THE FISSION TRANSMUTATION OF ^{237}Np IN A FIELD OF SPALLATION NEUTRONS EMITTED FROM THICK LEAD TARGET BOMBARDED BY 1-307 GEV PROTONS AND DEUTERONS

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Large-scale transmutation of radioactive waste, that might be in plenty produced in consequence of industrial reprocessing the irradiated nuclear fuel, would be based, most probably, on applying of intense neutron fluxes (both thermalized and fast) provided by fission reactors and accelerator-driven target-blankets, A lack of the reliable experimental data concerning some bothering nuclides makes difficult to ground practical expediency of the very transmutation approach.

So, up to recently consistent appraisals not always could be done even for rather elaborated systems with neutrons of thermal spectrum (say, transmuting rate of ^{137}Cs/^{90}Sr through capture of thermal neutron). Similarly, in the case of minor actinides (with relatively high fission thresholds) a scarcity of the relevant database enables to estimate, for instance, a burning down of these nuclides in complicated hard neutron spectra (including neutron fields around spallation target driven by ~GeV proton/deuteron beams of ~MW power), in essential, only on the basis of general considerations.

In order to narrow a gap between the experimental data spreading under an ideology of accelerator based large-scale neutron production (say) for purposes of nuclear fuel cycle needs), and calculational methods requiring such data for their validation and testing we performed a series of experiments on fission transmutation of some actinides (^{237}Np, ^{243}Am, ^{240,242}Pu extracted from irradiated fuel, and some reference actinides as well (^{232}Th, ^{235,238}U) in a field of spallation neutrons, Neutron were generated in very thick lead target bombarded with proton beams (in the ~GeV region), The measurements have been carried out at JINR synchrophasotron (Dubna).

The data of such *a* kind can be of interest, among the others, for those accelerator-based actinide transmute concepts that have deal with very hard neutron spectra [1,2], and for which one can appraise rather exactly some characteristic average values, say by analogy with [3], one-groupe fission cross section.

We studied the ^{237}Np fission rate (by now the data for ^{237}Np only are ready to be presented) in neutron field of Pb cylinder $\phi 20 \times 60$ cm, (fig.1) bombarded by protons/deuterons with energies 1.0,1.5, 2.0,2.5,3.17,3.65 GeV. Beam intensities on the front target surface were typically of the order of 10^8-10^9 ions/second, and beam spot size at the same position was 2,5+ 0.5 cm (FWHM) in both the vertical and horizontal directions. Beam monitoring was being done with multiwire proportional chambers and activation reactions $^{27}Al\ (^{p}_{d},\ X)^{24}\ Na$.

Fission detectors were usual solid state nuclear track detectors composed of 6-microns organic film (polyethyleneterephtalate, PETP) in tight contact with layers of actinides to be studied, size of layers being 11.3 mm in diam., ~ 1 mg/cm^2 thick. Tracks were counted automatically by spark counters AIST. The fission event detection efficiencies were calculated using fission cross sections of ENDF/B-V library and recently measured values for high-energy neutrons [4,5]; for Bi detector we used the data from [6]. Detectors were placed over target surface along beam direction and also at angles 10°,30°,60°,90°,120° and 150° relatively to beam axes at a distance 1 m from center of target.

As to the characteristics of spallation neutron field we modified the threshold detector technique having added high-threshold spallation detectors (thick Cu and Cd layers) and Bi to usual set of fission detectors. This improvement enabled to investigate and to measure enough precisely high-energy parts of neutron spectra; the method is described in detail in [7,8]. Some results of spectral measurements on above-mentioned lead target have been reported in [9].

Auxiliary time-of-flight experiments on shorter Pb target $\phi 20 \times 20$ cm showed that a yield of secondary protons from the target was one order of magnitude less than yield of neutrons of equal energies. Nevertheless, we corrected our results for virtual proton contribution into the fission data; efficiencies of spallation detectors (that have been measured on proton beam of various energies) were supposed to be independent of a type of incident nucleon.

The next step of the data processing was a reconstruction of neutron differential distributions after the results of integral measurements (obtained with threshold detectors) by means of an iterative procedure that was realized in computer code RESTOR [7], As a zero approximation of neutron energy spectrum we used the results of time-of-flight measurements for lead target $\phi 20$ x 20 cm. Integration of such reconstructed neutron energy distributions gives neutron yields exceeding by about 15% corresponding values provided from neutron moderation experiments (fig. lb). The difference seems to be quite reasonable taking into account features inherent to moderation technique.

In fig.2-3 normalized distributions of fission rate of ^{237}Np and some reference nuclides are presented. Integration of these distributions, say for ^{237}Np layer of 1 mg/cm^2 thick, fully surrounding Pb target of above-mentioned size, gives 7.09' 10-5 fissions per 1-GeV proton absorbed in lead; at proton energies 2.0,2.55,3.17 and 3.65 GeV the corresponding numbers of fission events amounted to 1.37'10-4, 1,750 10^{-4}, 2,02 "10-4 and 2.3" 10-4 per proton respectively. For deuterons as primary projectiles analogous values are by about 15-20% more what is apparently due to just the same rising of neutron yield when proton beam is substituted for deuteron one (of equal kinetic energy),

But it would be more properly to normalize the integral fission events on neutron yield, rather than on incident ion, Then the fission rates turn to be independent of bombarding particle initial kinetic energy, i.e. with a present level of accuracy of our data the yields, say, of ^{237}Np fissions per neutron emitted from Pb target stop to feel a change of ion initial kinetic energy in spite of the fact that average neutron kinetic energy changes quite noticeably (fig. 1c). additionally, integral yields of ^{237}Np as well as of reference nuclides are insensitive to a difference between neutron spectra generated by proton or deuteron of equal initial kinetic energies,

It means that in our experiments for each type of projectile the ratio

$$< \sigma_f >= \frac{\int \sigma_f(E_n)\varphi(E_n)dE_n}{\int \varphi(E_n)dE_n} \cong const$$

denominator being, by definition, the measured leakage neutron yield. So, spectrum-averaged fission cross sections $< \sigma_f >$ have been determined by experimentally measured values, the neutron yields were being used either from moderation technique measurements (with errors $\pm 3.5 \div 4\%$) and from threshold detector technique supported by time-of-flight method (summary errors $\pm 8 \div 12\%$). These mean cross sections $<\sigma_f>$ are presented in barns:

	^{235}U	^{237}Np	^{238}U	^{232}Th
	Proton			
moderation	1.57± 0.17	1.31± 0,14	0.38± 0.04	0.14± 0.02
TD+TOF	1.37± 0.14	1.14± 0,11	0.33± 0.03	0.12± 0.01
	Deuteron			
moderation	1.55± 0.25	1.32± 0.21	0.36± 0,06	0.14± 0.02
TD+TOF	1.35± 0.25	1.13± 0.23	0.31± 0.05	0.12± 0,02

The relationships between values of $<\sigma_f>$ for four these nuclides are approximately the same for proton and deuteron beams: -

	^{235}U	^{237}Np	^{238}U	^{232}Th
proton	1	0.84	0.24	0.09
deuteron	1	0.84	0.23	0.09

Now a new series of measurements is under way with some additional actinides, more perfect beam monitor devices being used, that will enable to improve essentially accuracy of the data and to understand better details in behavior of averaged cross sections.

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Figure legends

Fig. 1. General characteristics of neutron field around lead cylinder $\phi 20 \times 60$ cm bombarded with protons:

a) angular distributions of various neutron groups (1 - all neutrons, 2 - $E_n > 1$ MeV, 3 - $E_n > 6$ MeV, 4 - $E_n > 20$ MeV, 5 - $E_n > 50$ MeV, 6 - $E_n > 100$ MeV, 7- $E_n > 250$ MeV, 8- $E_n > 500$ MeV, proton energy $E_p = 2$ GeV) measured by threshold detector technique (SSNTD);

b) E_p - dependence of integral neutron yields measured by both moderation technique (□) and SSNTD technique (o); a curve refers to the former cumulative set of the data (that previously have been published by several groups: Oak Ridge-Chalk River, KfK-SIN/ KfK-Saclay, MRTI), which is quite well approximated by function

$$Y_p^m = -8.2(\pm 1.6) + 29.1(\pm 1.3)E_p^{0.75} \text{ n/p},$$

E_p is expressed in GeV; the values (o) were obtained by integration of full neutron spectra and may be described similarly:

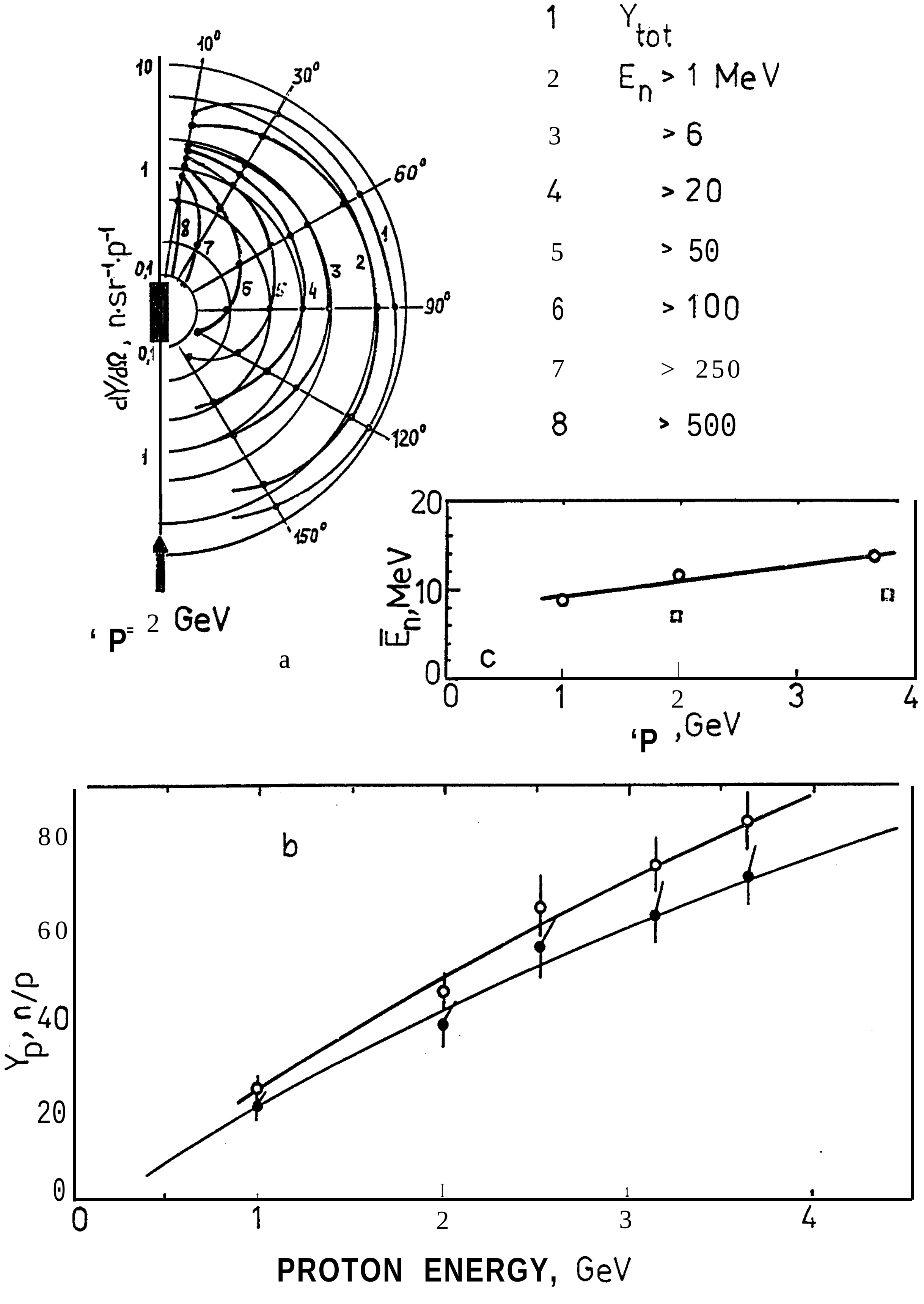
$$Y_p^t = -4.8(\pm 1.0) + 28.6(\pm 2.5)E_p^{0.85} \text{ n/p};$$

when the upper limit of the integration is 15 MeV, what is a reasonable boundary laid by the very moderation technique (upper index m/t means “moderation’’/’’threshold”) then the curves merge

c) average energy of neutrons emitted from Pb cylinder at various energies of incident protons (circles) or deuterons (squares).

Fig.2. Longitudinal (a) and angular (b) distributions of fission rates measured for ^{237}Np and some reference nuclides in neutron field presented in fig. 1, proton energy $E_p = 2$ GeV.

Fig.3. Analogous distributions but measured at proton/deuteron energies 3.7 GeV (squares) and 1.0 GeV (circles), clear symbols refer to deuterons.


1 Y tot
2 E n > 1 MeV
3 > 6
4 > 20
5 > 50
6 > 100
7 > 250
8 > 500
10°
30°
60°
90°
120°
150°
10
1
0,1
0,1
1
dY/dΩ , n·sr⁻¹·p⁻¹
1
2
3
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6
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'P = 2 GeV
a
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Ē n, MeV
c
0
1
2
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'P ,GeV
80
60
40
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Y p, n/p
b
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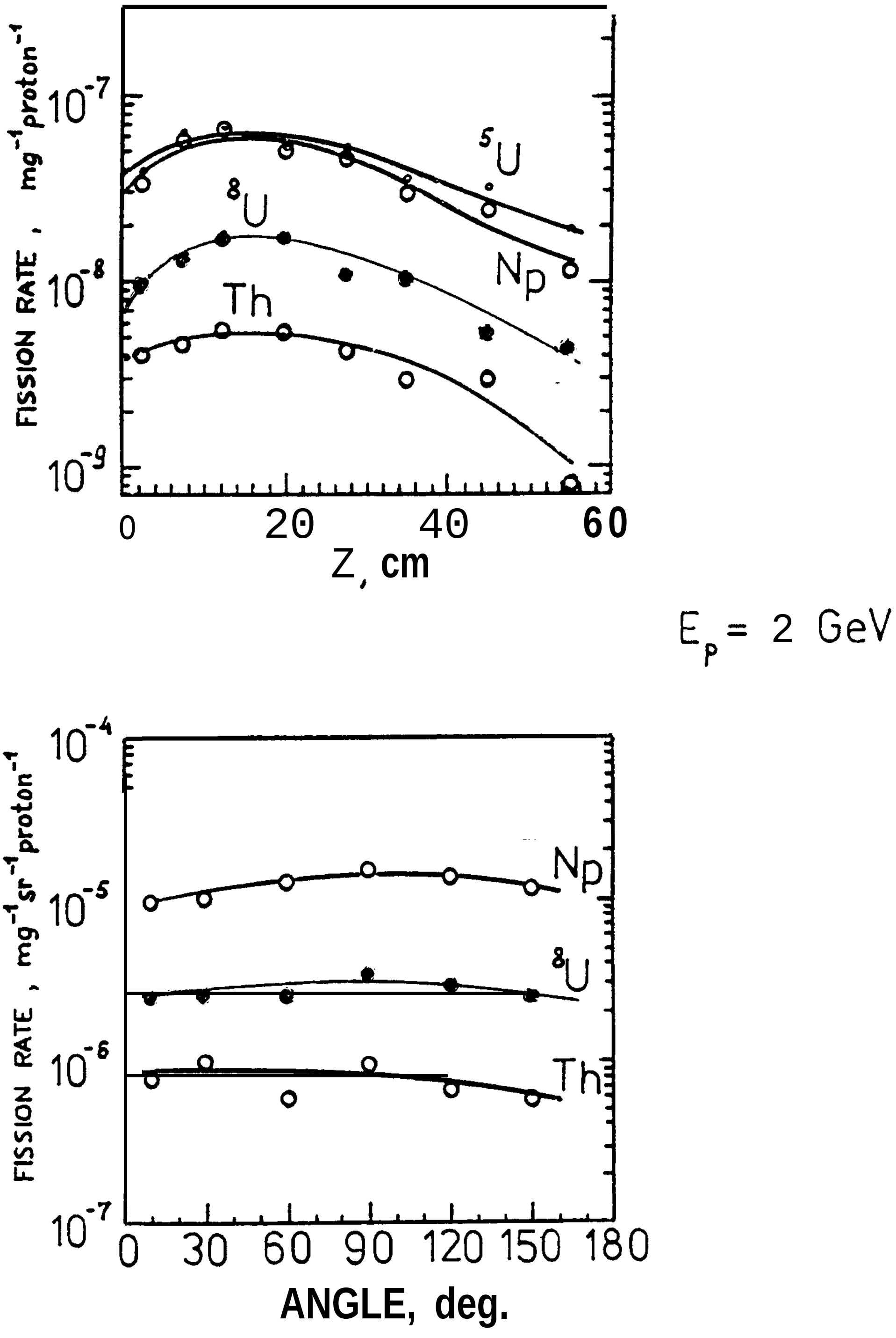
3
4
PROTON ENERGY, GeV


FISSION RATE , mg^{-1}proton^{-1}
10^{-7}
10^{-8}
10^{-9}
^{5}U
Np
^{8}U
Th
0
20
40
60
Z, cm
E_p = 2 GeV
FISSION RATE , mg^{-1}sr^{-1}proton^{-1}
10^{-4}
10^{-5}
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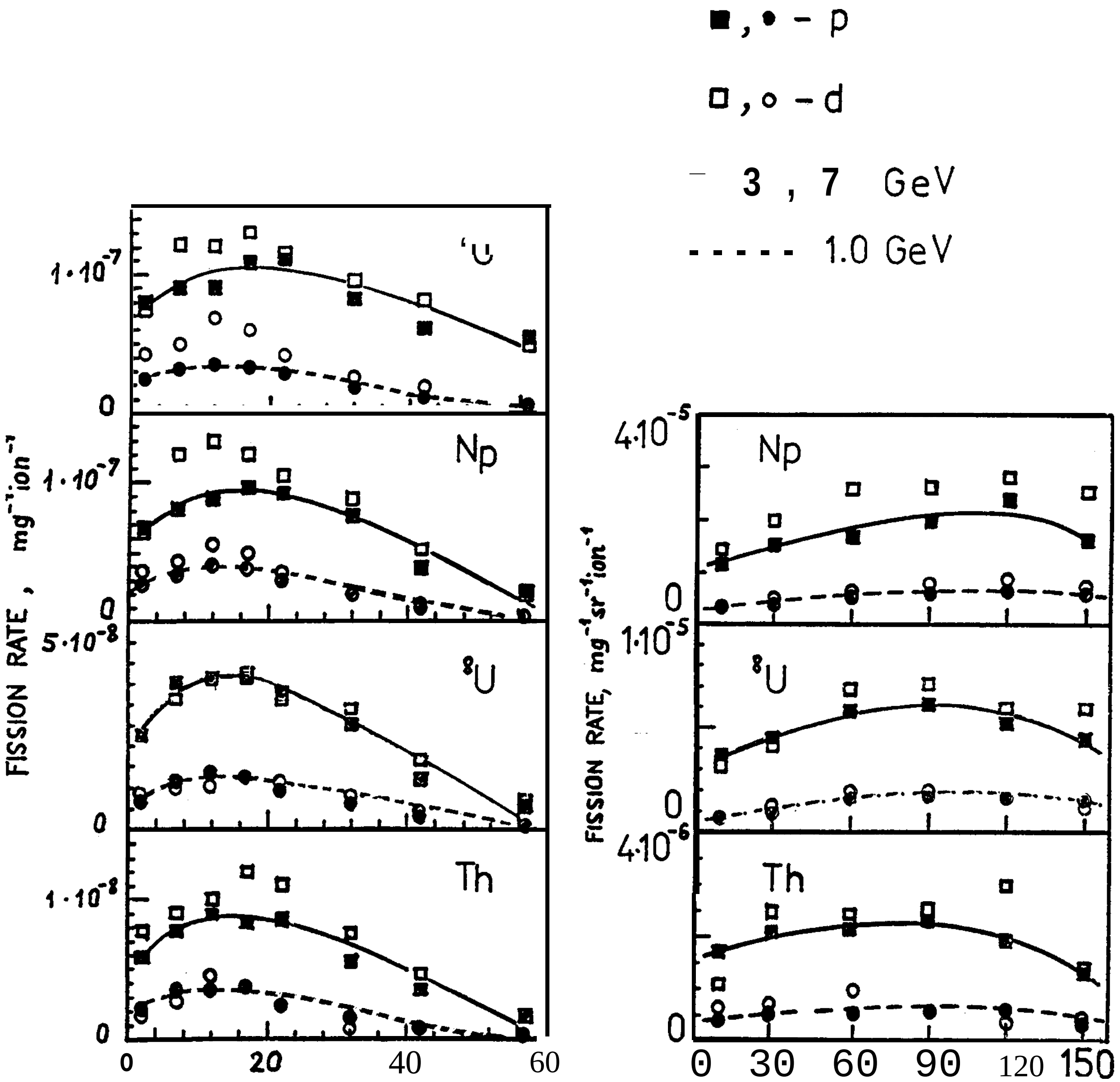
ANGLE, deg.


■, ● - p
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— 3, 7 GeV
- - - - - 1.0 GeV
FISSION RATE, mg⁻¹ion⁻¹
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'U
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5·10⁻⁸
⁸U
1·10⁻⁸
Th
0
20
40
60
Z, cm
FISSION RATE, mg⁻¹sr⁻¹ion⁻¹
4·10⁻⁵
1·10⁻⁵
4·10⁻⁶
0
30
60
90
120
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ANGLE, deg.