REVIEW OF THE DIFFERENT ACCELERATOR-BASED BNCT FACILITIES WORLWIDE AND AN ASSESSMENT ACCORDING TO THE ALARA CRITERION

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Abstract

Presently, there are a number of different facilities for Accelerator-Based Boron Neutron Capture Therapy (AB-BNCT) worldwide. They range from high-energy 30 MeV cyclotrons, medium-energy RFQ-DTL accelerators (8-10 MeV), low-energy electrostatic and RFQ machines (2.3-2.8 MeV), to a very low-energy electrostatic quadrupole accelerator (1.45 MeV). The paper analyses these installations from the point of view of activation. Since these facilities are intended to work in hospital environments one of the guiding criteria should be the ALARA (As Low As Reasonably Achievable) one.

A detailed analysis using MCNP simulations was made to evaluate neutron induced radioactivity produced in the Beam Shaping Assembly. Also, the activation of the target due to the primary beam is evaluated based on evaluated nuclear data bases.

Based on the IAEA RS-G 1.7 Safety Guide, Application of the Concepts of Exclusion, Exemption and Clearance the paper assess the long-term operation sustainability from the point of view of activation.

1. INTRODUCTION

A wide variety of accelerator-based facilities are envisaged worldwide for Accelerator-Based BNCT (AB-BNCT) worldwide. Some of them are already working and even treating patients, and some are under development and construction. They range from high-energy 30 MeV cyclotrons (using the ^{9}Be(p,n) reaction), medium-energy RFQ-DTL accelerators (at 8 and 10 MeV using likewise the ^{9}Be(p,n) reaction), low-energy electrostatic (both Tandem and single-ended), and RFQ machines (working on ^{7}Li(p,n) at about 2.5 MeV), to a very low-energy electrostatic quadrupole ESQ accelerator (working on ^{9}Be(d,n) or ^{13}C(d,n) at 1.45 MeV). Based on the neutron production reaction, target material and beam energy, all AB-BNCT facilities fit to one of the listed in Table 1.

Since these facilities are intended to work in hospital environments one of the guiding criteria should be the ALARA (As Low As Reasonably Achievable) one. Focusing on long-term operation sustainability and on the basis of the ALARA criterion, the production of residual radioactivity should be minimized as much as reasonably possible.

Although the common goal in all AB-BNCT facilities is to produce an, as pure as possible, epithermal neutron beam (in the range of 0.5 eV to 10 keV), production of high-energy neutrons and hence of induced radioactivity varies considerably from one to another. For instance, facilities working with the ^{7}Li(p,n) reaction produce mainly neutrons of less than 1 MeV. Hence, concerning long-term activation, only exothermic and very low-threshold neutron-induced reactions are relevant to assess activation of these facilities. On the other hand, the ones working with 30 MeV protons and the ^{9}Be(p,n) reaction generate neutrons with energies up to 28.1 MeV. In this case there are many more energetically possible neutron-induced reactions that may generate radioactivity on the subsystems of the facility. In an intermediate scenario are the facilities based on 1.45 MeV deuterons (for both ^{9}Be and ^{13}C targets) and those based on 8 MeV protons on ^{9}Be, where maximum neutron energies are, respectively, 5.76, 6.72 and 6.31 MeV. In particular, the deuteron induced reactions have a large proportion of the yield in the less-than-1 MeV energy region (see Table 1) which makes these reactions particularly attractive for BNCT.

The aim of this work is to quantitatively assess the residual radioactivity in a representative group of AB-BNCT facilities, both by the primary (due to the beam) and secondary (due to neutrons) activation.

TABLE 1. AB-BNCT FACILITIES WORLWIDE GROUPED BY NEUTRON PRODUCING REACTION AND BEAM ENERGY

Target-reaction	Beam Energy (MeV)	Beam current goal (mA)	Percentage of yield with energy ≤ 1 MeV at 0º (%)	Institute, Country
^{7}Li(p,n)^{7}Be	2.3-2.8	10-30	100-92 [1]	Helsinki Univ. Hospital, Finland [2] National Cancer Center, Japan [3, 4] Edogawa Hospital, Japan [5] Nagoya University, Japan [6] Shonan Kamakura Hospital, Japan [7] Soreq, Israel [8] Xiamen Humanity Hosp., China [9–11] IHEP, China [12] Budker Inst., Russia [9] CNAO, Italy [9] Birmingham University, UK [13, 14] Granada University, Spain [15]
^{9}Be(p,n)^{9}B	30	1	9 [16]	Kyoto University, Japan [5, 17] Kansai BNCT RC, Japan. [5, 17, 18] Southern Tohoku Hosp., Japan [5, 17, 19]
^{9}Be(p,n)^{9}B	8	10	21 [16]	Tsukuba University, Japan [20, 21] Gachon Univ.-Dawon Medax, Korea [22, 23]
^{9}Be(d,n)^{10}B	1.45	30	66 [24]	CNEA, Argentina [25–28] KIRAMS, Korea*
^{13}C(d,n)^{14}N	1.45	30	70 [29]	CNEA, Argentina [25–27, 30] KIRAMS, Korea*

*Cooperation agreement between CNEA and KIRAMS.

2. PRIMARY ACTIVATION

Primary activation is produced by interaction of the proton or deuteron beam with the different subsystems of the accelerator. This comes from nuclear reactions (p,X) or (d,X), were X stands for any open reaction channel with radioactive products. The radioactive product may be either the ejectile or the heavy product of the reaction. The largest fraction of the beam impinges directly on the target material and hence, this is the most relevant subsystem as far as primary activation is concerned. Table 2 summarizes the radioactive products and nuclear reactions relevant for each projectile and target material.

TABLE 2. NUCLEAR REACTIONS LEADING TO RADIOACTIVE PRODUCTS ON THE DIFFERENT AB-BNCT TARGETS

	Product, $T_{1/2}$	Reaction	Threshold Energy or Q (MeV)
^{7}Li+p	^{7}Be, 53.22 d	^{7}Li(p,n)^{7}Be	E_{thres}=1.88
^{9}Be+p	^{7}Be, 53.22 d	^{9}Be(p,t)^{7}Be	E_{thres}=13.432
		^{9}Be(p,d+n)^{7}Be	E_{thres}=20.4
		^{9}Be(p,p+2n)^{7}Be	E_{thres}=22.9
	tritium, 12.32 y	^{9}Be(p,t)^{7}Be	E_{thres}=13.432
^{9}Be+d	tritium, 12.32 y	^{9}Be(d,t)^{8}Be	Q=4.602
^{13}C+d	tritium, 12.32 y	^{13}C(d,t)^{12}C	Q=1.312
	^{14}C, 5700 y	^{13}C(d,p)^{14}C	Q=5.962

Facilities based on proton beams on lithium targets produce ^{7}Be as the heavy product in the neutron production reaction. This means that radioactive ^{7}Be nuclei are generated at the same rate than neutrons are. For protons on beryllium, there are two well-differentiated scenarios. Facilities working with high-energy 30 MeV protons generate tritium and ^{7}Be. Tritium comes as the ejectile of the (p,t) reaction while ^{7}Be comes as a heavy product. The threshold energy the ^{9}Be(p,t) reaction is 13.432 MeV, and there are no any other open channels producing radioactivity for protons below this energy, hence medium-energy (8-10 MeV) facilities do not produce long-lived radioactivity in the target. For higher proton energies more channels producing ^{7}Be open, being (n,d+n) at 20.4 MeV and (n,p+2n) at 22.9 MeV the most relevant ones [31].

Radioactivity on beryllium and ^{13}C targets is due to tritium produced through the exothermic (d,t) reactions on ^{9}Be and ^{13}C, respectively. On ^{13}C targets some activity of ^{14}C coming from the (d,p) reaction is also expected.

3. SECONDARY ACTIVATION

Secondary activation comes from neutron induced reactions (n,X) on any element exposed to the neutron flux. There are several subsystems that can be activated, such as the Beam Shaping Assembly (BSA), shielding, target assembly, irradiation room walls, the beamline, ancillary equipment, wires, and any other element exposed to neutrons.

Due to the proximity to the neutron source, the most exposed one and hence, potentially highly activated is the BSA. The main role of the BSA is to moderate the neutrons down to the epithermal regime and to efficiently guide them to the element to be irradiated. It also acts as a first shielding for neutrons and gamma rays. The structure, components, size, and materials, depend on the energy distribution of the neutrons produced in the target. A generic design is shown in Fig. 1.

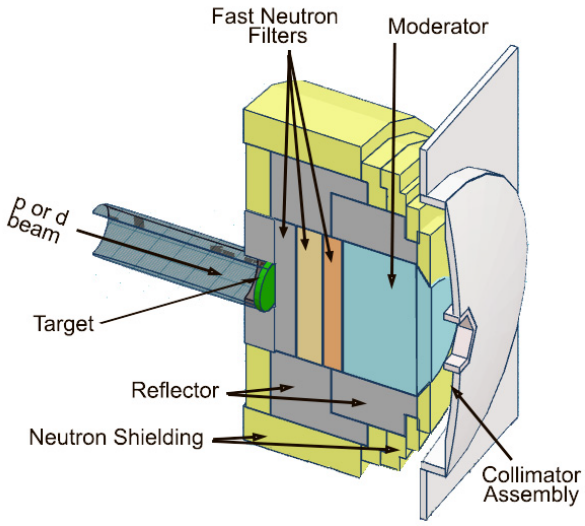

FIG. 1. Schematic view of a typical Beam Shaping Assembly (BSA).

To analyse neutron activation a first approach is to evaluate which of the (n,X) reactions are energetically allowed. Quantitatively, the activity concentration (Bq per gram of material) must be calculated by Monte-Carlo transport of neutrons through the BSA materials. Based on publications of the different AB-BNCT facilities worldwide, a representative group of BSA's was set up and simulated with MNCP 6.1 [32]. Table 3 summarizes their main components.

Each of the BSAs listed in Table 3 were simulated and the accumulated activity concentration after 1 year of operation was calculated. For each radionuclide, the activity concentration was compared to the levels of clearance given for materials in bulk in the IAEA Safety Guide RS-G1.7 [33]. These clearance levels represent the maximum activity concentration tolerable to release radioactive materials from regulatory control. The values are derived for each radionuclide in the IAEA Safety Report Nº44 [34], and correspond to the activity concentration values such that individual effective doses to a critical group (i.e., the public and workers) would be of the order of 10 μSv/a and would have only a very low probability of approaching an individual dose of 1

mSv/a. For materials containing a mixture of radionuclides, the normalized activity concentration (C) must be less than 1:

$$C = \sum_{i=1}^{n} \frac{C_i}{L_i} < 1 \quad (1)$$

where C_i is the concentration (in Bq/g) of the i^{th} radionuclide in the material, L_i is the respective activity concentration for clearance and n is the number of radionuclides present (ref paragraph 4.7 in [33]).

TABLE 3. MAIN COMPONENTS OF THE BEAM SHAPING ASSEMBLIES FOR A REPRESENTATIVE GROUP OF AB-BNCT FACILITIES

	Beam energy, current	Moderator	Fast Neutron Filter	Reflector	References.
^{7}Li+p	2.3 MeV, 30 mA	Fluental, MgF_2 or CaF_2	None	Pb	[35–38]
^{9}Be+p	8.0 MeV, 10 mA	MgF_2	Fe	Pb	[39]
^{9}Be+p	30 MeV, 1 mA	CaF_2	3: Pb+Fe+Al	Pb	[40]
^{9}Be+d	1.45 MeV, 30 mA	2: PTFE+Al	None	Pb	[28, 41]
^{13}C+d	1.45 MeV, 30 mA	2: PTFE +Al	None	Pb	[30, 42]

4. ACTIVITY CALCULATION

Daily operation is usually carried out by alternating hours of continuous irradiation and hours of downtime. During operation, the activity A accumulates as:

$$A(t) = A_0 + \frac{p}{\lambda} \cdot (1 - e^{-\lambda \cdot t}) \quad (2)$$

being t the irradiation time, p the activity production rate, and λ the decay constant of the radioactive residue. A_0 is the initial activity in the element to evaluate. The activity production rate (p, in Bq/h) is the product of the reaction yield (Y, in 1/mA-s), the beam current (in mA) and the decay constant (λ, in 1/h).

During downtime, the activity follows the radioactive decay law:

$$A(t + t') = A(t) \cdot e^{-\lambda \cdot t'} \quad (3)$$

where t' is the length of downtime.

For a given scheme of operation, the accumulated activity in the element lifetime can be calculated by recurrence applying Eqs. 2 and 3 alternatively. For a daily scheme of 8 hours of irradiation followed by 16 hours of downtime, the accumulated activity is:

$$A(N) = \frac{p}{\lambda} \cdot \left(1 - e^{-\lambda \cdot 8\,h}\right) \cdot \frac{1 - \left(e^{-\lambda \cdot 24\,h}\right)^{N}}{1 - e^{-\lambda \cdot 24\,h}} \quad (4)$$

Where N is the target lifetime (in days).

To evaluate primary activation on the target, the activity production rate (p) was calculated from reaction cross-sections $\sigma(E)$ as:

$$p = \lambda \cdot \dot{n} \cdot n_A \int S(E)^{-1} \cdot \sigma(E) \cdot dE \quad (5)$$

Where $\dot{n}$ (in 1/s) is the number of projectiles (protons or deuterons) per second impinging on the target, n_A is the atom density of the target, $S(E)$ is the stopping power. Cross-sections and stopping powers were taken from [31, 43–45]and [46] respectively.

For secondary activation, the group of BSA's listed in Table 3 were simulated with the MNCP 6.1 code [32]. Nuclear reactions leading to radioactive products were considered for each element of the BSA (moderator, fast neutron filters and reflector). All energetically allowed reactions with up to 4 light products were considered.

For a given reaction, the activity production rate p was calculated as

$$p = \lambda \cdot N_A \int \dot{\phi}(E) \cdot \sigma(E) \cdot dE \quad (6)$$

where $\dot{\phi}(E) \cdot dE$ (in 1/cm^2-s) is the neutron flux in the element to evaluate (moderator, fast neutron filters or reflector), $\sigma(E)$ and N_A are, respectively, the cross-section and the number of target atoms for the reaction that generates radioactivity in the element of interest. The integrals in Eq. 6 were calculated as MCNP F4 Tallies with the reaction cross-sections as the tally modifiers (DE and DF cards). Cross-sections were taken from different evaluated databases [43, 47, 48].

5. RESULTS

5.1. Target activation

Table 4 summarizes the radioactivity accumulated for different targets over 1 year of operation with a daily scheme of 8 hours of irradiation a day. The activities reported here are an average of different results based on the indicated reference data.

TABLE 4. RADIOACTITIVY INDUCED FOR DIFFERENT AB-BNCT TARGETS AT THE INDICATED BEAM ENERGY AND CURRENT (IN ORDER OF DECREASING TOTAL ACTIVITY)

	Beam energy, current	Radionuclide: activity	Reference data
^{7}Li+p	2.3 MeV, 30 mA	^{7}Be: 5.7 TBq/y	[43, 49, 50]
^{9}Be+p	30 MeV, 1 mA	^{7}Be: 1.2 TBq/y	[31]
		^{3}H: 51 GBq/y	[31,43]
^{9}Be+d	1.45 MeV, 30 mA	^{3}H: 57 GBq/y	[43,44]
^{13}C+d	1.45 MeV, 30 mA	^{3}H: 9.3 GBq/y	[43,44]
		^{14}C: 28 MBq/y	[43]
^{9}Be+p	8 MeV, 10 mA	Only prompt radiation	[31]

The ^{9}Be+p case for 8 MeV protons is the cleanest target. In this case, all reaction channels leading to radioactivity are energetically forbidden (see Table 2). Only short-lived light radionuclides, such as ^{9}Be(p,n)^{9}B (E_{thres}=2.057 MeV), ^{9}Be(p,d)^{8}Be (Q=570 keV) and ^{9}Be(p,p+n)^{8}Be (E_{thres}=1.850 MeV) are produced. The products are ^{9}B ($T_{1/2}$ = 0.54 keV, decays via proton emission to ^{8}Be) and ^{8}Be which rapidly breaks into two alpha particles with no gamma emission.

The highest activity was obtained for the lithium target. In this case, radioactive ^{7}Be ($T_{1/2}$=53.22 d) is generated through the ^{7}Li(p,n)^{7}Be reaction on the target. A 30-mA proton beam of 2.3 MeV generates 5.7 TBq of ^{7}Be after 1 year of operation of the target. This radionuclide emits gamma rays of 478 keV thus, represents a complication concerning dose to workers given tasks involving handling of the target (i.e., replacement).

In order of importance follows the beryllium target with 30 MeV protons. The activity of ^{7}Be is 1.2 TBq/y, coming from the ^{9}Be (p,t) , ^{9}Be(n,d+n) and ^{9}Be(n,p+2n) reactions (see Table 2). In addition to ^{7}Be, 51 GBq/y of tritium ($T_{1/2}$=12.32 y) are produced through the ^{9}Be(p,t) ^{7}Be reaction. This radionuclide decays emitting very low-energy beta particles (E<18.591 keV) with no gamma rays. These beta particles stop in less-than 1 cm of air, thus radiological risk only arises if inhaled, ingested, or absorbed through the skin.

With deuterons on beryllium and ^{13}C targets only low-energy pure beta emitters are produced. In the beryllium target, 57 GBq/y activity of tritium is generated through the ^{9}Be(d,t) reaction. Concerning the ^{13}C target, tritium and ^{14}C are generated. The activity of tritium (9.3 GBq/y) is considerably lower than in the ^{9}Be target (both with deuterons and 30 MeV protons). The activity of ^{14}C (28 MBq/y) comes from the ^{13}C(d,p) reaction. ^{14}C decay emitting low energy beta particles up to 156 keV (this radiation barely penetrates the outer protective dead layer of the skin) with no gamma rays. Figs. 2 and 3 show the accelerator and a 10 mA beam developed at the Atomic Energy Commission in Buenos Aires, to be used in conjunction with the ^{9}Be(d,n) and ^{13}C(d,n) reactions.

FIG. 2. Single-ended, modular ESQ accelerator. Operates in air for easy maintenance. This machine has 720 kV. The full size one of 1.45 MV is being constructed. Reprinted from Ref. [27]

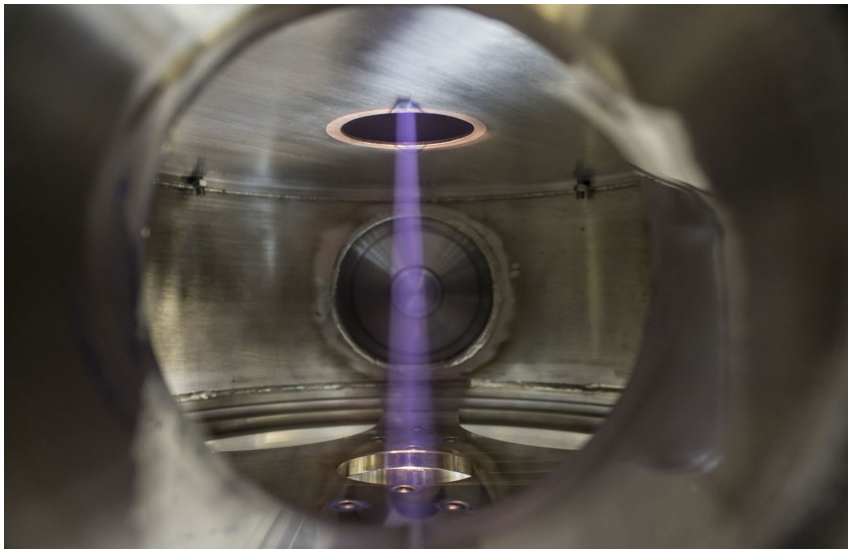

FIG. 3. 10 mA proton beam, as seen through the induced fluorescence in the residual gas in the accelerator column of the ESQ. Reprinted from Ref. [51]

5.2. BSA activation.

From lowest to highest neutron energy, first are the facilities based on the ^{7}Li(p,n) reaction. For protons of 2.3 MeV the maximum neutron energy is 573 keV. Hence, only exothermic and some very low-threshold reactions, are relevant for neutron induced activation in these facilities. These reactions are typically (n,γ) reactions, although for lead and materials containing ^{6}Li there are more reaction channels open (discussed below, §5.2.1).

Following are the facilities based on 1.45 MeV deuterons (both ^{9}Be and ^{13}C targets) and on 8 MeV protons on ^{9}Be. The highest neutron energies are, respectively 5.76, 6.72 and 6.17 MeV. For neutron activation, exothermic and reactions with threshold energies of a few MeV must be considered. For commonly used BSA materials, most (n,p), (n,α) and (n,d) become accessible for neutrons of these energies, although the cross-sections in general are rather low. Finally, facilities based on 30 MeV protons and beryllium targets produce neutrons of up to 28.15 MeV. For some commonly used BSA materials, (n,t) reactions become accessible at about 8-10 MeV. Moreover, above ~10 MeV reaction channels with multiple ejectiles (such as (n,2n), (n,n+p), (n,n+α) etc.) become accessible.

As a general trend, the higher the neutron energy, the higher the number of energetically possible reactions that may generate radioactive products. Activation of each BSA is discussed separately below.

5.2.1. ^{7}Li+p (2.3 MeV)

Moderator materials. Three different moderators were simulated: MgF_2, CaF_2 and Fluental (AlF_3: 69 w-%), metallic aluminium: 30 w-% and LiF:1 w-%).

The MgF_2 moderator does not activate. On natural Mg and F, the only energetically allowed reactions are ^{19}F(n,γ)^{20}F (Q= 6.601 MeV) and ^{26}Mg(n,γ)^{27}Mg (Q=6.443 MeV). The half-lives of ^{20}F and ^{27}Mg are, respectively, $T_{1/2}$=11.07 s and $T_{1/2}$=9.458 m. Due to the short half-lives, these radionuclides will rapidly decay after daily-stop and do not accumulate in the intermediate or long-term. All other energetically allowed reactions produce stable products.

In the CaF_2 moderator, ^{45}Ca ($T_{1/2}$=162.61 d) and ^{47}Ca ($T_{1/2}$=4.536 d) are produced coming from (n,γ) reaction on ^{44}Ca and ^{44}Ca, respectively. Over 1 year operation, the normalized activity concentration is C=101, and the cooling time (i.e., time required to decay below the clearance level) is ~3 y.

The Fluental moderator is the most critical one**.** Long-term radioactivity is due to tritium ($T_{1/2}$ =12.32 y) coming from the exothermic reaction ^{6}Li(n,α)^{3}H (Q=4.783 MeV). The activity concentration of tritium is 14.4 kBq per gram of Fluental and the cooling time is 88 y. Other short-lived radionuclides, such as ^{28}Al ($T_{1/2}$=2.245 m), ^{20}F ($T_{1/2}$=11.07 s), ^{8}Li ($T_{1/2}$=840 ms) are also produced, coming from activation of Al, F and ^{7}Li present in Fluental, with no impact on long-term radioactivity.

Reflector (Pb). There are several exothermic reactions with intermediate-lived radioactive products, as shown in Table 5. For neutrons of less than 573 keV, the reaction cross-sections are, except for ^{208}Pb(n,γ)^{209}Pb, very small. Moreover, simulations showed that the associated activities (of ^{204}Tl, ^{203}Hg, ^{200}Pt and ^{197}Pt) represent less than 0.00001 of the total activity, being all of them well below their clearance levels. The activity of ^{209}Pb ($T_{1/2}$=3.234 d) is the only significant contribution. After 1 year operation, the activity concentration of ^{209}Pb is 705 Bq/g. The IAEA Safety Guide RS-G1.7 does not establish a limit for this radionuclide, so the normalized activity concentration was not calculated. It is worth mentioning that this radionuclide emits beta particles only (no γ) with up to 197.5 keV. Thus, practically all Bremsstrahlung radiation due to the slowing down of beta particles is self-absorbed in the reflector, except a small fraction coming from ^{209}Pb in the outer shell of lead.

5.2.2. ^{9}Be+d and ^{13}C+d, 1.45 MeV

Moderator. The moderator is constituted by slabs of pure aluminum and PTFE. On the PTFE slabs, the long-term activation is due to ^{14}C ($T_{1/2}$=5700 y) only, coming from ^{13}C(n,γ)^{14}C. All other nuclear reactions generate stable or short-lived products ($T_{1/2}$ ~ s). Due to the small isotopic abundance of ^{13}C (1.07%) and to the long-life of the activation product, the activity production rate is expected to be small. In fact, simulations showed that after 1 y operation the accumulated activity of ^{14}C is 0.006 Bq per gram of PTFE, which is far below the clearance level of 1 Bq/g, and it would take about 170 y in operation to exceed this value.

On the aluminum slabs, there is no long-term accumulation of radioactivity. Only ^{28}Al ($T_{1/2}$=2.245 m) and ^{24}Na ($T_{1/2}$=14.997 h) and are produced, coming from (n,γ) and (n,α) reactions on ^{27}Al respectively. The activity of ^{28}Al rapidly decays in a few hours after the end of irradiation, hence does not accumulate. Concerning ^{24}Na, the half-life is comparable to the time in between irradiations, thus, some accumulation is produced. Simulations showed that after 1 year of operation the accumulated activity is 110 Bq per gram of aluminum and takes 4.2 days to decay below 1 Bq/g, the clearance level for ^{24}Na.

Reflector (Pb). As for the Li+p case, the main contribution is ^{209}Pb ($T_{1/2}$=3.234 d) produced through the ^{208}Pb(n,γ)^{209}Pb reaction. The activity after 1 year operation is 290 Bq per gram of lead. In addition to those given in Table 5, some endothermic reactions are accessible (see Table 6), although simulations show that none of the radionuclides exceed their clearance levels, and also, that the total activity is negligible compared to the activity of ^{209}Pb.

TABLE 5. INTERMEDIATE-LIVED RADIOACTIVE PRODUCTS FROM EXOTHERMIC NEUTRON INDUCED REACTIONS ON NATURAL LEAD

Product	$T_{1/2}$	Nuclear Reaction	Q-value	Clearance Level
^{209}Pb	3.234 d	^{208}Pb(n,γ)^{209}Pb	3937.4 keV	-
^{203}Hg	46.594 d	^{206}Pb(n,α)^{203}Hg	7130.1 keV	10 Bq/g
		^{207}Pb(n,n+α)^{203}Hg	392.3 keV	
^{204}Tl	3.783 y	^{204}Pb(n,p)^{204}Tl	18.6 keV	1 Bq/g
^{200}Pt	12.6 h	^{207}Pb(n,2α)^{200}Pt	7369 keV	-
^{197}Pt	19.8915 h	^{204}Pb(n,2α)^{197}Pt	8531 keV	1000 Bq/g

TABLE 6. INTERMEDIATE-LIVED RADIOACTIVE PRODUCTS FROM LOW-THRESHOLD NEUTRON INDUCED REACTIONS ON NATURAL LEAD

Product	$T_{1/2}$	Nuclear Reaction	Threshold Energy	Clearance Level
^{3}H	12.32 y	^{208}Pb(n,t)	6.4 MeV	100 Bq/g
		^{206}Pb(n,t)	6.3 MeV	
		^{204}Pb(n,t)	6.0 MeV	
^{199}Au	3.139 d	^{204}Pb(n,d+α)^{199}Au	3.5 MeV	100 Bq/g
		^{208}Pb(n,n+2α)^{199}Au	5.8 MeV	
^{202}Tl	12.31 d	^{204}Pb(n,t)^{202}Tl	6.0 MeV	10 Bq/g
^{203}Hg	46.594 d	^{204}Pb(n,2p)^{203}Hg	6.4 MeV	10 Bq/g
^{204}Tl	3.783 y	^{206}Pb(n,t) ^{204}Tl	6.3 MeV	1 Bq/g

5.2.3. ^{9}Be +p (8 MeV)

Fast neutron filter. The critical element of this BSA is the iron filter. Table 7 shows the energetically allowed reactions that lead to intermediate and long-lived radioactivity for this material. Also shown are the respective clearance levels. All radioactive products exceed their clearance level after 1 year operation, and the normalized activity concentration after 1 year operation is $C=9.6\times10^{5}$. Almost 98% of the normalized activity is due to ^{54}Mn ($T_{1/2}$=312.2 d), thus the cooling time is mainly determined by this residue. For 1 year of service this time is 18.8 y.

Moderator. A MgF_2 moderator was simulated. In this moderator, ^{24}Na ($T_{1/2}$=14.997 h) is generated due to the ^{24}Mg(n,p)^{24}Na reaction. Simulations showed that the activity concentration of ^{24}Na saturates at ~2 Bq/g after 4 days of operation, thus a longer period of service of the moderator does not lead to more radioactivity. Since the clearance level for ^{24}Na is 1 Bq/g, the cooling time is equal to 1 half-life, independently of the lifetime of the moderator.

TABLE 7. INTERMEDIATE AND LONG-LIVED RADIOACTIVE PRODUCTS FROM EXOTHERMIC AND LOW-THRESHOLD NEUTRON INDUCED REACTIONS ON NATURAL IRON

Product	$T_{1/2}$	Nuclear Reaction	Threshold Energy or Q	Clearance Level
^{54}Mn	312.2 d	^{54}Fe(n,p) ^{54}Mn	Q=85.5 keV	0.1 Bq/g
^{56}Mn	2.5789 h	^{56}Fe(n,p) ^{56}Mn	E_{thresh}=2964.7 keV	10 Bq/g
^{51}Cr	27.7025 d	^{54}Fe(n,α)^{51}Cr	Q=843.3 keV	100 Bq/g
^{59}Fe	44.495 d	^{58}Fe(n,γ)^{59}Fe	Q= 6581 keV	1 Bq/g
^{55}Fe	2.744 y	^{54}Fe(n,γ) ^{55}Fe	Q=9298 keV	1000 Bq/g

Reflector. As for the above mentioned cases (§5.2.1 and 5.2.2) the main contribution is ^{209}Pb ($T_{1/2}$=3.234 d) produced through the ^{208}Pb(n,γ)^{209}Pb reaction. The activity after 1 year operation is 14.3 kBq per gram of lead. The activity concentration of the rest of radionuclides is negligible and do not exceed (when applicable) their clearance levels.

5.2.4. ^{9}Be +p (30 MeV)

Fast neutron filters. There are 3 fast neutron filters in this BSA. Closest to the target is the filter of lead, followed by a second filter (of iron) and a third one of aluminum.

In the lead filter, the most important contribution to the total activity is from ^{203}Pb ($T_{1/2}$=51.92 h), as shown in Table 8. The normalized activity concentration is C=8.7×10^4. Concerning long-term activation, ^{204}Tl ($T_{1/2}$=3.783 y) is the most relevant residue to consider, since the cooling time is strongly determined by this isotope. Only for ^{204}Tl, it takes 36.6 y for clearance, and the time required for the whole mixture (C=1) is 37.8 y.

TABLE 8. RADIOACTIVITY INDUCED IN THE LEAD FILTER BY NEUTRONS FROM 30 MEV PROTONS ON A ^{9}BE TARGET, AFTER 1 YEAR OF OPERATION. ONLY RADIONUCLIDES EXCEEDING THE CLEARANCE LEVELS ARE LISTED

Product	$T_{1/2}$	Activity concentration	Clearance Level
^{203}Pb	51.92 h	8.6×10^5 Bq/g	10 Bq/g
^{203}Hg	46.594 d	2.0×10^3 Bq/g	10 Bq/g
^{204}Tl	3.783 y	812 Bq/g	1 Bq/g
^{202}Tl	12.31 d	512 Bq/g	10 Bq/g
^{3}H	12.32 y	164 Bq/g	100 Bq/g

In the iron filter, the normalized activity concentration is C=9.2×10^5. The most important contribution is from ^{54}Mn ($T_{1/2}$=312.2 d), as shown in Table 9. This radionuclide exceeds the clearance level 8.3×10^5 times and is the one that most contributes to the high value C. As far as long-term activation is concerned, the dominant contribution is ^{55}Fe ($T_{1/2}$=2.744 y). The cooling time for this radioisotope is 23.1 y, and the cooling time for the whole mixture (C=1) is 26.2 y.

TABLE 9. RADIOACTIVITY INDUCED IN THE IRON FILTER BY NEUTRONS FROM 30 MEV PROTONS ON A ^{9}BE TARGET, AFTER 1 YEAR OF OPERATION. ONLY RADIONUCLIDES EXCEEDING THE CLEARANCE LEVELS ARE LISTED

Product	$T_{1/2}$	Activity concentration	Clearance Level
^{54}Mn	312.2 d	8.3×10^4 Bq/g	0.1
^{56}Mn	2.5789 h	7.0×10^5 Bq/g	10
^{59}Fe	44.495 d	1.5×10^4 Bq/g	1
^{55}Fe	2.744 y	3.4×10^5 Bq/g	1000
^{52}Mn	5.591 d	3110 Bq/g	1
^{51}Cr	27.7025 d	3470 Bq/g	100
^{3}H	12.32 y	237 Bq/g	100

For the aluminum filter, the normalized activity concentration is C=7.9×10^4, mostly from ^{24}Na ($T_{1/2}$=14.997 h). This radionuclide decays in a relatively short time, thus for the long-term radioactivity only ^{22}Na ($T_{1/2}$=2.6018 y) and ^{3}H ($T_{1/2}$=12.32 y) are relevant. The cooling time for ^{22}Na and ^{3}H are 3.85 y and 5 y, respectively, and cooling time for the mixture (C=1) is 9.45 y.

TABLE 10. RADIOACTIVITY INDUCED IN THE ALUMINIUM FILTER BY NEUTRONS FROM 30 MEV PROTONS ON A ^{9}BE TARGET, AFTER 1 YEAR OF OPERATION

Product	$T_{1/2}$	Activity concentration	Clearance Level
^{24}Na	14.997 h	7.9×10^4 Bq/g	0.1
^{22}Na	2.6018 y	0.28 Bq/g	10
^{3}H	12.32 y	133 Bq/g	100

Moderator. A CaF_2 moderator was simulated. The normalized activity concentration is C=3007 due to ^{45}Ca ($T_{1/2}$=162.61 d), ^{47}Ca ($T_{1/2}$=4.536 d), ^{43}K ($T_{1/2}$=22.3 h) and ^{18}F ($T_{1/2}$=109.77 m). The most relevant one as far as long-term activation is ^{45}Ca. The activity concentration for this radionuclide is 58.4 kBq per gram of CaF_2 after 1 year operation. The time required to decay below the clearance level (100 Bq/g) is ~4 y.

Reflector (Pb). The normalized activity concentration in the reflector is C=1.2×10^3, where the main contribution from ^{203}Pb ($T_{1/2}$=51.92 h) (see Table 11). Also, above the clearance limit are ^{203}Hg ($T_{1/2}$=46.594 d) and ^{204}Tl ($T_{1/2}$=3.783 y), being the latter what determines the cooling time of the reflector (11.6 y).

TABLE 11. RADIOACTIVITY INDUCED IN THE LEAD REFLECTOR BY NEUTRONS FROM 30 MEV PROTONS ON A ^{9}BE TARGET, AFTER 1 YEAR OF OPERATION. ONLY RADIONUCLIDES EXCEEDING THE CLEARANCE LEVELS ARE LISTED

Product	$T_{1/2}$	Activity concentration	Clearance Level
^{203}Pb	51.92 h	1.2×10^4 Bq/g	10 Bq/g
^{203}Hg	46.594 d	112 Bq/g	10 Bq/g
^{204}Tl	3.783 y	8.4 Bq/g	1 Bq/g

6. SUMMARY AND CONCLUSIONS

A wide variety of accelerator-based facilities are envisaged worldwide for BNCT. They can be classified into high (30 MeV), medium (8-10 MeV), low (2.3-2.8 MeV), and very low-bombarding energies (1.45 MeV). Based on publications of the existing facilities, a representative case was assessed for each category from the point of view of activation.

High-energy facilities work with 1 mA of 30 MeV protons on a ^{9}Be target, producing neutrons through the ^{9}Be(p,n) reaction. Neutrons up to 28.15 MeV are generated. At this energy, there are many neutron-induced reactions that lead to the activation of the BSA materials. Simulations showed that after 1 year operation, cooling times of several decades are necessary for clearance, according to the established values in the IAEA Safety Guide. Primary activation of the target (due to the proton beam) will also be high.

For medium-energy facilities, the case of 8 MeV protons on a ^{9}Be target was assessed. This facility produces neutrons also through the ^{9}Be(p,n) reaction, with a beam current of 10 mA. The benefit of working with this energy is the absence of primary activation of the target. All proton induced reactions leading to intermediate or long-term radioactivity are forbidden at bombarding energies below 13.432 MeV. Moreover, the neutron energy is considerably lower than in the high-energy case, thus neutron induced activation is also lower. The only critical element concerning long-term activation is the fast neutron filter (of iron), that requires ~19 y of cooling time after 1 year operation.

For the low-energy group, a facility based on a 30-mA proton beam of 2.3 mA was analyzed. This facility produces neutrons through the ^{7}Li(p,n)^{7}Be. Although the maximum neutron energy is low (573 keV), long-term activation may be produced depending on the choice of the moderator. A Fluental moderator may need, after 1 year of lifetime, 88 y of cooling time due the activity of tritium produced by the activation of ^{6}Li. On the opposite

side, there is no radioactivity if the moderator material is MgF_2. The main drawback for the Li+p is the target activation. Radioactive ^{7}Be produced as the residue of the ^{7}Li(p,n)^{7}Be emits 478 keV gamma rays and may represent a complication for tasks involving handling of the target

For the very low-energy group, 1.45 MeV deuterons on ^{9}Be and ^{13}C targets were evaluated. Both targets produce neutrons through the (d,n) reaction using a deuteron current of 30 mA. The maximum neutron energies are, respectively, 5.76 and 6.72 MeV, but with a large proportion of the yield in the less-than-1 MeV energy region (see Table 1) which makes these reactions particularly attractive for BNCT. These options work with a moderator of pure aluminum and PTFE, with no intermediate or long-term activation. Concerning primary activation (target) only low-energy pure beta emitters are generated.

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