CONF-

790125--57

MASTER

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

RANGE CALCULATIONS USING MULTIGROUP TRANSPORT METHODS*

T. J. Hoffman

Computer Sciences Division

as

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
Union Carbide Corporation, Nuclear Division

M. T. Robinson

Solid State Division

H. L. Dodds, Jr.

The University of Tennessee, Knoxville, Tennessee 37916

By acceptance of this article, the publisher or recipient acknowledges the U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering the article.

26
REPRODUCED FROM THE U.S. GOVERNMENT PRINTING OFFICE

*Research sponsored by the Magnetic Fusion Energy Division, U.S. Department of Energy under Contract W-7405-eng-26 with the Union Carbide Corporation.

RANGE CALCULATIONS USING MULTIGROUP TRANSPORT METHODS*

T. J. HOFFMAN and M. T. ROBINSON

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, USA

H. L. DODDS, JR.

The University of Tennessee, Knoxville, Tennessee 37916, USA

Several aspects of radiation damage effects in fusion reactor neutron and ion irradiation environments are amenable to treatment by transport theory methods. In this paper, multigroup transport techniques are developed for the calculation of particle range distributions. These techniques are illustrated by analysis of Au-196 atoms recoiling from (n,2n) reactions with gold. The results of these calculations agree very well with range calculations performed with the atomistic code MARLOWE. Although some detail of the atomistic model is lost in the multigroup transport calculations, the improved computational speed should prove useful in the solution of fusion material design problems.

1. INTRODUCTION

The purpose of this work is to investigate the applicability of multigroup transport methods to the calculation of radiation damage by energetic ions in amorphous materials. In particular, we wish to evaluate the use of discrete ordinates and Monte Carlo multigroup computer codes on ion slowing down problems.

The interaction of charged particles with amorphous media was analyzed by Sigmund¹ with a Boltzmann transport equation model. He used a semi-analytical approach known as the moments method to solve the Boltzmann equation. The moments method is restricted to infinite medium problems in which the differential cross sections can be represented with simple analytical expressions. For many slowing down problems, these assumptions are not valid.

To treat finite medium problems with arbitrarily complex cross-section representations, numerical procedures are needed. The most widely used and highly developed method for numerical solution to the Boltzmann equation is the discrete ordinates method.²

The discrete ordinates method has been applied to electron scattering problems by Bartine, et al.³ However, several aspects of these calculations are not desirable for analysis of ion slowing down problems:

- a. The discrete ordinates computer code was modified to account for continuous slowing down of the electrons due to small-angle Coulomb scattering;

- b. Photon transport, although important for electron slowing down problems, has a negligible effect on ion slowing down calculations;
- c. Target atom transport, although important for some slowing down problems such as sputtering, was not treated in the electron transport work.

In a previous paper,⁴ we successfully used the discrete ordinates computer code, ANISN,⁵ to calculate sputtering yields by light ions incident on the inner wall of a controlled thermonuclear reactor (CTR). Although the usual application of this code has been to neutron and gamma-ray problems, many of the computational algorithms are applicable to ion transport problems. Our development has involved the appropriate manipulation of ANISN input data, i.e., the multigroup cross sections, rather than code modification. Therefore, any multigroup transport computer code can be used. The success of this multigroup transport method on sputtering problems has encouraged us to apply related methods to other atomic particle transport problems of interest in fusion energy applications. The calculation of the penetration distance distribution, the topic of this paper, is an example of such an application.

2. PROBLEM FORMULATION

2.1 The multigroup equation

Consider ions with energy E_0 normally incident on a plane ($x=0$) of a thick slab ($x=-a, a$). By considering a particle balance in differential phase space $dx dE d\mu$ about the distance into the slab, x , the particle energy, E , and the particle direction cosine relative to the x -axis, μ , a Boltzmann equation of the following form can be written for the ion flux, $\phi(x, E, \mu)$, in the slab^{1,3,4}:

*Research sponsored by the Magnetic Fusion Energy Division, U.S. Department of Energy under Contract W-7405-eng-26 with the Union Carbide Corporation.

$$\begin{aligned} \mu \frac{\partial \phi(x, E, \mu)}{\partial x} + \Sigma(E) \phi(x, E, \mu) &= \delta(x) \delta(E - E_0) \delta(\mu - 1) \\ &+ \frac{\partial [\Sigma(E) \phi(x, E, \mu)]}{\partial E} \\ &+ \int_0^\infty dE' \int_{-1}^1 d\mu' \Sigma(E' \rightarrow E, \mu' \rightarrow \mu) \phi(x, E', \mu'). \end{aligned} \quad (1)$$

The terms on the LHS of Eq. (1) represent losses due to leakage and elastic collisions; the terms on the RHS are gains due to ion production, slowing down by electronic interactions, and in-scatter by elastic collisions, respectively.

To obtain the multigroup form of Eq. (1), the energy range $(0, E_0)$ is divided into NG energy segments or "groups." The Boltzmann equation is then integrated over energy group G, i.e., from E_G to E_{G-1} . Within each group, the ion flux is assumed separable:

$$\phi(x, E, \mu) = \phi_G(x, \mu) W_G(E) \text{ for } E \in (E_G, E_{G-1}). \quad (2)$$

With this assumed separability, the multigroup form of the ion slowing down problem is

$$\begin{aligned} \mu \frac{\partial \phi_G(x, \mu)}{\partial x} + \Sigma_G^{(MOD)} \phi_G(x, \mu) &= Q_G(x, \mu) \\ &+ \sum_{G'=1}^{NG} \int_{-1}^1 d\mu' \Sigma_{G' \rightarrow G}(\mu' \rightarrow \mu) \phi_{G'}(x, \mu'), \text{ for } G=1, NG. \end{aligned} \quad (3)$$

All terms in Eq. (3) have their standard multigroup definitions as given in Refs. 3 and 4, except for the "modified total cross section," $\Sigma_G^{(MOD)}$, which is defined as

$$\begin{aligned} \Sigma_G^{(MOD)} &\equiv \Sigma_{tG} - S(E_{G-1}) W_G(E_{G-1}) \\ &+ S(E_G) W(E_G). \end{aligned} \quad (4)$$

In this equation, Σ_{tG} is the total multigroup cross section, S is the electronic stopping cross section, and W is the weight function of Eq. (2).

2.2 Normalization

The product, $S(E)W(E)$, can be viewed as a cross section for ions slowing down below energy E due to electronic interactions, e.g., the reaction rate

$$R_G^U(x, \mu) = \phi_G(x, \mu) S(E_{G-1}) W_G(E_{G-1}), \quad (5)$$

is the rate at which ions enter group G by slowing down below E_{G-1} and

$$R_G^L(x, \mu) = \phi_G(x, \mu) S(E_G) W_G(E_G), \quad (6)$$

is the rate at which ions leave group G by slowing down below E_G . Clearly, to conserve ions

$$R_{G-1}^L(x, \mu) = R_G^U(x, \mu), \quad (7)$$

i.e., the number of ions leaving group G-1 must equal the number entering the neighboring group, G.

Equation (7) places an additional constraint on the weight function. If this equation is not satisfied, particle sources or sinks will occur at the group boundaries and the problem will not be properly normalized. To satisfy this equation, an iterative procedure was adopted: fluxes were used as weight functions to generate the multigroup cross sections which, in turn, were used to calculate updated fluxes. Several iterations were required to achieve convergence of the fluxes and proper normalization of the calculation. The fluxes used in this iterative procedure were calculated with AMIS. An example of the convergence is given in Section 4.

2.3 The slowing down density

Of interest in the slowing down problem is the mean penetration distance of the ions, i.e., the "range," $\langle x \rangle$. To obtain a workable definition of $\langle x \rangle$, the following definition for the slowing down density, $p(x)$, was used:

$$p(x) dx \equiv \text{the number of ions that emerge from interactions in } dx \text{ with energy less than } 5 \text{ eV}. \quad (8)$$

Then,

$$\langle x \rangle \equiv \int_{-a}^a x p(x) dx. \quad (9)$$

Higher moments of $p(x)$ were also calculated.

The slowing down density is related to the ion flux as follows:

$$p(x) = \sum_{G=1}^{NG} \Sigma_{tG} f_{G \rightarrow 5 \text{ eV}} \int_{-1}^1 \phi_G(x, \mu) d\mu, \quad (10)$$

where $f_{G \rightarrow 5 \text{ eV}}$ is the probability that an ion experiencing a collision in group G will emerge from that collision with an energy less than 5 eV.

3. CALCULATIONAL PROCEDURES

3.1 Discrete and continuous interactions

In the development of Eq. (3), elastic interactions with the nuclei of the slab material were treated in the total interaction and in-scatter terms of the Boltzmann equation ("discrete interactions"). Inelastic interactions with the electrons were treated in the continuous slowing down term of the Boltzmann equation ("continuous interactions"). However, any interaction in which small directional changes of the incident particle occur can be treated as a continuous interaction. If all nuclear interactions are treated as discrete interactions, many terms are needed in the Legendre expansion of the differential cross sections. To avoid these costly calculations, all interactions in which the incident particle scatters through an angle less than a prescribed angle, θ_m , are treated as continuous interactions. For the calculations reported in this paper, θ_m was set to five degrees, and a P_9 Legendre

expansion was used. These values are based on our experience with the sputtering calculations.⁴ For light ion slowing down calculations, lower values of θ_m and higher P_2 expansions are necessary.

3.2 Data representation

In Sigmund's work,¹ the differential cross section for Thomas-Fermi interactions was represented by a power approximation,

$$d\sigma(E, T) = CE^{-1-m} dT, \quad (11)$$

where

$$C = \frac{1}{2} \pi \lambda_m a_{12}^2 \left(\frac{M_1}{M_2} \right)^m \left(\frac{2Z_1 Z_2 e^2}{a_{12}} \right)^{2m}$$

M_1, Z_1 = mass, charge of the incident particle

M_2, Z_2 = mass, charge of the recoil particle

$$a_{12} = 0.4683(Z_1^{2/3} + Z_2^{2/3})^{-1/2} \text{ \AA}$$

$$(m, \lambda_m) = \left(\frac{1}{3}, 1.309 \right) \text{ for } E_A < E < E_B$$

$$= \left(\frac{1}{2}, 0.327 \right) \text{ for } E_B < E < E_C$$

$$= (1, 0.5) \text{ for } E_C < E.$$

For lower energies ($E < E_A$), the differential cross section for Born-Mayer interactions was also represented with a power approximation of the form of Eq. (11) with $(m, \lambda_m) = (0, 24)$. The transition energies E_A, E_B, E_C were determined by continuity of the nuclear stopping power, i.e.,

$$S_n(E) = \int_0^{Y_1 E} T d\sigma(E, T) = \frac{1}{1-m} C Y_1^{1-m} E^{1-2m}. \quad (12)$$

In general, Eq. (11) describes approximately the scattering from a potential of the form $V(r) \propto r^{-1/m}$.

For screened Coulomb interaction between an ion and an atom or between two atoms, Lindhard et al.⁶ derived the following approximate form of the differential cross section:

$$d\sigma(E, T) = \frac{\pi a_{12}^2 f(t^{1/2})}{2t^{3/2}} dt, \quad (13)$$

where

$$t = \epsilon^2 T / Y_1 E,$$

$$\epsilon = \frac{M_2 E}{M_1 + M_2} \frac{a_{12}}{Z_1 Z_2 e^2},$$

and $f(t^{1/2})$ is a function that depends on the assumed form of the screening function. This function has been calculated by Lindhard et al.⁶ for the collision of neutral Thomas-Fermi atoms and is shown in Fig. 1 together with Winterbon's analytical approximation⁷ to $f(t^{1/2})$, i.e.,

$$f_A(t^{1/2}) = \lambda t^{1/6} [1 + (2\lambda t^{2/3})^{2/3}]^{-3/2}, \quad (14)$$

where $\lambda = 1.309$. Also plotted in Fig. 1 is the power approximation for the Thomas-Fermi cross section ($m > 0$) given by Eq. (11). As shown in this figure, Eq. (14) is a better representation of $f(t^{1/2})$ than Sigmund's power approximation.

Fig. 1. Reduced differential cross section for Thomas-Fermi potential.

For the moments method used by Sigmund,¹ simple expressions such as the power approximation of Eq. (11) are needed. However, for the numerical multigroup approach, more complex expressions for $d\sigma(E, T)$ can be used. In our work, we used Winterbon's analytic approximation to the Thomas-Fermi potential, i.e., Eqs. (13) and (14) at energies above E_A . For lower energies, we used Sigmund's approximation to the Born-Mayer potential, i.e., Eq. (11) with $m = 0$.

The stopping cross section, $S(E)$, is the energy loss between discrete collisions per unit track length of the particle. All energy losses not included in the discrete loss terms (total interaction and inscatter) of the Boltzmann equation must be included in the calculation of $S(E)$, i.e., the energy loss due to electronic interactions and the energy loss due to nuclear interactions in which the incident particle scatters through an angle less than θ_m .

Electronic losses are calculated with the following expression:⁸

$$S_e(E) = kN \left(\frac{Z_1 Z_2 e^2}{a_{12}} \right)^{1/2} \left(\pi a_{12}^2 \right) \left(\frac{4M_1}{M_1 + M_2} \right) \times \left(\frac{M_2}{M_1 + M_2} \right)^{1/2} E^{1/2}, \quad (15)$$

where

$$k = \frac{32}{3\pi} \left(\frac{m_e}{M_2} \right)^{1/2} \left(1 + \frac{M_2}{M_1} \right)^{3/2} \frac{Z_1^{2/3} Z_2^{1/2}}{(Z_1^{2/3} + Z_2^{2/3})^{3/4}}$$

N = atom density

m_e = mass of an electron.

The energy loss due to nuclear interactions in which the incident particle scatters through an angle less than θ_m was calculated with the following expression:

$$S_n(E) = N \int_0^{Y_1 E} T d\sigma(E, T), \quad (16)$$

where Y_1 is the fractional energy transfer associated with θ_m . For the power approximation to the differential cross section, this expression was evaluated analytically; for the other representations of $d\sigma(E, T)$, a numerical integration was used.

The integrations necessary to obtain the multigroup parameters were performed numerically. To illustrate the procedure, consider evaluation of the following quantity:

$$\bar{\sigma} = \frac{\int_{E_a}^{E_b} W(E) \sigma(E) dE}{\int_{E_a}^{E_b} W(E) dE},$$

where $W(E)$ is the weighting function of Eq. (2), and $\sigma(E)$ is an energy-dependent cross section. Let $W(E) = E^{Y_i}$ and $\sigma(E)$ go roughly as E^{-m} , where m is determined by Eq. (11). Then,

$$\bar{\sigma} = \frac{\int_{E_a}^{E_b} E^{Y_i} [E^m \sigma(E)] \frac{dE}{E^m}}{\left(\frac{1}{Y_i+1} \right) (E_b^{Y_i+1} - E_a^{Y_i+1})},$$

$$= \frac{(Y_i + 1)}{(Y_i + 1 - m)} \frac{\int_{E_a}^{E_b} [E^m \sigma(E)] d(E^{Y_i+1-m})}{(E_b^{Y_i+1} - E_a^{Y_i+1})}.$$

Since $E^m \sigma(E)$ is approximately constant, the integration in $d(E^{Y_i+1-m})$ phase space can be performed with fewer intervals, N , than the integration in dE phase space. Simpson's rule of integration was performed in $d(E^{Y_i+1-m})$ phase space with $N = 10$ for all numerical integrations in the cross-section processing routines. The Y_i 's were determined from the ANISN-calculated fluxes.

3.3 Multigroup computer codes

Two multigroup computer codes were used to analyze the ion slowing down problem: ANISN, a one-dimensional (spatially) discrete ordinates code, and MORSE,⁹ a three-dimensional (spatially) Monte Carlo code. ANISN was used to calculate one-dimensional effects of interest such as $p(x)$ of Eq. (10) and its moments. Also, as indicated earlier in this paper, ANISN was used in the iterative procedure for the multigroup cross-section generation. MORSE was used to calculate multidimensional parameters such as the vector range and the radial spread of the ion slowing down distribution. The product, $\Sigma_t g f \approx 5$ ev of Eq. (10), is treated as an "activity cross section" by ANISN and a "response function" by MORSE.

4. RESULTS

Table 1 shows the convergence of the ANISN fluxes and the normalization constant for a 100 keV Au-196 atom slowing down in gold. Also shown in the table is the mean range as a function of iteration number. These tabulated values indicate that meaningful results can be obtained for this problem after about three iterations. The total cpu time required for all data in this table is about 17.4 minutes on the IBM 360/91. This cross-section generation time

must be added to that of any multigroup slowing down calculation to evaluate the merits of the multigroup approach.

Table 1. Convergence of ANISN Fluxes ($\bar{\sigma}$) and the Normalization Constant During Multigroup Processing (100 keV, Au-196 in gold)

Iteration number Energy Group	1	2	3	4
1	8.35	8.33	8.34	8.34
2	61.7	56.5	57.3	57.3
3	27.7	25.8	26.0	26.0
4	17.8	16.6	16.8	16.7
5	12.3	11.4	11.6	11.6
6	8.66	8.07	8.16	8.16
7	6.20	5.79	5.86	5.86
8	4.28	3.99	4.03	4.02
9	3.70	3.45	3.49	3.49
10	2.99	2.79	2.82	2.82
11	2.77	2.59	2.61	2.61
12	2.67	2.49	2.52	2.52
13	2.59	2.41	2.44	2.44
14	2.50	2.34	2.36	2.36
15	2.41	2.25	2.28	2.27
16	2.31	2.15	2.18	2.17
17	2.18	2.04	2.06	2.06
18	2.04	1.92	1.94	1.94
19	1.88	1.76	1.80	1.80
20	1.71	1.63	1.66	1.66
21	1.52	1.47	1.50	1.49
Normalization Constant	1.038	0.986	1.002	1.000
Calculated Range, Å	135.7	126.1	128.7	128.4

Figure 2 illustrates some of the detail for the slowing down problem that can be obtained with the multigroup discrete ordinates code, ANISN. Shown in the figure is the slowing down density, $p(x)$, for Au-196 in gold. Also shown is the slowing down distribution as calculated by MARLOWE,¹⁰ an atomistic simulation code. Although MARLOWE uses scattering data based on the Molière approximation to the Thomas-Fermi potential (as opposed to the LSS theory⁶ of ANISN), reasonably good agreement is obtained.

Fig. 2. The slowing down density for 100 keV Au-196 atom in gold.

The multigroup Monte Carlo code, MORSE, was also used to analyze this problem (Au-196 incident on gold). The MORSE results for several integral parameters is shown in Table 2 along with the comparable results from ANISN and MARLOWE. With MORSE and MARLOWE (and unlike ANISN), multidimensional parameters can be calculated. Since MORSE is a multigroup code, unlike MARLOWE, much of the cross-section manipulation is performed by the multigroup cross-section generation code. As a result, Monte Carlo histories can be processed faster by MORSE than by MARLOWE -- roughly ten times faster for this problem.

Parameter	ANISN	MORSE ^a	MARLOWE ^b
$\langle x^0 \rangle$	1.000	0.991 $\pm$ 0.008	1.0
$\langle x^1 \rangle$, Å	128.4	125.5 $\pm$ 2.0	121.3 $\pm$ 0.7
$\langle x^2 \rangle$, Å ²	23407.	22676. $\pm$ 573.	19350.
Radial Spread, Å	b	86.08 $\pm$ 1.84	67.53 $\pm$ 0.42
Vector Range, Å	b	161.0 $\pm$ 2.7	144.1 $\pm$ 0.7
cpu time (IBM 360/91) ^c , min.	4.8	9.8 ^d	108.0 ^d

^aBased on 10,000 histories.
^bNot amenable to one-dimensional analysis.
^cThe ANISN and MORSE computation times do not include the 17.4 min. required on the IBM 360/91 to generate the multigroup cross sections.
^dRun on the IBM 360/195; a conversion factor of 2.0 was used to obtain equivalent IBM 360/91 cpu times.

5. CONCLUSION

Multigroup transport methods have been applied to the problem of Au-196 slowing down in gold. Although Sigmund's transport model is used in this work, the approximations in Sigmund's work that make it inapplicable to many slowing down problems are avoided by the multigroup solution method. Through modification of the multigroup cross-section definitions, standard computer codes, such as ANISN and MORSE, were used to solve charged particle interaction problems. The multigroup approach enables calculation of the ion range distribution and related quantities such as ion reflection coefficients, ion sputtering, and the behavior of ion beams penetrating thin foils. In fact, any problem concerning the interaction of energetic particles (ions and atoms) with structureless (amorphous and polycrystalline) media can probably be treated with these methods.

Thus far, work has been limited to one-component targets and to cross sections based on Lindhard-Scharff-Schiott theory. Efforts are currently being pursued to extend this work to multicomponent targets (such as alloys and compounds) and to the construction of cross-sections based on classical potential scattering.

REFERENCES

1. P. Sigmund, *Phys. Rev.*, **184**, 383 (1969).
2. B. G. Carlson, *Solution of the Transport Equation by the S_N Method*, LA-1891, Los Alamos Scientific Laboratory (1955); also, C. E. Lee, *The Discrete S_N Approximation to Transport Theory*, LA-2595, Los Alamos Scientific Laboratory (1962).
3. D. E. Bartine, R. G. Alsmiller, Jr., F. R. Mynatt, W. W. Engle, Jr., and J. Barish, *Nucl. Sci. Eng.*, **48**, 159 (1972).
4. T. J. Hoffman, H. L. Dodds, Jr., M. T. Robinson, and D. K. Holmes, *Nucl. Sci. Eng.*, **68**, 204 (1978).
5. W. W. Engle, Jr., *A User's Manual for ANISN, A One-Dimensional Discrete Ordinates Code with Anisotropic Scattering*, K-1693, Oak Ridge Gaseous Diffusion Plant (1967).
6. J. Lindhard, M. Scharff, and H. E. Schiott, *Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd.*, **33**, No. 14 (1963).
7. K. B. Winterbon, P. Sigmund, and J. B. Sanders, *Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd.*, **37**, No. 14 (1970).
8. J. Lindhard and M. Scharff, *Phys. Rev.*, **124**, 128 (1961).
9. M. B. Emmett, *The MORSE Monte Carlo Radiation Transport Code System*, ORNL-4972, Oak Ridge National Laboratory (1975).
10. M. T. Robinson and I. M. Torrens, *Phys. Rev. B*, **9**, 5008 (1974).