Influence of the Incident Particle Energy on the Fission Product Mass Distribution*

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produced, extracted, and separated continuously at a reasonable rate to feed the second accelerator-stage. The greatest concerns are the integrity of the target and the efficient removal of the radioisotopes of interest from the target material.

As mentioned above, two basic types of target configuration are under investigation. One is a low-density uranium carbide target which is directly bombarded by the particles of the first stage accelerator beam. The second type of target is a two-step target in which a target is bombarded by primary beam particles. In this process secondary particles are generated (mainly neutrons) which are going to interact with a separate target. The secondary target, low-density uranium carbide, is placed down stream from or around the primary target. The clear advantage of the two-step type of target is that the extraction of beam power deposited in the primary target is separated from the problems of isotope extraction from the secondary target. On the other hand, the direct target type has the advantage of being more compact and also allowing the use of direct fission between the particle beam and the uranium carbide.

III. ENERGY DEPENDENT $^{235}\text{U}$ FISSION CROSS SECTION

The magnitude of the cross section for different incident particles is an important aspect in assessing the fission product generation rate. In this section the cross section values obtained with the fission nuclear models included in the LAHET code are compared. The fission nuclear models compared are the ORNL [4] and the RAL [5] models. Figures 1 to 3 present the cross section values obtained with the two models for neutrons, protons, and deuterons, respectively. In Figure 1, which presents neutron cross sections, the data below and at 20 MeV is from the ENDF-BVI [6] and above 20 MeV the cross sections are calculated by the LAHET code, with either the ORNL or RAL model. It is interesting to notice that neither the ORNL nor the RAL nuclear models match the evaluated nuclear data for $^{235}\text{U}$ fission at 20 MeV. The RAL model produces a 34% smaller cross section at 20 MeV than the evaluated data, while the ORNL calculated cross section is 15% smaller than the evaluated data at that energy. The deviation between the models varies with the energy, being larger at lower energies than at higher energies. Figure 4 displays the comparison of the cross section data for neutron, proton, and deuteron calculated by the ORNL model and using ENDF-BVI data for neutron energies below 20 MeV. The overall fission yields for the three particle types are within a factor of two of each other at energies above 50 MeV.

IV. FISSION PRODUCT YIELDS AS A FUNCTION OF THE MASS NUMBER

In this section we compare the mass distribution of the fission products for $^{235}\text{U}$ bombarded by neutrons, protons, and deuterons at different energies. Generally, there are two well-separated mass peaks characteristic of asymmetric fission at lower energies, and the valley is filled in by symmetric fission at higher energies.

The analysis presented here is based on evaluated cross sections for neutron energies below 20 MeV and on the ORNL fission model evaluated via the LAHET code system for beam energies from 20 to 150 MeV. The ORNL fission model has been successfully benchmarked against fission isotopic yields for rubidium and cesium for proton beams of 50 and 156 MeV [7,8]. It was shown [7] that the RAL model predicts unphysical wide isotopic distribution for fission induced by protons on uranium at these energies.

Figure 5 displays the calculated $^{235}\text{U}$ fission product mass distribution for neutrons of several energies. The influence of the neutron energy on the distribution is very clear and it is more dramatic, as we can see, in the energy range from the fast fission to 20 MeV. As the energy of the incident neutron increases the rate of change in the distribution with the energy reduces considerably. From this plot we can say that the advantage of having a higher incident neutron energy is the possibility of producing a broader range, with significant yield, of fission product masses. On the other hand, as it is shown later, the evaporation stage of the fission process is more intense as the energy of the incident particle increases, resulting in fission products with less neutrons per atomic number. The optimal energy for the production of a specific isotope is determined by the interplay of these two phenomena.

Figure 6 shows a plot of a normalized mass distribution for protons and deuterons of 20 and 150 MeV. As it can be noticed, the mass distributions for deuterons are more peaked than the ones for protons. Also, the general trend of higher energy incident particles producing more isotopes within the intermediate mass range (from 100 to 130 amu) is clearly noticeable. If one makes a closer comparison between the neutrons' and the charged particles' fission product mass distribution, one would see that they do not differ too much for the energies considered in this analysis. However one has to keep in mind that the mass distribution presented in this section does not consider the atomic number of the fission products. This aspect of the problem will be addressed in the next section.
Figure 1. Comparison of the neutron-induced fission cross section for $^{238}$U.

Figure 2. Comparison of the proton-induced fission cross section for $^{238}$U.

Figure 3. Comparison of the deuteron-induced fission cross section for $^{238}$U.

Figure 4. Comparison of the neutron, proton, and deuteron $^{238}$U fission cross sections.

Figure 5. Influence of the neutron energy on the fission product mass distribution of $^{238}$U.

Figure 6. Influence of the charged particle's (protons and deuterons) incident energy on the fission mass distribution.
V. FISSION PRODUCT YIELDS PER ELEMENT

In this study we restrict ourselves to analyze only a few elements of interest and their predicted isotopic distribution as products of fission induced by neutrons, protons, and deuterons on "U and "U at different energies. The elements selected for this study are rubidium, cesium, xenon, tin, and krypton. The incident energies chosen were thermal (thermal fission reactor spectrum from 1.e-09 to 1.e-04 MeV), fast (fast breeder fission reactor spectrum from 1.e-04 to 5 MeV), D-T (14 MeV D-T fusion neutrons), 20, 50, 100, and 150 MeV for neutrons, and 20, 50, 100, and 150 MeV for protons and deuterons. The minimum energy for charged particles was set to 20 MeV due to the steep decrease in the fission cross section below that energy. The maximum energy of 150 MeV was set based on a preliminary design for a 200 MeV charged particle beam. In the "U target case, it is important to note that the use of the fast fission reactor spectrum makes the reaction rate to be relatively small because this spectrum is dominantly below the energy threshold for neutron induced fission of "U. Hence, in all the analysis that follows, if the neutron spectrum of interest, in the fast neutron energy region, has most of the neutrons in the energy range above 1 MeV, then the yields from the fast neutrons on "U should be renormalized to the same order of magnitude as the 14 MeV ones.

The total amount extracted of a particular isotope from the target depends on the isotopic mass distribution of the element under consideration and on the total production rate of the element itself. Table 1 and 2 provide a compilation of the results of the yields per element for "U and "U, respectively. The configuration consisted of a thin target (0.05 centimeter thick or 1 cm²) of either "U or "U, bombarded by neutrons, or, protons, or deuterons at different energies. The results are given per incident particle. Since this is a thin target configuration, the effect of neutron multiplication and production of secondary particles is not considered and they have no influence in the results presented.

The results for "U show the strong contribution that can be achieved by thermal neutrons in that case. Furthermore, the relative contribution of the 150 MeV particles to the yields is, generally speaking smaller than the one from the 50 MeV particles. This is basically due to the fact that the elements analyzed have atomic masses close to the fission peaks and, as shown before, as the energy of the incident particle increases, less pronounced are those peaks. In Table 2 it is possible to see the influence of the fast fission reactor spectrum on the production of these elements; there is a considerably smaller amount produced than it would be expected if the neutrons had energy between 1 and 2 MeV.

In these tables and all of the figures that follow the cross sections in millibarns are the yields presented multiplied by 4x10⁵. Also, in Tables 1 and 2 the yields tabulated for 20 MeV neutrons are calculated via LAHET code system.

VI. FISSION PRODUCT YIELDS PER ISOTOPE.

The mass distribution for each of the elements considered in this analysis is given in this section along with a few comments about the elements and their isotopes. For each element considered two figures are shown, one for "U and another for "U fission. These figures show only selected results regarding the energy of the incident particles. In the "U cases the results shown are the yields per incident particle for 100 MeV protons, deuterons, and neutrons, and thermal, fast, and 14 MeV neutrons. In the "U cases the results are presented for the same energies except for the thermal neutrons due to the negligible fission cross section for this case. All results are for a very thin target (0.05 centimeter thick) bombarded by particles at the indicated energies.

Neutron induced fission above and below 20 MeV are treated separately. Above 20 MeV the number of fission events and the mass distribution of the fission products is calculated by the LAHET code. Below 20 MeV the fission products yields are calculated based on the evaluated data sets [2] for three energies, namely, thermal, fast, and 14 MeV neutrons. The number of fission events in the system is calculated using the Monte Carlo MCNP code [9] with a source of neutrons having thermal, fast, or 14 MeV neutron spectrum. Then, a computer program calculates the mass distribution of the elements of interest based on the number of fission events per energy range and individual yields. The energy boundaries were arbitrarily set in such a way that thermal, fast, and high (14 MeV) fission product yields are used for neutrons below 100 eV, between 100 eV and 5 MeV, and above 5 MeV, respectively.

A. Krypton Isotopes from Fission.

Krypton is a noble gas and its isotopes are in the low-mass peak region of the fission product mass distribution. It has an atomic number of 36 and the naturally occurring isotopes of krypton have mass numbers between 78 and 86. Figure 7 and 8 display results of the calculated thin target production of krypton isotopes for "U and "U, respectively. For "U fission, the peak of the krypton isotopes mass distribution is at 91 for thermal, fast, and 14 MeV neutrons, and at 89 for 50 MeV protons and 100 MeV neutrons or deuterons.

On the other hand, for the "U fission the peak is at 92-93 for fast neutrons, and at 92 for 14 MeV neutrons.
The 100 MeV distributions for neutrons, and deuterons show some kind of odd-even effect, which produce peaks at 90 and 92 for neutrons and deuterons. The proton distribution for 100 MeV also presents an odd-even effect, but it is less pronounced. These results indicate that, for this element, the fission products from \(^{235}\text{U}\) fission are richer in neutrons than \(^{235}\text{U}\) fission, but both have more neutrons than the naturally occurring isotopes. The results also show that as the energy increases the mass of the fission products decrease, basically due to the evaporation of neutrons in the compound nucleus during the fission process. The isotopic yields from thermal neutrons on \(^{252}\text{U}\) are generally the largest except for the most neutron rich. For the most neutron rich isotopes, the yields from the 14 MeV neutrons become comparable.

### B. Rubidium Isotopes from Fission

Rubidium isotopes are in the low-mass peak region of the fission product mass distribution. Rubidium has an atomic number of 37 and the naturally occurring isotopes have mass numbers of 85 and 87. Figure 9 and 10 display results of the calculated thin target production of rubidium isotopes from \(^{235}\text{U}\) and \(^{238}\text{U}\) fission, respectively. The \(^{235}\text{U}\) calculated yields indicate that the isotope distribution for rubidium peaks at 94 for thermal and fast neutrons and at 94 for 14 MeV neutrons. The 100 MeV distributions for all particles peak around 91-92 amu. The \(^{238}\text{U}\) yields for fast and 14 MeV neutrons peak around 94-95 and 100 MeV distributions at 92. The 100 MeV distributions present a lower peak (or increase) at the 94-95 mass range. Again, for the most neutron rich isotopes, 14 MeV neutrons on \(^{238}\text{U}\) have the highest production rate.

### Table 1. Yields of Kr, Rb, Sn, Xe, and Cs per beam particle from \(^{235}\text{U}\) bombarded by protons, deuterons, and neutrons.

<table>
<thead>
<tr>
<th>Particle Type</th>
<th>Energy (MeV)</th>
<th>Calculated Yield per Element per gram/cm(^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Kr</td>
<td>Rb</td>
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<tr>
<td>Proton</td>
<td>20</td>
<td>3.91E-05</td>
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<tr>
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<td>Proton</td>
<td>150</td>
<td>2.30E-04</td>
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<td>Deuteron</td>
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</tr>
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<td>50</td>
<td>2.93E-04</td>
</tr>
<tr>
<td>Deuteron</td>
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<td>3.53E-04</td>
</tr>
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<td>Deuteron</td>
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<td>3.32E-04</td>
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<tr>
<td>Neutron</td>
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<td>6.49E-03</td>
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<tr>
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<td>1.e-04 to 5</td>
<td>6.13E-04</td>
</tr>
<tr>
<td>Neutron</td>
<td>14.</td>
<td>5.21E-04</td>
</tr>
<tr>
<td>Neutron</td>
<td>20.</td>
<td>3.12E-04</td>
</tr>
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<td>Neutron</td>
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### Table 2. Yields of Kr, Rb, Sn, Xe, and Cs per beam particle from \(^{238}\text{U}\) bombarded by protons, deuterons, and neutrons.

<table>
<thead>
<tr>
<th>Particle Type</th>
<th>Energy (MeV)</th>
<th>Calculated Yield per Element per gram/cm(^2)</th>
</tr>
</thead>
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<td>Neutron</td>
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<td>2.20E-04</td>
</tr>
<tr>
<td>Neutron</td>
<td>150.</td>
<td>2.07E-04</td>
</tr>
</tbody>
</table>
**Krypton from U-235 Fission**

Neutron/Proton/Deuteron

- $^6\text{U}-^235\text{p}.100\text{MeV} - Y=2.29\times 10^{-04}$
- $^6\text{U}-^235\text{p}.100\text{MeV} - Y=3.66\times 10^{-04}$
- $^6\text{U}-^235\text{n}.100\text{MeV} - Y=2.47\times 10^{-04}$
- $^6\text{U}-^235\text{thermal} (0.1) - Y=5.50\times 10^{-03}$
- $^6\text{U}-^235\text{n}.\text{fast} - Y=8.13\times 10^{-04}$
- $^6\text{U}-^235\text{n}.14\text{MeV} - Y=5.22\times 10^{-04}$

**Krypton from U-238 Fission**

Neutron/Proton/Deuteron

- $^6\text{U}-^238\text{p}.20\text{MeV} - Y=3.46\times 10^{-05}$
- $^6\text{U}-^238\text{p}.100\text{MeV} - Y=2.13\times 10^{-04}$
- $^6\text{U}-^238\text{d}.100\text{MeV} - Y=3.02\times 10^{-04}$
- $^6\text{U}-^238\text{n}.100\text{MeV} - Y=2.20\times 10^{-04}$
- $^6\text{U}-^238\text{n}.\text{fast} (10) - Y=1.15\times 10^{-04}$
- $^6\text{U}-^238\text{n}.14\text{MeV} - Y=2.55\times 10^{-04}$

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**C. Tin Isotopes from Fission**

Tin neutron-rich isotopes are in the high-mass peak region of the fission product mass distribution. Tin has an atomic number of 50 and the naturally occurring isotopes have mass numbers between 112 and 124. Figures 11 and 12 show the isotopic distribution for fission of $^{235}\text{U}$ and $^{238}\text{U}$, respectively. Figures 11 and 12 display the isotopic distribution of tin as a fission product. The calculated mass distribution for thermal and fast $^{235}\text{U}$-fission peaks around 131-132 amu. For $^{238}\text{U}$ fission, the fast and 14 MeV neutron distributions peak at 132 and 131, respectively. The $^{235}\text{U}$ fission for thermal neutrons yields more or about the same neutron rich isotopes than any other fission type for mass numbers up to 136 amu. In this particular case the differences between the below 20 MeV neutron energy and the high energy (above 50 MeV) fission is more pronounced. Again, for neutron rich isotopes such as $^{132}\text{Sn}$ and heavier, thermal neutrons on $^{235}\text{U}$ and 14 MeV on $^{238}\text{U}$ are both very productive.

**D. Xenon Isotopes from Fission.**

Xenon is, as krypton, a noble gas. Xenon has an atomic number 54 and naturally occurring isotopes in the mass number range of 124 to 136 amu. Figures 13 and 14 display the isotopic distribution of xenon as a fission product. The calculated $^{235}\text{U}$ fission product mass distribution for this element indicate that the peak of the isotope distribution is at 139 amu’s for thermal, fast, and 14 MeV neutrons bombarding $^{235}\text{U}$ target. The main peaks for 100 MeV distributions are at 136 for the $^{235}\text{U}$ target. For the $^{238}\text{U}$ target the fast and 14 MeV distributions peak around 140-141 amu. The 100 MeV distributions for $^{238}\text{U}$ target have the peak at 136 amu. Thermal neutrons on $^{235}\text{U}$ produce and 14 MeV neutrons on $^{238}\text{U}$ produce the most neutron-rich isotopes.
E. Cesium Isotopes from Fission.

Cesium has an atomic number of 55 and the naturally occurring isotope has a mass number of 133. Figures 15 and 16 are plots of the isotopic distribution for cesium. The cesium isotopes from fission have mass number in the range of the high-mass peak. The $^{235}$U fission with thermal and fast neutrons produce a mass distribution of cesium isotopes that peaks at 142 amu’s. The 14 MeV neutrons generate a peak at 141 and the 100 MeV incident particles produce double peaks at 137 and 139 amu’s. The thermal neutrons on $^{235}$U and 14 MeV neutron on $^{238}$U are the best for production of neutron rich isotopes.

VII. CONCLUSIONS

For $^{235}$U targets and the five elements considered here, the best yields of neutron-rich isotopes are obtained from neutrons in the 2-20 MeV range. High energy beams of neutrons, protons, and deuterons have comparable integral yields per element to neutrons below 20 MeV, but the distributions are peaked at lower neutron numbers. This is presumably due to a higher neutron multiplicity in the pre-equilibrium stage and/or the compound nucleus/fission stage.
For $^{235}$U targets there are high yields predicted especially for thermal neutrons, and also for the fast neutron spectrum. For the high energy neutrons, protons, and deuterons $^{235}$U has no advantage over $^{238}$U. A detailed comparison of the relative advantages of $^{235}$U and $^{238}$U for radioactive beam applications is beyond the scope of this study and will be addressed in the future.

The present work is the first step of a more detailed analysis of various possible one- and two-step target geometry calculated with the LAHET code system. It is intended to serve as a guide in choosing geometry and beams for future studies. It is desirable to extend this study to higher beam energies, e.g. 200 to 1000 MeV, but at this time there is very little data against which to benchmark the analysis. Additional data would also permit comparisons of isotope yields beyond the tails of the distributions presented here, to even more neutron rich isotopes.

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REFERENCES

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Figure 15. $^{235}$U fission isotopic distribution for Cs.
To illustrate relative yields in the tails of the distributions, this graph is plotted on a semi-log scale.

Figure 16. $^{238}$U fission isotopic distribution for Cs.
To illustrate relative yields in the tails of the distributions, this graph is plotted on a semi-log scale.