oscillator strength for energy losses in liquid water at about 20 eV. Collective states excited in the deposition of energy near the peak in the oscillator strength are assumed to decay by localizing this energy on electrons in molecules that may be nanometers away from a particle track. This has the effect of delocalizing the initial spatial pattern of energy deposition events. Another effect of the liquid structure is to change the excitation and ionization thresholds over those observed in a gas. The ionization threshold in liquid water may be as low as 8 eV compared to the isolated water molecule where it is 12.6 eV. These effects on the oscillator strength result in the inverse mean free paths of electrons (macroscopic cross sections), shown in Figure 6.4, being considerably larger in the gas phase than in the liquid phase for energy losses less than about 30 eV. This occurs because the majority of the oscillator strength for the interaction of charged particles is for small energy loss events where the gas-phase cross sections are largest. This difference in oscillator strengths leads to larger distances between interaction products in the liquid as does the delocalization caused by the collective modes of excitation in the liquid. The effect of the delocalization function used by Hamm *et al.* (1985) was shown to yield a peak in the lateral delocalization of about 0.2 nm, with some influence being seen as far out as 5 nm. There was little change in the position of the peak as a function of initial electron energy from 10 eV to 10 keV.

Because of the importance of the proximity of energy deposition events on subsequent chemical reactions and on the potential repairability of damage produced in biological targets, Paretzke *et al.* (1991) investigated the influence of the characteristics of liquid and gas phase targets on the nearest-neighbor distributions of events produced along the tracks of electrons. The results of their calculations are shown in Figure 6.5. The most obvious differences between the liquid and gas data shown in Figure 6.5 are that the gas yields a much higher frequency of very close

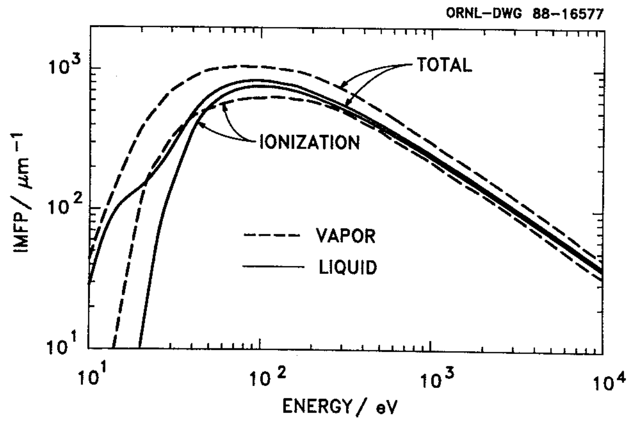

Fig. 6.4. Inverse mean free paths for electrons. ——, in liquid water; --, in water vapor (from Paretzke *et al.*, 1991).

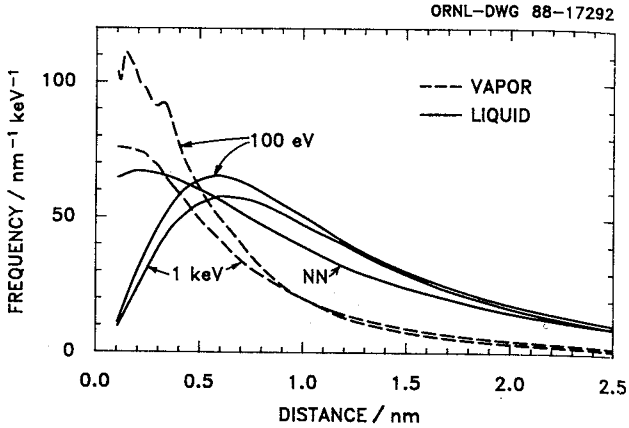

Fig. 6.5. Nearest neighbor distributions for all inelastic events for tracks of electrons. ——, in liquid water; --, water vapor; NN, liquid calculation without delocalization (from Paretzke *et al.*, 1991).

events and the liquid yields more interactions at larger separation distances. The delocalization function effectively eliminates the occurrence of nearest neighbor distances less than a few tenths of a nanometer, whereas the large cross sections for interactions with small energy loss in the gas code yields a maximum in the distributions there. It may be somewhat surprising that there are such large differences in the event frequencies at larger separations; these are expected to occur predominately from hard collisions (large energy loss), where one would not expect the phase of the medium to have much, if any, effect. The differences in the frequency distributions at the smallest nearest-neighbor distances are the most striking; however, these distances may have little significance in a condensed material. It is difficult to understand the meaning of localization of an energy deposition event to distances comparable to, or less than, the dimensions of the constituent molecules of the medium. Certainly it is very likely that the interaction will be with the electronic wave function of a larger collective entity than that of an isolated atom or molecule and the codes, as presently developed, cannot properly simulate this collective feature of the biological medium.

With our present understanding of the underlying physics, it is not clear precisely what is the best way to describe the condensed phase for applications of track structure in radiation biology, although there have, been several approaches (Hamm *et al.*, 1985; Zaider, 1991; Kaplan *et al.*, 1986), all with relatively untested results. One should recognize that even the existence of plasmons in pure water is still a matter of uncertainty and controversy (LaVerne and Mozumber, 1993); the physics is suggestive, but is much less so than is the case for plasmons in metals, where they have been observed (Inokuti, 1991). It should also be noted that even the "liquid" Monte Carlo transport codes have focussed on pure water as the medium of transport, which is far from the complex heterogeneous mixture of interest in the study of radiation biology. What we must not lose sight of, however, is that both the gas and liquid based track simulation codes can be powerful tools in the study of the *relative* biological effectiveness (RBE) of different radiations and in the search for mechanisms of radiation damage at the cellular and molecular level. The RBE is the ratio of the dose of a particular kind of radiation to the dose of a standard kind of radiation, when the two doses produce the same amount of a specified biological effect. Customarily one takes high-energy x rays as the standard kind of radiation.

One may also use the Monte Carlo calculations of charged particle tracks to obtain many of the more traditional quantities of radiological physics. For example, calculations of stopping power and W-values are commonly performed as consistency tests of the performance of such codes. For such calculations, however, the accuracy of the derived quantity is often much poorer than when the quantity is obtained by other methods because the Monte Carlo calculations are limited by the inherent accuracy of the collision cross sections used as input data; the related cross section measurements usually contain uncertainties of 20% or more. In addition, the Monte Carlo technique is computationally intensive; hence it is best used only when other computational techniques are unavailable and the stochastic nature of the calculation is important.

6.3 Microdosimetric Distributions

This subject is treated in ICRU (1983) and by Rossi and Zaider (1995). In this section, we briefly show how the spectra of secondary electrons are related to radiological parameters.

As was discussed earlier, proportional-counter techniques are the primary *experimental* methods of determining the stochastic distributions of energy deposited in small tissue-like volumes for dosimetric purposes. Proportional-counter techniques, however, have been limited to simulated volumes with dimensions greater than a few hundred nanometers because of the characteristics of gas multiplication. Because the stochastics of energy deposition in small volumes result from energy transport by secondary electrons, the Monte Carlo technique is ideally suited for the investigation of such distributions in sites smaller than can be investigated experimentally. In practice, calculations are first conducted for sufficiently large volumes to be tested by experiment (Wilson and Paretzke, 1980), then tested codes are used to explore the distributions in smaller sites. Wilson *et al.* (1988) published a detailed description of the systematics of energy deposition by protons in small volumes of simulated tissue; tissue, in this case, is simulated using water vapor cross sections. These calculations investigated energy distributions for protons passing through the simulated site as a function of site diameter, position of the passage of proton through the spherical volume, and proton energy. They investigated proton energies from 0.3 to 20 MeV. More recently, Wilson (1994) has presented similar calculations for protons that pass near, but not through, the site and deposit energy within the volume via secondary electrons. The significance of energy transport by secondary electrons *inside* the volume of interest from initial interactions of the radiation *outside* of that volume can be seen in the proportional counter data by Gross, *et al.* (1970) and by Glass and Roesch (1972). Such processes are much more pronounced in fast, heavy-ion collisions, where fast secondary electrons can transport energy over relatively large distances (see, for example, Kliauga and Rossi, 1976; Metting *et al.*, 1988; Toburen *et al.*, 1990a; Braby *et al.*, 1992).

6.4 Track Entities Derived from Electron Spectra

6.4.1 Track Entities in Radiation Chemistry

As discussed above, radiation chemists, in their effort to calculate chemical yields, were among the first to recognize the need for some means of accounting for the nonhomogeneous nature of energy deposition by a radiation field. The "string-of-beads" model of a track, developed by Samuel and Magee (1953), simply divided the track into spherical "spurs"; each spur was of a radius of 1 to 1.5 nm and contained an energy deposition of 40 eV each. The spurs were equally spaced along the particle's path at distances required to give the proper LET (stopping power) values. This model was used for the discussion of diffusion-controlled reactions in water and aqueous solutions. Ganguly and Magee (1956) extended this model by placing the spurs randomly along the track and accounting for variations in the LET as the particle slowed. The next extension to this model was an allowance for variations in the sizes of the spurs. Mozumder and Magee (1966a) argued that the distribution of spur sizes must be as important to the subsequent chemical reactions as the ratio of the energy deposited as spurs and δ-rays and defined additional entities based on the amount of energy deposited. They redefined the spur to contain energy originating from primary events up to 100 eV. Secondary electrons with sufficient energy to produce their own tracks were defined as δ-rays; these result from near head-on collisions of the primary electron with a target electron. They also noted that it is possible to produce a distribution of δ-rays with different energies; those with ranges less than the average spur separation distance were called "short tracks". They then reasoned that there was a range of electrons above 100 eV, but of insufficient energy for the second generation electron to leave its site of birth. This electron, and all others produced by it, were said to form an entity called a blob. These track features, the spur, the blob, and the short track were discussed earlier and illustrated in Figure 6.1. The distribution of these entities along a charged particle track as prescribed by Mozumber and Magee was based on the frequency of event sizes obtained from classical collision theory. This technique for describing the energy deposition was quite useful in calculations of chemical yields along the path of high-LET particles. A shortcoming of this technique, however, was that all of the interactions and their corresponding track entities were placed on the axis of the particle and there was no means to account for the less dense track of particles that have the same LET, but higher velocities.

The application of secondary electron spectra to the study of chemical yields induced by the passage of fast heavy charged particles was first explored by Miller and co-workers (Miller and West, 1977; 1981; Miller, 1981; Miller and Wilson, 1989a). They extended the basic method of Magee and his colleagues by using the Monte Carlo technique of computational track structure to derive the 3-dimensional spectrum of spurs along the track. This technique incorporated the effects of electron transport as a function of the charged particle velocity and provided chemical yields that were in agreement with experiment for particles of the same LET, but different velocities (Miller and Wilson, 1989a).

6.4.2 Track Entities in Radiation Biology

The use of electron spectra within Monte Carlo models of the spatial distributions of energy deposition by ionizing radiation has played an important role in the investigation of mechanisms for the production of biological damage and in the understanding of the RBE of different types of radiation. A major shortcoming in modeling radiation-induced biological damage is a lack of knowledge of the actual reaction pathways leading from energy deposition to fundamental endpoints such as DNA strand breaks, chromosomal aberrations, etc. In addition, little is known of the relationship of these endpoints to observed radiation-induced cell mutation and/or death. Although one can calculate energy deposition distributions in arbitrarily small volumes simulated as water vapor, or liquid water, one cannot readily convert these energy distributions into known chemical or biological products, such as DNA strand breaks and cellular mutation spectra. This is not to suggest that there have not been considerable advances in understanding in these areas (see for example the review by Magee and Chatterjee, 1986). However, for the most part the tests of the assumptions in the models have been made using track models that provide only descriptions of the average parameters of energy deposition, and information on the stochastic distributions of initial products is lacking. By comparing the stochastic track structure information with the measured chemical and biological products, one can begin to understand the nature of the critical volumes and energies that lead to biologically detectable damage.

The difference between the spatial and temporal distributions of energy deposited by low- and high-LET radiation in DNA was initially studied by Charlton and co-workers (1985) using the Monte Carlo track simulation techniques developed by Wilson and Paretzke (1981). By superimposing computer-generated tracks and randomly oriented cylinders with lengths and diameters chosen to approximate the elements of a DNA fiber, they compared energy distributions in chromatin by different radiations. Using this method, they showed that the energy deposition in simulated nucleosomes was several orders of magnitude greater for alpha particles than for low-LET radiation. A much more sophisticated model of DNA has been used to investigate the energy required to produce a break in one or both of the strands of DNA (Charlton and Humm, 1988; Miller and Wilson, 1989b). By comparing the energy deposited in the strands of DNA with the measured spectrum of strand breaks induced by the decay of incorporated ^{125}I, they obtained 17.5 eV as the energy deposition related to the production of single-strand breaks. Goodhead and Brenner (1982) also investigated the combinations of energy and deposited volume that are correlated with the RBE observed in cells irradiated with soft and 250 kV x rays. In their studies, they found a correlation between the deposition of about 100 eV in spherical volumes of approximately 3-nanometer diameter and the RBE for x rays of different energies. Although comparisons such as these, leading to information on the energetics and volumes of importance for strand breaks and cell killing, can be criticized for many assumptions in the models, they still aid radiation biologists in developing a better understanding of the mechanisms of radiation action.

6.5 Stopping Power and LET

The definition of LET and the basic ideas related to it are given in ICRU (1970). Many of the non-stochastic quantities relating to energy loss by charged particles in matter, or at least basic information relating to these quantities, can be obtained from a knowledge of the spectrum of electrons emitted as the particles interact with the atomic and molecular constituents of the medium. As discussed above, the cross sections for energy loss, $\sigma(\epsilon)$, to secondary electrons can be obtained from the DDCSs for ionization, $\sigma(\epsilon,\theta)$, by integration with respect to the angle of emission, Eq. 1.5. Likewise, the TICS, σ_i, can be obtained by integration over both the ejected electron energy and emission angle as illustrated in Eq. 1.7. In a similar fashion, one can obtain the mean and median energies of the ejected electrons from the DDCSs for electron emission; see Eqs. 6.1 and 6.2, respectively. The mean and median electron energies will vary depending on the projectile energy and species. For example, a 1.5 MeV proton in water vapor will eject electrons with a mean energy of 47 eV as calculated by Eq. 6.1, whereas the median energy obtained using Eq. 6.2 is 220 eV. The same quantities, when calculated for 0.5 MeV protons, yield a mean energy of ejected electrons that is approximately the same as in the case of the 1.5 MeV proton, about 55 eV, but the median energy, 136 eV, is smaller by nearly a factor of 2.

The spectrum of electrons produced by ionization can provide detailed information on the stopping power of heavy charged particles. Using the theoretical definition of the linear stopping power, S, (ICRU, 1993), Wilson (1972) wrote it as the sum of three contributions

$$S = n\left[\sum_{u} \sigma_u E_u + \sum_{w} \sigma_w E_w + \int_0^{\epsilon_{max}} \sigma(\epsilon)(B + \epsilon)\, d\epsilon\right], \tag{6.3}$$

where n is the number of target atoms or molecules per unit volume, $\sigma(\epsilon)$ is the ionization cross section per unit energy range for ejection of an electron of energy ϵ, σ_u is the cross section for excitation of state u of the target with energy E_u, and σ_w is the cross section for producing the residual ion in an excited state w with energy E_w. The first term in the sum is the contribution from target excitation, the second term contains energy loss that is converted to excitation energy of the residual ion, and the last term includes both the energy going into binding energy B to eject the electron and that contributing to the kinetic energy ϵ of secondary electrons. At lower incident ion energies, it would also be necessary to include energy that is lost by the proton in charge-changing collisions and at sufficiently low energy, one would also need to include energy lost in nuclear collisions. Wilson calculated each of the contributions to the stopping power described by Eq. 6.3 and plotted the fraction of total stopping power they represented for the case of protons passing through hydrogen gas. This plot is reproduced in Figure 6.6. From this data we see that the fraction of the energy loss going into free electron kinetic energy, F_ϵ, is approximately 60% for proton energies above about 100 keV. If the fraction of energy used to overcome the binding energy, F_B, is added to F_ϵ, this fraction, representing the total energy going into ionization, is nearly 80% of the stopping power in the proton energy region above 100 keV. Below about 100 keV charge transfer and

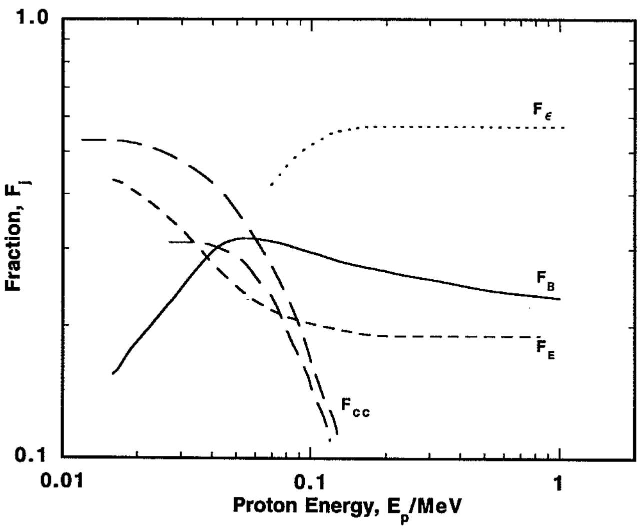

Fig. 6.6. Fraction of the total stopping power for protons in hydrogen attributed to binding energy for ionization (F_B), to kinetic energy of ionized electrons (F_ϵ), to excitation (F_E) and to charge transfer (F_{CC}) (after Wilson, 1972). The upper F_{cc} line is based on experimental cross sections while the lower one is calculated.

nuclear collisions become increasingly important contributions to the stopping power. This example illustrates the utility of secondary electron spectra in understanding the mechanisms of energy loss by energetic particles.

The significance of the linear energy transfer, often abbreviated as LET, as an index of the energy locally deposited in a specified volume has been extensively discussed in ICRU Report 16 (1970). The LET is the stopping power minus the energy carried away by secondary electrons of high energies to large distances; the meaning of "high energies" or "large distances" needs to be specified for the purpose of a particular consideration. For instance, Bartels and Harder (1990) consider the energy deposition in a nanometer region around a particle track, state that it is nearly independent of the energy and mass of the primary particle, and interpret certain radiobiological effects in terms of the LET.

So far, the evaluation of the LET has been largely based on the Rutherford cross section, Eq. (2.3). Data given in the present report will enable one to evaluate the LET or other track-structure quantities more accurately, and will help make related considerations more convincing.

6.6 Energy Per Ion Pair

A principal quantity of radiation dosimetry is the average energy necessary to produce an ion pair in a gas (ICRU, 1979). This quantity, denoted by W, is a function of the gas and the type and energy of the particle producing the ion pairs. This is another area where the spectra of electrons ejected in ionizing collisions are required in a theoretical treatment.

Following the development of Bichsel and Inokuti (1976), one can define the differential W-value (ICRU, 1979) for protons as

$$w(E) = \frac{S(E)}{j(E)}, \tag{6.4}$$

where $S(E)$ is the stopping power of protons at energy E and $j(E)$ is given, for unit molecular density, by

$$j(E) = j_1(E) + j_2(E), \tag{6.5}$$

with

$$j_1(E) = \sigma_i(E) \tag{6.6}$$

and

$$j_2(E) = \int \frac{\epsilon \sigma_i(\epsilon, E)\, d\epsilon}{W(\epsilon)}. \tag{6.7}$$

In these expressions, $\sigma_i(E)$ is the TICS by protons of energy E (and can be obtained from the measured DDCSs by Eq. 1.5), $\sigma_i(\epsilon, E)$ is the SDCS for ionization by protons of energy E (obtained from the measured DDCSs by Eq. 1.3), and $W(\epsilon)$ is the mean energy for the production of an ion pair by an electron of energy ϵ. In this calculation, j_1 is the number of ion pairs produced directly by the proton and j_2 is the number of ion pairs produced by the secondary electrons. If the quantity $w(E)$ can be established over the entire range of E, one can calculate the integral W-value from

$$\frac{E}{W(E)} = \int_B^E \frac{S(E')}{w(E')}\, dE'. \tag{6.8}$$

Using the data of Toburen (1971) for ionization of nitrogen by protons, Bichsel and Inokuti (1976) obtained a W-value of 32.4 eV using this technique. This is in rough agreement with the known W-value of 35 eV for nitrogen. Further development is given in Inokuti, *et al.* (1992).

6.7 Moments of Energy Loss Distributions

The secondary electron spectra provide a means of calculating the moments associated with energy loss by charged particles. From the integral of $\sigma(\epsilon)$ (given by Eq. 1.7) one can obtain the zeroth moment, M_0,

$$M_0 = \int_0^{\epsilon_{max}} \sigma(\epsilon)\, d\epsilon. \tag{6.9}$$

This is equivalent to the TICS given by Eq. 1.5. The first moment of the electron spectra, M_1, is related to the energy transfer to secondary electrons by

$$M_1 = \int_0^{\epsilon_{max}} \epsilon \sigma(\epsilon)\, d\epsilon. \tag{6.10}$$

This is the partial stopping cross section for providing kinetic energy to secondary electrons. Similarly the second moment, M_2, is related to the energy straggling distribution (see Section 6 of ICRU, 1993) and is given by

$$M_2 = \int_0^{\epsilon_{max}} \epsilon^2 \sigma(\epsilon)\, d\epsilon. \tag{6.11}$$

Further information on straggling is given by Bichsel (1988).

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