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Journal of Nuclear Materials 274 (1999) 336–340

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Letter to the Editors

Transmutation of technetium – results of the EFTTRA-T2 experiment

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Received 19 January 1999; accepted 26 March 1999

Abstract

Two rods of metallic technetium were irradiated in the high flux reactor (HFR) at Petten during 579.3 full power days (total neutron fluence 5.4×10^{26} m^{-2}). The extent of transmutation to ruthenium, the transmutation product, was determined to be 15–18%. Electron probe microanalysis showed an increase from 16% Ru in the centre to 30–40% Ru near the rim of the pellets. Metallographic examinations showed no changes in the microstructure compared to the unirradiated material.

1. Introduction

Recently we have reported the results of an irradiation test of the transmutation of the long-lived fission product ^{99}Tc [1–3]. This test (EFTTRA-T1) was performed in the high flux reactor (HFR) at Petten in 1994 in the frame of the EFTTRA collaboration [4]. Extensive post-irradiation examinations of the irradiated material were made at three different European laboratories [2]. The results showed that the extent of transmutation of ^{99}Tc to the stable ^{100}Ru isotope, with which it forms a continuous series of solid solution [5], was about 6 at.%. The post-irradiation examinations also revealed that no macroscopic or microscopic changes in the target material (technetium metal) occurred. These results indicate that the metal is a good material for targets for transmutation of technetium.

As a follow-up to the T1 experiment, we have re-introduced the target material of one of the three irradiation capsules in the EFTTRA-T2 experiment. The aim of this irradiation was to confirm the irradiation behaviour of the technetium metal at a significantly higher extent of transmutation. The results of this T2 experiment are reported in this letter.

2. Experimental

2.1. Samples

The irradiation capsule for the EFTTRA-T2 test contained two rods of technetium metal (designated A and B). The rods were previously irradiated in the EFTTRA-T1 experiment. After visual inspection and measurement of the dimensions, they were re-encapsulated in a 15.15 Ti stainless steel capsule for which the material was provided by CEA-Cadarache. The filling of the capsules was done in the hot cell laboratory in Petten. The capsules were filled with helium at atmospheric pressure and welded by laser welding using a specially designed exsiccator.

The rods were initially fabricated at the Institute for Transuranium Elements (ITU) in Karlsruhe. Details of the fabrication method are described in references [1,6]. The diameter of the rods was 4.8 mm and the length 25 mm, the density was higher than 99.9% of the theoretical density, and the ruthenium concentration in the metal was less than 1 ppm.

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0022-3115/99/$ – see front matter
PII: S0022-3115(99)00107-5

Table 1
The neutron fluences of the T1 and T2 experiments

Energy	Neutron fluence (m^{-2})		
	T1	T2	Cumulative
$E > 1.353$ MeV	3.36×10^{25}	6.32×10^{25}	9.68×10^{25}
67.4 keV $< E <$ 1.353 MeV	5.42×10^{25}	10.80×10^{25}	16.22×10^{25}
0.683 eV $< E <$ 67.4 keV	7.35×10^{25}	12.27×10^{25}	19.62×10^{25}
$E < 0.683$ eV	3.29×10^{25}	5.45×10^{25}	8.74×10^{25}
Total	19.42×10^{25}	34.74×10^{25}	54.26×10^{25}
$E > 0.1$ MeV	8.34×10^{25}	16.30×10^{25}	24.64×10^{25}
$E > 1$ MeV	4.10×10^{25}	7.81×10^{25}	11.91×10^{25}

2.2. Irradiation conditions

The irradiation capsules (samples) were positioned in a sample holder designed for a leg of a TRIO in-pile irradiation facility for the HFR [7]. The sample holder consisted of a ring-shaped aluminium block with peripheral holes in which the target was positioned. The sample holder was surrounded by three stainless steel containments with gas gaps between them that were flushed with inert gas (He or Ne) for temperature control.

The cumulative irradiation time of T1 and T2 is 579.3 full power days. During the irradiation, the temperature of the sample holder was measured by thermocouples positioned close to the samples. It varied around 700 K. The central temperature of the technetium metal in the irradiation capsules was calculated to be about 1100 K. The neutron fluence data, obtained from post-test analysis of gamma-scan wires and fluence detector sets (detector foils), are given in Table 1.

2.3. Post-irradiation examinations

Transverse cross-sections were cut at 5 mm from the bottom (section A1) and the top (section B2). At the same sections, thin slices of about 100 mg were cut for chemical analysis, X-ray diffraction analysis (XRD) and transmission electron microscopy (TEM).

The cross-sections were embedded in hysol, ground with SiC paper and polished with diamond paste. The sections were examined by optical microscopy for which they were etched by means of an equi-molar solution of HNO_3 and H_2O_2 containing a few drops of HF. Electron probe microanalysis (EPMA) was done on a CAMECA MS46R microprobe operating at 20 kV and 40 nA, using ruthenium metal as a standard.

For the chemical analysis by isotope dilution mass spectrometry (IDMS), the discs were dissolved in 7 M HNO_3 to which a standard solution was added. Ruthenium was distilled from the solution as ruthenium tetroxide and collected in a NaOH solution, in which it was precipitated as ruthenium hydroxide. The hydroxide was then subsequently dissolved in concentrated hydrochloric acid. The ruthenium concentration in this solution was measured by mass-spectrometry.

The X-ray diffraction analysis was done by mounting a disk (section B2) on a Nonius PDS120 diffractometer placed in a glove box. A mixture of silicon and tungsten metal powder was dispersed on the surface to serve as internal standard.

For the TEM a specimen was prepared by electrolytic thinning of a 1 mm disk of section A1, using a (0.2 H_2SO_4 + 0.8 methanol) mixture at a current of 38 V. The sample was studied in a Philips 301 TEM operated at 100 kV. Bright field and electron diffraction patterns were recorded.

3. Results

The results of the post-irradiation examinations can be summarised as follows:

- The measurements of the length and diameter of the rods, as listed in Table 2, show no significant change compared to the pre-test data and the results of the T1 test.

Table 2
Diameter and length of the technetium rods of the T1 and T2 experiments

Rod	Diameter (mm)			Length (mm)		
	Pre-test	T1	T2	Pre-test	T1	T2
A	4.80 ± 0.01	4.83 ± 0.01	4.83 ± 0.01	25.05	25.09	25.14
B	4.81 ± 0.02	4.84 ± 0.01	4.83 ± 0.01	25.05	25.12	25.16

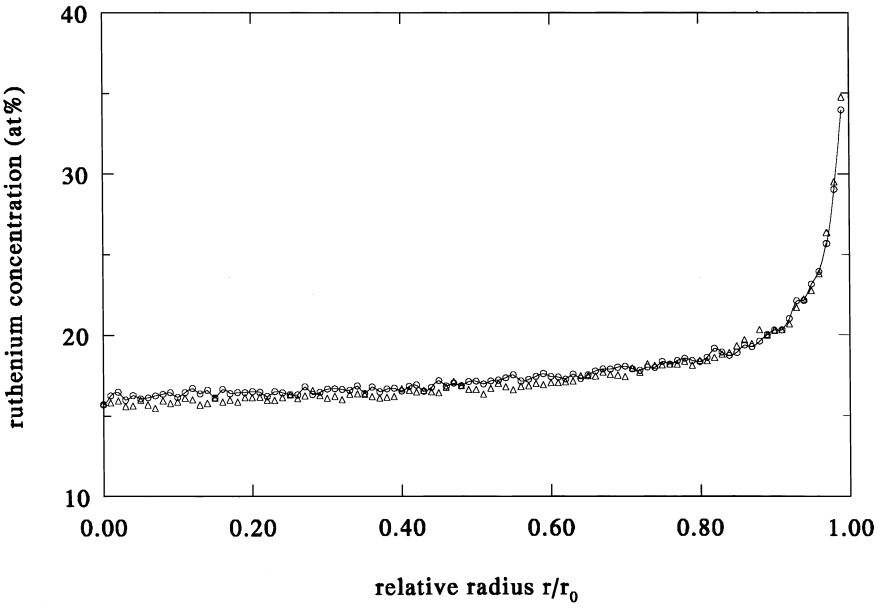

Fig. 1. Two perpendicular EPMA radial profiles of the ruthenium concentration in the technetium (section A1).

- The metallographic examinations of the irradiated samples revealed no changes in the microstructure as compared to that of the unirradiated material and that of the T1 irradiation.
- The Ru concentration profiles obtained by EPMA of the cross-section samples A1 and B2 showed an increase from about 16% in the centre to 30–40% in the outer 150 μm of the rods (Fig. 1). The pellet average values, obtained by a volumetric integration of the profiles, yielded 18.8 at.% for A1 and 18.2 at.% for B2.
- IDMS analysis of the sections A1 and B2 yielded (15.9 ± 0.3) at.% and (15.0 ± 0.4) at.% for the pellet-average Ru concentration, respectively.
- Analysis of the XRD pattern (Fig. 2 and Table 3) gave $a = 273.66(3)$ pm and $c = 437.24(10)$ pm for

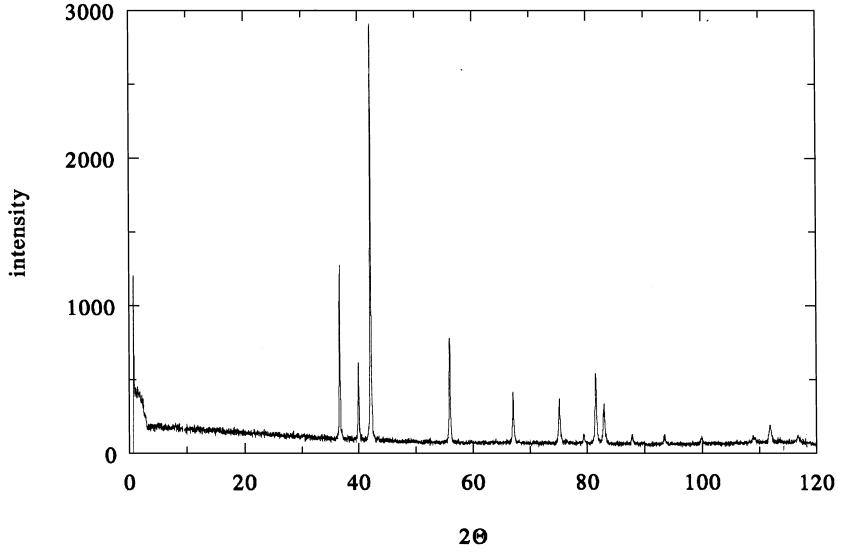

Fig. 2. The XRD pattern of the irradiated technetium (section B2).

Table 3
Experimental X-ray diffraction data of the irradiated technetium rod

d (pm)	$h\ k\ l$	I/I_0
237.20	1 0 0	16.4
218.46	0 0 2	32.1
208.32	1 0 1	100.0
160.74	1 0 2	14.5
136.80	1 1 0	6.0
124.15	1 0 3	14.2
118.52	2 0 0	2.0
115.99	1 1 2	11.0
114.35	2 0 1	7.3

the lattice parameter of the alloy. These values correspond to a cell volume of 28.357×10^{-30} m^3. Using the variation in the lattice parameters in the Tc–Ru solid solution as reported by Darby et al. [5] we derive that the Ru concentration is (18 ± 1) at.%. The error in this value is estimated (see below).

- TEM analysis showed the presence of some isolated dislocation loops.

4. Discussion and conclusions

The results of the ruthenium concentration measurements are summarised in Table 4. It can be seen that the results of the EPMA and XRD analysis of sections are in good agreement, but deviate from the results of the IDMS analysis, as was the case for the T1 test. As explained before [3] the results of the IDMS analysis are considered the most relevant because they represent the average value of the cylindrical volume. The Ru concentrations obtained by integration of a 'one-dimensional' EPMA profile are less representative for the volume concentration because variation exists in the Ru concentration near the pellet edge. The result of the XRD analysis is considered less accurate because it depends upon accuracy of the lattice parameter of the Tc–Ru solid solution. For the present work, the data were read from the graph in the paper by Darby et al. [5], in absence of numerical data. In addition, Darby et al. stressed that the compositions of alloy used in their work are approximate.

Table 4
The ruthenium concentration in samples of rod D from the T1 experiment (neutron fluence 2.0×10^{26} m^{-2}) and of rods A and B from the T2 experiment (neutron fluence 5.4×10^{26} m^{-2})

Technique	Ru concentration (at.%)			
	D1	D2	A1	B2
EPMA	6.7	7.1	18.8	18.2
IDMS	6.1 ± 0.1	6.4 ± 0.1	15.9 ± 0.3	15.0 ± 0.4
XRD				18 ± 1

The normalised Ru profiles are shown in Fig. 3, together with those of the sections D1 and D2 from the T1 experiment. The results of the T1 and T2 experiment exhibit exactly the same shape, showing that the effect of resonance shielding of the epithermal neutrons in the resolved energy range is identical for both irradiations.

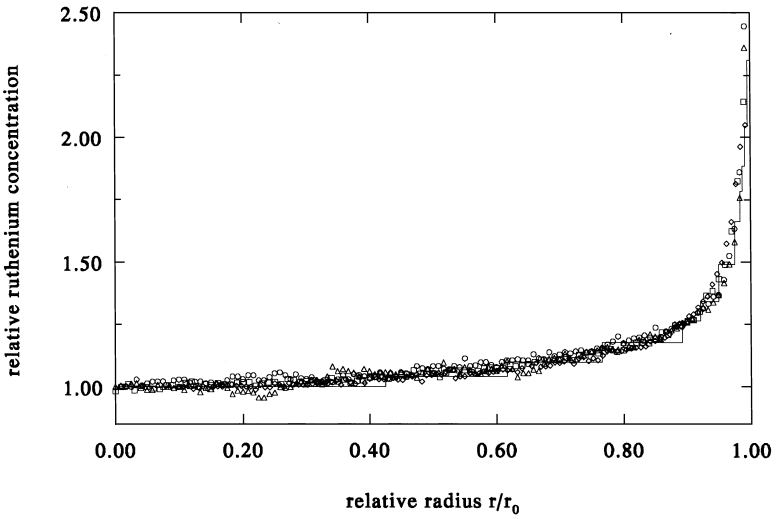

Fig. 3. The relative radial ruthenium concentration in the technetium samples D1 (○) and D2 (△) from the T1 experiment (neutron fluence 2.0×10^{26} m^{-2}) and A1 (□) and B2 (◇) from the T2 experiment (neutron fluence 5.4×10^{26} m^{-2}); the solid line shows the results of the MCNP calculations [3].

Also shown in Fig. 3 is the calculated profile obtained by post-test calculations for experiment T1, using the Monte Carlo code MCNP [3], which is in good agreement with the EPMA results for both irradiations.

The results of the T2 irradiation test of the transmutation of ^{99}Tc show negligible changes in the dimensions and microstructure of the metallic targets at an extent of transmutation of about 15–18%. They confirm the conclusion of our earlier study that there are no technical limitations to the use of metallic technetium as a target for transmutation.

Acknowledgements

The authors want to thank R. Belvroy, F. v.d. Berg, G. Dassel, J.G. v. Raaphorst en P. v. Vlaanderen for their technical support.

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