Inclusive Spectra of (p,xp) and (p,xd) Reactions on $^{90,92}$Zr and $^{92}$Mo Nuclei at $E_p=30.3$ MeV

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Abstract

New experimental data for the inclusive reactions (p,xp) and (p,xd) on isotopes of the nuclei $^{90,92}$Zr and $^{92}$Mo, have been measured at $E_p=30.3$ MeV, which has not been investigated in detail so far. We show the extension of the pre-equilibrium reactions to this energy region and interpret the results of these experiments. Moreover, we display the mechanism of the reaction and the level of energy-dependence. The adequacy of the theoretical models in explaining the measured experimental data is also discussed. In our theoretical analysis, the contributions of multi-step direct and compound processes in the formation of cross-sections are determined and we assert that the traditional frameworks are valid for the description of the experimental data.

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I. INTRODUCTION

Working out the pre-equilibrium decay mechanism in nuclear reactions, which reflects the dynamics of the formation of the excited system and its evolution to the equilibrium state, remains an actual problem of the nuclear reaction theory. The problem is largely connected with obtaining the new experimental data on double-differential cross-sections in \((p,\text{xp}), (p,\text{xd})\) etc. reactions with different proton energies. These reactions play a role in the applied researches on secure and wasteless nuclear power system creation (accelerator+subcritical reactor). In this respect, there is a problem of determining the spatial and power distribution of the secondary particles, generated not only during the transition of the primary proton beam of target assembly and neutron flow, but also of more composite \((^2^3\text{H}, ^3^4\text{He})\) particles, which can represent themselves as initiators of the reactions by emitting neutrons.

In this paper, the zirconium and molybdenum elements have been chosen as the objects of our investigation since they are the construction elements of a hybrid nuclear energy plant.

Actually, in the past, there have been experimental studies of the \((p,\text{xp})\) data on \(^{98}\text{Mo}\) at incident energies around 26 MeV by Watanabe \textit{et al.} \[1\]. This group has measured the energy spectra of emitted protons between angles 30° to 150°, at intervals of 10°. But, there are no experimental data at outgoing proton energies around 7 MeV or less because the protons have been stopped completely in the silicon surface-barrier \(\Delta E\) detector, which was 300 micron thick. They have analyzed the proton-induced reactions theoretically by using the FKK-GNASH code based on the FKK theory. The experimental nucleon emission spectra at 26 MeV have been reproduced well by the calculations including pre-equilibrium MSD and MSC emission, direct collective excitation to low-lying discrete levels, and Hauser-Feshbach equilibrium emission, in a quantum-mechanical way.

There are also the experimental \((p,\text{xp})\) spectra for \(^{98}\text{Mo}\) at 12, 14 and 16 MeV \[2, 3\] and for \(^{90}\text{Zr}, ^{54,56}\text{Fe}\) and \(^{93}\text{Nb}\) at 26 MeV \[4\] from the Kyushu group. In the first case, the target was the self-supporting metallic foil \(^{98}\text{Mo}\), with a thickness of 450 \(\mu\text{g/cm}^2\). Emitted protons were detected with a \(\Delta E-E\) counter telescope consisting of 75 \(\mu\text{m}\) and 2000 \(\mu\text{m}\) silicon surface detectors. Proton energy spectra were measured at intervals of 10°, from 30° to 160°. The \((p,\text{xp})\) spectra were analyzed on the basis of the exciton model and the Hauser-Feshbach model, in which isospin conservation was taken into account.

In the second case, they used self-supporting metal foils of \(^{54-56}\text{Fe}, ^{90}\text{Zr}\) and \(^{93}\text{Nb}\) with
a thickness of 500 µg/cm², at 26 MeV. The thicknesses of ∆E and E silicon detectors were 30, 200 and 5000 µm. The energy spectra were measured in the angle range 30-150°, at intervals of 10°. These data were analyzed pursuant to the FKK theory for the pre-equilibrium process by using the code FKK-GNASH and pursuant to the Hauser-Feshbach model for the compound process. The calculated results showed a good agreement with the experimental spectra for both cases.

In our experiment, we focus on investigating two issues: Firstly, there are no experimental data at outgoing proton energies around 7 MeV or less since, from Watanabe et al.’s measurement, the protons have been stopped completely in the silicon surface-barrier ∆E detector, which was 300 micron thick. In order to overcome this problem and to cover the soft part of the spectra, we have used a 50 micron ∆E-detector, therefore, the energy range is started from 2.5 MeV for our measurements. Secondly, the protons in the energy region of 30 MeV have not been studied in detail [1, 2, 3, 4] and extending the experiment in this direction allows us to view the mechanisms of the reaction and the level of energy-dependence in detail and to use these observations for adequate analysis within the framework of the FKK theory. Undoubtedly, the extraction of experimental information on channels of reactions with the emission of complex particles (deuteron, tritium, ⁃He and ⁴He) remains interesting.

In the next section, we present our experimental method, the details of the measurement and the experimental results. Section II is devoted to the theoretical analysis of the measured experimental data by the exciton model and quantum mechanical representations. Finally, Section IV gives our summary and conclusion.

II. EXPERIMENT AND RESULTS

The inclusive cross-section measurements of the reactions (p,xp), (p,xd), originated by protons with E=30.3 MeV, on isotopes of the nuclei ⁹⁰,⁹²Zr and ⁹²Mo, have been carried out on the cyclotron U-150M of the Institute of Nuclear Physics NNC Republic of Kazakhstan, within the range of 15-150°, at intervals of 15°. Typically, intensities between 20 and 40 nA have been utilized with a beam energy resolution of 0.3%.

For the registration and identification of reaction products in the whole energy range, a system has been designed with a three-detection telescope (ΔE₁ − ΔE₂-E₃), consisting
of two silicon surface-barrier ORTEC detectors and a scintillation detector CsI (Tl) with a total absorption of $E_3$. The thicknesses of detectors were $\Delta E_1=50\ \mu m$, $\Delta E_2=300\ \mu m$ and $E_3=25000\ \mu m$ correspondingly. The solid angle subtended by a telescope of detectors was equal to $\Omega=2.9\ 10^{-5}\ sr$. The spectra of protons have been registered from a threshold, which is defined by absorption in the first detector $\Delta E_1$, and by maximum energy absorption in the second detector $\Delta E_2$ in an interval, and these spectra have been identified by a matrix of coincidence ($\Delta E_1 \times \Delta E_2$). The second interval of registration has been determined by energy of fragments in the detector $\Delta E_2$, down to the energy of total absorption in the detector $E_3$, at which a matrix of identification ($\Delta E_2 \times E_3$) corresponds. Thus, the power spectrum of protons in the studied reactions has been measured within an energy range starting from a threshold of $E_p \approx 2.5\ MeV$ up to $\approx 30\ MeV$, and deuterons $\approx 2.5\sim 20\ MeV$.

The energy calibration of a spectrometer has been carried out on kinematics of levels of residual nuclei in the reaction $^{12}\text{C} (p,x)$ and protons of recoil. The base calibration $\Delta E_1 \times \Delta E_2$ is approximated by a straight line and it does not depend on any kind of fragments, whereas for events $\Delta E_2 \times E_3$ (detector CsI (Tl)), the base calibration represents a parabola for protons and a straight line for deuterons. This is shown in Figure 11 where the base calibration expresses the relation between the channel number in line spectra and the lost energy in the detector. It is found by subtracting the particle energy, lost in the target and the detectors, from the kinetic energy. The energy of the particle before hitting into the telescope of detectors is determined by using this base calibration and by restoring the losses in the detector. After that, we define the energy of the emitted particle by adding the losses in the target. The full energy resolution of the spectrometer has amounted to 800 keV for $\Delta E_2 \times E_3$ and $\approx 400\ keV$ for $\Delta E_1 \times \Delta E_2$.

The electronic scheme of the measurement is presented in Figure 2, the main bends of which are four-entrance spectroscopic amplifiers, multi-port (up to eight) analog-digital converters, discriminators and pulse counters. The main complexity during the set-up of the system is connected with the installation of a threshold of the discriminator SCA#3 for avoiding an overload by false coincidences with gamma-quantums, that leads to the formation of a slot between events $\Delta E_1 \times \Delta E_2$ and $\Delta E_2 \times E_3$, in the width of 2 MeV for protons and about 3 MeV for deuterons. The dead time of the system makes from 7 up to 1 % and is controlled by the Counters 1-2.

In the experiments, the self-supporting targets of isotopes $^{90,92}\text{Zr}$ and $^{92}\text{Mo}$ made by a
TABLE I: Characteristics of target nuclei

| Isotope | $^{90}\text{Zr}$ | $^{92}\text{Zr}$ | $^{92}\text{Mo}$ |
|---------|-----------------|-----------------|-----------------|
| Thickness, mg/cm$^2$ | 2.13 | 0.8 | 0.51 |
| Enrichment, % | 95 | 97 | 95 |

TABLE II: Experimental partial cross-sections (mb) of reactions (p,xp) and (p,xd)

| Reaction | OER$^a$ (MeV) | $^{90}\text{Zr}$ | $^{92}\text{Zr}$ | $^{92}\text{Mo}$ |
|----------|----------------|-----------------|-----------------|-----------------|
| (p,xp)   | 3-27           | 973.3 ± 8.5     | 839.6 ± 6.4     | 1229.0 ± 10.2   |
| (p,xd)   | 4-17           | 47.39 ± 0.59    | 28.83 ± 0.38    | 33.25 ± 0.51    |

$^a$OER denotes Outgoing Energy Range

method of electrochemical evaporation have been used, the characteristics the one of which is listed in Table I.

In Figures 3 and 4, the spectra of double-differential cross-sections ($d^2\sigma/dE_d\Omega$) are shown. The total cross-sections of (p,xp) and (p,xd) reactions, based on the integral ($d\sigma/dE$), are listed in Table 2.

In Figure 5, the experimental cross-sections of the studied reactions are shown. Moreover, three areas connected with different decay mechanisms for a channel with emitting protons are observed. The first one is the area of equilibrium components of a cross-section with a maximum at $E_p \approx 5$ MeV, the pre-equilibrium component ($E_p \approx 12-20$ MeV) and the range of $E_p > 20$ MeV, conditioned by direct mechanisms following it.

For a channel with emission of deuterons (p,xd), the situation is less determined. Typically for this channel, the cross-section of the equilibrium components is sharply overwhelmed ($E_d \approx 5$ MeV). It is remarkable that the form of deuteron spectra is different for zirconium isotopes and $^{92}\text{Mo}$ nuclei.
III. ANALYSIS AND RESULTS

There have been many theoretical approaches used to describe the equilibrium reactions data over a wide range of incident energies (see references [5, 6, 7, 8] for a detailed discussion). In this paper, we analyze the experimental cross-sections data of all the reactions pursuant to both the Hauser-Feshbach theory -while taking into account the multi-particle emission of both single-shot (protons, neutrons) and two-charging fragments (deuterons, α-particles)-, and the stringent quantum-mechanical theory (program EMPIRE II [9]). Thus, the contributions of statistical direct and compound processes for the reactions (p,xp) and (p,xd) have also been calculated.

The approach to statistical multi-step direct reactions is based on the multi-step direct theory of pre-equilibrium scattering to the continuum, originally proposed by Tamura, Udagawa and Lenske [10]. Since then, the approach has been revised especially in the part related to the statistical and dynamical treatment of nuclear structure.

The evolution of the projectile-target system from small to large energy losses in the open channel space is described in the MSD theory with a combination of direct reaction (DR), microscopic nuclear structure and statistical methods. As typical for the DR-approach, it is assumed that the closed channel space, i.e. the MSC contributions, have been projected out and can be treated separately within the multi-step compound mechanism.

The modelling of multi-step compound processes follows the approach of Nishioka et al. (NVWY) [11]. Like most of the pre-compound models, the NVWY theory describes the equilibration of the composite nucleus as a series of transitions along the chain of classes of closed channels in increasing complexity. In the present context, we define the classes in terms of the number of excited particle-hole pairs (n) plus the incoming nucleon, i.e. excitons. Thus, the exciton number is N = 2n+1 for nucleon induced reactions. Assuming that the residual interaction is a two-body force, only neighbouring classes are coupled (Δn = ±1).

In all cases, the parameter of level density had already been determined by Gilbert-Cameron parameterization [12]. For comparison, the calculation of the inclusive integral cross-section with the parameter of level density a=A/8 has been carried out, where A is the nuclear mass number.

The optical potential of Becchetti-Greenlees [13] for proton and neutron channels and the
optical potential of Perey-Perey \cite{14} for deuteron channel have been used in calculations of reaction transmission coefficients.

The results of these calculations are shown in Figure 6. It is shown that the contribution of multi-particle compound of the mechanism determines the emission of protons from 2.5 MeV up to 10 MeV, and the contribution of the multi-step direct process extends from 5 MeV up to the kinematic threshold. The shape of (p,xp) reaction integral spectra is determined by the multi-step direct processes. At the same time, the shape of spectra within the indicated theory that can be connected to the contribution of single-step direct mechanisms, which are not taken into account within this approach, is not to be described for the reaction (p,xd).

The cross-section contribution of the multi-step compound process for the (p,xp) reaction is by an order of magnitude smaller than the multi-step direct mechanism.

The quantum-mechanical theory allows to play back the shape of spectra and of the absolute cross-section for the reaction (p,xp). At the same time, however, this program does not calculate (p,xd) reaction cross-sections using the quantum-mechanical MSD and MSC model. We can use it only in the calculations of Hauser-Feshbach components.

In this connection, the calculations of the double differential cross-sections (Figure 3, 4) and integral spectra (Figure 7) have been carried out by using the program PRECO-D2 \cite{15}. This code uses the Griffin exciton model \cite{16} for pre-equilibrium nuclear reactions to describe the emission of particles with mass numbers of 1 to 4 from an equilibrating composite nucleus. A distinction is made between open and closed configurations in this system and between the multi-step direct and multi-step compound components of the pre-equilibrium cross sections \cite{17}. Additional MSD components are calculated semi-empirically to account for direct nucleon transfer reactions and direct knock-out processes involving cluster degrees of freedom. Evaporation from the equilibrated composite nucleus is included in the full MSC cross section. Output of energy differential and double differential cross sections is provided for the first particle emitted from the composite system. For that, there are additional subroutines in this program, which use the total MSD (including direct) and total MSC (including evaporation) cross sections to calculate the angular distributions for the emitted particles. This is done phenomenologically according to the paper by Kalbach and Mann \cite{18}.

The comparisons of the experimental integral cross-sections with PRECO-D2 calculations are shown in Figure 7. In all calculations, the configuration (1p0h) has been firstly used.
The Becchetti-Greenlees optical potential parameters for proton channel have been used to generate reaction transmission coefficients. The optical potential of Mani et al. \cite{19} has been used in the calculations for neutron channel and the potential of Cline \cite{20} for deuterons. The densities of levels have been parameterized as \( a = A/8 \). With the same parameterization, the total cross-section for reaction \((p,xp)\) and multi-particle component for reaction \((p,xd)\), calculated by the program EMPIRE, are demonstrated for a comparison in Figure \(7\). Figure \(3\), \(4\) and \(7\) show that the satisfactory theoretical description of the double-differential and integral cross-sections both for reactions \((p,xp)\) and \((p,xd)\) has been reached. The main difference between PRECO-D2 calculations and the experiment data is observed in a soft part of the spectra that is apparently connected with the neglect of the multi-particle emission within the code PRECO-D2. The high-energy part of experimental spectra consists of an elastic peak that can not be studied within the framework of this model.

The contributions of the MSD, the MSC, the equilibrium components and the single-step direct process of knock-out for the reaction \((p,xp)\) and one-nucleon transfer (pick-up of a neutron) for the reaction \((p,xd)\) have been well estimated. Two classes of single-step direct reactions which are not included in the Griffin model are nucleon transfer (pickup) and knock-out or inelastic-scattering processes which involve complex particle degrees of freedom. While taking into account the contribution of these direct processes, the description of the measured experimental data can be considerably improved by the program PRECO-D2. It follows from the theoretical results that the main contribution into the cross-sections of the reaction \((p,xd)\) is the introduction of the mechanism of one-nucleon transfer. At the same time, the contribution of the knock-out mechanism to the reaction \((p,xp)\) is much less, but the contribution of the MSD dominates and that is in agreement with the results of calculations in the quantum-mechanical theory. Also, it is possible to see that the relative contribution of the MSC mechanism calculated in the exciton model, correctly replicates the contribution from the MSC, within the program EMPIRE.

IV. SUMMARY AND CONCLUSIONS

We have measured the double-differential \(^{90,92}\text{Zr}, ^{92}\text{Mo}\,(p,xp),(p,xd)\) reaction cross-sections at \( E_p = 30.3 \pm 0.15 \text{ MeV} \) in order to investigate pre-equilibrium nucleon emission. Integral
dσ/dE spectra and partial reaction cross sections have also been deduced. The experimental data has been analyzed within the framework of the phenomenological exciton model of pre-equilibrium decay and microscopic theory of the MSD and MSC processes. Since there are no experimental data for the protons around 30 MeV, this experimental study is very important for the extension of the pre-equilibrium experiments in this direction to see the mechanism of the reaction and the level of energy-dependence. It is also important to observe the adequacy of the above-mentioned theoretical models to explain the measured experimental data.

For this purpose, we have presented that the satisfactory theoretical description of the double-differential and integral cross-sections both for reactions (p,xp) and (p,xd) has been reached. The main difference between the PRECO-D2 calculation and the experiment is observed in a soft part of the spectra. This might be due to the neglect of the multi-particle emission in the calculations. However, the contributions of the MSD, the MSC, the equilibrium components and the single-step direct process of knock-out mechanisms for the reaction (p,xp) and one-nucleon transfer (pick-up of a neutron) for the reaction (p,xd) have been well estimated. We have shown that it is also possible to improve the description of the measured experimental data considerably by taking into account the contribution of these direct processes. It follows from the theoretical results that the main contribution into the cross-sections of the reaction (p,xd) introduces the mechanism of one-nucleon transfer. At the same time, the contribution of the knock mechanism to the reaction (p,xp) is much less, but the contribution of the MSD dominates and that is in agreement with the results of calculations in the quantum-mechanical theory. It is now well established that the contribution of the multi-particle compound emission into cross-sections of reactions is determined by protons with energies from 2.5 up to 10 MeV, and from 5 MeV up to the kinematic limit in direct processes.

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FIG. 1: Calibration characteristics of the telescope $\Delta E_2 \times E_3$ for protons and deuterons
FIG. 2: The block-scheme of the registration system: Amp.#1,2,3 - Spectroscopic Amplifier; SCA#1,2,3 - Single Channel Analyzer; Coinc.#1,2 - Scheme of Coincidences; Counter#1,2 - Counter Scheme; ADC#1,2,3 - Analog-Digital Converter, Level Adapter - impedance matcher of levels.
FIG. 3: Double-differential cross sections of reactions $^{90}$Zr, $^{92}$Zr and $^{92}$Mo(p,xp)
FIG. 4: Double-differential cross sections of reactions $^{90}$Zr, $^{92}$Zr and $^{92}$Mo(p,xd)
FIG. 5: Integral cross-sections of reactions $^{90}$Zr, $^{92}$Zr, $^{92}$Mo(p,xp),(p,xd)
FIG. 6: Contribution MSC and MSD mechanisms to the formation of integral cross-sections of reactions (p,xp) and (p,xd) obtained by using the code EMPIRE.
FIG. 7: Contribution of MSC and MSD mechanisms in the formation of integral cross-sections of reactions (p,xp) and (p,xd) obtained by using the code PRECO-D2.