Analysis of Isotopic Yields of Primary Residues in 1 A GeV $^{208}$Pb+p Reactions

İskender Demirkol

K. Maraş Sütçüimam University, Faculty of Art and Science, Department of Physics, Kahramanmaraş, Turkey

(Received March 7, 2006)

In this study the production cross sections of heavy residual nuclides in the $^{208}$Pb(1 GeV/nucleon)+p reaction were calculated. The production of more than 350 different isotopes for incident reactions are presented. The calculations were made with the Cascade-Exciton Model including the pre-equilibrium effect, the Intranuclear Cascade Model, the empirical, and the semi-empirical parameterisation. The results of the cross sections obtained were compared with the available experimental data, and the relation between them was examined. It is seen that the precision of these models in estimating the residue production cross sections is still far from the performance required for technical applications. The models should be improved to obtain a deeper understanding of the reaction mechanisms involved in the production of isotopes.

PACS numbers: 24.10.-i, 25.40.Sc, 25.70.Mn

I. INTRODUCTION

Recently, spallation reactions have attracted considerable interest due to their importance in technical applications. They can, for example, be used for the production of neutrons in a spallation neutron source, and they can act as an intense neutron source in accelerator-driven subcritical reactors, capable of incinerating nuclear waste and of producing energy [1–5]. It is proposed that in the next generation radioactive beam facilities, projectile fragmentation at relativistic energies will be one of the major production methods [6]. The generation of residual product nuclei in proton interactions with a heavy target nucleus is due to the intranuclear processes of spallation, fission, fragmentation, and the evaporation of light nuclei and nucleons.

For some years now, heavier projectiles have become available, and investigations have been extended to the fragmentation of Krypton [7, 8], Xenon [9, 10], Gold [11], Lead [12], and Uranium [13]. The nuclide production yields of peripheral relativistic nuclear collisions are of considerable technical importance in several aspects [14]. One application is the production of mass-separated radioactive nuclides in on-line mass separators by the bombarding of heavy target materials with intense nuclear beams [15, 16]. Another field of interest, which is presently being discussed, is the technical application of the nuclear-collision processes for energy production and the transmutation of nuclear waste in a hybrid reactor [17, 18] which works as a fission reactor, where an additional high-energetic proton
or ion beam serves to increase the neutron flux in the reactor in a controlled way.

The current development of neutron generating targets for accelerator-driven nuclear facilities is aimed at extending the data available on the numerous reactions induced by high-energy neutrons and protons in target materials, including the generation of product nuclei within the range of the atomic numbers \( A \) from 1 up to the target atomic number plus. The generated nuclei may be either radioactive or stable. The nuclear characteristics may affect the rated performance and safe operation of a facility through the total activity of the target, the “poisoning” of the target, the accumulation of long-lived radio nuclides to be transmuted, the alpha-activity of the isotopes (e.g. \( \text{Po} \)) produced, the production of nuclides with a high vapor pressure (\( \text{T, He, Hg, etc.} \)), and the accumulation of chemically active nuclides, which reduces the corrosion resistance of the structural materials \([19, 20]\).

Spallation reactions have recently attracted more interest, due their technical applications as intense neutron sources for accelerator-driven subcritical reactors \([21]\) or as spallation neutron sources \([22, 23]\). It is proposed that in the next generation radioactive beam facilities, projectile fragmentation at relativistic energies will be one of the major production methods \([6]\). The mean spallation residue and fission-fragment kinetic energies are particularly important in the technical design of the accelerator-driven subcritical reactors, due to their ability to induce severe radiation damage in the structural materials. Nowadays, applications of Accelerator-Driven Systems (ADS) are usually tuned to the transmutation of transuranic elements and/or long-lived fission products, due to the higher transmutation capability and surmised enhanced safety \([24–27]\). The design of an accelerator-driven system requires precise knowledge of the nuclide production cross sections, in order to be able to predict the amount of radioactive isotopes produced inside the spallation target.

The precision of models in estimating the residue production cross sections is still far from the performance required for technical applications. This can be mostly ascribed to a lack of the complete distributions of all the produced isotopes, which will constrain the models \([28]\). From the production cross sections of long-lived isotopes, for example, it is possible to estimate the level of the long-term radiotoxicity problem of the target. In the present work we have described the production of more than 350 different isotopes from the reaction \( ^{208}\text{Pb} \text{ (1 GeV/nucleon) +p} \). The ability of existing spallation models to accurately estimate the residual production cross sections is still far less than the performance required for technical applications. Therefore this study is aimed at obtaining a deeper understanding of the reaction mechanisms involved in the production of isotopes. The calculations were made with the Cascade-Exciton Model, including the pre-equilibrium effect, the Intranuclear Cascade Model, and the empirical and the semi-empirical parameterisation. The theoretical calculations were compared with the experimental data from the literature, and the relation between them examined. This comparison shows that these incident models have poor predictive power.
II. THE MODELS

II-1. Cascade – Exciton Model (CEM)

Nucleon–nucleus reactions in the medium-energy range still attract much attention, because they present an opportunity to investigate the pre-equilibrium particle emissions [29]. The particle emission mechanism during the attainment of statistical equilibrium in an excited nuclear system is some intermediate between direct reactions and decays through the states of a compound nucleus. The development of the pre-equilibrium concept of the nuclear reactions allows one to understand the importance of this mechanism and its relation to the intermediate nuclear structure. The majority of the exciton models claim only to describe the shape of the angle-integrated energy spectra of secondaries, mainly of nucleons [30, 31]. At higher energies many features of nuclear reactions are fairly well reproduced within the intranuclear cascade model [32].

In the CEM (Cascade-Exciton Model) it is assumed that reactions occur in three stages. The first stage is the intranuclear cascade, in which primary particles can be rescattered several times prior to absorption by, or escape from, the nucleus. The excited residual nucleus remaining after the emission of the cascade particles determines the particle-hole configuration, which is the starting point for the second, pre-equilibrium stage of the reaction.

The subsequent relaxation of the nuclear excitation is treated in terms of the exciton model of pre-equilibrium decay, which includes the description of the equilibrium evaporative third stage of the reaction [33]. In a general case, the three components may contribute to any experimentally measured quantity, in particular, for the inclusive particle spectrum,

\[ \sigma(p) \, dp = \sigma_{\text{in}} \left[ N^{\text{cas}}(p) + N^{\text{preq}}(p) + N^{\text{eq}}(p) \right] \, dp. \]  

The inelastic cross section \( \sigma_{\text{in}} \) is not taken from the experimental data or independent optical model calculations, but is calculated within the cascade model itself [29]. Hence the CEM predicts the absolute values for calculated characteristics and does not require any additional data or special normalization of its results.

II-2. Pre-equilibrium decay of nuclei and intranuclear cascade

To understand the interrelation between the pre-equilibrium nuclear decay and intranuclear cascade models, the main physical assumptions and the resulting different equations describing the relaxation phenomena are briefly treated. Let an excited nuclear system be determined by the Hamiltonian \( \hat{H} = \hat{H}_0 + \hat{V} \), where \( \hat{H}_0 \) refers to the undisturbed nuclear constituents. Now choose the representation in which the undisturbed energy \( E \) is diagonal, \( \hat{H}_0 |E\alpha\rangle = E |E\alpha\rangle \). Here, all indices of a nuclear state, except for the energy, are included in \( \alpha \). Starting with the dynamical Liouville equation and using statistical mechanics methods, one can show that the diagonal elements of the density matrix \( P(E, \alpha, t) \), treated as a probability of finding a system at the time \( t \) in the \( E\alpha \) state, will satisfy the
master equation [34, 35],
\[
\frac{\partial P(E, \alpha, t)}{\partial t} = \sum_{\alpha' \neq \alpha} \left[ \lambda \left( E\alpha, E\alpha' \right) P \left( E, \alpha', t \right) - \lambda \left( E\alpha', E\alpha \right) P \left( E, \alpha, t \right) \right].
\] (2)

Here, the energy-conserving probability rate is defined in the first order time-dependent perturbation theory,
\[
\lambda \left( E\alpha, E\alpha' \right) = \frac{2\pi}{\hbar} \left| \left\langle E\alpha \right| \hat{V} \left| E\alpha' \right\rangle \right|^2 w_\alpha (E),
\] (3)

where the matrix element \( \langle E\alpha \left| \hat{V} \right| E\alpha' \rangle \) is believed to be rather a smooth function in energy, \( w_\alpha (E) \) is the density of final states of the system, and \( \alpha \) is the representation of occupation numbers, \( \alpha \rightarrow \nu_1, \nu_2, \ldots, \nu_A \) in the case of a short-range potential for constituents. The master equation (2) is the mathematical basis of a large class of pre-equilibrium decay models known as exciton models. Within these models, an excited nuclear state is completely defined by an excitation energy and a number of excited nuclear particles \( p \) and holes \( (n = p + h) \) is the number of excitons, that is \( \alpha \equiv n \). The Boltzmann equation is as follows [36]:
\[
\left( \frac{\partial}{\partial t} + \frac{p_k}{m} \nabla_r + F \nabla_{p_k} \right) f_k = \int \int dp_l d\Omega_{rel} \frac{d\sigma (\nu_{rel})}{d\Omega} \left( f_i f_j - f_k f_i \right).
\] (4)

The intranuclear cascade model is based on this equation, but it is preliminarily linearized in the following manner. The fast (cascade) particles and the target-nucleus nucleons which have not yet been involved in the interaction are considered as two different types of particles, and collisions between particles of the same type are neglected. The nuclear constitutions are believed to be described by the equilibrium distribution function \( f^T(r, p) \). Then, for the distribution function of cascade particles, \( f^{cas}(r, p, t) \), we get, from Eq. (4), the following equation:
\[
\left( \frac{\partial}{\partial t} + \frac{p_k}{m} \nabla_r + F \nabla_{p_k} \right) f^{cas}(r, p, t) = \rho^T(r) \langle \sigma_{\nu_{rel}} \rangle f^{cas}(r, p, t) + Q(r, p, t).
\] (5)

According to the normalization of the single-particle distribution function, \( \rho^T(r) = \int dp f^T(r, p) \) is the local number density. In Eq. (5) averaging is carried out over the distribution function of the target nucleus nucleons,
\[
\langle \sigma_{\nu_{rel}} \rangle = \frac{1}{\rho^T(r)} \int dp f^T(r, p) \sigma(\nu_{rel}) \nu_{rel},
\] (6)

and the cross section \( \sigma(\nu_{rel}) \) allows for the exclusion principle. The source function on the right-hand side of Eq. (5) is
\[
Q(r, p, t) = \int \int dp_i d\Omega_{rel} \frac{d\sigma (\nu_{rel})}{d\Omega} f^T(r, p_i) f^{cas}(r, p_j, t).
\] (7)
If the fast particle flux collides with the semi-infinite slab of nuclear matter, we have for the cascade particles,

\[
f^{\text{cas}}(r, p_j, t) = N_0 \delta (p - p_0) \exp \left[ - \int_0^t dt' \rho_T^T \langle \sigma \nu_{\text{rel}} \rangle \right] \\
+ \int_0^t dt'' \rho_T^T \rho^{\text{cas}} \left( r - \frac{p}{m} (t - t''), t'' \right) \\
\times Q \left( r, \frac{p}{m} (t - t''), p, t'' \right) \exp \left[ - \int_{t'}^{t''} dt' \rho_T^T \langle \sigma \nu_{\text{el}} \rangle \right],
\]

where \( \rho^{\text{cas}} (r, t) \) is related to \( f^{\text{cas}} (r, p, t) \) by an equation of the type (5). The probabilistic interpretation of Eq. (8) gives grounds for the cascade model following from an analogy between the interaction of fast particles with nuclei and high-energy radiation transport through matter [32].

The CEM95 code is intended to be used for the Monte Carlo calculation of nuclear reactions within the framework of the Cascade-Exciton Model. The CEM95 code allows us to calculate reactions, elastic, fission and total cross sections, nuclear fissionities, excitation functions, nuclide distributions, energy and angular spectra, double differential cross sections, mean multiplicities, mean energies, and production cross sections for ejectile yields.

Different models are incorporated in CEM95 to calculate the level density parameter.


A powerful method for calculating the nonelastic reactions of nucleons with complex nuclei has evolved over the years. This is the method of intranuclear cascade followed by evaporation. In this approach, the continual state transitions of high energy particles \((E \geq 100 \text{ MeV})\) on nuclei are treated as a two-step process. The first step is the fast cascade, where the reaction is described by a series of individual particle-particle reactions that occur within the nucleus; the second is the evaporation of particles from the excited nucleus remaining after the cascade. In this method Monte Carlo calculation techniques are generally employed [37]. The Medium-Energy Intranuclear Cascade Code System MECC-7 is the code used to calculate the results of nuclear reactions caused by a medium-high energy particle colliding with a nucleus. The code system is based on the assumption that nuclear reactions involving high energy particles can be described in terms of particle-particle collisions, the type of collision and the scattering angles for each collision are determined by statistical sampling techniques.

II-4. Empirical parameterisation of fragmentation cross sections

The EPAX formula is a parameterisation of measured production cross sections from high-energy targets and projectile fragmentation ranging from \( A = 40 \) to \( A = 232 \). It can be seen that this parameterisation gives a fair description of the overall distribution of the production cross sections [38]. The distribution of the projectile-near fragments is influenced by the remaining dependence on the proton-to-neutron ratio of the projectile [39]. The basic characteristics of the analytical description of high energy fragmentation
cross sections by the EPAX formula is as follows [40]:

(i) In the absence of systematic excitation-function measurements of heavy-ion-induced fragmentation reactions, the formula is valid only for the so-called “limiting fragmentation” regime, i.e., for projectile energies dependent. (ii) The EPAX formula is meant to describe the fragmentation of medium to heavy mass projectiles. (iii) For fragments that involve only a small mass loss from the projectile, the isotope distributions should be centered close to the projectile and their variance should be small.

As explained in detail in Ref. [38], the cross section of a fragment with mass \( A \) and charge \( Z \) produced by projectile fragmentation from a projectile \((A_p, Z_p)\), impinging on a target \((A_t, Z_t)\) can be written as

\[
\sigma(A, Z) = Y_A \sigma(Z_{\text{prob}} - Z) = Y_A n \exp \left( -R |Z_{\text{prob}} - Z|^U_p \right) .
\]  

(9)

The first term \( Y_A \) represents the mass yield, i.e., the sum of the isobaric cross sections with fragment mass \( A \). The second term describes the “charge dispersion” the distribution of elemental cross sections with a given mass around its maximum, \( Z_{\text{prob}} \). The factor \( n = \sqrt{R/\pi} \) simply serves to normalize the integral of the charge dispersion to unity. \( R \) is the width parameter. The exponent \( U_p \) is the charge dispersion parameter \((p\)-rich slope \( U_p \)). The mass yield curve is taken to be an exponential as a function of \( A_p - A \). The slope of this exponential, \( P \), is a function of the projectile mass. An overall scaling factor \( S \) accounts for the peripheral nature of the fragmentation reactions, and therefore depends on the circumference of the colliding nuclei:

\[
Y_A = SP \exp \left[ -P (A_p - A) \right] ,
\]

\[
S = S_2 \left( A_p^\frac{1}{2} + A_t^\frac{1}{2} + S_1 \right) \text{[barn]} ,
\]

\[
\ln P = P_2 A_p + P_1 .
\]

(10)

(11)

The most probable charge, \( Z_{\text{prob}}(A) \), is measured relative to the \( \beta \)-stable charge, \( Z_\beta(A) \), \( Z_{\text{prob}} = Z_\beta + \Delta \).

(12)

\( Z_\beta \) is approximated by the smooth function

\[
Z_\beta = \frac{A}{1.98 + 0.0155A^\frac{2}{3}} .
\]

(13)

\( \Delta \) is found to be a linear function of the fragment mass \( A \) for heavy fragments \((A \geq \Delta_4)\), and is extrapolated quadratically to zero:

\[
\Delta = \begin{cases} 
\Delta_3 A^2 & \text{if } A < \Delta_4 \\
\Delta_2 A + \Delta_1 & \text{if } A \geq \Delta_4
\end{cases} .
\]

(14)

The numerical values of the various constants can be found in Ref. [40].
Analysis of isotopic yields of

Vol. 44

(a)

(b)

(c)

(d)

(e)

(f)
FIG. 1: (a)-(s): Isotopic production cross sections of elements to $Z = 82$ from $Z = 65$, in the reaction $^{208}\text{Pb}$ (1 GeV/nucleon) +p versus mass number. The experimental data is taken from Ref. [12].
II-5. Semiempirical parameterisation of fragmentation cross sections

In the Silberberg formula, the procedure relies on the experimentally verified concept that projectile fragmentation cross sections obey the so-called weak factorisation property. In this concept the partial cross section for the production of fragment $f$ can be expressed \cite{41} as

$$\sigma_f = \Gamma_f \Gamma_{P,T},$$  \hspace{1cm} (15)

where $\Gamma_f$ is a factor which depends upon the species of the projectile and the fragment, and $\Gamma_{P,T}$ is a factor which depends only upon the species of the projectile and the target.

In this procedure, $\Gamma_f$ is taken to be proportional to the predicted p-nucleus cross section for the production of the projectile fragment $f$, i.e.,

$$\Gamma_f \rightarrow \gamma_f = \sigma_f (\text{proj + proton} \rightarrow f).$$  \hspace{1cm} (16)

Hence, the factorisation property can now be written as

$$\sigma_f = \gamma_f \gamma_{P,T},$$  \hspace{1cm} (17)

It follows from Eqs. (14)–(16) that $\Gamma_{P,T}$ is different from but geometrically related to, $\gamma_{P,T}$. The dimensionless factor $\gamma_{P,T}$ is developed with the participant-spectator model \cite{42} in mind.

III. RESULT AND DISCUSSION

A comparison of the calculated and experimental isotopic distributions of the production cross sections of elements in the reaction $^{208}\text{Pb} (1 \text{ GeV/nucleon}) + \text{p}$ is shown in
Figs. 1(a)–(s). Fig. 2 shows a comparison of the calculated and experimental cross sections of the spallation residues as a function of the mass loss for the $^{208}\text{Pb}(1\text{GeV/}\text{nucleon})+p$ reaction. The experimental data were taken from Ref. [12]. The design of an accelerator-driven system requires a precise knowledge of the production cross sections of residues inside the spallation target. It is possible to estimate the level of the long term radiotoxicity problem of the target from the production cross sections of long-lived isotopes. Since the spallation residues dominate the isotopic production, they are responsible for most of the radiation damage. Most of the presented distributions exhibit a Gaussian-like shape, where the neutron-proton evaporation competition determines the position of the maximum. The most significant deviations from this shape occur for the neutron rich fragments with masses close to that of the projectile. In the case of these residues, one or more neutron removal channels from low excited nuclei, created mainly in peripheral collisions, are responsible for the increased production cross sections.

Calculations performed with the different INC plus pre-equilibrium, evaporation models [29, 37], the semi empirical parameterisation of Silberberg et al. [41, 43], with the empirical parameterisation EPAX [38, 40] are shown together with the experimental data in Figs. 1(a)–(s), Fig. 2, and Fig. 3. The first two calculations were done with the CEM95 and MECC7 code systems. The shapes of the isotopic distributions obtained with both INC models differ significantly from the experimental data. This can be ascribed to the fact that the predictions of the neutron-proton evaporation competition in the CEM95 and MECC7 code systems are not satisfied. The magnitude of the measured and calculated cross sections is also quite different, especially in the case of the lighter elements. This effect is more visible on the mass distribution in Fig. 3. This discrepancy of the INC models is due to a distribution of excitation energies $E^*$ of the prefragments extending to too high values, which results in the evaporation of more particles, and finally the production
of lighter nuclides. On the other hand, in the region very close to the projectile mass, INC model calculations are in good agreement with the data. The third and fourth calculations in Figs. 1(a)–(s) were performed with the semi-empirical parameterisation of Silberberg et al. and the empirical parameterisation of EPAX. The EPAX calculation reproduces the shape of the experimental isotopic distributions much better than does the former ones. The Silberberg calculation is not in good agreement with the data. The most important contribution to the isotopic production is due to the heaviest elements just below lead in the incident reaction. The conclusion can be made that the reaction $^{208}$Pb (1 GeV/nucleon) +p is not optimumal for producing neutron-rich isotopes in the fission fragment element range. However, this reaction is a good candidate to produce very neutron-rich isotopes of heavier elements closer to the projectile. The magnitude of the cross sections is not always reproduced, the calculation underpredicts the production of the light isotopes. In addition, the main defect of this calculation is the underproduction of isotopes very close to the projectile, which represent an important part of the total cross section. This defect results in a poor prediction of the mass distribution.

Fig. 2 shows a comparison of the experimental data and calculated cross sections of spallation residues as a function of the mass loss. It can be seen in Fig. 2 that, for very small mass losses, there is a sharp decrease in the production cross sections. Then, with increasing mass loss, a nearly constant production followed by an exponential decrease is observed. It can also be seen in Fig. 2 that due to the limited excitation energy induced in the spallation process, the production of spallation residues dies out below $Z = 65$ and $N = 77$. This means that a proton of 1 GeV can not completely destroy a heavy nucleus like $^{208}$Pb. Only a part of the bombarding energy is used to break the nuclear bindings, while a larger fraction, which depends strongly on the impact parameter, is found in the kinetic energies of promptly emitted high-energetic particles.

A comparison with the experimental data clearly indicates that these incident models, the empirical and semi-empirical parameterisation, are not able to reproduce the experimental data with an accuracy that is required for the design and construction of different applications. Pre-equilibrium effects are also important for the calculation of cross sections and the developing of empirical formulations. This study therefore shows that the incident models need to be improved, before a deeper understanding of the reaction mechanisms involved in the production of isotopes can be obtained.

References

* Electronic address: idemirkol@ksu.edu.tr


