B: 184 (2001) 29–54

# History

- First « practical » OSL measurements have been made in 1985 with quartz and feldspar → via the determination of the environmental dose absorbed by these materials → determination of the age of archeological or geological materials
- The measure of the luminescence of the material is a measure of the dose absorbed since the last exposition to the light → by comparison with reference samples → dating
- For personal dosimetry → late development due to the absence of « good » materials (sensitivity to radiation, small Z<sub>eff</sub>, weak fading,...)
- Currently → development of « good » materials → Al<sub>2</sub>0<sub>3</sub>:C, quartz,...

# Advantages of OSL

- Techniques of reading simpler than for TLD  $\rightarrow$  only optical
- No heating  $\rightarrow$  no thermal quenching
- Possible multiple reading → no necessary to stimulate all traps during the first reading
- Fast reading process

### Escape probability p

• Thermoluminescence  $\rightarrow$ 

$$p = s \exp\left(-\frac{E}{kT}\right)$$

- Optical stimulation at a fixed wavelength  $\lambda \rightarrow$ 

$$p(E_0) = \Phi\sigma(E_0)$$

with  $\Phi$ , the optical stimulation intensity,  $\sigma$ , the photoionization cross section for interaction of the metastable state and  $E_0$ , the threshold optical stimulation energy required to release the e<sup>-</sup> from the trap

#### Photoionization cross section (1)

- $\sigma(E_0)$  has an energy threshold  $\rightarrow$  for  $E < E_0 \rightarrow \sigma(E_0) = 0$
- $\neq$  expressions have been derived for the dependence as a function of the frequency of  $\sigma(E_0) \rightarrow \sigma(h\nu, E_0)$  as a function of the potential energy in the vicinity of the defect
- For a shallow trap  $\rightarrow$  hydrogenic potential (and planewave for the e<sup>-</sup>)  $\rightarrow$   $(h\nu - E_0)^{3/2}$

$$\sigma(h\nu, E_0) \propto \frac{(h\nu - E_0)^{3/2}}{(h\nu)^5}$$

For a deep trap → delta-function potential for the defect (model of Lucovsky) with the assumption that the mass of the e<sup>-</sup> in the localized state is equal to the effective mass (m<sub>e</sub>) of the e<sup>-</sup> in the conduction band →

$$\sigma(h\nu, E_0) \propto \left[\frac{4(h\nu - E_0)E_0}{(h\nu)^2}\right]^{3/2}$$

#### Photoionization cross section (2)

 For a deep trap → model of Grimmeis and Lebedo considering a mass m<sub>0</sub> (rest mass) for the e<sup>-</sup> in the localized state →

$$\sigma(h\nu, E_0) \propto \frac{(h\nu - E_0)^{3/2}}{h\nu[h\nu - E_0(1 - m_0/m_e)]^2}$$

 Other models exist but for OSL → deep traps → the models of Lucovsky or of Grimmeis and Lebedo are most often used



 $\sigma(h\nu, E_0)$  for  $E_0 = 3.0$  and  $m_0/m_e = 2$  (normalized functions to reach  $\sigma_{max} = 1$ ): 1: hydrogenic, 2: Lucovsky, 3: Grimmeis and Lebedo

#### Modes of optical stimulation

- 1.  $\Phi$  = constant (and  $\lambda$  is constant also)  $\rightarrow$  optically stimulated luminescence in continuous mode (continuous-wave OSL: CW-OSL)
- 2. Introduction of a time-dependence for  $\Phi(t)$  or  $\lambda(t) \rightarrow$  example 1: linear increase in the intensity of optical stimulation at a fixed  $\lambda$ (with  $\beta_{\Phi} = d\Phi/dt$ )  $\rightarrow$  $\Phi(t) = \Phi_0 + \beta_{\Phi}t$

 $\rightarrow$  optically stimulated luminescence in linear mode (linear modulation OSL: LM-OSL)

3.  $\rightarrow$  example 2: pulsed stimulation  $\rightarrow$  optically stimulated luminescence in pulsed mode (pulsed OSL: POSL) with  $\Delta t$ , the pulse width and  $\tau$ , the period  $\rightarrow$ 

$$\begin{cases} \Phi(t) = \Phi_0 & \text{for } t_0 \le t < t_0 + \Delta t \\ \Phi(t) = 0 & \text{for } t_0 + \Delta t \le t < t_0 + \tau \end{cases}$$

# Schematic representation of the three main OSL stimulation modes



#### CW-OSL

- Most often used mode in practice because of its ease of application
- Simplest model: 1 trap 1 recombination center (as for TL)



• Additional complexities can be subsequently considered

# 1 trap – 1 recombination center: 1<sup>st</sup> order

- Quasi-equilibrium assumption (QE)
- Negligible retrapping
- Equation similar to equations obtained for TL  $\rightarrow$

$$I_{OSL}(t) = -\frac{dm}{dt} = -\frac{dn}{dt} = np$$
$$I_{OSL}(t) = n_0 p \exp(-tp) = I_0 \exp(-t/\tau_d)$$

with  $\tau_d^{-1} = p = \Phi \sigma$  with  $\tau_d$  the CW-OSL decay constant

- 1<sup>st</sup> order model and exponential decrease of the light intensity
- When all traps are empty  $I_{OSL} = 0$



•  $n_0$  is equal to the area of  $I_{OSL}(t) \rightarrow$ 

$$\int_{0}^{\infty} I_{OSL}(t)dt = n_0$$

• The initial intensity is  $\propto$  to  $n_0$  and  $\Phi \rightarrow I_{OSL}(0) = n_0 \Phi \sigma$ 

#### 1 trap – 1 recombination center: 2<sup>nd</sup> order

• Significant retrapping rate  $\rightarrow$ 

$$I_{OSL}(t) = np - n_c(N - n)A_n$$

• For  $N \gg n$  and  $R = A_n/A_m \gg n/(N-n) \rightarrow$  we obtain the 2<sup>nd</sup> order shape  $\rightarrow$  $n^2p \quad dn$ 

$$I_{OSL}(t) = \frac{1}{NR} = -\frac{1}{dt}$$

$$I_{OSL}(t) = I_0 \left(1 - \frac{n_0 p t}{NR}\right)^{-2}$$
with  $I_0 = \frac{n_0^2 p}{NR}$ 

1 trap – 1 recombination center: general-order

$$I_{OSL}(t) = I_0 \left(1 - \frac{n_0 p t}{NR}\right)^{\frac{b}{1-b}}$$

with 
$$I_0 = \frac{n_0^b p}{NR}$$

# Models with multiple traps and centers



2 traps including one deep trap not sensitive to the optical

2 traps including one shallow trap for which thermoluminescence is

2 recombination centers including a non-radiative center

#### Independent traps

 We consider 2 traps (1 and 2) optically sensitive and independent → we have the traps concentrations n<sub>1</sub> and n<sub>2</sub> and the escape probabilities p<sub>1</sub> = τ<sub>d1</sub><sup>-1</sup> and p<sub>2</sub> = τ<sub>d2</sub><sup>-1</sup> → superposition principle →

$$-\frac{dm}{dt} = -\frac{dn_1}{dt} - \frac{dn_2}{dt}$$

$$I_{OSL}(t) = n_{10}p_1 \exp(-tp_1) + n_{20}p_1 \exp(-tp_2)$$
  
=  $I_{01} \exp(-t/\tau_1) + I_{02} \exp(-t/\tau_2)$ 

• Superposition principle can be applied to more than 2 traps

## Additional deep trap without optical stimulation

- Additional deep trap without optical stimulation (trap 2) occurs for a stimulation with long wavelength and thus with insufficient energy to empty the deep traps → the deep trap is a competitor to the recombination center
- The OSL intensity can be written (with N<sub>2</sub>, n<sub>2</sub> and A<sub>n2</sub>, the concentration of available traps, of filled traps and the trapping probability for the deep trap) →

$$I_{OSL}(t) = n_{10}p \exp\left(-t/\tau_d\right) - n_c A_{n2}(N_2 - n_2)$$

- The intensity is reduced due to the trapping in the deep trap
- The decrease is no more exponential

#### Additional shallow trap with thermoluminescence

 $I_{OSL}(t) = n_{10}p \exp\left(-t/\tau_d\right) + n_2 s \exp\left(-E/kT\right) - n_c A_{n2}(N_2 - n_2)$ 

- The last 2 terms combine to produce a long-lived and temperature-dependent component to the OSL decay curve
- That may imply (as a function of the parameters) an initial increase of I<sub>OSL</sub> followed by a decrease → in such circumstances the terms « OSL decay curve » is inappropriate

# Second recombination center (non-radiative)

- A second recombination center (2) may exist and be nonradiative (or if radiative, outside the detector sensitivity window)
- Let be m<sub>1</sub> and m<sub>2</sub>, the h<sup>+</sup> concentrations at R<sub>1</sub> and R<sub>2</sub>
- As only the recombination  $e^{-}$  h<sup>+</sup> at 1 is radiative  $\rightarrow$

$$I_{OSL}(t) = -\frac{dm_1}{dt}$$
$$I_{OSL}(t) = np \exp\left(-t/\tau_d\right) - \frac{dm_2}{dt}$$

• Again decrease of I<sub>OSL</sub>



## More complex models

- In previously models → we add 1 deep trap <u>or</u> 1 shallow trap <u>or</u> 1 recombination center
- For realistic materials → they contain multiple traps, deep or shallow, and multiple recombination centres (radiative or not)
   → model with all these elements →



# Results for this complex model



# LM-OSL

Alternative method introduced in 1996 by Bulur → the intensity of the stimulation linearly increases

$$\Phi(t) = \Phi_0 + \beta_\Phi t$$

- The OSL response is a series of peaks, each peak corresponding to the optical escape of charge from a given type trap → the traps with the largest photoionization cross section are empty first
- The traps characterized by slow, medium or fast escape are more easily discriminate than for CW-OSL
- Not very often used in practice due to a larger technical complexity than for CW-OSL but also to a larger mathematical complexity

# 1 trap – 1 recombination center: 1<sup>st</sup> order

- 1<sup>er</sup> ordre → hypothèses de quasi-équilibre et de repiégeage négligeable
- Considérons comme forme pour  $\Phi(t) \rightarrow$

$$\Phi(t) = \gamma t$$

$$\frac{dn}{dt} = -\sigma\gamma tn$$

• Cette équation a comme solution une fonction gaussienne  $\rightarrow$ 

$$n = n_0 \exp\left(-\frac{\sigma\gamma}{2}t^2\right)$$
$$I_{OSL} = n_0 \sigma\gamma t \exp\left(-\frac{\sigma\gamma}{2}t^2\right)$$

#### Caractéristiques du 1<sup>er</sup> ordre

- The OSL curve shape starts from 0 at *t* = 0
- The curve shows a maximum at

$$t = t_{max} = \sqrt{1/\sigma\gamma}$$

- For known  $\gamma \rightarrow$  it is possible to obtain  $\sigma$
- The maximum intensity of the curve is  $\rightarrow$

$$I_{OSL}^{max} = \frac{n_0}{t_{max}} \exp\left(-\frac{1}{2}\right)$$

• The integration of *I*<sub>OSL</sub> gives n<sub>0</sub>

#### Examples of LM-OSL curves



#### Multiple independent traps

• The superposition principle can be applied  $\rightarrow$  for k traps characterized by their occupation  $n_i$  and their photoionisation cross section  $\sigma_i \rightarrow$ 

$$I_{OSL} = \gamma t \sum_{i=1}^{k} n_{0i} \sigma_i \exp\left(-\frac{\sigma_i \gamma}{2} t^2\right)$$

• OSL curve  $\rightarrow$  sum of k curves having each a maximum dependent on  $\sigma_i \rightarrow$ 



Characteristics at general-order

- QE but important retrapping rate
- For 1 type of trap  $\rightarrow$

$$\frac{dn}{dt} = -\frac{\sigma\gamma tn^b}{n_0^{b-1}}$$
$$I_{OSL} = n_0\sigma\gamma t \left[ (b-1)\frac{\sigma\gamma t^2}{2} + 1 \right]^{b/(1-b)}$$

• Maximum  $t_{max}$  for a value of  $I^{max}$   $\rightarrow$ 

$$t_{max} = \sqrt{\frac{2}{\sigma\gamma(b+1)}} \quad \text{and} \ I_{OSL}^{max} = \left(\frac{2n_0}{b+1}\right) \left(\frac{1}{t_{max}}\right) \left(\frac{2b}{b+1}\right)^{b/(1-b)}$$

 Attention → for several types of traps → retrapping implies that the traps are **not** independent → the superposition is not possible as for 1<sup>st</sup> order → complexity

### POSL

- POSL allows to efficiently separate luminescence and stimulation lights
- Important decrease of the background due to the scattered stimulation light
- Very powerful method for weak dose dosimetry

# Principles of POSL (1)

- We consider several stimulation pulses (supplied by LED or laser) of different intensities  $\Phi_i$  and durations  $T_i$  such that  $\Phi_i T_i = \Phi T = \text{constant}$  (fixed absorbed energy by pulse).
- By considering the 1<sup>st</sup> order model  $\rightarrow$

$$\Delta n = \int_0^T n\sigma \Phi dt$$

For a weak stimulation (∆n ≪ n → the concentration of charges released by pulse is negligible compared to the concentration of trapped charges) → ∆n ∝ ΦT → the charge released from the traps is approximately equal after each pulse

# Principles of POSL (2)

- Normally when writing rate equations → we consider that each recombination event leads **instantly** to a photon emission event
- In reality → the recombination center is first excited (luminescence center) before relaxation by light emission → existence of a built-in delay τ (lifetime of the excited state)
- If we have  $n_e$ , the concentration of excited states  $\rightarrow$

$$\frac{dn_e}{dt} = \frac{dm}{dt} - \frac{n_e}{\tau}$$

• One photon is emitted when the excited state relaxes  $\rightarrow$ 

$$I_{OSL} = \frac{n_e}{\tau}$$

# Principles of POSL (3)

- If  $\tau \ll T \rightarrow$  conditions of quasi-equilibrium  $\rightarrow dn_e/dt \approx 0$  and  $I_{OSL} \approx -dm/dt$  as usual (as for CW-OSL)
- If \(\tau \ge T \rightarrow \neq \text{ situation }\rightarrow \text{ a certain number of centers relaxe after the end of the stimulation pulse
- If τ ≫ T → most of the photons are emitted after the pulse (for Al<sub>2</sub>O<sub>3</sub>:C → 90% of the light emerges after the pulse if it is narrow enough)
- For  $t < T \rightarrow I_{OSL} \nearrow$  when  $t \nearrow$  (optical stimulation of the traps)
- For t > T  $\rightarrow$   $I_{OSL} \supseteq$  when t  $\nearrow$  (relaxation with a time constant  $\tau$ )

#### Schematic illustration of POSL



- POSL after pulses with Φ = 10<sup>3</sup>, 10<sup>2</sup> and 22 u.e./s → we have the corresponding T: T = 6.6, 66 and 300 ms (τ = 100 ms was chosen)
- The ratio of the areas after during the pulse ↗ for T ↘

**Experimental illustration of POSL** 



POSL for  $Al_2O_3$ :C stimulated by laser during 0.1 ms

Ratio of the areas after and during the pulse for  $Al_2O_3$ :C

## Determination of the dose by POSL

- The total integration of the POSL curve corresponding to 1 pulse is equal to the charges released from the traps and is thus  $\propto$  to  $\rm n_0$
- The integration of the curve after the end of the pulse is also  $\propto$  to the charges released from the traps and is thus also  $\propto$  to n<sub>0</sub> but the proportionality coefficient changes as a function of the width T of the pulses
- With ∆n ≪ n → only a few stimulation pulses are enough → to make several measurements is possible
- POSL especially applied with Al<sub>2</sub>O<sub>3</sub>:C but very promising
# $Al_2O_3:C$

- $Al_2O_3 \rightarrow as \alpha Al_2O_3$ : corundum or alpha alumina
- Al<sub>2</sub>O<sub>3</sub>:C is the material most often used in OSL
- Al<sub>2</sub>O<sub>3</sub>:C is the only material used in POSL

# $Al_2O_3$ :C: Structure of $\alpha$ - $Al_2O_3$

- The crystalline structure of α-Al<sub>2</sub>O<sub>3</sub> is a slightly distorted hexagonal-closepacked O<sup>2-</sup> lattice with Al<sup>3+</sup> ions occupying two out of every three octahedral interstices
- Each Al<sup>3+</sup> ion is surrounded by 6 octahedral nearest-neighbor O<sup>2-</sup> ions



## Defects and impurities

- The thermodynamic defects are oxygen or aluminium vacancies
   → centers of type F and V, respectively
- The O<sup>2-</sup> vacancy implies a charge compensation  $\rightarrow$ 
  - the occupancy of the vacancy by 2  $e^{-}$  gives rise to a neutral F-center
  - the occupancy of the vacancy by  $1 e^{-}$  gives rise to a positive F<sup>+</sup>-center
- The addition of carbon impurities causes significant increase the sensitivity of the material
- The divalent C<sup>2+</sup> carbon ion replaces a trivalent Al<sup>3+</sup> ion → charge compensation → creation of oxygen vacancies as F<sup>+</sup>center

# Configuration of an F<sup>+</sup>-center charge-compensated by a divalent carbon ion



#### **Emission mechanisms**

- The sensitivity of Al<sub>2</sub>O<sub>3</sub>:C depends on the concentration of F<sup>+</sup>centers that are the recombination centers
- The main emission band of the Al<sub>2</sub>O<sub>3</sub>:C is a quite large band centered at 420 nm and ascribed to a F-center of luminescence
- The e<sup>-</sup> are released from the traps and recombine with F<sup>+</sup>-center
   → creation of excited F-center relaxing and giving luminescence
- The lifetime of the excited F-center is 35 ms at ambient T

$$F^+ + e^- \longrightarrow F^* \xrightarrow{\tau = 35 \text{ ms}} F + h\nu_{420\text{nm}}$$

• The identity of the defects responsible for the capture of the charge carriers during the ionization is not known

## Emission spectrum of Al<sub>2</sub>O<sub>3</sub>:C



## Dosimetric properties of $Al_2O_3$ :C (1)

- The energy band-gap of Al<sub>2</sub>O<sub>3</sub> is wide (~ 9.5 eV) → allows to engineer deep and thermally stable traps and color centers (little sensitive to T)
- Al<sub>2</sub>O<sub>3</sub>:C is very sensitive to radiation (sensitivity → ratio between the number of emitted photons and the absorbed dose) because of the absence of thermal quenching and of very small noise (especially for POSL) → 60 × more sensitive than the TLD-100 which is the reference
- This very good sensitivity allows to make measurements at very small doses  $\rightarrow$  *D* = 1  $\mu$ Gy
- Al<sub>2</sub>O<sub>3</sub>:C shows a linear response to the dose in a large range → 1 µGy < D < 10 Gy → on 5 decades (supralinearity appears at 10 Gy and sublinearity at 100 Gy)

#### Dose response of Al<sub>2</sub>O<sub>3</sub>:C



« strong » and « weak » characterize the stimulation power (1.2 W and 0.01 W)

## Dosimetric properties of $Al_2O_3$ :C (2)

- Possible rereading of the dosimeter → due to the large sensitivity of Al<sub>2</sub>O<sub>3</sub>:C → in CW-OSL a measurement is precise enough if only a part of the traps is emptied → by considering the fraction of charges released during the 1<sup>st</sup> measurement, a second measurement is possible (or a 3<sup>th</sup>, etc.)
- In POSL, rereading is more obvious → only a few stimulations pulses are enough → such as ∆n ≪ n → measure could be redo
- Also possibility to reuse the OSL dosimeter after heating at high T to empty the traps
- Al<sub>2</sub>O<sub>3</sub>:C is not tissue-equivalent (Z<sub>eff</sub> = 10.2 ≠ 7.35) → energy dependence (overestimation) for E < 200 kEV → 1 filter could correct this problem</li>

## Fading

• Fading comparable to TLD-100 (5%/an)



 A transient signal appears for t < 10 min → due to shallow traps which are quickly empty → afterwards stable signal

## Characteristics of Al<sub>2</sub>O<sub>3</sub>:C and other OSL

	Rel. OSL	Linear dose	Stimulation wavelength	Main emission wavelength	Fading rate in	Effective Z (Tissue
USL material	Sensitivity	range	(nm)	(nm)	сагк	=7.4)
Al <sub>2</sub> O <sub>3</sub> :C (Landauer Inc.)	1.00	µGy-10 Gy	450-550	~420	<5% /y	10.2
Al <sub>2</sub> O <sub>3</sub> :C (BARC, India)	~0.3	50 μGy- 1 Gy	470	420	negligible in a month	10.2
**Al <sub>2</sub> O <sub>3</sub> :B	~0.17	100 µGy- 15 Gy	470	420	Negligible in a month	10.2
BeO sintered pellets (Germany)	~1.00	µGy-10 Gy	~435	~335	1% in six months	7.2
MgO:Tb	weak	100 µGy- 10 Gy	500-560	375,420,440, 470,500,650	43% in 1 <sup>st</sup> 36h and then nil	10.8
NaMgF <sub>3</sub> :Eu	~10.0	μGy- 100 Gy	~470	360	40% in 1 <sup>st</sup> 24h and then nil	10.4
Mg₂SiO₄:Tb	~0.11	30 µGy- 10 Gy	<532	370,420 and 440	30% in 1 <sup>st</sup> 10h and then nil	11.23
<sup>∗</sup> LiMgPO₄:Tb,B (BARC, India)	~1.00	20 µGy -1 kGy	470	380,417,440	16% in 4 days and stable thereafter	11.20

# Al<sub>2</sub>O<sub>3</sub>:C dosimeters in practice

- As monocrystal
  - $\rightarrow$  very sensitive  $\rightarrow$  excellent for the measurement of small doses
  - $\rightarrow$  may be heated  $\rightarrow$  deep traps are emptied  $\rightarrow$  may be indefinitely reused
  - → lack of uniformity between 2 crystals → large variability between dosimeters of this type → individual calibration
- As powder (Landauer Inc.) → crystals grinded into powder and embedded in a plastic matrix
  - ightarrow very good uniformity
  - → cannot be heated → deep traps are not emptied → loss of sensitivity in the end
- Microcrystals and optical fiber
  - ightarrow in-vivo dosimetry
  - ightarrow real-time dosimetry
  - ightarrow to be investigated



 $\phi$  = 300  $\mu$ m

#### **OSL** Readers: Basic properties

A basic installation requires:

- 1. A light source for the stimulation (LED, laser,...)
- 2. Filters to select the wavelength of the light source
- 3. Filters to select the wavelength of the luminescence light
- 4. A light detector (photomultiplier,...)
- 5. The associated electronics



### Wavelength of the stimulation light

- The more efficiency wavelength of the stimulation light is blue  $(\lambda \approx 470 \text{ nm})$  but it too close of the maximum of luminescence  $(\lambda = 420 \text{ nm})$
- In practice in dosimetry green is used ( $\lambda \approx 525$  nm)

## OSL reader in practice (1)

- OSL reader for research
  - $\rightarrow$  High-capacity automatization
  - $\rightarrow$  allows OSL or TLD measurements
  - ightarrow calibration source is often incorporated
  - ightarrow equipped with blue and/or green LEDs



## OSL reader in practice (2)

• OSL Landauer reader  $\rightarrow$  in POSL mode

→ OSL stimulated by laser Nd:YAG (532 nm) with short period → the OSL intensity is measured between pulses → as a function of the dose → ≠ stimulation powers

- OSL Landauer reader  $\rightarrow$  in CW-OSL mode
  - $\rightarrow$  short stimulation (1s) by green LEDs
  - → number of LEDs (and thus power) depending as a function of the dose to be measured

# OSL reader in practice (3)

- System with optical fiber
  - → the optical fiber delivers the OSL stimulation OSL and carries back the luminescence
  - $\rightarrow$  green laser for stimulation
  - $\rightarrow$  also POSL mode
  - $\rightarrow$  allows in-vivo and real-time measurements



## OSL dosimeter: In practice at ULB (1)

- Dosimeters Landauer IPLUS in Al<sub>2</sub>O<sub>3</sub>:C
- From crystals in aluminium oxide → aluminium oxide powder applied between 2 plastic films



### OSL dosimeter: In practice at ULB (2)

- For IPLUS → optical stimulation realized from a set of electro luminescent diodes emitting continuously a 532 nm light (green) → CW-OSL
- Signal emitted from detectors (blue light of about 420 nm) measured with a photomultiplier



## OSL dosimeter: In practice at ULB (3)

- Dosimeter re-analyze: The optical stimulation maintain more than 99% of information in the detector → possible multiple reading and storage of the dosimeter for later investigations
- No required calibration: The sensitivity of the detector is determined during the fabrication process → the sensitivity value carved on the detector is automatically taken into account during the reading process
- Stable sensitivity in time: Sensitivity defined for ever
- Very weak fading: Smaller than 1,5 % / month and <4% / year → the fading does imply to be corrected during the dose estimation
- **Response to neutrons:** No neutron sensitivity highlighted here

OSL dosimeter: In practice at ULB (4)

- Temperature and hygrometry: Range of use: -10°C to 40°C and from 0 % to 90%
- **Exposition to light:** no particular restriction in normal conditions
- Strong and light dosimeter: Strong, compact, light, fully customize dosimeters
- Types of radiations:
  - photon: energies from 16 keV to 6 MeV (with a maximum energy of 18 MeV)
  - beta: energies larger than 250 keV
- **Possible reutilization**: dose maximum of 100 mSv and maximum number of reutilizations: larger than 1000
- Homogeneity of the pack sensitivity: < 10% (generally < 2,4%)

## OSL dosimeter: In practice at ULB (5)

- Detector with :
  - A detector card with 4 pastilles in  $Al_2O_3$ :C
  - A filter holder box with a barcode printed in a legible form and composed of 4 filters: plastic, aluminium, titanium, tin





## Radiophotoluminescence dosimetry (RPL)

- Again luminescence process stimulated by light
- For radiophotoluminescence (RPL) → no ionization of the defect but excitation of the defect → during relaxation → emission of light
- Main advantage → the trapped carriers are preserved → the measurement can be indefinitely repeated → storage
- Process ≠ from OSL → another class of dosimeter (even with optical stimulation)
- Very recent development → this technology need to prove itself
- Reference:

T. Yamamoto, D. Maki, F. Sato, Y. Miyamoto, H. Nanto, T. Iida, *The recent investigations of rdiophotoluminescence and its application*, Radiations Measurements 46 (2011) 1554-1559

## History

- Discovery and first application in 1951 by Schulman
- Problem → Photoluminescence inherent to considered materials → emission of light without irradiation → important noise → brake on its use while properties are extremely interesting
- Recently → development of a pulsed laser system + perfectly controlled materials + improved electronics → decrease of the noise to a correct level
- Possibly a very valued method in the future (currently: Japan and IRSN in France)

# Simple model for RPL (1)



- We have n<sub>1</sub>, the concentration of e<sup>-</sup> trapped in 1 (ground state of the trap), n<sub>2</sub>, the concentration of e<sup>-</sup> trapped in 2 (excited state of the trap), α and β, the transition probabilities 1 → 2 and 2 → 1
- The material is exposed to a light source with short wavelength (UV) and with intensity I<sub>in</sub> (constant) → the result is that a light with a larger wavelength and with intensity I<sub>out</sub>

#### Simple model for RPL (2)

• The equations of the process are  $\rightarrow$ 

$$\begin{cases} \frac{dn_1}{dt} = \beta n_2 - \alpha I_{in} n_1 \\ I_{out} = \beta n_2 \end{cases}$$

• By considering stationary state  $\rightarrow dn_1/dt = dn_2/dt = 0 \rightarrow$ 

$$n_2 = \frac{\alpha I_{in} n_1}{\beta}$$

$$I_{out} = \alpha I_{in} n_1$$

#### Simple model for RPL (3)

• The important parameter linked to the dose is  $n_t = n_1 + n_2 \rightarrow n_1 + n_2 \rightarrow n_2 \rightarrow n_2 + n_2 \rightarrow n_2 \rightarrow n_2 + n_2 \rightarrow n_2$ 

$$n_{t} = n_{1} + \frac{\alpha I_{in} n_{1}}{\beta}$$

$$n_{1} = \frac{n_{t}}{1 + \alpha I_{in} / \beta}$$

$$I_{out} = \frac{\alpha I_{in}}{1 + \alpha I_{in} / \beta} n_{t}$$

- If the source is a powerful laser  $\rightarrow I_{in} > \rightarrow$  important denominator
- If the source is a LED  $\rightarrow I_{in} < \rightarrow I_{out} = \alpha I_{in} n_t$
- If signal present even though dose = 0 → I<sub>out</sub> = αI<sub>in</sub>(n<sub>t</sub> + n<sub>b</sub>) → effect of pre-dose (photoluminescence)

RPL dosimeter: Phosphate glass with silver impurities

- Phosphate glass  $\rightarrow$  type of glass in which SiO<sub>2</sub> is replaced by P<sub>2</sub>O<sub>5</sub>
- The P<sub>2</sub>O<sub>5</sub> mainly crystallize into P<sub>4</sub>O<sub>10</sub> → each P atom is surrounded by 4 O atoms as tetrahedron shape →



#### Phosphate glass: Silver impurities

- Several compositions exist but all of them contain Ag impurities (example: FD-7 → Na: 11.00%, P: 31.55%, 0: 51.16%, Al: 6.12%, Ag: 0.17%)
- The optimum Ag concentration is determined in a phenomenological way
- In the phosphate glass, Ag is stable as Ag<sup>+</sup> ion
- The e<sup>-</sup> and h<sup>+</sup> created by ionizing radiation in the glass are diffused in it and produce at the end the reactions →

$$\begin{cases} Ag^+ + e^- \to Ag^0 \\ Ag^+ + h^+ \to Ag^{2+} \end{cases}$$

#### Luminescence mechanism

• Ag<sup>0</sup> are Ag<sup>2+</sup> are the stable luminescence centers at ambient T



 When the glass is exposed to UV radiation (345 and 308 nm) → excitation of Ag<sup>0</sup> and Ag<sup>2+</sup> impurities and then relaxation of these impurities by visible light emission (460 nm: blue and 560 nm: yellow-orange)

#### Absorption and emission spectra

There is thus a combination between the 2 visible radiations emitted  $\rightarrow$  the emission spectrum is dependent on the absorption spectrum



### Build-up effect

 Particularly for the emission at 560 nm → a waiting time is needed after the irradiation by ionizing radiation to stabilize the RPL light intensity → « build-up » phenomenon



• It was suggested that, initially, the h<sup>+</sup> is trapped by a PO<sub>4</sub> tetrahedron  $\rightarrow$  only after a certain time: Ag<sup>+</sup> + hPO<sub>4</sub><sup>+</sup>  $\rightarrow$  Ag<sup>2+</sup> + PO<sub>4</sub>

## RPL dosimetry: In practice

- The dosimeters based on the radiophotoluminescence phenomenon are called RPLD
- For the FD-7 dosimeters:  $Z_{eff} = 12.0 \rightarrow$  not tissue-equivalent ( $Z_{eff} = 7.35$ )  $\rightarrow$  energy dependence for small energies (photoelectric effect)
- To correct this non-linearity → addition of filters (for instance: Sn, Al, Cu, PE,...)

#### Energy response



(a) Energy response for incident  $\gamma$  with  $\neq$  filters (b) corrected response of the dosimeter ( $R = \sum_i k_i R_i$ )

 $\rightarrow$  Linear response equal to 1 (with corrections)



• Linear response for small doses



- Linear response for high doses
- Linear response for 20  $\mu$ Gy < *D* < 10 Gy
## Fading - Reproducibility

- The fading data depend on the considered materials (and on measurement laboratories) but fading ≤ 1%/month is generally observed
- The measurements are reproduced several times after various waiting times (t < 10 days) and various doses (20  $\mu$ Gy < D < 10 Gy) give identical results (with variation of 1.5%)

TL dosimeters	OSL dosimeters	RPL dosimeters
High sensitivity of materials: LiF:Mg,Cu,P, LiF:Mg,Cu,Si, CaSO <sub>4</sub> :Dy	High sensitivity due to absence of thermal quenching and IR background: Al <sub>2</sub> O <sub>3</sub> :C and BeO	High sensitivity of materials such as silver-activated phosphate glass
Stable sensitivity possible	Stable sensitivity possible	Stable sensitivity possible
Easy handling (no light sensitivity using LiF:Mg,Cu,P, LiF:Mg,Ti.	Highly light sensitive, but can be managed with appropriate precautions during storage and use.	No light sensitivity for visible light (λ>366nm), as RPL emission is stimulated by UV light.
Dose re-estimation possible. Doses as low as 0.2 mGy can be re- estimated using PTTL in LiF:Mg, Cu,P*	Multiple dose re-estimations possible. Multiple measurements of OSL from Al <sub>2</sub> O <sub>3</sub> :C using either pulsed or continuous wave stimulation have been reported.**	Dosimeter can be read as many times as necessary without depleting the signal.
Flat photon energy response for LiF:Mg, Cu,P (100keV – 3MeV)	Flat photon energy response for BeO OSL dosimeters. (100keV – 3MeV)	Use of appropriate filters gives nearly flat photon energy response: 10 keV to 10 MeV.
Identification of static/dynamic exposure is somewhat difficult	Identification of static/dynamic exposure is possible.	Identification of static/dynamic exposure is possible.
Dose imaging difficult	Dose imaging possible.	Dose imaging possible.
Complex thermal annealing steps for some TL materials	Elimination of thermal annealing steps, as heating of samples is not required during OSL readout	The RPL dosimeter must undergo heat treatment at 70°C to stabilize the build- up of colour centers before readout.
Real-time measurements- not possible	Using Optical Fiber based OSL dosimetry system: real-time (on- line) measurements.	No published report on this aspect of use is available.
Readout time for commercial systems: acquisition time:13.3s per readout using hot air #	~1-2s per readout; 12-13s per 4-element dosimeter (using CW- OSL mode).	<1s per readout
Dose range: 1µGy to 100Gy	10µGy to 10 Gy	14µGy–10 Gy
Post irradiation fading: <2% in 6months	<5% per year	1% per month