

## SHORT COMMUNICATION

K. Takamiya, T. Imanaka, S. D. Egbert, W. Rühm

### **Amendments to $^{63}\text{Ni}$ production calculation for Hiroshima by Takamiya and coworkers and DS02 fluence data by Egbert and coworkers**

K. Takamiya (✉), T. Imanaka

Research Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka  
590-0494, Japan

e-mail [takamiya@rri.kyoto-u.ac.jp](mailto:takamiya@rri.kyoto-u.ac.jp)

Tel.: +81-724-51-2460

Fax: +81-724-51-2632

S. D. Egbert

Science Applications International Corporation, San Diego, California 92121, USA

W. Rühm

Helmholtz Zentrum München, German Research Center for Environmental Health,  
85764 Neuherberg, Germany

Recently,  $^{63}\text{Ni}$  has been measured in copper samples from Hiroshima in an effort to reconstruct the fast neutron fluence from the atomic bomb and to validate calculated neutron doses to the atomic bomb survivors (Straume et al. 2003, Straume et al. 2004, Straume et al. 2005, Rühm et al. 2007, Marchetti et al. 2009). The paper by Takamiya et al. (2008a) published in *Radiation and Environmental Biophysics* presented a calculation of  $^{63}\text{Ni}$  production by the Hiroshima atomic bomb using their experimental cross section values of the  $^{63}\text{Cu}(n,p)^{63}\text{Ni}$  reaction as well as the same neutron fluence as used in the radiation dosimetry system DS02 (Young and Kerr 2005) and given in Egbert et al. (2007). Takamiya and coworkers compared their results with the corresponding calculation given in the DS02 report that used another set of the cross section values from the ENDF/B-VI nuclear data file (1990). A tendency was observed in Takamiya et al. that their results for  $^{63}\text{Ni}$  production were larger by about 40% – 60% than those in DS02 near the hypocenter, while the differences decreased to about 0% – 20% beyond 1,000 m from the hypocenter. After the publication of Takamiya et al. (2008a), a series of discussions were held between Takamiya and Egbert. As a result of these discussions, the following two issues are described here as amendments to the previous papers (Egbert et al. 2007, Takamiya et al. 2008a).

One issue was typographical errors that appeared in several energy boundary values in the DS02 report (Table 7, Chapter 12) which were copied into Egbert et al. (2007) and used by Takamiya et al. (2008a). The recent DS02 errata (RERF 2009) correct errors in Egbert's Table 1, replaced in Table 1 of the present work in its entirety. These typographical errors had resulted in an incorrect neutron spectrum near 2 MeV and a slight underestimation of the  $^{63}\text{Ni}$  production in Takamiya et al. (2008a).

Another issue is an inappropriate assumption on the low-energy cross section values used by Takamiya et al. (2008a). Because of a lack of experimental data in the low neutron energies, the cross section values below 1 MeV were simply obtained by proportional extrapolation from the experimental cross sections at the lowest energy ( $E_n = 1.0$  MeV) to  $\sigma = 0$  barn at  $E_n = 0$  eV, but these were much larger than found in ENDF/B-VI. This evaluation caused an overestimation of the number of  $^{63}\text{Ni}$  nuclei produced, especially near the hypocenter. In order to estimate an approximate contribution of thermal neutrons to the  $^{63}\text{Cu}(n,p)^{63}\text{Ni}$  reaction, the approach mentioned in Rühm et al. (2007) was applied. In the revised calculation, the cross section values at the lower energy range were calculated following a procedure described by Nolte et al. (2006). The calculation was normalized to an experimental cross section value of 50  $\mu\text{b}$  (upper-bound estimate) at 25 meV for the  $^{63}\text{Cu}(n,p)^{63}\text{Ni}$  reaction, as mentioned in (Rugel 2002). And at energies between 0.1 and 1 MeV cross section values were

estimated by fitting of JENDL-3.3 with an additional extrapolation down to about 0.03 MeV. The cross sections of the reaction used in the revised calculation is shown in Fig. 1, and the cross section values adopted in the revised calculation are listed here in Table 2, which replaces Table 1 of Takamiya et al. (2008a) in its entirety. The experimental cross section values from which the upper and lower limit cross section values shown in Table 2 are estimated are listed in Table 3. The number of  $^{63}\text{Ni}$  nuclei was revised using the revised cross section values, and the resulting new values on  $^{63}\text{Ni}$  production are shown in Table 4, which replaces Table 2 of the previous paper in its entirety. The revised values are slightly lower at the hypocenter and are marginally higher beyond 1,000 m compared with the previous results, namely the number of  $^{63}\text{Ni}$  nuclei estimated on the basis of experimental cross section values becomes somewhat closer to that estimated in DS02. As can be seen in Table 4, however, there is still a substantial difference between the revised values and those of DS02 at the hypocenter, which reflects the differences between the revised Takamiya and ENDF/B-VI cross section values. Low energy neutrons, below about 1 MeV, play a non-negligible role in the production of  $^{63}\text{Ni}$ , especially close to the hypocenter: Table 5 indicates that the fraction of  $^{63}\text{Ni}$  nuclei produced by neutrons below 1 MeV is larger at small distances than at great distances from the hypocenter.

In order to reduce the uncertainty from this last issue, an experiment to measure the  $^{63}\text{Cu}(n,p)^{63}\text{Ni}$  cross section below about 1 MeV will be performed using a research reactor in the near future.

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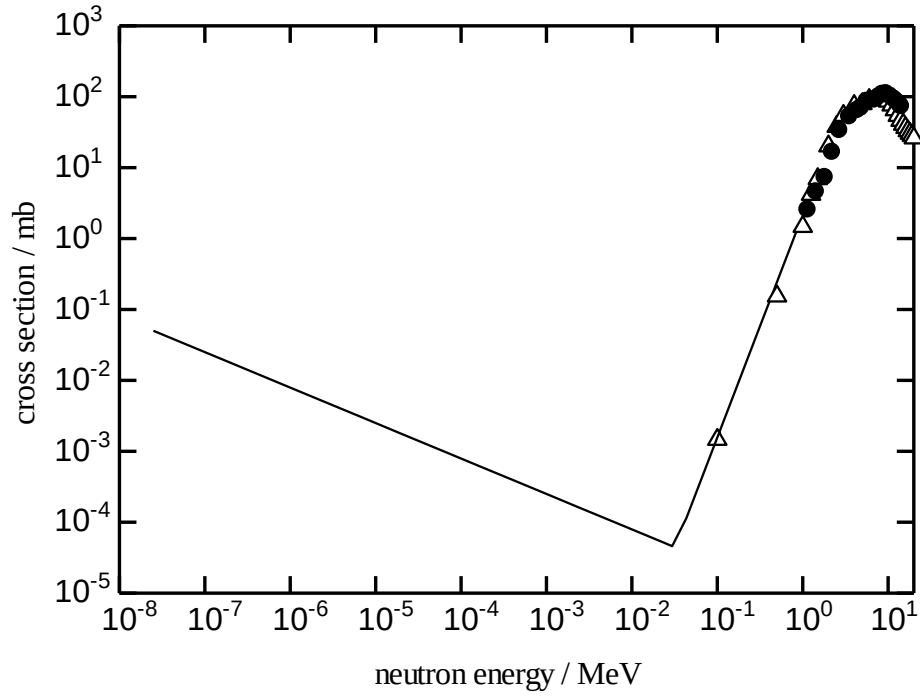
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**Fig. 1** Assumed excitation function of the reaction of  $^{63}\text{Cu}(n, p)^{63}\text{Ni}$  (solid line) that was used in the recalculation of the number of  $^{63}\text{Ni}$  nuclei produced in 1 g of copper. Circles and triangles show experimental cross section values (Takamiya et al. 2008b) and evaluated values from JENDL-3.3 (JENDL 2002), respectively.

**Table 1.** Energy group boundaries and fluence-to-kerma coefficients for 2002 calculations.

Corrected Egbert et al. table (Egbert et al. 2007) based on Errata (RERF 2009)

| Group # | Upper Energy<br>Boundary<br>(MeV) | Soft Tissue<br>Kerma<br>coefficient<br>(Gy cm <sup>2</sup> ) | Group #   | Upper Energy<br>Boundary<br>(MeV) | Soft Tissue<br>Kerma<br>coefficient<br>(Gy cm <sup>2</sup> ) |
|---------|-----------------------------------|--------------------------------------------------------------|-----------|-----------------------------------|--------------------------------------------------------------|
| neutron |                                   |                                                              | gamma ray |                                   |                                                              |
| 1       | 1.96×10 <sup>+1</sup>             | 6.90×10 <sup>-11</sup>                                       | 47        | 2.00×10 <sup>+1</sup>             | 3.78×10 <sup>-11</sup>                                       |
| 2       | 1.69×10 <sup>+1</sup>             | 6.73×10 <sup>-11</sup>                                       | 48        | 1.40×10 <sup>+1</sup>             | 3.03×10 <sup>-11</sup>                                       |
| * 3     | 1.49×10 <sup>+1</sup>             | 6.61×10 <sup>-11</sup>                                       | 49        | 1.20×10 <sup>+1</sup>             | 2.66×10 <sup>-11</sup>                                       |
| * 4     | 1.42×10 <sup>+1</sup>             | 6.57×10 <sup>-11</sup>                                       | 50        | 1.00×10 <sup>+1</sup>             | 2.28×10 <sup>-11</sup>                                       |
| 5       | 1.38×10 <sup>+1</sup>             | 6.47×10 <sup>-11</sup>                                       | 51        | 8.00×10 <sup>+0</sup>             | 1.98×10 <sup>-11</sup>                                       |
| 6       | 1.25×10 <sup>+1</sup>             | 6.38×10 <sup>-11</sup>                                       | 52        | 7.00×10 <sup>+0</sup>             | 1.78×10 <sup>-11</sup>                                       |
| 7       | 1.22×10 <sup>+1</sup>             | 6.33×10 <sup>-11</sup>                                       | 53        | 6.00×10 <sup>+0</sup>             | 1.61×10 <sup>-11</sup>                                       |
| 8       | 1.11×10 <sup>+1</sup>             | 6.04×10 <sup>-11</sup>                                       | 54        | 5.00×10 <sup>+0</sup>             | 1.43×10 <sup>-11</sup>                                       |
| 9       | 1.00×10 <sup>+1</sup>             | 5.83×10 <sup>-11</sup>                                       | 55        | 4.00×10 <sup>+0</sup>             | 1.21×10 <sup>-11</sup>                                       |
| 10      | 9.05×10 <sup>+0</sup>             | 5.54×10 <sup>-11</sup>                                       | 56        | 3.00×10 <sup>+0</sup>             | 1.03×10 <sup>-11</sup>                                       |
| 11      | 8.19×10 <sup>+0</sup>             | 5.45×10 <sup>-11</sup>                                       | 57        | 2.50×10 <sup>+0</sup>             | 8.96×10 <sup>-12</sup>                                       |
| 12      | 7.41×10 <sup>+0</sup>             | 5.12×10 <sup>-11</sup>                                       | 58        | 2.00×10 <sup>+0</sup>             | 7.53×10 <sup>-12</sup>                                       |
| 13      | 6.38×10 <sup>+0</sup>             | 4.71×10 <sup>-11</sup>                                       | 59        | 1.50×10 <sup>+0</sup>             | 5.84×10 <sup>-12</sup>                                       |
| 14      | 4.97×10 <sup>+0</sup>             | 4.58×10 <sup>-11</sup>                                       | 60        | 1.00×10 <sup>+0</sup>             | 4.25×10 <sup>-12</sup>                                       |
| 15      | 4.72×10 <sup>+0</sup>             | 4.43×10 <sup>-11</sup>                                       | 61        | 7.00×10 <sup>-1</sup>             | 2.86×10 <sup>-12</sup>                                       |
| 16      | 4.07×10 <sup>+0</sup>             | 4.18×10 <sup>-11</sup>                                       | 62        | 4.50×10 <sup>-1</sup>             | 1.92×10 <sup>-12</sup>                                       |
| 17      | 3.01×10 <sup>+0</sup>             | 3.55×10 <sup>-11</sup>                                       | 63        | 3.00×10 <sup>-1</sup>             | 1.03×10 <sup>-12</sup>                                       |
| 18      | 2.39×10 <sup>+0</sup>             | 3.33×10 <sup>-11</sup>                                       | 64        | 1.50×10 <sup>-1</sup>             | 5.30×10 <sup>-13</sup>                                       |
| * 19    | 2.31×10 <sup>+0</sup>             | 3.20×10 <sup>-11</sup>                                       | 65        | 1.00×10 <sup>-1</sup>             | 3.49×10 <sup>-13</sup>                                       |
| * 20    | 1.83×10 <sup>+0</sup>             | 2.89×10 <sup>-11</sup>                                       | 66        | 7.00×10 <sup>-2</sup>             | 3.16×10 <sup>-13</sup>                                       |
| * 21    | 1.42×10 <sup>+0</sup>             | 2.61×10 <sup>-11</sup>                                       | 67        | 4.50×10 <sup>-2</sup>             | 4.64×10 <sup>-13</sup>                                       |
| * 22    | 1.11×10 <sup>+0</sup>             | 2.50×10 <sup>-11</sup>                                       | 68        | 3.00×10 <sup>-2</sup>             | 1.07×10 <sup>-12</sup>                                       |
| 23      | 9.62×10 <sup>-1</sup>             | 2.22×10 <sup>-11</sup>                                       | 69        | 2.00×10 <sup>-2</sup>             | 3.51×10 <sup>-12</sup>                                       |
| 24      | 8.21×10 <sup>-1</sup>             | 2.06×10 <sup>-11</sup>                                       |           | 1.00×10 <sup>-2</sup>             |                                                              |
| 25      | 7.43×10 <sup>-1</sup>             | 1.93×10 <sup>-11</sup>                                       |           |                                   |                                                              |
| 26      | 6.39×10 <sup>-1</sup>             | 1.79×10 <sup>-11</sup>                                       |           |                                   |                                                              |
| 27      | 5.50×10 <sup>-1</sup>             | 1.63×10 <sup>-11</sup>                                       |           |                                   |                                                              |
| 28      | 3.69×10 <sup>-1</sup>             | 1.29×10 <sup>-11</sup>                                       |           |                                   |                                                              |
| 29      | 2.47×10 <sup>-1</sup>             | 1.02×10 <sup>-11</sup>                                       |           |                                   |                                                              |
| 30      | 1.58×10 <sup>-1</sup>             | 8.06×10 <sup>-12</sup>                                       |           |                                   |                                                              |
| 31      | 1.11×10 <sup>-1</sup>             | 5.64×10 <sup>-12</sup>                                       |           |                                   |                                                              |
| 32      | 5.25×10 <sup>-2</sup>             | 3.57×10 <sup>-12</sup>                                       |           |                                   |                                                              |
| 33      | 3.43×10 <sup>-2</sup>             | 2.61×10 <sup>-12</sup>                                       |           |                                   |                                                              |
| 34      | 2.48×10 <sup>-2</sup>             | 2.14×10 <sup>-12</sup>                                       |           |                                   |                                                              |
| 35      | 2.19×10 <sup>-2</sup>             | 1.49×10 <sup>-12</sup>                                       |           |                                   |                                                              |
| 36      | 1.03×10 <sup>-2</sup>             | 6.39×10 <sup>-13</sup>                                       |           |                                   |                                                              |
| 37      | 3.35×10 <sup>-3</sup>             | 2.21×10 <sup>-13</sup>                                       |           |                                   |                                                              |
| 38      | 1.23×10 <sup>-3</sup>             | 9.25×10 <sup>-14</sup>                                       |           |                                   |                                                              |
| 39      | 5.83×10 <sup>-4</sup>             | 4.49×10 <sup>-14</sup>                                       |           |                                   |                                                              |
| 40      | 2.75×10 <sup>-4</sup>             | 2.09×10 <sup>-14</sup>                                       |           |                                   |                                                              |
| 41      | 1.01×10 <sup>-4</sup>             | 1.04×10 <sup>-14</sup>                                       |           |                                   |                                                              |
| 42      | 2.90×10 <sup>-5</sup>             | 9.37×10 <sup>-15</sup>                                       |           |                                   |                                                              |
| * 43    | 1.07×10 <sup>-5</sup>             | 1.36×10 <sup>-14</sup>                                       |           |                                   |                                                              |
| 44      | 3.06×10 <sup>-6</sup>             | 2.28×10 <sup>-14</sup>                                       |           |                                   |                                                              |
| 45      | 1.13×10 <sup>-6</sup>             | 3.74×10 <sup>-14</sup>                                       |           |                                   |                                                              |
| 46      | 4.14×10 <sup>-7</sup>             | 1.50×10 <sup>-13</sup>                                       |           |                                   |                                                              |
|         | 1.00×10 <sup>-11</sup>            |                                                              |           |                                   |                                                              |

The upper energy boundaries of the energy groups marked by asterisk on the left side are revised.

**Table 2** Revised upper and lower experimental limits of the  $^{63}\text{Cu}(n,p)^{63}\text{Ni}$  cross section at the median energy of each 46 energy group.

| $E_n$ (MeV) | $\sigma$ (mb)  |                | $E_n$ (MeV) | $\sigma$ (mb)  |                |
|-------------|----------------|----------------|-------------|----------------|----------------|
|             | lower<br>limit | upper<br>limit |             | lower<br>limit | upper<br>limit |
| 2.07E-07    | *1.7E-02       | *1.7E-02       | 7.82E-01    | *1.0E+00       | *1.0E+00       |
| 7.72E-07    | *9.0E-03       | *9.0E-03       | 8.92E-01    | *1.5E+00       | *1.5E+00       |
| 2.10E-06    | *5.5E-03       | *5.5E-03       | *1.04E+00   | *1.8E+00       | *1.8E+00       |
| 6.88E-06    | *3.0E-03       | *3.0E-03       | *1.27E+00   | *3.4E+00       | *3.8E+00       |
| 1.99E-05    | *1.8E-03       | *1.8E-03       | *1.63E+00   | *5.9E+00       | *8.7E+00       |
| 6.50E-05    | *9.8E-04       | *9.8E-04       | *2.07E+00   | *1.2E+01       | *2.4E+01       |
| 1.88E-04    | *5.8E-04       | *5.8E-04       | *2.35E+00   | *2.2E+01       | *3.5E+01       |
| 4.29E-04    | *3.8E-04       | *3.8E-04       | 2.70E+00    | 3.4E+01        | 4.2E+01        |
| 9.07E-04    | *2.6E-04       | *2.6E-04       | 3.54E+00    | 5.2E+01        | 6.8E+01        |
| 2.29E-03    | *1.7E-04       | *1.7E-04       | 4.40E+00    | 6.4E+01        | 7.5E+01        |
| 6.83E-03    | *9.6E-05       | *9.6E-05       | 4.85E+00    | 6.9E+01        | 7.9E+01        |
| 1.61E-02    | *6.2E-05       | *6.2E-05       | 5.68E+00    | 8.6E+01        | 8.9E+01        |
| 2.34E-02    | *5.2E-05       | *5.2E-05       | 6.90E+00    | 9.0E+01        | 9.1E+01        |
| 2.96E-02    | *4.6E-05       | *4.6E-05       | 7.80E+00    | 9.9E+01        | 9.9E+01        |
| 4.34E-02    | *1.1E-04       | *1.1E-04       | 8.62E+00    | 1.1E+02        | 1.1E+02        |
| 8.18E-02    | *8.3E-04       | *8.3E-04       | 9.53E+00    | 1.1E+02        | 1.1E+02        |
| 1.35E-01    | *4.0E-03       | *4.0E-03       | 1.06E+01    | 9.9E+01        | 1.0E+02        |
| 2.03E-01    | *1.4E-02       | *1.4E-02       | 1.17E+01    | 8.9E+01        | 9.3E+01        |
| 3.08E-01    | *5.3E-02       | *5.3E-02       | 1.24E+01    | 8.2E+01        | 8.8E+01        |
| 4.60E-01    | *1.9E-01       | *1.9E-01       | 1.32E+01    | 7.4E+01        | 8.2E+01        |
| 5.95E-01    | *4.2E-01       | *4.2E-01       | *1.40E+01   | 6.3E+01        | 7.5E+01        |
| 6.91E-01    | *6.8E-01       | *6.8E-01       | *1.46E+01   | 6.0E+01        | 7.0E+01        |

The energy values and corresponding cross section values marked by asterisk on the left side are revised values.

**Table 3** The experimental  $^{63}\text{Cu}(\text{n,p})^{63}\text{Ni}$  cross section values obtained by Takamiya et al. (2008b).

| Energy range (MeV) |       | $\sigma$ (mb) | Error (mb) | Energy range (MeV) |       | $\sigma$ (mb) | Error (mb) |
|--------------------|-------|---------------|------------|--------------------|-------|---------------|------------|
| Lower              | Upper |               |            | Lower              | Upper |               |            |
| 1.52               | 1.58  | 5.3           | 0.3        | 3.10               | 3.34  | 59.2          | 1.5        |
| 1.60               | 2.00  | 7.8           | 0.4        | 3.28               | 3.60  | 60.5          | 0.8        |
| 1.67               | 1.87  | 9.7           | 0.3        | 3.30               | 4.00  | 54.6          | 1.1        |
| 1.75               | 1.95  | 7.4           | 0.3        | 3.35               | 3.85  | 60.4          | 1.3        |
| 1.77               | 1.99  | 7.8           | 0.2        | 3.40               | 5.80  | 75.2          | 1.2        |
| 1.80               | 1.96  | 13.9          | 0.3        | 3.61               | 3.95  | 74.0          | 0.7        |
| 1.86               | 2.06  | 19.3          | 0.6        | 3.74               | 3.98  | 70.3          | 1.4        |
| 1.96               | 2.19  | 19.3          | 0.4        | 3.85               | 4.30  | 74.8          | 1.6        |
| 1.98               | 2.30  | 13.2          | 0.3        | 3.95               | 4.31  | 70.5          | 0.6        |
| 2.17               | 2.37  | 31.4          | 0.9        | 4.00               | 4.76  | 64.0          | 1.3        |
| 2.19               | 2.52  | 28.0          | 0.6        | 4.31               | 4.69  | 71.0          | 0.9        |
| 2.30               | 2.74  | 29.1          | 0.6        | 4.78               | 5.04  | 73.2          | 1.4        |
| 2.35               | 2.57  | 39.2          | 0.9        | 5.05               | 5.41  | 73.3          | 2.0        |
| 2.52               | 2.90  | 35.1          | 0.7        | 5.11               | 5.39  | 85.9          | 1.7        |
| 2.73               | 3.31  | 41.6          | 0.9        | 5.40               | 5.74  | 86.0          | 2.3        |
| 2.73               | 2.99  | 43.9          | 0.5        | 5.43               | 5.71  | 88.8          | 1.7        |
| 2.83               | 3.05  | 42.5          | 1.1        | 12.99              | 13.87 | 71.9          | 1.6        |
| 2.90               | 3.35  | 51.4          | 1.1        | 13.66              | 14.48 | 61.5          | 1.2        |
| 2.99               | 3.29  | 53.0          | 0.6        | 14.20              | 15.50 | 58.6          | 1.3        |

**Table 4** Recalculated number of  $^{63}\text{Ni}$  nuclei produced in 1 g of copper free-in-air at 1 m above ground using the experimental lower- and upper-limit cross section values shown in Table 2. The two columns on the right show the ratios between the numbers of  $^{63}\text{Ni}$  nuclei calculated based on the DS02 approach and the present recalculation.

| Ground<br>range (m) | $^{63}\text{Ni}$ (nuclei per g Cu) |                |                | Ratio, present/<br>DS02 |                |
|---------------------|------------------------------------|----------------|----------------|-------------------------|----------------|
|                     | DS02                               | Lower<br>limit | Upper<br>limit | Lower<br>limit          | Upper<br>limit |
| 0                   | 1.14E+07                           | 1.38E+07       | 1.74E+07       | 1.2                     | 1.5            |
| 500                 | 3.58E+06                           | 3.98E+06       | 5.06E+06       | 1.1                     | 1.4            |
| 1,000               | 2.82E+05                           | 2.95E+05       | 3.75E+05       | 1.0                     | 1.3            |
| 1,500               | 1.61E+04                           | 1.67E+04       | 2.09E+04       | 1.0                     | 1.3            |
| 2,000               | 9.41E+02                           | 9.77E+02       | 1.21E+03       | 1.0                     | 1.3            |
| 2,500               | 5.77E+01                           | 6.01E+01       | 7.36E+01       | 1.0                     | 1.3            |

**Table 5** The production ratio of  $^{63}\text{Ni}$  at the energy ranges of 1) below 0.03 MeV, 2) 0.03 – 1 MeV, and 3) 1 - 15 MeV, as a function of the ground range. The ratios are calculated by the upper limit cross sections.

| Ground<br>range (m) | Production ratio of $^{63}\text{Ni}$ (%) |              |            |
|---------------------|------------------------------------------|--------------|------------|
|                     | < 0.03 MeV                               | 0.03 - 1 MeV | 1 – 15 MeV |
| 0                   | 7                                        | 12           | 81         |
| 500                 | 3                                        | 9            | 88         |
| 1000                | 1                                        | 5            | 94         |
| 1500                | 0                                        | 3            | 97         |
| 2000                | 0                                        | 2            | 98         |
| 2500                | 0                                        | 2            | 98         |