

DISINTEGRATION RATE MEASUREMENTS OF ^{51}Cr AND ^{54}Mn RADIOACTIVE SOLUTIONS

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ABSTRACT

This paper describes the procedure followed by the Nuclear Metrology Laboratory (LMN) of the IPEN - CNEN/SP, in São Paulo, for the standardization of ^{51}Cr and ^{54}Mn . The calibration was performed in a $4\pi\beta\text{-}\gamma$ coincidence system by the efficiency extrapolation method. The electronic system was the conventional, using the TAC method developed at LMN, for the registrations of the observed events.

1. INTRODUCTION

The Nuclear Metrology Laboratory (LMN) of the IPEN - CNEN/SP, in São Paulo, since its foundation, has been involved in the development of methods for the standardization of radionuclides, mainly those, which are suitable for the calibration of secondary standard systems, such as ionization chambers and spectrometers. This paper describes the procedure followed by LMN for the standardization of ^{51}Cr and ^{54}Mn radioactive solutions, selected to be used as tracers in the standardization of ^{55}Fe by the tracing method, or in the gamma-ray spectrometers for efficiency calibration.

These radionuclides decay by electron capture process followed by gamma-ray emission. The radionuclide ^{51}Cr decays 9.89 % by electron capture to the excited states of ^{51}V , with emission of a 320 keV gamma-ray and 90.11% by electron capture to the ground state of ^{51}V . Mn-54 decays 100 % by electron capture to the excited levels of ^{54}Cr , with the emission of a 834 keV gamma-ray. The calibration was performed in a $4\pi\beta\text{-}\gamma$ coincidence system by the efficiency extrapolation method. The electronic system was the conventional, using the TAC method developed at LMN [1] for the registrations of the observed events.

2. EXPERIMENTAL METHOD

2.1. Source Preparation

The $^{51}\text{CrCl}_3$ aqueous solution was obtained from the Radiopharmaceutical Center of IPEN and the $^{54}\text{MnCl}_2$ was purchased from Isotopes Laboratories Ltd. The radioactive sources of ^{51}Cr and ^{54}Mn were prepared by dropping known aliquots onto a Collodion film substrate $20 \mu\text{g cm}^{-2}$ thick. This film had been previously coated with a $10 \mu\text{g cm}^{-2}$ gold layer in order to make the film conductive. A seeding agent (CYASTAT SM) was used to improve the deposit uniformity; the sources were dried in a desiccator. The mass determination was performed using the pycnometer technique [2].

2.2. $4\pi\beta\text{-}\gamma$ Coincidence method

The $4\pi\beta\text{-}\gamma$ coincidence system consisted of a 4π proportional counter (PC) filled with P-10 gas, operated at 0.1 MPa, for detecting the X-rays and Auger electrons coming from the electron capture process decay. The PC counter was coupled to a pair of 76 mm x 76 mm NaI(Tl) scintillators for detecting gamma-rays. The measurements were performed selecting a gamma-ray window at the total absorption peak of 320 keV for ^{51}Cr and at 834 keV for ^{54}Mn . Collodion films $50\mu\text{g cm}^{-2}$ thick and aluminum foils $150 \mu\text{g cm}^{-2}$ thick were used as external absorbers, placed on both sides of the sources.

The formulae applied to the coincidence measurement is given by the well-known relationship [3] where the number of detected events in the proportional counter is given by:

$$N_{(X,A)} = N_0 \left[a_1 \epsilon_{(X,A)} P_1 + a_2 \epsilon_{(X,A)} P_2 + a_1 (1 - \epsilon_{(X,A)}) \frac{(\alpha_T \epsilon_{ce} + \epsilon_{\beta\gamma})}{1 + \alpha_T} \right] \quad (1)$$

The number of gamma-ray detected events in the scintillator events are given by:

$$N_\gamma = N_0 a_1 \epsilon_{\gamma 1} \frac{1}{1 + \alpha_T} \quad (2)$$

The coincidental events are given by:

$$N_c = N_0 \left[a_1 \epsilon_{(X,A)} P_1 \epsilon_{\gamma 1} \frac{1}{1 + \alpha_T} \right] \quad (3)$$

Therefore,

$$\frac{N_{(X,A)} N_\gamma}{N_c} = N_0 \left[a_1 + a_2 \frac{P_2}{P_1} + \frac{a_1 (1 - \epsilon_{(X,A)})}{\epsilon_{(X,A)}} \frac{(\alpha_T \epsilon_{ce} + \epsilon_{\beta\gamma})}{1 + \alpha_T} \right] \quad (4)$$

Where:

N_0 is the disintegration rate;

$N_{(X,A)}$, N_γ and N_c are the electron Auger or X-ray, gamma-ray and coincidence counting rates, respectively;

a_1 and a_2 are the probabilities of electron capture process emission to the excited level and ground state of ^{51}V , respectively;

P_1 and P_2 are the probabilities of electron capture, relative to the excited level and to the ground state, respectively;

$\epsilon_{(X,A)}$ is the electron Auger or X-ray detection efficiency;

$\epsilon_{\gamma 1}$ is the gamma-ray detection efficiency of NaI(Tl) spectrometer;

$\epsilon_{\beta\gamma 1}$ is the gamma-ray detection efficiency of proportional counter;

ϵ_{ce} is the conversion electron detection efficiency and

α_T is the internal conversion coefficient.

The observed counting rates $N_{(X,A)}$ and N_γ were corrected for background, dead time and decay in the usual way. The coincidence rate N_c was corrected for dead time and accidental coincidences using the Cox-Isham formalism [4], adapted by Smith [5].

3. RESULTS

Fig. 1 shows the extrapolation curve obtained for the measurements of ^{51}Cr . In this curve, the behavior of the ratio $N_{(X,A)} N_\gamma / N_c$ as a function of the $(1-N_c/N_\gamma)/(N_c/N_\gamma)$ parameter presents a slope closed to zero, showing the same behavior as obtained with electronic threshold discrimination [6].

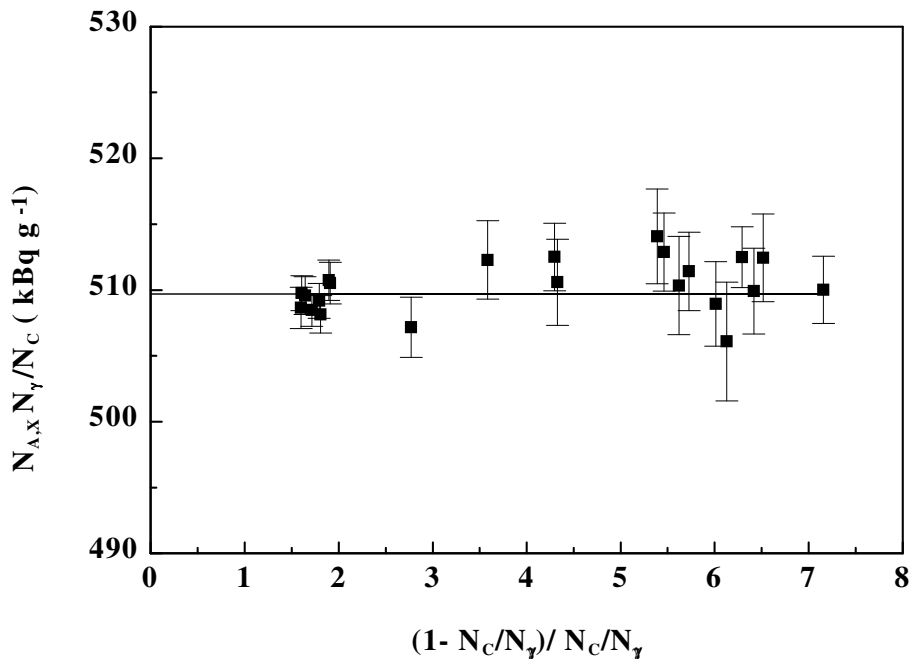


Figure 1. Extrapolation curve of ^{51}Cr obtained using external absorbers.

Fig. 2 shows the extrapolation curve obtained for the measurements of ^{54}Mn . The behavior of the ratio $N_{(X,A)} N_\gamma / N_C$, as a function of the $(1-N_C/N_\gamma)/(N_C/N_\gamma)$ parameter, presents a slope of (0.00337 ± 0.00035) , this slope is mainly due to detection of gamma-rays in the proportional counter. The value of $\varepsilon_{\beta\gamma}$ is around 0.3 % [7].

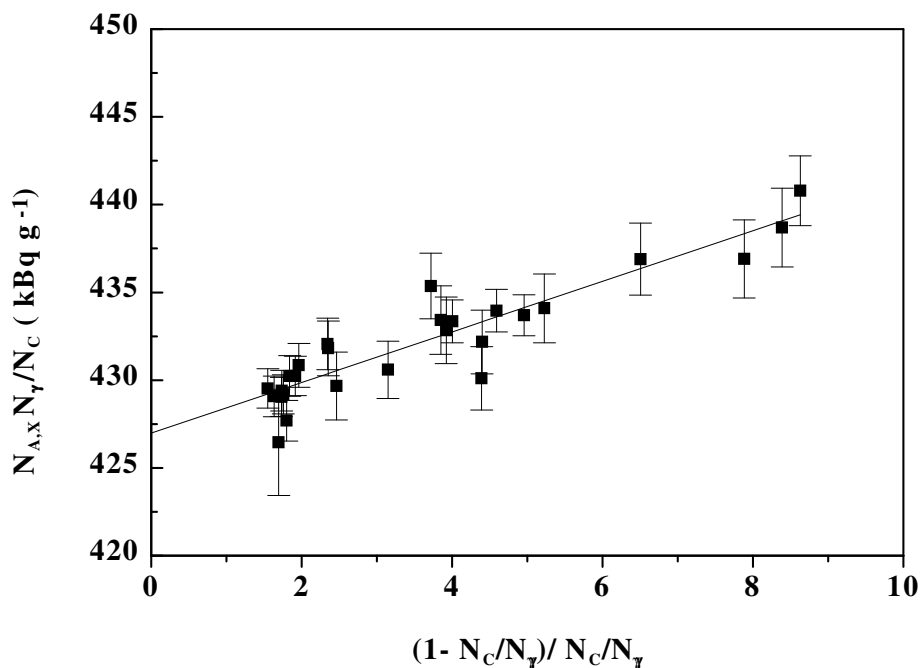


Figure 2. Extrapolation curve of ^{54}Mn obtained by using external absorbers.

The extrapolation curves were obtained by a linear least square fitting, using the LINFIT [8] code, which incorporates the covariance matrix methodology and takes into account all correlations involved. The specific activity of solutions were: (509.7 ± 1.4) kBq g⁻¹ and (426.98 ± 0.96) kBq g⁻¹ for ^{51}Cr and ^{54}Mn , respectively. The main uncertainties involved in the measurement are: counting statistics, weighing, dead time, half-life and extrapolation curve efficiency. All these uncertainties are included in the fitting.

4. CONCLUSIONS

These results present overall uncertainties around 0.2-0.3 %, showing that the method developed is adequate for the standardization of these radionuclides, and can be considered satisfactory as tracer solutions and for calibrating gamma-ray spectrometers.

ACKNOWLEDGMENTS

The authors are grateful to the Radiopharmaceutical Center for supplying the ^{51}Cr solution. We are also grateful to FAPESP (The State of São Paulo Research Foundation, Brazil) and to CNPq (National Council for Scientific and Technological Development) for partial support.

REFERENCES

1. M. F. Koskinas, E. A. Silva, I. M. Yamazaki, and M. S. Dias, "Standardization of ^{241}Am solution." *Appl. Radiat. Isot.* **64**, pp. 1238-1241 (2006).
2. P.J. Campion, "Procedures for accurately diluting and dispensing radioactive solutions." *Bureau International des Poids et Mesures, Monographie BIPM – 1*, (1975).
3. A. P. Baerg, "The efficiency extrapolation method in coincidence counting." *Nuclear Instruments and Methods.* **112**, pp.143 (1973).
4. D. R. Cox, V. Isham, "A bivariate point process connected with electronic counters." *Proc. Roy. Soc. A* **356**, pp.149 (1977).
5. D. Smith, Improved correction formulae for coincidence counting. *Nucl. Instrum. Methods.* **152**, pp. 505 (1978).
6. W. O. Lavras, M. F. Koskinas, M. S. Dias, K. A. Fonseca, "Primary Standardization of ^{51}Cr Radioactive Solution." *In: V Regional Congress on Radiation Protection and Safety*, (CD ROM) 2000.
7. L.P. Moura, "Generalized coincidence method for absolute activity measurements of radionuclides. Application for determining the internal conversion coefficient of Tl-203 279-keV transition". PhD Thesis, University of Campinas (1969).
8. M.S. Dias, *LINFIT: a code for linear least square fit with covariance analysis*. Internal Report, IPEN-CNEN/SP. (1999).