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BERYLLIUM-10: HALF-LIFE AND AMS-STANDARDS*

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Absolute AMS measurements of ^{10}Be require reliable standards for calibration. Among the existing standards, rather large differences have been observed. These differences were found partially to be due to the different half-life values which were assumed. Also for comparison of AMS data with activity measurements, it is necessary to know the ^{10}Be half-life as precisely as possible. Starting with 5 ml of the standardized ORNL-MASTER solution, a working solution with a well-defined ^{10}Be content was prepared. Its specific activity was determined by liquid scintillation counting. This measurement yielded a new value of (1.52 ± 0.05) My for the ^{10}Be half-life, which is in agreement with the previously reported values but is about three times more accurate. Two independent dilution series produced new AMS standards with $^{10}Be/^{9}Be$ ratios of the order of 10^{-10} and 10^{-11}. These standards were measured at the ETH/SIN AMS facility with high accuracy and are compared with other available ^{10}Be standards.

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1. Introduction

Beryllium-10 measurements have a large number of important applications (see, for example, ref. [1], and these proceedings). The absolute determination of isotopic ratios, however, is extremely difficult and gives rise to rather large errors. Therefore the unknown samples usually are measured relative to one or more known standards. The accuracy (standard deviation of the mean value) of such a relative measurement can be made better than 1% with some effort. The absolute accuracy of the measurement is then determined (and limited) by the uncertainty in the isotopic ratio of the standards.

Up to now, no internationally acknowledged standard for ^{10}Be was available. Recently, the National Bureau of Standards, Washington D.C., has been tackling this problem [2]. Meanwhile, almost all laboratories involved in ^{10}Be studies have developed their own interim standards. As these were found to differ, their use reduces the significance of absolute $^{10}Be/^{9}Be$ isotopic ratio measurements and renders interlaboratory comparison difficult.

Because most of these standards were calibrated with activity measurements, the problem cannot be dealt with independently of the ^{10}Be half-life. Previous measurements [3,4,5] of this half-life carry uncertainties exceeding 10%.

In this work, we present a more accurate value for the ^{10}Be half-life, describe the preparation of a new series of AMS-standards, and discuss the results of our comparison between these new and other available standards.

2. Experimental procedure

2.1 Origin of the ORNL-MASTER solution

Beryllium-10 was produced by the (n,γ) reaction on ^{9}Be at the Materials Testing Reactor (MTR) of the Idaho National Engineering Laboratory (INEL). It was purified by ion exchange and converted to BeO at the Los Alamos National

Laboratory (LANL). One hundred and ten grams of BeO with 570 ppm of ^{10}Be were shipped to the Oak Ridge National Laboratory (ORNL) for electromagnetic enrichment. About 1 mg of beryllium in 4 milliliters of 1 N HCl with estimated 60 atom percent of ^{10}Be was the starting material for the standardized solution. Its isotopic composition was measured by the Secondary Ion Mass Spectrometry (SIMS) techniques [6,7] and resulted in (36.55 ± 0.35) weight percent of ^{9}Be and (63.45 ± 0.35) weight percent of ^{10}Be. Dilution with water yielded 10 ml of the so-called ORNL-A solution, of which the Be concentration was found by the isotope dilution technique to be (104.9 ± 0.6) µg/g. About half of the ORNL-A solution was mixed with 24.8 mg of gravimetrically prepared, high purity inactive beryllium and further diluted with water to yield 25 ml of the ORNL-MASTER solution with the known contents of (961.6 ± 5.6) µg/g of ^{9}Be and (13.0 ± 0.2) µg/g of ^{10}Be. Five ml of that ORNL-MASTER solution were the starting material of our redetermination of the ^{10}Be half-life and the preparation of new AMS-standards. The flow chart describing the ORNL-A and ORNL-MASTER solutions is given as fig. 1 of ref. [2].

2.2 Dilution procedure

As a first step, 5 ml of the ORNL-MASTER solution were diluted with pure water to give a working solution (W) with isotopic concentrations of (46.12 ± 0.28) µg/g of ^{9}Be and (0.6235 ± 0.0096) µg/g of ^{10}Be. Aliquots of this working solution were used for the preparation of two independent dilution series (A and B) and for the specific activity measurements. Solutions with isotopic ratios of approximately 10^{-10} and 10^{-11} were obtained by gravimetrically diluting the working solution (W) with inactive ^{9}Be solutions made from beryllium sulphate tetrahydrate ($BeSO_4 \cdot 4H_2O$) and suprapure hydrochloric acid. This dilution procedure is shown schematically in fig. 1. All solutions were stored in acid-leached polyethylene bottles. Demineralized water was used throughout.

Based on our gravimetric analysis of the ^{9}Be solutions (as BeO), a relative error of 2% is assigned to the isotopic ratios of the diluted solutions (including the uncertainity in the composition of the inactive Be-solutions and weighing errors). Thus, the nominal values of the AMS standards prepared from those solutions have the same relative error of 2%.

2.3 Specific activity measurement

The specific activity of the working solution (W) was measured by the liquid scintillation counting technique. Mixtures containing 1% aqueous solution in Optifluor [8] were prepared gravimetrically for beryllium, calibration, and background samples. Portions of 10 g of these stack solutions were put into 20 ml polyethylene vials and counted. The detector was a commerical coincidence type liquid scintillation counter (Beckman β-mate II) with logarithmic amplification. The sum output pulses were fed into the ADC of a multichannel analyzer which was gated by the coincidence signals. The observed pulse spectra were stored on magnetic tape. Typical examples (compressed to 100 channel resolution and corrected for background) of spectra are shown in fig. 2. Standard solutions of ^{32}P, ^{204}Tl, and ^{147}Pm [9] were used to estimate the absolute efficiency for detecting ^{10}Be. The results of these measurements are shown in table 1.

Linear interpolation of the absolute counting efficiency as a function of the maximum energy of the beta decay yields a counting efficiency of ^{10}Be of (94.8 ± 2.2) %. Because the counting efficiency does not vary linearly with the maximum β-energy, this value is a lower limit of the efficiency. The calculation of the specific activity is therefore based on a counting efficiency of (97.5 ± 3.0) % for ^{10}Be, which lies between that lower and the trivial 100% upper limit. Thus the specific activity of the working solution (W) was found to be (543.1 ± 16.3) Bq/g.

3. Results and discussion

3.1 Half-life

The specific activity of our working solution (W) of (543.1 ± 16.3) Bq/g, together with its ^{10}Be content of (0.6235 ± 0.0096) µg/g (see section 2.1), yields a new value for the ^{10}Be half-life of (1.52 ± 0.05) My, which is in good agreement with previous values, but is three times more accurate. We compile these values in table 2.

3.2 Comparison of AMS-standards

These measurements were performed at the ETH/SIN AMS-facility which is described in detail elsewhere [10]. The two independent dilution procedures (A and B of fig. 1) led to five new standards ($^{10}Be/^{9}Be$ ratios given in parenthesis): BSZIA0 (≈ $7x10^{-9}$), BSZIA1 and BSZIB1 (≈ $1x10^{-10}$ each), and BSZIA2 and BSZIB2 (≈ $1x10^{-11}$ each). These five and other available standards were measured with high accuracy relative to our working standard BEST433 (see table 3). This inhouse working standard was produced several years ago by the (n,γ) reaction in the reactor belonging to the Swiss Federal Institute of Reactor Research, Würenlingen. The $^{10}Be/^{9}Be$ atom ratio in this standard was estimated then to be ≈ 10^{-10} on the basis of the known production cross section.

Assuming that the nominal values of the five new BSZI are correct (see table 3), the absolute value of our working standard BEST433 was derived as (93.4 ± 2.0) x 10^{-12}. With this calibration, the absolute AMS-values in table 3 were deduced from the relative measurements. The nominal values given in table 3 for the BSZI standards are based on the ORNL SIMS measurements and our dilution procedure; the nominal values for the remaining standards are those (except for a correction for the new half-life value) supplied by the originators of the respective standards. The latter values are given in table 3 with the understanding that these are interim working values pending the development of

global standards. In particular, detailed documentation on these interim values is unavailable to us. Nevertheless, it is clear from table 3 that the investigated standards agree with the newly developed ORNL-Bern standards to within ±7%.

Nominal values are not given in table 3 for the BSNB and BSNN standards. These standards arise from a dilution of the ORNL-A solution as discussed by Inn et al. [2]. Additional cross checks concerning the isotopic ratio, Be content, and specific activity of the ORNL-A solution are currently under way. These cross checks should be completed within a year and will provide an opportunity to reevaluate the standards listed in table 3.

4. Summary

Starting with the ORNL-MASTER solution, we have prepared five new ^{10}Be standards for use in accelerator mass spectrometry. The ^{10}Be content of the ORNL-MASTER solution was determined through secondary ion mass spectroscopy and isotope dilution techniques. Based on this content, our activity measurements yielded a ^{10}Be half-life value of (1.52 ± 0.05) My. The five new standards agree within ±7% with four other existing inhouse standards. The observed agreement is quite good considering the fact the documentation of these inhouse standards is generally scant.

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[8]: Optifluor is a trademark of the Packard Instrument Company for their high flash-point liquid scintillation cocktail.

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TABLE 1: Results of activity measurements

Nuclide	E(max) (MeV)	Measured activity (Bq/g)	Absolute activity[a)] (Bq/g)	Efficiency (%)
^{147}Pm	0.224	290.5 ± 0.6	335.3 ± 4.0	86.6 ± 1.1
^{204}Tl	0.763	519.1 ± 0.9	533.7 ± 6.3	99.9 ± 2.0
^{32}P	1.711	310.1 ± 0.6	306.0 ± 6.0	101.3 ± 2.0
^{10}Be	0.555	529.5 ± 0.7	543.1 ± 16.3	97.5 ± 3.0

a) The absolute activities of ^{147}Pm, ^{204}Tl, and ^{31}P are known from the vendor's assay. The activity for ^{10}Be is calculated from the measured activity and the assumed efficiency.

TABLE 2: Reported values for the ^{10}Be half-life

Reference(s)	Half-life [My]	Uncertainty [%]
[11], [3]	1.6	a)
[12], [13]	1.7 ± 0.4	23.5
[14]	1.3	a)
[15]	1.9	a)
[3]	1.6 ± 0.2	12.5
[4]	1.5 ± 0.3	20
[5]	1.48 ± 0.15	10.1
this work	1.52 ± 0.05	3.3

a) Uncertainty not given by the authors.

TABLE 3: Atomic $^{10}Be/^{9}Be$ ratios of different standards

Sample[a]	$R=^{10}Be/^{9}Be$ R / R(BEST433)	$^{10}Be/^{9}Be$ [10^{-12}] AMS[b]	nominal[c]	AMS/nominal
BSZIA0	72.91 ± 0.38	6810 ± 151	6881 ± 176	0.990 ± 0.022
BSZIA1	1.276 ± 0.003	119.2 ± 2.5	120.4 ± 3.1	0.990 ± 0.022
BSZIA2	0.0976 ± 0.0006	9.12 ± 0.20	9.24 ± 0.24	0.987 ± 0.022
BSZIB1	1.114 ± 0.004	104.0 ± 2.3	103.1 ± 2.6	1.009 ± 0.022
BSZIB2	0.0977 ± 0.0009	9.13 ± 0.21	9.13 ± 0.23	1.000 ± 0.023
BSNB01	87.97 ± 0.28	8217 ± 173		
BSRU01	11.154 ± 0.026	1042 ± 23	1121 ± 43	0.930 ± 0.041
BSNB02	0.3315 ± 0.0021	30.97 ± 0.70		
BSNN02	0.3292 ± 0.0016	30.75 ± 0.68		
BSNI01	0.0983 ± 0.0005	9.18 ± 0.20	9.46 ± 0.31	0.970 ± 0.039
BSWA01	0.0856 ± 0.0006	8.00 ± 0.18	7.59 ± 0.25	1.054 ± 0.041
BSNI02	0.0389 ± 0.0002	3.63 ± 0.09	3.82 ± 0.13	0.950 ± 0.039

a) The origins (starting material, characterization, dilution, and final preparation) of the different standards are as follows:
BSZI: ORNL-MASTER solution, SIMS, Bern (Zimmermann), Bern (Zimmermann)
BSNB: ORNL-A solution, characterization pending, NBS (Inn), Bern (Beer)
BSRU: ICN solution, β-counting, Rutgers (Herzog), Rutgers (Herzog)
BSNN: ORNL-A solution, characterization pending, NBS (Inn), UC La Jolla (Nishiizumi)
BSNI: ICN solution, β-counting, UC La Jolla (Nishiizumi), UC La Jolla (Nishiizumi)
BSWA: Unknown origin, β-counting, New York (Wahlen), New York (Wahlen).

b) Based on an absolute value of $^{10}Be/^{9}Be=(93.4 \pm 2.0)\times10^{-12}$ for BEST433 which value, in turn, is based on the five new BSZI standards.

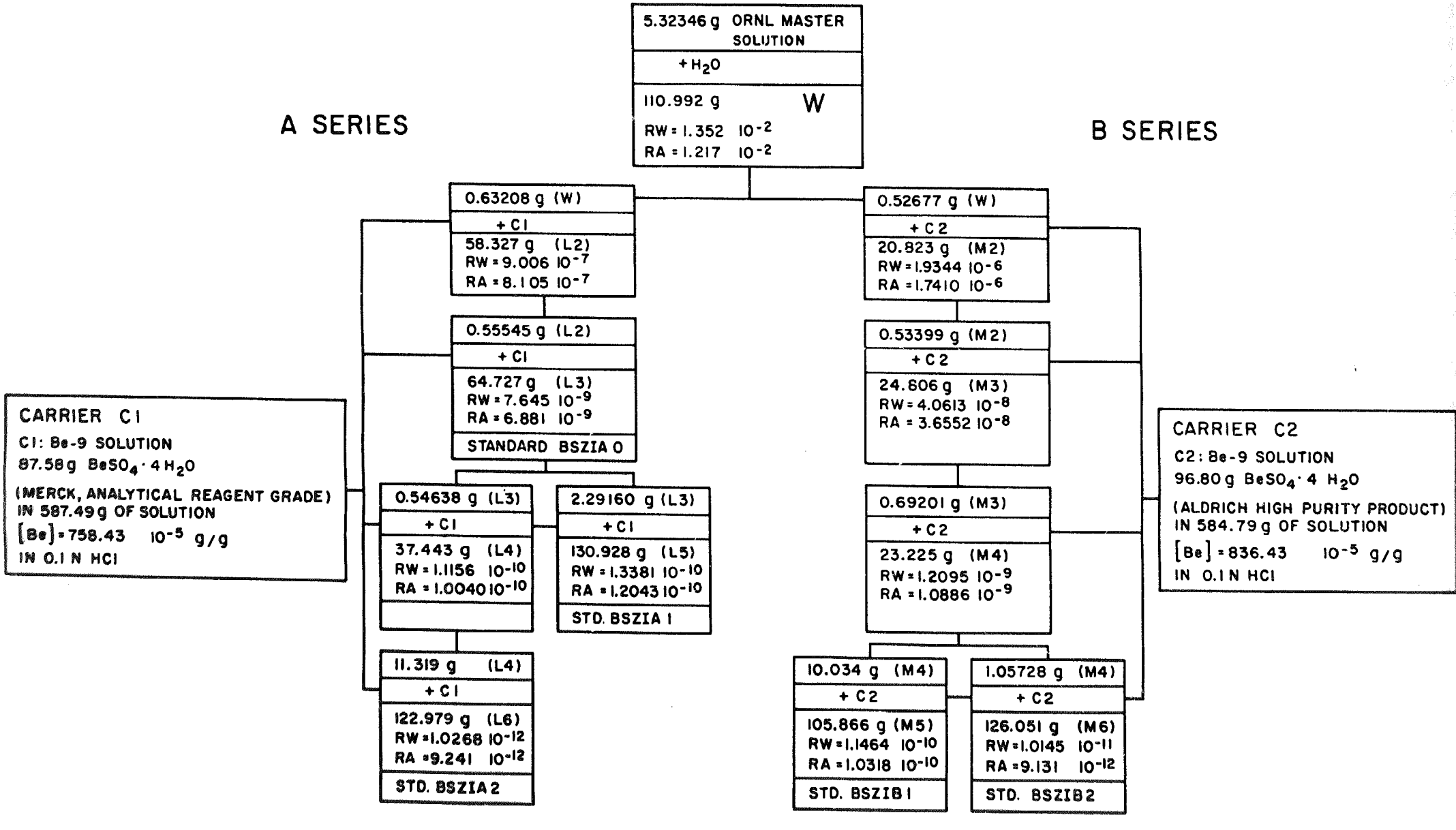
c) Value specified by the originator of the standards, but corrected if necessary, for the adopted half-life value of (1.52 ± 0.05) My.

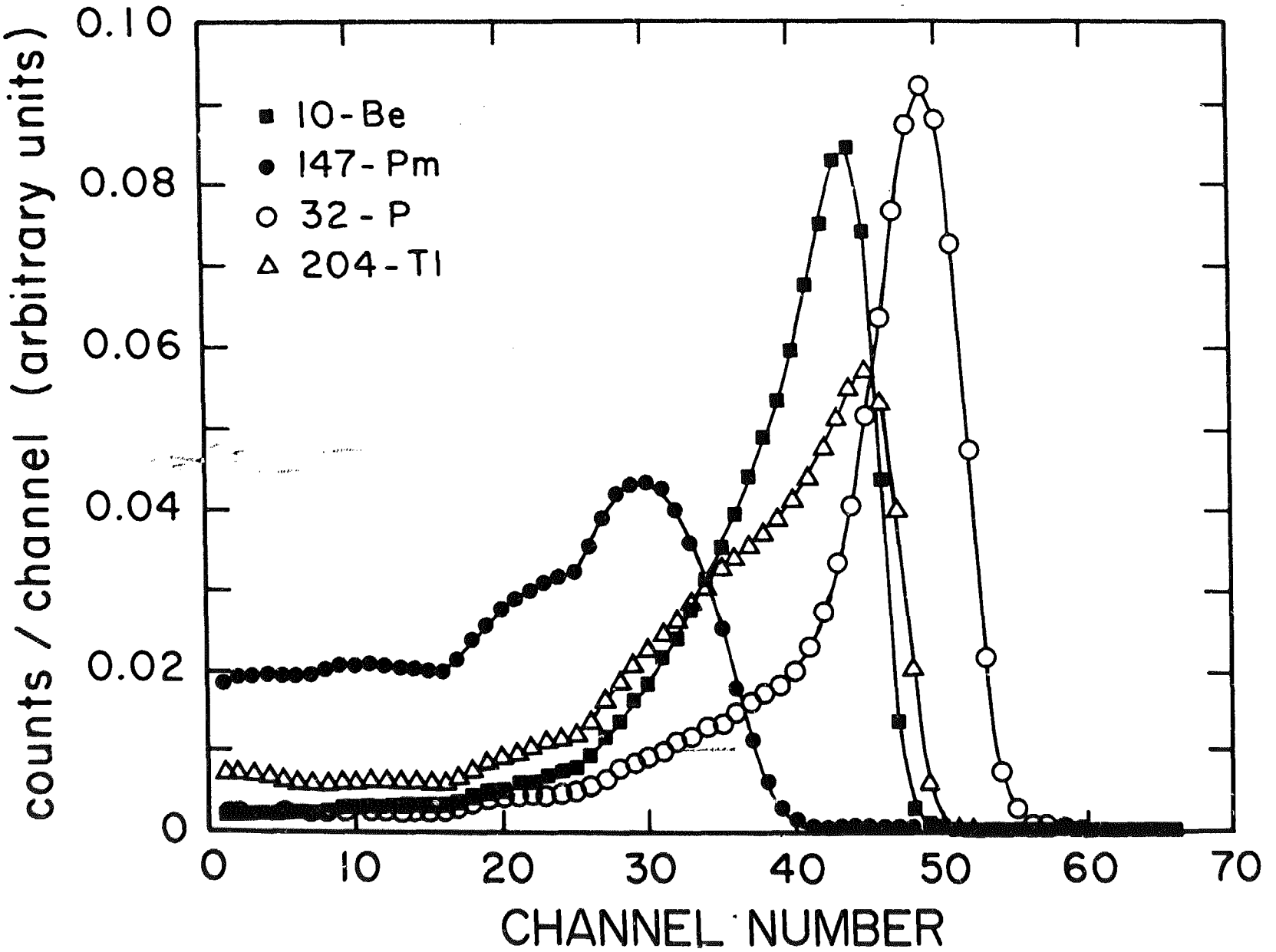
FIGURE CAPTIONS

Fig. 1: Schematic dilution procedure. The symbols RW and RA refer to the $^{10}Be/^{9}Be$ weight ratio and atomic ratio, respectively.

Fig. 2: Pulse-height spectra obtained with the liquid scintillation counter. When the channel numbers are converted to energy, the resulting scale is logarithmic.

DILUTION PROCEDURE

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counts / channel (arbitrary units)
10-Be
147-Pm
32-P
204-Tl
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CHANNEL NUMBER

DISCLAIMER