Air Activation in *Proteus*®*ONE* System

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Outline

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- Argon-41 production
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Introduction

- In a Proton Therapy (PT) system, the interaction of protons with matter (inside cyclotron, degrader, collimators and patients) leads to the production of a large flux of secondary neutrons in the cyclotron room and the treatment rooms.
- Unstable isotopes are produced in air because of nuclear interactions induced by these secondary neutrons, based upon different mechanisms:
  - Spallation reactions by fast neutrons interacting with N, O and Ar atoms present in air.
  - ^{41}Ar production by the capture of thermal neutrons on ^{40}Ar.
- The most frequently produced isotopes with a half-life > 1 min are [1,2]:

  ^{3}H (tritium), ^{7}Be, ^{11}C, ^{13}N, ^{14}O, ^{15}O and ^{41}Ar.
- These 2 processes are treated separately in this report.

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Spallation Products Induced by Neutrons (1)

- The interaction of fast neutrons with N and O atoms in air leads to the production of nuclides by spallation reactions:
  - $^{14}N(n,2n)^{13}N$ $^{14}N(n,^{3}H)^{12}C$
  - $^{16}O(n,2n)^{15}O$ $^{16}O(n,^{3}H)^{14}N$
- These reactions occur for fast neutrons with an energy above a few MeV but the cross section variations with energy are usually not very well known.
- The average cross sections for neutron-induced spallation reactions are given in references [1] and [2].
- These cross sections are integrated over the neutron energy and it is thus impossible to take into account the neutron flux variations with energy.
- In addition, the reaction threshold is not specified.
- These cross sections are summarized in Table 1 for information.

Spallation Products Induced by Neutrons (2)

Table 1: Cross sections for neutron-induced nuclide production in air (from [1,2])

Isotope	$T_{1/2}$	λ (s^{-1})	Parent	s (mb)
^{3}H	12.32 years	$1.8\ 10^{-9}$	N	30
			O	30
^{7}Be	53.22 days	$1.51\ 10^{-7}$	N	10
			O	5
			Ar	0.6
^{11}C	20.33 min	$5.68\ 10^{-4}$	N	10
			O	0.7
			Ar	0.7
^{13}N	9.96 min	$1.16\ 10^{-3}$	N	10
			O	9
			Ar	0.8
^{14}O	1.18 min	$9.79\ 10^{-3}$	O	1
			Ar	0.06
^{15}O	2.04 min	$5.7\ 10^{-3}$	O	40

Calculation of Saturated Activities for Spallation Products (1)

- We follow the methodology described in NCRP-144 [3].
- The production yield R_i for isotope I is given by [3]:

$$R_i = 100 \times Q_{HE} \times N_A \times A_i^{-1} \times f_w^i \times \rho_{air} \times \sigma_i \times L \qquad (1)$$

  where:
  - Q_{HE} = fast neutron emission yield (above production threshold for isotope i).
  - N_A = Avogadro number
  - f_w^i = mass fraction of the parent element i in air
  - A_i = atomic weight for the parent element i
  - ρ_{air} = air density
  - σ_i = production cross section for isotope i
  - L = neutrons path length in air (in meters)
- In the absence of air renewal, the saturated activity are equal to R_i.

Calculation of Saturated Activities for Spallation Products (2)

- In practice, the air is regularly renewed both in the cyclotron and treatment room and the nuclide activities never reach the saturation.
- To take into account the air renewal inside the rooms, one can replace the decay constant λ by an effective constant

$$\lambda' = \lambda + r, \qquad (2)$$

  where r represents the ventilation term, corresponding to the number of air renewal per unit of time [2].
- One can demonstrate that the saturated activity becomes [2]:

$$A'_{sat} = A_{sat} \times \lambda / \lambda' = A_{sat} \times \lambda / (\lambda + r) \qquad (3)$$

- In our estimations, we consider the following air renewals:
  - Every 10 minutes in the S2C2 vault ➔ $r_{S2C2} = 1/600\ s^{-1}$
  - Every 10 minutes in the treatment room (CGTR) ➔ $r_{CGTR} = 1/600\ s^{-1}$

Calculation of Saturated Activities for Spallation Products (3)

- In relationship (1), the isotope production yields depend upon the product $Q_{HE}.\sigma_i$.

- As the cross sections σ_i vary with the neutron energy, it is necessary to replace the term $\sigma.Q_{HE}$ by the integral $\int$⍰$(E_n).\sigma_i(E_n).dE_n$ computed for each isotope i.

- It is thus necessary to estimate the neutron fluxes $\Phi(E_n)$ obtained inside the S2C2 and CGTR rooms taking into account the various radiation sources existing in the *Proteus*®*ONE* system.

Calculation of Saturated Activities for Argon-41 (1)

- The nuclide Argon-41 ($T_{1/2}$ = 1.83 hours) is produced by a thermal neutron capture (n,γ) process on isotope Argon-40.
- To compute the reaction rate $^{40}Ar(n_{th},\gamma)^{41}Ar$, we follow the procedure described in publication NCRP-144 [3]:

$$R = \Phi_{th} \times N_A \times A^{-1} \times f_w \times \rho_{air} \times \sigma \qquad (4)$$

where:

N_A = Avogadro number
A = atomic weight for Argon-40
f_w = mass fraction of Ar in air = 0.013
ρ_{air} = air density
σ = cross section = 660 mb [2]
Φ_{th} = thermal neutron flux inside the room

Calculation of Saturated Activities for Argon-41 (2)

- The thermal flux is given by the relationship [3]:

$$\Phi_{th} = 1.25\, Q_F / S \qquad (5)$$

where Q_F is the total neutron yield (n/s) et S the inner surface of the room (cm^2).

- As for the spallation products, the saturated activity is computed by taking into account an air renewal every 10 minutes in S2C2 and CGTR rooms.

Basic Data

- Based upon the relationships (1), (4) and (5), a lot of basic data are required to perform the air activation determination:

  - Assumption on air composition and density;
  - Neutron path lengths, room volumes and wall surfaces;
  - Cross sections as a function of neutron energy for the spallation reactions;
  - Neutron fluences in the S2C2 and CGTR rooms considering the various radiation sources.

- We will present these various data in the following pages.

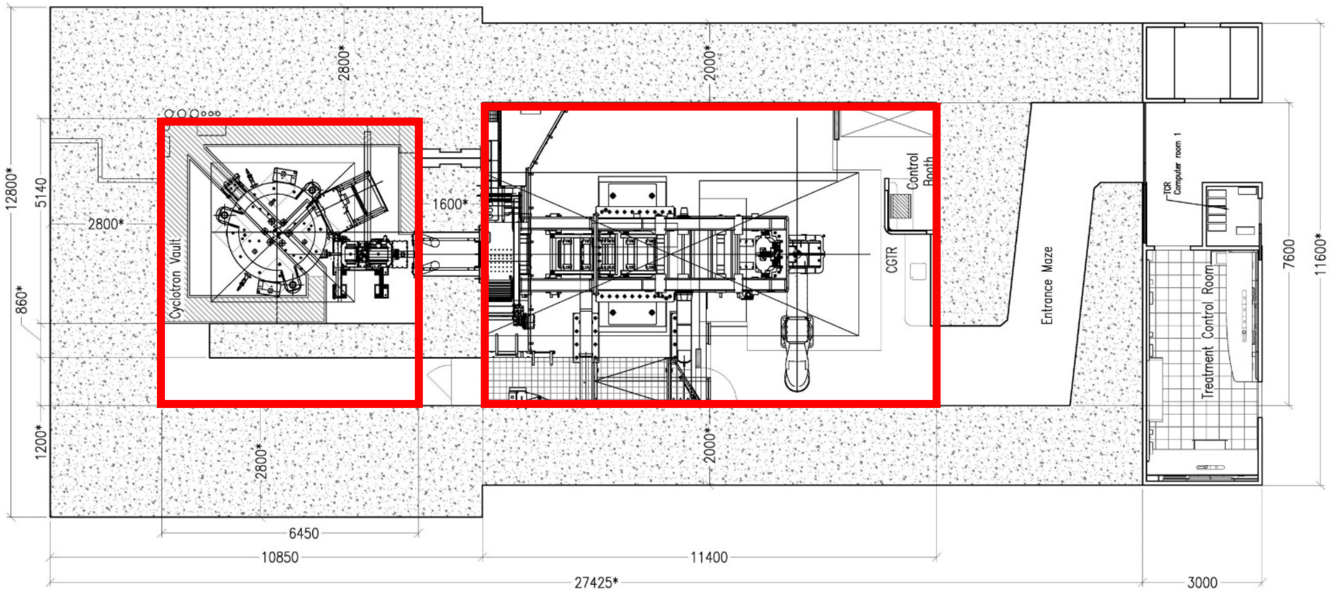
Basic Data: Air properties

- We use the following properties for air:
  - Density = 0.0013 g/cm³
  - Atomic composition

Isotope	A	f_W
N	14.0067	0.755
O	15.9994	0.232
Ar	39.948	0.013

Basic Data: Room Properties (1)

- We use the standard *Proteus®ONE* layout to derive the room volumes and wall surfaces.



Room	S2C2	CGTR
Room Volume (m³)	180	650
Wall Surface (m²)	55.5	288

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Basic Data: Room Properties (2)

- To evaluate the neutron path length, we make the assumption that all the radiation sources are located at the center of either the S2C2 or the CGTR rooms.
- This assumption is the truth for the S2C2, the momentum slit and the patient.
- On the contrary, the degrader, collimator and divergence slits are located on the border of their room. Then, the path length is two times longer but the neutron fluence must be divided by 2 as half of the neutrons will directly hit the 1.6 m thick separation wall between the S2C2 and CGTR rooms.
- Therefore, considering that all the radiation sources are located at the center of the room is a valid assumption.
- In addition, we replace the parallelepiped rooms by a sphere corresponding to the same volume and use the sphere radius as average path length.

Room	S2C2	CGTR
Neutron path length (m)	3.5	5.4

Cross Sections for Spallation Products Induced by Neutrons (1)

- It is possible to find detailed information on the cross sections for spallation reactions taking place in air in references [4] and [5].

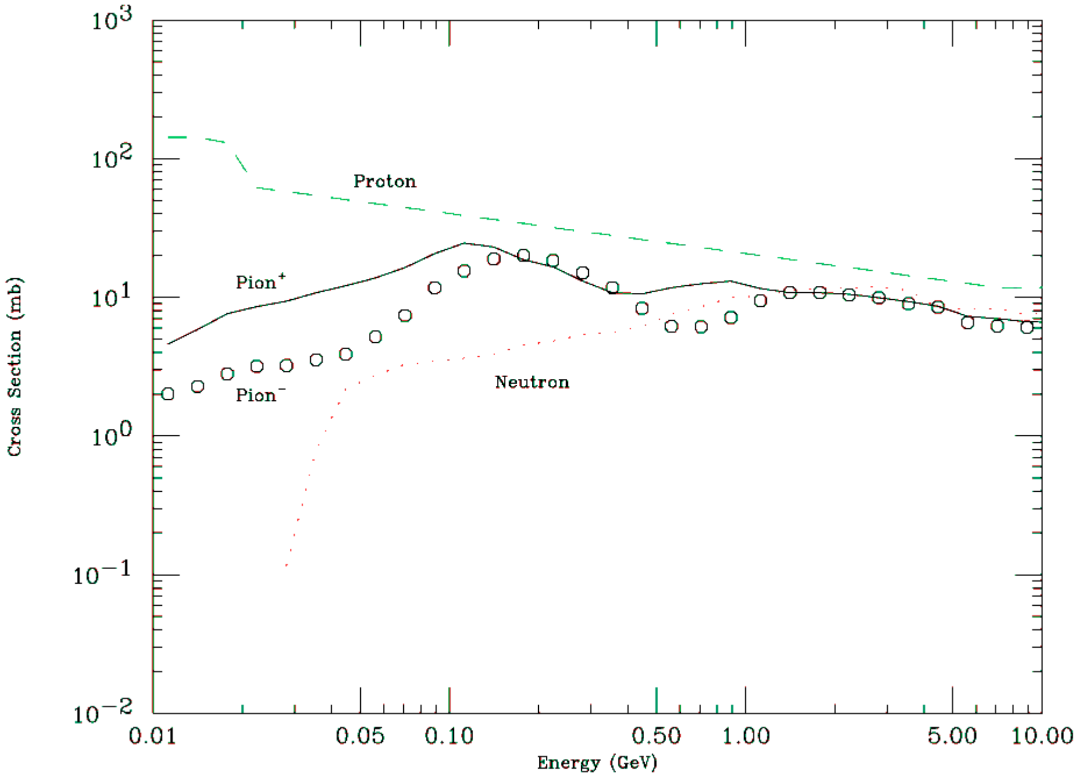
- These studies performed in the framework of LHC and NLC use either measured data when available or FLUKA simulations in the other cases.

- Data are available for the following reactions:

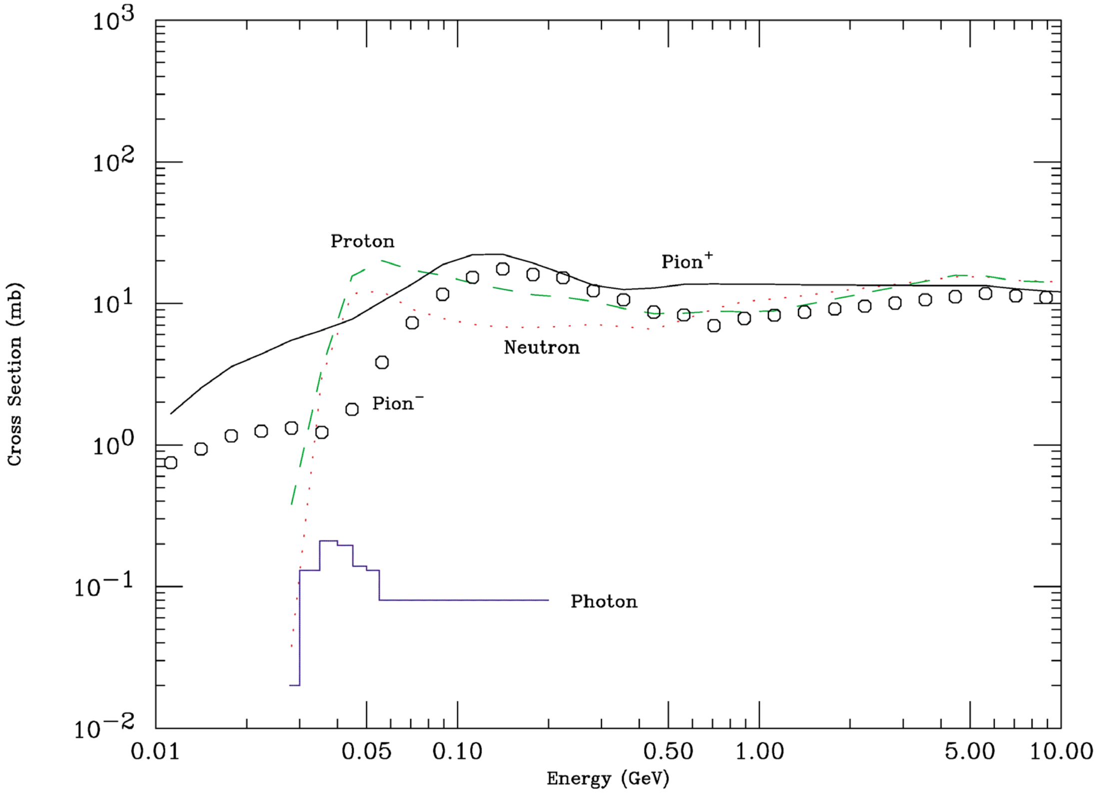
  - $^{16}O(n,X)^{11}C$ and $^{14}N(n,X)^{11}C$
  - $^{16}O(n,X)^{3}H$ and $^{14}N(n,X)^{3}H$
  - $^{16}O(n,X)^{7}Be$ and $^{14}N(n,X)^{7}Be$
  - $^{14}N(n,X)^{13}N$
  - $^{16}O(n,X)^{15}O$ and $^{16}O(n,X)^{13}N$

Cross Sections for Spallation Products Induced by Neutrons (2)

$^{14}N(n,X)^{11}C$



$^{16}O(n,X)^{11}C$



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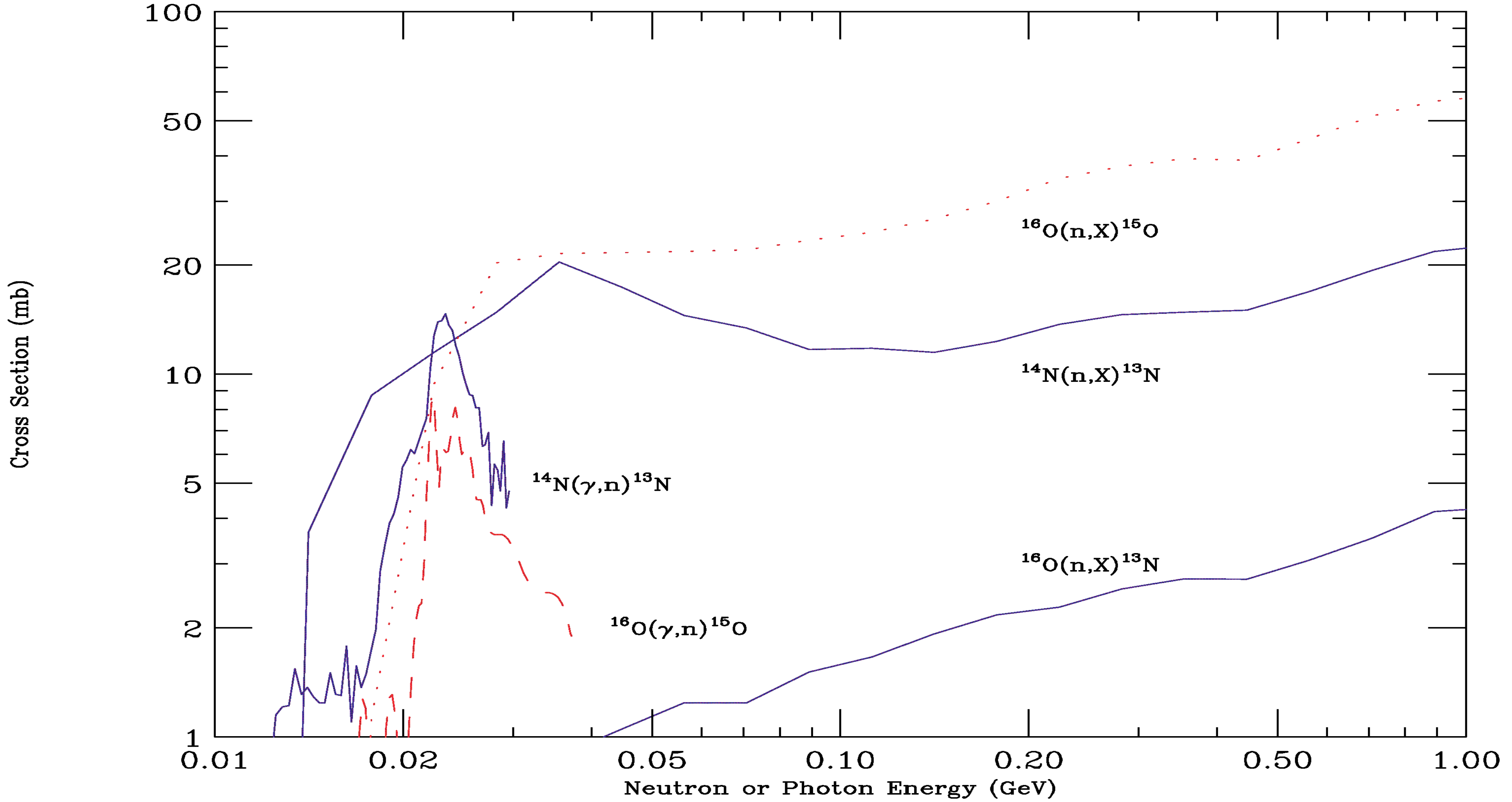
Cross Sections for Spallation Products Induced by Neutrons (3)

^{15}O and ^{13}N Production



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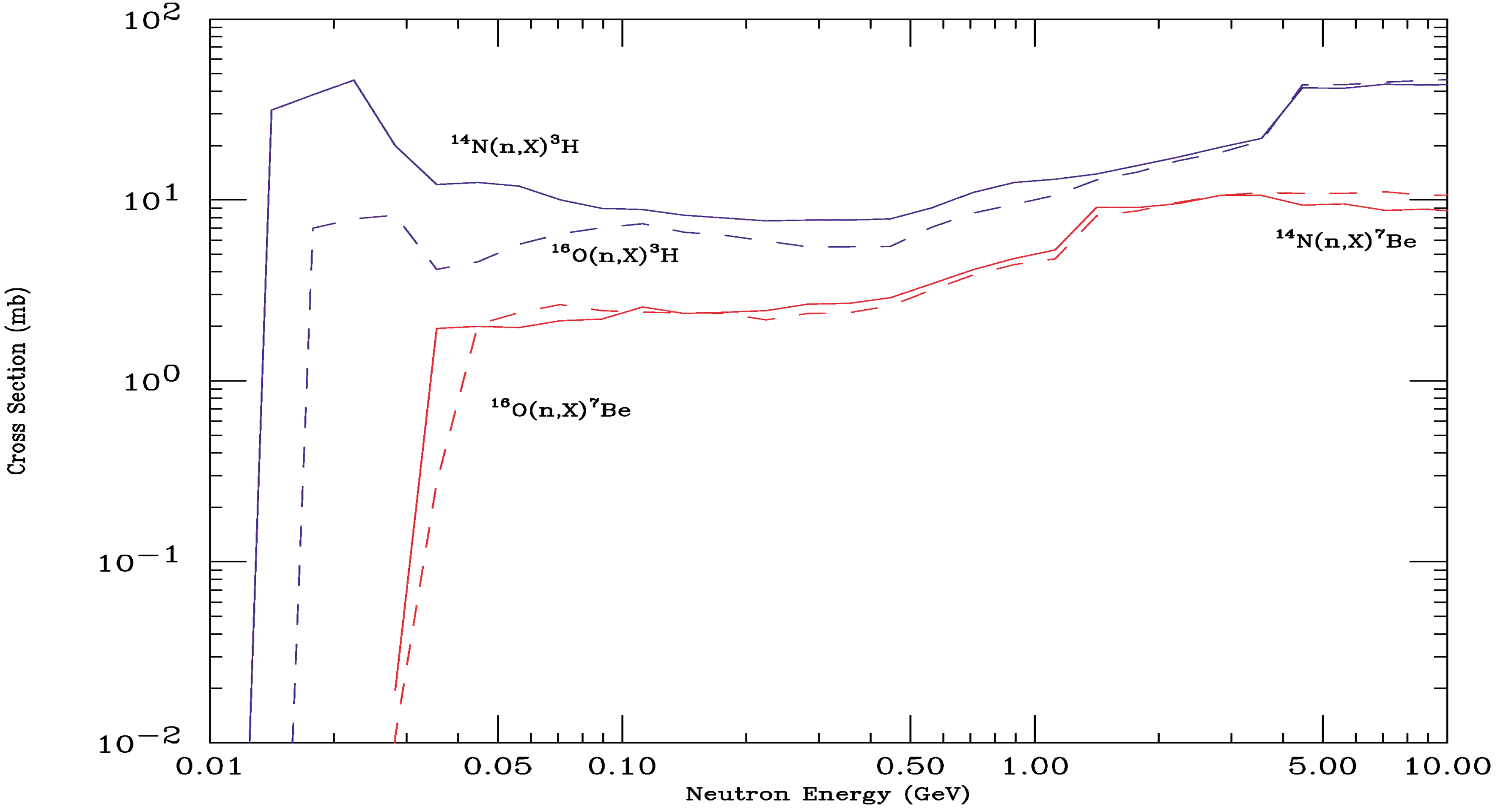
Cross Sections for Spallation Products Induced by Neutrons (4)

^{3}H and ^{7}Be Production

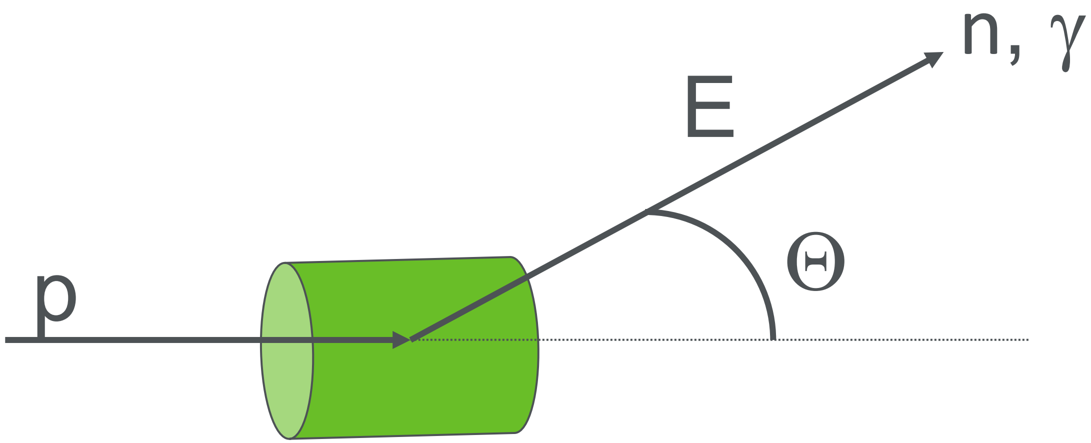


Determination of Neutron Fluences (1)

- The production yields for the different nuclides in air depend upon the fast and total neutron production yields (see relationships (1) and (5)).
- These yields are obtained from the average neutron fluences determined inside each room under the assumption that the neutrons are emitted from the centre of the room with an isotropic emission.
- One needs to take into account all the major sources active in the different rooms to compute these fluences, as describe in document « Radiation Sources in the *Proteus*®*ONE* System », F. Stichelbaut, M-ID 36694, September 2014.
- One than compute the integral $\int \square(E_n).\sigma_i(E_n).dE_n$ for each isotope i et use these values in the relationship (1).
- First, the neutron fluences are determined for a single proton using a simple model as a function of target material and beam energy ➔ pages 20 to 23.
- Next, these fluences are weighted by the annual workloads determined for each radiation sources ➔ pages 24 to 26.

Determination of Neutron Fluences (2)

- The fluxes of neutrons produced by protons are computed using Monte Carlo code MCNPX 2.7.0.
- The target is a cylinder with length = radius ≥ proton range (Thick target).



- MCNPX computes the energy (E) and the polar angle (Θ) of the emitted neutrons.
- The neutron/photon fluxes have been computed for the various targets and beam energies:
  - Degrader ➔ Beryllium (Be)
  - Collimator ➔ Tantalum (Ta)
  - Divergence and momentum slits ➔ Nickel (Ni)
  - Patient ➔ Human Tissues

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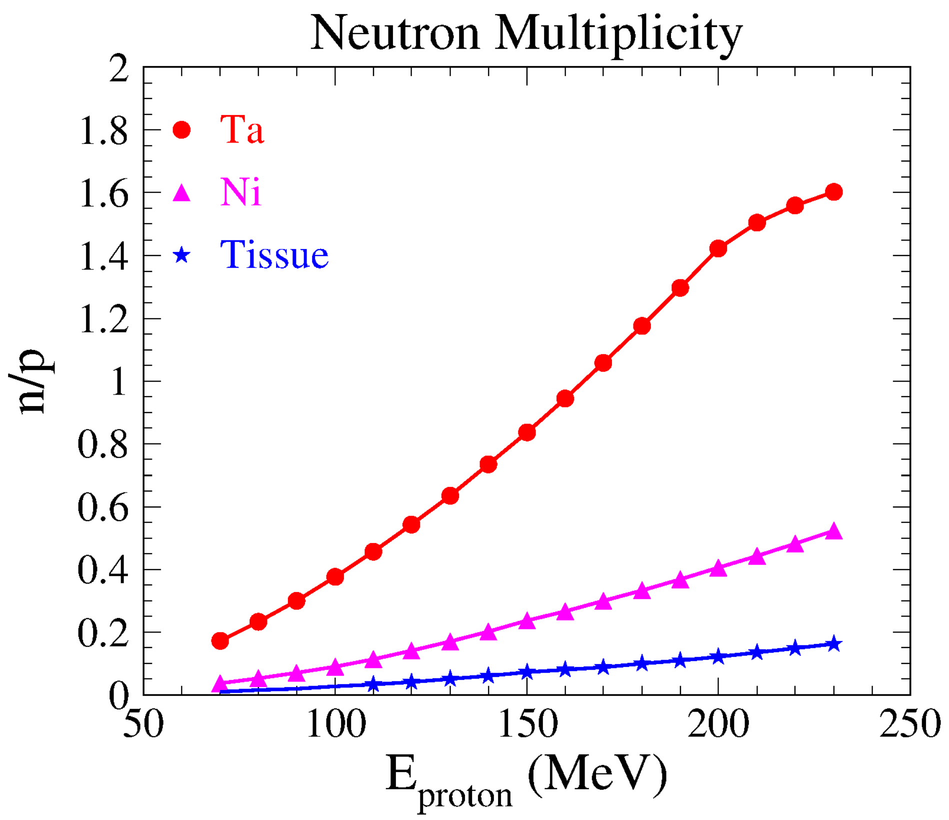
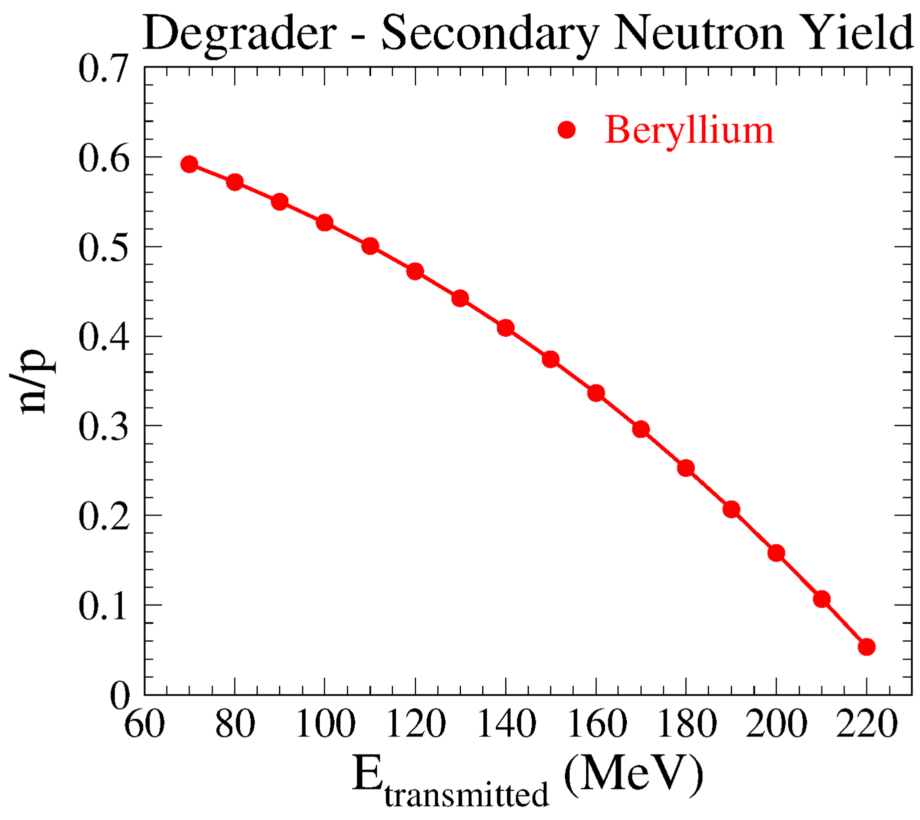
Determination of Neutron Fluences (3)

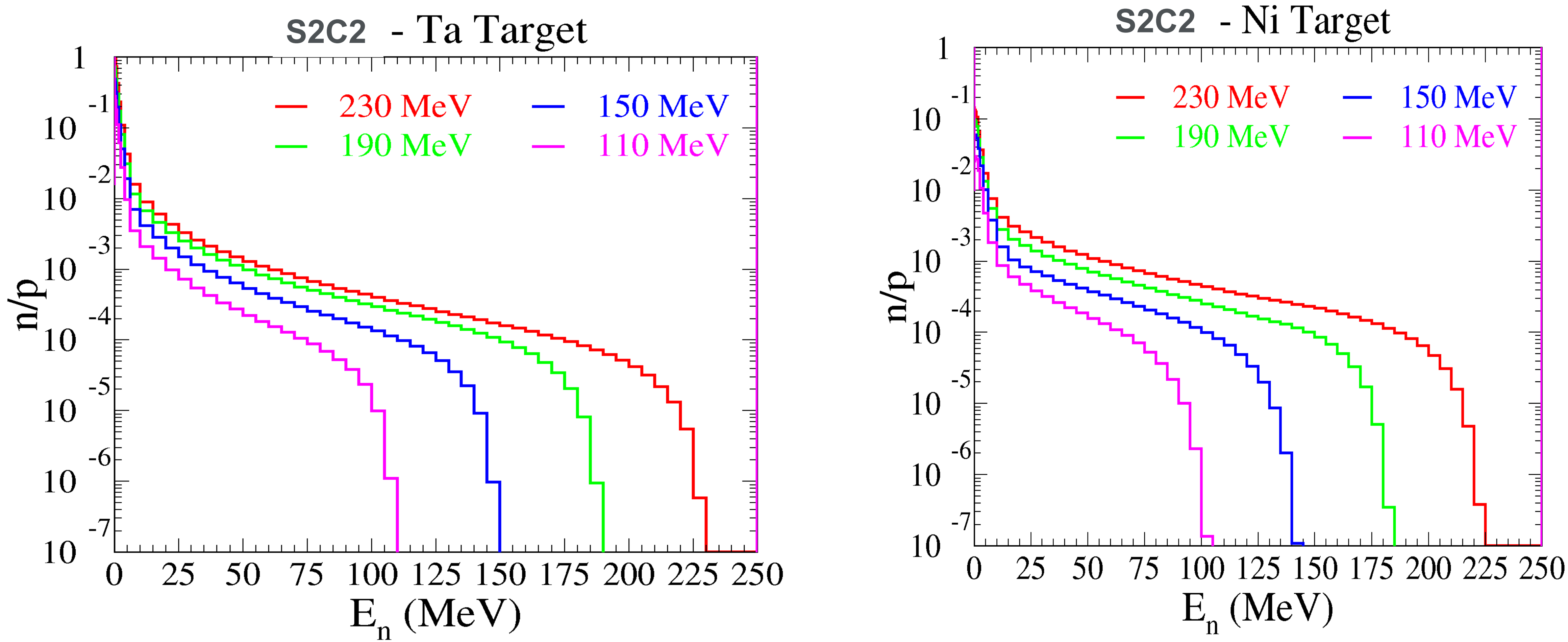
- Production rates for neutrons, n/p, strongly vary with target type and beam energy.
- Neutron production yields also strongly increase with Z.





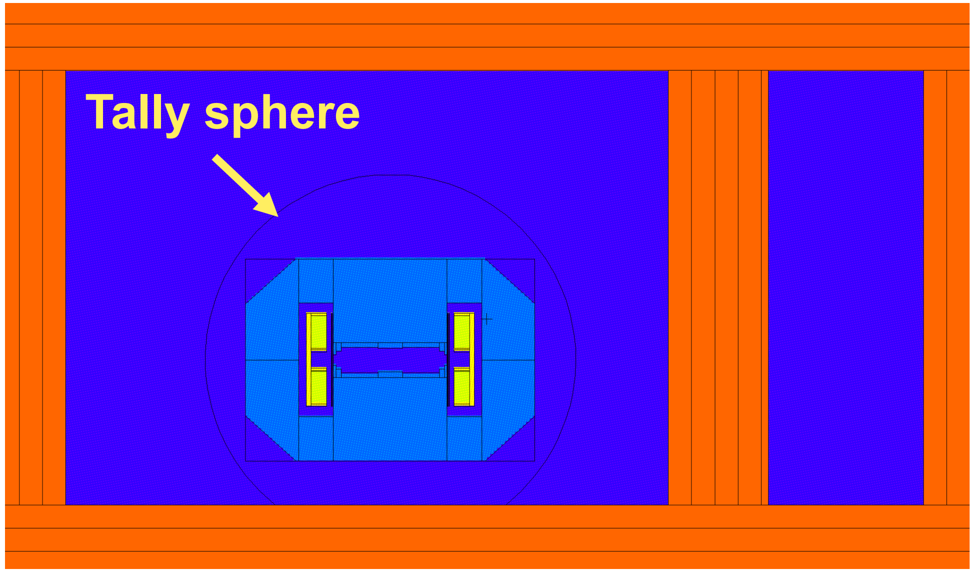
Determination of Neutron Fluences (4)

Global neutron spectra (Θ averaged) for Ta and Ni targets



Determination of Neutron Fluences (5)

- The model used for standard sources cannot be applied to the S2C2 case because neutrons produced inside the cyclotron need to cross the steel yoke before reaching the air. As this yoke has a minimal thickness of 45 cm, high-energy neutrons will be strongly slowed down due to inelastic interactions.
- To determine the neutron fluxes leaving the S2C2, we use a sphere surrounding the cyclotron and tally the neutron fluxes per lost proton.
- Both sources (septum and uniform losses) lead to the same spectra and the same yields n/p = 0.779.
- Therefore, the two sources are treated as a single one with a strength corresponding to 70% of the accelerated beam.



Workload Determination (1)

- The workloads represent the amount of beams impinging on the various beamline elements and generating secondary neutrons.
- The workloads are expressed in nA.h, with 1 nA.h = 3600 nC.
- The workloads used for air activation are based upon the workloads used for shielding design as described in M-ID 64063 (section 5).
- We start from the workloads delivered at the isocenter and corresponding to the patient case mix described in M-ID 64063.
- The shielding design is based on the use of complex SOBP's to describe the clinical indications, each SOBP corresponding to about 20 energy layers. To simplify the calculations of air activation while remaining conservative, we follow the following approach:
  - For the radiation sources downstream of the degrader, we select the distal energy for each SOBP and approximate it to the upper value in multiples of tens ➔ selected energies = 230, 210, 190, 170 and 130 MeV. We have demonstrated in M-ID 64063 Appendix 7 that the use of these distal energies leads to an overestimation of annual doses ranging between 11% and 45% depending upon the location.
  - For the S2C2 and the degrader, it is the use of the minimal energy of the SOBP's that leads to the most conservative approach. However, we have shown in Appendix 3 of M-ID 64063 that the use of the averaged SOBP energies leads to a more realistic situation, close to the real situation obtained by considering the full SOBP's. As for the shielding design, we use that approach based upon the average SOBP energies to evaluate the workloads for the S2C2 and the degrader.

Workload Determination (2)

- Workloads (in nA.h) derived for the different radiation sources after the energy degrader: collimator, divergence and momentum slits, patient.
- We start from the workloads at isocenter (W_{iso}) and divide them by the ESS transmission efficiency (ε(ESS)) to obtain an estimation of the workload extracted from the S2C2 ($W_{extract}$). Then, this $W_{extract}$ is multiplied by the amounts of beam losses in the collimator and slits to obtain the W_{coll} and W_{slit} workloads, respectively.
- The divergence and momentum slits are summed together as they are both made of Ni.

Table 2: Workloads for the radiation sources after the energy degrader

E (MeV)	W_{iso}	ε(ESS) (%)	$W_{extract}$	Loss(Coll) (%)	W_{coll}	Loss(Slit) (%)	W_{slit}
230	28.63	12.07	237.2	36.32	86.2	10.40	24.7
210	23.22	7.33	316.8	41.45	131.3	9.82	31.1
190	15.29	4.18	365.8	45.06	164.8	8.62	31.5
170	29.64	2.61	1135.6	47.15	535.5	6.79	77.1
130	39.63	1.12	3538.4	45.78	1619.9	3.24	114.6

Σ = 136.41 nA.h

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Workload Determination (3)

- Workloads derived for the energy degrader (W_{deg}) and the S2C2 (W_{S2C2}) based upon the average energy approach.
- The W_{S2C2} represent the amount of beam lost inside the machine, based upon an extraction efficiency of 30%. The septum and uniform losses are summed together (see M-ID 36694 for a description of these losses).

Table 3: Workloads for the S2C2 and degrader radiation sources

E (MeV)	W_{iso}	ε(ESS) (%)	W_{deg}	W_{S2C2}
180	5.25	5.91	88.86	207.34
170	24.04	4.96	484.68	1130.92
150	23.22	3.16	735.74	1716.73
140	4.59	1.98	231.82	540.91
110	10.04	1.43	702.10	1638.23
100	29.64	1.19	2486.58	5802.02
70	39.63	0.545	7271.56	16966.97

Σ = 136.41 nA.h

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Calculations of Air Activation (1)

- After detailing the data needed to compute the air activation, here is a description of the followed methodology.
- For the nuclides produced by spallation:
  - The MCNPX neutron fluences are folded with the energy-dependent cross sections to determine the integrals $\int$⍰$(E_n).\sigma_i(E_n).dE_n$ per incident proton for each source, each energy and each process i.
  - These integrals are then weighted by the corresponding workloads.
  - All the sources and energy values are summed together for each process to obtain the annual weighted flux $Q_{HE}.\sigma$ needed in relationship (1) and divided by the facility annual operating time T=4800 h/year to obtain the neutron flux in n/s.
  - Then relationship (1) is used to compute the nuclide production yields followed by relationship (3) to obtain the correct saturation activities for each production process.
  - Finally, the activities coming from different parents are summed together to obtain the total saturation activity for each nuclide type.
- The results are presented in Tables 4 and 5 for the S2C2 and CGTR rooms, respectively.

Calculations of Air Activation (2)

- For ^{41}Ar production:

  - We use the neutron multiplicities n/p weighted by the annual workloads for each source and each energy to derive the corresponding annual neutron fluences.

  - These fluences are summed together for each room and divided by the annual operating time T to obtain the quantity Q_F needed in relationship (5).

  - The thermal flux Φ_{th} is obtained thanks to relationship (5).

  - Finally, the ^{41}Ar production rates and saturation activities are derived using relationships (4) and (3), respectively.

- The results are presented in Table 6.

Results: Spallation Products in S2C2 Room

- Table 4: Activities obtained for the different nuclides produced by spallation inside the S2C2 room

Nuclide	Parent	Production yield (atoms/m³.s)	Saturated Activity (Bq/m³)	Total Activity (Bq/m³)
^{3}H	N	$7.03\ 10^{3}$	$7.60\ 10^{-3}$	$8.27\ 10^{-3}$
	O	$6.19\ 10^{2}$	$6.68\ 10^{-4}$	
^{7}Be	N	$6.12\ 10^{2}$	$5.54\ 10^{-2}$	$7.04\ 10^{-2}$
	O	$1.65\ 10^{2}$	$1.49\ 10^{-2}$	
^{11}C	N	$8.99\ 10^{2}$	$2.29\ 10^{2}$	$3.80\ 10^{2}$
	O	$5.97\ 10^{2}$	$1.52\ 10^{2}$	
^{13}N	N	$5.16\ 10^{3}$	$2.12\ 10^{3}$	$2.16\ 10^{3}$
	O	$1.04\ 10^{2}$	$4.25\ 10^{1}$	
^{15}O	O	$2.07\ 10^{3}$	$1.60\ 10^{3}$	$1.60\ 10^{3}$

Results: Spallation Products in CGTR Room

- Table 5: Activities obtained for the different nuclides produced by spallation inside the CGTR room

Nuclide	Parent	Production yield (atoms/m^3.s)	Saturated Activity (Bq/m^3)	Total Activity (Bq/m^3)
^{3}H	N	$2.23\ 10^{1}$	$2.41\ 10^{-5}$	$2.59\ 10^{-5}$
	O	$1.66\ 10^{0}$	$1.79\ 10^{-6}$	
^{7}Be	N	$1.43\ 10^{0}$	$1.29\ 10^{-4}$	$1.64\ 10^{-4}$
	O	$3.81\ 10^{-1}$	$3.45\ 10^{-5}$	
^{11}C	N	$1.95\ 10^{0}$	$4.95\ 10^{-1}$	$8.69\ 10^{-1}$
	O	$1.47\ 10^{0}$	$3.75\ 10^{-1}$	
^{13}N	N	$1.46\ 10^{1}$	$6.00\ 10^{0}$	$6.09\ 10^{0}$
	O	$2.10\ 10^{-1}$	$8.63\ 10^{-2}$	
^{15}O	O	$5.14\ 10^{0}$	$3.98\ 10^{0}$	$3.98\ 10^{0}$

Results: Argon-41 production

- Table 6: Saturated ^{41}Ar activity obtained in the S2C2 and CGTR rooms.

^{41}Ar	Production Yield (atoms/s.cm^3)	Saturated Activity (Bq/m^3)
S2C2	$1.50\ 10^{-2}$	$8.91\ 10^{2}$
CGTR	$9.01\ 10^{-6}$	$5.35\ 10^{-1}$

Release Limits (1)

- The derived saturation activities for the different unstable nuclides need to be compared to the release limits imposed by the legislation.
- As this legislation can vary from one country to another, it is difficult to validate our results.
- However, we can compare our results to the recent 'Ordonnance sur la radioprotection' (ORaP) published in April 2017 by the Swiss Confederation [6]. The appendix 3 of that publication provides a comprehensive list of radioprotection data for all the major nuclides, resulting from a compilation of IAEA safety guides and ICRP publications.
- Of particular interest are two quantities:
  - $\mathbf{e_{inh}}$ is the effective engaged dose coefficient for inhaled particulates (5 µm) in Sv/Bq.
  - **CA-20 mSv** is the activity concentration resulting in an effective engaged dose of 20 mSv for workers inhalating this air for 40 hours/week, 50 weeks/year (2400 m^3/y).
- We derive a limit **CA-1 mSv** corresponding to an effective engaged dose of 1 mSv/y for non-workers assuming a presence of 4800 h/year (the annual operating time for the PT center) ➔ CA-1 mSv = CA-20 mSv / 20 / 2.4

Release Limits (2)

- The CA-1 mSv limit can be directly compared to our saturated activities as they are expressed in Bq/m³.
- The e_{inh} coefficient can be used to estimate the effective engaged dose due to the inhalation of the air released by the PT center:

$$Ei = e_{inh} \times Ai \times F \times T \qquad (6)$$

where:

- Ai is the saturated activity for nuclide i (Bq/m³)
- F is the inhalated air volume = 1.2 m³/h
- T is the operating time per year = 4800 h/y

Release Limits (3)

- Table 7: Yearly effective doses and activity concentration limits

Room	Nuclide	T1/2	Saturated Activity (Bq/m^3)	e_{inh} (Sv/Bq)	Yearly Dose (µSv/y)	CA-20 mSv (Bq/m^3)	CA-1 mSv (Bq/m^3)
CGTR	H-3	12.32 y	$2.59\ 10^{-5}$	$1.8\ 10^{-11}$	$2.69\ 10^{-6}$	$5\ 10^{9}$	$1.04\ 10^{8}$
	Be-7	53.22 d	$1.64\ 10^{-4}$	$4.6\ 10^{-11}$	$4.35\ 10^{-5}$	$5\ 10^{5}$	$1.04\ 10^{4}$
	C-11	20.39 min	$8.69\ 10^{-1}$	$3.2\ 10^{-12}$	$1.60\ 10^{-2}$	$2\ 10^{5}$	$4.17\ 10^{3}$
	N-13	9.965 min	6.09		NA	$7\ 10^{4}$	$1.46\ 10^{3}$
	O-15	122.24 sec	3.98		NA	$7\ 10^{4}$	$1.46\ 10^{3}$
	Ar-41	109.61 min	$5.35\ 10^{-1}$		NA	$5\ 10^{4}$	$1.04\ 10^{3}$
S2C2	H-3	12.32 y	$8.27\ 10^{-3}$	$1.8\ 10^{-11}$	$8.57\ 10^{-4}$	$5\ 10^{9}$	$1.04\ 10^{8}$
	Be-7	53.22 d	$7.04\ 10^{-2}$	$4.6\ 10^{-11}$	$1.87\ 10^{-2}$	$5\ 10^{5}$	$1.04\ 10^{4}$
	C-11	20.39 min	$3.80\ 10^{2}$	$3.2\ 10^{-12}$	7.	$2\ 10^{5}$	$4.17\ 10^{3}$
	N-13	9.965 min	$2.16\ 10^{3}$		NA	$7\ 10^{4}$	$1.46\ 10^{3}$
	O-15	122.24 sec	$1.60\ 10^{3}$		NA	$7\ 10^{4}$	$1.46\ 10^{3}$
	Ar-41	109.61 min	$8.91\ 10^{2}$		NA	$5\ 10^{4}$	$1.04\ 10^{3}$

Release Limits (4)

- Comments:
  - Air released from the CGTR:
    - The effective dose resulting from the long-lived isotopes (^{3}H,^{7}Be, ^{11}C) is negligible.
    - For the other isotopes (^{13}N, ^{15}O, ^{41}Ar) the largest dose is due to immersion inside the released air and we can compare the released activities to the CA-1 mSv limit. As the released activities are more than 200 times smaller than the CA-1 mSv limit, we can assume that the air released from the CGTR does not represent any hazard for the surrounding population.
  - Air released from the S2C2 room:
    - The effective dose resulting from the long-lived isotopes (^{3}H,^{7}Be, ^{11}C) is small, with a maximal value of 7 µSv.
    - The saturated activities obtained for the 3 other isotopes ^{13}N, ^{15}O, ^{41}Ar are about the same magnitude as the CA-1 mSv values. Here, a more detailed analysis of the radiological impact of these air releases is required. This analysis must use a dilution model to evaluate at any location outside the facility the average annual concentration of these nuclides and compute their radiological impact. The saturated activities resulting from our study can then be used as source terms for this dilution analysis.

Conclusions

- Evaluation of the air activation in the standard *Proteus®ONE* system using the same assumptions as for the shielding design.
- Unstable nuclides are produced in air by secondary neutrons, either because of spallation reactions, resulting in the production of ^{3}H, ^{7}Be, ^{11}C, ^{13}N, ^{15}O, or by thermal neutron capture producing ^{41}Ar.
- The average saturated activities for these nuclides are computed on an annual basis using NCRP-144 recommendations, CERN-evaluated cross sections and MCNPX-based neutron fluences.
- The computed activities from the CGTR are very small, resulting in negligible effective doses for population leaving around the facility.
- The activities released from the S2C2 room are a factor 400 (spallation nucldies) to 1600 (^{41}Ar) larger than those coming from the CGTR. Here a more detailed analysis of the diffusion of the released air around the facility is needed and our results can be used as source terms.

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Thank You

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