# Neutron Activation as an Independent Indicator of Expected Total Yield in the Production of $^{82}Sr$ and $^{68}Ge$ with 66 MeV Protons

C. Vermeulen[a], G.F. Steyn[b,1], N.P. van der Meulen[a]

[a]*Laboratory of Radiochemistry and Environmental Chemistry, Paul Scherrer Institute, Villigen-PSI, SWITZERLAND*
[b]*Department of Nuclear Physics, iThemba LABS, Old Faure Road, P.O. Box 722, Somerset West 7129, SOUTH AFRICA*

## Introduction

A method based on neutron activation is being developed to assist in resolving discrepancies between the expected yield and actual yield of radionuclides produced with the vertical-beam target station (VBTS) at iThemba LABS.

The VBTS is routinely employed for multi-Ci batch productions of the radionuclide pairs $^{22}Na/^{68}Ga$ and $^{82}Sr/^{68}Ga$ using standardized $^{nat}Mg/^{nat}Ga$ and $^{nat}Rb/^{nat}Ga$ tandem targets, respectively [1]. The metal-clad target discs are bombarded with a primary beam of 66 MeV protons at an intensity of nominally 250 µA. The encapsulation materials are either Nb (for Mg and Ga) or stainless steel (for Rb) which serve to contain the molten target materials during bombardment and act as a barrier to the high-velocity cooling water which surrounds the targets in a 4π geometry. The $^{nat}Rb/^{nat}Ga$ targets are typically bombarded according to a two-week cycle while $^{nat}Mg/^{nat}Ga$ targets are bombarded on an ad-hoc basis, depending on a somewhat unpredictable $^{22}Na$ demand.

A too-large deviation between expected yield and actual yield has at times plagued this programme. These deviations can manifest both as an apparent loss or an apparent gain (relative to the expected yield) by up to about 15% in either direction. The resulting uncertainty of up to 30% (in the worst case) from one production batch to another can be costly and is unacceptable in a large-scale production regimen. This phenomenon is believed to be brought about by two types of problems:

(1) Production losses, e.g. during the radiochemical separation process or incomplete recovery of activated target material during the decapsulation step.

(2) Incorrect values obtained for the accumulated proton charge.

A problem of type (1) will always result in a loss of yield. A problem of type (2) can manifest as an apparent loss or gain. In an effort to get a handle on this second type of problem, neutron activation of suitable material samples, embedded in a target holder, is being investigated as an independent indicator of the total yield. For this purpose, samples of Co, Mn, Ni and Zn were activated during production runs and Co was found to be the most appropriate. Preliminary results will be presented after first discussing why the determination of the accumulated proton charge is a problem with the VBTS.

## Materials and Methods

The VBTS consists of a central region in which a target holder is located during bombardment as well as two half-cylindrical radiation shields which completely surround the target. The shields can be moved away from the central region on dedicated rails, e.g. when repairs or maintenance is required. Figure 1 shows the VBTS with the shields moved to the "open" position. As some components of the station are located below the vault floor, with the target position near floor level, it proved difficult to electrically isolate the VBTS as was done for the two horizontal-beam target stations at iThemba LABS [1]. The VBTS does not act as a Faraday cup like the other target stations. Instead, the beam current and accumulated charge is measured by means of a calibrated capacitive probe [1,2].

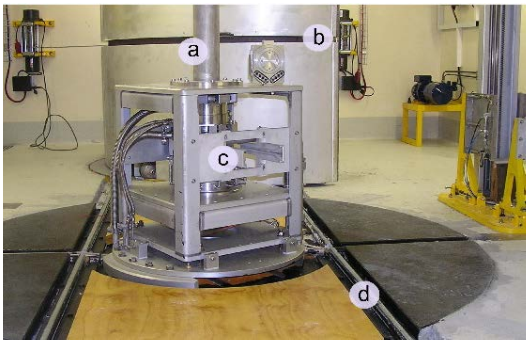


FIGURE 1. VBTS with radiation shields in the "open" position, showing (a) the vertical beamline, (b) one of the two half-cylindrical shields, (c) the target position in the central region, and (d) the rails on which the shields can move to gain access to the central region.

There appears to be a variation in the response of the capacitive probe, sensitive to the beam microstructure, in particular a dependence on the beam packet length. This problem is not yet fully resolved.

[1]Corresponding author, E-mail: deon@tlabs.ac.za

Figure 2 (a) shows the beamstop of a VBTS target holder with several Co samples mounted on the outside as well as one each of Ni, Mn and Zn. The samples are small "tablets" with a 10 mm diameter and 1 mm thickness. The reactions of interest are $^{59}Co(n,\gamma)^{60}Co$, $^{59}Co(n,3n)^{57}Co$, $^{nat}Ni(n,X)^{60}Co$, $^{nat}Ni(n,X)^{57}Co$, $^{nat}Zn(n,X)^{65}Zn$ and $^{55}Mn(n,2n)^{54}Mn$. The relevant half-lives are $^{60}Co$(5.271 a), $^{57}Co$(271.8 d), $^{65}Zn$(244.3 d) and $^{54}Mn$(312.2 d). The half-life should be long compared to the two-week cycle in order to reduce the dependence on the exact beam history, which is very fragmented over any production period. In this respect, $^{60}Co$ is considered to be particularly attractive as its long half-life of more than 5 years leads to a negligible effect by the beam history.

Note that the tandem targets, shown in Figure 2 (b), are mounted just upstream of the beamstop – in fact, the targets and beamstop form a single unit before being fitted into the target holder.

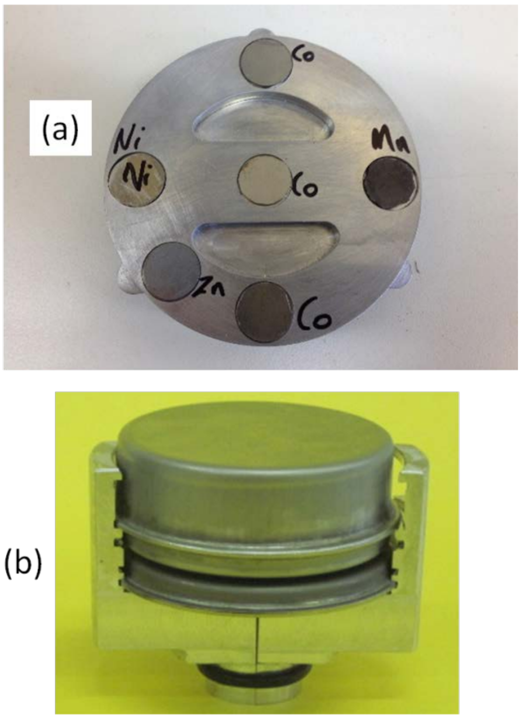


FIGURE 2. (a) Experimental VBTS target-holder beamstop showing the embedded Ni, Co, Mn and Zn samples to be neutron activated in a standard production run. (b) A Rb/Ga tandem target assembly which, together with the beamstop in (a), forms a single unit once assembled.

At the end of bombardment, all samples were assayed for their characteristic γ-emissions using standard off-line γ-ray spectrometry with an HPGe detector connected to a Genie 2000 MCA. Calculations of the neutron fluence density in the central sample volume on the beamstop were also performed using the Monte Carlo radiation transport code MCNPX. For these calculations, the entire VBTS, a Rb/Ga target and the vault walls were included in the model.

## Results and Conclusion

All samples activated significantly – copious amounts of $^{60}Co$ were detected in the Co discs after a two-week run.

The neutron fluence density for the case of a 250 μA, 66 MeV proton beam on a $^{nat}Rb/^{nat}Ga$ tandem target is shown in Figure 3. The dominance of low-energy neutrons is evident, which is in part due to the large amount of paraffin-wax shielding material in close proximity to the target. While reactions such as the (n,2n) and (n,3n) would be sensitive to the more energetic part of the neutron spectrum, the (n,γ) capture reaction benefits from the large low-energy component. This explains the copious amounts of $^{60}Co$ formed. It was therefore decided to only retain the central Co sample for subsequent bombardments, as shown in Figure 4.

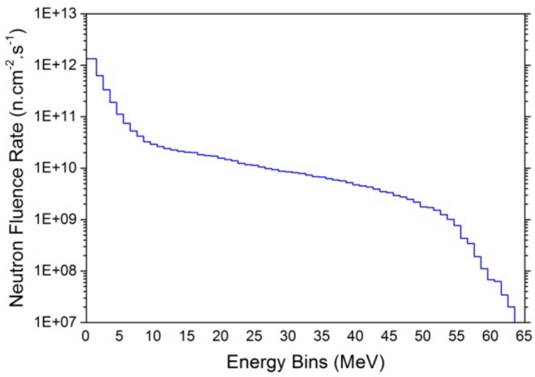


FIGURE 3. Neutron energy spectrum inside the central Co disc as calculated with MCNPX.

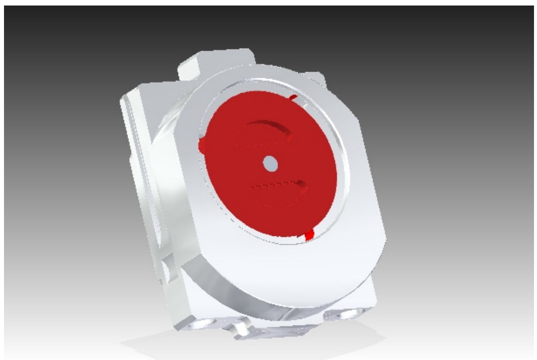

FIGURE 4. VBTS target holder. The beamstop (in red) shown with a centrally mounted Co disc.

The first results are shown in Table 1. The accumulated charge as obtained from the capaci-

[1]Corresponding author, E-mail: deon@tlabs.ac.za

tive probe ($Q$), the specific $^{60}$Co activity ($A$) at the end of bombardment (EOB), and their ratio ($A/Q$) are presented in the table, together with the deviation of individual ratios relative to their average for the case of the Mg/Ga tandem targets only. Note that all samples were counted until the statistical uncertainties were negligible. Any systematic uncertainties are ignored at this stage as they are considered to remain the same from one batch production to another.

| Target | $Q$ (mAh) | $A[^{60}$Co] (Bq/mg) | $A/Q$ | Deviation (%) |
|---|---|---|---|---|
| Rb/Ga | 20.01 | 590.09 | 29.49 | |
| Mg/Ga | 5.001 | 167.46 | 33.49 | +11.97 |
| Mg/Ga | 20.00 | 547.56 | 27.38 | -8.46 |
| Mg/Ga | 20.15 | 616.66 | 30.60 | +2.31 |
| Mg/Ga | 20.02 | 563.51 | 28.15 | -5.88 |
| Average | | | 29.91 | |

TABLE 1. Results from neutron-activated Co discs, based on $^{60}$Co activity measurements.

For the sake of argument, the average value of the ratio is taken as the expected value. A positive deviation of the $A/Q$ value is then indicative of a too-small value of the accumulated charge obtained from the capacitive probe, leading to a corresponding overproduction. Likewise, a negative value is indicative of a too-large value of the accumulated charge, leading to a corresponding underproduction.

It is certainly true that the data in Table 1 are currently very limited. It is envisaged, however, that with time the growing database of values will assist in reducing the uncertainty in determining the accumulated charge and reduce the discrepancies between predicted and actual yields significantly. Table 1 illuminates the underlying problem satisfactorily. The four Mg/Ga tandem target bombardments, on identical targetry, were performed successively. The neutron activation correlates well the with actual yields, pointing directly to the current integration as the main source of error.

The method already proves to be useful. An indication of an over or underprediction can be obtained prior to the target processing by recovering and measuring the Co disc. This information can be used to make a decision concerning the present batch production and/or the subsequent one. One can either add beam to the present production target and/or increase/reduce the total beam on the subsequent production target to compensate for an expected overproduction or shortfall.

In conclusion, we would like to stress that the capacitive probes show great promise and that better understanding and/or possibly some development of their signal processing algorithm may improve their ability to measure the accumulated charge to the desired accuracy. Segmented capacitive probes used at iThemba LABS and elsewhere for beam position measurement [1,3] are not affected by beam microstructure as only the ratios of the signal strengths on the different sectors are important. In this case, changes in response affect all sectors equally and the ratios are unaffected.

## References


1. G. F. Steyn et al., A vertical-beam target station and high-power targetry for the cyclotron production of radionuclides with medium energy protons. *Nucl. Instr. and Meth. A* **727,** pp. 131–144, 2013.
2. Z. Kormány et al., *Development of non-destructive beam current measurement for the iThemba LABS cyclotrons.* In: Proceedings of the 11th European Particle Accelerator Conference (EPAC 2008), pp. 1089–1091, 2008. Paper available at http://accelconf.web.cern.ch/AccelConf/e08/index.htm.
3. J. Dietrich et al., *Beam position monitor development for iThemba LABS cyclotron beamlines.* In: Proceedings of the 9th European Particle Accelerator Conference (EPAC 2004), pp. 2589–2591, 2004. Paper available at http://accelconf.web.cern.ch/Accelconf/e04/default.htm.


## Acknowledgements


The authors would like to thank the staff of the Cyclotron Department of iThemba LABS involved in the development of the capacitive probes. This work is based on research supported by the National Research Foundation of South Africa (UID 85507). The grant holder acknowledges that opinions, findings and conclusions or recommendations expressed in any publication generated with NRF supported research are that of the authors, and that the NRF accepts no liability whatsoever in this regard.



[1]Corresponding author, E-mail: deon@tlabs.ac.za