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Short note

STUDY OF THE INFLUENCE OF RADIONUCLIDE IMPURITIES IN RADIONUCLIDE METROLOGY*

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Abstract. *The absolute standardization of radionuclides is complex in the case of mixtures, such as is the case with the existence of radioactive impurities. The difficulty is still amplified in the case of radionuclides with very different emissions of gamma-rays, both as energy and intensity. One example is Co-57, which can contain impurities of isotopes Co-56 and Co-58. Even low contents of such impurities can influence in a significant way the final result as well as the activity of the measured radioactive source. In the present paper, an example of treatment of this mixture within the participation in the supplementary comparison code CCRI(II)-S6.Co-57, organized by the IAEA, within the Coordinated Research Project, CRP E.2.10.05 and in the key comparison code BIPM(II)-K1.Co-57, where the influence of impurities was underlined, will be presented.*

Key words: *Base radionuclide: Co-57; impurities: Co-56, Co-58; international comparisons*

1. INTRODUCTION

The radionuclide ^{57}Co is used in nuclear medicine directly to label the vitamin B12 for investigations of the gastrointestinal tract and for obtaining the product bleomycin-Co-57, used in tumor investigations. Recently, its use in obtaining radioactive sources for brachytherapy was reported [1]; it can be superior to ^{192}Ir used in afterloading units, due to its lower gamma-ray energy, the absence of beta radiations and longer half-life. Indirectly, it is used in the calibration and check of gamma cameras and radionuclide calibrators, see the document IAEA TRS454 [2], as a ^{99m}Tc mock standard or check source. In industrial applications, it is used for the production of Mössbauer radiation sources. In radionuclide metrology it is a basic radionuclide for the calibration of the gamma-ray spectrometers.

Due to the large list of applications, the precise measurement of ^{57}Co activity and the assurance of its metrological traceability is an important task of a Radionuclide Metrology Laboratory.

This was the reason for our participation in two international comparisons regarding its standardization, in the quality of IFIN-HH, of a Designated Institute within the International Committee of Weights and Measures – Mutual Recognition Arrangement (CIPM-MRA). The first comparison was organized within the Coordinated Research Project, CRP E.2.10.05 "Harmonization of quality practices for nuclear medicine radioactivity measurements", registered at the International Committee of Weights and Measures – Consultative Committee for Ionising Radiations Section II, Radionuclide Measurement [CIPM-CCRI(II)], as a supplementary comparison, code CCRI(II)-S6.Co-57, where ^{57}Co was used as a ^{99m}Tc mock standard. The ^{57}Co solution was received by the care of the International Atomic Energy Agency (IAEA) and was supplied by the QSA Global GmbH, at present time Eckert & Ziegler Nuclitec, Gieselweg 1, Braunschweig, Germany. The second comparison was a key comparison of the type BIPM.RI(II)-K1.Co-57 and deployed within the International Bureau of Weights and Measures (BIPM), International Reference System (SIR), using the solution standardized within the CCRI(II)-S6.Co-57.

2. EXPERIMENTAL PROCEDURE

2.1. Preparation of solutions and sources

The original solution was diluted gravimetrically, in order to standardize it. An ampoule containing standardized solution was sent to the BIPM for the SIR; others, radiopharmaceutical glass vials, types Schott 10R and penicillin P6, with nominal masses of 5.0 g of standardized solution were prepared and measured in the CENTRONIC IG12/20A ionization chamber, in order to calibrate it [3]. Solid sources were prepared gravimetrically on golded VYNS films for absolute standardization.

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2.2. Standardization of the sources

The radionuclide ^{57}Co decays by electron capture, followed by the prompt emission of gamma-rays; the complete decay scheme is presented in reference [4]. The solution was standardized by the 4π(PC)-γ coincidence method in the efficiency extrapolation variant, described in [5]. PC is a proportional counter, working with methane flow at normal pressure.

2.3. Measurement of impurity content

Impurity measurement. The reference [4] predicts as possible impurities ^{56}Co ($T_{1/2}$ = 77.24 d) and ^{58}Co ($T_{1/2}$=70.85 d). These impurities were detected also in our laboratory and were determined by the use of the high resolution γ-ray spectrometry, with a system provided with a HPGe detector. The result was: the activity of ^{58}Co was (0.050±0.035)% and that of ^{56}Co was (0.117 ±0.065)% from the activity of ^{57}Co, on the measurement of activity time; the combined standard uncertainties are given for the coverage factor (k=1).

Impurity correction. We calculated the activity reported in the two comparisons with an impurity correction factor, 1.00167, representing the sum of the two impurity contents on the time of measurement. The combined standard uncertainty was u_c=0.79%.

3. Reporting of data and the results of the two comparisons

3.1. The CCRI(II)-S6.Co-57, IAEA comparison

The IFIN-HH reported the value of QSA Global solution concentration, on the comparison reference time, corrected for the determined content of impurities. The comparison reference value (CRV), calculated only from primary standardization methods, such as reported in [6],

was CRV = (35.54±0.19)MBq g^{-1}.

The difference of our reported ^{57}Co activity concentration from the CRV was calculated as D_i= -0.79%.

3.2. The BIPM.RI(II)-K1.Co-57, key comparison

The same standardized, diluted solution, introduced in one SIR ampoule, calculated for decay, was sent to the BIPM. The reported values to the BIPM maintained the same impurity content: ^{58}Co – (0.050±0.035)% and ^{56}Co – (0.117 ±0.065)% from the activity of ^{57}Co, as in the IAEA comparison, not calculating their decay. The measurement in the ionization chamber of BIPM resulted in the following results, reported in [7]. As compared with the key comparison reference value (KCRV), the difference of IFIN-HH result was D_{i1} =+1.43%. After one year, when a significant decay of impurities occurred, our new BIPM calculated equivalent activity was lowered and the new difference was: D_{i2} = +0.42%, resulting a relative difference between the two evaluations of: D_{i1}-D_{i2} = 1.01%. Taking into account these results, we recalculated and reported to the BIPM the decay corrected content of impurities as: ^{58}Co – (0.026±0.018)% and ^{56}Co – (0.065 ±0.036)% from the activity of ^{57}Co, for the BIPM-SIR comparison reference time. We calculated the influence of impurities using measurement data from IFIN-HH for our CENTRONIC IG12/20A ionisation chamber.

The differences between these new calculated and reported and the initial impurity content values are respectively: ^{58}Co – (0.024 ±0.017) % and ^{56}Co – (0.052±0.029)%. In this case, we calculated the difference in the impurity influence measured at BIPM, which is different from the impurity activity versus ^{57}Co activity ratio, due to the higher contributions in the ionization current. We used the new reported impurity contents and ratio of ionization currents, according to the procedure largely described in the reference [5], as:

$$(A_{impCo-58} + A_{impC0-56}) / A_{Co-57} = (0.88 \pm 0.44)\%$$

This value is in very good agreement with the D_{i1}-D_{i2} =1.01% and fully explains the difference of the measurements in the BIPM-SIR system, based on the ionization chamber.

This result reconfirms the importance of the careful treatment of impurities, and mainly underlines their contribution in ionization chamber measurement, such as reported also in our papers [8 and 9].

4. Conclusions

A ^{57}Co standardized solution was used in two international comparisons: CCRI(II)-S6.Co-57 and BIPMRI(II)-K1.Co-57. The results were contradictory: in the first case our result was lower than the comparison reference value CRV, but in the second it was higher than the KCRV.

The solution contained the radionuclide impurities ^{58}Co and ^{56}Co.

This paper explains the reason for these differences, due to the important contribution of impurities in the ionization chamber measurement.

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