TRANSMUTATION OF TECHNETIUM IN THE PETTEN HFR: A COMPARISON OF MEASUREMENTS AND CALCULATIONS

J.L. Kloosterman, J.A. Hendriks and R.J.M. Konings
Netherlands Energy Research Foundation (ECN)
P.O. Box 1, NL-1755 ZG Petten, Netherlands
Tel: ++31 224 564402, Fax: ++31 224 563490
E-mail: kloosterman@ecn.nl

Abstract

Within the framework of the EFTTRA cooperation between CEA, ECN, EDF, FZK, IAM and ITU, six metallic ^{99}Tc rods have been irradiated in the Petten HFR for 193 effective full power days. During this irradiation, more than 6% of the ^{99}Tc has been transmuted to the stable ^{100}Ru. At ECN, one of the six rods has been examined in the hot cell laboratory. The ruthenium concentration in the rod measured by Isotope Dilution Mass Spectrometry reaches 6.4% at 5 mm from the bottom of the rod and 6.0% at 5 mm from the top. Also the axial and radial distributions of the ruthenium have been measured by Electron Probe Micro Analysis. The ruthenium concentrations calculated by the three-dimensional Monte Carlo code KENO reach 6.1% at 5 mm from the bottom of the rod and 5.7% at 5 mm from the top. These values are in reasonable agreement with the measured ones. However, the calculated radial distribution of the ruthenium concentration is not in agreement with the measurements. The radial profile calculated by the Monte Carlo code MCNP, which uses a point-wise cross-section library, agrees much better with the measurements.

To solve the remaining small differences between the measured and calculated ruthenium concentratrions in the rod, the thermal absorption cross section of ^{99}Tc will be measured in the Petten HFR in the course of this year.

1 Introduction

The long-lived fission products ^{99}Tc and ^{129}I are among the most important nuclides that dominate the beta activity of spent fuel after a hundred thousands of years. Because of their high solubility in (ground)water, technetium and iodine are easily transported to the biosphere once they are released from the deep-geological waste repository. To reduce the dose risks to future generations, technetium and iodine should be partitioned from the spent fuel and treated separately, e.g. transmuted in nuclear reactors or conditioned by chemical immobilization.

In 1992, the EFTTRA collaboration (Experimental Feasibility of Targets for TRAnsmutation) was founded between CEA, ECN, EDF, FZK, IAM and ITU, with the aim to investigate experimentally the behaviour of targets during irradiation in fast and thermal nuclear reactors and to demonstrate the applicability of scenarios for the transmutation of long-lived fission products and minor actinides [1].

This paper describes the irradiation and the results of the Post Irradiation Examinations (PIE) of one ^{99}Tc rod irradiated in the Petten thermal High Flux Reactor (HFR) within the framework of the EFTTRA cooperation. During the irradiation, the long-lived ^{99}Tc is transmuted to the stable ^{100}Ru. The measured ruthenium concentration and profiles are compared with results of three-dimensional Monte-Carlo calculations.

2 Target fabrication and Irradiation

Six metallic technetium rods of 4.8 mm diameter and 25 mm length were fabricated by ITU in Karlsruhe. Details of the fabrication method are described in reference [2]. The specific density was higher than 99.9% of the theoretical density. Analysis by glow-discharge mass spectrometry showed that the ruthenium concentration in the metal was less than 1 ppm. The rods were enclosed in stainless steel capsules (Phenix cladding material), provided by CEA. Each capsule contained two rods on top of each other.

The targets were positioned in peripheral holes of an aluminium sample holder. Three holes were occupied by the six ^{99}Tc rods (two rods on top of each other) and the other six holes were filled by iodine targets. The sample holder was placed in an aluminium filler element and loaded in the core position C5 of the Petten HFR. At this position, the total neuton flux in the targets exceeds 10^{15} $cm^{-2}s^{-1}$ with a thermal component higher than 2 10^{14} $cm^{-2}s^{-1}$. Close to the technetium and iodine targets, fluence detectors (^{59}Co and ^{56}Fe) and gamma scan wires were located to monitor the thermal and fast neutron fluences. Also nine thermocouples were installed to measure the temperature of the facility. The geometry of the irradiation facility is shown in figure 1.

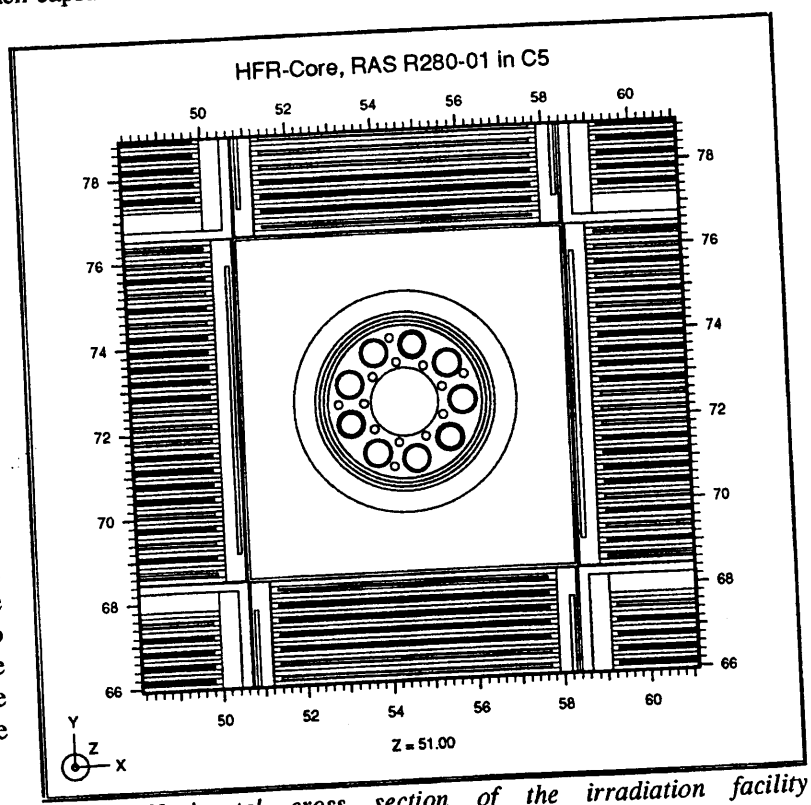

Fig. 1: *Horizontal cross section of the irradiation facility surrounded by HFR fuel assemblies.*

The irradiation lasted for eight cycles (192.95 effective full power days in total) and ended at the beginning of 1995. Due to gamma heating, the temperature of the rods exceeded 900 K. At the end of the irradiation, the thermal neutron fluence (E<0.7 eV) averaged over the technetium rods has reached a value of 3 10^{21} cm^{-2} and the total neutron fluence a value of 2 10^{22} cm^{-2}.

3 Post Irradiation Examinations

A joint PIE programme has been defined by CEA, ITU and ECN for four rods, whereas the other two rods are being re-irradiated again in the Petten HFR to reach a transmutation level of 20%.

Visual inspection after the irradiation showed that almost no irradiation damage and no swelling has occured. Measurements by means of a micrometer and micrographic examinations confirmed these conclusions. Results of these measurements are given in table 1.

Table 1: *Dimensions of some technetium rods before and after the irradiation in the Petten HFR [2].*

^{99}Tc	Diameter (mm)		Length (mm)	
sample	pre-irr	post-irr	pre-irr	post-irr
A	4.80±0.01	4.83±0.01	25.05	25.09
B	4.81±0.02	4.84±0.01	25.05	25.12
D	4.81±0.02	4.83±0.03		

Electron Probe Micro Analysis (EPMA) has been used to measure the radial profiles of the ruthenium concentration at two intersections of rod D: one located at 5 mm from the bottom (section D1) and one at 5 mm from the top (section D2). These intersections are shown in figure 2. The EPMA method has also been used to measure the axial profile along the rod axis at intersection D3 (see figure 2). The absolute values of the ruthenium concentration at D1 and D2 have been measured by Isotope Dilution Mass Spectrometry (IDMS), which is expected to be more accurate for absolute measurements.

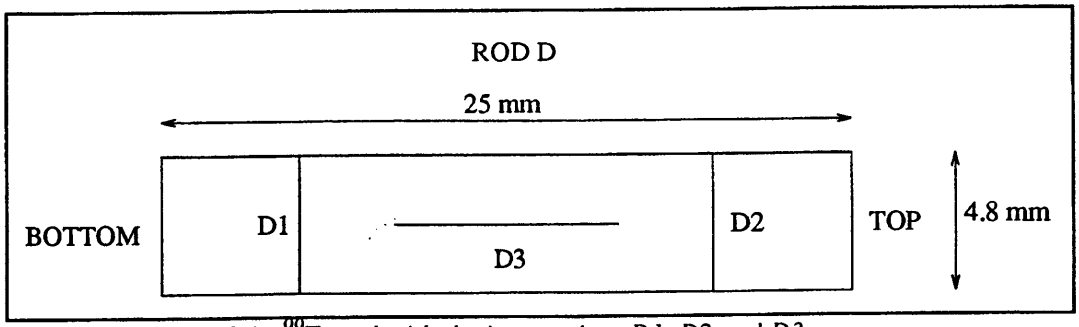

Fig. 2: *Drawing of the ^{99}Tc rod with the intersections D1, D2 and D3.*

The ruthenium concentration as a function of the cross-sectional area of the rod at intersection D1 is shown in figure 3. The two curves are the results of two measurements at different azimuthal angles perpendicular to each other. The vertical line at an area of 18.1 mm^2 denotes the outer edge of the rod with a diameter of 4.8 mm. The results at intersection D2 are very similar to the ones in figure 3 and shown in figure 4. In both figures, the so-called Rim effect caused by the resonance shielding of the neutrons is clearly visible. The difference between the profiles at the outer edge of the rod (area 18.1 mm^2) is most probably due to the position of the sample and the roughness of the surface of the rod.

The cross-sectional averaged ruthenium concentration measured by EPMA equals 7.2% at D1 and 6.8% at D2. However, as mentioned before, the EPMA method is not very accurate for absolute measurements. Therefore, two slices at each intersection D1 and D2 of the rod were dissolved in HNO_3 and the ruthenium concentration in each slice was measured several times by IDMS. The results are given in table 2.

Table 2: *Ruthenium concentrations at intersections D1 and D2 measured by Isotope Dilution Mass Spectrometry (IDMS).*

Slice	Ruthenium concentration (%)	
	D1	D2
1	6.39±0.15	5.98 ± 0.10
2	6.39±0.24	
Average	6.39±0.23	5.98 ± 0.10

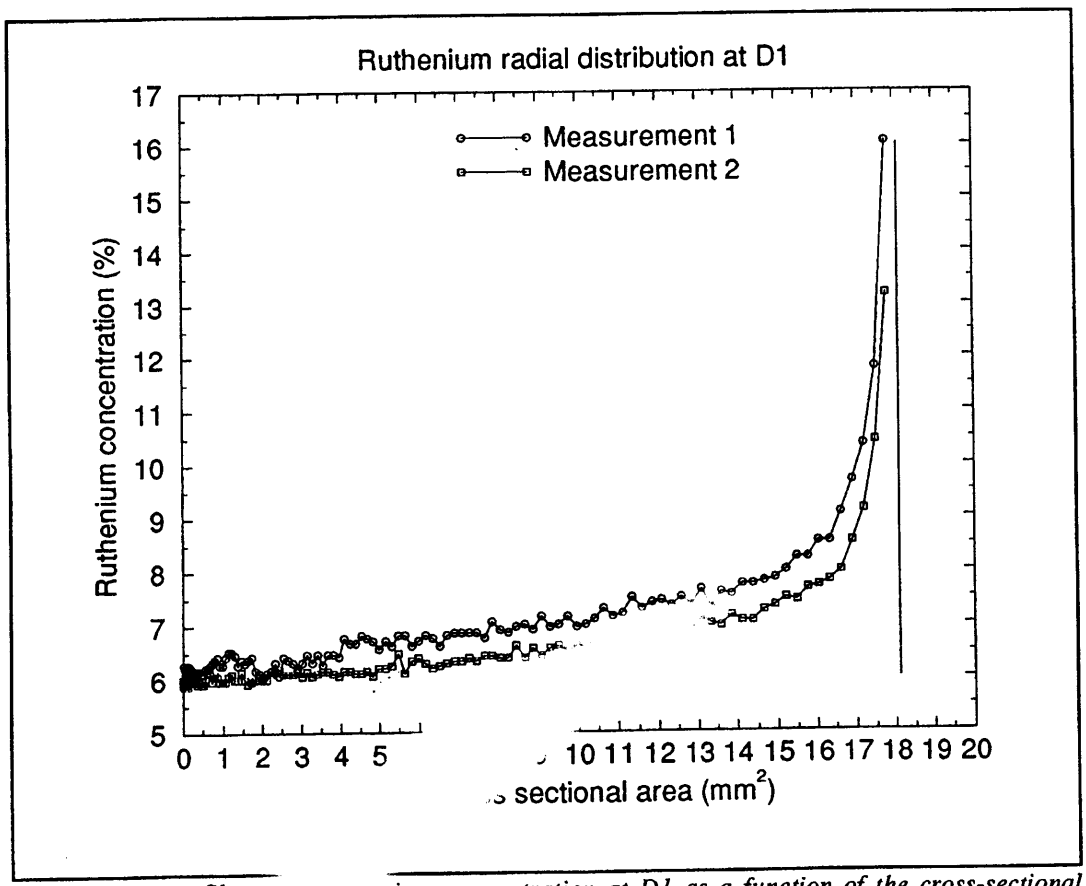

Fig. 3: *The profile of ruthenium concentration at D1 as a function of the cross-sectional area of the vertical line denotes the outer edge of the rod.*

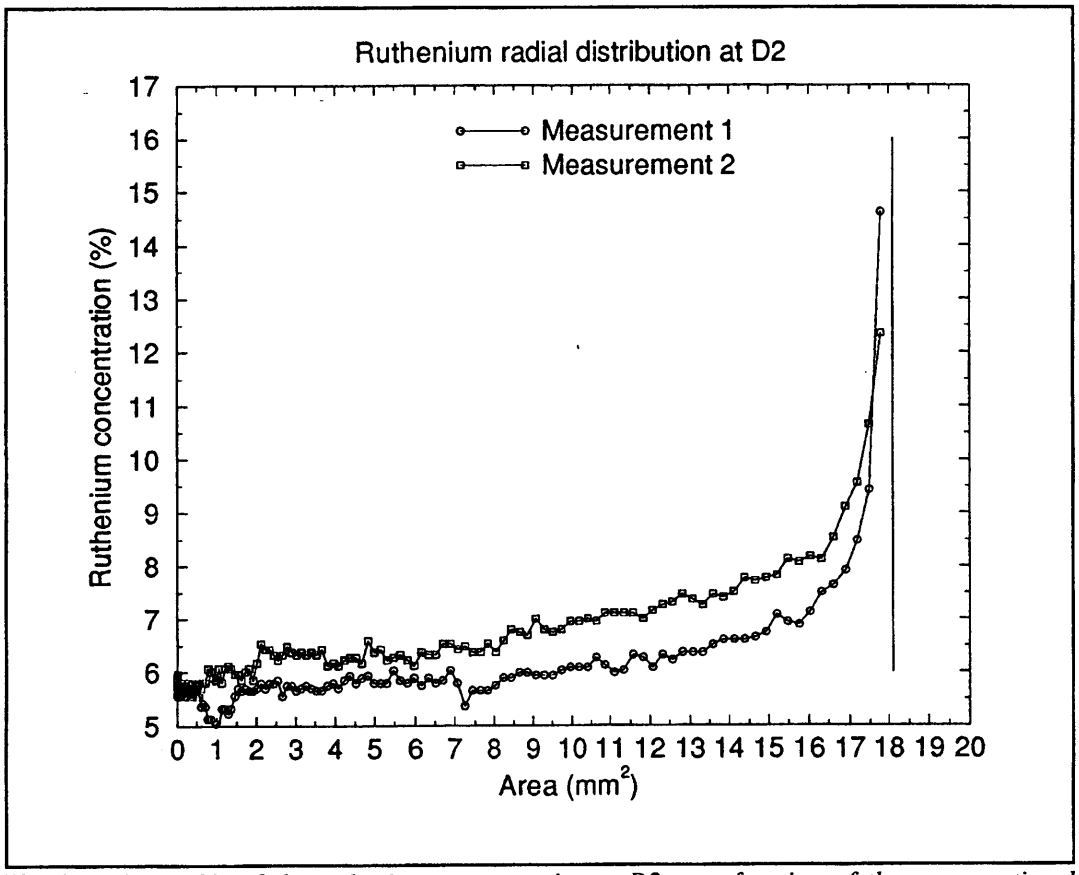

Fig. 4: *The profile of the ruthenium concentration at D2 as a function of the cross-sectional area of the rod. The vertical line denotes the outer edge of the rod.*

The ruthenium concentration at D3 as a function of the axial distance in the rod is shown in figure 5. The average ruthenium concentration is about 5.4%. Although there is a large spread in the results, the slope of the linear least squares fit indicates a small gradient of the ruthenium concentration as a function of the axial distance. According to this fit, the difference between the ruthenium concentrations at the intersections D1 and D2 along the rod axis would be 5.54-5.30=0.24%.

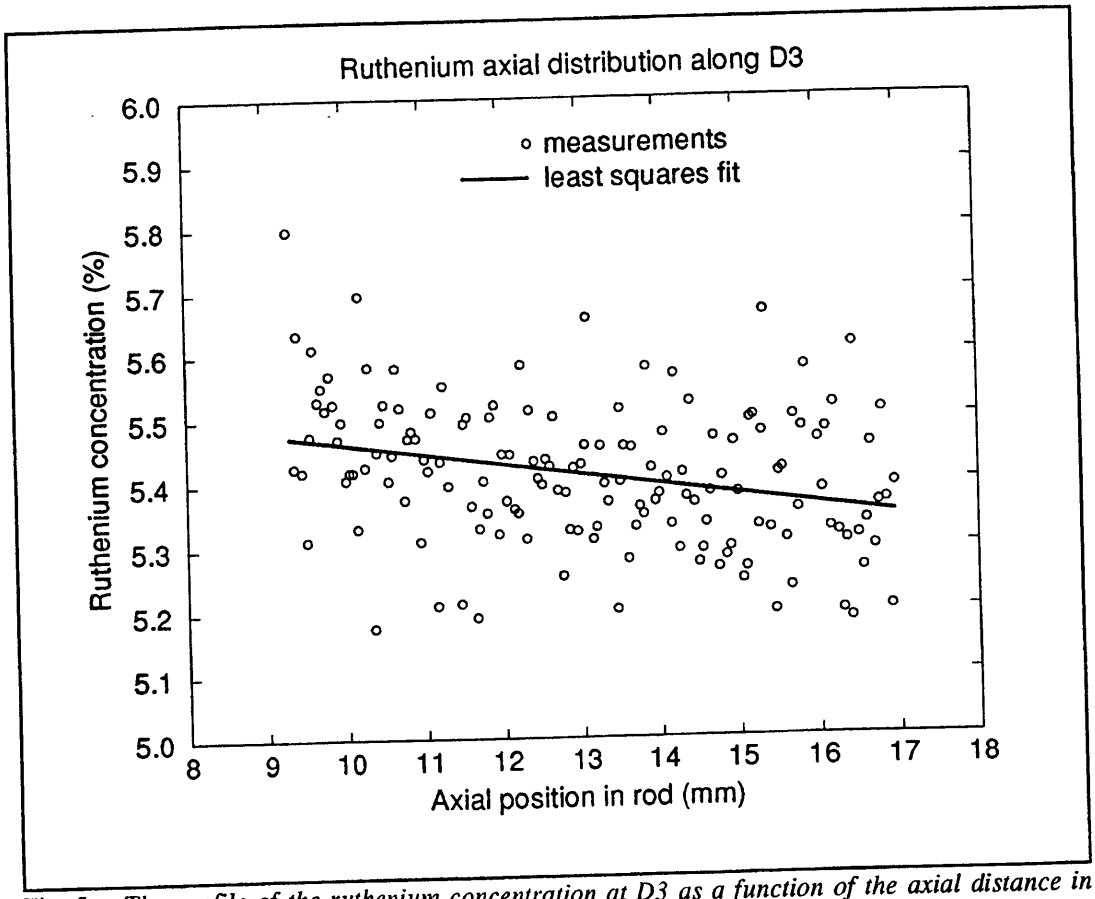

Fig. 5: *The profile of the ruthenium concentration at D3 as a function of the axial distance in the rod.*

4 KENO Monte Carlo Calculations

To assess the accuracy of the calculational methods and of the nuclear data files used in technetium transmutation studies, calculations have been performed by the Monte Carlo code KENO-Va [3] and associated nuclear data libraries based on the JEF2.2 evaluated file [4].

The standard three-dimensional core model of the Petten HFR has been used, which implies that the control members are at a fixed representative position and that a single type experimental facility was used to fill the experimental positions in the HFR model. This is done because it is too cumbersome to change the core model from one cycle to the other. The influence of these assumptions on the results is probably very small, because the experiment itself, which was loaded at position C5, has been modelled accurately, including the details on the fluence detectors (foils) and the gamma scan wires. Also the three-dimensional fuel distribution has been correctly represented. Figure 6 shows a horizontal cross section of the HFR core model with the experiment loaded at position C5, and figure 1 shows the irradiation facility in more detail.

In the calculations, the resonance shielding for all nuclides have been performed by the NITAWL-II code [5] (Nordheim method) in the resolved energy region and by the BONAMI-S code [6] (Bondarenko method) in the unresolved energy region, except for ^{99}Tc for which no resonance shielding in the unresolved region has been performed. This is due to the large background cross sections of ^{99}Tc in the unresolved region which may lead to negative cross sections when the resonance shielding is too large. Because the total contribution of the unresolved region to the capture rate is only 6.5%, this has no large effect on the results.

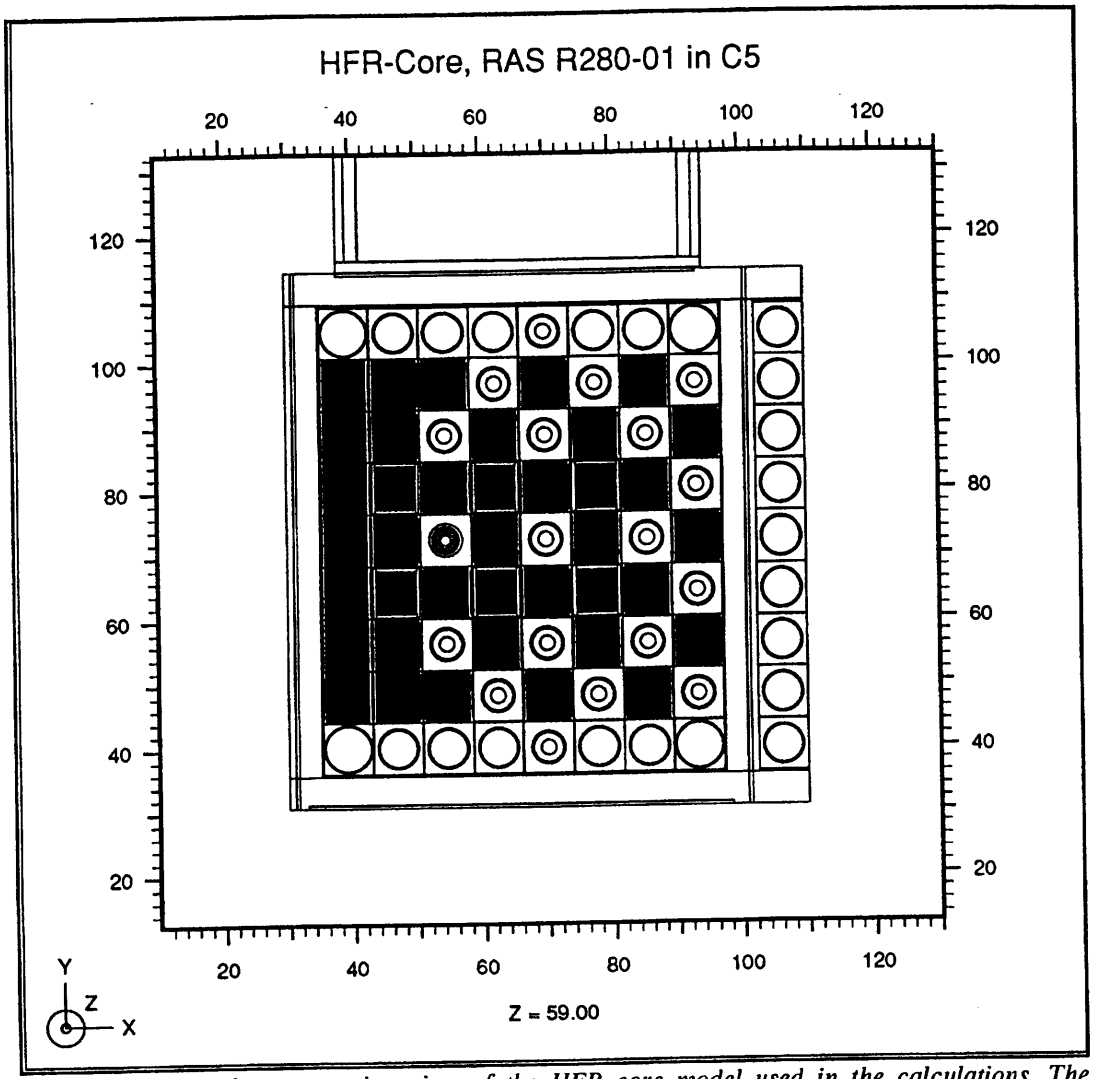

Fig. 6: *Horizontal cross section view of the HFR core model used in the calculations. The experimental facility is loaded at position C5 with (x,y) coordinates (55,73).*

The calculations yield the integrated number of $^{54}Fe(n,p)$ and $^{59}Co(n,\gamma)$ reactions in the detector foils, the ruthenium concentration averaged over each of the two rods, the axially averaged radial distribution and the radially averaged axial distribution in the two rods.

The integrated number of $^{54}Fe(n,p)$ and $^{59}Co(n,\gamma)$ reactions in the detector foils are given in table 3. It is seen that the calculated neutron spectra at the detector positions are quite accurate. The two foils closest to the investigated technetium rod (numbers 6 and 7) give results within 7% for the fast $^{54}Fe(n,p)$ reaction and within 1% for the thermal $^{59}Co(n,\gamma)$ reaction.

Table 3: *Calculated reactions in the ^{54}Fe and ^{59}Co detector foils and the associated C/E ratios.*

Detector foil	$^{54}Fe(n,p)$ (10^{21} cm^{-2})	C/E ratio	$^{59}Co(n,\gamma)$ (10^{21} cm^{-2})	C/E ratio
1	4.84±0.10	1.02±0.02	3.67±0.05	1.03±0.02
2	4.80±0.10	1.01±0.02	3.66±0.05	1.02±0.01
3	4.89±0.10	1.03±0.02	3.63±0.06	0.97±0.02
4	4.94±0.10	1.05±0.02	3.67±0.05	1.01±0.01
5	5.10±0.10	1.08±0.02	3.69±0.06	1.01±0.02
6	5.05±0.10	1.07±0.02	3.57±0.05	1.00±0.01
7	4.86±0.10	1.03±0.02	3.58±0.05	1.01±0.01
8	5.02±0.10	1.05±0.02	3.63±0.05	1.01±0.01
9	5.06±0.10	1.07±0.02	3.57±0.05	0.99±0.01
Average		1.05±0.02		1.01±0.02

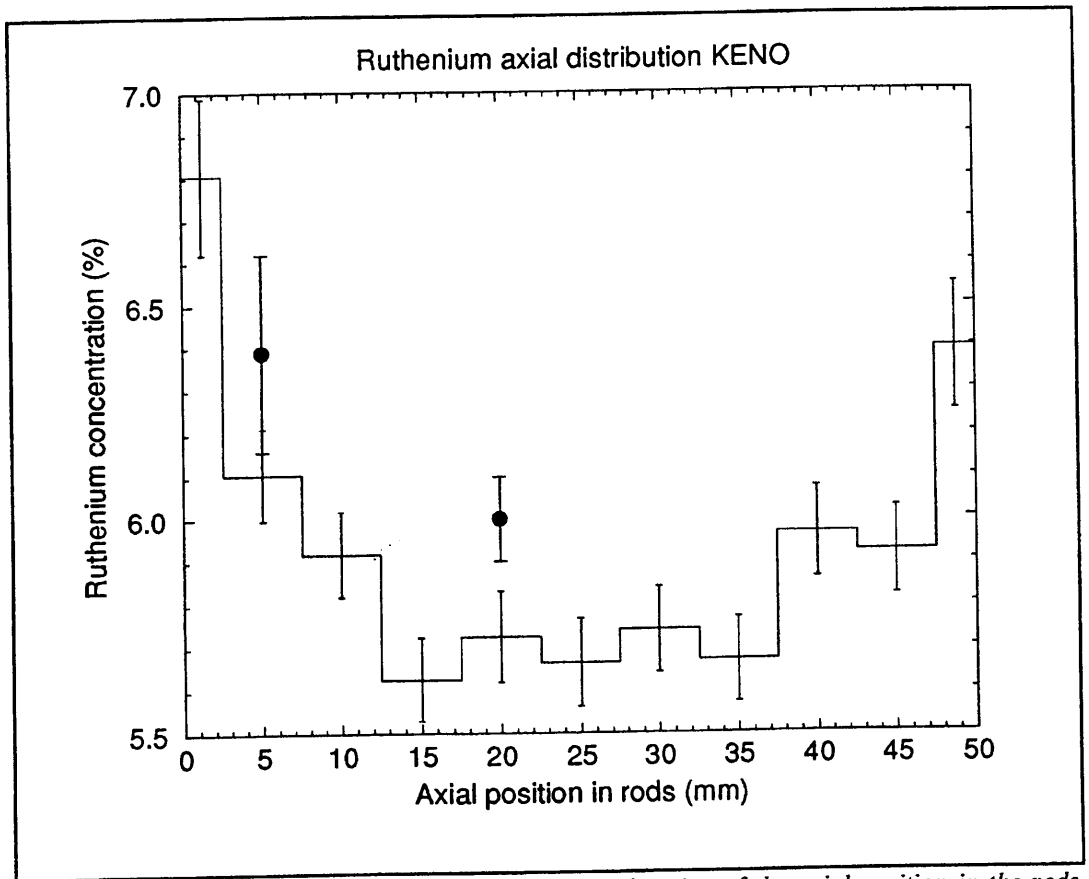

Fig. 7: *The calculated ruthenium concentration as a function of the axial position in the rods. The dots indicate the measurement values at D1 and D2.*

The axial distribution of the ruthenium concentration in the two rods calculated by the Monte Carlo code KENO is shown in figure 7 as a function of the axial position in the rods. Also the two measurement values at D1 and D2 are shown. The lower rod (0<z<25 mm) has been examined by ECN. The vertical bars at each interval denote one standard deviation due to the Monte Carlo process. Clearly the ruthenium concentration shows an axial gradient, but the volume averaged ruthenium concentration of the lower rod (5.92±0.05%) does not differ significantly from that of the upper rod (5.87±0.05%). These calculations support the measured axial gradient as shown in figure 5. The calculated ruthenium concentrations at intersections D1 and D2 have values of 6.10±0.11% and 5.72±0.10%, respectively, which are in reasonable agreement with the measured values given in table 2 (6.39±0.23% at D1 and 5.98% at D2).

The radial distribution of the ruthenium concentration averaged over the two rods in axial direction is shown in figure 8. Clearly, the calculated distribution is not in agreement with the EPMA measurements. Most probably, this is due to the resonance shielding method used in the resolved energy range. By use of the Nordheim method, pin-averaged goup cross sections are calculated which conserve the total neutron absorption. However, due to resonance absorption, the group cross sections change as a function of the distance to the moderator-rod interface.

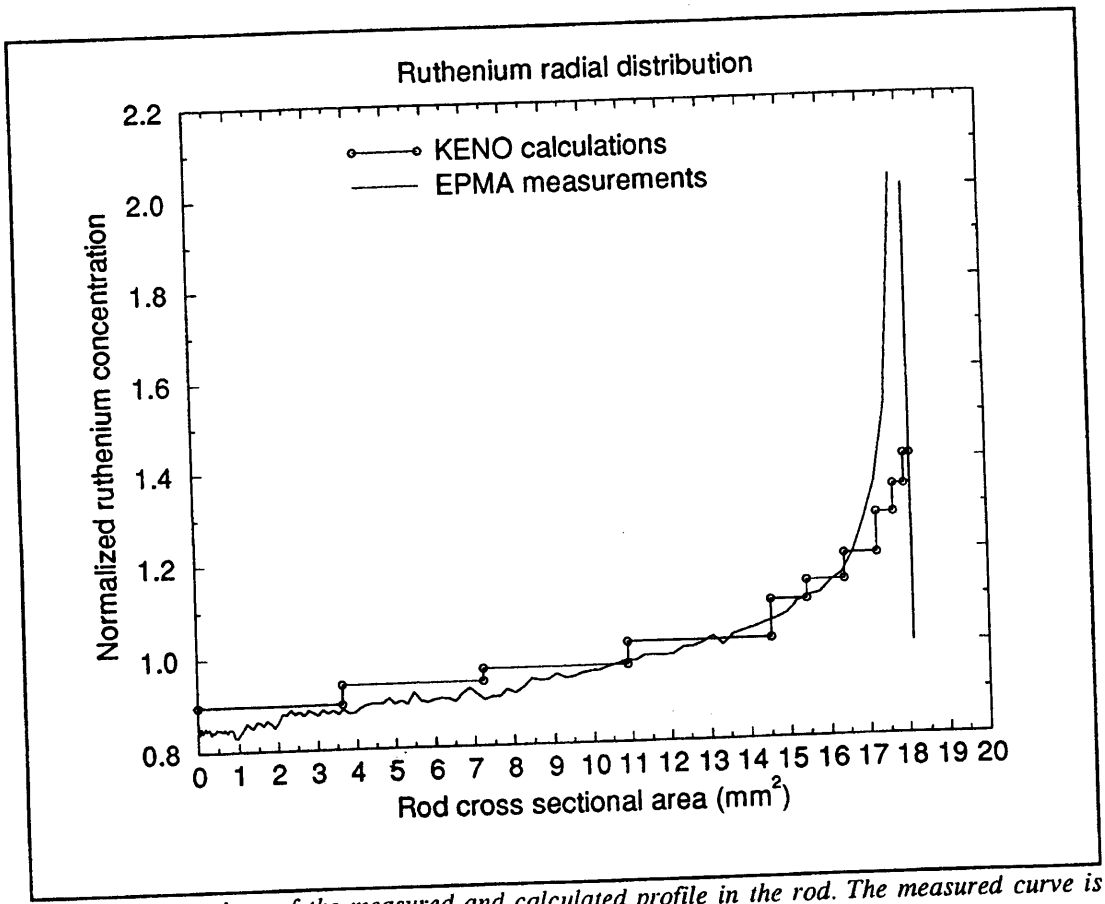

Fig. 8: *Comparison of the measured and calculated profile in the rod. The measured curve is the average of the four curves shown in figures 3 and 4.*

To demonstrate the inadequacy of the Nordheim resonance shielding method for these purposes, a simple one-dimensional model of the experiment has been defined and the radial distribution of the ruthenium concentration has been calculated by both the KENO and the MCNP-4A [7] Monte Carlo codes. The latter code uses a point-wise cross-section library also based on the JEF2.2 nuclear data file. The results are shown in figure 9, where the radial distribution of the ruthenium concentrations normalized to the same value are shown together with the results of the EPMA measurements. Clearly, the MCNP results are in much better agreement with the measurements than the KENO results, due to the point-wise sampling of the cross sections. It is emphasized, however, that the model used in this benchmark is only a simple one-dimensional mockup of the actual experiment. Therefore, the differences between the KENO and MCNP results are meaningful, while the (small) differences between the calculations and the EPMA measurements may be due to the simplified geometry.

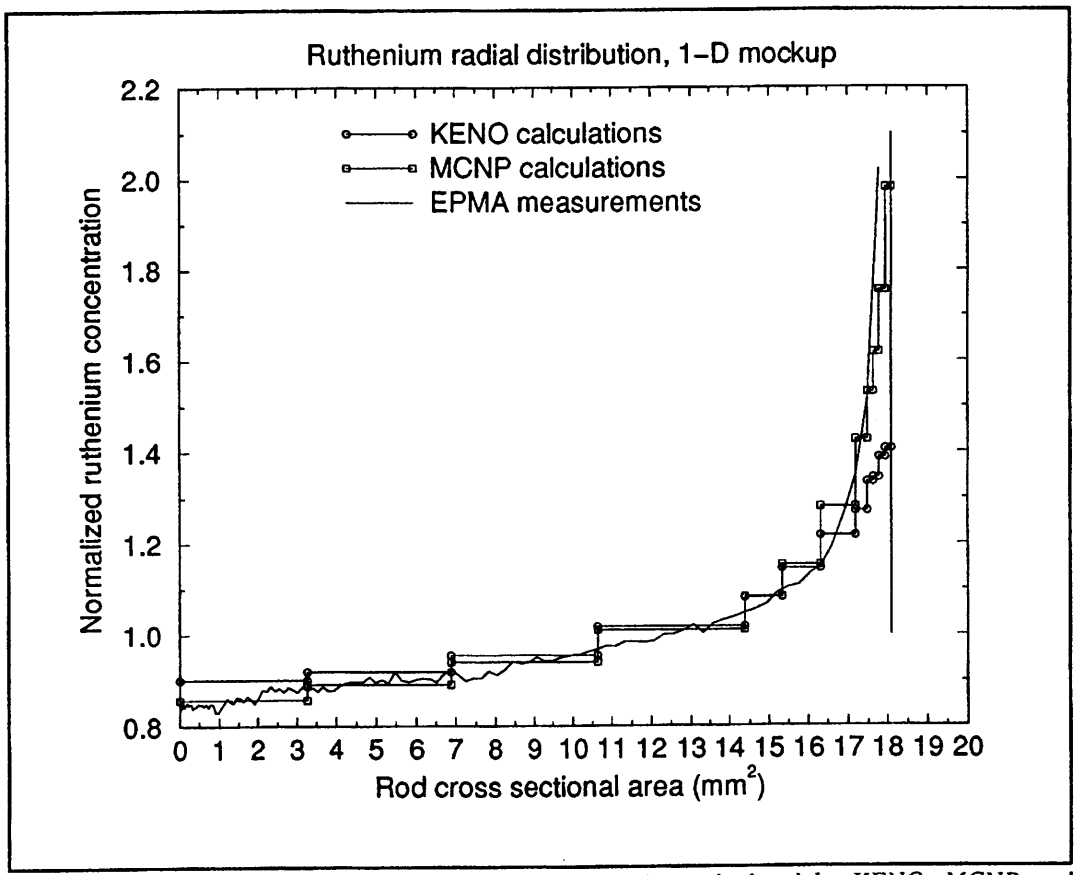

Fig. 9: *The radial profile of the ruthenium concentration calculated by KENO, MCNP and measured by EPMA.*

Conclusions

Six metallic ^{99}Tc rods have been irradiated in the Petten thermal High Flux Reactor with a total neutron fluence of 2 10^{22} cm^{-2}. About 6% of the ^{99}Tc has been transmuted to the stable ^{100}Ru.

The absolute ruthenium concentrations in the rod have been measured by Isotope Dilution Mass Spectrometry, and the radial and axial profiles of the ruthenium concentration have been measured by Electron Probe Micro Analysis (EPMA). The measured ruthenium concentration in the rod ranges from 6.4% at 5 mm from the bottom of the rod (intersection D1) to 6.0% at 5 mm from the top (intersection D2).

Calculations have been performed by the group-wise Monte Carlo code KENO in a detailed three-dimensional model of the HFR core. The calculated ruthenium concentrations of 6.1% at intersection D1 and 5.7% at D2 are in reasonable agreement with the measurements. This shows the validity of the HFR core model and of the cross sections used.

The group-wise Monte Carlo code KENO in combination with the cross-section generation code NITAWL-II (Nordheim resonance shielding method) cannot be used to calculate accurately the radial distribution of the ruthenium concentration in the rods. A Monte Carlo code using point-wise cross sections like MCNP appears to give much better results.

Further work on the transmutation of ^{99}Tc consists of the re-irradiation of two ^{99}Tc rods in the Petten HFR up to a transmutation level of 20%, and the measurement of the thermal absorption cross section. According to reference 8, the thermal capture cross section of ^{99}Tc is 10 to 15% higher than the value in the JEF2.2 evaluated nuclear data file (about 19 barn).

References

[1] J.F. Babelot *et al.*, "EFTTRA Irradiation Experiments for the Development of Fuels and Targets for the Transmutation", This Conference, September 1996.

[2] R.J.M. Konings *et al.*, "Transmutation of Technetium and Iodine - Irradiation Tests in the Frame of the EFTTRA Cooperation", submitted to Nuclear Technology, 1996.

[3] L.M. Petrie and N.F. Landers, "KENO-Va: An Improved Monte Carlo Criticality Program with Supergrouping", ORNL, Tennessee, USA, August 1990.

[4] R.C.L. van der Stad *et al.*, "EIJ2-XMAS: Contents of the JEF2.2 based Neutron Cross-Section Library in the XMAS Group Structure", Netherlands Energy Research Foundation (ECN), Petten, The Netherlands, Report ECN-CX-95-087 (confidential), February 1996.

[5] N.M. Greene *et al.*, "NITAWL-II, SCALE Module for Performing Resonance Shielding and Working Library Production, ORNL, Tennessee, USA, June 1989.

[6] N.M. Greene, "BONAMI-S, Resonance Self-Shielding by the Bondarenko Method", ORNL, Tennessee, USA, August 1981.

[7] J.F. Briesmeister, ed., "MCNP-4A: A General Monte Carlo Code for Neutron and Photon Transport", LANL, Los Alamos, USA, LA-7396-M, Rev. 2, 1986.

[8] H. Harada *et al.*, "Measurement of Thermal Neutron Cross Section and Resonance Integral of the Reaction $^{99}Tc(n,\gamma)^{100}Tc$", J. of Nuclear Science and Technology, vol.32, no.5, pp 395-403 (1995).