TECHNICAL ACTIVITIES

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Environment

Detector technology

Measurement methodology

RADIONUCLIDE METROLOGY

Nuclear Medicine

Nuclear data

Traceability

RADIOACTIVITY GROUP
IONIZING RADIATION DIVISION
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- **Photo-nuclear produced radioactivities:** The MIRF facility has been used to produce positron emitting radioactivities. Techniques have been developed to characterize those sources. Efficiencies for positrons which have been stopped and annihilated, have been developed and compared, when appropriate, with associated gamma-ray emission from the decay. Radioactivities characterized this last year included ^{18}F, ^{34m}Cl, and ^{126}I. The last radionuclide is of particular interest as a high gamma-ray energy emission rate standard for Ge detector calibration. $^{34}Cl^{m}$ has a 3303.6 keV gamma-ray with a probability per decay of 0.123. This abundant emission and readily prepared radioactivity allows the extension of the current efficiency curve to that energy. (F.J. Schima)

- **^{63}Ni Standardization and Decay Studies.** Standard solutions of ^{63}Ni have recently been prepared and will be disseminated as NIST SRM 4226C. The solutions were calibrated by $4\pi\beta$ liquid scintillation (LS) spectrometry with ^{3}H- standard efficiency tracing. This radionuclide is of interest to the nuclear reactor community because it is often found in reactor environments as the neutron activation product of nickel present in the steel used in construction of those facilities. Certain physical properties, namely a low β^- energy (66.945 $\pm$ 0.004 keV) and a relatively long half-life (101.1 $\pm$ 1.4 a) also make it attractive for radionuclidic metrology studies, as it tends to be a more sensitive indicator of effects in measurement technique and procedures than would a radionuclide with a higher β^- energy. In order to identify and quantify as many of these effects as possible, over 975 measurements involving 55 different LS samples were performed. Among the variables studied were aqueous fraction of LS sample, LS sample mass (volume) dependence, LS sample age (time between sample preparation and measurement) dependence, and effects due to different scintillation fluors. In addition, an exhaustive uncertainty analysis was performed. This SRM is gravimetrically related to two others previously prepared by NIST/NBS (SRM 4226, prepared in 1968 and calibrated by microcalorimetry and SRM 4226B, prepared in 1984 and calibrated with $4\pi\beta$ LS spectrometry with ^{3}H- standard efficiency tracing), allowing a comparison to be made between the three sources as a check of both measurement consistency and solution stability. After adjusting the data from the 1968 and 1984 measurements to include the latest available nuclear data for both ^{63}Ni and ^{3}H, the three measurements were found to be in agreement to within 0.3 %. Because of this remarkable consistency over a 27-year span, the three data sets provided enough data to make the first-ever determination by radioactive decay of the ^{63}Ni half-life. Using these data, the half-life was determined to be 101.06 $\pm$ 1.97 a. A critical review of measurements of the ^{63}Ni half-life was performed, resulting in a new recommended value of 101.1 $\pm$ 1.4 a. A series of articles addressing all of these topics is being prepared for publication in *Applied Radiation and Isotopes*, *Radioactivity and Radiochemistry* and the *Journal of Research of the National Institute of Standards and Technology*. (B. E. Zimmerman, R. Collé)

- **Development of Standards for the Palliative Therapy Radionuclide:** The American Cancer Society estimates that about 180,000 new cases of breast cancer and an equal number of cases of prostate cancer are diagnosed each year in the United States. New radiopharmaceuticals are being developed which have been shown to relieve pain that is unresponsive to narcotic treatment in many of these cases. NIST in collaboration with the Nuclear Energy Institute (NEI) has distributed standard reference materials (SRMs) for over 20 years which is an important first step in ensuring the accuracy of injected radiopharmaceuticals in the US.

Over the past two years NIST scientists in the Radioactivity Group have worked to standardize two new nuclides for bone palliation: ^{89}Sr (half life 50.5 days) and ^{117m}Sn (half life 14 days).

The ^{89}Sr used in SRM 4426A was produced in a fast-flux nuclear reactor in Obinsk, Russia and purified at NIST. The material was standardized by high-accuracy liquid-scintillation counting, and decay-scheme data were established by Ge(Li) gamma-ray spectrometry. The high-activity SRM 4426H-A was then distributed to the FDA's calibration laboratory and to North American radiopharmaceutical manufacturers, and a lower-activity SRM 4426L-A was distributed mainly to instrument companies and medical centers. (B.M. Coursey, D.B. Golas, and F.J. Schima)

As part of an increasingly active program to develop national standards for radionuclides of interest to the nuclear medicine community, this group has recently performed a calibration of ^{117m}Sn, which is currently under study for use in palliative therapy for pain associated with metastatic bone cancer. The calibration was performed using three techniques: γ-ray spectrometry with HPGe detectors, γ-ray spectrometry with a 4π 30- cm NaI(Tl) system, and $4\pi\beta$ liquid scintillation (LS) spectrometry. While the data are in the final stages of analysis, initial results indicate good agreement between the results obtained with each of the three methods. Data were also obtained for the re-determination of the halflife and probabilities per decay of the major emissions. A procedure for the direct standardization of this isomeric radioactivity based on sum coincidence peaks in a Ge spectrum of the ^{117m}Sn is under investigation. This appears to be one of those rare cases in which a ^{117m}Sn source can be calibrated by this method when using a Ge detector with an adequate resolution. This radionuclide is particularly exciting because of the greater uptake of ^{117m}Sn(4+)DTPA in bone tissue relative to the marrow. Compared to other commonly-used bone palliation radionuclides such as ^{32}P, ^{89}Sr, and ^{186}Re, there is as much as a 4-fold increase in the ratio of bone surface dose to bone marrow dose with the use of ^{117m}Sn(4+)DTPA. This suggests that a much higher dose can be given to the patient before marrow toxicity levels are reached, possibly leading to the ability to treat the metastases themselves. An additional advantage in using ^{117m}Sn is the presence of a 159- keV γ-ray, which allows the uptake and distribution of the radionuclide to studied with conventional imaging devices. (B. E. Zimmerman, J. T. Cessna and F.J. Schima)

• **A Mock "Soil" Calibration Standard for γ-Ray Spectrometry.** At the request of the Nuclear Energy Institute (NEI), a spiked, mixed-radionuclide, mock "soil" calibration standard (in a 1-L Marinelli beaker configuration) was gravimetrically prepared, and calibrated by high-resolution photonic-emission spectrometry. The matrix consisted of a well-characterized (in terms of densities, particle sizes, etc.) and blended standard Ottawa sand. The matrix was spiked with ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, and ^{60}Co. Source homogeneity and possible spike losses were evaluated for every spiking radionuclide. Measurements on aliquants (30 g samples out of 1850 g of spiked matrix) taken as a function of blending times (40 min to 400 min) indicated source homogeneity to within several percent. The only observed spike loss was for ^{203}Hg which is due to the chemical instability and volatility of deposited mercury compounds. A quantitative, verifiable, and efficacious spiking protocol was devised and developed as part of this work, and used to prepare the Marinelli-beaker calibration standard. The calibration obtained in this work will allow NIST to provide calibration services for other similar Marinelli beaker sources. Such sources are of vital importance for measurements made by the nuclear

energy industry. In addition, the developed protocol will be of great benefit to other laboratories for use in preparing their own Marinelli beaker calibration sources. A publication describing the protocol is in preparation. (R. Collé and F.J. Schima)

- **Calibrations for Tropospheric Radon Measurements.** As part of a collaboration with the Atmospheric Sciences Research Center (ASRC) of the State University of New York at Albany (Western Field Office, Moffett Field, CA), efforts are underway to calibrate and evaluate the performance (stability, linearity, transfer and detection efficiencies, etc.) of airborne instrumentation used to measure radon (^{222}Rn) concentrations in the troposphere. Atmospheric radon measurements, both at surface levels and at varying altitudes, are widely used by the metrology and atmospheric sciences communities in applications ranging from its use as an indicator of the presence of continental air masses over the oceans to its use as a tracer for identifying air of recent tropospheric origin in the lower stratosphere. It has also become a major tool in the development and verification of global circulation and chemical transport models. These are the complex models used to assess questions such as ozone depletion and the possible effect of supersonic and subsonic aircraft on the chemical state of the atmosphere. Recently, modelling groups from all over the world have begun to examine some of the significant discrepancies in the various models, and have identified the need for reliable radon measurement results. The present collaboration with ASRC will assure and enhance the quality of a significant database required by the modelers -- namely free tropospheric profiles as a function of altitude. The on-going ASRC tropospheric measurements will represent an order-of-magnitude increase in the size of the database previously available in this region, and the NIST contributions to the collaboration will undoubtedly improve their quality by nearly as much. Major breakthroughs in evaluating the transport component of the models are anticipated as a result of significant improvements in the quality of this radon measurement database. (R. Collé)

- **Final Results for the International Intercomparison of Marine-Atmospheric Radon Measurements.** The importance of various kinds of high quality radon measurement data to the world's atmospheric transport modelers was identified in the preceding highlight. In 1991-1992, NIST conducted an *in situ* calibration and intercomparison exercise for marine atmospheric radon measurements. The participating laboratories have been responsible for perhaps 95 percent of the available surface-level measurements gathered around the globe over the last decade. The results of this intercomparison exercise have at last been fully published in a series of articles that appeared in the *Journal of Geophysical Research* and *Journal of Research of NIST*. The intercomparison utilized a common standardized, *in situ*, reference basis (provided by NIST) that could be directly related to U.S. national, and internationally, recognized, ^{226}Ra and ^{222}Rn standards, and evaluated the performance of all principal instruments that are used to measure radon activity concentrations for marine-atmospheric studies. The findings will assist various users in the global modelling community in applying the available and future radon measurement data bases in a more reliable and effective manner. The work went beyond serving the needs of just this particular intercomparison. It also demonstrated the broader utility of the developed procedures, i.e., the calibration protocol and the methodology for providing *in situ* standardized samples. Most environmental measurement intercomparisons of field instruments in actual use merely rely on evaluating the relative performance of the participants, or some

comparison to the pooled results. This exercise demonstrated, for the very first time, the capability of providing a standardized reference basis even for such low-level, field-measurement intercomparisons. The developed methodologies could be adopted with slight modifications to cover other radon concentration ranges and other applications, and could be employed in many other types of radon environmental field-measurement intercomparisons. (R. Collé)

- **Radon Measurement Standards Program.** The primary objectives of this program are to maintain the national standards for ^{226}Ra and ^{222}Rn, to develop new transfer standards and measurement applications, and to disseminate standards and provide other mechanisms for insuring the quality of radon measurements. Highlights of this year's accomplishments primarily involved substantial efforts to evaluate and verify the continuing performance of our measurement systems and extant standards. These activities are critical for maintaining the internationally-recognized excellence of our radon measurement capability. The performance and long-term stability of both the radon-in-water standard generator and radon emanation capsule standards have been undergoing continuing evaluations (the former for now over the past 6 years). The NaI-based secondary radon measurement system was completely re-evaluated in terms of extending its dynamic range (by developing a new deadtime correction approach), confirming the calibration factors, and performing a more realistic uncertainty analysis. This work was largely done as part of the atmospheric radon measurement collaborations with ASRC. In order to achieve the low radon concentration ranges required for the atmospheric profiles, there was a need to verify that decay corrections for our sample bulbs could be made over 10 to 12 half-lifes. This verification in turn necessitated experiments starting at higher activity concentrations than previously used with the system. The deadtime corrections previously employed were found to be inadequate at these higher concentrations. The new approach can accurately handle samples from 20,000 counts per second down to background levels. Work was also performed on testing some aspects (mainly sample transfers) of the ongoing EPA traceability program that ensures the relatability of EPA measurements to U.S. national standards. This program, operating under an interagency agreement, tests the measurement proficiency of the two EPA laboratories that in turn conduct the national radon measurement proficiency testing program for commercial radon measurement vendors. (R. Collé, J. Cessna, P.A. Hodge, and J.M.R. Hutchinson)

- **Internal Gas Proportional Counting.** An intercomparison of tritiated water standards is being planned by ICRM involving NPL, PTB and NIST as the lead laboratories. This involves the conversion of the liquid standards to tritium gas and calibration in the NIST length-compensated internal gas proportional counters. A recalibration of the NIST tritiated water standards is planned. Tritiated samples of gas from private suppliers are calibrated upon request, about one every two months. (M.P. Unterweger)

- **Calibration of Large-Area Beta Sources.** Calibration of the $2\pi\beta$ emission rate of several large area sources have been completed and the effects of β-backscattering are under investigation in order to provide accurate values of the activities of these sources for use in calibrating β field monitors. A large number of large area sources will be calibrated for $2\pi\beta$ emission rate and for homogeneity for a private supplier. (M.P. Unterweger, P. Hodge)

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