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Partial Heterodyne Method to Study Spatial-Inhomogeneity in Acrylamide-co-Sodium Acrylate Hydrogel

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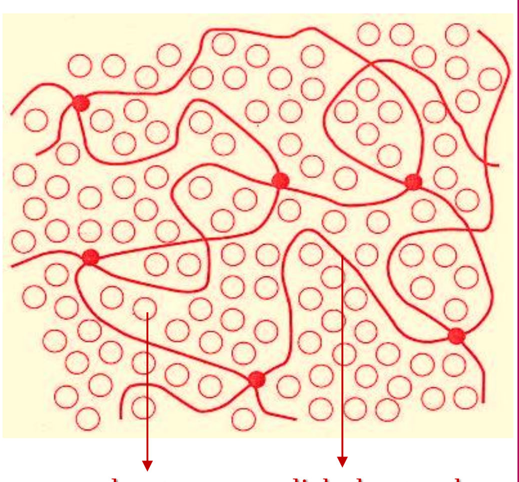
ABSTRACT

The dynamics of Acrylamide-co-Sodium acrylate hydrogels with the variation in sodium acrylate (SA) content was investigated by partial heterodyne approach using dynamic light scattering. With the addition of hydrophilic sodium acrylate into the polyacrylamide matrix, both fluctuating and non-fluctuating components decreased. Also the spatial inhomogeneities in the network was greatly suppressed as a result of rise in SA concentration. Cooperative diffusion coefficient decreased which may be attributed as the decrease in crosslinking efficiency of crosslinker with the addition of sodium acrylate.

BACKGROUND

- Hydrogels are three dimensional crosslinked polymer network swollen in solvent medium of water.
- Capable of absorbing substantial amount of water.
- Exhibits both solid and liquid like behaviour.

Common examples are: Polysaccharides, DNA/RNA, Mucin-lining the stomach, intestines etc.



Classification: Based on cross-linking types

Physical gel	Chemical gel
Formed by weak forces, like hydrogen bonds, Vander Waals force.	Formed by strong forces like covalent bonds.

Classification: Based on structure

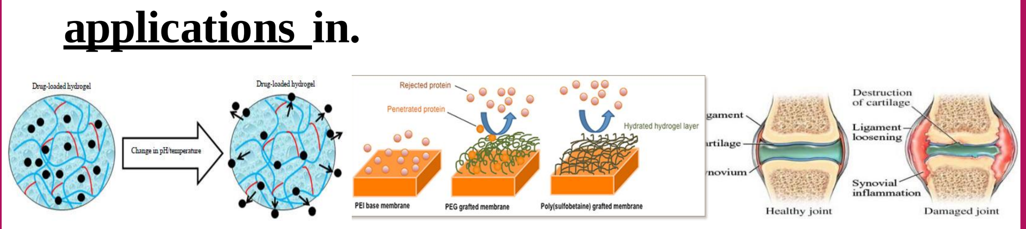
Homogeneous	Heterogeneous
Homogeneous polymer network.	Inhomogeneous polymer network.

Factors effecting hydrogel structure

- ✓ Polymer concentration
- ✓ Crosslink density
- ✓ Solution pH
- ✓ Temperature
- ✓ Solvent quality
- ✓ Salts

Why AAm/SA gel ?

✓ Excellent response to external stimuli with applications in.

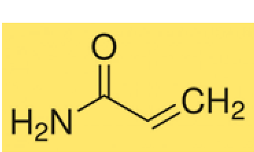


Drug Delivery Separation Technology Tissue Engineering

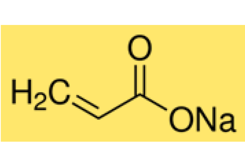
Objectives

1. To investigate the diffusive dynamics of hydrogels
2. Study the effect of comonomer content on dynamics of hydrogels

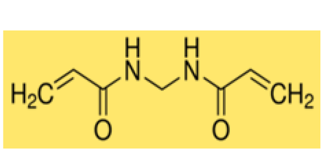
MATERIALS USED



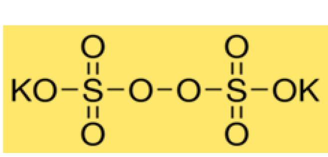
Acrylamide (AAm)



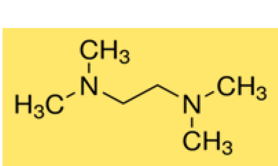
Sodium acrylate (SA)



N,N′-Methylenebis(acrylamide) (Bis)



Potassium persulfate (KPS)



N,N,N′,N′-Tetramethylethylenediamine (TEMED)

HYDROGEL PREPARATION

AAm+SA ✚ Crosslinker (Bis)

Pregel solution

Initiator (KPS)

Catalyst (TEMED)

AAm/SA hydrogel

DYNAMIC LIGHT SCATTERING

Dynamic light scattering (DLS) measures time averaged intensity correlation function (ICF) $g_T^{(2)}(q,t)$ which is practically same as ensemble averaged ICF, $g_E^{(2)}(q,t)$ for ergodic samples and is related by Siegert relation as

$$g_T^{(2)}(q,\tau) = \frac{\langle I(q,0)I(q,\tau)\rangle_T}{\langle I(q,0)\rangle_T^2} = 1 + \beta\left|g^1(q,\tau)\right|^2$$

However this is not the case for non-ergodic samples like Gels & Glasses.

Due to presence of frozen in structures, electric field has two contributions and thus $\langle I(q)\rangle_T$ has two contributions

$$\langle I(q)\rangle_T = I_c(q) + \langle I_F(q)\rangle_T$$

$\langle I_F(q)\rangle_T$-Intensities due to concentration fluctuations.
$I_c(q)$-Intensities due to frozen structures.

DLS setup in lab

The system dynamics is highly position dependent. For gels D_A as well as $\langle I(q)\rangle_T$ varies with sample position. $g_T^{(2)}(q,t)$ is measured at large number of positions (~100) and are compared with absolute intensity value, $\langle I(q)\rangle_T$ as a function of position.

Hence apparent diffusion coefficient is related to cooperative diffusion coefficient of gel by

$$D = D_A/2 - X_p \qquad X_p = \langle I_F(q)\rangle_T/\langle I(q)\rangle_T$$

For $X_P = 0$, completely heterodyne

For $X_P = 1$, completely homodyne.

The cooperative diffusion coefficient D_c and the fluctuating component of scattered intensity $\langle I_F(q)\rangle_T$ can be obtained by plotting a graph of $\langle I(q)\rangle_T/D_A$ vs $\langle I(q)\rangle_T$ from the equation

$$\frac{\langle I\rangle_T}{D_A} = \frac{2}{D_c}\langle I\rangle_T - \frac{\langle I_F\rangle_T}{D_c}$$

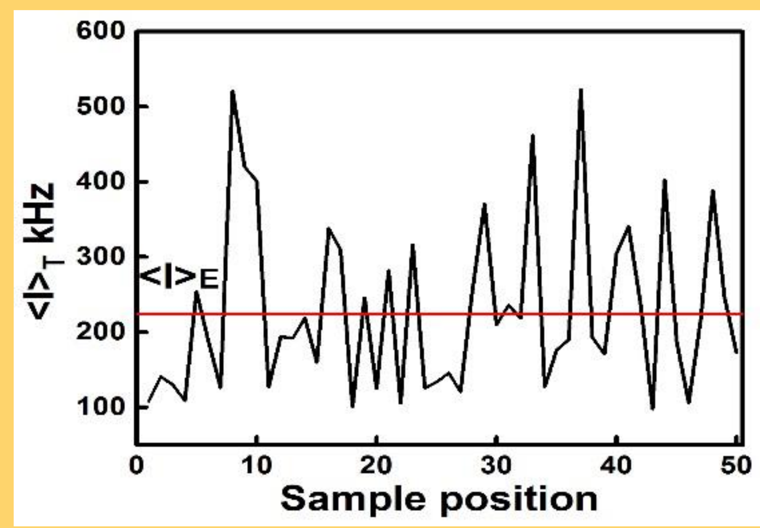
The correlation function was fitted with the equation of the form

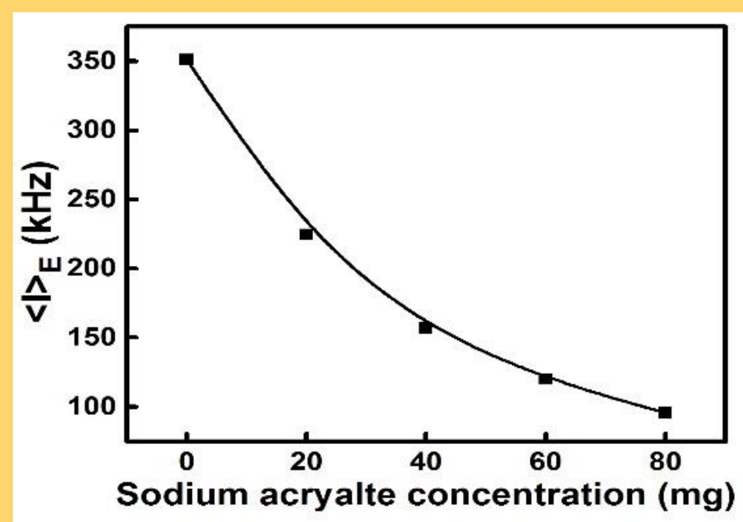
$$g_2(t) - 1 = A_f exp\left(-\frac{t}{\tau_f}\right) + A_s exp\left(-\frac{t}{\tau_s}\right)$$

Where A's are the amplitude of each mode, τ_f and τ_s are fast and slow relaxation modes respectively.

RESULTS & DISCUSSION

- $\langle I\rangle_T$ varies arbitrarily with sample position.
- Red line indicates the average equivalent to $\langle I\rangle_E$.



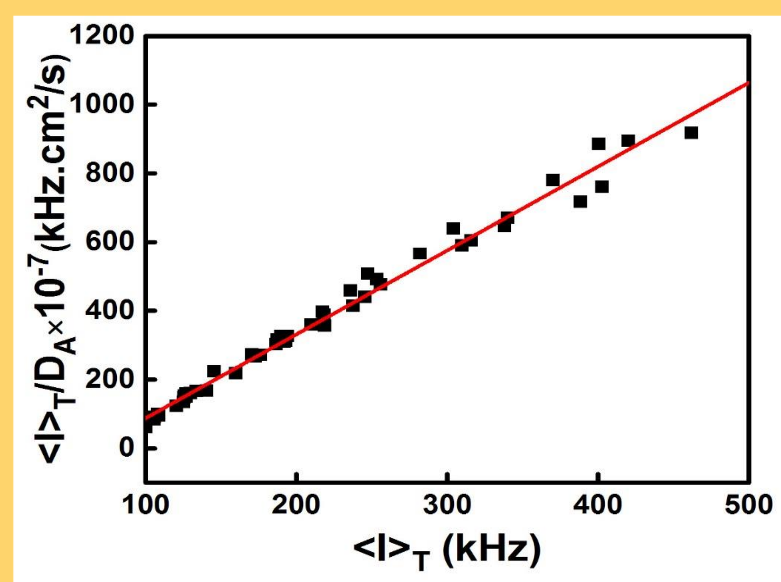


- $\langle I\rangle_E$ decreases with increasing SA concentration, spatial inhomogeneities in the gel network is systematically suppressed .

- The solid line is fitting to double exponential equation
- Inset shows the residual graph

- Intensity-intensity correlation function is plotted.
- Y intercept is less than one & position dependent, non-ergodic nature of gels

- The curves are fitted and data points collapse into a straight line.
- D_C and $\langle I_F\rangle_T$ are calculated from slope and intercept



CONCLUSIONS

- ✓ The dynamics of AAm/SA hydrogel was analyzed by employing partial heterodyne method.
- ✓ Both fluctuating and non-fluctuating components decreased with the addition of sodium acrylate.
- ✓ The cooperative diffusion coefficient D_C was seen to decrease with increase in sodium acrylate content.
- ✓ Thus by the introduction of the SA into PAAm network, the spatial inhomogeneities was greatly reduced.

REFERENCES

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2. *E. A. Kuru, N. Orakdogen and O. Okay, Eur Polym J. 43, 2913-2921 (2007).*

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