NBSIR 77-1283

Spectra From Energy Degradation of Fast Electrons in Water

Martin J. Berger

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SPECTRA FROM ENERGY DEGRADATION OF FAST ELECTRONS IN WATER*†

Martin J. Berger

ABSTRACT

This report gives data for energy degradation spectra in water vapor. Calculations utilized the Monte Carlo method and included Auger electrons. Results include 1) a new energy scaling principle at low energies, 2) values at different source energies for W, the mean energy loss per ion pair formed, 3) data on the electron range calculated by continuous-slowing-down approximation, and 4) data on stopping powers and restricted stopping powers as compared with results from the Bethe theory.

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*Work supported by Office of Naval Research, Arlington, Va. and U.S. Energy Research and Development Admin., Washington, D.C.

†This is an interim report and the work is continuing. The report is to be superseded by a future publication which will receive general distribution and should be cited as a reference. Please consult the NBS Office of Technical Publications to obtain the proper citation.

INTRODUCTION

Fast electrons lose energy to the surrounding medium by many kinds of interactions, including a) knock-on processes with individual electrons which follow approximately the Møller cross section, and b) glancing processes for which the electric pulse generated when a fast electron passes an atom can stimulate both excitation and ionization.

The cumulative result of these interactions is the progressive loss of energy, often called energy "degradation," by the fast electrons. A source of electrons can thus give rise to electrons of all lower energies; and these electrons include not only the "primaries" whose energies have been degraded, but also "secondaries" produced in ionizing interactions and defined to be the free electron of lower energy of the two which emerge from such interactions. The resulting spectrum is commonly called the "degradation" spectrum; and it has importance in many fields dealing with radiation effects on matter.

Since the early exploratory calculations of Spencer and Fano (1954), calculations of these spectra for a variety of materials have been made; although the requirement for a complete set of inelastic cross sections at all energies below the source energy T_o makes these calculations difficult. In this brief interim report we give many of our results on degradation spectra for water vapor. We have not yet completed comparisons with other calculations, including those for comparable materials; these will be included in the final publication on this subject.

SUMMARY DESCRIPTION OF THE CALCULATIONS

The Monte Carlo method has been applied to the calculation of energy degradation spectra in water vapor. Calculations have been made at a large number of energies between 100 keV and a few eV. The successive inelastic events which an electron makes in the course of slowing down in an extended medium have been simulated in direct analogy to the physical processes, and all generations of secondary electrons were followed. The number of electrons whose energy histories were simulated varied depending on the initial energy T_o of the primary electron; for example:

T_o, eV	No. of primaries followed	No. of secondaries followed per primary, on the average
100,000	500	3270
10,000	2,000	331
1,000	10,000	33
100		2.8

As far as possible, the various inelastic events were sampled according to cross sections derived from experiments. The sources of cross section data were as follows:

Excitation	Olivero, Stagat and Green (1972); Kutcher and Green (1976)
Dissociative excitation	Beenakker, de Heer, Krop and Möhlmann (1974)
Dissociative attachment	Compton and Christophorou (1967)
Ionization (total)	Schutten, de Heer, Moustafa, Boerboom and Kistemaker (1966); Maerk and Egger (1975)
Ionization (K-shell)	Glupe and Mehlhorn (1971)
Spectrum of Auger electrons	Moddeman, Carlson, Krause and Pullen (1971)
Ionization (differential)	Opal and Beaty (1972)

Altogether 27 possible modes of excitation were taken into consideration, and use was made of the very convenient parameterization of the excitation cross sections by Green and collaborators. The Opal-Beaty ionization cross section, differential in the secondary electron energy, was available only at 500 eV. It was fitted at that energy by a simple analytical formula which is a modification of the Møller cross section (for scattering by free electrons) and which takes into account exchange effects). Following Kim (1975) it was assumed that the spectrum of energy transfers in glancing collisions is determined predominantly by the oscillator-strength distribution, and is nearly the same at all energies. The simple analytical formula was therefore retained, but was renormalized at each energy to obtain agreement with the experimental total ionization cross section. At spectral energies from 200 to 600 eV the ionization cross section was made to go over into the Møller cross section. Ionization was assumed to be possible from five orbitals, with binding energies of 12.6, 14.7, 18.4, 32.2 and 540 eV. The relative probability of electron ejection from the K-shell (followed by emission of an Auger electron) was estimated from experimental data; the relative probabilities of ejection from the other orbitals was estimated from a Weizsacker-Williams calculation by Seltzer (private communication).

RESULTS

We summarize here certain significant conclusions that can be drawn from the analysis of results performed so far, and back them up with information in tabular form.

1. As shown in Table 1, the energy degradation spectra calculated with the use of detailed inelastic collision cross sections are in good agreement at high and intermediate energies with the results of the Spencer-Fano (1954) theory. The latter assumes energy transfers to be governed by the Møller cross section, adjusted with a cut-off on low-energy transfers to give the correct stopping power. It turns out that this approximation gives good results for spectral energies down to 200 eV; below this energy the Spencer-Fano theory is no longer applicable. This presumably holds not only for water vapor but also for other gases of similar atomic number. Below 200 eV, the new Monte Carlo results are the appropriate extension of the Spencer-Fano theory into the low-energy region of interest for radiation chemistry yield studies.

2. A new and useful scaling law has been discovered, which applies at low spectral energies, say, below a few hundred eV, and which greatly reduces, and at low energies practically suppresses the dependence of the energy degradation spectra on the initial electron energy. It has been customary to represent the energy degradation spectrum in terms of the differential tracklength distribution $y(T,T_o)$, defined such that $y(T,T_o)dT$ is the average tracklength of electrons with energies between T and $T+dT$. The new scaled quantity is

$$T\,\frac{dG}{dT} = T(100/T_o)(N\sigma/\rho)y(T,T_o) \quad .$$

Here T is the spectral energy and T_o the source energy, both in eV; N is the number of molecules per cm^3, σ is the total cross section for inelastic collisions, in cm^2, and ρ is the density of the medium, in g/cm^3; $y(T,T_o)$ is the differential tracklength distribution in g/cm^2 per eV. The quantity dG/dT can be interpreted as the differential yield of inelastic collisions per unit spectral energy, and is normalized to an energy input of 100 eV. The dimensionless quantity T dG/dT is given in Table 2 for initial electron energies T_o between 100,000 and 1000 eV. It can be seen that, as claimed, the dependence of T dG/dT on T_o becomes progressively weaker as the spectral energy T decreases. This suggests that energy degradation spectra for values of T_o much higher than 100 keV could be obtained without much trouble by using the Spencer-Fano theory down to 200 eV, and adding on the low-energy part of the spectrum assuming a universal spectral shape. Finally, we note that the yield $G_i(T_o)$ of events of type i can be calculated from the expression

$$G_i(T_o) = \int_o^{T_o} (dG/dT)(\sigma_i/\sigma)dT \quad ,$$

where σ_i is the cross section for the event of type i. Again, the yield is normalized to an energy input of 100 eV.

3. Auger electrons emitted after ionization from the K-shell make a significant contribution to the energy degradation spectrum at spectral energies below 500 eV. The magnitude of this contribution is given in Table 3. Many previous treatments of the energy degradation spectrum, including that of Spencer and Fano, have omitted the Auger electron contribution. Therefore the comparison given in Table 2 does not include Auger electrons.

4. In the course of the Monte Carlo calculation of the energy degradation spectra, the yields of various events have also been obtained, for example, the yield of ionizations. Table 4 gives the mean number of ionizations as a function of the initial electron energy, as well as the ionization fluctuations as summarized in terms of the Fano factor, which is the ratio of the mean square to the mean number of ionizations. If the fluctuations were governed by a Poisson distribution, the Fano factor would be unity. As can be seen from Table 4, this is the case only quite close to the ionization threshold (12.6 eV). At higher energies the Fano factor assumed a limiting value of 0.25.

5. The value of W (mean energy needed to produce an ion pair in water vapor) is shown in Table 5. It can be seen that W is almost constant down to energies of a few hundred eV, whereupon it rises at first slowly and then quite rapidly at lower energies. Between 10 keV and 100 keV there is a very slow rise of 1.1%. It would not be altogether surprising if this rise continued as the electron

energy is further increased. Table 6 compares our value of W with that obtained in other calculations, with differences presumably arising mainly from the different assumed cross sections, but to some extent also from the calculational techniques. The experimental W-value for water vapor is given as 29.9 eV by Adler and Bother (1965) and as 30.1 eV by Booz and Ebert (1963). These values differ by 1.3% and 0.7%, respectively, from the result at T_o = 1 keV of the present calculation, differences which are well within the combined limits of experimental and theoretical uncertainties.

6. The contribution of ionization events from different spectral regions of the energy degradation spectrum is indicated in Table 7. It can be seen that a significant contribution to the ionization is made by high-energy as well as by low-energy electrons. Moreover, the relative contributions from different spectral regions are seen to be rather insensitive to the initial energy of the primary electron, if scaling by T_o is used for the top half of the table.

7. From the inelastic scattering cross sections compiled for the Monte Carlo calculation, one can also derive the electron stopping power. Table 8 compares the results thus obtained with the values given by the Bethe stopping-power formula (assuming a mean excitation energy of 71.7 eV for water vapor). Between 100 keV and 20 keV, there is no difference. At lower energies, the present results fall below the results from the Bethe theory, by 1.3% at 10 keV, 6.8% at 5 keV, 8.3% at 2 keV and 11% at 1 keV.

8. The CSDA (continuous-slowing-down-approximation) range has been computed from the new stopping-power values in Table 7, and is given in Table 9. The range is computed as the pathlength which the electron would travel until it reaches a cut-off energy T_c, assuming that the energy loss along the entire path is always equal to the mean loss, i.e., the stopping-power. The results in Table 9 are for a cut-off T_c = 12.6 eV, equal to the lowest ionization potential in water vapor. Depending on the chosen value of T_c, different range values are obtained; for example:

	CSDA range, g cm^{-2}		
T_o, keV	T_c = 4.5 eV	T_c = 12.6 eV	T_c = 20.0 eV
100	1.522×10^{-2}	1.432×10^{-2}	1.423×10^{-2}
10	1.246×10^{-2}	2.828×10^{-4}	2.599×10^{-4}
1	0.926×10^{-4}	7.156×10^{-6}	6.500×10^{-6}

9. In radiological physics, it is often of interest ot know the "local" energy deposition in terms of restricted stopping power, i.e., a mean energy loss per unit pathlength from events in which the energy transfer from the electron to the molecule is smaller than some chosen cut-off energy Δ. In cavity ionization theory, the Δ is typically

10 to 15 keV, and the Bethe theory for the restricted stopping power can be applied above ~ 10 keV in water. In track structure theory in radiobiology, cut-off energies Δ as low as 100 eV are considered. As is shown in Table 10, the cross sections assumed in the present work then lead to significant departures from the predictions of the Bethe theory, even for electron energies as high as 100 keV.

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TABLE 1

$$\left[y(T,T_o)\ S(T)\right] \Big/ \left[y(T,T_o)\ F(T,T_o)\right]_{\text{Spencer-Fano}}$$

T/T_o	T_o = 100 keV	50 keV	20 keV	10 keV	5 keV	2 keV
$1/2^{-1/6}$	1.048	1.059	1.088	1.098	1.075	1.117
$1/2^{-2/6}$	0.988	0.996	1.012	1.003	0.983	1.012
$1/2^{-3/6}$	0.966	0.992	0.989	0.983	0.979	0.986
$1/2^{-4/6}$	0.963	1.000	0.988	0.989	0.982	0.968
$1/2^{-5/6}$	0.972	1.005	0.994	0.997	0.971	0.966
1/2	0.983	0.993	0.996	0.997	0.962	0.970
1/4	1.013	1.013	0.985	0.986	0.997	0.981
1/8	0.991	0.998	0.999	0.991	0.995	0.953
1/16	1.001	0.970	0.983	0.997	1.008	
1/32	0.964	0.966	0.997	1.010	0.890	
1/64	0.970	0.972	1.031	0.921		
1/128	0.964	1.001				
1/256	0.997	1.050				
1/512	1.068					

Comparison of energy-degradation spectra from the present work and the corresponding spectra from the Spencer-Fano theory. T_o is the source energy, T the spectral energy, $y(T,T_o)$ the differential tracklength distribution, S(T) the electron stopping power, and $F(T,T_o)$ the quantity which plays the role of a stopping power in the Spencer-Fano theory.

TABLE 2

$T\ dG(T,T_o)/dT$

T (eV) \ T_o =	100,000	50,000	20,000	10,000	5000	2000	1000 eV
50,000	1.03						
20,000	0.462	0.822					
10,000	0.274	0.475	1.08				
5000	0.217	0.300	0.589	1.14			
2000	0.200	0.230	0.345	0.552	1.00		
1000	0.222	0.243	0.309	0.405	0.613	1.35	
500	0.272	0.286	0.327	0.376	0.485	0.838	1.52
200	0.513	0.509	0.529	0.522	0.550	0.653	0.882
100	0.684	0.692	0.691	0.697	0.707	0.740	0.832
50	1.00	1.00	1.01	1.01	1.02	1.03	1.06
20	1.60	1.61	1.61	1.62	1.64	1.65	1.66
10	1.88	1.88	1.89	1.90	1.89	1.91	1.92
5	1.73	1.73	1.74	1.75	1.77	1.78	1.80

Scaled energy degradation spectra in water vapor. The quantity dG/dT is the differential yield, per spectral energy interval, of inelastic collisions produced by the primary electrons and all their secondaries. The differential yield is normalized to an energy input of 100 eV into the medium.

TABLE 3

ENERGY INTERVAL (eV)	PERCENT CONTRIBUTION OF AUGER ELECTRONS					
	T_o = 100	50	20	10	5	2 keV
500 - 490	55.0	50.8	44.1	35.4	25.1	8.4
490 - 480	39.3	35.3	30.4	22.4	12.7	4.6
400 - 380	27.9	26.0	22.4	15.6	11.5	3.4
300 - 280	20.9	19.2	16.1	11.1	8.9	3.3
200 - 190	13.3	12.3	11.3	8.1	5.4	2.4
100 - 96	7.1	6.5	6.1	5.2	3.6	2.0
50 - 48	5.7	5.1	4.4	3.0	2.4	1.3
30 - 28	5.0	4.6	4.0	2.8	2.3	1.0
20 - 19	4.9	4.4	3.9	3.0	2.4	1.1
10 - 9.6	4.9	4.4	4.0	2.8	2.4	0.8
5 - 4.8	4.9	4.4	3.8	2.6	2.4	1.1

Percent contribution by Auger electrons (and their secondaries) to the energy degradation spectrum. The contributions shown are averaged over the indicated energy intervals.

TABLE 4

T_o	$\overline{n_i}$	$\overline{n_i^2} / \overline{n_i}$
12.6	0.0	1.00
13	0.037	0.96
14	0.139	0.86
15	0.213	0.79
20	0.347	0.65
50	1.225	0.40
100	2.822	0.33
200	6.152	0.28
500	16.24	0.25
1000	32.98	0.24
2000	66.33	0.24
5000	165.9	0.24
10,000	330.7	0.25
20,000	659.1	0.25
50,000	1639.	0.25
100,000	3270.	0.25

Mean number of ions produced, $\overline{n_i}$, and Fano factor $f = \overline{n_i^2} / \overline{n_i}$, for electrons of initial energy T_o slowed down in water vapor.

TABLE 5

T_o (eV)	W (eV)
20	57.67
30	47.07
50	40.81
100	35.43
200	32.51
300	31.54
500	30.78
1000	30.32
2000	30.15
3000	30.17
5000	30.14
10,000	30.23_5
20,000	30.34_5
30,000	30.44_5
50,000	30.51_5
100,000	30.58

Average energy needed for the production of an ion pair, W, for electrons with initial energy T_o in water vapor.

TABLE 6

T_o (eV)	W, eV			
	OSG	KG	P	Present Work
20	73.6	82.5	44.6	57.7
40	33.3	43.2	36.9	43.2
60	26.9	37.9	34.6	39.0
80	25.4	35.8	33.4	37.0
100	25.0	34.9	32.7	35.4
200	25.0	33.2	31.2	32.5
400	25.0	31.7	30.6	31.0
600	25.0	30.9	30.5	30.6
800	25.0	30.6	30.4	30.4
1000	25.0	30.0	30.4	30.3

Value of W, the mean energy needed to produce an ion pair, as calculated by various authors.

OSG: Olivero, Stagat and Green (1972)

KG: Kutcher and Green (1976)

P: Paretzke (1975)

The results of Kutcher and Green are for liquid water; all the others are for water vapor.

TABLE 7

Percent	Spectral energy T, keV		
100	100.0	10.0	1.0
90	81.1	8.41	0.921
80	56.6	6.12	0.765
70	29.5	3.96	0.588
60	7.43	1.95	0.438
50	0.802	0.662	0.300
40	0.249	0.244	0.175
30	0.101	0.100	0.0921
20	0.0494	0.0490	0.0481
10	0.0276	0.0274	0.0270
0	0.0126	0.0126	0.0126
T_o	100	10	1 keV

Cumulative
/contributions of ionization events from various spectral regions of the energy degradation spectrum. For example, for an electron with initial energy T_o = 100 kEV, 50 percent of the ionization is produced by electrons (primary and secondaries) with energies less than 0.802 keV.

TABLE 8

T, keV	dE/dx, MeV/g cm^{-2} (a)	(b)	T, keV	dE/dx, MeV/g cm^{-2} (a)	(b)
100	4.14	4.14	0.3	200.	230.
50	6.65	6.65	0.2	221.	257.
30	9.73	9.73	0.1	221.	
20	13.3	13.3	0.05	130.	
10	22.5	22.8	0.03	67.7	
5	36.3	38.8	0.02	27.8	
3	53.5	56.9	0.01	2.86	
2	70.4	76.2	0.005	0.035	
1	110.	122.	0.003	0.0021	
0.5	165.	183.	0.002	0.013	

Stopping Power for Electrons in Water Vapor

(a) Present work; derived from detailed cross sections for ionization and excitation.

(b) From Bethe stopping-power formula, evaluated with mean excitation energy I = 71.7 eV.

TABLE 9

ELECTRON ENERGY (eV)	CSDA RANGE ($g\ cm^{-2}$)	ELECTRON ENERGY (eV)	CSDA RANGE ($g\ cm^{-2}$)
100,000	1.432×10^{-2}	1000	7.156×10^{-6}
80,000	9.723×10^{-3}	800	5.446×10^{-6}
60,000	5.913×10^{-3}	600	3.980×10^{-6}
50,000	4.303×10^{-3}	500	3.343×10^{-6}
40,000	2.911×10^{-3}	400	2.764×10^{-6}
30,000	1.756×10^{-3}	300	2.236×10^{-6}
20,000	8.635×10^{-4}	200	1.761×10^{-6}
15,000	5.234×10^{-4}	150	1.540×10^{-6}
10,000	2.828×10^{-4}	100	1.319×10^{-6}
8000	1.799×10^{-4}	80	1.223×10^{-6}
6000	1.096×10^{-4}	50	1.037×10^{-6}
5000	8.095×10^{-5}	40	9.509×10^{-7}
4000	5.615×10^{-5}	30	8.333×10^{-7}
3000	3.538×10^{-5}	20	6.103×10^{-7}
2000	1.890×10^{-5}	15	3.241×10^{-7}
1500	1.240×10^{-5}	12.6	0.0

Electron range in water vapor.

TABLE 10

Δ	100 keV			50 keV		
(eV)	(a)	(b)	% DIFF	(a)	(b)	% DIFF
400	0.6940	0.6672	3.9	0.7167	0.6855	4.4
200	0.6467	0.6001	7.2	0.6649	0.6160	7.3
100	0.5993	0.5267	12.1	0.6128	0.5408	11.8
50	0.5519	0.4551	17.5	0.5607	0.4674	16.6

Δ	30 keV			20 keV		
(eV)	(a)	(b)	% DIFF	(a)	(b)	% DIFF
400	0.7355	0.6982	5.1	0.7522	0.7088	5.8
200	0.6797	0.6265	7.8	0.6929	0.6352	8.3
100	0.6237	0.5499	11.8	0.6331	0.5574	11.9
50	0.5675	0.4750	16.3	0.5731	0.4812	16.0

Δ	10 keV			5 keV		
(eV)	(a)	(b)	% DIFF	(a)	(b)	% DIFF
400	0.7854	0.7393	5.9	0.8262	0.7854	4.9
200	0.7191	0.6616	8.0	0.7518	0.7029	6.5
100	0.6518	0.5804	11.0	0.6752	0.6168	8.6
50	0.5840	0.5006	14.3	0.5974	0.5316	11.0

Ratio of the restricted to the total electron stopping power in water vapor, as a function of the cut-off energy Δ .

(a) Calculated according to the Bethe theory

(b) Derived from the excitation and ionization cross sections used in the present work.

Note that the restricted stopping power excludes energy transfers in which the secondary electron acquires a kinetic energy greater than Δ .

NBS-114A (REV. 7-73)

U.S. DEPT. OF COMM. BIBLIOGRAPHIC DATA SHEET	1. PUBLICATION OR REPORT NO. NBSIR 77-1283	2. Gov't Accession No.	3. Recipient's Accession No.

4. TITLE AND SUBTITLE

Spectra from Energy Degradation of Fast Electrons in Water

5. Publication Date

6. Performing Organization Code

7. AUTHOR(S)

Martin J. Berger

8. Performing Organ. Report No.

9. PERFORMING ORGANIZATION NAME AND ADDRESS

NATIONAL BUREAU OF STANDARDS
DEPARTMENT OF COMMERCE
WASHINGTON, D.C. 20234

10. Project/Task/Work Unit No.
2401437/2401439

11. Contract/Grant No.

12. Sponsoring Organization Name and Complete Address *(Street, City, State, ZIP)*

Office of Naval Research, Arlington, Va. 22217
U.S. Energy Research & Development Admin., Wash., D.C. 20545

13. Type of Report & Period Covered
Interim report - Oct. 1976-July 1977

14. Sponsoring Agency Code

15. SUPPLEMENTARY NOTES

16. ABSTRACT *(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here.)*

This report gives data for energy degradation spectra in water vapor. Calculations utilized the Monte Carlo method and included Auger electrons. Results include 1) a new energy scaling principle at low energies, 2) values difference source energies for W, the mean energy loss per ion pair formed, 3) data on the electron range calculated by continuous-slowing-down approximation, and 4) data on stopping powers and restricted stopping powers as compared with results from the Bethe theory.

17. KEY WORDS *(six to twelve entries; alphabetical order; capitalize only the first letter of the first key word unless a proper name; separated by semicolons)*

Charged particle transport; electron energy degradation; electron penetration; slowing-down spectra; stopping power in water; W-value in water.

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