Stopping powers of energetic electrons penetrating condensed

matter— theory and application

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Abstract In this review article, the motivation of studying inelastic energy loss for energetic electrons penetrating through matter and the corresponding technological importance have been outlined. The theoretical development and method for the calculation of stopping powers are described. The stopping power data tables for a group of polymers and bioorganic compounds are presented, and the application aspects of the stopping power data are briefly discussed.

Keywords Stopping power, Electron inelastic scattering, Organic compounds, Bethe theory, Dielectric response theory

CLC number 0571.33

1 Introduction

The interaction of electron beam with materials is of especial importance and is the physical foundation for many fields of modern science and technology, such as scanning electron microscopy (SEM), electron probe microanalysis (EPMA), electron beam lithography (EBL), radiation biology and radiation therapy.^[1–5]

Using a beam of electrons with incident energy E to impact a specimen, various signals such as secondary electrons, Auger electrons, X-ray and backscattered electrons could be generated on the surface of the specimen due to the interactions between the penetrating electrons and the atoms in the specimen. By detecting the secondary electrons or backscattered electrons from the specimen, a high-resolution image of the sample surface can be presented in SEM. By use of the X-ray from the specimen, the quantitative information of the components in micro-region of the specimen can also be provided by EPMA. Electron beam lithography is an important technique for the fabrications of large scale and very large scale integrated circuits. In EBL, an incident e-beam depicts a pattern on resist film and the penetrating electrons interact with the resist film, depositing their energies

Received date: 2004-06-15

in the resist film and thus causing the breaking of bonds. Finally, the pattern contour can be obtained by suitable development steps.

In radiation biology, the interactions of radiation origin with biology material lead to various radiation effects in biology materials. These radiation effects are applied extensively to cell engineering, gene engineering and induced mutation in plant breeding.^[6-9] Electron beam is one of the important radiation sources. The interactions between other radiation beams and biology materials can result in secondary electrons with a wide energy distribution and these secondary electrons, in turn, interact with the biology materials. Therefore, the studies on the interactions of energetic electrons with biology materials play an especially important role in the radiation biology. In radiation therapy, electron beam is also an important radiation sources. Similarly, the interactions of other radiation sources with biology tissues can create secondary electrons, which will interact with the tissues as well.

As described above, in SEM, EPMA, EBL, radiation biology and radiation therapy, the interaction of energetic electron with material is the fundamental problem. This problem is also a complicated physical process and can be summed up as two types of scattering events, namely, the elastic and inelastic scattering. When a primary electron enters the solid, it transfers part of its energy to atomic electrons in the inelastic scattering events, resulting in ionization or excitations. Electron stopping power is a basic physical quantity for characterizing the inelastic interaction of electron with material and is essentially important for understanding basic physical process of the electron-material interaction. On the other hand, as the rapid development of computer technology, Monte Carlo simulation has become a main method to investigate the transport behavior of energetic electrons in materials.^[5,10] Electron stopping powers are the basic input data in Monte Carlo simulations of electron transport in matter. Therefore, the stopping power is of important significance not only for theoretical study but also for practical application in the technologies relating to electron beam.

In Section 2, two basic theoretical systems for the calculation of stopping powers, the non-relativistic Bethe theory and that based on linear dielectric response theory, and their developments are described. Due to the importance of stopping powers of electron inelastic scattering for organic compounds in many application fields, such as electron beam lithography, radiation biology and radiation therapy, the results of stopping powers obtained by our group for a set of polymers and bioorganic compounds are tabulated in Section 3. Finally, a brief summary on this review is made.

2 Theoretical development and calculation method

2.1 Bethe theory

In general, there are two main theoretical systems for the calculation of stopping power of energetic electrons in matter. One is the non-relativistic Bethe theory and the other is based on the linear dielectric response theory.

On the basis of Born approximation, Bethe^[11,12] derived the following formula to calculate the stopping power of energetic electrons in matter:

$$-\frac{\mathrm{d}E}{\mathrm{d}s} = 785 \frac{\rho Z}{AE} \ln \frac{1.166E}{J} \quad (\mathrm{eV/Å}) \tag{1}$$

where *E* is the energy of the penetrating electron (in eV), *s* is the path length along the trajectory (in Å), ρ is the density (in g/cm³)of the target material, *Z* is the atomic number of the target atom, *A* is the atomic weight of the target, and *J* is the mean ionization potential of the material (in eV). Commonly, the value of *J* can be evaluated according to Berger and Seltzer's formula^[13] as follows

$$\begin{cases} J = 9.76 + 58.5Z^{-0.19} \quad (eV) \text{ for } Z \ge 13 \\ J = 11.5Z \quad (eV) \text{ for } Z \le 12 \end{cases}$$
(2)

Bethe stopping power theory can well describe the energy loss of the penetrating electrons in matter at high energies. But this theory is, in general, invalid for low-energy electron due to the Born approximation used in this theory. For example, Bethe stopping power theory presents a decrease of the energy loss of electrons in Au at energies less than several keV and the energy loss becomes negative at energies less than about 1 keV. Obviously, these are in conflict with experimental facts. In spite of the invalidation mentioned above for Bethe theory, it is desirable to extend Bethe formula into low-energy region with an empirical or semi-empirical method because Bethe's expression is very simple and thus is easy to use in Monte Carlo model.

Rao-Sahib and Wittry^[14] extended Bethe's formula to low-energy region by using a parabolic extrapolation with the consideration that path length per unit energy loss approaches zero as the energy goes to zero, resulting in the following modified Bethe's expression (Rao.Sahib–Wittry equation)

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$$\begin{cases} -\frac{\mathrm{d}E}{\mathrm{d}S} = 785 \frac{\rho Z}{AE} \ln(\frac{1.166E}{J}) \quad (\mathrm{eV/\mathring{A}}) \quad \mathrm{for} \quad E > 6.338J \\ -\frac{\mathrm{d}E}{\mathrm{d}S} = 624 \frac{\rho Z}{A\sqrt{EJ}} \quad (\mathrm{eV/\mathring{A}}) \quad \mathrm{for} \quad 0 \le E < 6.338J \end{cases}$$
(3)

Rao-Sahib and Wittry applied Eq.(3) to the investigation of X-ray continuum from thick elemental targets with considerable success. Also, Kotera *et al.*^[15] used this modified Bethe formula to simulate the low-energy electron scattering in an aluminum target.

In the study of atomic number correction for quantitative electron-probe microanalysis, it is required to calculate the stopping power factor, which They wrote Bethe's expression in terms of the variable V=E/J, i.e.

$$-\frac{\mathrm{d}E}{\mathrm{d}S} = \frac{\rho Z}{JA} \times \frac{1}{f(V)} \quad (\mathrm{eV/Å})$$
(4)

where the function f(V) is in the form:

$$f(V) = \frac{V}{785\ln(1.166V)}$$
(5)

The function f(V) against V is approximately parabolic at high V and hence Love *et al.* expanded the f(V) in a series of $V^{1/2}$, i.e.

$$f(V) = A + BV^{1/2} + CV + DV^{3/2} + \dots$$
(6)

Based on the reality that dE/dS must tend to infinity as $V \rightarrow 0$, the f(V) tends to zero as $V \rightarrow 0$ and thus the coefficient A is equal to zero. They further determined the other coefficients B, C, D,... and finally gave f(V) in an approximate form as follows

$$f(V) = 1.18 \times 10^{-3} V^{1/2} + 1.47 \times 10^{-4} V \tag{7}$$

According to Eqs.(4) and (7), Love *et al.* expressed Bethe formula as

$$-\frac{\mathrm{d}E}{\mathrm{d}S} = \frac{\rho Z}{JA} \times \frac{10^3}{1.18V^{1/2} + 0.147V} \text{ (eV/Å)}$$
(8)

Using Eq.(8), Love *et al*.^[17] obtained a simple form for the stopping power factor, which was applied to the atomic number correction to EPMA with considerable success. Their equation has also been used in other applications, for example in quantitative studies of electron beam lithography.^[18]

From Lindhard's theory for an ion beam, Kanaya and Okayama^[19] derived a formula to calculate energy loss of electrons penetrating into a target material due to inelastic collisions with atomic electrons. This formula includes two adjustable parameters and can be written as

$$-\frac{\mathrm{d}\,E}{\mathrm{d}\,S} = \lambda_s \,\frac{\pi \cdot 2^{2/s-1} a^{2-2/s} e^{4/s} nZ}{1-1/s} \cdot E^{1-2/s} \quad (9)$$

where Z is the target atomic number, *n* is the atomic density of the target materials, $a = 0.8853a_0Z^{-1/3}$, a_0 is the Bohr radius, *e* is the electron charge, and *s* and λ_s are the unknown parameters. The parameters *s* and λ_s are so determined that the calculated electron range coincides with the experimental extrapolated range. For instance, Kotera *et al.*^[20] gave s = 1.3 and $\lambda_s = 0.115$, respectively, for Au target. However, it is clear that the determinations of both *s* and λ_s are a complex calculation process. Thus, Kanaya and Okayama's equation is not convenient to evaluate the stopping power of the penetrating electrons in matter.

Joy and Luo^[21] analyzed Rao-Sahib–Wittry equation and Love *et al.*'s equation in detail, and compared the corresponding stopping powers with those based on a detailed first-principles computation. Consequently, Joy and Luo pointed out that the Rao-Sahib–Wittry and Love *et al.*'s expressions have no inherent physical significance and also overestimate the stopping power by a factor of two or more for energies of a few hundred electron volts or less. Therefore, according to Rao-Sahib–Wittry and Love *et al.*'s equations, those quantities related to the stopping power, such as electron range, secondary electron and X-ray yields, will be in error.

According to several authors, e.g. Reimer and Stelter,^[22] Ritchie *et al.*,^[23] and Shimizu and Ichimura,^[24] Joy and Luo wrote a more general version of the Bethe equation in the form:

$$-\frac{\mathrm{d}E}{\mathrm{d}S} = 785 \frac{\rho Z}{AE} \sum_{i} \frac{Z_{i}}{Z} \ln \frac{E}{E_{i}}$$
(10)

where Z_i is the occupancy of the *i*-th shell as tabulated in the work of Reimer and Sletter, E_i is the corresponding binding energy.^[24] In Eq.(10), for a given energy *E*, the summation is only carried out over those terms *i*, for which $E > E_i$. The stopping powers derived from Eq.(10) are in excellent agreement with those computed from other models.^[25]

On the other hand, Joy and Luo regarded the quantity J in Eq.(1) as energy dependent rather than constant and wrote Bethe formula in the form:

$$-\frac{\mathrm{d}E}{\mathrm{d}S} = 785 \frac{\rho Z}{AE} \ln\left[\frac{1.166(E+kJ)}{J}\right] \qquad (11)$$

The equivalent form of Eq.(11) can be written as

$$\frac{\mathrm{d}E}{\mathrm{d}S} = 785 \frac{\rho Z}{AE} \ln \left[\frac{1.166E}{J'}\right] \tag{12}$$

where

$$J' = \frac{J}{1+k\frac{J}{E}}$$
(13)

Eq.(13) shows clearly that J' is dependent on energy.

For elements C, Al, Si, Ni, Cu, Ag and Au, by fitting Eq.(11) to the numerical data of the stopping powers calculated with Eq.(10), Joy and Luo gave the best fitted k values, which are 0.77, 0.815, 0.822, 0.83, 0.83, 0.852 and 0.851, respectively.

According to the dependence of these k values on atomic number Z, Gauvin *et al.*^[26] gave an empirical formula to calculate value of k, i.e.

$$k = 0.734 Z^{0.037} \tag{14}$$

The modified Bethe formula of Joy and Luo enables the calculations of stopping powers to be extended into low-energy region while maintaining the simplicity and generality of the original Bethe expression. So, this formula has been widely used.^[27–29]

In above three modified Bethe formulas, the formula by Love *et al.* is the only one with which the analytical expression of the Bethe range can be obtained by integration. On the other hand, it is desirable to be able to calculate the Bethe ranges of the penetrating electrons in Monte Carlo simulation studies on electron beam lithography, SEM and EPMA. By comparing the formula by Love et al. with that of Joy

and Luo, Tan and He^[30] proposed an improved Love *et al.* formula.

Let $U = (E/J)^{1/2}$, then Love *et al.*'s formula can be rewritten as

$$-\frac{\mathrm{d}E}{\mathrm{d}S} = \frac{\rho Z}{AE} \times \frac{10^3}{UG_L(U)} \tag{15}$$

where

$$G_L(U) = 1.18 + 0.147U \tag{16}$$

In addition, the calculated stopping power based on Joy and Luo equation is, in practice, not strongly sensitive to the value of k, and thus the formula of Joy and Luo can be written in the from:

$$-\frac{\mathrm{d}E}{\mathrm{d}S} = 785 \frac{\rho Z}{AE} \ln \left[1 + \frac{1.166E}{J}\right] \tag{17}$$

Tan and He further expressed Eq.(17) as

$$-\frac{\mathrm{d}E}{\mathrm{d}S} = \frac{\rho Z}{AE} \times \frac{10^3}{UG_I(U)} \tag{18}$$

where

$$G_J(U) = \frac{U}{0.785 \ln(1 + 1.166U^2)}$$
(19)

Let $D(U) = G_J(U) - G_L(U)$, by calculating and analyzing the values of D(U) with variable U in the range of $U \le 31$, which corresponds to the applicable range, $E/J \le 1000$, of the equation of Love *et al.*, Tan and He gave an improved formula of Love *et al.*, namely

$$\frac{\mathrm{d}E}{\mathrm{d}S} = \frac{\rho Z}{JA} \times \frac{10^3}{0.303(E/J)^{-1/2} + 1.16(E/J)^{1/2} + 0.147(E/J)}$$
(20)

With Eq.(20), the calculated stopping powers are in good agreement with those predicted by Joy and Luo,^[21] and Tung *et al.*^[25] Hence, the modified formula of Love *et al.* has solved the problems of overestimating energy loss of the penetrating electrons at low energies by Love *et al.*'s equation while retaining the desirable characteristic of being able to derive the analytical expression of the Bethe range by integration.

It should be pointed out that the above formulas

for calculating stopping power of the penetrating electrons in matter have different characteristics and thus are applicable in different cases.

2.2 Linear dielectric response theory

The inelastic interactions of the incident electron with a target material can be divided into two parts, i.e. the interaction of the incident electron with the valence electrons of a target atom and that between the incident electron and the core electrons. Therefore, the

$$(-dE/dS)_{t} = (-dE/dS)_{v} + (-dE/dS)_{c}$$
(21)

where the subscripts indicate the total, valence and core parts, respectively.

Based on the electromagnetic principle and free electron gas model, the dielectric response theory to characterize the inelastic interaction of the penetrating electrons with solid has been well developed since 1950's and the general descriptions of this theory have been presented by several well-known authors, e.g. Lindhard,^[31] Ritchie^[32] and Pines.^[33] Nowadays, it is considered that the dielectric response theory is perhaps the best approach to characterize the inelastic scattering of the incident electron in solids. According to the dielectric response theory, a basic expression, i.e. the electron inelastic differential cross section can be given as

$$\frac{\mathrm{d}^2 \sigma}{\mathrm{d}(\hbar\omega) \mathrm{d}q} = \frac{1}{\pi a_0 E} \operatorname{Im}[-1/\varepsilon(q,\omega)] \frac{1}{q} \quad (22)$$

where *E* is the kinetic energy of the incident electron, a_0 is the Bohr radius, $\hbar \omega$ and $\hbar q$ are the energy loss and the momentum transfer, respectively, from the incident electron into a medium described by the dielectric response function $\epsilon(q, \omega)$. Over the allowed values of $\hbar \omega$ and $\hbar q$, the stopping power corresponding to the interactions between the incident electron and the valence-electron system can be obtained from the relevant integrations of Eq.(22), i.e.

$$-\left(\frac{\mathrm{d}E}{\mathrm{d}S}\right)_{\nu} = \frac{1}{\pi a_0 E} \int_0^E \mathrm{d}(\hbar\omega)(\hbar\omega) \int_{q_-}^{q_+} \frac{1}{q} \mathrm{Im}[-1/\varepsilon(q,\omega)] \mathrm{d}q$$
(23)

where $q_{\pm} = \sqrt{2m/\hbar^2} \left(\sqrt{E} \pm \sqrt{E - \hbar\omega}\right)$. This expression is due to the conservation of energy and momentum, and is based on the assumption that the energy-momentum relation for a non-relativistic electron in the solid does not differ appreciably from that of a free electron in vacuum. The quantity Im[$-1/\varepsilon(q,\omega)$] is commonly referred to as the energy loss function.

From Eq.(23), in the frame of the dielectric response theory, it is clear that the key problem for calculating the stopping power is to derive the energy loss function Im[$-1/\varepsilon(q,\omega)$]. However, the well-known Lindhard dielectric function is only applicable for limited materials, so called nearly-free-electron materials. Therefore, many efforts have been devoted to the calculations of the energy loss function. A main method for obtaining the energy loss function is to use the optical energy loss function (OELF) Im[-1/ $\varepsilon(0,\omega)$] and to extend it into the area with q>0 by introducing a suitable dispersion relation. The optical energy loss function can be calculated from experimental optical data, the refractive index and the extinction coefficients, which are available for a number of materials.^[34,35] A practical approach to derive the energy loss function, $\text{Im}[-1/\varepsilon(q,\omega)]$, is to expand it or the imaginary part of the dielectric function, $\varepsilon(0,\omega)$, as a sum of Drude-type functions, which has been first proposed by Ritchie and Howie.^[36] Each term in the sum of Drude-type functions has three adjustable parameters. All the parameters in the sum of Drude-type functions will be determined by fitting the expression of the sum of Drude-type functions, in the limit $q \rightarrow 0$, to experimental OELF.

Kwei *et al.*^[37,38] expressed the imaginary part, $\varepsilon_2(0,\omega)$, of the dielectric function, $\varepsilon(0,\omega)$, as a sum of Drude-type functions, namely

$$\varepsilon_2(0,\omega) = \sum_j \frac{A'_j \gamma_j \omega}{(\omega_2 - \omega_j^2)^2 + \gamma_j^2 \omega^2} \qquad (24)$$

The parameters A_j' , ω_j and γ_j are determined by fitting ε_2 in the form of Eq.(24) to the values computed from experimental optical data in the range of $\hbar\omega$ less than several tens of eV.

The real part, $\varepsilon_1(0, \omega)$, of the dielectric function is further derived from the Kramers–Kronig relation. The result is

$$\varepsilon_{1}(0,\omega) = 1 - \sum_{j} \frac{A_{j}'(\omega^{2} - \omega_{j}^{2})}{(\omega_{2} - \omega_{j}^{2})^{2} + \gamma_{j}^{2}\omega^{2}}$$
(25)

Consequently, the optical energy loss function can be calculated by

$$\operatorname{Im}[-1/\varepsilon(0,\omega)] = \frac{\varepsilon_2}{\varepsilon_1^2 + \varepsilon_2^2}$$
(26)

Finally, by introducing a suitable dispersion relation $\omega_j = \omega_j(q)$, to extend Im[-1/ $\varepsilon(0,\omega)$] into the area with q>0 can be performed and thus the energy loss func-

tion, $\text{Im}[-1/\varepsilon(q,\omega)]$, is obtained. A commonly used dispersion relation is

$$\omega_q = \omega_p + \hbar q^2 / 2m \tag{27}$$

where ω_p is the plasmon frequency of the free-electron gas and *m* is the mass of electron. Kwei *et al.* applied the method described above to the calculation of stopping powers for semiconducting III-V compounds and other several solids. Emfietzoglou and Moscovitch^[39] have also used similar approach to calculate the electron stopping powers for liquid water.

To obtain the energy loss function by making use of the Drude-type function, another method is to directly expand $\text{Im}[-1/\varepsilon(q,\omega)]$ as a sum of the Drude-type function, i.e.

$$\operatorname{Im}[-1/\varepsilon(q,\omega)] = \sum_{j} \frac{A_{j}\gamma_{j}\hbar\omega}{(\hbar^{2}\omega^{2} - E_{j}^{2}(q))^{2} + \gamma_{j}^{2}\hbar^{2}\omega^{2}}$$
(28)

where $E_j(q)$ is the dispersion relation. Similarly, the parameters A_j , γ_j and $E_j(0)$ are determined by fitting the expression of the sum of Drude-type functions, in the limit $q \rightarrow 0$, to experimental optical energy loss function. This approach has been extensively used by many workers.^[40–42]

Using the methods described above, the stopping powers, $(-dE/dS)_v$, corresponding to the interaction of the incident electron with the valence electrons system of the condensed matter, can be calculated. For $(-dE/dS)_c$, the contributions from core electrons, are commonly calculated according to Gryzinski.^[43] Kwei and Tung^[37] have also applied the local plasma approximation for the evaluation of $(-dE/dS)_c$. One characteristic of the above methods for deriving $\text{Im}[-1/\varepsilon(q,\omega)]$ is to use experimental optical data. In addition, the numerical fitting process for the determination of all parameters in the sum of Drude type function is tedious. Particularly, most of the transitions for noble metals and some alkali halides present a very complex structure in $\text{Im}[-1/\varepsilon(\omega)]$, and thus many terms of Drude type function are required. As an example, the calculation of the optical energy loss function of KCl is presented in Fig.1. In this calculation, the optical energy loss function of KCl is expanded as a sum of the ten terms of Drude type function and the corresponding parameters are taken from Boutboul *et al.*'s calculation.^[41] The experimental optical energy loss function is also plotted in this figure for comparison. From this figure, it is shown clearly that the optical energy loss function of KCl presents a complex structure.



Fig.1 Experimental optical energy loss function of KCl (solid line) and that obtained by using fitted expansion formula of Eq.(28) (dashed line) versus the photon energy.

A method referred to as model insulator theory^[44] has been used to calculate the Im[$-1/\varepsilon(q,\omega)$] from valence electrons' response by Painter et al. and Ashley et al.^[45–47] In this method, the imaginary part, $\varepsilon_2(q,\omega)$, of the dielectric response function for the valence electrons is described with the model insulator theory and its adjustable parameters are fixed by fitting the theoretical expression of the imaginary part of the dielectric response function in the optical limit $(q \rightarrow 0)$ to experimental $\varepsilon_2(\omega)$ spectrum. The real part, $\varepsilon_1(q,\omega)$, of the dielectric response function is derived from the Kramers-Kronig relation. Also, the ionization contribution from inner shell electrons is calculated with the generalized oscillator strengths. This method has been applied to calculate the stopping powers for organic compounds and water with considerable success.

As mentioned above, the stopping powers are divided into two parts, i.e. contributions of the valence electrons and inner shell electrons, respectively. The calculation of the energy loss function from the valence electrons' response results in a tedious numerical fitting process. Another method is the so-called electron gas statistical model, pioneered by Lindhard and co-workers,^[48] which has been used to the calculation of stopping power for the charged-particle by many authors.^[36] In this method, a basic assumption is that the stopping medium is characterized by a space-varying electron density, n(r), and the contribution of the electrons in a small volume element d^3r at rto the stopping power or the inverse mean free path is the same as that of the same number of free electrons in an electron gas having the same density. So, the total stopping power or inverse mean free path is computed by averaging these functions of the electron density n(r) over the volume of the solids. Based on this assumption, Tung *et al.*^[48] presented the following approach for the inverse mean free path $<\mu(E)>$ and the electron stopping power <S(E)> resulting from the interactions of the incident electron with the total electron system of the solid :

$$\begin{cases} \langle \mu(E) \rangle = \int \mu[E, n(r)] d^3 r / \int d^3 r \\ \langle S(E) \rangle = \int S[E, n(r)] d^3 r / \int d^3 r \end{cases}$$
(29)

where the integration is over the volume of the Wigner-Seitz cell for a given solid, n(r) is considered in spherical symmetry, and

$$\mu[E, n(r)] = \frac{\hbar}{\pi a_0 E} \int_{0}^{(E-E_{\rm F})/\hbar} \mathrm{d}\,\omega \int_{k_{\rm T}}^{k_{\rm T}} \mathrm{Im}[-1/\varepsilon(k,\omega)] \frac{\mathrm{d}\,k}{k}$$
(30)

$$S[E, n(r)] = \frac{\hbar^2}{\pi a_0 E} \int_{0}^{(E-E_{\rm F})/\hbar} \omega \,\mathrm{d}\,\omega \int_{k_{\rm T}}^{k_{\rm T}} \mathrm{Im}[-1/\varepsilon(k, \omega)] \frac{\mathrm{d}\,k}{k}$$

where $E_{\rm F}$ is the Fermi energy, which is a function of $n(r), k_{\pm} = \sqrt{2m/\hbar^2} (\sqrt{E} \pm \sqrt{E - \hbar \omega})$. In this scheme the spherically symmetric electron density distribution n(r) was calculated by using a relativistic Hartree-Fock method. Based on the model described with Eqs.(29), (30) and (31), Tung et al. calculated the stopping powers for Al, Si, Cu, Ni, Ag and Au, and the calculated stopping powers are in good agreement with experimental results. In Tung et al.'s method, it is clearly shown that all electrons in solid are considered as one group but with a density varying from point to point in solid. There is, therefore, no distinction between valence and inner-shell electrons. Thus, Tung et al.'s method provides a systematic evaluation for the total stopping powers of the electron inelastic scattering in solid.

Based on the idea of statistical approximation

used by Tung *et al.*,^[48] Penn^[49] calculated the energy loss function Im[-1/ $\varepsilon(q, \omega)$] and derived Im[-1/ $\varepsilon(q, \omega)$] by averaging the energy loss function over a Wigner-Seitz cell, i.e.

$$\operatorname{Im}[-1/\varepsilon(q,\omega)] = \int \frac{\mathrm{d}^{3} r}{\Omega} \cdot \operatorname{Im}[-1/\varepsilon_{L}(q,\omega;r_{s}^{p}(\omega_{p}))]$$
(32)

where $\varepsilon_L(q,\omega; r_s^p)$ is the Lindhard dielectric function, the region of integration is a Wigner-Seitz cell of volume Ω , and

$$r_s^p(\omega_p) = \left[\frac{3}{4\pi n_p(r)}\right]^{1/3} a_0^{-1}$$
(33)

Here, $n_p(r)$ is a pseudo-charge-density chosen to ensure that

$$\operatorname{Im}[1/\varepsilon(0,\omega)] = \operatorname{Im}[1/\varepsilon(\omega)]$$
(34)

By further operating, Penn expressed the energy loss function as

$$\operatorname{Im}[-1/\varepsilon(q, \omega)] = \int_{0}^{\infty} \mathrm{d}\omega_{p} G(\omega_{p}) \operatorname{Im}[-1/\varepsilon_{L}(q, \omega; \omega_{p})]$$
(35)

where

(31)

$$G(\omega) = \frac{2}{\pi\omega} \operatorname{Im}[-1/\varepsilon(\omega)]$$
(36)

Penn's method shows that the energy loss function can be obtained from the optical dielectric response function $\varepsilon(\omega)$, provided a suitable dispersion relation is introduced. This method doesn't require a tedious numerical fitting process for deriving the energy loss function but directly uses the experimental optical data, and thus is convenient for use. Making use of the available experimental optical data and applying Penn's approach, Tanuma et al.[50-53] calculated electron mean free paths for a large number of elements and compounds. Also, starting with the doubly differential cross section given by Eq.(22) and using Penn's method, Ding and Shimizu^[54,55] derived an expression to calculate the excitation function and stopping power of electrons in metal solids and applied them to Monte Carlo simulations for electron probe microanalysis. Öztürk and Williamson Jr.[56] used Ding and Shimizu's method for the Monte Carlo simulations of ~ keV electron transport in solid media.

Constructing the Bethe surface by choosing a δ -oscillator representation of a free-electron gas, Ashley^[57] suggested a method for calculating the energy loss function from the optical data. This method is similar to Penn's method and directly uses the experimental optical data, thus referred to as optical data model. Ashley evaluated the stopping powers for many elements and compounds by using the optical data model. Furthermore, an important contribution of Ashley is to introduce, for the first time, the correction approach for taking into account the exchange effect between the incident electron and the electrons in the medium in an empirical way based on the non-relativistic Moller formula. By including the exchange effect correction, an obvious result is that the calculated stopping powers are substantially different from those of ignoring the exchange correction at low energies. Also, by means of a modified Born-Ochkur approximation,^[58] another method for incorporating the exchange correction into the electron inelastic scattering has been used.^[59] However, the differences between the calculated results based on these two

kinds of exchange correction methods need to be further compared.

3 Stopping power data tables and application aspects

In this section, based on the dielectric response theory and Penn's statistical model, we have presented a method^[60] for the calculation of stopping powers of electrons penetrating into a series of organic compounds in the energy range of $E \leq 10$ keV. We have incorporated the exchange effect corrections of Born-Ochkur into the calculations. Also, we have given an empirical method to obtain optical energy loss functions for a large number of organic compounds, for which optical data are not available. Further, the stopping power data tables for a group of important polymers and a group of important bioorganic compounds are presented and the applications of stopping power data are briefly discussed.

Based on the Penn's method, inserting Eq.(36) into Eq.(35), one finds that

$$\operatorname{Im}[-1/\varepsilon(q, \omega)] = \int_{0}^{\infty} \frac{2}{\pi\omega_{p}} d\omega_{p} \operatorname{Im}[-1/\varepsilon(\omega_{p})] \operatorname{Im}[-1/\varepsilon_{L}(q, \omega; \omega_{p})]$$
(37)

A single–pole approximation has also been applied in the work of Ritchie.^[36] In a single–pole approximation, the Lindhard's dielectric function can be expressed as

$$\operatorname{Im}[-1/\varepsilon_{\mathrm{L}}(q,\omega;\omega_{\mathrm{p}})] = \frac{\pi}{2} \frac{\omega_{\mathrm{p}}^{2}}{\omega_{q}} \delta(\omega - \omega_{q}) \quad (38)$$

where $\omega_q = \omega_q(q)$ is the dispersion relation.

Eq.(37) can be, therefore, written in the form

$$\operatorname{Im}[-1/\varepsilon(q,\omega)] = \int_{0}^{\infty} \frac{\omega_{p}}{\omega_{q}} \operatorname{Im}[-1/\varepsilon(\omega_{p})] \ \delta(\omega - \omega_{q}) d\omega_{p}$$
(39)

As adopted by many authors,^[36,37,42] we have used the simple dispersion relation given by Eq.(27), which depicts the reasonable behavior required for two extremes, i.e. $\omega_q \rightarrow \omega_p$ when $q \rightarrow 0$ and $\omega_q \rightarrow hq^2/2m$ as $q \rightarrow \infty$ corresponding to the Bethe ridge.

Incorporating Eq.(39) with the dispersion relation of Eq.(27) into Eq.(22) and applying Born-Ochkur's exchange correction method,^[58] the inelastic differential cross section can be written as

$$\frac{\mathrm{d}^2 \,\sigma}{\mathrm{d}(\hbar\omega) \,\mathrm{d}\,q} = \frac{1}{\pi a_0 E} \int_0^\infty \frac{\omega'}{\omega} \times \frac{1}{q} \,\mathrm{Im}[-1/\varepsilon(\omega')] \times \\\delta(\omega - \omega' + \frac{\hbar q^2}{2m}) C_{\mathrm{ex}} \,\mathrm{d}\,\omega'$$
(40)

where C_{ex} is a correction factor and is expressed as

$$C_{\rm ex} = 1 - \frac{Q}{E + \hbar\omega' - \hbar\omega} + \left(\frac{Q}{E + \hbar\omega' - \hbar\omega}\right)^2 (41)$$

where $Q = \hbar^2 q^2 / 2m$.

With Eq.(40) and over the allowed integration region, the SPs can be calculated by

$$-\frac{\mathrm{d}E}{\mathrm{d}S} = \frac{1}{2\pi a_0 E} \int_0^{E/2} (\hbar\omega) \operatorname{Im}[-1/\varepsilon(\omega)] v(\alpha) \,\mathrm{d}(\hbar\omega)$$
(42)

where $v(\alpha)$ is given as

$$v(\alpha) = \frac{2s}{(1+\alpha)(1+\alpha+s)} + \ln\{\frac{(1-\alpha^2)(1+\alpha)}{(1-\alpha-s)(1+\alpha+s)^2}\}$$
(43)

and $\alpha = \hbar \omega / E$, $s = \sqrt{1 - 2\alpha}$. It is obvious that the calculations of the SPs based on Eq.(42) involve only a single numerical quadrature, thus requiring less computational work.

Recently, much interest has been focused on the calculations of stopping powers for electrons in organic compounds, especially in bioorganic compounds, because these calculations are of obvious importance for the radiation biology, the analysis of polymers and the electron beam lithography. On the other hand, the methods described above need to use the experimental optical data, based on the dielectric response theory.

$$\operatorname{Im}[-1/\varepsilon(\omega)] = \begin{cases} f_1(\hbar\omega; a, b, c), \\ f_2(\hbar\omega; y_0(a), y_1, y_2), \\ f_3(\hbar\omega), \end{cases}$$

where $f_1(\hbar \omega; a, b, c)$ is a single Drude-type function and is given by

$$f_1(\hbar\omega; a, b, c) = \frac{a(\hbar\omega)}{[(\hbar\omega)^2 - b^2]^2 + c^2(\hbar\omega)^2}$$
 (45)

the parameters b and c are calculated with the following relations:

$$b = 19.927 + 0.9807 \overline{Z} \tag{46}$$

$$c = 13.741 + 0.3215 \ \overline{Z} \tag{47}$$

where \overline{Z} is the mean atomic number of the molecule (or monomeric unit) in each material, $f_3(\hbar\omega)$ is obtained numerically by use of atomic photoabsorption data of Henke *et al.*,^[61] and $f_2(\hbar\omega; y_0(a), y_1, y_2)$ is a parabolic interpolation function, with adjustable parameters $y_0(a)$, y_1 and y_2 , to link f_1 and f_3 smoothly. In $f_2(\hbar\omega; y_0(a), y_1, y_2)$, the parameters y_1 and y_2 are given by f_3 at energies $\hbar\omega = 50$ eV and 60 eV, respectively, and the parameter $y_0(a)$ is the value of f_1 at the energy $\hbar\omega = 40$ eV. Finally the parameter *a* and thus $y_0(a)$ are determined by the following *f*-sum rule, which the Im[-1/ $\varepsilon(\omega)$] given above must obey, in accordance with the dielectric response theory:

$$Z = \frac{2}{\pi \hbar^2 \Omega_{\rm p}^2} \int_0^\infty (\hbar\omega) \,\mathrm{Im}[-1/\varepsilon(\omega)] \,\mathrm{d}(\hbar\omega) \quad (48)$$

where $\Omega_{\rm p} = (4\pi n e^2/m)^{1/2}$, $n = N_{\rm av}\rho/M$ is the molecular density, $N_{\rm av}$ is Avogadro's number, ρ is the bulk den-

However, to our knowledge, there are only 14 organic compounds and water, for which the experimental optical data are available. These 14 organic compounds were listed in the work of Tanuma *et al.*^[53] Based on the structure features of Im[$-1/\varepsilon(\omega)$] of the organic compounds with available optical data and by means of the approach of Ritchie and Howie,^[36] we proposed an empirical approach^[60] to obtain the OELF for organic compounds in the energy range from 0 to infinity. This approach is briefly introduced as follows.

We express the OELF in the form:

$$\begin{aligned} &\hbar\omega \leqslant 40 \text{eV} \\ &40 \text{eV} \leqslant \hbar\omega \leqslant 50 \text{eV} \\ &\hbar\omega \geqslant 50 \text{eV} \end{aligned} \tag{44}$$

sity of the material and *M* is the molecule weight, *Z* is the total numbers of electrons per molecule, and *m* is the mass of electron. The $\text{Im}[-1/\varepsilon(\omega)]$ determined in this way not only has the structural feature of the OELF for organic compounds, but also satisfies the *f*-sum rule of the dielectric response theory, which is a standard check for the reliability of the OELF given.

Using our approach given by Eq.(44), the calculated values of $\text{Im}[-1/\varepsilon(\omega)]$ for the organic compounds listed in the work of Tanuma et al.[53] have been compared with the corresponding experimental data for photon energies lower than several tens of eV. Reasonable agreement between our theoretical values and the experimental data is obtained. Applying our methods of calculating the SPs by Eq.(42) and the evaluation for OELF by Eq.(44), we have calculated the SPs for the organic compounds mentioned above and compared them with other theoretical results.^[42,62] The comparisons show that our calculated SPs are in good agreement with the other theoretical results. In addition, our calculated SP values also converge to that predicted by Bethe-Bloch theory at high energy. These tests show the reliability of our methods.

The polymers (or macromolecules) are a large group of important materials having a great deal of applications. Using our method, we have performed the systematic calculations of SPs in the energy range of $E \leq 10$ keV for alanine and 9 polymers: polypropylene, polycarbonate, mylar, polyvinylalcohol, polyoxymethylene, polyacrylonitrile, polyvinylpyrrolidone,

polyvinylacetate and bakelite, for which experimental optical data have not ever been reported. These ten organic compounds and their composition, mass density, ρ (in g/cm³), M and \overline{Z} are presented in Table 1.

The calculated results of SPs are presented at selected energies in Table 2. In order to make these tables compact and clear, we use the code numbers, from 1 to 10, to represent the 10 compounds, respectively.

 Table 1
 The composition and parameters for 9 kinds of polymers and alanine

Material		Composition				$\alpha(\alpha/cm^3)$	М	$\overline{7}$
No.	Name	C H N O		p(g/cm)	111	L		
1	polypropylene	3	6			0.9	42	2.667
2	polycarbonate	16	14		3	1.2	254	4.06
3	mylar	10	8		4	1.4	192	4.545
4	polyvinylalcohol	2	4		1	1.3	44.05	3.428
5	polyoxymethylene	1	2		1	1.425	30.03	4
6	polyacrylonitrile	3	3	1		1.17	53.06	4
7	polyvinylpyrrolidone	6	9	1	1	1.25	111.14	3.529
8	polyvinylacetate	4	6		2	1.19	86.09	3.833
9	alanine	3	7	1	2	1.42	89.10	3.692
10	bakelite	44	36		6	1.4	660.7	4.047

Table 2	Stopping powers (in eV/	A) with Born–Ochkur exchange correction included for 9 kinds of p	polymers and alanine
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E(eV)	Material No.									
	1	2	3	4	5	6	7	8	9	10
20	0.1636	0.1409	0.1414	0.1707	0.1540	0.1427	0.1668	0.1411	0.1709	0.1652
30	0.4426	0.3750	0.3742	0.4577	0.4100	0.3799	0.4468	0.3765	0.4567	0.4398
40	0.9944	0.8372	0.8323	1.0259	0.9160	0.8486	1.0010	0.8419	1.0223	0.9820
50	1.7926	1.5784	1.5871	1.8994	1.7241	1.5974	1.8591	1.5776	1.9079	1.8507
60	2.4851	2.3185	2.3786	2.7176	2.5263	2.3405	2.6711	2.2956	2.7599	2.7169
80	3.2008	3.1619	3.3141	3.6084	3.4363	3.1837	3.5615	3.1003	3.7051	3.7030
100	3.4117	3.4568	3.6629	3.9124	3.7729	3.4746	3.8604	3.3841	4.0408	4.0472
150	3.2719	3.4410	3.7233	3.9010	3.8583	3.4414	3.8146	3.3970	4.0777	4.0243
200	2.9698	3.1848	3.4859	3.6226	3.6338	3.1748	3.5199	3.1646	3.8113	3.7206
300	2.4530	2.6850	2.9745	3.0678	3.1231	2.6660	2.9586	2.6886	3.2487	3.1321
400	2.0862	2.3085	2.5736	2.6443	2.7128	2.2869	2.5396	2.3215	2.8095	2.6905
500	1.8198	2.0273	2.2690	2.3260	2.3977	2.0054	2.2281	2.0443	2.4763	2.3615
600	1.6193	1.8126	2.0339	2.0815	2.1523	1.7910	1.9905	1.8307	2.2187	2.1106
800	1.3544	1.5264	1.7148	1.7464	1.8091	1.5034	1.6697	1.5386	1.8600	1.7781
1000	1.1762	1.3316	1.4958	1.5171	1.5718	1.3105	1.4527	1.3383	1.6150	1.5522
2000	0.7360	0.8464	0.9536	0.9571	0.9961	0.8327	0.9185	0.8476	1.0213	0.9872
3000	0.5480	0.6354	0.7177	0.7173	0.7491	0.6245	0.6880	0.6363	0.7665	0.7410
4000	0.4412	0.5140	0.5815	0.5798	0.6069	0.5049	0.5558	0.5149	0.6202	0.5993
5000	0.3716	0.4343	0.4918	0.4896	0.5132	0.4264	0.4692	0.4351	0.5240	0.5063
6000	0.3222	0.3775	0.4279	0.4255	0.4464	0.3705	0.4076	0.3782	0.4555	0.4401
8000	0.2565	0.3015	0.3421	0.3396	0.3569	0.2958	0.3253	0.3021	0.3639	0.3514
10000	0.2144	0.2525	0.2867	0.2844	0.2991	0.2477	0.2723	0.2531	0.3048	0.2943

Also, we have calculated the SPs for 10 kinds of bioorganic compounds: DNA, Adenine (C5H5N5), Guanine ($C_5H_5N_5O$), Thymine ($C_5H_6N_2O_2$), Cytosine $(C_4H_5N_3O),$ Uracil $(C_4H_4N_2O_2),$ Oleic acid (C₁₈H₃₄O₂), Linoleic acid (C₁₈H₃₂O₂), Arachidonic acid $(C_{20}H_{32}O_2)$ and Cellulose $(C_5H_{10}O_5)$. According to the compositions of the sodium salt of calf thymus DNA,^[63] we have formed the equivalent molecules (or monomeric units) of DNA, which consists of 58% A-T (Adenine-Thymine base pair) and 42% G-C (Guanine-Cytosine base pair). The parameters of these 10 bioorganic compounds are listed in Table 3. Table 4 gives the corresponding SP values calculated by our method with Born-Ochkur exchange correction included for electron energies from 20 eV to 10000 eV. Similarly, we use the code numbers, from 11 to 20, to represent these 10 bioorganic compounds, respectively.

The applications of electron stopping powers can be roughly divided into two aspects. One is the direct use in the technologies relating to electron beam. In SEM, the stopping power is used to evaluate the range of electron in solid and to calculate the yields of the secondary electrons.^[64] In quantitative electron-probe microanalysis, the stopping power is applied to calculate the stopping power factor,^[16] which is one of the quantities to determine the measured X-ray intensities from the specimen and the standard sample. In addition, the stopping power is often used for estimation of dose or other related quantities in radiation therapy.

As rapid development of computer technique and with increasing computational ability to resolve the complicated physical problems, Monte Carlo simulation has become a dominant method for the study of electron transport in matter. The electron stopping power is the basic input data for Monte Carlo simulation of energetic electron transport in matter. A typical example is the single scattering model^[65] in Monte Carlo simulation, which is widely applied in SEM, SAMP and EBL. In this model, the energy loss of the penetrating electrons in the inelastic scattering process is calculated by using stopping power, based on the continuous slowing down approximation. In addition, many quantities relating to electron scattering process are relevant to stopping power data. For example, the spatial distribution of energy dissipation is the most

important quantity for electron beam lithography and it determines the ultimate resolution of micro-fabrication by EBL. However, the energy dissipation in the resist film is closely related to the electron stopping power. Also, in the study of radiation induced DNA damage, the stopping power is used to calculate the energy dissipation in DNA.^[66] These dissipations of electron energies determine the numbers of DNA strand breakage.

The data of SPs given in Tables 2 and 4 can be used in the electron microscopy analysis for Monte Carlo simulations of energetic electrons transport and in the studies of various radiation effects for these important materials.

4 Summary

The interaction of energetic electrons with materials is the physical foundation for many fields of modern science and technology, such as scanning electron microscopy (SEM), electron probe microanalysis (EPMA), electron beam lithography (EBL), radiation biology and radiation therapy. Electron stopping power is a basic physical quantity for characterizing the inelastic interaction of electron with materials and is essentially important for better understanding of the basic physical process of electron-material interaction. Meanwhile, the electron stopping powers

 Table 3
 The material parameters for 10 kinds of bioorganic compounds

Comp	oound	$a(a/am^3)$	М	\overline{Z}	
No.	Name	$\rho(g/cm^2)$	M		
11	DNA	1.35	662	5.185	
12	Adenine	1.35	135.14	4.667	
13	Guanine	1.58	151.14	4.875	
14	Thymine	1.48	126.12	4.4	
15	Cytosine	1.3	111.11	4.462	
16	Uracil	1.32	112.09	4.833	
17	Oleic acid	0.89	282.45	2.926	
18	Linoleic acid	0.9	280.44	3	
19	Arachidonic acid	0.92	304.46	3.111	
20	Cellulose	1.35	162.14	4.095	

<i>E</i> (eV)	Compound No.									
	11	12	13	14	15	16	17	18	19	20
20	0.0811	0.1148	0.1911	0.1540	0.1366	0.1250	0.1454	0.1436	0.1421	0.1450
30	0.2742	0.3175	0.3814	0.4082	0.3619	0.3299	0.3922	0.3870	0.3825	0.3857
40	0.7226	0.7340	0.7559	0.9091	0.8055	0.7319	0.8808	0.8690	0.8585	0.8610
50	1.3690	1.4743	1.4680	1.7280	1.5331	1.4037	1.6034	1.5860	1.5730	1.6247
60	1.9928	2.3140	2.2976	2.5742	2.2898	2.1285	2.2466	2.2292	2.2210	2.3900
80	2.8274	3.1933	3.3405	3.5634	3.1784	3.0047	2.9232	2.9090	2.9112	3.2643
100	3.2240	3.5066	3.7858	3.9312	3.5067	3.3411	3.1318	3.1204	3.1282	3.5871
150	3.3684	3.5268	3.9334	4.0002	3.5545	3.4299	3.0344	3.0278	3.0403	3.6591
200	3.1922	3.2876	3.7144	3.7483	3.3225	3.2279	2.7713	2.7671	2.7805	3.4402
300	2.7613	2.7919	3.1917	3.2005	2.8293	2.7688	2.3050	2.3032	2.3157	2.9511
400	2.4139	2.4091	2.7697	2.7697	2.4448	2.4018	1.9679	1.9670	1.9783	2.5609
500	2.1458	2.1203	2.4460	2.4422	2.1537	2.1209	1.7207	1.7204	1.7305	2.2620
600	1.9353	1.8981	2.1945	2.1889	1.9290	1.9028	1.5337	1.5337	1.5430	2.0298
800	1.6371	1.5920	1.8443	1.8392	1.6180	1.5997	1.2843	1.2848	1.2936	1.7069
1000	1.4300	1.3877	1.6083	1.6014	1.4082	1.3931	1.1157	1.1167	1.1250	1.4840
2000	0.9156	0.8879	1.0312	1.0196	0.8968	0.8891	0.7003	0.7017	0.7080	0.9414
3000	0.6905	0.6684	0.7776	0.7669	0.6743	0.6697	0.5226	0.5238	0.5289	0.7081
4000	0.5604	0.5416	0.6307	0.6212	0.5461	0.5429	0.4213	0.4224	0.4267	0.5736
5000	0.4746	0.4581	0.5338	0.5253	0.4617	0.4594	0.3552	0.3561	0.3598	0.4851
6000	0.4133	0.3985	0.4646	0.4569	0.4015	0.3997	0.3082	0.3091	0.3123	0.4220
8000	0.3310	0.3186	0.3717	0.3653	0.3209	0.3197	0.2456	0.2463	0.2490	0.3373
10000	0.2778	0.2670	0.3117	0.3061	0.2689	0.2680	0.2054	0.2060	0.2082	0.2827

Table 4 SPs (in eV/Å) with Born–Ochkur correction included for 10 kinds of bioorganic compounds

are also basic input data in Monte Carlo simulation of energetic electrons transport in materials. In this review article, the developments of two theoretical systems, i.e. the non-relativistic Bethe theory and that based on linear dielectric response theory, for calculating stopping powers have been outlined. The stopping power data tables obtained by our group for a set of polymers and bioorganic compounds have been presented. These data can be used in the studies of various radiation effects for these materials.

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