LETTER TO THE EDITOR

A Light Scattering Method of Measuring Membrane Vesicle Number-Averaged Size and Size Dispersion

Dear Sir:

The Z-averaged diffusion coefficient and its relative dispersion obtained from the autocorrelation analysis of light scattered from dilute vesicle suspensions can be used to calculate vesicle number-averaged size and size relative dispersion. The equations necessary to calculate average size and size dispersion from the light scattering results are derived here.

A light scattering/intensity autocorrelation technique for measuring the vesicle Z-averaged diffusion coefficient, $<D>_Z$, and its relative dispersion, δ_Z, of dilute vesicle suspensions was presented in ref. 1. Here it is shown how $<D>_Z$ and δ_Z can be used to calculate the number-averaged vesicle radius, $<R>_N$, and its corresponding relative dispersion, δ_N. Except where noted, all symbols and abbreviations are those used in ref. 1.

Denoting Z-averaged and number-averaged quantities by Z and N subscripts, respectively, $<D>_Z$ may be written (2):

$$<D>_Z = \sum_i N_i F(KR_i) M_i^2 D_i \Big/ \sum_i N_i F(KR_i) M_i^2. \tag{1}$$

For scattering angles and vesicle sizes sufficiently small to satisfy criterion 18 of ref. 1, the vesicle structure factor, $F(KR_i) \simeq 1$ and is essentially constant for all vesicles. Then, by using an average vesicle membrane bulk density, ρ, and recognizing that the vesicle shell thickness, ΔR, is the same for all vesicles, the ith vesicle mass, $M_i = 4\pi\rho R_i^2 \Delta R$ so that Eq. 1 can be re-expressed in terms of number-averaged quantities as

$$\begin{aligned} <D>_Z &= \sum_i N_i M_i^2 D_i \Big/ \sum_i N_i M_i^2 \\ &= A \sum_i N_i R_i^3 \Big/ \sum_i N_i R_i^4 = A <R^3>_N / <R^4>_N, \end{aligned} \tag{2}$$

where the Stokes-Einstein relation, $D_i = A/R_i$, used in ref. 1 has been employed.

Similarly, δ_Z may be recast in terms of number-averages as

$$\delta_Z + 1 = \frac{<D^2>_Z}{<D>_Z^2} = \frac{<R^2>_N <R^4>_N}{<R^3>_N^2}. \tag{3}$$

When Eqs. 2 and 3 are multiplied together and the result inverted, an expression with the dimensions of length is obtained:

$$A/<D>_Z(1 + \delta_Z) = <R^3>_N/<R^2>_N. \tag{4}$$

By expanding R^3 and R^2 about R_N on the right-hand side of Eq. 4 and then number-averaging as indicated, Eq. 4 becomes

$$\frac{A}{<D>_Z(1 + \delta_Z)} = \frac{<R>_N^3 + 3\mu_{2N}<R>_N + \mu_{3N}}{<R>_N^2 + \mu_{2N}} \simeq <R>_N \frac{1 + 3\delta_N}{1 + \delta_N} \tag{5}$$

with μ_{2N} and μ_{3N} the size distribution second and third moments, respectively, and $\delta_N \equiv \mu_{2N}/\langle R\rangle_N^2$. In addition, it has been assumed that the size distribution is not too skewed so that μ_{3N} is small compared with other terms in the central expression of Eq. 5.

Eq. 5 is further simplified by relating δ_Z and δ_N as follows:

$$\frac{\delta_Z + 1}{\delta_N + 1} = \frac{\langle R^4\rangle_N \langle R\rangle_N^2}{\langle R^3\rangle_N^2} \simeq 1 + \frac{2\mu_{3N}}{\langle R\rangle_N^3 + 6\langle R\rangle_N \mu_{2N} + 2\mu_{3N}}, \quad (6)$$

where terms of the order of the size distribution fourth moment were considered negligible and ignored. Again, when the skewness is small, the second term on the right-hand side of Eq. 6 is small compared to unity and

$$\delta_N \simeq \delta_Z. \quad (7)$$

Now, Eq. 5 can be simplified to read

$$\langle R\rangle_N \simeq \frac{A}{\langle D\rangle_Z(1 + 3\delta_Z)}. \quad (8)$$

Eqs. 7 and 8 are those which can be used to calculate the vesicle number-averaged size and size dispersion from $\langle D\rangle_Z$ and δ_Z.

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REFERENCES

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