Centre for Energy Research

Dosimetry Systems and Procedures in Traditional Radiation Processing and Wastewater Treatment Technologies

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Eötvös Lóránd Research Network

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Introduction – Centre for Energy Research (EK)

It is one of the 18 research centres of **Eötvös Loránd Research Network** (Hungarian Academy of Sciences)

One of the **Technical Support Organizations** of the Hungarian Atomic Energy Authority

Main **Technical Consultant** of the **Paks NPP**

Nuclear Security Support Centre of IAEA NSSC Network together with the Hungarian Atomic Energy Authority

Operating a centralized **National Nuclear Forensics Laboratory** in Hungary (working together with the relevant national organizations and authorities)

IAEA Collaborating Centre for Nuclear Forensics

Governmental Decree (1035/2012) indicates Hungary's National Security Strategy and 490/2015 G.D. **delegates nuclear forensics to EK** (original: 17/1996)

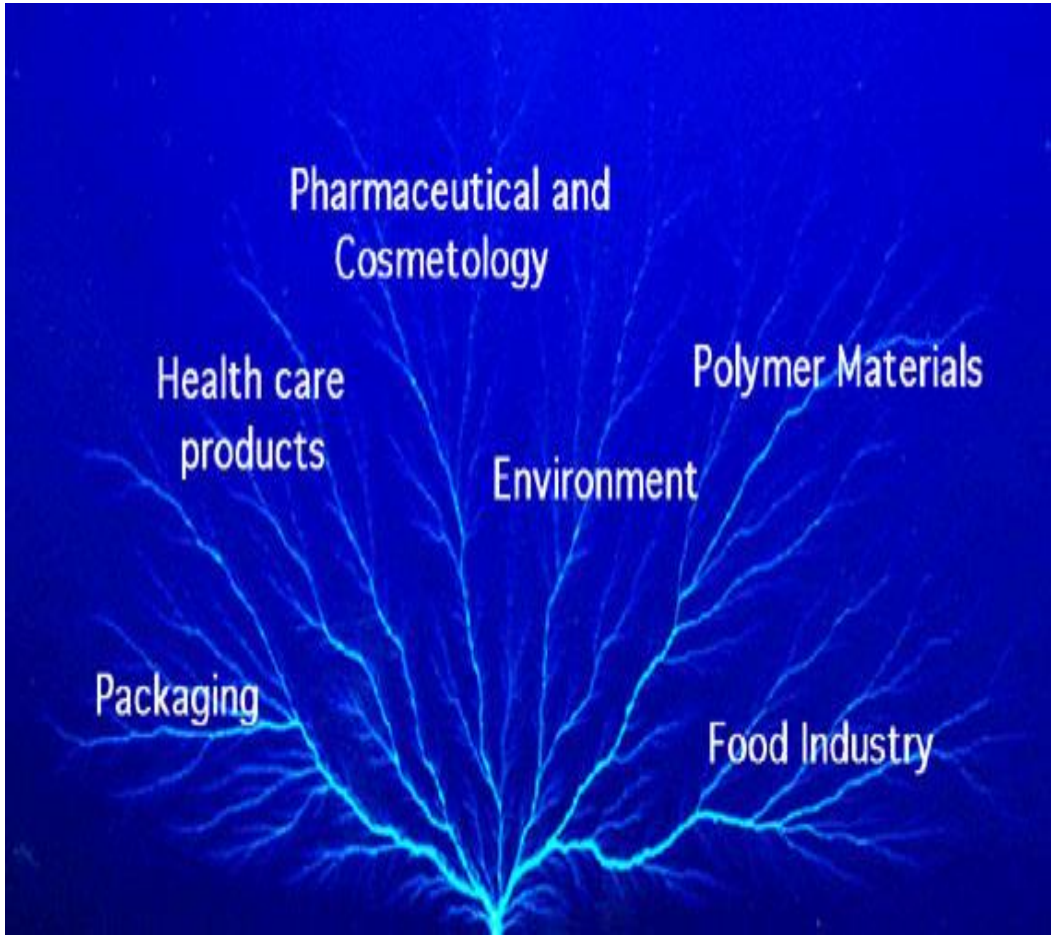
EK has about ~400 *employees (~65 % scientists)*

Age distribution is acceptable (50% of researchers are below 40)

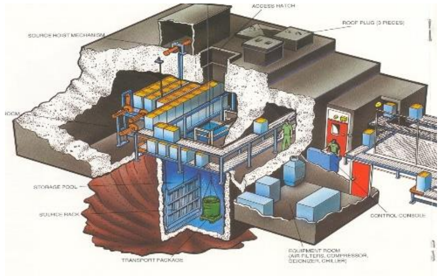
Overall budget is 10-15 M€

21 departments (laboratories)

Radiation Processing Applications



To assure, that



- the required (biological, chemical, physical) effect is achieved and
- the radiation technology is performed safely

the absorbed dose and dose distribution in the product

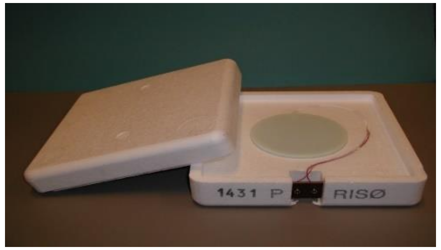
and

its relationship with irradiation facility parameters

(like irradiatio/dwell time, position of source rack, electron energy and current, conveyor speed, scanning width and homogeneity, etc.)

⇓

have to be measured and controlled with suitable dosimetry systems!



Basic Dosimetry Principles

- **Absorbed dose is the quantity of ionizing radiation energy imparted per unit mass of a specified material (*d´/dm)*, where *d´* is the mean incremental energy imparted by ionizing radiation to matter of incremental mass *dm*.**

- **In radiation processing, validation and process control (sterilization, polymer processing, food irradiation, environmental applications, etc.) depend on the measurement of absorbed dose.**

- **Measurements of absorbed dose shall be performed using a dosimetric system or systems having a known level of accuracy and precision.**

- The calibration of each dosimetric system shall be traceable to an appropriate national standard.

- Classification: *primary-, reference-, transfer standard and routine* systems*;*

Dosimetry Systems

Dosimeter system	Method of analysis	Useful dose range, Gy	Nominal precision limits	References
Fricke solution	UV – spectro-photometry	$3x10 – 4x10^2$	1 %	ASTM E 1026 - 04
Ceric – cerous sulphate	UV – spectro-photometry	$10^3 – 10^6$	3 %	ISO/ASTM 51205
Potassium dichromate	UV-VIS spectrophotometry	$5x10^3 – 4x10^4$	1 %	ISO/ASTM 51401
Ethanol-mono-chlorobenzene	Titration,or HF oscillometry	$4x10^2 –3x10^5$	3 %	ISO/ASTM 51538
L - alanine	EPR	$1 – 10^5$	0.5 %	ISO/ASTM 51607
Perspex systems	VIS - spectro-photometry	$10^3 – 5x10^4$	4 %	ISO/ASTM 51276
FWT – 60 film	VIS - spectro-photometry	10^3 - 10^5	3 %	ISO/ASTM 51275
B 3 film	VIS - spectro-photometry	10^3 - 10^5	3 %	ISO/ASTM 51275
Cellulose triacetate	UV – spectro-photometry	10^4 - 10^6	3 %	ISO/ASTM 51650
Calorimetry	Resistance/ temperature	$1.5x10^3 – 5x10^4$	2 %	ISO/ASTM 51631

Classification of Dosimetry Systems

a./ **Based on metrological properties: types I and II;**
(E 2628 – 09, ASTM Standard);

Type I:

- dosimeter of high metrological quality; its response is affected by individual influence quantities in a well defined way so, that it can be expressed in terms of independent correction factors; (Fricke, dichromate, ceric-cerous sulphate, ECB, alanine/EPR).

Type II:

- dosimeter, its response is affected by influence quantities in a complex way – cannot be expressed in terms of independent correction factors; (process calorimeter, CTA, Sunna, PMMA, FWT, B3, TLD, etc).

Classification of Dosimetry Systems

b./ Based on field of application:

- **Reference standard systems (type I);**

  Used to calibrate dosimeters for routine use, therefore high metrological qualities, low uncertainty and traceability to appropriate national or international standards are needed.

  +/- 3 % (k = 2);

- **Routine systems (type II);**

  Used for routine absorbed dose measurements (i.e. dose mapping and process monitoring). Traceability to national or international standards is needed.

  +/- 6 % (k = 2);

Classification of Dosimetry Systems

c./ Chemical and physical methods of dosimetry:

- **Chemical methods of dosimetry:**

Liquid systems:

- Aqueous dosimetry systems: Fricke, dichromate, ceric-cerous;
- Organic dosimetry system: ethanol-monochlorobenzene solution;

Solid systems:

Cellulose triacetate, polymethylmethacrylate, radiochromic films (FWT-60, B3 (GEX), GafChromic, Sunna, alanin/EPR;

- **Physical methods of dosimetry:**

Calorimeters: primary standard systems and process calorimeters;

Selection criteria

- **Determination of absorbed dose in product specific dosimeter systems;**

$\Downarrow$

- **The radiation absorption characteristics of the product and the dosimeter material should be similar in terms of atomic number;**

$\Downarrow$

absorbed dose is material dependent

- **According to the process to be controlled (i.e.: gamma, electron, X-ray);**

- **According to dosimeter characteristics;**

  (dose, dose range, energy, cost, reproducibility, resolution, stability, etc);

Dosimetry principles

Dose measurements depend on various methods (e.g.):

- Temperature increase (calorimeters);
- Colour change (perspex, radiochromic systems);
- Free radical concentration (alanine);
- Conductivity change (ECB, aqueous-alanine solution);
- Radiation chemical oxidation (Fricke);
- Radiation chemical reduction (dichromate, ceric-cerous);
- Optically stimulated luminescence (Sunna);

Primary standard systems:

- Dosimeter of the highest metrological quality, established and maintained as an absorbed dose standard by a national or international standards organization for calibration of radiation environments (fields);

- Application is based on measurement of basic physical quantities (ionization current and temperature);

- Most common systems: ionization chambers, calorimeters; (primary standard laboratories: e.g. NPL);

- No calibration is needed;

Reference standard systems:

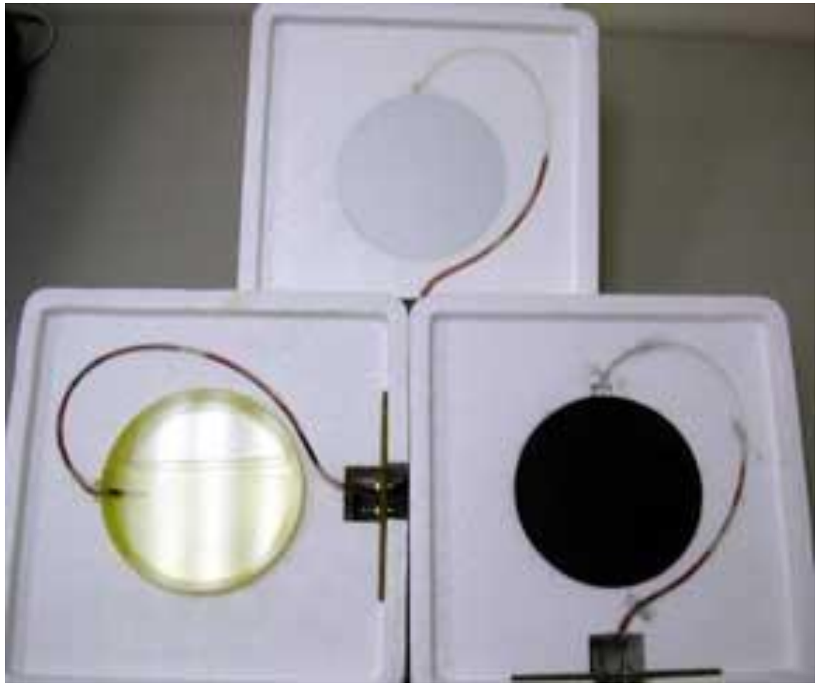
- Dosimeter of high metrological quality used as a standard to provide measurements traceable to measurements made by primary standard systems;

- These systems require calibration and are used to calibrate radiation environments and routine dosimeters;

- Solid phase dosimetry systems :
  alanine (pellet, rod, film);

- Liquid phase dosimetry systems :
  Fricke solution;
  potassium dichromate solution;
  ethanol-monochlorobenzene solution;
  ceric-cerous solution;

- Process calorimeters;



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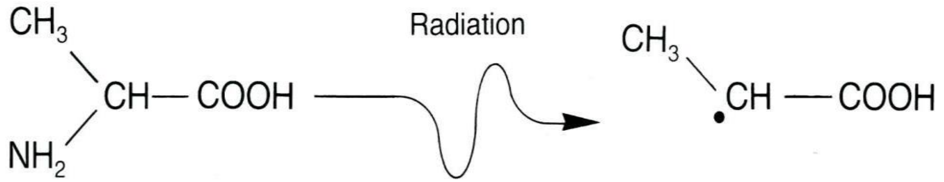
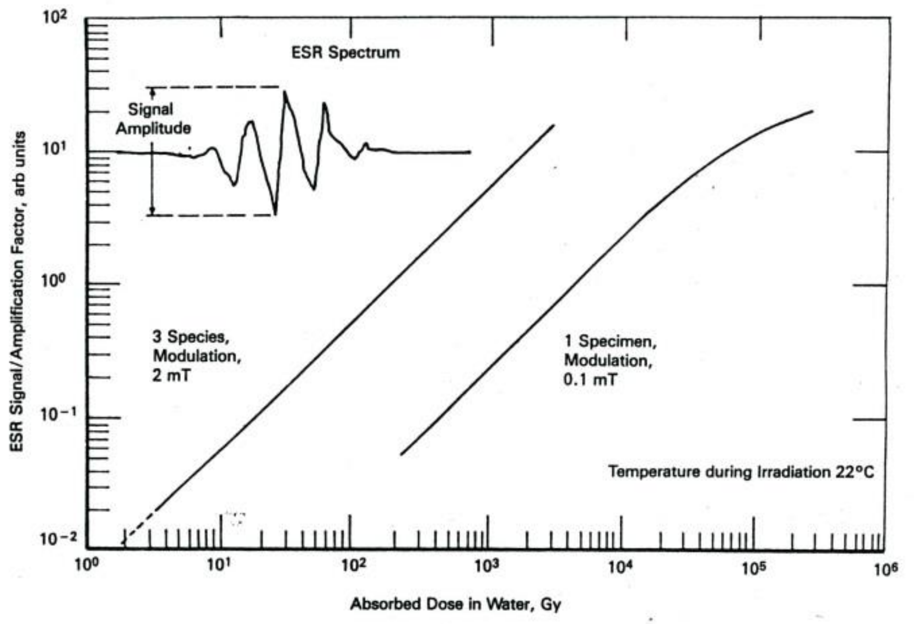
Alanine Dosimetry (ISO/ASTM 51607)

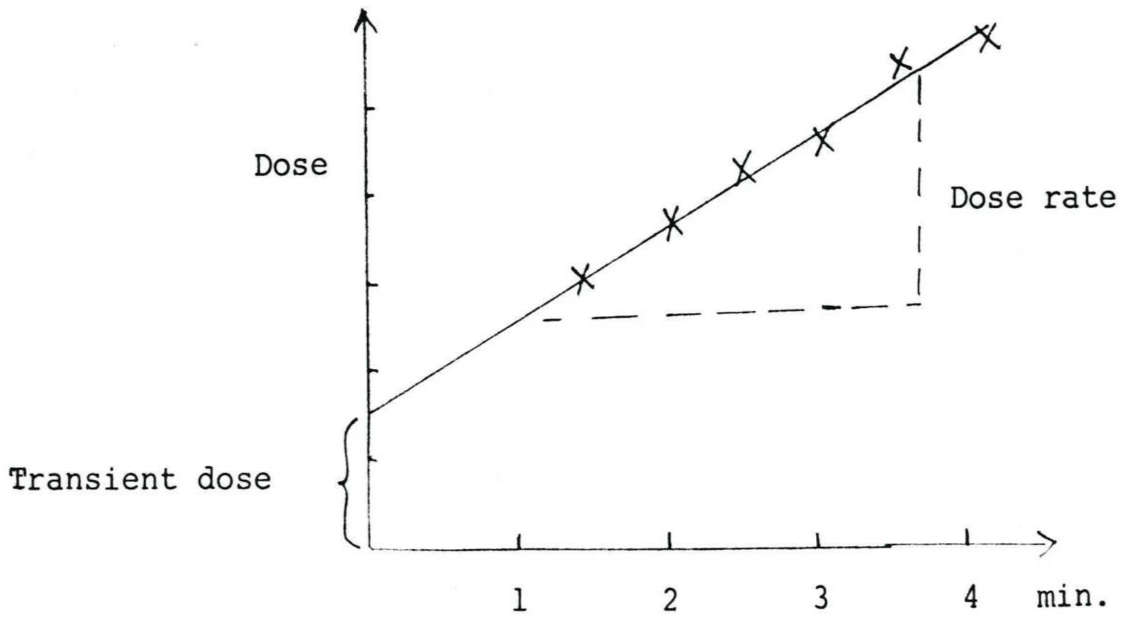
-ESR analysis: measures free radical concentration;
- Dose range: 10 Gy – 100 kGy;
- Reproducibility < 0.5 %;

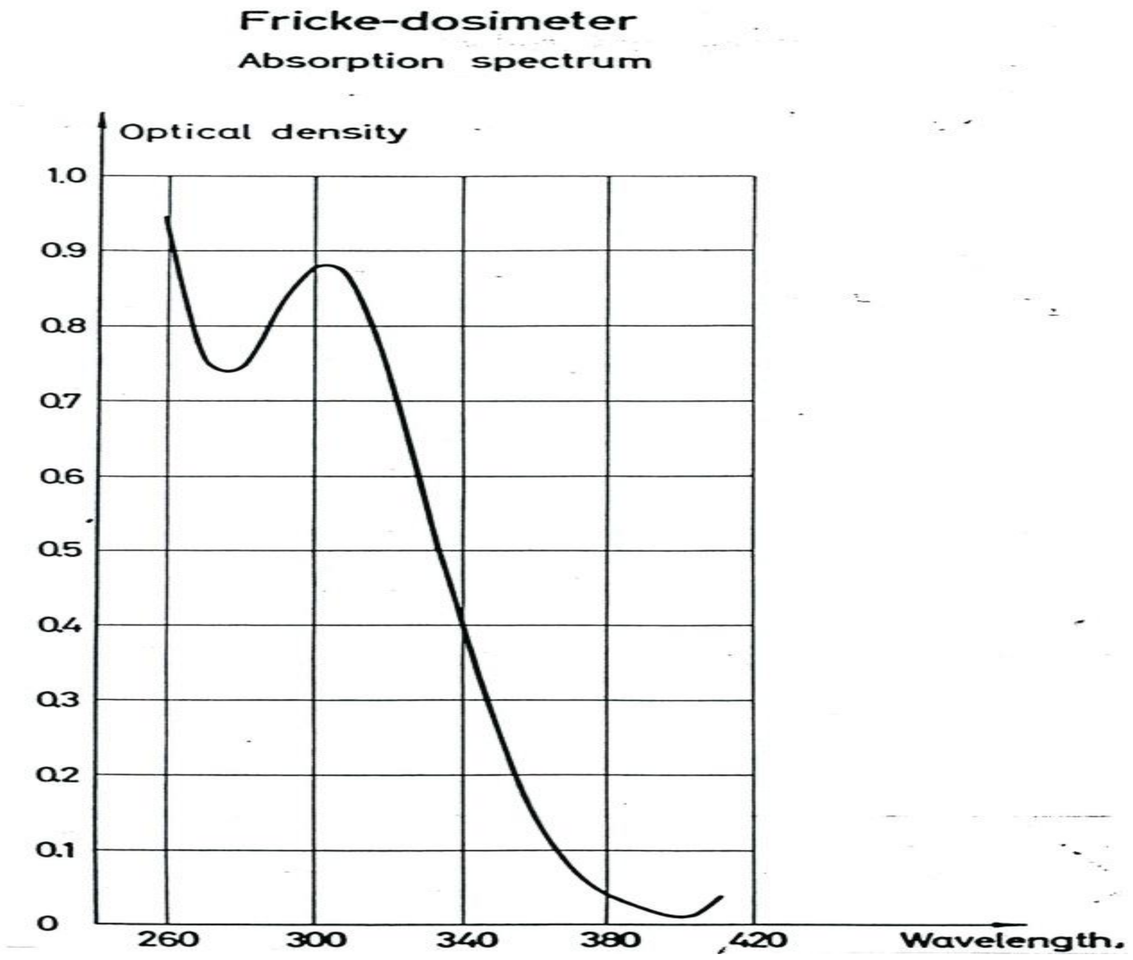




FRICKE Dosimetry (ASTM E 1026)

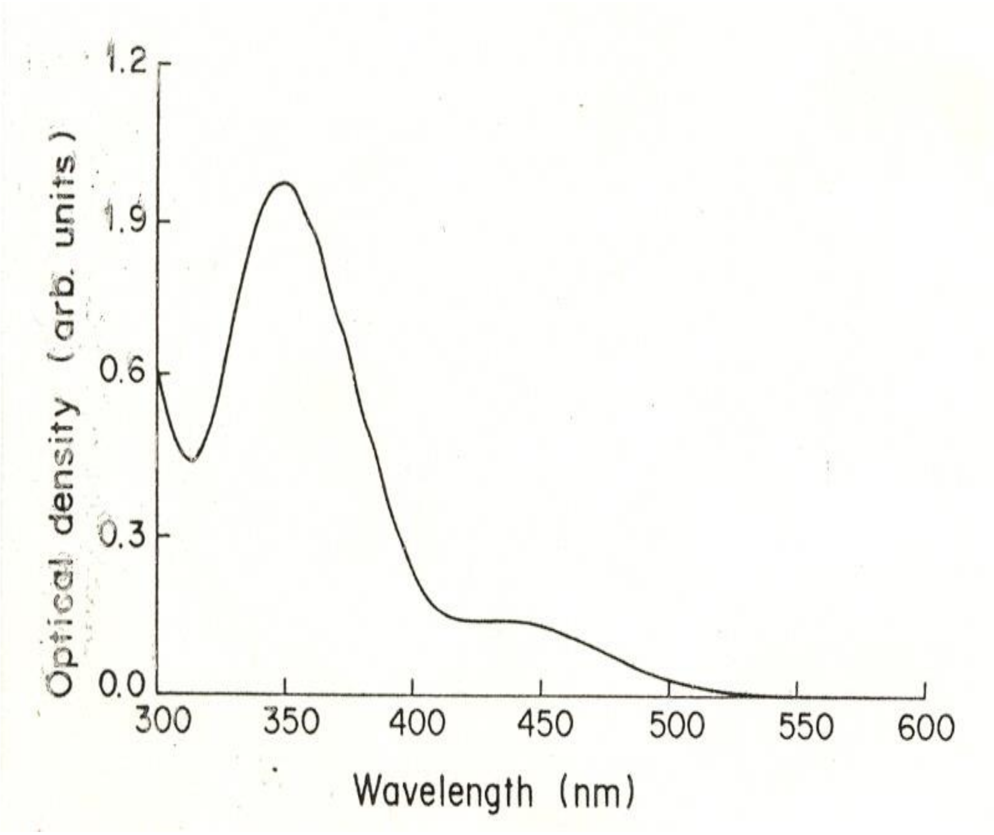
Radiation-induced oxidation of ferrous ions, Fe(II), to ferric ions, Fe(III), in acidic-aqueous solution: $G(Fe^{3+}) = 15.6$
Dose (Gy) = $2.74 \times 10^2 \times \Delta$ O.D. (25 ^oC)
Dose range: 30 – 400 Gy.
Spectrophotometric read-out at 304 nm.





Dichromate Dosimetry (ISO/ASTM 51401)

- Colour change measured by spectrophotometry (440 nm);
- Dose range: 10 – 50 kGy;
- Reproducibility < 0.5 %;

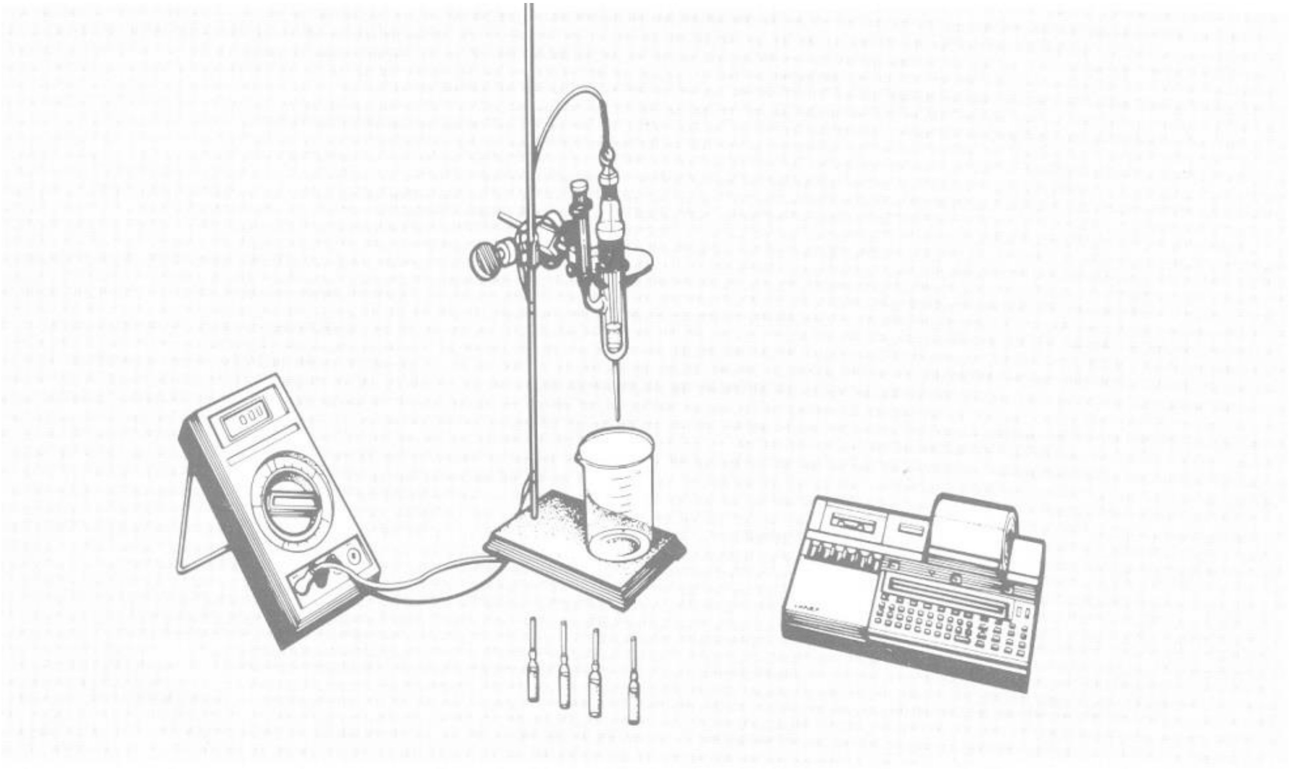


Ceric sulphate (or ceric-cerous sulphate) dosimeter (ISO/ASTM 51205)

Radiolytic reduction of the ceric ions (Ce^{4+}) to cerous ions (Ce^{3+}) in an aqueous - acidic solution.

1. Ceric sulphate solution:
Spectrophotometric read-out: 320 nm.
Dose range: 1- 200 kGy;

2. Ceric – cerous solution:
Potentiometric read-out:
Dose range: 0.5 - 5 / 5 – 50 kGy;



ECB Dosimetry (ISO/ASTM 51538)

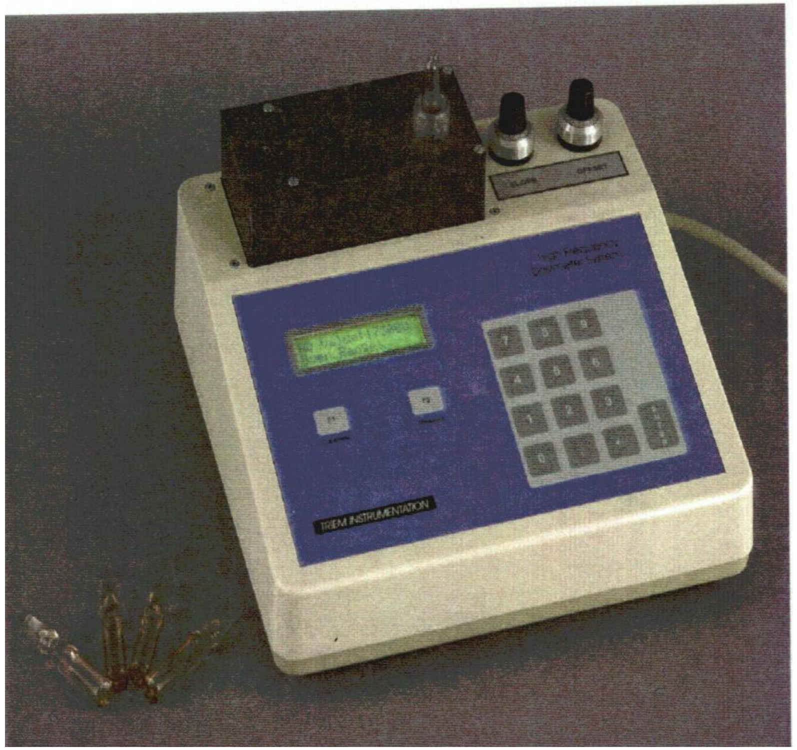
Ethanol-monochlorobenzene solution:

- HCl formation, its concentration is the measure of absorbed dose
- Evaluation:

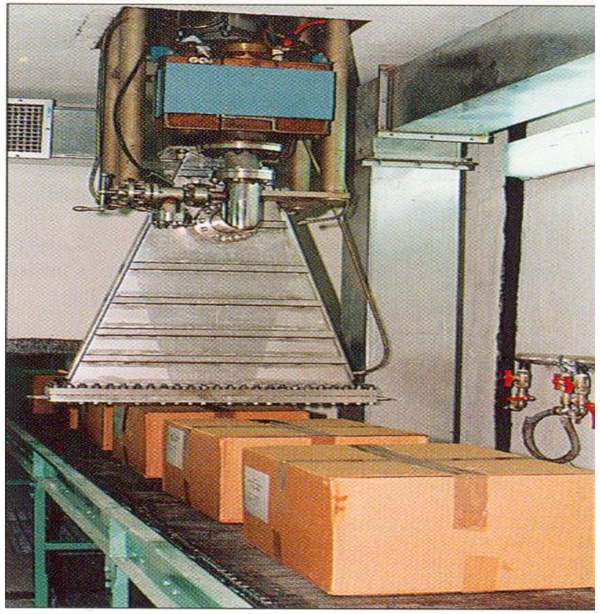
- Chemical titration – reference system:
- G(HCl) = 5.6;
  Dose range: 0.05 – 100 kGy;

- Oscillometry – routine system;
- non-destructive analytical method:
- reevaluation is possible years later;
  Dose range: 1 – 200 kGy;

- Reproducibility: +/- 3 – 5 %;



Process calorimeters ISO/ASTM 51631

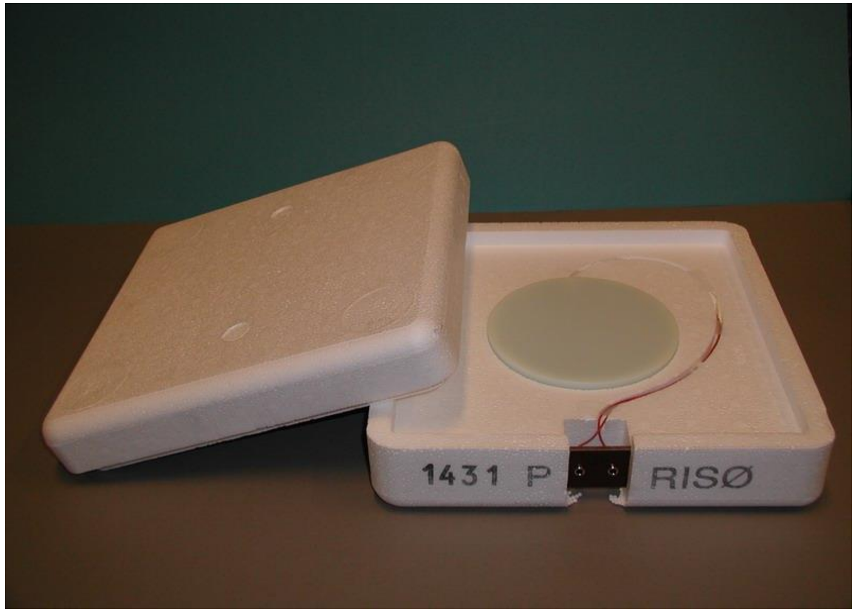


1.5 - 4 – 10 MeV:

- graphite, water, PS calorimeters (1.5 – 50 kGy);

- calibration, nominal dose measurements;

- reproducibility: less than 1 %;



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Dosimetry systems in radiation processing

Transfer standard systems:

- Intermediary system with high metrological qualities, suitable for **transferring dose information from an accredited/standard laboratory to an irradiation facility** to establish traceability (comparing absorbed dose measurements)$\Rightarrow$dosimetry intercomparison exercise;

- These systems require calibration;

- Dosimetry systems:
  - ***alanine;***
  - ***ethanol-chlorobenzene (ECB);***
  - ***potassium dichromate;***
  - ***ceric-cerous,***

Dosimetry systems in radiation processing

Routine systems:

-Dosimetry systems used in radiation processing facilities for **absorbed dose mapping and process monitoring;**

-Systems,capable of giving reproducible signals;

-These systems require calibration;

-Dosimeter systems:

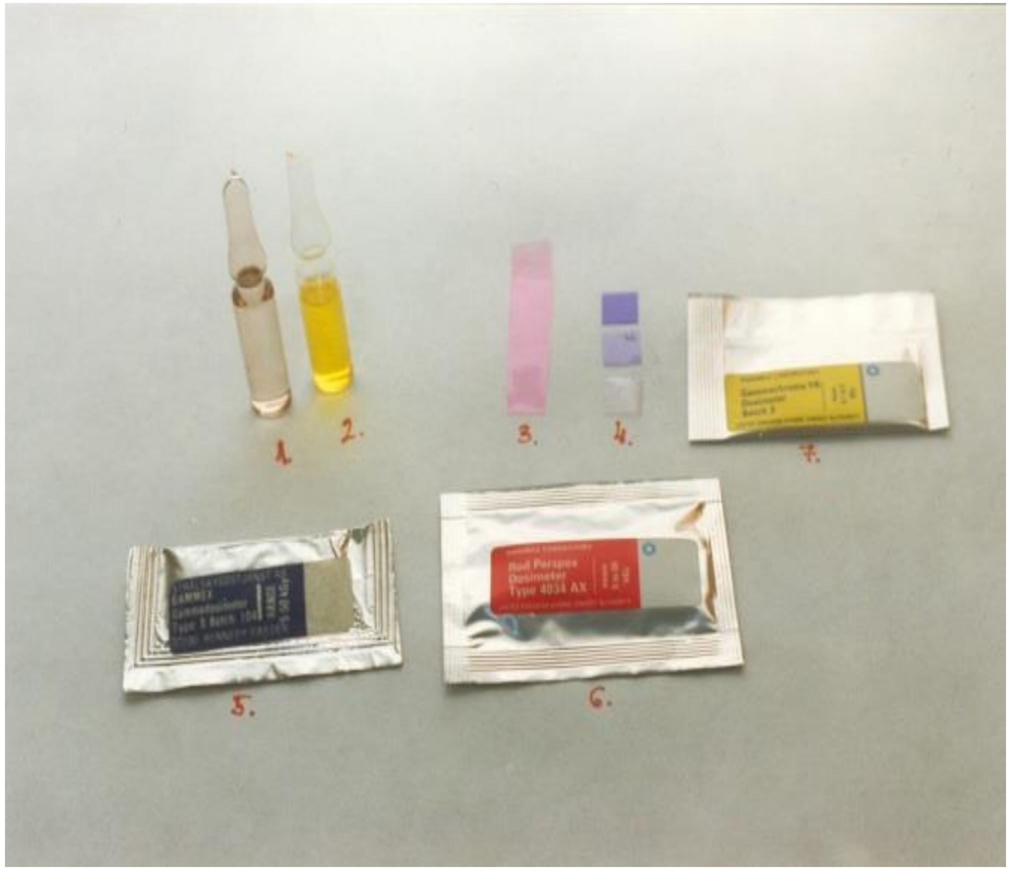
- ***Fricke solution;***

-Perspex (red-, amber-, Gammachrome);

-radiochromic films (FWT-60, B3 - Gex, Gafchromic, Sunna);

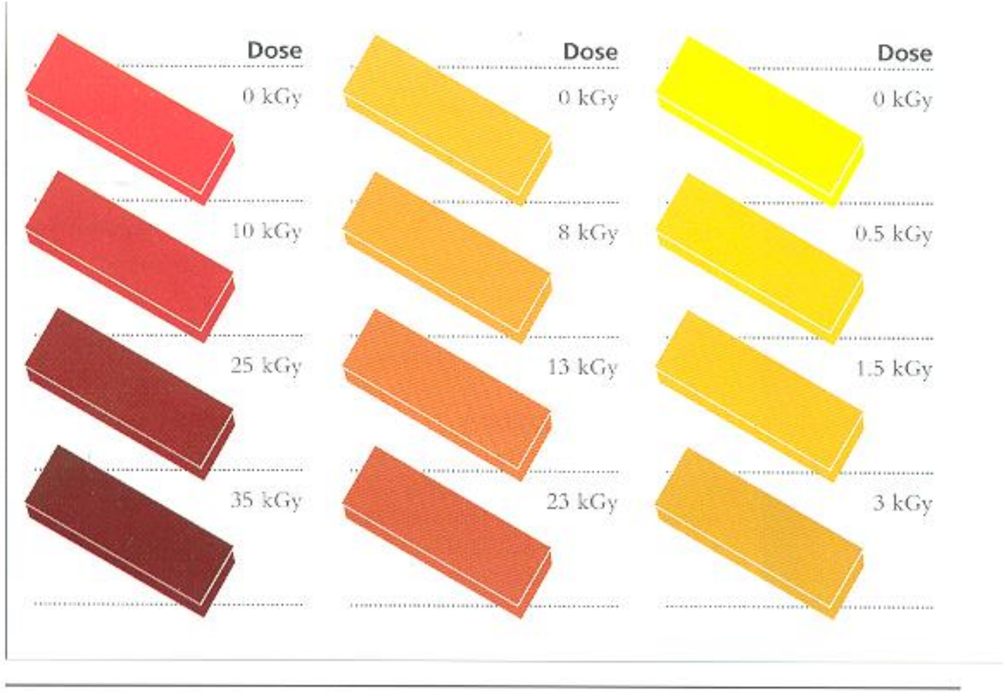
-ECB, ceric-cerous solutions;

-Process calorimeters (water, graphite, polystyrene);



Systems based on optical absorption measurement: Perspex dosimetry (ISO/ASTM 51276)

Colour change - spectrophotometry;
Dose range: 0.5 – 50 kGy;
Reproducibility < 3 %;
Post irradiation change of signal;



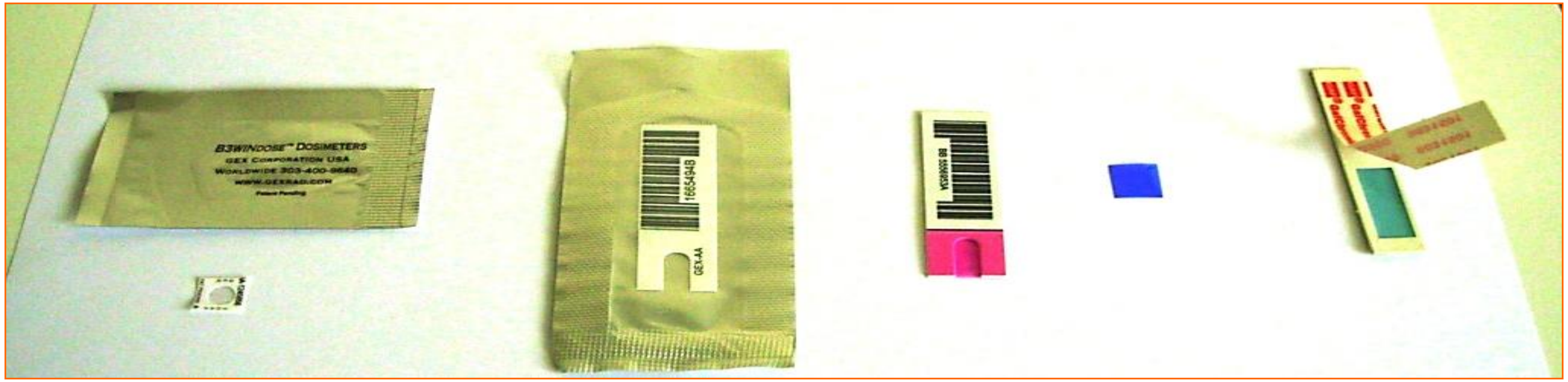
HARWELL
Dosimeters

Systems based on optical absorption measurement: Radiochromic films (ISO/ASTM 51275)

Spectrophotometric readout:

	GEX(B3)	FWT	Gafchromic
Dose range, kGy:	3 – 150	3 – 150	0.01 - 40
Wavelength, nm:	554	510, 605	670, 633, 580, 400

Stability: heat treatment after irradiation; packaging (UV);



System based on conductivity evaluation

Aqueous – alanine solution (1 – 100 kGy):

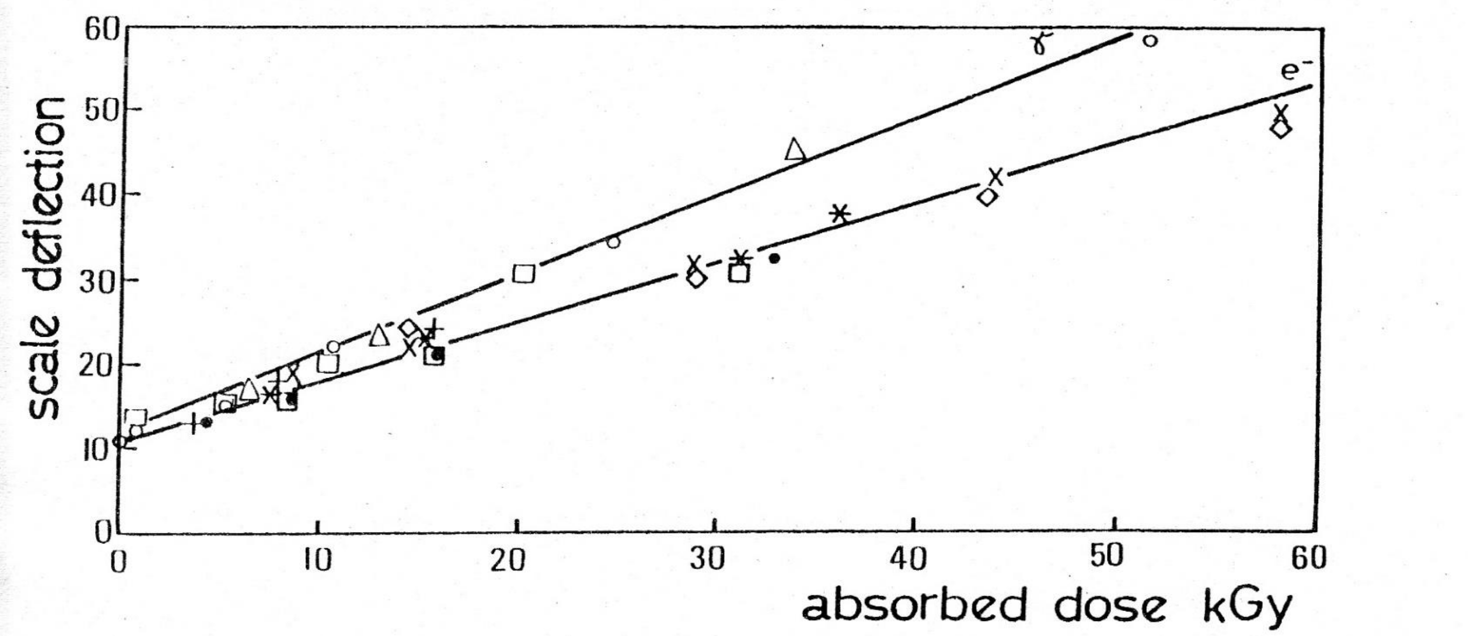
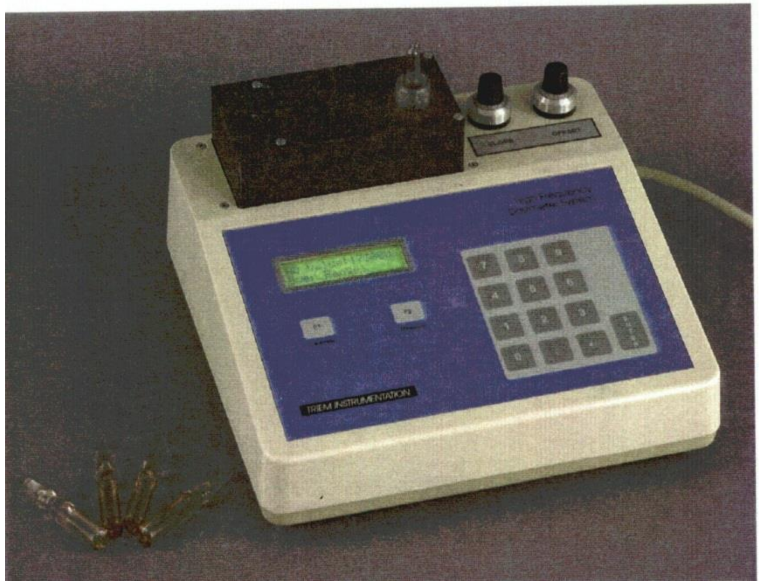


Fig. 2.

Oscillometric response of alanine solutions as a function of dose at various dose rates (^{60}Co irradiation), electron energies and currents (electron irradiation).
^{60}Co irradiation: 0.13 kGy/h: □ ;3.5 kGy/h: △ ;30 kGy/h: ○ ;
electron irradiation: 4 MeV, 2.6 μs, 13 μA: ◊ ; 4 MeV, 2.6 μs, 26 μA: × ; 10 MeV, 0.5 μs, 0.4 A: + ; 10 MeV, 1 μs, 1 A: ∗ ; 10 MeV, 4 μs, 1 A: • ;



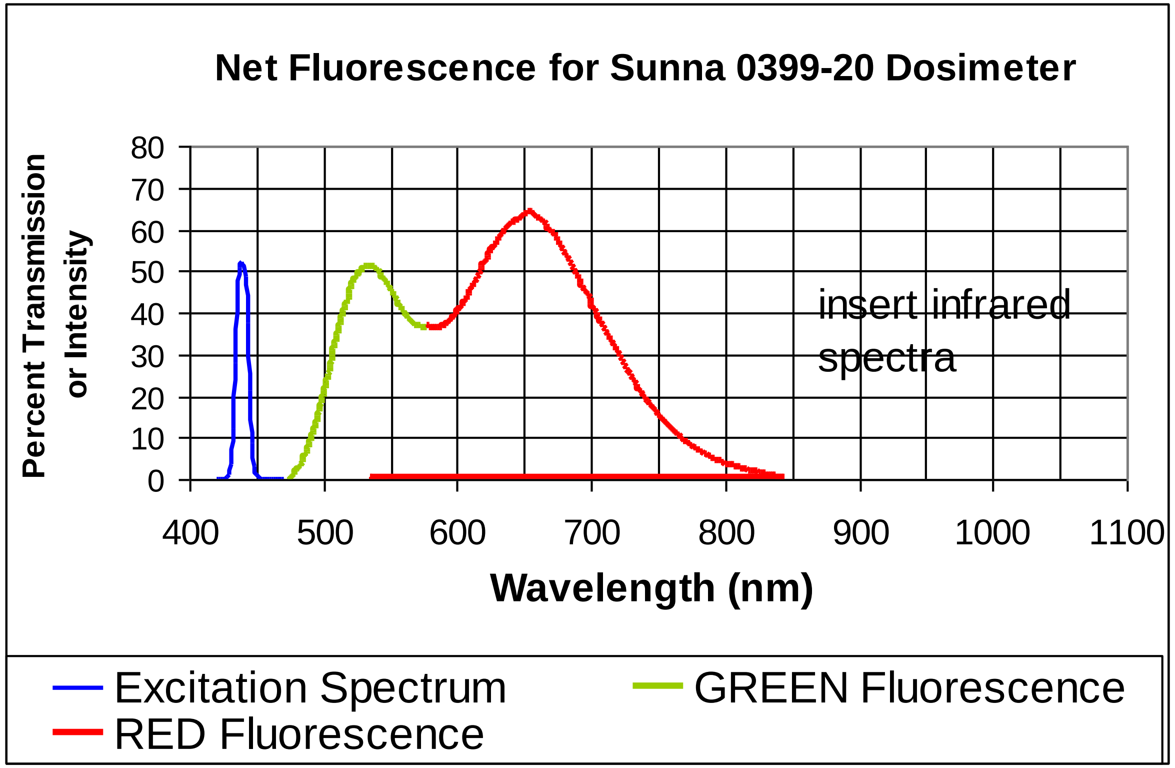
System based on fluorescent measurement – SUNNA dosimeter film

Principles:

LiF dispersed uniformly in PE
(1 cm x 3 cm x 0.4 cm);

Colour centers (F-, M-, N-, R centers) form due to ionizing radiation;

Red, green or IR OSL or UV absorption used for dosimetry;



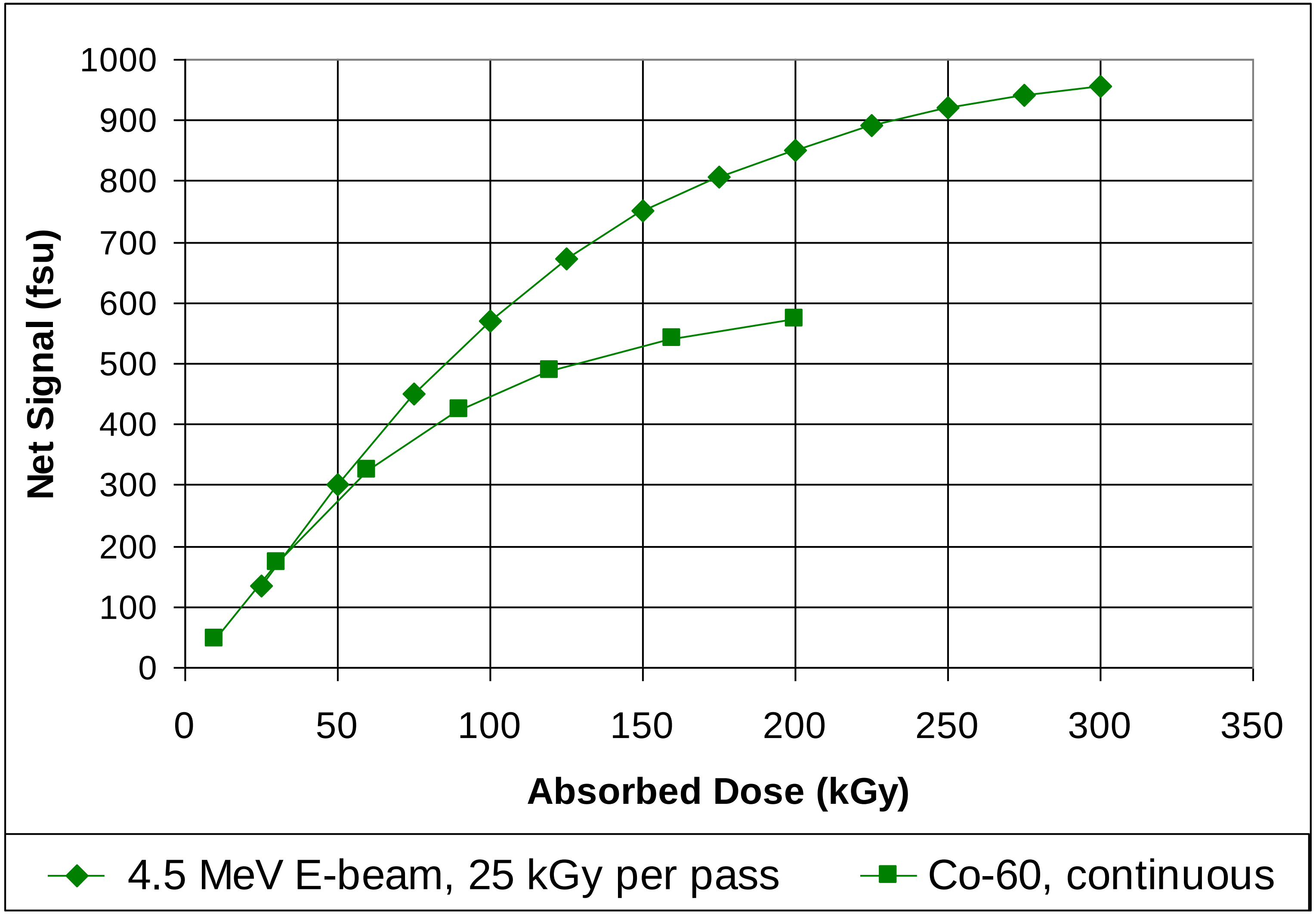
The Sunna dosimeter

Application possibilities:

Evaluation of green OSL (200 Gy – 250 kGy);

Evaluation of UV absorbance (5 – 100 kGy);

Evaluation of IR OSL (10 Gy – 10 kGy);



Environmental Effects on Dosimetry Systems

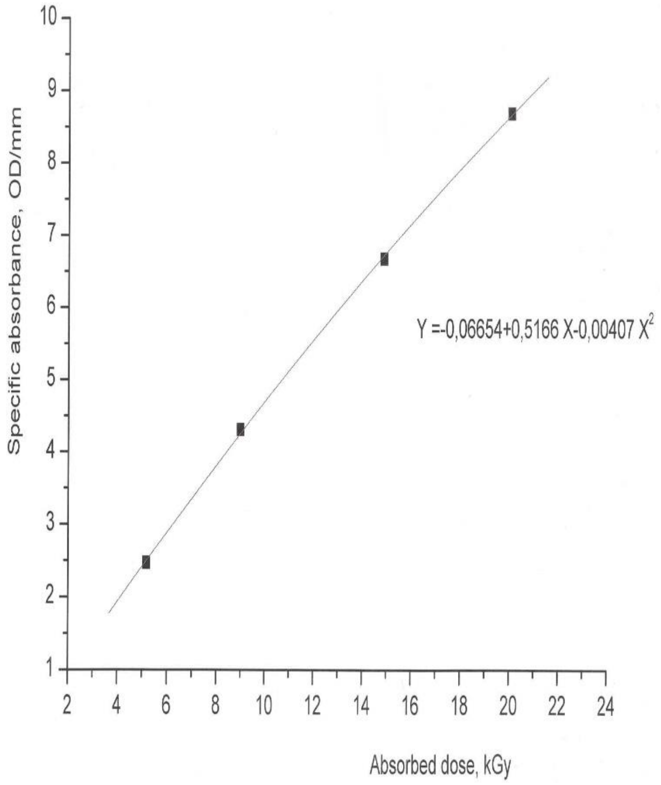
Dosimeter	Measurement time after irr.	Humidity	Dose rate ($Gy\ s^{-1}$)	Irradiation temp. coeff., $(^oC)^{-1}$
Alanine	24 hours	yes	$< 10^8$	+ 0.25 %
Dichromate	24 hours	no	$0.7 – 5x10^2$	- 0.2 %
Ceric-cerous	immediately	no	$< 10^6$	conc. dep.
ECB	immediately	no	$< 10^8$	+ 0.05 %
Calorimeters	immediately	no	$< 10^8$	-
Perspex	24 hours	yes	$< 10^5$	+ 1 %
FWT-60	5 min/60 ^oC	yes	$< 10^{13}$	+ 0.2 %
B3	5 min/60 ^oC	yes	$< 10^{13}$	+ 0.3 %
Sunna	20 min/70 ^oC	no	$< 10^{13}$	+ 0.2 %

Calibration of Dosimeters

Aim of calibration:

Determine relationship between response of a dosimeter and absorbed dose:

Traceability has to be achieved!



Calibration:

1. **Dosimeters;**
2. **Measuring instrument;**

Types of calibration:

1. **In a calibration facility – verification is needed;**
2. **Calibration in-plant – all routine irradiation and calibration irradiation conditions are the same;**

3. Use of calibration phantoms;

The steps of validation as described in the EN ISO 11137 Standard:

- Process definition: establishing maximum acceptable and minimum required dose;
- Installation qualification (IQ): the irradiation facility has been supplied and installed according to its specifications;
- Operational qualification (OQ): to demonstrate the capability of the equipment to deliver appropriate doses;
- Performance qualification (PQ): to measure dose map in real product;

In addition:

- Routine process control: to measure dose at monitoring positions;

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Installation Qualification

EB facility:

To determine beam characteristics

by dosimetry;

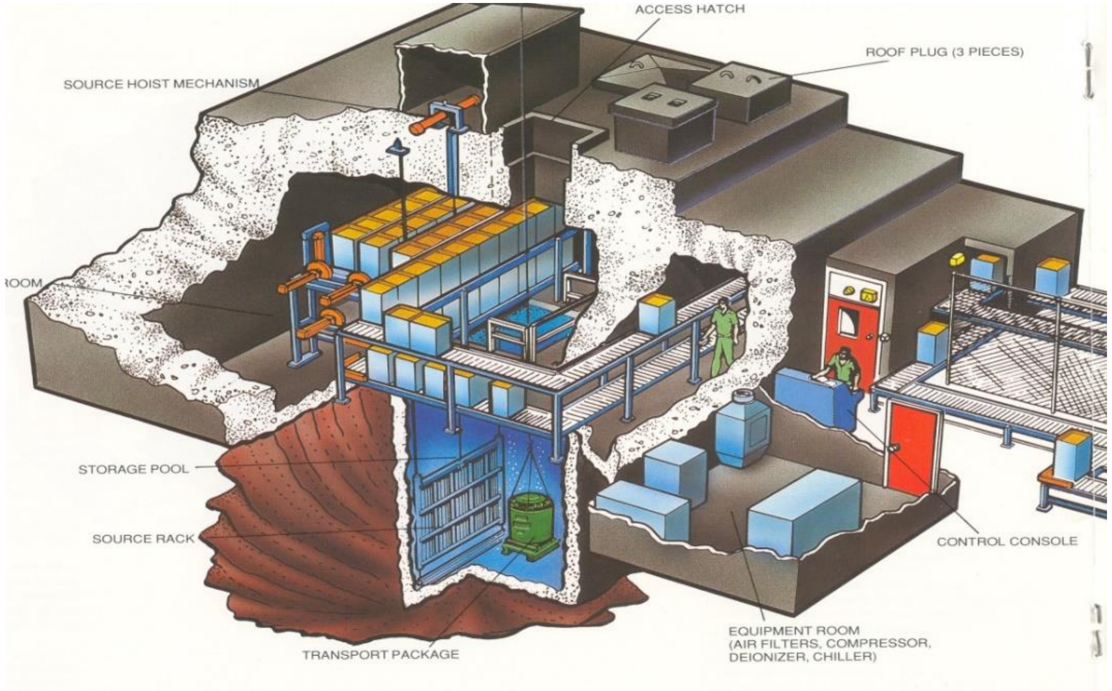
Dosimetry: Calorimeters, ECB, Sunna,

alanine, Gex (B3), FWT-60, dichromate;

Gamma facility:

No specific dosimetry requirements

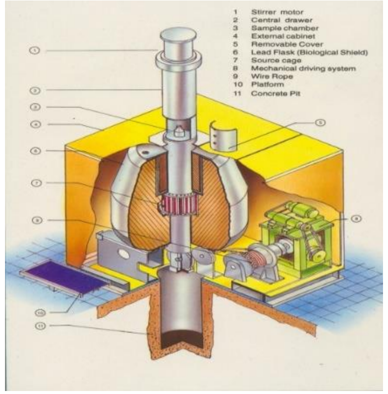
to verify operation within specification;

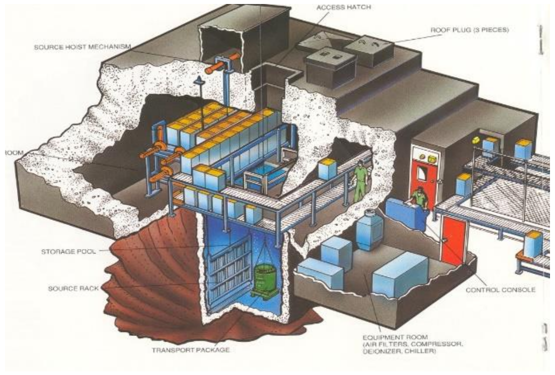


Operational Qualification (gamma facility)

To characterize the irradiation facility relating plant parameters to absorbed dose (in reference product):

Nominal dose vs. irradiation/dwell time*, *dose distribution, process interruption;
Dosimeters: Dichromate, ECB, ceric-cerous, Fricke, Gex (B3), FWT- 60, Perspex, Sunna, alanine;





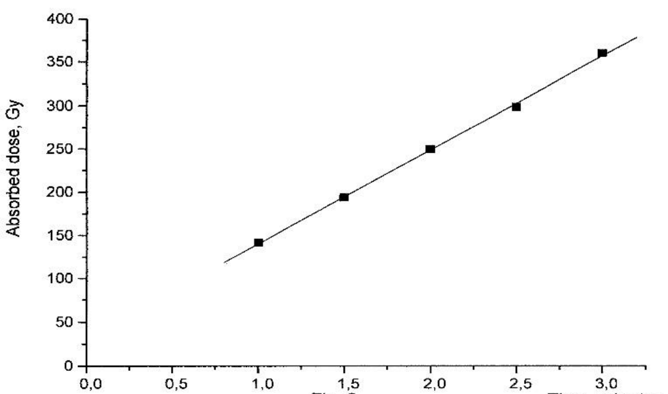
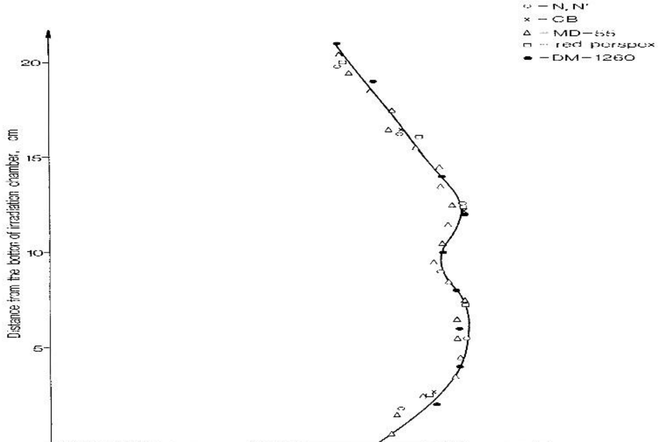
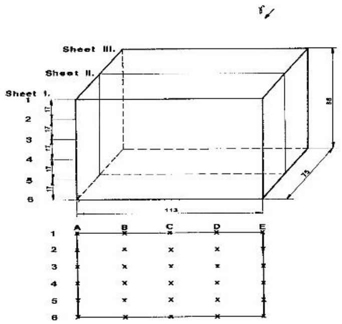
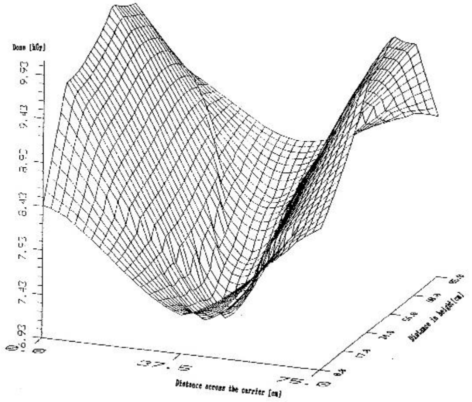


Fig. 2.





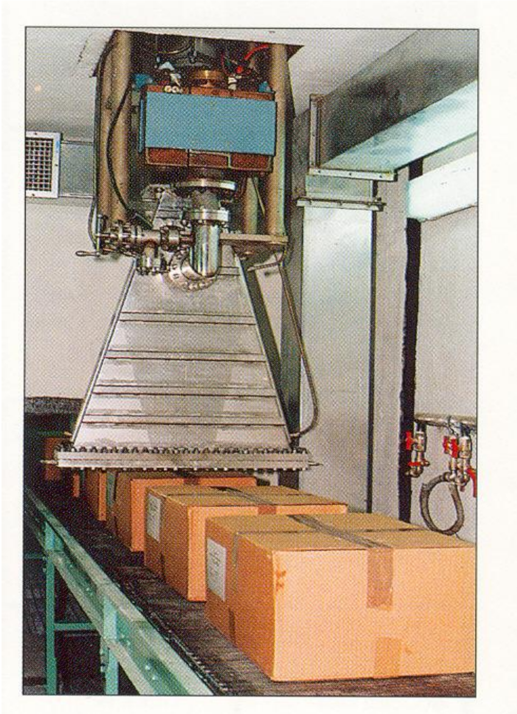


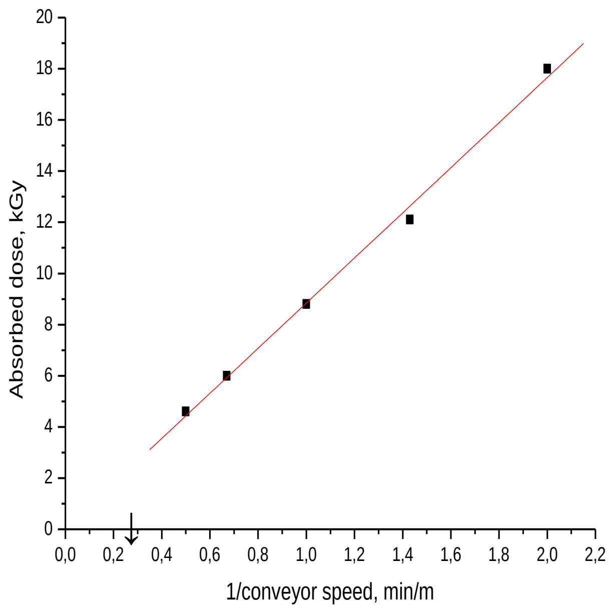
Operational Qualification (EB)

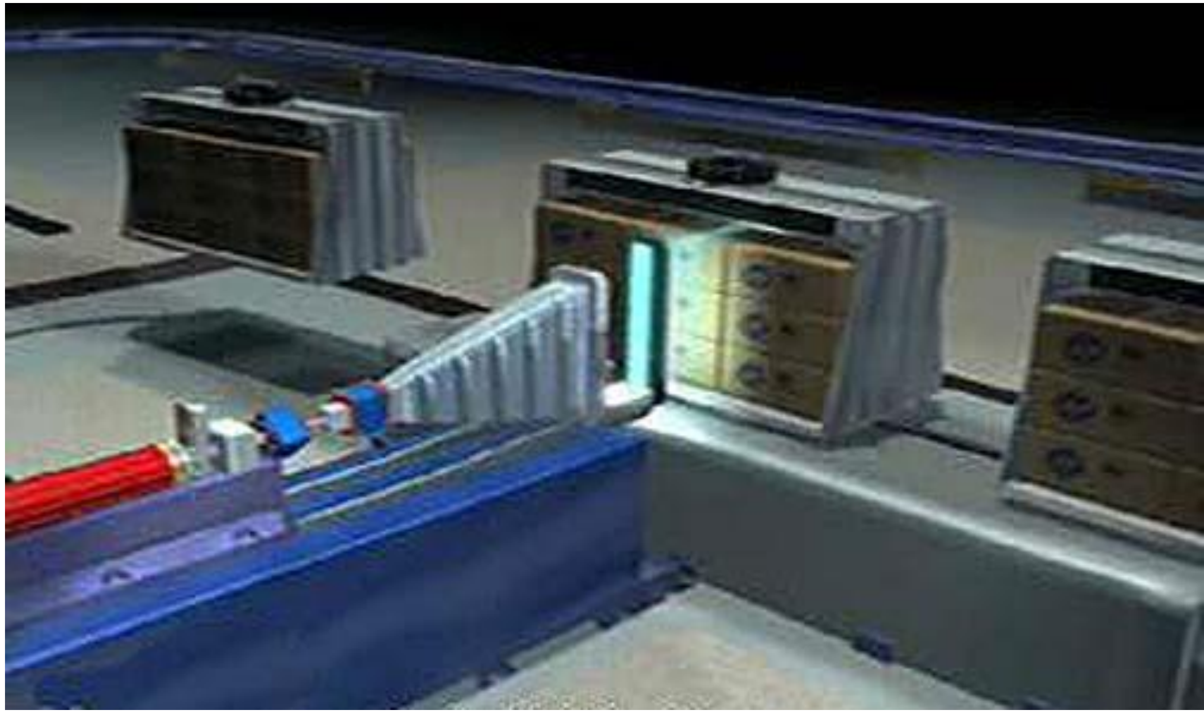
To characterize the irradiation facility relating plant parameters to absorbed dose (in reference product);

Nominal dose vs. conveyor speed/electron current/scanning width; beam homogeneity, scanning width; dose map in reference product, process interruption;

Dosimetry: Calorimeters, ECB, Sunna, alanine, Gex (B3), dichromate;







Operational Qualification (EB)

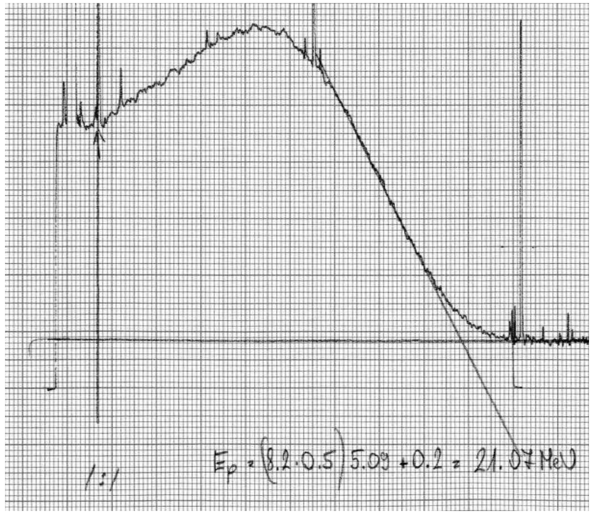
Aim:

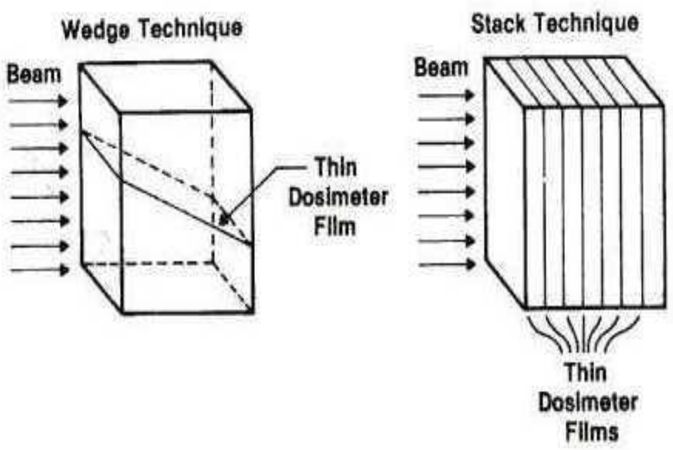
To characterize the irradiation facility relating plant parameters to absorbed dose (in reference product);

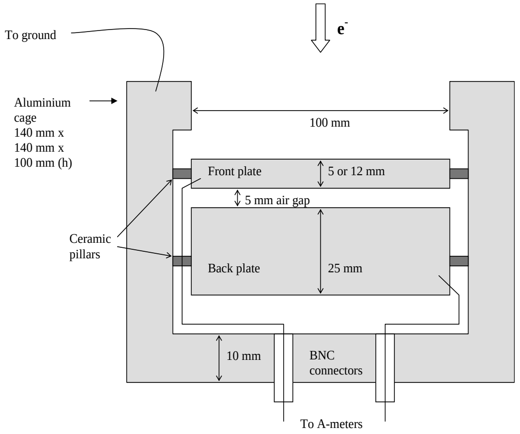
Electron beam facility:

Nominal dose vs. conveyor speed/electron current/scanning width; beam characteristics; dose map in reference product, process interruption, electron energy;

Dosimetry: Calorimeters, ECB, Sunna, alanine, Gex (B3), dichromate;







Performance Qualification

<u>Aim:</u>

1. **To measure <u>dose map</u> in real product in order to locate D_{min} and D_{max} and to establish irradiation conditions according to required specifications, i.e.:**

D(product) > D(required, e.g. sterilization dose)

and

D(product) < D(acceptable)

To map one product container and then to irradiate 8-10 containers with dosimeters in the minimum and maximum dose locations!

2. **To determine relationship between D_{min} and D_{max} and the dose at the routine monitoring position;**

3. **Mathematical modelling to optimize the positioning of dosimeters during dose mapping;**

↓

Process Control

Aim:

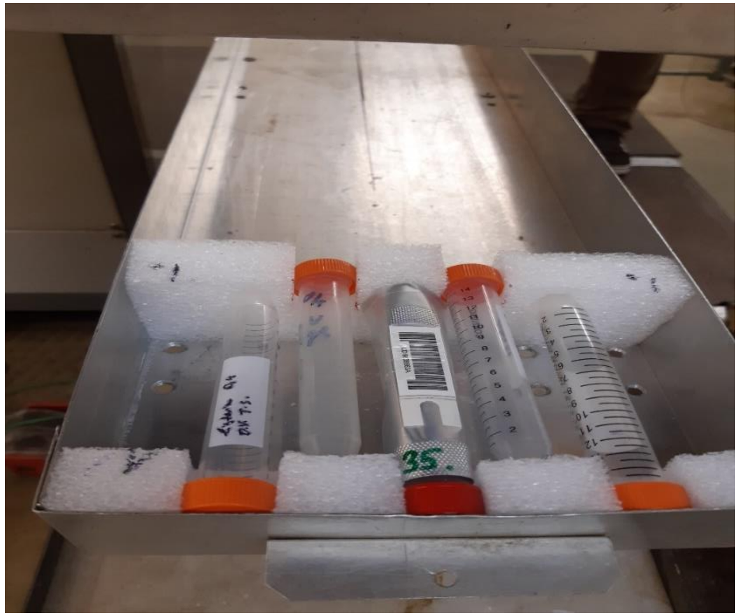
1. **Measurement of process parameters:**

To measure dose at the monitoring position to verify that the irradiation process is within established/required limits

$\Downarrow$

knowing the relationship between D_{min}, D_{max} and $D_{monitoring}$.

Dose measurement frequency: at beginning and end of run (min)



2. **Control and monitoring of operating parameters.**

Controlled parameters:

Electron beam facility:

- **Electron energy**
- **Beam current**
- **Scanned beam width**
- **Conveyor speed**
- **Routine dose**

↓

Gamma facility:

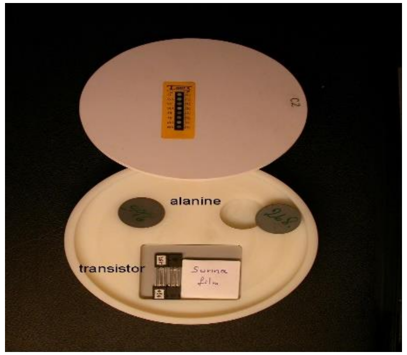
- **Timer setting**
- **Other type of products present**
- **Routine dose (on product boxes)**

Process Control (EB)

Application possibilities:

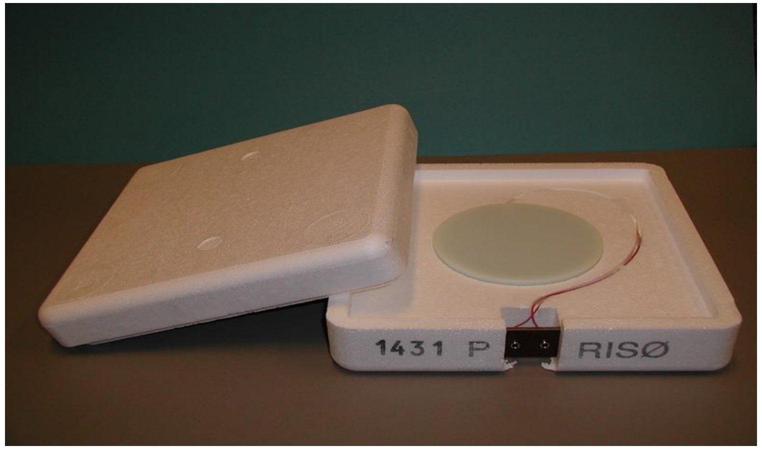
1. Measurement of routine/process dose - on conveyor - quick measurement soon after irradiation :

    - calorimeters;
    - ECB;
    - alanine;
    - Sunna film;
    - FWT-60, B3;



2. Measurement on product at reference location:

    - alanine;
    - FWT-60, B3;
    - Sunna film;

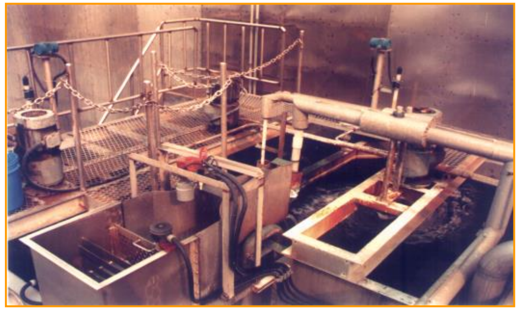


Disadvantage: effect of (varying) product location on the measured dose;

↓

Industrial and municipal waste water treatment;

Drinking water treatment;



Flue gas treatment:
(SO_2 (~ 95 %) and
NO_x (~ 80 %) removal);
(Calorimetry, MC, machine parameters);

Sludge decontamination;
(Perspex, Ceric-cerous solutions, ECB);

Medical hospital waste;
(Perspex, Ceric-cerous solutions, ECB);

Temperature measurement before and after irradiation – Calorimetry;

ESR measurement of non-soluble additives ($BaSO_4$);

Methylene blue bleaching;

Radiation induced decomposition of CCl_4;

Aqueous-alanine solutions – conductivity/oscillometry measurements (1 – 100 kGy);

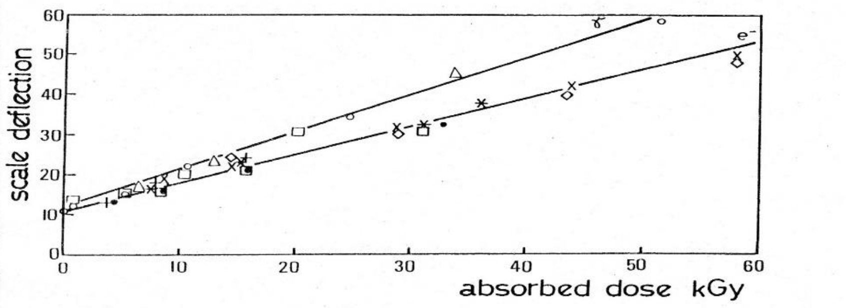
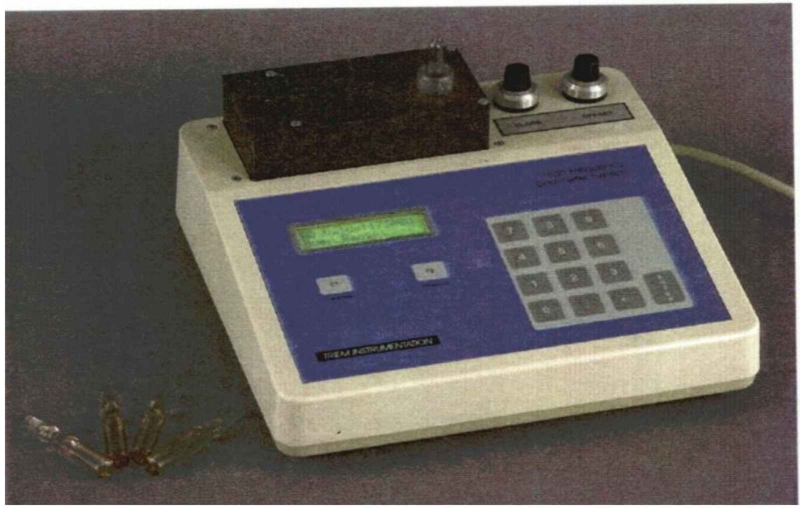


Fig. 2.

Oscillometric response of alanine solutions as a function of dose at various dose rates (^{60}Co irradiation), electron energies and currents (electron irradiation).
^{60}Co irradiation: 0.13 kGy/h: □ ;3.5 kGy/h: △ ;30 kGy/h: ○ ;
electron irradiation: 4 MeV, 2.6 μs, 13 μA: ◇ ; 4 MeV, 2.6 μs, 26 μA: × ; 10 MeV, 0.5 μs, 0.4 A: + ; 10 MeV, 1 μs, 1 A: * ; 10 MeV, 4 μs, 1 A: • ;



Centre for
Energy Research

Thank you for your attention!

Welcome to Hungary!