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A Theoretical Consideration of the Electron-scavenging Process in Liquid Hydrocarbons. IV. Integral Transforms in the Prescribed Diffusion Approximation

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The relations of the integral transforms and the integral form between various distribution functions have been discussed in the prescribed diffusion approximation for the Smoluchowski equation. The initial electron distribution from the central-ion cluster in a spur or a track, $\omega(r, 0)$; the time dependence of the central value of the electron distribution, $\omega(0, t)$; the time dependence of the space-averaged electron concentration, $N(t)$, after pulse irradiation, and the scavenger-concentration dependence of the relative G-value of electron scavenging, $G_s(k_sC_s)$, as a function of the scavenger concentration, C_s, and the rate constant, k_s, are shown to be connected by this diagram of the Laplace transforms and the integral form:

$$\begin{array}{ccc} \omega(0, t) & \xleftarrow[\text{Laplace transform}]{} & \omega(r, 0) \\ \text{Integral form} \downarrow & & \\ N(t) & \xrightarrow{\text{Laplace transform}} & G_s(k_sC_s) \end{array}$$

By the use of this relation, the various distribution functions are evaluated from the generalized Gaussian initial distribution. The square-root relationship of the $G_s(k_sC_s)$ to the scavenger concentration is discussed.

The experimental and theoretical approaches to the electron-recombination process in the radiolysis of hydrocarbons have indicated that the various quantities measured are interconnected. Hummel[1] and Schuler[2] proposed that the scavenger-concentration dependence of the G-value for electron scavenging was connected with the time dependence of the charged species after pulse radiolysis by means of a Laplace transform. Recently, Mozumder[3] extended the prescribed diffusion approximation of the Smoluchowski equation to the multiple-ion spur, blob, and track. He obtained various relationships, namely, the time dependence of the concentration of the charged species after pulse irradiation as well as the G-value of electron scavenging, as functions of the scavenger concentration, using the initial Gaussian distribution of the thermalized electrons. Sato and Oka[4] and Tachiya[5] obtained the relation between the G-value of electron scavenging and the initial spatial distribution of the thermalized electrons by ignoring the effect of diffusion. In a

1) A. Hummel, *J. Chem. Phys.*, **49**, 4840 (1968).
2) S. J. Rzad, P. P. Infelta, J. M. Warman, and R. H. Schuler, *ibid.*, **52**, 3971 (1970).
3) Mozumder, *ibid.*, **55**, 3020 (1971): **55**, 3026 (1971).
4) S. Sato and T. Oka, This Bulletin, **44**, 856 (1971).
5) M. Tachiya, *J. Chem. Phys.*, **56**, 6269 (1972).

previous paper,[6] in which the diffusion term of the Smoluchowski equation was neglected, the present authors showed a square-type flow chart of interconnection by means of Laplace transforms among four characteristic functions, the initial spatial distribution of thermalized electrons, the scavenger-concentration dependence of the relative G-value of electron recombination, the time dependence of the electron concentration after pulse irradiation, and the life-time spectrum of the decay of the electron concentration after pulse irradiation. In this paper it will be shown that a similar square diagram of the flow chart for the characteristic functions is present in the prescribed diffusion approximation of the Smoluchowski equation. Various explicit expressions of the characteristic functions for the generalized Gaussian distribution of the thermalized electrons will be given using these relations of the integral transforms in the flow chart.

Theory

The Extended Form of the Smoluchowski Equation and its Prescribed Diffusion Approximation. The extended form of the Smoluchowski equation[7] for the time-dependent spatial distribution of electrons without any electron scavenger in spherical geometry or in cylindrical geometry is expressed by:

$$\frac{\partial C}{\partial t} = D(1/r^n)\frac{\partial}{\partial r}\left(r^n\frac{\partial C}{\partial r}\right) + \frac{u}{r^n}\frac{\partial}{\partial r}\left(r^n C\frac{\partial \phi}{\partial r}\right) \quad (1)$$

($n=2$, spherical geometry; $n=1$, cylindrical geometry),

where $C(r, t)$ is the distribution function, D is the diffusion constant, u is the mobility, and ϕ is the static electric potential.

The electric-field strength is given thus for spherical geometry:

$$F_n = \frac{e\partial\phi(r,t)}{\partial r} = -e^2(N^+(r,t) - N(r,t))/\varepsilon r^2 \quad (2)$$

and thus for cylindrical geometry:

$$F_n = \frac{e\partial\phi(r,t)}{\partial r} = -2e^2(N^+(r,t) - N(r,t))/\varepsilon r \quad (3)$$

where the plus sign of the superscript denotes quantities for positive ions, ε is the dielectric constant, and $N^+(r,t)$ and $N(r,t)$ are given by:

$$N^+(r,t) = \int_0^r C^+(r,t)\mathrm{d}v \quad (4)$$

and:

$$N(r,t) = \int_0^r C(r,t)\mathrm{d}v \quad (5)$$

where $\mathrm{d}v$ is a volume element. For the sake of simplicity it is assumed that the positive ions are localized at the origin or in the z-axis:

$$C^+(r,t) = N(t)\delta(r) \quad (6)$$

where $\delta(\boldsymbol{r})$ means the Delta function of $\boldsymbol{r}$; the prescribed diffusion approximation is used for the distribution of electrons:

$$C(r,t) = N(t)\omega(r,t) \quad (7)$$

where $\omega(r, t)$ is a solution for the corresponding diffusion equation. Equations (2) and (3) may then be rewritten as:

$$F_n = \frac{e\partial\phi(r,t)}{\partial r} = -e^2N(t)\left(1-\int_0^r \omega(r,t)\mathrm{d}v\right)\Big/\varepsilon r^2, \quad (8)$$

$$(\mathrm{d}v = 4\pi r^2\mathrm{d}r)$$

and:

$$F_n = \frac{e\partial\phi(r,t)}{\partial r} = -2e^2N(t)\left(1-\int_0^r \omega(r,t)\mathrm{d}v\right)\Big/\varepsilon r, \quad (9)$$

$$(\mathrm{d}v = 2\pi r\mathrm{d}r)$$

According to the analysis of Mozumder,[3] the electron decay by recombination is to be expressed in terms of the central value of the electron distribution, $\omega(0, t)$, for spherical and for cylindrical geometry:

$$\frac{N(t)}{N(0)} = 1\Big/\left[1+4\pi Dr_c N(0)\int_0^t \omega(0,t)\mathrm{d}t\right] \quad (10)$$

where $N(0)$ is the initial number of electrons in a spur or in a unit length of track and where $r_c(=e^2/\varepsilon kT)$ is the Onsager length. The details of the more general calculation for evaluating Eq. (10) are given in the Appendix. Inversely, the central value, $\omega(0, t)$, in a spur or in a track is given by the electron-decay function for recombination:

$$\omega(0,t) = \frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{1}{4\pi Dr_c}\left(\frac{1}{N(t)} - \frac{1}{N(0)}\right)\right\} \quad (11)$$

Relations of Laplace Transforms. Since $\omega(r, t)$ is the solution of a diffusion equation, the central values of distributions in a spherical spur and in a cylindrical track are expressed in terms of the initial distribution:

$$\omega(0,t) = \frac{4\pi}{(2\sqrt{\pi Dt})^3}\int_0^\infty \exp(-r^2/4Dt)\omega(r,0)r^2dr \quad (12)$$

and:

$$\omega(0,t) = \frac{2\pi}{(2\sqrt{\pi Dt})^2}\int_0^\infty \exp(-r^2/4Dt)\omega(r,0)rdr \quad (13)$$

The relation between the initial distribution and the central value of distribution in spherical geometry can be conveniently described by the Laplace transform:

$$\omega(0, 1/4D\tau)\sqrt{\pi}/2\sqrt{\tau}^3 = \int_0^\infty \exp(-\xi\tau)\omega(\sqrt{\xi}, 0)\sqrt{\xi}\,\mathrm{d}\xi \quad (14)$$

and:

$$\omega(\sqrt{\xi}, 0)\sqrt{\xi} = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty}\omega(0, 1/4D\tau)\frac{\sqrt{\pi}}{2\sqrt{\tau}^3}\exp(\xi\tau)\mathrm{d}\tau \quad (15)$$

where τ and ξ are the substitutions of $1/4Dt$ and r^2 respectively, and where c is taken as having a larger real value than the conversion axis. Similarly, the Laplace transforms in cylindrical geometry are:

$$\omega(0, 1/4D\tau)/\tau = \int_0^\infty \exp(-\xi\tau)\omega(\sqrt{\xi}, 0)\mathrm{d}\xi \quad (16)$$

and:

$$\omega(\sqrt{\xi}, 0) = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty}\omega(0, 1/4D\tau)\frac{1}{\tau}\exp(\xi\tau)\mathrm{d}\tau \quad (17)$$

According to Hummel,[1] Schuler,[2] and Mozumder,[3] the G-value, G_s, of electron scavenging is expressed by the Laplace transform of $N(t)/N(0)$:[*1]

6) H. Yamazaki and K. Shinsaka, This Bulletin, **45**, 1335 (1972).
7) H. Yamazaki and K. Shinsaka, *ibid.*, **43**, 2713 (1970).

*1) In a previous paper,[6] the relative G-value of the electron recombination was used instead of $G_s(\beta)$, that of electron scavenging.

$$G_s(\beta) = \beta\int_0^\infty N(t)/N(0)\exp(-\beta t)dt \tag{18}$$

where β is the product of the rate constant, k_s, of the electron scavenging and the scavenger concentration, C_s. Using the inverse Laplace transform, $N(t)/N(0)$ is given by:

$$N(t)/N(0) = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty}\frac{G_s(\beta)}{\beta}\exp(\beta t)d\beta \tag{19}$$

As a summary of these transforms of the four characteristic functions, the following flow chart of the transformations is given by Fig. 1. A similar flow chart of the linear response of the four characteristic functions was given in a previous paper[6] using the potential control approximation.

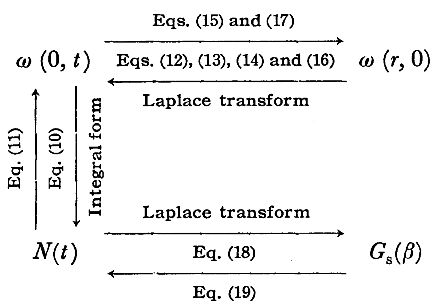

Fig. 1. Flow chart of Laplace trasform and integral form of the four characteristic functions: $\omega(r,0)$, $\omega(0,t)$, $N(t)$ and $G_s(\beta)$.

Examples of Distribution Functions. To evaluate the explicit form of the four characteristic functions, a generalized Gaussian distribution for the thermalized electrons in the spherical spur will be considered:

$$\omega(r,0) = A_m r^m \exp(-r^2/b^2) \tag{20}$$

where A_m is a normalization constant and where b is a constant of the spread of the distribution:

$$A_m = 1\Big/\int_0^\infty 4\pi r^{m+2}\exp(-r^2/b^2)dr \tag{21}$$

Similarly, the initial distribution in cylindrical geometry is expressed by Eq. (20) with the normalization constant of:

$$A_m = 1\Big/\int_0^\infty 2\pi r^{m+1}\exp(-r^2/b^2)dr \tag{22}$$

By using Eqs. (12), (13), (14), and (16), the central values in the distribution of electron can be obtained for spherical and cylindrical geometries:

$$\omega(0,t) = (4Dt)^{m/2}/\{\pi^{3/2}(4Dt+b^2)^{(m+3)/2}\} \tag{23}$$

and:

$$\omega(0,t) = (4Dt)^{m/2}/\{\pi(4Dt+b^2)^{(m+2)/2}\} \tag{24}$$

By means of Eq. (10) the time dependence of the concentration in spherical geometry is expressed in terms of the incomplete beta function:

$$\frac{N(t)}{N(0)} = 1\Big/\left\{1+\frac{r_c N(0)}{b}B\left(\frac{1}{2}, \frac{m+2}{2}; \frac{4Dt}{b^2}\right)\right\} \tag{25}$$

where the incomplete beta function is defined by:

$$B(p,q;x) = \int_0^x \frac{x^{q-1}}{(1+x)^{p+q}}dx \tag{26}$$

By the use of the Laplace transform, Eq. (18), the G-value of electron scavenging is obtained:

$$G_s(\beta) = \beta\int_0^\infty 1\Big/\left\{1+\frac{r_c N(0)}{b}B\left(\frac{1}{2}, \frac{m+2}{2}; \frac{4Dt}{b^2}\right)\right\} \times \exp(-\beta t)dt \tag{27}$$

For dilute concentrations of a scavenger, $G_s(\beta)$ can be expanded as the power series of β for the Gaussian distribution ($m=0$):

$$G(\beta) = \frac{1}{1+K} + \frac{\sqrt{\beta}K\sqrt{\pi}}{\sqrt{\alpha}(1+K)^2}\{\exp(\beta/\alpha)\ Erfc\ (\sqrt{\beta/\alpha})\} - \frac{\beta K^2}{\alpha(1+K)^3}[\exp(\beta/\alpha)\ Ei\{-(\beta/\alpha)\}] \tag{28}$$

where $Erfc$ and Ei are the error function and the exponential integral function,[8)*2)] and where K and α denote $2r_c N(0)/(\sqrt{b\pi})$ and $4D/b^2$ respectively. The leading term in the low concentration of the scavenger is $\beta^{1/2}$, which corresponds to the square-root dependence proved by various experiments. The coefficient of the $\beta^{1/2}$ term is:

$$\frac{K\sqrt{\pi}}{\sqrt{\alpha}(1+K)^2} = \frac{r_c N(0)}{\sqrt{D}(1+2r_c N(0)/\sqrt{\pi}b)^2} \tag{29}$$

which corresponds to α in Schuler's expression.[2)*3)]

Similarly, in cylindrical geometry $N(t)/N(0)$ is given for the Gaussian distribution ($m=0$) for the thermalized electrons by:

$$N(t)/N(0) = 1/\{1+r_c N(0)\ln(1+4Dt/b^2)\} \tag{30}$$

$G_s(\beta)$ is evaluated by[8)] means of the Laplace transform:

$$\begin{aligned} G_s(\beta) &= \beta\int_0^\infty\{1-r_c N(0)\ln(1+\alpha t)+r_c^2 N^2(0) \\ &\quad \times(\ln(1+\alpha t))^2\cdots\}\exp(-\beta t)dt \\ &= 1-\exp(\beta/\alpha)\ Ei\,(-(\beta/\alpha))+\cdots \end{aligned} \tag{31}$$

Therefore, the square-root law does not hold for the electron-recombination process with the Gaussian distribution of thermalized electrons in the cylindrical track. After a long interval, however, the cylindrical track is converted to the 'isolated' ion-pair geometry after recombination in the track. Therefore, the final state can be expressed by the spherical coordinate and

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8) For example, H. Beteman, "Table of Integral Transforms. I," McGraw-Hill Book Co., New York (1954).

*2) The error function and exponential integral function are given by:

$$Erfc\ (x) = \frac{2}{\sqrt{\pi}}\int_x^\infty \exp(-v^2)dv$$

and:

$$-Ei\,(-x)\int_x^\infty \exp(-v)/v\,dv$$

*3) In Schuler's expression[2)], $G_s(\beta)$ can be written as:

$$G_s(\beta) = g_{fi} + g_{gi}\{\sqrt{\alpha\beta/k_s}/(1-\sqrt{\alpha\beta/k_s})\}$$

where g_{fi} and g_{gi} are the relative G-values of free electrons and of geminate electrons:

$$g_{fi} = G_{fi}/(G_{fi}+G_{gi})$$

and:

$$g_{gi} = G_{gi}/(G_{fi}+G_{gi})$$

Schuler's expression can be expanded in the series of β:

$$G_s(\beta) = g_{fi} + g_{gi}\sqrt{\alpha\beta/k_s} + \cdots$$

From Eq. (28), the coefficients of the first and second terms are found to be:

$$g_{fi} = 1/(1+K)$$

and:

$$g_{gi}\sqrt{\alpha/k_s} = \frac{r_c N(0)}{\sqrt{D}(1+2r_c N(0)/\sqrt{\pi}b)^2}$$

the square-root law is valid.[3] The contribution of the spherical spurs to the whole process of recombination can be estimated by the factor of $\sigma(=1/\{(2r_cG_i)\text{-}(-\mathrm{d}E/\mathrm{d}x)\})$, where r_c is the Onsager radius, G_i is the G-value of the total ionization, and $(-\mathrm{d}E/\mathrm{d}x)$ is the average LET in the radiation track. For example, the α beam of 20 Mev and the $^{4+}$N beam of 80 Mev, which are used for the study of the LET effect in hydrocarbons,[9] the factor σ, is evaluated as 2×10^{-2} and 4×10^{-3} respectively, assuming that G_i is 3 and r_c is 100 Å. Therefore, the contribution of the square-root dependence is negligibly small in these cases.

In this paper, the integral transforms in the prescribed diffusion approximation have been discussed. The prescribed diffusion approximation, one of the most common approximations, has been used since the initial stage of the development of the theroy of radiation chemistry. However, the real importance of the integral transforms is that the transforms can also be applied to the other forms of approximation.[5,10–12] On the other hand, in their preliminary stages of development these approximations should be considered as of equal importance.

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Appendix

Introduction of a Finite Reaction Radius. It is reasonable to consider the finite radius, r_0, of the reaction between central-ion clusters and the surrounding electrons, because to consider ion clusters as a point or an axis is not a realistic view of the models, as has been discussed in connection with other approximations,[10,13] and the origin of coordinate is a singular point of the Smoluchowski equation. The solution of the diffusion equation, $\partial v/\partial t=D\Delta^2v+\delta(r-r')\delta(t)$, is given by,[14]

$$v(r,t)=\frac{1}{8(\pi Dt)^{3/2}}\exp\{-(r-r')^2/4Dt\} \quad \text{(A-1)}$$

If the reaction radius, r_0, is used, then the value, $\omega(r_0,t)$, at the reaction radius can be used instead of the central value, $\omega(0,t)$. In spherical coordinates, $\omega(r_0,t)$ can be expressed by the initial distribution, $\omega(r,0)$, by using the super position of the solutions of Eq. (A-1) outside of the ion cluster*4):

$$\omega(r_0,t)=\frac{2\pi}{(2\sqrt{\pi Dt})^3}\int_{r_0}^{\infty}\int_0^{\pi}\exp(-d^2/4Dt)\omega(r,0)r^2\mathrm{d}r\sin\theta\,\mathrm{d}\theta \quad \text{(A-2)}$$

where d is the distance between the ion and the electron:

$$d^2=r^2+r_0^2-2rr_0\cos\theta \quad \text{(A-3)}$$

Similarly, in the cylindrical geometry of a track in cylindrical coordinates, $\omega(r_0,t)$ is given by:

$$\omega(r_0,t)=\frac{1}{(2\sqrt{\pi Dt})^2}\int_{r_0}^{\infty}\int_0^{2\pi}\exp(-d^2/4Dt)\omega(r,0)r\mathrm{d}r\mathrm{d}\theta \quad \text{(A-4)}$$

After integration, Eqs. (A-2) and (A-4) are transformed to:

$$\begin{aligned}\omega(r_0,t)&=\frac{2\pi}{(2\sqrt{\pi Dt})^3}\exp(-r_0^2/4Dt)\int_{r_0}^{\infty}\exp(-r^2/4Dt)\\&\quad\times\omega(r,0)r^2\frac{2Dt}{rr_0}\left\{\exp\left(\frac{rr_0}{2Dt}\right)-\exp\left(-\frac{rr_0}{2Dt}\right)\right\}\mathrm{d}r\\&=\frac{1}{\sqrt{\pi Dt}}\exp(-r_0/4Dt)\int_{r_0}^{\infty}\exp(-r^2/4Dt)\\&\quad\times\omega(r,0)\frac{r}{r_0}\sinh\left(\frac{rr_0}{2Dt}\right)\mathrm{d}r\end{aligned} \quad \text{(A-5)}$$

and

$$\begin{aligned}\omega(r_0,t)&=\frac{1}{4Dt}\exp(-r_0^2/4Dt)\int_{r_0}^{\infty}\exp(-r^2/4Dt)\\&\quad\times\omega(r,0)I_0\left(\frac{rr_0}{2Dt}\right)r\mathrm{d}r\end{aligned} \quad \text{(A-6)}$$

where I_0 denotes the modified Bessel function. For the outside of the ion cluster $(r>r_0)$, $N^+(r,t)$ and $N(r,t)$ can be expressed by:

$$N^+(r,t)=N^+(t) \quad \text{(A-7)}$$

and

$$N(r,t)=N(t)\Delta+N(t)\int_{r_0}^{r}4\pi\omega(r,t)r^2\mathrm{d}r \quad \text{(A-8)}$$

or

$$N(r,t)=N(t)\Delta+N(t)\int_{r_0}^{r}2\pi\omega(r,t)r\mathrm{d}r$$

where Δ is a correction factor of the electron probability in the ion cluster and is given by:

$$\Delta=4\pi\int_0^{r_0}\omega(r,t)r^2\mathrm{d}r \quad \text{(A-9)}$$

or

$$\Delta=2\pi\int_0^{r_0}\omega(r,t)r\mathrm{d}r$$

Then, by the use of Eqs. (2) and (3), Eqs. (8) and (9) can be corrected by:

$$\frac{\partial\psi}{\partial r}=-\frac{e}{\varepsilon r^2}N(t)\left\{1-\Delta-\int_{r_0}^{r}\omega(r_1,t)4\pi r_1^2\mathrm{d}r_1\right\} \quad \text{(A-10)}$$

in the spherical geometry and:

$$\frac{\partial\psi}{\partial r}=-\frac{2e}{\varepsilon r}N(t)\left\{1-\Delta-\int_{r_0}^{r}\omega(r_1,t)2\pi r_1\mathrm{d}r_1\right\} \quad \text{(A-11)}$$

in the cylindrical geometry. Considering that $\omega(r,t)$ is a solution of the diffusion equation:

$$\frac{\partial\omega(r,t)}{\partial t}=\frac{D}{r^n}\frac{\partial}{\partial r}\left(r^n\frac{\partial\omega}{\partial r}\right) \quad \text{(A-12)}$$

where $n=1$ for the cylindrical geometry and $n=2$ for the spherical geometry, the Smoluchowski equation can be expressed by:

$$\begin{aligned}\frac{\mathrm{d}N(t)}{\mathrm{d}t}\omega(r,t)&=-\frac{N(t)u}{r^2}\frac{\partial}{\partial r}\left\{\omega(r,t)\frac{e}{\varepsilon}N(t)\right.\\&\quad\left.\times\left(1-\Delta-\int_{r_0}^{r}\omega(r_1,t)4\pi r_1^2\mathrm{d}r_1\right)\right\}\end{aligned} \quad \text{(A-13)}$$

*4) It is possible to use the solution of the diffusion equation with the boundary condition:

$$(\partial\omega(r,t)/\partial r)_{r=r_0}=0$$

in the prescribed diffusion approximation. In this case, however, the solution assumes a more complicated expression, so, for the sake of simplicity, the present solutions for the diffusion equation were used instead.

9) M. Matsui, T. Karasawa, and M. Imamura, The 15th Conference of Radiation Chemistry at Osaka, Oct. (1972).
10) H. Yamazaki and K. Shinsaka, This Bulletin, **44**, 2611 (1971).
11) G. C. Abell and A. Mozmder, *J. Chem. Phys.*, **56**, 4079 (1972).
12) G. C. Abell, A. Mozmder, and J. L. Magee, *ibid.*, **56**, 5422 (1972).
13) M. G. Robinson and G. R. Freeman, *ibid.*, **55**, 5644 (1971).
14) For example, H. S. Carslaw, and J. C. Jaeger, "Conduction of Heat in Solids," Clarendon Press, Oxford (1959), p. 256.

in the spherical geometry and:

$$\frac{\mathrm{d}N(t)}{\mathrm{d}t}\omega(r,t) = -\frac{N(t)u}{r}\frac{\partial}{\partial r}\left\{\omega(r_1,t)\frac{2e}{\varepsilon}N(t)\times\left(1-\Delta-\int_{r_0}^{r}\omega(r_1,t)2\pi r_1\mathrm{d}r_1\right)\right\} \quad \text{(A-14)}$$

in the cylindrical geometry.

By integration outside of the ion cluster, Eqs. (A-13) and (A-14) become:

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{1}{N(t)}\right) = \left\{4\pi u\Big/\left(4\pi\int_{r_0}^{\infty}\omega(r_1,t)r_1^2\mathrm{d}r_1\right)\right\}\times\left\{\omega(r_0,t)\frac{e}{\varepsilon}(1-\Delta)\right\} = 4\pi ue\omega(r_0,t)/\varepsilon \quad \text{(A-15)}$$

and:

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{1}{N(t)}\right) = \left\{2\pi u\Big/\left(2\pi\int_{r_0}^{\infty}\omega(r_1,t)r_1\mathrm{d}r_1\right)\right\}\times\left\{\omega(r_0,t)\frac{2e}{\varepsilon}(1-\Delta)\right\} = 4\pi ue\omega(r_0,t)/\varepsilon \quad \text{(A-16)}$$

After integration over time, Eqs. (A-15) and (A-16) are transformed to the same expression for both the spherical and cylindrical geometries. $N(t)$ can be obtained from $\omega(r_0, t)$ instead of $\omega(0. t)$:

$$\frac{N(t)}{N(0)} = \frac{1}{1+4\pi Dr_c N(0)\int_0^t\omega(r_0,t_1)\mathrm{d}t_1} \quad \text{(A-17)}$$

If r_0 is converged to zero, Eq. (A-17) is reduced to Eq. (10).