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Effect of cumulated dose on gas emission and oxygen consumption from polyethylene irradiated under oxidative atmosphere using Swift Heavy Ions

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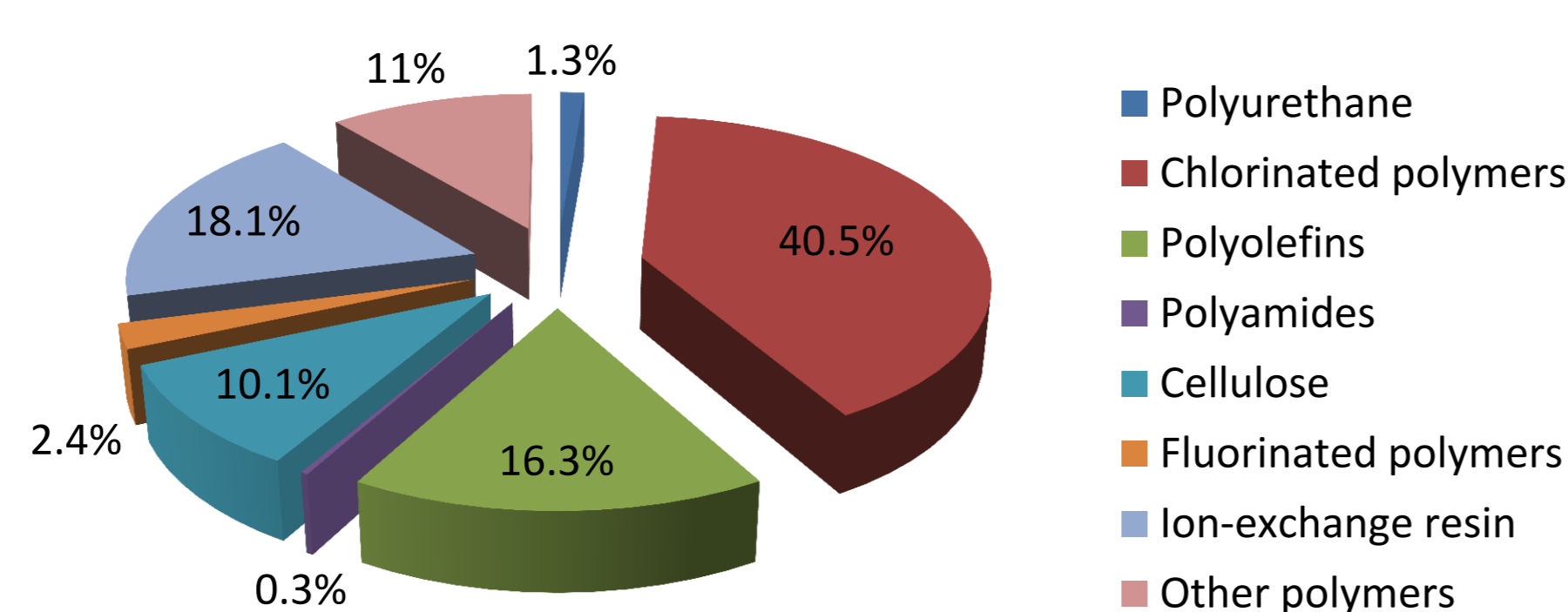
Context

- Intermediate Level Long Lived Waste (IL-LLW) : various organic materials, including polymers, in presence of radionuclides
 - Production of potentially explosive (hydrogen, methane...) or corrosive (hydrogen chloride, fluoride...) gases
- In the French nuclear safety context: development of a 500-meter deep disposal concept for IL-LLW
 - Quantification of the gaseous compounds release, as a function of the polymer
 - Doses higher than tens or one hundred of MGy => chemical composition of organic materials highly modified

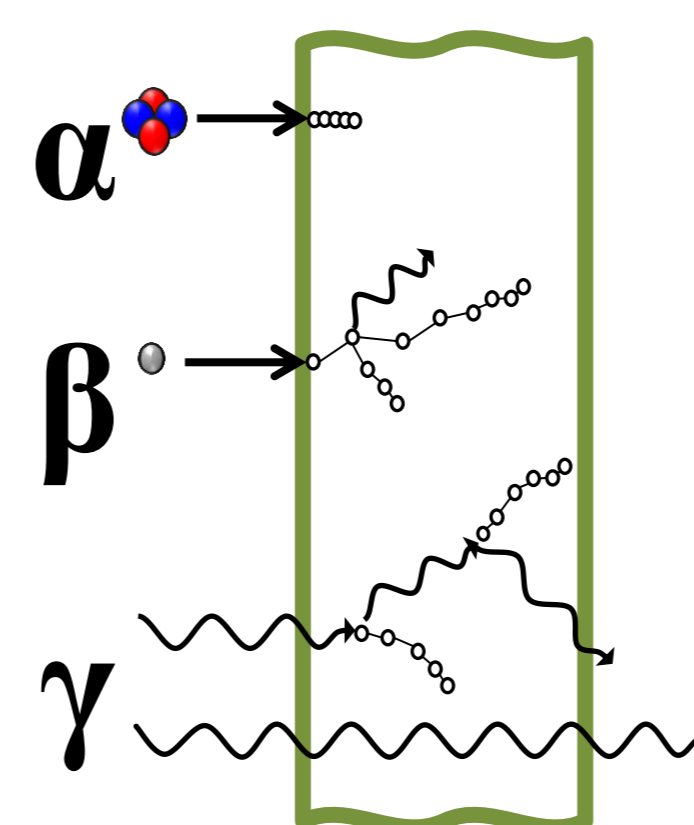
Aim

- Study of the influence of the irradiation dose on gas emission and oxygen consumption
 - Doses consistent with IL-LLW [1]

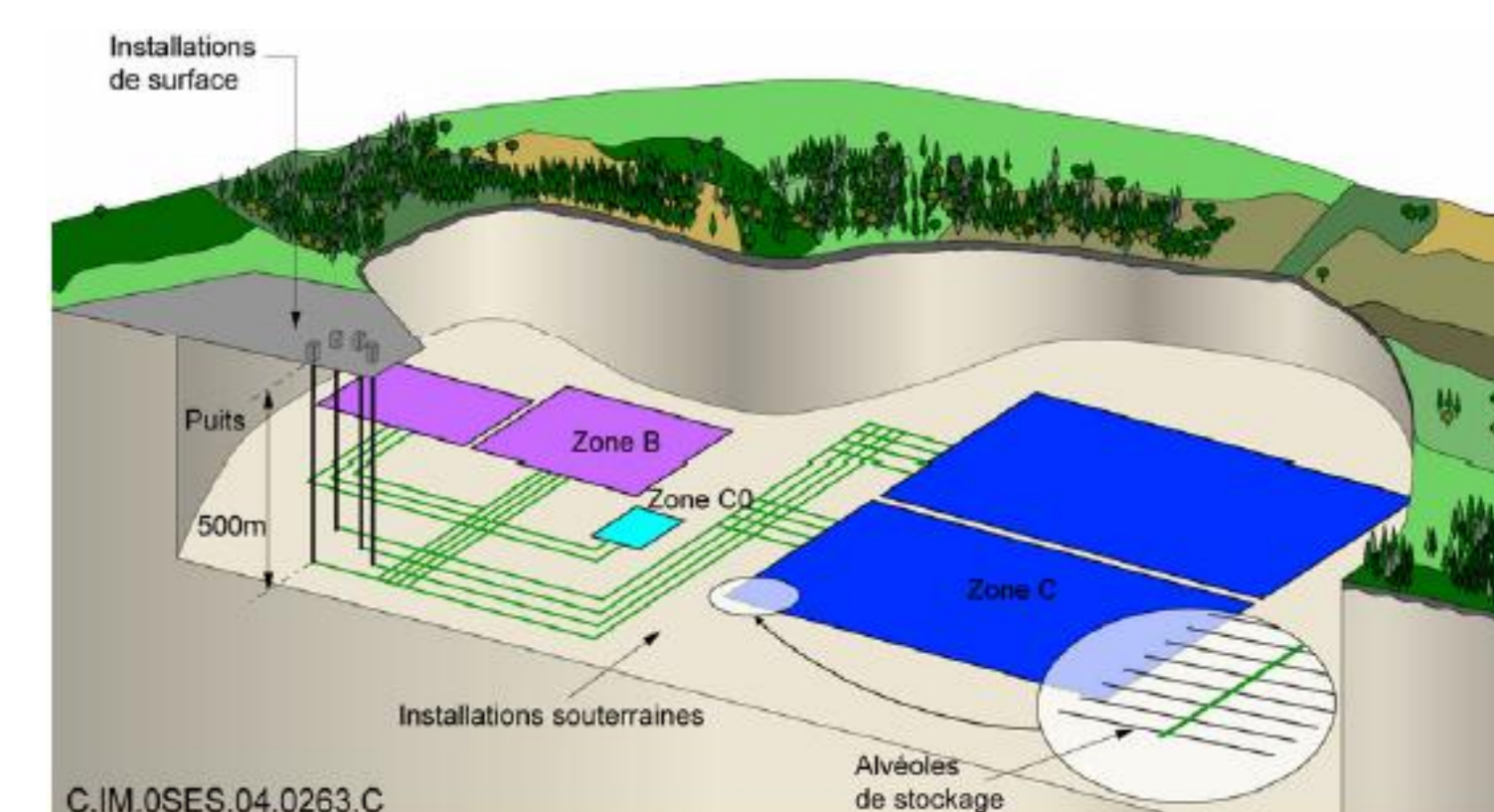
Polymers used in the French electronuclear industry



Ionizing radiations encountered in IL-LLW



Projected deep underground geological repository



IL-LLW tank

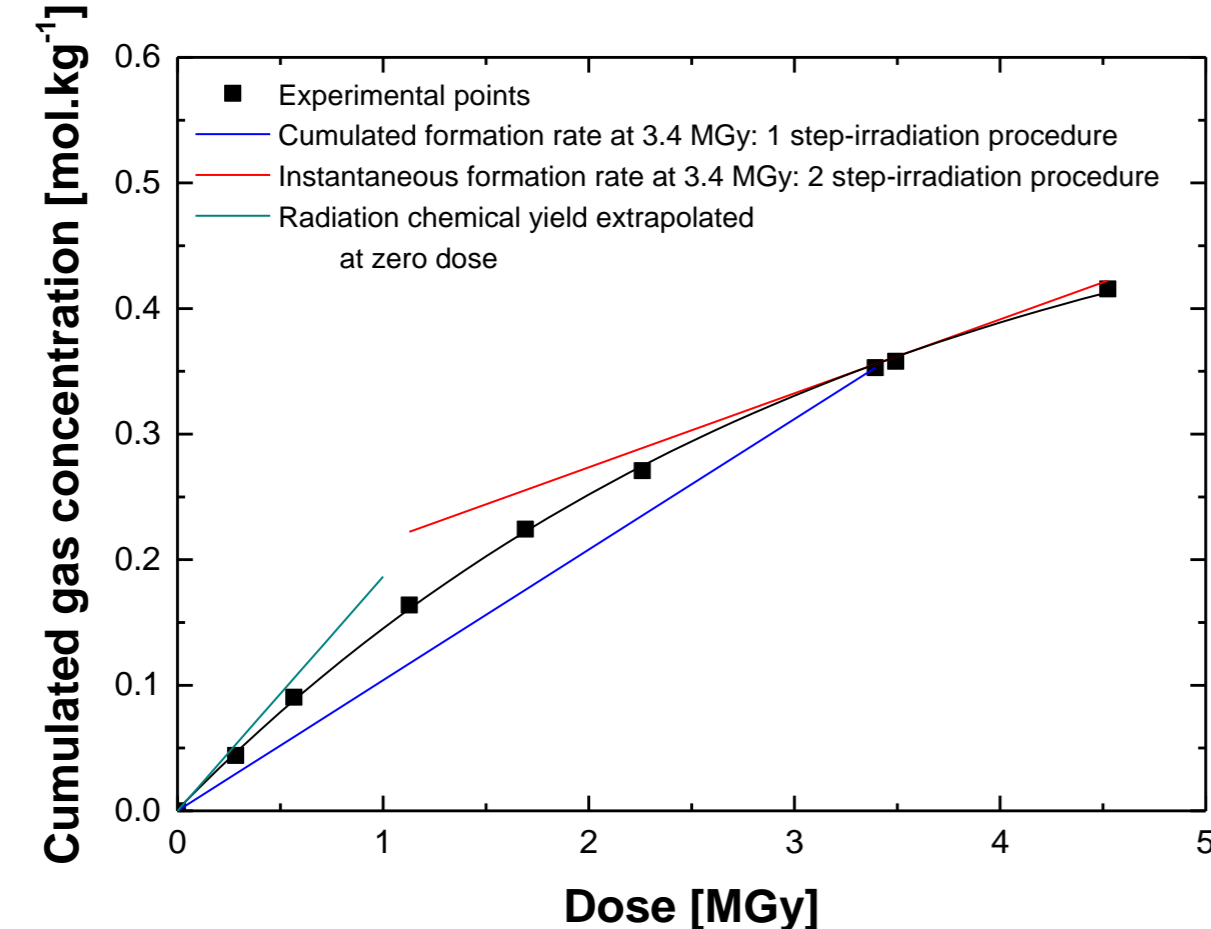


Material

- HDPE (High Density Polyethylene)
- Thin films prepared by high pressure molding
 - To ensure homogeneous oxidation profile in the sample thickness

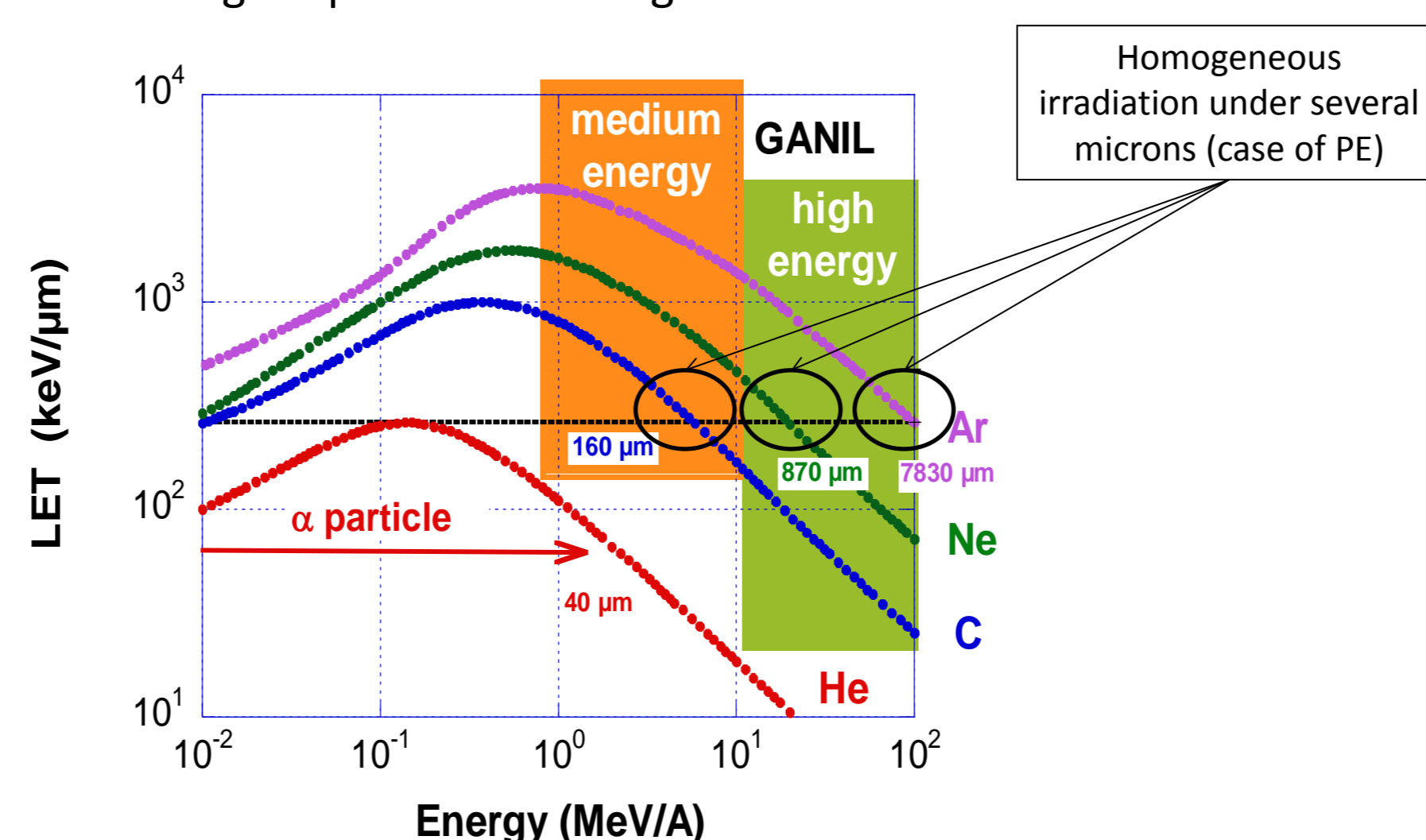
Irradiation conditions : a 2 step-irradiation procedure [2]

- Pre-aging: irradiation at high and various doses, under oxygen, without gas analysis
- Gas emission and oxygen consumption quantification: irradiation at low dose in closed container, along with gas analysis
- Storage conditions: pre-aged polymers stored under inert atmosphere and in the dark
- Result analysis

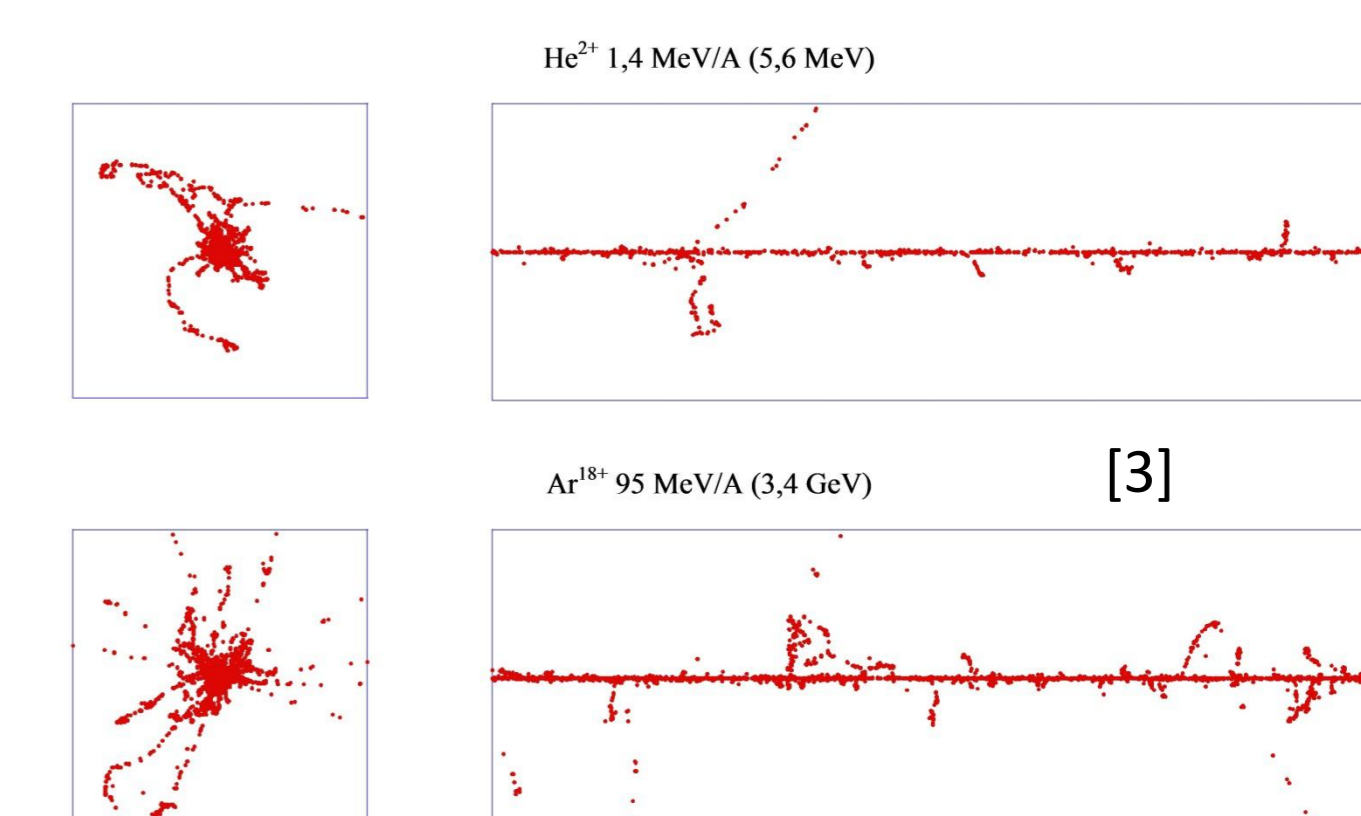


Ion beams to simulate α irradiations

- Why SHI (Swift Heavy Ions) instead of α ?
 - LET (Linear Energy Transfer) equivalent to radionuclides-emitted α
 - Higher penetration range



- Pre-aging: ¹³C (10 MeV/A) beams
 - Medium Energy beam line of GANIL (3.6·10⁸ ions.cm⁻².s⁻¹ up to 1.2·10¹³ ions.cm⁻²)
- Gas emission and oxygen consumption quantification: ³⁶Ar (95 MeV/A) beam
 - High Energy beam line of GANIL (3.6·10⁸ ions.cm⁻².s⁻¹ up to 2.6·10¹² ions.cm⁻²)
 - => Necessity to go through glass walls of the closed containers
 - => Necessity to ensure constant LET through samples thickness

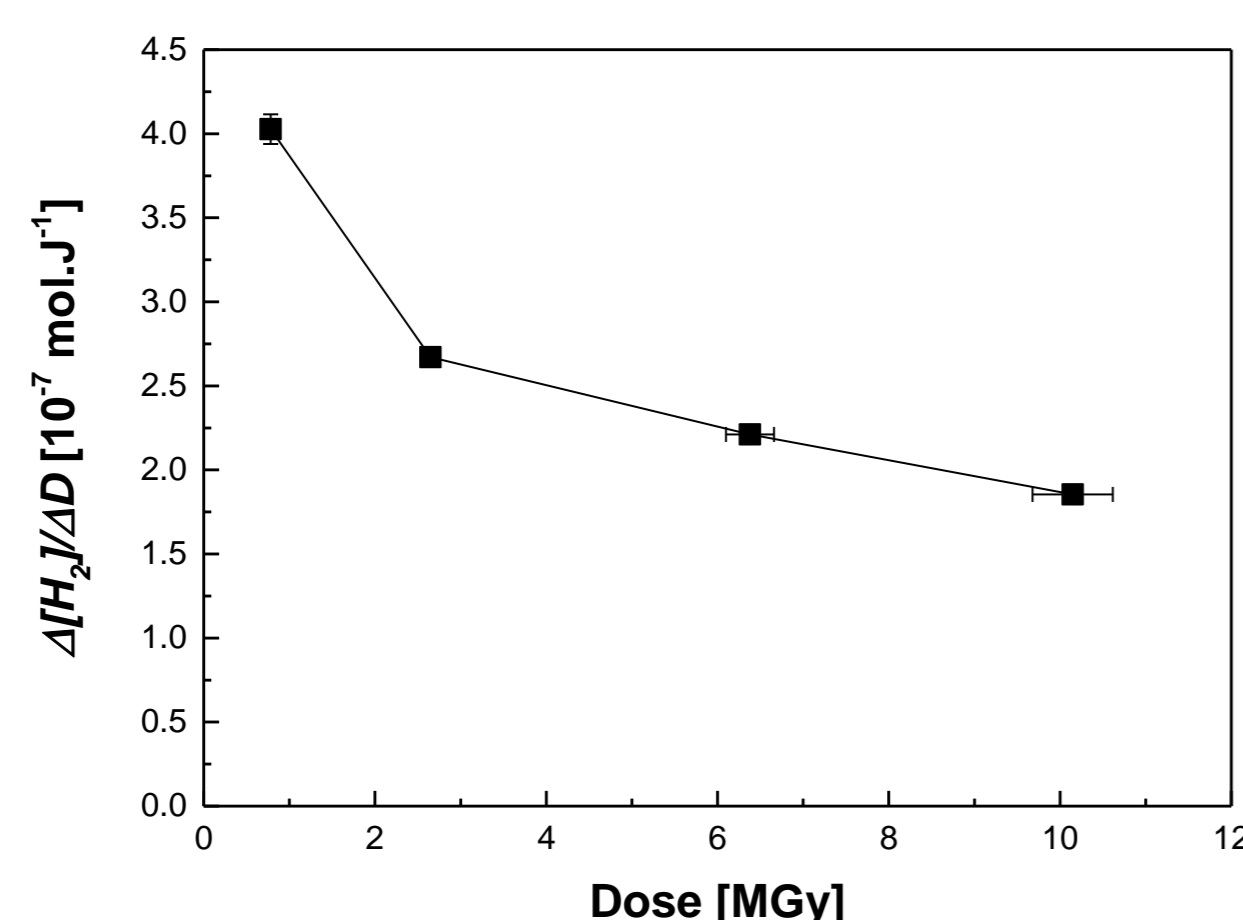


- In HDPE :

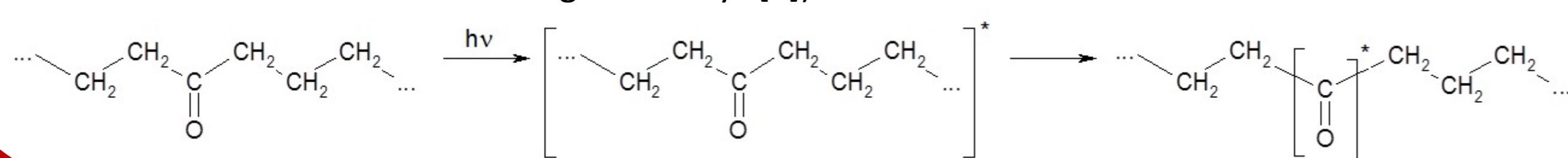
- Equivalent excitation densities along the ion track with both ion beams
- But: difference in radial repartition of dose
- No LET effect on the formation of ketone defects (in the LET range explored) [4]



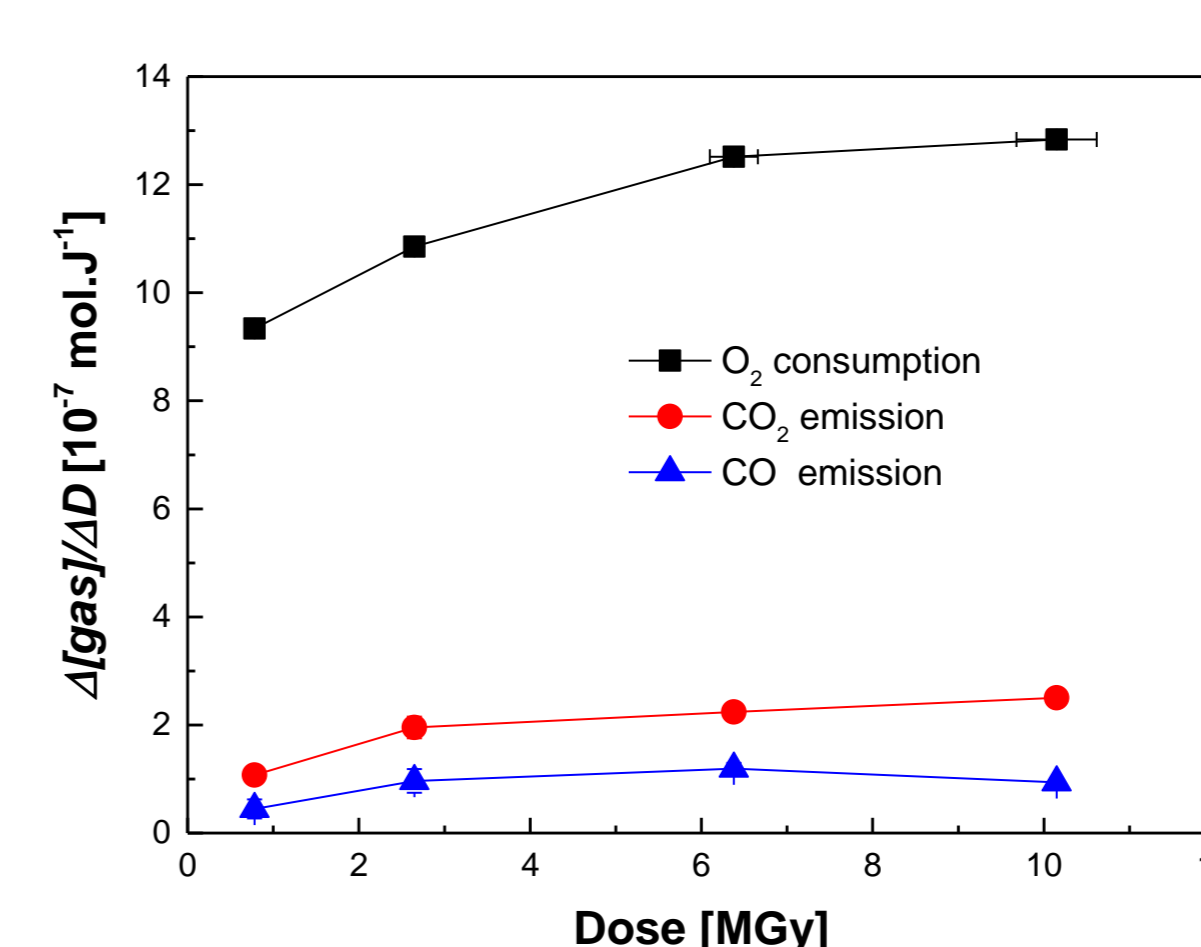
Hydrogen emission



- H₂ instantaneous emission rate ↘ when dose ↗
 - Previously observed by Seguchi [5]
 - => Under vacuum and gamma rays up to 2 MGy
 - => Assigned to energy transfers towards radiation-induced C=C double bonds
 - In this work, first evidence of radiation protection
 - => Under oxidative atmosphere and ion beam up to doses as high as 10 MGy
 - => Assigned to energy transfers towards oxidized defects (for ketone defects, observed by Slivinskas & Guillet under gamma-rays [6])



Oxygen consumption and carbon oxides emission



- When dose ↗: O₂ consumption and CO and CO₂ emission instantaneous rates ↗ and then →
- Competing reactions
 - ↘ in radiation-induced radical formation on the non-modified polymer
 - => Evidenced by the decrease of hydrogen formation with dose
 - ↗ of oxidized defects degradation
 - => Direct action of irradiation
 - => By energy transfer on these oxidized defects (like hydroperoxides) acting as energy sinks
- At high doses: overlapping of protection spheres
 - => Constant O₂ consumption and CO and CO₂ emission instantaneous rates

Conclusions and ongoing studies

- Conclusions
 - H₂ emission ↘ and oxygen consumption and CO₂ emission ↗
 - Oxidized defects created upon radio-oxidation act as energy sinks
 - Energy transfers towards radiation-induced defects are also effective under ion beam and under oxidative atmosphere
- Better understanding of aging mechanisms involved at very high doses, of primary importance for the deep underground repository
- Ongoing studies
 - Correlation between macromolecular defects and gas emission ?
 - => Better understanding of in-film modifications (FTIR, TGA, DSC...) in HDPE
 - Energy transfers evidenced in polyethylene: what about the other polymers?

Bibliographic references

- [1] C. Lamouroux, S. Esnouf & F. Cochin, GLOBAL conference, Salt Lake City, USA (2013)
 [2] D. Lebeau et al., J. Nucl. Mater. 460 (2015), 130
 [3] B. Gervais & S. Bouffard, Nucl. Instrum. Methods Phys. Res., Sect. B 88 (1994), 355
 [4] Y. Ngono-Ravache et al., Polym. Deg. Stab. 111 (2015), 89
 [5] T. Seguchi, Nucl. Instrum. Methods Phys. Res., Sect. B 185 (2001), 43
 [6] J.A. Slivinskas & J.E. Guillet, J. Polym. Sci. : Polym. Chem. Ed. 12 (1974), 1469

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