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Simultaneous production of ^{57}Co and ^{109}Cd in cyclotron

L. Landini,* J. A. Osso Jr.**

Instituto de Pesquisas Energéticas e Nucleares, Travessa R, 400 – Cid. Universitária, 05508-900, São Paulo - SP, Brazil

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^{57}Co and ^{109}Cd simultaneous production was studied, using composite targets of ^{nat}Ni and ^{nat}Ag. The targets were irradiated at the CV-28 Cyclotron, with proton beams of 24 MeV. The average production yields of ^{57}Co and ^{109}Cd were 1179.93 kBq/μA·h, produced by direct and indirect reactions (11.31 days after the EOB) and 71.41 kBq/μA·h (EOB), respectively. The chemical separation procedure was developed in order to obtain a mixed calibration source of ^{57}Co and ^{109}Cd, with a separation yield higher than 80%. The gamma spectroscopy technique was used for the radioactive analysis, using a HPGe detector. The stable elements were identified by atomic absorption spectrophotometry.

Introduction

The radioisotopes produced at cyclotrons are mainly employed in "in vivo" diagnosis in nuclear medicine. They can be administered intravenously, orally or inhalated, after being incorporated into a proper chemical compound or pharmaceutical. High quality images can be obtained in the region or organ of interest, using techniques such as: single photon emission tomography (SPET) and positron emission tomography (PET).

Some radioisotopes can be used in other applications such as calibration sources of X-rays and γ-rays detectors or radioactive tracers. ^{57}Co and ^{109}Cd are examples of such applications.

^{57}Co has a half-life of 271.3 days and decays by electron capture to ^{57}Fe with the emission of γ-rays of 14.40 keV (relative intensity of 7.8%), 122.56 keV (84.8%), 136.43 keV (11.4%) and 692.10 keV (0.2%), along with the characteristic X-ray from the K level of Fe, with energy of 6.5 keV (48%).[1]

^{57}Co can be produced in cyclotrons and in nuclear reactors and it is widely used as calibration source of detectors such as: Ge(Li), Ge(HP), NaI(Tl) and dose calibrators (well type detectors). Besides these applications, ^{57}Co flood sources are used to test the response uniformity of gamma cameras, in nuclear medicine. This is a critical quality control procedure to ensure that the camera response is uniform over the total head area(s). The photon energies of ^{57}Co simulate ^{99m}Tc ($T_{1/2}$ = 6 h; E_γ = 140.5 keV – 85%).[2] Other uses can be named, as: Mössbauer spectroscopy; Schilling test (for studies of anaemia related to depletion and ill absorption of B_{12} vitamin) and labeling of Bleomicyn for tumor detection.

A compact variable energy cyclotron, model CV-28, from The Cyclotron Corporation (TCC), is installed at the Instituto de Pesquisas Energéticas e Nucleares (IPEN-CNEN/SP). Nowadays, this machine accelerates only proton beams with energy between 2 and 24 MeV. ^{57}Co can be produced at a cyclotron using the 4 main particles (p, d, ^{3}He^{++} and α) and among the reactions with protons, i.e., Co+p,[3] Fe+p[4] and Ni+p,[5] the last method is the one that gives the highest production yield and it is the more suitable for use in this cyclotron. The best proton energy range for production of ^{57}Co is 24 to 14 MeV.

^{109}Cd has a half-life of 462.6 days and decays by electron capture to ^{109}Ag with the emission of a γ-ray of 88.03 keV (3.79%) along with the characteristic X-ray from the K level of Ag, with energy of 22.54 keV (102%).[1]

This radioisotope can be employed as calibration source of X-ray and γ-ray detectors and as a radioactive tracer of Cd, an environment pollutant. It can also be used in the energy dispersion X-ray fluorescence (EDXRF) technique.

The production of ^{109}Cd with a proton beam can be performed by two reactions: In+p,[6] that needs energy of about 96 MeV and Ag+p,[7] with an energy range between 14 and 9 MeV, ideal for the cyclotron used in this work.

The objectives of this work were the evaluation of the simultaneous production yields of ^{57}Co and ^{109}Cd using a composite target of Ni and Ag, together with the yields of the radionuclidic impurities of the process and to develop a simultaneous chemical separation of ^{57}Co and ^{109}Cd, in order to prepare a mixed calibration source.

Experimental

All the targets were irradiated in the external beam position at the CV-28 cyclotron of IPEN-CNEN/SP, with a wobbling system and the beam hitting the targets perpendicularly. A 2π water cooling system was used in the target holder.

* E-mail: llandini@net.ipen.br
** E-mail: jaosso@net.ipen.br

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Irradiations of Ni and Ag (composite target)

The experimental production yields measure the quantity of radioisotope (in the case, ^{57}Co and ^{109}Cd) and the main radionuclidic impurities produced, for a fixed charge deposited in the target. ^{nat}Ni and ^{nat}Ag (Goodfellow Metals Ltd.) foils were used in all the irradiations, with the isotopic composition as follows: ^{58}Ni (natural abundance - 68.27%); ^{60}Ni (26.10%); ^{61}Ni (1.13%); ^{62}Ni (3.59%) and ^{64}Ni (0.91%); ^{107}Ag (51.83%) and ^{109}Ag (48.17%), respectively.[8] The area of each foil was 13×13 mm^2. A ^{nat}Cu (^{63}Cu – 69.17% and ^{65}Cu – 30.83%)[8] foil (Goodfellow Metals Ltd.) with a thickness of 50 μm was used in one irradiation, in order to monitor the beam current and energy. The monitor radionuclides analyzed were: ^{62}Zn ($T_{1/2}$ = 9.3 h; E_γ = 596.63 keV – 25.70%) and ^{65}Zn ($T_{1/2}$ = 5856 h; E_γ = 1115.55 keV – 50.75%), produced by the ^{63}Cu(p,2n)^{62}Zn and ^{65}Cu(p,n)^{65}Zn nuclear reactions. The data from KOPECKÝ[9] were used in the calculations. The energy degradation in each foil was estimated using the range equation according to JANNI.[10]

The Ni and Ag foils were irradiated superposed, the Ni foil being the first in the relation to the beam direction. The thickness of Ni and Ag varied between 607 and 662 μm and 228 and 328 μm, respectively. The proton beam degradation was in average from 22.5 to 14 MeV (ideal for ^{57}Co production) in the Ni foil and from 14 to 9 MeV (ideal for ^{109}Cd production) in the Ag foil. The irradiation times varied from 10 minutes to 1 hour and the average beam currents were between 0.77 and 1.5 μA.

Chemical separation of Ni and Ag (composite target)

The method developed for the simultaneous chemical separation of ^{57}Co and ^{109}Cd, from the composite target of Ni and Ag, is decribed as follows.

The targets of Ni (~0.9 g) and Ag (~0.4 g) were dissolved in 20 ml of concentrated HNO_3 and the solution evaporated to dryness and further dissolved with 20 ml of H_2O. The dryness process was repeated up to the solution reached pH between 2 and 7. About 1.25 g of eletrolitic metallic Cu were added and the solution was stirred for 15 minutes. Cu reduces Ag^+ to Ag^0. The reduced Ag and the excess of Cu were filtered by quantitative filter paper and the filtrate, containing ^{57}Co, ^{109}Cd, Ni^{2+}, Cu^+ and traces of Ag^+, was heated to dryness and further dissolved with 8N HCl. This last step was repeated twice. The final solution (10 ml in 8N HCl) was percolated into an anion exchange resin Dowex 1X8 (50–100 mesh, 7 cm high × 1 cm diameter), previously conditioned with 8N HCl. In this condition, Ni^{2+}, Cu^+ and Ag^+ are not adsorbed on the resin while ^{57}Co and ^{109}Cd are. These radioisotopes were later eluted in 0.1N HCl. The elution samples were analyzed by gamma spectroscopy and Cu and Ni were analyzed by atomic absorption spectrophotometry (AAS) in the final solution.

Gamma-spectroscopy

The radioactivity of each radionuclide produced in the irradiation of the targets and in the samples from the chemical separation was measured using HPGe detector, model GX1518, from Canberra, with resolution of 1.8 keV (FWHM), at the 1332 keV peak related to ^{60}Co.

The physical decay characteristics of the nuclides analyzed are shown in Table 1.[1]

Atomic absorption spectrophotometry (AAS)

The analyses were performed in the atomic absorption spectrophotometer model Z-5000 from Hitachi. Calibration curves were obtained with standards of 1, 3, 5, 7, 10 and 20 ppm of Ni and Cu.

Results and discussion

Irradiation of Ni and Ag (composite target)

The experimental production yields of the radioisotopes found in the irradiated foils, ^{55}Co, ^{56}Co, ^{57}Co, ^{58}Co, ^{56}Ni and ^{57}Ni, in the activation of Ni and ^{107}Cd, ^{109}Cd and ^{106m}Ag, in the activation of Ag, are shown in Table 2. The average production yield of ^{57}Co, produced by both direct and indirect (via the decay of ^{57}Ni) reactions, was 1180±221 kBq/μA·h (32± 6 μCi/μA·h), 11.31 days after the EOB. This value is in good agreement with the ones obtained by REIMER and QAIM,[5] SPELLERBERG et al.[11] and RYLOV et al.[12] at the EOB, that used enriched targets: 1400.08 kBq/μA·h (37.84 μCi/μA·h), 370 kBq/μA·h (10 μCi/μA·h) and 370 kBq/μA·h (10 μCi/μA·h), respectively. The average production yield of ^{57}Co through the indirect reaction was 259±35 kBq/μA·h (7.0±1.0 μCi/μA·h), 11.31 days after the EOB. The results obtained for ^{57}Ni had good reproducibility, with value of 48271±6610 kBq/μA·h (1304±178 μCi/μA·h).

Table 1. Physical decay properties of the radionuclides identified in the irradiated samples of ^{nat}Ni and ^{nat}Ag[1]

Radionuclide	Half-life	E_γ, keV	I_γ, %
^{55}Co	17.54 h	931.10	75
^{56}Co	78.76 d	846.77	99.9
^{57}Co	271.3 d	122.56	84.8
^{58}Co	70.78 d	810.77	99.4
^{56}Ni	6.1 d	158.38	98.8
^{57}Ni	36 h	1377.63	81.7
^{107}Cd	6.5 h	22.1	89
^{109}Cd	462.9 d	88.03	3.61
^{106m}Ag	8.46 d	717.3	29.1

Table 2. Average experimental production yields (R), of the radionuclides produced in the ^{nat}Ni(p,*x*) and ^{nat}Ag(p,*x*) reactions, corrected to EOB

Reaction	Radionuclide	R_{dir}, kBq/μA·h	R_{ind},* kBq/μA·h	$R_{dir+ind}$,* kBq/μA·h
^{nat}Ni(p,*x*)	^{55}Co	33410 ± 4700	–	0.74 ± 0.00
	^{56}Co	16.5 ± 3.3	2.6 ± 0.7	16.2 ± 3.3
	^{57}Co	948 ± 240	259 ± 35	1180 ± 221
	^{58}Co	37 ± 8.5	–	33 ± 7
	^{56}Ni	45 ± 12	–	12.5 ± 3.3
	^{57}Ni	48271 ± 6610	–	259.7 ± 35.5
^{nat}Ag(p,*x*)	^{107}Cd	150244 ± 52832	–	–
	^{109}Cd	71.4 ± 5.5	–	–
	^{106m}Ag	0.37 ± 0.00	–	–

* 11.31days after EOB, time for the activity of ^{57}Co reaches its maximum, from the decay of ^{57}Ni.

The total level of impurities of ^{57}Co, ^{56}Co and ^{58}Co, was about 3% and 1.6%, 60 days after the EOB, taking into account the direct and indirect reactions and only the indirect reaction, respectively.

The average production yield of ^{109}Cd measured in this work was 71.4±5.5 kBq/μA·h (1.93±0.15 μCi/μA·h) and it is in good agreement with MENGATTI et al.,[7] 81.4 kBq/μA·h (2.2 μCi/μA·h).

The radionuclidic impurities of ^{109}Cd are easily eliminated after 7 days of decay.

The relative deviation between the value given by the cyclotron's charge and the one obtained in the analyses of the monitor foil was 10%.

Chemical separation of Ni and Ag (composite target)

The chemical separation of the composite target of Ni and Ag had good results. The separation yield of ^{57}Co and ^{109}Cd was higher than 80% in 60 ml of 0.1N HCl. The concentration of Ni and Cu in the final product was (1.6±4.4%) ppm and (9.8±1.3%) ppm, respectively. The chemical separation yields of Ni and Cu were then calculated, with values of 99.99% and 99.95%, respectively. Ni was eluted with 50 ml of 8N HCl, and Cu with more 150 ml of the same elutant. The elution of Ag was followed by measuring the tracer ^{106m}Ag, that was eluted in the first 150 ml of 8N HCl. No ^{106m}Ag was detected in the final solution of ^{57}Co and ^{109}Cd.

Conclusions

The production yields of ^{57}Co and ^{109}Cd obtained in the irradiation of the composite targets of Ni and Ag showed the viability of its use and the resulting optimization of the use of the proton beam, reducing the irradiation time and consequently the costs for production of ^{57}Co and ^{109}Cd. The good results of the chemical separation make it possible to prepare a mixed calibration source of ^{57}Co and ^{109}Cd.

IPEN acquired a new cyclotron, the Cyclone 30, from IBA (Ion Beam Applications) that is capable of accelerating protons with energies between 15 and 30 MeV. The present work will continue with experiments using this new cyclotron.

References

1. E. BROWNE, R. B. FIRESTONE, Table of Radioactive Isotopes, V. S. SHIRLEY (Ed.), John Wiley and Sons, New York, 1996.
2. A. DIAS-NETO, C. P. G. SILVA, J. A. OSSO Jr., E. SOKOLE, Proc. XV Congresso da Assossiação Latino Americana de Sociedades de Biologia e Medicina Nuclear, Lima, Peru, 1997.
3. P. JOHNSON, M. C. LAGUNAS-SOLAR, M. J. AVILA, Intern. J. Appl. Radiation Isotopes, 35 (1984) 371.
4. S. TAKÁCS, F. SZELECSÉNYI, F. TÁRKÁNYI, Proc. Conf. on Cyclotrons and Their Applications 98, Caen, 14-19 June 1998, p. 246.
5. P. REIMER, S. M. QAIM, Radiochim. Acta, 80 (1998) 113.
6. F. M. NORTIER, S. J. MILLS, G. F. STEYN, Appl. Radiation Isotopes, 42 (1991) 1105.
7. J. MENGATTI, E. M. SCAMBATTI, J. B. SUMIYA, V. SCIANI, J. A. OSSO Jr., I.V. CGEN, Vol. 1., Rio de Janeiro, 5–9 July, 1992, p. 145.
8. W. SEELMANN-EGGEBERT, G. PFENNIG, H. MUNZEL, H. KLEWE-NEBENIUS, Karlsruher Nuklidkarte, 1981.
9. P. KOPECKÝ, Intern. J. Appl. Radiation Isotopes, 36 (1985) 657.
10. J. F. JANNI, Proton Range – Energy Tables 1 keV–10 GeV, Atomic Data and Nuclear Data Tables, 27 (4/5), 1982.
11. S. SPELLERBERG, P. REIMER, G. BLESSING, H. H. COENEN, S. M. QAIM, Appl. Radiation Isotopes, 12 (1998) 1519.
12. A. A. RYLOV, I. R. GULAMOV, A. P. PORTYANKO, B. S. YULDASHEV, Proc. 14th Intern, Conf. on Cyclotrons and Their Applications, J. CORNELL (Ed.), Cape Town, 8–13 October 1995, p. 556.