the dielectric constant may rise with increasing temperature because of the effect of temperature on the relaxation time. There is a frequency region in which the two effects tend to cancel, making the dielectric constant insensitive to changes in temperature. This unique property of polar liquids suggests uses in matched terminations or in calorimetric measurement of power where dielectrics are desired whose properties do not change with temperature.

As for the frequency limitation on nitrobenzene as a Kerr medium, the behavior of the dielectric constant indicates that dipole polarization above 1000 mcps falls away rapidly with increasing frequency.

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Theory of Radiation Chemistry. III. Radical Reaction Mechanism in the Tracks of Ionizing Radiations*

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This paper develops a model for the dissipation of the tracks of high-energy particles. It is assumed that all chemical effects are due to one kind of radical and that there are no effects of overlapping of neighboring tracks, i.e., that there is a sufficiently high concentration of scavenger to prevent such overlapping. The model of Samuel and Magee is used and extended to take into account the interaction of randomly distributed spurs along the track. Calculated values of the extent of scavenger reactions and radical reactions are presented in graphical form. General trends of experimental results to be expected according to the model are indicated. Quantitative correlation with experiments was not attempted because of the uncertainty in the values of various parameters used and because of the serious limitations of the one-radical model of tracks.

INTRODUCTION

Recent papers[1–5] have developed the theory of diffusion and reaction of radicals in the tracks of ionizing particles in liquids. These treatments generally follow the lines set forth by Jaffe[6] in his pioneering study of the ionization density in columnar tracks. Lea[7] pointed out that high-energy electrons do not form columns, but more or less isolated "spurs,"† and this fact has been recognized in most subsequent work.

In this paper the authors have studied a model for a particle track which consists of a series of spurs randomly spaced along the linear trajectory of the particle. A Gaussian distribution for the position of the radicals in a spur is assumed, and all spurs are assumed to have the same number of radicals. The treatment used here only applies to the "low background" case, i.e., the case in which sufficient "scavenger" is present to react with the radicals of a track before they intermingle with radicals from adjacent tracks. The intermingling of radicals from adjacent spurs of the same track is, however, taken into account and the concentration of the scavenger is also varied.

A chemical reaction mechanism for the one-radical track can be given as

$$R+R\rightarrow R_2, \quad \text{(a)}$$

$$R+S\rightarrow RS, \quad \text{(b)}$$

where R and S stand for radical and scavenger, respectively.

The emphasis in this paper is upon a study of the method for treatment of the competition of reactions (a) and (b) in a track which is expanding by diffusion. Although this is a one-radical model, it may be possible that the radiation chemistry of water can be understood essentially on the basis of this model.[2–5] We have used only "average" spurs of six radicals each and a study has not been made of the effect of actual spur-size distribution. Recently Gray[8] has made a study along different lines in which there was a significant effect attributed to the highly ionizing delta rays.

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TRACK STRUCTURE

The track is taken as a linear arrangement of spurs. Radicals in each spur are in spherically symmetrical Gaussian distribution round the center of the spur. It is assumed further that the Gaussian distribution is maintained throughout the lifetime of a spur. This is the use of "prescribed diffusion."[3] All spurs have the same initial number of radicals, formed by the absorption of the same amount of primary energy (ϵ). In every region of the track the spurs occur randomly and their average spacings along the finite length of the track decreases exponentially with the residual range [see Eq. (3) below]. From the experimental range-energy relations of Geiger[9] with α-particles, Katz and Penfold[10] with β particles, and Wilson's[11] approximate range-energy relation for protons derived from Bethe's stopping power formula, a general expression for range and energy for an ionizing particle can be written in the form,

$$1-\frac{\rho}{R}=\left(\frac{E_\rho}{E_0}\right)^\eta, \tag{1}$$

where R is the mean range of the particle of energy E_0 and $R-\rho$ is the residual range of the same when the energy is reduced to E_ρ, η is a not-too-sensitive function of energy E_0 and can be evaluated from experimental values of range. Energy density along the track is then given by

$$\frac{dE_\rho}{d\rho}=\frac{E_0}{\eta R}\cdot\frac{1}{[1-(\rho/R)]^{(\eta-1)/\eta}}, \tag{2}$$

and the average spacing Z_1 of spurs at any point ρ on the track is given by

$$Z_1=\epsilon\frac{d\rho}{dE_\rho}=\frac{\epsilon\eta R}{E_0}\left(1-\frac{\rho}{R}\right)^{(\eta-1)/\eta} \tag{3}$$

Taking the axis of the track along the z direction, the concentration of radicals $c(r,z,\tau)$ at a distance r from the point z on the axis at any time τ equal or larger than a certain initial time t_0 is given by

$$c(r,z,\tau)=\frac{\exp(-r^2/4D\tau)}{(4\pi D\tau)^{\frac{3}{2}}}\sum_{i=1}^{n}\nu_i\exp\left(-\frac{(z-z_i)^2}{4D\tau}\right), \tag{4}$$

where ν_i is the total number of radicals that originated in the ith spur surviving up to the time τ, z_i is the coordinate of the center of the spur from the same arbitrary origin, n is the total number of spurs in the track [equal to (E_0/ϵ)], and D is the diffusion coefficient of the radicals. At the initial time $t=t_0$ the radical concentration is

$$c(r,z,t_0)=\frac{\exp(-r^2/r_0^2)}{\pi^{\frac{3}{2}}r_0^2}\sum_{i=1}^{n}\nu_i\exp\left(-\frac{(z-z_i)^2}{r_0^2}\right), \tag{4a}$$

which is the assumed initial distribution. The relation between t_0 and r_0 is thus given by $r_0^2=4Dt_0$.

A natural consequence of the use of the expression (4) is that the spurs overlap at all times, even initially.

CALCULATION OF SCAVENGER REACTION

The track equation is:

$$\partial c/\partial t=D\nabla^2c-kc^2-k_sc_sc.$$

The scavenger concentration, c_s is assumed to remain uniform and constant for all times, and k and k_s are taken as the reaction rate constants for the reactions a and b, respectively. Integration over all space yields

$$\int\frac{\partial c}{\partial t}dv=-k\int c^2dv-k_sc_s\int cdv$$

since

$$\int D\nabla^2cdv=0$$

or

$$-\frac{dW_t}{dt}=k\int c^2dv+k_sc_sW_t, \tag{5}$$

where

$$W_t=\int cdv=\sum_{i=1}^{n}\nu_i. \tag{5a}$$

A convenient expression,

$$\int c^2dv=\frac{1}{(8\pi Dt)^{\frac{3}{2}}}\times\left[\sum_{i=1}^{n}\nu_i^2+2\sum_{\substack{i,j\\i<j}}^{n}\nu_i\nu_j\exp\left(-\frac{(z_i-z_j)^2}{8Dt}\right)\right], \tag{6}$$

is derived in Appendix I, where $t=t_0+\tau$. We define a volume V_t as

$$V_t=\frac{\left[\int cdv\right]^2}{\int c^2dv}=\frac{W_t^2}{\int c^2dv}. \tag{6a}$$

The physical significance of V_t will be apparent in the sequel. Substitution of (6a) into (6) gives

$$\frac{W_t^2}{V_t}=\frac{1}{(8\pi Dt)^{\frac{3}{2}}}\times\left[\sum_{i=1}^{n}\nu_i^2+2\sum_{\substack{i,j\\i<j}}^{n}\nu_i\nu_j\exp\left(-\frac{(z_i-z_j)^2}{8Dt}\right)\right]. \tag{7}$$

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[11] R. R. Wilson, Phys. Rev. 71, 385 (1947).

These equations lead to the following expression for V_t:

$$\frac{1}{V_t}=\frac{1}{n(8\pi Dt)^{\frac{3}{2}}}\left[1+\frac{n(8\pi Dt)^{\frac{1}{2}}}{(2-\eta)\eta R}\right]. \tag{8}$$

The derivation of expression (8) is given in Appendix II.

A measure of the volume of a spur is given by $(8\pi Dt)^{\frac{3}{2}}$; it is apparent from (7) and (8) that for the case in which the spurs do not overlap appreciably, i.e., when $\exp(-(z_i-z_j)^2/8Dt)\sim 0$, we have $V_t\approx(8\pi Dt)^{\frac{3}{2}}\cdot n$. That is to say that for small enough time V_t increases in n isolated expanding spheres, and for large time V_t expands as a cylinder [see Eq. (8)]. The reciprocal of the factor within the square bracket in Eq. (8) may be taken as a measure of the extent of spur overlapping in a track at any time. Equation (7) indicates that at very large time when $\exp(-(z_i-z_j)^2/8Dt)\sim 1$, V_t expands as a sphere. Equation (8) is not valid for such a large expansion and for most practical cases with moderate concentration of scavenger the radicals in the track are found to be well exhausted long before they outlive the elliptical stage of expansion. Exceptions can be found in tracks of low-energy primary particles and also for very low scavenger concentration.

Equation (5) can now be written in the form:

$$-\frac{dW_t}{dt}=\frac{kW_t^2}{V_t}+k_s\cdot c_s\cdot W_t.$$

The solution of the equation can be written in the form:

$$\frac{1}{W_t}=\exp\left(\int_{t_0}^{t}k_sc_sdt\right)\cdot\left[\int_{t_0}^{t}\frac{k\exp\left(-\int_{t_0}^{t}k_sc_sdt\right)}{V_{t'}}dt'+\frac{1}{W_0}\right], \tag{9}$$

where W_0 is the total number of radicals in the track at time t_0. We make the substitution $x=t/t_0$ and $q=k_sc_st_0$. The fraction of the radicals surviving any time x is then given by

$$\frac{W_x}{W_0}=\frac{1}{\exp[q(x-1)]\cdot\left[1+kW_0t_0\int_1^x\frac{\exp[-q(x'-1)]}{V_{x'}}dx'\right]}, \tag{10}$$

where

$$\int_1^x\frac{\exp[-q(x'-1)]}{V_{x'}}dx'=\frac{2}{n(8\pi Dt_0)^{\frac{3}{2}}}-\frac{2\exp[-q(x-1)]}{n(x)^{\frac{1}{2}}(8\pi Dt_0)^{\frac{3}{2}}}$$
$$-\frac{4\cdot q^{\frac{1}{2}}\cdot\exp(q)}{n\cdot(8\pi Dt_0)^{\frac{3}{2}}}\int_{q^{\frac{1}{2}}}^{(qx)^{\frac{1}{2}}}\exp(-y^2)dy+\frac{\exp(q)\cdot\int_q^{qx}\exp(-y)\cdot dy/y}{(2-\eta)\eta R\cdot 8\pi Dt_0}. \tag{10a}$$

The fraction S of the radicals reacting with scavenger is given by

$$S=\frac{k_sc_st_0}{W_0}\int_1^\infty W_xdx$$

or

$$\frac{dS}{dx}=q\cdot\frac{1}{\exp[q(x-1)]\left[1+kW_0t_0\int_1^x\frac{\exp[-q(x'-1)]}{V_{x'}}dx'\right]}. \tag{11}$$

One can then calculate S by numerical integration.

NUMERICAL CALCULATIONS IN AQUEOUS SYSTEMS

The following values for the parameters occurring in Eq. (11) were adopted for calculation from the estimates of previous authors[2,3,5]:

$$D=2\times10^{-5}\text{ cm}^2/\text{sec},\qquad t_0=1.25\times10^{-10}\text{ sec},\qquad k=10^{-11}.$$

On the average, 6 radicals are assumed to be present in each spur at the initial time t_0 formed by the absorption of 100 ev of primary energy. Substitution of these values in Eq. (11) and with the use of Eq. (10a) the general

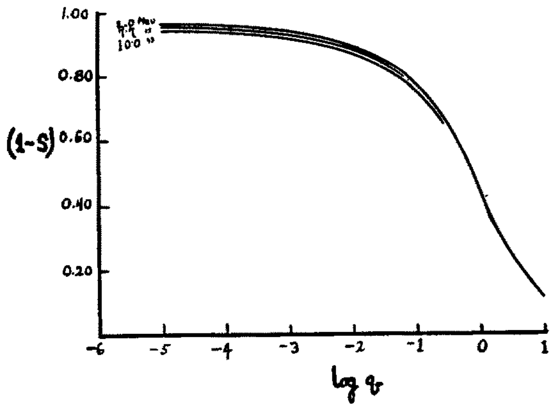

FIG. 1. Radical recombination $(1-S)$ in α-particle tracks of different energy as function of scavenger concentration (q). Graph for the 5.3-Mev track is not shown in the figure.

TABLE I. Range R and η values for particles of different energies in aqueous system.

Particle	Energy in Mev	R in μ	η
α particle	10.00	108.4	1.66
α particle	7.68	67.2[a]	1.60
α particle	5.30	38.1[a]	1.50
α particle	2.00	10.1	1.12
Protons	10.00	1211	1.77
Protons	5.00	355	1.73
Protons	1.00	23	1.57
Protons	0.50	8.81[b]	1.45
Protons	0.10	1.105[b]	1.14
Electrons	0.50	1748[c]	1.40
Electrons	0.10	141.2	1.70
Electrons	0.05	42.7	1.75
Electrons	0.01	2.52	1.75

[a] Taken from Carvalho and Yodoga, reference 13.
[b] Calculated from Wenzel and Whaling's data (reference 14).
[c] Extrapolated from Lea's Table (reference 12). The rest of the range values are taken from Lea.

form of the equation in aqueous systems is

$$\frac{dS}{dx}=q\frac{1}{\exp[q(x-1)]\left[1.9525-\dfrac{0.9525\exp[-q(x-1)]}{x^{\frac{1}{2}}}-1.9048q^{\frac{1}{2}}\exp(q)\cdot\displaystyle\int_{q^{\frac{1}{2}}}^{(qx)^{\frac{1}{2}}}\exp(-y^2)dy+\dfrac{1.1937\cdot10^{-7}\cdot n\exp(q)}{(2-\eta)\eta R}\displaystyle\int_{q}^{qx}\frac{\exp(-y)}{y}dy\right]}. \tag{12}$$

The results obtained with α particle, proton, and electron tracks are discussed in the following. Values of R are taken from Lea,[12] except in the cases cited, and η values are calculated therefrom.

For the range of α particles of energy 5.3 Mev and 7.68 Mev, the more accurate experimental data of Carvalho and Yogoda[13] were taken and η calculated from their values of molecular stopping power.

The range of protons of energy below 1 Mev were computed from the results of stopping power for protons obtained with D_2O ice by Wenzel and Whaling[14] and also η was calculated therefrom. Table I gives a collection of R and η values for particles of different energies passing through aqueous systems.

An arbitrary criterion for the extent of time x up to which the condition for cylindrical expansion of the track [Eq. (8)] is approximately fulfilled, may be expressed as

$$\frac{(8\pi Dt_0)^{\frac{1}{2}}\cdot x^{\frac{1}{2}}}{R}\leqslant 1,$$

or,

$$x\leqslant 1.6\times10^{13}\cdot R^2. \tag{13}$$

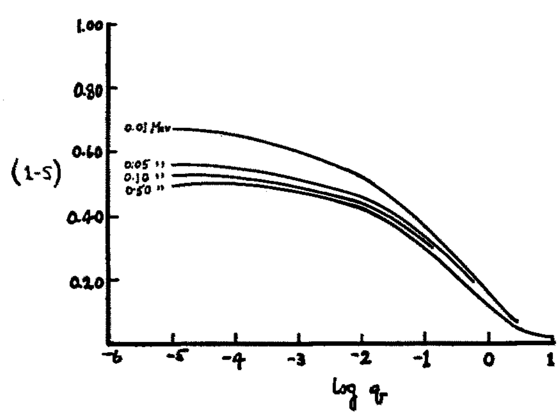

FIG. 2. Radical recombination $(1-S)$ in β-particle tracks of different energy as function of scavenger concentration (q).

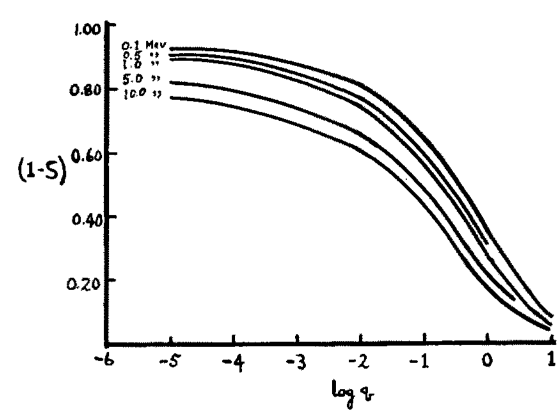

FIG. 3. Radical recombination $(1-S)$ in proton tracks of different energy as function of scavenger concentration (q).

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[13] H. G. Carvalho and H. Yogoda, Phys. Rev. 88, 273 (1952).
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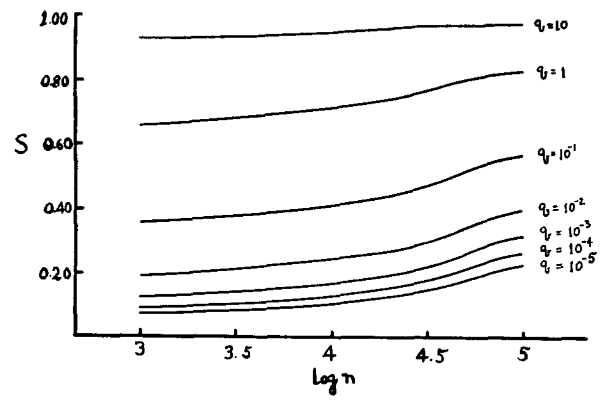

FIG. 4. Scavenger reaction (S) in proton tracks as function of the total number of spurs (n), for different values of scavenger concentration (q).

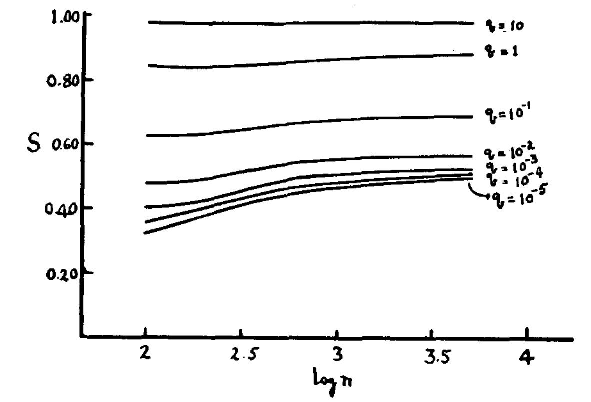

FIG. 5. Scavenger reaction (S) in electron tracks as function of the total number of spurs (n) for different values of scavenger concentration (q).

The lowest range for the particles chosen here is of the order of 1μ, which offers the poorest case for the applicability of Eq. (8). From the relation (13) it is seen that Eq. (8) will continue to hold in such tracks for time x up to 1.6×10^5. Calculations show that for the case $q=10^{-5}$ and $R\sim1\mu$, 80% of the reaction is over by that time, and for values of $q>10^{-5}$ the condition (13) is much better fulfilled up to the time when the reactions are reduced to insignificance.

RESULTS AND DISCUSSION

In Figs. 1–3 calculated values of $1-S$ (which give the extents of radical recombination) are plotted against $\log q$ for different energies of the primary particles, as indications of the general nature of the dependence of S on q (see references 4 and 5). In all cases it is apparent that at low values of q the net radical recombination attains limiting values. The value of S for $q\approx0$ can be taken as a measure of the radicals which escape recombination.[2] It should be noted that according to Eq. (11), at zero concentration of scavenger ($q=0$), $S=0$, i.e., given infinite time, none of the radicals would escape recombination. The limiting values of S, as obtained in the figures at very low values of q, give a measure of radicals which in effect escape recombination in tracks and these can be compared with experimental "radical yields." In dense α-particle tracks, for the energy range considered, most of the reaction is radical recombination ($\sim$95%) at low scavenger concentrations. For electron tracks, radical recombination is much lower, and proton tracks are intermediate as one would expect. It can also be noted from the curves that, in the range considered, scavenger effects increase with increase in particle energy.

Figures 4 and 5 show the dependence of scavenger reaction on primary particle energy for selected scavenger concentrations. These curves are all monotonic. Magee in a recent paper[3] developed a "two-stage" diffusion model for tracks and found that such curves were predicted to have minima. The present treatment, however, has an assumption which limits its validity to relatively high energies. The development of Appendix II is valid only for a large number of spurs.

The results of this treatment are in good qualitative agreement with previous diffusion model treatments. This detailed consideration of the track structure, of course, brings in more parameters than simpler treatments. Since an effect is calculated for the intermingling of the radicals from neighboring spurs even for lightly ionizing particles such as an electron having an energy of approximately 1 Mev, a consideration of the distribution of spur sizes is expected to be necessary for further refinement of the model.

ACKNOWLEDGMENTS

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APPENDIX I. DERIVATION OF A CONVENIENT EXPRESSION FOR THE INTEGRAL $\int c^2dv$

With use of Eq. (4) we can write

$$\int c^2dv=\frac{1}{(4\pi Dt)^{\frac{1}{2}}}\int_0^\infty\frac{\exp(-2r^2/4Dt)}{4\pi Dt}\cdot2\pi r dr\int_{-\infty}^{\infty}\left[\sum_{i=1}^{n}\nu_i\exp\left(-\frac{(z-z_i)^2}{4Dt}\right)\right]^2\frac{dz}{(4\pi Dt)^{\frac{1}{2}}}$$

$$=\frac{1}{2(4\pi Dt)^{\frac{1}{2}}}\left[\int_{-\infty}^{\infty}\sum_{i=1}^{n}\nu_i^2\exp\left(-\frac{2(z-z_i)^2}{4Dt}\right)\frac{dz}{(4\pi Dt)^{\frac{1}{2}}}+2\int_{-\infty}^{\infty}\sum_{i,j}^{n}\nu_i\nu_j\exp\left(-\frac{(z-z_i)^2+(z-z_j)^2}{4Dt}\right)\frac{dz}{(4\pi Dt)^{\frac{1}{2}}}\right]$$

$$=\frac{1}{2(4\pi Dt)^{\frac{3}{2}}}\left[\frac{1}{\sqrt{2}}\sum_{i=1}^{n}\nu_i^2+2\sum_{\substack{i,j\\i<j}}^{n}\nu_i\nu_j\exp\left(-\frac{(z_i-z_j)^2}{8Dt}\right)\int_{-\infty}^{\infty}\exp\left(-\frac{[\sqrt{2}z-(z_i+z_j/\sqrt{2})]^2}{4Dt}\right)\frac{dz}{4\pi Dt}\right]$$

$$=\frac{1}{(8\pi Dt)^{\frac{3}{2}}}\left[\sum_{i=1}^{n}\nu_i^2+2\sum_{\substack{i,j\\i<j}}^{n}\nu_i\nu_j\exp\left(-\frac{(z_i-z_j)^2}{8Dt}\right)\right]$$

or,

$$=\frac{1}{(8\pi Dt)^{\frac{3}{2}}}\left[\sum_{i=1}^{n}\nu_i^2+\sum_{\substack{i,j\\i\neq j}}^{n}\nu_i\nu_j\exp\left(-\frac{(z_i-z_j)^2}{8Dt}\right)\right]. \tag{I}$$

APPENDIX II. DERIVATION OF AN APPROXIMATE EXPRESSION FOR THE TRACK VOLUME (V_t)

By substitution of the expression (I) in Appendix I for the integral $\int c^2dv$, one gets from Eq. (6a):

$$\frac{W_t^2}{V_t}=\frac{1}{(8\pi Dt)^{\frac{3}{2}}}\left[\sum_{i=1}^{n}\nu_i^2+\sum_{\substack{i,j\\i\neq j}}^{n}\nu_i\nu_j\exp\left(-\frac{z_{ij}^2}{8Dt}\right)\right],$$

where $z_{ij}^2=(z_i-z_j)^2$. With the use of (5a) this can be written

$$\frac{(\sum_i\nu_i)^2}{V_t}=\frac{1}{(8\pi Dt)^{\frac{3}{2}}}\left[\sum_{i=1}^{n}\nu_i^2+\sum_{\substack{i,j\\i\neq j}}^{n}\nu_i\nu_j\exp\left(-\frac{z_{ij}^2}{8Dt}\right)\right].$$

If all the ν's are taken as equal, this becomes

$$\frac{n^2\nu^2}{V_t}=\frac{1}{(8\pi Dt)^{\frac{3}{2}}}\left[n\nu^2+\nu^2\sum_{\substack{i,j\\i\neq j}}^{n}\exp\left(-\frac{z_{ij}^2}{8Dt}\right)\right]. \tag{IIa}$$

We now evaluate the expression

$$Q=\sum_{\substack{i,j\\i\neq j}}^{n}\exp\left(-\frac{z_{ij}^2}{8Dt}\right).$$

First expressing the double summation as a series of single summations,

$$Q=2\left[\sum_{i=1}^{n-1}\exp\left(-\frac{z_{i,i+1}^2}{8Dt}\right)+\sum_{i=1}^{n-2}\exp\left(-\frac{z_{i,i+2}^2}{8Dt}\right)+\sum_{i=1}^{n-3}\exp\left(-\frac{z_{i,i+3}^2}{8Dt}\right)+\cdots\right]. \tag{IIb}$$

For a random distribution of spurs the probability function for adjacent neighbors is given by $\exp(-\chi/Z_1)\cdot d\chi/Z_1$ for a spur to fall within χ and $\chi+d\chi$, Z_1 being the average spacing. For alternate neighbors the function is given by $\exp(-\chi/Z_1)\cdot(\chi/1!Z_1^2)d\chi$ and so on.

Therefore the expression (IIb) can be written as

$$Q=2\int_0^R\int_0^{\infty}\left[\exp\left(-\frac{\chi}{Z_1}-\frac{\chi^2}{8Dt}\right)\frac{d\chi}{Z_1}\cdot\frac{dn}{d\rho}\cdot d\rho+\exp\left(-\frac{\chi}{Z_1}-\frac{\chi^2}{8Dt}\right)\cdot\frac{\chi d\chi}{1!Z_1^2}\cdot\frac{dn}{d\rho}\cdot d\rho+\exp\left(-\frac{\chi}{Z_1}-\frac{\chi^2}{8Dt}\right)\cdot\frac{\chi^2 d\chi}{2!Z_1^3}\cdot\frac{dn}{d\rho}\cdot d\rho+\cdots\right], \tag{IIc}$$

neglecting the negative terms of the series. The terms of the series (IIb) which take into account the widely separated spurs, contribute insignificantly to the total sum. The series converges rather rapidly; i.e., before the negative numbers in the upper limits of summations in the series (IIb) become comparable with n, the sums themselves become vanishingly small. This corresponds to the physical situation that only the neighboring spurs in a track contribute to their overlapping and the distant spurs do not overlap for all practical purposes. Hence, we may write

$$Q=2\int_0^R\int_0^{\infty}\exp\left(-\frac{\chi}{Z_1}-\frac{\chi^2}{8Dt}\right)\frac{1}{Z_1}\times\left[1+\frac{\chi}{Z_1}+\frac{\chi^2}{2!Z_1^2}+\frac{\chi^3}{3!Z_1^3}\cdots\right]d\chi\frac{dn}{d\rho}d\rho$$

$$=2\int_0^R\int_0^{\infty}\frac{1}{Z_1^2}\exp\left(-\frac{\chi^2}{8Dt}\right)d\chi d\rho$$

since

$$\frac{dn}{d\rho}=\frac{1}{Z_1}.$$

Substitution for Z_1, as obtained in Eq. (3) and integration gives

$$\sum_{\substack{i,j\\i\neq j}}^{n}\exp\left(-\frac{z_{ij}^2}{8Dt}\right)=\frac{n^2\cdot(8\pi Dt)^{\frac{1}{2}}}{(2-\eta)\eta R}.$$

Hence, from (IIa) we have

$$\frac{1}{V_t}=\frac{1}{n(8\pi Dt)^{\frac{3}{2}}}\left[1+\frac{n(8\pi Dt)^{\frac{1}{2}}}{(2-\eta)\eta R}\right],$$

which is Eq. (8).