TRANSPORT CALCULATION OF THE NEUTRON MULTIPLICITY MOMENTS WITH ENERGY DEPENDENCE

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Abstract

In recent work we have developed a general one-speed transport theory model for the calculation of the multiplicity moments, used in nuclear safeguards. Quantitative results were obtained for spheres, cylinders of various shapes, and shells. In all these works, similarly to the point model, the only neutron reaction was assumed to be fission. Since the quantitative results for highly multiplicative systems were not in agreement with recent experiments, we extended the model to include isotropic elastic scattering without energy loss of the neutrons. In the present work, we generalise the model further, to include inelastic scattering, which requires the introduction of energy dependence and anisotropic scattering. In the paper, the significance of the elastic scattering, as well as the need for including inelastic scattering is demonstrated, and the extension to energy dependent transport theory is described and illustrated with one example.

1 Introduction

The general one-speed transport theory of the multiplicity moments for arbitrary geometries was elaborated recently as a first step of calculating the multiplicity moments beyond the point model [1]. The arising integral equations for the factorial moments of the neutrons leaving the item, induced by a single neutron as well as by a source event (spontaneous fission or (α, n) event) were solved numerically with a collision number expansion. Quantitative results were obtained in simple geometries with high order of symmetries, i.e. spheres [1], cylinders of various aspect ratios [2], and shells [3]. Both spatially distributed and localised sources were treated. The extension to spherical shells and localised sources was motivated by recent measurements on the Rocky Flats Shells during the MUSIC campaign conducted by Los Alamos National Laboratory and assisted by the University of Michigan [4, 5, 6]. It was shown that in all cases, the space-dependent model predicted larger factorial moments, and also larger fission rates, than the corresponding point model.

However, when comparing the results of the space-dependent model with the measurements, the calculated values were significantly smaller (in magnitude) than the measurements. It was clear that the relatively moderate increase of the space dependent model as compared to the point model was not sufficient to reconstruct the measured results.

One natural suggestion to explain the large differences between the measured and calculated results was that similarly to the point model, in the one-speed space-dependent model that far, the only reaction the neutrons were supposed to undergo was fission. All other reactions, such as absorption and scattering, were neglected. Neglecting absorption is fully justified due to its extremely small cross section at energies of the fission neutrons. On the other hand, elastic scattering has a cross section about three to four times as large as that of fission, depending on the fissile isotope and the neutron energy. In the point model, elastic scattering cannot be taken into account, but a space-dependent theory can account for scattering. Since scattering changes the direction of the neutrons, it will affect the leakage multiplication (basically acting as a reflector). Especially in highly multiplicative systems, close to critical, the presence of scattering can influence the results substantially.

Therefore we extended the model to include elastic scattering. In case of elastic scattering of neutrons on heavy elements, one can disregards the slight anisotropy and energy loss of neutrons, this extension is therefore quite straightforward [3]. The quantitative results were now in much better agreement with the measurements. However, the good agreement was possible to achieve only by accounting for inelastic scattering in an implicit and heuristic way, suggested in Ref. [7] for criticality calculations . Namely, instead of using the cross sections and the induced fission multiplicities at the average energy 2 MeV of source neutrons, all nuclear data used corresponded to 1 MeV, the average energy of neutrons after one inelastic scattering event.

Even though by this procedure, the agreement with the measurements was surprisingly good, this is neither a final or a perfect solution. Instead of emulating the effect of inelastic scattering by a one-speed theory with a "tuned" energy, the proper energy dependence of the fisison chain in the item should be taken into account. Also, the agreement achieved in this particular case between measurements and calculations was good, but not perfect, and it could also be improved

To this order, in this paper we extend the formalism to include inelastic scattering. This necessitates also the inclusion of anisotropic scattering, since the angular distribution of elastically scattered neutrons is slightly anisotropic. Introducing energy dependence requires the treatment of energy dependent cross sections and scattering functions, as well as energy dependent fission neutron number factorial moments. In the following the conceptual theory of multiplicity with energy dependence and inelastic scattering will be given, and a simple illustration of the effect of the energy dependence is given.

2 General considerations

First we recapitulate the essential formalism of the one-speed theory and its extension to include elastic scattering. For simplicity, (α, n) reactions will be omitted (corresponding to pure metallic items) and the formalism will be applied for a solid sphere. Extension to cylinders or spheres is a mere technicality.

The starting point is a master equation for the generating function $g(z|r, \mu)$ of the single particle induced probability distribution $p(n|r, \mu)$ of n neutrons leaving the item by a starting neutron with coordinates (r, μ) [1, 3]. It was also shown in the cited publications that due to isotropic emission of neutrons both from spontaneous and induced fission, it is sufficient to handle only the angularly integrated ("scalar") generating function g(z|r), and its moments. The equation for the scalar generating function reads as

$$g(z|r) = \frac{z}{2} \int_{-1}^{1} d\mu \, e^{-\ell(r,\mu)} + \frac{1}{2} \int_{-1}^{1} d\mu \, \int_{0}^{\ell(r,\mu)} ds \, e^{-s} \, q_f \left[g(z|r'(s)) \right]. \tag{1}$$

Here r'(s) is the radial position from r along the direction μ at a path length s, $q_f(z) = \sum_{k=0}^{\infty} f_k z^k$ is the generating function of the number distribution f_k of the induced fission

neutrons, the quantity in the square brackets being its argument, and $\ell(r, \mu)$ is the distance to the boundary of the sphere from point r along the direction μ . All distances are in dimensionless optical units, i.e. in units of the mean free path.

The equations for the first three factorial moments n(r), m(r) and w(r) of the number of neutrons leaving the item for one single neutron starting from the radial position r and with isotropic angular distribution read as follows. For the first moment one has

$$n(r) = n_0(r) + \frac{\nu_{f,1}}{2} \int_{-1}^1 d\mu \int_0^{\ell(r,\mu)} ds \, e^{-s} \, n(r'(s)).$$
⁽²⁾

with

$$n_0(r) = \frac{1}{2} \int_{-1}^1 e^{-\ell(r,\mu)} \,\mathrm{d}\mu,\tag{3}$$

 $\nu_{f,1}$ being the mean number of neutrons emitted in induced fission. For the second moment one has,

$$m(r) = A(r) + \frac{\nu_{f,1}}{2} \int_{-1}^{1} d\mu \int_{0}^{\ell(r,\mu)} ds \, e^{-s} \, m(r'(s)) \tag{4}$$

with

$$A(r) = \frac{\nu_{f,2}}{2} \int_{-1}^{1} \mathrm{d}\mu \int_{0}^{\ell(r,\mu)} \mathrm{d}s \, e^{-s} \, n^{2}(r'(s)), \tag{5}$$

and for the third moment

$$w(r) = B(r) + \frac{\nu_{f,2}}{2} \int_{-1}^{1} \mathrm{d}\mu \, \int_{0}^{\ell(r,\mu)} \mathrm{d}s \, e^{-s} \, w(r'(s)), \tag{6}$$

where B(r) is defined as

$$B(r) = \frac{1}{2} \int_{-1}^{1} d\mu \int_{0}^{\ell(r,\mu)} ds \, e^{-s} \left\{ \nu_{f,3} \, n^{3}(r'(s)) + 3 \, \nu_{f,2} \, n(r'(s)) \, m(r'(s)) \right\}. \tag{7}$$

What regards the statistics of the number of neutrons leaving the sample due to a spatially homogeneously distributed isotropic source event (spontaneous fission), for its generating function G(z) one has the expressions

$$G(z) = \frac{3}{R^3} \int_0^R r^2 q_s \left[g(z|r) \right] dr,$$
(8)

where $q_s(z)$ is the generating function of the number of neutrons per spontaneous fission. The first moment N (expectation) of the number of neutrons emitted from the item due to a source emission event is obtained from G(z) as

$$N = \frac{3\nu_{s,1}}{R^3} \int_0^R r^2 n(r) \,\mathrm{d}r \tag{9}$$

where $\nu_{s,1}$ is the first moment (expectation) of the number of neutrons emitted in a source event (spontaneous fission). For the second (M) and third (W) moments one has

$$M = \frac{3}{R^3} \int_0^R r^2 \left\{ \nu_{s,2} n^2(r) + \nu_{s,1} m(r) \right\} \,\mathrm{d}r \tag{10}$$

and

$$W = \frac{3}{R^3} \int_0^R r^2 \left\{ \nu_{s,3} n^3(r) + 3 \nu_{s,2} n(r) m(r) + \nu_{s,1} w(r) \right\} \, \mathrm{d}r,\tag{11}$$

respectively. It is these formulae which will be compared with their energy dependent counterparts.

3 Accounting for elastic scattering in a one-speed model

Elastic scattering can be simply included into the one-speed transport theory model if the small energy loss and slight anisotropy of the neutrons scattered elastically on heavy nuclei is neglected. In that case, scattering can be treated as a fission event resulting in one neutron. This simply means to replace the probability distribution $p_f(k)$ with the number distribution $p_r(k)$ of the neutrons arising from a *reaction* (either fission or scattering), such as

$$p_r(k) = c_f \, p_f(k) + c_{el} \, \delta_{k,1} \tag{12}$$

where

$$c_f \equiv \frac{\Sigma_f}{\Sigma_T}; \quad c_{el} \equiv \frac{\Sigma_{el}}{\Sigma_T} \quad \text{and} \quad \Sigma_T = \Sigma_f + \Sigma_{el}.$$
 (13)

This means that the moments $\nu_{f,i}$, i = 1, 2, 3 of induced fission have to be replaced by the moments $\nu_{r,i}$ of the number of secondaries per reaction:

$$\nu_{r,i} = c_f \,\nu_{f,i} + c_{el} \,\delta_{k,1} \tag{14}$$

Correspondingly, the optical path has to be scaled by the total cross section, instead of the fission cross section as in the preceding work.

A comparison between the preliminary results of the MUSIC measurements on the Rocky Flats shells and the calculations with elastic scattering included [3] showed an improved agreement, but it was still not satisfactory. The most obvious reason for the disagreement was that the effect of inelastic scattering was not taken into account. Inelastic scattering has approximately the same macroscopic cross section as elastic scattering, both of them about three times as large as that of fission at 2 MeV. Inelastic scattering has the effect that the mean energy of inelastically collided neutrons will be lower than the source energy, and at the lower energies, both the ratio of the scattering to fission cross section, as well as the induced fission multiplicities are different (essentially leading to lower multiplication).

In order to account in a rough way for the presence of inelastic scattering, we followed the recommendation of Ref. [7], namely to perform the one-speed calculations with cross sections and fission neutron multiplicities corresponding to 1 MeV. The results of such calculations, and a comparison with the MUSIC measurements, taken from [3], is shown in Fig. 1. The calculated results when elastic scattering is not accounted for are also shown. Fig. 1 demonstrates that a one-speed transport model, which accounts for elastic scattering nearly exactly, and for inelastic scattering in an empirical way, yields good agreement with the measurements. The Figure also shows that by not accounting for scattering, the difference between calculations and measurements is much larger.



Figure 1: Comparison of measured and calculated first, second and third moments of the number of neutrons emitted from the Rocky Flats Shells for four different outer radii. In the calculations, the material properties corresponding to those of the isotopic composition of the Rocky Flats Shells (93.5% enriched 235U) were used at the neutron energy of 1 MeV

4 Accounting for inelastic scattering

It is clear that, despite the relative good agreement between calculations and experiments, the approximate way of accounting for elastic scattering, and in particular the empirical way of accounting for inelastic scattering is not fully satisfactory. In order to describe the effect of these processes properly, one needs to extend the model to include energy dependence. This will be described below.

4.1 Single particle induced distributions and moments

We need to introduce the energy dependent cross sections

$$\Sigma_f(E), \qquad \Sigma_{el}(E) \quad \text{and} \quad \Sigma_{in}(E)$$
 (15)

and the total cross section $\Sigma_T(E) \equiv \Sigma(E)$ as

$$\Sigma(E) = \Sigma_f(E) + \Sigma_{el}(E) + \Sigma_{in}(E)$$
(16)

as well as the energy dependent fission number distribution $p_f(n, E)$. Further, the scattering densities for both the elastic and inelastic scattering, as well as for the fission process, are needed:

$$f_{el}(\mu \to \mu', E \to E'), \qquad f_{in}(\mu \to \mu', E \to E') \quad \text{and} \quad \chi(E \to E')$$
(17)

Unlike in ordinary transport theory, the elastic and inelastic and fission scattering functions cannot be combined into one common scattering function, because they have different number distributions¹. Putting it another way, unlike the energy-independent case (Sect. 3), the elastic (and inelastic) scattering number distributions cannot be accounted for by modifying the fission neutron number distributions, because they have different scattering functions.

The specific forms of the elastic and inelastic scattering functions are as follows. For the elastic scattering, assuming energy conservation with scattering on free nuclei with atomic number A and isotropy in the center of mass system, the scattering function of elastic scattering can be written as [8]:

$$f_{el}(\mu \to \mu', E \to E') = \frac{\Delta(E' - \alpha E)}{(1 - \alpha) E} \delta\left(\mu' - S(E, E')\right); \qquad E' \le E$$
(18)

with

$$\alpha = \left[\frac{A-1}{A+1}\right]^2 \quad \text{and} \quad S = \frac{1}{2}\left[(A+1)\sqrt{\frac{E'}{E}} - (A-1)\sqrt{\frac{E}{E'}}\right] \tag{19}$$

On the other hand, similarly to fission neutrons, the angular distribution of inelastic scattering from heavy nuclei can be regarded isotropic in the laboratory system. Thus,

$$f_{in}(\mu \to \mu', E \to E') = \frac{1}{2} f_{in}(E \to E'); \qquad E' \le E$$
 (20)

Because of the separate treatment of the scattering and fission reactions, it is no longer practical to use optical units, hence the corresponding cross sections will appear in the equations. In addition, due to the special form (18) of the elastic scattering function, it is not possible to write down an equation directly to the scalar quantities, one has to keep the angular form, and calculate the scalar quantities only after the solution is found. By using (18) and (20) and by performing the angular integrals in each term on the right hand side, the master equation for the angular, energy dependent generating function will read as

$$g(z|r, \mu, E) = z e^{-\ell(r,\mu)\Sigma(E)} + \frac{\Sigma_{el}(E)}{(1-\alpha)E} \int_{0}^{\ell(r,\mu)} ds e^{-s\Sigma(E)} \int_{\alpha E}^{E} dE' g(z|r'(s), S(E, E'), E') + \Sigma_{in}(E) \int_{0}^{\ell(r,\mu)} ds e^{-s\Sigma(E)} \int_{0}^{E} dE' f_{in}(E \to E') g(z|r'(s), E') + \Sigma_{f}(E) \int_{0}^{\ell(r,\mu)} ds e^{-s\Sigma(E)} q_{f} \left[\int_{0}^{E_{max}} dE' \chi(E \to E') g(z|r'(s), E') \right]$$
(21)

where, similarly to the one-speed case,

$$g(z|r, E) = \frac{1}{2} \int_{-1}^{1} d\mu' g(z|r, \mu E)$$
(22)

is the "scalar" (angularly integrated) generating function.

¹The elastic and inelastic scattering could be combined together, but it would not have advantages, because then the specific properties of these scattering functions could not be utilized.

The equations for the factorial moments can be derived from Eq. (21) in the usual way. The first moment, i.e. mean number of neutrons $n(r, \mu, E)$ leaving the item obeys the equation

$$n(r, \mu, E) = e^{-\ell(r,\mu)\Sigma(E)} + \frac{\Sigma_{el}(E)}{(1-\alpha)E} \int_{0}^{\ell(r,\mu)} ds \, e^{-s\,\Sigma(E)} \int_{\alpha E}^{E} dE' \, n(r'(s), S(E, E'), E') + \Sigma_{in}(E) \int_{0}^{\ell(r,\mu)} ds \, e^{-s\,\Sigma(E)} \int_{0}^{E} dE' \, f_{in}(E \to E') \, n(r'(s), E') +$$

$$\nu_{f,1}(E) \, \Sigma_{f}(E) \, \int_{0}^{\ell(r,\mu)} ds \, e^{-s\,\Sigma(E)} \, [\int_{0}^{E_{max}} dE' \, \chi(E \to E') \, n(r'(s), E') \equiv e^{-\ell(r,\mu)\Sigma(E)} + \widehat{\mathbb{M}}(r,\mu, E) \, n(r,\mu, E)$$

$$(23)$$

where the integral transport operator $\widehat{\mathbb{M}}(r,\mu,E)$ was introduced by the last equality. It is also indicated that the neutron multiplicities, and hence the factorial moments $\nu_{f,i}$ of the induced fission, are now energy dependent.

With the operator $\widehat{\mathbb{M}}(r, \mu, E)$, the equation for the second factorial moment $m(r, \mu, E) = \langle (n(r, \mu, E))((n(r, \mu, E) - 1)) \rangle$ reads as

$$m(r, \mu, E) = A(r, \mu, E) + \widehat{\mathbb{M}}(r, \mu, E) m(r, \mu, E)$$
(24)

with $A(r, \mu, E)$ already being known from the solution for the first moment:

$$A(r,\mu,E) = \nu_{f,2}(E) \Sigma_f(E) \int_0^{\ell(r,\mu)} \mathrm{d}s \, e^{-s \, \Sigma(E)} \left[\int \mathrm{d}E' \, \chi(E \to E') \, n(r'(s),E') \right]^2 \tag{25}$$

The equation for the third factorial moment $w(r, \mu, E)$ can be written down in an analogous manner as

$$w(r, \, \mu, \, E) = B(r, \mu, E) + \widehat{\mathbb{M}}(r, \, \mu, \, E) \, w(r, \, \mu, \, E)$$
(26)

with

$$B(r,\mu,E) = \Sigma_{f}(E) \int_{0}^{\ell(r,\mu)} \mathrm{d}s \, e^{-s \,\Sigma(E)} \Big\{ \nu_{f,3}(E) \left[\int \mathrm{d}E' \,\chi(E \to E') \, n(r'(s),E') \right]^{3} + 3 \,\nu_{f,2}(E) \left[\int \mathrm{d}E' \,\chi(E \to E') \, n(r'(s),E') \right]^{2} \left[\int \mathrm{d}E' \,\chi(E \to E') \, m(r'(s),E') \right] \Big\}$$
(27)

The equations for all moments can be numerically solved with the same collision number expansion technique as in the previous publications for the simpler cases. Compared to the one-speed calculations for a sphere, the difference is the appearance of one more parameter, the energy E in the equations, with one more nested loop of integrals in the collision number expansion. In principle, in computing effort this is equal with the calculations made for a cylinder [2], where in addition to the radial position r and the cosine μ of the polar angle, also the azimuthal angle φ appeared.

The bigger challenge will be to incorporate the energy dependent quantities, such as cross sections, scattering functions, which are only available in tabulated form, and the induced fission factorial moments $\nu_{f,i}(E)$. The precise values of these, or rather the probability distributions from which these are calculated, are found in the literature with a sparse spacing in energy (1 MeV). Therefore, most likely approximative analytical formulae will have to be used for the energy dependent number distributions to calculate the latter with sufficient fine energy mesh.

4.2 Source emission induced generating function

The generating function G(z) of the number distribution of neutrons leaving the item for a source event can easily be obtained from the generating function of the single-neutron induced generating function by quadrature. In the energy dependent case, assuming isotropic angular distribution of the source neutrons, one needs to account for the energy dependence of the source neutrons, through the (normalised) energy spectrum $\chi_s(E)$. Hence, in the energy dependent case one has

$$G(z) = \frac{1}{V} \int_{V} \mathrm{d}\boldsymbol{r} \, q_{s} \left[\int g(z \, | \boldsymbol{r}, E) \, \chi_{s}(E) \, \mathrm{d}E \right]$$
(28)

It is easy to show that for the moments N, M and W of G(z), this means that the single particle induced moments n(r), m(r) and w(r) in the energy dependent case have to be replaced by their fission energy spectrum weighted integrals when energy dependence is taken into account. Define the spontaneous fission spectrum weighted single particle induced factorial moments as

$$\overline{n}(r) \equiv \int n(r, E) \,\chi_s(E) \,\mathrm{d}E; \qquad \overline{m}(r) \equiv \int m(r, E) \,\chi_s(E) \,\mathrm{d}E$$
and
(29)

$$\overline{w}(r) \equiv \int w(r, E) \,\chi_s(E) \,\mathrm{d}E.$$

Then, for a spherical item, we obtain expressions in perfect analogy with those of the one-speed case, Eqs (9) - (11) as

$$N = \frac{3\nu_{s,1}}{R^3} \int_0^R r^2 \,\overline{n}(r) \,\mathrm{d}r,$$
(30)

$$M = \frac{3}{R^3} \int_0^R r^2 \left\{ \nu_{s,2} \,\overline{n}^2(r) + \nu_{s,1} \,\overline{m}(r) \right\} \,\mathrm{d}r \tag{31}$$

and

$$W = \frac{3}{R^3} \int_0^R r^2 \left\{ \nu_{s,3} \,\overline{n}^3(r) + 3 \,\nu_{s,2} \,\overline{n}(r) \,\overline{m}(r) + \nu_{s,1} \,\overline{w}(r) \right\} \,\mathrm{d}r. \tag{32}$$

In order to evaluate (30) - (32), one only needs the energy dependent single neutron induced moments n(r, E), m(r, E) and w(r, E), as well as the spontaneous fission energy spectrum $\chi_s(E)$.

5 Results

We illustrate the use of (30) - (32) in a simple example. We consider a spherical ²³⁹Pu item, with a spontaneous fission source of ²⁴⁰Pu. However, instead of using the true energy dependent single-particle induced moments, we perform calculations of five one-speed cases at the energies E = 0.5, 1, 2, 3 and 4 MeV, with elastic scattering included without energy loss of the neutrons, as it was described in Section 3. These energies cover the significant part of the ²⁴⁰Pu spectrum reasonably well. Inelastic scattering is not included. There is no coupling between the one-speed equations for different energies, the only circumstance which distinguishes the equations is that each has its own fission and scattering cross section at the particular energy selected, as well as the induced fission multiplicities at the same energies. The cross sections were taken from the ENDF/B-VIII library, whereas the ²⁴⁰Pu spectrum and the energy dependent factorial moments $\nu_{f,i}(E)$ of induced fission were taken from runs with MCNPX-PoliMi [9].

The energy averaged values of the single particle induced factorial moments $\overline{n}(r)$, $\overline{m}(r)$ and $\overline{w}(r)$ of Eq. (29) were calculated by a simple weighted average with weights

$$c_{i} = \frac{\chi_{s}(E_{i})}{\sum_{i=1}^{5} \chi_{s}(E_{i})}$$
(33)

and these were then used in (30) - (32) to calculate the factorial moments of the source event induced emission. Two different spherical items were considered, one with R = 2.5 cm, and another one with R = 3.2 cm. The results are shown in Table 1.

	m R=2.5~cm			m R=3.2~cm		
E [MeV]	Ν	М	W	N	М	W
0.5	3.31	19.45	243.9	4.10	42.33	1081
1	3.49	23.53	353.5	4.43	55.26	1757
2	4.00	39.41	953.1	5.72	136.6	9057
3	4.11	44.22	1204	6.07	172.8	13955
4	4.19	48.72	1465.4	6.36	208.5	19709
Average	3.67	29.87	620.2	4.94	92.9	5682

Table 1: Factorial moments for five one-speed calculations, and the energy-averaged moments according to Eqs (30) - (32) for two different item sizes.

It is seen that the factorial moments calculated by accounting for the energy distribution of the spontaneous fission neutrons in an approximate, but still representative way, lie between those of the one speed calculations with 1 and 2 MeV, respectively, for both item sizes (coloured with yellow in the Table). Although for different reasons, this points towards the same conclusion as was stated in Refs [3] and [7], that when performing one-speed calculations, one gets results closer to the real one if instead of the average neutron energy of the fission spectrum, 2 MeV, one uses data corresponding to a lower energy. The main reason is of course the presence of inelastic scattering, which so far has not been included in our quantitative work. The calculations with full energy dependence are ongoing.

6 Conclusions

The general theory of the multiplicity moments in a full space-angle-energy dependent setting was presented, which is suitable to incorporate elastic and inelastic scattering with anisotropic scattering. Work is going on to obtain quantitative solutions in a full spaceenergy-angle-dependent description, where both elastic and inelastic scattering are included without simplifications. The model will supply quantitative result of sufficient high fidelity, that it will be possible to use it to generate training data for an artificial neural network (ANN) to unfold the parameters of an unknown item, in particular both the ²³⁹Pu and the ²⁴⁰Pu content simultaneously. Work is going on in this directions, even if at present only at the level of one-speed theory [10]

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