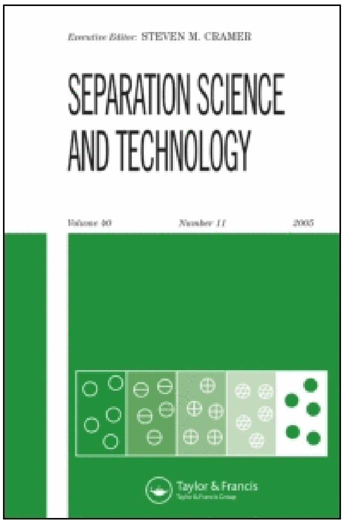

Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Separation of No-Carrier-Added ^{188}Re from Neutron-Irradiated Natural Tungsten

M. Akhtar[a]; A. Mushtaq[a]; H. M. A. Karim[a]; M. Zafar Iqbal[b]
[a] NUCLEAR CHEMISTRY DIVISION, PAKISTAN INSTITUTE OF NUCLEAR SCIENCE & TECHNOLOGY, ISLAMABAD, PAKISTAN [b] INSTITUTE OF CHEMISTRY, UNIVERSITY OF THE PUNJAB, LAHORE, PAKISTAN

To cite this Article Akhtar, M. , Mushtaq, A. , Karim, H. M. A. and Iqbal, M. Zafar(1996) 'Separation of No-Carrier-Added ^{188}Re from Neutron-Irradiated Natural Tungsten', Separation Science and Technology, 31: 14, 1997 — 2000
To link to this Article: DOI: 10.1080/01496399608001026
URL: http://dx.doi.org/10.1080/01496399608001026

SEPARATION SCIENCE AND TECHNOLOGY, 31(14), pp. 1997–2000, 1996

TECHNICAL NOTE

Separation of No-Carrier-Added ^{188}Re from Neutron-Irradiated Natural Tungsten

M. AKHTAR, A. MUSHTAQ, and H. M. A. KARIM
NUCLEAR CHEMISTRY DIVISION
PAKISTAN INSTITUTE OF NUCLEAR SCIENCE & TECHNOLOGY
P.O. BOX 1482, ISLAMABAD, PAKISTAN

M. ZAFAR IQBAL
INSTITUTE OF CHEMISTRY
UNIVERSITY OF THE PUNJAB
LAHORE, PAKISTAN

ABSTRACT

Rhenium-188 has been determined to be a suitable therapy radiolable due to its unique half-life and β^- emission associated with 155 keV photons and chelation properties. ^{188}Re is obtained from the decay of parent ^{188}W in the form of a generator system while ^{188}W is produced by double neutron capture of ^{186}W. The yield of generators using alumina for separation is limited due to the low sorption capacity of sorbent. We have exploited precipitation as hydrous tungsten oxide in acid media and redissolution in alkaline media to separate ^{188}Re from tungsten. The optimized experimental conditions indicate the feasibility of a generator system based on this method.

INTRODUCTION

The use of ^{186}Re and ^{188}Re has increased substantially in radioimmunotherapy as radiolabels of tumor-specific antibodies (1, 2). ^{186}Re is characterized by a reasonably long half-life of 90 hours, strong β^- emission (1.07 MeV), a single γ emission of 137 keV (9%), and decay into a stable nuclide of Os. ^{188}Re decays by β^- emission of 2.11 MeV (79%) and 1.96 MeV (20%) followed by a γ photon of 155 keV (15.8%) (3). These characteristics make them ideal labels for internal radiotherapy. Imaging with an Auger camera is simultaneously feasible to monitor the biodistribution pattern of therapeutic agents. Recent advances in the chemistry of Tc and Re have also contributed to their widespread use (4). To get the desired specific activity, ^{186}Re is obtained by reactor irradiation of enriched ^{185}Re. No-carrier-added production of ^{188}Re is possible through reactor irradiation of tungsten by the double neutron capture of ^{186}W (5):

$$^{186}\mathrm{W}(\mathrm{n},\gamma)^{187}\mathrm{W}(\mathrm{n},\gamma)^{188}\mathrm{W} \xrightarrow[t_{1/2}=69\ \mathrm{d}]{\beta^-} {}^{188}\mathrm{Re} \xrightarrow[t_{1/2}=17\ \mathrm{h}]{\beta^-,\gamma} {}^{188}\mathrm{Os}$$

Due to the longer half-life of ^{188}W and the shorter half-life of ^{188}Re, a generator can be developed for periodically separating the daughter from the parent nuclide. The growth of ^{188}Re is maximum after 114 hours of separation.

A ^{188}W/^{188}Re generator has been developed mainly using chromatographic separation of two nuclides (5–7). The loading capacity of W(VI) on alumina has been studied in detail by Klofutar et al (7). Despite good separation factors achieved for W and Re, the adsorption capacity of alumina poses limitation for its use on a production scale (maximum capacity of 4.41 mg WO_3/g alumina). Brodskaya et al. studied the separation of W and Re on organophosphorous resins (5). Anion-exchange resins have been exploited for the separation of these two elements (8, 9). Solvent extraction of rhenium with pyridine gives low decontamination factors (10). The present study covers the separation of ^{188}Re from ^{188}W using repeated precipitation and dissolution of tungsten.

EXPERIMENTAL

A target of Na_2WO_4 (1 g) was irradiated in a PARR-1 at a neutron flux of $\sim 10^{14}$ $\mathrm{cm}^{-2}\cdot\mathrm{s}^{-1}$ for a period of 72 hours followed by a decay period of 10 days. The irradiation target was dissolved in water and its volume was adjusted to 10 mL (Solution I).

To 2 mL of the above solution was added dropwise 8 mL of 3 N HCl with continuous stirring to obtain a precipitate of tungsten(VI) oxide which was allowed to digest at room temperature for 4 hours. The precipitate was separated by centrifugation, washed with 10 mL distilled water, and dissolved in 0.1 N NaOH and set aside to generate ^{188}Re by the decay of ^{188}W for a 5-day interval. The process of precipitation and separation was then repeated. The supernatant solution was collected (Solution II) while the precipitate was dissolved in 0.1 N NaOH (Solution III). Aliquots of Solutions I, II, and III were counted on a calibrated Canberra 85 series γ-spectrometer coupled to a Ge (intrinsic) detector for peaks corresponding to ^{187}W and ^{188}Re.

Solution II containing ^{188}Re was treated with 1–2 drops of H_2O_2 followed by boiling to remove excess H_2O_2. The pH of the solution was adjusted to 6–7. The last traces of tungsten were removed by passing the solution through alumina (7).

RESULTS AND DISCUSSION

The intensity of 155 and 480 keV of ^{188}Re and ^{187}W respectively were measured to quantify the amount of ^{188}Re and tungsten in the mixture and the separated fractions. Using counts of Solution I as the standard, the following results were obtained in decant (Solution II) and redissolved tungsten(VI) oxide (Solution III):

	Solution II	Solution III
^{188}Re	>98%	<2%
^{187}W	<1%	>99%

The above results show that about 98% ^{188}Re accompanied by 1% W is separated by this method. The efficiency of the separation procedure depends upon the time of digestion of the precipitate as given in Table 1.

The oxide of tungsten produced by this method contains about 2.7 molecules of water per formula unit WO_3 (11). This oxide has been selected for separation of Re and W due its simple preparation and ease of dissolution. Methods involving precipitation of W as tungstates of metals were also studied for separation of Re. However, no tungstate has been found suitable for repeated precipitation and dissolution. $CaWO_4$ separates ^{188}Re almost quantitatively but it is not easy to redissolve. $ZnWO_4$ precipitate is accompanied by a large amount of hydrolyzed Zn^{2+} at a pH above 5, and about one-third of the ^{188}Re remains adsorbed on the precipitate.

TABLE 1

Time of digestion	Amount of W precipitated (%)	Amount of ^{188}Re recovered (%)
15 minutes	80	88
30 minutes	85	91
1 hour	93	94
2 hours	98	98
3 hours	99	98
4 hours	99	98

The yield of ^{188}W was determined by counting the ^{188}Re in equilibrium with ^{188}W using the counting efficiency curves of the measuring equipment after irradiation and cooling as given above. Under the given conditions the maximum activity of ^{188}Re obtained at equilibrium (the time to reach equilibrium is 4.75 days) is 0.5 mCi/g W, which is also the yield of the parent nuclide produced. The radiochemical purity of the product is >95%.

REFERENCES

1. S. M. Quadri and B. M. Wessels, *J. Nucl. Med.*, *13*(4), 447–451 (1986).
2. J. L. Vanderheyden, A. R. Fritzberg, T. N. Rao, S. Kasina, A. Srinivasar, J. M. Reno, and A. G. Morgan, *Ibid.*, *28*(4), 656 (1987).
3. E. Browne and R. B. Firestone, *Table of Radioactive Isotopes*, Wiley, New York, 1988.
4. E. Deutsch, K. Libson, J. L. Venderhyden, A. R. Ketring, and H. R. Maxon, *Nucl. Med. Biol.*, *13*(4), 465–477 (1986).
5. G. A. Brodskaya, O. U. Gapurova, and E. Gureev, *Radiokhimiya*, *32*(6), 56–61 (1990).
6. A. P. Callahan, D. E. Rice, D. W. McPherson, S. Mirzadeh, and F. F. Knapp Jr., *Appl. Radiat. Isot.*, *43*(6), 801–804 (1992).
7. C. Klofutar, F. Krasovec, and A. Kodre, *J. Radioanal. Chem.*, *5*, 3–10 (1970).
8. M. Pirs and R. J. Magee, *Talanta*, *8*, 395–399 (1961).
9. J. Blachot, J. Horment, and A. Moussa, *Int. J. Appl. Radiat. Isot.*, *20*(6), 467–478 (1969).
10. H. Grosse-Rukyen and H. G. Doge, *Talanta*, *12*, 73–80 (1965).
11. A. K. De and N. D. Chowdhury, *Chromatographia*, *11*(10), 586–590 (1978).

Received by editor August 15, 1995