High-Dose Standardization Study For γ-rays radiation processing at NIM

ZHANG Yanli, GAO Junchen, YANG Yuandi.

Division of Metrology in Ionizing Radiation and medicine, National Institute of Metrology,

No.18, Bei San Huan Dong Lu, Beijing 100013, China

Abstract

The high dose standard and dissemination system of γ-rays has already established at NIM. Fricke dosimetry is accepted as primary standard. The transfer standards include alanine/ESR dosimetry and dichromate solution dosimetry. Those dosimeters possess many favorable characteristics, such as good linearity, long shelf life, reproducible and stable radiation response and equivalence to water in terms of radiation absorption properties.

In order to guaranteeing the product quality of radiation processing and promoting the progress of radiation techniques. On April 18, 1990, SBTS and SCST had jointly issued "the Provisional Regulation of Metrological Supervision and Management for Radiation processing". This regulation stipulates that all the enterprises and institutions dealing with radiation processing should apply for "the Measurement License for Radiation Processing" from the local metrological administrative department of provincial government.

1. Introduction

In recent years, the radiation processing with γ-rays and EB has been extensively developed in China. At the present more then 150 γ-rays irradiation facilities and more than 80 EB irradiation facilities spreading all over the country have been set up, which are for plot demonstration or commercial scale, and for multipurpose, such polymer modification, medical product sterilization and food preservation. With the growing, the problem on control of the quality of irradiation products was placed on the order of the day. It is well know that the key element for control of the quality is reliable dosimetry in radiation processing.

In 1983, National Institute of Metrology (NIM) initiated a larger research project. The title of the project is "To establish Metrological standards for dosimetry of ^{60}Co γ-rays used in medicine, industrial irradiation and agriculture". Its focus point is on the industrial irradiation. At that time, the standardization on dosimetry of industrial radiation processing made the first step in China. When the research was completed in 1988, several new primary and secondary metrological standards for dosimetry had been built up. These dosimetric standards include Fricke system, alanine-EPR system, potassium (silver) dichromate system, and silver dichromate system Together with the old standards, graphite calorimetry system and graphite ionization chamber system, there is a more complete group of metrology systems for high-dose dosimetry at NIM.

In order to guaranteeing the product quality of radiation processing and promoting the progress of radiation techniques in China, the State Bureau of Technical Supervision (SBTS) and the State Commission of Science and Technology (SCST) have jointly issued" the Provisional Regulation of Metrological Supervision and Management for Radiation Processing". This is an administrative rule, which provides the regulation of radiation measurement license for radiation processing plants. SBTS has decided to establish the State Office of Technical Examination for Measurement License of

Radiation Processing which is attached to the National Institute of Metrology (NIM). Its main tasks include establishment of examination plans for radiation processing license, organization of technical examination and other works related to the measurement regulation and performance of measurement supervision and regulation in radiation processing fields.

The program contains following works:

1. To establish national primary and secondary dosimetry standards in radiation processing;
2. To choose the NIM transfer standard dosimetry system;
3. To train the technical personal in dosimetry for irradiation facilities and to help them to set up their own routine dosimetry system; and
4. To draw up concerned technical regulations.

2. DOSIMETRY SYSTEMS

The high dose measurement system for γ-rays, including primary standards, transfer standards and working dosimeters, has been established. The schema of dosimetry tractability to primary standards for γ -rays is illustrated in Fig.1.

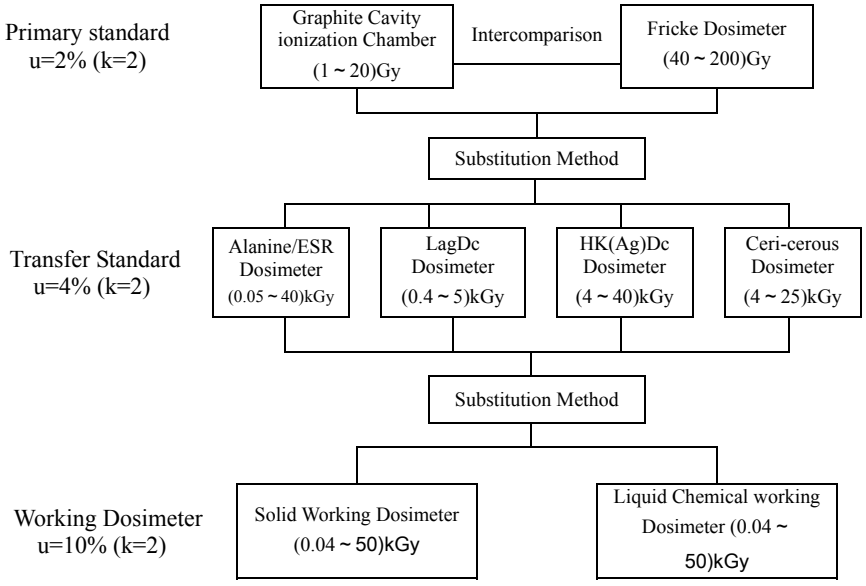

FIG.1. The schema of dosimetry tractability to national primary standards for γ-rays

2.1. Fricke Dosimeter (primary standard)

The Fricke dosimeter is used as the national absorbed dose standard of irradiation processing in China to calibrate the standard dosimeters and transfer the absorbed dose, and the act as the standard or working dosimeter and to calibrate the γ-ray irradiation field. It also can measure the dose mapping in the space and confirm the dose delivered to the products, and check and accept the freshly built or replenished source and the newly start-rising dose. It has given service to more than a hundred provincial standard laboratories or irradiation processing installations.

The methods used for preparation, measurement and calculation of results are essentially as described in ASTM standard E 1026 [1], except that the solution is aging and preirradiated and is dispensed into a standard flame-sealed 2 ml pharmaceutical ampoule of diameter 12 mm and overall height not exceeding 55 mm.

We use Fricke dosimeter to be primary standard of our country. This may be considered as a absolute method, since the value of chemical yield of ferric ions is well documented (G = 1.61 mJ^{-1}) and molar linear absorption coefficient of ferric ions has been measured accurately to be 216.7 ± 0.9 (m^2mol) with Cary 4E UV/vis spectrophotometer. which has good linear related factor (r>0.999990) given by using liner regression analysis. when absorbance plotted against ferric ions concentration in 0.01 to 1.30 Abs range. In addition, by using the technique of aging and preirradiation in preparing dosimetric solution, the microimpurities can be controlled effectively, and the yield will not be affected, so we can conveniently use the cheap natural glass ampoule as irradiation vessels in mall comparison and calibration services. This dosimetry has a reproducibility within ±1.0% in absorbed dose measurement and the total systematic uncertainty is estimated to be ±1.9% at 95% confidence level.

2.2. Dichromate Dosimeter (transfer standard)

The Dichromate Dosimeter is used by NIM during product dose mapping studies for critical process parameter determination; during sterilization dose determination for product qualification studies; and for routine process control where a high degree of accuracy is required. These dosimeters are sold on a supply and measurement basis to users and suppliers of commercial irradiation services in China to verify the response of their routine dosimetry systems or confirm the dose delivered to their products. The dosimeters are also used as a transfer standard during the in-plant calibration of the routine dosimetry system used by China's only commercial irradiation facilities.

The methods used for preparation, measurement and calculation of results are essentially as described in ASTM standard E 1401 [2] using the potentiometric method except for the following. A low range Dichromate dosimeter (hereafter called low-range dosimeter LAgDc) is prepared and calibrated over the range 0.5-5 kGy using concentrations of 0.35 mmol• L^{-1} silver dichromate ($Ag_2Cr_2O_7$) in 0.1molL^{-1} aqueous perchloric acid ($HClO_4$), and a high range dosimeter (hereafter called high-range dosimeter HKAgDc) is prepared and calibrated over the range from 5 to 40 kGy contains 2 m(molL^{-1} potassium dichromate ($K_2Cr_2O_7$) and 0.5 mmolL^{-1} silver dichromate ($Ag_2Cr_2O_7$) in 0.1 molL^{-1} aqueous perchloric acid ($HClO_4$) air saturate both solutions before use. They are contained in standard flame sealed 2-ml pharmaceutical ampoule of diameter 12 mm and overall height not exceeding 55 mm. The radiation chemical yield G or the dose-response conversion factor k of dichromate dosimeters for γ-rays irradiation is calibrated with NIM's Fricke dosimeter.

Position the dosimeters in the radiation field in a defined, reproducible location and irradiate them to dose in the range of utilization. Record geometric position, the time of irradiation, and the temperature of the dosimeter during irradiation, temperature variation, and the date. The total systematic uncertainty is contributed by many factors. The combined uncertainty of an absorbed dose determined by the both of dichromate dosimetry system is estimated within ±4% at a 95% confidence level.

2.3. Alanine/ESR System (as a transfer Standard in NDAS)

NIM has studied alanine/ESR dosimetry system since 1983 and utilized it as a transfer standard to establish a program of National Dose Assurance Service (NDAS) program since 1988, which is used to unify quantity of absorbed dose in high level dosimetry all over china.

We prepare the alanine dosimeter for the NDAS program at the NIM. One dosimeter consists of three cylindrical samples with 3mm diameter, 8mm long and 65mg mass and a Perspex capsule with 12.5 mm in diameter and 36mm in length. The composition of the sample is 80% DL- **α**-alanine

reagent and 20% paraffin. The numbers are carved on the end of the capsule. In deciding the material and the dimensions of both sample and capsule, the following factors have been considered: low preparation cost, sensitivity of the sample to dose and its lower detective limit, measurement accurate, water equivalency, secondary electron equilibrium, protection from moisture, convenience for calibration and easy to mail etc.

A type of JEF-FA200 ESR spectrometer has been used for determining the alanine sample's absorbed dose by measuring its ESR spectrum. The spectrometer operates in X-band at room temperature. The corresponding microwave frequency is about 9.8 GHz with 100 kHz modulation and automatic frequency locking. The principle in choosing setting parameters of the instrument is to obtain higher ESR amplitude spectrum of irradiated alanine sample. The decided settings are RF modulation amplitude 0.63 mT, microwave power 2 mW, sweep time 4 min, time constant 0.3s, but the receiver gain is variable according to absorbed dose. The accuracy ratios among the receiver gains had been determined with a group of alanine samples irradiated with different doses.

The dose-response conversion factor k of the alanine/ESR dosimeters for γ-rays irradiation is calibrated with NIM's Fricke dosimeter.

2.4. Dosimetry intercomparison, calibration and NDAS Program.

In Oct. 1986, intercomparison of exposure and absorbed does standards was conducted between NIST and NIM, using five NIST's transfer standard graphite ionization chambers. The results evenly reasonable agreement for the exposure primary standards of X-rays and gamma rays in air, with deviation from -0.1% to 0.7%. For the absorbed dose in water of gamma rays, the agreement of comparison between calorimeter (NIST) and Fricke dosimeter (NIM) is also very good, for which the deviation is about 0.3%.

In July 1998, NIM participated in CCRI comparison of standards of absorbed dose to water for $^{60}C_o$ at high dose levels among primary dosimetry laboratories organized by IAEA in collaboration with the BIPM and conducted using the NIST alanine/ESR dosimeter system and the NPL alanine/ESR dosimeter system. The results of comparison between the NIM Fricke dosimeter system and NIST alanine/ESR dosimeter system are given in Table 1 and those and NPL alanine/ESR dosimeter system in Table 2.

TABLE 1. Results of absorbed-dose estimates for NIST transfer dosimeter

Lab	$D_{w,lab}$ /kGy	$D_{w,issue}$ /kGy	R_{dosim}	R_{dosim}	S_{level}	R	s_R	u_R
NIM	5.035	5.149	0.978	0.9871	0.0092	0.9871	0.0029	0.019
	5.035	5.094	0.988					
	4.980	5.084	0.980					
	4.930	5.057	0.975					
	15.12	15.41	0.981	0.9830	0.0038			
	15.12	15.39	0.982					
	14.96	15.06	0.993					
	14.81	15.19	0.975					
	30.23	30.39	0.995	0.9981	0.0031			
	30.23	30.38	0.995					
	29.90	29.68	1.007					
	29.60	29.74	0.995					
BIPM	1.163	1.166	0.997	0.9949	0.0017	0.9949	0.0017	0.012
	1.158	1.160	0.998					
	0.940	0.947	0.993					
	0.935	0.943	0.991					

TABLE 2. Results of absorbed-dose estimates for NPL transfer dosimeter

Lab	$D_{w,lab}$ /kGy	$D_{w,issue}$ /kGy	R_{dosim}	R_{dosim}	S_{level}	R	s_R	u_R
NIM	5.035	5.048	0.990	0.9971	0.0024	1.0037	0.0025	0.019
	5.035	5.029	1.001					
	4.980	4.983	0.999					
	4.930	4.944	0.997					
	14.93	14.93	1.000	1.0027	0.0014			
	14.93	14.91	1.001					
	14.77	14.67	1.007					
	14.62	14.58	1.003					
	30.23	30.13	1.003	1.0113	0.0051			
	30.23	29.46	1.026					
	29.90	29.62	1.010					
	29.60	29.42	1.006					
BIPM	1.163	1.157	1.005	1.0029	0.0014	1.0029	0.0014	0.012
	1.158	1.152	1.005					
	0.940	0.936	0.999					
	0.935	0.938	1.002					

Several methods of calibration are accepted. Among the NIM primary standards, the Fricke dosimeter is chosen to make dose calibration for NIM dichromate dosimeter alanine dosimeter. The calibration is carried out using a 3.7×10^{15} Bq opened beam and Barrel source at China Institute of Atomic Energy Sciences (CIANES). The 30×30×30 (cm^3) dimension water phantom is fixed in the radiation filed in a defined, reproducible location. The irradiation facilities dosimeters may be sent to NIM for irradiation in an accurately standardized radiation field, and the readout of dose may be made at NIM and at the facility. Alternatively transfer standard dosimeters may be provided by NIM for irradiation at the facility, to calibrate a reference point in radiation field of the facility, which may be used for calibrating of working dosimeter. So far about for 70% irradiation facilities have established traceability to national standard through direct calibration and NDAS program.

As far as the procedure is concerned, the NDAS program is similar to IDAS program by IAEA in 1985. Each γ-rays radiation processing facility, who participates in the NDAS program, will receive a set of two alanine dosimeters as transfer standard with an information sheet from the NIM every six months by post.

Two irradiation patterns for the participant. The one is that the alanine dosimeters are irradiated along with facility's own routine dosimeters simultaneously under similar conditions. The other is that both alanine and routine dosimeters are irradiated one-by-one under the same condition at the fixed position, e.g. at the reference point in the radiation field. In the latter pattern, the irradiation duration for the two kinds of dosimeters can be either the same or different. Based on the irradiated dose or doses of the routine dosimeters, the nominal dose of the alanine dosimeters is estimated by the participant. Then the alanine dosimeters along with the information sheet filled in are returned to the NIM for measurement by ESR spectroscopy to obtain the evaluated dose. On the basis of the nominal and evaluated doses, the percent deviation of the participant's dosimetry against the NIM dosimetry is calculated. Finally, the assessed result with the deviation is mailed to the participant in a signed and sealed certificate. Up to now, the NDAS program has been implemented for more than ten years, and the number of participants is near 60. By the end of August 2006, altogether 1200 dose checks had been carried out. 790 of 1200 checks fall on the deviation range less than ±5%, making up 65.9% of all checks. 240 and 167 checks fall on the range from ±5% to ±10% (20.2%) and on the range more

than ±10% (13.9%), respectively. The NDAS has already provide a useful method to ensure the working dosimeters trace to the national standard and take an important role to control quality of radiation processing in China.

3. Metrological Supervision and Management.

3.1 Administrative regulation

In order to guaranteeing the product quality of radiation processing and promoting the progress of radiation techniques. On April 18, 1990, The State Bureau of Technical Supervision (SBTS) and State Commission of Science and Technology (SCST) had jointly issued "the Provisional Regulation of Metrological Supervision and Management for Radiation processing". This regulation stipulates that all the enterprises and institutions dealing with radiation processing should apply for "the Measurement License for Radiation Processing" from the local Metrological administrative department of provincial government, and whoever applying for the license of radiation processing should meet the following requirements.

In order to guaranteeing the product quality of radiation processing and promoting the progress of radiation techniques in China, the State Bureau of Technical Supervision (SBTS) and the State Commission of Science and Technology (SCST) have jointly issued" the Provisional Regulation of Metrological Supervision and Management for Radiation Processing". This is an administrative rule, which provides the regulation of radiation measurement license for radiation processing plants. SBTS has decided to establish the State Office of Technical Examination for Measurement License of Radiation processing which is attached to the National Institute of Metrology (NIM). Its main tasks include establishment of examination plans for radiation processing license, organization of technical examination and other works related to the measurement regulation and performance of measurement supervision and regulation in radiation processing fields.

(1) Measuring instruments, verification equipments and accompanying facilities should fit
The processing ability and should be verified and qualified;

(2) Metrological personnel in charge of Metrological verification and testing should be suitable for radiation processing work, and should be examined and qualified;

(3) Working environment should satisfy the processing requirement and guarantee the normal work of measurement and testing inspection

(4) Working and management regulations should be set up to safeguard the accuracy of absorbed dose of radiation processing products.

This is an administrative rule, which provides the regulation of radiation measurement license for radiation processing plants. SBTS has decided to establish "the State Office of Technical Examination for Measurement License of Radiation Processing", which is attached to NIM.. Its main tasks Include establishment of examination plans for radiation processing license, organization of technical examination and other works related to the measurement regulation and performance of measurement supervision and regulation irradiation processing fields.

3.2 Standard and technical norm

For implementing the Metrology Law and Provisional Regulation, we exercise compulsory verification of working instruments and radiation processing. The Metrological verification shall be conducted pursuant to the verification system, and the verification regulation shall be applied when conducting verification.

Nine national standards and eight technical norms have been issued, witch are following

(1) GB 139-89 Standard method for using the ferrous sulfate (Fricke) dosimeter to measure absorbed dose in water.

(2) GB/T 15053-94 Standard method for using the radiochromic film and polymethylmethacrylate dosimetry to measure absorbed dose

(3) GB/T 15447-95 Conversion method of absorbed dose in different materials irradiated by X, γ–rays and electron beams

(4) GB/T 15446-95 Terminology relating to radiation processing dosimetry

(5) GB/T 16334-96 Practical guide of dosimetry in a gamma irradiation facility for food processing.

(6) GB/T 16509-96 Standard guides for estimating uncertainties in dosimetry for radiation processing.

(7) GB/T 16640-96 Standard guides for selection and calibration of dosimetry systems for radiation processing.

(8) GB/T 16639-96 Alanine-ESR dosimetry system for radiation processing.

(9) GB/T 16841-97 Guide for dosimetry in an electron beam facility for radiation processing at energies between 300kev and 25 Mev.

(1) JJG 591-89 Ray Radiation Source (for Radiation Processing).

(2) JJG 1018-90 Standard Method for Using the Potassium (Silver) Dichromate Dosimeter to Measure γ-rays Absorbed Dose in Water.

(3) JJG 1020-90 Monitoring Method of Dose Assurance for Radiation Processing with γ-rays.

(4) JJG 735-91 Verification Regulation of the Standard Dosimeter of Water Absorbed Dose for γ-rays (Radiation Processing Level).

(5) JJG 1028-91 Standard Method for Using the Silver Dichromate Dosimeter to Measure γ-rays Absorbed Dose in Water.

(6) JJG775-92 Verification Regulation of the Working Dosimeter γ-rays Radiation Processing.

(7) JJG 772-92 Electron Beam Radiation Source (for Radiation Processing).

(8) JJG 851-93 Verification Regulation of the Working Dosimeters for Electron Beam Radiation Processing.

These regulations will provide the technical basis for the regulatory approval of irradiation facility and the standardization of high dose measurement.