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Separation of no-carrier-added rhenium from bulk tantalum by the sodium malonate–PEG aqueous biphasic system

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HIGHLIGHTS

- Radioisotopically pure ^{183}Re was obtained by α particle bombardment on tantalum.
- Sodium malonate–PEG 4000 ABS was used for first time for metal ion partitioning.
- Nca ^{183}Re was separated from bulk tantalum by sodium malonate–PEG 4000 ABS.
- Temperature and salt concentration of ABS were found to optimize the separation.

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ABSTRACT

The aqueous biphasic system (ABS) involving sodium malonate–polyethylene glycol (PEG) phases has been applied for the first time for separation of no-carrier-added ^{183}Re ($T_{1/2}$=70 d) from α-particle irradiated bulk tantalum target. The various ABS conditions were applied for investigating the separation by varying pH, temperature, PEG-molecular weight, concentration of salt. The extraction pattern was hardly affected by change in pH and the molecular weight of PEG. One step separation of nca ^{183}Re from Ta was achieved at the optimal conditions of (i) 50% (w/w) PEG-4000—2 M sodium malonate, 40 °C and (ii) 50% (w/w) PEG-4000—3 M sodium malonate, room temperature (27 °C).

1. Introduction

In recent decades Re radionuclides, like ^{186}Re (3.7 d) and ^{188}Re (17.0 h) have emerged as potential choices in therapeutic nuclear medicine (Mausner and Srivastava 1993; Knapp et al., 2001; Oh et al., 2003; Syed et al., 2006, Christensen and Petersen, 2012). Production rates of these radionuclides from charged particle activation are low (Ruth, 2009), leaving only practical production route via neutron capture in a reactor. However, the reactor produced radioisotopes are not carrier-free and have low specific activity which encouraged alternative accelerator based production such as proton and deuteron bombardment on enriched ^{186}W target in order to produce high specific activity ^{186}Re through ^{186}W (p, n)^{186}Re reaction (Bonardi et al., 2010). On the other hand, other radioisotope of rhenium like ^{181}Re has suitable moderate half-life (19.9 h) and high intensity γ-lines (365.6 keV, Iγ 56%), thus can serve as a potential alternative radionuclide in the field of nuclear medicine or for biodistribution and biokinetic studies. Besides, relatively long-lived radionuclide like ^{183}Re (70.0 d) with principle γ-line 162.3 keV (Iγ 23.3%) is beneficial for trace/ultra-trace scale geochemical research regarding the Re–Os isotopic system (http://nucleardata.nuclear.lu.se/nucleardata/toi/). The nuclear properties of various Re radionuclides mentioned in this paper have been tabulated in Table 1.

Among the cyclotron-based production routes for $^{180-184}$Re isotopes, alpha particle bombardment on tantalum is a convenient choice according to several theoretical as well as the experimental literature. In 1968 Scott et al. (1968) reported experimental cross-section of ^{181}Ta(α, xn)$^{181-184}$Re and ^{181}Ta(^{3}He, xn)$^{180-183}$Re. In the same year, Newton (1968) produced ^{183}Re by ^{181}Ta(α, 2n) reaction. Hermes et al. (1974) also measured the cross-section values of ^{181}Ta(α, 2–8n)$^{177-183}$Re for a wide projectile energy range of 15–103 MeV. Abe et al. (1984) bombarded various metals including

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Table 1
The nuclear properties of various Re radionuclides.

Radionuclide	Decay mode	Half-life (h)	Principal γ-lines in keV (% γ intensity)
^{181}Re	ε	19.9	109.97 (2.7), 360.7 (20.0), 365.6 (57), 639.3 (6.5), 661.65 (3.0), 805.11 (3.0), 953.43 (3.6), 1000 (3.4), 1009.38 (2.5)
^{182}Re	ε	64.0	100.11 (16.4), 113.67 (4.9), 130.8 (7.5), 133.77 (2.39), 152.43 (8.5), 156.39 (7.2), 169.15 (11.3), 172.88 (3.57), 178.46 (2.26), 179.39 (3.01), etc.
^{182m}Re	ε	12.7	100.11 (14.3), 152.43 (7.0), 229.32 (2.5), 470.32 (2.0), 894.88 (2.1), 1121.3 (32.0), 1189.05 (15), 1221.41 (24.8)
^{183}Re	ε	70.0 d	107.93 (2.2), 109.73 (2.97), 162.33 (24.0), 208.81 (3.05), 291.73 (2.7)

Table 2
The cross-section values of $^{nat/181}$Ta(α, xn)$^{181-184}$Re nuclear reactions in some earlier studies.

Nuclear reaction	Maximum cross-section (mb)	Projectile energy (MeV)	Reference
^{181}Ta(α, n) ^{184}Re	44.7	21.0	Scott et al. (1968)
^{181}Ta(α, 2n) ^{183}Re	869.6	27.6	
^{181}Ta(α, 3n) ^{182}Re	1266.3	36.6	
^{181}Ta(α, 4n) ^{181}Re	1286	47.8	
^{181}Ta(^{3}He, 3n) ^{181}Re	400	24	
^{181}Ta(^{3}He, 3n) ^{181}Re	672.8	31	
^{181}Ta(α, 2n) ^{183}Re	786	26.3	Hermes et al. (1974)
^{181}Ta(α, 3n) ^{182}Re	1177	37.6	
^{181}Ta(α, 4n) ^{181}Re	1045	49.1	
^{181}Ta(α, 2n) ^{183}Re	1061.3	26.3	Ismail and Divata (1988)
^{181}Ta(α, 4n) ^{181}Re	988.6	47.7	
^{181}Ta(α, 2n) ^{183}Re	801.44	27.137	Rao et al. (1991)
^{181}Ta(α, 4n) ^{181}Re	984.23	45.929	
^{181}Ta(α, n) ^{184}Re	15.74	18.7	Ozafran et al. (1993)
^{181}Ta(α, n) ^{184}Re	34	19.5	Singh et al. (1994)
^{181}Ta(α, 2n) ^{183}Re	1100	26.5	
^{181}Ta(α, 4n) ^{181}Re	1040	47.8	
^{181}Ta(α, 2n) ^{183}Re	772.58	27.8	Ismail (1998)
^{181}Ta(α, 2n) ^{183}Re	1040	27	Patel et al. (1999)
^{181}Ta(α, 4n) ^{181}Re	1150	44.8	
^{nat}Ta(α, n) ^{184}Re	34.7	19.3	Tárkányi et al. (2003)
^{nat}Ta(α, 2n) ^{183}Re	934.68	27.2	
^{nat}Ta(α, 3n) ^{182}Re	1016.2	35.7	
^{nat}Ta(α, 3n) ^{182m}Re	312.6	35.7	
^{nat}Ta(α, 2n) ^{183}Re	~800	25	Maiti (2011)[a]
^{nat}Ta(α, 3n) ^{182}Re	~1300	~36	
^{nat}Ta(α, 4n) ^{181}Re	1300 <	~46	

[a] Theoretical calculation.

tantalum with 30 MeV α-particle beam. They reported thick target production yield of long lived $^{183-184}$Re radionuclides in α-particle irradiated tantalum matrix. Alpha particle induced reaction on tantalum was reported by Gadiolo et al. (1985), Mohan Rao et al. (1991) and Ismail (1998). Production of ^{184}Re via ^{181}Ta(α, n) reaction was investigated by Ozafran et al. (1993), Denisov et al. (1993) and Santos et al. (2000). In 2003 excitation functions for the ^{nat}Ta(α, *x*n) reaction were evaluated by Tárkányi et al. (2003). Recently the theoretical cross-section data on production of proton-rich radionuclides calculated by Maiti (2011) reveals that Ta(α, *x*n) nuclear reaction is best suited for the production of nca $^{181-184}$Re isotopes. The cross-sections of some of important reactions described above have been depicted in Table 2.

Several reports on the separation of no-carrier-added (nca) rhenium nuclides from the bulk target matrix are available in the literature. One of the sources of ^{188}Re is ^{188}W–^{188}Re generator system where the parent ^{188}W is produced by double neutron capture of ^{186}W through ^{186}W(n,γ)^{187}W(n,γ)^{188}W reaction. Earlier, the separation of nca rhenium was attempted from the alumina column based ^{188}W–^{188}Re generator system (Knapp et al., 1994). Other methods like thermochromatography (Novgorodov et al., 2000), electrochemical separation technique (Chakravarty et al., 2009), etc., were also proposed for separation of ^{188}Re from ^{188}W. In 2001, Lahiri et al. (2001a) separated ^{181}Re from ^{16}O irradiated Tm_2O_3 target by liquid–liquid extraction (LLX) using cation exchanger di-(2-ethylhexyl) phosphoric acid (HDEHP). Experimental simulation on separation of nca rhenium from tantalum target was carried out by Banerjee et al. (2000) with the HNO_3–trioctylamine (TOA) LLX system. Lahiri et al. (2001b) also reported an indirect method of obtaining nca ^{183}Re. They produced ^{183,183m}Os (13.0 h, 9.9 h) isotopes by ^{7}Li bombardment on natural tantalum and separated nca ^{183,183m}Os by the HCl–TOA LLX system. The short-lived ^{183,183m}Os decayed by electron capture to produce ^{183}Re. Recently, we have carried out a simulation study on separation of nca rhenium from bulk Hf and W using both cation exchanger HDEHP and anion exchanger TOA from HNO_3 medium (Maiti et al., 2011).

ABSs are formed in contact of two mutually incompatible water-soluble components; mainly with three kind of combinations:(i) polymer–polymer (e.g., dextran and poly ethylene glycol or PEG), (ii) polymer–inorganic salt (e.g., PEG and ammonium sulfate) and (iii) salt–salt combination (e.g., potassium phosphate and 1-butyl-3-methyl imidazolium chloride or [C$_4$mim]Cl). Among these ABSs the polymer–salt combination are the most widely used for large-scale extraction due to rapid biphasic disengagement, stability of system and low cost in processing.

ABSs are attractive choices for selective metal ion partitioning. The hydrophobic polymer polyethylene glycol (PEG) has been predominantly used along with mostly phosphate and sulfate salts of sodium/potassium for these separations (Rogers and Zhang 1996; Roy et al., 2008a, 2008b; Lahiri and Roy, 2009). Even in some studies, PEG based ABS systems were applied for developing green methods for synthesizing gold and gold–palladium bimetallic nanoparticles (Roy and Lahiri, 2006, Roy and Lahiri, 2008c). However, high concentrations of these salts in the effluent streams (Hustedt, 1986) are not desirable as per environmental concerns (WHO Guideline, 1993; Arai, Sparks 2001, Mavrov et al., 1997).

Some authors proposed citrate (Zafarani Moattar and Hamidi (2003)) and tartrate (Malpiedi et al., 2008) salts as substitutes of these inorganic salts. The ABSs formed by these salts offers similar characteristics with PEG+ phosphate/sulfate salt. Additionally, due to their biodegradability and non-toxicity, they can be discharged into biological waste water treatment plants.

Recently, we demonstrated potential applicability of PEG based ABSs with citrate and tartrate salt for separation of nca ^{183}Re from bulk tantalum (Dutta et al., 2013). The present study continues this attempt by reporting the first time use of environmentally benign biodegradable sodium salt of naturally occurring malonic acid, to form the ABS system with PEG and application of this system for metal partitioning and developed separation methodology for separating no-carrier-added ^{183}Re from α-particle irradiated bulk tantalum target.