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A Method to Calculate Reactivity Worth by

Integral Transport Theory+)

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1. Introduction

Material worth measurements belong to the standard experiments in fast critical assemblies. A sample of the material under study is substituted for some standard material (e.g. fuel, aluminum, or void), mostly in the center of the reactor, and the change in reactivity is measured. These experiments can be carried out with good precision. However, the methods used so far for the interpretation are rather inadequate.

Most of the experimental results are compared to calculations by first order perturbation theory. If the concept of composition-dependent resonance self-shielding factors, as described by Abagyan et al. /1/, is used, a first order perturbation (FOP) calculation is carried out in the following way: A composition is defined, which contains core material plus a small amount of the sample material. The reactivity worth of this composition versus core material is calculated. Obviously, the self-shielding factors involved are those of the core material, and the results are valid essentially for an infinitely thin sample in the homogeneous core.

The samples used in actual measurements are usually plates, mostly with a thickness of less than 1 cm. Thus the optical thickness is about 1/5 mean free paths outside the resonances, but many mean free paths in a large resonance. There are two types of self-shielding effects which cause a dependence of the material worth per unit mass on the thickness of the sample.

One is the flux depression in an absorber or flux peak in a fissile material ("self multiplication") at energies where the cross sections are smooth. The other one is the dependence of the resonance self-shielding on the geometry of the sample.

Several authors have worked out corrections for the first effect /2,3/ using a collision probability formalism. However, they cannot account for the second effect, which in many samples is at least as large as the first one.

In the present report a method of analysis will be described which takes both effects into account. It will be called "Integral Transport Perturbation" Method (ITP) because it is based on a perturbation solution of the integral transport equation. The solution will be given for a 2-zone-configuration which consists of the sample and the surrounding core. The method was worked out only for the center of the reactor, where there is no leakage contribution to the reactivity worth. The escape probability from the sample is calculated using an improved rational approximation

$$1 - P(E) = \frac{1}{1 + \frac{1}{a} \Sigma_{t}(E)}$$
(1)

which was first suggested by Levine /4/. 1 is the mean chord length, and a is a geometry dependent factor. This approximation is usually satisfactory if the factor a is suitably chosen; it could be easily further improved. More basic limitations of the method are that, being a perturbation method, it is not valid for very large samples, and that no account is taken of the heterogeneous structure of the surrounding core.

2. Outline of the Theory

The theory is described in some detail in /5/, and will only be briefly outlined here. We start from the integral transport equation which reads in the multigroup formalism

$$S_{i}(\mathbf{r}) = \sum_{j \leq i} \int d\mathbf{V}' S_{j}(\mathbf{r}') \left\langle \frac{\Sigma^{j \rightarrow i}}{\Sigma_{t}} (\mathbf{r}, E) p(\mathbf{r}' \rightarrow \mathbf{r}, E) \right\rangle_{j} + \chi_{i} \sum_{j} \int d\mathbf{V}' S_{j}(\mathbf{r}') \left\langle \frac{\nabla \Sigma_{f}}{\Sigma_{t}} (\mathbf{r}, E) p(\mathbf{r}' \rightarrow \mathbf{r}, E) \right\rangle_{j}$$
(2)

The variable $S_i(r)$ is the emission density of neutrons per cm³ and sec, $p(r' \rightarrow r, E)$ is the probability that a neutron born in r' will suffer its next collision in r.

The reactivity change upon insertion of a sample in the reactor can be calculated using a perturbation solution of equation (2) for a configuration which consists of the sample and the surrounding core. The reactivity of the sample versus vacuum is given by two terms, the first of which reads

$$\frac{\delta k_{1}}{k} = \frac{v_{1}}{D} \sum_{ij} \left[(s_{i}^{+} - s_{j}^{+}) \left\langle F(E) \frac{\Sigma_{s1}^{j \to 1}}{\Sigma_{t1}} \right\rangle_{j} - s_{j}^{+} \delta_{ij} \left\langle F(E) \frac{\Sigma_{a1}}{\Sigma_{t1}} \right\rangle_{j} + \chi_{i} s_{i}^{+} \left\langle F(E) \frac{v\Sigma_{f1}}{\Sigma_{t1}} \right\rangle_{j} \right]$$
(3)

where F(E), the collision density in the sample, is given by

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$$F(E) = P(E)S_{1j}^{2} + (1-P) \frac{\Sigma_{t1}}{\Sigma_{t2}} S_{2j}$$
(4)

The second contribution to the reactivity is

$$\frac{\delta k_{2}}{k} = \frac{V_{1}}{D} \sum_{i,j} \left\langle (1-P) \left(s_{1j} - \frac{\Sigma_{t1}}{\Sigma_{t2}} s_{2j} \right) \left[\left(s_{i}^{+} - s_{j}^{+} \right) \frac{\Sigma_{s2}^{j+i}}{\Sigma_{t2}} - s_{j}^{+} \delta_{ij} \frac{\Sigma_{a2}}{\Sigma_{t2}} + \chi_{i} s_{i}^{+} \frac{\nabla\Sigma_{f2}}{\Sigma_{t2}} \right] \right\rangle_{j} + \frac{V_{1}}{D} \sum_{j} \chi_{j} \sum_{i} \left[\left(s_{i}^{+} - s_{j}^{+} \right) \left\langle \frac{\Sigma_{s2}^{j+i}}{\Sigma_{t2}} \right\rangle_{j} - s_{j}^{+} \delta_{ij} \left\langle \frac{\Sigma_{a2}}{\Sigma_{t2}} \right\rangle_{j} + \chi_{i} s_{i}^{+} \left\langle \frac{\nabla\Sigma_{f2}}{\Sigma_{t2}} \right\rangle_{j} \right] \left(5 \right)$$

The nomenclature used in these equations is

^S 1j ^{,S} 2j	emission density of neutrons in the sample (core) in energy group j
s ⁺ j	adjoint emission density for the unperturbed case (sample volume void)
v ₁	sample volume
D	perturbation integral
subscripts	

1	sample
2	core

Whereas term (3) describes the effect connected with the reaction rates in the sample, term (5) obviously takes account of the reaction rates in the surrounding core. The theory gives $X_i=0$ in equation (5); this case shall be discussed first. The expression in the square brackets is just the adjoint equation. Thus, if the cross sections are free of resonances, the expression (5) is zero. On the other hand, if a sample with a purely non-resonant cross section is measured in a core that contains resonance absorbers, the average over resonances includes a factor $1/\Sigma_{t2}(E)$, so that (5) is non-zero. This is obviously the effect that the sample reduces self-shielding for the resonance absorbers in the core. If the cross section of the sample material has resonances too, (5) describes the fact that neutrons leaving the sample have a different resonance structure, and therefore different reaction probabilities, from those neutrons moderated in the core. It should be noted that (5) is unchanged if the quantities X; take on any arbitrary values, because they are multiplied by zero. The X, will be chosen later on so as to simplify the calculations.

The procedure is to define in a first step "effective" perturbation cross sections from equations (3) and (5), and to evaluate them. In a second step, these perturbation cross sections are used together with fluxes and adjoints taken from a diffusion calculation to obtain central reactivity worths by a standard perturbation calculation.

The effective perturbation cross sections are obtained from (3) and (5) if the reaction rates are written as "perturbation cross section times flux"; thus, for the reaction type x, equation (3) leads to the perturbation cross section

$$\Sigma_{xj}^{(1)} \Phi_{j} = \left\langle (PS_{1j}^{+} + (1-P) \frac{\Sigma_{t1}}{\Sigma_{t2}} S_{2j}) \frac{\Sigma_{x1}}{\Sigma_{t1}} \right\rangle_{j}$$
(6)

and equation (5) leads to

$$\Sigma_{xj}^{(2)} \Phi_{j} = \left\langle (1-P) \left(S_{1j} - \frac{\Sigma_{t1}}{\Sigma_{t2}} S_{2j} \right) \frac{\Sigma_{x2}}{\Sigma_{t2}} \right\rangle_{j} + X_{j} \left\langle \frac{\Sigma_{x2}}{\Sigma_{t2}} \right\rangle_{j}$$
(7)

In order to evaluate these expressions, the unperturbed group fluxes Φ_j and the emission densities $S_{2j} = \Phi_j / (1/\Sigma_t 2)_j$ are needed. They are obtained from a zero-dimensional calculation.

Furthermore, one needs the perturbed emission densities S_{1j}. They are obtained from the equations

$$S_{1i} = \sum_{j} \left\langle \frac{\sum_{s1}^{j \rightarrow i} + x_{i} \nu \sum_{f1}}{\sum_{t1}} P(E) \right\rangle_{j} S_{1j} + \sum_{j} \left\langle \frac{\sum_{s1}^{j \rightarrow i} + x_{i} \nu \sum_{f1}}{\sum_{t2}} (1-P) \right\rangle_{j} S_{2j}$$
(8)

by an iterative procedure, which converges very fast.

If the rational approximation (1) (where 1 will be written for 1/a) is introduced, one obtains

$$\Sigma_{xj}^{(1)} = \left\langle \frac{\Sigma_{x1}}{\Sigma_{t1}+1/1} \right\rangle_{j} \frac{S_{1j}}{\Phi_{j}} + \left\langle \frac{\Sigma_{x1}}{1\Sigma_{t2}(\Sigma_{t1}+1/1)} \right\rangle_{j} \tilde{\Sigma}_{t2j}$$

$$\Sigma_{xj}^{(2)} = \left\langle \frac{\Sigma_{x2}}{1\Sigma_{t2}(\Sigma_{t1}+1/1)} \right\rangle_{j} \frac{S_{1j}}{\Phi_{j}} - \left\langle \frac{\Sigma_{t1}}{1\Sigma_{t2}^{2}(\Sigma_{t1}+1/1)} \right\rangle_{j} \tilde{\Sigma}_{t2j} + \frac{X_{j}}{\Phi_{j}} \left\langle \frac{\Sigma_{x2}}{\Sigma_{t2}} \right\rangle_{j}$$

$$\tilde{\Sigma}_{t2j} = \tilde{\Sigma}_{t2j}^{(2)} + \tilde$$

where Σ_{t2} is the self-shielded total cross section. According to /1/, the self-shielded cross sections are (in the "narrow resonance approximation") defined by

$$\tilde{E}_{x} = \frac{\langle \Sigma_{x} / \Sigma_{t} \rangle}{\langle 1 / \Sigma_{t} \rangle} , \quad \tilde{\Sigma}_{t} = \frac{1}{\langle 1 / \Sigma_{t} \rangle}$$

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It will now be demonstrated that in the limit of very small samples the perturbation cross section of the ITP, as defined by (9) and

$$\Sigma_{\rm x}^{\rm ITP} = \Sigma_{\rm x}^{(1)} + \Sigma_{\rm x}^{(2)},$$
 (9a)

is identical with the perturbation cross section obtained from FOP.

The FOP method, as described above (small amount of sample material added to the core material), leads to the following definition of the perturbation cross section

$$\varepsilon \Sigma_{\mathbf{x}}^{\text{FOP}} = \frac{\left\langle \frac{\Sigma_{\mathbf{x}2}^{+}\varepsilon\Sigma_{\mathbf{x}1}}{\Sigma_{\mathbf{t}2}^{+}\varepsilon\Sigma_{\mathbf{t}1}} \right\rangle}{\left\langle \frac{1}{\Sigma_{\mathbf{t}2}^{+}\varepsilon\Sigma_{\mathbf{t}1}} \right\rangle} - \frac{\left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{\mathbf{t}2}} \right\rangle}{\left\langle \frac{1}{\Sigma_{\mathbf{t}2}} \right\rangle}$$
(1o)

where ϵ is assumed to approach zero. This expression can be split up and written as follows

$$\varepsilon \Sigma_{\mathbf{x}}^{\text{FOP}} = \varepsilon \frac{\left\langle \frac{\Sigma_{\mathbf{x}1}}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle}{\left\langle \frac{1}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle} + \frac{\left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle}{\left\langle \frac{1}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle} - \frac{\left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{\mathbf{t}2}^{+\varepsilon}} \right\rangle}{\left\langle \frac{1}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle}$$
$$= \varepsilon \frac{\left\langle \frac{\Sigma_{\mathbf{x}1}}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle}{\left\langle \frac{1}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle} + \frac{\varepsilon}{\left\langle \frac{1}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle} \frac{d}{d\varepsilon} \left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle} - \frac{\varepsilon}{\left\langle \frac{1}{\Sigma_{\mathbf{t}2}} \right\rangle} \frac{d}{d\varepsilon} \left\langle \frac{1}{\Sigma_{\mathbf{t}2}^{+\varepsilon\Sigma}\mathbf{t}1} \right\rangle}$$
(10a)

If equation (10a) is carried to the limit, one obtains

$$\Sigma_{\mathbf{x}}^{\text{FOP}} = \tilde{\Sigma}_{t2} \left[\left\langle \frac{\Sigma_{x1}}{\Sigma_{t2}} \right\rangle - \left\langle \frac{\Sigma_{t1}\Sigma_{x2}}{\Sigma_{t2}^{2}} \right\rangle + \left\langle \frac{\Sigma_{x2}}{\Sigma_{t2}} \right\rangle \left\langle \frac{\Sigma_{t1}}{\Sigma_{t2}^{2}} \right\rangle \tilde{\Sigma}_{t2} \right]$$
(11)

This is the result of the FOP method. The corresponding result of the ITP method can be obtained from (9), if 1 is set equal to zero

$$\Sigma_{\mathbf{x}}^{\mathrm{ITP}} = \tilde{\Sigma}_{t2} \left\{ \left\langle \frac{\Sigma_{\mathbf{x}1}}{\Sigma_{t2}} \right\rangle + \left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{t2}} \right\rangle \frac{S_{1}}{\tilde{\Sigma}_{t2}\Phi} - \left\langle \frac{\Sigma_{t1}\Sigma_{\mathbf{x}2}}{\Sigma_{t2}} \right\rangle + \left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{t2}} \right\rangle \frac{X}{\tilde{\Sigma}_{t2}\Phi} \right\}$$
(12)

A comparison of the expressions $(1 \ 1)$ and (12) shows that they are identical if X is taken to be

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$$X = -S_1 + \left\langle \frac{\Sigma_{t1}}{\Sigma_{t2}^2} \right\rangle \tilde{\Sigma}_{t2} S_2, \qquad (13)$$

for 1 equal to zero. Thus, it has been demonstrated that ITP, in the limit of an infinitely thin sample, is equivalent to FOP within the frame of the general theory discussed so far. However, it will turn out later that this equivalence is no longer valid if the approximation of the self-shielding factors is introduced.

Equation (13) would lead to complicated expressions if the self-shielding factors are to be introduced. Therefore, the following choice for the quantities X_i is preferred for practical calculations

$$x_{j} = -\left\langle 1-P \right\rangle \quad (s_{1j} - \frac{\Sigma_{t1}}{\widetilde{\Sigma}_{t2}} s_{2j}) \quad .$$
(14)

Then the perturbation cross section $\Sigma_{x}^{(2)}$ takes on the form

$$\Sigma_{\mathbf{x}j}^{(2)} = \left\langle \frac{1}{1(\Sigma_{t} \mathbf{1}^{+1/1})} \left(\frac{\Sigma_{\mathbf{x}2}}{\Sigma_{t2}} - \left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{t2}} \right\rangle_{j} \right) \right\rangle_{j} \frac{S_{1j}}{\Phi_{j}}$$
$$- \left\langle \frac{1}{1(\Sigma_{t} \mathbf{1}^{+1/1})} \left(\frac{\Sigma_{t} \mathbf{1}^{\Sigma_{\mathbf{x}2}}}{\Sigma_{t2}^{2}} - \frac{\tilde{\Sigma}_{t1}}{\tilde{\Sigma}_{t2}} \left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{t2}} \right\rangle_{j} \right\rangle_{j} \tilde{\Sigma}_{t2j}$$
(15)

If the cross sections are free of resonances, the expression (15) is zero.

3. Introduction of the Self-Shielding Factors

The above equations are rigorous within the narrow resonance approximation. In order to evaluate them numerically, the concept of the self-shielding factors (or f-factors) will now be introduced, which, of course, implies a certain approximation. According to Abagyan et al. /1/, the f-factor for the isotope n in a given composition, and for the reaction type x, is defined by

$$f_{x}(\sigma_{on}) = \tilde{\sigma}_{xn} / \langle \sigma_{xn} \rangle$$
(15)

where σ_{o} is the cross section of the other isotopes per atom of isotope n. It is given by

$$\sigma_{\rm on} = \frac{1}{Nn} \sum_{n' \neq n} N_{n'} \tilde{\sigma}_{tn'}$$
(17)

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It is important to note that (16) and (17) define the σ_{on} uniquely for a given composition, though they have to be calculated in an iterative way. Only if this iteration has been carried out, the quantity $\langle 1/\Sigma_t \rangle$ is defined uniquely; i.e. the expression

$$\left\langle \frac{1}{\Sigma_{t}} \right\rangle = \frac{1}{N_{n}(\sigma_{on} + \tilde{\sigma}_{tn})}$$

is independent of the isotope index n.

It must be pointed out that, once the self-shielding factors are introduced, (10) is no longer rigorously consistent with (12). The reason is the following:

The two expressions (10) and (11) are equivalent within the general theory. In the FOP method, the self-shielding factors for each isotope are introduced in equation (10). If then the same manipulations which lead to (11) are carried out, one obtains for the last term in (11)

$$\sum_{n} N_{1n} \tilde{\sigma}_{x2n} \left[1 + \frac{N_{2n} (\sigma_{o1n} - \sigma_{o2n})}{\tilde{\Sigma}_{t2}} (1 + \frac{d\tilde{\sigma}_{t2n}}{d\sigma_{o2n}}) \right]$$
(11a)

The notation is explained in the appendix. This expression, however, cannot be written in the form

$$\left\langle \frac{\Sigma_{x2}}{\Sigma_{t2}} \right\rangle \left\langle \frac{\Sigma_{t1}}{\Sigma_{t2}^2} \right\rangle \tilde{\Sigma}_{t2}^2$$

so that (10) is no longer equivalent with (11) once the self-shielding factors are introduced. However, if (11) is no longer a valid expression of the FOP method, it is impossible to demonstrate that ITP for small samples is identical with FOP.

However, in most practical cases, the results obtained by both methods are practically indistinguishable. An exception is U238, where in a typical case (see section 4) FOP is about 8% lower. It can be concluded that FOP is not adequate to calculate the reactivity worth of U238, not even in the limit of a very thin sample. This is due to the approximation which is implicit in the concept of the self-shielding factors. The concept works consistently only for a homogeneous composition, but not for a sample in a homogeneous medium.

The effective perturbation cross sections (9) and (15) can be expressed through the self-shielding factors, as shown in the appendix. A FORTRAN II program was written for the IBM-7074 which calculates the effective perturbation cross sections from these equations. In a second step, the size-dependent reactivity worth is evaluated from these cross sections using unperturbed fluxes and adjoints from a diffusion calculation.

4. Numerical Results and Comparison with Experiments

A series of experiments to study the sample size effect was carried out by W.J. Oosterkamp /6/ on SNEAK-3B-2. In this section, the experimental results will be compared with calculations by the ITP method. The assembly 3B-2 in the fast critical assembly SNEAK was a zoned core, where the composition of the central zone was typical of a PuO_2UO_2 -fueled, steam cooled fast reactor. The assembly is described in detail elsewhere /7/.

The results of the experiments and of calculations by the ITP method are shown in Fig. 1 to 4. The central reactivity worths of different materials are given as a function of the sample thickness. However, for the Pu samples, which are diluted with Al, and for Blo, the surface density (g/cm^2) is a more suitable variable. The calculations were done with the 26-group SNEAK SET, which is described in /8/. The experimental values were converted into absolute reactivity units using $\beta_{eff} = 0.00497$, as given in /7/.

Most calculated reactivity effects are too high. In all cases, the calculation gives the proper sign of the sample size dependence, but the magnitude is usually underpredicted.

The size effect is most pronounced for absorber materials which are not present in the core (Ta, Mo, Nb, Blo). For the resonance absorbers Ta and Mo, the worths, including the self-shielding effect, are predicted fairly well; for Nb, there is probably an error in the cross sections which leads to a large disagreement.

The predicted strong increase in the worths of these materials for thin samples could not be verified because the samples used in the experiment were too thick. The size effect of Blo is predicted well for 3B-2. For comparison, results obtained in assembly 3A-2, which was purely uranium fueled, are also shown. The size effect of Blo is not predicted as well as in 3B-2.

The scattering materials CH_2 , C, and Al show a rather strong size effect, which is fairly well predicted for CH_2 and for Al. For C, the measured values

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are small and show large uncertainties, which explains in part the strong disagreement.

The fuel isotopes U235, U238, and Pu (the samples contained 8 % of Pu240) may be classified as resonance absorbers which are also present in the surrounding core. Therefore, U238 has a much weaker size effect than, for example, Ta; it is underpredicted by the theory. For U235 and Pu, the size dependence is very weak, and agrees with predictions within the error limits.

It should be mentioned that the values calculated by the ITP method agree well in the limit of infinitely thin samples with the values obtained by FOP method, except for U238, where FOP is about 8 % too low.

5. Conclusions

The "Integral Transport Perturbation" method of analysing reactivity worth measurements gives a certain improvement over the standard first order perturbation method, because it allows for self-shielding effects (including resonance effects) in the sample. On the other hand, the heterogeneous structure of the environment is not accounted for. The comparison with experiment shows that the size dependence is generally underpredicted by the ITP. The improvement over FOP is substantial for certain materials (Ta, Blo, CH₂), but rather small for the fissile isotopes.

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<u>APPENDIX</u>: Evaluation of the Perturbation Cross Sections Using Self-Shielding Factors

The expression (9) for $\Sigma_{\mathbf{x}}^{(1)}$ and (15) for $\Sigma_{\mathbf{x}}^{(2)}$ can be written in the form

$$\Sigma_{\mathbf{x}}^{(1)} = \left\langle \frac{\Sigma_{\mathbf{x}1}}{\Sigma_{\mathbf{t}1}^{+1/1}} \right\rangle \frac{S_{1}}{\Phi} + \mathbb{R}2 \cdot \tilde{\Sigma}_{\mathbf{t}2}$$
(A-1)

$$\Sigma_{\mathbf{x}}^{(2)} = \left(\mathbb{R}3 - \frac{1}{1 \ \overline{\Sigma}_{\mathbf{t}1}} \right) \left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{\mathbf{t}2}} \right\rangle \frac{S_{1}}{\Phi}$$
(A-1)

$$- \left(\mathbb{R}4 - \frac{\tilde{\Sigma}_{\mathbf{t}1}}{1 \ \overline{\Sigma}_{\mathbf{t}1}\tilde{\Sigma}_{\mathbf{t}2}} \left\langle \frac{\Sigma_{\mathbf{x}2}}{\Sigma_{\mathbf{t}2}} \right\rangle \right) \tilde{\Sigma}_{\mathbf{t}2}$$
(A-2)

where

$$\overline{\Sigma}_{t1} = \sum_{n} N_{1n} \overline{\sigma}_{t1n} + 1/1$$
$$\widetilde{\Sigma}_{t1} = \overline{\Sigma}_{t1} - 1/1$$
$$\widetilde{\Sigma}_{t2} = \sum_{n} N_{2n} \overline{\sigma}_{t2n}$$

The quantities R2, R3, R4 can be evaluated using the self-shielding factors in the following way

$$\left\langle \frac{\Sigma_{x1}}{\Sigma_{t1}^{+1/1}} \right\rangle = \sum_{n} \frac{\sigma_{x1n}}{\overline{\sigma}_{o1n}^{+}\sigma_{t1n}}$$

$$R2 = \left\langle \frac{\Sigma_{x1}}{1\Sigma_{t2}(\Sigma_{t1}+1/1)} \right\rangle = \sum_{n} \left\langle \frac{\sigma_{xn}}{1N_{2n}(\overline{\sigma}_{01n}+\sigma_{tn})(\sigma_{02n}+\sigma_{tn})} \right\rangle$$
$$= \frac{1}{1\widetilde{\Sigma}_{t2}} \sum_{n} \frac{\widetilde{\sigma}_{x2n}}{\overline{\sigma}_{01n}-\sigma_{02n}} - \frac{1}{1\overline{\Sigma}_{t1}} \sum_{n} \frac{N_{1n}\widetilde{\sigma}_{x1n}}{N_{2n}(\overline{\sigma}_{01n}-\sigma_{02n})}$$

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$$R3 = \left\langle \frac{\overline{\Sigma}_{x2}}{1\overline{\Sigma}_{t2}(\overline{\Sigma}_{t1}+1/1)} \right\rangle = \sum_{n} \left\langle \frac{\sigma_{xn}}{1\overline{N}_{1n}(\overline{\sigma}_{01n}+\sigma_{tn})(\sigma_{02n}+\sigma_{tn})} \right\rangle$$

$$= \frac{1}{1 \cdot \overline{\Sigma}_{t2}} \sum_{n} \frac{N_{2n} \sigma_{x2n}}{N_{1n}(\overline{\sigma}_{01n}-\sigma_{02n})} - \frac{1}{1\overline{\Sigma}_{t1}} \sum_{n} \frac{\sigma_{x1n}}{\overline{\sigma}_{01n}-\sigma_{02n}}$$

$$R4 = \left\langle \frac{\overline{\Sigma}_{t1}\overline{\Sigma}_{x2}}{1\overline{\Sigma}_{t2}^{2}(\overline{\Sigma}_{t1}+1/1)} \right\rangle = \sum_{n} \left\langle \frac{(\sigma_{tn}+\overline{\sigma}_{0n}-\frac{1}{1\overline{N}_{1n}})\sigma_{xn}}{(n_{2n}(\sigma_{02n}+\sigma_{tn})^{2}(\overline{\sigma}_{01n}+\sigma_{tn})} \right\rangle$$

$$= \frac{1}{N_{1n}N_{2n}(\overline{\sigma}_{01n}-\sigma_{02n})^{2}} \left\langle \frac{\sigma_{xn}}{\sigma_{02n}+\sigma_{tn}} - \frac{\sigma_{xn}}{\overline{\sigma}_{01n}+\sigma_{tn}} \right\rangle$$

$$+ \frac{\sigma_{02n}-\overline{\sigma}_{01n}}{1\overline{N}_{2n}(\overline{\sigma}_{01n}-\sigma_{02n})^{2}} \left\langle \frac{\sigma_{xn}}{\sigma_{02n}+\sigma_{tn}} - \frac{\overline{\sigma}_{xn}}{\overline{\sigma}_{02n}+\sigma_{tn}} \right\rangle$$

$$= \frac{1}{1^{2}\overline{\Sigma}_{t2}} \sum_{n} \frac{\sigma_{n}}{N_{1n}(\overline{\sigma}_{01n}-\sigma_{02n})^{2}} - \frac{1}{1^{2}\overline{\Sigma}_{t1}} \sum_{n} \frac{\sigma_{n}}{N_{2n}(\overline{\sigma}_{01n}-\sigma_{02n})^{2}}$$

$$+ \frac{1}{1\overline{\Sigma}_{t2}} \sum_{n} \left(\frac{1}{1N_{1n}(\overline{\sigma}_{01n}-\sigma_{02n})} - 1 \right) \left[\frac{d\sigma_{x2n}}{d\sigma_{02n}} - \frac{N_{2n}\sigma_{x2n}}{\overline{\Sigma}_{t2}} \left(1 + \frac{d\sigma_{t2n}}{d\sigma_{02n}} \right) \right]$$

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