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EPR DOSIMETRY—PRESENT AND FUTURE

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Abstract

In the past, IAEA has played a central role in stipulating research and development in EPR high-dose standardisation as well as co-ordinating and organising international dose intercomparison programs, within the Member States of the United Nations from the mid-seventies till today. The future tasks of EPR dosimetry seem to tend towards different subjects such as biomarkers, biological radiation effects, post-accident dose reconstruction in the environment, and retrospective human dosimetry. The latter may be considered a promising tool for epidemiology on the way to re-define radiation risk of man for chronicle radiation exposures, based on e.g. South Ural civil population and radiation workers. There are on-going international activities in the field of standardising high-level dosimetry by the American Standards on Testing and Materials (ASTM), and the International Organisation of Standards (ISO) as well as those of the International Commission on Radiation Units and Measurements (ICRU) considering the establishment of relevant recommendations concerning industrial radiation processing, but also human dose reconstruction.

1. INTRODUCTION

High-level application of ionising radiation from 10^3 to 10^6 Gy was described in the early seventies. The growing field of industrial radiation processing, under the aspects of safety and economy, was soon recognised to require a reliable and accurate dosimetry [1]. There were dosimetry techniques available at that time, but traceability to primary standards, access to national calibration services or availability of international recommendations on standardisation of high dose dosimetry were missing.

It was in 1977 that the Agency took initiative and invited international experts to discuss new methods of dosimetry and standardisation in radiation processing at the occasion of a Consultants' Meeting on High-Dose Standardisation and Intercomparison for Industrial Radiation Processing [2]. Decision was taken to accept a proposal of the author and implement a "new" technique based on radiation induced generation of radicals in an amino acid, i.e. alanine [3]. Electron paramagnetic resonance (EPR) spectrometry was chosen for quantification of the radical concentration. Within the subsequent IAEA co-ordinated research programme, EPR became a scientific tool for dosimetric application in routine and metrology [4,5]. Later, the Agency established the International Dose Assurance Service (IDAS) to the member states of IAEA using the meanwhile standardised alanine/EPR technique. IDAS was operated jointly between IAEA and GSF [6]. Since 1991 the Agency continues to offer IDAS successfully as an exclusive service supplier [7,8].

This paper does not deal with archaeological and geological dating [9,10] nor with EPR imaging [11]. It should however be noted that it is particularly the task of geological dating for human sciences that stipulated great interest and initial progress in EPR dosimetry [12].

2. PRESENT STATUS

2.1.The Alanine/EPR system

Alanine/EPR dosimetry uses organic crystalline amino acids (e.g. alanine, CH_3-$CHNH_2$-COOH) as a sample material [3]. This method is applicable to dosimetry of different types of radiation as well as, within limits, to dosimetry in mixed radiation fields, e.g. x and gamma rays, beta radiation, accelerator electrons, protons [13], neutrons [14], and ions [15-18].

The free radicals in crystalline biomolecules are relatively stable interim products in a chain of events, that similar to tissue, start with the absorption of radiation energy. Since free radicals take key positions in the chain of events which lead to biological damage in cell structures, the quantification of free radicals in alanine can even be used for a biologically relevant dosimetry.

Accurate EPR dosimetry requires the availability of an appropriate "system" as well as qualified personnel with know-how, experience and care. The essential system components are: Alanine detectors of a high purity production and metrological quality, a high performance EPR spectrometer and an air-conditioned laboratory, calibration facilities in the respective dose range, as well as dosimeters of secondary standard or reference quality level, whose calibration is traceable to the primary standards of national laboratories (Fricke dosimetry, calorimetry, ionisation chamber dosimetry). Essential dosimetric properties for GSF alanine/EPR dosimetry system are compiled in Table I.

2.2. Radiation processing: Reference and transfer dosimetry

Meanwhile a number of acknowledged national laboratories world-wide started to use the alanine/EPR technique for quality control programs in radiation processing. The alanine/EPR technique has been introduced for quality control also in therapy [19] using the alanine/EPR technique similar to the IDAS programme [21,22]. Future applications will probably be based on a new EU Medical Devices Directive [20].

TABLE I. CHEMICAL, PHYSICAL AND DOSIMETRIC PROPERTIES OF GSF ALANINE/EPR DOSIMETRY SYSTEM

Composition	Alanine CH_3-$CHNH_2$-COOH (85 %) and paraffin (15 %)
Effective atomic number	$Z_{eff} \cong 7.2$ (tissue: $Z_{eff} \cong 7.4$)
Specific gravity	$\cong 1.15$ g/cm^3
Dimensions	4.9 mm in diameter × 10 mm length
Measuring quantity, D_w	Absorbed dose to water; otherwise on request
Measuring range	$0.5 \text{ Gy} < D_w < 5 \cdot 10^5$ Gy
Detection threshold	0.05 Gy
Dose rate dependence	Not detectable till to 10^{11} Gy·h^{-1}
Energy dependence	Approx. independent, for $E_{ph} \geq 100$ keV and $E_e \geq 1$ MeV
Irradiation temperature, θ	Negligible or correctable in the range $-90°C < \theta < +70°C$ ($k_\theta = 0.0018$ °C^{-1} for $D \leq 40$ kGy)
Fading (signal loss) within 2 years	< 1 % at 22°C and <70 % r.h.
Interspecimen scattering	1 s.d. $\leq \pm$ 0.5 %, within a batch
Interbatch scattering	≤ 1 %
Precision	u ≤ ± 1.5 % at 95 % confidence level, approved by a national laboratory intercomparison protocol
Electron/photon ratio of response	1.0, for $E_e > 1$ MeV
Neutron/photon ratio of response	about 0.6, for $E_n > 0.1$ MeV

2.3. Therapy-level dosimetry

The properties of alanine/EPR dosimetry have always been studied with an eye focusing on radiation therapy [23]. The advantages of alanine/EPR dosimetry in this field are evident, e.g. the dynamic dose range, the archival character of dose information based on a non-destructive readout that allows for repeated EPR measurements and sample storage, and the tissue equivalency of detector samples. The latter property makes the method applicable also in high-energy radiation dosimetry without the impact of perturbation and displacement effects as known from ionisation chamber dosimetry. A mathematical method based on Fast Fourier Transform has meanwhile been developed capable to filter simultaneously background and noise in the frequency domain of EPR spectra [24]; it provides significantly higher resolved alanine/EPR signals, and this down to about 50 mGy.

It was again the Agency which at an early stage had organised an alanine/EPR intercomparison in the therapy-level range [25]. Also, it recently convened a consultants' meeting to review the current status of use of alanine for therapy [26]. Complementary investigations are reported on the use of alanine EPR dosimetry in proton therapy [27,28].

2.4. Identification of irradiated food

For a long period of time there was a lack of appropriate identification methods of irradiated foodstuffs, however the situation has changed during the past decade [29]. A variety of methods has become available to evaluate on the status of radiation processing, or even roughly estimate the dose level, valid for irradiated spices and spice products, herbs, dried vegetables, some kind of fruits, coffee beans, different meats and meat products as well as fish, shell-fish, shrimps, etc. [30]. EPR spectroscopy is applicable to the identification of irradiated foods, for discrimination against unirradiated foods or exclusion of two- or more-fold food irradiations [31-36]. Some food allows an EPR dose assessment within ≤ 5 % [37]. For a relevant Food Control Dosimetry Network based on EPR, standard EPR spectra from the above foods should be available to be transferred for calibration and measurement, between laboratories and authorities involved by internet.

2.5. Ultra high-level dosimetry

Since material fatigue of radiation sensitive components is an imported factor in nuclear fuel safety, it should carefully be checked before the components are implemented in the facility construction, and be under continued control during operation. Dose control by an appropriate dosimetry technique is inevitable for this task. For quantification of doses up to 10^8 Gy and above, EPR spectroscopy based on crystalline detectors was developed for irradiation temperatures of up to several hundred degrees Celsius [38,39] and verified by Monte Carlo based simulation of the radiation transport [40].

2.6. Standards and recommendations

Principles, procedures and quantities used in the alanine/EPR technology are described in an ASTM standard [41] which is now also a recognised ISO standard [42]. ICRU is presently considering to establish a task group on "Dosimetry in Industrial Uses of Ionising Radiation" [43,44]. An ICRU recommendation in this field would be of great interest and need, for public acceptance, benefit and health as far as dosimetry is concerned for radiation preserved food, sterilised pharmaceuticals, and medical and health care products [29]. IAEA and other international and national bodies, e.g. WHO, FAO, CEC and FDA, should be invited to contribute to the recommendation with their great expertise and the available technical documents, particularly in the field of EPR dosimetry.

2.7. EPR spectrometry

Alanine/EPR dosimetry uses sophisticated research EPR spectrometers in the X band microwaves. The state of the art of this equipment, in course of time, and its future potential are described by Ettinger [45], Pilbrow [46] and Ikeya [47]. Routine dosimetry makes use of meanwhile available

table-top EPR spectrometers with permanent magnets. Interesting parametric improvement of equipment as well as procedures has recently been proposed [48,49]. A pitch-activated alanine detector type [50] or the use of an in-cavity Mn^{++} standard [51] offer approaches to introduce individual calibration factors for alanine samples. The use of a continuously rotating goniometer reduces the effect of response anisotropy and results into reduced detection limits [52]. Large-scale alanine dosimetry services in national or international quality control may profit from automatic sample changing devices by robot [53] or magazine type mechanisms [54]. Future *in situ* oriented biophysical dosimetry will make use of EPR spectroscopy applying different microwave bands and magnetic field strengths [55,56].

3. FUTURE TASKS

EPR spectroscopy started playing a role in tooth dosimetry in the late seventies addressed to victims of Hiroshima and Nagasaki. Since scientists started in the nineties to work on dose histories of individuals from the early nuclear production plants of the former Soviet Union, remarkable progress has been achieved in extending the lower detection limit for this material.

3.1. Biophysical dosimetry

So far, the most known biological dosimetry technique is based on chromosome analysis in peripheral human lymphocytes serving in cases of acute exposures, dose estimates above 100 mGy, and a number of victims for evaluation [57]. Further improvement is expected from the so-called fluorescence in situ hybridisation (FISH) method based on translocations [58,59].

Today the biophysical EPR dosimetry using tooth (and bone) samples from exposed victims may represent a potential completion of the biological dosimetry for reasons of accuracy, reliability, dose individuality and procedural simplification [60,61]. EPR dosimetry is based on radiation induced radicals in hydroxyapatite, which is the mineral phase of teeth and bones. The most stable radical known is CO_2^- [62], whose life time has been reported to be 10^7 years (at 25°C) [63]. At present, doses from about 50 mGy to above 100 Gy can be evaluated, which is the relevant range of potential accidental doses [64,65], including photon and electron radiation as well as potentially α-particles and ions [66].

3.2. Environmental dose reconstruction and accident dosimetry

Management of dose assessment from objects of man's environment can profit from the free radical generation in and EPR spectroscopy of many deserted objects [67], e.g. sugar [68], pharmaceuticals [69], egg shells [70,71], cellulose [72] and pot-scale [73]. In future, also the mineral phase of soil and bricks [74,75], building materials [76] and appropriate tissues of animals [77] will be considered more intensively. Subsequently, accidental doses of individuals can be derived from the material doses to the human environment by computational conversion if not directly assessed from teeth and bone samples [78-80]. Recently doses were evaluated using EPR methodology to individuals exposed to radioactive sources or radioactive scrap metal in public areas or in possession of private persons [81].

3.3. Retrospective dosimetry

Biophysical EPR dosimetry is on the way to become a tool of increasing interest for retrospective dosimetry, i.e. evaluation of individual exposures that occurred years or decades and more ago [82]. It was first described by Ikeya et al. for the atomic bomb survivors of Hiroshima and Nagasaki [83], later for victims of the Chernobyl accident [84,85], nuclear workers of the PO Mayak [64], and residents of the Techa river valley, the latter both in the Southern Ural region [86]. A valuable survey is given in [87]. ICRU has meanwhile established a working group in the field of retrospective dosimetry; the IAEA also has established a co-ordinated research project in the same field.

3.4. Uncertainties in retrospective dosimetry

Individual and environmental retrospective dosimetry may serve, e.g. for verification of dose records as a basis for epidemiological studies. Comparisons with risk data from Hiroshima and Nagasaki require careful consideration of uncertainties. The uncertainties cover, e.g., the EPR spectrometer, sampling, sample treatment and evaluation procedures, the impact of influence parameters (e.g. ultraviolet light, diagnostic x-rays; etc.), calibration, and the statistical treatment of data [88,89]. Future research tasks in this domain should focus unbiased scientific attention to identify component uncertainties and evaluate overall uncertainties [90-93].

4. PERSPECTIVES

4.1. Emergency network for dosimetry

The capability of EPR to contribute to emergency response dosimetry was recently under consideration [94] demonstrating a rapid analysis method for screening deciduous teeth of children in the days and weeks following a radiation accident [95]. Such a response could conceivably aid in medical and social decision making.

Also of importance are the savings of time and manpower and the increase in reliability which could be achieved in dose reconstruction if identification and collection of biological and environmental dosimetry materials were promptly undertaken. The complexity of reconstructing doses from a population years after an accident would be greatly reduced if consideration were given to the task immediately following an accident [96]. Protocols for such response are currently not available [97].

4.2. Basis for juridical consequences

EPR based dose reconstruction will compete and equally complement the established biological dosimetry in the future, particularly in accidental or emergency dosimetry cases. Contrary to biological methods, the biophysical methods will allow to evaluate doses reliably for all those exposures of humans which occurred decades and more ago. The achievements to be expected represent new perspectives for labour inspection authorities in cases of radiation accidents at work and for the juridical consequences in cases of prosecution or legitimate claims for compensation.

4.3. Epidemiology and redefinition of radiation risk

Interest in the new perspectives of EPR dosimetry will hold particularly also for retrospective dosimetry of individuals with long-term exposures where the traditional biological methods may be of limited use only, e.g. for the determination of exposures of radiation workers from nuclear centres starting in the forties. Individual retrospective dosimetry based on tooth tissues and bones, also for the verification of corresponding film dosimetry records, may help us together with available health records of those radiation workers, to redefine radiation risk for - contrary to the acute exposure in Hiroshima and Nagasaki - chronicle exposure of man. First evaluations showed a reduced risk for leukaemia [98].

4.4. *In vivo* and space dosimetry

On the horizon we may, in not a too far future, discover new techniques of EPR or magnetic resonance (MR) spectrometry for human *in vivo* dose evaluation [99] and local dose imaging after accidents, e.g. *non invasive* dosimetry from teeth, skeleton, cell membranes and other tissues capable to produce and store radiolysis products detectable with EPR spectrometry. This might probably lead to a presently still visionary development of a "whole body dose scanner", equivalent to the established whole body counter. The human body appears to represent a still widely undiscovered field for dosimetry - and an exciting reservoir for radiation research also in future [100].

Such dosimetry techniques lead to the further vision of an individual biodosimetry, e.g. for space missions, providing dosimetry that can not be lost, inappropriately handled or manipulated by an individual, and a permanent record of cumulative doses due to different radiation types [101,102].

5. CONCLUDING REMARKS

Present dosimetry techniques in radiation processing, including EPR, will in future be supplemented by new, e.g. radiofluorescence type, techniques for routine and quality control programmes [103,104]. For treatment planning of products the experimental dosimetry may be supplemented by dose computations based on source-target modelling. The necessary product parameters for absorption and scatter can be transferred from a central data bank, by the world wide web [105]. Its implementation shows new perspectives to exchange information between processing plants, trade companies, and authorities, and will make the radiation processing level of a product traceable. The internet can equally serve to check the local dosimetry quality of a processing plant by a future telemetrology, whose technique and logistics has still to be developed and established [106]. The internet will facilitate the exchange of dosimetry results, calibration and traceability certificates as well as of acceptance passports for products - to be verified by EPR dosimetry.

By contrast, the importance of EPR for biophysical dosimetry promises to expand in future. Unification and standardisation will allow individual dose reconstruction in emergency cases which tool was not available earlier [81, 107-115]. Apart from the mineral tooth enamel, it is the metabolic dentine which can probably provide additional EPR information on e.g. bone seekers. Also this potential will continue to keep scientific interest focused on EPR biophysical dosimetry in future [116,117].

The present duties of the world-wide SSDL network for radiation protection and radiation application in medicine could in principle be considered a basis for expansion to biophysical - accident and retrospective - dosimetry and a first step towards a concept of a world-wide operating "Integrated Retrospective Dosimetry Network" [94,118]. It could provide permanent assistance and expertise in biophysical dosimetry, probably in co-operation with other national or regional laboratories, that can offer biological dosimetry, whole body counting and bioassays, as well as thermally and optically stimulated luminescence dosimetry techniques for objects taken from an incidental environment (Fig. 1).

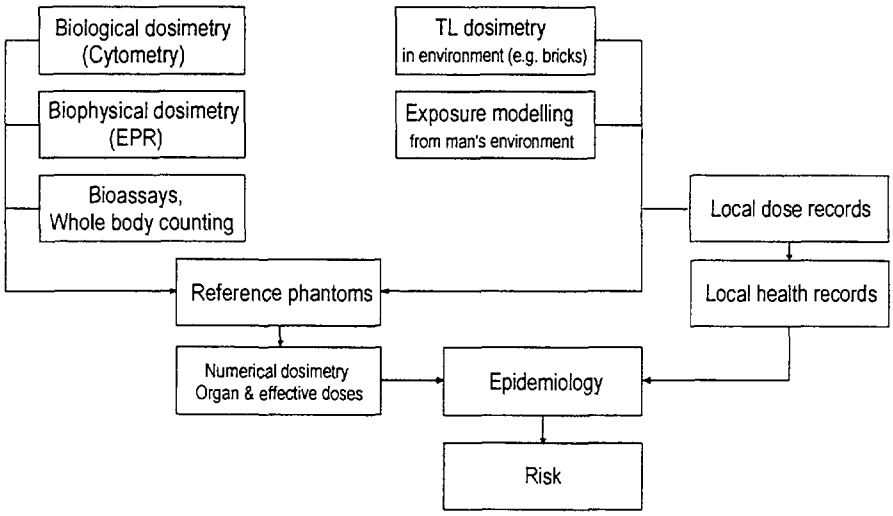

FIG. 1. Implementation of EPR biophysical dosimetry into a still visionary integrated retrospective dosimetry network, for further discussion [94,118].

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Part of the references refer to manuscripts presented at the recent "International Conference on Biodosimetry and 5th International Symposium on ESR Dosimetry and Applications", Obninsk/Moscow, Russia, 22-26 June 1998"; they are under process of peer-reviewing, and planned to be published in the International Journal of Applied Radiation and Isot. in 1999. Extended abstracts are available from the Conference Book of Abstracts (Prof. A. Tsyb, or Dr. Y. Skoropad, Medical Radiological Research Center of Russian Academy of Medical Sciences, Obninsk, email address: mrrc@obninsk.ru). A condensed review on the development and achievements of EPR dosimetry and applications in the past two decades can be found in: "ESR Dosimetry and Applications", Appl. Radiat. Isot. **40** (1989) 829-1246; **44** (1993) 1-468; and **47** (1996) 1151-1687, as well as in: "Retrospective Dosimetry - Physical and Biological Aspects", Radiat. Prot. Dosim. **77** (1998) 1-138.

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