ALANINE-EPR DOSIMETRY SYSTEM. WHY WE LIKE IT?

Zofia Stuglik

Institute of Nuclear Chemistry and Technology,
Laboratory for Measurement of Technological Doses,
Warsaw, Poland

What we should do to develop a new high-dose dosimeter?

(1) To find material with radiation effect monotonically (*if possible lineary*) dependent on an absorbed dose

(2) To investigate its dosimetric characteristics (*sensitivity, dose range, repeatability, accuracy, post-efects*)

(3) To evaluate economical parameters of new method (*cost and availability of dosimetric material, cost of analytical instrument and its services)*

(4) To evaluate operational features of new dosimeter (*sensitivity for environmental conditions, time from irradiation to the read-out, other ones..)*

(5) To perform a calibration curve = functional dependence between radiation effect (dosimetric signal) and absorbed dose

Short history of EPR α-alanine dosimetry
(EPR ≡ ESR)

1960 - ESR signals in single crystal of alanine [*I.Miyagawa and W.Gordy,J.Phys.Chem. 32 (1960) 255-264*]

1962 – First use of alanine as EPR dosimeter [*W.W.Bradshaw, D.G.Cadena, G.W.Crafford, H.A.Spetzler, Rad.Res., 17 (1962) 11-21*]

1982 – Re-validation of alanine-EPR dosimetry system [D.F.Regulla, U.Deffner, Int.J.Appl.Radiat.Isot., 33 (1982) 1101-1114]

1983 – alanine-EPR dosimeter as transfer dosimeter in radiation processing [D.F.Regula, U Deffner, Radiat.Phys.Chem. 22 (1983) 305-309]

1988 - A polymer-alanine film for dose distribution measurement [I.Janovsky, J.W.Hansen, P.Cernoch, Appl.Radiat.Isot., 39 (1988) 651-659

Standard Practice for Use of an Alanine-EPR Dosimetry System

1/ 1994 → E 1607 - 94 ASTM

1a/ International Standard **ISO 15566:1998(E)**

2/ 1996 → E 1607 - 96 ASTM

3/ 2002 → ISO/ASTM 51607:2002(E)

4/ 2004 → <u>ISO/ASTM 51607:2004(E)</u>

ISO/ASTM 51607:2004(E)

Standard Practice for Use of an Alanine-EPR Dosimetry System[1]

This standard is issued under the fixed designation ISO/ASTM 51607; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision.

1. Scope

1.1 This practice covers materials description, dosimeter preparation, instrumentation, and procedures for using the alanine-EPR dosimetry system for measuring the absorbed dose in the photon and electron irradiation processing of materials. The system is based on electron paramagnetic resonance (EPR) spectroscopy of free radicals derived from the amino acid alanine.[2] It is classified as a reference-standard dosimetry system (see ISO/ASTM Guide 51261)).

1.2 This practice covers alanine-EPR dosimetry systems for dose measurements under the following conditions:

E 668 Practice for Application of Thermoluminescence-Dosimetry (TLD) Systems for Determining Absorbed Dose in Radiation-Hardness Testing of Electronic Devices

2.2 *ISO/ASTM Standards:*[4]

51204 Practice for Dosimetry in Gamma Irradiation Facilities for Food Processing

51261 Guide for Selection and Calibration of Dosimetry Systems for Radiation Processing

51400 Practice for Characterization and Performance of a High-Dose Gamma Radiation Dosimetry Calibration Laboratory

What is alanine?

Simple amino-acid (**$C_3H_7O_2N$**) which can exist in two structural forms: α and β

α-alanine: CH_3-$CH(NH_2)$-$COOH$

dosimetric material

~~β-alanine~~: $CH_2(NH_2)CH_2$-$COOH$

unconvenient for dosimetry because <u>low stability of radicals</u>

α-alanine: CH_3-CH(NH_2)-COOH exist in three structural forms:

- L-α-alanine (optically active, **557 Euro/kg**)
- D-α-alanine (optically active, **3220 Euro/kg**)
- DL-α-alanine (racemat, **113 Euro/kg**)

All forms can be used in technological dosimetry but because of economical reasons the most suitable is DL-α-alanine.

DL : L : D = 1 : 5 : 28

Crystalline α-alanine

$\cancel{CH_3\text{-}CH(NH_2)\text{-}COOH}$

$CH_3\text{-}CH(NH_3^+)\text{-}COO^-$

zwitterion

very stable structure, melting point: 314,5 ℃

FW 89,06

$CH_3\text{-}CH(NH_3^+)\text{-}COO^-$

chemical bond	energy (eV)
C-H	4,3
N-H	4,0
C-O	3,6
C-C	3,5
C-N	3,0

The weakest bond is C-N and it is broken during the radiolysis giving the NH_3 (*amonium, stable molecule*) and SAR radical

microcrystalline α-alanine

$$CH_3\text{-}CH(NH_3^{+})\text{-}COO^{-}$$

$$\Big\downarrow \; e^{-} \text{ fast electron}$$

$$CH_3\text{-}C^{*}H\text{-}COO^{-} \quad + \quad NH_3$$

SAR

Stable **A**monium **R**adical

(paramagnetic specie)

AMMONIUM

stable chemical molecule

On the base of this very stable (years) SAR radical generated in crystalline α-alanine it is established

an alanine-EPR dosimetry system.

EPR signal in microcrystalline α-alanine

magnetic field [mT]

Dosimetric signal

EPR signal presented before is the first derivative of the absorption band.

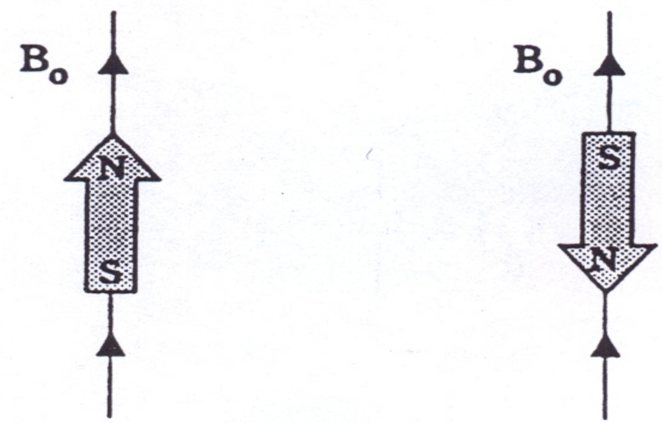
The quantity proportional to an absorbed dose is number of radicals. Number of radicals is proportional to double integral of presented before EPR signal.

Nevertheless, double integration of EPR signal is not accurate procedure and in dosimetry practice an amplitude of the highest line of EPR signal is used as dosimetric signal.

EPR measurements

The method was developed at 1941 year by Zavojskij

and is connected with Zeeman effect.



Minimum and maximum energy orientations of μ with respect to the magnetic field B_0.

Zeeman effect

Unpaired electron *(for instance in radical)* has a magnetic moment (spin).

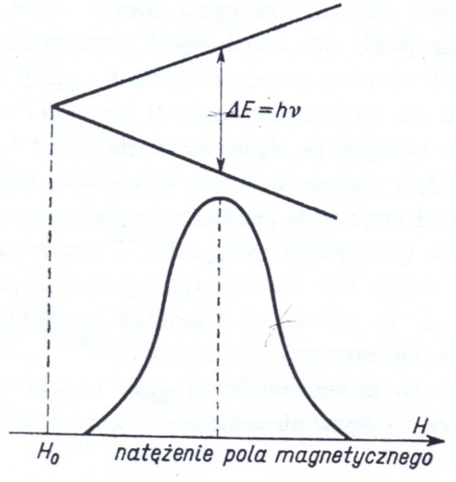
In a magnetic field B_o the moment of electron μ

- can be aligned to the magnetic field (a state with lowest energy),
- can be aligned against to the magnetic field (a state with highest energy)

The difference between them depends linearily on the magnetic field.

EPR

When we use not only magnetic field but also microwave field with the frequency ~10 GHz (X range, λ~3cm) we can observe an absorption of microwave energy by spin population with lower energy → EPR signal generation.

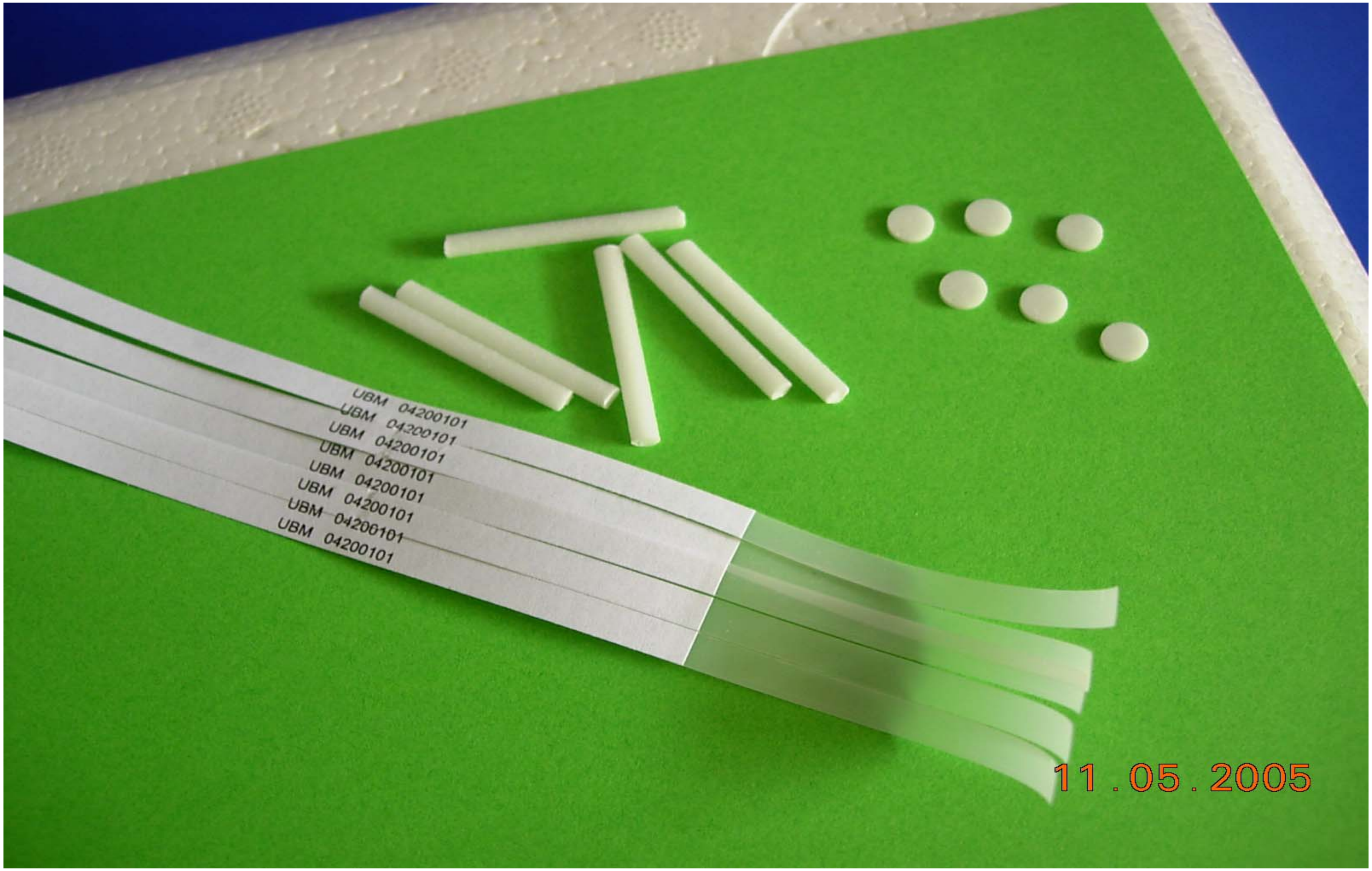


Powstawanie absorpcyjnego widma rezonansowego spinów elektronowych

What is necessary to establish dosimetry system ?

1. Radiation sensitive material =dosimeter
2. Analytical instrument for dosimetric signal measurement
3. Calibration curve = signal-to-dose dependence

Different kinds of alanine-EPR dosimeters

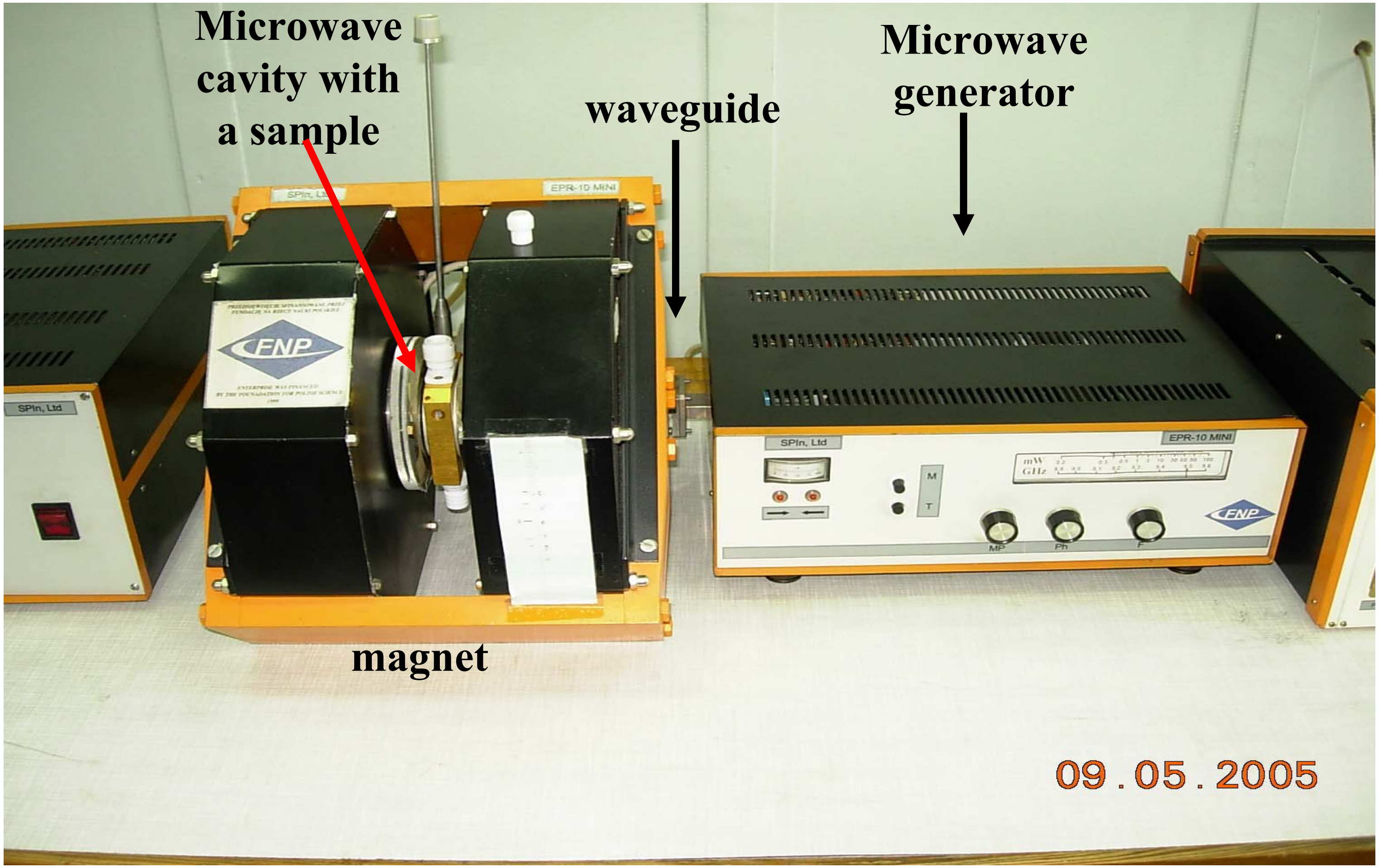


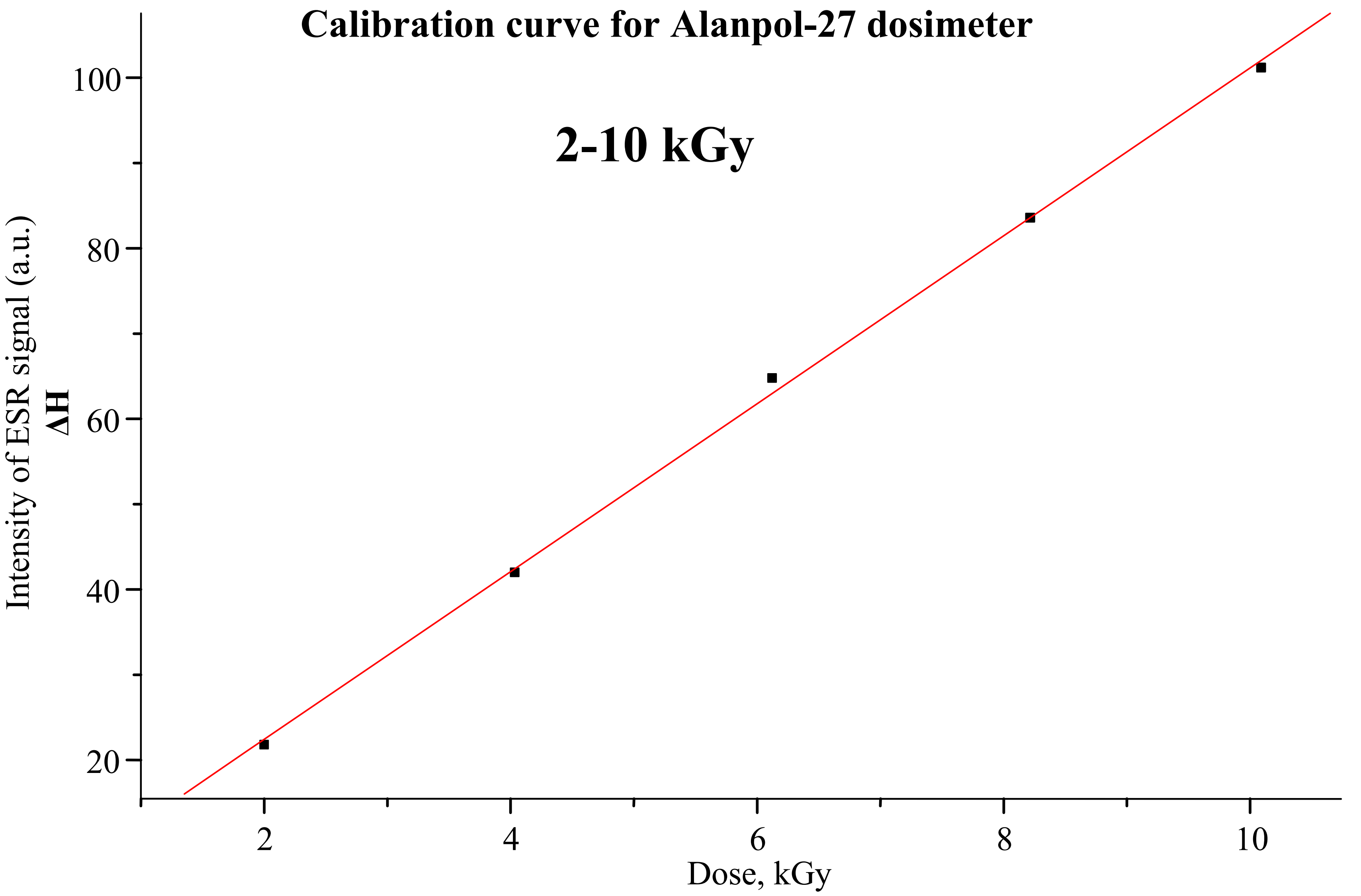
Kodak alanine-EPR film dosimeter

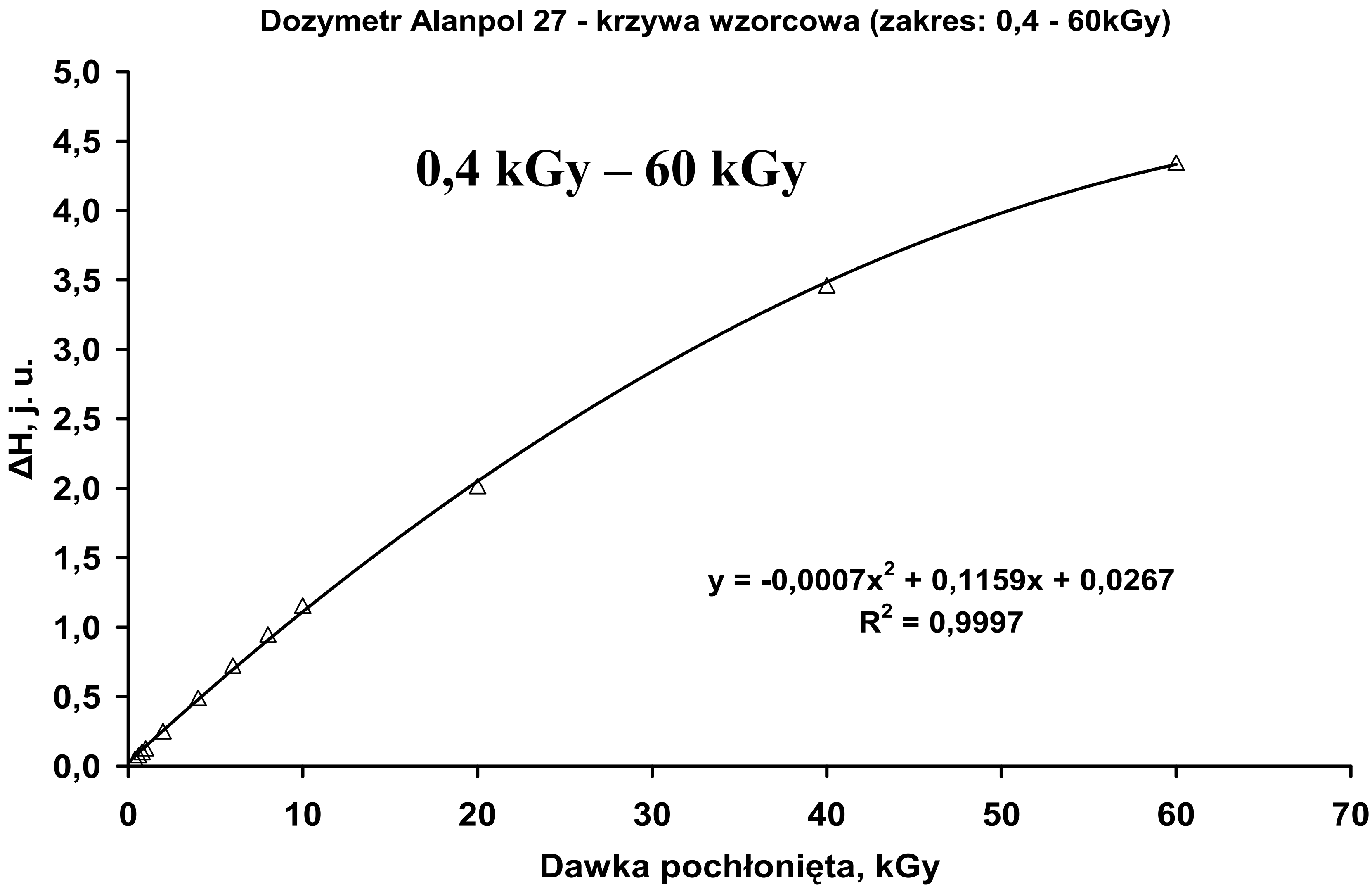
Spectrometer

of **E**lectron **P**aramagnetic **R**esonance

(**EPR** ≡ ESR)




Calibration curve for Alanpol-27 dosimeter
2-10 kGy
Intensity of ESR signal (a.u.)
ΔH
100
80
60
40
20
2
4
6
8
10
Dose, kGy



Dozymetr Alanpol 27 - krzywa wzorcowa (zakres: 0,4 - 60kGy)
0,4 kGy – 60 kGy
y = -0,0007x2 + 0,1159x + 0,0267
R2 = 0,9997
ΔH, j. u.
Dawka pochłonięta, kGy
5,0
4,5
4,0
3,5
3,0
2,5
2,0
1,5
1,0
0,5
0,0
0
10
20
30
40
50
60
70

Advantages of alanine-EPR dosimetry system

- dosimetric signal is stable during very long time (years)
- wide dose detection range (1Gy - 100 kGy)
- linear signal-to-dose dependence (≤ 10 kGy)
- Dosimetric signal is energy and dose rate independent (*gamma and electron beams*)
- accuracy
- non-destructive and fast detection method
- low temperature coefficient of irradiation
- different shapes of dosimeters, also films
- dosimeters are not very expensive
- easy to handle
- non-toxic
- chemical composition of dosimetric material is similar to the chemical composition of organic matter

Disadvantages of alanine-EPR dosimetry system

EPR signal

- Sensitive to light (for long time illumination)
- Sensitive to water and humidity

EPR spectrometer

- Expensive
- Market monopolized by one producer

Small EPR spectrometers for dosimetry (20 000 - 60 000 Euro)

RZECZPOSPOLITA POLSKA

Urząd Patentowy Rzeczypospolitej Polskiej

(12) **OPIS PATENTOWY** (19) **PL** (11) **194369**

(13) **B1**

(21) Numer zgłoszenia: **338997**

(22) Data zgłoszenia: **14.03.2000**

(51) Int.Cl.
G01T 1/04 (2006.01)

(54) **Alaninowo-polimerowy dozymetr promieniowania jonizującego**

(43) Zgłoszenie ogłoszono:
24.09.2001 BUP 20/01

(45) O udzieleniu patentu ogłoszono:
31.05.2007 WUP 05/07

(73) Uprawniony z patentu:
Instytut Chemii i Techniki Jądrowej,Warszawa,PL

(72) Twórca(y) wynalazku:
Zofia Stuglik,Warszawa,PL
Teresa Bryl-Sandelewska,Warszawa,PL
Krzysztof Mirkowski,Legionowo,PL

(57) 1. Alaninowo-polimerowy dozymetr promieniowania jonizującego w kształcie pręta, **znamienny tym**, że stanowi jednorodną mieszaninę DL-α-alaniny, polietylenu i wosku polietylenowego.

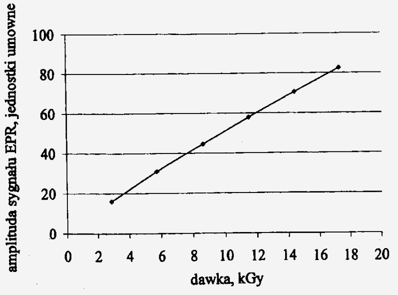


Fig.1

PL 194369 B1

IAEA Training Course, 3-7 December 2007, Warsaw, Zofia Stuglik

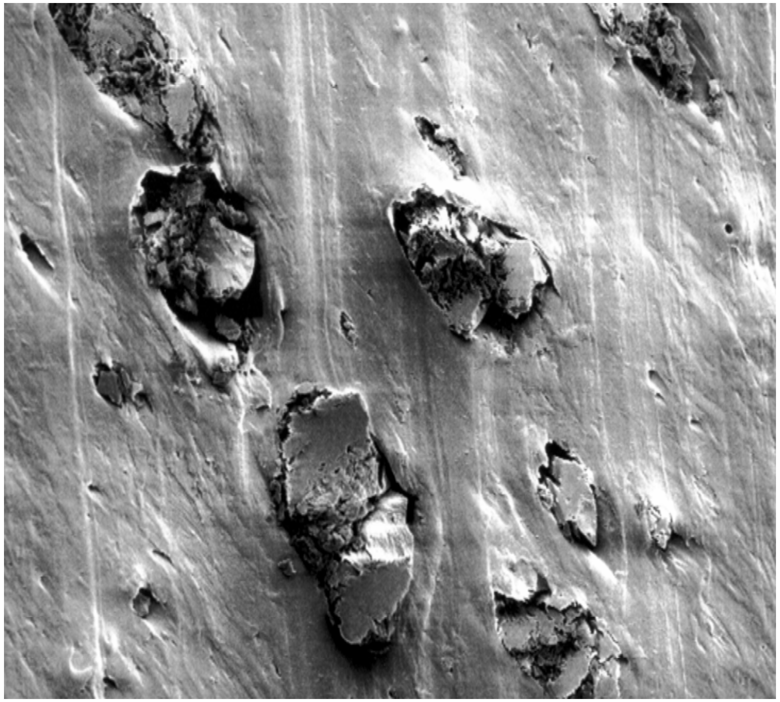
Alanpol dosimeter

Characteristic features:

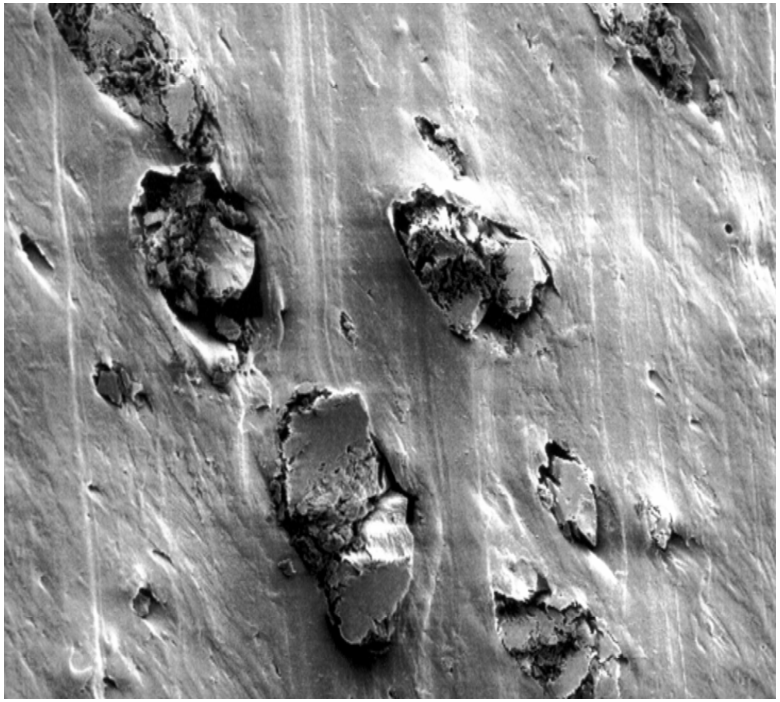
•Low concentration of DL-α-alanine (10 - 30%)

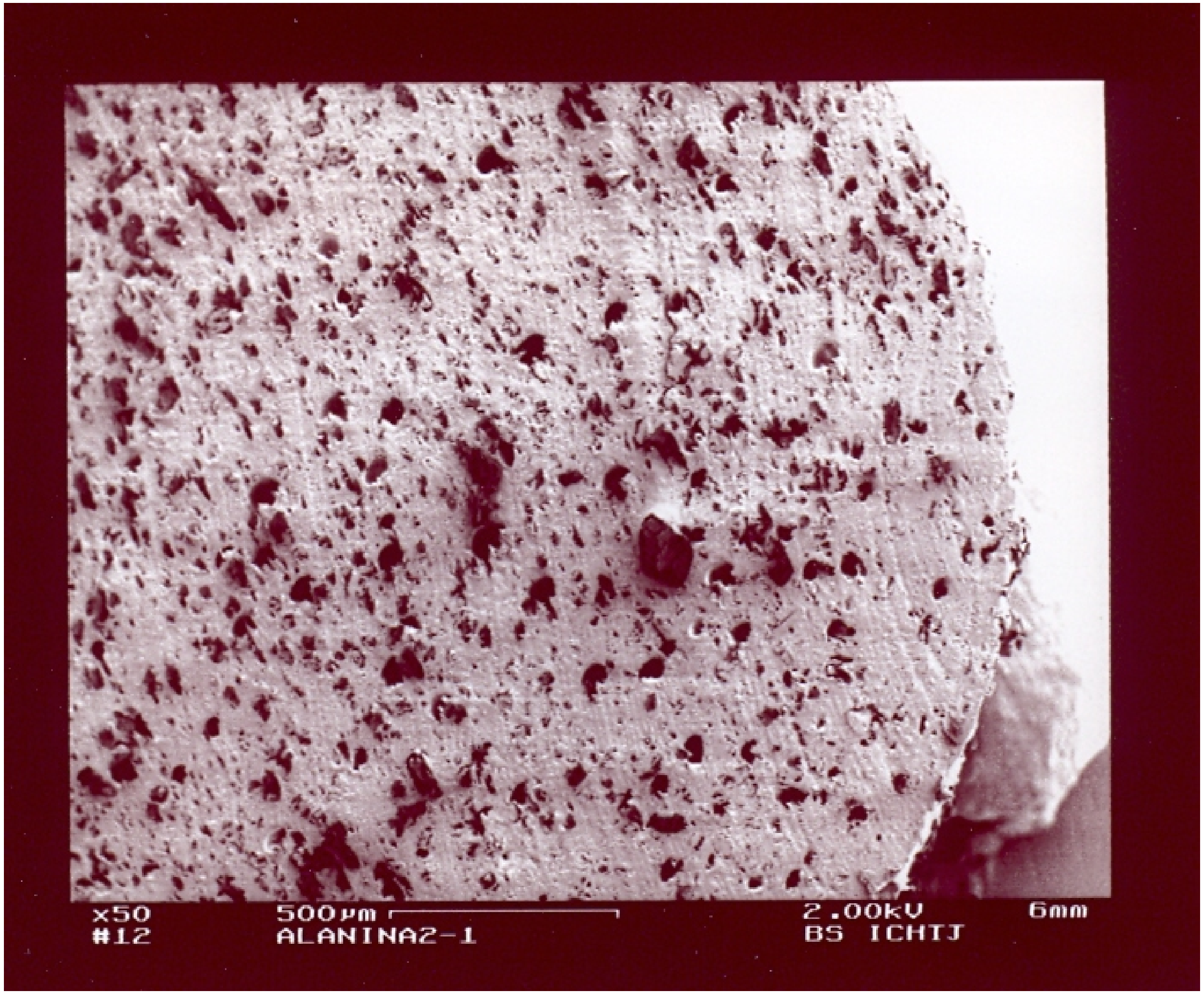
•Hydrofobic polymer as a matrix

Alanpol dosimeter



Alanpol dosimeter




x50
#12
500µm
ALANINA2-1
2.00kV
6mm
BS ICHTJ

Ana Belchior

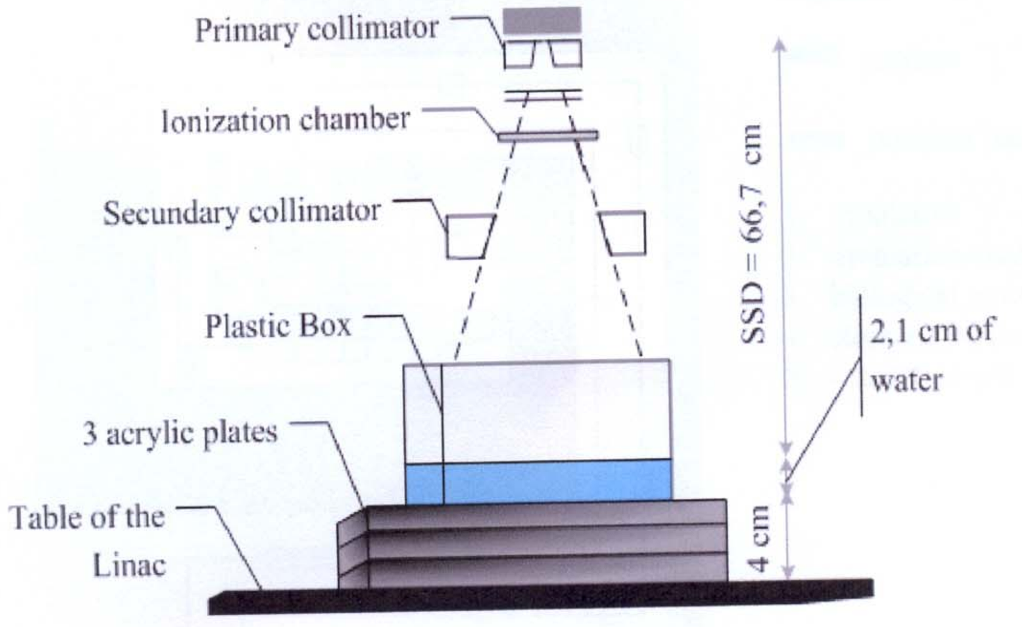


Figure 2 – Scheme of irradiation

Ana Belchior

maximum dose rate is 0, 69[1] kGy/h. The values chosen for this experiment were established by the minimum absorbed dose of ALANPOL® (0.4 kGy) and by the maximum operating time of the LINAC, which corresponds to a maximum dose of about 2, 5 kGy.

Set Package: The three sticks of ALANPOL® were packaged separately inside a plastic recipient (20 x 20 cm^2)(figure 1) to form a triangle in order to maximize the uniformity of the dose.

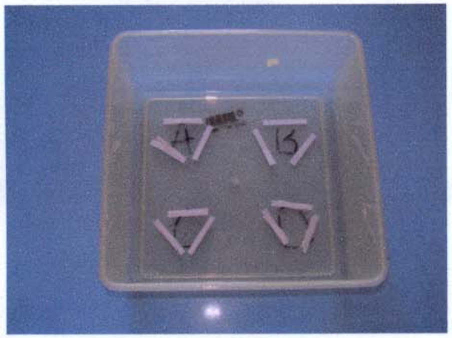


Figure 1 – Illustration how the dosimeters were placed in the plastic recipient.

To obtain a dose rate of 0,69 kGy/h the sets of ALANPOL® were irradiated inside of water which depth was approximately of 2 cm^2. Because of the backscattering radiation effect, 3 acrylic blocks were placed under the plastic recipient. Figure 2 illustrates the scheme of irradiation, including the main components of the LINAC, other devices used in the irradiation processes, the distance between the source and surface (SSD), and the height of the acrylic plates.

INSTITUTE OF NUCLEAR CHEMISTRY AND TECHNOLOGY
LABORATORY FOR MEASUREMENTS OF TECHNOLOGICAL DOSES
ul. Dorodna 16, 03-195 Warsaw, Poland
tel. (+4822) 811 30 21 ext.1053; fax: (+4822) 811 15 32
e-mail: lpdt@orange.ichtj.waw.pl; stuglik@ichtj.waw.pl

AB 461

PROTOCOL No 6-05-E

DOSE MEASUREMENTS BY MEANS OF ALANPOL®-A TRANSFER DOSIMETER IN ACCORDANCE WITH ISO/ASTM 51607:2002 STANDARD AND INCT/LMTD PROCEDURES: PT-7 AND PT-8

Data of issue	23.01.2006
Customer	Dr. M.Luisa Botelho Instituto Technologico e Nuclear, Sector da Fisica Estrada Nacional No.10 Apartado 21 2686-953 Sacavem Portugal
Transfer dosimeters	ALANPOL®-A, batch I, No 1a, 1b, 1c, 2a, 2b, 2c, 3a, 3b, 3c, 4a,4b, 4c and 0a, 0b, 0c (control)
Date of departure	16.06.2004
Date of irradiation	24.05.2005
Date of arrival	24.08.2005
Date of read-out	27.09.2005
Read-out operator	Sławomir Fabisiak, Ing.
Identification	Irradiated dosimeters were delivered in envelopes on which the irradiation conditions was described. Before the read-out, an identification numbers (1a, 1b...) were written on the dosimeter sticks with the waterproof ink.
Irradiation	Linear Accelerator (Precise™ - Elekta™) located in CUF-Descobertas - Lisbon, Portugal and used for clinical purposes. The energy of electron beam: 10 MeV. The maximum dose rate: 0,69kGy h^{-1}.
Irradiation temp.	not measured

Checked by: 2P

Irradiat. conditions: ALANPOL®-A dosimeters were irradiated in water filled plastic vessel, under ~2cm layer of water. 3 acrylic plates were placed under the plastic vessel to minimize the backscattering effect. The target doses were 0,5; 1,0; 1,5 and 2,5kGy. Dose rate was determined by ionization chamber and confirmed by Fricke dosimeter.

Results

Table 1 Absorbed doses measured by means of ALANPOL®-A dosimeters

Time of iradiation h	D_{nom} kGy	D_t kGy h^{-1}	Contact with water	No	D_{raw} kGy	D kGy	D-D_{nom} %
0,72	0,5	0,69	1h 31min	1a	0,497		
				1b	0,497		
				1c	0,484		
				mean value	**0,493**	**0,508**	**1,6**
				standard deviation	**0,008**		
				CV(%)	**1,6**		
1,44	1,0	0,69	2h 40min	2a	0,976		
				2b	0,972		
				2c	0,989		
				mean value	**0,979**	**1,009**	**0,9**
				standard deviation	**0,009**		
				CV(%)	**0,9**		
2,17	1,5	0,69	3h 42min	3a	1,470		
				3b	1,477		
				3c	1,426		
				mean value	**1,458**	**1,503**	**0,2**
				standard deviation	**0,027**		
				CV(%)	**1,9**		
3,62	2,5	0,69	5h 47min	4a	2,385		
				4b	2,430		
				4c	2,423		
				mean value	**2,412**	**2,487**	**-0,5**
				standard deviation	**0,024**		
				CV(%)	**1,0**		

D_t = dose rate
D_{nom} = target dose
D_{raw} = absorbed dose (dose to water) before correction of systematic error
D = measured dose (dose to water) after correction of systematic error

Checked by: Zf

An experiment

After lunch we will have small experiment with different kinds of alanine-EPR dosimeters

We will started with 9 alanine-EPR dosimeters Alanpol and 9 film dosimeters from Gamma Service, East Germany

All dosimeters were irradiated with 10 MeV electrons to the same dose 25 kGy

Each group will have 3 dosimeters of each kind

Group I is going with me to EPR Laboratory to observe EPR measurement of dosimetric signal

An experiment

Group II is going with Mr. S.Fabisiak to LMTD. This group will place its dosimeters in exicator (humidity ~100%) (1/2 h)

Group III is also going with Mr. S.Fabisiak to LMTD. This group will put its dosimeters to glass of water (½ h)

After half of hour the dosimeters both groups will go with Mr. Fabisiak to Department VII, to EPR Laboratory for EPR measurements

EPR measurements will be done by **Dr. Jarosław Sadło**

The results of all measurements will be given to the participants today or tomorrow.

You will able to observe that the sensitivity of different alanine dosimeters for environmental condition is different

We expected that signal in Alanpol will be practically the same for all dosimeters.

EPR signals in GS films will strongly depend on the storage conditions.