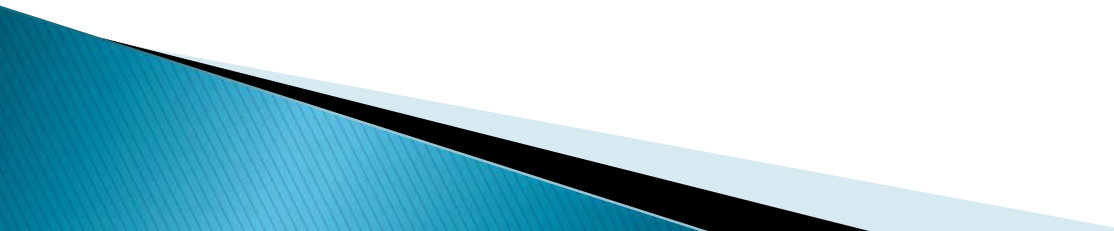


Applications of Radiation Processing in Japan -The History and Future Prospects-

Masakazu Washio

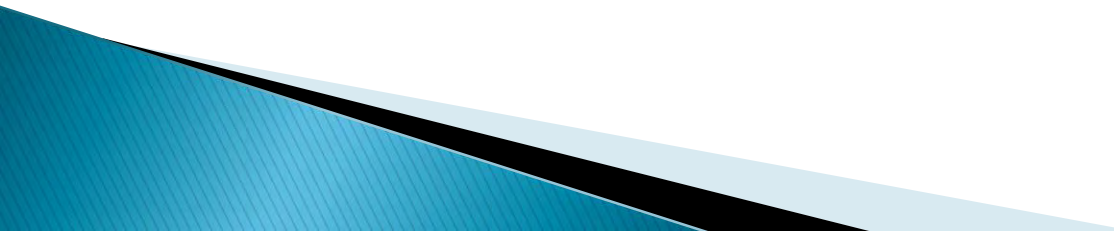
Cooperative Major in Nuclear Energy,
Graduate School of Advanced Science and Engineering,
Faculty of Science and Engineering,
Waseda University



Preface

In this talk, I would like to give the present status of radiation processing in Japan as objective looks as possible to the social implementation, which were accumulated by many stakeholders.

And finally I would like to show the expectations and prospects for the future of Radiation Processing.



Presentation Outline

- Overview of the History of Radiation Application and Recent Economic Scale
- **Processing overviews for Electron, X-ray(γ -ray), Ion Beam**
 - ✓ **Electron beam applications**
 - ✓ **X-ray(γ -ray) applications**
 - ✓ **Ion beam applications**
- **Future Prospects of the Radiation Processing**

Overviews of the History of Radiation Applications

1895	W. C. Röntgen	Discovery of X-ray
1897	J. J. Thomson	Discovery of electron
1898	A. Rutherford	Discovery of α -ray
1898	M. Curie	Discovery of Ra and Po
1911	A. Rutherford	Proposal of the Model of Atoms
1931	E. O. Lawrence, D. H. Sloane	Success of Linear Acc. Operation
1932	E. O. Lawrence	Invention of Cyclotron
1932	J. Cockcroft & E. Walton	Success of Electro-static Acc.
1932	J. Chadwick	Discovery of Neutron
1932	C. Anderson	Discovery of Positron
1933	R. J. Van de Graaff	Invention of Van de Graaff Acc.
1935	H. Yukawa	Meson theory
1948	M. Dole	Literature of Radiation-induced Crosslinking
1952	A. Cherslby	First Achievement of Radiation-induced Crosslinking of PE
1956	Ethicon Co.	EB Sterilization of Catgut sutures
1957	TE Connectivity (Raychem)	Development of Polyethylene Heat Shrinkable Tubing
1960	GE	Crosslinked Polyethylene Tape
1961	SEI	Manufacture of Electron Irradiated Polyethylene Wire, and Heat Shrinkable Tubing

Overviews of the History of Radiation Applications (2)

- 1963 **Establishment of Japan Atomic Energy Research Institute (JAERI)**
Takasaki Establishment (Present QST) Center of Radiation
Application in Asia
- 1969 **First Gamma Irradiation Contracted facility @Tochigi**
- 1970 **Approval of γ -sterilization of Disposable syringe & Needle**
- 1970~ **Production started for Tire, Form Plastics, and Curing for release coatings by using EB**
- 1971 **Radiation Sterilization was listed in the Eighth Revised Japanese Pharmacopoeia**
- 1972 **Approval of potato sprouting Control by γ -irradiation**
- 1973 **Nihon Medi-Physics Co., Ltd. Established. Radiopharmaceutical Supply started in Japan**
- 1977 **The first IMRP (@ Puerto Rico)**
- 1980~ **Declaration of Food irradiation soundness by FAO/IAEA/WHO (<10kGy)**
- 1985 **First Cyclotron application service @Ehime**
- 1986~ **Practical Use stated for Food Irradiation by EB in French**
First Production of Floppy Disc by EB processing by TDK (Production Terminated)
- 1989 **First Contracted EB Irradiation Facility @Tsukuba**

Overviews of the History of Radiation Applications (3)

- 1990~ Practical Realization of various kinds of EB Application**
Wire Harness, Tunnel interior plate, introduction of defect to Semiconductor, Degradation of PTFE, Battery Separator, Grafting Membranes, Bio-degradable Polymer
- 1991 SiC fiber by EB Crosslinking (Hi-Nicalon™ TypeS) Market launched**
- 1991 First Approval of EB Sterilization for Medical Devices by Ministry of Health and Welfare**
- 1992 γ -ray processing for the Lymphocyte Activity Control of Blood Products**
- 1997 Abolition of Upper Limit(10kGy) for Food Irradiation by WHO(<75kGy)**
- 2000~ VOC, Dioxin Elimination Tests by EB**
- 2002 PET Insurance Application (for Glucose Metabolism Diagnostic Agent)**
- 2006 First Approval of EB sterilization for Pharmaceutical(Eye drops)**
- 2007 Ultra-low Energy EB (50kV) Machine for Production Market launched**
- 2010 First **Inline EB Sterilization for PET bottle** (S company @Kumamoto)**
- 2012 CERN found Higgs Boson finding (2013 Nobel Prize in Physics)**
- 2015 T. Kajita Awarded for the Nobel Prize in Physics by finding Neutrino Oscillation**

Medical / Health Care Use

Diagnosis by Radiation : ¥1,460bn.
Radiotherapy: ¥140 bn.
Breast Cancer Therapy : ¥20 bn.

Particle Therapy: ¥20 bn.



PET · CT Cancer Diagnosis: ¥15 bn.



Agricultural Use

Development of Mutated Breed



Sprouts Control of Potatoes



· Food Irradiation
· Radiation Analysis etc.

Equipment Device: ¥390bn.

- Accelerators
- Radiation Equipment for Diagnosis
- Medical Radiation-related Equipment & Products for Medical

Industrial Use

Semiconductor ¥1,230 bn.



Radiation Sterilization ¥310 bn.

- Hypodermic Syringe & Needle
- Vacuum Blood Collecting Tube
- Artificial Joint &

Measurement & Inspection ¥180 bn.

- Radiation measuring Instruments
- Non Destructive Inspection etc.

Polymer Processing such as Radial Tire Manufacturing ¥110 bn.



Electron Beam Processing

Application field list of electron beam (Typical)

- Heat resistance and non-combustibility of wire (graft technology)
- Heat shrinkable tube and sheet productions (crosslinking technology) → memory effect
- Production of foamed plastic (crosslinking technology) → Fine foaming
- Cross-linked rubber (green rubber strength enhancement) → For tire manufacturing etc.
- Coating film hardening (UV competition) → polymerization technology)
- Separator for batteries (grafting technology) → mass production of electrolyte membrane
- Sterilization, Disinfection (DNA double strand break) → Medical device sterilization
PET bottle in-line sterilization
- Food irradiation (except for Japan DNA double strand break) → sterilization of spices
- Desulfurization and denitrification of flue gas (using ion molecular reaction) → Pollution control
- Cleaning of water (DNA double strand break, ionization reaction)
- Crosslinking and modification of plastic, (crosslinking) → improve heat resistance and
mechanical properties
- Semiconductor custom-made control (lattice defect generation) → After processing
- Crosslinking of Teflon → Improvement of radiation resistance, and sliding characteristics

Merits for EB processing

- **Very low energy consumption (high efficiency)**
- **Processing time is very short (in seconds)**
- **No need pollution measures**
- **The degree of freedom of reactions such as polymerization reaction and crosslinking reaction is high**
- **The color of the material does not matter at all when absorbing the energy**
- **Available reaction via ionization**
- **Various reaction environments can be selected (high and, low temperature reaction, etc.)**

How good is Irradiation of Ionizing Radiation Efficiency?

When considering the reaction amount of Ionizing radiation (EB), how much energy is absorbed by the substance Is it collected ?

Its unit is absorbed dose (Gy)

Amount of energy of Ionizing radiation absorbed per unit mass is defined by
absorbed dose Gy (gray)

(Gy is equivalent to J/kg)

If the material is given an absorbed dose of 10 kGy (= 10 kJ / kg), what is the amount of heat (temperature rise)? (Absorbed doses of 10-30 kGy are used in many radiation processes)

If 10kGy of absorbed energy is given to water (specific pressure specific heat: 4.2 J / Kg), this corresponds to a temperature rise of 2.4 °C.

Why is ionizing radiation highly efficient ?

The chemical (covalent) bond in the molecules is relatively weak.

It is governed by the interaction between electrons in atoms.

→ The state should be changed by several eV (4.3 eV for CH) of energy input.

Even for the chemical reagents and heat, they induce the state changes in the materials, which are due to the electronic state change.

The most direct way to change the molecules is direct injection of ionizing radiation !

For this reason, the effect given by ionizing radiation is significantly more efficient than the chemical reagents and heat!

Electron Beam Application Technology (polymer crosslinking)



Heat Shrinkable Tubing

Electron Beam Application Technology (polymer crosslinking)



Heat Resistant Insulating Wire

USA (Voyager) 22145 C
USA 150 1000000

Super heat resistant silicon carbide fiber

Ref. Washio and Maekawa
Material creation and application development
using EB technology, CMC 2016, pp.171-179

(Adoption to an aircraft engine (by GE) ; **Hi-Nicalon™ TypeS**)

Advanced Materials Development / Ceramic Composites

Fabrication of SiC fiber

Si-containing polymer fiber

↓ EB irradiation up to 10MGy

Cured polymer fiber

↓ Pyrolysis in inert gas (~1500°C)

SiC Fiber

- Diameter : 14μm
- Tensile strength: 3GPa (300kg/mm²)
- Heat resistant : 1700°C
- Density : 2.7g/cm³



SiC fiber (Hi-Nicalon®)

Application of SiC fiber



Ceramic Matrix
Composites



Space plane materials

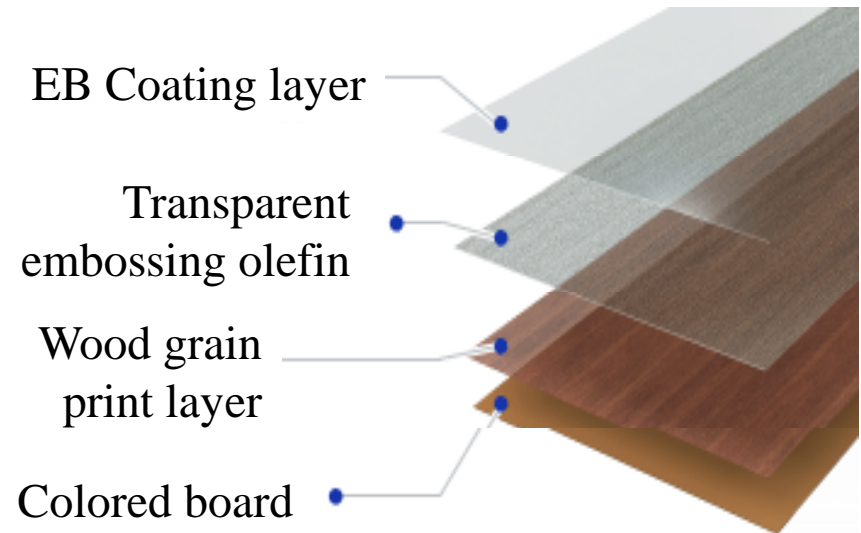
EB applied technology (polymerization) : Decorative plate (Building materials)

▶ Non wax floor sheet 「EB-F」

DNP Product

EB coating technology: Technology to cure resin by irradiating electron beam. It is resistant to scratches, dirt, sunlight, etc., highly durable, practical performance, and stable in quality compared to UV coatings and urethane. This is an energy-saving manufacturing process, which can reduce the CO₂ emissions, and next-generation environment-friendly technology capable of solvent-free coating.

Ref. Washio and Maekawa
Material creation and application development
using EB technology, CMC 2016, pp.182-188



Ref. DNP HP

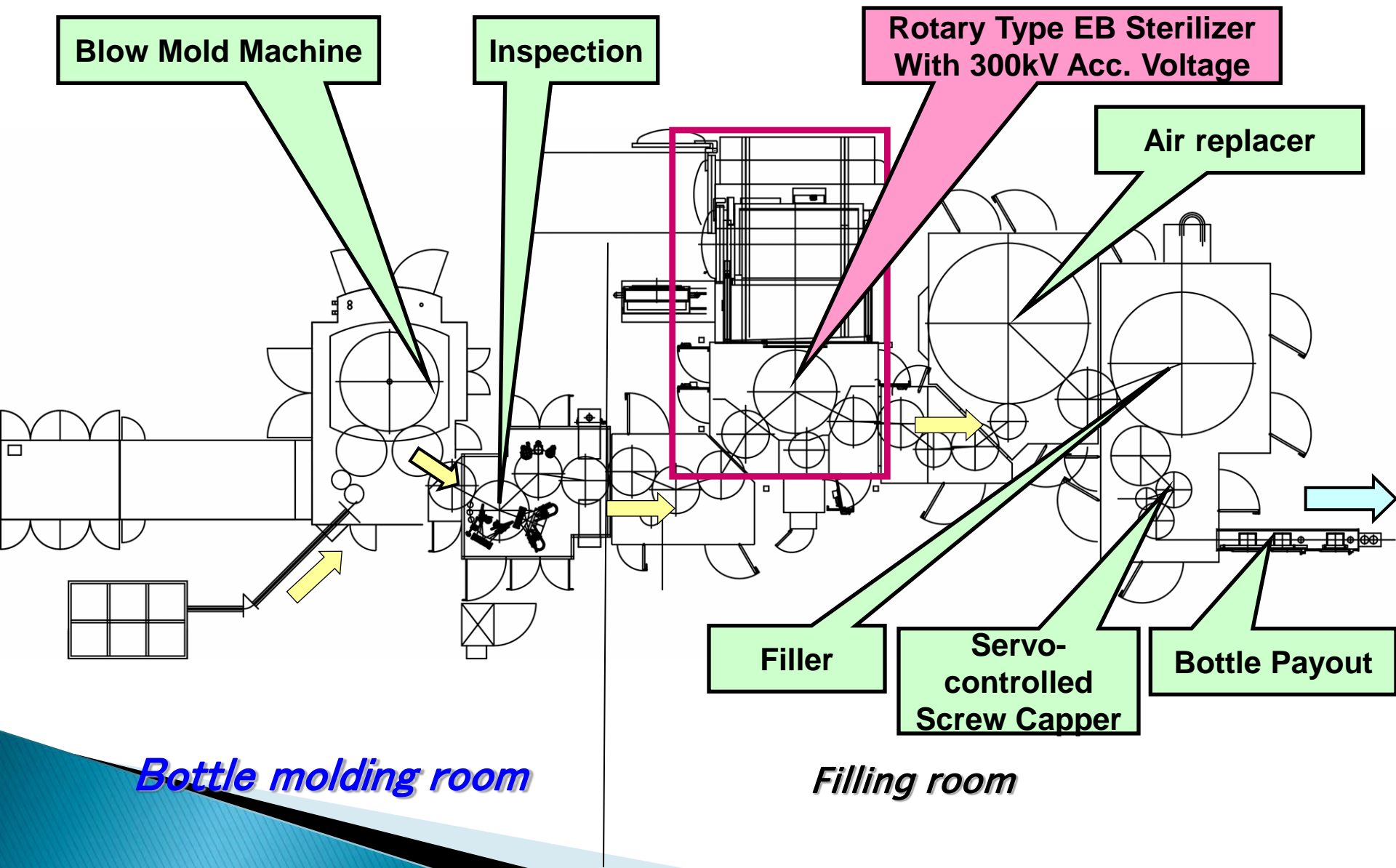
<http://www.dnp.co.jp/kenzai/product/brand/brand05/>

Germ free filling system by EB sterilization system



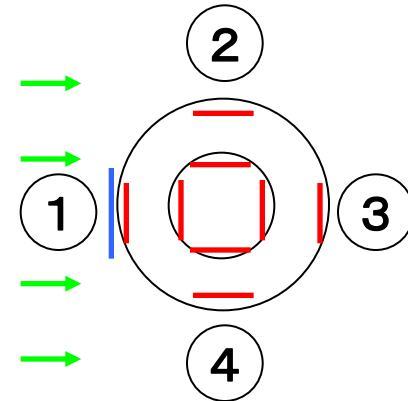
澁谷工業株式会社
Shibuya Kogyo

Layout for Aseptic filling system by EB Sterilization



▶ Absorbed dose at Bottle mouth and Cold Spot

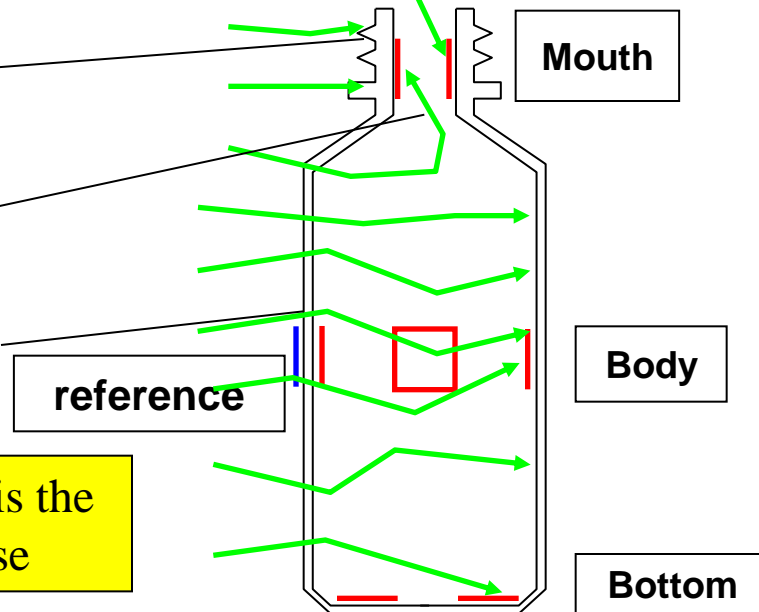
	①	②	③	④
Mouth inside	8. 8kGy	8. 8kGy	8. 7kGy	9. 1kGy
Body inside	44kGy	30kGy	45kGy	30kGy
Bottom inside	32kGy	26kGy	32kGy	33kGy
reference	80kGy (Body outside)			



Mouth: Thickness of 1mm
Electron beam can not penetrate

Mouth: Scattered electron will irradiate on this part

Body: Thickness of 0.1~0.3mm
Electron beam can penetrate



The inner surface of the bottle mouth is the cold spot with the lowest absorbed dose

Improvement in irradiation efficiency at bottle mouth part by magnetic field

Side view



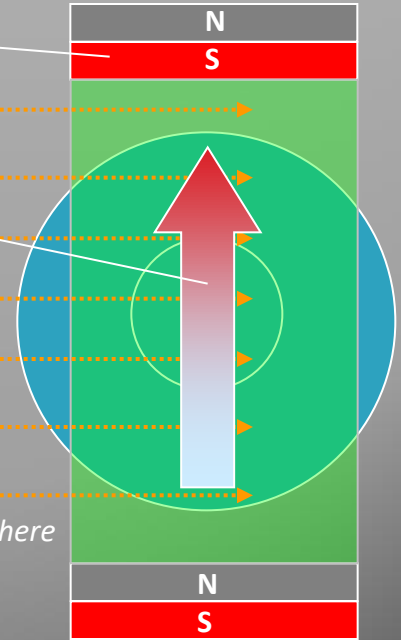
Magnets are placed face-to-face at the side of bottle upper part

Direction of magnetic field

Electron beam is bent in the magnetic field, which enables to irradiate electron beam on bottle mouth part highly efficiently.

Magnet	Not placed	Placed
Current (mA)	169	89
Dose on mouth part (kGy)	10.1	10.7
Dose on body part (kGy)	79.5	43.5

Top view



Vacuum Atmosphere

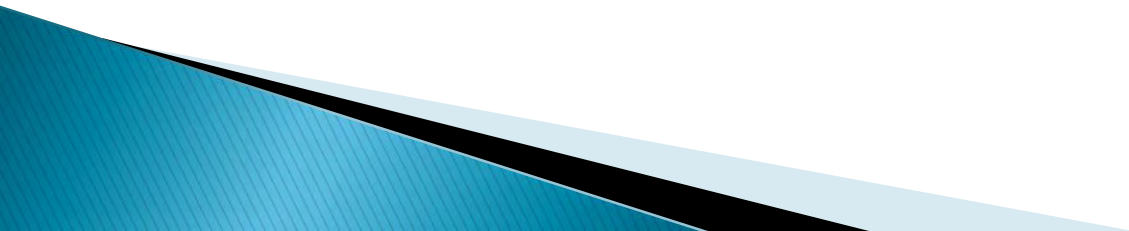
Vacuum Atmosphere

Titanium foil

→ Possible to improve sterilization efficiency with avoiding shrinkage and deformation of bottles.

→ Possible to ensure enough dose at mouth part even by low electric current.

Application of Synchrotron Radiation



As features of X-ray use

- High penetration ability, because it is an electromagnetic wave
- High quality of beam can be achieved (SR)
- Energy selection can be done
- We can use refraction, interference effect
- Good at imaging capability
- Applicable to processing by etching (example will be shown)

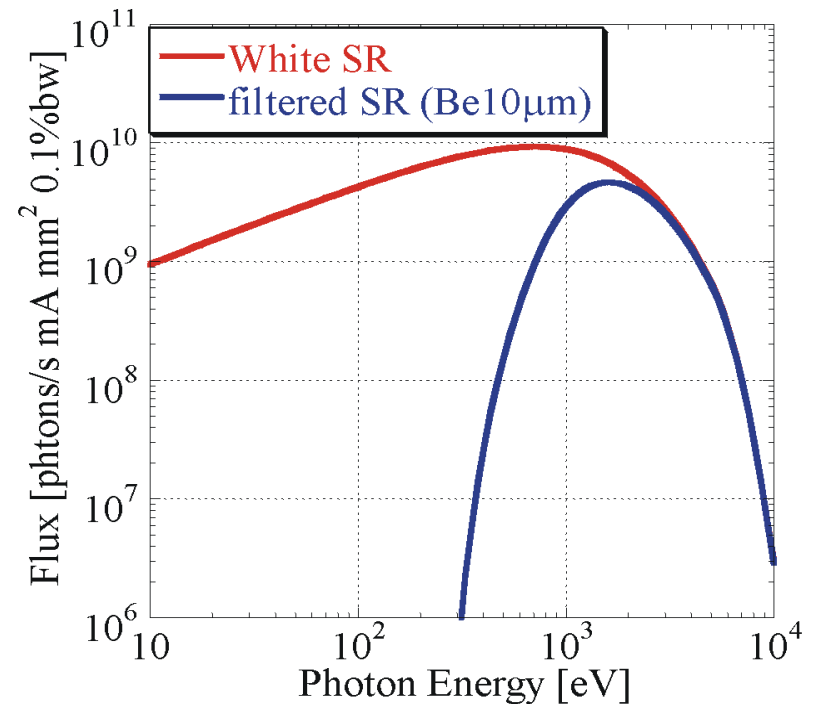
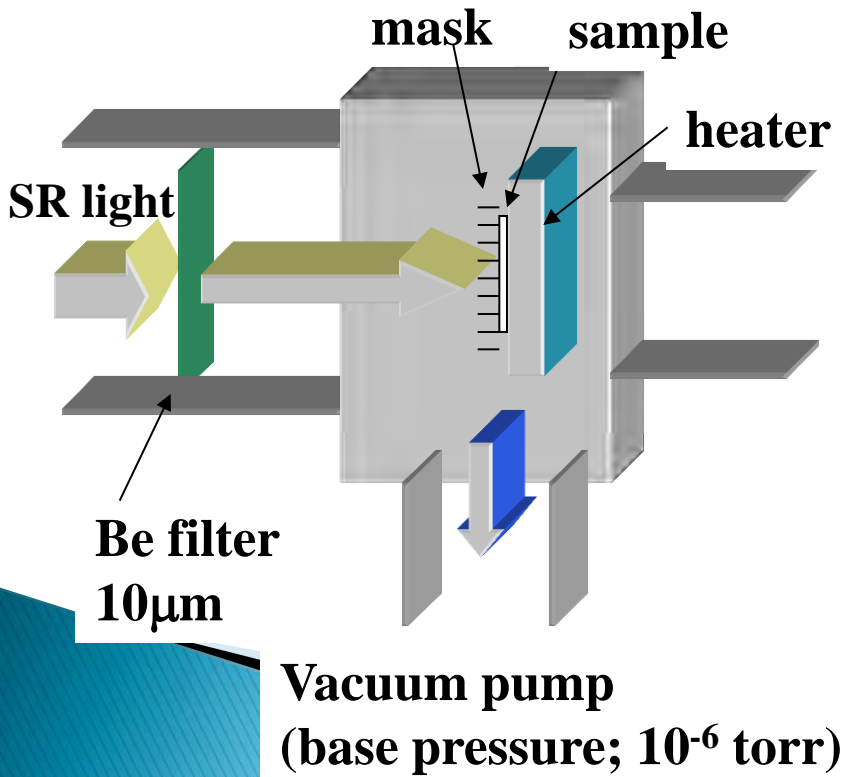
By Using these features, X-ray has a major impact on the development of analytical techniques, and also is applicable to the processing such as direct etching, and EUV lithography, etc.)

Light source

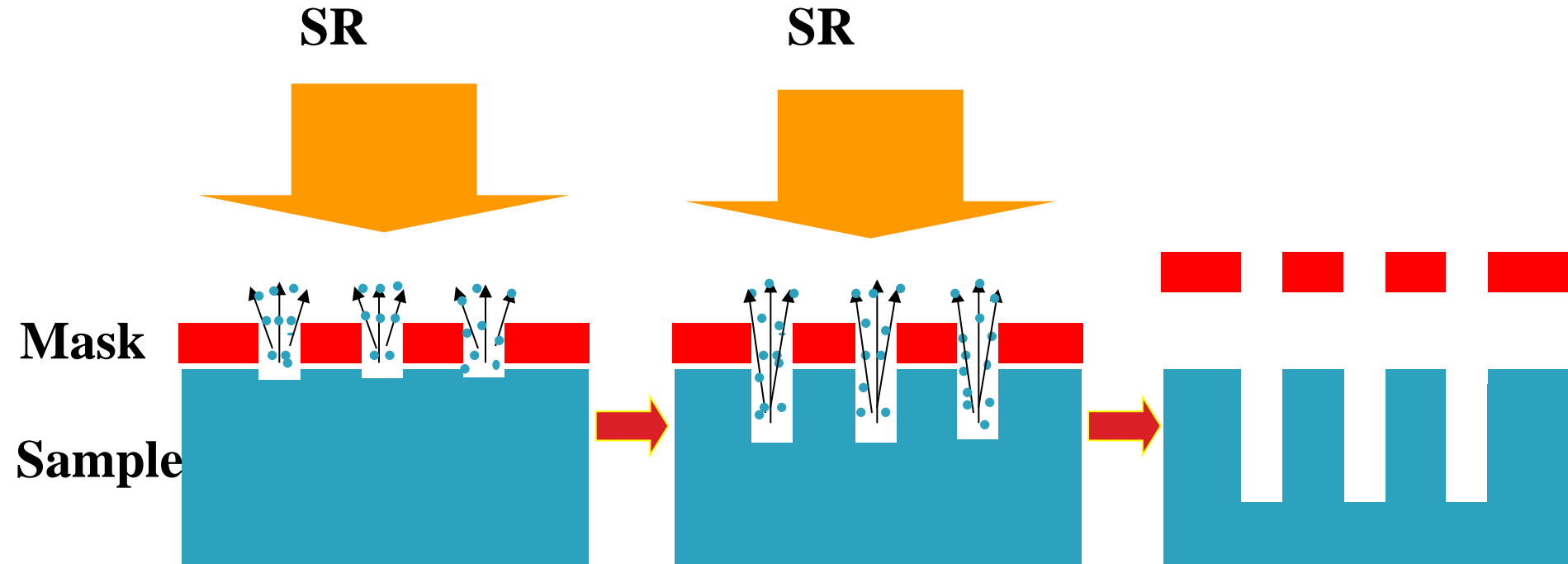
AURORA-2S

(Sumitomo Heavy Industries)

- Max. Beam Current : 600 mA
- Electron Energy : 700 MeV



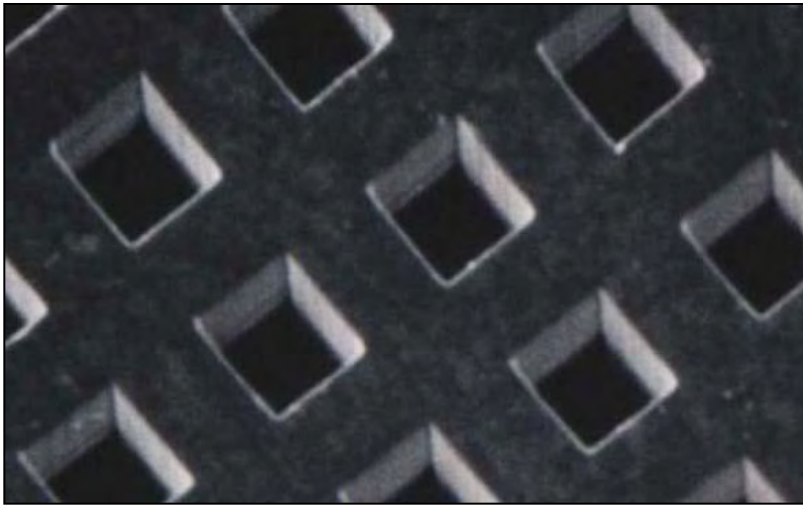
Microfabrication (TIEGA™)



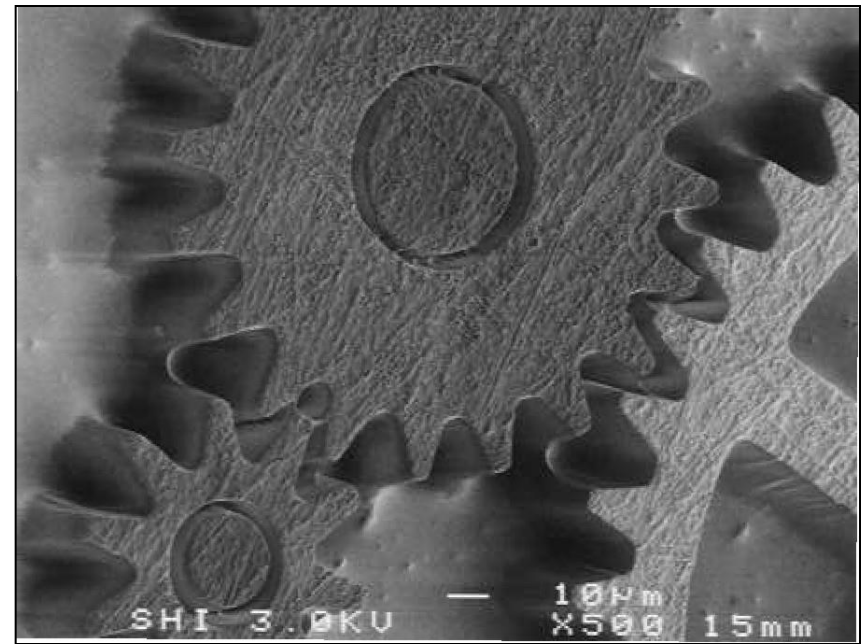
Concept of direct photo-etching using SR

SR exposed region of sample *decomposes and desorbs consequently*. As a result, it is possible to create microstructures of fluorinated polymers by SR **without any chemicals and in single process** !

Example of Microfabrication



— 100 μ m
(a) Micro-filter



— 10 μ m
(b) Micro-gear

SEM images of Micro-parts made by SR irradiation for
RX-PTFE

Application of Ion Beam Irradiation



Abrasion measurements for Automobile Engine by the Thin layer Radio-activation method

(Toyota Central R & D Labs)

Technique

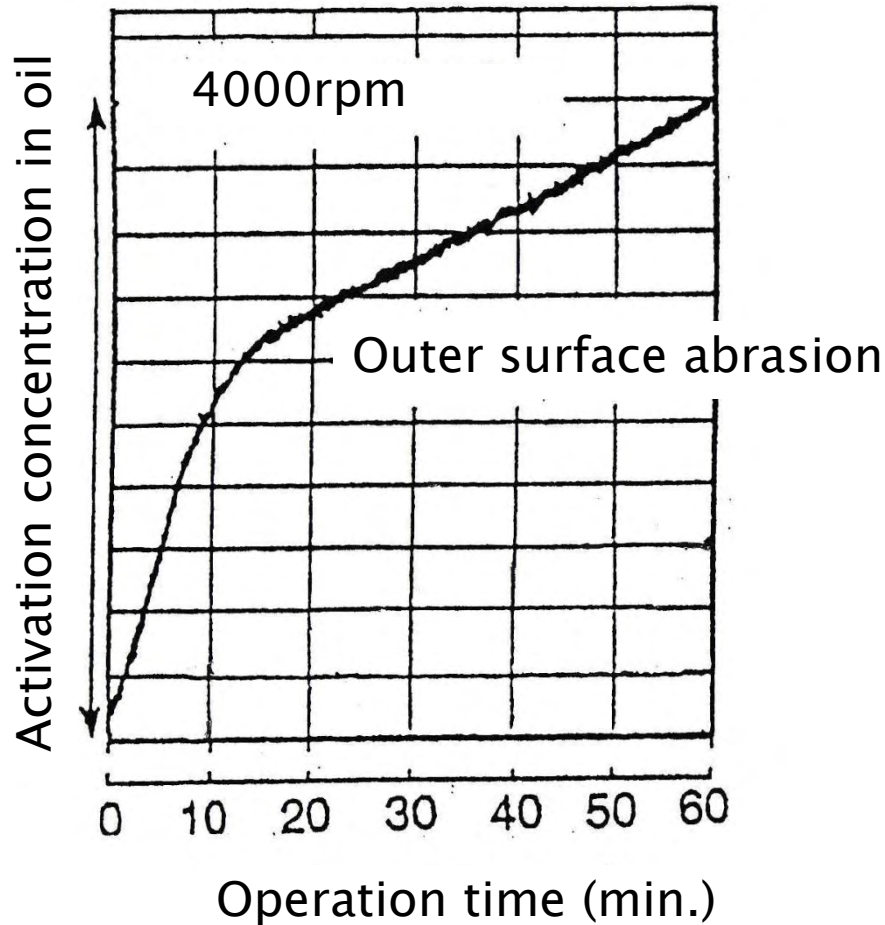
Radio-activation of metallic materials by proton, deuteron, He ion beams from a cyclotron

Objectives

- Piston ring abrasion ⇒ Elucidation of abrasion characteristics in minutes
- Engine valve abrasion ⇒ Elucidation of operation speed dependency of abrasion
- Engine oil development ⇒ High temperature high shear viscosity optimization

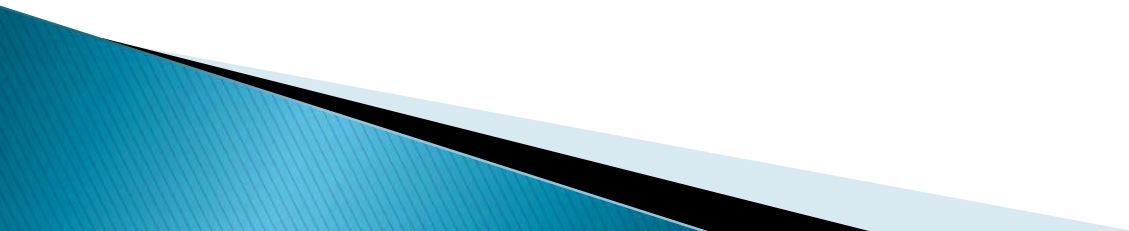
Target in developing engine of automobile

Example of the application to the measurement of outer peripheral surface abrasion of piston ring (top ring)



Piston ring abrasion measurement chart

Application for Agriculture



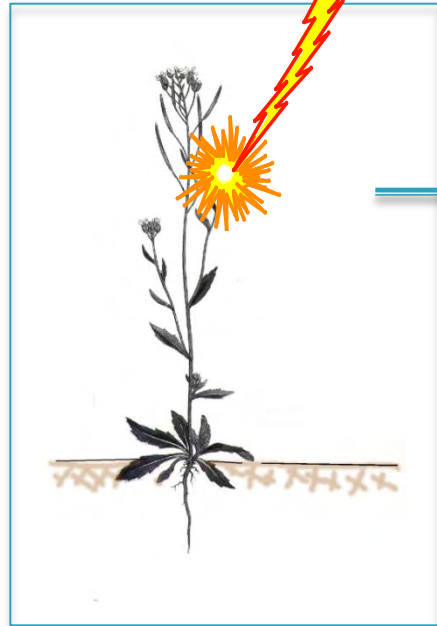
Ion beam mutagenesis and ion beam breeding

Ion beams

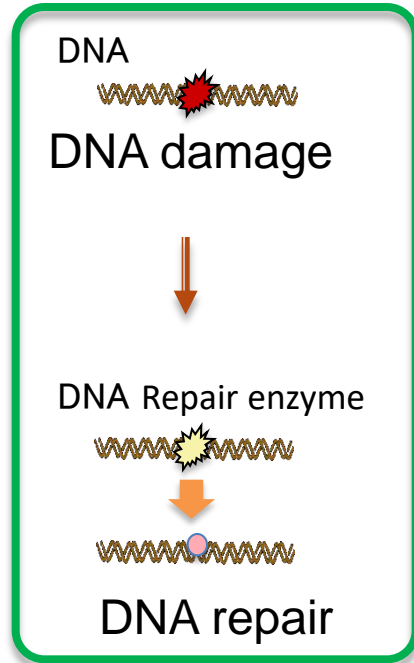
DNA damage

DNA repair

Mutation



Cells



Correctly repaired

Wrong repair
Mutation DNA

⇒ Mutant gene

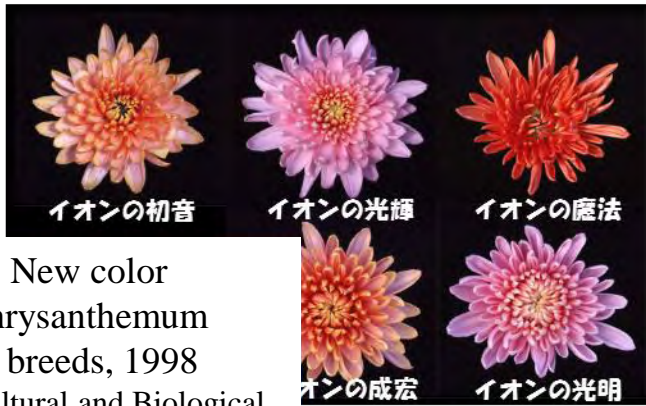
Mutant



Creature

Creation and
breeding of
useful varieties

Ion beam breeding



New color
chrysanthemum
5 breeds, 1998

Agricultural and Biological
Resources Research Institute



New color Carnation
8 breeds, 2002~ Kirin



No side branch chrysanthemum
3 breeds, 2005~ Kagoshima



New color Osteospermum
2 breeds, 2007~
Gunma



Ficus pumila with
Environmental purification
2007 Hiroshima Univ.



Low Cd absorption rice
2012 Agricultural Environmental
Technology Research Institute



Low temperature growth
greenhouse melon
2011 Shizuoka



New color Fragrance
cyclamen 2 breeds,
2012~ Saitama



Sweet smell Sake yeast
2012 Gunma

Future prospects of radiation processing

1. Looking back on history, What has been running all the time? ...

Polyethylene crosslinking is still a major item

Crossing of tire rubber is also a big item

Why did these survive?

Easy process, no achievement of cost and quality in any other methods

Life cycle does not differ from conventional products

Technology that has died or hardly moves

Floppy disk manufacturing (demand has been lost)

Tunnel interior steel plate (demand is limited)

Transfer printing technology (alternatives have evolved or no longer needed)

Oji Paper's Super Mirror (Problems after use)

etc



Future prospects of radiation processing (2)

2. Surviving technology, survival technology ?

Supporting the bottom of social infrastructure

Alternative technologies are expensive or have poor performance

3. What kind of processing will survive?

What can almost be done only by using radiation

⇒ polyethylene foams and crosslinked films

Things that can not be done without radiation

⇒ Medical etc.

Materials used continuously

Technology with low environmental impact (limited water use, etc.)

⇒ PET bottle production line

Huge scientific projects

J-PARC Project (JAEA & KEK)

⇒ Proton + Neutron Science

RI Beam Factory (RIKEN)

⇒ Unstable nucleus, new element

X-FEL (RIKEN Harima)

⇒ Ultra-short pulsed X-ray

Super-KEKB (KEK)

⇒ electron-positron collision

⇒ Beyond standard theory

Coming ILC (International Linear Collider)

Japan & all over the world

Superconductivity technology,

Material technology Vacuum technology,

High power technology

International Research Center,

etc.

Finally

Really new radiation processing technology will be born from cutting edge science.

Acknowledgment

Many people gave me advices, materials, etc. when giving this lecture.

In particular, Many people,

Takanori Kato of Sumitomo Heavy Industries,

Yutaka Yamase from Sumiju Atex,

To everyone in Shibuya (especially Mr. Naka managing director)

All of Dai Nippon Printing

All of Environmental Purification Research Institute Inc.

Mr. Yamamoto of TPR (Imperial Piston Ring)

The members of the Washio Lab.

I received many materials, knowledge, and information.

I will express my sincere thanks to you here.



Thank you very much for your attention.

