ESR Dosimetric Properties of Sodium Glutamate

S. Eid, S. Ebraheem and A. Sobhy

Radiation Protection and Dosimetery, Dept., National Centre for Radiation Research and Technology (NCRRT). P. O. Box; Nasr City, Egypt.

MONO SODIUM GLUTAMATE powder and rods (3x 10 mm) were studied to be a radiation sensitive material for ESR dosimetry. Samples were irradiated with ⁶⁰Co γ - rays. The developed signal after irradiation increases with the increase of the mono sodium glutamate in the rods. The prepared powder can be used in the dose range from (10-90) kGy, whereas the rods are useful in the range from (10-120) kGy. The obtained number of free radicals per 100ev (G value) was found to be 0.201 ± 0.01 . The g factor is 2.0113 ± 0.0001 . The rods have the advantage of negligible humidity effects during irradiation. The pre and post irradiation stability was found to be satisfactory.

Keywords: Mono sodium glutamate, Electron paramagnetic resonance, Radiation dosimetry.

Ionising radiation (gamma rays form ⁶⁰Co and ¹³⁷Cs sources, x-ray and electron beam) has been used for a long time in order to sterilize medical devices to improve hygienic quality of food (Gancheva et al., 2006) or for sterilization of drugs and cosmetics (Regulla and Deffner, 1983 and Talbi et al., 2004). Electron paramagnetic resonance (EPR) was proposed as a method to measure radiation induced radicals. One of the advantages of EPR dosimetry is that the readout does not affect the spin concentration; the sample can be evaluated many times. Therefore, the signal to noise ratio (S/N) can be improved by repeated reading of the sample (IKeya, 1993). The Application of EPR to radiation dosimetry yields several materials such as tissue equivalent alanine, which is convenient for use in transfer dosimetry at high and intermediate dose. Efforts to use EPR dosimetry in health physics have resulted in a minimum detectable dose of 10mGy (Nakajima, 1988, Nakajima, 1995 and Nakajima et al 1990). Lithium lactate was used to measure the low dose from 0.1 to 10 Gy by applying a modulation field of 0.25mT and using a sharp signal of g= 2.0035 (Hassan and Ikeya, 1997). A high modulation field of 1 mtT was used to measure intermediate and high dose by using the quartet signal of Li-lactate (Hassan et al., 1998).

A number of solid materials, including amino acids, in which free radical populations are formed by irradiation, have been suggested for high dose dosimetry by EPR analysis. A useful method of high dose measured is the use of ESR spectroscopy of irradiated amino acids, in particular L-alanin, CH3CH(NH2).COOH purified in polycrystalline form (Bermann *et al.*, 1970, Beshir *et al.*, 2012, Bradshow *et al.*, 1962 and Regulla and Deffner, 1982). Poly (methylmetacrilate)-maghemite (PMMA-gamma-Fe2O3) hybrid material was studied by the electron stimulated ion desorption techniques coupled with time-of-flight mass spectrometry (TOF/MS) and theoretical investigation about its fragmentation were carried out (Rocha *et al.*, 2014). The objective of the present study is preparation of a new EPR routine dosimeter by simple technique in the laboratory by mixing mono sodium glutamate with both vinyl acetate co-polymer and paraffin wax. The radiation induced radical in glutamate powder and rods were investigated from dosimetric point of view.

Experimental

Materials

Mono sodium glutamate H₂o with molecular formula $C_5H_8NNaO_4$. H₂O. M. wt 187.13 ,melting point 232°C, water solubility $\geq 10g/100$ ml at 200 °C as well as melt stick adhesive based on ethylene vinyl acetate copolymer (Tec-Bond32/12, Power Adhesives Limited, England) and paraffin wax (congealing point 65-71°C, BHD) were selected as binding materials for mono sodium glutamate rod preparation. This binder was previously selected in preparation of alanine/EPR rods (3x 10mm) for high dose radiation dosimetry in the range 1-125 kGy (Abdel-Fattah *et al.*, 2004).



Fig.1. Structure formula of monosodium glutamate.

An equal weight mixture of paraffin wax and Ethylene Vinyl acetate (EVA) co-polymer was melt at $85-95^{\circ}$ C in a water bath 5, 10, 20 and 40% fine powdered mono sodium glutamate was added to the hot mixture solution and completely mixed by mechanical stirrer for about 15min at the same temperature to obtain homogeneous mixture and then sucked into polypropylene tubes (inner diameter 3mm) and was left to solidify by cooling. Mono sodium glutamate mixture rod was obtained by removing the poly propylene tube then cut into rods (3x 10mm) the average mass of prepared rods was found 0.08g± 0.002. It was found that, the use of EVA/paraffin as a binder did not show any interference or noise in the EPR signal before and after irradiation by our investigation.

Irradiation of the prepared sample

⁶⁰Co irradiation facility was used for Irradiation of the powder and the prepared rods. The absorbed dose rate was about 3.0639 kGy/h overall the time of the experimental part. Three rods of monosodium glutamate for each dose point were irradiated together at the central position of gamma cell using a specially designed holder made from polystyrene to ensure electronic equilibrium. Samples were irradiated at various absorbed doses from (10-120 kGy) to establish the response curve.

EPR measurements

EPR spectra of unirradiated and irradiated mono sodium glutamate were recorded at ambient temperature with a Bruker EMX spectrometer (X-band); product of Bruker, Germany the cavity used was the standered Bruker ER 4102 rectangular cavity. The operating conditions for the EPR spectrometer were as follow: microwave frequency: 9070GHz; microwave power 0.796mV; modulation frequency 100 kHz; modulation amplitude 5 Gauss; time constant: 81.92 ms; sweep time: 20.97 s/scan. The bottom of the EPR tube was adjusted at a fixed position to ensure reproducible and accurate positioning of the rods in the sensing zone of the cavity to obtain higher reproducibility and accuracy of the EPR results. The intensity of each irradiated rod dosimeter was recorded at two orientations in the EPR cavity (0° and 90°) by changing the position of EPR tube in the cavity. In addition, all of SH reading was corrected to SH of the reference standard material (SH_r) of 2, 2- diphenyl-1-picrylhydrazyl (DPPH).

The DPPH was measured before and after each series of measurements in the same rectangle cavity at the same identical measurement conditions in order to correct for change in the spectrophotometer sensitivity. After that, the dose response curve was established in terms of (SH_s) divided by sample wt corrected by (SH_r) as a function of absorbed dose, where corrected SH= SH_s/ SH_rx wt).

Results and Discussion

Spectral features

Fig. 2. shows the EPR spectrum of irradiated mono sodium glutamate powder, shows sextet signal with g factor 2.0325 ± 0.0013 for S₁, 2.02188 ± 0.0019 for S₂, 2.01135 ± 0.0009 for S₃, 2.00533 ± 0.001 for S₄, 1.9932 ± 0.0022 for S₅ and 1.9847 ± 0.001 for S₆. The spectral line width= 90.7 gauess. S₃ with the magnetic field 3459.18 gauss was chosen to perform all the work in this study due to its high sensitivity.



Fig. 2. EPR spectrum of irradiated mono sodium glutamate powder irradiated at 90 kGy.

Power dependence

Fig. 3. shows the microwave power dependence of the mono sodium glutamate signal intensity. The intensity of EPR signals increases in proportion to $p^{1/2}$ up to high microwave power. Appropriate setting of power level is necessary for the EPR measurements. The signal intensity increases as a function of power up to 0.7969 mw without reaching saturation.



Fig. 3. Relationship between square root of microwave power (mw ¹/₂) and EPR signal intensity of gamma irradiated mono sodium glutamate.
Effect of gamma radiation on mono sodium glutamate powder

Fig. 4. represents the spectra of unirradiated and irradiated mono sodium glutamate. The powder wt is 0.1286 g and irradiated doses are 8, 20, 45, 70, 90 kGy. From the figure it is clear that the signal intensity increases with the increase of irradiation dose without any change in its shape.



Fig. 4. ESR spectra of mono sodium glutamate powder at different doses.

It can be seen that no signal has detected for the unirradiated mono sodium glutamate. As we know, when ionizing radiation passes through a material it impart some of its energy to that material. The imparted energy may be high enough to cause a break in bonds inside the molecules or between molecules or both, in such cases, free radicals are created. Behaviour of radiation induced radicals may lead to further understanding of molecular interactions or molecular (radicals) dynamics, or may lead to a decision on the preference of a material for a specific applied from a dosimetric point of view. Also organic radicals are characteristic by large line- width which due to super-hyperfine interaction with nearby protons in neighbouring molecules (Lendzian, 2005).

Fig. 5. represents the response curve of mono sodium glutamate powder in terms of peak to peak amplitude normalized to dosimeter mass (peak high/mass) versus the absorbed dose. It can be seen that the radiation sensitivity of the mono sodium glutamate increases with the increase of the absorbed dose. This attributed to the increase in number of free radicals induced by radiation.



Fig. 5. Response curve of mono sodium glutamate powder at different doses.

Short term decay of mono sodium glutamate powder

0.0813 g of mono sodium glutamate powder was irradiated to a dose of 5 kGy and fixed in cavity centre and measure for 5 h. Fig. 6. shows the obtained fading behaviour of the EPR signal. It can be seen that the free radicals

undergoes random orientation during the first 30min then tend to be stable after losing their extra energy until the end of the storage period.



Fig. 6. Decay of EPR line of irradiated mono sodium glutamate powder at a dose 5 kGy.



Fig. 7. Post-irradiation stability of mono sodium glutamate powder to 25 kGy.

Post irradiation stability of mono sodium glutamate powder

0.1287g mono sodium glutamate was irradiated to a dose of 25kGy and measures the signal intensity at different time intervals of 60 days as in Fig. 7. The peak intensity value shows slight increase in the first 10 days and then tends to be stable to the end of the storage period.

Radiation sensitivity

The efficiency of a dosimeter is expressed by a G-value *i.e.*, the number of radicals or ions produced by the absorbed radiation per 100ev. The radical formation efficiency was determined by double integration of the first derivative spectra of DPPH and compared with those of irradiated materials. The absolute spin concentration was estimated according to Hassan *et al.* (2000) and Ikeya (1985) by using the following equation:-

$$n = \frac{A_{sample} \times^{n} strong \, pitch}{A_{strong \, pitch} \times Dose(Gy) \times m(g)} = 6.25 \times 10^{13}. \, G(Gy^{-1}g^{-1})$$

where, A_{sample} , $A_{\text{strong pitch}}$, $\pi_{\text{strong pich}}$ and m are the areas of integrated signals of sample and strong pich, number of spin in strong pitch, number of spin in strong pitch and the mass of sample, respectively. The G-value of the whole area was found to be 0.2019 ± 0.01 .

Effect of gamma radiation on mono sodium glutamate rods

Four different concentrations of mono sodium glutamate namely 5, 10, 20, 40% irradiated to a dose of 40kGy were shown in Fig. 8. from which it could be concluded that the peak intensity increases with the increase of mono sodium glutamate in the rods.



Fig. 8. EPR spectra recorded for mono sodium glutamate rods of different concentrations irradiated to a dose of 40kGy.



Fig. 9. EPR spectra of mono sodium glutamate (20%) irradiated to 10, 20, 40, 60, 80, 100 and 120 kGy.

Fig.9. represents the EPR spectra of unirradiated and irradiated mono sodium glutamate rods (20%) were recorded at a series of absorbed doses 10, 20, 40, 60, 80, 100, 120 kGy. From the Fig. 9. it can be seen that the EPR signal begins to develop upon irradiation and its amplitude increases with increasing absorbed dose of gamma ray photons without any change in its shape.

Dose Response of Rods with different concentration of mono sodium glutamate

Fig. 10. represents the response curves of 4 sets of mono sodium glutamate (5, 10, 20,40%) to different absorbed dose range from 5-120 kGy.



Fig. 10. Dose response curve of irradiated different concentration of mono sodium glutamate rods.

The response curves obtained for the irradiated sample in terms of average peak to peak amplitude normalized to dosimeter mass (peak high/ mass) versus the absorbed dose. It can be seen that the radiation sensitivity increases with the increase of mono sodium glutamate in the rods. This increase is attributed to the increase of the free radicals induced by radiation.

Post-irradiation stability at different storage conditions

The EPR signal stability has been studied for three sets of mono sodium glutamate rods (20%) stored at different conditions (dark, light, and at 40 $^{\circ}$ C). The stability was investigated as a relative to that value immediately after irradiation as shown in Fig. 11. The signal intensity decreases at the beginning of storage time then tends to be stable at the end.



Fig. 11. Relative peak to peak signal light as a function of storage time at different storage conditions (concentration 20%, dose 25 kGy).

Humidity during irradiation

The effect of relative humidity during irradiation on the response of mono sodium glutamate of 20% concentration was investigated by the irradiation of the rods to 25 kGy at different humidity values(0-95%). To establish the RH levels, the rods were suspended over various saturated salt solutions in tightly closed vials (Wexler and Hasegawa, 1954). The rods were stored before

irradiation at room temperature for a period of 72 h to maintain equilibrium moisture content in the rod during irradiation. Fig. 12. shows the variation of the EPR intensity as a function of relative humidity during irradiation relative to the response value at 33% relative humidity. It can be concluding that mono sodium glutamate rods have negligible humidity effect not exceed 5% in the range of humidity from0-100%



Fig. 12. Variation of the EPR response of mono sodium glutamate rods (conc. 20%) as the function of relative humidity during irradiation (Absorb dose=25kGy).

Conclusion

From the presented data of the study, the following conclusion can be drawn: Mono sodium glutamate powder is a suitable dosimeter to a dose range 0-90 kGy while mono sodium glutamate rods are 5-120 kGy. The dosimetric characteristic of rods containing different concentrations of mono sodium glutamate was recorded. The rods have significant EPR signal which develops upon irradiation and intensity of the signal increases upon the increase in irradiation dose. These rod dosimeters have insignificant dependence on the change of relative humidity during irradiation. It is recommended to calibrate these dosimeters at conditions of use of humidity value less than 60%. The properties of the prepared mono sodium glutamate rods suggest their useful application for food irradiation, medical and industrial application. This

dosimeters are fairly stable before and after irradiation and show little fading between 5-10 % over 60 days storage period.

References

- Abdel-Fattah, A. A., Eel-Din. H. and Abed-Rehim F. (2004) New alanine/ EPR dosimeter using EVA copolymer/ paraffin as a binder for high-dose radiation dosimtry: performance characterization. *Int. J. Polym. Mater.*, 53, 927.
- Bermann, F., De Choudens, H. and Descours, S. (1970) In Advances in Physical and Biological Radiation Detector, Proc. Symp. Vienna, IAEA Publication STI/PUB/269 (Vienna, International Atomic Energy Agency) p. 311.
- Beshir, W. B., Abedel-Fattah, A. A., Abedel-Rehim, F. and Hassan, H. M. (2012) EPR dosimetric properties of radiation-Formed radicals in arginine mono hydrochloride. J. Potochem. Photobiol., 116, 1.
- Bradshaw, W. W., Cadena, D. G., Crawford, E. W. and Spetzler, H. A. W. (1962) The use of Alanine as a solid dosimeter. *Radiat. Res.*, 17, 11.
- Gancheva, V., Sagstuen, E., and Yordonov, N. D. (2006) Study on the EPR/ dosimetric properties some substituted alanine. *Radiat. Phys. chem.*, **75**, 329.
- Hassan, G. M. and Ikeya, M. (1997) Radical formation of lithium lactate for ESR dosimetry. J. Nucl. Sci. Technol., 34, 1185.
- Hassan, G. M., Ikeya, M. and Toyoda, S. (1998) Lithium lactate as an ESR dosimeter. *Appl. Radiat. Isotop.*, **49**, 823.
- Hassan, G. M., Ulku, W. and Ikeya, M. (2000) Radical Formation in Lithium and magnesium oxalate. *Jap. J. Appl. Phys.*, **39**, 6236.
- Ikeya, M. (1985) Techniques of Radiation Dosimetry Electron Spin Resonance. In: Mahesh, K., Viji, D.R. Wileyb eastern, New Delhi, Chap. 15.
- Ikeya, M. (1993) New Applications of electron spin resonance-dating, dosimetry and microscopy. World Scientific, Singapare, p.395.
- Lendzin, F. (2005) Structure and interactions of amino acid radicals in class 1 ribonucleotide reductase studied by ENDOR and high-field EPR spectroscopy. *Biochim. Biophys. Acta*, **170**, 67.
- Nakajima, T. (1988) Sugar as emergency populace dosimeter for radiation accident. *Heal. Phys.*, 55, 951.
- Nakajima, T. (1995) ESR of sugar as personal monitor for radiation emergencies. *Appl. Radiat. Isotop.*, 46, 819.
- Nakajima, T. and Ohtsuki, T. (1990) Dodimetry for radiation emergencies: radiation induced free radicals in sugar of various countries and effect of pulverizing on the ESR signal. *Appl. Radiat. Isotop.*, **41**, 359.
- Regulla, D. F. and Deffner, U. (1983) System transfer dosimetry in radiation processing. *Radiat. Phys. Chem.*, 22, 305.

- Regulla, D. F. and Deffner, U. A. (1982) dosimetry by ESR spectroscopy of alanine. *Int. J. Appl. Radiat. Isotop.*, 33, 1101.
- Rocha, M. V., Carvalho, H. W., Lacerda, L. C., Simoes, deSouza, G. G. and Ramalho, T. C. (2014) Ionic desorption in PMMA-gamma-Fe2O3 hybrid materials induced by fast electrons: an experimental and theoretical investigation. Spectrochim. Acta A Mol. Biomol. Spectrosc., 117, 276.
- Talbi, S., Raffi, J., Arena, S., Colombani, J., Piccerelle, P., Prinderre, P. and Dolo, J. M. (2004) EPR study of gamma induced radicals in amino acid powders. *Spectrochimica Acta*, part A, 60, 1335.
- Wexler, A. and Hasegawa, S. (1954) Relative humidity-temperature relationships of some saturated salt solutions in the temperature 0°C to 50°C. J. Res. NBS, 53, 1920.

(Received: 01/06/2014; accepted: 17/07/2014)

استخدام الصوديوم جلوتامات كمقياس للجرعه الاشعاعية باستخدام الرنين المغناطيسي المغزلي

سيده عيد و سيف الدين ابراهيم و عصماء صبحى

قسم الوقاية و الجرعات الإشعاعية ، المركز القومي لبحوث وتكنولوجيا الإشعاع ص. ب : ٢٩ مدينة نصر ، مصر.

تم فى هذا البحث در اسة استخدام مادة الصوديوم جلوتامات على شكل مسحوق أو قطبان قطر ها (١٠x ١ سم) كمادة ذات حساسيه للرنين المغناطيسى. تم تشعيع هذه العينات بواسطة الكوبلت ٢٠ و قد تبين أن شدة الموجات تزيد بزيادة الجرعه الممتصه من اشعة جاما و تبين أن المدى الاشعاعى لمسحوق الصوديوم جلوتامات يتراوح من ٢٠-١٠ كيلو جراى بينما كانت القضبان لها حساسيه فى المدى الاشعاعى ٢٠-١٢ كيلو جراى ز ثم أيضا حساب الناتج الاشعاعى و الذى يعنى عدد الشقوق الحره الناتجه عن ٢٠ الكترون فولت و وجد أن قيمتها ٢٠٢ ر . بالاضافه الى إجراء اختبارات الثباتيه لهذه الماده و التى تبين منها انها لا تتأثر بالرطوبه خلال التشعيع و أيضا تتمتع بثباتيه عاليه قبل و بعد التشعيع.