# On the radioanalytical methods used to assay stainless-steel-encapsulated, ceramic-based $^{90}$Sr–$^{90}$Y intravascular brachytherapy sources

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## Abstract

Very quantitative radiochemical procedures for the destructive assay of stainless-steel-encapsulated, ceramic-based $^{90}$Sr–$^{90}$Y intravascular brachytherapy sources (termed 'seeds') have been devised. These seeds, developed and provided by Bebig Isotopentechnic und Umweltdiagnostik (Berlin, Germany) in collaboration with the Novoste Corporation (Norcross, GA), are intended for use in the prophylactic treatment of restenosis following balloon angioplasty in heart-disease patients. The procedures were applied to the radionuclidic assay of both the bare-ceramic source materials (of proprietary composition) contained within the seeds and to the stainless-steel (SS) sealed sources. The approach consisted of extracting some arbitrary fraction of the $^{90}$Sr activity from the ceramic-like material and assaying the resulting solution by $4\pi\beta$ liquid scintillation (LS) spectrometry with $^{3}$H-standard efficiency tracing. The fraction of extracted activity was determined by ionization current measurements before and after the chemical extraction. All of the ionization current and LS-based activity determinations were made under the experimentally-verified conditional that $^{90}$Y was in radioactive equilibrium with $^{90}$Sr. For the assay of the SS-jacketed seeds, the encapsulation was initially dissolved and the resultant solution was also assayed by LS spectrometry to determine the amount of activity removed by the SS dissolution step. The developed protocol included provisions for accounting for all possible losses of $^{90}$Sr activity in the chemical and source-handling procedures, for the unrecovered activity in the extracted source material and for any residual activity in the solution-transfer and source-handling tools. These destructive assays were required for relating radiochromic-film measurements of the absorbed dose spatial distributions for the seeds to theoretic dose modelling and for establishing calibration factors for subsequent non-destructive radionuclidic measurements on the seeds. Published by Elsevier Science Ltd.

## 1. Introduction

The use of various high-energy, pure β-emitting radionuclides in intravascular brachytherapy sources for the radiotherapeutic reduction of restenosis at bal-loon-angioplasty sites has been under considerable investigation by the cardiology and radiation oncology research communities (Coursey et al., 1998). The radioactivity content of such sources (termed 'seeds') must be known in order to unequivocally relate measurements and calculations of their absorbed dose spatial distributions. These dose determinations are critical for evaluating the efficacy of the radiation-induced prevention of restenosis and for meaningful interpretation of

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results from various preclinical studies and clinical trials. Uniform and consistently-determined dose profiles are particularly important since the restenosis treatments may use a variety of radionuclides and differing dose delivery systems. The absence of monoenergetic radiative signatures (e.g. γ rays) for these types of β-emitting seeds requires that their radionuclidic content be assayed by destructive radioanalytical methods.

Collé (1999), for example, recently reported on the techniques that were employed for the chemical digestion and radionuclidic assay of TiNi-encapsulated $^{32}$P intravascular brachytherapy sources. This earlier work discussed the importance and use of these sources for the prophylactic treatment of restenosis, as well as the critical need for such destructive radiochemical analyses. The results of the assays were used to link Monte-Carlo-based theoretic modelling of the absorbed dose spatial distributions of the sources to radiochromic-film dosimetric measurements and to establish calibration factors for subsequent assays of the sources by non-destructive methods (Collé et al., 1999).

Strontium-90 (in radioactive equilibrium with $^{90}$Y) is another high-energy, pure β-emitting, candidate nuclide whose use in intravascular brachytherapy sources has been proposed and may be promising. A stainless-steel-encapsulated $^{90}$Sr/$^{90}$Y-laded seed having a 'ceramic' internal source material has been developed and is under investigation by a consortium between Bebig Isotopentechnic und Umweltdiagnostik (Berlin, Germany) and Novoste Corp. (Norcross, GA). This paper reports on the radioanalytical methods, developed at the National Institute of Standards and Technology (NIST), that were used to assay these seeds and to establish ionization-chamber-based calibration factors.

## 2. Experimental considerations

### 2.1. The Bebig/Novoste $^{90}$Sr seeds

The seeds, manufactured by Bebig, consist of $^{90}$Sr–$^{90}$Y-containing source material that is encapsulated in a thin-walled, stainless-steel (SS) jacket. Each has a nominal length of 2.5 mm and a 0.6-mm outer diameter and contains of the order of 100 MBq of $^{90}$Sr. For therapeutic use, trains of upwards of a dozen such SS seeds are inserted into suitable artery catheters.

The internal source material is a highly refractory, ceramic-like matrix and is designated herein as a 'ceramic'. Further information on its composition and production method is proprietary and can not be disclosed. The radiochemical procedure (see Section 2.2)

was devised, however, in consideration of this source composition.

Four sources were submitted to NIST by Bebig for radionuclidic assay of their $^{90}$Sr activity content. Two consisted of bare 'ceramics', arbitrarily labelled 'C1' and 'C2', which were obtained from the manufacturing process prior to being sealed in SS. The other two, designated 'Q10' and 'Q11', were completed SS-jacketed seeds. The assay results on both kinds of sources could be used to establish calibration factors for in-house quality control measurements by Bebig. In addition, the activity assay results coupled to ionization current measurements on the initially submitted sources established calibration factors that could be used by NIST for any subsequent non-destructive assays on either type of source.

### 2.2. The overall radiochemical protocol and the basis of the $^{90}$Sr determinations

The devised radiochemical procedure required two principal considerations.

The first, and more trivial, involved finding a method to dissolve the SS jacket while ensuring the quantitative recovery of any $^{90}$Sr activity that might be removed during this dissolution process.

The second involved finding a way to get the $^{90}$Sr activity out of the 'ceramic' matrix and into solution by some digestion process. The nature of the internal source material precluded finding any single agent (e.g. an acidic solvent or fusion flux) that was likely to completely digest the entire seed. Therefore, an alternative approach was sought.

Based on the known production process and chemical composition of the internal source, it seemed likely that some substantial fraction of the $^{90}$Sr activity might be extracted (or leached) from the ceramic-like material by application of an appropriately chosen acid. Furthermore, it seemed apparent that the fraction of unextracted activity (of the total contained in the ceramic) might be adequately accounted for by making ionization current measurements of the source's bremsstrahlung-emission rates before and after the chemical extraction. The total $^{90}$Sr activity $A$ in the source could then be determined from the simple relation:

$$A = A_s/(1 - \Gamma_a/\Gamma_b) \tag{1}$$

where $A_s$ is the $^{90}$Sr activity extracted and brought into solution (and to be subsequently assayed), $\Gamma_b$ is an initial ionization current measurement of the source before the extraction and $\Gamma_a$ is the source's ionization current measurement after the extraction (refer to Section 2.3).

The Eq. (1) approach, of course, involves several, significant, inherent assumptions as to the conditions
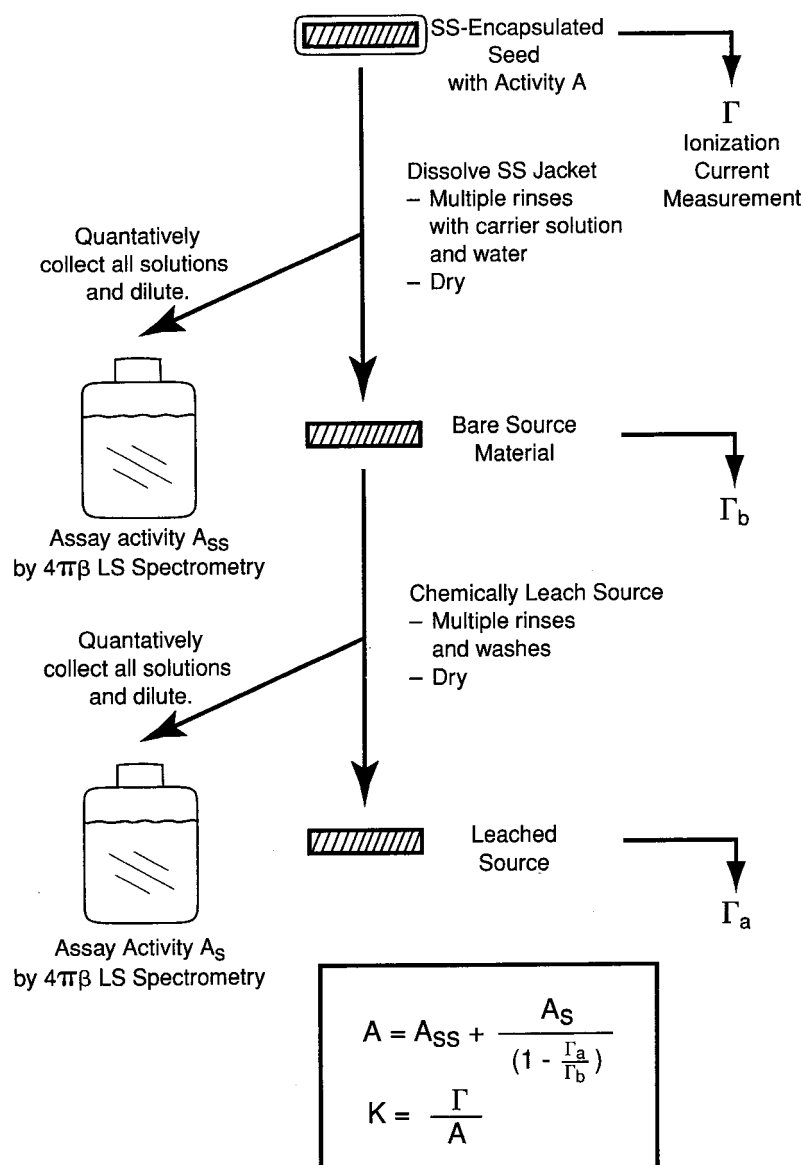
Fig. 1. An overall schematic of the radioanalytical method used to assay the seeds.

under which $\Gamma_b$ and $\Gamma_a$ are determined: viz., (i) that the two determinations are made under as nearly identical conditions as possible; (ii) that there are no appreciable activity losses in the handling of the sources that are not accounted for within $A_s$; (iii) that the source composition and geometry for the two measurements are not significantly different so as to not appreciably alter the bremsstrahlung yield per unit activity; (iv) that the two measurements are obtained under identical states of radioactive equilibrium between ^{90}Y and ^{90}Sr and (v) that the ionization cur-

rents are appropriately decay corrected to the reference time used for $A_s$ and $A$.

To explore the feasibility and efficacy of this avenue of approach, NIST designed some preliminary extraction experiments which were performed by Bebig (Heß, 1998). These experiments demonstrated that extractions with several different mineral acids (of varying concentration) could remove anywhere from a few percent to over 90% of the ^{90}Sr activity. There was no evidence of any significant physical changes in the sources after the extractions. Yet, the gravity of the

approach's assumptions required that there be some experimental verification that the relative bremsstrahlung yield (as obtained from ionization current measurements) was independent of the fraction of extracted activity (see Sections 3.6 and 3.7).

For the assay of the SS-encapsulated seeds, the above extraction procedure on a bare ceramic source could easily be joined to a previous SS removal step. In this case, the activity $A_{ss}$ (as subsequently assayed) that is removed during the SS dissolution is added to the right-hand side of Eq. (1) so that the total $^{90}$Sr activity in the encapsulated seed is determined from:

$$A = A_{ss} + A_s/(1 - \Gamma_a/\Gamma_b) \qquad (2)$$

This overall protocol is outlined in the schematic of Fig. 1.

The above procedure could, of course, be repeated over several such extractions. Source Q10, in fact, was assayed by performing three sequential extractions (after the removal of the SS jacket) which resulted in four master solutions ($A_{ss}$, $A_{s1}$, $A_{s2}$ and $A_{s3}$) and three sets of 'before' and 'after' relative ionization current measurements ($\Gamma_b$ and $\Gamma_{a1}$, $\Gamma_{a1}$ and $\Gamma_{a2}$ and $\Gamma_{a2}$ and $\Gamma_{a3}$). From this, the total $^{90}$Sr activity in the seed could be determined from any one (or all) of the relations:

$$A_1 = A_{ss} + A_{s1}/(1 - \Gamma_{a1}/\Gamma_b)$$

$$A_2 = A_{ss} + A_{s1} + A_{s2}/(1 - \Gamma_{a2}/\Gamma_{a1})$$

$$A_3 = A_{ss} + A_{s1} + A_{s2} + A_{s3}/(1 - \Gamma_{a3}/\Gamma_{a2}) \qquad (3)$$

All of the $A$ and $\Gamma$ variables on the right-hand sides of the above Eqs. (1)–(3) are given at a common reference time and with $^{90}$Y in secular equilibrium with $^{90}$Sr. Initially, the sources would be sufficiently aged so that one could consider that $^{90}$Y is in radioactive equilibrium with its $^{90}$Sr parent. However, if they are not in equilibrium directly after the extraction during the measurements of $\Gamma_a$ (even if it was for the determination of $\Gamma_b$) and for the assay of $A_s$, then the above simple equations become more complicated and require knowledge of the $^{90}$Y/$^{90}$Sr ratios and each nuclide's relative measurement response (refer to Sections 2.3 and 2.4). It seems possible that Y may be extracted differently than the Sr from the sources (maybe more or maybe less depending upon their chemical states within the 'ceramic') and one can not a priori assume that equilibrium has been maintained.

To minimize solution transfer losses (during the SS dissolution and ceramic extraction steps) and source manipulations (for the ionization current measurements), a novelly-designed and specially-fabricated 'handling assembly' was employed for this work. From the onset of the overall assay procedure to its com-
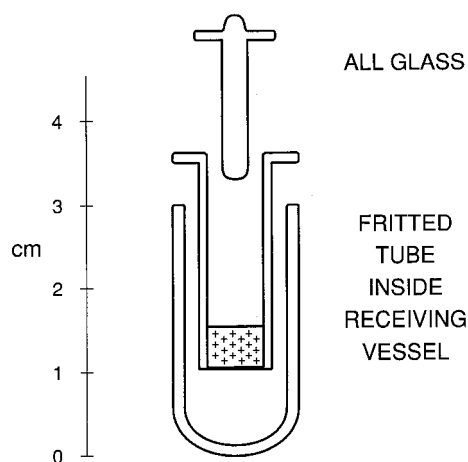


Assembly used to hold seed/source for ionization current measurements, and to perform chemical dissolution/extraction.

Fig. 2. The 'handling assembly' that was used both as a 'chemistry kit' and as a source holder for the ionization current measurements of the seeds.

pletion, each source was contained within its own unique handling assembly. These handling assemblies were used both as source holders for all ionization current measurements and as the apparatus (a kind of small chemistry kit) in which all of the radiochemical procedures were performed. The handling assembly, illustrated in Fig. 2, consisted of four all-glass parts:

1. An outer, round-bottomed, thin-walled tube (nominal 3.5 cm length, 1.2 cm o.d.).
2. An inserted, inner, right-circular-cylindrical tube with a fritted-glass bottom (nominal 2.5 cm length, 0.5 cm o.d.) which held the source and which was used to drain digesting solutions into the outer tube.
3. A rod (nominal 2 cm long) that fit into the fritted tube and acted as a stopper.
4. A second 'clean' tube (identical to (a)) that was used to hold the fritted tube and stopper for the ionization chamber measurements that followed any of the 'wet chemistry' steps.

The assembly's components were sufficiently small so that they could easily be evaluated for any residual activity (on completion of the assay) by immersion in a scintillant within conventional 22-ml liquid scintillation vials.

The specific experimental details that were employed for the assay of each source are given in Section 3.4. It may be useful, however, to provide a generalized synopsis for the overall procedure as it applies to the

assay of a SS seed. The steps are: (i) following all pre-liminary evaluations, such as for surface contamination and photon-emitting impurity analyses (see Sections 3.1 and 3.2), the seed is placed inside its handling assembly for an initial ionization current measurement $\Gamma$ (which will be subsequently used to obtain an ioni-zation chamber calibration factor $K = \Gamma/A$). (ii) A few ml of warmed, concentrated HCl is added to the hand-ling assembly to dissolve the SS over the course of sev-eral hours. (iii) After dissolution, the resultant solution is transferred (by pycnometer) to a large-volume mas-ter solution (called 'ss') that contains an acidic $Sr^{+2}$–$Y^{+3}$ carrier solution. (iv) The bared ceramic source and handling assembly is then repetitively rinsed with large volumes of distilled water and dried for several hours under an infrared heat lamp. All of the rinses are added to the master 'ss' solution. (v) The master 'ss' is diluted with additional carrier solution and assayed by LS spectrometry. (vi) The inner fritted tube and stopper (containing the dried bared seed) is trans-ferred to the 'clean' tube for the ionization chamber determinations of $\Gamma_b$. (vii) Following these measure-ments, the bared seed is extracted by the addition of HCl to the handling assembly (after the fritted tube and stopper is returned to the original outer tube). (viii) After extraction, the resultant solution and all rinses are transferred to a master solution 's'. (ix) The master 's' is also diluted with carrier solution and assayed by LS spectrometry. (x) After drying, the inner assembly is again transferred to the 'clean' tube for ionization current measurements of $\Gamma_a$. (xi) The orig-inal outer tube of the handling assembly (used for the 'wet chemistry' steps) and transfer pycnometers for the 's' and 'ss' masters are assayed by LS spectrometry for residual activity.

### 2.3. Ionization current measurements

All ionization current measurements (to determine $\Gamma$, $\Gamma_b$ and $\Gamma_a$) were performed with the NIST 'chamber A', an Ar-filled, pressurized (2 MPa), reentrant '$4\pi\gamma$' ionization chamber. The chamber, its electronic system and its operation have been described by Calhoun (1987) and Unterweger et al. (1992). For these measurements, the source-containing handling assem-blies were placed within capped, 1-mm wall, lucite vials that in turn just fit inside the standard source holder for 'chamber A'. Relative ionization currents were obtained with respect to three NIST, internal-standard, $^{226}$Ra reference sources: identified as RRS20, RRS50 and RRS100, whose numerical suffixes corre-spond roughly to their $^{226}$Ra content in units of micro-grams.

The ionization current measurements are given in terms of background- and decay-corrected ionization current ratios $\Gamma_{j(i)} = I_i/I_j$ at some $t = 0$ reference time

where the index $i$ identifies the specific source and where $j$ refers to a specific radium reference source. For each $\Gamma_{j(i)}$ determination, paired ionization current measurements of $I_i$ and $I_j$ (as well as a background current) were periodically performed over a sufficient time interval to experimentally verify that the $^{90}$Y com-ponent was in radioactive equilibrium with $^{90}$Sr to within its cited uncertainty. These intervals ranged from a minimum of 4 to over 20 days.

The equilibrium condition was evaluated by examin-ing the constancy of the quantity $\Gamma_t(\exp(\lambda_{Sr}t))$ as a function of time $t$. The time-dependent, background-corrected, relative ionization current $\Gamma_t$ obtained for a given source at some measurement time $t$ may be con-sidered to consist of the sum of two components: the relative current resulting from the $^{90}$Sr activity and that due to $^{90}$Y, such that

$$\Gamma_t = K_{Sr}A_{Sr} + K_Y A_Y \qquad (4)$$

where $K_{Sr}$ and $K_Y$ are proportionality constants (cali-bration factors) that relate the chamber's relative re-sponse to each nuclide and where $A_{Sr}$ and $A_Y$ are the $^{90}$Sr and $^{90}$Y activities in the source at time $t$. With $A_{Sr} = A_{Sr(0)}\exp(-\lambda_{Sr}t)$ for $^{90}$Sr decay constant $\lambda_{Sr}$ and with $Q_t = A_Y/A_{Sr}$, Eq. (4) may be immediately rewritten as

$$\Gamma_0 = \Gamma_t[\exp(\lambda_{Sr}t)] = A_{Sr(0)}[K_{Sr} + Q_t K_Y] \qquad (5)$$

where $A_{Sr(0)}$ is the $^{90}$Sr activity at a reference time $t = 0$ and where $\Gamma_0$ is the decay-corrected (to $t = 0$ only for $^{90}$Sr decay) relative ionization current.

For sources in which $^{90}$Y is in equilibrium with $^{90}$Sr at the measurement time $t$, the activity ratio $Q_t$ assumes a constant value $Q = \lambda_Y/(\lambda_Y - \lambda_{Sr})$ given by the respective $^{90}$Sr and $^{90}$Y decay constants. In this case, the quantity $(K_{Sr} + Q_t K_Y)$ in Eq. (5) becomes a true, time-independent, proportionality constant

$$K = K_{Sr} + Q K_Y = K_{Sr} + [\lambda_Y/(\lambda_Y - \lambda_{Sr})]K_Y \qquad (6)$$

that relates $\Gamma_t$, or the decay corrected $\Gamma_0 = \Gamma_t(\exp(\lambda_{Sr}t))$, to the $^{90}$Sr activity in the source

$$K = [\Gamma_t/A_{Sr}] = [\Gamma_0/A_{Sr(0)}] \qquad (7)$$

The constant $K$ may be viewed as a calibration fac-tor for determining the $^{90}$Sr activity in a source from measurements of the source's relative ionization cur-rent (for the given chamber and defined source geome-try) under the conditional that the $^{90}$Y child is in radioactive equilibrium with its $^{90}$Sr parent.

It is easy to demonstrate that Eqs. (6) and (7) are not valid under $^{90}$Sr-$^{90}$Y nonequilibrium conditions. In this case, in contradistinction, the activity ratio $Q_t$ in Eq. (5) is not a time-independent constant, but instead is a function of $t$ and depends on some initial

$Q_0 = A_{Y(0)}/A_{Sr(0)}$ activity ratio, such that

$$Q_t = Q[1 - F(t)] + Q_0 F(t) \tag{8}$$

where $F(t) = \exp(-(\lambda_Y - \lambda_{Sr})t)$. From the above, it is clear that the absence of $^{90}\text{Sr}-^{90}\text{Y}$ equilibrium during any ionization chamber measurements of the seeds would require knowledge of the individual $K_{Sr}$ and $K_Y$ calibration factors as well as some determination of the state of disequilibrium ($Q_t \neq Q$).

Ancillary experiments were also performed to evaluate the variability in the ionization current response due to source positioning within a given handling assembly and between various sets of handling assemblies. The individual components of variance $v_p^2$ for the positioning variability and $v_s^2$ for the source-holder variability (in terms of relative standard deviations $v$) were evaluated using a nested-uncertainty-treatment model that has been described by Collé and Kishore (1997). The approach is based on determining the precision estimators $v_0$ for $n_m$ measurements of the ionization current $I_i$ for a given source $i$ in the same position and in the same source holder, $v_1$ for $n_p$ positionings of the same source in the same holder and $v_2$ for the same source in $n_s$ different source holders. These estimators are linked through the relations: $v_2^2 = v_1^2 + n_s v_s^2$, $v_1^2 = v_2^2 + n_p v_p^2$ and $v_0^2 = v_i^2 + n_m v_m^2$ where $v_i^2$ can be considered to be some inherent statistical (Poisson) variance and where $v_m$ is the relative standard deviation for an additional component of variance for the measurement process. Similarly, $v_1$ can be considered to be comprised of $v_0$ and additional component of variance $v_p$ for the positioning differences and $v_2$ is composed of $v_1$ and a component of variance $v_s$ for the dispersion amongst different holders. The components $v_0$, $v_1$ and $v_2$ were directly obtained from measurement data of replications; whereas $v_p$ was derived from $v_0$ and $v_1$ and $v_s$ is derived from $v_1$ and $v_2$. Collé and Kishore (1997) may be consulted to find an illustrated, representative example of such nested uncertainty analyses.

For these measurements, it was found, in general, that the positioning variability $v_p$ largely dominated the uncertainty in any ionization current measurement. Typically $v_0/\sqrt{n_m}$ was nearly negligible with respect to $v_p$ and $v_s < v_p$ such that $v_1/\sqrt{n_p} \cong v_p$ and that $v_2 \cong v_1$. This finding is largely attributed to the fact that the small 2 mm seeds have considerable freedom of lateral movement within the fritted tube of the source holder (i.e. the handling assembly in Fig. 2). In addition, the overall uncertainties in the decay-corrected ratios $\Gamma_{j(i)}$ were largely dictated by the imprecision evidenced by repeated measurements (in different positions) over the sufficiently long time intervals that were needed to verify the $^{90}\text{Y}-^{90}\text{Sr}$ equilibrium conditional.

## 2.4. The LS efficiency-tracing methodology for the assay of the master solutions

The resultant large-volume master solutions 's' and 'ss' and various residual-activity vials were assayed for $^{90}\text{Sr}-^{90}\text{Y}$ by an efficiency-tracing $4\pi\beta$ LS spectrometric method that is somewhat routine for this laboratory. It is based on well-established procedures for the assay of gravimetrically-determined aliquants of solutions containing β-emitting radionuclides (Collé, 1997a,b,c; 1999a,b; Collé and Zimmerman, 1996a,b, 1997; Zimmerman and Collé, 1997a,b,c).

For a given LS cocktail formed with an aliquant of mass $m$ of a $^{90}\text{Sr}-^{90}\text{Y}$ solution in some suitable scintillant, the total number of counts $C_i$ detected in a counting interval $T$ can be given by

$$C_i/mT = Æ_{Sr(0)}\epsilon_{Sr}\exp(-\lambda_{Sr}t_i) + Æ_{Y(0)}$$
$$\epsilon_Y \exp(-\lambda_Y t_i) + Æ_{Sr(0)}[\lambda_Y/(\lambda_Y - \lambda_{Sr})]$$
$$\times [\exp(-\lambda_{Sr}t_i) - \exp(-\lambda_Y t_i)]\epsilon_Y + B_i/T \tag{9}$$

where $Æ_{Sr(0)}$ and $Æ_{Y(0)}$ are the *massic* activities of $^{90}\text{Sr}$ and $^{90}\text{Y}$ in solution (activity per unit mass of solution) at some $t_i = 0$ reference time, respectively; where $\epsilon_{Sr}$ and $\epsilon_Y$ are the respective $4\pi\beta$ LS detection efficiencies (at time $t_i$ if they are temporally dependent); where $\lambda_{Sr}$ and $\lambda_Y$ are the $^{90}\text{Sr}$ and $^{90}\text{Y}$ decay constants, respectively; and where $B_i/T$ is the estimation of the background counting rate for a composition-matched blank cocktail at time $t_i$. In the assay of the residual-activity vials, the aliquant mass $m$ is irrelevant and $Æ_{Sr(0)}$ and $Æ_{Y(0)}$ in eq. (9) then represent the *total* activities in the vials. The counting intervals $T$ are always $1/\lambda_{Sr} \gg T \ll 1/\lambda_Y$. The $\epsilon_{Sr}$ and $\epsilon_Y$ efficiencies are evaluated by efficiency tracing with composition-matched $^3\text{H}$-standard cocktails using the CIEMAT/NIST methodology (outlined below). The first two terms of Eq. (9) correspond to those parts of the $^{90}\text{Sr}$ and $^{90}\text{Y}$ activities that were initially present at time $t = 0$ that are still present at time $t$, while the third term is the ingrowth of the $^{90}\text{Y}$ as a child of $^{90}\text{Sr}$. The background-corrected massic counting rate $R_i = (C_i - B_i)/mT$ at time $t_i$ at the midpoint of the counting interval $T$ is, therefore, just

$$R_i = Æ_{(0)}\{\epsilon_{Sr}\exp(-\lambda_{Sr}t_i) + Æ\epsilon_Y[\lambda_Y/(\lambda_Y - \lambda_{Sr})$$
$$\times [\exp(-\lambda_{Sr}t_i) - \exp(-\lambda_Y t_i)]\}$$
$$+ Æ_{Y(0)}\epsilon_Y\exp(-\lambda_Y t_i) \tag{10}$$

which is easily transformed into a linear form

$$Y = Æ_{Sr(0)} + XÆ_{Y(0)} \tag{11}$$

with

$$Y = R_i/\{\epsilon_{Sr} \exp(-\lambda_{Sr}t_i) + \epsilon_Y[\lambda_Y/(\lambda_Y - \lambda_{Sr})]$$

$$\times [\exp(-\lambda_{Sr}t_i) - \exp(-\lambda_Y t_i)]\} \quad (12)$$

and

$$X = [\epsilon_Y \, exp(-\lambda_Y t_i)]/\{\epsilon_{Sr} \exp(-\lambda_{Sr}t_i) + \epsilon_Y[\lambda_Y/(\lambda_Y$$

$$- \lambda_{Sr})][\exp(-\lambda_{Sr}t_i) - \exp(-\lambda_Y t_i)]\}. \quad (13)$$

With sufficient $(R_i, t_i)$ counting data pairs, $\text{Æ}_{Sr(0)}$ and $\text{Æ}_{Y(0)}$ can be obtained as constant parameters by fitting the $(R_i, t_i)$ data to the functional form of Eq. (10). The linearized Eq. (11), in which the intercept of $Y$ at $X = 0$ is equal to the $^{90}$Sr massic activity $\text{Æ}_{Sr(0)}$ and the slope $dY/dX$ is equal to the $^{90}$Y massic activity $\text{Æ}_{Y(0)}$, was used as a powerful graphical tool for examining the quality of the input $(R_i, t_i)$ measurement data, for determining the degree of radioactive equilibrium of $^{90}$Y with the $^{90}$Sr parent and for evaluating the possible presence of radionuclidic impurities (Collé, 1997a,1999c).

In the somewhat usual case where $^{90}$Y and $^{90}$Sr are in secular equilibrium at both the measurement time $t$ and at the reference time $t = 0$, $\text{Æ}_{Y(0)}$ assumes the value $\text{Æ}_{Sr(0)}(\lambda_Y/(\lambda_Y - \lambda_{Sr}))$ and Eq. (10) becomes independent of $_{Y(0)}$:

$$R_{eq(i)} = \text{Æ}_{Sr(0)}[\exp(-\lambda_{Sr}t_i)]\epsilon_{Sr+Y} \quad (14)$$

with $\epsilon_{Sr+Y} = \epsilon_{Sr} + \epsilon_Y(\lambda_Y/(\lambda_Y - \lambda_{Sr}))$, in which the linearizing transform

$$Y_{eq} = R_{eq(i)}/\{[\exp(-\lambda_{Sr}t_i)]\epsilon_{Sr+Y}\} \quad (15)$$

yields a constant $Y_{eq} = \text{Æ}_{Sr(0)}$. This was the approach used to verify the existence of $^{90}$Sr–$^{90}$Y equilibrium in the LS data. Simple graphical use of these transforms could readily identify any Eq. (11) non-linearities due to short-lived impurities or Eq. (15) inconstancies due to an impurity or a $^{90}$Sr–$^{90}$Y disequilibrium state.

The acronym CIEMAT/NIST refers to the two laboratories that collaborated in developing the employed LS spectrometry tracing methodology; viz. the Centro de Investigaciones Energeticas, Medioambientales y Technologicas (CIEMAT) and NIST. The method's principal features have been described in numerous publications by the CIEMAT/NIST originators (Grau Malonda and Garcia-Toraño, 1982; Coursey et al., 1985; 1986; 1989; 1991; 1994). Some of its more important aspects as employed for this work are given in the references that follow. Specific details on the method's practical application, as it is presently invoked in this laboratory, are given by Collé and Zimmerman (1996a,b, 1997)) and Zimmerman and Collé (1997a,b,c)). A good summary review and overview of

the calculational protocol is also available (Collé and Zimmerman, 1996, 1997). This protocol employs various updated and revised versions of the CIEMAT-developed EFFY code (Garcia-Toraño and Grau Malonda, 1985; Grau Malonda et al., 1985; Garcia-Toraño, 1993) to determine the detection efficiencies for cocktails of the traced radionuclide (under known and varying quench conditions) by following the experimentally-determined efficiencies for closely-matched cocktails of $^3$H. Tritium ($^3$H) is recommended to serve as the matched (in terms of cocktail composition and quenching) standard since extrapolations to the lower-energy portions of the β spectra are more sensitive than that obtained with higher-energy β-emitting standards like $^{14}$C (Coursey et al., 1986, 1989). The importance of cocktail matching between the standard cocktails and those of the traced radionuclide has been treated by Collé (1997c). Similarly, the need to understand the cocktail-composition systematics, such as the effects of the aqueous mass fraction (Collé and Zimmerman, 1996) or ionic loading (as it affects cocktail stability and what has been termed cocktail 'tractability'), is critical (Collé, 1999b). Collé (1997c), Collé and Zimmerman (1997) and Zimmerman and Collé (1997) have also identified and addressed some of the critical underlying assumptions regarding the method's applicability and have developed and presented a detailed measurement uncertainty model for the CIEMAT/NIST method (Collé and Zimmerman, 1996).

## 2.5. On the LS cocktail preparations, LS spectrometers and measurements

The routine gravimetric, source-preparation procedures used in our laboratory as they apply to the preparation of LS counting sources (such as those prepared for this work), as well as estimations of their associated uncertainties, have been treated at length previously (Collé, 1995; Collé et al., 1995; Collé and Kishore, 1997). Mass determinations for the aliquants have relative standard uncertainties of about $\pm 0.05$ %. The LS counting sources were prepared in all cases with a di-isopropylnapthalene (DIPN)-based, 'ready-to-use', commercially-prepared scintillant (namely, 'Ultima Gold AB' (Packard Instrument Co., Meriden, CT)) and were contained in conventional, 22-ml, low-potassium glass vials with screw caps having polyethylene vee-cone liners. The total cocktail mass in every counting source was kept constant at 10.5 g to within about 0.2 g (refer to Zimmerman and Collé (1997) for cocktail volume (mass) effects) and each had an aqueous mass fraction of at least $f_w \geq 0.05$ to insure cocktail stability (Collé and Zimmerman, 1996).

The $^3$H-standard cocktails used for the $^{90}$Sr and $^{90}$Y efficiency tracing were prepared with aliquants of care-
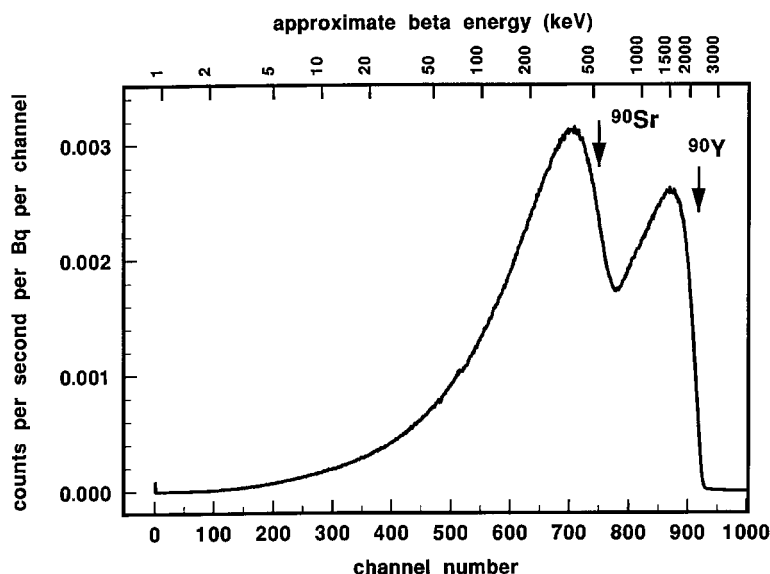
Fig. 3. Experimental LS spectrum for an equilibrium mix of $^{90}$Y and $^{90}$Sr. The spectrum was obtained with the NIST 'system B' spectrometer which has logarithmic pulse amplification so that the difference between any two channel numbers $(c_2 - c_1)$ is proportional to the logarithmic energy ratio $\log(E_2/E_1)$.

fully-prepared gravimetric dilutions of a NIST $^3$H (tritiated-water) standard (NIST, 1991). Efficiency variations for the tracing were achieved by either using varying aliquant sizes (and hence variable aqueous mass fraction and quenching) or by the use of a 10 % solution of $CH_3NO_2$ in ethanol as an imposed quenching agent. The $^3$H cocktails were in all cases composition matched to the $^{90}$Sr–$^{90}$Y cocktails by the addition of proportionate amounts of the blank carrier solutions that were used to prepare the $^{90}$Sr–$^{90}$Y master solutions.

Measurements on the prepared cocktails were mainly performed with one of our two LS spectrometers, namely the NIST 'system B'. A few measurements were also performed with our 'system P'. The principal operating characteristics of the two spectrometers have been outlined previously (Collé and Kishore, 1997; Collé and Zimmerman, 1997). Descriptions of these operating characteristics, their respective performance and use within this laboratory for various radionuclidic calibrations are available (Collé, 1993, 1995, 1997b, 1999; Collé and Thomas, 1993; Collé et al., 1995; Collé and Kishore, 1997; Collé and Zimmerman, 1996, 1997). Use of the two spectrometers on the identical set of cocktails provides the benefit of being able to verify that the efficiency tracing (and hence the accuracy of the assay) is indeed instrument independent. The two instruments have some similar and some substantially different operating characteristics and have decidedly different detection thresholds (refer particu-

Table 1
Relevant nuclear data used for the $^{90}$Sr–$^{90}$Y assays and as obtained from ENSDF (1997)

| Nuclide | Half-life, $\ln(2)/\lambda$ | $E_{(\max)}^{\beta}$ [a] | $E_{(\mathrm{mean})}^{\beta}$ [b] | $I^{\beta}$ | β transition |
|---|---|---|---|---|---|
| $^{90}$Sr | $(28.78 \pm 0.04)$ a | $(546.0 \pm 1.6)$ keV | $(195.8 \pm 0.8)$ keV | 1.0000 | 1st forbidden unique $J^{\pi} = 0^+ \rightarrow 2^-$ |
| $^{90}$Y | $(64.10 \pm 0.08)$ h | $(2281.5 \pm 2.5)$ keV | $(933.7 \pm 1.2)$ kev | $0.999885 \pm 0.000014$ | 1st forbidden unique $J^{\pi} = 2^- \rightarrow 0^+$ |
| | | 521.3 keV | $(185.6 \pm 1.0)$ keV | $0.000115 \pm 0.000014$ | 1st forbidden unique $J^{\pi} = 2^- \rightarrow 0^+$ |
| $^3$H | $(12.32 \pm 0.06)$ a | $(18.619 \pm 0.011)$ keV | $(5.69 \pm 0.04)$ keV | 1 | Allowed $J^{\pi} = (1/2)^+ \rightarrow (1/2)^+$ |

[a] The effect of the $E_{\beta(\max)}$ uncertainties on the EFFY4-code efficiency calculations, in terms of relative standard uncertainties, are 0.006, 0.0002 and 0.044% for $^{90}$Sr, $^{90}$Y and $^3$H, respectively.

[b] The values of $E_{\beta(\mathrm{mean})}$ are not required as input into the EFFY4 code, but rather are calculated by EFFY4 from input values of $E_{\beta(\max)}$. The $E_{\beta(\mathrm{mean})}$ values tabulated here are those obtained from ENSDF (1997). For comparison, those obtained from the EFFY4 code (using the tabulated $E_{\beta(\max)}$ values) are $E_{\beta(\mathrm{mean})} = 195.7$ keV for $^{90}$Sr, $E_{\beta(\mathrm{mean})} = 934.0$ keV for $^{90}$Y and $E_{\beta(\mathrm{mean})} = 5.70$ keV for $^3$H.
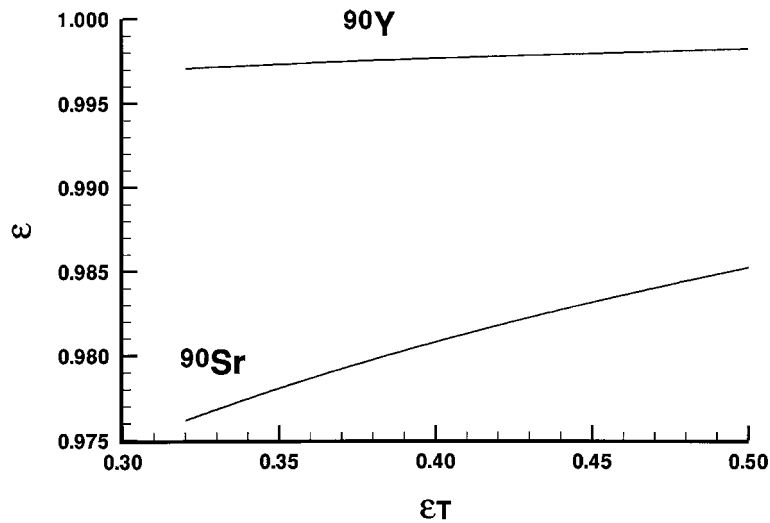
Fig. 4. EFFY4-code calculations for the $^{90}$Sr and $^{90}$Y LS detection efficiencies ($\epsilon_{Sr}$ and $\epsilon_Y$) as a function of that for $^3$H over the range of typical experimental efficiencies $0.3 \leq \epsilon_T \leq 0.5$.

larly to Collé et al. (1994)). A typical $^{90}$Sr–$^{90}$Y β spectrum obtained with the system B spectrometer is shown in Fig. 3.

The quench indicating parameter (QIP) used to account for small quenching differences between the matched $^3$H-standard and $^{90}$Sr–$^{90}$Y cocktails was an instrument-derived Horrocks number $H$ for the 'system B' spectrometer and a 'transformed spectral index of the external standard' (*tSIE*) for 'system P'. The calculated QIP-adjusted $^{90}$Sr and $^{90}$Y efficiencies, $\epsilon_{Sr}$ and $\epsilon_Y$, as obtained from tracing with the experimentally-

determined $^3$H efficiencies, were obtained with the EFFY4 code (Garcia-Toraño, 1993). Table 1 includes the nuclear data (ENSDF, 1997) for the $E_{\beta(max)}$ values that were used as input for the EFFY4-code calculations, as well as the half-lifes that were used for decay corrections. Fig. 4 shows the slight dependence of the $\epsilon_{Sr}$ and $\epsilon_Y$ efficiencies as a function of the $^3$H efficiency $\epsilon_T$. For cocktails with $^{90}$Y in equilibrium with $^{90}$Sr, the combined efficiency, given as in Eq. (14) by
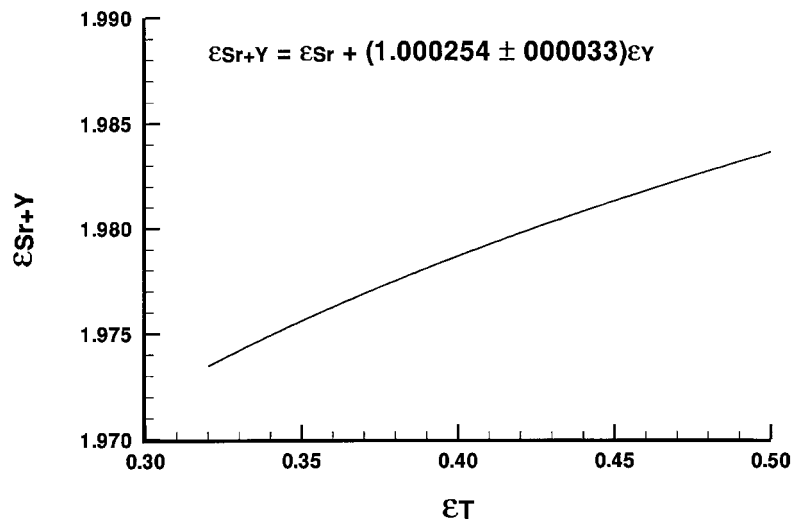


Fig. 5. The overall $\epsilon_{Sr+Y}$ LS detection efficiency as a function of the $^3$H efficiency $\epsilon_T$ (from EFFY4-code calculations) for cocktails having $^{90}$Y in radioactive equilibrium with $^{90}$Sr.

$$\epsilon_{Sr+Y} = \epsilon_{Sr} + \epsilon_Y[\lambda_Y/(\lambda_Y - \lambda_{Sr})]$$

$$= \epsilon_{Sr} + (1.000254 \pm 0.000033)\epsilon_Y,$$

as a function of $\epsilon_T$ is shown in Fig. 5.

Each $Æ_{Sr}$ master-solution assay (Eq. (10)) typically used more than one set of prepared cocktails that were measured over periods of weeks to evaluate the $^{90}Sr$–$^{90}Y$ state of equilibrium. Some of the experimental details (e.g. for the number of cocktails used, the cocktail 'ages' and the time intervals over which the measurements were made) for each assay are summarized in Section 3.4. Other details (e.g. for the solution aliquant sizes $m$ (Eq. (9)), the cocktail masses, aqueous mass fractions $f_w$, the imposed quenching agent to vary efficiencies, the efficiency ranges for $^3H$, the counting time intervals $T$, the number of measurements on each cocktail) were either given above or are very similar to typical experimental conditions used by this laboratory for standardizations by $4\pi\beta$ LS spectrometry. A complete specification and tabular summary of these conditions for somewhat similar kinds of assays, which are very representative of those used for the present work, has been presented in Collé (1997) and Collé and Zimmerman (1997).

The total $^{90}Sr$ activity $A_{ss}$ or $A_s$ for each 'ss' or 's' master solution was derived from the massic activity determination of $Æ_{Sr(ss)}$ or $Æ_{Sr(s)}$ along with a careful, gravimetrically-determined total solution mass $M_{ss}$ or $M_s$ (for the final, diluted solution and after application of any subsequent dilution factor); i.e., $A_{ss} = Æ_{Sr(ss)}M_{ss}$ or $A_s = Æ_{Sr(s)}M_s$, where $A_{ss}$ or $A_s$ was also corrected for any residual activity in the solution-transfer and source-handling tools.

### 2.6. Methods used to evaluate residual activities

The residual activity corrections to either $A_{ss}$ or $A_s$ were used to account for possible activity losses during the various steps of the radiochemical procedure and were obtained from LS measurements on suitably-prepared counting sources that contained all of the solution- and source-handling tools that were used for each individual assay. These typically included: (i) the aspirating pycnometer that was used to transfer solution (and washes) from the handling assembly to the master solution bottle; (ii) the pycnometers that were used to replenish fresh solution into the handling-assembly bath between the procedure's steps, such as between sequential extractions or washes; (iii) various LS vials that had been used as intermediate vessels that held the various bath-replenishment solutions, e.g. those for the dissolution, extraction, washings, or carrier; and (iv) the outer tube of the handling assembly. These represented essentially every object in direct contact with the radioactive solutions, excepting the inner

fritted tube whose residual activity would be partially accounted for by the $\Gamma_a$ ionization current measurement (refer to Eq. (1)). Subsequent evaluations of these tubes (after the extracted seeds were removed at the conclusion of the assay) indicated that the magnitudes of their residual activity content were negligible with respect to either $\Gamma_a$ or $A_s$. For example, in the assay of seed Q10, the residual activity in the empty tube at the end of the procedure was found to have less than a 0.05% effect on any of the $\Gamma_a$ measurements and was less than 0.007% of the $A_s$ activity.

The LS counting-source preparation methods for the residual activity determinations on the tools was similar to that previously used by Collé (1999). Assays of these residual activity vials were based on relative LS counting rate measurements (with comparisons made to cocktails prepared with the master solution) with corrections for efficiency differences. The LS detection efficiencies for these vials were estimated from either $^3H$ efficiencies obtained with comparably-prepared $^3H$-spiked blanks or by standard-addition methods. In the latter case, the standard additions were performed by firstly measuring the decay- and background-corrected LS counting rate $R_x$ for the vial (which contains a $^{90}Sr$ residual activity $A_x$), by then adding a small aliquant of mass $m_s$ of the master solution (having massic activity $Æ_s$) and finally remeasuring the corrected rate $R_{x+s}$. Under the assumption that the $^{90}Sr$ detection efficiencies for the two measurements are equal, the residual activity could be obtained from the relation:

$$A_x = [R_x/(R_{x+s} - R_x)]/m_s Æ_s \tag{16}$$

and the ratio of $A_x$ to the total $^{90}Sr$ activity $A_s$ of the master solution (having total mass $M_s$) is just

$$A_x/A_s = (m_s/M_s)/[(R_{x+s}/R_x) - 1]. \tag{17}$$

### 3. Results and discussion

Unless specifically noted otherwise, all uncertainties and uncertainty intervals cited within this paper correspond to derived 'standard uncertainties' or propagated 'combined standard uncertainties'. 'Expanded uncertainties' with 'coverage factors' of $k = 2$ are only used to report measurement values that have been certified by NIST. Refer to Taylor and Kuyatt (1994) for internationally-accepted definitions of these uncertainty terms.

### 3.1. Surface-contamination and source-leakage evaluations

The SS-encapsulated sources Q10 and Q11 were examined for initial surface contamination. This was

Table 2
Initial ionisation current measurements on the four submitted sources at a reference time of 1200 EST 1 April 1998 and in terms of the ratio $\Gamma_{ji} = I_i/I_j$ between the ionization current $I_i$ for each source compared to that for Radium Reference Sources RRS20, RRS50 and RRS100 (i.e. with $j = 20$, 50 and 100). The relative ionization current measurements were performed with the NIST re-entrant ionization chamber 'A' with the source contained within the 'handling assembly' which was in turn placed inside the chamber's source holder (see Section 2.3).The cited uncertainty interval on each $\Gamma_{j(i)}$ value corresponds to a standard uncertainty (see text)

| Source ($i=$) | $\Gamma_{20(i)}$ | $\Gamma_{50(i)}$ | $\Gamma_{100(i)}$ |
|---|---|---|---|
| Sealed seeds | | | |
| Q10 | $1.7042 \pm 0.0021$ | $0.7073 \pm 0.0013$ | $0.4158 \pm 0.0005$ |
| Q11 | $1.7636 \pm 0.0045$ | $0.7355 \pm 0.0011$ | $0.4292 \pm 0.0011$ |
| Bare ceramics | | | |
| C1 | $1.6755 \pm 0.0057$ | $0.6940 \pm 0.0023$ | $0.4078 \pm 0.0014$ |
| C2 | $1.6602 \pm 0.0037$ | $0.6884 \pm 0.0019$ | $0.4033 \pm 0.0020$ |

done by washing them with gentle agitation in a dilute $EDTA^{-4}$ (ethylenediaminetetraacetate) solution and assaying the resultant wash solution by LS spectrometry. The surface contaminations were found to be $\leq 5 \times 10^{-7}\%$ for Q10 and $(7 \pm 3) \times 10^{-6}\%$ for Q11 in terms of the relative $^{90}$Sr activity in each source (on an equivalent activity basis). The sources were subsequently rinsed with large volumes of distilled water and dried.

The two small glass vessels containing the ceramic sources and the SS seeds (as used for shipping the sources) were examined in a similar fashion and were found to have relative $^{90}$Sr residual activities of $(0.24 \pm 0.03)\%$ for the ceramic-source vessel and $(5 \pm 2) \times 10^{-6}\%$ for the SS-seed vessel (where each cited value was normalized to the total $^{90}$Sr activity in both sources). The substantially greater residual activity in the former vessel can probably be attributed to activity losses from mechanical action on the bared ceramic material during shipping.

From these findings, one could conclude that the sources had sufficient integrity and that potential source-leakage losses during their initial handling did not have to be of substantive concern.

### 3.2. Radionuclidic impurity analyses

Prior to the initiation of the assays, impurity analyses for possible photon-emitting impurities were performed on source Q10 by high-resolution, high-purity germanium (HPGe) γ-ray spectrometry (Schima, 1997). None were detected. Energy-dependent upper limits for any unobseved photons of energy $E_\gamma$ were:

$< 8 \times 10^{-6}$ γ s$^{-1}$ per Bq of $^{90}$Sr for 45 keV $< E_\gamma <$ 507 keV,
$< 10^{-4}$ γ s$^{-1}$ per Bq of $^{90}$Sr for 515 keV $< E_\gamma <$ 1455 keV and
$< 8 \times 10^{-4}$ γ s$^{-1}$ per Bq of $^{90}$Sr for 1465 keV $< E_\gamma <$ 3300 keV

at the 1200 EST 1 April 1998 reference time.

From the time-dependent LS data (typically accumulated over many weeks), there was no evidence to suspect the presence of any other pure β-emitting impurity. Upper limits for any such impurity could be established from analyses of the $^{90}$Sr decay-corrected data. The upper limit for an impurity activity $A_x$ with half-life $T_x$ (in units of days) compared to the master solution $^{90}$Sr activity $A_s$ from ceramic source C1 was $A_x/A_s < 0.004$ (as of 11 February 1998) provided that the hypothetical, unobserved, pure β-emitting impurity had a LS detection efficiency of 0.7 and that it had a half-life $T_x \leq 1000$ d.

### 3.3. Initial ionization current measurements

Initial ionization current measurements were performed on all four sources (in their respective handling assemblies) for the purpose of being able to establish (after the destructive activity determinations) nondestructive assay calibration factors for both the ceramic source and SS-encapsulated seed configurations (see Section 3.7).

The results of these determinations, in terms of background- and decay-corrected ionization current ratios $\Gamma_{j(i)} = I_i/I_j$ at a reference time of 1200 EST 1 April 1998 are given in Table 2 where the index $i$ identifies the specific source and where $j = 20$, 50 and 100 refers to radium reference sources RRS20, RRS50 and RRS100, respectively (refer to Section 2.3). The cited uncertainty interval on each $\Gamma_{j(i)}$ value in Table 2 corresponds to a standard uncertainty with $\nu_{eff} > 80$ effective degrees of freedom. These uncertainties include components due to: replicate measurement precision for the radium reference source (which includes both source-positioning and temporal dependencies); replicate measurement precision for the same source in the same handling assembly (in the same position over a relatively short measurement time interval); the pre-

cision due to replicate positioning within the same handling assembly (at other measurement times); the variability due to the same source within different handling assemblies; background corrections; decay corrections; and estimated limits for the effects due to possible radioactive disequilibrium between $^{90}$Y and $^{90}$Sr.

### 3.4. Employed experimental procedures

#### 3.4.1. For bare ceramic source C1

For the assay of source C1, the procedures were not yet very routinized and involved many steps that were subsequently compressed for the assays of other sources. To extract source C1 (following the $\Gamma_b$ determination), a few ml of 4.2 mol l$^{-1}$ HCl were added to the handling assembly containing the source (as used for the initial ionization current measurements) and mildly heated to a temperature in the range of 40–50°C for a period of about 44 h. Crude bremsstrahlung-emission measurements with a NaI(Tl) ratemeter on the resulting solution revealed that less than 10% of the activity had been extracted. The solution was transferred to a master solution 'ss' that consisted of 1.0 mol l$^{-1}$ HCl with 50 μg Sr$^{+2}$ and 50 μg Y$^{+3}$ per gram of solution. The source and handling assembly were then repetitively rinsed with large volumes of distilled water which were also added to master solution 's'. The extraction was continued with 8.1 mol l$^{-1}$ HCl at about 60°C over the next 46 h. Following solution transfers and rinses and drying of the source (at about 120°C for 4 h), ionization current measurements indicated that a fraction of $0.73 \pm 0.01$ of the source's activity had been extracted. Subsequent ionization current measurements over the next five days exhibited evidence of disequilibrium of $^{90}$Y with $^{90}$Sr. The temporal dependence was consistent with a slight excess of $^{90}$Y. Two further extractions were sequentially performed. Each was made with 8.1 mol l$^{-1}$ HCl at 60°C for a period of about one day. The extracting solutions (and washes) from both were transferred and also added to master solution 's'. Solution 's' was then diluted to a total mass of $M_s = 642.81 \pm 0.32$ g. The source (still within the handling assembly) was again dried over a period of about 5 h at 120°C. Periodic relative ionization current measurements to obtain $\Gamma_a$ were performed over the following 37 d. The initial ionization current data again exhibited a slight excess of $^{90}$Y. The effect (a rapidly decreasing $\Gamma_a$ over the first 8 d) was roughly one third of that observed after the previous extraction. The fraction of extracted activity (at $^{90}$Y–$^{90}$Sr radioactive equilibrium) was found to be $(1 - \Gamma_a/\Gamma_b) = 0.9889 \pm 0.0004$. Two sets of LS cocktails were prepared from dilutions of the master solution. The two dilutions had dilution factors of $21.577 \pm 0.011$ and $10.8964 \pm 0.0054$. The first set of seven cocktails was prepared with varying aliquants (to vary the quenching for the efficiency tracing) ranging from 0.012 to 0.14 g of the diluted solution. The second set of five cocktails, prepared 22 d after the first, used nearly constant aliquants of 0.04 g. Quench variations for the second set were achieved by addition of varying quantities of a 10 % solution of nitromethane in ethanol. The two sets of cocktails were measured with NIST 'system B' spectrometer on three occasions over time periods of 61 and 39 d, respectively. The resultant, efficiency-traced, activity $A_s$ was corrected by a factor of $(0.097 \pm 0.020)\%$ for the residual activity on the solution transfer tools.

#### 3.4.2. For bare ceramic source C2

Source C2 was extracted with 11.5 mol l$^{-1}$ HCl at a temperature in the range $68 \pm 8$°C over the course of 3 d. Over this period, five sequential solution transfers (and rinses) were performed. After each (except the last), the handling assembly was replenished with fresh 11.5 mol l$^{-1}$ HCl for the next extraction. These transfers were separated in time by a minimum of 6 h. All of the transferred solutions and rinses were added to the same master solution 's' (which used the same carrier solution described above in part (*i*)). The source was dried at a temperature of about 110°C for 14 h. Ionization current measurements to determine $\Gamma_a$ were performed over the course of 21 d. There was no evidence of disequilibrium between $^{90}$Y and $^{90}$Sr. The master solution was diluted to a total mass of $(606.03 \pm 0.30)$ g. Two sets of counting sources were prepared for the LS assay of $A_s$: one of three cocktails from the master solution 's' and another of four cocktails from a dilution of 's' (with a dilution factor of $5.7737 \pm 0.0029$). Quench variations were obtained by addition of varying aliquants of the blank carrier solution. Both sets of cocktails were measured with the 'system B' spectrometer on two occasions over a period of 13 d. The correction to the resultant $A_s$ for the residual activity on the transfer tools was $(0.012 \pm 0.002)\%$.

#### 3.4.3. For SS-encapsulated seed Q10

For the digestion and extraction of seed Q10, a few ml of 11.5 mol l$^{-1}$ HCl were added to the assembly containing the seed (as used for the ionization chamber measurements) to dissolve the SS jacket. The bath was heated to a temperature of approximately 70°C. After approximately 5 h the jacket was not wholly removed and the HCl-seed mix was allowed to sit (without heating) another 13 h (overnight). By that time, there was no evidence of any remaining stainless steel. Bremsstrahlung-emission measurements with a NaI(Tl) ratemeter on the resulting solution revealed that a substantial quantity of the activity was extracted since the bared seed had undoubtedly sat for a considerable

Table 3
Results of the $^{90}$Sr activity determinations for bare ceramic sources C1 and C2

| Radionuclide | Strontium-90 | |
|---|---|---|
| Source identification | C1 | C2 |
| Reference time | 1200 EST 1 April 1998 | |
| $^{90}$Sr activity A | 146.2 MBq | 145.7 MBq |
| Expanded uncertainty in $A$ ($k = 2$) | 2.4 MBq | 1.8 MBq |
| Fraction of extracted activity $(1-\Gamma_a/\Gamma_b)$ | $0.9889 \pm 0.0004$ | $0.9721 \pm 0.0003$ |

time in the HCl after the jacket had dissolved. The solution in the assembly was quantitatively transferred to a master solution, labelled 'ss'. This was followed by eight rinses of equal or greater volumes of distilled water, which were also transferred and added to solution 'ss'. On dilution with carrier solution, the total mass of 'ss' was $(242.50 \pm 0.12)$ g. The assembly and bared ceramic source was dried under an infrared heat lamp at a temperature of $(110 \pm 4)°C$ for approximately 4 h. Periodic ionization current measurements of $\Gamma_b$ were performed over the next 8 d to verify the existence of radioactive equilibrium between $^{90}$Y and $^{90}$Sr. Following this determination of $\Gamma_b$, the bared ceramic was sequentially extracted three times to derive

the three master solutions with activities $A_{s1}$, $A_{s2}$ and $A_{s3}$ and the three 'after' relative ionization currents $\Gamma_{a1}$, $\Gamma_{a2}$ and $\Gamma_{a3}$ for the extracted source. Each of these extractions were performed with 11.5 mol $l^{-1}$ HCl (at temperatures typically in the range of $65 \pm 10°C$) over the course of two to four days and employed multiple solution transfers and HCl replenishments (typically three). The determinations of $\Gamma_{a1}$, $\Gamma_{a2}$ and $\Gamma_{a3}$ were obtained from periodic measurements over 18, 1 and 7 d, respectively. Neither $\Gamma_{a1}$ nor $\Gamma_{a3}$ exhibited any evidence of $^{90}$Y–$^{90}$Sr disequilibrium over their respective measurement time intervals. The master solutions were diluted with the HCl/Sr$^{+2}$/Y$^{+3}$ carrier solution to total masses of $M_{s1} = (606.10 \pm 0.30)$

Table 4
Assessment of the uncertainty in the $^{90}$Sr activity for the ceramic sources C1 and C2

| Uncertainty component (and type) | Relative standard uncertainty (in percent) | |
|---|---|---|
| | source C1 | source C2 |
| Relative ionization current measurements to determine the fraction of unextracted activity (A) | 0.041 | 0.028 |
| Effect of possible $^{90}$Y–$^{90}$Sr disequilibrium on ionization current measurements (A) | 0.0003 | 0.0003 |
| $^{90}$Sr decay corrections for ionization current measurements (B) | 0.00002 | 0.00002 |
| Master solution gravimetric dilution (B) | 0.07 | 0.09 |
| Residual activity corrections and possible losses during extractions and solution transfers (A) | 0.011 | 0.004 |
| LS measurement precision for the decay-corrected massic activity of the master solution (within-cocktail-series reproducibility) (A) | 0.52 | 0.17 |
| LS measurement precision (quench dependence) for the decay-corrected massic activity of the master solution (between-cocktail-composition reproducibility) (A) | 0.29 | 0.42 |
| Gravimetric preparation of LS sources (B) | 0.05 | 0.05 |
| Counting livetime and timing interval determinations for the LS measurements (B) | 0.1 | 0.1 |
| $^{90}$Sr decay corrections for LS measurements (B) | 0.0008 | 0.0005 |
| Effect of possible $^{90}$Y–$^{90}$Sr disequilibrium on the LS measurements (A) | 0.33 | 0.018 |
| LS cocktail stability, composition, and mismatch effects (A) | 0.1 | 0.05 |
| $^{90}$Sr and $^{90}$Sr LS detection efficiency calculations, including uncertainties due to: the employed $^3$H standard; quench indicating parameter (QIP) measurements; assumed beta maximum endpoint energies; calculational step sizes; fitted relations between the calculated efficiencies, QIP determinations and the EFFY4-code figures of merit; ionization quenching model assumptions and phototube response asymmetry (B) | 0.43 | 0.37 |
| Combined standard uncertainty | 0.82 | 0.61 |
| Expanded uncertainty ($k = 2$) | 1.6 | 1.2 |

Table 5
Results of the three determinations of the $^{90}$Sr activity for SS-encapsulated source Q10

| Radionuclide | Strontium-90 | | |
|---|---|---|---|
| Source identification | | Q10 | |
| Determination index | $A_1$ | $A_2$ | $A_3$ |
| Reference time | | 1200 EST 1 April 1998 | |
| $^{90}$Sr activity $A$ | 128.8 MBq | 129.7 MBq | 129.7 MBq |
| Expanded uncertainty in $A$ ($k = 2$) | 1.9 Mbq | 1.1 MBq | 0.9 MBq |
| Overall fraction of extracted activity $(1-\Gamma_a/\Gamma_b)$ | $0.4815 \pm 0.0024$ | $0.8119 \pm 0.0010$ | $0.9583 \pm 0.0002$ |
| Mean $^{90}$Sr activity (weighted with $k = 2$ uncertainty) | | $(129.6 \pm 0.7)$ MBq | |

g, $M_{s2} = (754.28 \pm 0.38)$ g and $M_{s3} = (662.34 \pm 0.33)$ g. Single sets of five cocktails were prepared from nearly constant aliquants (in the range of 0.03–0.1 g) of each of the four master solutions. Varying quantities of the blank carrier solution were added to the sets to vary the quenching. Each set of cocktails (along with their composition-matched $^3$H-standard cocktails) were measured on either two or three occasions over time intervals ranging from 8 to 15 d. Residual activity corrections for the resulting four efficiency-traced $A_{ss}$, $A_{s1}$, $A_{s2}$ and $A_{s3}$ determinations were $(2.74 \pm 0.54)$, $(0.039 \pm 0.008)$, $(0.047 \pm 0.010)$ and $(1.47 \pm 0.29)$ %, respectively.

### 3.5. $^{90}$Sr assay results

#### 3.5.1. For the bare ceramic sources

Table 3 gives the results of the radionuclidic assays of sources C1 and C2 as certified by NIST (1998). The uncertainty assessment for the $^{90}$Sr activity of the two sources is summarized in Table 4. The combined standard uncertainty is multiplied by a coverage factor of $k = 2$ to obtain the expanded uncertainty which is presumed to provide a level of confidence of 95%.

#### 3.5.2. For the SS-encapsulated seeds

The radionuclidic assay for source Q11 could not be completed because of an accidental spill during the extraction process. Some unknown portion of the activity in the extracted solution was lost when part of the solution spilt from the handling assembly during a solution transfer.

The results for the three determinations of the $^{90}$Sr activity of source Q10 is given in Table 5. The uncertainty assessments are summarized in Tables 6 and 7. The results for these three determinations were obtained from the measurements of:

$A_{ss} = (0.6925 \pm 0.0055)$ Mbq,
$A_{s1} = (61.66 \pm 0.33)$ MBq,
$A_{s2} = (42.90 \pm 0.25)$ MBq,
$A_{s3} = (19.03 \pm 0.12)$ MBq,
$\Gamma_{a1}/\Gamma_b = 0.5185 \pm 0.0024$,
$\Gamma_{a2}/\Gamma_{a1} = 0.3628 \pm 0.0018$,
$\Gamma_{a3}/\Gamma_{a2} = 0.2218 \pm 0.0013$,

where each cited uncertainty interval corresponds to a combined standard uncertainty.

Table 6
Assessment of the uncertainty in the $^{90}$Sr activity determinations for the SS-encapsulated source Q10

| Uncertainty component | Relative standard uncertainty (in percent) | | |
|---|---|---|---|
| | determination $A_1$ | Determination $A_2$ | determination $A_3$ |
| Relative ionization current measurements to determine the fraction of unextracted activity, including $^{90}$Sr decay corrections and the effect of possible $^{90}$Y–$^{90}$Sr disequilibrium | 0.49 | 0.15 | 0.032 |
| $^{90}$Sr activity for master solution $A_{ss}$ (see Table 7) | 0.0043 | 0.0043 | 0.0043 |
| $^{90}$Sr activity for master solution $A_{s1}$ (see Table 7) | 0.52 | 0.25 | 0.25 |
| $^{90}$Sr activity for master solution $A_{s2}$ (see Table 7) | – | 0.31 | 0.20 |
| $^{90}$Sr activity for master solution $A_{s3}$ (see Table 7) | – | – | 0.12 |
| Combined standard uncertainty | 0.72 | 0.43 | 0.35 |
| Expanded uncertainty ($k = 2$) | 1.4 | 0.86 | 0.70 |

Table 7
Assessment of the uncertainty in the $^{90}$Sr activity for the four master solutions $A_{ss}$, $A_{s1}$, $A_{s2}$, and $A_{s3}$ used for the assay of SS-encapsulated source Q10

| Uncertainty component (and type) | Relative standard uncertainty (in percent) | | | |
|---|---|---|---|---|
| | $A_{ss}$ | $A_{s1}$ | $A_{s2}$ | $A_{s3}$ |
| Master solution gravimetric dilution (B) | 0.09 | 0.07 | 0.07 | 0.07 |
| Residual activity corrections and possible losses during extractions and solution transfers (A) | 0.54 | 0.008 | 0.010 | 0.29 |
| LS measurement precision for the decay-corrected massic activity of the master solution (within-cocktail-series reproducibility) (A) | 0.10 | 0.038 | 0.056 | 0.046 |
| LS measurement precision (quench dependence) for the decay-corrected massic activity of the master solution (between-cocktail-composition reproducibility) (B) | 0.35 | 0.35 | 0.35 | 0.35 |
| Gravimetric preparation of LS sources (B) | 0.05 | 0.05 | 0.05 | 0.05 |
| Counting livetime and timing interval determinations for the LS measurements (B) | 0.1 | 0.1 | 0.1 | 0.1 |
| $^{90}$Sr decay corrections (B) | 0.001 | 0.0008 | 0.0008 | 0.0005 |
| Effect of possible $^{90}$Y–$^{90}$Sr disequilibrium on the LS measurements (A) | 0.012 | 0.010 | 0.015 | 0.015 |
| LS cocktail stability, composition, and mismatch effects (A) | 0.06 | 0.02 | 0.16 | 0.06 |
| $^{90}$Sr and $^{90}$Sr LS detection efficiency calculations (see Table 4 for components) (B) | 0.44 | 0.37 | 0.42 | 0.39 |
| Combined standard uncertainty | 0.80 | 0.53 | 0.59 | 0.62 |
| Expanded uncertainty ($k = 2$) | 1.6 | 1.1 | 1.2 | 1.2 |

### 3.6. Confirmatory measurements

Five-g aliquants of all of the master solutions used in this work were dispensed into NIST standard 5-ml, flame-sealed, glass ampoules and were used for secondary, less-accurate, confirmatory measurements. Comparisons of ionization current measurements on these ampoules confirmed, on a relative basis, the internal consistency amongst the various master solution dilutions and the LS assay results. The agreements (in terms of ratios of the ampoules' ionization currents compared to their determined massic activity ratios) were in all cases within a few percent and, for higher-activity ampoules, were more typically within a few tenths of 1%.

In addition, the internal consistency of the assays could be evaluated by comparisons of the ratios of the relative ionization currents $\Gamma$ and the derived activities remaining in the bared ceramic sources (see Section 3.7].

### 3.7. Determination of calibration factors for non-destructive assays

NIST 'chamber A' calibration factors were determined for both the bare ceramic sources and the SS-encapsulated seed Q10. The calibration factors, given in Table 8, can be used for subsequent non-destructive assays of comparable $^{90}$Sr–$^{90}$Y sources (in the same source holder geometries). For example, based on the calibration factor obtained from the assay of seed Q10

and the initial ionization current measurements of seed Q11, the $^{90}$Sr activity of seed Q11 was $(134.1 \pm 1.1)$ MBq at the 1200 EST 1 April 1998 reference time and where the cited uncertainty interval is an expanded uncertainty with $k = 2$.

The calibration factor $K = \Gamma/A$ is defined by the ratio of the relative ionization current $\Gamma$ (normalized to some radium reference source) and the $^{90}$Sr activity $A$ in the source, where both $\Gamma$ and A are decay corrected to a common reference time and for the conditional that $^{90}$Y is in radioactive equilibrium with $^{90}$Sr. In addition to the calibration factor determinations for sources C1 and C2, bare ceramic $K$ values were also obtained from the results of the intermediate steps in the assay of seed Q10. In this case, the calibration factor for the bared ceramic derived after each step was:

$$K = \Gamma_b/(A - A_{ss}) = \Gamma_{a1}/(A - A_{ss} - A_{s1})$$

$$= \Gamma_{a2}/(A - A_{ss} - A_{s1} - A_{s2})$$

$$= \Gamma_{a3}/(A - A_{ss} - A_{s1} - A_{s2} - A_{s3}) \qquad (18)$$

where $A = (129.6 \pm 0.7)$ MBq corresponds to the $^{90}$Sr activity in the original intact seed Q10.

## 4. Concluding notes

This work established a primary standardization for the Bebig/Novoste $^{90}$Sr–$^{90}$Y brachytherapy sources

Table 8
Calibration factors for NIST 'chamber A' at a reference time of 1200 EST 1 April 1998. The relative ionization current measurements were performed with the NIST re-entrant ionization chamber 'A' with the source contained within the 'handling assembly' which was in turn placed inside the chamber's source holder. The uncertainty intervals given for $A$, $\Gamma$ and $K$ are expanded uncertainties with a coverage factor of $k = 2$

| Source | $^{90}$Sr activity A in the source | Ionization current $\Gamma$ (relative to RRS20) | Calibration factor $K = \Gamma/A$ $(MBq^{-1})$ |
|---|---|---|---|
| Sealed seed | | | |
| Q10 | $(129.6 \pm 0.7)$ MBq | $1.7042 \pm 0.0042$ | $(0.01315 \pm 0.00008)$ |
| Bare ceramics | | | |
| C1 | $(146.2 \pm 2.4)$ MBq | $1.6755 \pm 0.0114$ | $(0.01146 \pm 0.00020)$ |
| C2 | $(145.7 \pm 1.8)$ MBq | $1.6602 \pm 0.0074$ | $(0.01140 \pm 0.00015)$ |
| Q10 (after SS removal) | $(128.9 \pm 0.7)$ MBq | $1.4876 \pm 0.0102$ | $(0.01154 \pm 0.00010)$ |
| Q10 (after 1st extraction) | $(67.25 \pm 0.97)$ MBq | $0.7713 \pm 0.0047$ | $(0.01147 \pm 0.00018)$ |
| Q10 (after 2nd extraction) | $(24.35 \pm 1.10)$ MBq | $0.2798 \pm 0.0022$ | $(0.01149 \pm 0.00053)$ |
| Q10 (after 3rd extraction) | $(5.32 \pm 1.13)$ MBq | $0.06206 \pm 0.00055$ | $(0.0117 \pm 0.0025)$ |

that are intended for use in the prophylactic treatment of restenosis. Its findings provided definitive $^{90}$Sr activity determinations that were critical for providing a link between the necessary dosimetric measurements and calculations for these sources and for establishing an ionization-chamber-based calibration factor for a relative, secondary measurement method. The latter can now be used to perform subsequent nondestructive assays which will be decidedly simpler to conduct, less time and labor intensive and more cost effective than the difficult and laborious efforts required for the presently-described destructive radioanalytical method. In the same vein, the results of this standardization could be used by the manufacturer to establish in-house calibration factors for quality control measurements for their source production methods.

The results presented here, of course, apply only to the Bebig/Novoste sources. The study, however, went beyond merely serving the interests and calibration needs for the application of this one particular kind of radionuclidic source. Much more significantly, the general method developed for the present work may have much broader application for the destructive assay of other kinds of sources that are not amenable to complete digestive procedures.

The coupling of ionization current measurements that account for the unextracted fraction of activity $(\Gamma_a/\Gamma_b)$ with conventional radionuclidic assays for the extracted fraction $(A_s/A)$ to account for the total activity $A = A_s/(1 - (\Gamma_a/\Gamma_b))$ may have application to other materials that are difficult (if not impossible) to dissolve. The method may have utility even for cases in which the ionization current response $\Gamma_a$ is not constantly proportional to the remaining activity $(A - A_s)$ in the source after extraction, but is instead a function

$f$ of the amount of extracted activity, such that $K \neq (A - A_s)/\Gamma_a = f(A_s/A)$. In this case, sequential extractions (with successive $A_{s(i)}$ and $\Gamma_{a(i)}$ determinations) may be used to reveal the $f(A_s/A)$ functional form so that $A$ becomes determinable from extrapolations of the serial $\Sigma_i A_{s(i)}$ values to $\Gamma_a/\Gamma_b = 0$.

ject; and the author's esteemed colleague and boss, Bert Coursey, who, however often additional experiments were proposed, never failed to provide timely encouragements.

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