Report of the CCRI(II) Transfer Instrument Working Group

2005 - 2007

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The SIR Transfer Instrument (TI) project for measuring short-lived radionuclides was proposed at the 2005 CCRI(II) meeting and is described in document CCRI(II)/05-08. The TIWG used this document as a starting point and met in November 2005 to discuss the experimental detail and the design of the system. No further meeting was organized because communication by e-mail was found to be sufficient.

1. Summary of decisions made at the WG meeting in November 2005

Although other setups were discussed, the working group agreed to continue the project along the lines defined in the CCRI(II) proposal: well-type NaI(Tl) (3” × 3”, well diameter of 2 cm) and use of a $^{93}\mathrm{Nb}^{\mathrm{m}}/^{94}\mathrm{Nb}$ source (irradiated wire from the IRMM) on the one hand to define the electronic threshold (using the $^{93}\mathrm{Nb}^{\mathrm{m}}$ x-ray emission) and on the other hand for long term stability checks (enabled by the long half-life of $^{94}\mathrm{Nb}$). Each short-lived radionuclide measurement would be relative to a Nb measurement and equivalent activities A_{E} would be defined similarly to those of the SIR.

To measure the counting rate, a live-time NIM module and scalers were identified as being more reliable than using a MCA with its internally provided live time. Nevertheless, it was decided that an appropriate limiting activity would be recommended in the comparison protocol. In addition, due to decay during measurement, the counting intervals should also be time-limited.

The need for lead shielding was discussed to reduce the background and backscattering from the surroundings but it was rejected, initially because it would make the transport of the whole system more difficult.

The LNE-LNHB volunteered to simulate the detector using Monte Carlo simulations in order to obtain the efficiency curve to enable the calculation of impurity corrections.

It was agreed to test the whole system with $^{99}\mathrm{Tc}^{\mathrm{m}}$ solutions from the LNE-LNHB and the NPL. The measurement of the same solutions in the SIR would enable the linking factor $A_{\mathrm{e}} / A_{\mathrm{E}}$ to be calculated, which is necessary to link the future $^{99}\mathrm{Tc}^{\mathrm{m}}$ TI measurements to the SIR. The TI measurement would follow the SIR measurement after waiting for several half-lives so that

the ^{99}Tc^{m} activity is sufficiently reduced. The relative uncertainty on the decay correction factor would remain below 10^{-3} and so would not be a limiting factor.
In order to limit the influence of the chemistry of the solutions on the results, the participants agreed that the use of NaCl in water should be recommended in the protocol for ^{99}Tc^{m}.

Other radionuclides were considered as other good candidates for the TI system (^{18}F, ^{11}C and ^{56}Mn) and the participants recommended including them in the TI programme as soon as possible.

The BIPM asked the participants to consider sending secondees to work on the project at the BIPM for a few months.

2. Additional decisions made after the 2005 meeting

When making the long term stability check measurements with the Nb source, it was noted that the ^{93}Nb^{m} x-rays contribute significantly to the total counting rate and this would need a correction factor that changes with time (^{93}Nb^{m} half-life is ca 16 a). In order to eliminate this problem, it was agreed to use a 1 mm thick brass liner in the NaI well. The chemical composition of the brass was selected carefully to avoid high Z elements that could emit high energy fluorescence x-rays.

It was noted that for the ^{99}Tc^{m} measurements, the ^{99}Tc^{m} x-ray peak (18 keV) is close to the threshold adjusted at ca 17 keV using the ^{93}Nb^{m} x-ray peak. Consequently, any small drift in the electronics causing a change of threshold position by a few channels could have a significant effect on the counting rate. It was then agreed to use the same brass liner described above also for the ^{99}Tc^{m} measurements.

The LNE-LNHB proposed the use of their MTR2 live-time NIM module which is commercially available (although momentarily out of stock). The working group accepted this proposal and the LNE-LNHB donated one module to the BIPM.

Once the detector and electronics were available at the BIPM, the background counting rate was measured as ca 300 s^{-1}. This was considered too high as it would limit the dynamic range of the system. A cylindrical shielding was designed at the BIPM, made of 5 mm of Pb, 3 mm of Sn and 2 mm of Cu, with an external protective layer of PVC, for a total mass of 9.5 kg. The shielding reduced the background rate to ca 65 s^{-1} and this was deemed acceptable (see Figure 1). No Pb or Sn x-ray fluorescence could be identified in the NaI energy spectra.

3. Developments and tests of the SIR TI at the BIPM

The LNE-LNHB obtained an efficiency curve of the SIR TI by Monte-Carlo simulation using PENELOPE and produced a detailed report (Note Technique LNHB 2006/48). However, the brass liner and the lead shielding were subsequently added to the design of the system and consequently, the simulation now needs to be updated.

A plexiglass holder for the Nb wire was discussed and designed in collaboration with the IRMM. The holder is dismountable so that the source could be used in another holder. Several such holders were produced at the BIPM workshop and were sent to IRMM for source insertion. Two Nb sources in these holders are now available at the BIPM. They were measured in the NaI well, initially producing a counting rate of ca 12 000 s^{-1} which reduced to 8 500 s^{-1} with the brass liner. The liner appeared to be efficient in absorbing all $^{93}Nb^{m}$ x-rays (see Fig. 2a and 2b).

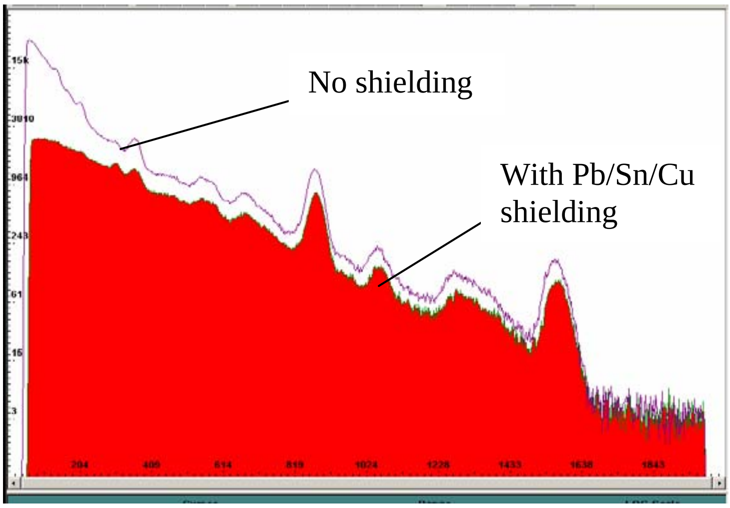

Figure 1. Background energy spectrum of the SIR TI with and without shielding.

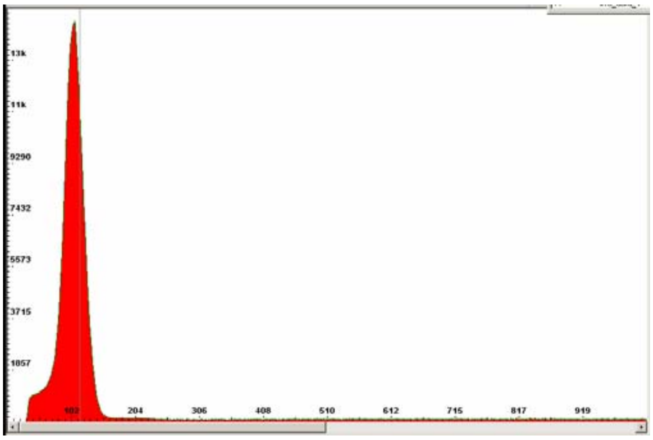

Figure 2a. Low-energy spectrum of the Nb source in the NaI well. The spectrum is dominated by the $^{93}Nb^{m}$ x-ray peak.

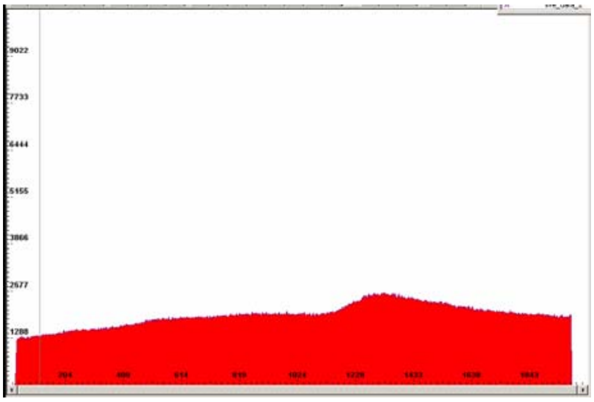

Figure 2b. Low-energy spectrum of the Nb source in the NaI well with the brass liner that absorbs the ^{93}Nb^{m} x-rays. The Compton continuum is from the ^{94}Nb γ- rays.

The electronic scheme is shown in Figure 3. The short shaping time of 0.5 µs at the amplifier was selected for better performance at high counting rate. This shaping only slightly alters the energy resolution of the system. The FWHM is 6.5 % at 662 keV, 9.7 % at 122 keV and 25 % at 22 keV.

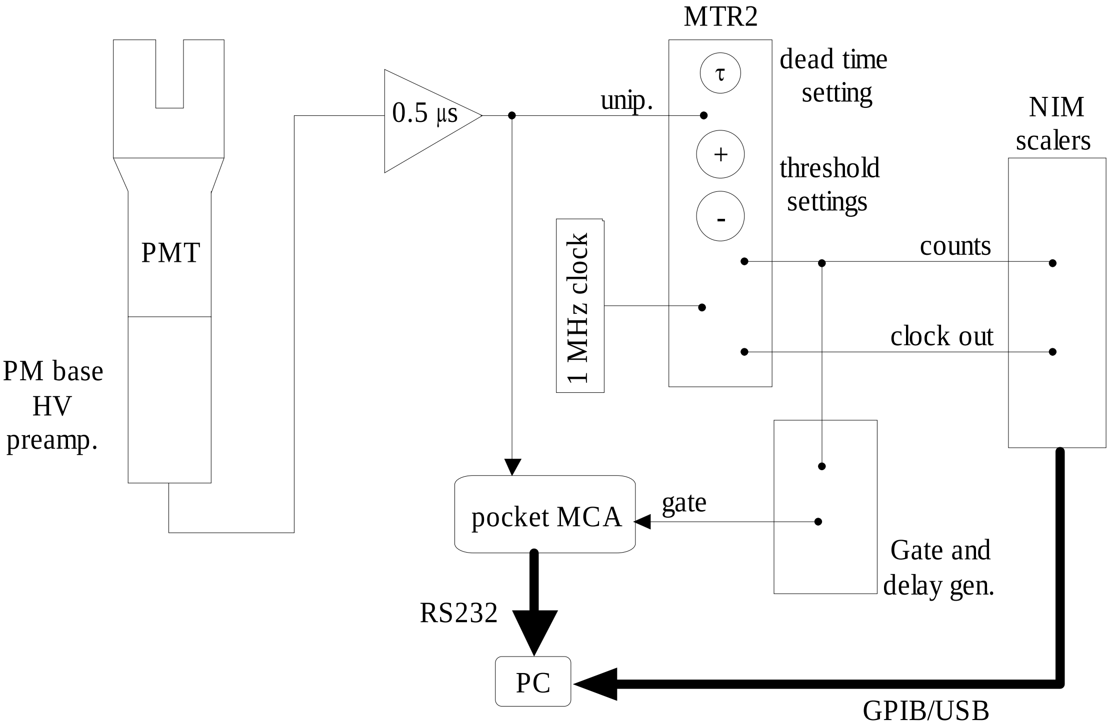

Figure 3. Electronic scheme of the SIR TI

The scalers are read through a GPIB/USB controller under LabView. The measurement duration is controlled internally by the scaler time base (quartz). Overflow counting is managed by software in LabView. The scalers and their readings have been tested extensively using a 10 MHz frequency originating from an atomic clock. The observed 10 MHz readings differ from the atomic frequency by 8×10^{-6} as a maximum (in relative terms), which complies with the typical frequency precision and stability of a quartz.

A pocket clock of 1 MHz frequency was produced at the BIPM. The frequency was selected to be at least ten times the maximum NaI counting rate. This ensures a negligible contribution from the clock frequency on the live time correction uncertainty (Baerg *et al.*, 1976). The short term (15 min) stability of the frequency is better than 3×10^{-7}. The long term stability (over 5 weeks) observed to date is about 5×10^{-6}.

The response of the amplifier to highly saturating pulses was observed using the oscilloscope: the pulse width is enlarged and followed by a long undershoot. In order to have these long periods covered by the live-time correction, an extending dead time of 30 µs is necessary for the MTR2.

The stability of the gain of the electronic chain was verified: peak shifts by a few 10^{-3} are observed over 24 h periods. The influence of such shifts on the counting rate was quantified by measuring the counting rate as a function of the threshold position. The results are sensitivity coefficients of 8.8×10^{-5} per channel for ^{94}Nb and $< 10^{-5}$ per channel for ^{57}Co. This means that for the ^{94}Nb measurements, a change of threshold position by a few channels can produce a non-negligible effect. It is thus important to check the threshold position (using the ^{93}Nb^{m} x-ray peak) before each ^{94}Nb measurement. On the other hand, for a radionuclide like ^{57}Co (and ^{99}Tc^{m} with the liner) where most counts are concentrated in the full-energy peak, the threshold position is not crucial and gain shifts during series of measurements should not affect the results. This study was carried out in a room where temperature and humidity are controlled. Tests should perhaps be made to verify the behaviour of the system in a non-controlled environment.

The behaviour of the SIR TI at high counting rate was studied. A shift of the peak position versus rate was observed (see Figure 4). The reason for this effect has not been clearly identified. A similar effect was observed when using a high-performance spectroscopy amplifier. Consequently, it is advisable to limit the counting rate of future measurements to 75 000 s^{-1}.

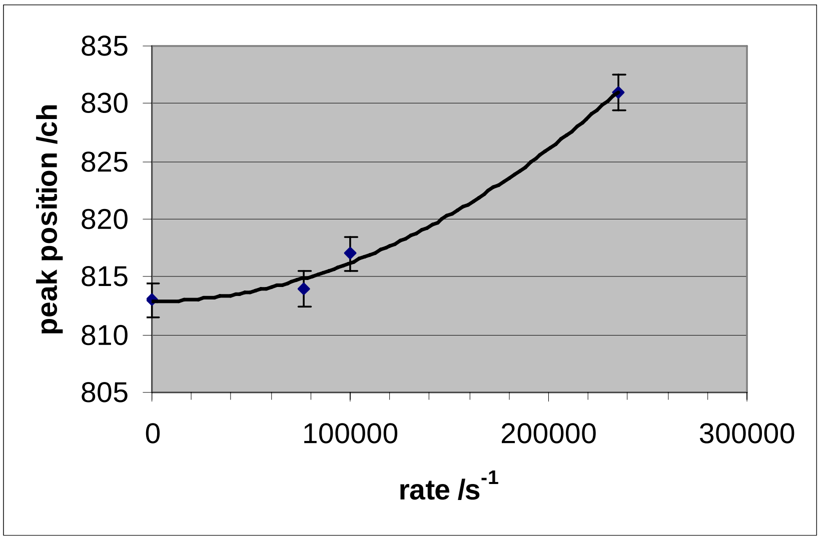

Figure 4. ^{57}Co peak position versus counting rate corrected for live time.

Series of tests have enabled the following preliminary uncertainty budget to be deduced for a ^{94}Nb measurement in the SIR TI:

Uncertainty source	**Standard uncertainty / 10^{-2}**
Reproducibility *	< 0.02
Backscattering from surroundings	< 0.01
Clock frequency *	0.001
Decay correction for the next 20 years	< 0.01
Statistical uncertainty for a 2 h measurement	0.05

* in a room with air conditioning

A suitcase for the air transport of the SIR TI as hand luggage was purchased and the internal layout is in preparation. The lead shielding should be shipped to the participant NMI separately, well in advance. The BIPM proposes to produce a series of identical shielding cylinders so that each participating NMI would keep one such shield on permanent loan in preparation for future participations in the SIR TI comparisons.

4. First measurement of ^{99}Tc^{m} in the SIR TI at the BIPM

When measuring a short-lived radionuclide, the well-known correction for decay during measurement should be applied. However, it is necessary also to consider the effect of decay on the live-time correction, which in theory is deduced for a constant count rate. This has been studied in detail by (Chauvenet, 1990) where a correction formula in agreement with an approximation deduced by (Baerg *et al.*, 1976) is given. Using this study, maximum measurement durations were deduced for ^{99}Tc^{m} such that the correction factor is lower than 10^{-4} and thus can be neglected. Examples of maximum durations are 600 s for a true rate of 10^{5} s^{-1} and 2000 s for a true rate of 10^{4} s^{-1}.

Two ^{99}Tc^{m} ampoules were prepared at the LNE-LNHB and submitted to the SIR on the 6 March 2007. The ampoules were measured 5 times in the SIR over 48 h. One ampoule was measured with the BIPM Ge(Li) to quantify the ^{99}Mo activity. The measurement of the two ampoules in the SIR TI started on the 8 March and the decay was followed for 72 h. Figure 5 shows a ^{99}Tc^{m} energy spectrum measured with the NaI.

The objectives of this first measurement were to verify the linearity of the SIR TI versus count rate and to determine the maximum ^{99}Tc^{m} activity acceptable for future measurements. A further objective is to evaluate the linking factor to the SIR. This data analysis is in hand.

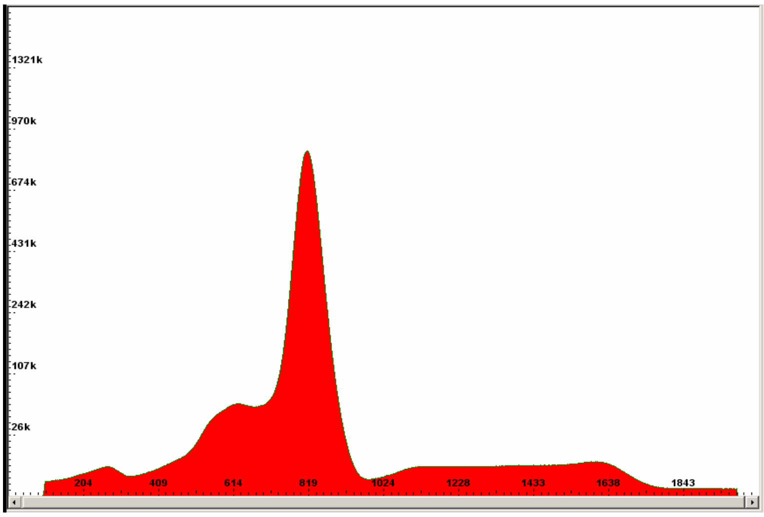

Figure 5. Energy spectrum (NB: sqrt vertical scale) of ^{99}Tc^{m} in the NaI well with the brass liner. The true counting rate is ca 35 000 s^{-1}.

5. Future plans

Some of the tests described above will need to be repeated with the other short-lived radionuclides proposed to be used in the SIR TI.

The Monte Carlo simulation of the NaI should be updated to include the brass liner and the external shielding. This would then enable the response of the SIR TI to ^{99}Mo to be evaluated as this is needed for the analysis of any ^{99}Tc^{m} measurement. Another possibility could be for the BIPM to purchase a ^{99}Mo solution and carry out a calibration of the SIR TI using the SIR as the reference.

Studies of the influence on the SIR TI response of the filling height of the ampoule, the solution density and the ampoule wall thickness should also be made.

References

Baerg A.P. *et al*., 1976, Live-Timed Anti-Coincidence Counting with Extending Dead-Time Circuitry, *Metrologia* **12**, 77-80.

Chauvenet B., 1990, Utilisation du temps actif pour la mesure d'un radionucléide de courte période avec temps mort cumulative, Rapport interne LPRI/90/101/Novembre, CEA Saclay.