ALANINE/ESR DOSIMETRY FOR MEASUREMENT OF ELECTRON DOSES IN INDUSTRIAL APPLICATIONS*

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ABSTRACT

In this report, the results of preliminary research on L-alanine/ESR electron dosimeters are described. They include the designs of modelling tools and experimental capsules, the preparation and irradiation of very thin dosimeters, the measurement of ESR signal spectrum and the calibration method. The physical, chemical and dosimetric properties of the alanine free radical dosimeters, such as the smallest thickness, average density, lowest detectable limit, repeatability of the method, reproducibility resulted in directivity and location in resonance cavity, linearity regions as well as relative scattering of the response to identical doses, are summarized.

Keywords: Alanine/ESR dosimeter Electron dose

1 INTRODUCTION

The great objective of the IAEA has been encouraging to develop the peaceful uses of atomic energy. Among other things the industrial radiation processing is becomming more and more increasing in the world at present. The Accelerator Department of the China Institute of Atomic Energy therefore decided to reform the three kinds of available accelerators in 1986: a 600 kV Cockoroft-Walton Generator, a 2.5 MV Van de Graff, and a 14 MeV Electron LINAC. In the past, these accelerators had only been used for research in low energy nuclear physics.

So far as the radiation processing is concerned, the measurement of absorbed dose at a high level is quite important. Though as early as 1970's an ionization chamber system satisfying the needs of high level gamma dosimetry for production of artificial radioisotopes was developed[1], the electron dose in radiation processing has not been involved. For development of routine and transfer dosimeters used in the field, the present project on alanine/ESR free radical dosimetry-a new dosimetric method of resonant absorption of electromagnetic energy in paramagnetic substances, had been

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approved and supported by the IAEA and IAE of China in 1987. The results are here in summarized as follows.

2 OUTLINES OF PRINCIPLE FOR THE ALANINE/ESR DOSIMETRY

Alanine comes close to tissue equivalent with respect to atomic compositions. So that such a dosimeter may be considered a tissue equivalent (TE) chamber.[2] There are two kinds of alanines. One of them is β－alanine. Its radiation chemical yield is very low and ESR (electron－spin resonance) spectrum varies with the time after irradiation.[3] Hence the L－ or DL－ α alanines are used for radiation dosimetry. The L－alanine dosimeter has a composite structure of L－alanine with paraffin. The sensitive material to ionizing radiation is L－alanine, and paraffin is binder. The principle of detection is the effect of radiation on the disruption of covalent bonds formed by electrons with opposite spin in paramagnetic substances. This result in two paramagnetic parts each with an unpaired electron. These parts are called free radicals. They are produced in alanine by ionizing radiations, their types and amount generally depend on the crystal structure of the material interested, temperature as well as absorbed dose. The free radicals $CH_3-\dot{C}H-COOH$ formed from the disruption of C－H bonds are predominant at room temperature, and have a long lifetime. Hence, the stability of the free radical dosimeter in organic solid crystalline alanine is excellent. On the basis of bond－breaking effect the relative amount of the radicals in an alanine/ESR dosimeter can be evaluated, so as to determine the absorbed doses.

3 PROCEDURES OF THE EXPERIMENT

The alanine/ESR electron dosimetry system includes four major components, i.e. preparation of alanine detectors, irradiation, evaluation by an ESR spectrometer and appropriate calibration procedure.

This project includes either the design of modelling tools and sample capsules, the exposure of alanine dosimeters to different dose levels, the measurement of first derivative spectra of paramagnetic absorption by means of an ESR spectrometer, and the calibration for electron absorbed doses.

3.1 The design of modelling tools

Table 1

The hole dimensions of modelling tools used for preparation of the alanine dosimeters

Size	Diameter (mm)	Area (cm^2)	Hight (cm)
1	4.45±0.0015	1.56±0.01	2.0
2	3.67±0.0017	1.06±0.01	2.0
3	2.78±0.0025	6.06±0.01	2.0
4	1.87±0.015	2.76±0.01	2.0

For preparing the alanine dosimeter the first step was to try to design the

modelling tools. Their dimensions depend on radial and axial extensions of microwave cavity of the ESR spectrometer. At present the common ESR spectrometers permit to evaluating the samples with diameters less than 5 mm. Four sizes can be chosen freely and are listed in the Table 1. The features of modelling tools show in Fig.1.

3.2 The design of sample capsules

The lateral thickness of sample capsules must stop the scattered electrons from outside. It is well known that the most energy of an electron is depleted in first one-third path as it passes throughout any material. The thickness of 1.5 cm has been chosen so as to stop the scattered electrons with energies up to 3 MeV from outside. Depending on the energy of incidence electrons the frontal thicknesses have

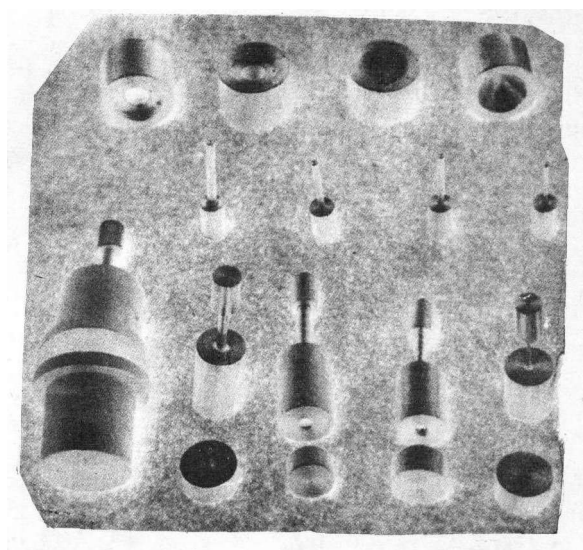

Fig.1 The modelling tools

Table 2

The typical physical and chemical properties of the present alanine dosimeter

Sensitive material to radiation: L-alanine 80 % by weight
Effective atomic number: $Z=7.2$ (approximate to tissue $Z=7.46$ and water $Z=7.42$).
Predominant radical type generated by radiation: $CH_3-\dot{C}H-COOH$
G-value for gamma and electron radiations: 3 free radicals per 100 eV absorbed energy.
Binder: Paraffin with ceresin used for embedding and sectioning, 20 % by weight, melting point 60—62 °C.
Practical mass density of the detectors: 1.15 g/cm^3.

alterability. The minimum layer is a black plastic film. Various thicknesses of black plexiglass can be piled in layers and placed in front of the alanine detectors in order to obtain the absorbed doses at different depths in materials of interest. The hole's diameter of the capsule equals to that of the dosimeters. The features of sample capsules show in Fig.1.

3.3 The preparation of L— alanine dosimeters

This reagent in form of crystalline powder is commercially obtained from the Reagent Manufacture No.3, Shanghai. From its specifications the purity is chromatographic grade and rotation qualification. The L-alanine is used without further purification. The typical physical and chemical properties are listed in the Table 2.

The preparation of the dosimeters includes three steps: Firstly, the pure polycrystalline L-alanine powder is grained and sieved. Then the two kinds of components (alanine and paraffin with appreciate proportions) are mixed up in a oven with appreciate temperature in order to get homogenious mixture and act as a basic material. Finally, the quantitative blended material by weighing is filled and compressed manually in the modelling tools. Different proportions between the L-alanine and paraffin as well as workability during preparation may lead to different mass densities (i.e. different electron densities) and surface smoothness of the detectors. This results in different sensitivities. It was finally found 80 % L-alanine and 20 % paraffin by weight are suitable. For measurement of electron absorbed dose the thinner dosimeters, i.e. smaller dimensions in the direction of incidence radiations, are preferred. But it brings about a lot of problems during the preparation and appears a poor mechanical rigidity and stability. Hence the effort has been made to overcome these difficulties in the preparation of thin dosimeter. Again, the amount of free radicals for the thinner dosimeters is much less, and the ESR signals therefore are smaller. The lowest limit would be restricted by ratio of the signal to noise of ESR spectrometer. From these criteria the thicknesses of alanine dosimeters used for measurement of electron absorbed dose have been chosen to be 0.5—1 mm. Its accurate dimensions were determined by a depth thousandth dial indicator. By careful treatment with appropriate contents of L-alanine the minimum thickness of about 0.5 mm is obtained. Because the decrement during preparation procedure was in existence, these detectors prepared must be weighed, and classed according to their mass for an identical diameter.

3.4 Calibration and measurement

Irradiation of the alanine dosimeters is performed with a $^{90}Sr/^{90}Y$ source of 1.0 GBq, which has been calibrated in terms of absorbed dose rate in polystyrene material by an extrapolation ionization chamber. The absorbed dose rates in different conditions and electron spectrum of the rotating radiation field are all well known. The fluence rates are relative uniform and precisely reproductable. The maximum dose rate is about 400 Gy/h.

The dosimetry method based on alanine/ESR is a relative method. The signal-to-dose conversion doesn't need to provide the values of absolute spin concentrations. As the experiments have indicated that the ESR spectra at different dose levels are in the same shape as shown in Fig.2, secondary radical reactions are

negligible and no any change in the hyperfine structure of the ESR spectrum with absorbed doses. The later can be directly derived from relative amplitude of the first derivative signal of ESR spectrum to that of a reference dosimeter calibrated in a radiation field with known dose rates. The reference detector has such identical properties and dimensions as those in routine use. Thus the simplification of measurement is achieved.

Hight of the peak-to-peak value represents absorbed dose. The measurement of electron spin resonance spectrum is carried out with a Bruker ESR Spectrometer. The normal operation conditions are as follows: operating in x-band, central magnetic field intensity 348 mT, field scan range 4 mT, microwave power 2 mW, field modulation intensity 0.1 mT, time constant 100 ms, scan time 100 s. All the measurements are made at room temperature (22—24 °C).

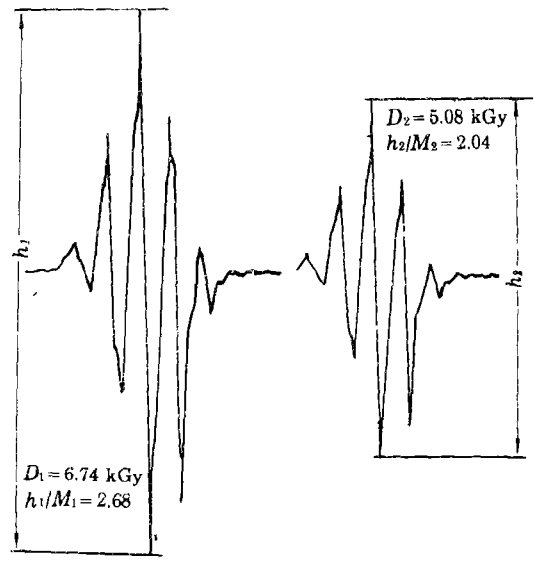

Fig.2 The first derivative spectra of two L—alanine dosimeters irradiated at dose levels of 5.08 kGy and 6.74 kGy

D_1/D_2= 1.326 $(h_1/XM_1):(h_2/XM_2)$= 1.310

4 EXPERIMENTAL RESULTS

4.1 Homogeneity of the sample density

The consistency for the sample mass density has been checked. The spread in densities for 45 dosimeters chosen from three kinds of diameters in several batches of the samples prepared is shown in Fig.3. The average density is 1.15 g/cm^3 and standard deviation is about less than 5 % for the sample pellets with total masses 5—20 mg.

4.2 The lowest detectable dose corresponding to the hight of ESR zero signal for the electron dosimeters

Non-irradiated dosimeter didn't give out any detectable ESR signal. The low limit of the alanine dosimeter has been determined by the spread degree in zero reading values and the noise of ESR spectrometer The results obtained from 10 pieces of different alanine dosimeters have indicated that the lowest detectable dose corresponds roughly to 60 Gy. The standard deviation from the average value is about 15 %. This is higher than that of cylindrical samples used for measurement of gamma radiation (4.5). As such thinner detectors, the amount of generating free radicals at

low dose is few. The lowest detectable dose for electron dosimetry is therefore higher than that of gamma radiations. Fig.4 shows the result of zero readings from 10 sample dosimeters.

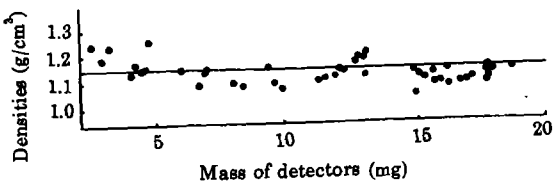

Fig.3 Distributions of densities for 45 pellets of L— alanine dosimeters

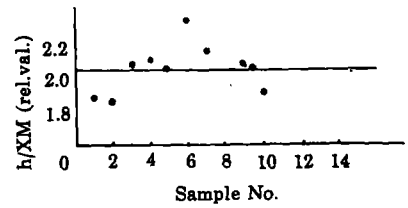

Fig.4 Distributions of ESR signals corresponding the lowest detectable dose (60 Gy)

4.3 Repeatability of measurement for the ESR spectrometer

For an individual alanine dosimeter the repeatability of successive measurements (1.0×10^4, 1.0×10^4, 9.97×10^3, 9.98×10^3, 9.98×10^3, 1.0×10^4, 1.0×10^4, 9.95×10^3, 9.97×10^3, 9.96×10^3 Gy) is about 0.5 %.

4.4 The reproducibility resulted in directivity and position of the dosimeters in the cavity

The reproducibility of ESR signal height using the peak–to–peak method depends on the stability of ESR spectrometer in short or long time, and on the directivity and positions of the detectors inside microwave cavity. These affecting factors all together have been tested. During the testing the operating conditions of ESR spectrometer were not changed. The irradiated dosimeter was taken out from the microwave cavity after each measurement. Its orientation in the cavity was successively changed and then inserted again. The average value among the measured signal heights and standard deviation are shown in the Table 3.

Table 3

Effect of directivity and position for the alanine dosimeters

Directions	E	E–S	S	W–S	W	W–N	N	E–N
Amplitudes (rel.val.)	2.56	2.56	2.52	2.52	2.60	2.60	2.60	2.60
Average value: 2.57,				Relative SD: 1.3 %				

4.5 Calibration curves of the electron dosimeters

For completing the dose–response curves, the solid state alanine dosimeters in the shape of thin pellet were irradiated with a $^{90}Sr/^{90}Y$ source calibrated by an extrapolation ionization chamber. Then the absorbed doses in the dosimeters were evaluated by a Kurker spectrometer. All peak values of the ESR signals are corrected using the gain of spectrometer, mass of the detector, and zero dose reading. As an

example, a straight line relationship between the peak-to-peak amplitudes and absorbed doses has been obtained in the range of 36 Gy to 20 kGy. For $n=5$, correlation coefficient is 0.9957, the equation obtained on log-log plot is $\log D = \log 43.5 + 1.158 \log (h/XM)$.

It is shown that the peak-to-peak values (h) of ESR signals increase linearly with absorbed dose (D) up to about 2×10^4 Gy. X is the gain of spectrometer and the M is the mass of the dosimeter.

4.6 Relative scattering of responses for alanine interspecimen irradiated by an identical dose

Table 4 shows an example for eight pieces of samples irradiated to 5080 Gy. In most cases the relative scattering of responses to an identical absorbed dose is less than 5 %.

Table 4

Relative scattering of responses to identical dose for L— alanine dosimeters (D=5080 Gy)

No.	L1	L2	L3	L4	L5	L6	L7	L8
h/XM (rel.val.)	6.493	6.877	6.924	7.102	7.040	6.980	7.035	7.537
Average value h/XM= 6.998				Relative SD = 3.9 %				

4.7 Sum effect of the evaluations

The four pieces of alanine dosimeters irradiated were measured respectively. Then, they all together were placed in a quartz-tube and measured. The results indicate that the relative diviation between the average value of them and average value of the sum effect over these samples is 3.8 %, and the relative diviation of the twice sum effects is 0.5 %.

4.8 Fading characteristic

Fading of response was measured for 150 d after irradiation under the storage at room temperature (about 15—28 °C). The fading rate with time after irradiation can be described by the formula, $D=D_0 e^{-\lambda t}$. Here $\lambda = 0.00015$, t is the time after irradiation in days.

5 CONCLUSION

It is reported that among isomers of alanine the L-alanine is found to be the best material for radiation dosimetry. With regard to atomic composition, effective atomic number and mass density it is approximate to tissue equivalent. These properties combining a high yield of free radicals after irradiation with a very low prior-dose corresponding with a zero signal and high precision in the measurement by electron spin resonance spectrometer make it a useful dosimeter for both routine use and transfer dosimeter.

In the report, the results of preliminary research on home-made alanine/ESR

electron dosimeters are summarized.

a. L-alanine dosimeters with different diameters and thicknesses have been successfully prepared. The smallest thickness of 0.5 mm suited for measurement of electron dose at high level has been obtained.

b. The average density is 1.15 g/cm^3 (alanine 80 % and paraffin 20 % by weight), and the standard deviation is less than 5 % for 45 pieces of dosimeters. The peak-to peak height of ESR signal has been corrected by mass of each detector itself.

c. The lowest detectable limit of the thin dosimeters for measurement of electron absorbed dose corresponds to about 60 Gy. Using the sum effect of evaluation and pilling the samples it is possible that the low limit is extended to lower doses.

d. Repeatability of the instrumental measurement is about 0.5 %.

e. Reproducibility resulted in directivity, locations as well as the repeatability of spectrometer is less than 2 %.

f. Linearity regions cover more than 4 decades (20Gy—20 kGy).

g. Relative scattering of responses to an identical dose of electrons, in most cases, is less than 5 %.

Alanine/ESR dosimetry is a new technique in high dose determination. It has a lot of advantages over traditional methods which have been described elsewhere in literature. This report has dealt with some other problems, their influence factors will be published.

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REFERENCES

[1] Chen Lishu, Jin Zaikun. *Nuclear Protection*, 1980, (1):64.

[2] Regulla D F, Deffner U. *Int J Appl Radiat Isot*, 1982, 33:1101.

[3] Svein prydz, Thormod Henriksen. *Acta Chemica Scandinavia*, 1961, 15:791.

[4] Bartolotta A, Indovina P L, Onori S *et al. Radiation Protection Dosimetry*, 1984, 9(4):277.

[5] Bartolotta A, Caccia B, Indovina P L *et al.* Proceedings of an international symposium on high-dose dosimetry. IAEA-SM-272-41. 1984.