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# Activation of Trace Elements in Concrete Walls of the Solid Target Room at Cyclotron Accelerator at NRCAM

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**ABSTRACT:** Concrete as a construction material is used widely in nuclear facilities. The walls of the solid target room of cyclotron accelerator at Nuclear Research Center for Agriculture and Medicine (NRCAM) are made of concrete with mass density of 2.3 g/cm3. In routine production of radiopharmaceuticals 201Tl and 67Ga at Cyclotron Dept. at NRCAM, which are, respectively, based on the 203Tl (p,3n) 201Pb 201Tl and 68Zn(p,2n) 67Ga nuclear reactions, in addition to the main products 201Pb and 67Ga, a number of neutrons are also produced as a by-product. The collision of emitted neutrons with the equipments and concrete walls of the target room can cause them to become activated. In this paper the neutron-induced activity of the elements in the concrete, with a reasonable half life, after a period of 30 years of cyclotron operation were investigated. Among the elements constituting the concrete, the isotopes 151Eu, 59Co, 153Eu, 133Cs, and 40Ca which turn respectively to the radioisotopes 152Eu, 60Co, 154Eu, 134Cs and 41Ca were considered. Our calculations show that the estimated specific activity of the above mentioned radioisotopes, after 30 years of cyclotron operation are 23.059 Bq/kg, 6.186 Bq/kg , 0.511 Bq/kg, 0.241 Bq/kg and 0.055 Bq/kg espectively.
**Keywords:** activity, trace elements, concrete, cyclotron, radiopharmaceutical.

## INTRODUCTION

At cyclotron department of Nuclear Research Center for Agriculture and Medicine (NRCAM), the radiopharmaceuticals $^{201}$Tl and $^{67}$Ga are produced weekly through the following nuclear reactions respectively [7]:

$^{203}Tl\,(p,3n)\,^{201}Pb \xrightarrow[EC,\beta^+]{} {}^{201}Tl$  Ep = 28.5 MeV

$^{68}Zn\,(p,2n)\,^{67}Ga$  Ep = 19 MeV

Bombarding time of thallium target is 10 hours continuously, once a week and for that of gallium target is the same but once every two weeks. The target assembly in target room surrounded by concrete walls with mass density of 2.30 g/cm$^3$ and minimum thickness of 1 m.

Our previous works [9], showed that the neutrons emitted from the above nuclear reactions have high intensity in the order of $10^{13}$ n/s. The collision of emitted neutrons with the concrete walls of the target room can cause the elements constituting the concrete to during decommissioning of the cyclotron accelerator and from waste management point of view.

The main purpose of this paper is to estimate the neutron-induced activity of the concrete walls after 30 years operation of cyclotron accelerator at NRCAM. To do so, the group neutron fluxes at the surface of the closest concrete wall to the target assembly, and at different depths of concrete wall are calculated using one dimensional computational code ANISN/PC [6] and group cross section libraries IRAN4.LIB [3-5] and EXFOR [8]. The elements constituting concrete are H, C, O, Mg, Al, Si, K and Ca. In addition, the elements Eu, Co, and Cs are present in concrete as trace elements in the order of ppm[10].

Among the elements mentioned above, only isotopes $^{151}$Eu, $^{59}$Co, $^{153}$Eu, $^{133}$Cs and $^{40}$Ca will become activated through (n,γ) reaction with reasonable half lives( compare to the total cyclotron operating time which assumed to be 30 years), while the other elements either have short half life (e.g. $^{28}$Al in $^{27}Al + n \rightarrow {}^{28}Al + \gamma$ reaction with a half life of 2.24 min.) or have very long half life (e.g. $^{40}$K with a half life of 1.28E+9 years) or will become stable(e.g. $^{2}$H in $^{1}H + n \rightarrow {}^{2}H + \gamma$ reaction). In this work the activity of the radioisotopes $^{152}$Eu, $^{60}$Co, $^{154}$Eu, $^{134}$Cs, and $^{41}$Ca which are produced from isotopes $^{151}$Eu, $^{59}$Co, $^{153}$Eu, $^{133}$Cs and $^{40}$Ca through (n,γ) reaction are calculated. The activities of the trace elements in the concrete have also been investigated by some authors [1, 2].

Calculation of activity based on the basic formula $A = \varphi(\varepsilon).\ \sigma(\varepsilon).\ N$, where N is the number of isotope under consideration, $\varphi(\varepsilon)$ and $\sigma(\varepsilon)$ are the neutron flux and reaction cross section respectively at energy ε.

## MATERIAL AND METHODS

**Neutron source:** The neutrons as a by-product in radiopharmaceutical production are produced through the following nuclear reactions:

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EC,$\beta^+$

$^{203}Tl$ (p,3n) $^{201}Pb$ ⟶ $^{201}Tl$ Ep = 28.5 MeV

$^{68}Zn$ (p,2n) $^{67}Ga$ Ep = 19 MeV

The intensities and energy spectra of emitted neutrons from the two above reactions have already been investigated [9]. In this work, the same method applied to obtain the neutron source parameters. Our calculations showed that the neutron intensity from thallium target is $3.75\times10^{13}$ n/s and for that of gallium target is $1.16\times10^{13}$ n/s. The neutron energy spectra from the two above reactions are shown in Fig. 1.

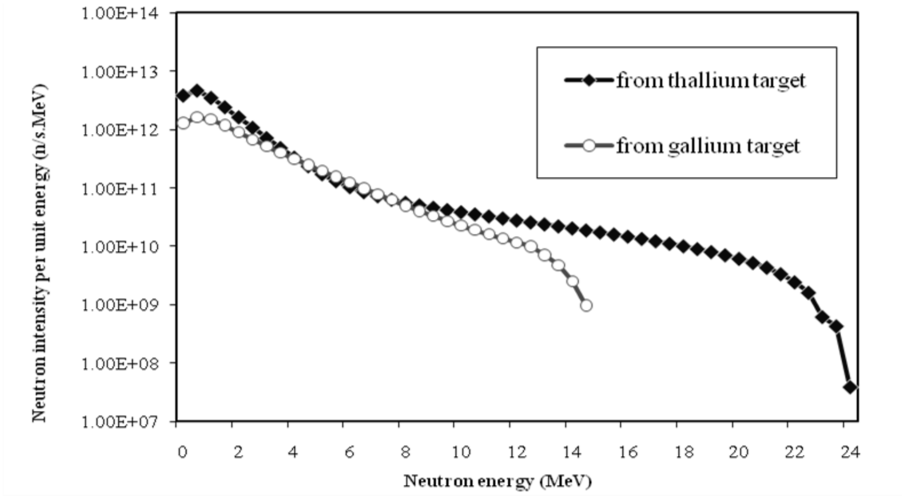


**Fig 1**. Neutron energy spectra from thallium and gallium targets

**Concrete wall.** The solid target assembly (neutron source) at cyclotron department is surrounded by concrete walls with mass density 2.30 gr/cm$^3$. Schematic diagram of the target assembly and irradiation target room is shown in Fig. 2.

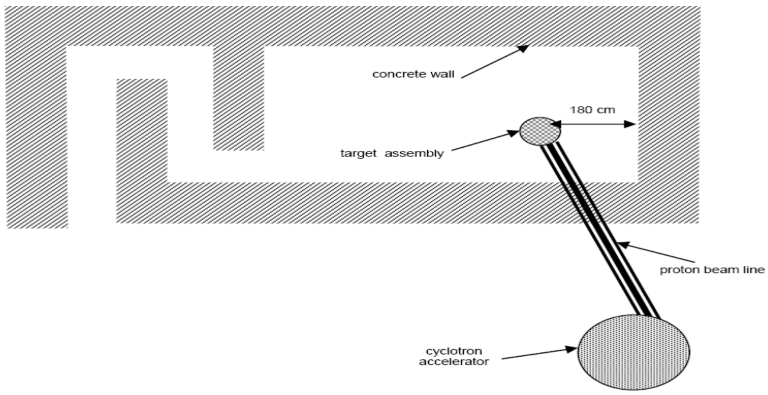


**Fig. 2**. Schematic diagram of target room and solid target assembly (not to scale). The other Accessories (e.g. target carrier) are not shown.

## RESULTS

The calculation of activity was performed for the closest wall to the target assembly which is 180 cm. The elements and their weight percents constituting the concrete are shown in Table 1.

The isotopes $^{1}H$, $^{12}C$, $^{16}O$, $^{24}Mg$, $^{28}Si$ and $^{56}Fe$ under neutron capture are converted to isotopes $^{2}H$, $^{13}C$, $^{17}O$, $^{25}Mg$, $^{29}Si$ and $^{57}Fe$ which are stable, therefore they are not being considered. On the other hand the isotopes $^{27}Al$ (by neutron capture converts to $^{28}Al$, with a short half life of 2.24 min.) and $^{39}K$ (converts to $^{40}K$ with a very long half life 1.28E+9 years) are not also being considered. Among the elements constituting the concrete, there are also trace elements such as $^{59}Co$, $^{133}Cs$, $^{151}Eu$ and $^{153}Eu$. Although, the isotopic abundances of these isotopes in the concrete constitution are very low but due to their reasonable half-lives and high neutron capture cross section, their activities in long term might be noticeable. In addition to the trace elements, the weight percent of isotope $^{40}Ca$ in concrete is not low and it converted to $^{41}Ca$ through neutron absorption, therefore the activity of $^{41}Ca$ is also taken into account. General speaking, in interaction of neutrons with matter and depending on the neutron energy, reactions such as (n,γ), (n,p) (n,α ), (n,2n), (n,n′),…. may occur, but in this work only (n,γ) reaction were investigated in Table 2. The radioisotopes which have been considered and their corresponding half-lives are shown.

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**Table 1:** Elements and their weight percents in concrete[a]

| Elements in concrete | Weight percent % | Atomic density($cm^{-3}$) |
|---|---|---|
| **H-1** | 1.00 | $1.385x10^{22}$ |
| **C-12** | 0.1 | $1.154x10^{20}$ |
| **O-16** | 52.91 | $4.581x10^{22}$ |
| **Mg-24** | 0.20 | $1.154x10^{20}$ |
| **Al-27** | 3.38 | $1.734x10^{21}$ |
| **Si-28** | 33.70 | $1.667x10^{22}$ |
| **K-39** | 1.30 | $4.618x10^{20}$ |
| **Ca-40** | 4.40 | $1.524x10^{21}$ |
| **Fe-56** | 1.40 | $3.463x10^{20}$ |
| **Co-59** | $25x10^{-6}$ ppm[b] | $5.87x10^{17}$ |
| **Cs-133** | $3 x10^{-6}$ ppm[c] | $3.13x10^{15}$ |
| **Eu-151** | $1.005x10^{-7}$ ppm[d] | $9.22x10^{15}$ |
| **Eu-153** | $1.095x10^{-7}$ ppm[e] | $9.92x10^{15}$ |
| a: Data taken from http://physics.mist.gov/cgi-bin/compos.pl?manto=144<br>b,c,d,e: Data taken from http://www.chemistryexplained.com/index.html | | |

**Table 2**: Half-lives of the radioisotopes [11]

| **Radioisotope** | **Nuclear reaction** | **Half life** |
|---|---|---|
| **Ca-41** | $^{40}Ca(n,γ)^{41}Ca$ | 1.3 E+5 years |
| **Co-60** | $^{59}Co(n,γ)^{60}Co$ | 5.27 years |
| **Cs-134** | $^{133}Cs(n,γ)^{134}Cs$ | 2.065 years |
| **Eu-152** | $^{151}Eu(n,γ)^{152}Eu$ | 13.5 years |
| **Eu-154** | $^{153}Eu(n,γ)^{154}Eu$ | 8.5 years |

**Table 3.** Neutron energy groups

| **Energy Group No.** | **Lower limit (eV)** | **Upper limit (eV)** |
|---|---|---|
| **1** | 5.22E+06 | 1.73E+07 |
| **2** | 1.00E+06 | 5.22E+06 |
| **3** | 4.90E+05 | 1.00E+06 |
| **4** | 1.00E+05 | 4.90E+05 |
| **5** | 9.10E+03 | 1.00E+05 |
| **6** | 5.30E-01 | 9.10E+03 |
| **7** | 0.00 | 5.30E-01 |

**Neutron flux calculations:** In order to calculate the activity of the concrete it is necessary to have neutron energy spectrum and group neutron flux at the point of interest. For this reason, and on the basis of the neutron intensity which was obtained from our previous work, the group neutron fluxes at the surface of the concrete wall were calculated. Then by applying one-dimensional computational code ANISN/PC together with the group cross section libraries IRAN4.LIB and EXFOR, the group neutron fluxes at different depths of the concrete wall were calculated. The neutron energy groups used in this work are shown in Table 3.

The thickness of concrete wall assumed to be 100 cm and the calculations were performed for every 5 cm step in the concrete. By applying the above mentioned data the group neutron flux distribution in the concrete wall from thallium and gallium targets neutron source were calculated and the results are shown in Figs. 3 and 4.

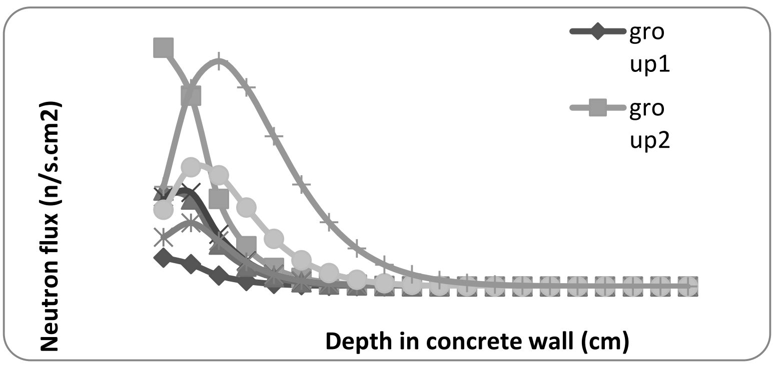


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**Fig. 3**. Group neutron flux distribution in the concrete wall. Thallium target is the neutron source

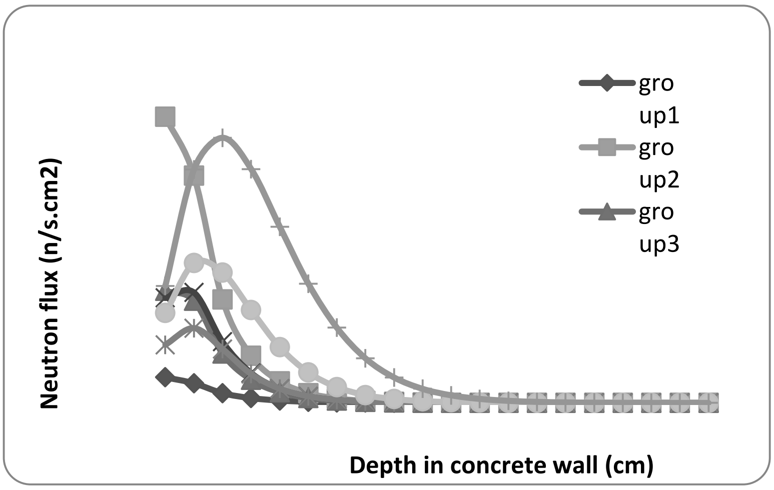


**Fig. 4**. Group neutron flux distribution in the concrete wall. Gallium target is the neutron source

**Calculation of activity:** The activity of each radioisotope is calculated on the basis of activity's general formula: $A_0 = \varphi(\varepsilon). N. \sigma(\varepsilon)$ (1)

where $A_0$ is the saturated activity (disintegration/s.cm$^3$) of any isotope, $\varphi(\varepsilon)$ is the neutron flux at energy group ε (n/s.cm$^2$), N is atomic density of the stable isotope in the concrete (cm$^{-3}$) and $\sigma(\varepsilon)$ is the average (n,γ) reaction cross section at energy group ε (cm$^2$). If the cyclotron accelerator is run for $t_1$ sec. and then to be shut down for $t_2$ sec., then the activity of the induced radioisotope at the end of shutting down time will be:

$A_1 = A_0 (1 - e^{-\lambda t_1}) e^{-\lambda t_2}$ (2)

where $A_1$ is the activity of induced radioisotope after one cycle of cyclotron operation and λ is its decay constant. One cycle of cyclotron operation, T, is defined as the run time plus shut down time i.e. $T = t_1 + t_2$. The activity after the second cycle of operation will be:

$A_2 = A_0 (1 - e^{-\lambda t_1}). e^{-\lambda t_2} + A_1 e^{-\lambda T}$ (3)

where the first term on the R.H.S. is the activity after one cycle of operation and the second term on the R.H.S. is the activity remained from the first operating cycle. By combining Eqs. 2 and 3 we can write:

$A_2 = A_1 + A_1 e^{-\lambda t_2}$ (4)

with the same argument, the activity after the third operating cycle will be:

$A_3 = A_0 (1 - e^{-\lambda t_1}). e^{-\lambda t_2} + A_2 e^{-\lambda T}$ (5)

where the second term on the R.H.S. is the activity remained from the second operating cycle. Substituting Eq. 4 into Eq. 5 and using the definition of $A_1$ from Eq. 2, one obtains:

$A_3 = A_1 + A_1 e^{-\lambda T} + A_1 e^{-2\lambda T}$ (6)

with the same procedure mentioned above, the activity after n operating cycles, which covers the 30 years of cyclotron operation, will be:

$A_n = A_1. \sum_{m=0}^{n-1} e^{-m\lambda T}$ (7)

in driving equation 7 it is assumed that in all operating cycles, the atomic density of the isotopes , N , do not vary, that is, the decrement of atomic density due to neutron absorption is ignored. Furthermore, the run time, $t_1$, and shut down time, $t_2$, remain constant in every operating cycle, i.e. the value of $A_1$ remains constant in all operating cycle.

Equation 7 was used to calculate the activities of the isotopes $^{152}$Eu, $^{60}$Co, $^{154}$Eu $^{134}$Cs and $^{41}$Ca. The other isotopes in Table 1 due to either short half-life (e.g. $^{28}$Al) or very long half-life (e.g. $^{40}$K), or stable isotopic product (e.g. $^{2}$H) were not being considered. The calculations were performed once for the neutron source resulting from thallium target and once for the neutrons from gallium target. In the first case it is assumed that the thallium target is bombarded by 28.5 MeV proton particles once a week for 10 hours continuously for a period of 30 years, i.e. $t_1$ for thallium is 10 hours and T is 168 hrs. (One week), whereas gallium target is bombarded by 19 MeV proton particles once every two weeks for 10 hours continuously, that is, $t_1$=10 hrs. and T=336hrs. (Two weeks). The total activity of each radioisotope at any time is simply the summation of the activity induced in both cases. The results of the activity as a function of time during 30 years of cyclotron operation are shown in Figs. 5 and 6 and the activity at the end of 30 years as a function of depth in concrete wall are shown in Figs. 7 and 8.

From the figures 5 and 6 it is seen that the activity after 30 years of cyclotron operation is quite low and can be neglected. The low activity is due to (a) low isotopic concentration; (b) low neutron flux; (c) long shutting down time in one cycle of operation, compare with the running time and (d) the half-lives of the radioisotopes (except $^{41}$Ca) are few times less than the total operating time of the cyclotron. It means that the activity curve reaches the region of saturation value after 30 years. This is more pronounced for the activity of $^{134}$Cs with a half-life of 2.065 years which reaches its saturation value in nearly 10 years.

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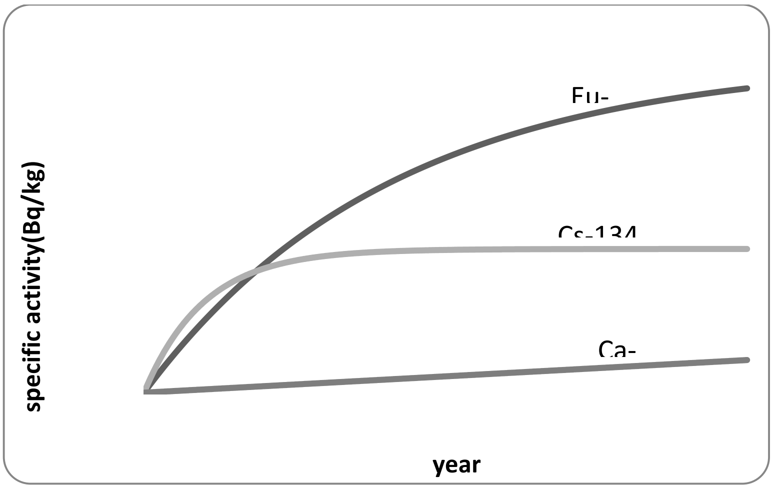


**Fig. 5** Specific activity of 134Cs, 154Eu and 41Ca in concrete wall during 30 years of cyclotron operation

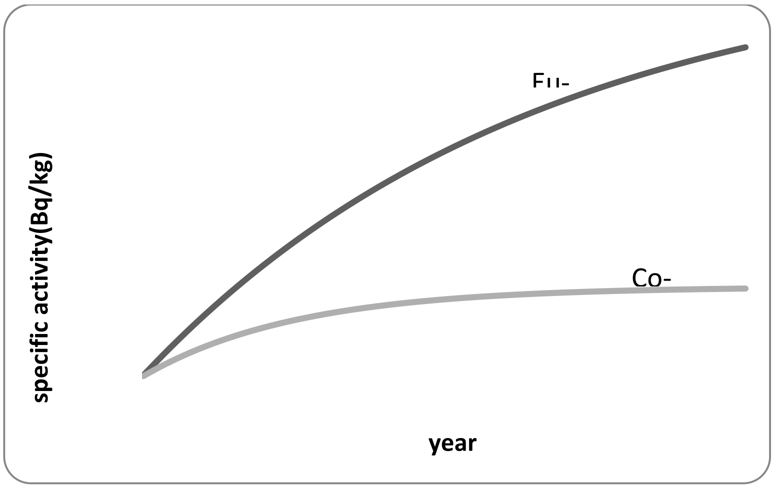


**Fig. 6**: Specific activity of 60Co and 152Eu in concrete wall during 30 years of cyclotron operation

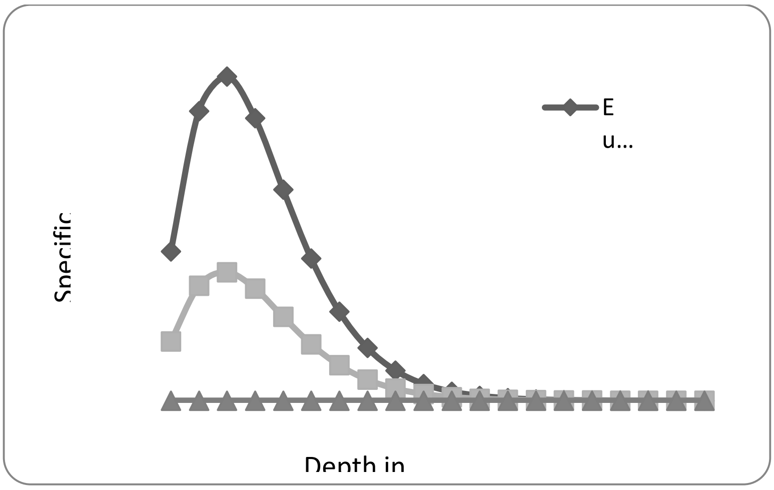


**Fig. 7**: Specific activity of $^{154}$Eu, $^{134}$Cs and $^{41}$Ca as a function of depth in concrete wall

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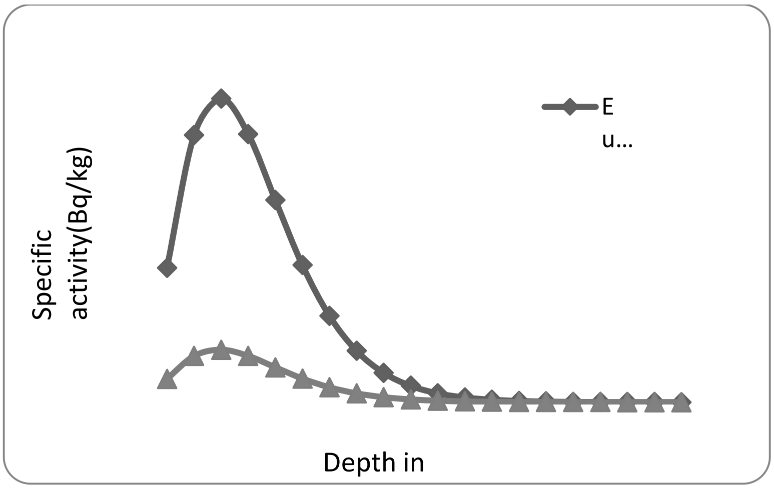


**Fig. 8**: Specific activity of $^{152}Eu$ and $^{60}Co$ as a function of depth in concrete wall

Also from the Figs. 7 and 8 it is seen that the activity mainly concentrated at depth of 15 cm in the concrete wall which is due to the thermal neutrons that have its maximum value at the depth of 15 cm from the wall surface(see Figs. 3 and 4).

## DISCUSSION AND CONCLUSION

Concrete as a structural material was used in construction and applied as shielding material at cyclotron department at NRCAM. Routine production of radiopharmaceuticals $^{201}Tl$ (once a week for 10 hrs. continuously) and $^{67}Ga$ (once every two weeks for 10 hrs. continuously) which are based on the bombarding of targets $^{203}Tl$ and $^{68}$ Zn by proton particles are accompanied by emission of high energy neutrons from the bombarded targets. As a result of the collision of emitted neutrons with the concrete walls, surrounding the target assembly, the walls become activated.

In this paper the neutron-induced activities in the closest concrete wall to the target assembly was investigated. The neutron intensities from the $^{203}Tl$ and $^{68}Zn$ targets were calculated to be $3.75\times10^{13}$ n/s and $1.16\times10^{13}$ n/s respectively. By using the one dimensional computational code ANISN/PC and group cross section data libraries IRAN4.LIB and EXFOR, the group neutron flux at different depth the concrete wall was calculated. As expected, the most neutron intensity in the concrete wall belongs to the thermal neutrons which have a peak at the depth of 15 cm from the wall surface. The elements constituting the concrete become activated as the incoming neutrons interact with them. In this work the activities of the trace elements $^{152}Eu$, $^{60}Co$, $^{154}Eu$, $^{134}Cs$ and $^{41}Ca$ ($^{40}Ca$ concentration in concrete is 4.4%) which have high (n,γ) reaction cross sections and reasonable half-lives were calculated. Our calculations show that the corresponding activity of the above mentioned radioisotopes, after 30 years of cyclotron operation, are 23.059 Bq/kg, 6.186 Bq/kg ,0.511 Bq/kg, 0.241 Bq/kg a 0.055 Bq/kg respectively. The values obtained are too low to be considered as a serious radiation hazard and may be ignored.

The assumptions made in calculation of activities were: (1) only (n,γ) reaction was considered and the other neutron reactions were not considered; (2) attenuation of neutron intensity traveling the target assembly wall was ignored; (3) the concentration values of the elements taken from the literature (and not experimentally). The results can be improved if: (a) two or three-dimensional computational code (instead of one-dimensional code) is applied to determine group neutron flux in the concrete, (b) finer group reaction cross sections, particularly in the thermal region, are used and finally (c) more reliable, experimental value for concentration of elements in the concrete is used.

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