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

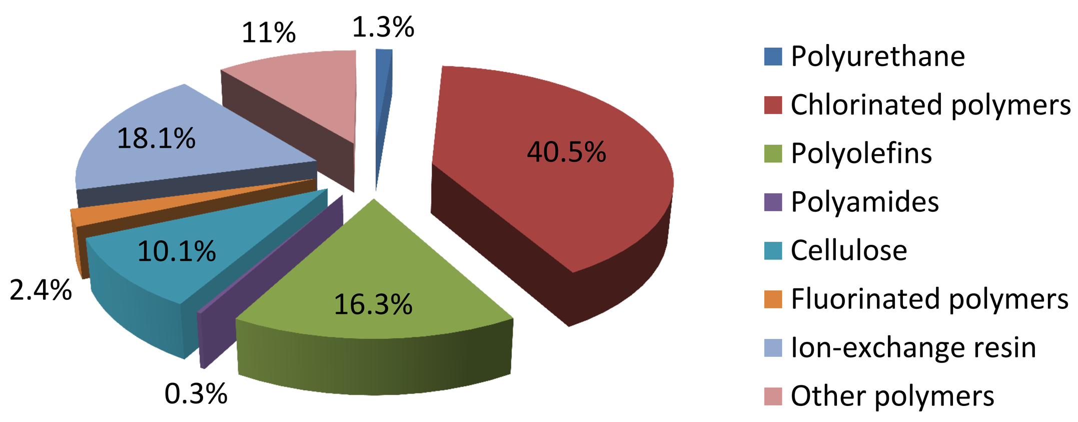
- Intermediate Level Long Lived Waste (IL-LLW) : various organic materials, including polymers, in presence of radionuclides
  - Production of potentially explosive (hydrogen, methane…) or corrosive (hydrogen chloride, fluoride…) gases
- In the French nuclear safety context: development of a 500-meter 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

IL-LLW tank

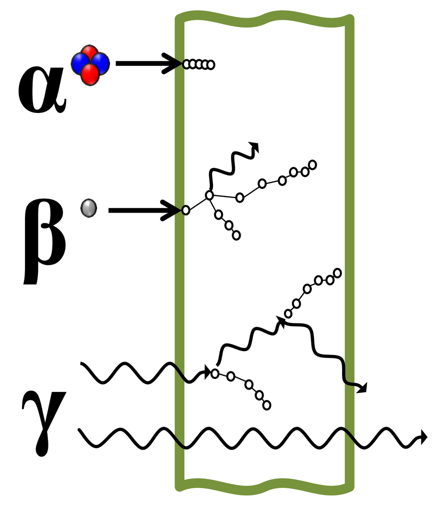
Aim

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

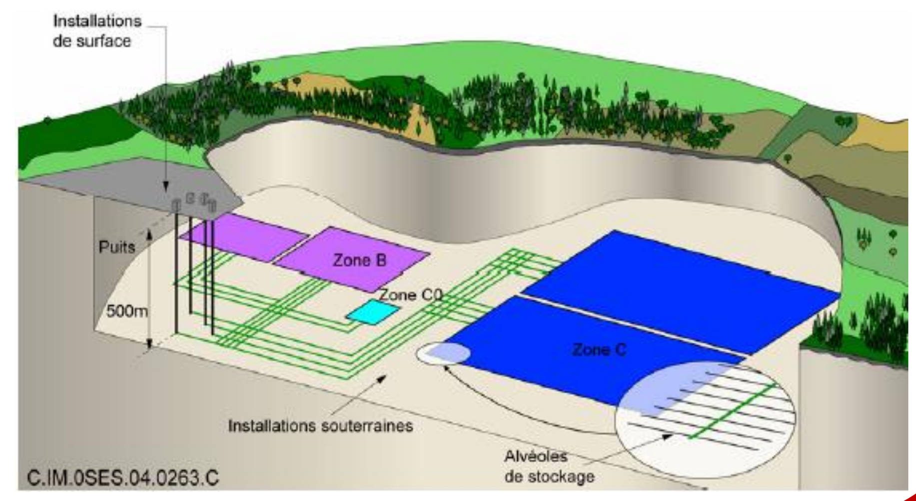
Polymers used in the French electronuclear industry



Ionizing radiations encountered in IL-LLW



Projected deep underground geological repository

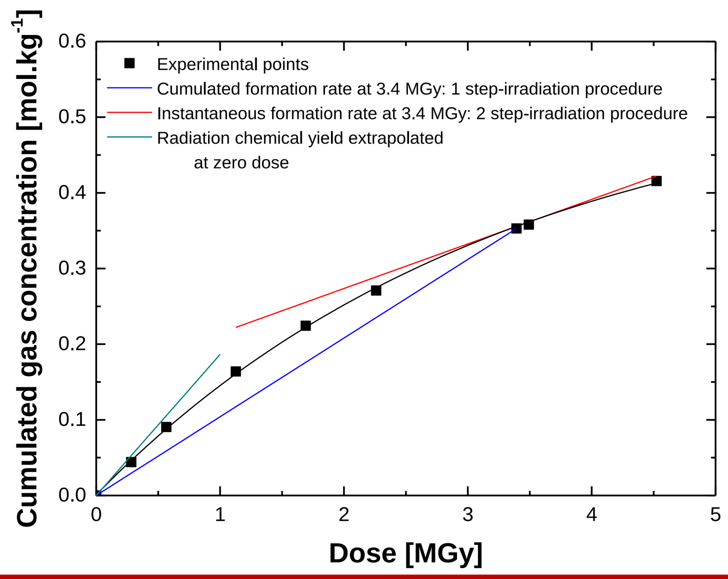


Material

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

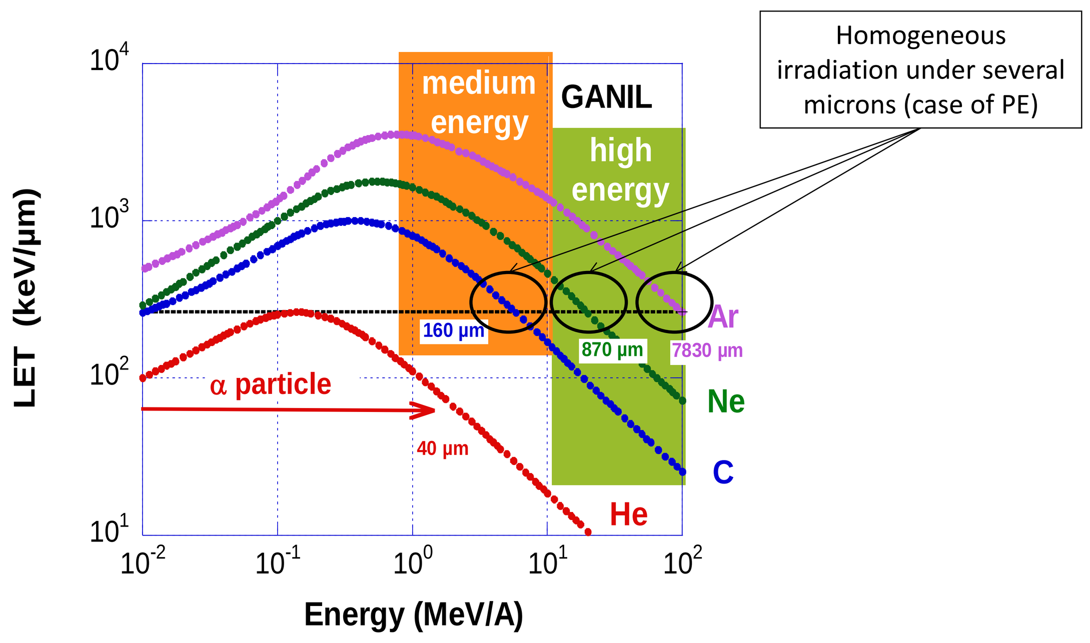
Irradiation conditions : a 2 step-irradiation procedure [2]

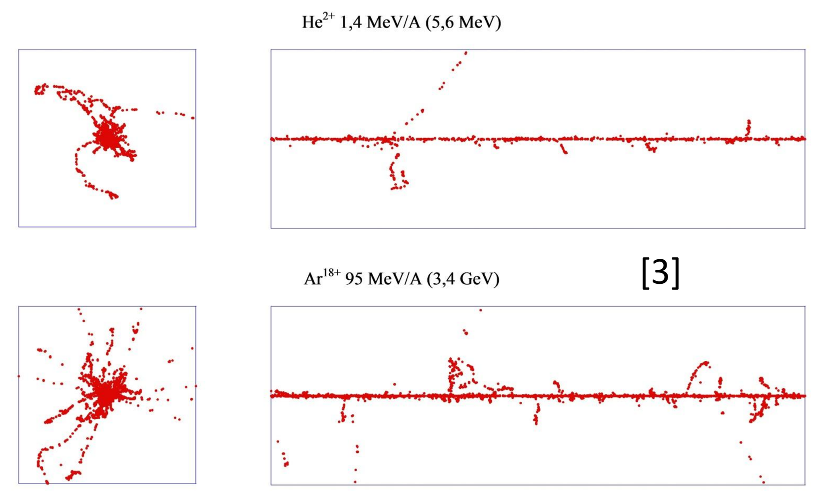
- Pre-aging: irradiation at high and various doses, under oxygen, without gas analysis
- Gas emission and oxygen consumption quantification: irradiation at low dose in closed container, along with gas analysis
- Storage conditions: pre-aged polymers stored under inert atmosphere and in the dark
- Result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

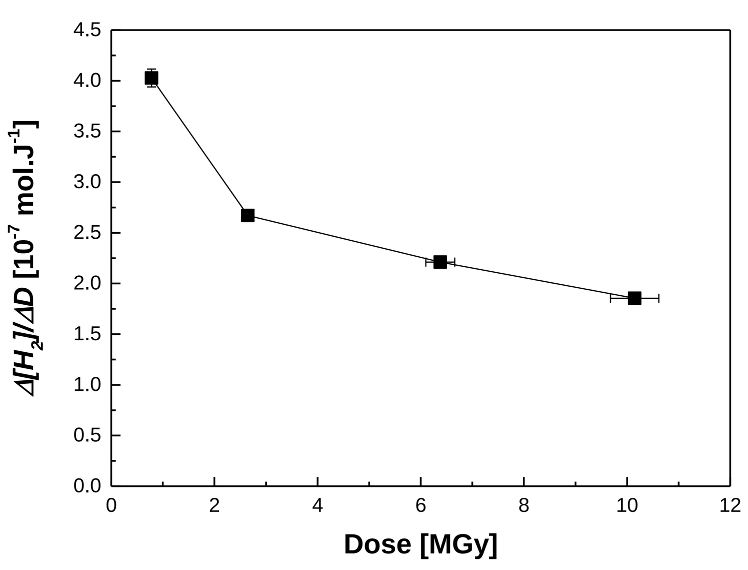




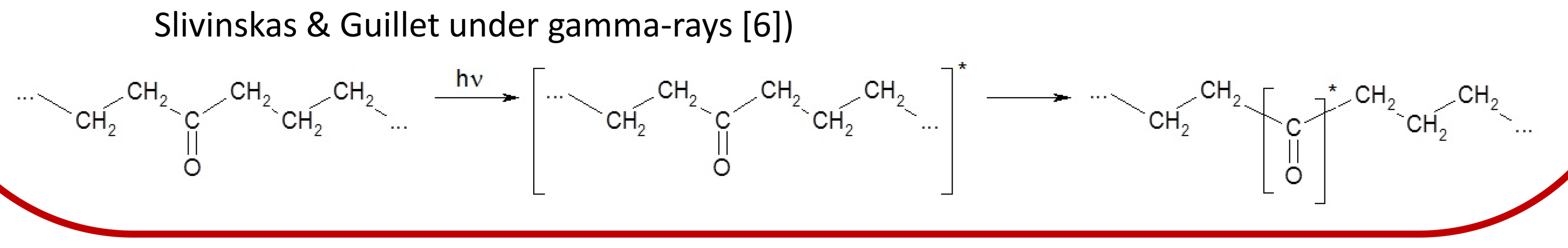
- In HDPE :
  - Equivalent excitation densities along the ion track with both ion beams
  - But: difference in radial repartition of dose
- No LET effect on the formation of ketone defects (in the LET range explored) [4]

- Pre-aging: ^{13}C (10 MeV/A) beams
  - Medium Energy beam line of GANIL ($3.6{\cdot}10^{8}$ ions.cm^{-2}.s^{-1} up to $1.2{\cdot}10^{13}$ ions.cm^{-2})
- Gas emission and oxygen consumption quantification: ^{36}Ar (95 MeV/A) beam
  - High Energy beam line of GANIL ($3.6{\cdot}10^{8}$ ions.cm^{-2}.s^{-1} up to $2.6{\cdot}10^{12}$ ions.cm^{-2})
    - => Necessity to go through glass walls of the closed containers
    - => Necessity to ensure constant LET through samples thickness

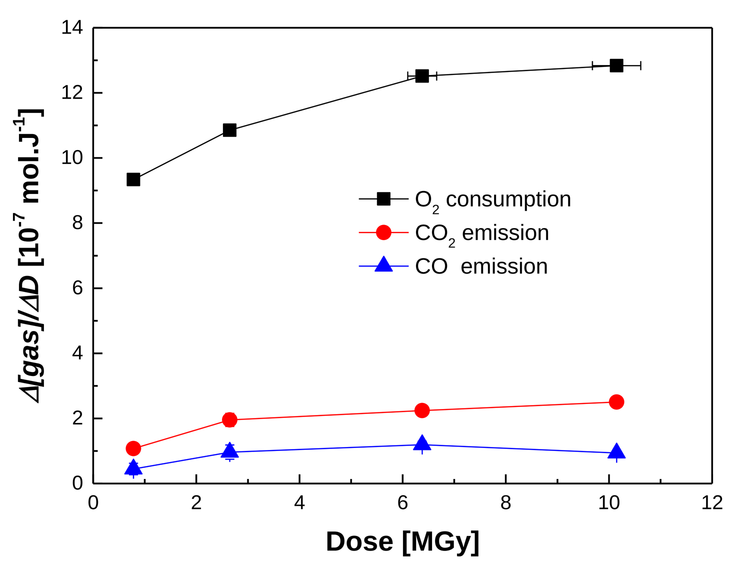
Hydrogen emission



- H_2 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 ↗: O_2 consumption and CO and CO_2 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 O_2 consumption and CO and CO_2 emission instantaneous rates

Conclusions and ongoing studies

- Conclusions
  - H_2 emission ↘ and oxygen consumption and CO_2 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?

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