Decay Nature of Radionuclide Released From Triga Mark-II Reactor

M. Ashaduzzaman, D.M.S. Zaman and T. Akhter

Abstract: Considering a hypothetical accident, the deposition of radio krypton ($^{85}$Kr) has been studied that is released from TRIGA MARK-II reactor by its decay behavior. The measurement of radiological ground concentration leads to the study of the emission process of $^{85}$Kr nucleus, and the site-specific data related to this measurement have been analyzed later on. In this work, the radioactivity in the reactor core and release rate as well as Gaussian diffusion factor have also been considered. It is observed from the data analysis that the maximum concentration of $^{85}$Kr in ground is 1.115E+3 Bq/m$^2$ in South (S) direction. Here, the nuclear binary fission has been presented with the greater probability of production of magic nuclei where the reactions found to be endoergic where a nuclear reaction occurs with the absorption of energy. Doubly magic nuclei like $^4$He, $^{16}$O, $^{40}$Ca, and $^{48}$Ca have also been identified as fission fragments that follow the nuclear shell closure. The results of this work will be an important guide in the study of radionuclide splitting into several nuclei as well as for accidental scenerio.

Index Terms: Radiological Concentration, TRIGA Reactor, Nuclear Reaction, Magic Nuclei.

I. INTRODUCTION

CALCULATION of atmospheric dispersion from accidental decays of radioactive elements are essential. It is needed to select a proper site for reactor by considering international criteria [1]. As well as volatiles are also an important parameter for such selection. Generally nuclear reactor does not emit radioactive elements. But, in accidental cases there would be an emission of radioactive materials from the destruction of reator parts. In such scenerio, radionucl can be deposited to human body and on the ground. This can occur by various ways, for instance external irradiation, inhalation, etc. [2, 3].

Immediately after an accident, $^{85}$Kr isotope present the most serious radiological hazard [4]. Excessive inhalation of the isotope may create dizziness, nausea, vomiting, loss of consciousness, and death of humans. The pollution and atmospheric effect of $^{85}$Kr and its meteorological impacts has been presented by R.G. Harrison in 1994 [5]. The analzyation of site-specific data is an crucial factor. Several considerations and methods have been studied for this purpose. Several methods are established by IAEA for the study of the data in the determination of radiologocal doses [6, 7].

Due to the low ratio of neutrons and protons, $^{85}$Kr isotope will decay by $\beta$- emission by the following way:

$$^{85}_{36}Kr\rightarrow ^{85}_{37}Rb+^{0}_{-1}e^-$$

where Rubidium-85 nucleus will generate as a decay product. Radiological concentration has been calculated by developing a computational program. Emission rates from several gases and halogens has been found out by IAEA [7]. The determinations has been done by taking data from Bangladesh Meteorological Department. Several significant values has been also used from other works for this calculation [6].

II. SOURCE TERM AND ACCIDENT SCENARIO

A number of parameters are needed for the calculation of radiological concentration such as activity and considered variables.

A. Source Term Calculation

Several variables can be used to measure source term that reflects decayed amount of radioactive materials. Activity can be calculated with the help of fission yield ($\gamma$), decay constant ($\lambda$), time duration ($T$), and thermal power ($P$). The equation for calculating activity can be presented [8] below:

$$A_i(t) = 0.82\gamma P (1 - e^{-\lambda_i T}) \times e^{-\lambda_i (t-T)}$$

(1)

Where, $A_i(t)$ is the activity of an isotope i at time t after the start of irradiation ($t=0$).
The total activity is found with the help of fractional release ($F_p$), source term ($\lambda_l$), leak rate parameter ($\lambda_l$), and decay constant ($\lambda_r$). This can be calculated by following equation [9]:

$$Q_i(\tau) = F_p F_B A_i (t) \left( \frac{\lambda_l}{\lambda_l + \lambda_r} \right) \left[ 1 - e^{-(\lambda_l + \lambda_r) \tau} \right]$$  \hspace{1cm} (2)

where $Q_i(\tau)$ is total activity of isotope i released over time $\tau$.

B. Assumed Parameters

The deposition of radionuclide around the reactor mainly depends on the source term. Release of radiokrypton ($^{85}$Kr) is considered after the accident. One of the assumed variables is the power of the reactor (3 MW). Another parameters are total operating time (10 days), radioactive decay time after accident (2 hours), and fraction emission [10]. The activity of the radionuclide has been determined with the help of Eq.1 and fission yield were also calculated by using above assumptions. Table 1 represents the obtained parameters for the above mentioned parameters.

### Table 1. Determined parameters of radioactive krypton.

<table>
<thead>
<tr>
<th>Radioactive Nucleus</th>
<th>Fission yield</th>
<th>Overall activity in core ($C_i$)</th>
<th>Decay rate ($Bq$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{85}$Kr</td>
<td>0.0031</td>
<td>13.351</td>
<td>$5.716 \times 10^4$</td>
</tr>
</tbody>
</table>

III. GAUSSIAN PLUME MODEL (GPM)

Gaussian plum model [10] is mainly used for the measurements of atmospheric dispersion and concentration.

A. Ground Deposition

The deposition rate ($d_i$) depends on dry and wet deposition coefficients ($V_d$ and $V_w$) and can be calculated by following equation:

$$d_i = (V_d + V_w) C_A$$  \hspace{1cm} (3)

The sum of $V_d$ and $V_w$ is the total deposition coefficient ($V_T$). Determination of precipitation rate [11] is also essential parameters for this calculation. $V_T$ =1000 m/d can be considered for screening purposes [12, 13, 14] for radionuclide.

B. Ground Concentration

The ground concentration ($C_{gr}$) can be measured with the help of effective rate constant ($\lambda_{Ei}$), duration of the discharge ($t_D$), and full ground deposition rate ($d_i$) by the following equation [6],

$$C_{gr} = \frac{d_i \left[ 1 - \exp(-\lambda_{Ei} t_D) \right]}{\lambda_{Ei}}$$  \hspace{1cm} (4)

IV. NUCLEAR FISSION

The splitting of a nucleus into several other nucleuses is known as nuclear fission. This emission process was revealed in 1938 [15, 16]. After that liquid drop model (LDM) [18] was used to elaborate the process. This type of fission can occur by radioactive decay as well as in nuclear reaction. Later on, several works were done to establish the reaction dynamics of the process. [19-21]. The two newly produced nuclei in the binary decay process also known as primary fission fragments or products from fissioning nucleus.

A. Energetics of Nuclear Fission Process

The energy released or absorbed is known as $Q$ value of the reaction which can be obtained by applying energy conservation law. When energy is released from a reaction, then this reaction is known as exothermic reaction. Energy is produced if total energy of the projectile and target is greater than the total energies of fragments. Generally, produced energy is released in the form of heat energy. On the other hand, when a reaction requires external energy to proceed the reaction then it is said as an endothermic reaction since the kinetic energy of the products is less than the kinetic energy of the reactants.

V. RESULTS AND DISCUSSION

For the measurement of the concentration, a computer program has been demonstrated. This simpler code has been written by using MathCAD Software. The results of the calculations are presented in the following portions.

A. Concentration In Different Pathways

1) Utmost Air Concentration for several Directions:

Release rate is one of the essential parameters on which Air concentration of a radionuclide depends on.

Fig. 1: Air concentration for $^{85}$Kr.

Air concentrations were calculated with respect to the distance in 8 cardinal directions using the release rate mentioned above. The utmost air concentration for various directions were measured at 110 m distance from the core of the reactor. After this point, the air concentrations are reduced exponentially by GPM (Eq. 4). The maximum value was determined in the southern (S) side. For all other seven directions, the values are found nearer to S-direction.
2) Ground Concentration:

There are several factors on which the amount of radioactive element deposited in the ground depends on. The significant cases are the dry and wet deposition on the ground surface. Eight directions have been considered for concentrations calculation which is shown in Fig. 2. Concentrations have been calculated immediately after the accident in all directions and noticed as highest values. Table II shows the calculated concentrations for various directions. Among all values, maximum ground concentration was seen in S-direction and the values is 1.115E+3 Bq/m² for ⁸⁵Kr. As the time increased from accident time, concentrations are seen to reduce exponentially.

**TABLE II:** Highest ground concentration for various directions just after the accident.

<table>
<thead>
<tr>
<th>Direction</th>
<th>N</th>
<th>NE</th>
<th>E</th>
<th>SE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ground concentration (Bq/m²)</td>
<td>385.47</td>
<td>260.23</td>
<td>276.92</td>
<td>489.75</td>
</tr>
<tr>
<td>Direction</td>
<td>S</td>
<td>SW</td>
<td>W</td>
<td>NW</td>
</tr>
<tr>
<td>Ground concentration (Bq/m²)</td>
<td>1.115E+3</td>
<td>292.91</td>
<td>388.65</td>
<td>536.06</td>
</tr>
</tbody>
</table>

Fig. 2: Ground concentration of ⁸⁵Kr in eight directions.

B. Decay Nature of ⁸⁵Kr

The distribution of Q values for the fragmentation of ⁸⁵Kr isotope has been presented in Fig. 3 which shows the decay nature of the radionuclide is the endothermic process. The system requires very little energy to decay into secondary fragments and around 20 MeV of energy is the maximum possible value for the emission.

Fig. 3: Q-value distribution in the decay process of ⁸⁵Kr.

The distribution of charges (proton) of the primary fragments generated from the ⁸⁵Kr has been shown in Fig. 4. The distribution shows that the possibility to decay into products nuclei and charges in between 15 to 25 (Phosphorus to Manganese) has the greater possibility in the emission process.

Fig. 4: Charges distribution of the fragments in the decay process of ⁸⁵Kr.

Fig. 5 represents the distribution of neutron numbers of the primary fragments produced from the ⁸⁵Kr. Fragments having neutron numbers in between 16 to 34 has the greater possibility to be created in the initial decay process.

Fig. 5: Neutron numbers distribution of the fragments in the decay process of ⁸⁵Kr.
The amount of energy needed for the system to get decayed has been presented in Fig. 6 with respect to fragment nucleons numbers. The minima in the center region reflects the highest possible fragment having charge number 20 and neutron number 28 which is $^{48}\text{Ca}$.

Fig. 6: Energetics ($Q$-value) of the decay process with respect to nucleons numbers.

By considering energetics in the emission process and the shell closure effects on the magic number (2, 8, 20, 28, 50, 82, 126) nucleon, we have come to a conclusion that some specific fragments can get decayed in the initial process very rapidly and these fragments are presented in Fig. 7. The doubly magic nuclei $\text{He}$, $\text{O}$, and $\text{Ca}$ has the greater possibility to be generated in the decay process.

Fig. 7: Selective decay products on the emission of $^{85}\text{Kr}$.

The Q values has been calculated for possible produced products by using the following relation:

$$Q = m_{\text{Kr}} - \sum_{i=1}^{2} m_{i}$$

Where $m_{\text{Kr}}$ is mass excess of the $^{85}\text{Kr}$ and $m_{i}$ indicates the fragments, which are expressed in MeV. $Q$ value has been calculated for most probable fragmentations of $^{85}\text{Kr}$ these are presented in Table III.

TABLE III: Most Probable produced fragments.

<table>
<thead>
<tr>
<th>Parent Nucleus</th>
<th>Product Nuclei</th>
<th>$Q$ (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{85}\text{Kr}$</td>
<td>$^{4}\text{He} + ^{31}\text{Se}$</td>
<td>-7.50789</td>
</tr>
<tr>
<td></td>
<td>$^{16}\text{He} + ^{35}\text{Se}$</td>
<td>-58.50751</td>
</tr>
<tr>
<td></td>
<td>$^{40}\text{O} + ^{40}\text{Ni}$</td>
<td>-16.75768</td>
</tr>
<tr>
<td></td>
<td>$^{32}\text{S} + ^{48}\text{Ca}$</td>
<td>-10.35828</td>
</tr>
<tr>
<td></td>
<td>$^{48}\text{Ca} + ^{42}\text{S}$</td>
<td>-42.64407</td>
</tr>
</tbody>
</table>

The energetics of the process shows that $^{4}\text{He}$ and $^{32}\text{Se}$ are the easily decayed products in the emission process. Other than this, $^{48}\text{Ca}$ accompanied with $^{35}\text{S}$ and $^{40}\text{O}$ accompanied with $^{56}\text{Ni}$ has also the greater possibility to be created.

VI. CONCLUSION

A computational code has been written for the concentration measurements. Radionuclide concentration are found from a imagined accidental case of TRIGA MARK-II research reactor. Based on the assumed hypothetical accidental case, activity of the radio krypton ($^{85}\text{Kr}$) in the reactor core and decay rate was measured. The maximum concentration in ground of $^{85}\text{Kr}$ was found to be $1.115 \times 10^7$ Bq/m$^3$ in South (S) direction among 8 directions as a function of time. The concentration in the various directions is very low in other mentioned 8 directions, because of less frequency of the wind in these directions and hence the determination of the concentrations along these pathways was out of consideration in this study. The decay nature of radio krypton has also been analyzed with respect to the energy needed for the decay, which shows the number of decay products created in the emission process. Nucleus having doubly magic number needs very low energy to get decayed. $^{4}\text{He}$, $^{16}\text{O}$, $^{48}\text{Ca}$, and $^{48}\text{Ca}$ have been measured as products nuclei with greater possibility to be released.

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