COMPONENT ACTIVATION OF A HIGH CURRENT RADIOISOTOPE PRODUCTION MEDICAL CYCLOTRON

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Abstract

During routine operation of medical cyclotrons intense flux of fast neutrons and gamma rays are produced. The neutrons interact with the cyclotron parts, causing radio activation. The activated cyclotron parts pose considerable radiological hazard during maintenance and waste disposal procedures. In this report the mechanism of cyclotron component activation and the long and short-term prediction thereof have been analysed and related important radiological safety aspects highlighted.

INTRODUCTION

In July 1992 a 30 MeV high current (maximum proton current: 400 micro A) H^--ion medical cyclotron Cyclone30 (IBA Belgium) has been installed at Royal Prince Alfred Hospital (RPAH), the prime teaching hospital of University of Sydney, jointly operated by ANSTO (Australian Nuclear Science and Technology Organisation). In March 1992 the cyclotron commenced its routine production of PET radio radiopharmaceuticals $^{11}CO_2$, $^{13}NH_3$, and ^{18}FDG in aqueous form as well as the SPECT radioisotopes ^{201}Tl, ^{67}Ga and ^{123}I.

During routine operation of medical cyclotrons high flux of fast neutrons are produced at cyclotron targets, where the energetic proton beams interact with the target material to produce the radioisotope of interest. The primary fast neutrons suffer multiple collisions with the vault wall, roof and floor surfaces resulting in their thermalisation via slowing down process [1]. These thermal neutrons fill up the cyclotron, in particular the target vault. Evidently, the cyclotron parts, "immersed" in the thermal neutron cloud suffer activation. In course of long term cyclotron operation, the accumulated activation of the cyclotron parts could cause considerable radiological risk, in particular during the maintenance, waste disposal and handling (i.e. cyclotron part replacement) processes. We have estimated the integrated thermal neutron flux at spots located at selected cyclotron part using cobalt activation pellets at different cyclotron operation conditions, scaled by the integrated target current in the beam-stopper. The pellets were assayed using a gamma spectrometer and the neutron fluences evaluated. Using the neutron fluence data and isotopic composition of the building martial we have calculated the activity of the radioactive species generated in the cyclotron part of interest and reconstructed the gamma dose rate at 1m.

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A practical method for the prediction of component activation and associated gamma dose rates of newly installed cyclotrons has been highlighted. The footprint of the 30 MeV medical cyclotron at RPAH/ANSTO is shown elsewhere [2].

MATERIALS AND METHODS

Estimation of Neutron Fluence

Tiny cobalt (^{59}Co, isotopic abundance 100%) palettes (d = 8mm, t = 1mm, w = 447 mg) were wrapped in polyethylene satchel and attached at selected spots of the cyclotron target-station parts situated in target vault. The locations of the cobalt activation pellets on the cyclotron parts are depicted in Figure 1.

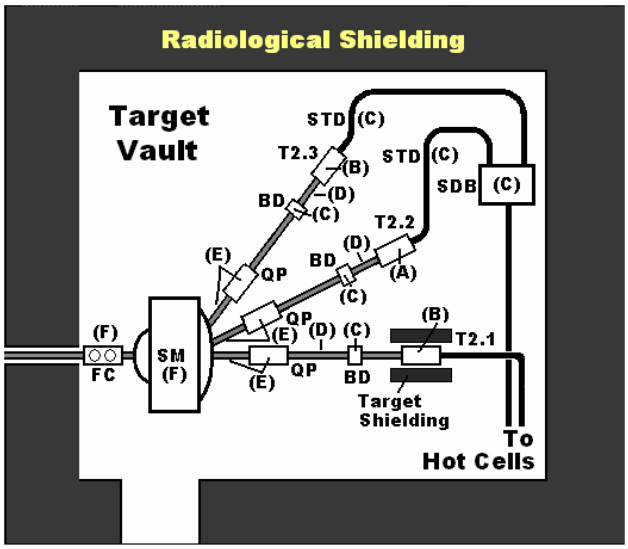

Figure 1: Highlighting the locations of neutron fluence measurement using Cobalt activation pellets: Faraday cup (FC), Switching magnet (SM), Quadrupole lenses (QP), Beam diagnostic ports (BD), Shuttle transfer duct (STD), Shuttle distribution box (SDB) as well as the I-123 (T2.1) and SPECT (TR2.2 and TR2.3) isotope production target stations. The letters in the "( )" brackets indicate the evaluated neutron fluence category as shown in Figure 3.

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The Cobalt pellets were exposed to parasitic neutrons produced during twelve days routine radioisotope production run. During the entire cyclotron operation period the

proton current bombarding the targets was monitored in real-time using the Health Physics Watchdog [3] and the data was stored in a database. The integrated proton current (at Faraday Cup) and irradiation time were estimated to be 15489 μAh and 118 h respectively.

The activation pellets were retrieved after 72 hours cool down time (cyclotron shut down period) and assayed using a high purity germanium (HPGe) detector The activity of the ^{60}Co in the cobalt pellet produced via the thermal neutron capture reaction ^{59}Co(n, γ)^{60}Co was estimated. The thermal neutron fluence rate Φ [cm^{-2}s^{-1}] was evaluated using the formula described elsewhere [4].

$$\Phi = Q\eta^{-1}\sigma^{-1}N^{-1}[1-\exp(-\lambda ti)]^{-1}\exp(-\lambda td)^{-1}10^{-24} \quad (1a)$$

Where

Q = γ-ray count rate of the irradiated cobalt pellet (s^{-1})
σ = thermal neutron capture cross section for ^{59}Co = 37 barn (1barn = 10^{-24} cm^2)
N = number of ^{59}Co atoms in the pellet (equation 1b)
λ = decay constant (equation 1c)
ti = total irradiation time = 118h (Table 1)
td = elapsed time between the end of irradiation and counting begin (cool down time) = 72 h

$$N = Lpkwa^{-1} \quad (1b)$$

Where

L = Avogadro's number = 6.02 × 10^{23} (atoms/mol)
p = fraction of element of in a mixture (for ^{59}Co, p =1)
k = isotopic abundance (for ^{59}Co, k =1)
w = sample weight (g) (for ^{59}Co pellet, w = 0.447 g)
a = atomic weight of sample (for ^{59}Co, a = 59)

$$\lambda = 0.693/T_{1/2} \quad (1c)$$

Where

$T_{1/2}$ = reaction product half life (for ^{60}Co, $T_{1/2}$ = 5.6 y)

The average thermal neutron fluence ϕ_m at seven categories A, B, C, D, E and F were calculated to be 1.5×10^4, 3.4×10^3, 1.8×10^3, 1.1×10^3 , 8.1×10^2 and 6.0×10^2 [cm^{-2}s^{-1}/μAh] respectively.

Building Material of Cyclotron Components

We undertook a thorough check up of all cyclotron components located in the target vault (Figure 1) and found following materials as most important building materials: (a) Copper [5], (b) Aluminium-type 5083 [6], (c) stainless steel-types 304, 316, [7], (d) Brass-types 83600, 86300 [8]. The elemental compositions (percent) of the building materials are summarised in Table 1.

Activity Calculations of Cyclotron Components

Important nucleonic properties major cyclotron building materials used in this report are presented in Table 3. The activities (A [s^{-1}]) generated in the cyclotron components via the thermal neutron capture is given as:

$$A = \Sigma_n \sigma_n N_n(1-\exp(-\lambda_n ti))\exp(-\lambda_n td)\Phi \quad (2a)$$

Table 1: Elemental compositions (percent) of the main cyclotron building materials considered in this work.

Mat.	Copper Pipe	Brass (83600)	Brass (86300)	Steel (304)	Steel (316)	Alu (5083)
Al	---	---	7.5	---	---	93
C	---	---	---	0.08	0.08	---
Cr	---	---	---	20	18	0.25
Cu	99.9	84	66	0.35	0.35	0.1
Fe	---	0.3	4	69	70	0.4
Mg	---	---	---	---	---	4.9
Mn	---	---	5	2	2	1
Mo	---	---	---	0.35	3	---
N	---	---	---	0.1	0.1	---
Ni	---	1	1	10.5	14	---
P	0.1	0.05	---	0.45	0.45	---
Pb	---	6	0.2	---	---	---
S	---	0.08	---	0.03	0.03	---
Sb	---	0.25	---	---	---	---
Si	---	0.005	---	0.75	0.75	---
Sn	---	6	0.2	---	---	
Ti	---	---	---	---	---	0.15
Zn	---	6	28	---	---	0.25

Where

σ_n = thermal neutron capture cross section for nth element in the cyclotron part of interest
N_n = number of the atoms of the nth element (equation 1b)
λ_n = decay constant of the nth reaction (thermal neutron capture) product (equation 1c)
ti = irradiation time [h]
td = cool down time [h]
Φ = thermal neutron fluence rate (equation 1a)

$$\Phi = \phi_m I \quad (2b)$$

Where

ϕ_m = normalised thermal neutron fluence rate of mth category and I = integrated Faraday Cup current [μAh]

A Real Life Example

We have calculated the induced radioactivity in a piece of copper pipe, removed from the beam diagnostic port (BD) of target station T2.1 (Figure 1) after a continuous cyclotron operation (isotope production) time (ti) of 28 days, the integrated (Faraday Cup) current (I) was estimated to be 38800 μAh. The copper pipe (w = 500g) was taken out from the target station 2.2 (Figure 1) after a cool down time (td) of 24 hours. The normalised thermal neutron fluence (ϕ_m) for category A was estimated as 3.4×10^3 [cm^{-2}s^{-1}/μAh]. By substituting the values of ϕ_m and I in equation 2b, the thermal neutron fluence rate at the location of copper pipe was calculated as:

$$\Phi_{Cu\text{-}Pipe} = 1.32\times10^8 \text{ [cm}^{-2}\text{s}^{-1}\text{]} \quad (3)$$

By using the list of cyclotron building materials (Table 1), the isotopic abundance of nuclide species (Table 2) in the material of interest and the formula (equation 1b) we estimated the number of ^{63}Cu (N_{63Cu}), ^{65}Cu (N_{65Cu}) and

^{31}P (N_{31P}) in the 500g copper tube to be 3.27×10^{21}, 1.46×10^{21} and 9.72×10^{18} respectively. Furthermore, by substituting the numerical values of $\Phi_{Cu\text{-}Pipe}$, ti, td, σ (Table 2) and the number of N_{63Cu} (3b), N_{65Cu} (3c) N_{31P} atoms in equation 2a, the activities of the thermal neutron reaction products A_{64Cu} , A_{68Cu} and A_{31P} were calculated as 10.8×10^{8}, 0.0 and 10.2×10^{3} Bq respectively. By using the gamma dose conversion factor Γ [9] of ^{64}Cu to be 3.6×10^{-5}[$mSvh^{-1}MBq^{-1}m^{-1}$] (Table 2) and the activity of ^{64}Cu (10.8×10^{8} Bq), the gamma dose equivalent rate at 1m from the copper pipe was calculated to be 39μSv.h^{-1}. Evidently there was no contribution of ^{32}P (a beta emitter) and ^{68}Cu ($T_{1/2}$ = 5.1 min), which has completely decayed to nil during the 48 hours cool down period. The gamma dose equivalent rate at 1m from the centre of the copper pipe was estimated with a radiation survey meter was found to be 48μSv.h^{-1}.

Table 2: Important nucleonic properties of the main cyclotron building materials including nuclide species, abundance (a), thermal neutron capture cross sections (σ), daughter product of the (n, γ) reaction, half life and the gamma dose conversion factor Γ [$mSv.h^{-1}MBq^{-1}.m^{-1}$].

Nuclide Species	a [%]	σ [b]	React. Prod	Half Life	Γ (Dose Const)
^{27}Al	100	0.232	^{28}Al	2.25m	2.4×10^{-4}
^{13}C	1.1	0.001	^{14}C	5730y	0
^{50}Cr	4.3	15.9	^{51}Cr	27.7d	6.3×10^{-4}
^{54}Cr	2.4	0.36	^{55}Cr	3.5m	0
^{63}Cu	69.2	4.5	^{64}Cu	12.7h	3.6×10^{-5}
^{65}Cu	30.8	2.17	^{66}Cu	5.1m	0
^{54}Fe	5.9	2.3	^{55}Fe	2.73y	0
^{58}Fe	0.28	1.2	^{59}Fe	44.5d	1.8×10^{-4}
^{26}Mg	11	0.036	^{27}Mg	9.5m	1.4×10^{-4}
^{55}Mn	100	13.3	^{56}Mn	2.58h	2.5×10^{-4}
^{92}Mo	14.84	0.019	^{93}Mo	6.9h	8.0×10^{-5}
^{98}Mo	24.13	0.14	^{99}Mo	2.75d	3.0×10^{-5}
^{100}Mo	9.63	0.195	^{101}Mo	14.6m	2.4×10^{-4}
^{58}Ni	68.27	4.6	^{59}Ni	7.6×10^{4} y	0
^{62}Ni	3.59	14.5	^{63}Ni	100y	0
^{64}Ni	0.91	1.58	^{65}Ni	2.5h	8.0×10^{-5}
^{31}P	100	0.18	^{32}P	14.3d	0
^{204}Pb	1.4	0.66	^{205}Pb	1.5×10^{7} y	6.8×10^{-5}
^{206}Pb	52.4	0.49	^{207}Pb	3.3h	0
^{34}S	4.21	0.29	^{35}S	87.2d	0
^{36}S	0.02	0.15	^{37}S	5.05m	0
^{121}Sb	57.4	5.9	^{122}Sb	2.7d	8.2×10^{-5}
^{123}Sb	42.6	4.15	^{124}Sb	60.3d	2.9×10^{-4}
^{30}Si	3.1	0.107	^{31}Si	2.62h	1.3×10^{-7}
^{112}Sn	0.97	0.98	^{113}Sn	115d	4.8×10^{-5}
^{120}Sn	32.59	0.14	^{121}Sn	27h	0
^{122}Sn	4.63	0.18	^{123}Sn	129d	1.0×10^{-6}
^{124}Sn	5.79	0.13	^{125}Sn	9.6d	4.73×10^{-5}
^{50}Ti	5.4	0.177	^{51}Ti	5.8m	7.1×10^{-5}
^{64}Zn	48.6	0.76	^{65}Zn	265d	8.9×10^{-5}
^{68}Zn	18.8	1.0	^{69}Zn	56m	1.2×10^{-9}
^{70}Zn	0.6	0.083	^{71}Zn	2.4m	0

By applying the above method we have estimated the activities in 1kg aluminium (Type 5083) chunks induced by thermal neutrons during 1 year period (integrated Tgt.current 3.7×10^{5} μAh) presented in Table 3.

Table 3: Predicted thermal neutron induced activation [Bq] in 1 kg aluminium chunks (Test Tag) for area categories A, B, C, D, E and F.

	Cr-51	Zn-65	Cu-64	Fe-55	Fe-59
A	4267	1159	473	295	29
B	1051	285	116	73	7
C	446	121	49	31	3
D	327	89	36	23	2
E	245	66	27	17	2
F	178	48	20	12	1

SUMMARY AND CONCLUSIONS

A simple experimental method for the prediction of neutron induced component activation in high-current radioisotope production cyclotrons using tiny cobalt (^{59}Co) activation pellets is presented. The inner space of the target vault was divided in six categories based on thermal neutron fluence rate level. Using the thermal neutron fluence rate (evaluated from ^{60}Co activities) and integrated target current we were able to predict the induced radioactivity generated in different cyclotron parts, commonly made of Copper, Aluminium, Brass and Steel. The gamma dose rate near the activated cyclotron part was calculated. The predicted induced activity (Bq) and the resulting gamma dose rate [μSv.h^{-1}] generated in a 500g copper pipe and 1kg aluminium chunks (test tag) exposed in the target vault was confirmed by measuring the gamma dose rate with a radiation survey instrument. This report ignored the activation products of fast neutron and proton induced reactions like, (n, 2n), (n, α), (p, n) and (p, 2n), prevalent in close proximity of targets, like collimator, Haver-foil window and the target station itself.

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