

NEW CALCULATIONS OF PROTON CYCLOTRON PRODUCTION CROSS
SECTIONS FOR SOME MEDICAL RADIOISOTOPES AND TARGET NUCLEI
USED ON THE SPALLATION NEUTRON SOURCES

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Abstract: In this study, the new calculations on the excitation functions of $^{18}\text{O}(p,n)^{18}\text{F}$, $^{68}\text{Zn}(p,n)^{68}\text{Ga}$, $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$, $^{112}\text{Cd}(p,2n)^{111}\text{In}$, $^{209}\text{Bi}(p,n)^{209}\text{Po}$ and $^{232}\text{Th}(p,n)^{232}\text{Pa}$ reactions have been carried out in the 5- 50 MeV incident proton energy range. In these calculations, the pre-equilibrium and equilibrium effects have been investigated. The pre-equilibrium calculations involve hybrid model, geometry dependent hybrid model, the cascade exciton model and full exciton model. Equilibrium effects are calculated according to Weisskopf–Ewing model. The calculated results are compared with the experimental data taken from the literature.

Key words: Medical radioisotope, Positron emitting radioisotope, Accelerator Driven Systems, Pre-equilibrium reactions, Proton cyclotron

BAZI TIBBİ RADYOİZOTOPLAR VE SPALLASYON NÖTRON
KAYNAKLARINDA KULLANILAN HEDEF ÇEKİRDEKLER İÇİN PROTON
SİKLOTRON ÜRETİM TESİR KESİTLERİNİN YENİ HESAPLAMALARI

Özet: Bu çalışmada, 5-50 MeV proton gelme enerji aralığında; $^{18}\text{O}(p,n)^{18}\text{F}$, $^{68}\text{Zn}(p,n)^{68}\text{Ga}$, $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$, $^{112}\text{Cd}(p,2n)^{111}\text{In}$, $^{209}\text{Bi}(p,n)^{209}\text{Po}$ ve $^{232}\text{Th}(p,n)^{232}\text{Pa}$ reaksiyonlarının uyarılma fonksiyonlarının yeni hesaplamaları yapılmıştır. Bu hesaplamalarda, denge ve denge-öncesi etkiler incelenmiştir. Denge öncesi hesaplamalar için; hibrid model, geometri bağımlı hibrid model, cascade exciton model, ve full exciton model kullanılmıştır. Denge etkileri de Weisskopf–Ewing modeline göre hesaplanmıştır. Hesaplanan sonuçlar literatürden alınan deneysel verilerle karşılaştırılmıştır.

Anahtar kelimeler: Tıbbi radyoizotop, Pozitron yayıcı radyoizotop, Hızlandırıcı sürümlü sistemler, Denge-öncesi reaksiyonlar, Proton siklotron

INTRODUCTION

A cyclotron can accelerate alpha particles to 28-30 MeV and also it can principally accelerate proton particles to energies higher than 30 MeV. Consequently, higher

reaction processes such as (p,4n) or generally (p,xn) or even (p,xn,yp) processes are possible ($x = 1,2,3,\dots$ and $y = 1,2,3,\dots$). Such a multipurpose cyclotron with the option of high particle beam intensity and well developed tools for beam diagnosis and a certain variation of particle beam energy is an excellent universal instrument supporting commercial isotope production and Research and Development (R&D) in the field of medical isotope application for diagnosis and therapy. Research and Development needed for development of alternative technologies producing carrier-free radioisotope preparations for therapy (BEYER 2006). Today, by using cyclotrons and nuclear reactors can be produced a lot of radioisotopes (QAIM 2001a, TARKANYI et al. 2001, QAIM 2002a, QAIM et al. 2002b, SCHOLTEN et al. 2002, QAIM 2004). These isotope production alternatives are spallation reaction, high energy proton induced fission, isotope separation etc. (BEYER 2006).

Today, radioisotopes are produced using both nuclear reactors and cyclotrons (QAIM 2001a, TARKANYI et al. 2001, QAIM 2002, QAIM et al. 2002, SCHOLTEN et al. 2002, QAIM 2004). In the last decade, a big success has been provided on production and usage of these radionuclides. The radioisotopes obtained from using charged particles play an important role in medical applications (QAIM 2001a, QAIM 2004, SCHOLTEN et al. 2002). A medical radioisotope can be classified as a diagnostic or a therapeutic radionuclide, depending on its decay properties. These radionuclides are used in diagnostic studies via emission tomography, i.e. Positron Emission Tomography (PET) and Single Photon Emission Computed Tomography (SPECT), and in endoradiotherapy (internal therapy with radio nuclides). In general, the diagnostic radioisotopes can also be classified into two groups; namely β^+ – emitters (^{11}C , ^{13}N , ^{15}O , ^{18}F , ^{62}Cu , ^{68}Ga , etc.) and γ – emitters (^{67}Ga , ^{75}Se , ^{123}I , etc.). The use of positron emitting radioisotopes such as ^{11}C , ^{13}N , ^{15}O and ^{18}F together with PET offers a highly selective and quantitative means for investigating regional tissue biochemistry, physiology and pharmacology (PHELPS et al. 1982). PET has been developing with the increasing number of clinical facilities raising interest in the use of PET in routine practice. The consequences of this fact, not only for the technology of PET but also for cyclotron application and radiopharmaceutical production, necessitate a review of what is possible given today's state-of-the-art (WOLF & JONES 1983, WOLH & FOWLER 1985).

There are several new technological application fields of fast neutrons such as accelerator-driven incineration/ transmutation of the long-lived radioactive nuclear wastes (in particular transuranium nuclides) to short-lived or stable isotopes by secondary spallation neutrons produced by high-intensity, intermediate-energy, charged-particle beams, prolonged planetary space missions, shielding for particle accelerators. Rubbia succeeded in a proposal of a full scale demonstration plant of the Energy Amplifier (EA) (RUBBIA et al. 1995). The application fields for 14-15 MeV neutron incident energy are Accelerator-Driven subcritical Systems (ADS) for fission energy production and hybrid reactor systems which are a combination of the fusion-fission processes). The design of an accelerator-driven system (ADS) requires precise knowledge of nuclide production cross sections in order to predict the amount of radioactive isotopes produced inside the spallation target (RUBBIA & RUBIO 1996). The spallation targets can be *Pb*, *Bi*, *W* etc. isotopes and these target material can be liquid or solid. The precision of models to estimate residue production cross sections is

still far from the performance required for technical applications. One applied field in the ADS systems is the production of neutrons from spallation reactions. The production of neutrons in spallation neutron source from ADS systems has recently gained considerable interest due to their importance in technical applications. ADS systems can be used for production of neutrons in spallation neutron source and they can act as an intense neutron source in accelerator-driven subcritical reactors, capable of incinerating nuclear waste and of producing energy (TAKIZUTA et al. 1995). Another field of interest, which is presently discussed, is the technical application of nuclear-collision processes for the energy production and the transmutation of nuclear waste in hybrid reactor systems which work as a fission reactor where an additional high-energetic proton or ion beam serves to increase the neutron flux in the reactor in a controlled way. The design of a fusion-fission (hybrid) reactor and ADS systems potentialities require the knowledge of a wide range of better data. ^{232}Th and ^{238}U are important as fissile material in hybrid and ADS reactor systems. Thorium and Uranium (HAN 2006) are nuclear fuels and Lead (TEL et al. 2004a, TEL et al. 2006), Bismuth, Tungsten are the target nuclei in these reactor systems (DEMİRKOL et al. 2004, ŞARER et al. 2006).

In the radioisotope production programmers, nuclear reaction data are mainly needed for optimization of production routes. This process involves a selection of the projectile energy range that will maximize the yield of the product and minimize that of the radioactive impurities (SCHOLTEN et al. 2002). Recently, many evaluated excitation functions of commonly used production reactions can be found in the literature (TARKANYI et al. 2001, QAIM 2001b). Nuclear reaction calculations which are based on standard nuclear reaction models can be helpful for determining the accuracy of various parameters of nuclear models and experimental measurements. Calculations which are based on nuclear reaction models play an important role in the development of reaction cross sections (CHADWICK 2001). In this manner, in the present study, production routes of medical isotopes used for PET i.e. ^{18}F , ^{67}Ga , ^{68}Ga , ^{111}In and ^{209}Bi , ^{232}Th used for spallation neutron source target nuclei were investigated in a range of 5 – 50 MeV energy. Excitation functions for pre-equilibrium calculations were newly calculated by using full exciton model, hybrid model, geometry dependent hybrid model and cascade exciton model. The reaction equilibrium component is calculated with a traditional compound nucleus model by Weisskopf-Ewing (WEISSKOPF & EWING 1940) method. Calculation results are also compared with the available excitation functions measurements in literature.

EQUILIBRIUM AND PRE-EQUILIBRIUM EXCITON REACTION MODELS

Equilibrium emission is calculated according to Weisskopf and Ewing (WE) model neglecting angular momentum (WEISSKOPF & EWING 1940). In the evaporation, the basic parameters are binding energies, inverse reaction cross section, the pairing and the level-density parameters. The reaction cross section for incident channel a and exit channel b can be written as;

$$\sigma_{ab}^{\text{WE}} = \sigma_{ab}(E_{\text{inc}}) \frac{\Gamma_b}{\sum_{b'} \Gamma_{b'}} \quad (1)$$

where E_{inc} is incident energy, $\Gamma_b = \frac{2 s_b + 1}{\pi^2 \hbar^2} \mu_b \int d\varepsilon \sigma_b^{\text{inv}}(\varepsilon) \varepsilon \frac{\omega_1(U)}{\omega_1(E)}$ and the total single-particle level density is taken as,

$$\omega_1(E) = \frac{1}{\sqrt{48}} \frac{\exp\left[2\sqrt{\alpha(E-D)}\right]}{E-D} \quad (2)$$

where σ_b^{inv} is the inverse reaction cross section, E is the excitation energy of the compound nucleus, D is the pairing energy and g is the single particle level density and $\alpha = \frac{6}{\pi^2} g$.

The exciton model assumes that after the initial interaction between the incident particle and the target nucleus the excited system can pass through a series of stages of increasing complexity before equilibrium is reached, and emission may occur from these stages giving the pre-equilibrium particles (GRIFFIN 1966). The exciton model uses a unified model based on the solution of the master equation (CLINE & BLANN 1971) in the form proposed by Cline (CLINE 1972) and Ribansky, et al (RIBANSKY et al. 1973). Integrating the master equation over time,

$$-q(n, t=0) = \lambda^+(E, n+2) \tau(n+2) + \lambda^-(E, n-2) \tau(n-2) - \left[\lambda^+(E, n) + \lambda^-(E, n) + W_\ell(E, n)\right] \tau(n) \quad (3)$$

Where $q(n, t=0)$ is the initial condition on the process. $\tau(n)$ is the solution of the master equation which represents the time during which the system remains in a state of n excitons. The $\lambda^+(E, n)$ and $\lambda^-(E, n)$ are the internal transition rates and the use of master Eq. (3), which includes both the probabilities of transition to equilibrium $\lambda^+(E, n)$ and probabilities of return to less complex stage $\lambda^-(E, n)$. $W_\ell(E, n)$ is the emission rate for a state with an n -exciton configuration.

CASCADE-EXCITON MODEL

The Cascade-Exciton Model (CEM) assumes that the nuclear reactions proceed through three stages: INC, pre-equilibrium and equilibrium (or compound nucleus). Generally, these three components may contribute to any experimentally measured quantity (BARASHENKOV & TONEEV 1972, GUDIMA et al. 1983, BARASHENKOV et al. 1969).

$$\sigma(p)dp = \sigma_{\text{in}} \left[N^{\text{cas}}(p) + N^{\text{prq}}(p) + N^{\text{eq}}(p) \right] dp \quad (4)$$

The CEM combines essential features of the intranuclear cascade model (INC) with the exciton model. The inelastic cross section σ_{in} is not taken from the experimental data or independent optical model calculations, but it is calculated within the cascade model itself. Hence the CEM predicts the absolute values for calculated characteristics and does not require any additional data or special normalization of its results.

HYBRID AND GEOMETRY DEPENDENT HYBRID MODEL

The hybrid model for pre-compound decay is formulated by Blann (BLANN & VONACH 1983) as

$$\frac{d\sigma_v(\varepsilon)}{d\varepsilon} = \sigma_R P_v(\varepsilon)$$

$$P_v(\varepsilon) d\varepsilon = \sum_{\substack{n=n_0 \\ \Delta n=+2}}^{\bar{n}} \left[{}_n\chi_v N_n(\varepsilon, U) / N_n(E) \right] g d\varepsilon \left[\lambda_c(\varepsilon) / (\lambda_c(\varepsilon) + \lambda_+(\varepsilon)) \right] D_n \quad (5)$$

where $P_v(\varepsilon)d\varepsilon$ represents number of particles of the v (neutron or proton) emitted into the unbound continuum with channel energy between ε and $\varepsilon + d\varepsilon$. The quantity in the first set of square brackets of Eq.(5) represents the number of particles to be found (per MeV) at a given energy (with respect to the continuum) for all scattering processes leading to an "n" exciton configuration. It has been demonstrated that the nucleon-nucleon scattering energy partition function $N_n(E)$ is identical to the exciton state density $\rho_n(E)$. The second set of square brackets in Eq. (5) represents the fraction of the v type particles at energy, which should undergo emission into the continuum, rather than making an intranuclear transition. The D_n represents the average fraction of the initial population surviving to the exciton number being treated. Early comparisons between experimental results, pre-compound exciton model calculations, and intranuclear cascade calculations indicated that the exciton model gave too few pre-compound particles and that these were too soft in spectral distribution for the expected initial exciton configurations. The intranuclear cascade calculations results indicated that the exciton model deficiency resulted from a failure to properly reproduce enhanced emission from the nuclear surface. In order to provide a first order correction for this deficiency the hybrid model was reformulated by Blann and Vonach (BLANN & VONACH 1983). In this way the diffuse surface properties sampled by the higher impact parameters were crudely incorporated into the pre-compound decay formalism, in the geometry dependent hybrid model (GDH). The differential emission spectrum is given in the GDH as

$$\frac{d\sigma_v(\varepsilon)}{d\varepsilon} = \pi \tilde{\lambda}^2 \sum_{l=0}^{\infty} (2l+1) T_l P_v(l, \varepsilon) \quad (6)$$

Where $\tilde{\lambda}$ is reduced de Broglie wavelength of the projectile and T_l represents transmission coefficient for l th partial wave.

RESULTS AND DISCUSSIONS

The present paper describes new calculations on the excitation functions of $^{18}\text{O}(p,n)^{18}\text{F}$, $^{68}\text{Zn}(p,n)^{68}\text{Ga}$, $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$, $^{112}\text{Cd}(p,2n)^{111}\text{In}$, $^{209}\text{Bi}(p,n)^{209}\text{Po}$ and $^{232}\text{Th}(p,n)^{232}\text{Pa}$ reactions carried out in the 5- 50 MeV proton energy range. The pre-equilibrium calculations involve the hybrid model, the geometry dependent hybrid model, the cascade exciton model and full exciton model. Equilibrium reactions have been calculated according to Weisskopf-Ewing model.

The pre-equilibrium calculations on the excitation functions were carried out with PCROSS code (CAPOTE et al. 1991) for full exciton model, ALICE91 code (BLANN

& BISPLINGHOFF 1982) for hybrid model and the geometry dependent hybrid model, and CEM95 code (MASHNIK 1980) for cascade exciton model. The reaction equilibrium component in PCROSS code is calculated with a traditional compound nucleus model by Weisskopf-Ewing (WEISSKOPF & EWING 1940, CAPOTE et al. 1991) method neglecting angular momentum.

In the calculations of exciton model (PCROSS code), we used the initial exciton number as $n_0=1$ (1 proton, 0 hole), thus taking into account the direct gamma emission. We used the standard Weisskopf-Ewing theory for equilibrium calculations and full exciton model for pre-equilibrium calculations. Equilibrium exciton number is taken equal to $\sqrt{1.4gE}$, as was suggested by F.C. Williams (WILLIAMS 1971). Single particle level density parameter g is equal to $A/13$ in the exciton model calculation, where A is the mass number and E is the excitation energy of the compound nucleus.

In the calculations of pre-equilibrium model, the hybrid model and the geometry dependent hybrid model use the initial exciton number as $n_0=3$ (1 proton, 1 neutron and 1 hole). This model requires the initial proton (p) and neutron (n) exciton numbers. A detailed description of the ALICE91 can be found in Ref. (BLANN & BISPLINGHOFF 1982).

The CEM95 code is intended for the Monte Carlo calculation of nuclear reactions in the framework of the Cascade-Exciton Model. The CEM95 code allows us to calculate reaction, elastic, fission and total cross sections; nuclear fissilities, excitation functions, nuclide distributions, energy and angular spectra, double differential cross sections, mean multiplicities, mean energies and production cross sections for ejectile yields. Different models are incorporated in the CEM95 to calculate the level density parameter. The cascade exciton model calculations have been made by using CEM95 code with the level density parameter, and a detailed description of the CEM95 can be found in Ref. (MASHNIK 1980).

The calculated results are compared with the experimental data taken from the literature. The decay data for the investigated radionuclides were taken from the NUDAT databases (NUDAT) that are collected in Table 1.

Table 1. The decay data for investigated radionuclides

Reaction Product	Half life	Mode of decay (%)	E_γ (keV)	I_γ (%)
^{18}F	1.83 h	EC + β^+ (100)	0.52	0.01795
^{67}Ga	3.2617 d	EC + β^+ (100)	393.527	4.56
^{68}Ga	67.63 m	EC + β^+ (100)	1077.35	3
^{111}In	2.8047 d	EC + β^+ (100)	245.350	94.10

CONCLUSIONS

The new calculations on the excitation functions of $^{18}\text{O}(p,n)^{18}\text{F}$, $^{68}\text{Zn}(p,n)^{68}\text{Ga}$, $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$, $^{112}\text{Cd}(p,2n)^{111}\text{In}$, $^{209}\text{Bi}(p,n)^{209}\text{Po}$ and $^{232}\text{Th}(p,n)^{232}\text{Pa}$ reactions have been carried out using nuclear reaction models. In general, the used all model codes are well in agreement with the measurements data on target nuclei in Figs.1-6. The Weisskopf-Ewing model and the full exciton model calculations are better in agreement with the measurements up to 14-16 MeV. The geometry dependent hybrid model and hybrid model calculations are in good harmony with the experimental data above the incident energy 14-15 MeV. Similar calculated results assessed before if the incident particles were neutrons for (n,2n) reaction cross sections (TEL et al. 2004b, TEL et al. 2004c). The cascade exciton model calculations are in very good agreement with the measurements data above the incident energy 8-10 MeV. Also, the production ^{18}F , ^{67}Ga , ^{68}Ga and ^{111}In radioisotopes can be employed at small-sized cyclotron.

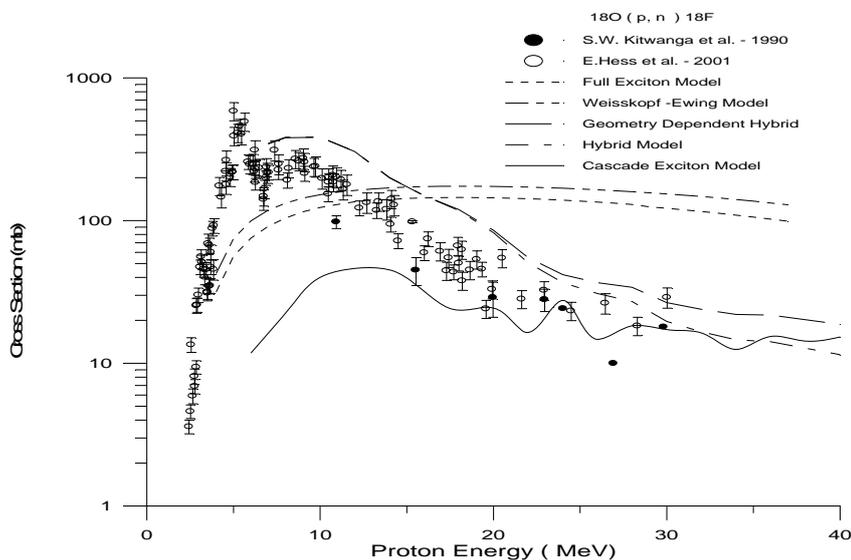


Figure 1. The comparison of calculated excitation functions of $^{18}\text{O}(p,n)^{18}\text{F}$ reaction with the values reported in literature

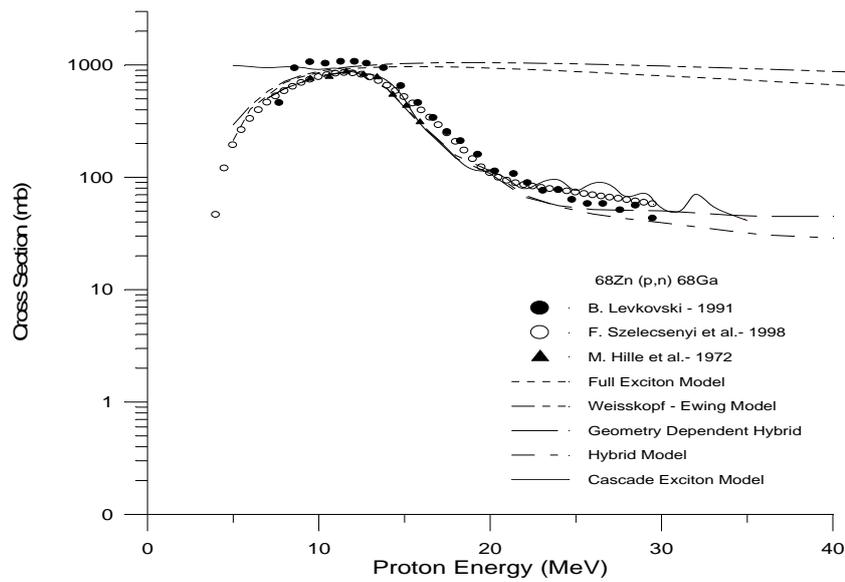


Figure 2. The comparison of calculated excitation functions of $^{68}\text{Zn}(p,n)^{68}\text{Ga}$ reaction with the values reported in literature

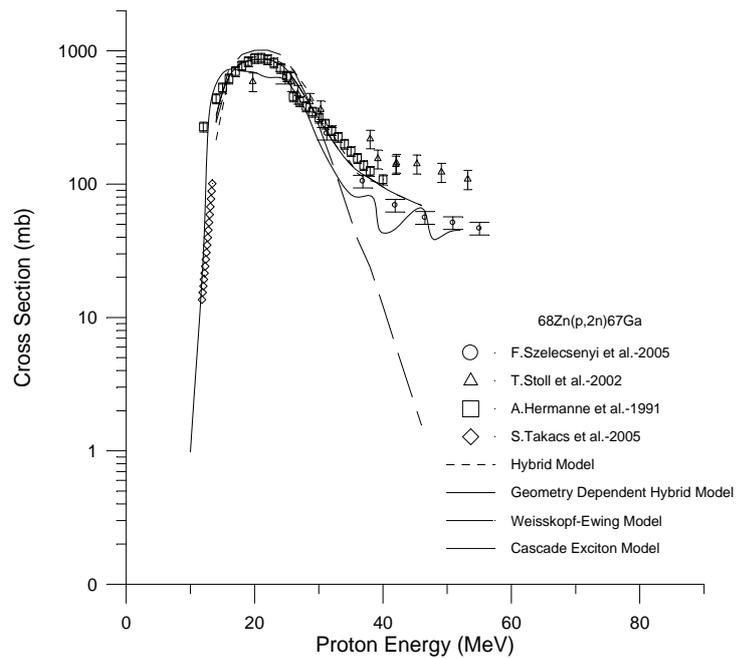


Figure 3. The comparison of calculated excitation functions of $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$ reaction with the values reported in literature

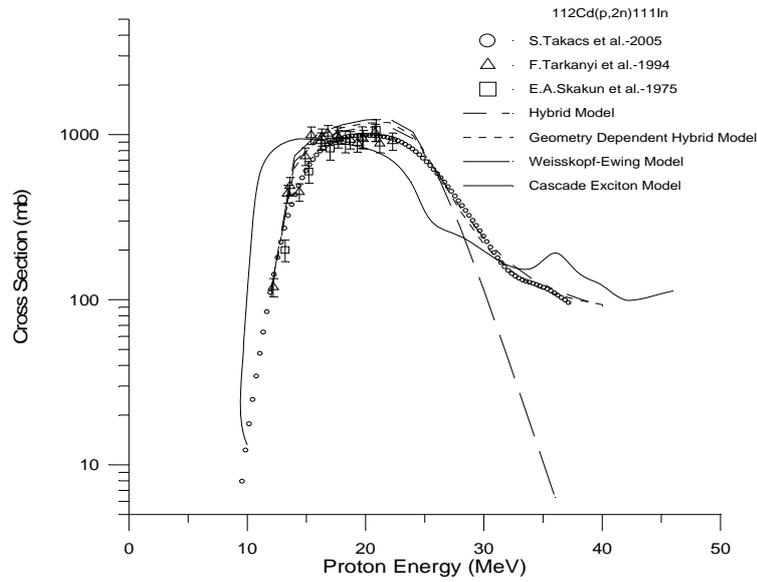


Figure 4. The comparison of calculated excitation functions of $^{112}\text{Cd}(p,n)^{111}\text{In}$ reaction with the values reported in literature

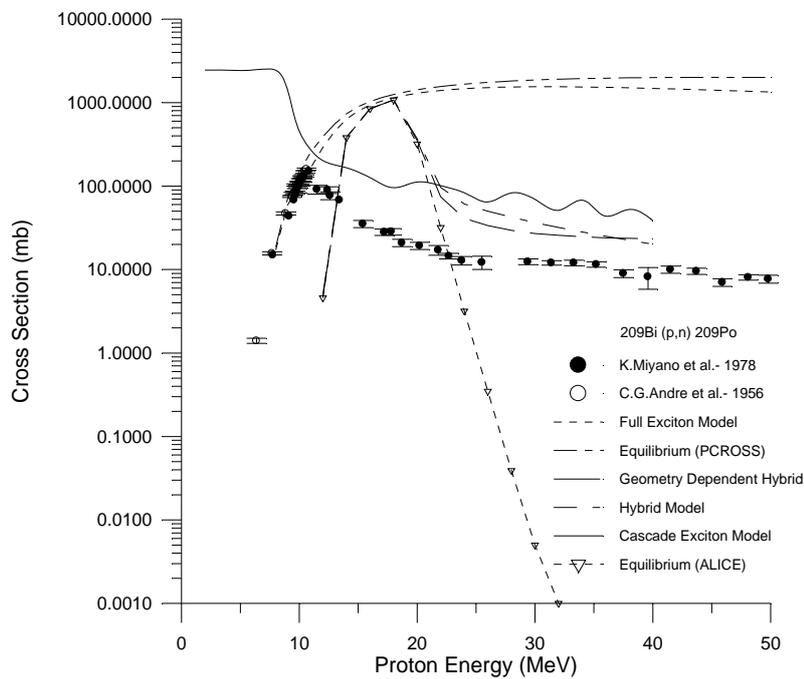


Figure 5. The comparison of calculated excitation functions of $^{209}\text{Bi}(p,n)^{209}\text{Po}$ reaction with the values reported in literature

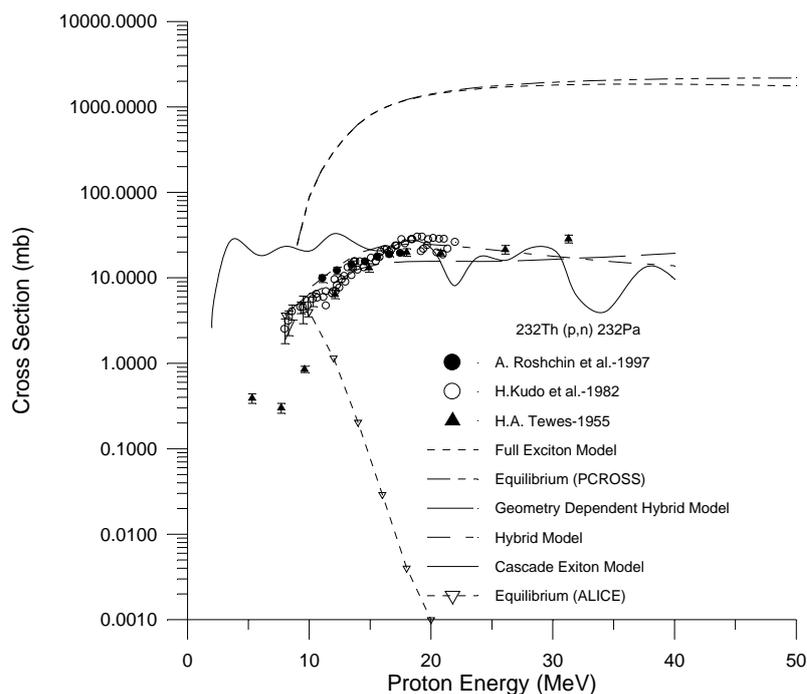


Figure 6. The comparison of calculated excitation functions of $^{232}\text{Th}(p,n)^{232}\text{Pa}$ reaction with the values reported in literature

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