SERESSA 2022

5th to 9th of December at CERN, Geneva

Introduction to ‘Radiation to Materials’: methodologies and examples

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Outline

Materials in high radiation areas: context

General **outline**:

- Electronics and **non-metallic materials** are increasingly used out of necessity in high radiation areas;
- Non-metallic materials **higher sensitivity to radiation in comparison to metals** – lower than electronics;
- The **selection of radiation tolerant components** is crucial for reliable operation, upgrades and development of **integrated system solutions**.
- The **risk of failures** in operation must be minimized.

GENERAL APPRECIATION OF RADIATION DAMAGE TO MATERIALS

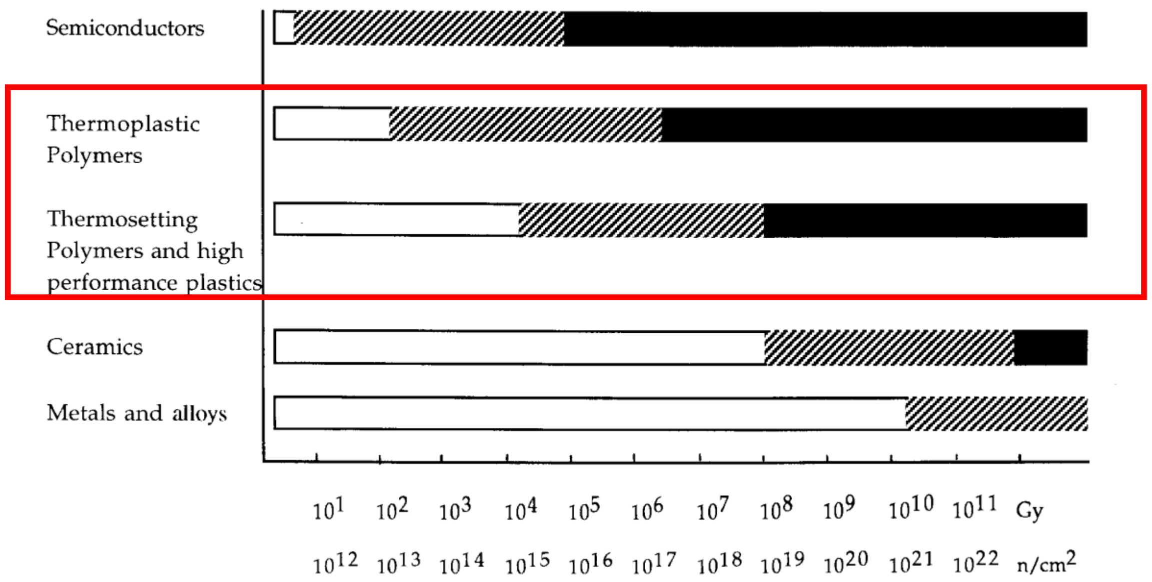


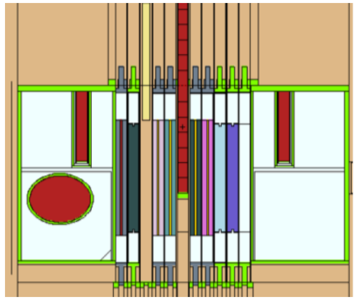
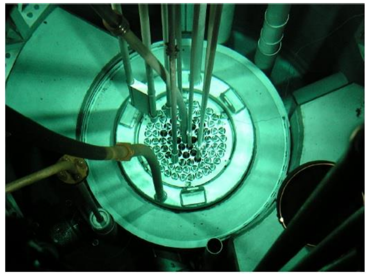
Figure 1: General appreciation of radiation damage to materials. Shown are ranges where materials undergo no damage (blank) mild to severe damage (shaded) or destruction (black).

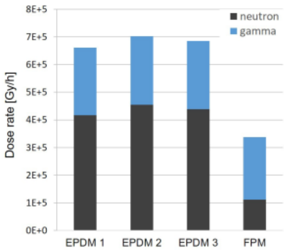
➢ H. Shönbacher, M. Tavlet, *Workshop on Advanced Materials for High Precision Detectors*, 1994

Multi disciplinary multi-scale approach

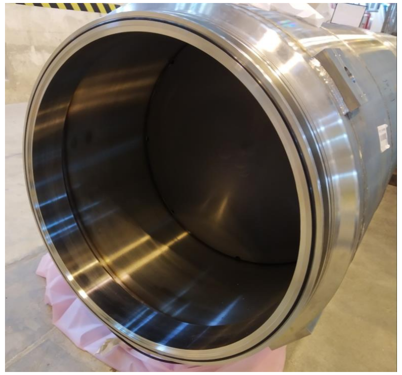
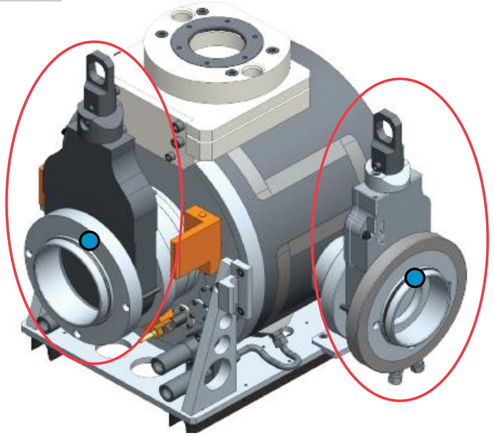
Main involved disciplines, including but not limited to:

- Radiation physics;
- Radio-chemistry;
- Dosimetry (experimental, Monte Carlo);
- Chemistry;
- Materials science;
- Mechanical engineering;
- …others depending on specific materials and application



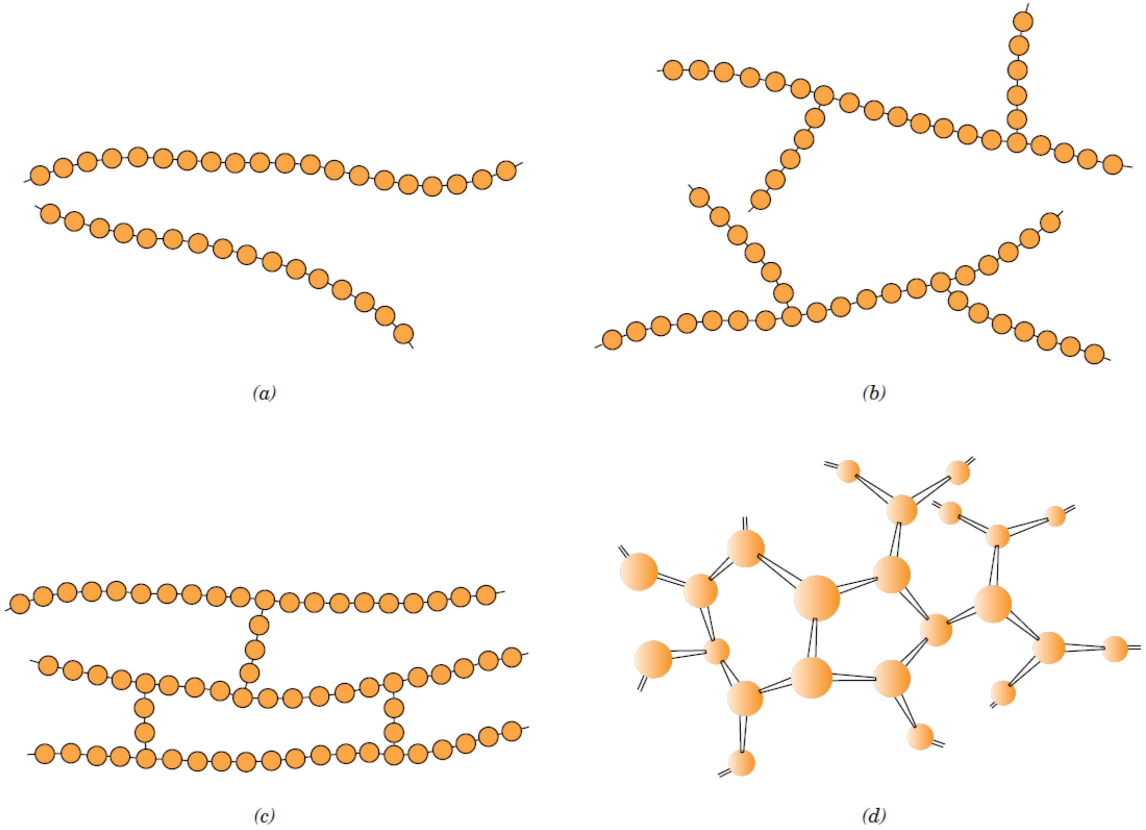


Today: overview

Polymeric Materials

Polymeric materials: structure (overview)



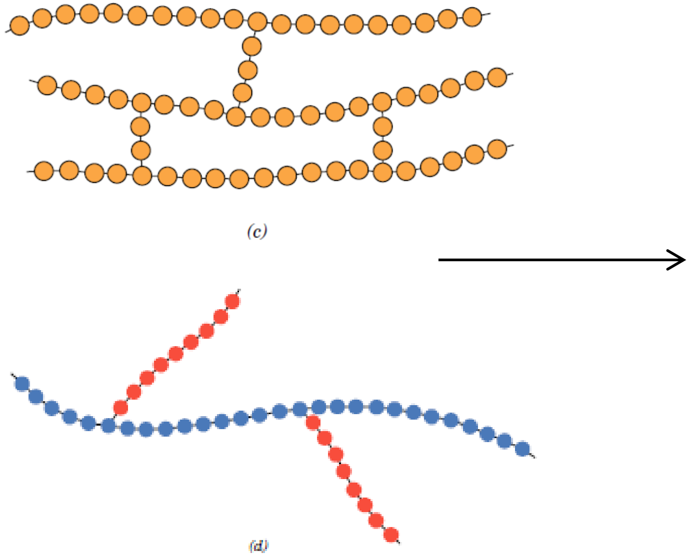
Simplified 2D polymeric structure: an example

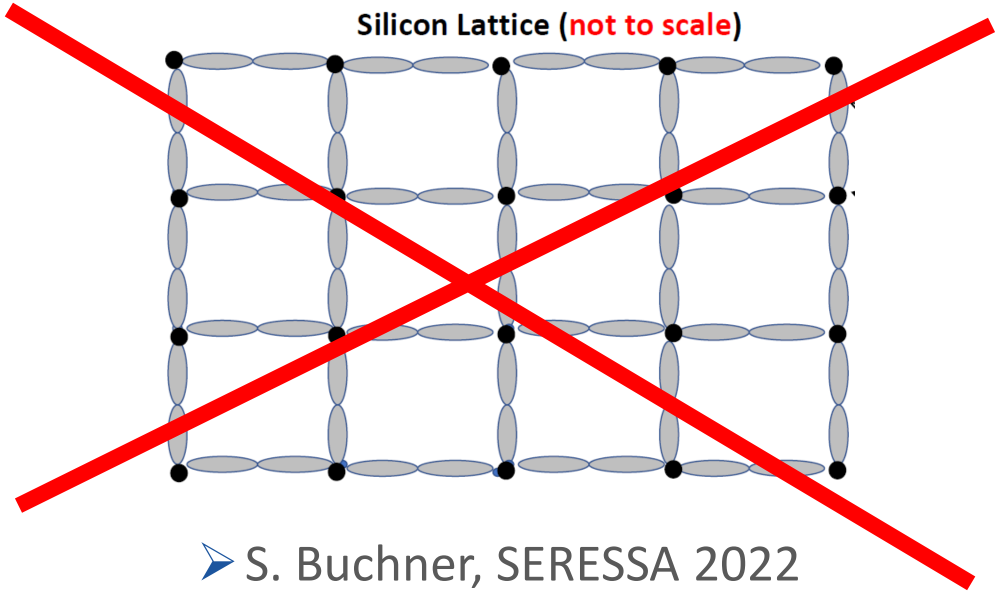
Image: Materials Science and Engineering: An Introduction - Callister 2010 8th Edition

Polymer - Definition

- ✓ A polymer is a substance composed of **macromolecules**, molecule of high relative molecular mass, the structure of which essentially comprises the **multiple repetition of units**. [*IUPAC definition*]
- ✓ Polymers include the familiar **plastic and rubber materials.** [*Callister 2010*]

From base polymer to commercial materials





➢ S. Buchner, SERESSA 2022

NO REGULAR LATTICE

- Complex multi-phase materials;
- Irregular and entangled/crosslinked structure;
- Different degrees of crystallinity or amorphous structures.

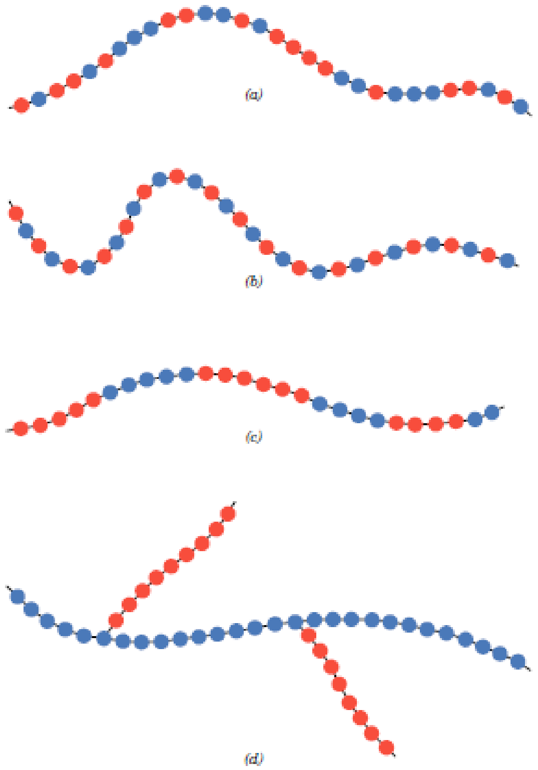
Polymers: main ingredients

Main ingredients:

- **Base polymer(s)** – huge variety of chemistry (natural, synthetic…) and of structures and arrangements;
- **Additives**: key role in determining the final properties;

But also:

- **Manufacturing** techniques;
- **Quality control** aspects;
- **Reliability** of manufacturers;
- …



Simplified polymer: an example

Callister 2010 8th Edition

Same ingredients, different arrangement.

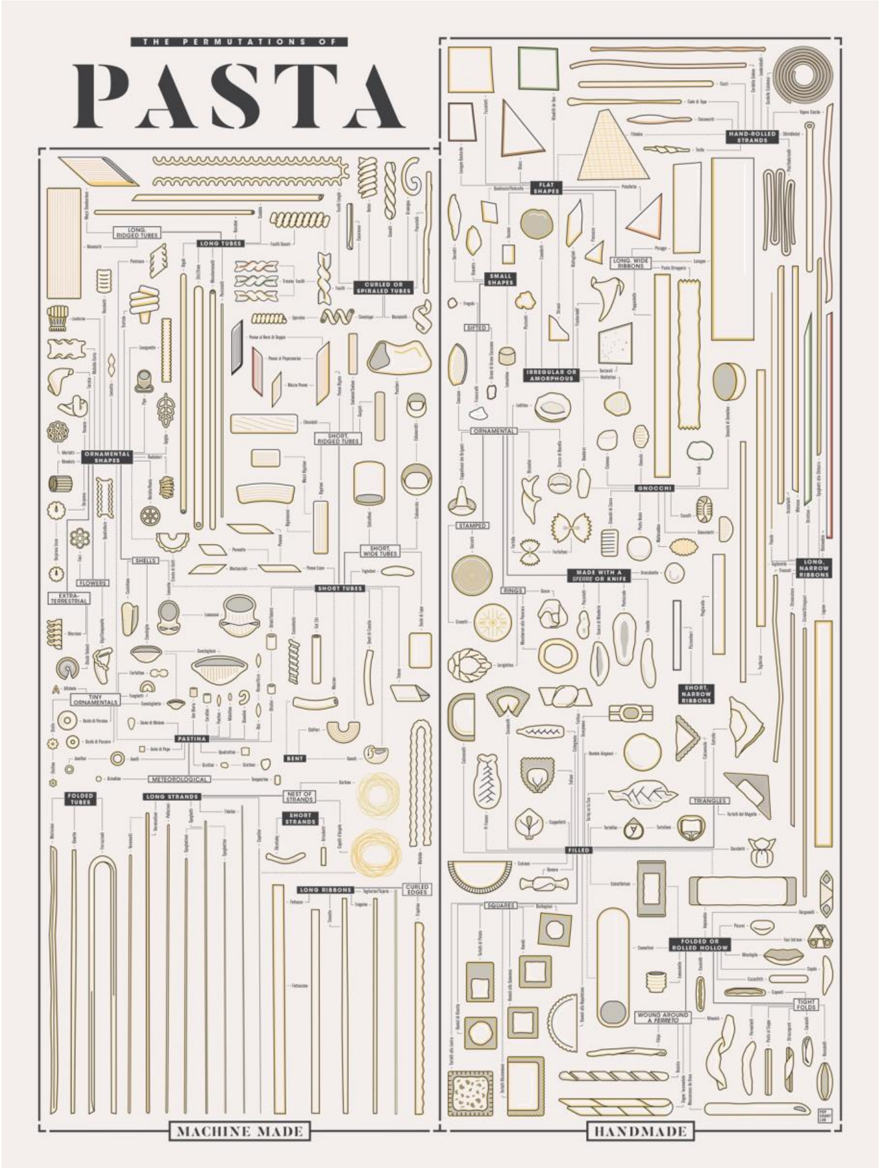
Are the final properties the same?

The pasta example: format

Same raw material
but

Different shapes;
Different use;

Completely different experience



https://www.prestelli.com/2016/02/23/tipi-di-pasta/

Pop Chart Lab, *The Permutations of Pasta*
(source: popchartlab.com)

The pasta example: recipes

Same raw material;
Similar shape
but
Different additives;
Completely different taste!

Additives: huge role in determining final properties!

The pasta example: the chef counts!

- Ratatouille, Disney

Same raw material;
Same additives;
Same recipe;
but

Different cooking technique

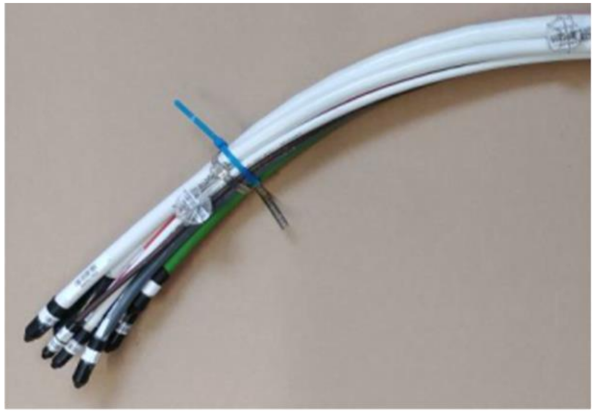
Completely different taste!

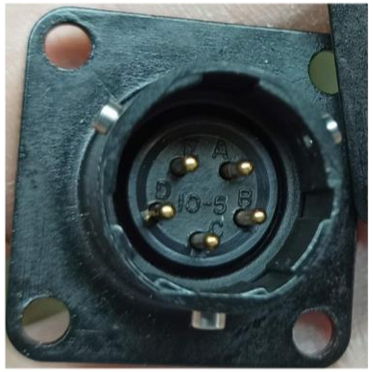
Polymer-based materials and components

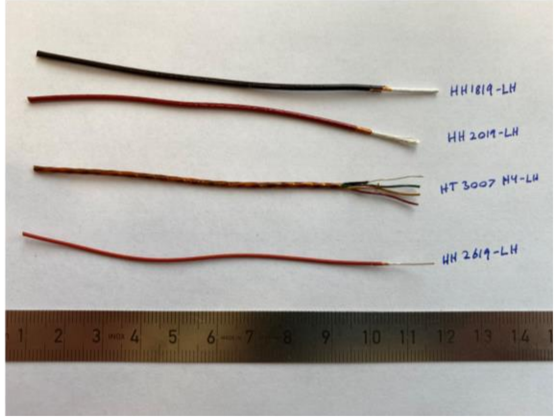
Examples of commercial components used in high-radiation areas:

- Lubricants (oils and greases)
- Elastomeric materials (O-rings, seals…)
- Insulators (cable components)
- Glues / adhesives
- Resins
- Structural materials
- Plastic components of commercial devices
- **Assemblies / complete systems**

➢ CERN *EDMS* R2M Archives







Interaction between polymers and radiation

Ionizing radiation

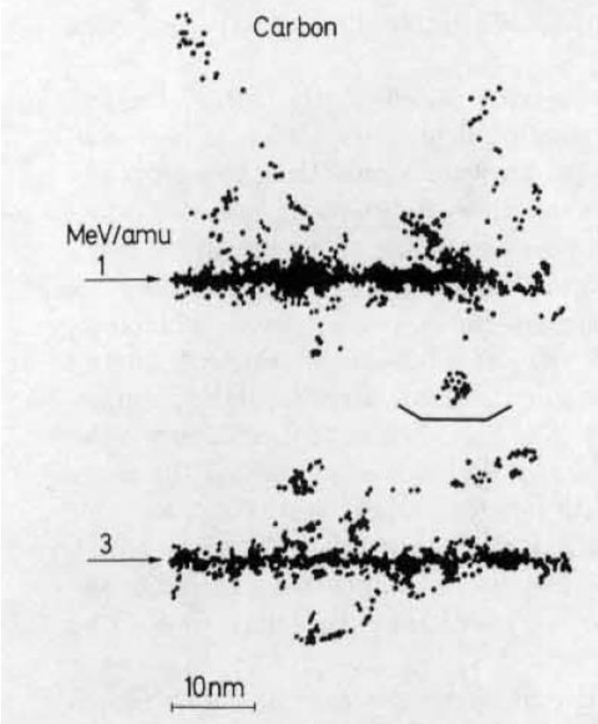


Figure 4.17. Simulated track structures. Each dot represents an energy deposition event (from PARETZKE 1980)

Ionizing radiation IR - Definition

- ✓ Radiation **having enough energy to ionise** (extract electrons from) atoms and molecules.
- ✓ Different molecules and atoms ionise at **different energies** (typically starting between 10 eV - 33 eV).
- ✓ **No clear boundary between IR and NIR** (in the tens /hundreds eV region).
  [Wikipedia]

Types of IR / Energy range / in specific applications

Reference unit: absorbed dose

Main effect: **ionisations**

Absorbed dose – Definition

- ✓ The total (radiation) energy absorbed per unit mass
- ✓ $Dose = \frac{E}{m}$ [Gy = J/kg]

Dose rate - Definition

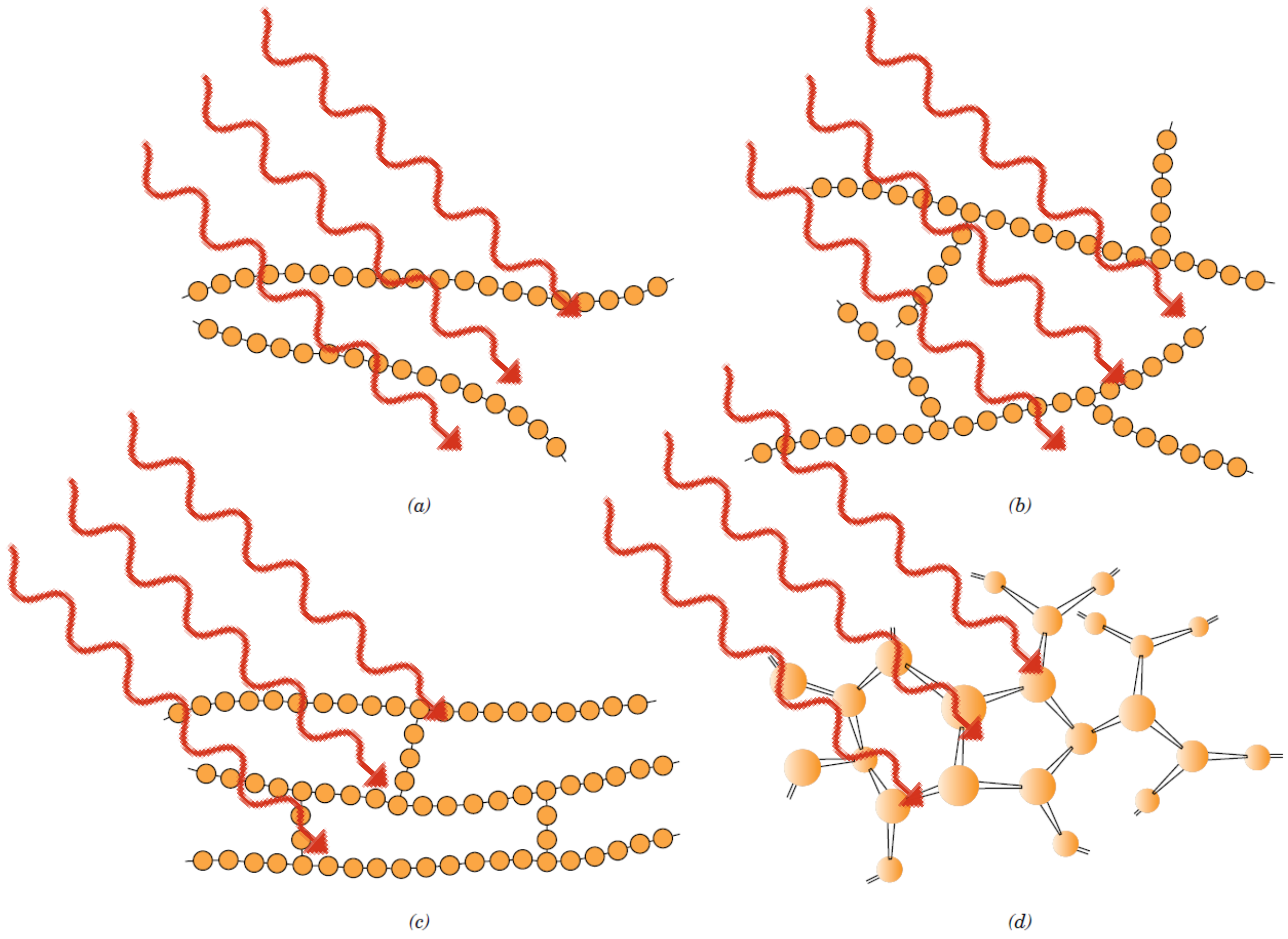
- ✓ The total (radiation) dose absorbed per unit of time
- ✓ $Dose\ rate = \frac{D}{t}$ [Gy/s]

Which **dose ranges** are relevant for materials?

What about displacement damage?

PS: many other dose-related quantities do exist! (equivalent dose, effective dose…)

Interaction between radiation and materials



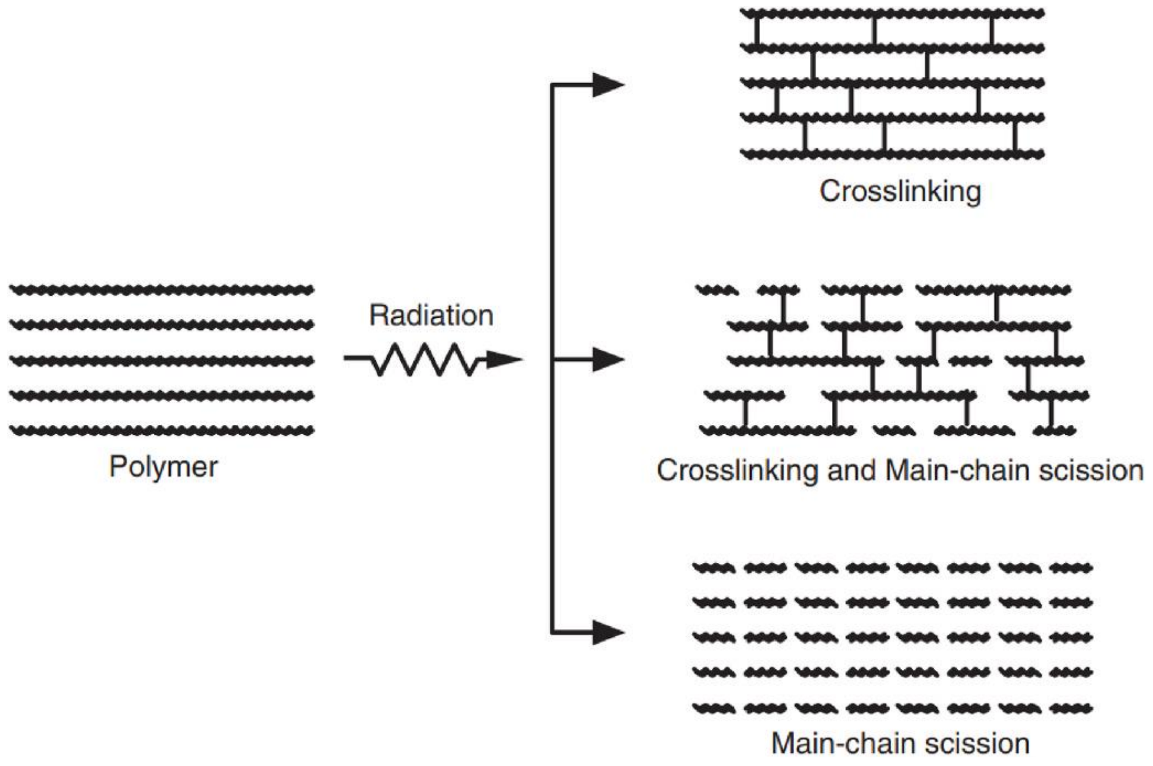
VERY COMPLEX SCENARIO

Dominant effects (physical-chemical phase):

- **Chain cleavage and scission** broken molecular bonds. Reactive species (**radicals**) and **fragments** are produced. They can diffuse/recombine
- **Cross-linking, polymerization**: **recombination** of broken chains, **Cross-linking points** and/or longer molecules are generated.

RE-ARRANGEMENT!

Crosslinking vs scission



Effect of radiation on polymers: schematic representation

- J.Zimmermann et al.,

International Journal of Adhesion & Adhesives 117 (2022) 103014

Somehow **competitive processes** (usually one is dominant) but **occurring in parallel**.

In which scenario the material becomes **softer**?

In which scenario the material becomes **harder**?

Macroscopic examples of radiation damage

Visible radiation effects in materials

MICRO
- **Chain scission**
- **Cross-link**

MACRO
- **Hardening**
- **Softening**

Left: ordinary commercial grease

Right: grease irradiated in reactor mixed field **0.45 MGy**

Fluidisation of the grease!

Structural and functional modification

➢ M.Ferrari et al., Heliyon 5 (2019) e02489

Dripping grease
0.45 MGy irradiation
Mixed field
STRUCTURAL FAILURE

- M.Ferrari et al., Heliyon 5 (2019) e02489

Other common radiation effects

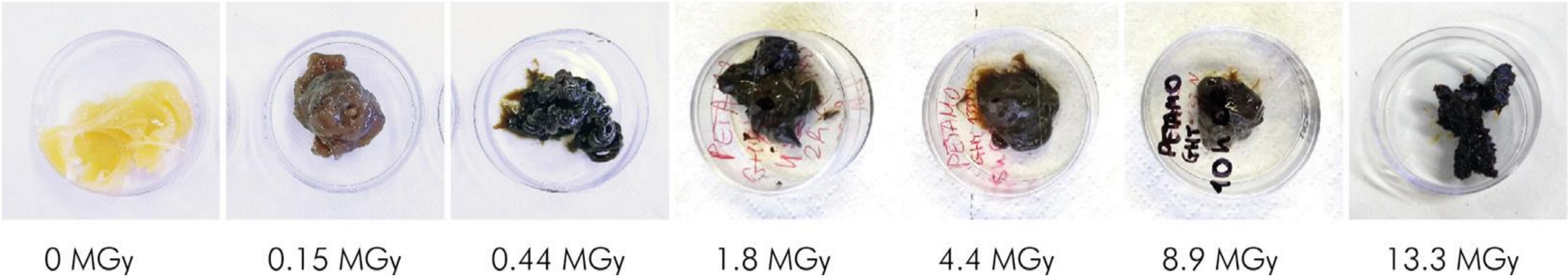
MACROSCOPIC EFFECTS:

- Softening, fluidization;
- Hardening / embrittlement;
- Acid products;
- Oxidation / corrosion;
- Gas production / swelling;
- Deformation;
- Bubble production;
- Colour darkening;
- Production of fragments;
- …

Fluorinated grease **1 MGy** irradiation – Al corrosion

➢ M.Ferrari et al., Heliyon 5 (2019) e02489

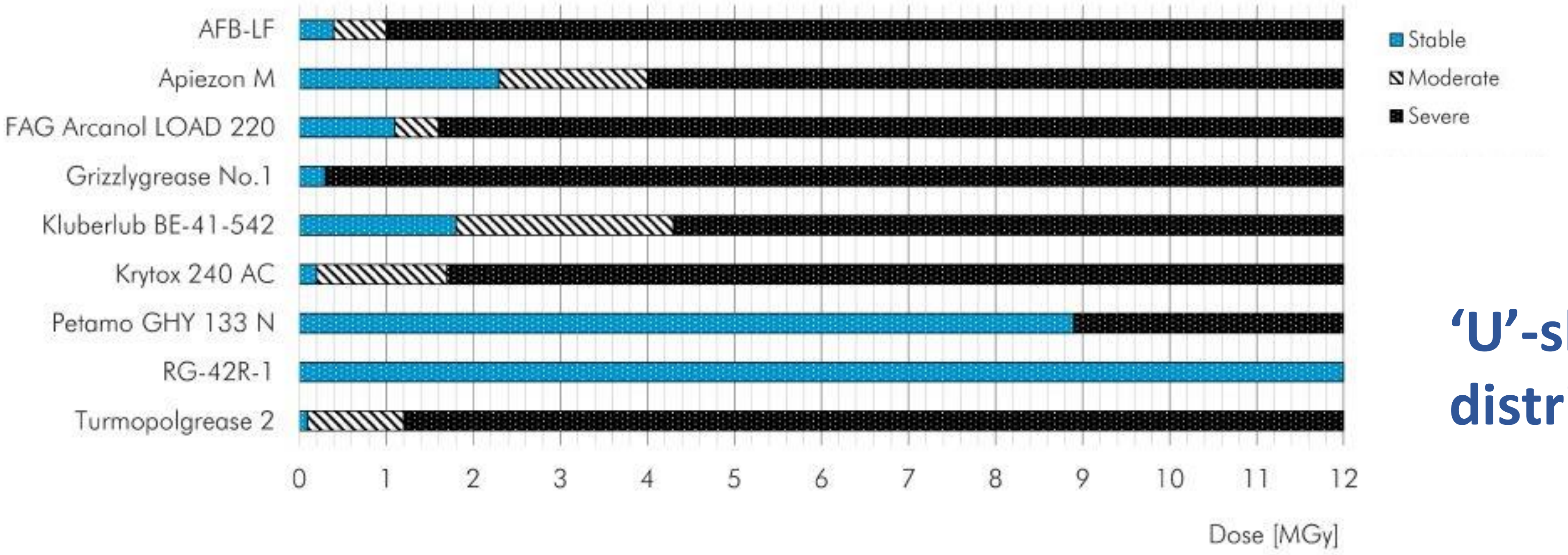
Radiation damage example: greases



- M.Ferrari et al., Heliyon 5 (2019) e02489
- M.Ferrari et al., *Nuclear Materials and Energy, vol 29,* 101088 (Dec 2021)

Color change and grease hardening (structural failure)
Grease failure at approx. 9 MGy

Thresholds of damage for commercial greases



'U'-shaped distribution?

- M.Ferrari et al., [Heliyon 5 (2019) e02489]
- M.Ferrari et al., [*Nuclear Materials and Energy, vol 29,* 101088 (Dec 2021)]

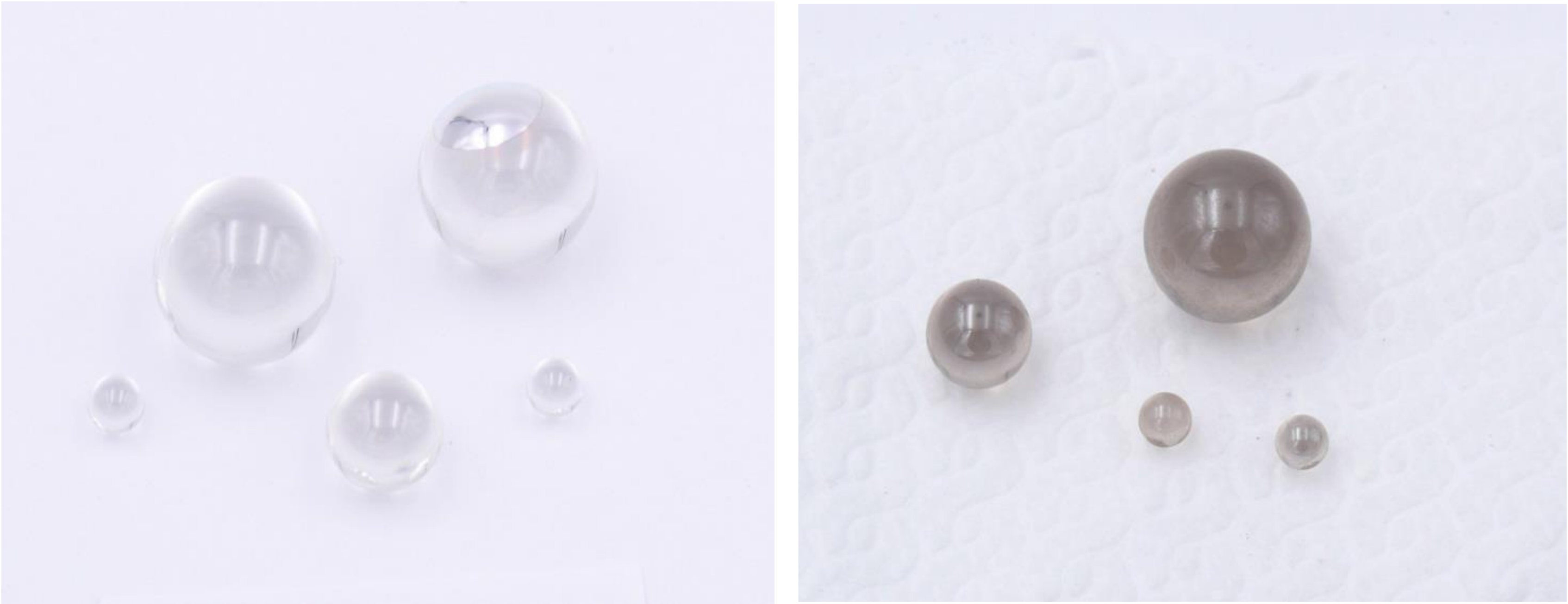
2 orders of magnitude of difference in radiation tolerance!

EPDM Elastomeric O-ring
3 MGy irradiation

STRUCTURAL FAILURE

- A.Zenoni et al., Rev. Sc. Inst. 88, 113304 (2017);

Radiation damage example: glass



Glass balls, precision components for accelerator alignment system
Non-irradiated (left) vs **5 MGy i**rradiation (right)

- M.Ferrari et al, IEEE TNS 2022 (*submitted for publication*)

Radiation-induced darkening

Radiation damage example: protective covers

Protective covers for magnets – accelerator applications
Non-irradiated (left) vs **10 MGy irradiation** (right)

- M. Ferrari et al, IEEE TNS 2022 (submitted)

Radiation-induced deformation and swelling
Structural failure

Dose

Lubricant oil

7.8 MGy irradiation

Mixed field

VISCOSITY INCREASE due to polymerisation

- M. Ferrari et al., NIM B, Vol. 497, pp. 1-9 (2021).

Selection criteria for high-radiation applications

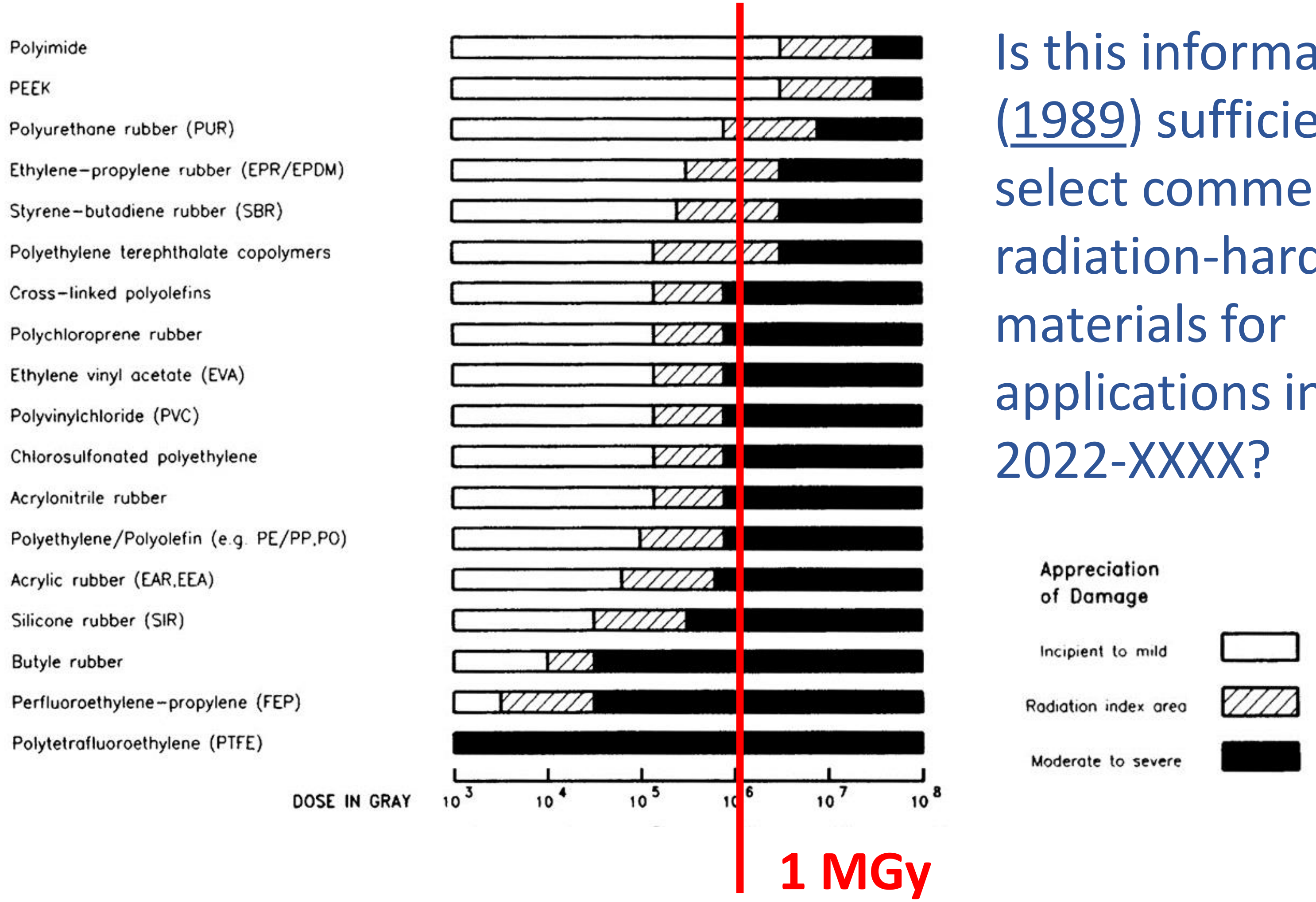
How to select rad-hard polymers?

LITERATURE:

General indication (o.o.m.) of the radiation tolerance of base polymer families (broad categories of commercial items)

Rad hard polymers:

Resist to MGy of dose



Is this information (1989) sufficient to select commercial radiation-hard materials for applications in 2022-XXXX?

➢ H.Schoembacker, M.Tavlet, **CERN Yellow Report** CERN 89-12 (1989).

Rad-hard lubricating oils? Example

Literature:

General indication on the radiation tolerance of base polymer families:

- Polyphenyls
- Polyphenyl ethers

…

+ additives, curing, quality control…

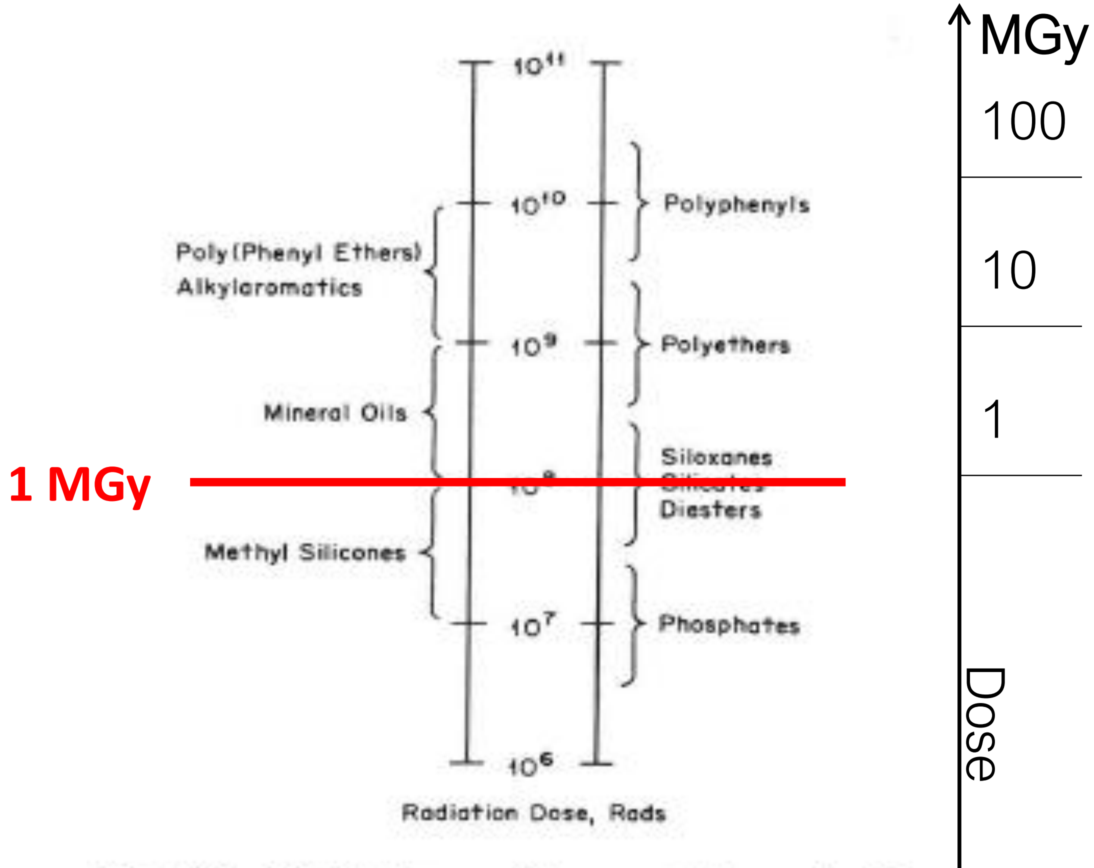


FIG. 9.3 Radiation resistance of base fluids.

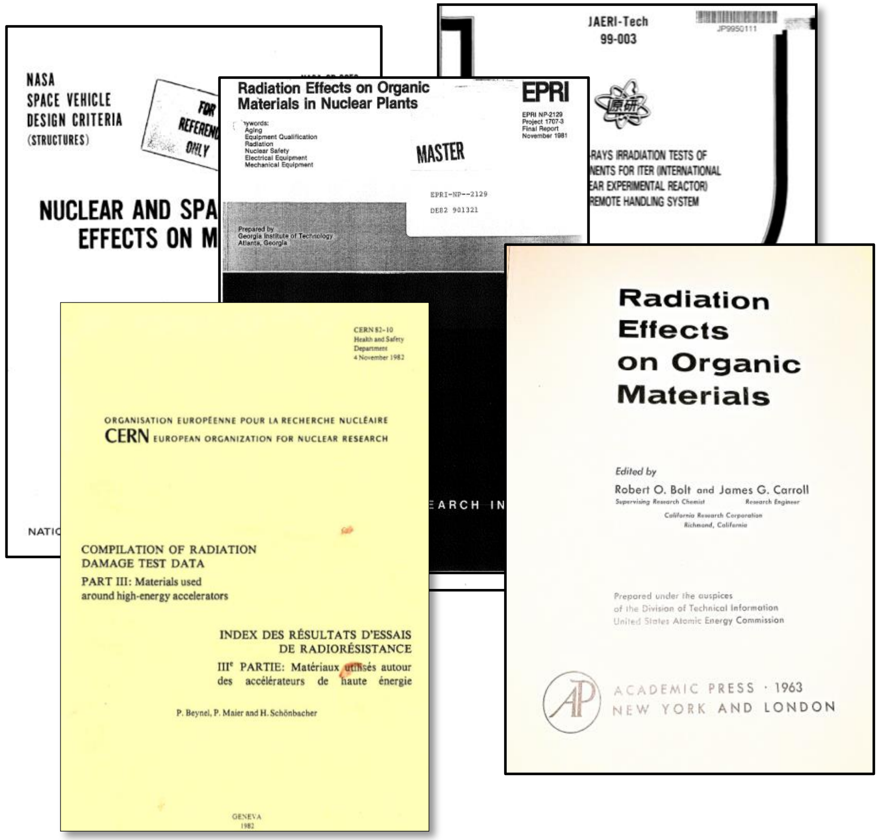
Indication that a commercial oil **might be** radiation tolerant

➢ R.O. Bolt, J.G. Carrol, Radiation Effects on Organic Materials (Academic Press, New York, **1963**).

Rad-hard commercial materials: literature

Literature:

Extensive data collected **between 50's and 2000's** on commercially available materials for the needed applications (accelerators, nuclear, military, space…)



Is this information sufficient?

Main limitations:

- **Old** information, in some cases obsolete;
- **Old**, discontinued products;
- Unverified assumptions
- **New** applications and technologies;
- **Lack** of scientific understanding;

➢ Example of references: CERN's Yellow Reports, NASA reports, studies for ITER…

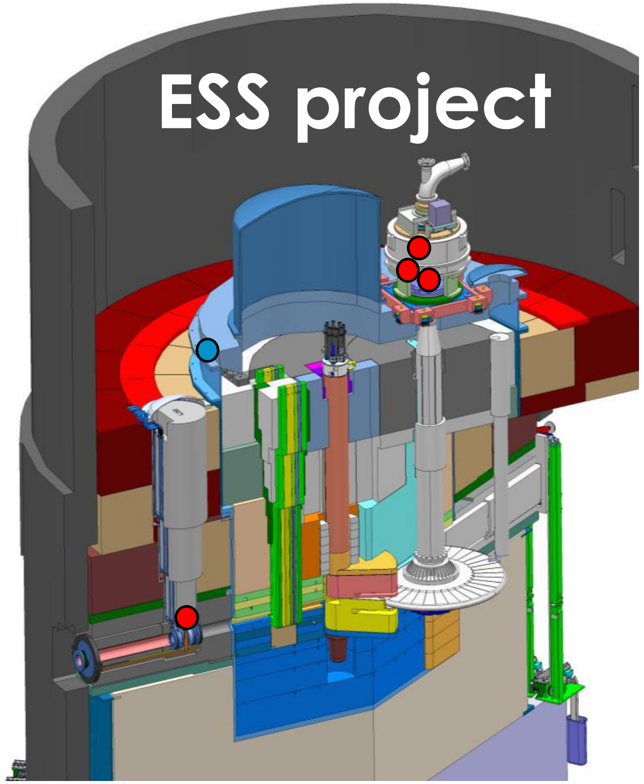
Is this technology still usable in this form for current space challenges?

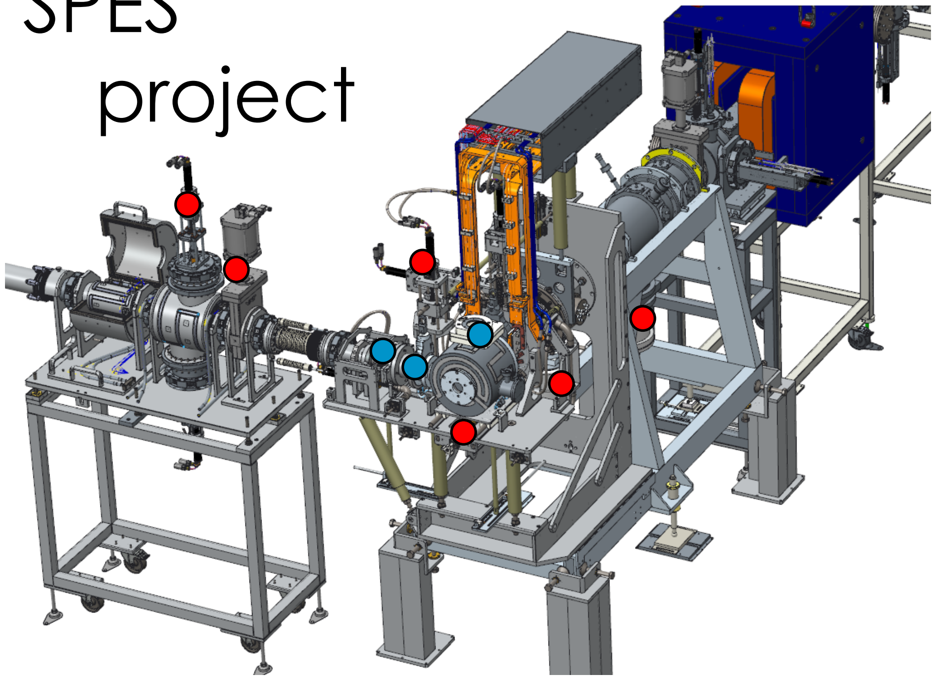
- First man on moon, 1969. Credits: NASA

Technologies being developed now: new needs!

Applications such as:

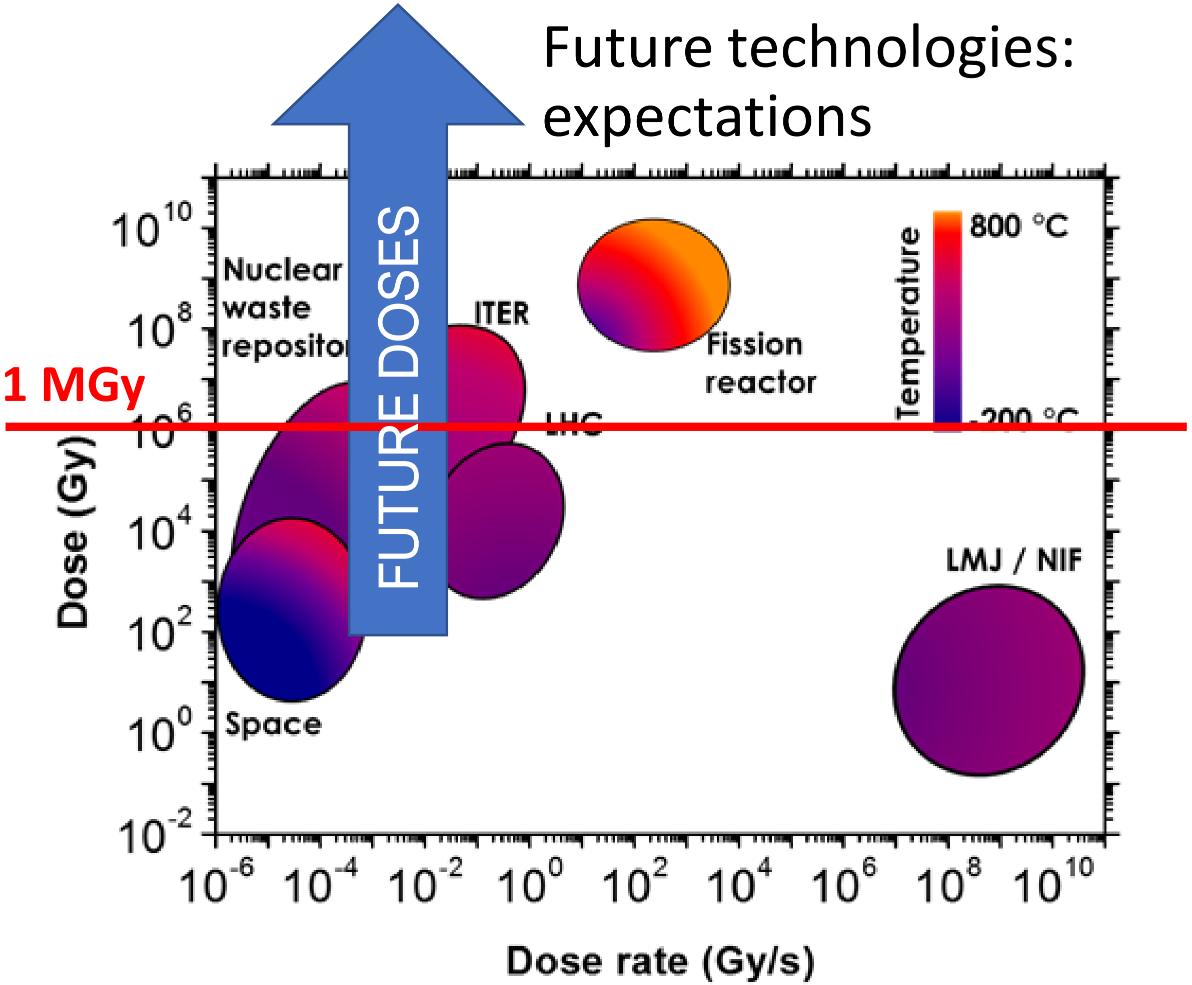
- Particle accelerators;
- High-power targets;
- Spallation sources;
- Fission reactors;
- Fusion technology;
- Radioactive waste;
- Space applications;
- Medical physics;
- …





- Elastomeric O-rings
- Greases and lubricants

Map of applications: dose vs dose rate overview



Upgrades, developments, new facilities, new technologies: **Doses are increasing and will keep on increasing**

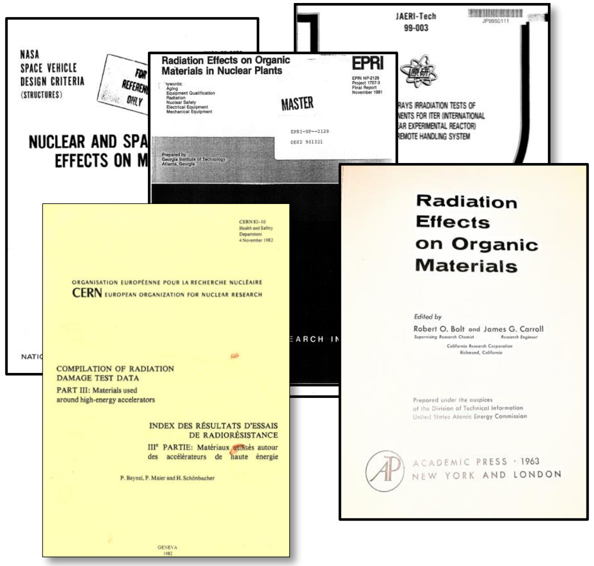
Map originally drawn for Optical Fiber application in high-radiation environments to be replicated for materials

https://laboratoirehubertcurien.univ-st-etienne.fr/en/teams/materials-for-optics-and-photonics-in-extreme-radiation-environments.html

Need for new radiation damage data

Radiation effects depend on:

- Total absorbed dose
- Dose rate
- Radiation energy spectrum
- Particle type
- Oxygen concentration
- Temperature
- Mechanical stress
- Other ageing factors



Current knowledge to be updated…

Irradiation and testing methodologies

What is radiation tolerance? Wrong answers only

Radiation tolerance – unacceptable definitions

A material / component is radiation tolerant because:

- a test on a somehow similar chemistry was performed in the 70's.
- its chemical composition is promising for radiation tolerance.
- it is chemically stable.
- it has been used in a certain application for several years (or in any other radiation environment) and no major failure was reported.
- we have always used it in this application.
- it was irradiated, tested and NOTHING HAPPENED.
- the producer declares so and the declaration is not supported by evidence.
- It is just a grease/O-ring, what can go possibly wrong?
- It has been irradiated once (in unknown irradiation conditions).

lack of scientific, systematic and complete approach

What can possibly go wrong?

What is radiation tolerance?

Radiation tolerance – *my tentative definition*

✓ A material or component is radiation tolerant up to a certain dose level when its mechanical, structural and/or functional **properties of interest are stable*** as a function of the absorbed dose. This is **experimentally** assessed** (irradiation in a specified set of irradiation conditions.)

- ***Stability** of a property: relative variation within a certain percentage of the unirradiated value and/or retain of a certain absolute value.
- **Nowadays, models are not sufficiently complete to allow radiation effects on a commercial product /material to be predicted without performing an **experimental test**. Assumptions might work relatively well with very pure materials and in standard irradiation conditions.

Data need to be collected to define ‘radiation stability’

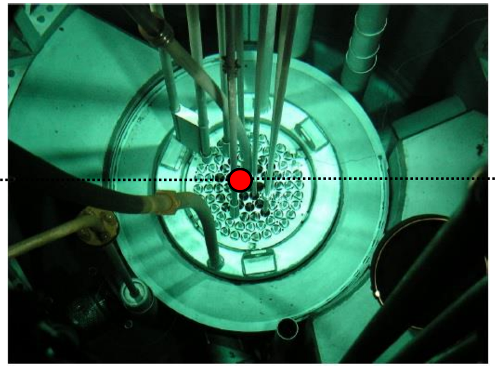
Are we happy with this definition? Need for new standards

Methodologies to be developed

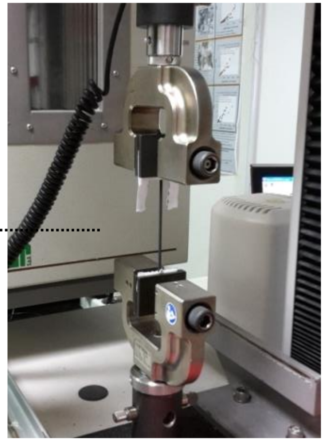
Main steps – iterative:

- **Selection** of commercial materials;
- **Characterization** of non irradiated samples;
- **Definition** of irradiation plan;
- **Irradiation** (in a set of irradiation conditions);
- **Post-irradiation characterization;**
- **Results** interpretation.

O-ring



Irradiation facility



Post-irradiation characterization

- A.Zenoni et al., [Rev. Sc. Inst. 88, 113304 (2017);]

General lack of standards & guidelines, system approach often missing

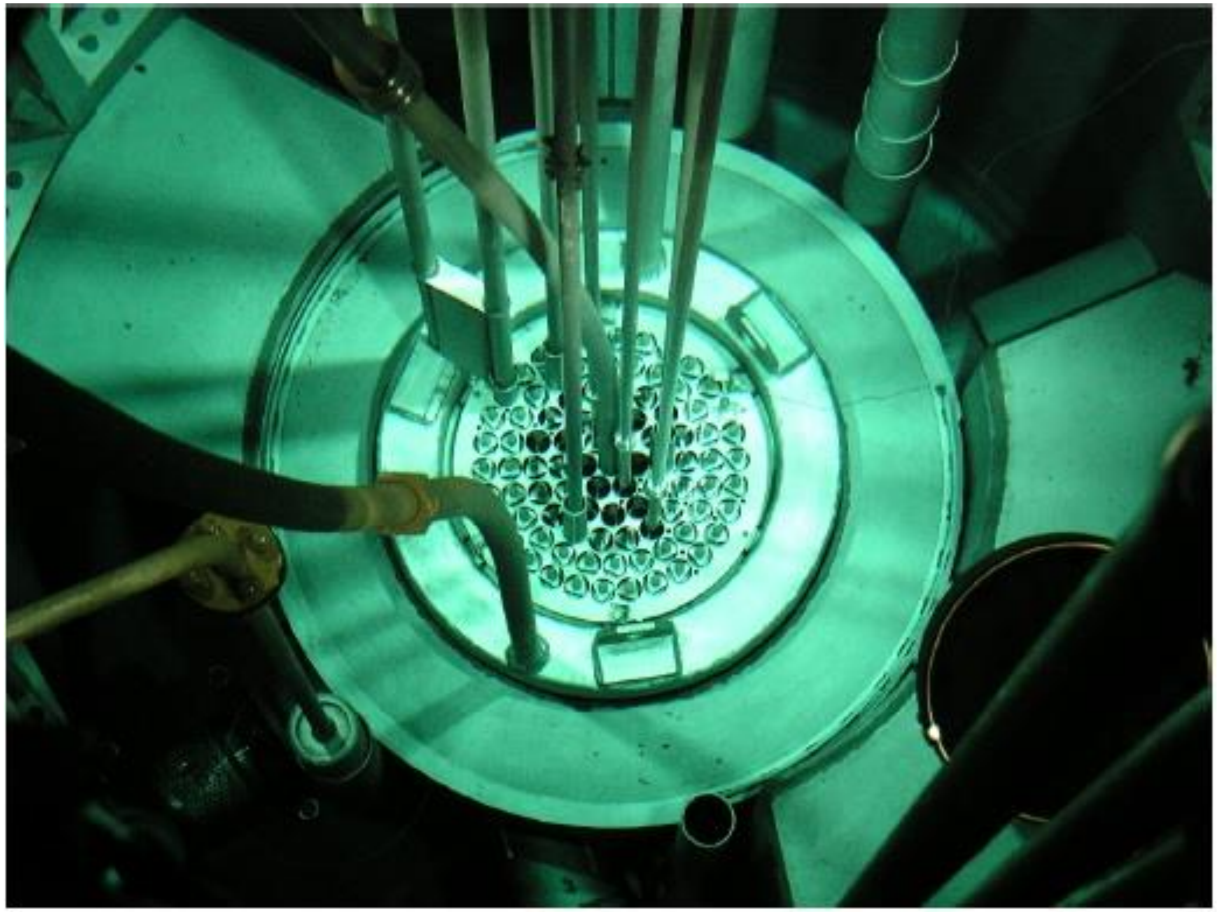
Irradiation facilities for materials damage

Specific technical needs:

- High doses: **MGy range;**
- High rates: **kGy/h** to **MGy/h**;
- Dose **homogeneity** for macroscopic samples;
- High Level **Dosimetry**;
- **Radiation protection** (for neutron/proton irradiation);
- Control/monitoring of **irradiation conditions**;
- very difficult **access.**

Difficult to achieve!

Need for coordination



➢ TRIGA Mark II research nuclear reactor, UniPv

A new irradiation station for materials at CERN

Main features:

- 24 sample positions
- dosimeters
- **since July 2021**
- dose: **1 – 2.5 MGy/y**
- mixed field: high energy neutrons and photons
- robotic handling
- radiation protection system
- elastomers and greases

‘Parasitic’ research station

➢ M.Ferrari et al., [Phys. Rev. Accel. Beams 25, 103001, 2022]

- M.Ferrari et al., Phys. Rev. Accel. Beams 25, 103001 20 October 2022

An example:
radiation effects
in elastomeric materials

An example: elastomeric EPDM O-rings studies

SELECTED MATERIALS:

- Different commercial EPDM based O-rings

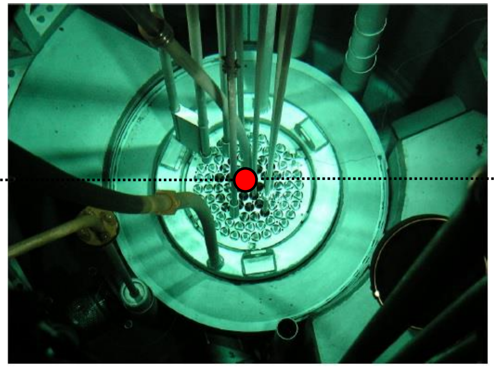
Samples:

- O-ring slices

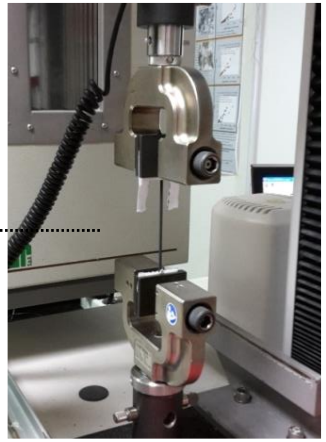
Irradiation:

- in-core facility, nuclear research reactor

O-ring samples



Irradiation facility



Post-irradiation characterization

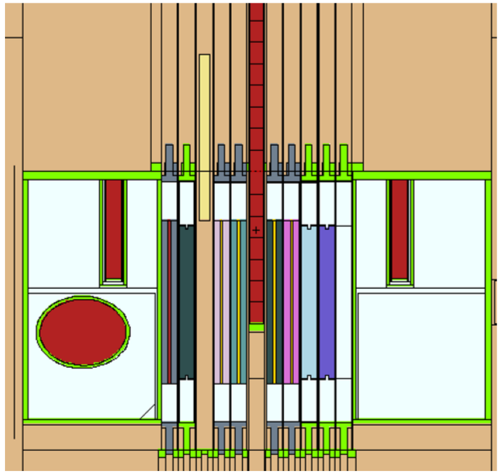
➢ A.Zenoni et al., Rev. Sc. Inst. 88, 113304 (2017);

Measured quantities: multi-scale approach

MECHANICAL TESTS
- Uniaxial Tensile test
- Compression set test

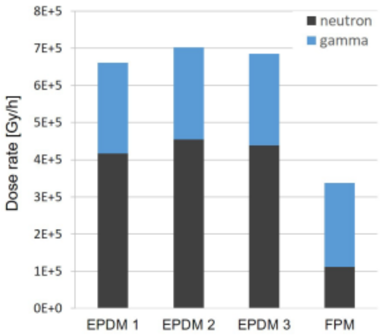
STRUCTURAL
- Swelling test
- DSC calorimetry
- DMTA
- Density

FUNCTIONAL
- Leakage test
- FEM analysis



Montecarlo simulations

Compression set test



Dose simulation

Swelling test

Tensile test

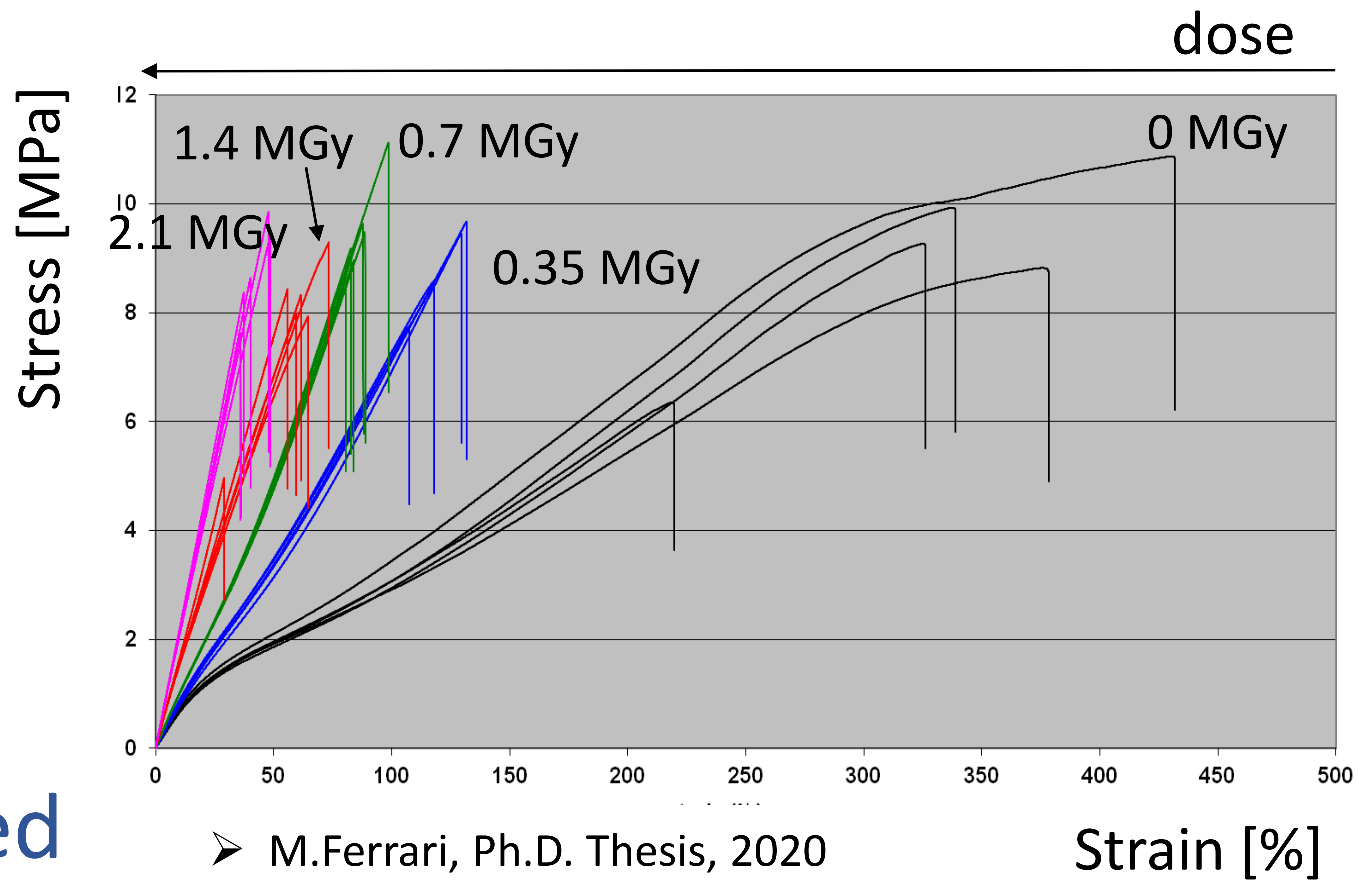
Irradiated EPDM: STRESS vs STRAIN curve

DOSE

- neutron 64%
- gamma 36%

Dose rate

- 0.7 MGy/h



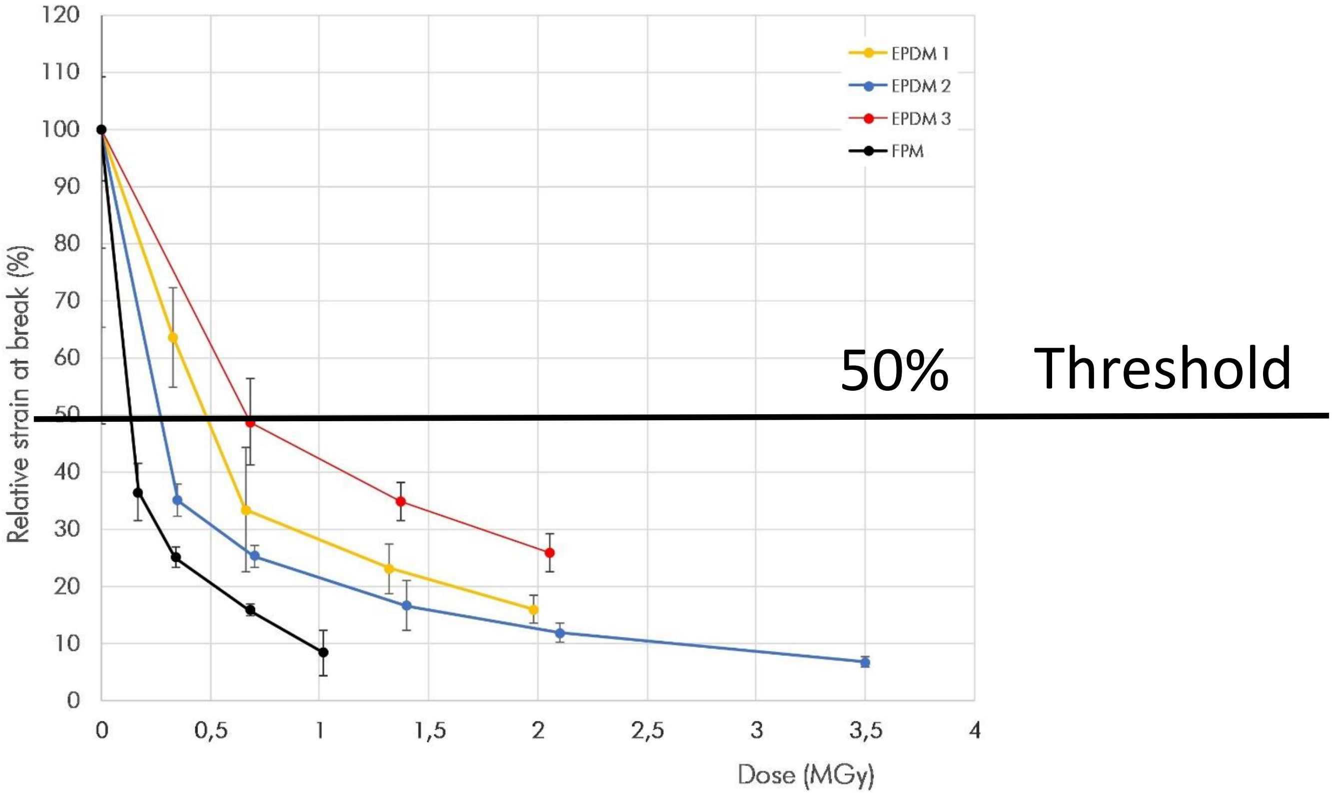
Tensile machine

- M.Ferrari, Ph.D. Thesis, 2020

Radiation-induced material hardening

Strain at break: comparison between materials

0 MGy 2.1 MGy



Radiation-induced embrittlement

Limited mobility of molecular chain

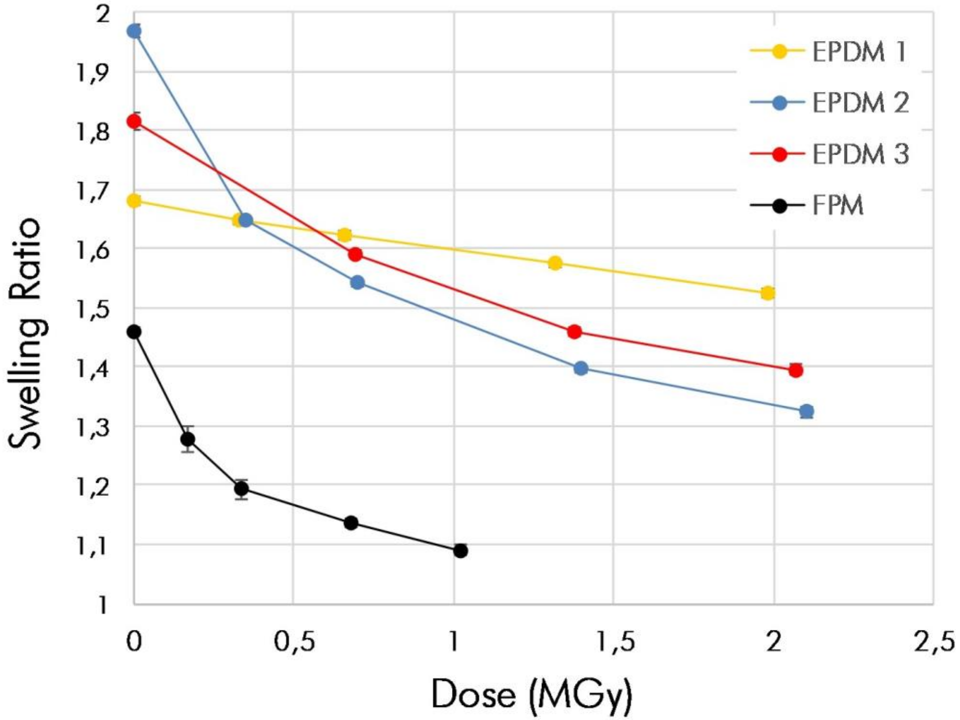
Swelling Test: different EPDMs

➢ M.Ferrari, Ph.D. Thesis, 2020

Swelling Ratio - Definition

Equilibrium swelling ratio (in solvent)

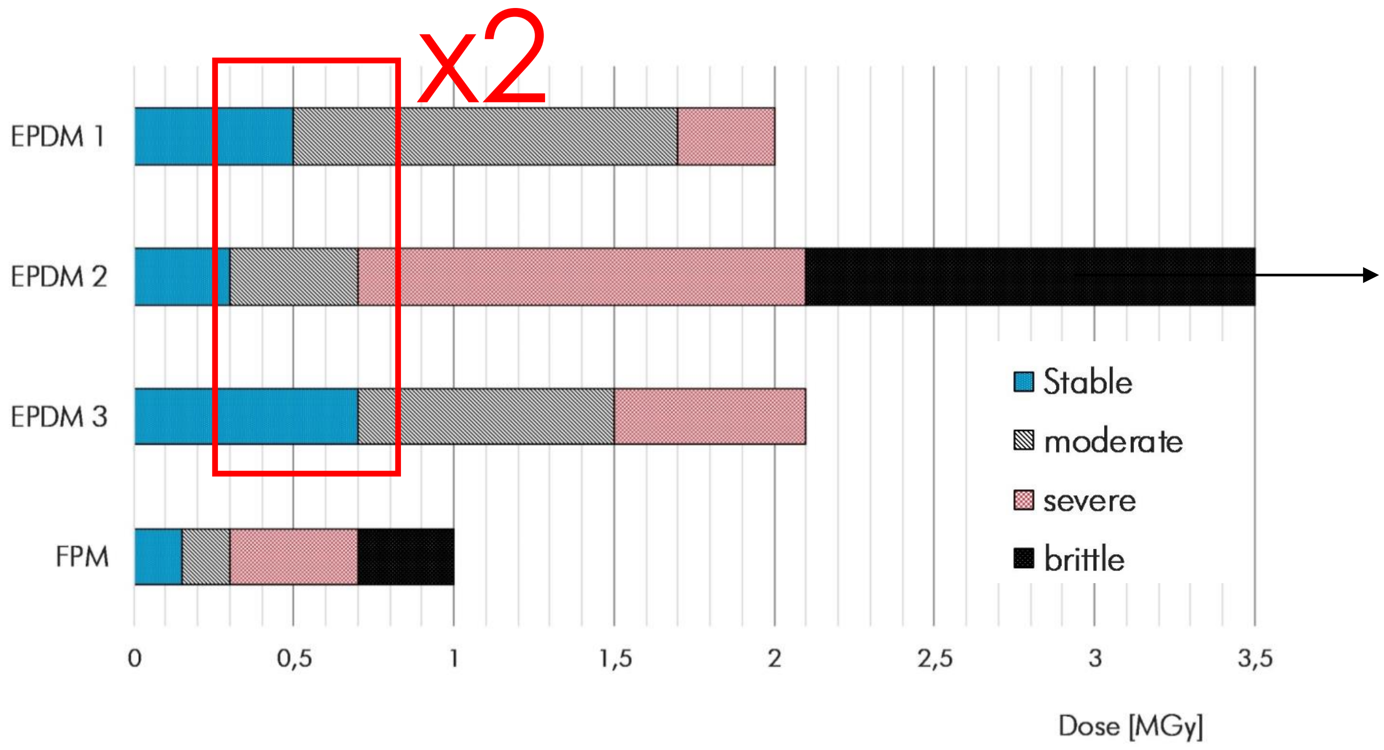
- ✓ SR $= \frac{m\ swollen}{m\ dry}$
- ✓ SR increase: cross-linking is the prevalent radiation-induced effect



Radiation-induced cross-linking

SR well correlated with mechanical evolution

Results: general dose thresholds and endpoints

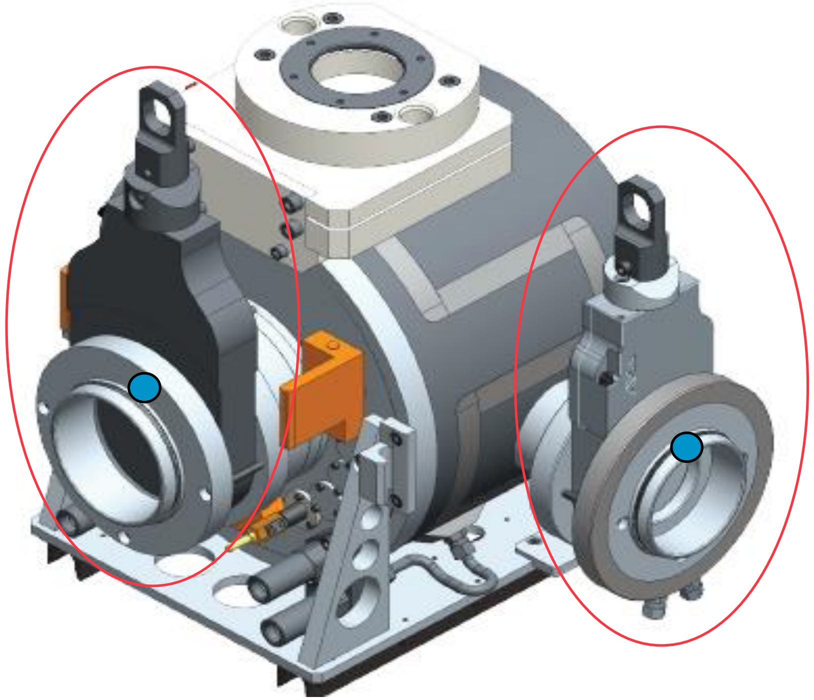


- M.Ferrari, Ph.D. Thesis, 2020

Do all 'EPDM-based' materials have the same radiation resistance?

Commercial EPDMs: large differences

Application #1: gate valve of SPES facility



CASE STUDY in a specific set of operation conditions

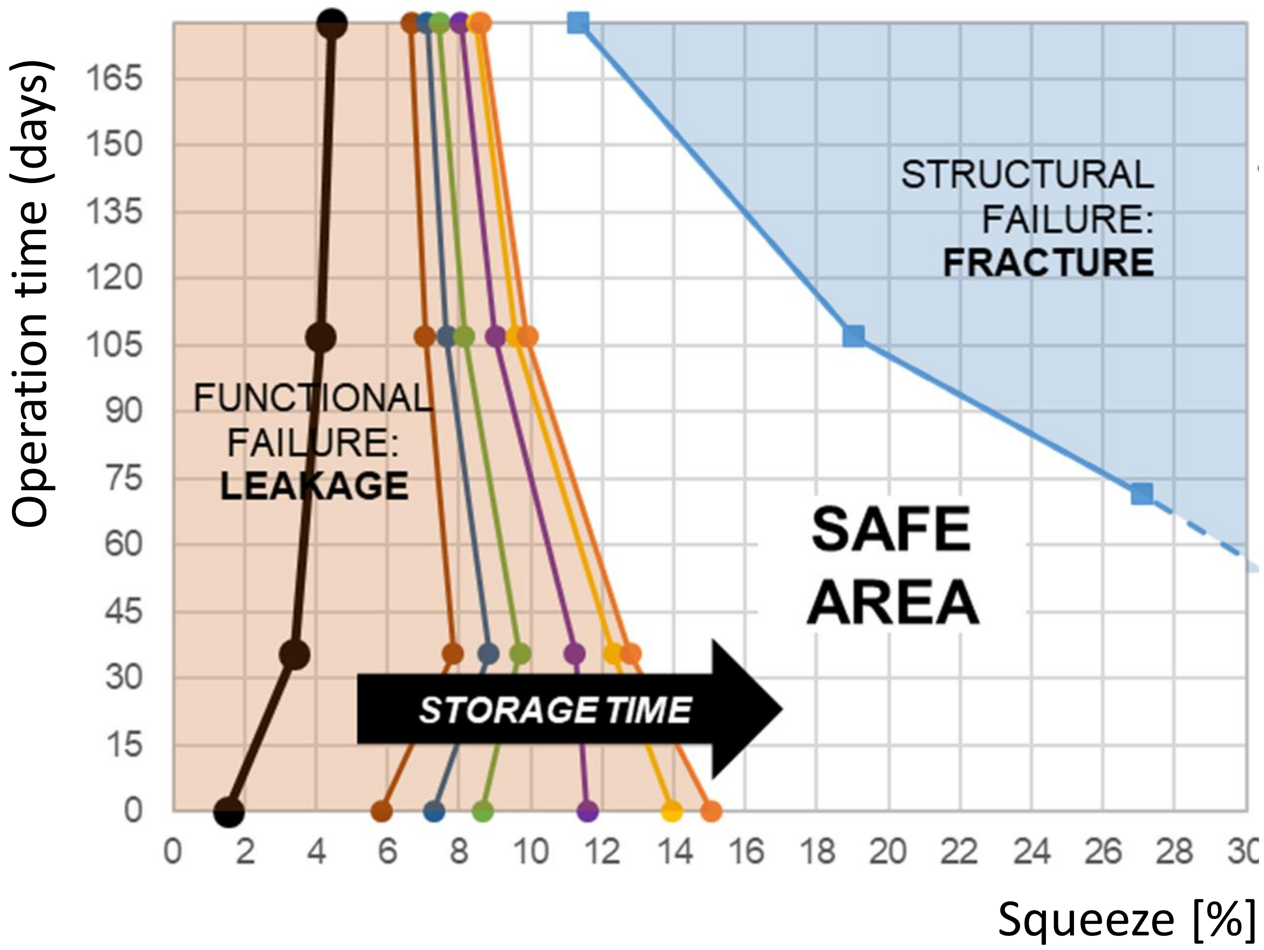
- Dynamic use
- 0.7 MGy expected (15 days)
- In vacuum
- Max temperature 85°C
- 5 more years of operation (sealing) after use

Can these conditions be replicated in a testing irradiation facility?

➢ D.Battini et al., Materials and Design vol. 156, 514-527 (2018)

System with an elastomeric O-ring with critical function

Prediction map of degradation in operation



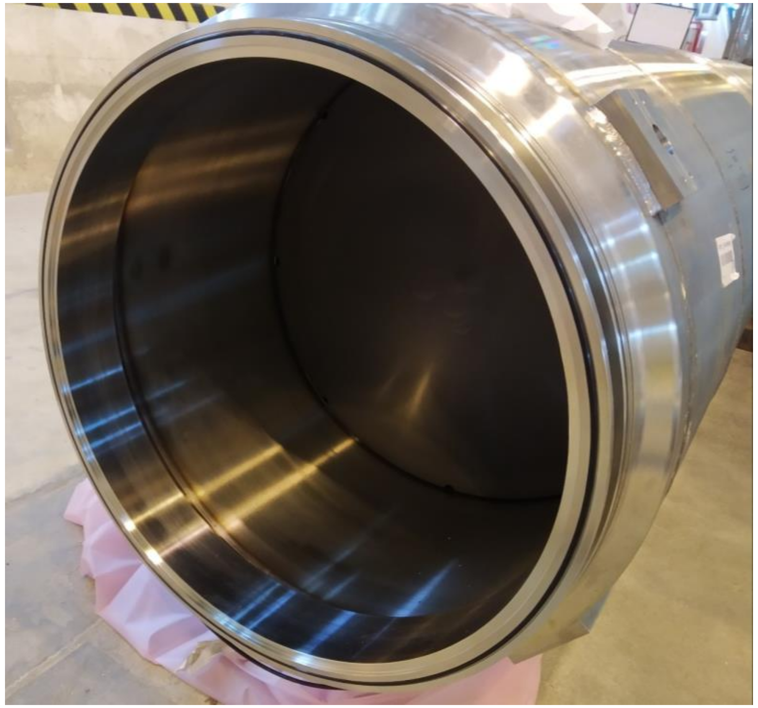
➢ D.Battini et al., Materials and Design vol. 156, 514-527 (2018)

CONSIDERED PARAMETERS:

- Dose in service;
- Squeeze degree;
- post-irradiation storage

Identification of safe usability areas for a specific use

Application #2: O-ring of LHC dump



CONSIDERED CRITERIA:

- **Dose**: 0.12 MGy expected (several years)
- **Maintenance**: impossible/very difficult
- **Failure impact**: accelerator shutdown

Can the LHC dump lifetime be affected by radiation damage on O-rings?

➢ J. Maestre et al., *JINST*, Vol. 16, P11019, (2021).

System with an elastomeric O-ring with critical function

Conclusions

Take-home message

MAIN POINTS:

- Despite their sensitivity, n**on-metallic materials** (such as lubricants, O-rings…) are fundamental for the development of integrated systems and complex devices for several high-radiation applications.
- Radiation damage: **scission and/or recombination** of long molecular chains.
- How to select a radiation tolerant material: **tests** in a well-defined set of irradiation conditions are necessary. **New data need to be produced**.
- **Commercial materials** do not generally have the same properties of base polymers! Specific commercial materials need to be considered.
- Radiation resistant materials typically get damaged **in the MGy dose** range.
- **Multi-scale and interdisciplinary approaches** are necessary. New methodologies need to be developed.

Thanks for your attention!

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