



CaLIPSO: Photon detector for brain PET imaging

Measurement of ionization parameters of the medium of detection

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15TH VIENNA CONFERENCE

ON INSTRUMENTATION



Context: Functional cerebral imaging

	MRI	PET	
Imaging	Structural and functional	Functional	
Spatial resolution	1 mm ³	(3 mm) ³	
Biochemical processes	differences in blood flow	uptake of the tracer (200 tracers)	
Sensitivity	10 ⁻⁴ mol	10 ⁻¹² mol	
Irradiation	no	yes	

<u>Motivations</u>: Technological **breakthrough** for PET detector Quantify the activity of brain cells down to **1 mm³**



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CaLIPSO: Detection medium

Liquid Trimethylbismuth (TMBi)

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High-density liquid (2.3 g/cm<sup>3</sup>)
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- $Z_{Bi} = 83$: heaviest non radioactive element
- Photoelectric conversion efficiency = 49 %

Transparency : $\lambda > 400$ nm Refraction index ~ 1.6

Double detection : ionization and light signals

Patent, Ref: FR 1 052 047, WO2011/117158 A1, 2010.

D. Yvon et al. IEEE Trans Nucl Sci, 61(60), 2014.

CH₃

E. Ramos et al. J Instrum, 11(11):P11008, 2016.

CaLIPSO: Double detection



CaLIPSO: Computed performances

Ionization properties of TMBi have never been studied.

Benchmark liquid : tetramethylsilane (TMSi)



Time resolution

 \sim 150 ps (FWHM)



3D positioning ~ 1 mm³ if $\mu = 90$ cm².V⁻¹.s⁻¹ (TMSi)



Energy resolution $\sim 10 \%$ (FWHM) if $G_{fi}^{0} = 0.6$ (TMSi)

• We need to measure the properties of TMBi

Preliminary work: Liquid purification

Remove electron-attaching molecules by adsorption:

- periodic micro-pores: molecular sieve
- polar molecules adsorbed first





Purification system

Free ion yields of TMSi and TMBi

Free ion yield

We need to measure the properties of TMBi \rightarrow number of charges produced by a photon: free ion yield G_{fi}



with :

I(E) current induced by a radioactive source (A)

- E applied electric field (V/m)
- *e* elementary charge (C)
- $\Delta \epsilon$ energy deposited in the liquid per second (eV/s)

Ionization current measurement



Ionization chamber assembly

Parallel-plate ionization chamber

Active volume of liquid : 1,6 cm³ Gap between electrodes : 12 mm

Irradiation by a ⁶⁰Co source

Charges drifted by an electric field Induced a weak current (< 200 fA for 0.7 MBq)

\rightarrow Low-noise measuring device



Ionization chamber



Low-noise current measurement



<u>Pico-ammeter</u> : Keithley 6517 B <u>Power supply module</u> : Caen N470 * equivalent electrical resistance due to ceramics composing the chamber

Low-noise current measurement



- signal output

liquid volume

- chosen materials
- clean assembly
- control moisture levels
- power inverter
- screened isolation transformer
- local ground

Accuracy \sim 5 fA under high voltage (< 8 kV)

Measured ionization current



M. Farradèche et al. J Instrum, 13(11):P11004, 2018.

Free ion yield



with :

- I(E) current induced by a radioactive source (A)
- E applied electric field (V/m)
- *e* elementary charge (C)
- $\Delta \epsilon$ energy deposited in the liquid per second (eV/s)

Energy deposited in the liquids

GATE/GEANT 4 Monte Carlo simulation

- simulation of the liquid volume and environment
- reconstruction of energy deposition



S. Jan et al. Phys Med Biol, 49(19):4543, 2004.

Spectra of energy deposited in the liquids



Free ion yield



with :

- I(E) current induced by a radioactive source (A)
- E applied electric field (V/m)
- *e* elementary charge (C)
- $\Delta \epsilon$ deposited energy in the liquid per second (eV/s)

Measured free ion yield



Onsager theory

Escape probability of an electron (from *initial* recombination) Brownian movement of an electron under the influence of:

- \rightarrow the Coulomb attraction of its parent ion only
- \rightarrow an additional electric field

$$G_{fi}(E) = G_{fi}^{0} (1 + \alpha E)$$

with:

1 predicted parameter: α = $e^{_3}$ / $8\pi\;\epsilon_{_0}\epsilon_{_r}\;(kT)^{_2}$

1 free parameter: G_{fi}^{0}



Assumption: every pair is independent of the other

- mean distance between ionization pairs ~ 3000 Å (hydrocarbon liquid)
- mean thermalization length ~ 166 Å (TMSi)
- L. Onsager. Phys Rev, 54(8):554, 1938.
- J. Engler. J Phys G Nucl Part, 22(1): 1, 1996.

Measured free ion yield



Measured free ion yield

Same shape of the curve than TMSi

α parameter non predicted by
Onsager theory

 Lower G_{fi}⁰ than other organometallic liquids (TMSi, TMGe, TMSn...)



Quantum chemistry calculation

Model and calculations by J-P. Dognon (CEA Saclay, DRF/NIMBE)

Fukui functions

local changes in the electronic density in case of capture (f^+) or loss (f^-) of an e⁻

- f^+ : reflects the ability of a molecule to accept an e^-
- f^- : reflects the ability of a molecule to donate an e^-

dual $f = f^{+} - f^{-}$

dual f : describe molecules which tend to accept (> 0) or loose (< 0) an e^{-}

	TMSi	ТМВі	TMPb
dual f	- 0.038	+ 0.017	- 0.030
	and the second s		

Deviation from Onsager theory

Suggested interpretation



Phenomenon analogous to initial recombination Not taken into account in Onsager theory

Charge pulses

Quantification of electron lifetime and mobility

Preliminary work: Low-noise charge amplifiers

Institut für Kernphysik, Münster (Ge)

noise = 79 e⁻ RMS with post-amplifier gain 50 shaping time 3 μ s

rise time = 218 nswithout post-amplifier IRFU, CEA Saclay (Fr)

noise = 162 e⁻ RMS with post-amplifier gain 20 shaping time 1 μ s

rise time = 52 nswithout post-amplifier

Impulse response (amplifier alone)





Charge pulses from γ source - Work in progress



- Low-noise charge amplifier (Münster)
- Improve signal/noise : amplifier shaper

By comparing measured and simulated Compton edges: attenuation length $\lambda \sim 4.7 \pm 0.4$ mm electron lifetime $\tau = \lambda / \mu E \sim 1.05 \pm 0.08 \mu s$

Amplitude spectra (in number of electrons) in TMSi



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Charge pulses from muon - Work in progress

Study of **signal induction** (rise time):

- without amplifier shaper
- fast charge amplifier needed (Saclay)

Current induced by a muon:

$$I(t) = \frac{Ne}{t_d} \left(1 - \frac{t}{t_d}\right) e^{-t/\tau}$$

with:

$$t_d = \frac{d}{\mu E}$$

2 free parameters to measure:

- electron lifetime τ
- electron mobility μ



Charge pulses from muon - Work in progress



To smooth the signals:

- sum of \sim 1000 impulse responses of the system
- sum of ~ 400 measured signals

Convolution of impulse response with theoretical current induced by a muon

electron lifetime $\tau \sim 1.1 \pm 0.2 \ \mu s$ (for $\mu = 90 \ cm^2/V/s$)

Design of a purification system based on molecular sieve for chemically reactive liquids

Development of a low-noise measuring device of radiation-induced currents

Measurement of the free ion yields of TMSi and TMBi

the G_{fi}⁰ of TMBi is 7 times lower than other organometallic liquids

Quantum chemistry computations to understand this discrepancy

ability of TMBi molecule to capture electrons

additional trapping mechanism for ionization electrons near their parent cations

Work in progress - To test these assumptions quantitatively: Measurements of single interaction charge pulses

BOLD-PET: three-year project in collaboration with University of Münster for prototyping high resolution (time and spatial) PET detector

Back-up slides

When an electron is released : thermal equilibrium by inelastic collisions

If Coulomb attraction < thermal energy kT of the medium:

- \rightarrow Diffusion of the electron in the medium
- \rightarrow Probability of **volume** recombination with another ion
- \rightarrow Dominant phenomenon in gases (low density)

In a liquid, thermal equilibrium occurs in the Coulomb field of its parent ion: \rightarrow Probability of **initial** recombination predominant

Ionization chamber : e^- – ion pairs separated by an electric field

In a gas

For a high enough electric field \rightarrow every pairs are separated: *saturation current*

In an organometallic liquid Initial recombination always present \rightarrow no saturation: *linear behaviour* Current induced by a muon as a function of:



Red solid line

Onsager model

$$G_{fi}(E) = G_{fi}^0(1 + \alpha E)$$

Blue dashed line

General collection efficiency model

$$G_{fi}(E) = G_{fi}^{0}(1 + \alpha E) \left[1 + \frac{C(1 + \alpha E)}{E^{2}}\right]^{-1}$$

J. Pardo-Montero, F. Gómez. Phys Med Biol, 54(12):3677, 2009.

3

4

2

1

0

0.7

0.6

0.5

0.4

0.3

0.2

0.1

0

0

Gfi, electrons/100 eV

voltage, kV

TMSi

TMBi

6

7

5

eletric field, kV/cm

5

3

8

Analysis of charge pulses from muon



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IDeF-X ASICs

In the final pixelated detection device: IDeF-X ASICs as charge amplifier

Considering: • detector capacitance ~ 0.1 pF

dark current < 1 pA

\rightarrow Equivalent noise charge \sim 35 electrons RMS

Signal produced by a 500-keV photon in TMBi \sim 825 electrons (7 kV/cm)

 \rightarrow Signal-to-noise ratio > 20

From: O. Gevin et al. Nucl Instrum Meth A, 695:415, 2012.



Fig. 7. ENC as a function of peak time, measured with gain 200 mV/fC, measured at different values of external capacitance C_{ADD} .

Energy absorbed in the liquids



<u>TMBi</u>

PET detectors

	scintillation detector (LSO)	Semi conductor (CdTe)	<i>Potential</i> CaLIPSO (TMBi)
Photoelectrique attenuation coeffcient at 511 keV	0.26 cm ⁻¹	0.09 cm ⁻¹	0.16 cm ⁻¹
Energy resolution at 511 keV	~ 15 %	~ 2 %	$\sim 10 \%$ if $G_{fi}^{\ o} = 0.6$
Time resolution	~ <mark>300 ps</mark> (jusqu'à 100 ps)	$\sim 10~000$ ps	\sim 100 ps
Interaction positionning	—	3D	3D