



Sviluppo di dosimetri con materiali avanzati per radioterapia clinica

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M. Bruzzi, INFN Firenze CdS 2 Marzo 2020

Outline



- 1. Motivation
- 2. Advanced materials and devices in clinical radiotherapy dosimetry
- epitaxial silicon
- SiC
- polycrystalline diamond
- **perovskites PERO2**
- 4. Conclusions

Esperimenti finanziati nel corso degli anni da INFN CSN5 presso la sezione di Firenze per lo sviluppo di dosimetri per radioterapia clinica

• Diamante policristallino

IDDD (96-98) / CANDIDO (99-02) /

DIAPIX (11-13) / IRPT MIUR (14-17) / 3DOSE (17/19)

- Si ossigenato, SiC, diamante CONRAD (03-05)
- Si epitassiale

PRIMA (04-09)

perovskite inorganica

PERO/PERO2 (19-21)









Motivation

Radiotherapy with external beams with high energy (X, γ , electrons, protons, ions) produced by particle accelerators





•immobilization techniques allow for positioning patient during the treatment with always increasing accuracy,

•accelerators equipped with beam modifying devices able to get dose distributions closely shaped on the target volume.



Advanced, highly accurate imaging and dose verifications systems must match with increased accuracy of irradiation techniques

Intensity Modulated Radiation Therapy (IMRT)

To spare surrounding healthy tissues released dose shaped along an irregular field conformed to the tumor volume



Intensity Modulated Radiation Therapy (IMRT) : a few radiation beams, generally from 2 to 9, produced by the same linear accelerator and directed towards the tumor from different angles, in order to concentrate the dose released on the volume of the tumor.





Multi Leaf Collimators (MLC)

Dose conformation obtained using Multileaf Collimators with sets of mobile lamellas in W mounted externally on the LINAC head













Example: IMRT irradiation in step and shoot modality, the total dose is released as a sum of nine segments, each corresponding to a particular arrangement of the MLC lamellas (left). Beam is off during their movement.

Total dose is obtained as the sum of the dose released by each segment (right).







VMAT (Volumetric Modulated Arc Therapy)

□ Able to focus more accurately at tumor tissues, ensuring greater preservation of healthy ones.



- Modulating not only the amplitude and velocity of the MLC, but also rotation speed of the Gantry and Linac dose-rate.
- continuous rotation of the accelerator head during irradiation for maximum focusing of radiation on tumor tissues, which are thus affected by all possible angles.
- □ significantly reducing duration of treatments compared to IMRT: about 5-7 minutes compared to traditional times which are around 20 minutes per session.
- □ useful when treatment focus must be maximum to preserve nearby organs: tumors of the head / neck, as larynx, pharynx and oral cavity; tumors of the pelvis, as prostate and rectum; tumors of the lung and breast.

Working principle of a on-line dosimeter



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[nC/Gymm³]

Dosimetry Challenges

Highly accurate imaging and dose verifications systems must match with increased accuracy of the irradiation techniques .



- **Response independent of energy** often Charged Particle Equilibrium (CPE) lacks (a phenomenon associated with the range of secondary particles and hence dependent on the beam energy, composition and density of the medium);
- small volume and high sensitivity to get enough spatial resolution;
- **response independent of dose rate,** continuously changing during VMAT;
- **Real time invivo detectors** european community require dose delivery to be verified experimentally **directly during irradiation** (Article 56 of COUNCIL-DIRECTIVE-2013/59/EURATOM).

State-of-art commercial dosimetric devices used in clini radiotherapy

	radiotherapy			INFN	
	IC (AIR)	SILICON	DIAMOND		
ρ [g/cm ³]	1.29x10 ⁻³	2.33	3.52		
E _i [eV]	34.00	3.60	16.20	-	
S [nC/Gymm ³]	0.038	647.22	217.28	-	
Area [mm ²]	25.00	0.64	3.80	-	
thickness [mm]	5.00	0.03	0.001		
volume [mm ³]	125	0.019	0.0038		
Array	OCTAVIUS	MAPCHECK	-	-	
	РТW	SunNuclear			
Detector	729 PTW	SunPoint®	microDiamond	-	
		Diode Detector	type 60019 PTW		
Reference	APL Mater. 7,	051101 (2019); doi:	10.1063/1.5083810		



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OCTAVIUS 729	Specifications

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OCTAVIUS Detector 729

Detector type:	Plane-parallel vented ionization chambers		
Detector design:	cubic		
Number of detectors:	729		
Detector size:	0.5 cm x 0.5 cm x 0.5 cm (0.125 cm³)		
Detector spacing:	10 mm center-to-center, 5 mm edge-to-edge		
Max. field size:	27 cm x 27 cm		
Reproducibility:	≤ ± 0.5%		
Dead time:	zero		
Repetition rate:	200 ms		
Measured quantities:	absorbed dose to water (Gy), absorbed dose rate to water (Gy/mir		
Resolution:	0.1 mGy or 0.1 mGy/min		
Measurement range:	(0.5 48 Gy/min)		
Reference point:	7.5 mm below the surface of the array		
Housing material:	GRP		
Dimensions:	30 cm x 42 cm x 2.2 cm (W x D x H)		
Weight:	5.7 kg		
Power supply:	(100 240) VAC; (50 60) Hz		
PC connection:	Ethernet, RS232		
Part No.:	L981378		



OCTAVIUS $^{\textcircled{B}}$ 729 - 2D ionization chamber array for patient and machine QA with OCTAVIUS 2D/4D



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Helical Detector Grid

Detectors are arranged on a HeliGrid[™] which increases the sampling rate and reduces BEV detector overlap and shadowing.

An ArcCHECK 10 x 10 cm² area contains 221 detectors; equivalent to the detector density in a MapCHECK® 2 Entrance and exit dose are measured, effectively doubling the detector density in the measurement field

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Beam Delivery



ArcCHECK Detector



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The Silicon Choice

Advantages:

- High sensitivity (18000 times higher than IC).

- Well developed manufacture technology.
- high spatial resolution.
- work in null bias mode (in-vivo).

Drawbacks:

- Sensitivity decrease with accumulated dose due to increase of concentration of recombination centers (recalibrations needed).

- Dose rate dependency due to centers saturation at high dose rates.

- Energy dependence: Si not "water equivalent".

Radiation Resistant ?

p-type radiation harder material









The neutral region contributes (diffusion of minority carriers into the depletion region) to the signal

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Radiation damage in standard Si dosimeters (thickness 300um)

Sensitivity as a function of trap concentration in a Si dosimeter





M. Bruzzi, NIMA 809, 2016 Novel Silicon Devices for Radiation Therapy Monitoring, 105-112

Pre-irradiation (\approx 10kGy) to reduce the dependence of the signal on the dose.

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Material engineering concepts have been applied also to Silicon dosimeters for radiotherapy

Improved radiation hardness of DOFZ Si



Decrease in sensitivity with the accumulated dose due to the generation of a dominant trap acting as lifetime killer.

$$1/\tau - 1/\tau_0 = \sigma v_{th} N_t$$
, $N_t = a \phi$; $a = trap generation rate$

 σ capture cross section ; v_{th} carrier thermal velocity. N_t trap concentration. $a_{DOFZ} < a_{SFZ} \implies$ increased radiation hardness of the device to radiotherapic beams.

$$a_{DOFZ} = 5.0 \times 10^7 \text{ cm}^{-3} \text{Gy}^{-1}, a_{STFZ} = 8.1 \times 10^7 \text{ cm}^{-3} \text{Gy}^{-1}$$

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Our recipe: epitaxial p-type Si on MCz substrates to limit active thickness

Active region is limited in any direction to a value shorter than L_n at the highest dose of interest. Epitaxial Layer is used to limit active depth, guard-ring to limit active area.





Si bidimensional dosimeter

Matrix: 21x21pixels

Pixel: 2x2mm Pitch: 3mm Detector size: 6.3x6.3cm²

Covered area 20×20 cm² ~4k channels

Measured time structure of dose segments



IMRT Large area covered mosaic by composition and/or modules shifting along x-y axes.

Dose map of an IMRT field for prostate cancer as measured by the Epi-Si 2D silicon dosimeter.

United States Patent Bruzzi et al.

M. Bruzzi, INFN Firenz (45) Date of Patent:



C.Talamonti, M.Bruzzi et al. 2011 Nucl. Instr. Meth A, vol. 658, p. 84-89.





Oct. 22, 2013

19

The SiC choice

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Low Leakage Current Working without applying bias Very low active volume Fast Response More tissue equivalent than Si Radiation resistance ? Schottky Contact



Ohmic Contact

Z
7.78
7.51
7.64
6.46
12.31
6
14
~10

High bandgap semiconductors: generation always negligible \rightarrow intrinsically radiation hard

Si

SiC

Diamond

10¹⁶



Dosimetric Characterisation

Stable signal - high S/N ratio - no priming effects



M. Bruzzi, F. Nava, S. Pini, S. Russo, App. Surf. Sci, 184 (2001) 425-430

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Comparison between Epitaxial SiC and standard dosimeters

Device	bias [V]	Vol. [mm ³]	S [nC/Gy]	S per unit volume
Standard Farmer Ionisation chamber	300	600	21.5	[nC/(Gy·mm ³)] 0.036
Miniature Farmer Ionisation chamber	300	50	1.38	0.028
Scanditronix GR-p BS Silicon	0	0.295	140	474
Scanditronix SFD stereotactic Silicon	0	0.017	6	353
Epitaxial SiC diode	0	0.0415	14.1	340

Problem: SiC is not defect tolerant



M. Bruzzi, INFN Firenze CdS 2 Marzo 2028 ruzzi et al. NIMA 579 (2007) 754-761

Microscopic radiation damage si responsible of the reduced charge collection of SiC with respect to Si and Diamond

- n epilayer 7µm
- ♦ N_{eff} ~7×10¹⁵cm⁻³
- Schottky Barriers Ti or Ni
- Ohmic contacts Ti/Ni/Ag
- Deep levels by C-DLTS
- ♦ 6.5MeV p up to 6.4x10¹³cm⁻²

Six traps detected after irradiation

◆E = 0.18 - 1.22eV

• $\sigma = 10^{-13} \cdot 10^{-18} \text{ cm}^{-2}$

• $N_t = 10^{11} - 5x10^{14} \text{ cm}^{-3}$



Data from A. Castaldini, A. Cavallini et al.

The Diamond Choice





- it is almost water equivalent
 - it doesn't perturb the radiation field \rightarrow small fields
 - the energy is absorbed as in the water \rightarrow no correction factors
- high radiation hardness → long term stability
- In high density \rightarrow high sensitivity \rightarrow small dimensions
- non toxic
- it can be used as TL dosimeter (off-line) or for on-line applications
- high defect density priming effects instability of the signal
 - high voltage required
 - high production costs

Specifications Learn more

60019

axis

Nominal sensitive volume: 0.004 mm³, radius 1.1 mm, thickness 1 µm

1 nC/Gy

0 V

waterproof, disk-shaped, sensitive

on detector axis, 1 mm from detector

volume perpendicular to detector

absorbed dose to water

100 keV ... 25 MV photons

(6 ... 25) MeV electrons

(70 ... 230) MeV protons

BNT, TNC or M

(1 x 1) cm² ... (40 x 40) cm²

tip, marked by ring

Nearly as good as water.

microDiamond

Key Features

Type No.

Design:

Measuring quantity:

Reference point:

Detector bias:

Field size:

Connectors:

Nominal response:

Radiation quality:

- Nearly water equivalent for all beam energies
- Very small sensitive volume (0.004 mm³) perfect choice for small field dosimetry
- Suitable for all field sizes up to 40 cm x 40 cm
- Precise, accurate measurements in photon, electron and proton fields
- Excellent radiation hardness, minimal energy, temperature and directional dependence
- No high voltage required. Suitable for all connecting systems (BNT, TNC, M)

microDiamond Synthetic Diamond Detector







microDiamond Detector Design

Single Crystal Diamond Dosimeter

device manufactured by Università di Roma Tor Vergata



•HPHT p+ boron doped substrate - 300 µm thick

- •35 µm thick p+ buffer layer (CVD)
- •Active epilayer 17 µm thick

•Al 100 nm ϕ 2mm upper electrode

•Buffer layer contacted at the perifery with Ag paste

Problem : No large area array available

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The Polycrystalline diamond Choice

it is almost water equivalent



it doesn't perturb the radiation field → small fields
 the energy is absorbed as in the water → no correction factors
 high radiation hardness → long term stability

- high density \rightarrow high sensitivity \rightarrow small dimensions
- non toxic

Natural diamond

very high production costs, difficult to select stones with proper dosimetric response

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Neutron irradiation in pCVD diamond beneficial as removes dominant defect and increase recombination centres, bringing to faster dynamics

Measurements of pCVD diamond photoresponse under different cycles of X-ray irradiations - deactivation of the peak at 1eV probably responsible of the current instability.



M. Bruzzi et al., Appl. Phys. Lett, (2002)

Fig. 1. TSC signal vs. temperature for non-irradiated and irradiated samples, after filling with UV xenon lamp for 20 min. The samples are biased with 100 V. The heating rate is 0.15 K/s. The neutron fluences are indicated in the legend.

450

Temperature [K]

500

550

600

400

Problem in polycrystalline diamond: priming and instability effects due to defects at grain boundaries



High voltage application



Current response of the unirradiated sample (Au contact) showing the priming effect during the first 6 successive irradiations.

Our Solution to get STABLE SIGNAL

- back to back schottky barriers
- low/null bias



Diamond & Related Materials 20 (2011) 84-92

Zero-bias operation of polycrystalline chemically vapour deposited diamond films for Intensity Modulated Radiation Therapy

M. Bruzzi ^{a,*}, C. De Angelis ^b, M. Scaringella ^a, C. Talamonti ^c, D. Viscomi ^b, M. Bucciolini ^c

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Priming and instability effects due to trapping and polarization in pCVD are dominant at high voltage with ohmic contacts and high electric field applied.



C. De Angelis et al. / Nuclear Instruments and Methods in Physics Research A 583 (2007) 195–203

Polarization is negligible at low/zero bias operation when using Schottky contacts.



Fig.5 Time structure of the IMRT segments as measured by one of the pCVD diamond pixels under an IMRT prostate cancer treatment.

M. Scaringella et al. / Nuclear Instruments and Methods in Physics Research, A, 796, 2015, 89-92

M. Bruzzi et al. / Diamond & Related Materials 20 (2011) 84-92

polycrystalline diamond segmented dosimeter prototype made in Florence

- Material

- Up to three polycrystalline diamond films 2.5x2.5cm² active area each, 300µm thick;
- Premium Detector Grade Element Six, UK

- Contacts

- Schottky Barriers produced @ University of Florence
- •12 x 12 matrix, pixel size: 1.8x1.8 mm² \rightarrow 288 pixels in total

- Read Out Electronics

four 64 channels 20 bit current-input analog to digital converter chips able of measuring currents from fAs to mAs; 160μs-1s integration time (50ms)
custom printed circuit board;

•semi-rigid silver-polymer pin-contacts produced by us connecting each pixel of the 144 matrix connecting vias on PCB.

-Measurement

- •Low voltage to get fast and reproducible signals;
- •Device can be moved in x-y directions to cover a
- •wider radiation field area. M. Bruzzi, INFN Firenze CdS 2 Marzo 2020





pCVD Diamond test under linac

RENZE



Performance under conventional and IMRT radiotherapy beam

Current response of all pixels in a conventional X-ray beam ($V_{app} = 1V$) Dose-rate 50 Mu/min



Current response of one pixel under an IMRT beam in step and shoot modality

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Bartoli et al. 2017 JINST 12 C03052
✓ negligible dark current → high S/N
✓ negligible polarization effects → stable response , fast dynamics

Fig. 5. Time structure of the IMRT segments as measured by one of the pCVD diamond pixels under an IMRT prostate cancer treatment.

M. Scaringella et al.

Nuclear Instruments and Methods in Physics Research A 796 (2015) 89-92



First IMRT map with Diamond Device





(GT = gantry target direction; LL = lateral-lateral direction) Grid spacing 3 mm.

M. Scaringella et al. / Nuclear Instruments and Methods in Physics Research A 796 (2015) 89-92

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The Perovskite Choice

Perovskites are emerging materials for large-area and flexible optoelectronic device systems

 AMX_3 (X = oxide, halide anion such as Cl, Br and I, M = metal cation).

Organo-metallic halide perovskites recently proposed for printable large area flexible X-ray detectors in medical imaging applications (<u>Nature</u> 2017 550(7674): 87-91; Physics in Medicine, Vol. 5, 2018 20-23.).

All-inorganic perovskites, CsPbX₃ (X=Cl, Br, and I) attracting alternatives due to their improved chemical stability, they can be produced in form of single crystals, nano-crystals and micro-crystalline thin films.



A 15% perovskite solar cell made in University of Oxford



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The CsPbBr₃ Choice

- More sensitive than silicon •
- **Easy deposition methods**
- **Potentially defect tolerant** •

Sample preparation and characterization in Florence

	SILICON	CsPbBr3	
Eg [eV]	1.12	2.30	
ρ [g/cm³]	2.33	4.55	
E _i [eV]	3.60	5.30	
S [nC/Gymm³]	647.22	860	

and formation of perovskite



of CsBr and PbBr, in DMSO (1:1)

New: single crystals cm size PMMA cover to protect against moisture

- Possibility to deposit directly on any kind of substrates, even flexible
- Spin coating, magnetron sputtering, dropcast polycrystalline and single crystals



Defect Tolerance of CsPbBr₃

Conventional semiconductors as GaAs, CdSe, InP, are defect intolerant as they are prone to mid-gap states very active as trap states.





Kovalenko et al., Science 358, 745-750 (2017)



J. Kang, L.-W. Wang, J. Phys. Chem. Lett. 2017, 8, 489–493.

CsPbBr₃ potentially defect tolerant: mostly shallow and intermediate defect states.

Shallow levels: The hydrogen-like model

Donor (e.g. P in Si) is seen as a complex Si + H, an hydrogen atom is added at a site of the silicon lattice.



$$E_{Dn} = -\frac{e^4 m^*}{2(\varepsilon_0 \varepsilon_r)^2 \hbar^2} \frac{1}{n^2} \approx 10-60 \text{meV}$$

→ fully ionized at moderate temperatures → Doping

$$a*_{n} = \frac{\varepsilon_{0}\varepsilon_{r}\hbar^{2}}{m*e^{2}}n^{2}$$

$$a_o = 0.529$$
 Å Bohr radius, $a_{Si} \sim 30$ Å ; $a_{Ge} \sim 80$ Å

→ delocalized wave functions



A level e.g. that in silicon is a few tens of meV reduces to few meV in CsPbBr₃ ($m^* = 0.13m_0$ and $\epsilon_r = 29.37$)





→Active thickness reduced due to defects



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Open Issue \rightarrow higher rise/decay times due to defects

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Lateral-lateral direction [mm]



Slower rise/decay times when radiation switches. This effect should be minimized to increase dose measurement RACEUMER OVEN Firenze CdS 2 Marzo 2020



- Peak due to charge emitted from trap only during heating;
- Background due to thermal generation rate, same during heating and cooling.

$$I_{TSC}(T) = e \mu_p F A \tau_{eff} p_{t0} e_p(T) e^{-\frac{1}{\beta} \int_{T_i}^{T} e_p(T) dT}$$

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New results: Main defect active at RT 0.45eV. Present only in polycrystalline samples



Mara Bruzzi, Naomi Falsini, Nicola Calisi, Anna Vinattieri, **Electrically active defects in polycrystalline and** single crystal metal halide perovskite, submitted to Energies, feb. 2020, section: Advanced Energy Materials, issue: Progress in Inorganic Halide Perovskites: Information Perovskites: 2020

Open issues

- Reproducibility;
- Radiation Hardness;



- Single crystal performance under X-ray beam
- Schottky barrier devices to increase S/N



Conclusions

- Advanced imaging and dosimetry systems needed to get increased accuracy in radiotherapy treatment plans;
- State-of-art segmented detectors made with silicon (epitaxial, Italian patent)
- Diamond potentially best suited for X-ray radiotherapy due to energy independence of its response (tissue equivalence); segmented dosimeters not yet available commercially; Florence prototype shows excellent performances under IMRT and VMAT treatment plans;
- Volumetric dosimetry : next generation of high sensitivity flexible geometry systems based on perovskite materials under development,
 - open issues: optimizing deposition techniques and device engineering, influence of defects on dosimetric properties, radiation hardness.