DOSIMETRY AS AN INTEGRAL PART OF RADIATION PROCESSING

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Abstract

Different connections between high-dose dosimetry and radiation processing are discussed. Radiation processing cannot be performed without proper dosimetry. Accurate high dose and high dose rate dosimetry exhibits several aspects: first of all it is the preservation of the quality of the product, then fulfillment of legal aspects and last but not the least the safety of processing. Further, seldom discussed topics are as follow: dosimetric problems occurring with double-side EB irradiations, discussed in connection with the deposition of electric charge during electron beam irradiation. Although dosimetry for basic research and for medical purposes are treated here only shortly, some conclusions reached from these fields are considered in dosimetry for radiation processing. High-dose dosimetry of radiation has become a separate field, with many papers published every year, but applied dosimetric projects are usually initiated by a necessity of particular application.

1. INTRODUCTION

Dosimetry, i.e. the determination of absorbed dose in a chemically and physically defined object of particular geometry, is applied for two different purposes: in basic research, being the basis of calculation of radiation yields of products or effects, and in radiation processing of materials as a control of desired technological effect. Working in both fields requires the great responsibility: firstly, because radiation yields enable to draw important conclusions and generalizations; secondly, because it is connected with the quality of the product and the economy of processing. Whereas the choice and the performance of dosimetry in the first field of dosimetric applications is a sole responsibility of the researcher, the dosimetry in the second case is the duty of the irradiation plant management and is the subject of standards (e.g. ASTM, ISO and national organizations) and supervisions, sometimes of international scope. In the transient zone, i.e. between basic research and applications, the responsibility of the researcher in the field of dosimetry is also important. The error in determination of the radiation yield by 0.1 μmol J^{-1} and/or negligence in estimation of yields as the function of dose can turn a highly promising technology into an economic disaster.

Connections between radiation processing and dosimetry were always very close. On the one hand the radiation processing was defining problems for solution, on another the dosimetry and results obtained from measurements strongly influenced the techniques of irradiation and technologies of the processing. That is well recognised in one of the first textbooks on dosimetry [1] and in another one of the more recent books on dosimetry, in which the key word "radiation processing" has appeared in the title of the book [2].

2. DOSIMETRY IN BASIC RESEARCH

Dosimetry applied in strictly basic research will not be discussed in the present paper. However, there is a thin boundary between basic and applied research. For instance, very often the basic research reveals the existence of chain reactions. There is an extreme sensitivity of yields of chain reactions to the dose rate which has to be confirmed with proper dosimetry. For instance, reactions of oxidation running with the radiation yield of the order of hundreds per 100 eV of absorbed energy (order of 10 μmol J^{-1}) in the γ-radiation field lower their yield to a few units if high power electron beam is used [3]. The same applies to chain reactions initiated in systems where polymerization takes place.

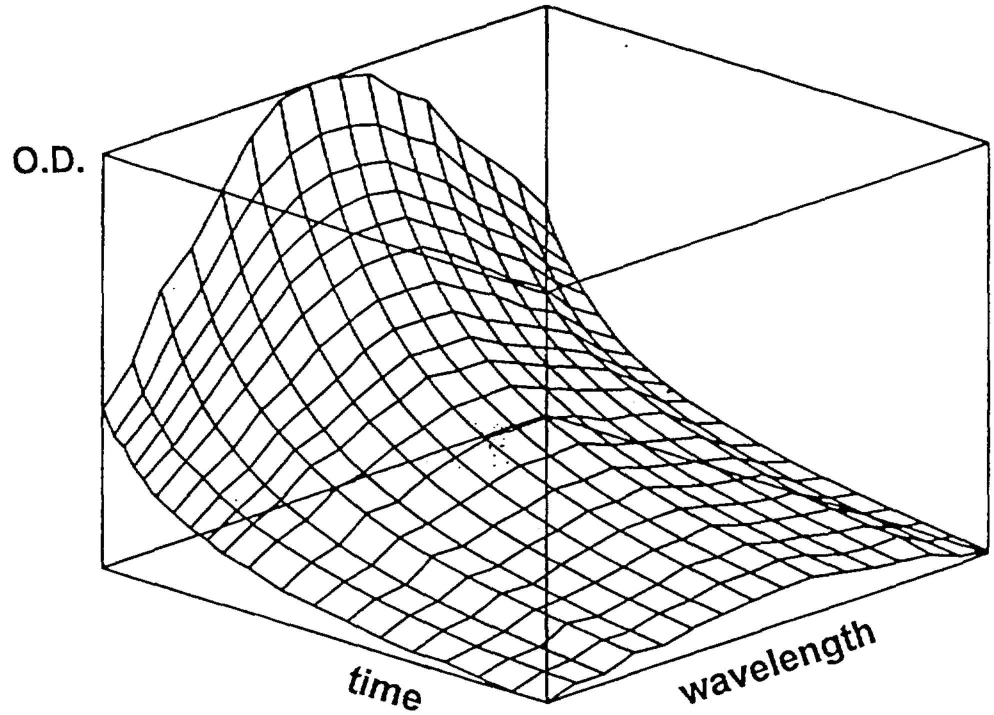

FIG. 1. 3-D diagram of spectra of $(SCN)_2^-$ decay in pulse radiolysis dosimetry. The dose is calculated from the optical density extrapolated to the zero time of beginning of the decay.

Dosimetry in pulse radiolysis experiments has been developed to the specific form taking into account high heterogeneity of the dose distribution unaccounted for in radiation processing. Nevertheless it reflects the true delivery of energy, because it averages the dose in exactly identical way, as it is in the case of experiment which follows the dosimetric measurement. The dose is measured with the same equipment as used in time-resolved measurement of concentration of a transient. The radiation-induced dosimetric reaction is also time-resolved, i.e. the determined product of reaction, (e.g. the $(SCN)_2^-$ radical anion) decays, but its concentration is extrapolated to the duration of the pulse (Fig. 1). Transient character of dosimeter is accepted in that case because it integrates the inhomogeneous dose in the same way as at experiment on unknown species. One can say that the specific dosimetry in pulse radiolysis is the integral part of this technique and it cannot be applied in other fields of radiation chemistry. Vice versa, other methods of dosimetry can hardly be applied in the pulse radiolysis.

3. DOSIMETRY FOR MEDICAL PURPOSES

The dosimetry developed for medical irradiations will not be fully considered as well. Similarly as in dosimetry used in the pulse radiolysis, medical dosimetry deals with very inhomogeneous distributions of dose, e.g. delivered by a single pulse of the straight beam of electrons. In medical applications the collimated beam of gamma radiation is used, or more frequently the straight beam of electrons, protons or heavy ions, always with the intention to concentrate the energy on a well defined spot with the minimum dose deposited around it. The approach at radiation processing is opposite to this in medical irradiations, i.e. the beam is scanned, resulting in the broad beam geometry with a characteristic depth dose curve, substantially different to that at narrow beam geometry [4]. Nevertheless, some experience with phantoms is useful for radiation chemists. All irradiations have a common basis in general radiation physical chemistry (e.g. mass absorption coefficients) which is considered in radiation processing.

4. DOSIMETRY FOR PREPARATION OF GENERAL SCHEME OF RADIATION PROCESSING

Dosimetry plays the especially important role in industrial radiation processing, in the preparation of the plan of routine irradiations, involving instructions for a producer of irradiated goods. The case of gamma irradiation is comparatively simple, as it deals with a relatively high homogeneity of dose distribution. This is the case when the unit operation of irradiation is done in large installations, where objects of irradiation are moving around the sources, collecting and averaging absorbed dose. Nevertheless, the mapping of the dose distribution should be made, considering possible irregularities if all boxes are not filled identically with the irradiated material, or the simulation of contents of boxes is not possible. The situation is even more complicated at the case of electron beam irradiations, where practically only one-step irradiation is made. Split-dose or double-side irradiations are possible only in exceptional cases. The latter technique (of double side irradiation) may improve the homogeneity of the dose, but brings the danger of a deposition of charge and a damage of irradiated objects by violent discharge, in the shape of sparks. It is discussed in separate paragraph below.

It is obvious, that the heterogeneity of dose distribution has a strong connection to the economy of processing. If higher heterogeneity can be tolerated (i.e. a larger difference between the highest and the lowest dose in the material), the cheaper is irradiation to the required average dose. The radiation resistance of the irradiated material and the radiation microbiology in the case of radiation sterilization require the defined upper and lower limits of the dose distribution. It can be shown on many examples how much may be saved of the expensive radiation energy with the proper adjustment of the maximal and minimal doses, as seen from the careful dosimetric mapping of the whole conglomerate of objects irradiated in a standard box. A complete dosimetric analysis of the dose distribution can be very expensive and requires an application of sophisticated dosimetric approaches, e.g. to solve the problem of highly increased doses in a polymer layer, which is placed close to a metallic object like a surgical needle. Such situation demands sometimes a research to be done, for particular kind of polymer, because a local overdose sometimes can be tolerated.

The margins of free choice of dose distribution are rather narrow, especially at radiation sterilization. The minimum dose delivered in particular place of the box with medical supplies must be higher than sterilization dose determined by required level of inactivation of microorganisms. On the other hand the maximal dose in the site of the highest dose must be lower than this dose allowed by the resistance of a material of the device towards radiation. The limits are set by requirements of sterility and by undesired chemical changes, induced by radiation in the material. Other limits are established by the presence of parts of the object, which exhibit energy absorption characteristics very different to the main material. These are e.g., as mentioned metallic parts like needles. The producer of devices is advised to make such parts as thin as possible.

Simple cases, e.g. uniform systems like rubber latex to be vulcanized by radiation, transported in ready-to-sell containers of the size and geometry adjusted to the energy of electrons, scanning width and conveyer construction, are very rare indeed. In this case, and in the case of any viscous liquid, especially thixotropic one which does not flow easily if not mixed, they represent cases of highest use of beam energy, where the thickness of the irradiated layer of the material is equal to the distance between the identical entrance and exit doses. In connection with latex one can notice, that pumping of liquids like that is difficult and the dosimetry in such case is extremely difficult. Due to non-uniform flow, especially close to the walls, doses received by the viscous liquid moved by pressure are different in comparison to the expected distribution of doses measured by dosimeters placed in proper places of the irradiation site. Therefore we have found the irradiation of latex on the typical conveyor best than other methods, from the point of view of both - technology and dosimetry. The application of well tried technique shows the advantage of realization of unit operation principle as the method of choice [5].

Multi-component objects of a complicated geometry, i.e. the majority of medical supplies are far from the ideal latex situation described above. Although only 10% of radiation energy is utilized in that cases, dosimetry can help to improve this value, and at the same time, also the economy.

The question arises to what extend the dosimetry of objects of composed geometry and chemical composition has to be repeated in routine irradiations. Full dosimetry, like that applied during the preparation of irradiation plan, is impossible and certainly too expensive. The management of every large irradiation plant has elaborated its own strategy in dealing with the problem. The minimum requirement are the semi-quantitative dosimetric labels, preferably with the possibility of reading the absorbed dose as the optical density with a hand-held optical DRS spectrophotometer. Traditional go-no-go labels are too primitive and not up to the state of the art.

The wide gap between the real dose distribution inside the box and the simple semi-quantitative reading from the label attached to the upper and lower surface of the box can be overcome by using electronic system of control. It consists of a continuous record of the accelerator performance, traceable to the particular box position, as well as the measurement, if possible of the power passing through the box. The latter measurement is difficult to perform with a vertical beam of electrons hitting the horizontal conveyer. There is an advantage of horizontal beam accelerators in that respect, that the dose at hanging boxes can be measured indirectly, with a detector just behind the irradiated object.

The split dose irradiation is a necessity if required doses are of the order of 100 kGy and higher, e.g. in the case of cross-linking of polyethylene. Single irradiation to that dose is technically possible but cannot be applied because of intolerable increase of temperature of the object [5]. As there is the necessity to cool down the object and letting injected electrons to dissipate, the dosimeter may be attached for every subsequent passage of the material under the electron window. There are no satisfactory dosimeters, which would integrate the dose of, e.g. 200 kGy and indicate the accumulated dose properly. Electronic control and the registration of the history of irradiation is also needed in this case of very high accumulated doses.

In many places of the present paper the difference between γ and EB irradiation is shown. Basic differences in both sources of ionization energy are projected on dosimetry. One of the basic features of electron beam processing is the congestion of isodose distributions in comparison to gamma fields,

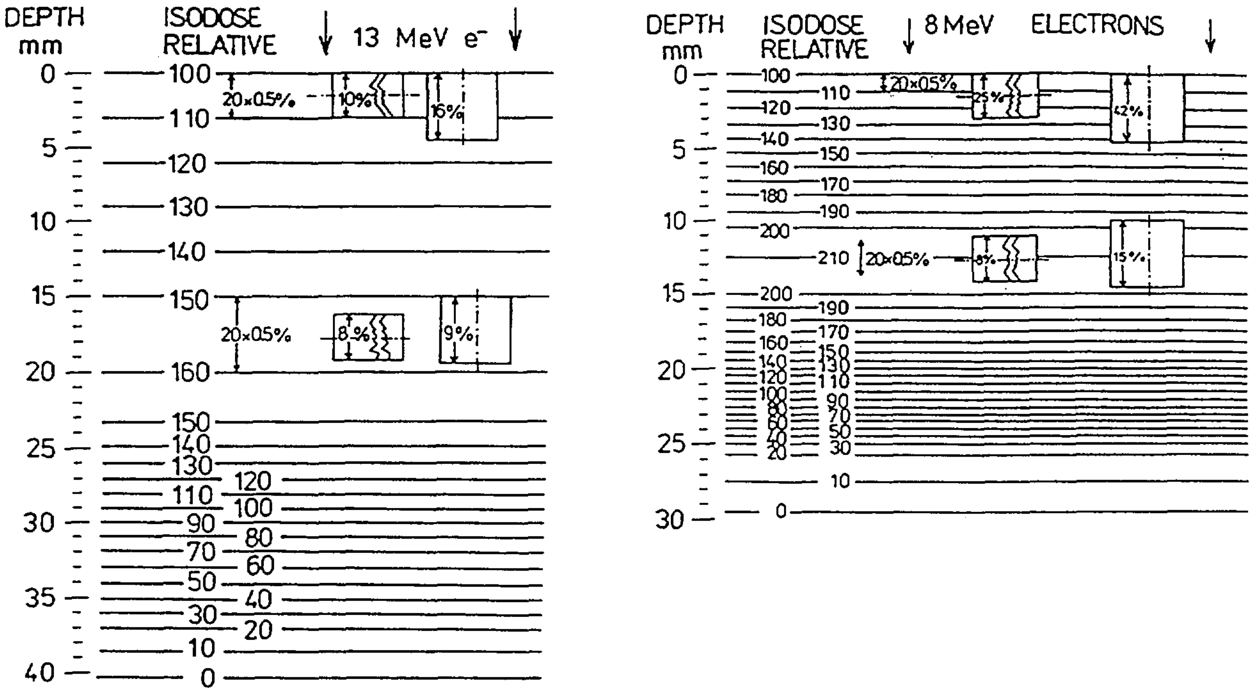

FIG. 2. Isodose layers in scanned electron beam irradiation of 13 MeV (a) and 8 MeV (b) energy. Object and dosimeter made of PCV. The surface dose (electron entrance dose) is taken as 100. Sizes of typical dosimeters are introduced with expected non-uniformities of dose distributions.

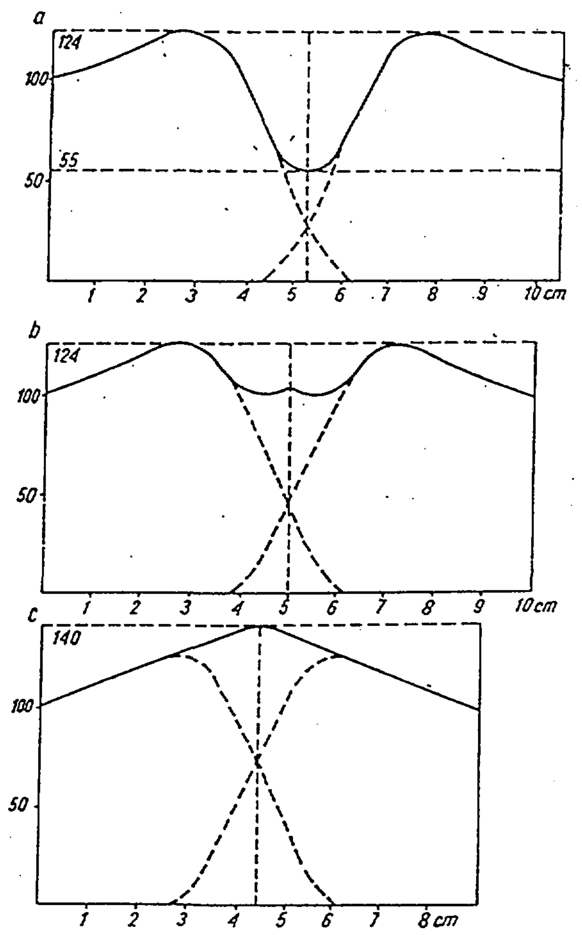

FIG. 3. Depth dose curves obtained in double sided irradiations as the function of thickness of material in the box. Entrance dose of 12-MeV electrons taken as 100%. There are three cases of thickness: a) Too high thickness of the material resulting in too low (55%) dose inside the object, b) Optimal thickness (24% overdose in relation to the minimum (entrance) dose, c) The case of too low thickness of the material (overdosage of 40% inside the object).

especially those in large cobalt-60 irradiation installations, where objects are moved across fields of very different dose rates. In the case of EB, the most convenient one step irradiation under the window passing the beam of electrons creates the dose distribution shown in Fig. 2 a and b. (Dose distribution in the shape of isodose curves is recalculated from depth dose curves describing absorption of electron beam in chosen medium, e.g. [4]). The congestion is already inconvenient at the sometimes allowed optimum of energy of 12 MeV, but is changing from bad to worse in the case of lower energies (8 MeV), not to mention such low energies as 2 MeV. The irradiated object obtains doses different in not very distant places. Geometric shapes and sizes of dosimeters which are excellent in γ-irradiation exhibit not acceptable behaviour at EB irradiation. The only solution is the application of thin layer dosimeters (see Ref. [7]).

The low range and the resulting congestion of doses has suggested the application of double side irradiations and for this purpose sophisticated mechanical constructions have been proposed, turning the boxes upside down on the conveyer, before the second run under the beam. That technique is acceptable under conditions of precise filling of boxes with the material to be irradiated. Figure 3 shows what happens if this condition is not fulfilled: substantial, easy measurable under- and over-exposures may occur. Special dosimetric control is advised in such cases, if severe errors in radiation processing have to be avoided. Application of wrong doses is not the only danger involved in double side irradiation.

For EB-radiation processing the problem of dosimetry of deposited charge arises. Deposition of electric charge in objects irradiated by electron beam is seldom taken into account. That negligence is justified when the thickness of irradiated material is limited, as usual to the „entrance dose-equal-exit-dose" treatment. Figure 4 shows the typical depth-dose curve, supplemented by the curve showing the depth distribution of deposited electrical charge. The maximum of charge is shifted towards the end of the range of penetration of electrons. Therefore the most of electrons are absorbed by the conveyor and lead to earth. The deposited charge can no longer be neglected when the object is thicker than the range of electrons in particular material. If the electrical conductivity of the material is low, the accumulated charge can reach high voltage potential, sometimes close to million of volts. It is easy to initiate the discharge, which rapidly takes the form of high intensity streams of current which creates paths of destroyed polymer. The phenomenon is used for decorative art, creating nice 'trees' in transparent polymers like PMMA [6]. In the case of radiation processing, e.g. of thick blocks of polymer the deposited charge can cause fire, which can be disastrous for the accelerator. Even without fire, the discharge destroys the material by burning holes in it. One can encounter that in techniques which are providing, sometimes with the help of sophisticated mechanical devices, the double side irradiation to improves the yield of radiation energy. There is no 'dosimetry' of the deposited charge in the sense used in the present paper. The function of dosimetry is played by theoretical preparation of the technology, considering electrical conductivities of the irradiated systems in the region of deposition of charge. After preparation of phantoms the experiment should be performed with increased doses of radiation. Careful inspection of the irradiated object for the presence of holes and/or 'trees' of discharges might convince the supervisor of production that the radiation processing in this particular case is safe for the integrity of product and the operation of irradiation is safe from the point of view of fire protection.

The deposition of electric charge has to be taken into account in the case of preparation of depth dose curves in thick phantoms (wedges, stacks etc.). If they are made of materials of poor conductivity and no charge collectors are provided, complications can occur. Phantoms well conducting the electric current i.e. all metals and polymers in which radiation induces the conductivity, like poly(vinyl chloride) presented in paper [4], are safe in that respect.

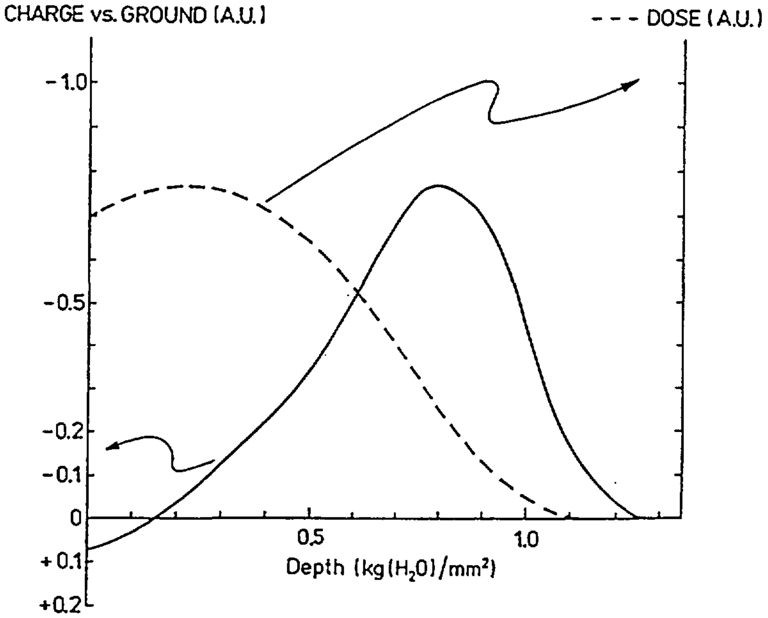

FIG. 4. Depth-dose curve, compared with the depth-charge curve for electron beam irradiation of 10 MeV energy, showing the accumulation of excess electrons at the end of the range of the beam, i.e. in the zone not used in the case of one-sided irradiation. The shift of the curve is substantial: the surface of the object is depleted of electrons.

5. SPECIAL CASES OF RADIATION PROCESSING

The question is asked sometimes if dosimetry is needed at all in the case of some exotic irradiations, far from usual radiation processing, represented, e.g. by radiation sterilization. Among seldom used applications there is a technology of perforation of thin plastic layers with a beam of heavy ions of high energy. Process of etching destroys chemically the burnt tracks. The result is a film with random holes and the product may be used for filtration of gases or liquids. In this technology the dosimetric control of the process is difficult, and the only quality assurance involves drawing attention to proper thickness of the film and its chemical quality and to keeping the parameters of heavy ion accelerator on the level previously tested as the optimal one. Computer programmes like TRIM help to determine the dose inside tracks, which often is between 10 and 100 MGy. Experimental dosimetry of heavy ion irradiations is controversial in general, because only a few percent of the absorbed energy is deposited in single ionization spurs, for which useful dosimetric methods have been developed. Most of energy is deposited in multi-ionization spurs, which represent rather the phenomenon of thermal spikes. It destroys the material completely, as it is in the case of holes and channels mentioned above. Dosimetry of energy deposited in tracks could be based on determination of debris of polymer extracted in the process of etching, but details of such method are not yet published.

6. CONCLUSIONS

Close cooperation between specialists of dosimetry and radiation processing is advised. High dose dosimetry became almost separate field in recent years. The quest for new dosimetric techniques has produced new systems, usually of no application. However, there were as well remarkable achievements in the field, probably impossible without wide front of research and new dosimetric proposals. The early confrontation with realities of large scale radiation processing may help in more efficient development of useful dosimeters. This applies in particular to high absorbed dose, high dose-rate dosimeters applied at high power electron beam irradiations. Two features of that kind of EB radiation processing have to be taken into account: the adiabatic delivery of energy, resulting in the increase of temperature of the object during irradiation and deposition of electric charge. Electron beam irradiations produce very congested isodose fields, demanding application of thin dosimeters both for the preparation of depth-dose curves and for reliable dose-mapping. Many dosimeters thick in shape, excellent for dosimetry in gamma radiation facilities, are useless at EB irradiations [7]. Dosimeters of thickness which cannot be reduced, are showing the average dose in a layer covering different doses. The dosimeter which averages the dose over unacceptable range of isodoses is of no help in detection of over- and under-irradiated regions in the object, what is the basic requirement in radiation processing for the purpose of sterilization of medical supplies.

Proper dosimetry applied to radiation processing creates a new branch of technology. Philosophy of the unit operation principle, useful in radiation processing [5] may be applied also to systematics of high dose and high dose-rate radiation dosimetry.

As the radiation processing is not supported economically by state organizations, even in former communist controlled regions with central steering, arbitrarily controlled economy [8], the proper dosimetry helps to keep the price for radiation processing in reasonable limits and promoting competition with other technologies, very often of inferior abilities as compared to radiation processing.

Dosimetry applied in radiation processing is in a constant development and revision. One can conclude that from efforts of American Society for Testing and Materials (ASTM) (recently in collaboration with ISO) in which the Subcommittee E10.01 prepares standards, e.g. E1261 'Guide for Selection and Calibration of Dosimetry Systems for Radiation Processing' (it will be published in 1999, as ISO 15556); E1707 'Guide for Estimating Uncertainties in Dosimetry for Radiation Processing' (it will be published in 1999 as ISO 15572); 'Practice for Dosimetry in a Gamma Irradiation Facility for Radiation Processing' (to be published in 1999 as ISO 15571); E1649 'Practice for Dosimetry in an

Electron Beam Facility for Radiation Processing at Energies between 300 keV and 25 MeV' (it will be published in 1999 as ISO 15569 with a revised scope); E1608-98 'Practice for Dosimetry in an X-Ray (Bremsstrahlung) Facility for Radiation Processing'. All standards refer to many related standards serving the quality of radiation processing.

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