Catalytic Activity in Vesicles

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Dynamic Light Scattering Sol to Gel States

Gelatin Solution



DLS & Photon Correlation Spectroscopy (PCS)

- Solid State Laser Requirements
 - Low Noise
 - Vertical Polarization





Dahneke, Barton E. (1983), Measurement of Suspended Particles by Quasi-elastic Light Scattering 34,50

DLS & PCS



Laser Intensity Control

Solid State Laser



DLS & PCS

Correlation Function for Intensity $g^{(2)}(t) \equiv \langle I(t)I(0) \rangle \equiv$ Intensity Correlation Function For Gaussian Scattered Light $g^{(2)}(t) \equiv 1 + |\langle E(t)E(0) \rangle|^2 = 1 + |g^{(1)}(t)|^2$ **Field Correlation** Function $g^{(2)}(t)$ $\tau_{c}^{-1} = 2q^{2}D = 2q^{2}k_{B}T(6\eta\pi r)^{-1}$ 2 $q = 4\pi n\lambda^{-1}\sin(\theta/2)$ 1.5 $au_{
m c}$ $r = (16\pi/3)(n/\lambda)^2 k_B T \eta^{-1} \sin^2(\theta/2) \tau_c$ Time

Sorensen, Christopher (1987), Photon Correlation Spectroscopy and its Application to Aerosol Systems 4

g² Function v. Tau



 g^2

au (ms)

Data Interpretation

Field Correlation Function

 $g^{1}(t) = [[\phi(t) + 1 - \sigma]^{0.5} - (1 - \sigma)^{0.5}] / [1 - (1 - \sigma)^{0.5}]$ $\phi(t) = g^{2}(t) - 1$ $\sigma = \phi(0) = g^{2}(0) - 1$



au (ms)

g¹ Function v. Tau



au (ms)

Relevance to Polymerization in Vesicles

- One method for analysis concerning polymerization in a vesicle
- Useful to characterize the statistical & coherent properties of electric fields



12 µm

3 um

Catalytic Activity in a Vesicle

Requires a greater understanding of Potential and Electric Fields within and around the vesicle and reaction rate

• $k \sim e^{(-E_a/(k_bT))}$



Reaction Coordinate

Vesicle Potentials

 $V_{in} = 3\sigma 2 \sigma 3AEorb3cos(\theta)$ $V_{mid} = \sigma 3AEob3(2\sigma 2 + \sigma 1)rcos(\theta) + \sigma 3AEoa3b3(\sigma 2 - \sigma 1)cos(\theta)/r2$ $V_{out} = -Arcos(\theta) + Ab^3 r^{-2}(1 + A\sigma_3 b^3(2\sigma_2 + \sigma_1) + A\sigma_3 a^3(\sigma_2 - \sigma_1)cos(\theta)$

Where: $A = 3*[2a^{3}(\sigma_{2}-\sigma_{1}) (\sigma_{2}-\sigma_{3}) -b^{3}(2\sigma_{2}+\sigma_{1}) (2\sigma_{3}+\sigma_{2})$ Length (microns)



Vesicle Electric Fields

0.03

0.02

0.03

$$\begin{split} E_{in} &= -3\sigma_2 \ \sigma_3 A E_o b^3 \cos(\theta) \\ E_{mid} &= \sigma_{-3} A E_o b^3 (2\sigma_2 + \sigma_1) \cos(\theta) + 2\sigma_3 A E_o a^3 b^3 (\sigma_2 - \sigma_1) \cos(\theta) / r^3 \\ E_{out} &= A \cos(\theta) + 3A b^3 \ r^{-3} [(1 + A \sigma_3 b^3 (2\sigma_2 + \sigma_1) + A \sigma_3 a^3 (\sigma_2 - \sigma_1)] \cos(\theta) \end{split}$$

Where:

A= 3*[2a³(σ_2 - σ_1) (σ_2 - σ_3) -b³(2 σ_2 + σ_1) (2 σ_3 + σ_2) 0.02 Length (microns) 0.00



0.03 0.02 0.00 0.02 0.03 Width (microns)

Self-Reproducing Model

Vesicle Heating



Self-Reproducing Model

- Take DPPC and/or DLPE Lipids and produce vesicles
 - Cycle the temperature from 35°C and 42°C and back again



Sakuma & Imai (2011), Model System of Self-Reproducing Vesicles Figure 1

Results of Initial Tests Using DLPE:DPPC Vesicles

• DLPE:DPPC Vesicles (3:7)

• Buffer Solution Only (no gelatin)



T=24.8° C

T=29.7° C

T=35.5° C

Results of Initial Tests Using DLPE:DPPC Vesicles

• DLPE:DPPC Vesicles (3:7)

• Buffer Solution Only (no gelatin)



T=37.1° C

T=42.8° C

Simultaneous Dynamic Light Scattering & Imaging



Kaplan, Trappe, & Weitz (1999), Light Scattering Microscope 2

Future Directions of Research

- Self-Reproducing Model of Vesicles (DLPE/DPPC lipids)
 - Temperature Cycling
 - Potential and Electric Vector Fields inside and outside of the vesicles
- Simultaneous DLS and Imaging Microscopy
 - While attempting to reproduce vesicles

Acknowledgements

• Special Thanks to:

Dr. Bret N. Flanders & Dr. Christopher M. Sorensen, Raiya Ebini, Jordan Morris, S.Z. Ren, Krishna Panta



