REACTIONS OF DEEP SPALLATION OF Fe NUCLEI BY 150-MeV PROTONS

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The main features of the process of spallation of Fe nuclei by protons of $E_p = 150$ MeV energy were studied. The majority of product nuclei were found to be grouped near the bottom of the stability valley. The production cross sections of the spallation products are described by Eq. (1). The weighted mean numbers of the emitted neutrons and protons are, respectively, 2.9 and 2.7. The cross section for the inelastic interaction of 150-MeV protons with Fe nuclei is 568 ± 162 mb. Comparison of σ_{inel} with optical-model calculations yields $R(Fe^{56}) = 1.21 \times 10^{-13}$ cm. The dependence on E_p of the formation cross sections for various nuclei from Fe^{56} is explained by the increase of the nuclear excitation energy as a result of the production and reabsorption of π mesons at $E_p \geq 200$ MeV.

In order to investigate the effects of nuclear reactions produced in iron meteorites by fast nucleons of cosmic radiation, a detailed radiochemical investigation of the final products of spallation of Fe nuclei by 660-MeV protons has been carried out earlier.[1,2] To understand volume effects in the distribution of cosmogenic nuclides in meteorites it was of great interest to study nuclear reactions of protons of lower energy. For this purpose we investigated the main features of the process of Fe spallation by protons of 150-MeV energy, which corresponds to the average energy of secondary particles formed by the interaction of primary cosmic radiation of 0.2—15 BeV energy with complex nuclei.[3] The results of this investigation are given below and are compared with the data for $E_p = 660$ MeV.

EXPERIMENTAL TECHNIQUE

Iron metal powder was used as the target material. Quantitative chemical analysis of the iron detected traces of several elements: 0.0001—0.0003% Mg, 0.007% Si, 0.0003% Ti, 0.003% Cr, 0.01% Mn, 0.005% Co, 0.001% Ni, 0.003% Cu. The iron powder was irradiated in a packet of aluminum foil of 1 × 10 × 30 mm dimensions by the internal proton beam of the Joint Institute synchrocyclotron at an orbit radius corresponding to the energy $E_p = 150$ MeV. The proton flux in the beam was monitored by the Na^{24} activity formed in aluminum by the reaction (p, 3pn). Aluminum foil of 99.9% purity was used as the monitor.

The excitation function of the reaction $Al^{27}(p, 3pn)Na^{24}$ has been investigated in detail from the threshold energy to $E_p = 6$ BeV.[4] The cross section of this reaction at $E_p = 150$ MeV is 9.2 mb.[5] The proton flux, calculated from this value of cross section, varied in different experiments within the limits 3×10^{11}—2×10^{12} protons/cm^2 sec. To compensate for the loss of recoil nuclei from the total number of Na^{24} nuclei formed in the aluminum,[6] the monitoring foil was placed between inner and outer aluminum foils on each side of the iron target. The inner aluminum foil served to absorb the recoil nuclei from the iron target. The possible formation of Na^{24} nuclei by the reaction $Al^{27}(n, \alpha)Na^{24}$ with secondary neutrons was practically avoided because of the small cross section of this reaction for targets thinner than 1 g/cm^2.[4] The target weight was usually 100—500 mg and the duration of irradiation was 0.5—1 hour.

After the target had been dissolved, isotope carriers were used to separate out the radioisotopes of the majority of elements from Be to Co. The activity of the samples was measured with an end-window counter using aluminum filters. The nuclides were identified by their decay half-life, the form of radiation and the energy of β particles and γ quanta. For this purpose a simplified β spectrometer and a scintillation γ spectrometer with a multichannel pulse analyzer were used. The techniques of chemical separation, radionuclide identification, and determination of the formation cross sections were similar to those described earlier.[2]

RESULTS AND DISCUSSION

Identified products of the spallation of Fe nuclei by 150-MeV protons and their formation cross sec-

Identified product nuclei and their production cross sections in the spallation of Fe nuclei by 660- and 150-MeV protons

Nucleus	$E_p = 660$ MeV		$E_p = 150$ MeV	
	σ_{meas}, mb	σ_{calc}, mb	σ_{meas}, mb	σ_{calc}, mb
1	2	3	4	5
$_4Be^7$	2±0,3	—	0,23±0,03	—
$_6C^{11}$	0,15±0,02	—	0,04±0.01	—
$_9F^{18}$	0,20±0,01	0.2	0,014±0,003	—
$_{11}Na^{22}$	0,36±0,03	0,29	0.03±0.01	—
Na^{24}	1,2±0,2	0.62	0,065±0.011	—
$_{12}Mg^{28}$	0,08±0.02	0,11	0,005±0,001	0,003
$_{14}Si^{31}$	0,9±0,2	1.1	0,026±0,013	0.043
$_{15}P^{32}$	2.3±0.2	2,4	0,2±0,1	0.12
P^{33}	1,2±0,23	1.6	0,065±0,032	0,16
$_{16}S^{35}$	1,7±0,4	2,0	0,18±0,09	0,4
S^{38}	0,05±0.01	0,01	—	—
$_{17}Cl^{34m}$	0,6±0,07	0,7	0.11±0.03	
Cl^{38}	0.8±0.07	1,36		0,19
Cl^{39}	0.3±0,04	0.24	0,024±0,008	0.067
$_{19}K^{42}$	2,6±0.23	3,5	0,25±0.05	0.65
K^{43}	0,67±0,09	1.35	0,11±0,04	0,15
$_{20}Ca^{45}$	1.2±0,13	1.26	0.36±0.06	0,33
Ca^{47}	0.07±0,01	0,0067	0,007±0,002	0,0027
$_{21}Sc^{43}$	3±1	5,8	2,5±0,2	1.8
Sc^{44}	9±2,6	13,5	5,9±0,4	4.4
Sc^{46}	5,8±0,9	8.5	3,0±0,6	2,9
Sc^{47}	1,2±0,2	2,2	0,7±0,2	0,8
$_{22}Ti^{45}$	4,6±0,3	6,5	4,5±1,0	3,0
$_{23}V^{47}$	6,1±0,7	7,5	5,9±1,9	4,7
V^{48}	15±2	22	15±2	13,5
V^{49}	25±4	31	33±5	18,5
$_{24}Cr^{48}$	1,1±0,3	1,3	0,5±0,1	1,2
Cr^{49}	7,3±0,5	8,7	6.1±1.7	7,1
Cr^{51}	35±6,2	42	63±19	34,5
$_{25}Mn^{51}$	6.2±1.2	9,8	5.8±1.2	5,4
Mn^{52}	16±2,1	35	14±3	40
Mn^{54}	34±9,8	44	36±16	49
Mn^{56}	2,6±0,8	2,7	0.7±0.2	—
$_{26}Fe^{52}$	1.8±0,3	1,4	5.2±1,4	2,5
Fe^{53}	31±8,7	11	30±2	19
Fe^{55}	60±20	74	110±10	120
$_{27}Co^{55}$	0.8±0.2	—	1,7±0,4	—
Co^{56}	1.3±0.3	—	1,6±0.3	—

tions are listed in the table. The table also gives the production cross sections of these nuclei from Fe^{56} at $E_p = 660$ MeV. The experimental values of the cross sections σ are averages of several separate determinations, and their errors are the rms values. It follows from the table that the products of Fe spallation represent a wide spectrum of radionuclides in the range A = 7—56 with decay half-lives from 8 min to 3 years.

Figure 1 gives a chart of the identified radionuclide products of Fe spallation by 150-MeV protons in coordinates σ_i and A_i. The characteristic feature of the dependence $\sigma_i = f(A_i)$ is, as for $E_p = 660$ MeV, the continuous decrease of the cross sections on increase of $\Delta A = A_{target} - A_i$ along isodialines with $J = N - Z$ = const.

By interpolating the values of the yields of radionuclides which were not observed and of stable nuclides using the nuclide chart it was possible to find the distribution of the sums of cross sections σ_Σ for nuclides with the same atomic

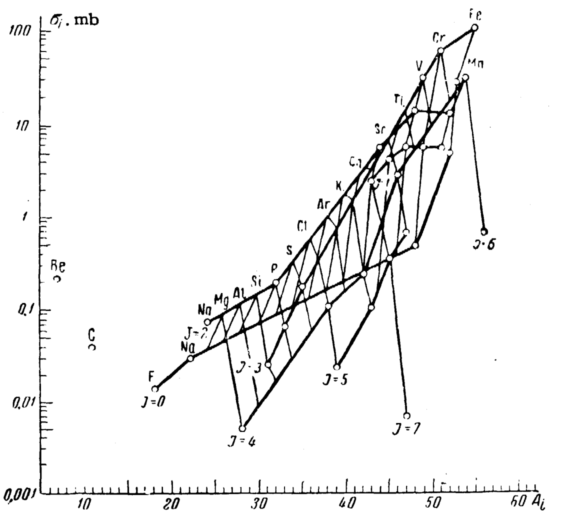


FIG. 1. Chart of identified products of the spallation of iron by 150-MeV protons.

number as a function of the atomic number Z_i (Fig. 2). It follows from Fig. 2 that these sums

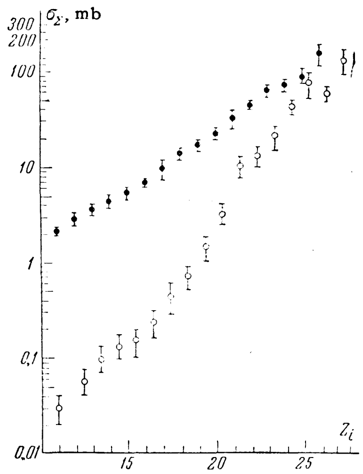
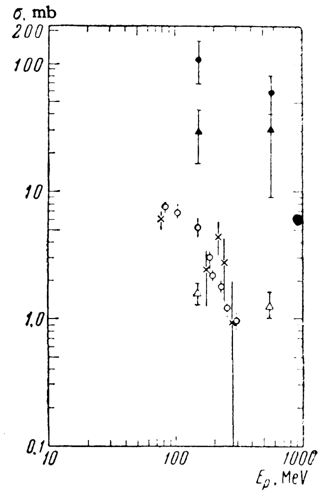


FIG. 2. Distribution of the sums of the formation cross sections of nuclides of given atomic number as a function of Z_i: o – E_p = 150 MeV; • – E_p = 660 MeV.

FIG. 3. Cross sections of the reactions (p, pn), (p, p3n) and (p, n) as a function of the incident proton energy: • – $Fe^{56}(p, pn)Fe^{55}$ reaction, present work; ▲ – $Fe^{56}(p, p3n)Fe^{53}$, present work; Δ – $Fe^{56}(p,n)Co^{56}$, present work; o – $Ni^{64}(p,n)Cu^{64}$, experimental data of Metropolis et al.;[10a] x – $Ni^{64}(p,n)Cu^{64}$, calculations of Koch and Turkevich.[10b]



of cross sections σ_Σ decrease as the difference $\Delta Z = Z_{target} - Z_i$ increases; the decrease ranges over three orders of magnitude from nuclei which are neighbors of the target to light nuclei ($\Delta Z_{max} = 20$). For E_p = 660 MeV this decrease ranges over two orders of magnitude.

The considerable difference between the distributions of the products at E_p = 150 MeV and E_p = 660 MeV is probably due to the formation, scattering, and absorption of mesons, which increase considerably the probability of transferring large excitation energy to a nucleus at E_p = 660 MeV. If the nuclear radius constant is $r_0 = 1.3 \times 10^{-13}$ cm, then the radius of the Fe^{56} nucleus is $R = 4.97 \times 10^{-13}$ cm. The total cross section for the π^+-p interaction for E_{π^+} = 150 MeV is 140 mb.[7] This corresponds to a mean free path of a meson in nuclear matter equal to $\lambda = 6.4 \times 10^{-14}$ cm. Consequently the probability of meson emission from the nucleus is small, and the probability of transferring to neighboring nucleons (by scattering) of an energy averaging 20–30 MeV per collision is high. The cross section for π-meson formation at E_p = 150 MeV is negligibly small.

Let us consider several characteristic groups of spallation products in the light of the nuclear reaction mechanisms of their formation. The most likely products of spallation are the nuclides differing by several mass units from the target nucleus. They are produced by the development of a nucleon cascade and subsequent evaporation in reactions of the type (p, xpyn), where $x+y \leq 6$. The cross section for their production is, for example for the sum of the Cr, Mn, and Fe isotopes, 0.58 and 0.74 of the total cross section for the inelastic interaction at E_p = 660 MeV and 150 MeV respectively. It was shown earlier that on spallation of Cu nuclei by protons of 680-MeV[8] and 340-MeV[9] energies the sum of the production cross sections of the isotopes of Cu, Ni, and Co is respectively 0.6 and 0.8 of the absorption cross section.

Among reactions of this type the reaction $Fe^{56}(p, pn)Fe^{55}$ is of special interest as representing the process of direct knocking out of a neutron on interaction of an incident proton with a nucleon near the nucleus surface. After the collision between the incident proton and the neutron both particles escape from the nucleus, leaving the latter in an excited state but the excitation energy is insufficient for the further evaporation of nucleons. The cross section of the reaction $Fe^{56}(p, pn)Fe^{55}$ decreases by approximately a factor of 2 as the proton energy increases from 150 to 660 MeV (Fig. 3), indicating an increase of the probability of cascade-evaporation reactions with increase of E_p.

Further development of a cascade may lead to reactions of the type (p, pyn) with $y > 1$, for example $Fe^{56}(p, p3n)Fe^{53}$. The cross section of this reaction is the same at E_p = 150 and 660 MeV (Fig. 3). The contribution of the reaction (p, pn) in Fe^{54} (5.84%) amounts to 0.2 and 0.1 of the cross section of this reaction in Fe^{56} at E_p = 150 and 660 MeV respectively.

The charge-exchange reaction (p, n) is also a direct result of the occurrence of a cascade when one neutron is knocked out and the excitation energy of the nucleus is less than the energy necessary for evaporation of further nucleons. Figure 3 shows the calculated and experimantal data for the reaction $Ni^{64}(p, n)Cu^{64}$.[10] The cross sections of the reaction $Fe^{56}(p, n)Co^{56}$ at proton energies of 150 and 660 MeV are also given by this dependence, to within the experimental error.

The majority of spallation products are nuclei formed on emission of ten or more nucleons from the target nucleus. In this region of spallation products we observed the equality of the formation cross sections of the nuclides Sc^{44}, Ti^{45}, Sc^{46}, V^{47}, Cr^{49}, Mn^{51}, Ar^{39}, K^{40}, Ca^{42} at proton energies of 660 and 150 MeV. The spallation products formed on emission of more than 15 nucleons from the target nucleus show a rapid decrease of yield with decrease of the mass number, particularly for E_p = 150 MeV. The formation of nuclides in this region involves considerable excitation energy of the nuclei. Consequently the formation of product nuclei with $40 > A_i > 20$ is energetically more favorable by the process of fission than by the emission of a series of separate nucleons. However, in the region $E_p \leq 100$ MeV the fission cross section is ≤ 0.01 mb, as was found for Cl^{34m-38} on irradiation of V[11] and Co[12] by protons of 60 MeV energy. For nuclides with $A_i \leq 31$, namely Si^{31}, Mg^{28}, Na^{24}, Na^{22}, and F^{18} an increase of yield is observed on going over from the proton energy of 150 MeV to 660 MeV, the increase being by one order of magnitude or more as a result of an increase of the excitation energy of the nuclei by the absorption of π mesons formed at E_p = 660 MeV.

However, in spite of the variety of Z_i and A_i of the observed products of the Fe spallation, the majority of them are grouped near the bottom of the stability valley. The relative probability of the formation of stable nuclides and of nuclides differing from them by one unit of A_i amounts to 0.85 and 0.81 of the total inelastic interaction cross section at E_p of 660 and 150 MeV respectively. The dominant influence in the formation cross section of these isotopes is the presence of an approximately equal number of protons and neutrons in nuclei of medium atomic weight ($_{26}Fe^{56}$, Z = 26, N = 30). Moreover, in the first stages of evaporation the emission of protons and neutrons is equally probable because of the lowering of the Coulomb nuclear barrier at high excitation energies. This is confirmed by the ratio of the weighted mean numbers of the emitted neutrons and protons $\bar{N}_{n,p} = \Sigma(n_i, p_i)\sigma_i/\Sigma\sigma_i$, namely:

	$\bar{N}_n$	$\bar{N}_p$	$\bar{N}_n/\bar{N}_p$
E_p = 660 MeV:	4.1	3.7	1.1
E_p = 150 MeV:	2.9	2.7	1.07,

i.e., the number of neutrons is slightly larger than the number of protons.

The distribution of isotopes of a given element according to mass number is in the form of peaked symmetric curves with peaks at Na^{24}, Mg^{26}, Al^{28}, Si^{30}, P^{32}, S^{34}, Cl^{36}, Ar^{38}, K^{40}, $Ca^{42(43)}$, Sc^{45}, Ti^{47}, V^{49}, Cr^{51}, Mn^{54}, Fe^{55}. The region of nuclides with maximum yield remains constant for E_p = 660 MeV and E_p = 150 MeV (Fig. 1). Using some interpolated values of σ_i from Fig. 1 and experimental values of σ_i, we can obtain the distribution of the cross sections for isobars in the range A_i = 42—48 when E_p = 150 MeV. The yield of isobaric nuclei on both sides of the maximum decreases according to a Gaussian curve, and in coordinates of ln σ, Z it is a parabola given by the equation $\ln \sigma_i = aZ_i^2 + bZ_i + c$ where Z_i is the atomic number of a nuclide with a given cross section σ_i. The parameter a represents the width of the parabolic distribution. The value of the most likely charge Z_0 for a given mass number and the corresponding maximum values of the cross sections are determined from the coordinates of the parabola vertex: $Z_0 = -b/2a$, $\sigma(A, Z_0) = (4ac - b^2)/4a$. The most probable charge of a nucleus with maximum yield at given A may be represented by a linear function of A passing through the origin of coordinates (A_0, Z_0) representing the target nucleus, and having a slope equal to the magnitude of the parameter S in the plot of Z(A) (Fig. 4). A similar dependence is observed for isobars in the range of mass numbers A_i = 42—48 for E_p = 660 MeV.

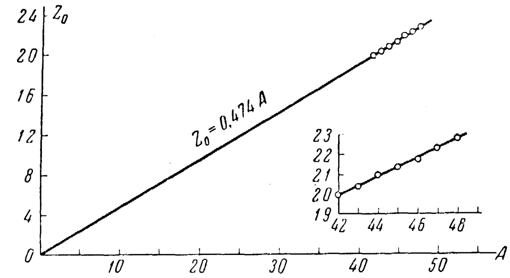


FIG. 4. Distribution of the most probable charge Z_0 as a function of A_i = 42–48.

The method of least squares was applied to the experimental values of the independent cross sections for the formation of nuclei in order to calculate the parameters P, Q, and R in the empirical equation

$$\ln \sigma_i = PA_i - Q - R(Z_i - SA_i)^2. \quad (1)$$

Using this equation we can determine the formation cross section of any product of the Fe spallation by high-energy protons, accurate to within a factor of 2. The calculated values of the parameters P, Q, R, and S are given below for proton energies 150 and 660 MeV. The quantity e^{ϵ} represents the average ratio between the measured and calculated cross sections.

	P	Q	R	S	e^{ε}
$E_p = 660$ MeV:	0.145±0.007	3.91±0.19	1.65±0.08	0.472±0.001	1.35
$E_p = 150$ MeV:	0.308±0.027	12.07±1.06	1.61±0.14	0.474±0.001	1.65

The values of the parameters P, Q, R, and S listed above for the spallation of Fe^{56} by 150 and 660-MeV protons are in good agreement with the results of other work on the spallation products of neighboring elements.[13] The table (columns 3 and 5) lists the cross sections calculated using Eq. (1).

By adding the experimental and interpolated values of σ_i we can estimate the total cross section for the inelastic interaction of iron nuclei:

$$\sigma_{\text{inel}} = 565 \pm 137 \text{ mb for } E_p = 660 \text{ MeV},$$
$$\sigma_{\text{inel}} = 568 \pm 162 \text{ mb for } E_p = 150 \text{ MeV}.$$

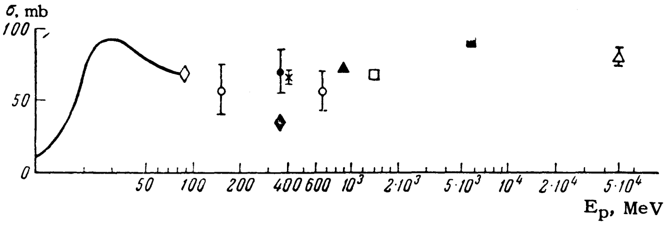


FIG. 5. Dependence of σ_{inel} on $E_{p,n}$ for Fe, Co, and Cu nuclei: ○ – present work; ■ – [4]; × – [14]; ● – [15]; ♦ – [16]; ◇ – [17]; □ – [18]; △ – [19]; ▲ – [20].

Comparison of our data with the results of others is given in Fig. 5 in the form of the dependence of the total absorption cross section for Fe, Co, and Cu nuclei on the energy of incident nucleons (protons and neutrons) in the energy range $E_p = 0.09$–50 BeV. In this range of energies σ_{inel} for target nuclei with Z = 26–29 is considerably smaller than their geometrical cross section, indicating a considerable transparency of nuclear matter (15–25%). It should be noted that the equality of the values of σ_{inel} for iron nuclei at $E_p = 150$ and 660 MeV, obtained in our work, is in agreement with the results of other authors.[21]

Comparison of the total cross section for the inelastic interaction of Fe nuclei with protons of 150 and 660-MeV energy with calculations based on the optical model allows us to estimate the radius of the Fe^{56} nucleus:

$$\sigma_{\text{inel}} = \pi R^2 \left[1 - \frac{1 - (1 + 2KR)\, e^{-2KR}}{2K^2R^2} \right],$$

where $K = 0.36 \times 10^{13}$ cm^{-1} is the absorption coefficient for protons in nuclear matter, and R is the nuclear radius. It follows that $R = 1.21 \times 10^{-13}$ cm and the transparency of nuclear matter is $\sigma_{\text{inel}} / \pi r_0^2 A^{2/3} \times 10^{-26} = 16\%$.

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