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Production of Z + 1 and A + 1 isotopes in proton-induced reactions on ${}^{A}Z$ nuclei

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Abstract

It is pointed out that the production of certain isotopes with mass and charge close to the target ones can be attributed to specific channels and sometimes to specific mechanisms in terms of binary collisions. Among the most illustrative cases is the production of A + 1 isotopes, which can be obtained through (p, π) channels only. It is indicated that, if the reaction proceeds through incoherent collisions, the production mechanism corresponds to a single inelastic scattering, with very special kinematical constraints, inhibiting further interaction. Other isotopes are also identified, which can only be produced with a concomitant pion. The contribution of pion-producing channels is also underlined for the production of Z + 1 isotopes, although these channels are not exclusive in this case. The description of the production of all these isotopes can be considered as a good testing ground for intranuclear cascades concerning the treatment of the pionic degrees of freedom. Data for the proton-induced reactions on ²⁰⁸Pb and ²⁰⁹Bi are compared with the predictions of the recent version of the INCL4 model, which has especially been improved in the pion sector. Good agreement is generally obtained. This shows that the (p, π) reactions are satisfactorily described by our model, leaving little room for the contribution of a coherent process. It is however stressed that the agreement is obtained owing to the energy-dependence of the nucleon mean field, which is inspired from the one of the real part of the phenomenological optical-model potential. Arguments are given to indicate why this energy-dependence shows up in these special reactions and not in most of the other observables. © 2009 Elsevier B.V. All rights reserved.

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1. Introduction

Most of the isotopes produced in spallation reactions induced by nucleons in the GeV range have a mass and a charge smaller and sometimes sizably smaller than the original mass and charge of the target nucleus, respectively. This is consistent with the current models used to describe these reactions, namely the intranuclear cascade + evaporation or the cascade + preequilibrium + evaporation models. According to these models, most of the time, the incident nucleon expels a few energetic nucleons from the target nucleus, leaving the latter in a moderately excited state, which thereafter emits other (slow) nucleons by pre-equilibrium processes and evaporation. However, occasionally, the mass of the so-called residue exceeds the target mass number, by one mass unit. This is possible when the incident nucleon is absorbed and when a non-baryonic particle is emitted. At low energy, the latter can be a photon. As incident energy increases, the radiative capture cross section goes down. At sufficiently high incident energy, the emitted particle can be a pion (or another meson at still higher energy). This (p, π) process is quite infrequent, but it has been measured, with a cross section reaching a few microbarns for heavy targets. In the following, we will restrict the discussion to pionic channels.

Residues with a charge one unit larger than the target charge can be produced by conventional (p, xn) reactions, except when their mass number exceeds the target mass number by one unit, in which case a (p, π^0) process is at work. An interesting experimental study of the Bi isotopes produced in $p - {}^{208}$ Pb collisions at 1 GeV has been published in Ref. [1]. The authors were not able to measure the 209 Bi isotope which can be produced by the (p, π^0) reaction only.

Some other isotopes can be produced only with an accompanying pion in the final state. This is illustrated in Fig. 1. For instance, isotopes with two extra charges compared to the target one are produced through (p, π^-) or $(p, xn\pi^-)$ reactions. Similarly, isotopes with an extra neutron can be produced only with the concomitant emission of a positive pion. Summarizing, some isotopes at the boarder of the allowed region in the (N, Z) plane can be produced owing to pion production only. Cross sections are in general small because the emission of nucleons should be avoided or very limited. Fig. 1 also suggests that pion production can also contribute significantly to the formation of isotopes which are lying close to the border of the allowed domain, in addition to the conventional mechanism (emission of nucleons).

This set of reactions are quite interesting, because they involve the emission of a single pion or of a pion with a very low number of nucleons. Several interesting questions are then raised. The bulk of the spallation reactions is well described by the intranuclear cascade + pre-equilibrium + evaporation model, which pictures the first stage of the reaction process as a sequence of binary collisions followed by the pre-equilibrium and evaporation stages, that involves also incoherent processes. Can this model be extended to the production of A + 1 residues or is the accompanying pion produced by another mechanism? In other words, can the production of A + 1 residues be described by incoherent processes, as embodied by the INC model, or is it the result of a coherent process? This question has been raised occasionally in the past, in particular for light targets [2,3].

Other interesting issues are linked with (p, p) or (p, n) reactions. These reactions leave a specific fingerprint in the neutron or proton spectra at forward angles under the form of a socalled quasi-elastic peak at large nucleon energy. This peak is usually interpreted as arising from a single collision between the incident nucleon and a nucleon of the target, emitting one of these nucleons without any further interaction. Paradoxically, INC models generally underestimate the size and the width of these quasi-elastic peaks while reproducing rather well the remaining part of the nucleon spectra, arising likely from several nucleon–nucleon collisions. This is par-



Fig. 1. Schematic representation in the (N, Z) plane of the residues neighbouring the target nucleus ${}^{A}Z$ (corresponding to the square with the bold contour) with indications of the main reactions leading to these residues. Colour squares correspond to residues that can be produced owing to pion production only. A + 1 residues are depicted by the yellow squares. Pink squares correspond to other Z + 2 (top) and N + 1 (right) residues. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

ticularly the case for the neutron spectra in proton-induced reactions [4]. Although production of A-residues is not strictly limited to the one-collision mechanism, it is interesting to know whether INC models suffer from the same default for the prediction of the isotope production rate. In Ref. [1], the authors were able to measure the velocity of the ²⁰⁸Bi residues (in reverse kinematics) in $p + {}^{208}$ Pb systems and identify the excitation of Δ resonance resulting from a quasi-inelastic process. This process can also contribute to the production of residues of mass A, but is accompanied by the emission of a pion, as illustrated in Fig. 1. It is interesting to know what is the importance of the quasi-inelastic process to the A-residue formation and to know whether INC models can reproduce it.

These considerations show that the production of pions plays an important (if not crucial) role for the production of A + 1 and of A residues. It is expected that pion production may still play a role for slightly lower mass residues, diminishing progressively as the mass loss is increasing. Production of residues close to the target thus provides with a good test for the pion dynamics of INC models. We have recently [5] improved this feature in the INCL4 model [4], which has been shown to give, when coupled with the ABLA evaporation-fission code [6,7], a fairly good description of a large amount of data concerning proton-induced spallation reactions in the 200 MeV–2 GeV range [4,8–13]. In this paper, we want to apply our INCL4 model, modified according to Ref. [5] and to other further investigations (see Section 2), and test its predictions for residue production close to the target mass region. Occasionally, we will compare our predictions for more extended residue mass regions. Our work is also motivated by radio-protection problems posed by the production of Po isotopes in Pb or Pb–Bi spallation sources. The investigation of this issue will be the object of a separate publication [14].

The paper is organized as follows. In Section 2, we briefly describe the standard INCL4 model, as well as the improved version used in this work. We compare in Section 3 the predictions of the INCL4 model, coupled to the ABLA evaporation code, for residue production to the experimental data concerning proton-induced reactions on ²⁰⁹Bi. Section 4 is devoted proton-induced reactions on ²⁰⁸Pb. Section 5 contains a discussion of our results. Finally, we give our conclusion in Section 6.

2. A brief description of the INCL4 model

We refer to Ref. [4] for a detailed description of the standard INCL4 model. It is sufficient here to remind the salient features. The INCL model is a time-like intranuclear cascade model. In the initial state, all nucleons are prepared in phase space. Target nucleons are given positions and momenta at random in agreement with a Saxon–Woods and a Fermi sphere distributions, respectively. They are moving in a potential well, describing the nuclear mean field. The incident nucleon is given the appropriate energy and an impact parameter at random. All nucleons are then set into motion and followed in space–time. They are assumed to travel along straight-line trajectories until two of them reach their minimum relative distance of approach or until a particle hits the nuclear surface. In the first case, the two nucleons can scatter if the relative distance is shorter than the square root of the total particle–particle reaction cross section (at the appropriate energy) divided by π . The outgoing momenta are then chosen at random in accordance with the experimental angular distributions and with the energy–momentum conservation law. In the second case, nucleons are transmitted or reflected, according to their energy and transmission probabilities for plane waves on a potential step. After the possible modification of the motion is applied, straight line motion is resumed until a new possibility occurs, and so on.

Although classical in nature, the model accounts for some quantum aspects: Pauli blocking of collisions, quantum transmission through the nuclear surface, stochastic determination of the final states in NN collisions and existence of a mean field. Finally, we want to stress that the model does not include free parameters. There are, of course, parameters such as those characterizing the initial distribution or those entering the procedure for evaluation of the phase space occupancy, but they have been determined once for all. There is no adjustable parameter left to the user.

Although the standard INCL4 model is quite successful [4], it has been improved on several points during the last years. We refer to Ref. [15] for a general discussion. The improved version used in this work brought modifications on three points:

- (i) The introduction of an isospin- and energy-dependent mean field for nucleons, as described in Ref. [16]. The value of the potential depth closely follows the phenomenology of the real part of the nucleon optical-model potential.
- (ii) The introduction of nuclear and Coulomb potentials for pions, as described in Ref. [5]; here also the numerical values are largely consistent with the phenomenology of the pion optical model. Additional minor changes, also quoted in the same reference, deal with pion– nucleon cross sections (the parametrization has been extended up to 5 GeV) and the mass of the Δ -resonances (a phase space factor is introduced in the mass distribution for very low masses).
- (iii) A strict Pauli blocking is applied to the first collision.

The last point requires some explanation. In the standard INCL4 model, Pauli blocking of nucleon–nucleon collisions is implemented on a statistical basis: phase space density around the final nucleon states is evaluated by counting nucleons in the neighbourhood of the representative points of the nucleons and the collision is accepted or avoided according to the probability given by the estimated blocking factors. This procedure allows to track the effects of the depletion of the Fermi sea as the collision process develops. However, since the initial Fermi sea in any particular event is represented by point particles taken at random, "holes" may be present in the (phase space) Fermi sea. The importance of these holes diminishes with the evolution of the collision process, but they allow sometimes collisions that would be forbidden by a continuous uniform Fermi sea and reducing the effect of the holes, is obtained when a so-called strict Pauli blocking (i.e. accepting only collisions with final momenta above the Fermi momentum) is applied to the first collision.

As far as numerical codes are concerned, the standard INCL4 model is embodied in the INCL4.2 code and the improved version used in this work is sometimes referred as 4.4.3 in the specialized literature.²

The INCL4 model, like other INC models, should be supplemented by an evaporation model. An original feature of the INCL4 model is that the stopping time, i.e. the time at which the cascade process is stopped to give place to evaporation is determined self-consistently, as explained in Ref. [4]. In the following we will use the ABLA code as the evaporation code to be coupled to INCL4 (actually the so-called KHSv3p version, see Ref. [4] for detail). The ABLA model is described in Refs. [6,7]. We will here limit ourselves to recall that light particles are emitted according the Weisskopf–Ewing model with rather conventional parameters for level density, Coulomb barrier and inverse cross sections. The ABLA model contains a sophisticated fission module, where fission fragmentation is based on microscopic potential energy surfaces and where transient effects are introduced, on the basis of the Fokker–Planck approach for the fission process.

3. Residue production in proton-induced reactions on ²⁰⁹Bi

3.1. The A + 1 case

An example is provided by the measurement of the cross section for ²¹⁰Po production in $p + {}^{209}$ Bi reactions, as illustrated in the first panel of Fig. 2. The latter production is possible at low energy (indicated by the open triangles) by the (p, γ) reaction. The production cross section decreases regularly as the incident energy is increasing, but rises when the latter reaches ~180 MeV, indicating the opening of the (p, π^0) reaction. We remind that the effective threshold energy is much lower than the threshold energy in free space nucleon–nucleon kinematics (~280 MeV), due to Fermi motion. The detail of this region is given in Fig. 3. It can be seen that the standard version of INCL largely overestimates the cross section below 400 MeV, whereas our modified version gives a more satisfactory account of the cross section, although it has a tendency to underestimate it. In the same figure are given the relative importance of the modifications to the standard INCL model brought in this work. One can see that the introduction of

 $^{^2}$ Actually, the version 4.4.3, as first defined in Ref. [18], does not include the application of the strict Pauli blocking on the first collision.



Fig. 2. Cross sections for production of various Po isotopes (indicated in each panel) in $p + {}^{209}$ Bi reactions, as functions of the kinetic energy T_p of the incident proton. Black or open symbols correspond to experimental data of Refs. [19–37]. Blue crosses give the predictions of the standard INCL4 model and red squares give those of our modified model. Note that all cross sections for 210 Po have been multiplied by 100. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

an isospin-dependence of the potential depth for nucleons (results indicated by the green curve in Fig. 3) does not bring an important modification. On the contrary, the further addition of the energy-dependence for this potential (blue curve) yields an important reduction of the cross section. The subsequent introduction of an average potential for the pions (indicated by the purple curve) does not really change the results. Finally, the introduction of a strict Pauli blocking for the first collision (yielding the red curve) brings an additional reduction.

Let us try to interpret these results. First of all, one has to realize that the (p, π^0) reaction requires some special conditions. Indeed, in our model, the production of a pion goes through the $NN \rightarrow N\Delta$, $\Delta \rightarrow \pi N$ sequence. In addition, the pion should escape further interaction and the two involved nucleons should acquire an energy which is just above the Fermi energy. Otherwise, these nucleons will bring too much excitation energy by further interaction or escape from the target nucleus, leading so to other final channels. We checked that these conditions can indeed be realized provided the incident nucleon strikes a nucleon with a momentum close to the Fermi momentum but oriented approximately in the direction opposite to the one of the incident proton and provided the Δ particle is formed with a light mass.³ In other words, the (p, π^0)

³ In our model the Δ resonance is assigned a definite mass which is taken at random according to a Breit–Wigner distribution. See Ref. [4] for detail.



Fig. 3. Cross section for production of ²¹⁰Po isotopes in $p + {}^{209}$ Bi reactions, as functions of the kinetic energy T_p of the incident proton. Black symbols correspond to the experimental data of Ref. [34], after removal of the (p, γ) background. The theoretical curves correspond to various versions of the INCL4 model, with the standard version in black dashes and the 4.4.3 version in red. See text for the meaning of the other curves. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

reaction requires the sequence of reactions indicated above with special kinematics and with no further interaction, leaving the target almost unexcited. It is then understandable that the cross section is very small. Given this mechanism, it is also understandable that the existence of an energy-dependent mean field for the nucleons has an important impact on the results. Indeed, during nucleon–nucleon collisions, the conservation of energy implies kinetic (E) as well as potential (V(E)) energies and can thus be written:

$$E_1 + V(E_1) + E_2 + V(E_2) = E'_1 + V(E'_1) + E'_2 + V(E'_2) + (\Delta m)c^2,$$
(1)

where the last term accounts for the possible difference of masses (in inelastic collisions). When the average potential does not depend upon the energy, the potential terms cancel out and collisions proceed as in free space. The energy-dependent potential that we use closely follows the phenomenology of the real part of the optical-model potential [5]. It has a depth of ~ 50 MeV around the Fermi level, and decreases roughly linearly when the energy increases until $E \sim 200$ MeV, where it basically vanishes, as well as at higher energies. Let us consider the kinematics required for a (p, π^0) reaction, as discussed above. In an inelastic collision, producing a Δ -isobar, the kinetic energy in the final state is reduced compared to the initial state. The use of the energy-dependent potential introduces in Eq. (1), a mismatch of ~ 50 MeV between the initial and final potential energies. This is roughly equivalent to having a collision with a negative O-value of the same size (in addition to the mass difference).⁴ As a consequence, the cross section for the (p, π^0) reaction is sizably reduced, as shown in Fig. 3. The other curves in this figure are also easy to explain. For the special conditions required for (p, π^0) reactions, it can be shown that the pion issued from the Δ -decay has always an important momentum, with respect to the target. Therefore the presence of rather shallow pion potential (around 20-30 MeV, see Ref. [5]) does not make a real difference. Often, the effect of the pion potential is compensated

⁴ Reasoning on a NN \rightarrow NN π process leads to the same conclusion.

by the one of the isospin-dependence of the nucleon average potential (see Ref. [5] for more explanation). On the contrary, since it is required that the two involved nucleons be lying around the Fermi level in the final state, the use of a strict Pauli blocking, compared to our standard statistical implementation, can make a non-negligible difference.

It is interesting to note that the influence of the energy-dependence of the average nucleon potential is particularly visible in the kind of reactions investigated here. In Ref. [16], where we introduced such a potential for the first time, we stated that, for most observables, the effect of this introduction was quite small. Of course, many of the observables imply processes with several unconstrained collisions, where the mismatch of potential energy prior and after collisions is small (and probably fluctuating from collision to collision). In the same reference, we mentioned that the effect of the energy-dependence is the most effective on the quasi-elastic peak, which basically demands a single scattering. It is therefore not surprising that the effect is even more visible on (p, π^0) reactions, which require a single (inelastic) scattering with stronger conditions on the final state.

Our results and discussion indicate that the (p, π^0) reaction is consistent with the incoherent process picture. The underprediction yielded by our fully modified model leaves some room to a possible contribution of coherent process. The dominance of the incoherent processes was already suggested in Ref. [34], where a rather crude model was used, using an unsatisfactory level density parameter and assuming somehow arbitrarily excitations to particle unbound states up to 10 MeV. Our conclusion is in keeping with the results of previous works [19,38–40], but the effect of the energy-dependent nucleon potential was not investigated before our work.

The fact that the experimental cross section remains sizable well above the threshold is coming mainly from the Fermi motion. There is some discussion in Ref. [34] about the apparent increase of the cross section above 400 MeV, which departs from the trend of INCL4. This may be attributable to a decreasing absorption of the fast pions, which is not properly accounted for in our model.

3.2. The Z + 1 case

We consider first the production of ²⁰⁹Po and ²⁰⁸Po in $p + ^{209}$ Bi reactions, i.e. the production of the heaviest Z + 1 isotopes not corresponding to A + 1. Experimental data are displayed in Figs. 2 and 4. The large cross sections (compared to the ²¹⁰Po case indicate that the production of these isotopes is dominated by the usual (p, n) and (p, 2n) processes. We compare with the predictions of INCL4 in the same figures. One can see that the theoretical results are considerably improved by the modifications brought to the standard version. This is particularly spectacular for the production of ²⁰⁹Po, which also reflects on the ²⁰⁹Po/²⁰⁸Po ratio.

It is interesting to discuss the effect of the various new ingredients of the INCL4 model, in connection with the dominant reaction mechanisms in this model. Production of ²⁰⁹Po has to proceed primarily through the (p, n) channel. In the cascade approach, this production essentially corresponds to a single charge exchange collision, but leading to a small excitation energy (smaller than nucleon separation energy). Production of ²⁰⁸Po is slightly more complicated. It is due either to a single charge exchange collision (with emission of the neutron) leading to excitation energy sufficient for evaporating one more neutron but too small for allowing the evaporation of two neutrons, or to two collisions, one elastic and the other charge exchange, each of them emitting a neutron in the cascade stage. One of these collisions may be a hard collision, but then the hit nucleon should leave the nucleus without depositing much excitation energy. The other collision should allow the emission of a neutron while the remaining excitation energy is below



Fig. 4. Production cross sections of ²⁰⁸Po (upper left) and of ²⁰⁹Po (upper right) isotopes in proton-induced reactions on ²⁰⁹Bi, as a function of the incident proton kinetic energy T_p . Black symbols correspond to the experimental data taken from Refs. [19,22,24,33,34,36]. The theoretical curves correspond to various versions of the INCL4 model, with the standard version in black and the 4.4.3 version in red. See text for the meaning of the other curves. The lower panel gives the comparison for the ²⁰⁹Po/²⁰⁸Po ratio, with the same convention. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the emission threshold. Let us now discuss the effects of the new ingredients. The introduction of an isospin-dependent potential (from black to green curves, in Fig. 4) does bring a rather moderate modification of the results, but larger for the ²⁰⁹Po case. This is in keeping with the fact that the production of this isotope is basically due to a single (p, n) scattering and the observation made in Ref. [16] concerning the effect of an isospin-dependent mean field on the quasi-elastic peak in neutron spectra. The introduction of an energy-dependent average potential makes no difference, in agreement with our arguments above (in the first elastic scattering initiated by the incoming nucleon, the momentum transfer is generally small; consequently, the kinetic energy remains the same, as well as the potential energy). Pion average potential does not bring any modification since the pion channels are not contributing much (see below for a discussion). The last new ingredient, strict Pauli blocking on the first collision, makes a sizable reduction of the cross sections, more important for the ²⁰⁹Po case. This is consistent with the importance of the first collision, being the only one, for this case.

The relative importance of purely nucleonic and pion-producing channels for the production of 208 Po and 209 Po isotopes, as predicted by INCL4, is illustrated in Fig. 5. One can see that, in the last case, above the effective threshold, the $(p, n\pi^0)$ channel contribution is about one order



Fig. 5. Upper panel: contributions of the (p, 2n), $(p, 2n\pi^0)$ and $(p, pn\pi^-)$ channels to the production of ²⁰⁸Po in proton-induced reactions on ²⁰⁹Bi, as predicted by INCL4 (black dashes for the standard version and red curves for the improved version). Lower panel: same for the contributions of the (p, n), $(p, n\pi^0)$ and $(p, p\pi^-)$ channels to the production of ²⁰⁹Po. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

of magnitude smaller than for the (p, n) channel one. Again, this is explained by arguments similar to those produced above. The production a ²⁰⁹Po isotope through the $(p, n\pi^0)$ channel requires the same $NN \rightarrow N\Delta$, $\Delta \rightarrow \pi N$ sequence, as for the production of ²¹⁰Po, but, this time, only one of the two final nucleons is required to have a final momentum close to the Fermi momentum. Compared to the ²¹⁰Po case, the inelastic single scattering is much less constrained. Accordingly, the production cross section is much larger: a few tenths of millibarns compared to a few microbarns. It is nevertheless interesting to see that the (p, n) process, corresponding to a single elastic scattering, give a much larger contribution than the $(p, n\pi^0)$ process, which corresponds to the single inelastic process. This is due to several factors. First, the inelastic



Fig. 6. Cross sections for various channels in proton-induced reactions on 209 Bi, as predicted by INCL4 (standard version in black and the 4.4.3 version in red; see text for the meaning of the other curves). Note that some cross sections have been multiplied by an indicated factor in order to keep the same vertical scale for all panels. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

nucleon-nucleon cross section is smaller than the elastic one in the energy range considered here. Second, isospin conservation selects only some of the final pion channels. Third and more importantly, the constraint of a small excitation energy is stronger for the second channel than for the first one, since the creation of a Δ is an efficient process for stopping and since the created pion should avoid further interaction. Concerning the production of ²⁰⁸Po (upper part of Fig. 5), the relative importance of (p, 2n) and $(p, 2n\pi^0)$ contributions can be understood at the light of the discussion above. The $(p, pn\pi^-)$ contribution, not shown in Fig. 5, has roughly the same shape as the $(p, 2n\pi^0)$ contribution and is roughly a factor 2 smaller.

The production of other Z + 1 isotopes is investigated in Fig. 2. One can see that both the standard and the modified version of INCL4 reproduce rather well the experimental cross sections, except for details near threshold for ²⁰³Po. There is not very much difference between the standard and the modified versions of INCL, for the series of isotopes lighter than ²⁰⁸Po. This is understandable on the basis of the discussion above. The modifications that we brought in are expected to bring significant changes if single scattering is important, which is not really the case for the production of these isotopes.

Some of the considerations above are also illustrated by Fig. 6, which depicts the calculated cross sections for various channels. We have already commented on (p, π) channels and on

the (p, xn) channels, where the difference in the cross sections of the standard and modified versions, and the effect of the strict Pauli blocking on the first collision diminish as the neutron loss increases. Concerning the $(p, xn\pi^0)$ channels, the different modifications bring effects of similar size. One can recognize that the introduction of an energy-dependent mean field for the nucleons has the most important influence at low energy. The effect of the strict Pauli blocking on the first collision has a meaningful effect for the $(p, n\pi^0)$ channel only. Similar considerations can be made concerning the contributions of the channels with a negative pion.

Let us comment of the shape of the excitation functions. All (p, xn) and (p, π) cross sections are decreasing with the incident energy. This is due to the competition with the increasing number of other channels essentially. On the other hand, the $(p, xn\pi)$ excitation functions are displaying a plateau in the energy range of interest. In our opinion, this results from a combination of the competition of other channels (which tends to decrease the cross sections) and the fact that as energy increases the kinematical constraint discussed above are less and less effective as energy increases (which tends to increase the cross sections).

3.3. The Z + 2 case

This refers to the production of Astatine isotopes for the $p + {}^{209}$ Bi system under interest. As illustrated in Fig. 1, the production of these isotopes is only possible if a negative pion is emitted, possibly accompanied with neutrons. The generic features of our predictions are displayed in Fig. 6. A comparison with available experimental data will be the subject of a forthcoming separate publication. It is nevertheless worthwhile to comment on our results. The (p, π^{-}) process, leading to the A + 1 isotope, has a very small cross section, comparable with the (p, π^{0}) one, the ratio of the two being compatible with isospin symmetry. As the latter, it is sensitive to the energy-dependence of the nucleon mean field. The $(p, xn\pi^{-})$, leading to lighter At isotopes have larger and larger cross sections with increasing number of neutrons. These cross sections, at least for x = 1, are also sensitive to the implementation of the Pauli blocking on the first collision.

3.4. The other isotopes

Although it is not in the main stream of this paper, we display the predictions of our model for the production of some other typical lighter isotopes in Fig. 7. There is a general agreement between the predictions of both versions of INCL4 and the experimental data. When they differ, predictions of the modified version are often better than those of the standard version (see the heaviest Bi isotopes). Surprisingly there are noticeable disagreement for some of fission isotopes (mainly given in the lower panel of Fig. 7), in the energy domain extending from ~100 MeV to ~800 MeV. This contrasts with our results on the $p + {}^{208}\text{Pb}$ system at 1 GeV, for which a good agreement for all the fission isotopes is obtained (see Ref. [4] for detail).

4. Residue production in proton-induced reactions on ²⁰⁸Pb

4.1. The Z + 1 case

We concentrate first on the measurements of Ref. [1] concerning the production of Bi isotopes at 1 GeV. They offer a good systematics with good precision, as they are obtained by the inverse kinematics method. They are shown in Fig. 8 and compared with our predictions. One can see that our modified version considerably improves the results for the heaviest isotopes. The reason



Fig. 7. Production cross sections of various isotopes in proton-induced reactions on ²⁰⁹Bi, as a function of the incident proton kinetic energy T_p . Black symbols correspond to the experimental data taken from Refs. [19,20,23,24,31,32]. Blue crosses give the predictions of the standard version of the INCL4 model, and red squares give those of the modified version. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



Fig. 8. Production cross sections of Bi isotopes in proton-induced reactions on 208 Pb at 1 GeV, as a function of their mass *A*. Black symbols correspond to the experimental data of Ref. [1]. The dotted line gives the predictions of the standard version of the INCL4 model, and the full line gives those of the modified version.

is that the production of the heaviest isotopes is possible under the kinematical constraints that we have discussed above and which are more properly accounted for with our modifications. Note that the authors of Ref. [1] have not measured the production of ²⁰⁹Bi, corresponding to the (p, π^0) channel. For lighter isotopes (below A = 205), the yield is not changed very much by our modifications. This is in keeping with our arguments above: these nuclei are predominantly formed in events with several collisions that are thus less kinematically constrained than for the heaviest isotopes. The fact that we somehow underestimate the yield for isotopes in A = 200-205range indicates that there are still problems in the 4.4.3 version with the distribution of excitation energy.

Let us comment of the shape of the distribution in Fig. 8. As we indicated in Section 3, the cross section is decreasing for the largest mass numbers because the kinematical constraints are more and more effective. Of course, the cross section is bound to decrease at small mass number for two reasons: the excitation energy is limited and as *A* decreases the neutron evaporation is more and more in competition with proton evaporation (or other charged particle evaporation).

4.2. The other isotopes

The excitation functions of the formation cross sections for several representative isotopes are shown in Fig. 9, along with the predictions of the standard and modified versions of the INCL4 model. Grossly speaking, the predictions are similar for the two versions of the model, except for the two heaviest Bi isotopes, in which case the modified version brings an important improvement. It is perhaps worthwhile to point out that the production of some of the fission isotopes are underpredicted in the ~100 MeV-800 MeV range, as for the $p + {}^{209}$ Bi case. The discrepancy is however larger in the present case. To determine whether this discrepancy is due to the cascade model or the fission-evaporation model requires some further investigations.



Fig. 9. Production cross sections of various isotopes in proton-induced reactions on 208 Pb, as functions of the incident proton kinetic energy T_p . Black symbols correspond to the experimental data of Refs. [19,26,27,29,35,36,41–43]. Red crosses give the predictions of the standard version of the INCL4 model, and blue squares give those of the modified version. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

5. Discussion

Production of A + 1 isotopes in $p + {}^{A}Z$ reactions is interesting because it corresponds solely to the (p, π) reactions, except for the isotopes with one extra charge, for which the (p, γ) process provides an alternative to the (p, π^{0}) process. These two processes are however dominant in different ranges of incident energies: the (p, γ) process at low energy and the (p, π^{0}) process above ~200 MeV. Strictly speaking, other processes like the emission of two pions cannot be ruled out, but it is evident that they would correspond to even smaller cross sections.

It is shown in Section 3 that the production of such isotopes (in $p + {}^{209}\text{Bi}$ reactions) are consistent with our modified version of the INCL4 model. The basic premise of this model is the assumption of independent, well separated collisions. Within such a picture, the (p, π) reaction can almost surely be identified with the following process: a $pN \rightarrow N\Delta$, followed by the decay of the Δ , and the emission of the pion. Of course, as already pointed out, the pion should be emitted and the two involved nucleons should be captured. This can be realized with the following kinematical constraints: the pion and the two involved nucleons should not undergo further collision and the two nucleons should have acquired energies which are lying just above the Fermi energy. Lower values are forbidden by the Pauli blocking and larger values would lead to their emission. The typical range of allowed values is a few MeV broad. Of course, one can imagine that the three particles re-interact with target nucleons, but then the hit nucleons should be subject to the same constraints. This kind of scenario is much less probable.

We have indicated that the rather good agreement with the experimental data (see Figs. 2, 3) is mainly obtained owing to two modifications of our standard model, namely the introduction of an energy-dependent mean field for nucleons and the implementation of a strict Pauli blocking on the first collision. The energy-dependence is chosen in accordance with the phenomenology of the real part of the optical-model potential [44], as mandatory [45]. The substantial influence of this modification on the production of 210 Po in $p + ^{209}$ Bi reactions is to be contrasted with the minor effect it brings to the INC description of the bulk of the spallation data [16]. The explanation is that the energy-dependence is most fully effective in collisions implying one nucleon at high kinetic energy and the other (initial or final) particles at low energy. This is indeed the case in an inelastic collision with the excitation to a Δ resonance. But it is very likely that if such a collision is followed (or preceded) by other collisions, the final effect of the energy-dependence is largely washed out.

The application of a strict Pauli blocking on the first collision may appear as an inconsistency in our model. In Section 2, the procedure for the implementation of the Pauli blocking in our standard model is briefly described. An somehow opposite alternative is provided by the strict Pauli blocking, in which the final momenta of the nucleons, after any collision, should lie above the Fermi momentum. The advantage of the first implementation is that it takes account of the depletion of Fermi sphere in events with several collisions. The disadvantage arises from the presence of unavoidable "holes" in sampling the initial state by point-like objects, which allows for collisions that would be forbidden in an ideal (uniform) Fermi sphere. A reasonable compromise is provided by the implementation of a strict Pauli blocking on the first collision and of a statistical Pauli blocking on the following ones. This option has been shown to give the best results in Refs. [17,46]. It is of course understandable that this modification may bring non-negligible effects on reaction channels that are dominated by single-collision processes such as the production of A + 1 isotopes or, even more, by the (p, n) reactions.

One of the questions we addressed in this paper is to know whether the (p, π) reactions are due to incoherent processes or not. We are not able to give a decisive answer. First our numerical

results do not match experimental results perfectly. Furthermore, even if INC models have continuously improved in the last years, they still rely on the idealization of the physical situation: well-separated collisions, ideal Fermi gas momentum distribution, static mean fields are examples of this idealization. Therefore, our results are still subject to uncertainty. Our experience of the field allows us to believe that this uncertainty can amount to a few tens of percent. Yet, the mere fact that a simplified model such as INCL can give the right order of magnitude for a reaction with a so low branching ratio is already extremely remarkable. A possible contribution of coherent processes is not to be excluded. In view of our results and our discussion, their contribution may be of comparable (though smaller) size as the one of the incoherent processes described here. More studies are needed to clarify this problem.

We want to comment here on a somewhat related question. There is a longstanding debate about the necessity of introducing an intermediate pre-equilibrium module between INC and evaporation [4,47–50]. Our model does not seem to require such a module, whereas, for instance, Mashnik's CEM model does (the introduction of such a module is optional in many particle transport codes). The question remains open. We just want to emphasize that for the production of isotopes very close to the target nucleus, and in particular for A + 1 isotopes, the reaction mechanism reduces to a very small number of rather hard collisions. It is then expected that pre-equilibrium does not play a role, since it departs from INC when the energy of the participants is rather low.

We now consider the production of the ${}^{A}(Z + 1)$ isotopes. They are formed by the simplest process involving nucleons, that can be identified with a quasi-elastic charge exchange single collision. As we have said, the latter is subject to some kinematical constraint in order to lead to the indicated final channel. These isotopes can be formed also through the $(p, n\pi 0)$ or $(p, p\pi -)$ reactions, which can be identified with a single inelastic collision, subject to kinematical constraints, which are however less stringent than for the A + 1 isotopes. At low energy, just above the effective threshold, the contribution of the processes involving pions is an order of magnitude lower the one of the purely nucleonic process. As energy increases, the contribution of the pionic channels remains basically the same and the one of the pure nucleonic process decreases continuously, basically because it is more and more probable that large excitation is injected into the nucleus, favoring so the decay into other channels with extra neutrons. In pionic channels, the formation of a pion limits severely the excitation energy. Consequently, the emission of extra neutrons is less probable.

A surprising result of our analysis is the fact that the ratio of the contribution of the $(p, xn\pi 0)$ channels to the contribution of the (p, xn) channel is not decreasing very fast when going from x = 1 discussed above to x = 2, 3 (see Fig. 6), where it reaches values between 0.5 and unity. This remark applies to high incident energies, around 1 GeV. Of course this ratio is much smaller at low energy due to threshold effects for pionic channels. It would be nice to have direct measurements of these channels to check our results.

Finally, we want to mention that we concentrated our discussion on results concerning heavy targets. However, we obtain similar trends for lighter targets, as explained in Ref. [18].

6. Conclusion

In this paper, we have identified the interest of the production of A + 1 and Z + 1 isotopes in proton-induced reactions on ^AZ nuclei. The A + 1 isotopes can be made through (p, π) reactions only. The physics issue is to know whether this reaction proceeds through coherent or incoherent processes. The ^A(Z + 1) isotope is made by (p, n) reaction mainly. The interest here is to know

whether the $(p, n\pi^0)$ and $(p, p\pi^-)$ channels contribute also to the production of the ${}^A(Z + 1)$ isotope and whether the (p, n) reaction corresponds to a single quasi-elastic scattering. We also pointed out the interest of the production of Z + 2 isotopes, since they automatically involve the production of a negative pion, as well as the interest of the production of N + 1 isotopes, which similarly involves the emission of a positive pion. In summary, for these sometimes infrequent events, the production of a pion is a dominant feature. They provide thus with a good testing ground for the pion dynamics in reaction models.

The main purpose of this paper was precisely to apply our intranuclear cascade model, that we recently improved in the pion sector, to this kind of events. In addition we compared our results to the experimental results of Ref. [34] in order to try to determine whether the (p, π) reaction is due to a coherent process. The third purpose of this work was to see whether the improved version of INCL4 could reproduce also the bulk of isotope production cross sections, not necessarily restricted to small mass losses.

Our results can be summarized as follows:

- (i) Our improved version can reproduce rather well the production of ²¹⁰Po in $p + {}^{209}Bi$ reactions (A + 1 case).
- (ii) The agreement is mainly due to the introduction of the energy-dependence of nucleon average potential and, to a lesser extent, to the implementation of a strict Pauli blocking on the first collision.
- (iii) The production of the A + 1 isotopes is consistent with a picture in terms of incoherent processes; yet, a non-negligible contribution of coherent processes cannot be ruled out in view of our calculations.
- (iv) The smallness of the corresponding cross section is interpreted in terms of kinematical constraints on single collision events.
- (v) The production of ${}^{A}(Z + 1)$ isotopes is mainly due to (p, n) processes. The evaluated contribution of $(p, n\pi)$ channels is about one order of magnitude smaller. These results has been interpreted in terms of single elastic or inelastic collision events.
- (vi) The importance of pionic channels for lighter isotopes has been shown to be non-negligible. It, of course, decreases with the mass of the isotopes.

We think we have clarified the mechanisms of production of special $(A + 1, {}^{A}(Z + 1), Z + 2$ and N + 1) isotopes. For these ones, our method consisting of identifying reactions channels and attaching to these channels particular mechanisms in terms of number (and type) of collisions, seems to be promising. More data especially on production of Z + 2 and N + 1 isotopes are highly desired to check the method, before it can be extended to the production of isotopes with larger mass loss.

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