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Deuteron Bombardment Of ^{103}Rh: A New Promising Pathway For The Production Of ^{103}Pd

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Excitation functions for the deuteron induced reactions on ^{103}Rh as alternative pathway for the production of the medically used ^{103}Pd were studied by the stacked foil technique. Experiments made at incident energies of 15.3 and 20.5 MeV allowed reliable determination of the cross sections for ^{103}Pd and data near threshold for the production of ^{101m,101g}Rh and ^{102m,102g}Rh. The experimental values constitute the first cross section measurements for these reactions. Measurements for ^{103}Pd were done using the X-lines as well as the E_γ= 357.7 keV line. Thick target yields were calculated from the fitted cross section values that are in good agreement with earlier published directly measured values. They show the predicted 100% yield increase over the proton induced reaction in the optimal energy range.

KEYWORDS: ^{103}Pd production, deuteron cross sections, thick target yield, prostate cancer.

I. Introduction

The proportion of patients suffering from prostate cancer treated by permanent brachytherapy is rapidly increasing (more than 40000 in 1998 in the USA) and is expected to reach 50% in 2006[1]. Regular updates on recommendations to sustain the favourable results are published by the American Brachytherapy Society[1,2]. Two radionuclides, ^{125}I and ^{103}Pd, are used for this technique. The accelerator production method for ^{103}Pd nowadays is based on irradiation of rhodium with 18 MeV protons via the reaction ^{103}Rh(p,n)^{103}Pd, followed by a chemical separation of ^{103}Pd.

In a recent study of this reaction we reviewed existing data, improved the status of the cross section database[3], confirmed yields for ^{103}Pd production and established optimal irradiation conditions. Higher yields derived from thick target deuteron irradiations were published by Dmitriev[4] and Mukhammedov[5]. No information was found on cross sections for this deuteron reaction.

The possible contaminants during the production, limiting the upper bombarding energy, are ^{101m,101g,102m,102g}Rh. These nuclides can be separated from ^{103}Pd at the stage of target dissolution, but the high activities involved and their long half-lives could hamper recycling of the target material. Knowledge of the relevant nuclear data is hence important.

II. Experimental Techniques and Evaluation of Data

The cross sections were determined using the stacked foil technique. Experimental set-ups and data evaluation methods were similar as described in earlier publications[3,6].
Only features specific to this new study are discussed here

Two stacks containing high purity 12.3 and 6.8 μm thick Rh foils (>99.9% pure, Goodfellow) and high purity 12.1 μm thick Ti monitor foils (Goodfellow, purity > 99.6%) were activated in the external beam of the VUB-Cyclotron (CGR-560). Both stacks were irradiated in a Faraday-cup-like target holder equipped with a long collimator (effective beam diameter on target is 5 mm) and a secondary electron suppressor. The charge on target was initially derived from the Faraday cup measurement. Less than 6% adaptations in beam current were performed after analysing the results of the monitor foils for the ^{nat}Ti(p,x)^{48}V reaction and using the recommended cross sections of IAEA-TECDOC[7].

The incident energies, with an uncertainty of 0.2 MeV, were derived from our earlier calibrations based on a combination of time of flight and activation techniques[8] and on the results of separate monitoring experiments[6].

High-resolution X- and gamma-spectrometry using 3 types of detectors (LEGe, Extended Range HPGe and HPGe) coupled to Canberra Accuspec boards and Genie 2000 acquisition/analysing software was used to determine the activity of irradiated Rh and Ti foils. No chemical treatment of the samples took place. Several measurements, from shortly after EOB up to after several weeks, allow study of the decay of isomeric states and minimisation of statistical uncertainty for longer lived nuclides.

All cross sections were calculated using the well-known activation formula and are direct formation cross sections taking into account decay during the experiment. Errors due to recoil effects were assessed and are negligible (<1%).

The decay data used for assessment of the activity in the Rh and Ti foils, taken from Browne and Firestone[9] (X-rays and gamma rays), except for ^{102,102m}Rh where we relied on the recently published data of [10], are summarised in **Table 1**. Data for abundances and energies of X-rays of ^{152}Eu, ^{241}Am and ^{129}I, needed for calibration of X-ray detectors (LEGe, Ext Range), were taken from [9,11] after critical compilation of the available literature. Nuclear data needed for the efficiency calibration at higher energy were taken from [9].

Uncertainty on each cross section (8-13%) is estimated as the positive square root of a sum of the contributing sources in quadrature: number of target atoms (5%), nuclear data (5%), beam intensity (3%), efficiency calibration (3- 6%), statistical and peak fitting uncertainty (2-10%).

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Table 1 Decay data of radionuclides, Q-values of processes

Nuclide	$E_{X,g}$ (keV)	I (%)	Contributing reactions	Q (MeV)
^{103}Pd	K_a 20.1+20.2	64.7	^{103}Rh(d,2n)^{103}Pd	-3.6
16.97d	K_b 22.7+23.3	12.3		
	39.75	0.068		
	294.95	0.003		
	357.5	0.022		
	497.1	0.004		
^{102m}Rh	695.6+697.6	45.7	^{103}Rh(d,t)^{102m}Rh	-3.1
2.9y	766	34	^{103}Rh(d,dn)^{102m}Rh	-9.4
	1046.6	33	^{103}Rh(d,p2n)^{102m}Rh	-11.6
^{102g}Rh	468.6	2.6	^{103}Rh(d,t)^{102m}Rh	-3.1
207d	556	1.9	^{103}Rh(d,dn)^{102m}Rh	-9.3
	1103	2.89	^{103}Rh(d,p2n)^{102m}Rh	-11.5
		0.23	IT decay of ^{102m}Rh	
^{101m}Rh	306.8	86	^{103}Rh(d,tn)^{101m}Rh	-10.7
4.34d	545	4	^{103}Rh(d,d2n)^{101m}Rh	-16.9
			^{103}Rh(d,p3n)^{101m}Rh	-19.1
^{101g}Rh	127.2	73	^{103}Rh(d,tn)^{101}Rh	-10.5
3.3y	197.9	70.8	^{103}Rh(d,d2n)^{101}Rh	-16.8
	325	13.4	^{103}Rh(d,p3n)^{101}Rh	-19.0
		7.2	IT decay of ^{101m}Rh	

The effective particle energy in the foils was calculated by the polynomial approximation of Andersen and Ziegler[12]. The uncertainty was estimated taking into account the cumulative errors influencing the calculated energy (incident energy, inhomogeneity and different thickness of the foils, beam straggling).

III. Results and Discussion

1. ^{103}Rh(d,2n)^{103}Pd

The most direct way to assess the activity of ^{103}Pd is measurement of the intense, not resolved, X-ray complexes of the de-excitation of the ^{103}Rh daughter atoms ($K_{\alpha 1}+K_{\alpha 2}$: 64.7%; $K_{\beta 1}+K_{\beta 2}$: 12.34%). Contaminating contributions of the short lived ^{103m}Rh isomer have to be minimised by a waiting time of at least 24 hours. Above the threshold of the (d,dn) reactions the radionuclides ^{102g,102m}Rh can be formed and at higher energies ^{101g,101m}Rh becomes important. The production rate of ^{101g,101m}Rh and ^{102g,102m}Rh in the energy range studied here (see section III 3.) is however low. As also the internal isomeric transition ratios for ^{101m}Rh and ^{102m}Rh, giving rise to Rh X-rays, are only a few percent or less, we can ignore the possible contribution of this isomeric decay mode. Decay of these nuclei to stable Ru gives rise to the emission of K_α and K_β complexes that are shifted 1 keV downward with respect to the Rh X-lines. In this study the contribution is however negligible so that only above 25 MeV any deformation of the peaks or inaccuray in photopeak area determination can be expected.

We probed the possible use of the low abundance γ-rays measurements of ^{103}Pd for determination of cross sections. Obtaining consistent results for X- and γ-measurements requires special care for a number of problems. The most important are: accurate detector calibration over a wide energy range, relevance of measured signals to the determination of the radionuclide of interest, correction for self absortion and reliability of nuclear data.

As the accuracy of the detection efficiency calibration at 39.7 keV is poor for the HPGe used, as the 294.9 keV and 497.1 keV lines are weak and possibly subject to rather high background signals, we only consider here results obtained at 357.5 keV. We investigated the possible presence of contaminating nuclides to the signal at this energy and could not positively identify any systematic contribution.

Repeated measurements of background signals were executed allowing reliable corrections at all energies.

Energy specific counting rates were obtained by the peak fitting algorithm provided by the GENIE 2000 software.

For all measurements of X-lines the calculated cross sections for the 2 energy lines show differences lower than 1%. No significant differences in cross section value for measurement at different times after EOB are observed.

Also for the cross sections determined from the γ-measurement at different times on the detectors Extended Range and HPGe no important scatter is observed indicating that no contamination with radionuclides having a half-life much different from 16.97d is present.

Self-attenuation for the X-lines was taken into account using the mass attenuation coefficients published in [13] (16.0 cm^2/g at 20.2 keV; 11.7 cm^2/g at 23 keV). The correction factors calculated are 1.127 respectively 1.07 for 12.5 μm and 6.5 μm at 20.2 keV, 1.092 respectively 1.05 at 23 keV.

The excitation curve obtained for stacks a and b as determined from the averaged calculated cross sections derived from X-measurements and from the evaluation at E_γ= 357.5 keV show an overall similar behaviour in the energy range studied but differences in absolute values between both measurements are noticed. Over the energy range studied a nearly constant 1.25 ratio for σ_γ/σ_X exists between values obtained from the different measurements.

This behaviour was already remarked in our previous proton experiments[8]. A possible influence of correction factors for X-ray self-absorption is nearly excluded in these new experiments by using thin foils. No difference is found in ratio of σ_γ/σ_X for 12.3 or 6.8μm thick foils.

Although we are aware that all abundances are derived from the decay level scheme we think that the very low abundance for the γ-lines is open to question.

In **Table 2** we hence give 2 cross sections values for the ^{103}Rh(d,2n)^{103}Pd reaction: one derived from the weighted (by γ-line intensity) mean of values obtained on the X-lines, a second one based on the E_γ= 357.5 keV measurements.

A comparison of the X-lines results with a model calculation performed by the Obninsk group using the Alice-IPPE code[14] is shown in **Fig. 1**. The general agreement is surprisingly good but a discrepancy in shape is seen above 12MeV, probably due to improper modelling of the precompound part of the reaction. Threshold and maximal value are well represented.

Table 2 Cross sections of the ^{103}Rh(d,2n)^{103}Pd reaction as function of energy derived from X-lines and E_γ=357.7keV

Cross sections of the ^{103}Rh(d,x)^{101m,101g,102m,102g}Rh reactions as function of energy

Energy (MeV)	^{103}Pd X-lines	^{103}Pd γ-line	^{102m}Rh	^{102g}Rh	Energy (MeV)	^{103}Pd X-lines	^{103}Pd γ-line	^{102g}Rh
20.2 ± 0.2	473 ± 52	565 ± 62	15 ± 1.6	39.3 ± 4	13.4 ± 0.7	997 ± 110	1273 ± 140	13.7 ± 1.5
19.8 ± 0.2	514 ± 56	631 ± 69	13 ± 1.4	33.8 ± 4	13.0 ± 0.3	1091 ± 120	1298 ± 143	14.7 ± 1.6
19.4 ± 0.3	561 ± 62	664 ± 73	11 ± 1.2	31.9 ± 4	12.9 ± 0.7	1034 ± 114	1192 ± 131	12.7 ± 1.4
19.1 ± 0.3	581 ± 64	669 ± 74	7.7 ± 0.8	24.3 ± 3	12.5 ± 0.3	1085 ± 119	1148 ± 126	16.9 ± 1.9
18.8 ± 0.3	606 ± 67	740 ± 81	7.1 ± 0.8	27.7 ± 3	12.4 ± 0.7	1045 ± 115	1367 ± 150	12.3 ± 1.4
18.5 ± 0.3	644 ± 71	808 ± 89	5.2 ± 0.6	26.3 ± 3	12.0 ± 0.4	1008 ± 111	1216 ± 134	10.7 ± 1.2
18.1 ± 0.4	678 ± 75	832 ± 92	4.5 ± 0.5	23.5 ± 3	12.0 ± 0.7	968 ± 106	1150 ± 126	8.5 ± 0.9
17.7 ± 0.4	725 ± 80	923 ± 102	3.4 ± 0.4	23.7 ± 3	11.6 ± 0.7	985 ± 108	1159 ± 128	9.2 ± 1.0
17.3 ± 0.4	787 ± 87	988 ± 109	2.6 ± 0.3	22.9 ± 3	11.5 ± 0.4	972 ± 107	1204 ± 132	10.2 ± 1.1
17.0 ± 0.4	851 ± 94	977 ± 107	1.7 ± 0.2	20.3 ± 2	11.0 ± 0.4	861 ± 95	1132 ± 125	8.2 ± 0.9
16.7 ± 0.5	869 ± 96	1082 ± 119	1.7 ± 0.2	20.4 ± 2	10.4 ± 0.4	906 ± 100	971 ± 107	7.2 ± 0.8
16.3 ± 0.5	916 ± 101	1094 ± 120	1.3 ± 0.1	18.9 ± 2	9.9 ± 0.5	773 ± 85	943 ± 104	5.1 ± 0.6
15.9 ± 0.5	941 ± 104	1123 ± 123	1.4 ± 0.2	17.1 ± 2	9.3 ± 0.5	606 ± 67	881 ± 97	2.6 ± 0.3
15.4 ± 0.5	945 ± 104	1171 ± 129	0.9 ± 0.1	17.0 ± 2	8.7 ± 0.6	682 ± 75	746 ± 82	2.0 ± 0.2
15.0 ± 0.6	1064 ± 117	1292 ± 142	0.9 ± 0.1	17.7 ± 2	8.0 ± 0.7	456 ± 50	508 ± 56	
14.9 ± 0.2	1010 ± 111	1173 ± 129	0.6 ± 0.1	16.2 ± 2	7.3 ± 0.7	388 ± 43	442 ± 49	
14.7 ± 0.6	1036 ± 114	1203 ± 132		18.0 ± 2	6.8 ± 0.8	219 ± 24	268 ± 29	
14.4 ± 0.2	1127 ± 124	1408 ± 155		15.5 ± 2	6.2 ± 0.8	152 ± 17		
14.3 ± 0.6	1027 ± 113	1341 ± 147		18.0 ± 2	5.6 ± 0.9	71 ± 8	67 ± 7	
14.0 ± 0.3	1022 ± 112	1290 ± 142		15.5 ± 2	5.1 ± 0.9	38 ± 4	54 ± 6	
13.8 ± 0.6	1077 ± 119	1384 ± 152		14.5 ± 2	4.6 ± 1.0	15 ± 2		
13.5 ± 0.3	1044 ± 115	1246 ± 137		15.3 ± 2	4.1 ± 1.0	4 ± 0		

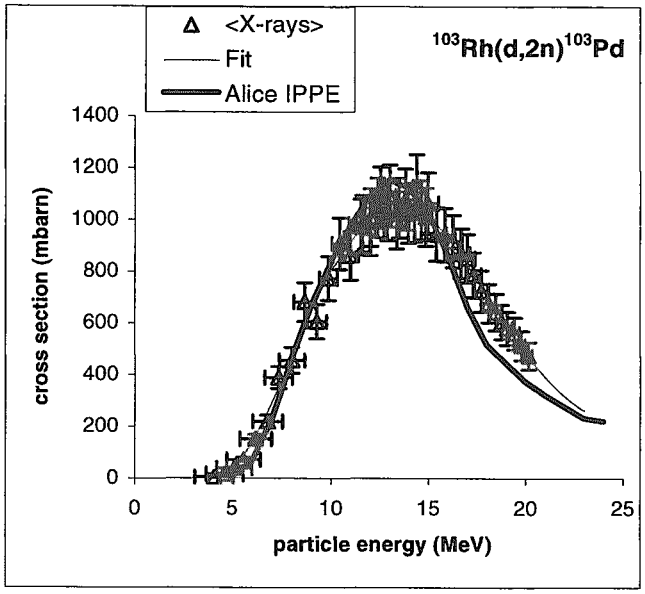

Fig. 1. Excitation function for the ^{103}Rh(d,2n)^{103}Pd reaction: values derived from X-ray measurements, fit to these values and results of the Alice-IPPE model calculation.

2. ^{103}Rh(d,x)$^{102m,\ 102}$Rh

The specific γ-lines emitted allow separate quantification of the activity of both isomers at any time after corrections for the decay of the metastable state into the ground state.

Numerical cross section values for both isomers (weighted values for principal γ-lines) are given in Table 2.

Although the Q-value of the (d,t) reaction is rather low, it is only above 12 MeV (when the (d,p2n) channel contributes fully) that significant amounts of the ground state of this nuclide are formed. Measurable production of the longer lived isomer could only be noticed above 17 MeV.

3. ^{103}Rh(d,x)$^{101m,\ 101}$Rh

As can be expected from the high threshold values, the production of these nuclides is very low in the whole energy region studied (cross section lower than 0.6 mbarn.)

The high threshold of the ^{103}Rh(d,4n)^{101}Pd reaction implies that no measurable activity of this Pd nuclide was found.

4. Metal Contaminants

Special attention was paid to identification of metal radionuclides in this work. Neither traces of activation products of noble metals (Ag, Pt, Au), of the most probable impurity in the foils (In) or of Co radionuclides induced on Fe were found.

IV. Thick Target Yields

A fit to our experimental cross sections derived from X-line measurements for ^{103}Pd is shown in **Fig. 2** together with the measured X-line derived values. The numerical values are given in **Table 3** as are the thick target yields calculated from these values. The comparison of our yields to datapoints directly measured by Dmitriev[4] and Mukhammedov[5] is shown in Fig 2. Our calculated yield at 20 MeV (21.4 MBq/μAh) is marginally lower than the 23.5 MBq/μAh value published in[5] for this energy and the same tendency can be seen over the whole energy range studied.

V. Conclusions

In this work we studied the excitation functions influencing the production of the medically important ^{103}Pd nuclide by deuteron bombardment of pure Rh targets. We determined consistent values for the cross sections over a wide energy range. A 25% difference exists between X-line

Table 3 Calculated thick target yield (MBq/μAh) and fitted cross section (mbarn)

Incident Energy (MeV)	Yield MBq/μAh	fit (mb)	Incident Energy (MeV)	Yield MBq/μAh	fit (mb)
20.0	21.40	495	12.0	6.15	1021
19.5	20.71	549	11.5	5.22	973
19.0	20.00	604	11.0	4.42	937
18.5	19.20	660	10.5	3.60	875
18.0	18.35	716	10.0	2.93	805
17.5	17.47	771	9.5	2.28	727
17.0	16.53	824	9.0	1.75	643
16.5	15.55	875	8.5	1.29	556
16.0	14.53	921	8.0	0.92	467
15.5	13.47	962	7.5	0.62	378
15.0	12.43	997	7.0	0.39	292
14.5	11.34	1025	6.5	0.21	211
14.0	10.30	1045	6.0	0.11	138
13.5	9.20	1056	5.5	0.04	77
13.0	8.17	1056	5.0	0.01	31
12.5	7.15	1045	4.5		4

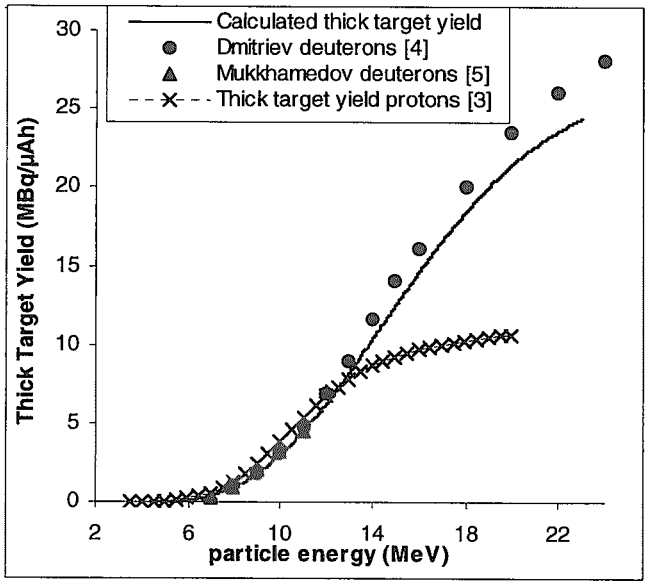

Fig. 2. Calculated thick target yields (total stopping of the beam) at indicated incident energy and comparison with published results for protons and deuterons [3,4,5]

and gamma measurements, probably due to a discrepancy in published values for decay data. Thick target yields derived from our fitted cross sections agree well with those measured earlier[4, 5] and are nearly double at 20 MeV particle energy as those found for proton irradiations[3,4]. For ^{103}Pd production increase of the incident deuteron energy above 20 MeV seems not to be profitable due to two reasons: additional yield at higher energy is negligible (about 10% increase between 20 and 22 MeV) and the contamination with ^{101m,101g,102m,102g}Rh becomes important as their production cross sections rise rapidly above 20MeV. Although both contaminants will be separated from the ^{103}Pd at the stage of recuperation of the expensive Rh target material, accumulation of these long lived nuclides may occur through recycling of targets.

A lower energy limit (also determining the optimal target thickness) could be set at about 9 MeV as only 10% of the theoretical total stopping yield is lost. A practical yield of 19.8 MBq/μAh in the energy range 20-9 MeV deuterons can be expected as compared to 8.8MBq/μAh for an optimal proton energy range of 18-7.5 MeV

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