Radioactivity generation in Pb target by protons — A comparative study from MeV to GeV

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In an Accelerator Driven Subcritical System (ADSS), choice of the target is decided by several factors like neutron yield, heat generation, ease of cooling, possibility of fire hazard, generation of chemically toxic elements and radioactive nuclides, running cost of the accelerator, etc. Lead (Pb) is one of the probable targets for an ADSS. In the present work, we have estimated induced activity in a Pb target by primary proton beam in the energy range of 20 MeV up to 2.0 GeV using reaction model codes ALICE-91, TALYS-1.2, EMPIRE-2.19 and QMD. The energy range studied spans the entire energy interval used for target property study to practical application of an ADSS. At several hundreds of MeV, some of the major contributors to induced activity are projectile-like fragments, such as, 3H. The maximum activity produced is of the order of 10^6-10^7 MBq over the whole energy range. Some chemically toxic elements like Xe, Hg are also formed in significant amount.

Keywords: Induced activity, ALICE-91, TALYS-1.2, EMPIRE-2.19, QMD, Thick target, Saturation activity

1 Introduction
Accelerator Driven Subcritical Systems (ADSS) has emerged as one of the preferred choices in nuclear power technology. ADSS employs high current proton beam induced spallation reactions at energies of several GeV. Choice of target in ADSS is dictated by several factors like high neutron yield, low risk of fire hazard in operating system, ease of cooling, low chemical and radio-toxicity production, relatively low running cost of the system. Studies on several target systems revealed that lead (Pb) is one of the preferred targets which produces good neutron yield and the neutron cost is optimized around and above 1 GeV. A typical ADSS employing a Pb target would be operated at these energies with a beam current of 1-3 mA. Production of induced radioactivity in such a system plays a key role in deciding final operating parameters. Also studies on target properties are carried out at much lower energies where protection of personnel against undue radiation exposure calls for estimation of induced activity in the system. In the present work, we have estimated the radioactivity induced in an ADSS employing a Pb target in the energy range of 20 MeV to 2 GeV. For this purpose, we have calculated the formation cross-section of product radioisotopes using nuclear reaction model codes and converted it to thick target yield.

2 Method of Calculations
The induced activity generated in lead (Pb) by proton induced reaction in the energy range of few tens of MeV up to 2 GeV, has been calculated. We have determined the formation cross-sections of various radioisotopes produced in Pb target using nuclear reaction model codes ALICE-91, TALYS-1.2 and EMPIRE-2.19 for proton energies up to ~200 MeV and using the code Quantum Molecular Dynamics (QMD) for higher energies. Using these calculated cross-sections, the yield of each of these isotopes and activity due to them, have been estimated.

2.1 ALICE-91
ALICE-91 is a well known code developed by Blann to calculate the reaction cross-section for nucleon and alpha particle induced reactions up to 200 MeV incident energy. This code calculates pre-compound emission cross-section in the framework of hybrid/geometry-dependent hybrid model (GDHM) where pre-emission energy distribution of excited particles is taken into account to determine the emission probability at that stage. In GDH version, the effect of longer mean free path of the particle at surface has been considered. Compound nuclear reaction cross-section is calculated in the framework
of Weisskopf-Ewing evaporation model. Level density of the residual nucleus after evaporation can be determined using Fermi gas or Gilbert Cameron formalism. In the present work, we have used optical model sub-routine to calculate cross-section of the reverse reaction channel. Fermi gas level density formalism, with level density parameter \( a = A/9 \), has been considered for residual nuclei where \( A \) is the mass number of the nucleus.

2.2 TALYS-1.2

The code TALYS\(^2\) calculates direct (DIR), pre-equilibrium (PEQ) and compound nuclear (EQ) emission cross-sections for nucleon induced reactions. DIR cross-sections are calculated using one of the different formalisms like distorted wave-Born approximation (DWBA), rotational or vibrational coupled channel analysis and giant resonances. Two-component exciton model estimates PEQ particle emission and the angular distribution of these PEQ particles is determined using Kalbach systematics. Compound nuclear emission is calculated in the framework of Hauser-Feshbach formalism in competition to fission. Here we have used different PEQ models along with different pairing options viz:

1. **PREEQMODE=2**: Exciton model; numerical transition rates with energy dependent matrix element, **PAIRMODEL=1**: Fu’s pairing energy correction.
2. **PREEQMODE=3**: Exciton model; numerical transition rates with optical model for collision probability, **PAIRMODEL=2**: Compound nucleus pairing energy correction.

2.3 EMPIRE-2.19

EMPIRE-2.19\(^3\) code calculates DIR, PEQ and EQ reaction cross-sections for neutron and proton induced reactions up to \(~150\) MeV above which pion production becomes significant. The code has provision to use any of the several PEQ models like multi-step direct (MSD), multi-step compound (MSC), exciton and hybrid Monte-Carlo simulation (HMS) to calculate PEQ cross-sections. It uses the statistical Hauser-Feshbach theory to calculate compound nuclear emission. Level density of residual nucleus which strongly influences evaporation spectra which can be determined using different formalisms like Fermi gas, Gilbert Cameron and Bardeen, Cooper, and Schrieffer model (BCS) + Fermi gas.

2.4 QMD

The code QMD\(^4,5\) calculates nuclear reaction through Monte Carlo simulation of the relaxation process. It uses a quantum molecular dynamics (QMD) approach to quantify initial high energy part of the equilibration process followed by a statistical decay approach of composite nucleus\(^4,6\). Transition from high energy cascade reaction to the evaporation of equilibrated compound nucleus occurs in a self-consistent manner through an intermediate PEQ stage. QMD calculations are done for 10000 histories, 1 fm/c time step with 7 fm maximum impact parameter.

2.5 Induced activity estimation

In an ADSS, full utilization of high energy beam is ensured by the use of a thick, large mass of target which fully stops the projectile beam. In such a thick target, an isotope is produced by the interaction of the incident particle at different degrading energies from incident energy down to reaction threshold\(^7\). The total yield of isotope is obtained by summing over the yield of the isotope over this entire energy range. The activity of a radioisotope ‘i’ is given by:

\[
A_i = \sum_j N \sigma_j(E_j) \phi_j(E_j) (1 - e^{-\lambda t_i})
\]

where \( A_i \) = activity of radioisotope ‘i’

\( N \) = number of target atoms available for reaction

\( \sigma_j(E_j) \) = cross-section for production of radioisotope ‘i’ at projectile energy \( E_j \)

\( \phi_j(E_j) \) = projectile flux at energy \( E_j \)

\( \lambda \) = decay constant of radioisotope ‘i’

\( t_i \) = irradiation time

3 Results and Discussion

The generation of induced activity by primary beam in an ADSS employing \(^{208}\)Pb target operating in the energy range of 800 MeV to 2.0 GeV has been studied. Radioactivity induced in target at much lower beam energies, \(~20-250\) MeV, which might be used for target property analysis, have also been studied. We have considered a beam current of 1 mA, an irradiation period of 1-30 days and a cooling time of \(10^4\) to \(10^7\) s. Figure 1 shows the activity induced in a Pb target due to \(^{205,206,207,208}\)Bi and \(^{204,207}\)Tl, by proton induced reaction at 20 MeV (ALICE-91). The induced activities are calculated using ALICE-91, TALYS and EMPIRE 2.19. The activity calculated by ALICE91 and TALYS agrees with each other well while for some product isotopes EMPIRE calculated values are a little higher (TALYS and EMPIRE results not shown here). This may be due to the fact that EMPIRE calculates DIRECT reaction and direct emission at 20 MeV proton energy is likely to
populate the residual nuclei close to the target nucleus below their particle emission threshold. Maximum activity of the order of $10^6$ MBq is produced for $^{206}$Bi ($t_{1/2} = 6.24$ d) and activity reaches its saturation value for an irradiation period of 30 days. Considerable activity is also produced for $^{205}$Bi ($t_{1/2} = 15.31$ d) ~$10^5$MBq but is less than that for $^{206}$Bi. Yield of $^{207}$Bi ($t_{1/2} = 31.55$ y), $^{208}$Bi ($t_{1/2} = 3.68 \times 10^5$ y) and $^{204}$Tl ($t_{1/2} = 3.78$ y) are much less than the maximum activity and are of the order of $10^4$, $10^5$ and $10^5$ MBq, respectively. Saturation activity is not reached for any of these isotopes in the irradiation time taken as they are much longer lived isotopes.

In Figs 2 and 3, we have shown activity induced in the Pb target due to some isotopes at proton energies of 100 MeV and 250 MeV, respectively. From Fig. 2, it is observed that activity of $^{206}$Tl and $^{207}$Bi are of the order of $10^5$ and $10^6$ MBq, respectively, much higher than that produced at 20 MeV proton energy. Other isotopes of Hg and Tl, further away from the target nucleus have larger contribution to the total induced activity generated. In Fig. 3, we have compared induced activities for $^{205}$Bi and $^{207}$Bi calculated using three codes ALICE-91, TALYS-1.2 and EMPIRE-2.19. For $^{207}$Bi, activities calculated by three codes agree well. In the case of $^{205}$Bi, EMPIRE calculations give a lower value than those from other two formalisms. This may be attributed to the fact that if DIR emissions are over-predicted, then this may result in a lower value of the production cross-section of an isotope further away from composite system.

As we move on to higher operating energies, the activity induced in an ADSS employing a Pb target and a proton projectile in the energy range of 800 MeV to 2.0 GeV is estimated using the code QMD. In order to validate the code QMD, we have compared in Table 1 the cross-section for production of $^{200}$Tl and $^{198}$Au from $^{p+p_{nat}}$Pb at 660 MeV and 1.5 GeV, calculated by QMD with experimentally measured values. From Table 1, it is seen that calculated values are fair approximation of measured cross-sections. In absence of any other convenient tool, QMD code can be used to approximately estimate production cross-section, and hence induced activity, in this energy range.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Energy (MeV)</th>
<th>Cross-section (mb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{nat}$Pb (p, X) $^{200}$Tl</td>
<td>660</td>
<td>$28.0 \pm 8^{\dagger}$</td>
</tr>
<tr>
<td>$^{nat}$Pb (p, X) $^{198}$Au</td>
<td>660</td>
<td>$1.8 \pm 0.3^{\dagger}$</td>
</tr>
<tr>
<td>$^{209}$Bi(p,7n5p) $^{198}$Au</td>
<td>1500</td>
<td>$0.4 \pm 0.1^{\dagger}$</td>
</tr>
</tbody>
</table>
Figures 4 and 5 show the growth of activity of light-mass fragments for 1 mA p + 208\(^{\text{Pb}}\) at 800 MeV and 1.6 GeV, respectively.

In this high energy range, the dominant contribution to the induced activity comes from projectile-like and low mass fragment along with that from target-like fragments unlike in the case of low energy reaction where only nuclei close to the target mass are predominant.

In Figs 4 and 5, we see that the highest activity is produced for \(^{32}\text{P}\) at both the energies and is of the order of \(10^7\) MBq. \(^{32}\text{P}\) is a \(\beta\)-active isotope with a half-life of 17.3 days. Hence, it does not pose long-term hazard and should be protected against skin and internal contamination after the system shut-down. But these two Figs 4 and 5 show that significant activity of \(^3\text{H}\) and \(^{60}\text{Co}\) are produced. \(^3\text{H}\) is a long-lived \(\beta\)-emitter with a half-life of 12.33 years and poses a problem of ground water contamination. \(^{60}\text{Co}\) has a half-life of 5.27 years with two characteristic gammas at 1.17 and 1.33 MeV. Precaution should be taken against external exposure from these sources. It is seen that saturation activity is not reached for any of the isotopes shown here.

In Fig. 6, we have compared activity of \(^3\text{H}\), \(^{14}\text{C}\) and \(^{60}\text{Co}\) at 800 MeV and 1.6 GeV produced after 30 days of irradiation. Fig.6 shows that \(\sim10^8\) MBq activity of \(^3\text{H}\) is produced while that of \(^{14}\text{C}\) is much lower and is \(\sim14 - 43\) MBq.

In Fig. 7, we have shown the activity produced in Pb target due to \(^{200}\text{Tl}\), \(^{203}\text{Pb}\) and \(^{207}\text{Bi}\) at 800 MeV and 1.6 GeV proton energies. All these product nuclides are close to target and represent the target-like component. Fig.7 shows that for \(^{200}\text{Tl}\) and \(^{203}\text{Pb}\) activity \(\sim10^8\) MBq is produced in 30 days of irradiation with an 1 mA beam current. In case of \(^{207}\text{Bi}\), this activity is \(\sim10^4\)MBq. For all these three

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**Fig. 4** — Activity build-up of light-mass fragments, calculated using QMD, due to 1 mA p + 208\(^{\text{Pb}}\) reaction at 800 MeV

**Fig. 5** — Activity build-up of light fragments, calculated using QMD, due to 1 mA p + 208\(^{\text{Pb}}\) reaction at 1.6 GeV

**Fig. 6** — Activity of projectile-like and light mass fragments, calculated by QMD, for 1 mA p + 208\(^{\text{Pb}}\) at 800 MeV and 1.6 GeV

**Fig. 7** — Activity of target-like and heavy mass fragments, calculated by QMD, for 1 mA p + 208\(^{\text{Pb}}\) at 0.8 and 1.6 GeV
isotopes induced activity generated at 1.6 GeV is slightly less than that at 800 MeV. Both the above observations can be explained from the fact that as we go to higher beam energies more reaction channels open up and nuclei further away from target+projectile composite nuclei are likely to be formed.

In Fig. 8, we compared the induced activity generated for isotopes in the mass range of $A = 125-195$ in $^{208}$Pb target by 1 mA proton beam for incident energies of 1.2 GeV and 2.0 GeV after irradiation for 30 days. Fig. 8 shows that in this mass range maximum activity produced is $\sim 2 \times 10^6 - 4 \times 10^6$ MBq for $^{195}$Au (186.0 d).

Activity of $^{125}$I (60.2 d) and $^{127}$Xe (36.4 d) are also in the range of $10^5 - 10^6$ MBq. Xe is a chemically toxic gas. Proper precaution should be taken to protect personnel and equipments against exposure to Xe. Minimum activity is produced for $^{194}$Hg and is of the order of $4 \times 10^3$ MBq.

4 Conclusions

The present study shows that in an ADSS employing a $^{208}$Pb target significant activity $\sim 10^6$ MBq of $^3$H and $^{60}$Co are formed. $^3$H poses a threat of environmental radiation hazard through ground water contamination. $^{60}$Co being a relatively longer lived isotope should be properly handled and disposed to minimize external radiation hazard. It is observed that for a $^{208}$Pb target the generation of induced activity increases for some of the radioactive products while it decreases for others with increasing beam energy. Since one of the major applications of ADSS is as source for generation of neutrons, a balance must be struck between the neutron economy and chemical and radio-toxicity generation. Further study in this direction is required for characterization of the ADSS target.

In the present paper, the radioactivity generated in the Pb target by the primary beam only, has been presented. But for a practical system, the total inventory includes the activity produced by the secondary neutrons in the target as well as in the surrounding material and should be taken into account.

References

3 Herman M, EMPIRE nuclear reaction model code (2.19 Lodi) (IAEA, Vienna, Austria) 2005.