Study of pre-equilibrium emission of light complex particles from Fe and Bi induced by intermediate energy neutrons

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Abstract. We have measured double differential cross sections (DDX) for emission of hydrogen- and helium-isotopes in the interaction of 175 MeV quasi-monoenergetic neutrons with Fe and Bi using the Medley setup at the The Svedberg Laboratory (Uppsala, Sweden). We compared experimental DDX with calculations with the TALYS code, which includes exciton model and Kalbach systematics; the code fails to reproduce the emission of complex light ions, generally overestimating it. We propose an correction for the application of the Kalbach phenomenological model in the TALYS code by introducing a new energy dependence for the nucleon transfer mechanism in the pre-equilibrium emission region. Our results suggest also evidence for multiple pre-equilibrium emission of composite particles at 175 MeV.

1. Introduction

Newly developed nuclear methods in civil applications, as Accelerator Driven Systems for transmutation of nuclear waste and production of energy, require nuclear data for energies up to 1 GeV. Above 200 MeV models as the intra-nuclear cascade model work well and can be used to estimate the needed reaction data, however at lower energies the incident nucleon have a de Broglie wavelength corresponding to the radius of the target nucleus. This implies that the entire structural properties of the nucleus may participate in the nuclear reaction, making it more complex to model. Traditional nuclear data libraries answered the data needs in fission reactors (below 8 MeV) and in fusion research (below 14 MeV), and contain extensive experimental data. However there is a need of experimental data in the 20 to 200 MeV region. Recently evaluated data libraries have been released to cover this region and to meet the needs of the applied nuclear physics community. The TENDL-2009 [1] evaluated data library has been built based on calculations made with the TALYS code [2]. In a previous work we presented preliminary
double differential cross sections (DDX) for production of light ions [3] in the interaction of 175 MeV quasi-monoenergetic neutrons with Fe and Bi; there we observed that the production of protons was well predicted by TALYS, whereas the production of light complex particles (LCP) was largely overestimated. TALYS describes the pre-equilibrium emission of single particles, following the two-component exciton model (EM) as described by Koning and Duijvestijn [4]. The EM describes the wide pre-equilibrium emission region and proved to be successful in the prediction of reactions involving single nucleons and with energies up to 200 MeV. However this phenomenological model cannot account for reactions having complex particles in the entrance or in the exit channels. Kalbach [5] proposed a phenomenological model to supplement the EM; this model was based on experimental data up to 90 MeV (63 MeV in the case of neutron induced reactions). Koning et al. [2] included the Kalbach systematics in the TALYS code to complement the EM in predicting LCPs production up to 200 MeV.

In the present work we investigated the discrepancies between our experimental data and TALYS calculations with focus on LCPs production in the pre-equilibrium region; we also compared TALYS calculations with neutron induced experimental data at 96 MeV and with proton induced experimental data in the 120-200 MeV region. These data seem to show a systematic overestimation by the Kalbach systematics for emission of LCPs; our study shows that this overestimation is energy dependent and possibly mass dependent. We suggest a phenomenological correction for the application of the Kalbach systematics in TALYS, which goes in the direction to improve the predictive power of the code.

2. Light-ion production measurements at 175 MeV

We reported elsewhere the details of the experimental setup [6], thus a brief description will be given here. At the The Svedberg Laboratory (TSL) in Uppsala, Sweden, a quasi-monoenergetic neutron (QMN) source is available with energies up to 175 MeV. A cyclotron is accelerating protons up to 180 MeV; these are extracted from the cyclotron and made to interact with a 23.5 mm-thick $^7$Li target. The produced neutrons are shaped in a beam by a set of iron collimators, while the residual proton beam is deflected by a bending magnet into a beam dump where it is integrated in a Faraday cup in order to monitor the beam current. The neutron beam is then transported inside the Medley reaction chamber. Medley is a spectrometer system consisting of eight three-elements telescopes, placed at angles from 20° to 160°, in steps of 20°, and a reaction target positioned inside an evacuated 90 cm-diameter aluminum chamber. Each telescope consists of two fully depleted surface barrier silicon detectors serving as $\Delta E$ detector and a CsI(Tl) scintillator serving as E detector. Particle identification is thus performed using the $\Delta E$-E technique. To allow a low energy threshold we installed a relatively thin silicon detector at the front of each telescope, with thicknesses varying between 50 and 60 $\mu$m, while the second silicon detector was 1000 $\mu$m-thick in order to obtain a good separation of the detected particles. The 100 mm-long, 50 mm-diameter CsI(Tl) scintillators were designed to fully stop all the charged particles produced in the measured reactions. The light from the scintillators is collected by a photodiode. The signals from each telescope were processed using the data acquisition system described in [6, 7]. In the present experiment we used a 375 $\mu$m-thick iron target and a 0.5 mm-thick bismuth target; to normalize the measured data to absolute cross section values we measured also the production of protons from a 1.0 mm-thick polyethylene ($\text{CH}_2$) target. The relative neutron beam intensity was monitored by both an ionization chamber and a thin film breakdown counter.

The data reduction procedure is the same as in previous measurements of light-ion production measurements performed with the Medley setup and is based on the $\Delta E$-$\Delta E$-E technique [6, 7]. The energy calibration of the detectors was obtained using the data themselves. Events in the $\Delta E$-E bands were fitted with respect to the energy deposition in the silicon detectors; energy losses of light-ions in the $\Delta E$ were computed and we assumed linear response of the silicon
detectors. To calibrate the scintillator however we should consider a non-linear relationship between light output and energy deposition. An approximate quadratic expression was applied for hydrogen isotopes, while for helium isotopes we had also to consider quenching effects at very dense ionization which give a bending of the calibration curve at low energies. Time-of-flight (TOF) measurements were used in the off-line analysis to reduce the contribution from the neutrons in the low energy tail of the QMN spectrum. Data were corrected for thick target particle and energy loss, for subtraction of background events and the account for reaction losses in the CsI(Tl) scintillator.

3. Pre-equilibrium emission in TALYS

The TALYS code was developed to analyze and predict nuclear reactions involving neutrons, photons and light charged particles (A≤4) in the 1 keV - 200 MeV energy range for target nuclei heavier than carbon [2]. Several state-of-the-art models are included in TALYS. The default model to describe the pre-equilibrium process in TALYS is the two-component exciton model (EM); in this model the time evolution of the nuclear state is described by the total energy of the system and the total number of particles (protons and neutrons) above the Fermi surface and corresponding holes below it. Reference [4] offers a detailed description of the model. This model provides a good predictive power for nucleon induced reactions, however when LCPs are present in the entrance and/or exit channel of the reaction, two direct-like mechanisms play an important role and should be considered: the nucleon transfer (NT) and the knock-out (KO) of preformed clusters. TALYS includes the phenomenological model proposed by Kalbach [5] to take in account the NT and the KO reactions not covered by the exciton model. The total pre-equilibrium (PE) cross section is given as sum of three contributions:

\[
\frac{d\sigma^{PE}}{dE} = \frac{d\sigma^{EM}}{dE} + \frac{d\sigma^{NT}}{dE} + \frac{d\sigma^{KO}}{dE}
\]  

Three parameters in TALYS can be used to control how the NT and the KO contributions are added. The preeqcomplex flag allows to switch on or off the use of the Kalbach model for NT (pickup, stripping) and KO reactions, in addition to the exciton model, in the pre-equilibrium region. Cstrip and the Cknock are two adjustable parameters, respectively for the NT and the KO processes, to scale the complex-particle pre-equilibrium cross section per outgoing particle. The scaling factor can vary between 0 and 10.

4. Results and discussion

In two previous works we have compared DDX for production of hydrogen and helium isotopes from Fe and Bi at eight angles in the laboratory system with TALYS calculations using the default parameters [3]. We have observed that TALYS generally overestimates the production of LCPs over the wide pre-equilibrium emission region. To investigate this discrepancy we performed several calculations with TALYS varying different parameters affecting the pre-equilibrium emission. The version of the code used in this work was TALYS-1.2. Calculations performed with TALYS varying the following parameters did not significantly affect the computed DDX: pairmodel (pairing correction for pre-equilibrium model), preeqspin (spin distribution for the pre-equilibrium population of the residual nuclides), preeqsurface (flag to use surface corrections in the exciton model), mpreeqmode (model for multiple pre-equilibrium reactions). TALYS allows to choose between three different exciton models for the pre-equilibrium reactions, via the preeqmode parameter: no significant difference emerges from the use of the three models. Switching off the Kalbach contribution via the preeqcomplex flag parameter reduces the computed DDX for LCPs production by several orders of magnitude. TALYS does not allow fine tuning over the Kalbach phenomenological model: the Cstrip and
the $C_{knock}$ parameters are overall multiplication factors for the $\frac{d\sigma_{NT}}{dE}$ and the $\frac{d\sigma_{KO}}{dE}$ term respectively. These multiplication factors can vary between 0 and 10.

4.1. 96 MeV neutron induced data and 120, 160, 175 and 200 MeV proton induced data
We compared the TALYS calculations with neutron induced data at 96 MeV on Fe and Pb measured with the Medley setup by Blideanu et al. [8]; TALYS well reproduces the pre-equilibrium emission of proton, deuteron and triton (Pb) with default parameters, while a better fit is obtained with $C_{strip}=0.5$ for triton (Fe) and helium isotopes (in the case of $\alpha$ particle production we set also $C_{knock}=0.5$).

TALYS calculations were also compared with proton induced data at 120, 160 and 200 MeV for production of helium isotopes from Co and Au [9]; the best fit for the data is obtained setting the $C_{strip}$ parameter to 0.5 - 0.75 (120 MeV), 0.5 - 0.25 (160 MeV) and 0.25 - 0.10 (200 MeV). The same values were applied to the $C_{knock}$ parameter for $\alpha$ particle production.

Proton induced data for LCPs production from Ni at 175 MeV were recently measured by Budzanovski et al. [10]; we compared these data with TALYS calculations and found that the best fit is obtained for $C_{strip}$ and $C_{knock}$ set to 0.25.

4.2. Quasi-monoenergetic 175 MeV neutron induced data
The present neutron induced data are obtained from a QMN source: the spectrum includes 175 MeV monoenergetic neutrons and a low energy tail with neutrons down to 85 MeV. To compare model calculations with the experimental data it is necessary to fold the calculations with the accepted neutron spectrum. Since the comparison with proton induced data showed an energy dependence in the best fitting values for the $C_{strip}$ and the $C_{knock}$ parameters, we decided to use a simple linear energy dependence for the parameters in the folded calculations: for $E \leq 90$ MeV $C_{strip}=1$, while for 90 MeV $\leq E \leq 180$ MeV

$$C_{strip} = 1.9 - \frac{E}{100 MeV}$$

(2)

In this preliminary work we assumed the same energy dependence for the $C_{knock}$ parameter, however further investigation would eventually lead to a different energy dependence for this scaling factor.

The results for LCPs production from Fe are presented in Fig. 1. We observe that deuteron and triton pre-equilibrium production is better described by the calculations with reduced Kalbach contribution (solid line) than by default TALYS calculations (dashed line), while production of helium isotopes is still largely overestimated by TALYS. This suggest that the Kalbach contribution should be further reduced to describe helium isotopes, compared to hydrogen isotopes production, in agreement with the observations made for the 96 MeV data.

In Fig. 2 we report LCPs production from Bi. Deuteron production in the pre-equilibrium region is well described by TALYS when we reduce the contribution of the pick-up mechanism (NT) via the $C_{strip}$ parameter. Triton production at forward angles is better described by the modified calculations, while at backward angles default calculations are in better agreement with the experimental data. We observe that below 50 MeV deuteron and triton production shows a wide emission peak, wider that the expected evaporation peak; this can be explained with a possible multiple pre-equilibrium emission: after a first particle is emitted in the pre-equilibrium region, a second LCP could be emitted before the compound system reaches equilibrium. This mechanism is included in TALYS only for proton emission, but it is not prescribed for complex particles. Our data seems to suggest evidence for this process.

Production of $^3$He is well predicted by the modified TALYS calculations at all emission angles but at 140°, where large error bars do not give univocal evidence. Production of $\alpha$ particle is better described reducing the pickup and knock-out mechanisms as prescribed by the Kalbach
systematics. We observe also in the case of α particle production the evidence of multiple pre-equilibrium emission process in the low energy part of the pre-equilibrium spectrum.

Figure 1. Production of LCPs in the interaction of 175 MeV QMN with Fe. Experimental data at 20°, 60° \((10^{-1})\), 100° \((10^{-2})\) and 140° \((10^{-3})\) in the laboratory system are compared with default TALYS calculations (dashed line) and with TALYS calculations modified reducing the Kalbach contribution to the pre-equilibrium emission as described in the text (solid line).

Figure 2. Same as Fig. 1 but for the interaction of 175 MeV QMN with Bi.
5. Conclusions
We have compared production of hydrogen and helium isotopes in the interaction of 175 MeV quasi-monoenergetic neutrons with iron and bismuth, with model calculations with the TALYS code; we have observed a large overestimation for the production of LCPs in the pre-equilibrium emission region. TALYS computes pre-equilibrium DDX via the EM and includes the Kalbach phenomenological model to describe direct-like mechanisms in the production of LCPs. We showed that reducing the contribution of the Kalbach systematic in the DDX calculated by TALYS improved the overall predictive power of TALYS. We observe that the new parametrization is more successful in the case of Bi data, whereas in the case of Fe the improvement is still not satisfactory: this suggest the need of introducing a target-mass dependence in the parametrization. Neutron induced data at 175 MeV QMN from C, Si, O and U are currently under analysis, and when ready will provide more information on this possible target-mass dependence. To determine the energy dependence of our parametrization, we studied proton induced measurements in the same energy range (120 to 200 MeV) with TALYS and we derived a dependence in the scaling factor for the Kalbach contribution. Comparison with 96 MeV neutron induced data showed that a scaling factor should be introduced also at this energy to describe helium isotopes emission. However at this stage we applied the same parametrization to all emitted particles.

We showed also that there is possible evidence for multiple pre-equilibrium emission of LCPs at 175 MeV.

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