
²⁰⁴Tl Source Replacement and Associated Potential Effects on Dosimetry

Purpose

This bulletin is provided to inform clients of the Pacific Northwest National Laboratory (PNNL) who request/receive dosimeter irradiations using ²⁰⁴Tl of potential biases associated with the implementation of a new source and geometry.

Scope

The Calibration Laboratory for Ionizing Radiation (CLIR) at PNNL irradiates dosimeters using beta particle sources, including ²⁰⁴Tl. We have become aware of problems with the responses of certain types of dosimeters that have been exposed to a recently installed ²⁰⁴Tl source configuration.

This bulletin briefly addresses

- the criteria for ²⁰⁴Tl beta particle source characteristics
- how PNNL calibrates and characterizes its beta particle sources
- the specific characteristics of the former and new sources
- what effects on dosimetry have been observed
- what is being done and what can be considered to help resolve noted disparities.

Background

The CLIR replaced a high-activity ²⁰⁴Tl beta particle source (Reference No. 318-192), which had been in use since 1990. This source was initially calibrated and used with a flattening filter that was supplied for use with a 0.5-mCi ²⁰⁴Tl beta particle source used prior to source 318-192. The earlier source and flattening filter were manufactured and marketed via Buchler and Amersham in Germany and was distributed with certified traceability via the Physikalisch Technische Bundesanstalt (PTB), the national physical

laboratory of Germany. The same flattening filter has been used for each of the beta sources described here. The newest ²⁰⁴Tl beta source (Reference No. 318-360) was manufactured by the same U.S. manufacturer that produced source 318-192, but it differs in its encapsulation and the specific activity of the source material. Characteristics and tolerance for suitable beta source reference fields are provided in various standards of the International Organization of Standardization (ISO), the U.S. Department of Energy (DOE) and the American National Standards Institute (ANSI) in conjunction with the Health Physics Society (HPS). The table on Page 6 identifies each of the pertinent characteristics associated with the formerly used source and the new source. Each of the beta sources has been characterized using the same criteria, and each was confirmed to be consistent with the requirements of those criteria. However, subtle changes in encapsulation may potentially affect the spectrum of beta radiation received at the normal position of dosimeter irradiations. The further implication is that dosimetry devices irradiated using ²⁰⁴Tl in Categories V and VII as specified in ANSI/HPS N13.11-1993 (ANSI/HPS 1993) and DOE/EH-0027 (DOE 1986), or Category V of ANSI/HPS N13.32-1995 (ANSI/HPS 1995), may display a biased shallow dose response to the new source if the spectral differences are of sufficient magnitude.

The calibration and characterization of beta source reference fields provided at the CLIR are primarily guided by ISO 6980 (ISO 1984), with additional criteria specified in dosimetry performance test standards ANSI/HPS N13.11 (ANSI/HPS 1993), ANSI/HPS N13.32 (ANSI/HPS 1995) and DOE/EH-0027 (DOE 1986). The key parameter that is used to characterize the energy of the beta radiation field is the *Residual Maximum Energy* (E_{res}). The intent of evaluating this parameter is to assure that neither encapsulation nor filtration excessively lowers the maximum energy of the beta particle fluence spectrum present at the calibration position. A second key factor is the ratio of the in-phantom dose rate transmitted through 20-mg/cm² and 7-mg/cm² of tissue-like media.

Background (contd)

All of the measurements required by various test standards and recommended by ISO 6980, were performed prior to using the new beta source (318-360) for dosimeter irradiations. These measurements verified that the characteristics of the new source, with the identical flattening filter used with source 318-192, were within the tolerance of the various standards. The E_{res} of the new source was lower than the formerly used source but was still within the limit of acceptability (see table on Page 6) specified by ISO 6980 (ISO 1984). Therefore, our initial decision was to use this source in the same configuration as the source it would replace. We anticipated that certain dosimeter models could be sensitive to almost any perceptible change in beta energy or geometry. This is a primary concern when any reference field is replaced, but especially when dealing with low energy radiation. In order to evaluate possible changes in dosimeter response, we measured the response of thermoluminescent (TL) chips (Harshaw TLD-700) with areal density of 40-mg/cm² that were placed in a specially designed jig beneath nine layers of aluminum filters ranging in areal density from 13-mg/cm² to 1100-mg/cm². From this evaluation of the two sources, we observed no differences in response that were considered statistically significant.

The laboratory always approaches any change-out of sources with caution and typically performs case-specific irradiation trials to identify potential differences in dosimeter response. Additional characterization measurements for the new beta source were in progress in October 2000, but before these measurements could be completed, the encapsulating window of source 318-192, began to lose integrity. This forced us to abandon its use and proceed with implementation of the new source that was already verified to have energy characteristics consistent with the criteria specified in ISO 6980 (ISO 1984).

The failure of source 318-192 also placed additional time pressure on the determination of field uniformity for the new source. ISO 6980 (ISO 1984) specifies that a flattening filter be used if the field non-uniformity is greater than $\pm 5\%$. An assessment of source 318-192 was performed using a variety

of measurement techniques and it was determined that the source satisfied the criterion; however, uncertainties associated with these measurement techniques were nearly as high as the criterion itself. This large component of uncertainty was assigned to the uniformity of the field and was the major constituent of the expanded uncertainty for the reference field. Field uniformity was also measured for the new source. These measurements revealed that the existing flattening filter appeared to be over-correcting the radiation field for source 318-360, and was perhaps not the ideal filter for the newer source. Furthermore, we suspected that a decision in the early 1990s to perform irradiations at a distance of 35-cm, instead of the previously used 30-cm, also *may* have contributed to the field non-uniformity by reducing the effective area "shadowed" by the filter. This could have also contributed to the uncertainty in uniformity measurements performed on the previous source.

The flattening filter was removed and the field uniformity was re-evaluated. The results from multiple measurement methods showed good consistency and demonstrated that the radiation field was uniform to within $\pm 5\%$ over the typical area on the phantom used for irradiating dosimeters. At that point, we considered that ISO-6980 (ISO 1984) does not preclude the use of ²⁰⁴Tl without a flattening filter under these conditions of uniformity. Furthermore, use without the filter had the potential to compensate for the decrease in E_{res} , relative to source 318-192, that was observed in the earlier assessment with the flattening filter. Upon reevaluation of the reference field characteristics without the flattening filter in place, we found that the E_{res} and the 20- and 7-mg/cm² transmission factors were nearly identical to the characteristics of the old source.

Following confirmation of the consistency of the new and old sources with the criteria specified in ISO 6980 (ISO-1984), the new source without the flattening filter was implemented for general use in October 2000. At this same time, we initiated another dosimetry-based intercomparison using the multi-filtered jig and 40-mg/cm² LiF TL chips used earlier. Measurements were performed using the new source with and without the flattening filter. This intercomparison produced some measurement results with increased response at certain depths, but these results did not

Background (contd)

indicate a problem with removing the flattening filter. The increased response did not appear to trend with the filter depth and the measurements had poor precision. We also realized that this evaluation was weighted to represent thicker filters and did not correspond to some of the common beta window thicknesses of widely used dosimeter types.

While these measurements were underway, we began to receive feedback from several clients that there appeared to be elevated dosimeter responses as compared to those found when we used the previous beta source and configuration. The new source and configuration had also been used for performance testing within the National Voluntary Laboratory Accreditation Program (NVLAP) with due caution and scrutiny of results. In the course of investigating the claims of elevated response, we examined the performance testing results and could find no evidence of statistical bias. However, the majority of the performance testing results were from participants using thin TLD phosphors. The specific problems of elevated responses had been identified by clients that were using dosimeters with thicker sensitive elements. When this became evident, we initiated a detailed evaluation of flattening filter dependence for several thicknesses of TL material behind various thicknesses of polyethylene terephthalate (PET) filters ranging from 0- to 100-mg/cm². The selection of PET filters in this evaluation permit the use of smaller filter thicknesses to better simulate more commonly used beta dosimeter windows. This study is still in progress and is anticipated to include a similar set of measurements at the National Institute of Standards and Technology (NIST) and the Radiological and Environmental Sciences Laboratory (RESL), the Department of Energy Laboratory Accreditation Program (DOELAP) testing facility in Idaho Falls, Idaho, using their respective irradiation geometries, both of which employ a flattening filter.

Theory

Based on the information gained to date, it is assumed that the observed differences in dosimeter sensitivity are

generally related to the number of beta particles entering the dosimeter at high oblique angles relative to the number following a more direct path from the source to the detector. When a flattening filter is in place, the number of obliquely incident beta particles tends to be larger than when the filter is absent. Since the beta particles incident at high oblique angles are likely to be lower in energy and less penetrating, it is possible that different thicknesses of dosimeter elements would respond differently to the two irradiation conditions. Beta particles from the “no-filter” geometry would generally be expected to penetrate further into the dosimeter material, resulting in more signal during processing.

The primary device used to measure the characteristics of the beta fields is a thin-window extrapolation ionization chamber that is relatively insensitive to changes in beta particle energy. Because this chamber is designed to yield the dose at zero (0) chamber depth, it would not likely be as sensitive to the angular distribution of the incident beta fluence. With these assumptions, thicker TL chips might be expected to respond higher, relative to the extrapolation chamber-determined dose rate, without the flattening filter present in comparison to the relative response with the flattening filter in place. This is consistent with our observations to date.

Timeline

The old source with the flattening filter was last used on October 3, 2000. The new source has been in use without the flattening filter since October 16, 2000. Unless specifically requested/arranged, all ²⁰⁴Tl (point source) exposures performed on or subsequent to that date have been executed without the use of the flattening filter.

Applicability

At this time, we suspect that the most significant potential effects will be on dosimeters that employ relatively thick sensitive elements (e.g., chips of the 40- or 100-mg/cm² variety). Although the affect upon thin phosphors has not been ruled out, noteworthy evidence of an influence has not yet been documented. Film dosimetry is currently being investigated.

Near-Term Status

Because this source/geometry has already been in use for over one year, we have been reluctant to reinstate the general use of the flattening filter. Until we complete our current investigations and acquire measurements from NIST to aid in making a better-informed decision, we would like to avoid further complicating the issue. We are, however, open to providing irradiations using the flattening filtered geometry on a case-by-case basis. In such cases, we would consider the nature of the dosimeter being used and what we have observed in our intercomparison evaluations to date.

As an alternative, we also would consider providing no-cost irradiations of a limited size “calibration” set that would allow your lab to identify a new or modified correction factor related to the current source/geometry. In such cases, we also ask that you inform us of the identified differences in response to enable us to establish a better understanding of this issue.

Long-Term Resolution:

We realize that, for some dosimeter types, this issue has created biases in periodic confirmatory-type measurements, which has, in turn, demanded investigations and explanations by processing staff. We empathize with the difficulties of this awkward situation. Although the cause appears to be evident; the most appropriate resolution is not yet clear. The following actions and their potential effects are being considered:

- *Reinstate the existing flattening filter.* While this may offer some advantages for the type of dosimeters currently affected by the absence of the flattening filter, it is a reasonable possibility that other dosimeters may experience biases due to the decrease in maximum residual energy (i.e., in comparison to the formerly used source). Furthermore, the difficulty in resolving the uniformity of the flattened field – coupled with the magnitude of the potential non-uniformity itself – contradicts the need for the flattening filter, as identified in ISO 6980.

- *Reinstate the existing flattening filter and adjust the irradiation distance.* This alternative is predicated on the idea that a decreased source-phantom distance may enable the filter to create a more uniform field and would potentially increase the maximum residual energy. Unfortunately, a change in phantom distance may create another potential bias in dosimeter response due to the offset of each dosimeter from the phantom surface. Given the difficulty of moderate energy beta dosimetry, the effects of such a change would need to be evaluated for a multitude of dosimeter types. There is also no guarantee that moving to an alternate distance would aid in establishing the desired uniformity (i.e., $\forall 5\%$) across the area of dosimeter placement on the phantom.
- *Replace the existing flattening filter with a more applicable design.* In our measurements of the field uniformity, we found that the existing flattening filter is potentially overcorrecting the field uniformity. As such, the most reasonable approach may be to redesign and apply a modified filter that could specifically complement the new source and our particular irradiation distance. This is not necessarily a straightforward task. Modeling could be of some use in redesigning the filter; however, identifying the source geometry (i.e., that of the active area) may be difficult. Empirically based design of the new filter would be time consuming and, upon completion and implementation, may still induce an effect (i.e., bias) upon various dosimeter types.

We will consider these options, as well as any additional alternatives, as we continue to evaluate the situation. We value input from our clients and/or proficiency testing beneficiaries – even if not directly affected by this issue. If you would like to provide input or simply express questions or concerns, please contact us via the following channels:

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Source Characteristics

Characteristic	Criteria^a	Old Source (318-192)	New Source (318-360)
Source Material	“Should be stable with time” (6980)	Electroplated thallium metal (thallic iodide)	Thallium salts in film matrix
Source Diameter	# 16-mm (6980)	8-mm	10 mm
Front Encapsulation (Window)	“..sufficiently robust and stable to withstand normal use without damage to the source and leakage of the radioactivity...” (6980)	6.6-mg/cm ² (1-mil) glass (inner) 8.5-mg/cm ² (0.3-mil) Kapton (outer)	0.1-0.2-mg/cm ² acrylic (inner) 9-mg/cm ² Kapton (outer)
Reference Distance	∃ 30-cm (13.11 / 0027)	35 cm	35 cm
Maximum Residual Energy	0.53 # E _{res} # 0.763-MeV (6980)	0.608-MeV (w/Flattening Filter)	0.557-MeV (w/Flattening Filter) 0.602-MeV (No Flattening Filter)
20:7-mg/cm ² Dose Rate	0.80 ∇ 0.05 (13.11 / 0027)	0.838 (w/Flattening Filter)	0.780 (w/Flattening Filter) 0.842 (No Flattening Filter)
Expanded Uncertainty ^b	none	11.8%	11.8% (w/Flattening Filter) 3.6% (No Flattening Filter)

a Criteria and/or recommendations for Tl-204 beta fields. Specific reference is identified in parenthesis: 6980 = ISO-6980 - *Reference Beta Radiations for Calibrating Dosimeters and Doseratemeters and for Determining Their Response as a Function of Beta Radiation Energy*, 13.11 = ANSI Standard HPS N13.11-1993 – *Personnel Dosimetry Performance – Criteria for Testing*, 0027 = DOE/EH-0027 – *Department of Energy Standard for the Performance Testing of Personnel Dosimetry Systems*.

b Quoted for the dose delivered to dosimeters placed on the phantom.