# Proton Activation Analysis for Tracer Kinetic Studies with Stable Isotopes of Zirconium

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## 1. Introduction

In internal dosimetry, biokinetic models are used to assess doses after intakes of radioactive material. Model parameters must be measured directly or estimated from animal experiments or other sources of data. The quality of such estimates depends on the often unknown similarity between animal and human metabolism.

Enriched stable isotopes, which are chemically similar to radioisotopes of the same element, can be used as tracers to investigate the uptake, distribution, retention and excretion of an element without the risk of ionizing radiation. By administering tracers orally and intraveneously, valid data from human subjects can be obtained to verifiy or improve biokinetic models. Measurement methods for body fluid samples must be capable of quantifying isotope concentrations as low as a few ng $ml^{-1}$.

Zirconium is an interesting element for such studies because the parameters of its biokinetic model as proposed by the International Commission on Radiological Protection (ICRP) are based on only few experiments with rodents and one single human radiotracer experiment. Radionuclides of zirconium are produced by uranium fission as well as by neutron activation of stable zirconium used e.g. in uranium fuel rod cladding. They may be accidentally incorporated both in regular nuclear technology or in large-scale accidents.

## 2. Method

Proton activation analysis (PNA) and thermal ionization mass spectrometry (TIMS) were selected as suitable measurement methods for the tracer experiments performed at the GSF. The current setup for PNA was used earlier in measurements at the Paul Scherrer institute in Villigen, Switzerland. After reconfiguration at the MLL, four irradiations were performed in 2006.

| Reaction | Product half-life | Main $\gamma$ energy |
|---|---|---|
| $^{90}$Zr(p,n)$^{90}$Nb | 14.6 h | 141, 1129 keV |
| $^{96}$Zr(p,2n)$^{95}$Nb | 35.0 d | 766 keV |
| $^{51}$V(p,n)$^{51}$Cr | 27.7 d | 320 keV |

Table 1: Reactions used for tracer quantification

Dried samples were pressed in tablet form, encased in aluminum plates, and mounted on a disc, which is rotated at approximately 70 rpm and cooled during irradiation to ensure uniform activation without sample loss. Samples were irradiated by 20 MeV protons with a beam current of up to 2.5 $\mu$A for up to 48 h. Activated samples were removed 8 h after end of irradiation and transported to the GSF. After removal of the aluminum shielding, activation product activity was measured by gamma spectrometry and compared to co-irradiated standard samples by means of $^{51}$V as internal standard.

## 3. Results

Apart from delivering data from tracer experiments as shown in the figures below, PNA can serve as an efficient means of quality control for TIMS since both methods have different sources of errors. The deviations between TIMS and PNA data shown in figure 1 between 0 and 200 minutes together with further TIMS reference measurements led to a substantial reduction of memory effects in sample preparation for TIMS. TIMS is now used as routine measurement method due to higher sample throughput and smaller uncertainties, with ongoing cross-checks of selected tracer experiments by PNA. Future irradiations will also include ruthenium tracer measurements.

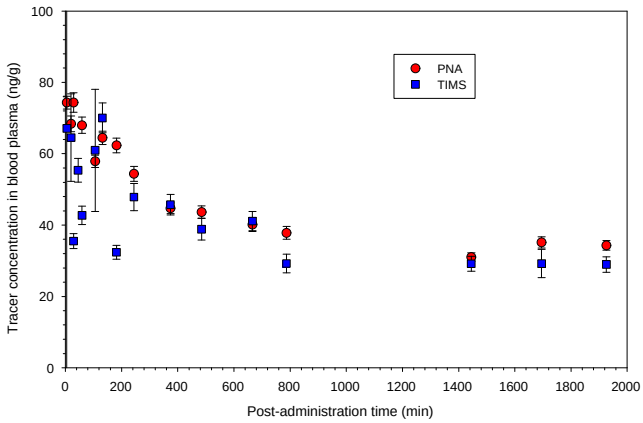


Fig. 1: Concentrations of intraveneous tracer (97.7 % $^{90}$Zr)

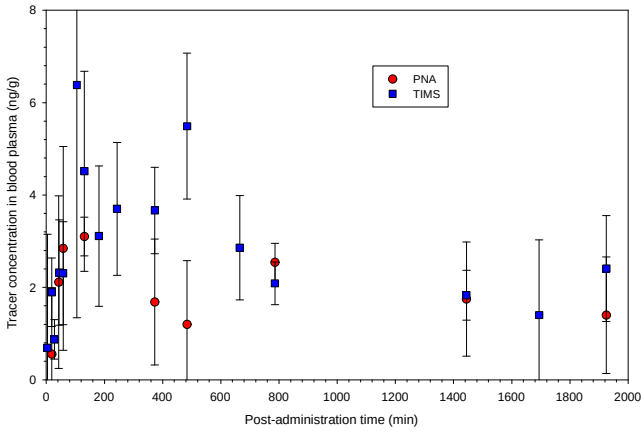


Fig. 2: Concentrations of oral tracer (86.4 % $^{96}$Zr)