Interaction of Yttrium with Protons of Energy between 60 and 240 Mev*

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Absolute cross sections are reported for a number of nuclides produced by the interaction with yttrium of protons of 60, 100, 150, 180, and 240 Mev. At low energies the yields can be accounted for by direct interaction or knock-on processes. At higher energies the knock-on cascade model together with evaporation appears to explain the observed yields.

INTRODUCTION

IN an earlier paper[1] the yields were reported of a number of products from the spallation of yttrium with 240-Mev protons. This paper reports the excitation functions between 60 and 240 Mev of 23 of the product nuclides from yttrium. Data of this type are of interest in trying to understand the mechanisms of nuclear reactions[2] at medium and high energies and indicate the differences from the low-energy compound nucleus mechanism. Similar studies have been carried out on vanadium,[3] cobalt,[4] and cesium[5] in this same energy region.

The results of the interaction of yttrium with 240-Mev protons are in agreement with the currently held idea that the mechanism of such nuclear reactions involves a knock-on cascade process as a primary step, subsequently followed by evaporation of a number of nucleons to remove the termal excitation. Such a mechanism would result in a distribution of product nuclides ranging many mass numbers from the target. The experimental distribution of cross sections of products has the characteristic of being quite flat for some 7 to 10 mass numbers from the target nucleus and then dropping off quite rapidly with decreasing mass number. Such a yield spectrum is an indication of the broad distribution of excitation energies available in these reactions.

EXPERIMENTAL

Spectroscopically pure yttrium oxide[6] was bombarded in the internal proton beam of the Rochester 130-inch synchrocyclotron at radii corresponding to energies of 60, 100, 150, 180, and 240 Mev. The oxide powder, 20–100 mg in weight, was contained in a 5-mil aluminum foil envelope 3 mm×3 mm square in cross section and 1 cm long, this latter dimension vertical and perpendicular to the beam. All targets could be considered thin since the expected energy loss was about 3 Mev at 240 Mev. The chemical separations and methods of identification of nuclides have already been described.[1]

Cross sections of the various nuclides were determined by comparison with the production of Na^{24} by the reaction $Al^{27}(p,3pn)Na^{24}$, the cross section for which has been determined at a number of energies.[7,8]

Pairs of 1-mil aluminum monitor foils were placed before and after the yttrium target envelope. In the determination of Na^{24}, only the inner two foils were used in order to compensate for Na^{24} recoils. In one series of experiments, performed at 60, 175, and 240 Mev, uniform mixtures of aluminum oxide and yttrium oxide were bombarded and the Na^{24} produced from aluminum was chemically separated. These experiments were performed in order to determine whether account need be taken of the radial attenuation of the proton beam through the leading edge of the aluminum envelope. No difference was observed in the results obtained from the two types of targets. All other experimental details were the same as given earlier.[1]

It should be noted that the energy resolution of the Rochester synchrocyclotron decreases with decreasing energy. Thus, at 60 Mev the energy spread of protons actually reaching the target could be as large as about 20% of the nominal energy. This introduces a fairly large error in the various cross sections, decreasing in magnitude, however, as the energy is increased until at 240 Mev the energy spread is less than 2%. Thus, although the individual cross sections of the various nuclides produced at the lowest energy are all plotted at 60 Mev, these values actually represent an integral cross section over an energy band several Mev wide. These effects were estimated in the assignment of errors.

RESULTS AND DISCUSSION

The experimental cross sections for 23 nuclides produced by bombardment of yttrium with 240-, 180-, 150-, 100-, and 60-Mev protons are listed in Table I.

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[1] A. A. Caretto, Jr., and E. O. Wiig, Phys. Rev. **103**, 236 (1956).

[2] See, *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley & Sons, Inc., New York, 1953), Vol. II, p. 141.

[3] C. G. Heininger, Jr., and E. O. Wiig, Phys. Rev. **101**, 1074 (1956).

[4] G. D. Wagner and E. O. Wiig, Phys. Rev. **96**, 1100 (1954).

[5] R. N. Fink and E. O. Wiig, Phys. Rev. **96**, 185 (1954).

[6] The yttrium oxide, supplied through the courtesy of Ames Laboratory, Iowa State College, Ames, Iowa, was >99.9% pure.

[7] L. Marquez and I. Perlman, Phys. Rev. **81**, 953 (1951); L. Marquez, Phys. Rev. **86**, 405 (1952).

[8] N. M. Hintz and N. F. Ramsey, Phys. Rev. **88**, 19 (1952).

TABLE I. Cross sections for the formation of nuclides from the bombardment of yttrium with protons of various energies.

Nuclide	Cross sections in millibarns 240 Mev	180 Mev	150 Mev	100 Mev	60 Mev
Zr^{89}	6.1±0.7	9.3±0.9	9.1±0.9	19.3±2.0	44±4
Zr^{88}	25.6±2.0	38.0±4.0	41.0±5.0	71.0±8.0	140±20
Zr^{87}	9.2±1.0	···	15±3.0	44±5.0	···
Zr^{86}	0.8±0.2	2.0±0.5	2.0±0.5	2.9±0.5	72±20
Y^{88}	93.0±15	120±20	94±12	150±20	420±50
Y^{87}	23.0±4	35±5	49±7	41±7	82±15
Y^{87m+86}	28.2±5	38±5	30±5	97±10	90±10
Y^{85}	15.8±3	17.5±4	···	30±5	4.0±1.0
Y^{82}	0.08±0.04	1.4±0.5	0.77±0.1	···	···
Sr^{87m}	5.0±0.5	6.0±0.5	···	18.4±3.0	54.0±8.0
Sr^{83}	27.5±4	18.5±3.0	11.0±2.0	8.9±1.0	3.5±0.8
Sr^{82}	9.3±5	11.0±3.0	7.0±1.0	2.0±0.5	···
Rb^{86}	4.3±1	1.7±0.5	0.41±0.1	0.2±0.02	···
Rb^{84}	71.4±12	68.0±12	27.0±5.0	9.4±1.0	3.9±0.2
Rb^{82}	37.2±15	40.0±7	25.0±4.0	7.0±1.0	···
Br^{80}	2.7±0.7	2.1±0.7	0.92±0.1	0.50±0.08	0.14±0.05
Br^{77}	4.3±1.0	5.9±1.0	0.66±0.07	···	···
Br^{76}	4.4±1.0	5.3±1.0	2.9±0.4	···	···
Br^{75}	3.8±1.0	2.7±0.5	0.33±0.03	···	···
Se^{73m}	3.5±1.0	2.9±0.7	0.25±0.07	0.001±0.0003	···
Se^{72}	0.96±0.3	0.39±0.1	0.027±0.007	0.0007±0.0002	···
As^{74}	0.77±0.3	0.70±0.2	0.31±0.1	0.027±0.01	···
As^{72}	2.3±0.5	1.4±0.3	0.22±0.05	0.009±0.0003	···
Total observed cross section	379±74	428±70	316±46	502±64	980±129

These values are the averages of two to three runs except at 240 Mev where the values are the averages of three to six experiments. The precision estimate is the average deviation in each case.

The excitation functions for the various nuclides investigated can be roughly divided into three classes. Those nuclides within about four mass units and two units of Z of the target are characterized by a rather rapid decrease in cross section between 60 and about 100 or 120 Mev and then a gradual levelling off or slight decrease in cross section between 100 and 240 Mev. These are shown in Figs. 1 and 2. The second class of excitation functions, for nuclides more than about four mass units from the target, is characterized by a rapid increase in cross section followed apparently by a levelling off as the energy is increased. Such yields are shown in Figs. 3 and 4. The third class of excitation functions is represented by Y^{85} and possibly Y^{86} and is intermediate between the first two classes. These excitation functions show a rapid increase, a maximum, and then a decrease as the energy is increased.

Also listed in Table I is the sum of the experimentally observed cross sections at each energy. At 60 Mev this is a maximum, about 980±129 mb. For a nuclear radius parameter r_0 between 1.2 and 1.4×10^{-13} cm, the geometric cross section for yttrium has values between 900 and 1200 mb. It can thus be concluded that at 60 Mev the observed cross sections account for roughly 100% of the interactions, within experimental error. As pointed out in the earlier paper,[1] the total cross section, including interpolated values for stable nuclides, for 240-Mev protons on yttrium is 773±177 mb, corresponding to approximately 25% nuclear transparency.

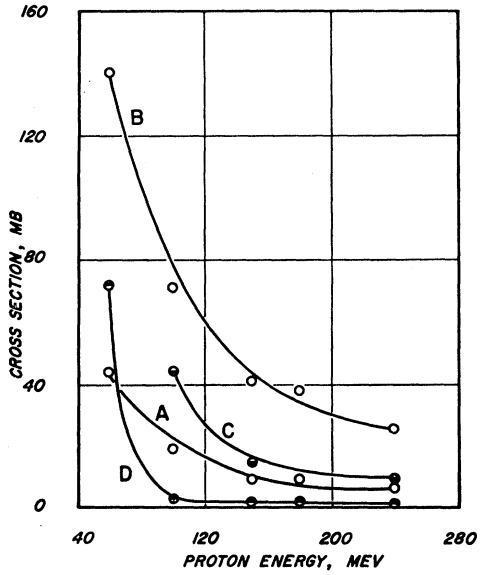

FIG. 1. Cross sections of zirconium isotopes produced from yttrium by protons of various energies: *A*, Zr^{89}; *B*, Zr^{88}; *C*, Zr^{87}; *D*, Zr^{86}.

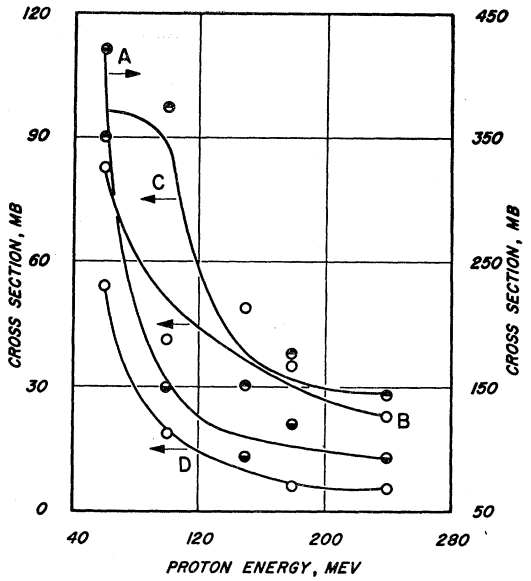

FIG. 2. Cross sections of nuclides produced from yttrium by protons of various energies: *A*, Y^{88}; *B*, Y^{87}; *C*, Y^{87m+86}; *D*, Sr^{87m}. Curves *B*, *C*, *D*, ordinates at left; curve *A*, ordinates at right.

The Monte Carlo calculations of Metropolis *et al.*,[9] for $r_0=1.3\times10^{-13}$ cm show transparencies of 21 and 15% for Cu^{64} and Ru^{100}, respectively, at 235 Mev.

The large cross sections observed at 60 and 100 Mev, as shown in Figs. 1 and 2 for nuclides which are close to the target and which are made in high cross section at low energies, may be accounted for in large part by direct interaction or knock-on of the incident particle.

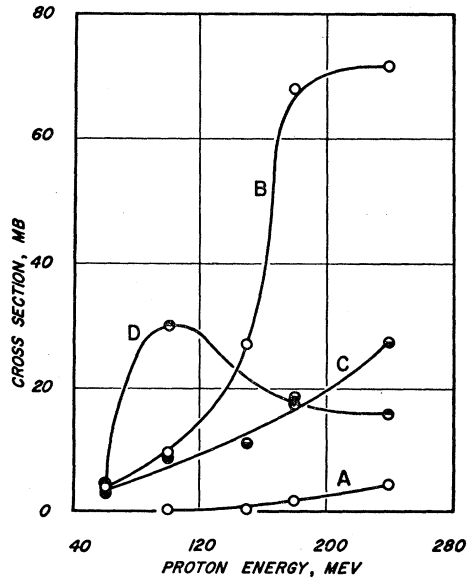

FIG. 3. Cross sections of nuclides produced from yttrium by protons of various energies: *A*, Rb^{86}; *B*, Rb^{84}; *C*, Sr^{83}; *D*, Y^{85}.

[9] Metropolis, Bivins, Storm, Miller, Friedlander, and Turkevich, Phys. Rev. **110**, 185 (1958).

Compound nucleus formation may contribute appreciably to the cross sections at 60 Mev. The Monte Carlo calculations[9] of Metropolis *et al.* show that for Ru^{100} and 82-Mev protons, 20%[10] of the incident protons undergo compound nucleus formation. Even at quite low energies, for 13.2-Mev[11] or 13.5-Mev[12] neutrons on various elements where the major contribution to the (n,p) reaction is made by decay of the compound nucleus, considerable direct interaction occurs. Cohen[13] has also observed direct-interaction effects for 23-Mev protons on a number of target elements. Similarly, Elton and Gomes[14] suggest that in the inelastic scattering of 31-Mev protons only minor roles are played by compound nucleus formation and knock-on events throughout the nuclear volume whereas

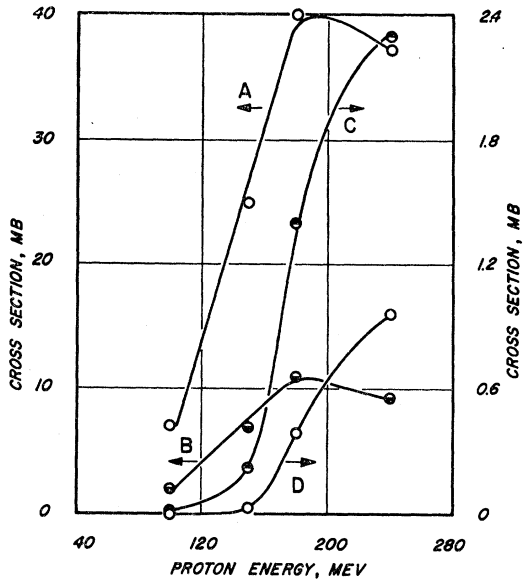

FIG. 4. Cross sections of two isobaric pairs of nuclides produced from yttrium by protons of various energies: *A*, Rb^{82}; *B*, Sr^{82}; *C*, As^{72}; *D*, Se^{72}. Curves *A*, *B*, ordinates at left; curves *C*, *D*, ordinates at right.

the main effect is due to scattering by almost free nucleons in the extreme rim of the outer nucleus.

As may be seen from Table I, for proton energies of 60–240 Mev the cross sections for Y^{88} production are approximately two to three times as large as those for Zr^{88}. Y^{88} may be produced in the following ways: a $(p,p'n)$ cascade leaving a nucleus insufficiently excited to evaporate a nucleon, a (p,p') cascade followed by evaporation of a neutron, and a (p,n) cascade followed by evaporation of a proton. The pickup process may also contribute to Y^{88} production, par-

[10] A. Turkevich (private communication).
[11] Brown, Morrison, Muirhead, and Morton, Phil. Mag. **2**, 785 (1957).
[12] P. V. March and W. T. Morton, Phil. Mag. **3**, 143 (1958).
[13] B. L. Cohen, Phys. Rev. **105**, 1549 (1957).
[14] L. R. B. Elton and L. C. Gomes, Phys. Rev. **105**, 1027 (1957).

ticularly at the lower energies. Zr^{88} may result from only two processes: a (p,nn) cascade yielding a nucleus with insufficient energy to evaporate a nucleon, and a (p,n) cascade followed by evaporation of a neutron.

The Monte Carlo calculations[9,10] already referred to show that for protons of energy 83, 159, and 234 Mev on Ru^{100} the number of $(p,p'n)$ cascades increases slightly with energy while the number of (p,p') cascades leaving a nucleus with sufficient energy to evaporate one neutron decreases slightly, their sums remaining almost constant. While the data for the (p,nn) cascade were not available, the number of such cases would seem to decrease slightly as the $(p,p'n)$ cascades increase slightly, since these are the only cascade processes leading to mass $A-1$ and the Monte Carlo calculations[9] show that the sum of these two processes for protons on Ru^{100} is essentially constant over the energy range 150–325 Mev. The (p,n) cascade may be followed by evaporation of one neutron to yield Zr^{88} or by evaporation of one proton to yield Y^{88}. The ratio of these two processes would not be expected to change appreciably with bombarding energy since the probability of deposition of sufficient energy to evaporate only one neutron or one proton would change only slowly in the energy range investigated. Thus, the constancy of the ratio of the yields of Y^{88} to Zr^{88} is not surprising, since the various processes contributing to the net yields of these two nuclides do not change appreciably relative to one another in our energy range.

The rather rapid increase with energy of the cross sections of nuclides several mass numbers from the target can be explained by the fact that as the incident energy increases, the probability of leaving behind more excitation energy increases, thus making possible the production of nuclides far from the target. The excitation functions of Sr^{83}, Rb^{86}, and Rb^{84} are shown in Fig. 3 and indicate increases in cross sections by a factor of 10 between 60 and 240 Mev.

This same general trend is observed for the isobaric pair Sr^{82}-Rb^{82}, as shown in Fig. 4. Between 100 and 240 Mev the yield of Rb^{82} remains 3.5 to 4 times as large as that of Sr^{82}. The constancy of this isobaric yield ratio seems to suggest that the same mechanism is operative at all energies. That the yield of Rb^{82} is larger than that of Sr^{82} may be due in part to emission of alpha particles during either the nucleonic cascade or the evaporation step, as observed, for example, by Hodgson[15] for 50- to 125-Mev protons on silver and bromine. If the excited nucleus were an yttrium nuclide, then Rb^{82} could be made by loss of neutrons and an alpha particle. However, if the reaction path involved an excited strontium nuclide, the last evaporation step would involve a neutron or proton from an excited Sr^{83} to produce either Sr^{82} or Rb^{82}. Since the last neutron of Sr^{83} is more tightly bound than the last proton, production of Rb^{82} would be favored. Again, Rb^{82} might be expected to be favored since the higher level density of the odd-odd nucleus[16] may enhance the reaction path toward that nucleus. From the data it is difficult to decide unambiguously which reaction path is predominantly involved.

Also shown in Fig. 4 are the excitation functions of the isobaric pair As^{72}-Se^{72}. Measurable cross sections for these nuclides were observed even at 100 Mev, far below the threshold for production of these nuclides by emission of individual protons and neutrons. This suggests that the production mechanism in this case must involve the emission of alpha particles or other fragments larger than protons and neutrons. As was shown by Hodgson[15] in a study of stars produced in photographic emulsions by protons between 50- and 125-Mev energy, there is no appreciable variation in the ratio of the number of alpha particles to all other particles as the energy is increased. The excitation curves for these isobars show a change in the ratio of As^{72} to Se^{72} from about 13 to about 3 in going from 100 to 240 Mev. This would seem to indicate that at the higher energies other reaction paths, not involving alphas or other large fragments, become operative.

Excitation functions of several other nuclides far from the target are not shown but the trend in their cross sections with change in proton energy may be seen in Table I. Some of these show increases in cross section by factors as large as 1000 with increasing energy. Again the threshold for production of these nuclides by emission of single nucleons is larger than the lowest energy at which they were observed and indicates reaction paths involving deuterons, alpha particles, or other fragments. The significant cross section for Br^{80} at 60 Mev, and the more gentle increase in its excitation function, suggest that the reaction path may involve the loss of two alpha particles and a proton and a neutron. As the bombarding energy increases, these excitation functions increase rapidly due to the increase in the number of reaction paths available for their production. This results from the larger excitation energies deposited in the nucleus.

The maximum in the excitation function of Y^{85}, occurring at about 100 Mev (Fig. 3), represents an interesting case of competition between available reaction paths. At low energies the cross section of Y^{85} is low due to the low probability of the specific reaction path $(p,p4n)$ necessary for its production. As the incident energy increases, this reaction path becomes more favorable and reaches a maximum some 20–30 Mev above threshold. The decrease above 100 Mev is then due to the rapid increase in the number of competing

[15] P. E. Hodgson, Phil. Mag. **45**, 190 (1954).

[16] S. N. Ghoshal, Phys. Rev. **80**, 939 (1950); E. Belmont and J. M. Miller, Phys. Rev. **95**, 1554 (1954); Miller, Friedlander, and Markowtiz, Phys. Rev. **98**, 1197 (1955).

reactions leading to other products. Spallation products might be expected in general to show similar excitation functions, though the maximum can be obscured where there are available several reaction paths leading to the same product.

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Number of Prompt Neutrons Emitted per Thorium-232 Fission

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The number of prompt neutrons emitted per Th^{232} fission [$\nu_{Th^{232}}$] is compared to the number emitted per U^{238} fission [$\nu_{U^{238}}$]. At a bombarding neutron energy of 1.4 Mev the ratio $\nu_{Th^{232}}/\nu_{U^{238}}=0.98\pm0.08$.

INTRODUCTION

THE number of prompt neutrons emitted per Th^{232} fission has been measured at 14.2 Mev[1] ($\nu_{Th^{232}}=4.64\pm0.2$) and at an effective neutron energy of 3.5 Mev[2] ($\nu_{Th^{232}}=2.35\pm0.07$). These two measurements lead to a value of $d\nu/dE$ considerably larger than that found for any other fission process.[3] Furthermore they suggest that $\nu_{Th^{232}}<\nu_{U^{238}}$ at neutron energies near the fission threshold. These conclusions are not encouraging to fast thorium reactor concepts.

In order to extend the existing measurements and to obtain information at neutron energies of interest in reactor design, this experiment was undertaken.

EXPERIMENTAL METHOD

Th^{232} and U^{238} samples were contained within a large fission chamber. Neutrons which originated in fission events within the chamber were detected by an adjacent Hornyak[4] button. Throughout the experiment it was assumed that the detection efficiency of the button was identical for both Th^{232} and U^{238} fission neutrons. This assumption is valid if the fission neutron spectra of Th^{232} and U^{238} are similar as is suggested by present knowledge of fission spectra.[5–8] The fission chamber and Hornyak button were irradiated with (1.4±0.08)-Mev neutrons. These neutrons were obtained from the $Li^7(p,n)$ reaction utilizing a Van de Graaff accelerator.

Electronic circuitry simultaneously recorded the following quantities:

(a) the total number of events in the neutron detector coincident with thorium fissions ($N_{Th}{}^t$),
(b) the total number of neutron events coincident with uranium fissions ($N_U{}^t$),
(c) the contribution of chance coincident events to the above two quantities ($N_{Th}{}^{ch}$,$N_U{}^{ch}$), and
(d) the number of fissions occurring in the uranium and in the thorium ($N_{Th}{}^f$,$N_U{}^f$).

The ratio $\nu_{Th^{232}}/\nu_{U^{238}}$ is related to the above experimental quantities through

$$\frac{\nu_{(Th^{232})}}{\nu_{(U^{238})}}=\frac{(N_{Th}{}^t-N_{Th}{}^{ch})/N_{Th}{}^f}{(N_U{}^t-N_U{}^{ch})/N_U{}^f}.$$

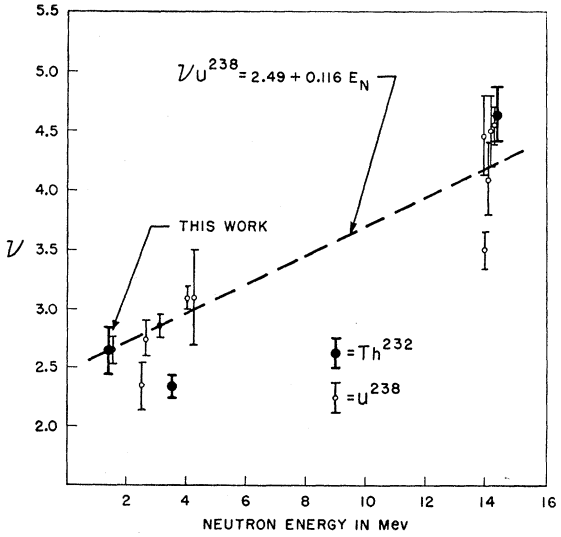

FIG. 1. The value of $\nu_{Th^{232}}$ obtained from this experiment is compared with the results of other workers. Measured values of $\nu_{U^{238}}$ are also shown.

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[1] P. Billaud *et al.*, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, September, 1958* (United Nations, Geneva, 1958), p. 1186.
[2] B. D. Kuzminov *et al.*, Atomnaya Energ. 4, 187 (1958) [translation: Soviet J. Atomic Energy 4, 250 (1958)].
[3] R. B. Leachman, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, September, 1958* (United Nations, Geneva, 1958), p. 2467.
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