# Fission and spallation induced by 7-GeV protons on U, Bi, Pb, Au, W, Ho, and Ag

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Thin targets of U, Bi, Pb, Au, W, Ho, and Ag were bombarded with 7-GeV protons in order to measure the fission and spallation cross sections. Muscovite mica, Makrofol-N, Makrofol-E, and Daicel track detectors were employed in  $4\pi$  and  $2\pi$  geometry arrangements. The binary- and ternary-fission cross sections ( $\sigma_b$  and  $\sigma_t$ ) and the value of fissility ( $\sigma_{\rm fission}/\sigma_{\rm inelastic}$ ) increase with increasing value of  $Z^2/A$  of the target. The spallation cross section ( $\sigma_{\rm spall}$ ) shows an increase with increasing mass number of the target. However,  $\sigma_t/\sigma_b$  and F/B (forward to backward) ratios decrease with increasing value of  $Z^2/A$ .

## I. INTRODUCTION

High energy proton interactions produce a spectrum of reaction products suggesting the occurrence of a number of competing processes. Attempts have been made to identify these processes.<sup>1,2</sup> It was of particular interest to separate the fission events from the spallation products. The radiochemical methods are inherently limited for this purpose.<sup>3-6</sup> Emulsions, on the other hand, create background problems due to the presence of Ag and Br. The use of mica,<sup>7-9</sup> glass,<sup>10</sup> and Makrofol<sup>11-15</sup> solid state nuclear track detectors (SSNTD's) produced some useful results in this direction. These detectors therefore have been employed<sup>7-15</sup> to study the interaction of protons having energies of (a) 0.6–3.0 GeV, (b) 13, 18, 23, and 29 GeV, and (c) 200 and 300 GeV with thin targets of U, Bi, Pb, Au, Ag, etc. Little work has been done (a) in the proton energy range of 3–10 GeV and (b) with targets other than the ones listed above.

The present paper describes the use of four different types of solid state nuclear track detectors for studying the interaction of 7 GeV protons with thin targets of U, Bi, Pb, Au, W, Ho, and Ag. Both  $4\pi$  and  $2\pi$  geometry arrangements were employed. These studies were carried out with a view to obtain (a) the cross sections for binary, ternary, and (possibly) higher fission modes, (b) spallation cross sections, and (c) information regarding the forward momentum transfer to the target nucleus. Variation of the above-mentioned reaction parameters as a function of  $Z^2/A$  of the target has also been studied. Particular attention has been paid to the study of track length distributions of the reaction products emitted in ternary fissions. The results have been compared with some of those obtained previously by other workers.

# **II. EXPERIMENTAL DETAILS**

Newly cleaved Muscovite mica sheets (thickness ~150  $\mu$ m, Z threshold ~16), and fresh pieces of Makrofol-N (thickness ~100  $\mu$ m, Z threshold ~8), Makrofol-E (thickness ~200  $\mu$ m, Z threshold ~2), and Daicel (thickness ~100  $\mu$ m, Z threshold=protons) were carefully cleaned and annealed so as to remove the background

tracks, if any. These detectors were then coated with thin targets of U, Bi, Pb, Au, W, Ho, and Ag under vacuum. The thickness of the target layers varied from 10 to 200  $\mu$ g/cm<sup>2</sup>. The targets so prepared were sandwiched by using blank sheets (having equal dimensions) of the same detectors and by gluing them on one side. Each sandwich was then contained in a thin envelope of Mylar, which was evacuated in order to compress the sandwich through atmospheric pressure. Stacks containing the same target material but having different combinations of track detectors were prepared with a view to attempt classification of the reaction products from one particular type of target from registration thresholds characteristic of different track detectors.

Ten stacks containing different target detector combinations were exposed to 7 GeV protons obtained from the NIMROD proton accelerator at Rutherford Laboratory (U.K.). Exposures were carried out at 90° with respect to the detector surface. During the exposure, care was taken to use only the central area of the beam. Thin aluminum foils were incorporated with every stack for monitoring of the proton flux by using the well-known <sup>27</sup>Al(p,3pn)<sup>24</sup>Na reaction.<sup>16,17</sup> Proton fluences of the order of 10<sup>11</sup> protons/cm<sup>2</sup> (with an accuracy of ±10%) were obtained.

After the exposures, the targets were removed by dissolving them in HCl or HNO<sub>3</sub> or aqua regia. The mica detectors were etched for 20 min in 48% HF kept at 22 °C. The etching of Makrofol-N, Makrofol-E, and Daicel was carried out in a 6N aqueous solution of NaOH (kept at 50±1°C) for various time intervals.

The etched detector foils (from the sandwich arrangements) were correctly assembled along with a graduated mesh system for ease of analysis and cross checking. Scanning of the reaction products was carried out at a magnification of  $500 \times$  while the actual analysis of the individual events/tracks was performed at a total magnification of  $1250 \times$  using a Zeiss (Universal) optical microscope. The optical arrangement was such that both the inner surfaces of the respective detectors of the sandwiches were in focus so that the correlated and uncorrelated tracks (most probably due to residual spallation products of target nuclei or possibly from heavy constituents of mica) could be easily separated and analyzed. The

projected lengths of the correlated tracks and their depths were analyzed along with the gaps between them. The events were traced out using Zeiss tracing system. Calculations were made to determine (a) the physical ranges of the individual tracks and (b) the point of origin. Only those tracks with physical lengths greater than 2  $\mu$ m were taken as genuine tracks.

## **III. RESULTS AND DISCUSSION**

Figure 1 is a set of photomicrographs of typical fission events. The figure shows (a) a binary and (b) an indirect ternary (or a binary event in which a few protons and alphas are emitted in the hemisphere not containing the fission fragments so that the momentum of the system is balanced) fission event in Ho registered in a mica track detector. Parts (c) and (d) show a binary and ternary fission event, respectively, in a Bi target obtained by using a Daicel plastic track detector. Studies of interaction with a uranium target, besides yielding a large number of binary and ternary events, showed some events which could qualify for quaternary fission events. Parts (e) and (f) are typical examples of such quaternary fission events in uranium as observed by using Muscovite mica [part (e)] and Daicel [part (f)] track detectors. Dots (as seen in the above fig-



FIG. 1. Set of photomicrographs showing (a) a binary fission event and (b) an "indirect" ternary fission event (or a binary event in which some energetic light particles were emitted in the hemisphere opposite to that containing the fission fragments, in order to balance the momentum of the system) produced in <sup>165</sup>Ho and registered in the mica detector. (c) and (d) show twoand three-prong events produced in bismuth and observed by using the Daicel track detector. (e) and (f) are photomicrographs of quaternary fission events produced in uranium and registered in mica and Daicel, respectively.

ures) most probably represent the background. These were therefore neglected in our analysis. Due to better sensitivity of the Daicel detector, many high multiplicity events could be observed [part (f)]. In a rather rare case even mica showed a quaternary (four heavy reaction products) event.

Binary and ternary fission cross sections for targets of U, Bi, Pb, Au, W, Ho, and Ag were obtained by using four different types of track detectors. Figures 2 and 3 show the results for Muscovite mica and Daicel detectors. Both cross sections show an increase with increasing value of  $Z^2/A$  of the target. Generally, our results are close to the ones obtained by (a) Debeauvais et al.<sup>15</sup> by using Makrofol-E for 200 and 300 GeV protons, (b) Katcoff and Hudis<sup>9</sup> by using a mica detector for 0.6-300 GeV protons, and (c) others using mica and other methods.<sup>18-23</sup> However, our cross-section values obtained by using the Daicel detector are higher than the mica values. This can be explained on the basis of the lower threshold value of the Daicel detector than that of mica. The  $\sigma_{t}$  values of Debeauvais et al.<sup>15</sup> (300 GeV) are higher than ours; those of Hudis and Katcoff<sup>7</sup> (3 and 13 GeV) are lower. Figure 3 shows a rapid increase in the cross sections for  $Z^2/A > 33$ . Thus it would be interesting to extend these experiments to targets heavier than uranium.

The variation of the ratio of the ternary fission to binary fission cross sections  $(\sigma_t/\sigma_b)$  as a function of  $Z^2/A$  of the target material shows a gradual decrease with increasing  $Z^2/A$  value (Fig. 4). The significant difference in the response of Daicel and Muscovite mica is



FIG. 2. Variation of the binary fission cross section  $(\sigma_b)$  as a function of  $Z^2/A$  of the target for 3 GeV (Ref. 9), 13 GeV (Ref. 9), 300 GeV (Ref. 15), and 7 GeV (the present work).



FIG. 3. Ternary fission cross section  $(\sigma_t)$  versus  $Z^2/A$  of the target for protons of various energies.



FIG. 4. Variation of  $\sigma_t/\sigma_b$  as a function of  $Z^2/A$  of the target for protons having energies of 2 and 3 GeV (Ref. 9), 13 and 29 GeV (Ref. 9), 200 and 300 GeV (Ref. 15), and 7 GeV (the present work).

noteworthy. The difference can be explained on the basis of the much higher efficiency of Daicel than of the mica detector for the registration of light charged particles. Since the probability of the emission of a light fragment (reaction product) is higher in the case of a ternary fission event than a binary fission event, a relatively fewer number of ternary fission events will be revealed by etching in mica, which has a lower sensitivity than Daicel. This is supported by our observations in the mica detector combination, which reveals almost 50% more V-shaped events (two correlated tracks having an angle less than 180° between them). This considerable difference in the light particle detection sensitivities of the above-mentioned two detectors produces the difference in the  $\sigma_t/\sigma_b$  ratio. Our Daicel values are higher than the values obtained at 2, 3, 13, and 29 GeV,<sup>9</sup> but lower than those at 200 and 300 GeV.<sup>15</sup> On the other hand, our mica values lie close to those obtained by others.9

F/B, i.e., the ratio of the number of fragments going in the forward direction to those going in the backward direction, is a useful parameter for the determination of the momentum transfer to the struck nucleus.<sup>2,9,15</sup> F/Bresults in the present studies have been summed up in Fig. 5. Higher momentum transfer for lighter target nuclei is quite evident. The values of F/B obtained by the two different detection systems are almost in agreement for  $Z^2/A < 31$ . Our results are fairly close to the values given by Hudis and Katcoff.<sup>7,9</sup>

It was observed that in certain cases both fragments of a binary fission event were registered in the forward detector of the sandwich. Useful information about the momentum transfer in the interaction can be obtained from (a) the number of pairs going in the forward hemi-



FIG. 5. F/B (forward to backward ratio) of reaction products versus  $Z^2/A$  of the target material for protons having energies of 3 GeV (Ref. 9), 7 GeV (the present work), and 13 GeV (Refs. 7 and 9).

sphere as a fraction of the total number of pairs produced, and (b) the angle between the two fragments of the pair. Our results along with some of those previously reported have been summed up in Fig. 6. The present results obtained by using two different detectors (Muscovite mica and Daicel) are found to be in quite good agreement. The only other value available is from Hudis and Katcoff.<sup>9</sup> The comparison of these reported values with our results shows that our results are (a) in agreement with the values for 3 GeV protons<sup>9</sup> and (b) higher than the values quoted for 13 GeV protons,<sup>9</sup> thus showing higher momentum transfer at 3 GeV. Detailed experiments were carried out concerning the distributions of opening angles between the two fragments (A and B) emitted in binary fission. Such angular measurements were made both in the plane of the beam  $(\theta_{AB})$  and in the plane of the detector  $(\alpha_{AB})$  for binary fission events in the Daicel plastic detector. Figure 7 is a set of eight angular distributions obtained for uranium, lead, tungsten, and holmium targets. The mean and median of the  $\theta$ -frequency distributions decrease with decreasing Z number of the target atoms, thus showing that the cone angle between the two fragments is smaller for lighter target nuclei than for the heavier ones, thus confirming our previous results that the momentum transfer to the struck nucleus increases with decreasing Z number of the target atom. On the other hand, the  $\alpha$ -frequency distributions are fairly similar.

Single unpaired tracks having projected lengths greater than 2  $\mu$ m were taken to be caused by spallation reaction products. The variation of the spallation cross section as a function of the mass number A of the target nuclei for 7



FIG. 6. Plot of the ratio of fragment pairs in the forward hemisphere to the total number of pairs versus  $Z^2/A$  of the target material for 3 and 13 GeV (Ref. 9) and 7 GeV (the present work).



FIG. 7. Frequency distributions of angles ( $\alpha$ , in the plane of the detector, and  $\theta$ , in the plane of the beam) between the two fragments of binary fission events.

GeV protons as obtained by using mica and Daicel detectors, along with some previously reported results, has been summarized in Fig. 8. The spallation cross section increases with increasing values of A of the target nuclei. The increase is quite slow in the beginning. It becomes quite rapid for  $A \ge 220$ . The values obtained by using Daicel are higher than those obtained by using the Muscovite mica detector. A comparison of our results with those of Hudis and Katcoff<sup>9</sup> indicates that 7 GeV spallation cross sections are higher than those obtained at 3 GeV proton energy.<sup>9</sup> On the other hand, spallation



FIG. 8. Variation of the spallation cross section as a function of mass number of the target for protons of various energies.

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| Target                          | Detector   | $\sigma_b$ (mb) | $\sigma_t$ (mb)   | $\sigma_t/\sigma_b$ (%) | F/B  | $\sigma_{\rm spall}$ (mb) | $\sigma_f/\sigma_{\rm inelastic}$ (%) |
|---------------------------------|------------|-----------------|-------------------|-------------------------|------|---------------------------|---------------------------------------|
| <sup>238</sup> 92               | mica       | 1035 ±31        | 7.1 ±2.5          | 0.69                    | 0.95 | 652±22                    | 53.55                                 |
|                                 | Makrofol-N | 1021 ±39        | $7.3 \pm 2.1$     | 0.72                    | 0.99 | 669±20                    |                                       |
|                                 | Makrofol-E | $1117 \pm 38$   | $8.3 \pm 2.0$     | 0.74                    | 0.86 | $679 \pm 18$              |                                       |
|                                 | Daicel     | 1196 ±32        | $10.5 \pm 1.8$    | 0.88                    | 0.92 | 895±23                    | 62.00                                 |
| <sup>209</sup> Bi               | mica       | $196 \pm 8$     | $1.47 {\pm} 0.53$ | 0.75                    | 1.29 | 192± 9                    | 11.17                                 |
|                                 | Makrofol-N | $202 \pm 11$    | $1.74 \pm 0.41$   | 0.86                    | 1.21 | $186 \pm 10$              |                                       |
|                                 | Makrofol-E | $201 \pm 7$     | $1.99 \pm 0.43$   | 0.99                    | 1.35 | $206 \pm 13$              |                                       |
|                                 | Daicel     | $227 \pm 7$     | $2.63 {\pm} 0.51$ | 1.16                    | 1.37 | $253 \pm 11$              | 12.99                                 |
| <sup>208</sup> <sub>82</sub> Pb | mica       | 141 ±14         | $0.96 {\pm} 0.31$ | 0.68                    | 1.20 | $195 \pm 10$              | 8.29                                  |
|                                 | Makrofol-N | 143 ±11         | $1.16 \pm 0.29$   | 0.81                    | 1.26 | $198 \pm 11$              |                                       |
|                                 | Makrofol-E | 146 ±13         | $1.27 \pm 0.26$   | 0.87                    | 1.32 | $206 \pm 14$              |                                       |
|                                 | Daicel     | 159 ±12         | $1.72 \pm 0.28$   | 1.08                    | 1.36 | $258 \pm 12$              | 9.39                                  |
| <sup>197</sup> <sub>79</sub> Au | mica       | $103 \pm 7$     | $0.88 \pm 0.33$   | 0.85                    | 1.05 | 178± 9                    | 7.44                                  |
|                                 | Makrofol-N | 98 ± 8          | $0.94 \pm 0.30$   | 0.96                    | 1.09 | 187± 9                    |                                       |
|                                 | Makrofol-E | $112 \pm 8$     | $1.21 \pm 0.32$   | 1.08                    | 1.15 | $193 \pm 11$              |                                       |
|                                 | Daicel     | $121 \pm 10$    | $1.51 \pm 0.26$   | 1.25                    | 1.11 | $226 \pm 13$              | 8.78                                  |
| $^{184}_{74}$ W                 | mica       | 60 ± 6          | 0.56±0.14         | 0.93                    | 1.55 | $150 \pm 12$              | 5.3                                   |
|                                 | Makrofol-N | $63 \pm 7$      | $0.68 \pm 0.15$   | 1.08                    | 1.53 | $158 \pm 11$              |                                       |
|                                 | Makrofol-E | $66 \pm 5$      | $0.81 \pm 0.12$   | 1.23                    | 1.48 | 171± 9                    |                                       |
|                                 | Daicel     | $71 \pm 5$      | $1.02 \pm 0.10$   | 1.44                    | 1.50 | 192± 8                    | 6.3                                   |
| <sup>165</sup> Ho               | mica       | $25 \pm 6$      | 0.30±0.13         | 1.20                    | 2.19 | 119±13                    | 3.51                                  |
|                                 | Makrofol-N | $26 \pm 8$      | $0.42 \pm 0.12$   | 1.62                    | 2.03 | $126 \pm 10$              |                                       |
|                                 | Makrofol-E | $27 \pm 6$      | $0.45 \pm 0.12$   | 1.67                    | 1.99 | 139± 9                    |                                       |
|                                 | Daicel     | $29 \pm 4$      | $0.61 \pm 0.11$   | 2.10                    | 2.10 | 155± 9                    | 4.1                                   |
| <sup>108</sup> <sub>47</sub> Ag | mica       | $3.1\pm 1.1$    |                   |                         | 3.8  | 69± 8                     | 0.87                                  |
|                                 | Makrofol-N | $3.0\pm 1.2$    |                   |                         | 3.6  | 64±13                     |                                       |
|                                 | Makrofol-E | $3.8 \pm 1.0$   |                   |                         | 3.3  | 77±11                     |                                       |
|                                 | Daicel     | 4.3± 1.1        | 0.11±0.06         | 2.6                     | 3.7  | 92±12                     | 1.21                                  |

TABLE I. Parameters obtained by using all four threshold track detectors.

cross-section values for 13 GeV protons<sup>9</sup> are higher than those obtained for 3 and 7 GeV (mica) values, thus showing that the spallation cross section increases with increasing proton energy. The higher sensitivity of Daicel is expected to introduce an error in the spallation cross-section values because of the inclusion of uncorrelated tracks due to some interactions from impurities. This is partially supported by the fact that the experimentally observed total cross section (by using the Daicel detector) exceeds the total calculated inelastic cross section  $\sigma_{\text{inelastic}}$ . However, since the binary and ternary events are always obtained from the correlated events, the use of the Daicel detector is not expected to introduce any appreciable error in deducing the binary and ternary fission cross sections.

The variation of fissility (defined as the ratio of the fission cross section to the total inelastic cross section) as a function of  $Z^2/A$  of the target nuclei is shown in Fig. 9. The values of  $\sigma_{\text{inelastic}}$ , the total inelastic cross sections, were derived from Refs. 24–26 and obtained by following the method outlined in Ref. 15. The results of both the mica and Daicel detection systems show an increase in fissility value with increasing  $Z^2/A$  of the target atoms. The rate of increase becomes higher for  $Z^2/A > 33$ , which suggests that it would be interesting to perform these measurements for targets heavier than uranium. Comparison of the present results obtained for 7 GeV proton energy with those reported previously at 300 GeV proton energy<sup>15</sup> shows fair agreement for uranium and lead. The parameters obtained by using all four threshold track detectors have been summarized in Table I.

The study of three-prong events produced interesting results. The three-prong events were analyzed individually. Total lengths of the three tracks of all the three-prong events were obtained by using (a) Muscovite mica, (b) Makrofol-N, (c) Makrofol-E, and (d) Daicel track detectors for a uranium target using 7 GeV protons. In every three-prong event, the longest track was labeled  $l_{max}$ , while the two smaller ones were called A and B. For every three-prong event, the ratios  $A/l_{max}$  and  $B/l_{max}$  were obtained. Figure 10 shows the  $A/l_{max}$  vs  $B/l_{max}$  plots as obtained by using the above-mentioned four detection systems for a uranium target. The distributions for mica and Makrofol-N are almost uniform. However, there exist two well-defined dense areas or "clusters" of points for the Daicel detector. The clusters are located symmetrical to the central line, and the  $A/l_{max}$  and  $B/l_{max}$  coordinates



FIG. 9. Fissility ( $\sigma_{\text{fission}}/\sigma_{\text{inelastic}}$ ) versus  $Z^2/A$  of the target material for 7 GeV (the present work) and 300 GeV (Ref. 15).

of their centers are approximately (0.2, 0.8) and (0.8, 0.2), respectively. There seems to be some evidence of the existence of the above-mentioned two dense areas in the distribution obtained by using the Makrofol-E detector (the sensitivity of which lies between those of the Makrofol-Nand Daicel detectors). The existence of the two humps in the histogram of the distribution of Daicel shows that in the majority of the three-prong events, the lengths of the tracks are such that two prongs are quite similar in length while the length of the third one is roughly one fifth that of the other two. It was also observed that in almost all the cases contributing to the dense areas, (a) the shorter track was found to lie in the direction of the beam and (b) the two longer tracks were roughly normal to the beam direction. The combination of two long tracks and a short track appears at long etching times only in Daicel, which

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FIG. 10. Distributions of  $B/l_{\text{max}}$  vs  $A/l_{\text{max}}$  of three-prong events produced in uranium and registered in Muscovite mica, Makrofol-N, Makrofol-E, and Daicel track detectors.  $l_{\text{max}}$  is the longest track in a certain ternary event, while A and B are the remaining two shorter tracks (see the text for details).

is capable of registering both light and heavy charged particles. Moreover, even in Daicel, the small tracks appear only after prolonged etchings. Less sensitive detectors (such as mica, Makrofol-N, and Makrofol-E) do not show such a combination of two long and one short track. These observations suggest that the three-prong events having the above-mentioned combinations in Daicel are perhaps two-prong events in other (less sensitive) detectors.

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