Evaluation of Radiation Resistance for Organic Materials Used in Atomic Energy-related Facilities

Japan Atomic Energy Agency
Quantum Beam Science Directorate
Advanced Ceramic Group

Akira Idesaki, Akihiko Shimada,
Norio Morishita, Masaki Sugimoto, Masahito Yoshikawa

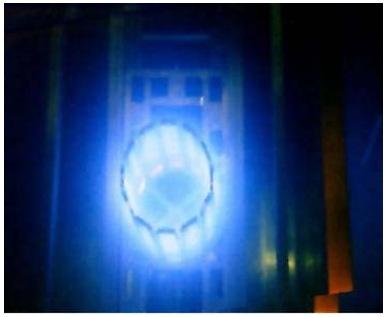
Takasaki Advanced Radiation Research Institute of JAEA

Electron beam irradiation facility

Acceleration Voltage :0.5～2.0MeV
Beam Current : 0.1～30mA

Co-60 gamma-ray irradiation facility

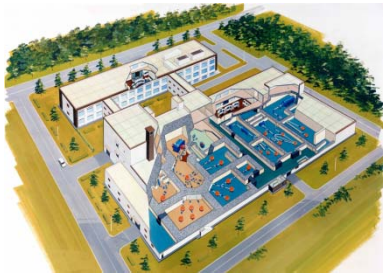
Dose rate : 0.2Gy/h ～ 20kGy/h



Ion beam irradiation facility

TIARA
(Takasaki Ion Accelerator for Advanced Radiation Application)

- ●AVF Cyclotron : H, He, Xe, Os, etc
  (～several hundred MV)
- ●3MV Tandem Accelerator : H, C, Ni, etc
- ●3MV Single-ended Accelerator : H, He, etc
- ●400kV Ion Implanter : H, Ar, etc

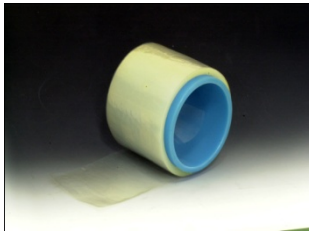


Takasaki-Daruma

Please visit ! → http://www.taka.jaea.go.jp/

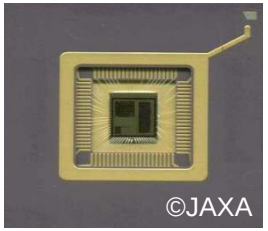
Research Activities for Quantum Beam Applications

Development of novel functional materials



Polymer electrolyte membrane for fuel cells

Silicon carbide fiber



Evaluation of semiconductor devices for space use

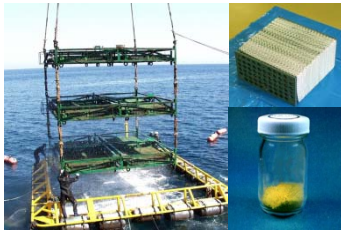
Application for biotechnology and medical use

Creation of carnations with variety of colors

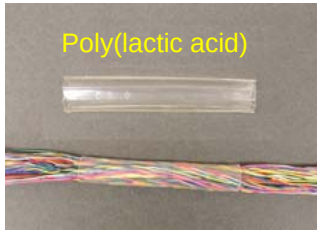
Novel DNA-repair protein

Creation of UV-resistant plants

R&D for environmental preservation and resource security technology



Fibrous adsorbent for metals



Biodegradable plastics

Decomposition of air pollutants

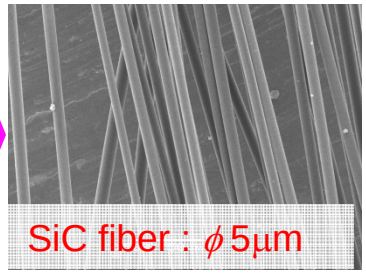
Quantum Beam Science Directorate　Advanced Ceramics Group

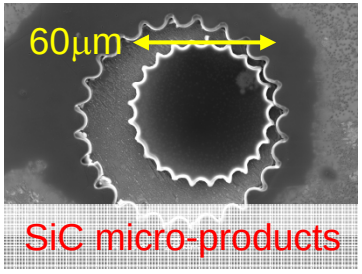
■ Fabrication of ceramic materials from Si-polymers with radiation curing

Forming : fiber, molding, etc.

Radiation curing : EB, gamma-rays

Firing : ～1200℃



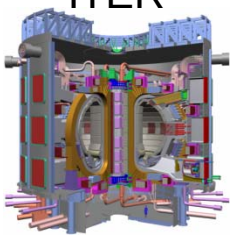


Spacecraft, Fusion reactor
Chemical plant, Microreactor

■ Evaluation of radiation resistance for organic materials used in atomic energy-related facilities

J-PARC

ITER



- Power cables
- Vacuum components
- Sensors
- Superconducting magnets etc.

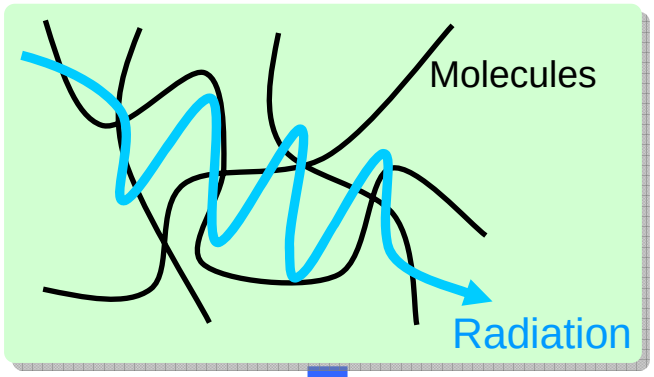
Organic materials are used as electrical insulator and structural materials.

Evaluation of radiation resistance for organic materials

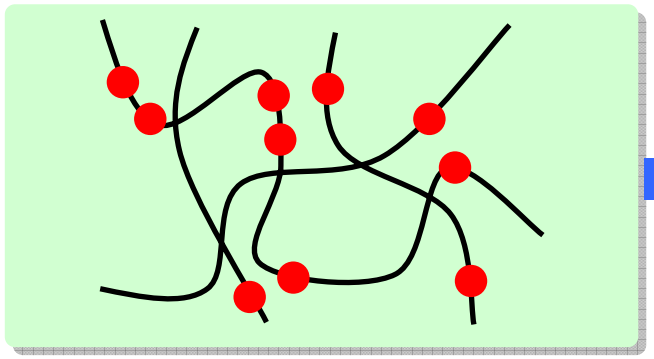
Safe and stable operation of facilities

Irradiation Effects on Organic Materials

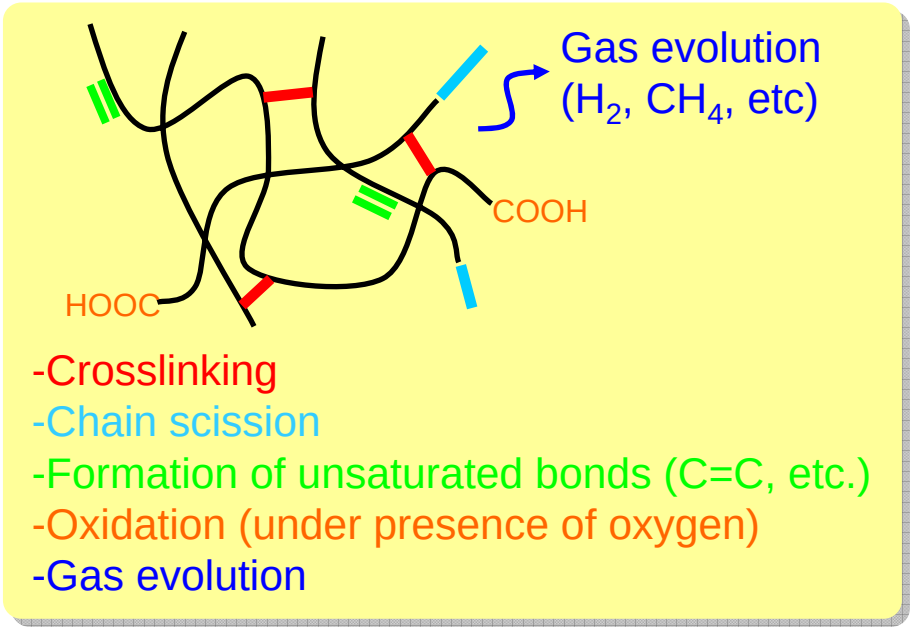
■Scheme of interaction between organic materials and radiation



Production of free radicals



Reaction of free radicals



Change of molecular structure

Modification

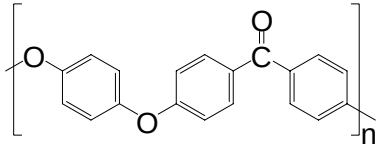
Degradation

- Irradiation temperature
- Irradiation atmosphere (presence of oxygen)
- Additives

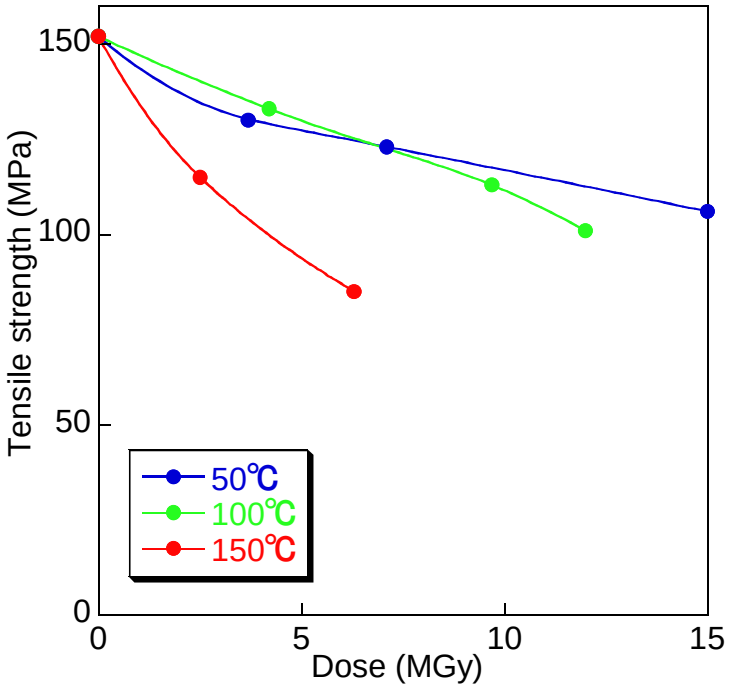
Irradiation Effects on Organic Materials

■Radiation resistance of organic materials : Effect of irradiation temperature

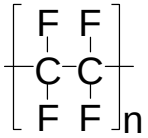
●Polyether-ether-ketone (PEEK)

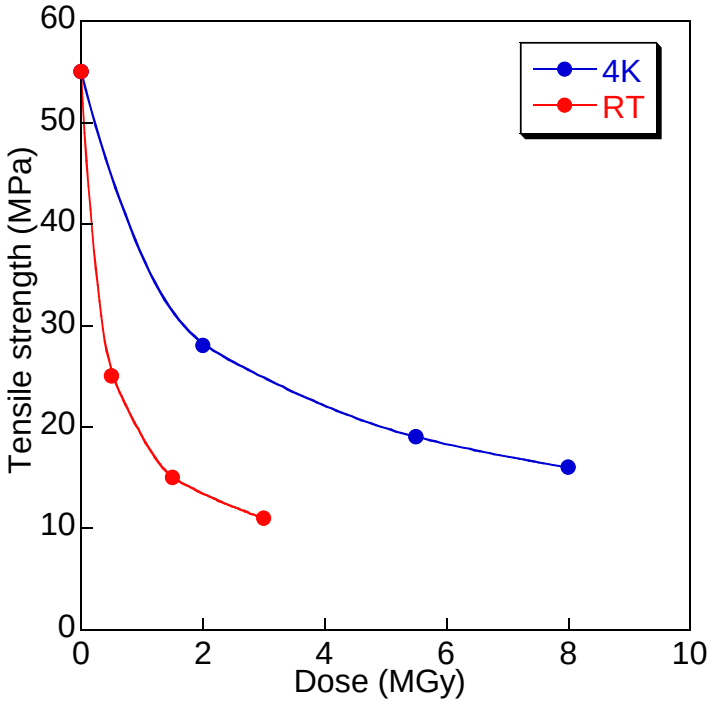


Glass transition temperature : about 140℃



●Polytetrafluoroethylene (PTFE)





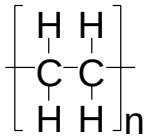
Degradation of organic materials is accelerated at high temperature.

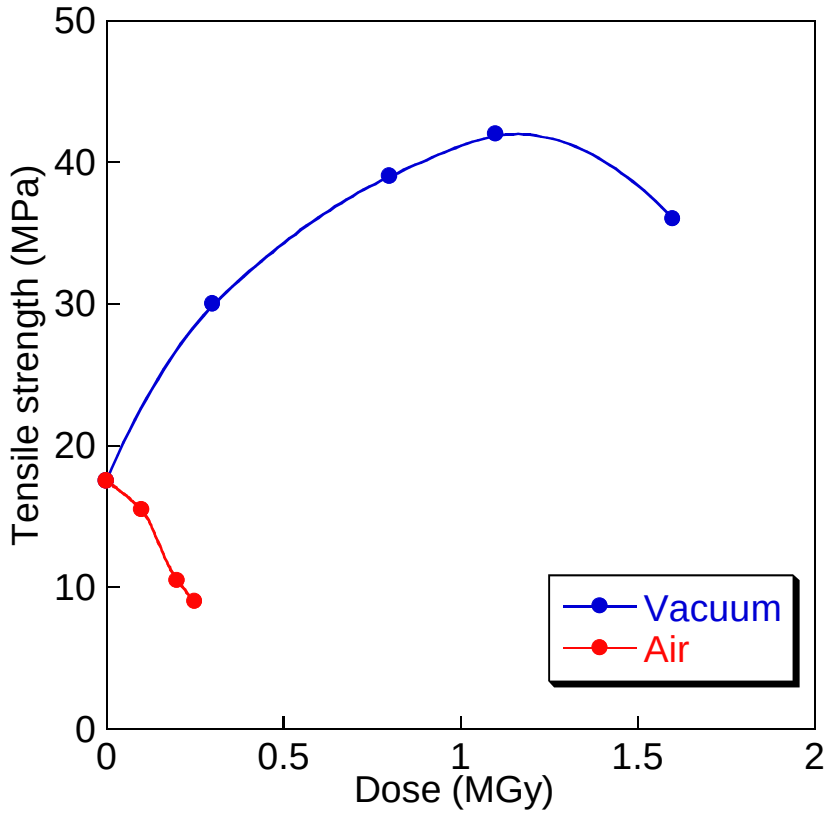
Reference : “Radiation Resistivity of Polymeric Materials with Data Tables” (in Japanese)
JAERI-Data/Code 2003-015

Irradiation Effects on Organic Materials

■Radiation resistance of organic materials : Effect of irradiation atmosphere

●Polyethylene (PE)





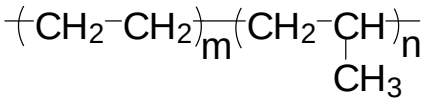
Radiation oxidation accelerates degradation of organic materials.

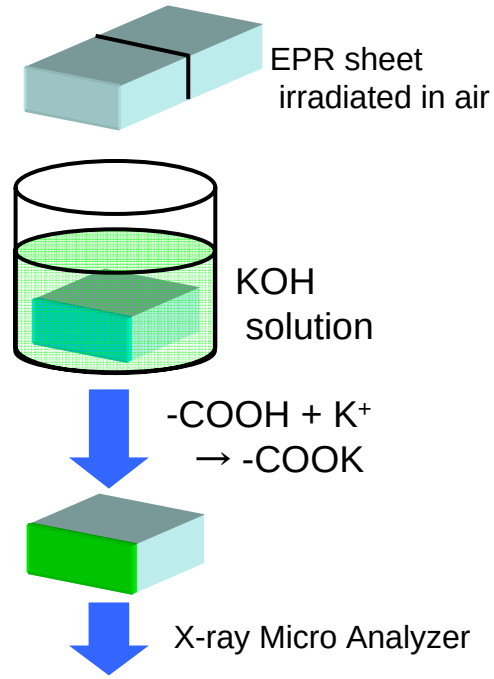
Reference : “Radiation Resistivity of Polymeric Materials with Data Tables” (in Japanese)
JAERI-Data/Code 2003-015

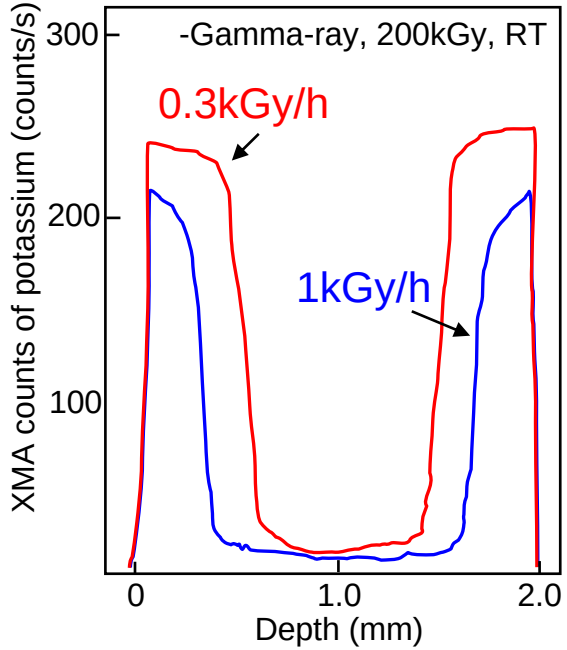
Irradiation Effects on Organic Materials

■Radiation resistance of organic materials : Radiation oxidation of organic materials

●Ethylene-propylene rubber (EPR)







Radiation oxidation is accelerated in the case of low dose rate.

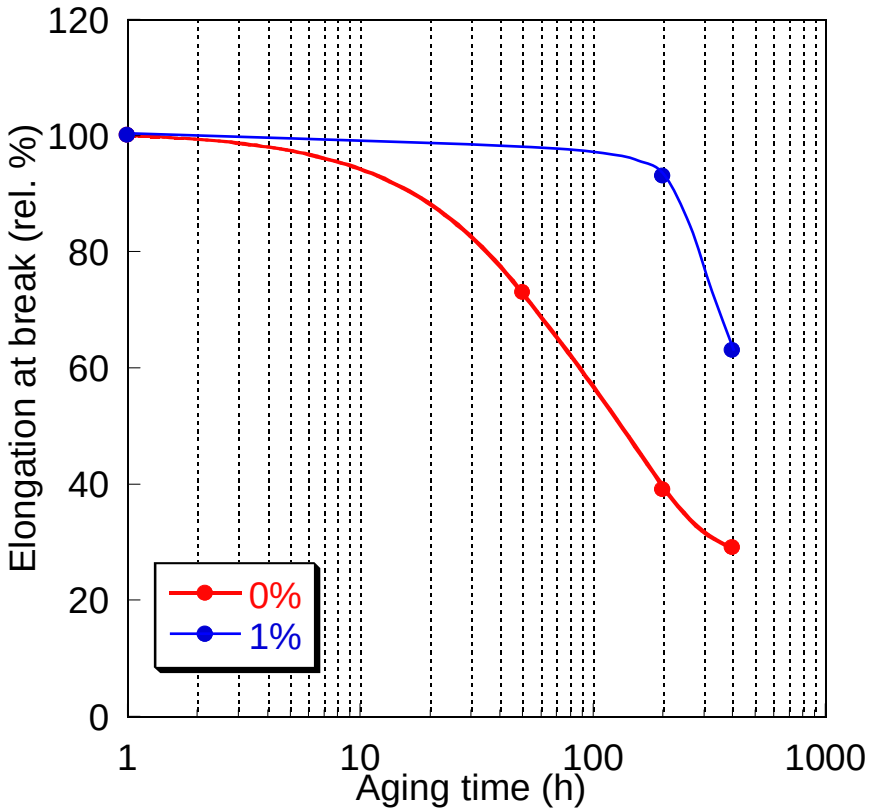
Reference : “Radiation Resistivity of Polymeric Materials with Data Tables” (in Japanese)
JAERI-Data/Code 2003-015

Irradiation Effects on Organic Materials

■Radiation resistance of organic materials :Effect of additives

●Crosslinked polyethylene (XLPE)

- XLPE with (1wt%) and without antioxidant
- Gamma-ray irradiation with dose rate of 1kGy/h in air at 100℃



Even 1wt% of antioxidant is effective for suppression of degradation.

Reference : T. Seguchi et al, *Rad. Phy. Chem*., 80(2011), pp.268-273.

Irradiation Effects on Organic Materials

■Measurements and analyses for evaluation of radiation resistance

●Chemical properties

- Electron spin resonance (ESR) : Quantification (and qualification) of free radicals
- Gas chromatography : Quantification and qualification of evolved gases
- FT-IR (Fourier-transformed infrared spectroscopy) : Analysis of chemical bonds
- GPC (Gel permeation chromatography) : Measurement of molecular weight
- Thermal analysis : Measurement of melting point, decomposition temperature
- Elasticviscosity : Measurement of glass transition temperature

●Mechanical properties

- Tensile test
- Bending test

●Electrical properties

- Breakdown voltage
- Electrical resistance

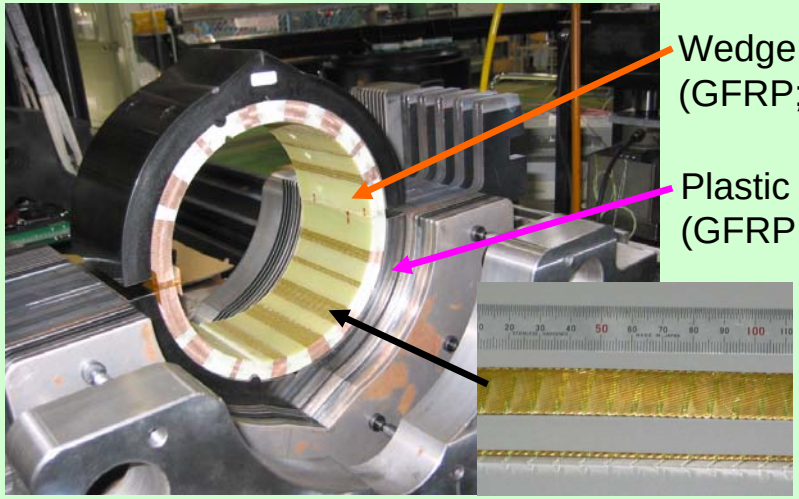
➡ Properties should be evaluated according to the purpose for use.

Evaluation under conditions close to practical conditions as possible

●Structural materials : Mechanical properties
●Electrical insulator : Electrical properties
●Materials for vacuum components : Outgas, mechanical properties
●Adhesives and paints : Detachment, coloring, outgas

Evaluation of radiation resistance for organic materials used in J-PARC

■Materials used in superconducting magnet system at J-PARC neutrino beam line



Wedge
(GFRP; Glass fiber/Epoxy resin)

Plastic spacer
(GFRP; Glass fiber/Phenolic resin)

Insulation of superconducting coil
(Film; Polyimide/Epoxy resin)

●Service condition:
- Temperature of liquid helium(4K)
- Irradiation of γ-rays and neutron
  Dose rate : 30kGy/year
  Operating time : 4000h/year

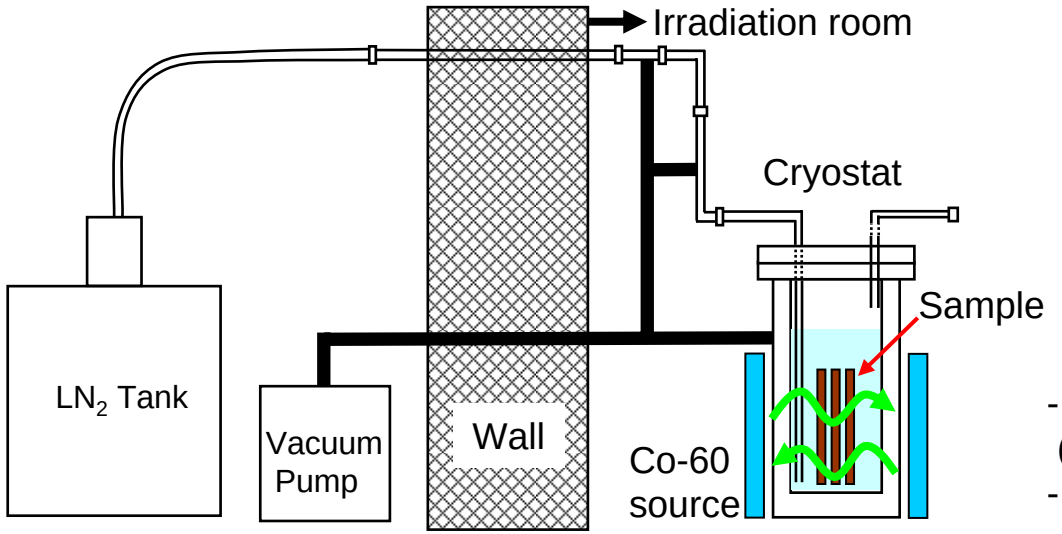
Gamma-ray irradiation at temperature of liquid nitrogen (77K)

Evaluation of radiation resistance
- Mechanical properties
- Behavior of gas evolution

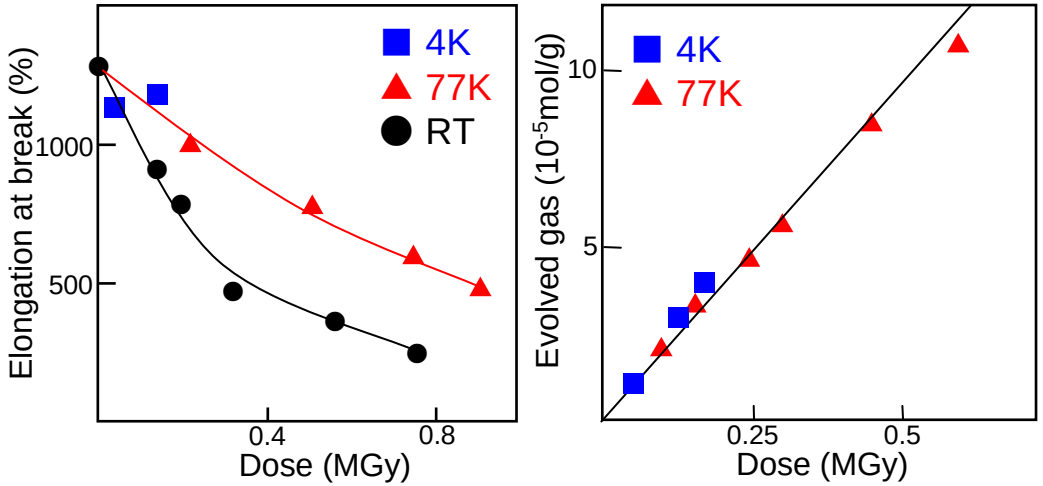
Reference : A. Idesaki et al, *Advances in Cryogenic Engineering*, Vol. 52, 2006, pp.330-334.
T. Nakamoto et al, *Advances in Cryogenic Engineering*, Vol. 52, 2006, pp.225-232.

Evaluation of radiation resistance for organic materials used in J-PARC

■Low temperature irradiation vessel



- Gamma-ray irradiation at 77K (4K is not available…)
- Dose rate : ～30kGy/h



Effect of irradiation temperature on tensile property and gas evolution.
(High Density Polyethylene; HDPE)

Difference between 4K and 77K is almost negligible.

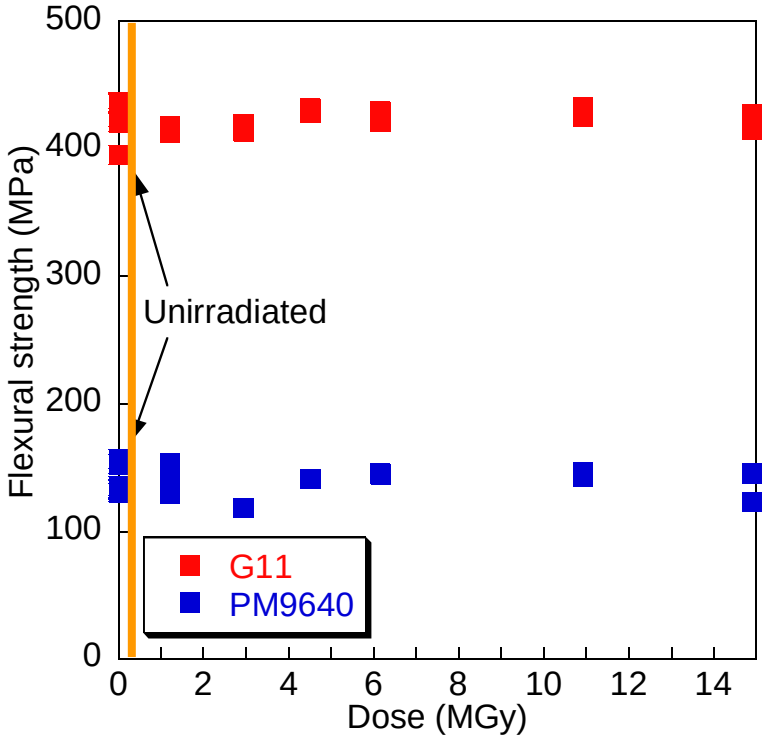
Condition of 4K can be simulated by irradiation at 77K.

Reference : H. Kudoh et al, *Rad. Phy. Chem.*, 48(1996), pp.89-93.

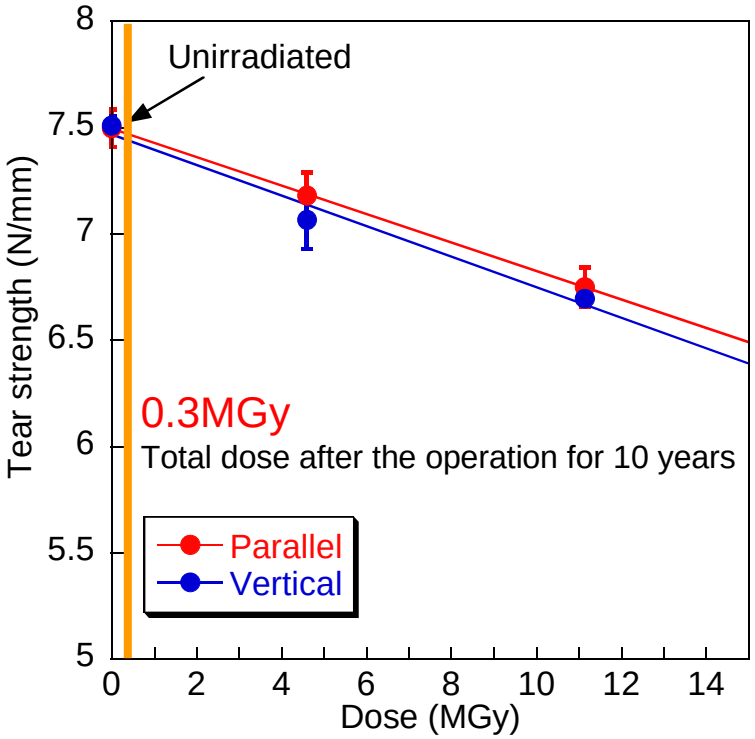
Evaluation of radiation resistance for organic materials used in J-PARC

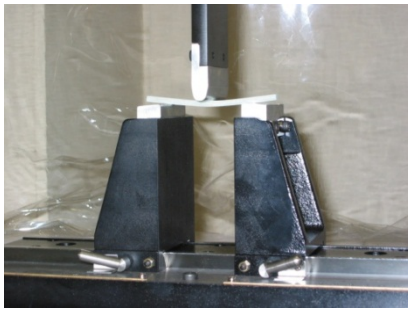
■Mechanical tests after γ-ray irradiation at 77K

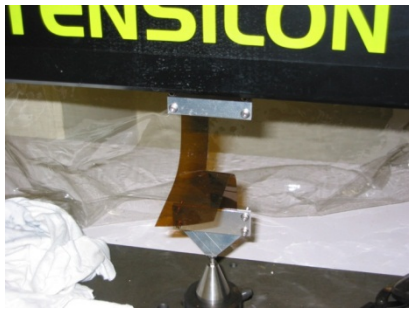
●Three-point bending test for GFRP
- Glass fiber/Epoxy resin (G11)
- Glass fiber/Phenolic resin (PM9640)



●Tear test for film
- Polyimide/Epoxy resin



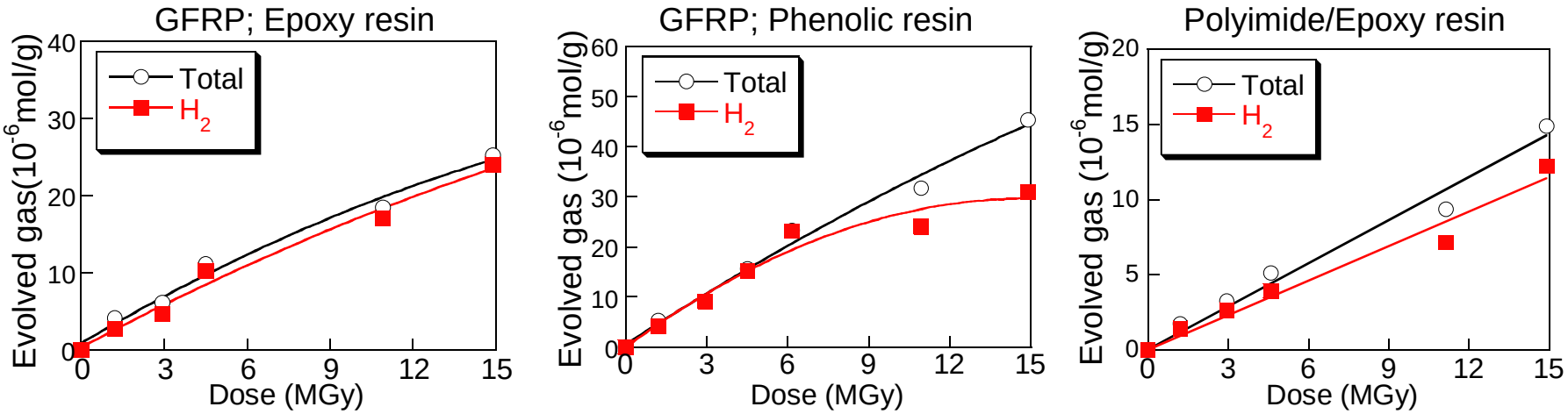




The materials show sufficient radiation resistance.

Evaluation of radiation resistance for organic materials used in J-PARC

■Gas analysis after γ-ray irradiation at 77K



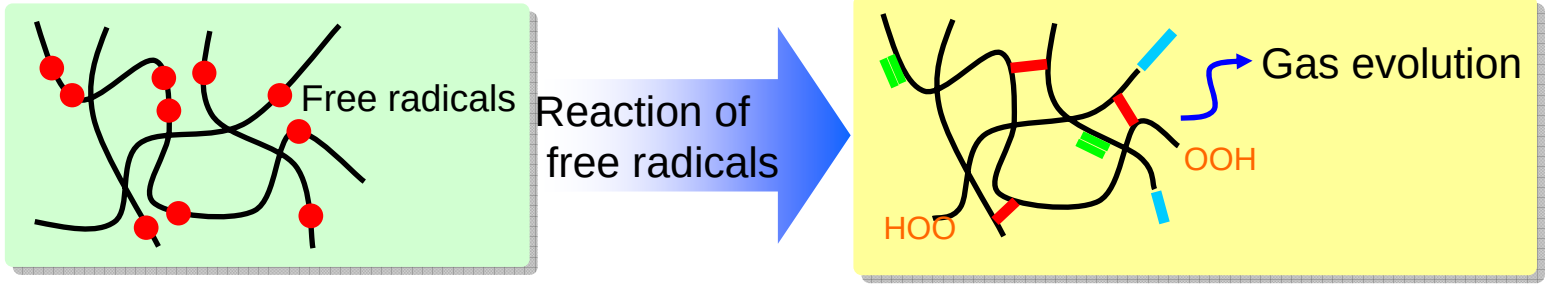
- Hydrogen should be removed to keep stable operation of the magnet system.
- Estimation of the amount of hydrogen from the whole magnet systems (28 magnets)

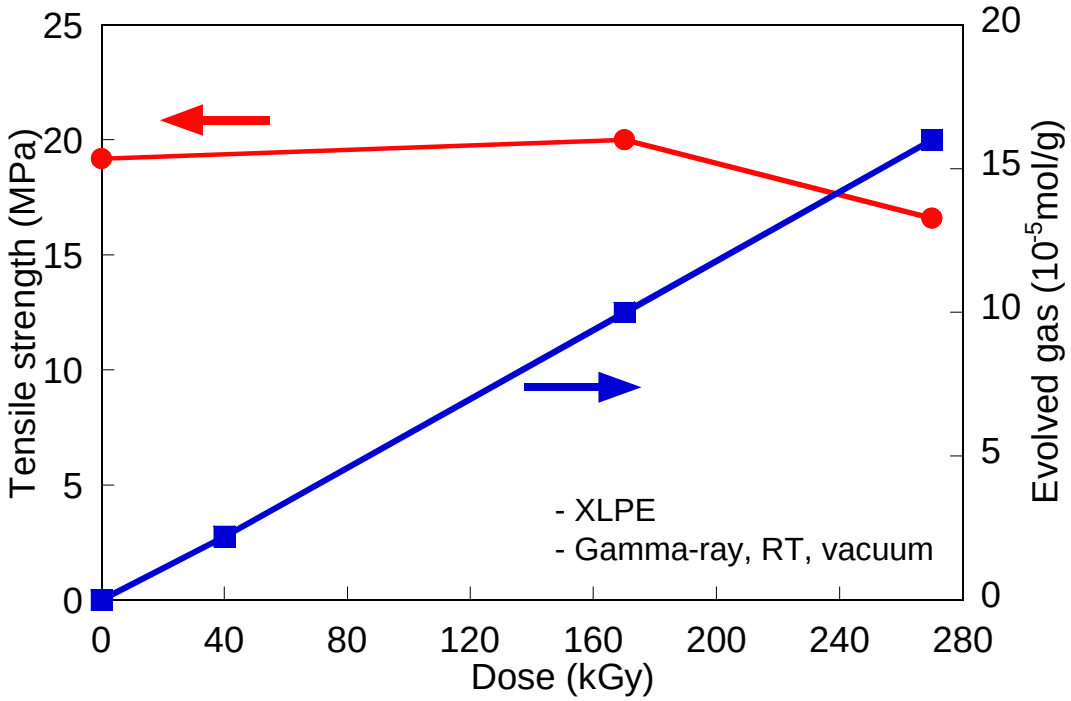
→ 0.37mol/year of hydrogen (0.01L/year as liquid hydrogen)
= Negligible for the capacity of hydrogen-absorber

Sufficient radiation resistance of organic materials was proved.

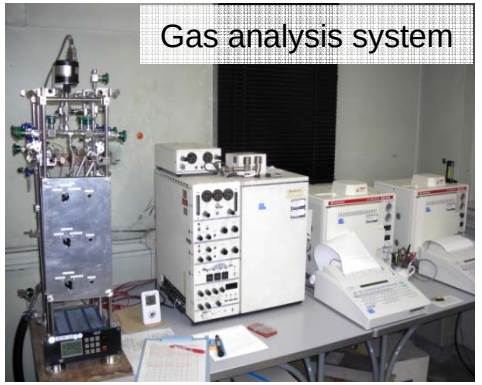
Beam supply has started since Jan 2010.

Gas analysis for evaluation of radiation resistance





Change of tensile strength and the amount of evolved gases by γ-ray irradiation for XLPE.

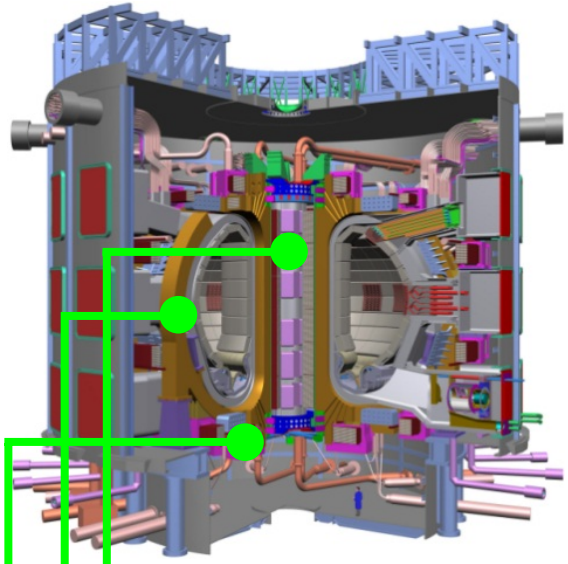


Gases evolved from organic materials by irradiation can be detected with low dose where deterioration of mechanical property is negligible.

Gas analysis is a good tool for:
- Selection of materials
- Evaluation of radiation resistance

Selection of organic materials used as electrical insulator for ITER superconducting coil

■Fabrication of electrical insulator for ITER superconducting coil

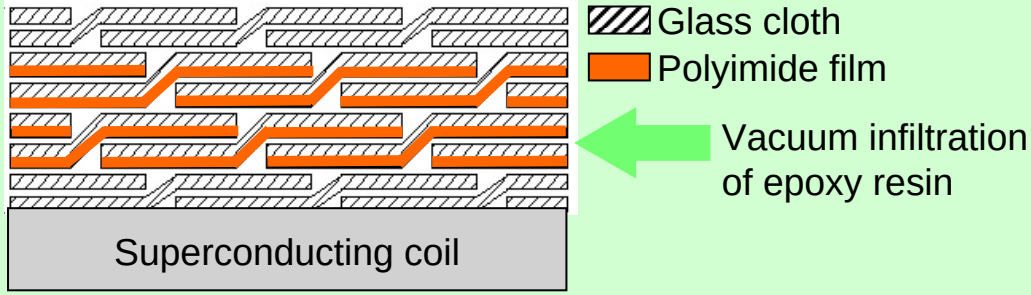


Central Solenoid (CS) Coil
for Ignition of Plasma

Toroidal Field (TF) Coil
for Confining of Plasma

Poroidal Field (PF) Coil
for Adjustment of Position
and Shape of Plasma

Glass cloth/Polyimide/Epoxy resin



●Required properties for epoxy resin
- Viscosity : 200mPa･s at 60℃
- Pot life time : 40h (time to increase the viscosity)
- Radiation resistance : 10MGy

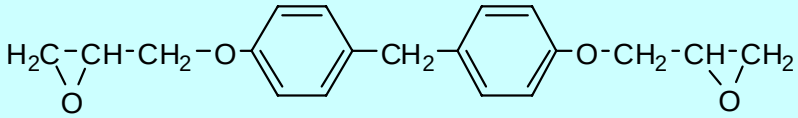
●Candidates
-Epoxy resin (DGEBF) + Cyanate ester (CE)
-Epoxy resin (DGEBF) + Epoxy resin (TGDDM)

Selection of organic materials used as electrical insulator for ITER superconducting coil

■Candidates of electrical insulator for ITER superconducting coil

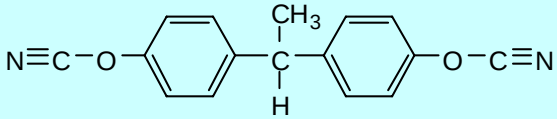
●DGEBF

Di-Glycidyl Ether of Bisphenol F



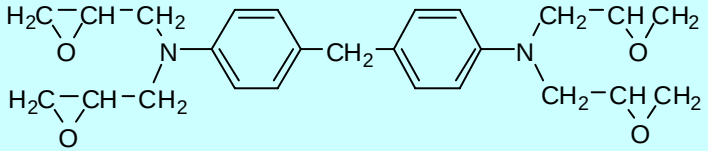
●CE

CyanateEster

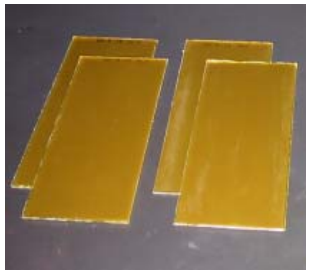


●TGDDM

Tetra-Glycidyl-Diamino-DiphenylMethane



Glass cloth/Polyimide/Epoxy resin



Resin of

●Sample A : DGEBF + CE

●Sample B : DGEBF + TGDDM

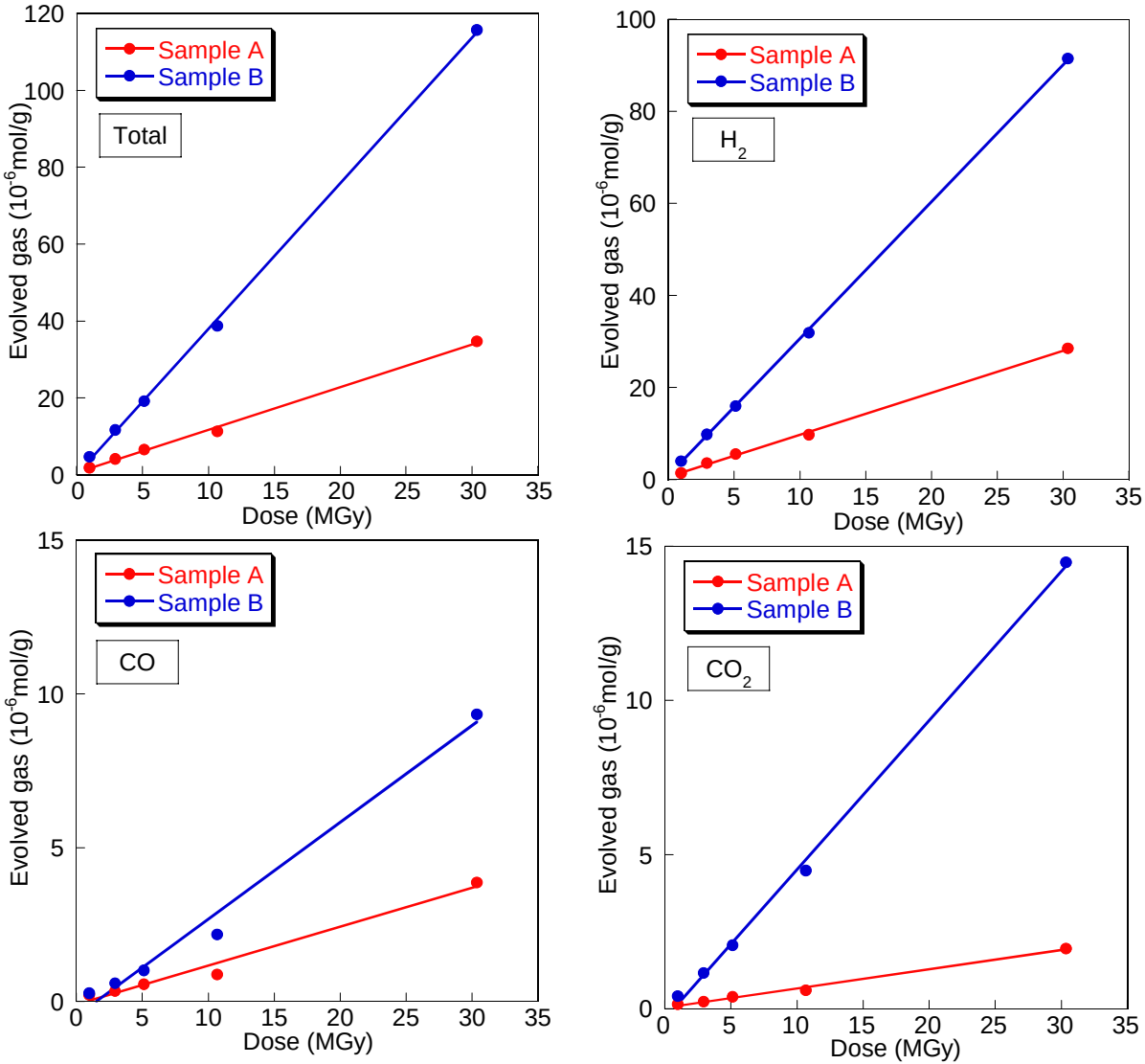
Gamma-ray irradiation at 77K

●Selection of insulation material

- Gas analysis
- Interlaminer shear strength

Selection of organic materials used as electrical insulator for ITER superconducting coil

■Gas analysis after γ-ray irradiation at 77K



Sample A : DGEBF + CE
Sample B : DGEBF + TGDDM

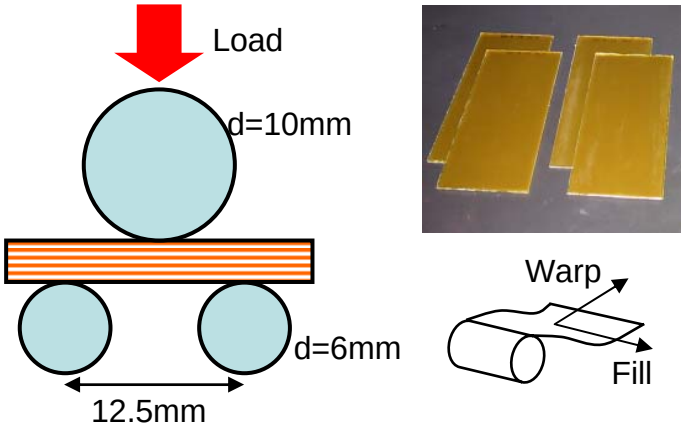
●Main components of the evolved gas : H_2, CO, CO_2
●Radiation resistance of Sample A is superior to that of Sample B.

Evolved gases from laminated materials by γ-ray irradiation at 77K

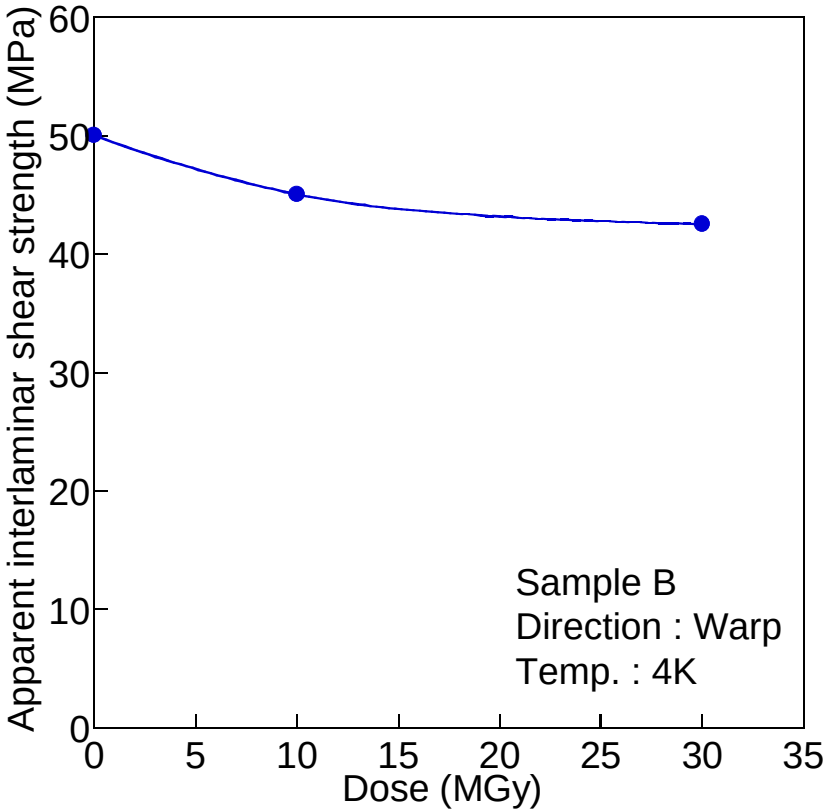
Reference : A. Idesaki et al, *Advances in Cryogenic Engineering*, Vol. 54, 2008, pp.169-173 .

Selection of organic materials used as electrical insulator for ITER superconducting coil

■Mechanical tests after γ-ray irradiation at 77K



Scheme of short-beam shear test.



Apparent interlaminar shear strength of the irradiated Sample B (warp specimens).

Sample A (Epoxy resin + Cyanate ester) was selected as electrical insulator for ITER superconducting coil.

Reference : Y. Shindo et al, *Cryogenics*, 50, 2010, pp. 36-42.

Development of organic material with high radiation resistance

■Irradiation effect on epoxy resins with different hardeners
(Collaboration with University of Hyogo (Prof. Kishi) and KEK)

Epoxy/Cyanate ester resin showed high radiation resistance.
・・・・・The reason has not been clarified.

Fabrication process of epoxy resin

Epoxy resin + hardener + (catalyst) → Heat treatment

- Acid anhydrides
- Amines
- Phenols
- Cyanate esters

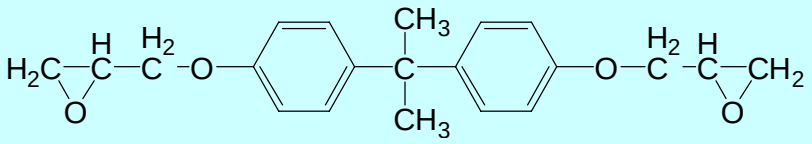
Relationship between chemical structure of hardener and radiation resistance

Development of organic material with high radiation resistance

■Examined materials

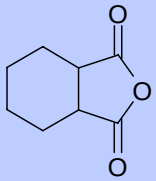
●Epoxy resin

DGEBA
Di-Glycidyl Ether of Bisphenol A

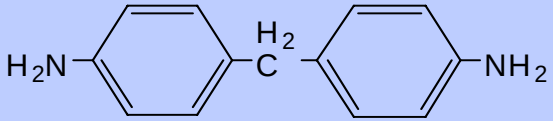


●Hardeners

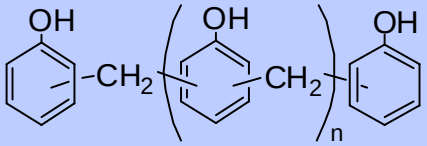
-Acid anhydride
HHPA (Hexahydrophthalic anhydride)



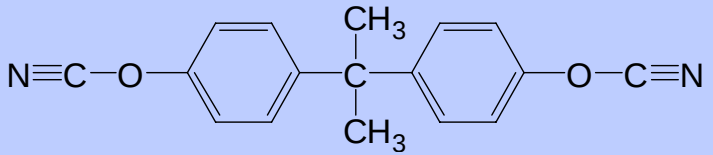
-Amines
DDM (Diaminodiphenylmethane)



-Phenols
PN (Phenolnovolac resin)

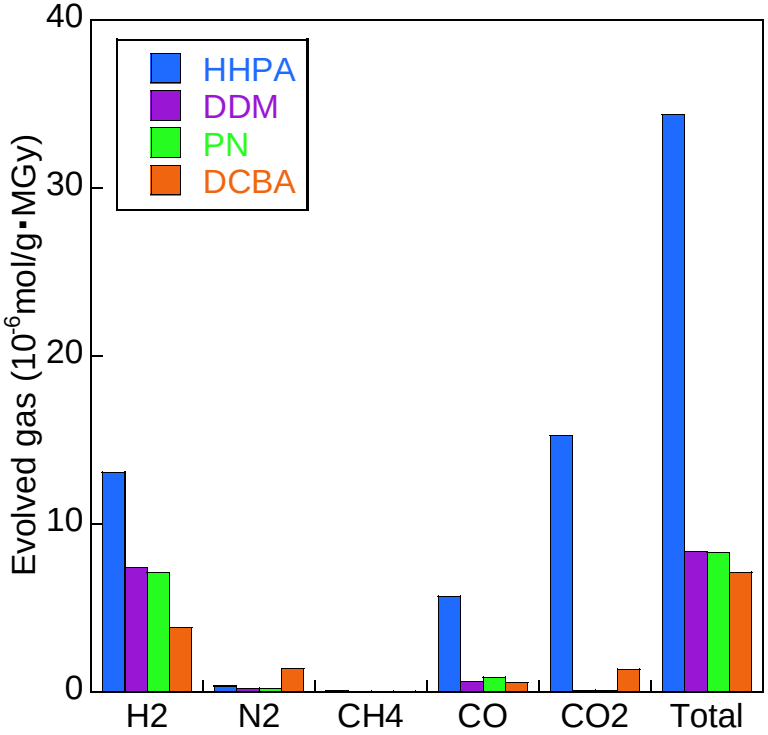


-Cyanate esters
DCBA (Di-Cyanate ester of Bisphenol A)



Development of organic material with high radiation resistance

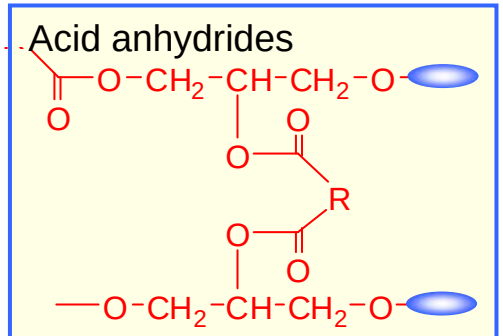
■Gas analysis after γ-ray irradiation



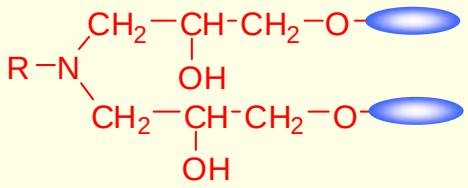
●Crosslinking structure is collapsed by radiation.
●Hardeners which include benzene ring and/or C-N bonds suppresses gas evolution.
●Cyanater esters, which forms very complex crosslinking structure, leads the highest radiation resistance.

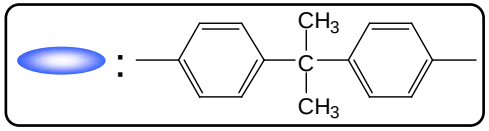
Crosslinking structure between epoxy and hardener

Acid anhydrides

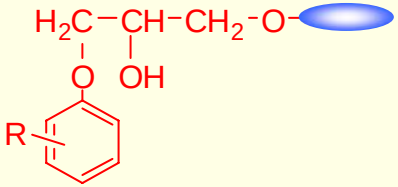


Amins

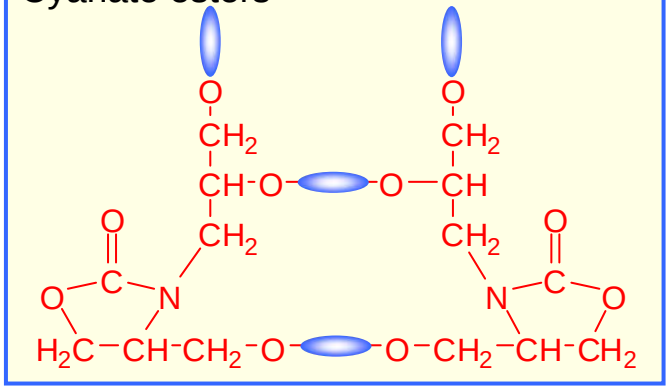




Phenols



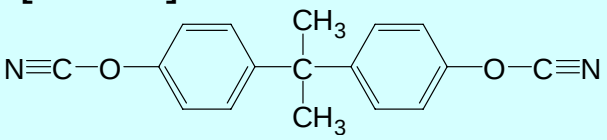
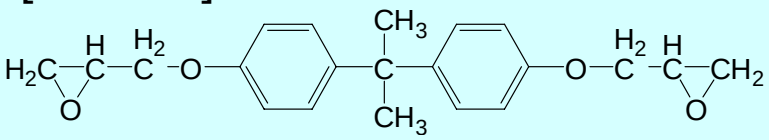
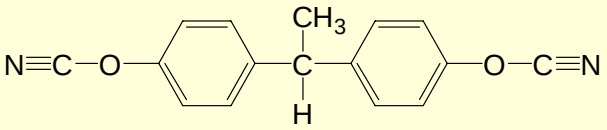
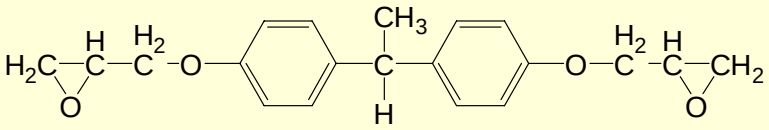
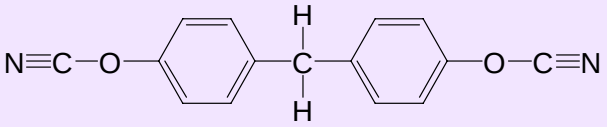
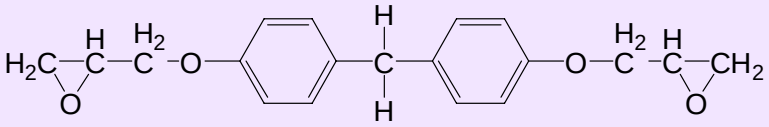
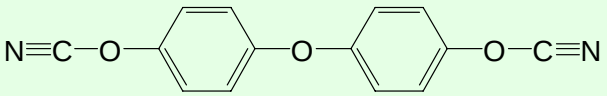
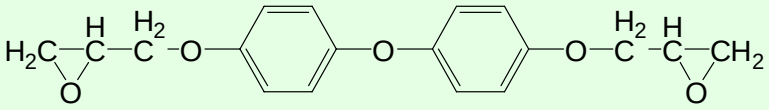
Cyanate esters



Bond-dissociation energy

	(kJ/mol)
C-H	334.7
O-H	424.4
C-C	599
O-N	626.8
C-N	745

Development of organic material with high radiation resistance

	Cyanate	Epoxy
Bis-A type	[DCBA] 	[DGEBA] 
Bis-E type	[DCBE] 	[DGEBE] 
Bis-F type	[DCBF] 	[DGEBF] 
Diphenyl ether type	[DCDPE] 	[DGEDPE] 

Development of Epoxy/Cyanate ester resin with higher radiation resistance

- Combination
- Composition

→ J-PARC, CERN LHC Upgrade, etc.

Summary

- The performance of instruments used in atomic energy-related facilities depends on the radiation resistance of organic materials used.

- Radiation degradation of organic materials depends on temperature, atmosphere and additives. Especially, much attention should be paid to the dose rate in case of evaluation under presence of oxygen.

- It is important to evaluate the materials under the conditions close to practical conditions as possible.

- Gas analysis can be a good tool for selection of materials.

- A novel epoxy/cyanate ester resin is under development in order to put it to practical use for J-PARC, CERN LHC Upgrade, etc.