

CALCULATIONS of THE MAIN FREE PATH on NEUTRON EMISSION CROSS-SECTION for SPALLATION REACTION of TARGET and FUEL NUCLEI

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ABSTRACT

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. Especially, accelerator driven subcritical systems (ADS) can be used for fission energy production and /or nuclear waste transmutation as well as in the intermediate-energy accelerator driven neutron sources, ions and neutrons with energies beyond 20 MeV, the upper limit of existing data files that produced for fusion and fission applications. In these systems, the neutron scattering cross sections and emission differential data are very important for reactor neutronics calculations. The transition rate calculation involves the introduction of the parameter of mean free path determines the mean free path of the nucleon in the nuclear matter. This parameter allows an increase in mean free path, with simulation of effect, which is not considered in the calculations, such as conservation of parity and angular momentum in intra nuclear transitions. In this study, we have investigated the multiple pre-equilibrium matrix element constant from internal transition for Uranium, Thorium, (n,xn) neutron emission spectra. The neutron-emission spectra produced by (n,xn) reactions on nuclei of some target (for spallation) have been calculated. In the calculations, we have used the geometry dependent hybrid model and the cascade exciton model including the effects of the pre-equilibrium. The pre-equilibrium direct effects have been examined by using full exciton model. All calculated results have been compared with the experimental data. The obtained results have been discussed and compared with the available experimental data and found agreement with each other.

Keywords: Cross-section, pre-equilibrium models, exciton model, mean free path and emission spectra

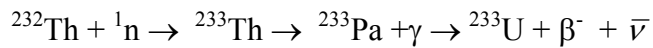
INTRODUCTION

The fusion-fission hybrid is a combination of the fusion and fission processes. The fusion plasma is surrounded with a blanket made of the fertile materials (U^{238} or Th^{232}) to convert them into fissile materials (Pu^{239} or U^{233}) by transmutation through the capture of the high yield fusion neutrons. The fertile materials may also undergo a substantial amount of nuclear fission, especially, under the irradiation of the high energetic 14.1 MeV- (D,T) neutrons. In addition to that some of the bred fissile material burns in the hybrid blanket “*in situ*” (Şahin et al., 1986, 2002a, 2002b, 2003, 2005; Übeyli and Tel, 2003). In an experimental hybrid blanket geometry concept, a line neutron source in a cylindrical cavity simulates the fusion plasma chamber. A first wall made of stainless steel, type SS-304, surrounds the latter. The fissile zone made up of natural- UO_2 or ThO_2 fuel. The fuel zone is cooled with pressurized helium gas coolant for a fast blanket or with light water for a thermal reactor. The cladding of the fuel rods is made of stainless steel, type SS-304, as the first wall. The radial reflector is made of Li_2O for production of tritium (T) and graphite in sandwich structure. This measure reduces the neutron leakage drastically and leads to a better neutron economy (Şahin, et al., 1984, 1986).

Thorium, like uranium, is a nuclear fuel, but the use of thorium fuel, unlike the use of uranium, has nearly been forgotten. In particular, ^{232}Th is important as fissile material in the new generation reactors. World thorium reserves are estimated to be about three times more abundant than the natural uranium reserves. The possibility of the production of ^{233}U in a fusion-fission (hybrid) reactor has been investigated by some early work. In addition, a neutronic analysis for a thorium fusion breeder with a special task of burning minor actinide's and production of ^{233}U , ^{238}Pu , ^{242m}Am and ^{245}Cm of a high isotopic purity for spacecraft application has been performed in this work. Calculations have been conducted using a (D,T)

fusion neutron driver for the hybrid reactor (Moir, 1979; Ragheb et al., 1979; Teller, 1981; Berwald et al., 1982; Lee et al., 1982; Greenspan, 1984; Moir et al., 1985; Moir and Lee, 1986).

The process of the main-breeding of the ^{233}U from the ^{232}Th by using the neutrons,



The design of a fusion-fission (hybrid) reactor and the study of the thorium cycle potentialities require the knowledge of a wide range of better data. Today the databases are relatively well filled for neutron energies below 20 MeV and for nuclei involved in the uranium cycle, but for those involved in the thorium cycle and for high neutron energies the situation is not satisfactory. ^{232}Th is important as fissile material in the hybrid and ADS reactor systems. In the ADS systems when heavy target elements are surrounded by a blanket assembly of nuclear fuel (Uranium, Thorium or Plutonium etc.), there is a possibility of sustaining fission reaction (Rubbia, et al., 1995,1996). It has been investigated in detail for $^{232}\text{Th}(n,xn)$, $^{238}\text{U}(n,xn)$ and some target nucleus in previous paper (Tel, et al., 2004,2006; Han, 2006; Demirkol et.al.2004; Şarer et al, 2006). In the present paper, the experimental neutron-emission spectra produced by (n,xn) reactions for ^{232}Th nuclei have been compared with experimental $^{238}\text{U}(n,xn)$ neutron-emission spectra at the bombarding energies 6, 14.1 and 18 MeV. Equilibrium angle-integrated cross sections in neutron induced reactions on targets ^{232}Th have been calculated at the bombarding energies at 14.1 MeV. The calculation results of the differential cross sections at 14.1 MeV for target nucleus ^{184}W , ^{207}Pb and ^{209}Bi have been given. A good description of experimental data has been achieved. By analyzing (n,xn) reaction on these nuclei with the incident energy from 6 MeV to 18 MeV, the importance of multiple pre-equilibrium emission can be seen clearly. All calculated results have been

compared with experimental data. The obtained results have been discussed and compared with the available experimental data and found agreement with each other

EQUILIBRIUM AND PRE-EQUILIBRIUM NUCLEAR REACTION MODELS

The equilibrium emission is calculated according to the Weisskopf-Ewing (Weisskopf and Ewing, 1940) model neglecting angular momentum. In the evaporation, the basic parameters are binding energies, the 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, $\Gamma_b = \frac{2s_b + 1}{\pi^2 \hbar^2} \mu_b \int d\varepsilon \sigma_b^{\text{inv}}(\varepsilon) \varepsilon \frac{\omega_1(U)}{\omega_1(E)}$ and the total single-particle level density is taken as,

$$\omega_1(E) = \frac{1}{\sqrt{48}} \frac{\exp\left[2\sqrt{\alpha(E-D)}\right]}{E-D}; \quad \alpha = \frac{6}{\pi^2} g \quad (2)$$

where σ_b^{inv} is the inverse reaction cross section, E is the excitation energy of the compound nucleus, U is the residual nucleus excitation energy, D is the pairing energy and g is the single particle level density .

It has been established that pre-equilibrium processes play an important role in nuclear reactions induced by light projectiles with incident energies above about 10 MeV. Starting with the introduction of exciton model by Griffin (Griffin, 1966) a series of semi classical models have been developed for the calculating and the evaluating particle emissions in the continuum (Betak, E., 1975). Then Blann and Vonach formulated the hybrid model for pre-

compound decay (Blann and Vonach, 1983). It was also shown that with some freedom in the choice of parameters, these models could give reasonable fit to the observed energy and angular distributions of the emitted particles. More recently, researchers have formulated several quantum–mechanical reaction theories (Hauser and Feshbach, 1952; Chadwick, 1989; Chadwick and Young, 1993) that are based on multi-step concepts and in which statistical evaporation at lower energies is connected to direct reactions at higher energies.

The exciton pre-equilibrium model for nuclear reactions is a phenomenological model based on phase space arguments. The equilibration in energy of a composite nucleus is followed in terms of the creation and destruction of pairs of particle and hole degrees of freedom. Exciton model is based on the solution of the master equation in the form proposed by Cline and Blann (Cline and Blann, 1971; Cline, 1972) and Ribansky et. al.(Ribansky, et al., 1973). Integrating the master equation over time, we can write as

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

where $q(n, t=0)$ is the initial condition, $\tau(n)$ is the time during which the system remains in a state of n excitons, W_l is the total particle decay probability of the n excitons state per unit time, E is the excitation energy of the compound nucleus, λ^+ is the probability of transition $n \rightarrow n+2$, and λ^- is the transition rate of $n \rightarrow n-2$ transition. The use of master equation (3), which includes both the probabilities of transition to equilibrium $\lambda^+(E, n)$ and the probabilities of return to less complex states $\lambda^-(E, n)$, enables us to calculate in a unified manner the pre-equilibrium and equilibrium emission spectrum in accordance with:

$$\frac{d\sigma_{ab}}{d\varepsilon_b}(\varepsilon_b) = \sigma_{ab}^r(E_{inc}) D_{ab}(E_{inc}) \sum_n W_b(E, n, \varepsilon_b) \tau(n) \quad (4)$$

where $\sigma_{ab}^r(E_{inc})$ is cross section of the reaction (a, b). $W_b(E, n, \varepsilon_b)$ is the emission probability of a particle of type b with energy E_b . The dimensionless depletion factor $D_{ab}(E_{inc})$ accounts for reduced population of each state due to the particle emission from simpler states with smaller n .

The theoretical calculations have also been made in the framework of the Full- exciton model using PCROSS computer code (Capote, et al., 1991). We used the standard Weisskopf-Ewing theory for equilibrium calculations. In the Full exciton model calculations, we used the initial exciton number as $n_o = 1$ of 1 neutron and 0 hole: single particle level density parameter g is equal to $A/13$ in the exciton model calculation, where A is the mass number. Level density expression given by Dilg et. al. (Dilg et. al., 1973) was used in the evaporation model calculation. Particle-hole state density expression reported by Williams (Williams, 1971) was used in the pre-equilibrium model calculation. The reaction cross sections and the inverse cross sections were obtained using the optical potential parameters. The octupolar and quadrupolar oscillations are only considered. The ω_2, β_2 values for even nuclei have been taken from Atomic Data & Nuclear Data Tables (Raman et al., 1987). In the case of odd nuclei, on the assumption of a weak bond, the values corresponding to the neighboring even nucleus are used. The ω_3 value has been taken from Table of Isotope (Lederer and Shirley, 1978). The octupolar deformation parameters have been calculated from $\beta_3^2 = (2\lambda + 1) \omega_3 [\text{MeV}] / 1000$. We have replaced the function $\delta(U - \omega_\lambda)$, which relates the excitation energy of the residual nucleus to the energy and the emission energy by Gaussian whose semi-width is chosen to take into account the experimental energy resolution.

There is strong dependence of the calculated emission spectra on the parameter of matrix element K if the parameterizations proposed by Kalbach (Kalbach and Mann, 1981) are used in the calculations of internal transitions rates $\lambda^i(i = +, -)$ and it is sought to achieve a good adjustment of the theoretical calculations to the experimental spectra. In order to avoid such a strong dependence, we preferred to use the parameterization of transition rate proposed by Blann and Vonach, which is based on existing experimental data on nucleon scatter and considers the Pauli principle in an approximate way (Blann and Vonach, 1983). Using this parameterization and considering the particle-hole state densities from the Williams formula

(Williams, 1971), we can obtain for internal transition rates the expressions found by Machner (Machner H., 1981):

$$\lambda^+(E, n) = \frac{1}{k_{mfp}} \left(1.4 \times 10^{21} E' - \frac{2}{n+1} 6 \times 10^{18} E'^2 \right)$$

$$\lambda^-(E, n) = \frac{1}{k_{mfp}} \frac{(n-1)(n-2)ph}{(gE')^2} \left(1.4 \times 10^{21} E' - \frac{2}{n-1} 6 \times 10^{18} E'^2 \right) \quad (5)$$

The PCROSS code takes into account an additional factor 3/8 multiple the right-hand side of (5), which was introduced by Gupta (Gupta, 1981) in deriving expressions for calculation by the exciton model from its bicomponent formulation, and includes approximate consideration of the Pauli principle by replacing E' by $E - [p(p+1) + h(h+1)]/4$ in λ^+ and by $E - [p(p-1) + h(h-1)]/4$ in λ^- . Blann's parameterization for internal transition rate calculation involves the introduction of the parameter of mean free path k_{mfp} so called because its value determines the mean free path of the nucleon in the nuclear matter. This parameter allows an increase in mean free path, with simulation of effect, which are not considered in the calculations, such as conservation of parity and angular momentum in intra nuclear transitions. As the direct excitations of levels of a collective nature are considered in the PCROSS code, the hard part of emission spectrum can be described satisfactorily with a global value of $k_{mfp} = 1.3$, which is smaller than reported earlier (Capote, et al., 1991).

SUMMARY AND CONCLUSIONS

In this paper, all calculated results using equilibrium and pre-equilibrium reaction mechanisms at the bombarding energies 6, 14 and 18 MeV have been presented and compared with the experimental data, and the following conclusions can be summarized as follows:

1. The experimental (n,f) fission cross section of the ^{232}Th nuclei is similar to ^{238}U (both of them can be make a fission with fast neutrons) so, ^{232}Th nuclei is important as fissile material in the new generation reactors (in Fig 1-2).
2. The experimental neutron-emission spectra cross section of the ^{232}Th nuclei has been lower than ^{238}U nuclei at the bombarding energies 6, 14 and 18 MeV. However, these experimental spectral shapes of the neutron-emission spectra are very similar to each other (in Fig 3-5).
3. The experimental neutron-emission spectra cross section of the ^{232}Th nuclei is very similar to ^{238}U so, ^{232}Th nuclei can be used as fissile material in the new generation reactors.
4. For the ^{232}Th neutron-emission spectra, the equilibrium calculations of the Weisskopf-Ewing have shown only good agreement with the experimental data up to 4-6 MeV (in Fig 6).
5. The summation contribution consists of the Weisskopf-Ewing theory for equilibrium and exciton model for pre-equilibrium contributions have shown good agreement with the experimental data (in Fig 8-9).

6. For the ^{232}Th , while the mean free path constant 2 and 3 correspond to the experimental values for 14.1 and 18 MeV neutrons, the mean free path constant 6 and 7 correspond to the experimental values for 6 MeV neutrons (in Fig 7-9).

7. The pre-equilibrium neutron-emission spectra cross section values of ^{207}Pb and ^{209}Bi are higher than ^{184}W . Therefore, ^{207}Pb and ^{209}Bi are good targets for using the spallation neutron sources (in Fig 10).

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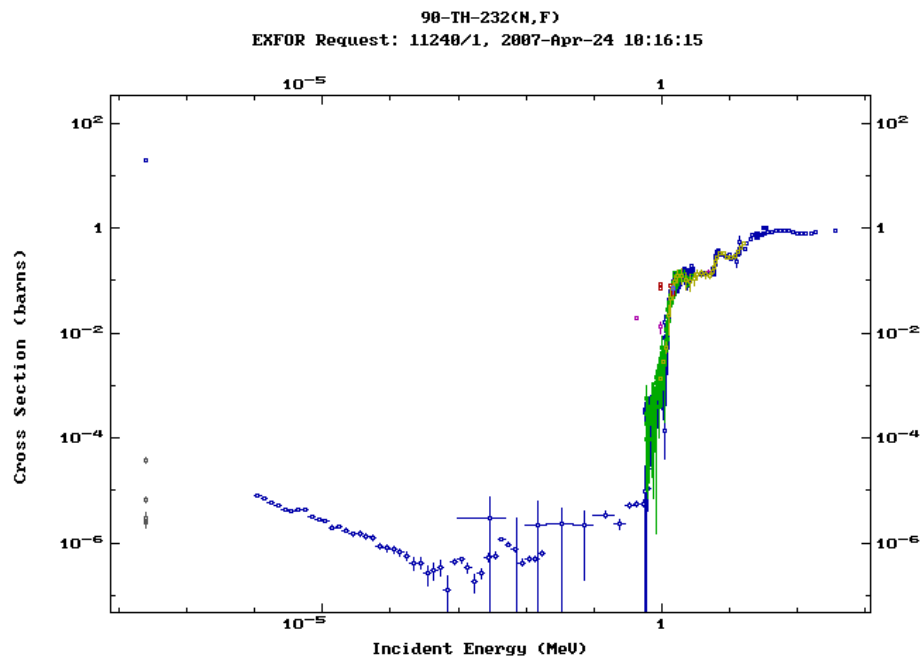


Fig. 1

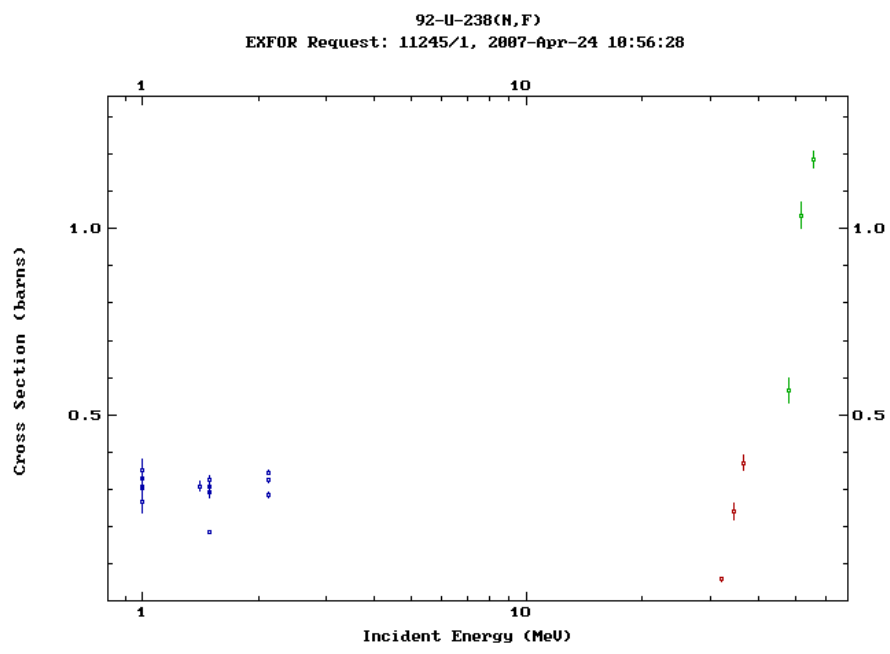


Fig. 2

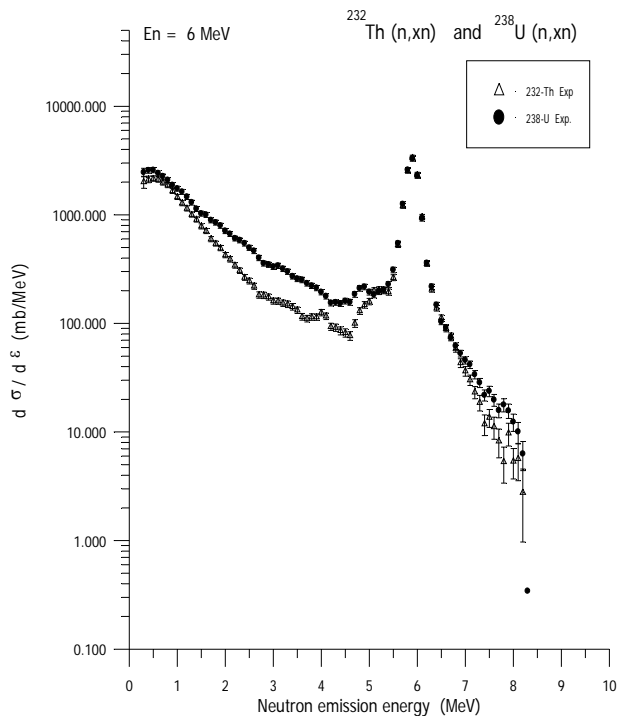


Fig. 3.

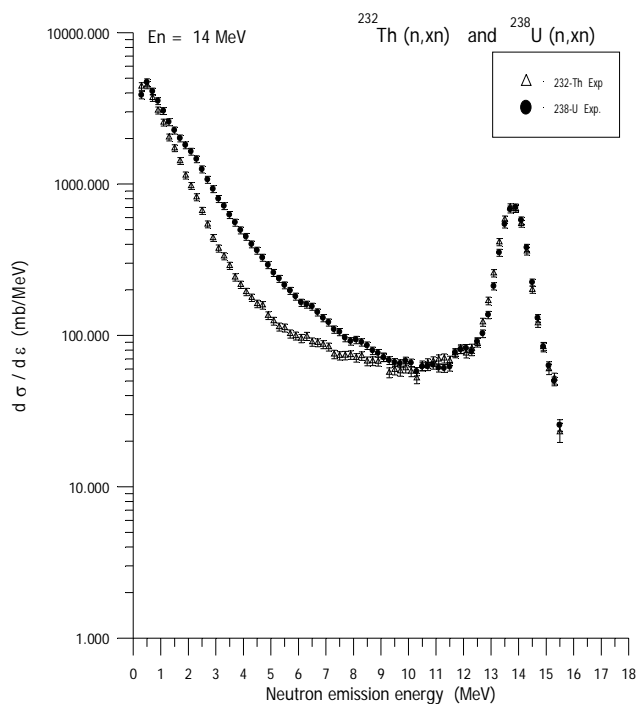


Fig. 4.

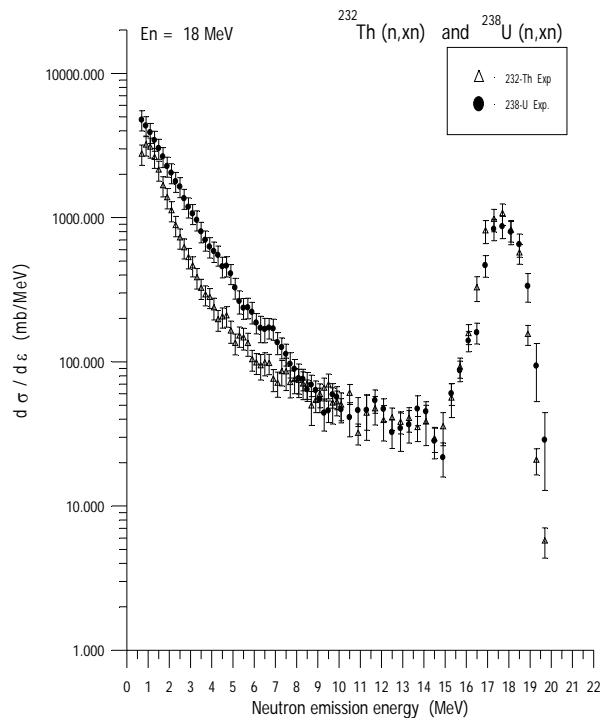


Fig. 5.

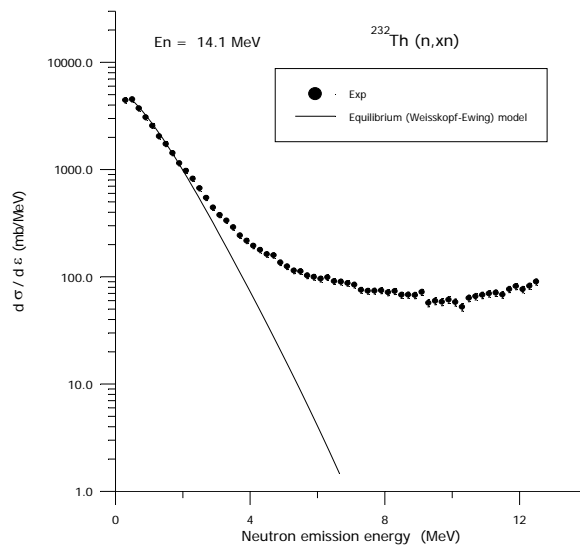


Fig. 6.

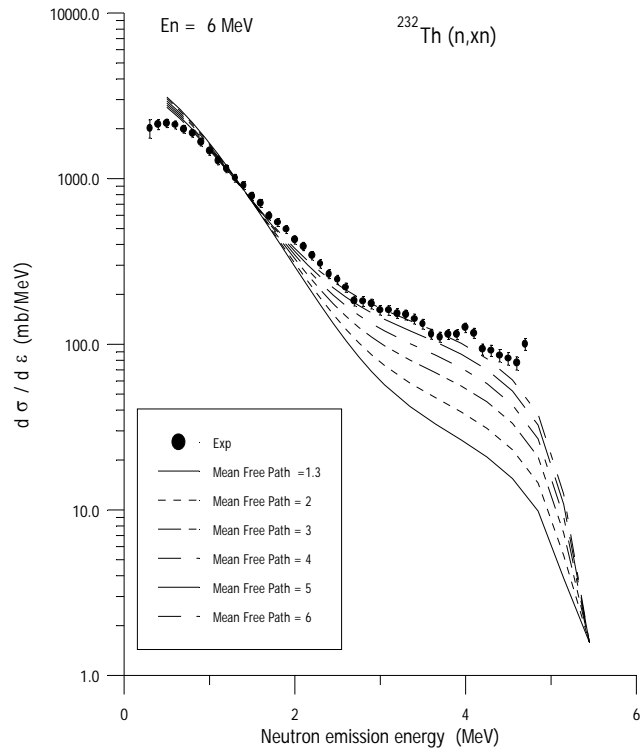


Fig. 7

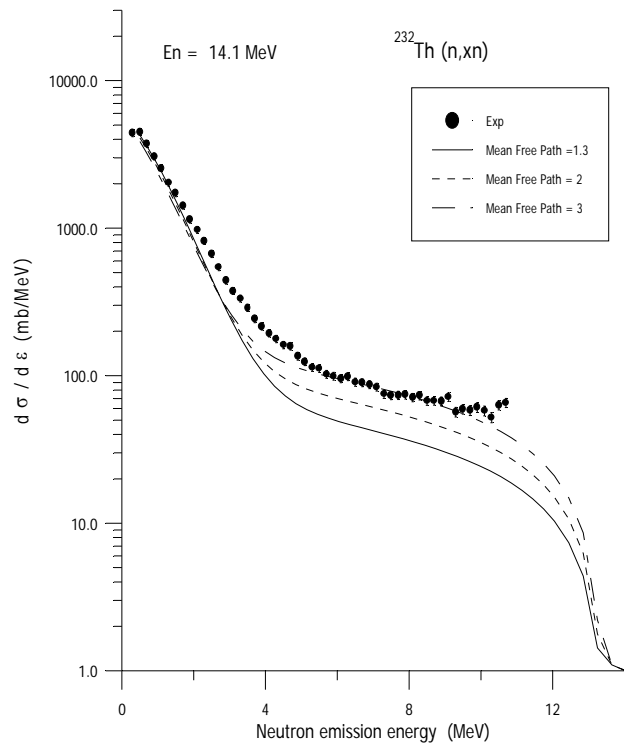


Fig. 8

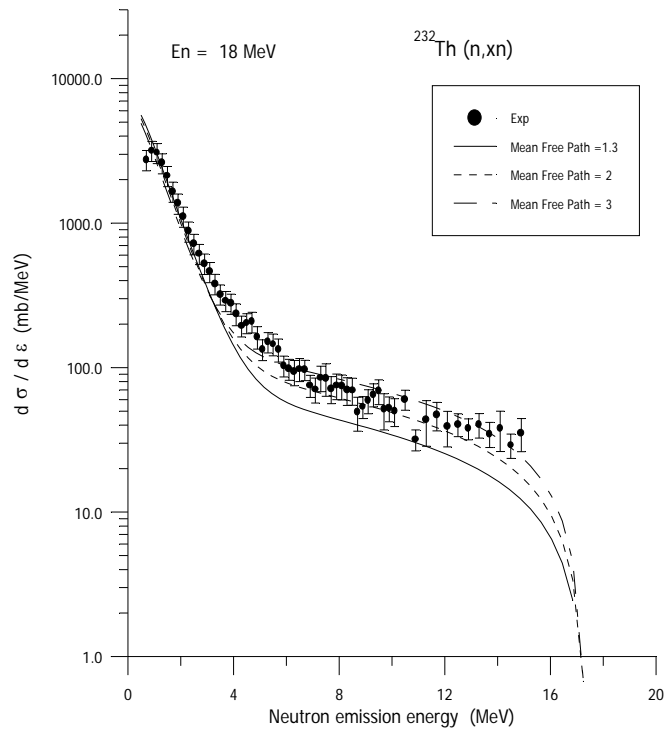


Fig. 9

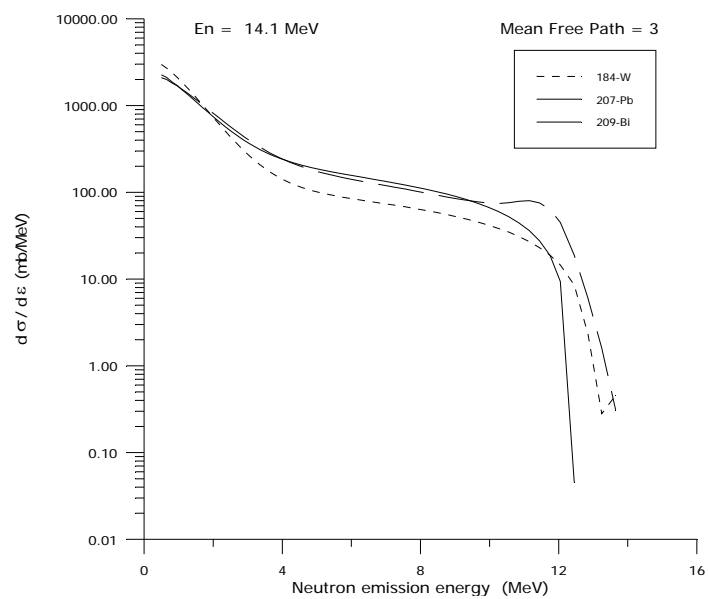


Fig. 10

Figure Captions

Fig. 1 The experimental values of the (n,f) reaction cross-sections of ^{232}Th . Experimental values were taken from Ref. Mclane, 1997.

Fig. 2 The experimental values of the (n,f) reaction cross-sections of ^{238}U . Experimental values were taken from Ref. Mclane, 1997.

Fig. 3 The comparison of the measured neutron spectra from the reaction of $^{232}\text{Th} + n$ and $^{238}\text{U} + n$ at 6 MeV. Experimental values were taken from Refs. Baba, 1989; Mclane, 1997.

Fig. 4 The comparison of the measured neutron spectra from the reaction of $^{232}\text{Th} + n$ and $^{238}\text{U} + n$ at 14 MeV. Experimental values were taken from Refs. Baba, 1989; Mclane, 1997.

Fig. 5 The comparison of the measured neutron spectra from the reaction of $^{232}\text{Th} + n$ and $^{238}\text{U} + n$ at 18 MeV. Experimental values were taken from Refs. Matsuyama, 1990; Mclane, 1997.

Fig. 6 The comparison of the calculated and measured neutron spectra from the reaction of $^{232}\text{Th} + n$ at 14.1 MeV. Experimental values were taken from Refs. Matsuyama, 1990; Mclane, 1997.

Fig. 7 The comparison of the calculated and measured neutron spectra from the reaction of $^{232}\text{Th} + n$ at 6 MeV. Experimental values were taken from Refs. Matsuyama, 1990; Mclane, 1997.

Fig. 8 The comparison of the calculated and measured neutron spectra from the reaction of $^{232}\text{Th} + n$ at 14.1 MeV. Experimental values were taken from Refs. Matsuyama, 1990; Mclane, 1997.

Fig. 9 The comparison of the calculated and measured neutron spectra from the reaction of $^{232}\text{Th} + n$ at 18 MeV. Experimental values were taken from Refs. Matsuyama, 1990; Mclane, 1997.

Fig. 10 The comparison of the calculated neutron spectra from the reaction of $^{184}\text{W} + n$, $^{207}\text{Pb} + n$ and $^{209}\text{Bi} + n$ at 14.1 MeV.

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