Evolution of neutron source strength measurements intercomparisons

N.N. Moiseev, I.A. Kharitonov
D.I. Mendeleyev Institute for Metrology
Saint-Petersburg, Russia

1. Introduction

Some attempts have been made to arrange international comparisons of radionuclide neutron source emission rate measurements since early 1950's.

Results of the same *Ra-Be* source emission rate measurement sequentially obtained by six laboratories in 1951 and 1954, were published in 1954 /1/. Spreading of the results was 20% in 1951, and it decreased to 10% in 1954.

In 1958, six laboratories participated in the comparisons as well, the results of which were published /2/. The spread of the results was already less than 3.5 %, while the uncertainties evaluated by the participants were from ±1.6 % to ±7 %.

The first international comparisons of radionuclide neutron source emission rate measurements, performed according to a decision of the CCRI Working Group on neutron measurements were performed in 1959 – 1965.

A *Ra-Be*(α,*n*)-source provided by the NRC (Canada) was circulated round eleven laboratories in 1959 – 1963, and results of the comparisons were published in 1966 /3/. The results showed the total spread of 4.7% and the reported uncertainties varied from ±0.9% to ±3.1%.

In 1959 – 1963, the participants of the comparisons used various methods of neutron source strength measurements. Among them, there were the Mn-bath method, gold activation in the $MnSO_4$ solution, small volume of $KMnO_4$ solution activation, integration of thermal neutrons distribution curves in various moderators (water, graphite, oil), recoil protons and associated particles counting, etc. Each of the laboratories used, at least, two different methods.

Fifteen laboratories took part in the comparisons carried out in 1979 - 1984 /4/. The aim of those comparisons was to determine of ^{252}Cf based spontaneous fission neutron source emission rate. The ASMW, BIPM, BLC, ENEA, ETL, IMM, LMRI, NBS, NPL, PTB and VGKRI measured the emission rate of a source provided by NBS and designated as SR144. The BARC, IAEB and NPL measured the emission rate of a CVN1054-source provided by NPL especially to permit the participation of China and India. To incorporate INEL measurements into the comparisons, the third source designated as NZ90 was measured by NPL.

Twelve laboratories used the Mn-bath method only, PTB used the gold foils activation method, BLC used the number of spontaneous fissions counting method, IMM used three methods, i.e. the Mn-bath, gold foils activation and associated particles counting. The spread of the results was 4.7 % (excluding the BLC result), the uncertainties evaluated by the participants were in the range from ±0.3 to ±1.3 %.

Eight laboratories participated in the CCRI(III)-K9 comparisons carried out during the period from 2000 till 2005. They were CIAE, LNE-LNHB, NIST, VNIIM, CMI, KRISS, LMRI and NPL. The main goals of the comparisons were the determination of a total emission rate and radiation asymmetry of the AMN-22 type *Am-Be*(α,*n*)-source provided by the NPL

All the eight laboratories used the same method, namely the Mn-bath method. The VNIIM additionally used the associated particles counting method. As we can see in the first Draft Report, the results spreading is 4.1% (excluding the LNE-LNHB result), while the uncertainties evaluated by the participants are in the range from ±1% to ±2.2%.

The above allows to make the following conclusions:

- results spreading keeps on being at the same level (about 4%) for fifty years and has no trends to decreasing;

- if we consider the ratios between the results of any pair of laboratories that participated in all the comparisons (for example, NPL and VNIIM), we can see that this ratio practically remains the same for fifty years; in another words, an expected closeness of results has not been achieved;

- all the many-sided measurement techniques used in previous comparisons is reduced itself to the only method, i.e. to the *Mn*-bath.

2. *Mn-bath* technique evolution

Analyzing the Mn-bath measurement procedure we can see that the difference of results can be caused by the following factors:

1. Using the wrong N_H/N_{Mn} value caused by errors in determining the $MnSO_4$ solution concentration. In the comparisons made in 1979 – 1984, this problem was solved by the following way. All the participants had to send the samples of the $MnSO_4$ solution to the BCNM and VGKRI. The results submitted by the participants were corrected taking into account the solution concentration determined by the BCMN /5/, the level of impurities determined by the BCMN, and the level of rare-earth element impurities determined by the VGKRI.

2. Errors in determining the gamma-detector efficiency

To minimize the influence of these factors in the comparisons of 1979-1984, a high-pressure ionization chamber and radium check source were sent to each laboratory. The measured Mn-bath efficiencies were corrected taking into account the information deduced from the ionization chamber measurements.

3. Errors in determining the correction for the following effects:

– neutron leakage outside the moderator;
– neutrons capture in the source material and structural materials;
– neutrons capture by the sulfur and oxygen nuclei;
– neutrons capture by the impurities nuclei.

In the comparisons made in 1979-1984, the correction presented by the participants were changed taking into account the calculation results for different solution concentrations and different bath dimensions based on the most up-to-date nuclear data /6/. An exception was the correction on neutron capture in the source material and mounting assemblies, which strongly depended on the design of a source holder including the cavity presence and dimensions. It should be remarked that no attention was drawn to the fact the leakage value depended on cavity dimensions too.

4. Difference of used values of the thermal neutron cross-section by manganese, hydrogen and gold (for gold foil activation method). In the 1979-1984 comparisons, the values presented by participants were changed on the unified values of nuclear data including the resonance capture and decay constant.

5. Measurement procedure errors. The evaluator cannot neutralize this uncertainty component.

Some results were significantly changed due to the correction (PTB - by 1.6%, LMRI - by 2.8%) nevertheless the results spreading remained at the level of 4%. This fact makes allows to conclude the main reasons of results differences are both the measurement procedures and the corrections on the neutron absorption in the source material. It seems the same reasons remains in the comparisons CCRI(III)-K9.

3. Discussion

The logic of 1979-1984 comparisons realization makes us try to standardize correction used by participants of CCRI(III)-K9 comparisons, i.e. to calculate the probability of neutron capture by Mn nuclei in the bath per neutron emitted from the source according the /7/ for all the participants. By other hand the total unification all the corrections within the only method (in our

case *Mn*-bath) is dangerous since in the long run instead of neutron source strength measuring we will measure the ^{56}Mn-solution specific activity.

If the main purpose of comparisons is to obtain the best correspondence of results rather than to obtain a true value of neutron source strength, probably it would be easier and cheaper to develop an artificial measurement standard of neutron source strength. It could be for example ^{248}Cm spontaneous fission neutron source. If the main purpose of metrology laboratories keeps in improvement of neutron sources and field characteristic measurements techniques it would be logical to promote the development of new absolute method, which is free from using any ambiguously determined parameters.

For example, using the "absorber in an absorber" method (gold activation in $MnSO_4$ solution), we need to measure the counting rate only (certainly, with correspondence corrections on dead-time and resolution time of measuring system).

1. D.Huges. Nuclonics, No12, 26 (1954)
2. K.Larsson. J.Nucl. Energy, 6, 322 (1958)
3. V.Nagguar. Recueil de Travaux du BIPM.Vol. 1, 1966-1967.
4. Metrologia, 23, 129-144 (1986/1987)
5. BCMN Report GE/R/AS/08/84 (1984)
6. Chen Lin. Thesis, Columbia University, New York, January 1982.
7. Briesmeister J F, Ed., MCNP – A General Monte Carlo N-Particle Transport Code, Version 4C (LA-13709-M), 2000