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Neutron multiplicity measurements in 2 GeV p and ^{3}He induced reactions on thin targets of heavy materials*

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One of the key issues for an efficient transmutation of long-lived wastes -both minor actinides and fission products- is the achievement of high neutron fluxes. Such fluxes are not accessible in current nuclear reactors and it has been proposed to make use of the neutrons generated by an intense beam of 1 to 2 GeV protons impinging on a very thick target of heavy material[1-3]. As it is well known, the many particles (nucleons, composite particles, π and K mesons) emitted in one single nuclear collision act as secondary projectiles and are able to generate in a thick target several tens of neutrons for each initially accelerated proton. It seems therefore crucial to study in great detail the processes responsible for neutron production in order to find out the most economical way for generating the largest number of neutrons, usable for transmutation, from any accelerated proton or light nucleus.

Two complementary approaches have been followed thus far to tackle the neutron production issue. The first one, more fundamental, implies the bombardment of thin targets to allow for a detailed study of all elementary processes which are involved. The neutron measurements for intermediate-energy proton-induced reactions are rather scarce[4-5] and they are essentially aimed at studying the characteristics of energetic neutrons to test the Intra-Nuclear Cascade (INC) step of the reaction[6]. The second approach, of more practical concern, considers very thick targets, similar to those which could be used in the future in genuine transmutation plants. It aims at measuring the average number of neutrons following one initial proton interaction[7]. It can be used to check the validity of those models including both the Intra-Nuclear Cascade and the Transport of the secondary emitted particles[8].

Our approach to the neutron problem is quite different from the previous ones. Instead of considering inclusive observables such as the doubly differential cross sections of any type of emitted particle, charged or neutral, or the distribution of residual nuclei we have tried to characterize each individual measured event as completely as possible. In order to do so, we measure event-wise the number of emitted neutrons and, in addition, the characteristics of light charged particles (p, d, t, He), intermediate-mass fragments (Li, Be, B, C...) and fission fragments emitted in coincidence with these neutrons. All these correlated data are expected to allow more stringent tests on the reaction processes than ordinarily done via inclusive data. Some aspects of this work have been already published elsewhere[9] and we will mostly consider in this contribution those aspects more relevant for the issues linked to transmutation.

A 4π liquid scintillator detector, loaded with gadolinium (ORION II), has been used at SATURNE in order to measure the number of neutrons emitted for every event. The neutron are first thermalized before being captured by the Gd nuclei. The capture time is long enough and broad enough for the simultaneously emitted neutrons (on a time scale of 10^{-18}s) to be seen by the phototubes surrounding the scintillator tank at well separated instants (on a time scale of 10^{-8} s). Thus, the neutrons can be numbered event by event. This type of detectors is well known for its very good detection efficiency for low-energy neutrons [10](Σ>80% for En>20 MeV) but fails to register most of the more energetic neutrons as well exemplified in Fig.1.The energy spectrum of neutrons emitted in a 2 GeV proton interaction with a thin Au target has been simulated using the HETC[11] computer code (solid dots) before being folded by our detector efficiency (open dots). The clear output of these calculations is the strong reduction of registered neutrons arising from the first steps of the INC process (essentially neutrons with more than 20 MeV).

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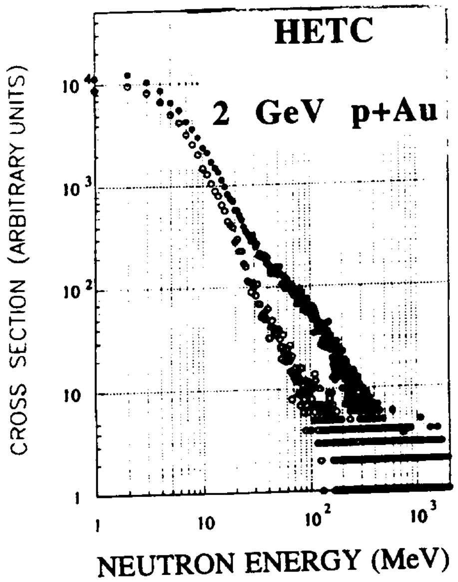

Fig.1 Energy spectrum of all emitted neutrons as simulated with HETC for 2 GeV p+Au (black dots) and after folding by the 4π detector efficiency (open dots)

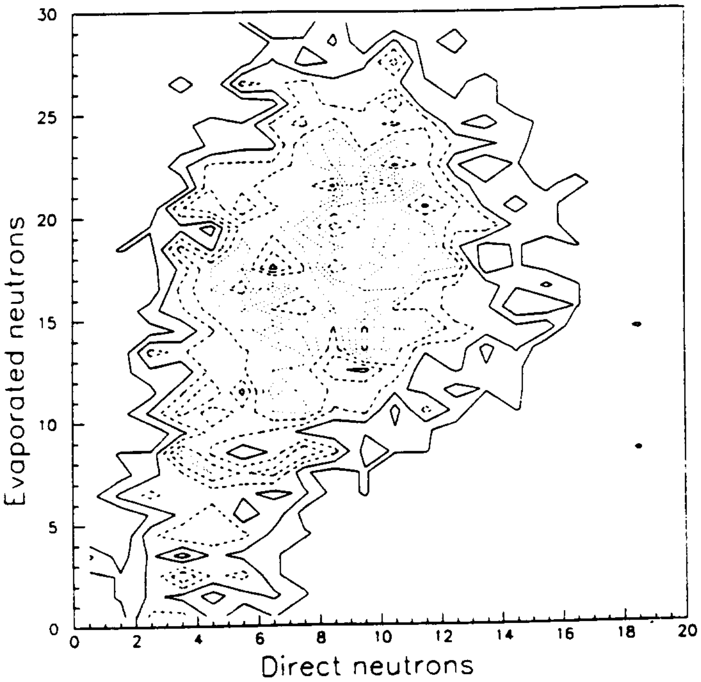

Fig.2 Correlation contours between the number of evaporated neutrons and the number of directly emitted ones (INC step) as simulated for 2 GeV protons on Au.

We have also investigated how this experimental cut-off could bias our perception of the physical processes. Another simulation has thus been run with a different computer code$^{12)}$ which distinguishes those neutrons emitted at high energy in the INC step from those which can be simply considered as evaporated neutrons when the struck nucleus has reached statistical equilibrium. It is shown in Fig.2 that there exists a definite correlation between pre-thermal (direct) neutron and post-thermal (evaporated) neutron multiplicities and that a strong attenuation in the registration of the former component cannot distord severely the apprehension of the whole process. This will be best shown in the following.

The first type of experimental data is shown in Fig.3; the neutron multiplicity distributions are given as obtained after background correction for three types of projectiles and a series of targets$^{9)}$. They all exhibit a similar pattern with a pronounced maximum at low multiplicity resulting from peripheral collisions and a broad bump at high multiplicity arising from a wide range of more central collisions. It can be noticed that, when considering a proton projectile at different energies, the average neutron multiplicity increases with energy, showing the benefit of higher energies for neutron production. On the other hand it is shown that, insofar as neutron production is concerned, protons or ^{3}He of the same total energy of 2 GeV lead to very similar measured distributions. Should this be confirmed on thick targets, it would suggest that proton might not be the unique projectile to be retained in transmutation projects.

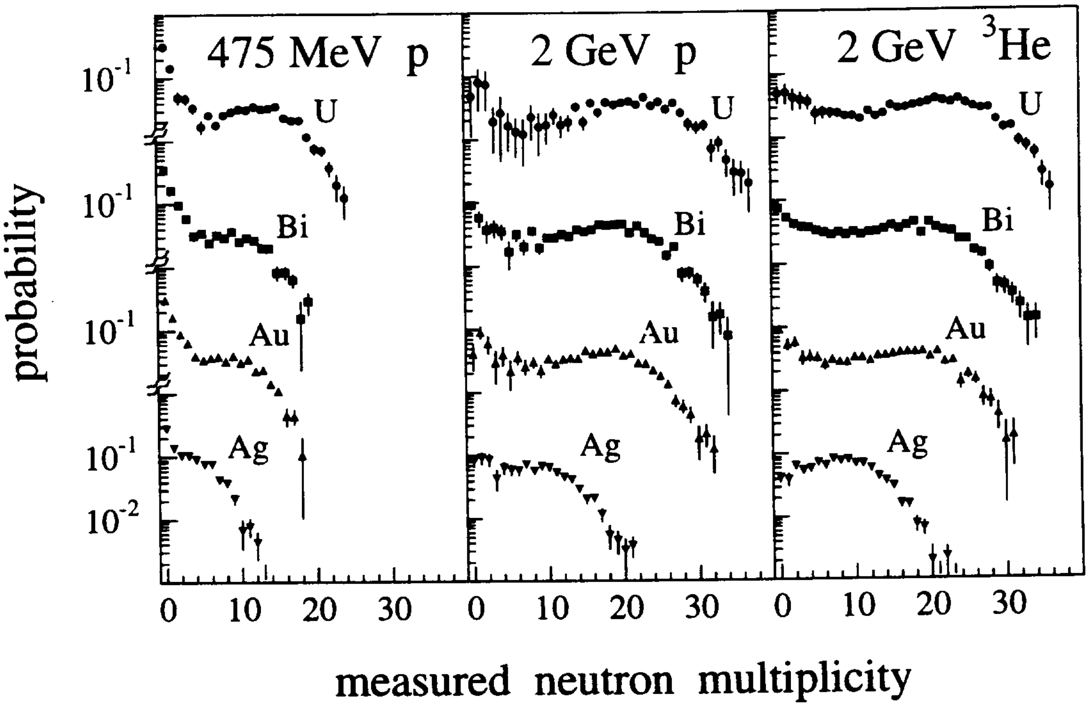

Fig.3 Neutron multiplicity distributions as measured for 3 different types of projectiles impinging on U, Bi, Au and Ag targets

The 2 GeV proton data on the gold target have been considered as quite representative of all data shown in Fig.3 and compared with model calculations. Two types of simulations have been performed: the one developed by Cugnon$^{12)}$, and the other one using the standard HETC (High Energy and Transport Calculation) code$^{11)}$. In the first case, two distinct steps are considered, an Intra-Nuclear Cascade process followed by an evaporation step, giving the opportunity to isolate the nuclei as a function of the temperature reached and follow their decay properties$^{9)}$. The results of this simulation, once corrected by the detector efficiency, are given in Fig. 4, in comparison with the experimental data (top). A reasonnable agreement is found, with slightly larger measured multiplicity values than calculated ones. This difference could be due to some neutrons created by secondary emitted particles after interaction in surrounding materials (scattering chamber walls, cave walls, ...). Estimates have been done using GEANT$^{13)}$, showing that 10 to 20% at most of the recorded neutrons could be "spurious" neutrons, in the sense that they do not arise from the very thin (1mg/cm^2) target.

It is shown on bottom of Fig.4 that the computed evaporated-like neutrons, as they should be measured, are much more abundant than those released during the Intra Nuclear Cascade step, thus showing that our 4π detector acts essentially as a thermometer: the larger the number of recorded neutron, the longer the evaporation chain and the hotter their nuclear emitter appears to be. It is indeed well known that for such heavy nuclei, heat is evacuated through neutron emission primarily and very little by charged particle evaporation$^{14)}$. It is estimated that for nearly 10% of the interactions, the nucleus is left with a temperature larger than 5 MeV when it reaches thermal equilibrium, after the INC process.

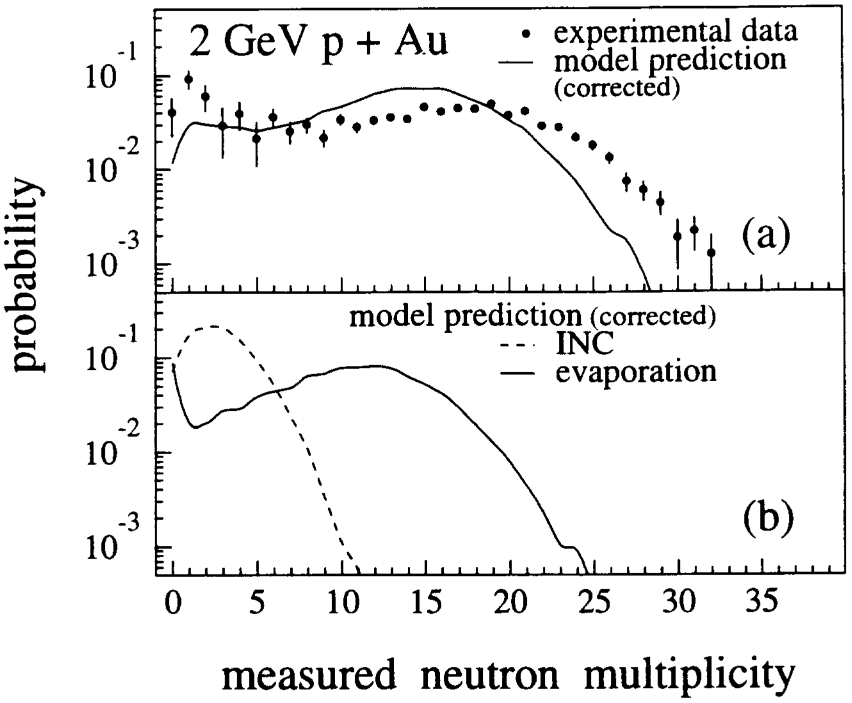

Fig.4 a) Measured neutron multiplicity distribution (dots) as compared to model predictions, corrected for detection efficiency (solid line).
b) Detailed distribution of INC-type neutrons and evaporative ones.

I It is also shown in Fig.5 that, due to the sectorization of the 4π detector, we have even more constraints than provided globally by the total measurement. It seems that it is close to 90° (sector C) and slightly forward of 90° (sector D) that the number of measured neutrons exceeds by the largest values the number of those calculated (the sectors are labeled from A to E, corresponding to the most backward one (A), the most forward one (E) and the intermediate B, C, D). Although this is not yet understood, this example demonstrates the benefit of the sectorization of the 4π detector in such studies.

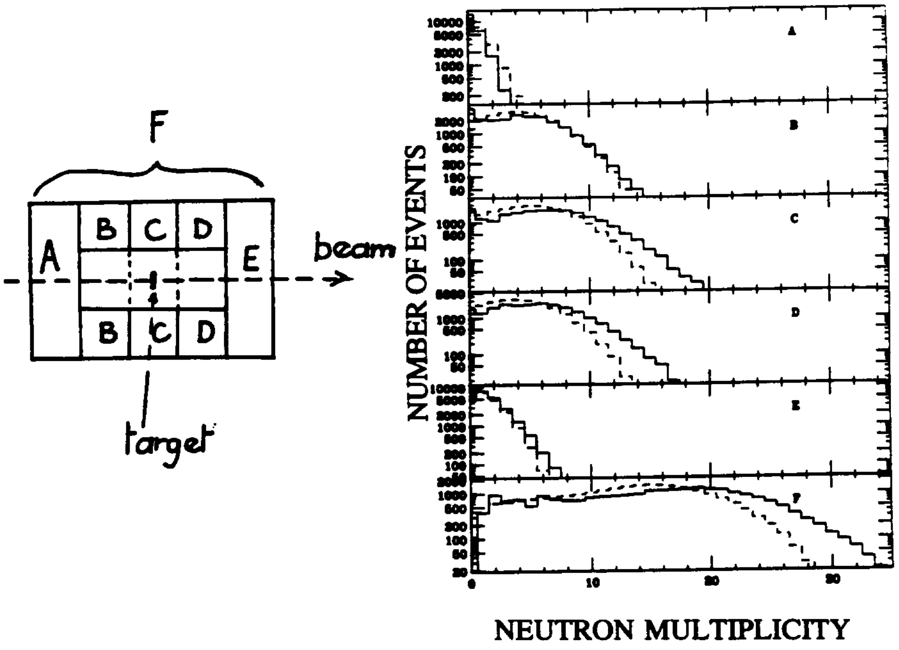

Fig.5 Distributions of neutron multiplicities in the 5 sectors (A-B-C-D-E) of the 4π neutron detector and their sum (F) as measured (solid lines) and simulated (dashed lines) for 2GeV protons on Au. The cross section of the 4m^3 detector is sketched on the left hand side.

In Fig.6, the HETC code has been utilized[11], showing first the expectation for the neutron multiplicity distribution (solid dots), the filtered distribution after folding by the detector efficiency (open dots) and the comparison with measured data (squares). A satisfactory agreement is found as it was in Fig.4 with the INC code.

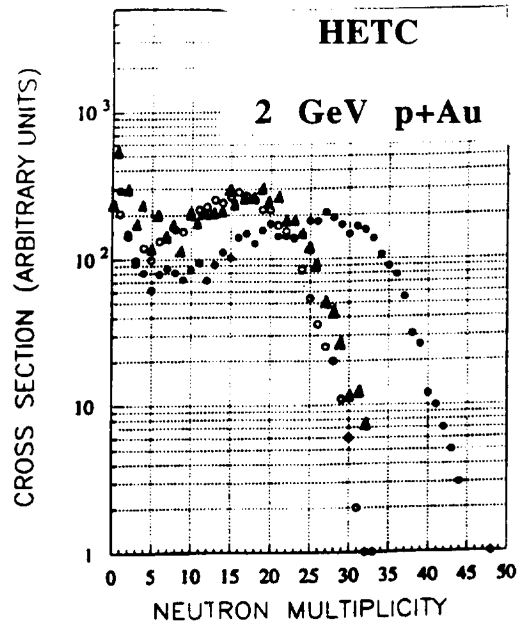

Fig.6 HETC simulations of the neutron multiplicity distribution for 2 GeV protons on Au (solid dots) and after folding by the detector efficiency (open dots). The experimental data are given by solid triangles.

More stringent tests have been performed with additional and more exclusive data than the neutron multiplicity data alone. Indeed, in conjunction with the latter, evaporated-like charged particles have been also measured which provide a unique signature of the temperature of the nuclei they are issued from. This signature is two-fold: first it is printed in the pattern of their energy spectra and then in their multiplicity. Most of the alpha-particles at backward angles have been produced via evaporation. Their energy spectrum, described as Maxwell-Boltzmann distributions, gives a direct access to the temperature T through an exponential relationship $((E-B)^{1/2}exp^{-(E-B)/T})$. It can be shown, selecting different gates of neutron multiplicity, that T increases with neutron multiplicity as expected[15]. For instance, for the highest selected neutron multiplicity gate (25 to 34 detected neutrons), a temperature of T>5 MeV can thus be inferred from the alpha-particle spectra, in fair agreement with what comes out of the model calculations for the stage where the nuclei are considered as thermalized.

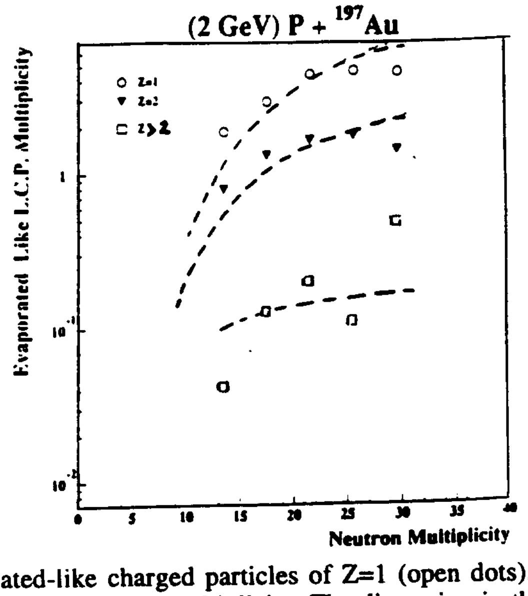

Fig.7 Multiplicity of evaporated-like charged particles of Z=1 (open dots), Z=2 (triangles) and Z>2 (sqares) as a function of measured neutron multiplicity. The dispersion in the later data is due to poor statistics. The corresponding model calculations are given by dashed lines.

Another severe test of the model calculations can be done, using the multiplicity of evaporated-like particles of Z=1, 2 and Z>2 detected as a function of measured neutron multiplicity. As shown in Fig.7, the experimental data are reasonnably accounted for with a calculation including consistently the INC step followed by an evaporation step[16](same conditions as for Fig.4).

To summarize, most of the neutron data or data taken in conjunction with the neutrons that we have presented in this contribution seem rather well reproduced, using current and rather simple concepts underlying the model of Intra Nuclear Cascade followed by thermal equilibrium and evaporation. A systematic investigation with other codes has still to be performed. Other data can not be understood with existing models. In particular the observation of composite particles, different from evaporative particles, such as $^{2-3}$H, $^{3-4}$He, Li, Be, ...and other nuclei of Intermediate-Mass are not explained by such models. The creation of such complex and energetic particles has been long known[17] but as far as we know they have never been satisfactorily explained, using for instance a coalescence model. There are other aspects which are far from being elucidated at present but might received some new pieces of information from our experiment[18]. For instance, the fate of the strongly excited and heavy nuclei is not well understood: the competition between evaporation residue formation, binary fission and possible multiple fragmentation seems to be an important issue. Indeed, this is what is going to determine the distribution of wastes in the thick target of heavy material.

As a last comment, and coming back to the neutron multiplicity measurements, it is worth noting that such measurements could be extended to very thick "production" targets with 4π detectors similar, in their design, to the one utilized in the thin target experiment performed at SATURNE. However, these detectors need to be adapted in order to house very massive and heavy targets or voluminous arrangements of heavy targets and light moderators and to handle much larger neutron multiplicity values than those recorded with thin targets. The neutron energy spectra from thick target being softer than those from thin targets, the overall detection efficiency should be notably improved. Also, it seems feasible to combine integrated 4π-neutron multiplicity measurements with complementary velocity measurements by designing a scintillator tank with small openings allowing TOF measurements[19]. This makes the sectorized, 4π-liquid scintillator detector a very powerful instrument well dedicated for further investigations required in designing any type of plant -spallation neutron source, transmutation plant, power plant[20] or hybrid- driven by an accelerator.

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