SPALLATION REACTIONS: CALCULATIONS*

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Current methods for calculating spallation reactions over various energy ranges are described and evaluated. Recent semi-empirical fits to existing data will probably yield the most accurate predictions for these reactions in general. However, if the products in question have binding energies appreciably different from their isotropic neighbors and if the cross section is ~ 30 mb or larger, then the intranuclear-cascade-evaporation approach is probably better suited.

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INTRODUCTION

There are two general theoretical approaches to the evaluation of spallation reactions. One is a parametric fitting of existing data, and the other is the utilization of various models to calculate the spallation cross sections.

PARAMETRIC STUDY

One of the most widely known parametric studies is that of Rudstam,¹ who attempted to fit experimental data for targets that ranged from vanadium ($Z = 23$) to uranium and for incident protons and alpha particles with energies that varied from 60 MeV to 30 GeV. He found that of the four expressions he examined, an expression of the form

$$\sigma(A,Z) = \sigma \exp[PA - R|Z - Z_p|^{3/2}] \quad (1)$$

with

$$Z_p = SA - TA^2 \quad (2)$$

best fit the data. There are five adjustable parameters in this expression - namely, σ , P , R , S , and T . $\sigma(A,Z)$ is the spallation cross section for producing the residual nucleus (A,Z) and Z_p is the most probable Z , given an A . No product nearer to the target than mass 2 is considered.

Sample results from expression (1) are given in Table I which contains the average ratio of the experimental to calculated quantities for the number of residual nuclides listed. Considering the energy range involved, the results are impressive for iron but relatively poor for bismuth. The comparison suggests that a user should utilize Rudstam's equation with caution.

Table I
Average Ratio of $\sigma_{\text{exp}}/\sigma_{\text{calc}}$ for Spallation Yields^a

Target	Proton Irradiation	No. of Products	$\langle \sigma_{\text{exp}}/\sigma_{\text{calc}} \rangle$
Fe	130 MeV	5	2.18 ± 1.21
	208	5	1.68 ± 0.72
	297	5	0.71 ± 0.27
	340	21	0.64 ± 0.11
	396	5	0.59 ± 0.18
	500	6	0.71 ± 0.10
	590	5	0.82 ± 0.04
	600	6	1.49 ± 0.23
	730	17	0.92 ± 0.13
	24 GeV	18	0.66 ± 0.11
	25	4	1.20 ± 0.10
	29	4	1.03 ± 0.15
Bi	380 MeV	34	1.44 ± 0.23
	480	17	3.69 ± 2.01
	660	21	6.45 ± 1.82

a. G. Rudstam, Z. Naturforschg. 21a, 1027 (1966).

Equally well known is the more recent work of Silberberg and Tsao,² who fitted data over a broader mass range with an expression of the form

$$\sigma = \sigma_0 f(A) f(E) \exp(-P\Delta A - R|Z - SA - TA^2|^V) \Omega \eta \xi \quad (3)$$

where ΔA is the mass difference between the target and residual nucleus. This expression is of the same general form as (1) above, but there are 11 adjustable parameters if one includes $f(A)$ and $f(E)$. Ratios of $\sigma_{\text{calc}}/\sigma_{\text{exp}}$ are generally within the range of 1 ± 0.5 using this method, which probably makes this parametric formulation more accurate for predicting spallation yields than any others at this time.

MODELS

The models that can be used in the alternative theoretical approach were not formulated specifically to predict spallation data but were intended to investigate the underlying mechanisms in nuclear reactions involving continuum-state transitions. They are semiclassical in nature rather than quantum mechanical.

At very low energies, i.e., of the order of the Coulomb barrier, a compound nucleus evaporation model can be used, but it will not be discussed here because of the limited energy for which it might be useful. In the energy region from the Coulomb barrier to about 50 MeV, various pre-equilibrium models might be employed, two of which are discussed below.

Master equation approach: In one such approach (the HMB model), a Boltzmann-like master equation is used to govern the time evolution of the occupation numbers of single-particle energy states within excited nuclei.^{3,4} The changes in the occupation numbers occur via internal two-body scatterings. Included in the equation is a term that describes the probability of emission for the particles that occupy each energy state.

Exciton models: In other models,⁵⁻¹¹ based primarily on the concepts of Griffin,¹² the excited nucleus is characterized by the number of excitons (i.e., excited particles within the nucleus plus holes), and the evolution toward equilibrium takes place via two-body interactions within the nucleus which successively changes the exciton number by ± 2 . Again, at each state described by the exciton number, the probability that a particle will be emitted is calculated.

Evaluation

Regarding the ability of these models to calculate spallation products, they suffer from the fact that a total reaction cross section must be supplied. Furthermore, the initial configuration, be it occupation number or exciton number, must be assumed. Both objections might be circumvented⁴ by initially utilizing an intranuclear-cascade calculation (described below). However, it should be kept in mind that both models were originally formulated to predict the spectra of emitted particles, rather than spallation data, for 10- to 60-MeV nonelastic nuclear reactions.

Intranuclear-Cascade Calculations

These calculations have been used at energies as low as 15 MeV¹³ and as high as 1000 GeV.¹⁴ However, at these energy extremes no attempt was made to calculate cross sections for spallation reactions. Rather, comparisons were made with experimental data for secondary-particle spectra and multiplicities. Results for these comparisons are encouraging. Spallation-reaction cross sections were calculated and compared to experimental data over the energy range from 25 MeV to about 3 GeV, and these comparisons will be discussed below.

Monte Carlo methods are employed in these calculations. The basic assumption is that the initial phase of the calculations (called the cascade) can be represented by a series of sequential two-body interactions. These two-body reactions are specifically calculated, where such things as the spatial point of each interaction, the type of struck particle within the nucleus (neutron or proton), the vector momentum of the struck particle, the final products of the two-body reaction (nucleons or nucleons and pions), and the vector momenta of these products are determined by the Monte Carlo selection procedures along with kinematics calculations that conserve energy and momentum.

In the HMB and exciton models, probabilities for two-body interactions are calculated and summed over all possible successive interactions. In the Monte Carlo approach, the specific life histories of each incident particle and all collision products from the two-body interactions are followed. The summing is replaced by calculating a sufficient number of life histories such that the probability distributions will have been adequately sampled and the normalization is made in terms of the total number of trials. Other important differences are that in the Monte Carlo calculation spatial locations of collision sites and trajectories are identified, whereas in the preequilibrium models they are not, and collision probabilities are governed by transport-type calculations, i.e., $\exp(-\Sigma x)$, rather than by available energy states or phase-space states, as they are in the pre-equilibrium models.

In the cascade calculations, after all reaction products have escaped or have been captured by the nucleus and with the nucleus in a highly excited state, the final phase of the reaction is calculated by evaporation

methods.¹⁵ It is at this stage that the preequilibrium models might be substituted to more realistically represent the completion of the reaction⁴ since the Monte Carlo calculations supply the reaction cross sections and the "state" of the nucleus prior to subsequent decay.

Intranuclear-cascade calculations have been extensively developed at three centers: Oak Ridge National Laboratory (ORNL),¹⁶⁻¹⁸ Brookhaven National Laboratory (BNL)-Columbia University,¹⁹⁻²³ and at the Joint Institute for Nuclear Research (JINR) in Dubna, USSR.^{14,24-27} The mass-yield distributions for reaction energies of 150- and 300-MeV protons on aluminum and tantalum were compared for early versions of these models, and the results were in remarkable agreement even though there were large discrepancies in the excitation-energy distributions of the residual nuclei following the cascade reaction.²⁸

The three calculational versions are similar in that the diffuse edge of the nucleus, the exclusion effects, the motion of the bound nucleons within the nucleus, the spatial variations of the single-particle nuclear potential inside the nucleus, and pion production and pion-nucleon interactions are all taken into account. They differ in that reflection and refraction effects and a velocity-dependent nuclear potential are included only in the BNL-Columbia version; the ORNL version is the only one that does not prohibit two or more interactions from taking place within the same spatial "volume" (a volume equivalent to that occupied by a bound nucleon) inside the nucleus, although efforts to include this prohibition are underway at ORNL; and the JINR version is the only one that permits two or more cascade particles to interact simultaneously with a single nucleon inside the nucleus. Also, the JINR version is the only one of the three

that assigns specific locations for the nucleons of the nucleus, with the locations selected from appropriate charge distributions each time a new incident particle is to be traced.²⁷ Recent versions of the BNL-Columbia model^{22,23} have included isobar²⁹ (excited nucleon states) interactions with nucleons but have not included reflection and refraction effects or a velocity-dependent potential.

For the calculational data that are available, the ORNL version and the BNL-Columbia version that includes reflection and refraction, a velocity-dependent potential, and nucleon correlations (i.e., preventing cascade particles from interacting twice or more in the same nucleon "volume") appear to give the best agreement with each other and with experimental data in general at energies ~ 200 MeV. As already mentioned, these versions also agree with results from JINR with regard to mass yields.²⁸ Similarly, results from the BNL-Columbia version that includes isobar interactions, the JINR version, and those from ORNL appear to agree at energies ~ 1 GeV.

COMPARISON WITH EXPERIMENTAL DATA

Since differences among results from the three intranuclear-cascade models do not appear significant, comparisons with experimental data for spallation yields will be illustrated for only the ORNL results. In order to ascertain systematic differences, if any, comparisons for the energy range of ~ 25 MeV to ~ 400 MeV for incident protons on targets with increasing mass are illustrated first. Results for simple reactions (p,pn) and more complicated reactions (p,xpyn) in this energy range for each target are shown. This is followed by comparisons for incident protons with energies from ~ 500 MeV to ~ 3 GeV on a few targets, and these data, in turn, are followed by comparisons for incident pions from 25 MeV to 1.6 GeV.

Figures 1-8 illustrate the comparison for incident protons over the energy range from 25-400 MeV.³⁰ Except for the $^{12}\text{C}(p,3p3n)^7\text{Be}$ reaction (Fig. 2), the agreement is moderate to good. The ORNL version has been consistently poor in predicting cross sections for producing beryllium in spallation reactions, and the reasons for this are not understood.

Figures 9 and 10 and Tables II and III illustrate comparisons for incident protons at energies ranging from 500 MeV to 2.9 GeV,¹⁶ and the agreement varies from poor to excellent. The latest BNL-Columbia version²³ gives better agreement for the production of ^{24}Na and 150 from 0.6 and 1 GeV protons on ^{27}Al (Fig. 9).

Figures 11 and 12 and Table IV illustrate comparisons for incident π^- with energies ranging from 25 MeV to 2.6 GeV, with reasonable agreement in general. In the energy range shown in Fig. 11, the BNL-Columbia calculation gives better agreement with experimental data for the (π^-, π^-n) reaction on carbon. Comparison of the ORNL results with recently measured $^{12}\text{C}(\pi^\pm, \pi N)^{11}\text{C}$ reactions in the same energy range³¹ (not illustrated) indicate that the model overestimates the $^{12}\text{C}(\pi^-, \pi^-n)^{11}\text{C}$ cross section by about 30% at the peak value and underestimates the $^{12}\text{C}(\pi^+, \pi N)^{11}\text{C}$ cross section by about 20%, which leads to a calculated ratio for $\sigma(\pi^-)/\sigma(\pi^+)$ of 2.6 compared to a measured ratio of 1.55. Effects of nucleon charge exchange, offered as an explanation of this long-standing discrepancy,³² are, in fact, included with little approximation in the intranuclear-cascade calculations. Therefore, these effects should not be major contributors to the discrepancy.

The discrepancy between the predictions and the experimental data at higher energies for the $^{12}\text{C}(\pi^-, x)^{11}\text{C}$ reaction can be attributed to the fact that multiple pion production in the two-body reactions is not included

Fig. 1. Cross section for the $^{12}\text{C}(p,pn)^{11}\text{C}$ reaction vs incident proton energy.

ORNL-DWG 65-11540R3

Fig. 2. Cross section for the $^{12}\text{C}(p,3p3n)^7\text{Be}$ reaction vs incident proton energy.

ORNL-DWG 65-11537R4

EXPERIMENT

- J.B. CUMMING, *NUCL. PHYS.* 49, 417 (1963)
- △ FROM COMPILATION BY E. BRUNINX, CERN 61-1 (1961)
- ▽ BRUN, LEFORT, AND TARRAGO,
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- ▲ I. LEVENBERG *et al.*, *NUCL. PHYS.* 51, 673 (1964)
- ▼ V. PARIKH, *NUCL. PHYS.* 18, 638 (1960)
- V. PARIKH, *NUCL. PHYS.* 18, 646 (1960)

Fig. 3. Cross section for the $^{27}\text{Al}(p,3pn)^{24}\text{Na}$ reaction vs incident proton energy.

EXPERIMENT

▲ N.M. HINTZ AND N.F. RAMSEY, *PHYS. REV.* 88, 19 (1952)

▼ V. PARIKH, *NUCL. PHYS.* 18, 638 (1960)

□ FROM COMPILATION BY E. BRUNINX, CERN 61-1 (1961)

◆ J.B. CUMMING *et al.* *PHYS. REV.* 128, 2392 (1962)

Fig. 4. Cross section for the $^{27}\text{Al}(p,5p5n)^{18}\text{F}$ reaction vs incident proton energy.

ORNL-DWG 65-11544R4

Fig. 5. Cross section for the $^{65}\text{Cu}(p,pn)^{64}\text{Cu}$ reaction vs incident proton energy.

Fig. 6. Cross section for the $^{65}\text{Cu}(p,2p7n)^{57}\text{Ni}$ reaction vs incident proton energy.

Fig. 7. Cross section for the $^{238}\text{U}(p,pn)^{237}\text{U}$ reaction vs incident proton energy.

ORNL-DWG 71-12307

Fig. 9. Spallation cross sections for protons on aluminum.

Fig. 10. Spallation cross sections for the isotopes of mass 72 from 2.9-GeV protons on arsenic. Experimental data are from S. Kaufman, Phys. Rev. 126, 1189 (1962). The solid line, from Kaufman, is the Gaussian curve that best fits the experimental points. He takes $Z_A = 32$.

Table II
Experimental^a and Theoretical Cross Sections for the Formation
of Iodine Isotopes From 720- and 2000-MeV Protons on ^{127}I

Proton Energy (MeV)	Cross Sections (mb)							
	$^{127}\text{I}(p,pn)^{126}\text{I}$		$^{127}\text{I}(p,p2n)^{125}\text{I}$		$^{127}\text{I}(p,p3n)^{124}\text{I}$		$^{127}\text{I}(p,p6n)^{121}\text{I}$	
	Exp.	Theor.	Exp.	Theor.	Exp.	Theor.	Exp.	Theor.
720	55 ± 14	78 ± 7	15 ± 4	29 ± 4	19 ± 5	20 ± 4	13 ± 3	10 ± 3
2000	59 ± 15	61 ± 6	20 ± 5	26 ± 4	18 ± 5	22 ± 4	11 ± 3	7 ± 2

a. I. Ladenbauer and L. Winsberg, Phys. Rev. 119: 1368 (1960).

Table III
Experimental^a and Theoretical Cross Sections for the Formation
of Tellurium Isotopes From 720- and 2000-MeV Protons on ¹²⁷I

Proton Energy (MeV)	Cross Sections (mb)					
	¹²⁷ I(p,2p9n) ¹¹⁷ Te + ¹²⁷ I(p,2p10n) ¹¹⁶ Te		¹²⁷ I(p,2p8n) ¹¹⁸ Te		¹²⁷ I(p,pπ ⁺) ¹²⁷ Te	
	Exp.	Theor.	Exp.	Theor.	Exp. ^b	Theor.
720	23.6 ± 4.0	43 ± 5	31.1 ± 7.8	18 ± 3	< 3.2 ± 1.1	0.7 ± 0.7
2000	7.0 ± 1.5	12 ± 3	18.1 ± 4.5	12 ± 3	< 3.2 ± 1.1	1.3 ± 0.9

a. I. Ladenbauer and L. Winsberg, Phys. Rev. **119**: 1368 (1960).

b. These values represent the sum of the stable and metastable states of ¹²⁷Te.

ORNL-DWG 66-3898R

Fig. 11. Cross section for the $^{12}\text{C}(\pi^-, \pi^- n)^{11}\text{C}$ reaction vs incident pion energy.

Fig. 12. Cross sections for the production of ${}^{11}\text{C}$ from incident π^- mesons. The experimental error in the data of Kaufman and Hower is about the size of the symbols.

Table IV
Experimental and Theoretical Radiochemical Cross
Sections for π^- on Carbon and Argon

π^- Energy (MeV)	Cross Sections (mb)					
	$^{12}\text{C}(\pi^-, \pi^- n)^{11}\text{C}$		$^{40}\text{Ar}(\pi^-, \pi^- p)^{39}\text{Cl}$		$^{40}\text{Ar}(\pi^-, \pi^- pn)^{38}\text{Cl}$	
	Exp. ^a	Theor.	Exp. ^b	Theor.	Exp. ^b	Theor.
600			24.5 ± 1.2	47 ± 3	14.3 ± 0.9	20 ± 2
800	16.1 ± 1.0	19 ± 2				
1100			20.8 ± 1.1	33 ± 3	13.0 ± 1.6	16 ± 2
1600	20.5 ± 0.8	34 ± 3				

a. S. Kaufman and C. O. Hower, Phys. Rev. 154: 924 (1967).

b. C. O. Hower and S. Kaufman, Phys. Rev. 144: 920 (1966).

in the calculations. Such reactions would increase the complexity of the cascade phase, making it less likely that ^{11}C is produced as a final product from a ^{12}C target.

Table V illustrates comparisons from another cascade model (RENO), which is still under development.³³ This model employs a discrete nucleon representation for the target nucleus, as does the JINR version,^{13,23-27} but it can also do the same for complex incident particles. Typical results are shown in Table V, where there is reasonable agreement between the predictions from the model and experimental data.

Finally, results from a preliminary version of a heavy-ion reaction model³⁴ are shown in Table VI. In this model, the individual nucleons of complex incident nuclei are treated independently as they generate cascade reactions within the target nucleus. The total reaction is considered to be the sum of the individual contributions. The data in the middle column for the theoretical results, labeled "smaller," are those that should be compared with the experimental results. The agreement is fair.

Table V

Comparisons of Predictions from the RENO-II Model with
Experimental Data for 150-MeV Protons on ^{16}O

Residual Nuclide	Cross Section (mb)	
	Experimental Data ^a	RENO-II
$^{10}\text{B} + ^{10}\text{C}$	10	16 ± 2
$^{11}\text{B} + ^{11}\text{C}$	25	30 ± 3
^{12}C	30	36 ± 3
$^{13}\text{C} + ^{13}\text{N}$	25	17 ± 2
^{14}O	33	29 ± 3
$^{15}\text{N} + ^{15}\text{O}$	52	51 ± 4

- a. R. Silberberg and C. H. Tsao, *Astrophys. J.* 25, Suppl. 220, 315 (1973).

Table VI

Cross Section (mb) for the Production of Various Fragments with Velocities Within $\pm 2\%$ (for A Values of 12 and 13) and $\pm 4\%$ (for $A \leq 11$) of the Incident ^{14}N Velocity and Emitted into the Angular Interval of 0-4 mrad from the Reactions of ^{14}N at 2.1 GeV/Nucleon on ^{12}C

Fragment	Theoretical Results for the Indicated Nuclear Configurations			Experimental Data ^a
	Standard	Smaller	Smallest	
^7_3Li	0	0.35 ± 0.18	0.30 ± 0.15	9.1 ± 3
^7_4Be	0	0.09 ± 0.09	0.07 ± 0.07	8.0 ± 1.0
^9_4Be	0	0.09 ± 0.09	0.30 ± 0.15	3.0 ± 0.3
^8_5B	0	0	0	5.1 ± 0.6
$^{10}_5\text{B}$	13 ± 3	12 ± 1	9.8 ± 0.9	14.4 ± 2.9
$^{11}_5\text{B}$	8.5 ± 2.1	6.5 ± 0.8	4.9 ± 0.6	10.7 ± 1.3
$^{12}_5\text{B}$	0.5 ± 0.5	2.2 ± 0.4	1.3 ± 0.3	1.9 ± 0.4
^9_6C	0	0.18 ± 0.12	0	0.14 ± 0.02
$^{10}_6\text{C}$	0	0.35 ± 0.18	0.37 ± 0.17	0.96 ± 0.12
$^{11}_6\text{C}$	7.4 ± 2.0	5.4 ± 0.7	4.0 ± 0.5	11.3 ± 1.1
$^{12}_6\text{C}$	68 ± 6	58 ± 2	54 ± 2	46 ± 9
$^{13}_6\text{C}$	36 ± 4	21 ± 1	17 ± 1	9.2 ± 1.8
$^{12}_7\text{N}$	0.5 ± 0.5	0.79 ± 0.26	0.22 ± 0.13	0.63 ± 0.13
$^{13}_7\text{N}$	17 ± 3	11 ± 1	10 ± 1	7.7 ± 1.5
Sum	151 ± 9	118 ± 3	102 ± 3	128.13 ± 10.4
Total Reaction Cross Section	1112	966	918	
% $\frac{\text{Sum}}{\text{Total}}$	14%	12%	11%	

a. H. H. Heckman *et al.*, "Fragmentation of ^{14}N Nuclei at 29 GeV: Inclusive Isotope Spectra at 0°," Phys. Rev. Letters 28, 926 (1972).

SUMMARY

If one is interested in the accurate predictions of spallation yields, the parametric fitting procedure of Silberberg and Tsao² is probably the most appropriate. One can have greater confidence in this method if the reaction energy and target are within the limits of the experimental data that were used to establish the parameters and if the binding energy of the spallation nucleus in question differs little from that of its neighbors. When there are significant differences in binding energies among the neighboring products, any of the intranuclear-cascade models would probably be more reliable, particularly if these cross sections represent at least 10% of the total reaction cross section.³⁰

The suggestion that the state of the excited residual nucleus, as determined by intranuclear-cascade calculations, be used as the starting point for preequilibrium calculations⁴ is surely worthwhile, but it has never been fully explored.

The validity of other approaches^{33,34} for the calculation of spallation yields from reactions between complex nuclei cannot be evaluated until these programs are completed and extensive comparisons with experimental data are made. However, preliminary results are encouraging.

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