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# A study of proton-induced spallation reactions by the improved quantum molecular dynamics model plus statistical decay models

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**Abstract**
The recent GSI data for proton-induced spallation reactions by using inverse kinematics are analyzed by the improved quantum molecular dynamics model (ImQMD05) merged with the generalized evaporation model (GEM2) and GEMINI model. We find that the model of ImQMD05+GEM2 reproduces the experimental data of mass and charge distributions for proton-induced spallation reactions on heavy targets ($^{208}$Pb, $^{238}$U and $^{197}$Au) well and the model of ImQMD05+GEMINI reproduces the ones on light targets ($^{56}$Fe) well. The experimental data for double differential cross sections of emitted neutrons and protons in intermediate energy proton-induced spallation reactions can also be reproduced well with the same models and this shows that they are not very sensitive to the merged statistical model.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

In the last century, a great number of studies on the spallation reactions were made both experimentally and theoretically due to their wide applications in material science [1], biology [2], surgical therapy [3, 4], space engineering [5] and cosmography [6]. Interest in the spallation reactions has recently been renewed because of the importance of intense neutron sources for various applications, such as spallation neutron sources for condensed matter and material science [7–9], accelerator-driven subcritical reactors for nuclear waste transmutation

  

[10, 11] or energy production, as well as for medical therapy. So there is a growing need for nuclear data for spallation reactions at intermediate energies up to 1 GeV for targets not only the neutron production materials such as Pb, Bi, W, but also for surrounding structural materials such as Al, Fe, Ni, Zr and biological elements such as C, O, Ca. Experimental data are important for the application of spallation reactions. However, it is both physically and economically impossible to measure all necessary data [12]. Therefore, theoretical model predictions are very important. At present, the simulation codes are generally Monte Carlo implementations of intra-nuclear cascade (INC) or the quantum molecular dynamics (QMD) model followed by de-excitation (principally evaporation/fission) models. The International Atomic Energy Agency (IAEA) and the Abdus Salam International Center for Theoretical Physics (ICTP) have recently organized an international inter-comparison of spallation models and codes. More than 16 models, including CEM03.03, LAQGSM03.03 [13], BUU [14], QMD [15], JAM [16], JQMD [17], INCL4 [18], ISABEL [19], Bertini [20], Geant4 [21] *et al*, merged with various statistical decay models such as GEM [22, 23], GEMINI [24], SMM [25] *et al*, have been involved. The outcome of the project is significant. The neutron, proton, pion emissions and isotopic distributions can be overall well described by most of the given models. But the data for double differential cross sections for composite particle production are not able to be reproduced well and the discrepancy in the prediction of the excitation functions of residues between most of the models was shown clearly. So there is still a necessity to search for a better theoretical model which can reproduce known nuclear data more exactly and also can predict both the spectra of emitted particles and the yields of residual nuclei well, and is also of good foundation in respect of the model itself. A very detailed review can be found in [26].

In [27, 28] we calculated the double differential cross sections of emitted neutrons by the improved quantum molecular dynamics model (version ImQMD05) plus the statistical decay model and the calculation results were in good agreement with experimental data. In [29] the fragment distributions of proton-induced reactions at intermediate energies were studied by using the quantum molecular dynamics model plus fission models, while experimental data for mass and charge distributions of products in spallation reactions were limited at the time. Since 1996, an experimental program devoted to reaching a full comprehension of the proton-induced spallation reactions has been carried out at GSI. A large number of data for cross sections for the production of practically all possible isotopes from interactions of $^{208}$Pb [30] and $^{238}$U [31] at 1 AGeV, $^{197}$Au at 800 AMeV [32, 33], $^{208}$Pb [34, 35] at 500 AMeV and $^{56}$Fe [36] at 300, 500, 750, 1000 and 1500 AMeV with liquid $^{1}$H have been accumulated up to now. These new data provide a good opportunity for us to test the theoretical models. In this work we apply the ImQMD05 model merged with various statistical models to analyze proton-induced reactions on Fe, Pb, Au and U targets at incident energies from 256 MeV to 1 GeV.

The paper includes the following parts. In section 2, we briefly describe the model adopted in this work. In section 3, we present the calculation results and the comparison with experimental data. A summary and conclusions are presented in section 4.

## 2. Theory model

It is well known that spallation reactions are usually described by three-step processes, i.e. the dynamical non-equilibrium reaction process leading to the emission of fast particles and an excited residue, followed by pre-equilibrium emission, and by the decay of the residue. The first process can be described by microscopic transport theory models, the pre-equilibrium is usually optional in different approaches, and the last one can be described by a statistical model. In this work, we apply the ImQMD05 to describe the dynamic reaction process and the pre-equilibrium reaction process is partially included within this model, and apply two statistical models, i.e. the generalized evaporation model (GEM2) by Furihata [22, 23] and GEMINI by Charity [24], to describe the de-excitation and the fission of residues.

### *2.1. The ImQMD05 model*

A detailed description of the ImQMD model and its new version ImQMD05 and their applications can be found in [37–39]. Here, we only mention that in this model the full Skyrme potential energy density is adopted with just the spin–orbit term omitted, which reads

$$V_{\mathrm{loc}} = \frac{\alpha}{2}\frac{\rho^2}{\rho_0} + \frac{\beta}{\eta+1}\frac{\rho^{\eta+1}}{\rho_0^{\eta}} + \frac{g_{\mathrm{sur}}}{2\rho_0}(\nabla\rho)^2 + \frac{g_{\mathrm{sur,iso}}}{\rho_0}[\nabla(\rho_{\mathrm{n}}-\rho_{\mathrm{p}})]^2 + (A\rho^2 + B\rho^{\eta+1} + C\rho^{8/3})\delta^2 + g_{\rho\tau}\frac{\rho^{8/3}}{\rho_0^{5/3}}, \tag{1}$$

where $\rho$, $\rho_{\mathrm{n}}$, $\rho_{\mathrm{p}}$ are the nucleon, neutron and proton density, and $\delta = (\rho_{\mathrm{n}}-\rho_{\mathrm{p}})/(\rho_{\mathrm{n}}+\rho_{\mathrm{p}})$ is the isospin asymmetry. The parameters that appeared in the potential energy density functional are all obtained from the parameters of standard Skyrme interactions. In this work the SkP [40] Skyrme interaction is adopted because we found that the SkP was successful in the calculations of double differential cross sections of emitted neutrons in proton-induced spallation reactions [27, 28], and also of some observables in heavy ion reactions at intermediate energies [39, 41, 42]. In the collision term, isospin-dependent nucleon–nucleon scattering cross sections [43] are used and the Pauli blocking effect is treated more strictly [39, 44].

It is of crucial importance to have the initial nuclei in the real ground state because considerable excitation of initial nuclei will lead to unreal nucleon emission and affect the low energy part of the neutron spectrum. We check carefully not only the binding energy and the root-mean-square radius of the initial nuclei but also their time evolution. The average binding energy per nucleon of initial nuclei is required to be $E_{\mathrm{g.s.}} \pm 0.1$ MeV, where $E_{\mathrm{g.s.}}$ is the binding energy of nuclei in the ground state. Only those initial nuclei with no spurious particle emission are taken to be good initial nuclei, and then are applied in the simulation of the reaction process.

The switching time from the ImQMD05 to GEM2 or GEMINI is taken to be 100 fm/$c$. At the end of the ImQMD05 calculations, clusters are constructed by means of the coalescence model widely used in the QMD calculations. The particles with relative momenta smaller than $P_0$ and relative distances smaller than $R_0$ are coalesced into one cluster. Here $R_0 = 4.5$ fm and $P_0 = 300$ MeV/$c$ are adopted. Then we calculate the total energy of each excited cluster in its rest frame and its excitation energy is obtained by subtracting the corresponding ground-state energy from the total energy of the excited cluster.

### *2.2. The GEM2 model*

A very detailed description of the GEM2 can be found in [22, 23]. Therefore, we only mention some modifications and parameters adopted in this work.

Furihata used GEM2 coupled either with Bertini's [20] intranuclear cascade (INC) or with the ISABEL [19] INC code, which differ from the ImQMD05 model and do not include the emission of pre-equilibrium particles, while the pre-equilibrium process is partially considered in ImQMD05 although the system still does not achieve a complete equilibrium state at the switching time. So, naturally, the parameters adjusted by Furihata should be readjusted for merging with the ImQMD05 model to get the best calculation results.

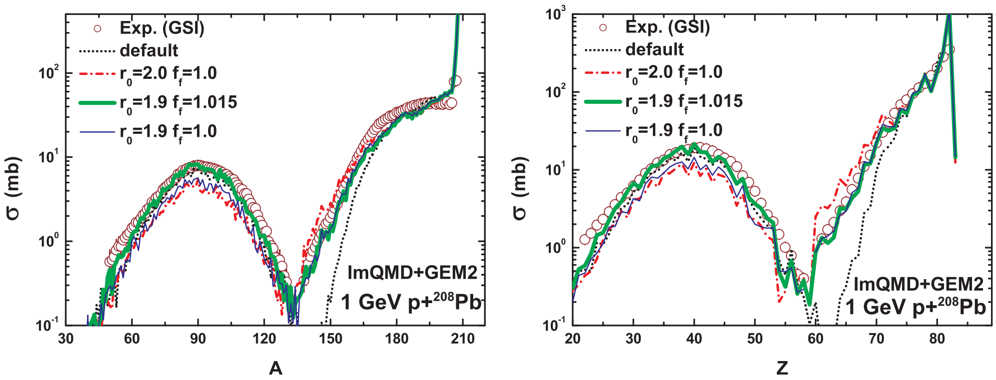


**Figure 1.** Comparison of the experimental mass and charge distributions of the nuclei produced in the reaction 1 GeV p+$^{208}$Pb [30] (circles) with the calculations by ImQMD05+GEM2 with different GEM2 parameters adopted.

In GEM2 Dostrovsky's formula is used to calculate the inverse cross sections for all emitted particles and fragments with total kinetic energy $\epsilon$:

$$\sigma_{\text{inv}}(\epsilon) = \sigma_g \alpha \left(1 + \frac{\beta}{\epsilon}\right), \tag{2}$$

which is often written as

$$\sigma_{\text{inv}}(\epsilon) = \begin{cases} \sigma_g c_n (1 + b/\epsilon) & \text{for neutrons} \\ \sigma_g c_j (1 - k_j V/\epsilon) & \text{for charged particles} \end{cases}$$

where $\sigma_g = \pi R_b^2$ (fm$^2$) is the geometrical cross section, and

$$V = Z_j Z_d e^2 / R_c \tag{3}$$

is the Coulomb barrier in MeV and $j$, $d$ denote the emitted particle and daughter nucleus. There are two kinds of parameter sets to be chosen in the calculation of inverse cross sections. One is the 'precise' parameter set as default option which can be found in [22, 23], and the other is the 'simple' parameter set adopted in this work, which is given as $c_n = c_j = k_j = 1$, $b = 0$ and $R_b = R_c = r_0\left(A_j^{1/3} + A_d^{1/3}\right)$ (fm), where $r_0$ should be adjusted according to the incident energy. The fission width for nuclei with $70 \leqslant Z_j \leqslant 88$ is calculated as

$$\Gamma_f = \frac{(s_f - 1)e^{s_f} + 1}{a_f}. \tag{4}$$

When $Z_j \geqslant 89$, the fission width is calculated by the approximate expression

$$\log(\Gamma_n / \Gamma_f) = C(Z_i)[A_i - A_0(Z_i)]. \tag{5}$$

As Mashnik *et al* did in their CEM model calculations work [45], in order to merge the GEM statistical model here we also readjust $r_0$ to fit the spallation product cross sections and $a_f$, the level density parameter, to fit the fission product cross sections by using Atchison's fission model. For the fission of elements with $Z_i \geqslant 89$, we find that the default parameter is appropriate and does not need re-adjustment. Now let us illustrate the reaction of 1 GeV p+$^{208}$Pb [30] calculated by ImQMD05+GEM2 with different GEM2 parameters, for example, to explain the parameter adjustment process. In figure 1 we show the calculation results by the ImQMD model plus GEM2 with and without parameter adjustment and the comparison with the experimental data for the mass and charge distributions of the products in the reaction.

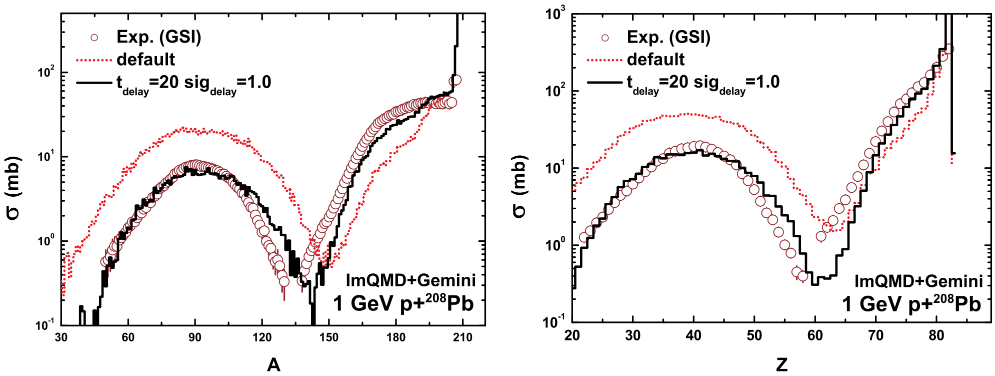


**Figure 2.** The same as figure 1 but for the calculation results of ImQMD05+GEMINI with different GEMINI parameters adopted. Experimental data are taken from [30].

One can see from the figure that the fission product cross sections are reproduced quite well but the light spallation product cross sections are too low to reproduce the experimental data if we only apply GEM2 without any modification of parameters (see the black dashed line in figure 1). Then we adopt the simple parameter set to calculate inverse cross sections with readjusted $r_0$; one sees that a good agreement with experimental data for spallation product cross sections is obtained as shown by the orange dashed dotted line and royal blue thin solid line in figure 1, where $r_0 = 1.9$ fm is adopted. But the fission product cross sections are depressed as the spallation product cross sections are enhanced now due to the competition between spallation and fission. So we have to readjust certain parameters to improve the fission product cross sections. Here a reduced factor $f_f$ is introduced, i.e. $a_f = f_f a_f$. Finally, for 1 GeV p+$^{208}$Pb, the experimental data for mass and charge distributions can be reproduced with the combination of $r_0 = 1.9$ and $f_f = 1.015$ (see the green thick solid line in figure 1).

All the default options in GEM2 except the parameters mentioned above, i.e. $r_0$ and $a_f$, are adopted in our calculations to analyze the recent GSI experimental data of intermediate energy proton-induced reactions in inverse kinematics.

### 2.3. The GEMINI model

A very detailed description of GEMINI can be found in [24] and references therein. The level density parameter is taken as Grimes case B modified form, i.e. $aden\_type = -23$. We use here the default version of the GEMINI model, without any changes of fitting parameters, except for the value of the delay time for fission (GEMINI input parameters $t\_delay$ and $sig\_delay$). The modification of these parameters was first studied in a version of CEM merged with GEMINI [46]. The parameter $t\_delay$ is used to control the competition between the spallation and fission, and the parameter $sig\_delay$ is used to adjust the mass/charge distributions of fission products. With non-zero $t\_delay$, fission and intermediate mass fragment decay is inhibited for a time of

$$t_{\mathrm{delay}} \exp\left(\frac{-\eta^2}{2\sigma^2_{\mathrm{delay}}}\right) \times 10^{-21}(\mathrm{s}), \tag{6}$$

where $\eta = (A_2 - A_1)/(A_2 + A_1)$, with $A_1$ and $A_2$ being the mass of the two fission fragments.

Figure 2 presents the comparison between the calculated results for the mass and charge distributions of the products in the reaction 1 GeV p+$^{208}$Pb by ImQMD05+GEMINI with

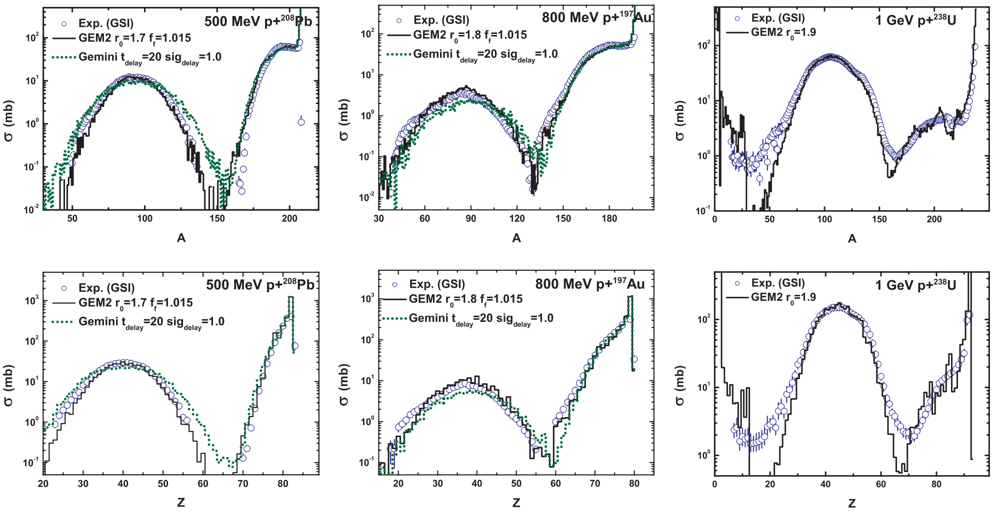


**Figure 3.** Comparison between the ImQMD05+GEM2 model and ImQMD05+GEMINI model calculation results and experimental data for mass and charge distribution cross sections of products in 500 MeV p+$^{208}$Pb [34, 35], 800 MeV p+$^{197}$Au [32, 33] and 1 GeV p+$^{238}$U [31], respectively. The parameters used in the GEM2 and GEMINI calculations are also shown in the figures.

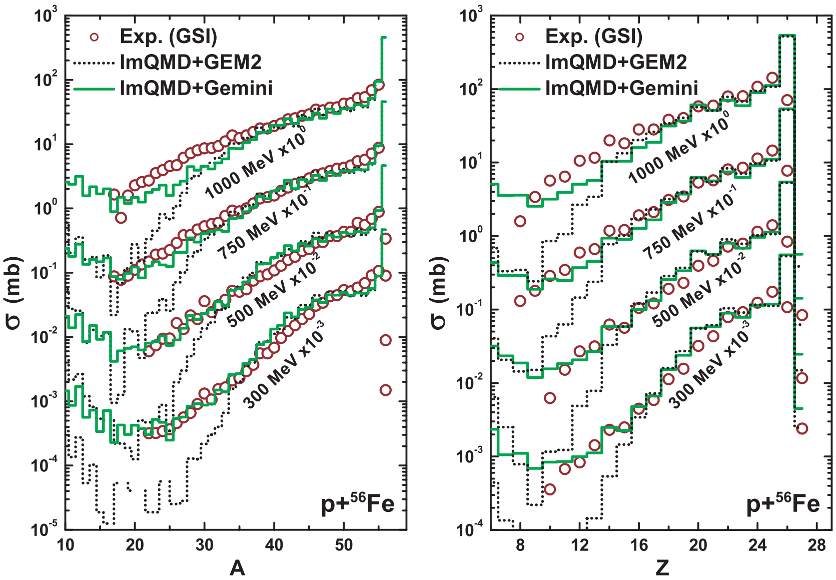


**Figure 4.** The same as figure 3 but for 300, 500, 750 and 1000 MeV p+$^{56}$Fe. The experimental data are taken from [36]. Default options are used in both GEM2 and GEMINI.

different GEMINI parameters and experimental data. One can see that with the default option, GEMINI provides too many fissions and too wide a fission product distribution. By varying the $t\_delay$ and $sig\_delay$ we find that the calculation results with $t\_delay = 20$ and $sig\_delay = 1.0$ fit the experimental data best.

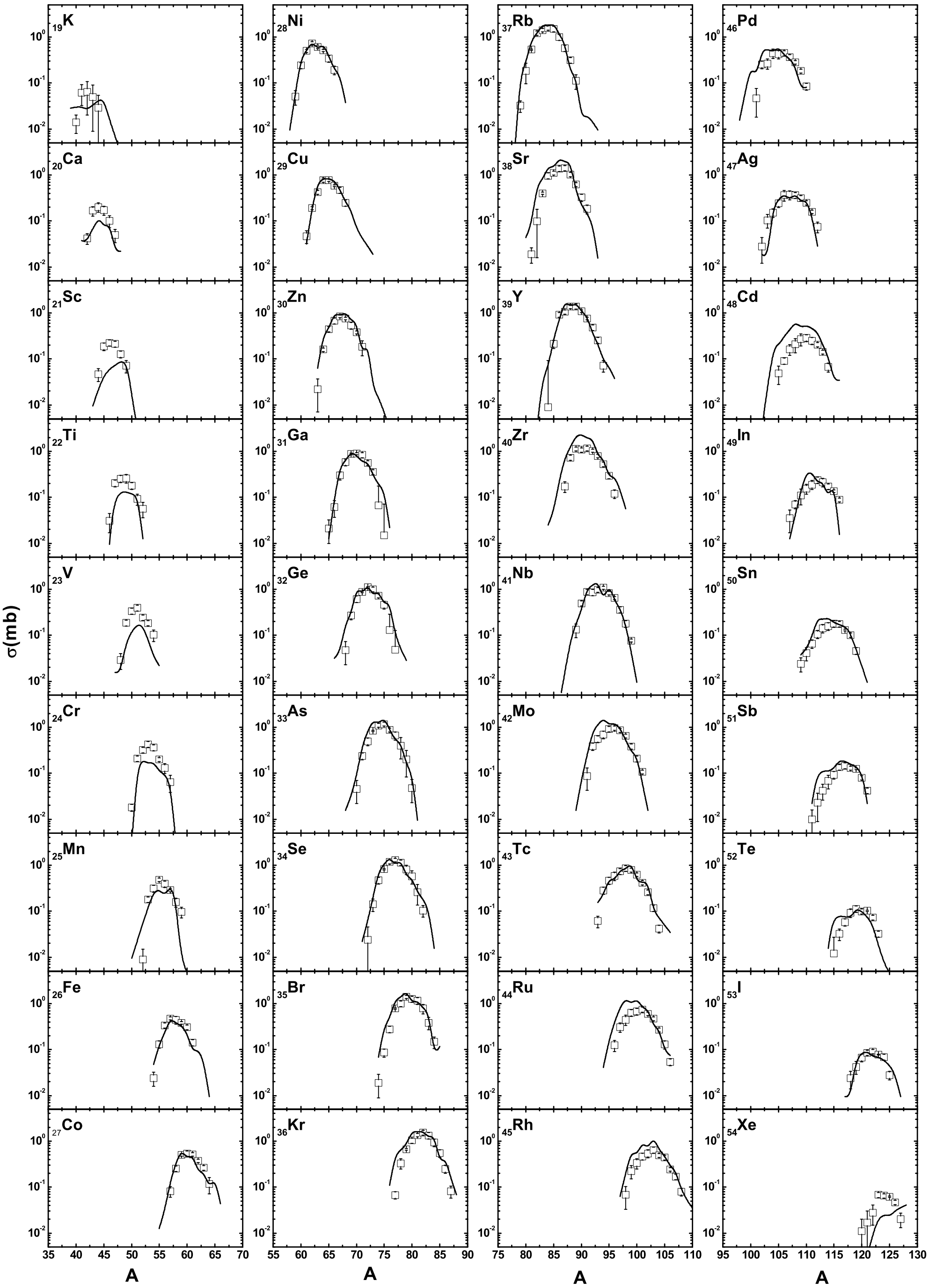


**Figure 5.** Comparison of all measured cross sections of fission products (open squares) from the reaction of 800 MeV proton on $^{197}$Au [32, 33] with ImQMD05+GEM2 calculation results (lines).

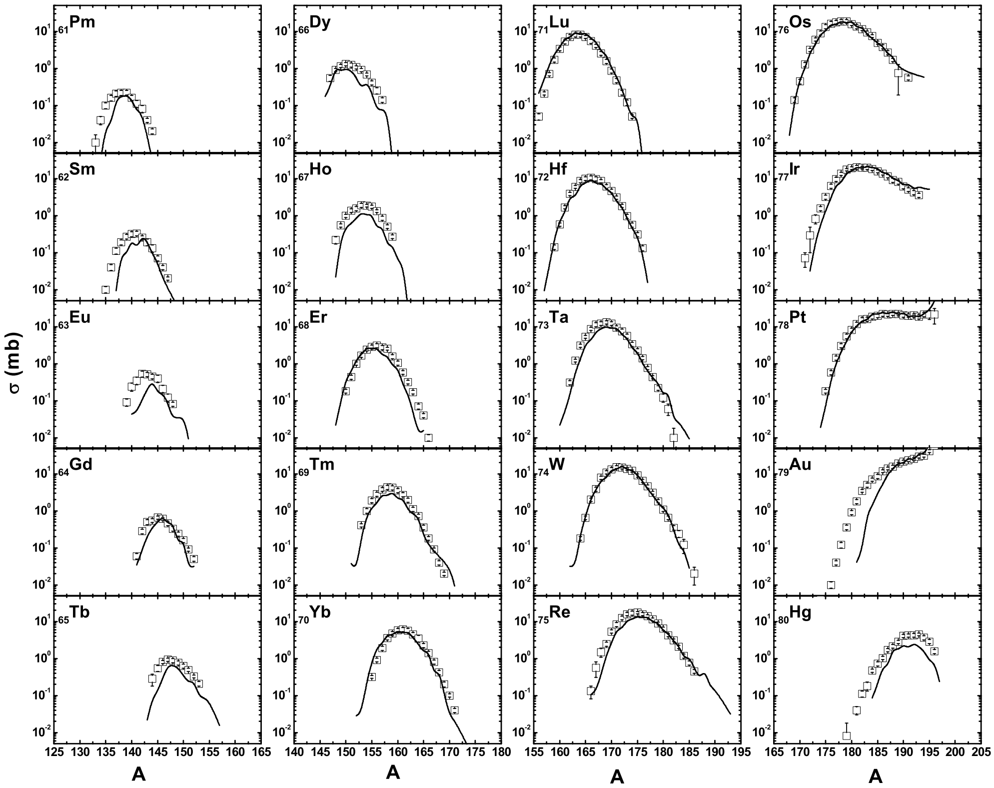

**Figure 6.** The same as figure 5 but for the spallation products. The experimental data are taken from [32, 33].

## 3. Calculation results

Figure 3 presents the comparison between the calculation results with ImQMD05+GEM2 and ImQMD05+GEMINI and the experimental data for the reactions of 500 MeV p+$^{208}$Pb [34, 35], 800 MeV p+$^{197}$Au [32, 33] and 1 GeV p+$^{238}$U [31], respectively. The reaction of p+$^{238}$U is only calculated with the ImQMD05+GEM2 because the current version of GEMINI does not work well for actinides. One can see from the figures that the calculation results with both ImQMD05+GEM2 and ImQMD05+GEMINI with modified parameters reproduce the experimental data quite well. Since in the GEM2 model, the parameters $r_0$ and $f_f$ for evaporation and fission products can be adjusted independently, it is relatively easier to handle. It seems to us that the value of the parameter $r_0$ should have some relation with the excitation energy of the residue and therefore should also have some relation with the incident energy. From the present study (only with three different incident energies) we find that there is a clear tendency that the parameter $r_0$ of the best fitting to the experimental data increases with incident energy monotonically. From proton energy $E_{\mathrm{p}} = 500$ MeV to 1000 MeV the value of $r_0$ increases almost linearly from 1.7 to 1.9 and the increasing slope is about 0.04 per 100 MeV. Following more precise experimental data becoming available, we can extract the more exact relationship between $r_0$ and excitation energy of residue (or incident energy) within this approach. And we find that the reduced factor $f_f$ is not very sensitive to the incident energy studied here. For the GEMINI model, the parameters of $t\_delay = 20$ and

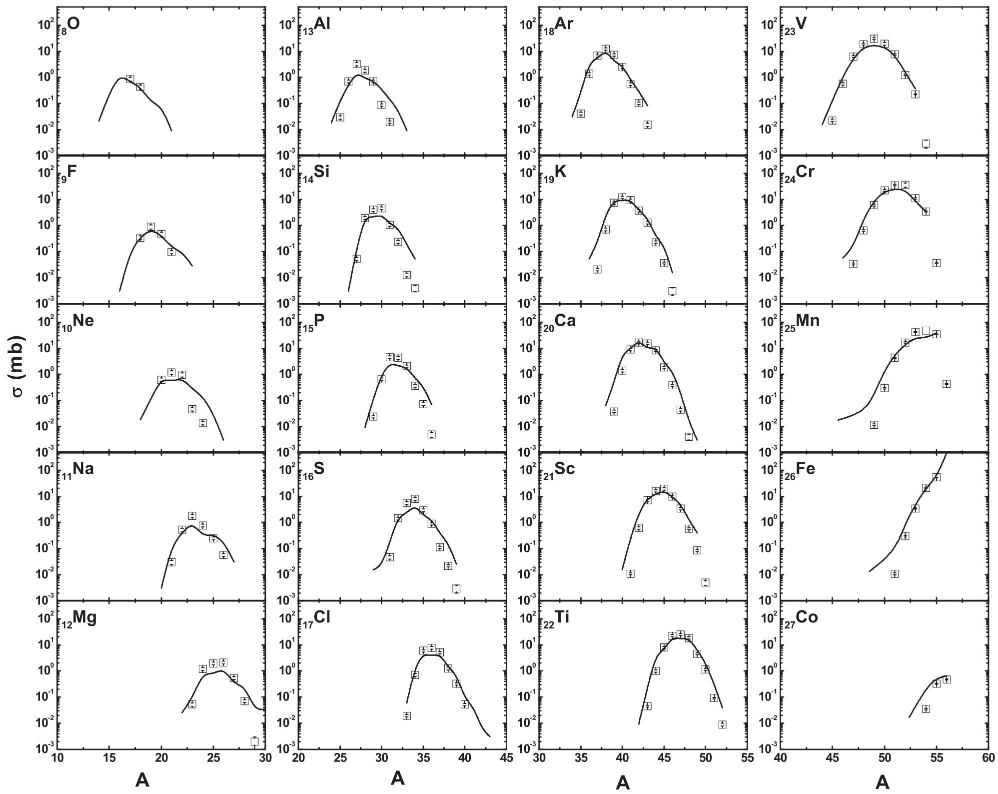


**Figure 7.** Comparison of all measured cross sections of products (open squares) from the reaction 750 MeV proton on $^{56}$Fe [36] with ImQMD05+GEMINI calculation results (lines).

$sig\_delay = 1.0$ are common ones for both fission and evaporation products and for various targets and energies studied in this work. However, the fission fragment distributions given by GEMINI are a little too broad and are shifted toward the neutron-rich side compared to experimental data.

Figure 4 presents the comparison between the calculation results with different models and the experimental data [36] for 300, 500, 750 and 1000 MeV p+$^{56}$Fe, respectively. From the comparison one can see that the GEMINI model describes the cross sections for products in proton-induced reactions on light targets quite well, but the GEM2 model cannot reproduce the yields of elements with $Z \leqslant 16$ even after readjustment of parameters.

The isotope distributions of residues produced in the reactions are also analyzed. All calculation results are found to be in nice agreement with experimental data. Here we only present the results for the reactions of 800 MeV proton on $^{197}$Au with ImQMD05+GEM2, and 750 MeV proton on $^{56}$Fe with ImQMD05+GEMINI, as examples. Figure 5 is for fission products and figure 6 is for spallation products in 800 MeV proton on $^{197}$Au, respectively. Figure 7 is for the reaction of 750 MeV proton on $^{56}$Fe.

Moreover, the double differential cross sections for emitted neutrons in spallation reactions of 113, 256, 597 and 800 MeV proton on targets $^{16}$O, $^{27}$Al, $^{56}$Fe and $^{208}$Pb studied by the ImQMD05 model plus the statistical decay model (SDM) in [28], are also recalculated by the ImQMD05+GEM2 model and the ImQMD05+GEMINI model. We find that the ImQMD05 model plus GEM2 or GEMINI model can reproduce the data equally well. That means that

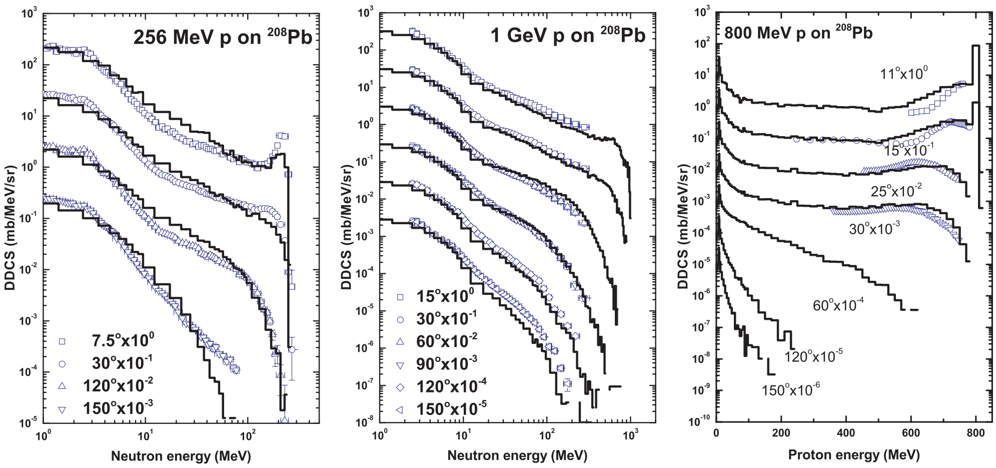


**Figure 8.** Comparison of ImQMD05+GEM2 calculation results (lines) with experimental data (open symbols) for double differential cross sections of emitted neutrons in 256 [47] and 1000 MeV [48] proton on $^{208}$Pb and emitted protons in 800 MeV proton on $^{208}$Pb [49] respectively.

the double differential cross sections of emitted protons and neutrons are not so sensitive to the parameters used in the merged statistical models. As an example, in figure 8 we show the comparison between the calculated results with the ImQMD05+GEM2 model and measurements for the double differential cross sections for emitted neutrons in the reactions of 256 [47] and 1000 MeV [48] proton on $^{208}$Pb and emitted protons in the reaction of 800 MeV proton on $^{208}$Pb [49].

In general, from the above investigation, one sees that the calculation results with the ImQMD05 model plus both the GEM2 and GEMINI models can reproduce the experimental data very well, and there are no fitting parameters in the calculations of the ImQMD05 model, which clearly demonstrates that the ImQMD05 model is very useful for studying proton-induced spallation reactions.

## 4. Summary and conclusion

We have analyzed the proton-induced spallation reactions with the ImQMD05 model merging the GEM2 and GEMINI models, respectively. By readjusting two respective parameters in GEM2 and GEMINI, the cross sections for products in proton-induced reactions on heavy targets can be reproduced quite well by both models, especially by the ImQMD+GEM2 model. However, the GEM2 model is not very competent to describe the reactions on light targets compared with the GEMINI model which can describe these reactions quite well. Furthermore, the double differential cross sections of emitted protons and neutrons in intermediate energy proton-induced spallation reactions are reproduced well by the ImQMD model plus statistical models, we find that they are not so sensitive to the merged statistical models. For the future, with more precise experimental data becoming available, we expect that the systematic adjustable parameters in GEM2 and GEMINI can be obtained with the present approach. However, there is still some work to be done in order to achieve a universal description for spallation reactions with arbitrary targets and arbitrary incident energy.

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