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

A. Giussani [a], M. C. Cantone [b], W. Carli, U. Gerstmann [a], M. Greiter [a], R. Hertenberger, V. Höllriegl [a], K. Leopold [a], U. Oeh [a], I. Veronese [b], and H. G. Paretzke [a]

[a] Helmholtz Zentrum München, Dt. Forschungszentrum f. Gesundheit u. Umwelt, 85764 Neuherberg
[b] Università degli Studi di Milano and INFN, Sezione di Milano, 20133 Milano (Italy)

## 1. Introduction

In internal dosimetry, biokinetic models are used to assess doses after intakes of radioactive material. Ruthenium is an important element in radiation protection because the radioisotopes $^{103}$Ru (half-life: 38.3 d) and $^{106}$Ru (half-life: 374 d) are produced by uranium fission and may be released in significant amounts in the environment in case of accidents. The biokinetic model proposed by the International Commission on Radiological Protection (ICRP) is however based on only few animal data and one single human study with administration of $^{103}$Ru in different chemical forms.

Stable isotopes can be used as tracers to investigate the uptake, distribution, retention and excretion of an element directly in humans, without exposing the investigated subjects to the risk of ionizing radiation. By administering tracers orally and intravenously, valid data from human subjects can be obtained to verify or improve biokinetic models. Measurement methods for body fluid samples must be capable of quantifying isotope concentrations as low as a few ng·ml$^{-1}$.

## 2. Method

Tracer kinetic studies using solutions isotopically enriched either in $^{99}$Ru or in $^{101}$Ru were performed at GSF (now Helmholtz Zentrum München) according to the protocol approved by the Ethical Committee of the TU München. Activation analysis with protons was used in order to detect the single isotopes in the blood and urine samples collected at scheduled time during the studies. The biological samples had to be properly treated, in case of urine samples including a complex wet pressure digestion procedure, in order to obtain solid tablets which can be encased in aluminium supports and mounted in the irradiation chamber. Up to 40 samples can be fixed onto a disc, which is rotated at a speed of approximately 70 rpm, in order to enable the simultaneous activation of the samples under the same experimental conditions.

Three runs of irradiation were conducted in 2007, including some tests for defining the best experimental conditions for activation of the stable isotopes of ruthenium. Samples were irradiated using 20 MeV protons with a beam current of up to 2.5 $\mu$A for up to 72 h. Activated samples were removed 8 h after the end of irradiation and transported to the GSF. After removal of the aluminium shielding, the activation product activities in the sample were measured by gamma spectrometry using HPGe detectors. Quantitative determination of the isotopes in the sample was achieved by comparison with the activities in standard samples, containing known amounts of the isotopes of interest.

## 3. Results

Table 1 shows the reactions chosen for the tracer quantification, on the basis of the preliminary tests conducted. Vanadium-51 is the isotope which is added to each sample as internal spike.

| Reaction | Product half-life | $\gamma$-lines employed |
|---|---|---|
| $^{99}$Ru(p,n)$^{99}$Rh | 16.1 d | 90, 528 keV |
| $^{101}$Ru(p,n)$^{101}$Rh | 4.34 d | 307 keV |
| $^{51}$V(p,n)$^{51}$Cr | 27.7 d | 320 keV |

Table 1: Reactions used for tracer quantification

The urine samples analyzed originated from studies aimed to investigate differences in the biokinetics between various chemical forms of administered ruthenium. The percentage of injected tracer excreted in urine over 48 hours was about 25% when ruthenium was administered in inorganic form and ranged between 55% and 75% when complexed to citrate. For comparison, the current ICRP model predict a 48-hour excretion of 16%. The results confirm previous observations that the plasma clearance of Ru-citrate was significantly faster than that of inorganic ruthenium (biological half-lives of 17 minutes and 23 hours, respectively).

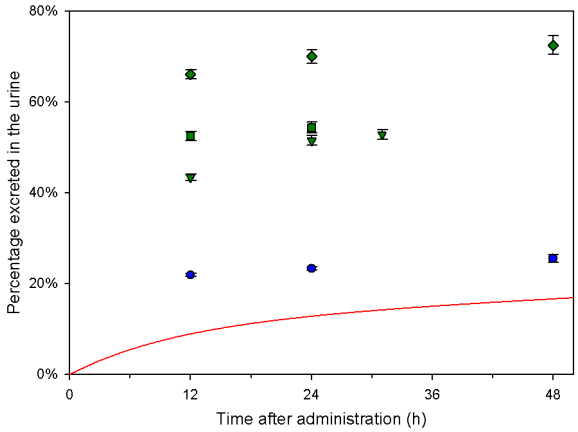


Fig. 1: Urinary excretion after intravenous administration of stable isotopes of ruthenium. Blue symbols: inorganic Ru; Green symbols: citrate complex; Line: prediction of ICRP model.

In another study, the intestinal absorption of ruthenium used as an external marker of solid food (salad) was evaluated to be equal to $(4.4\pm1.7)\cdot10^{-3}$. This value is one order of magnitude lower than the current recommendation of ICRP ($5\cdot10^{-2}$). Results from previous studies, where ruthenium had been administered as an aqueous solution, had shown that the absorbed value ranged between $7\cdot10^{-3}$ and $1.1\cdot10^{-2}$.