Effect of surfactant addition on dynamics of gellan gum gels: Non-ergodic approach

Mithra K[1] and Sidhartha S. Jena [1]
[1]Department of Physics and Astronomy, National Institute of Technology, Rourkela, 769008, India
mithra.k90@gmail.com

ABSTRACT SUMMARY

The dynamics of gellan gum hydrogels with addition of surfactant was studied by dynamic light scattering. A non-ergodic treatment was used for analysis. Results reveal that CMC plays a vital role in determining diffusion coefficients. The present study highlights the scope of studying structural properties of gel so that they can be tailored to suit drug delivery applications.

INTRODUCTION

Hydrogels have gained potential interest in recent decades both in industries for their versatile applications and in research to understand the structure and fundamental features. Hydrogels are of great importance in wide variety of applications including food industries, personal care, pharmaceutics, drug delivery etc[1]. Regardless of all these applications their use in drug delivery formulations is still emergent. Sometimes in order to suit specific applications, additives such as surfactants can be added. Surfactants allow modification of structure, solubilization capacity and drug release profiles of hydrogels. Thus to understand polymer surfactant interaction better, in the present study we report dynamic light scattering study of effect of sodium dodecyl sulfate (SDS) surfactant addition on dynamics of gellan gum hydrogels.

EXPERIMENTAL METHODS

To study the dynamics of gels, non-ergodic treatment was used. Time averaged intensity correlation function (ICF) $g_T^{(2)}(q,t)$ was measured for 30 minutes at a single position which also gives $\langle I(q)\rangle_T$ using a 7004- ALV multitau correlator. Ensemble averaged scattered intensity $\langle I(q)\rangle_E$ was measured by rotating the sample cell at a speed of 6rpm using a stepper motor. In the case of hydrogels because of the presence of non-ergodicity time averaged ICF is not equal to ensemble averaged ICF, the ensemble-averaged dynamic structure factor $f(q,t)$ was determined from $g_T^{(2)}(q,t)$ using Pusey and van Megan method [2.]

$$f(q,t) = \frac{Y-1}{Y} + \frac{(g_T^{(2)}(q,t) - \sigma_I^2)^{1/2}}{Y} \quad (1)$$

Where σ_I^2 is the mean square intensity fluctuation and $Y = \langle I(q)\rangle_E / \langle I(q)\rangle_T$

RESULTS AND DISCUSSION

The ISF $f(q,t)$ was constructed using equation 1 and is plotted in figure 1. The ISF for all the hydrogel samples decays from an initial value of 1 at short time. However at long times, due to the presence of frozen-in structures a non-decaying component is seen and the value saturates to different values. The long time ISF $f(q,\infty)$ which is a measure of frozen in component is analyzed with varying surfactant concertation.

All the curves are fitted with the standard equation. The values obtained from the fitting are tabulated for further analysis. The fast relaxation time is seen to shift to smaller values with increase in SDS concentration but below critical micellar concentration in this case. However above CMC, relaxation times increases. But for stretched exponential function a clear comparison is not possible due to the value of β. Nonetheless below CMC slow relaxation time decreases.

CONCLUSION

The interaction of gellan gum hydrogels with SDS was studied by use of dynamic light scattering. It was seen that surfactant as well as CMC plays a vital role in deciding the dynamics of hydrogels.

REFERENCES

1. Peppas, N A et al. Eur J Pharm Biopharm. 2000, 50, 27-46.
2. Pusey, P.; Van Megan, W. Physica A. 1989, 157, 705-741.

ACKNOWLEDGEMENTS

SJ acknowledges BRNS for funding. MK acknowledges DST for INSPIRE fellowship.

1st Controlled Release Asia (CRA) Meeting, September 24-25,2018 Singapore

Effect of surfactant addition on dynamics of gellan gum gels: Non-ergodic approach

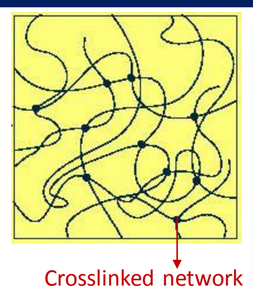
Mithra K* and Sidhartha S Jena

Department of Physics & Astronomy, National Institute of Technology Rourkela, Rourkela 769008, Odisha, India

*Email: mithra.k90@gmail.com

1. INTRODUCTION

- Hydrogels are hydrophilic crosslinked 3D polymer network.
- Capable of absorbing substantial amount of water.
- Examples : Polysaccharides DNA/RNA, Mucin-lining the stomach, intestines.



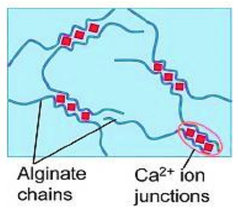
Crosslinked network

Classification

(a) Crosslinking

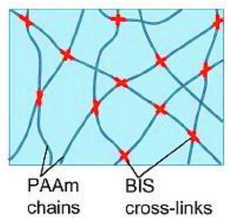
Physical gel

Formed by weak forces, hydrogen bonds, Vander Waals force.



Chemical gel

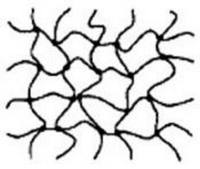
Formed by strong forces like covalent bonds.



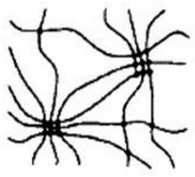
(b)Structure

Homogeneous

High polymer chain mobility



Strong inter polymer interaction, polymer chains immobile.



Heterogeneous

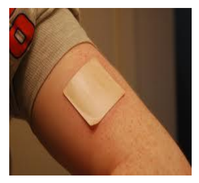
Factors effecting hydrogel structure

- Polymer concentration
- Crosslinking density
- Solution pH
- Solvent quality
- Aging of gels
- Temperature
- Presence of salts

Why Gellan gum?

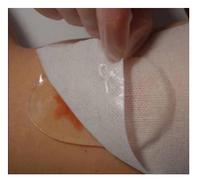
- One of the most potential in-situ gelling polymers in human body.
- Used in drug delivery vehicles.
- Used in food industry.
- Used in protein immobilization media.
- Biocompatible.
- Non-toxic.
- Structure can be altered to suit specific applications

Applications of Hydrogels

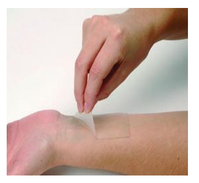


Drug delivery

Toe implants

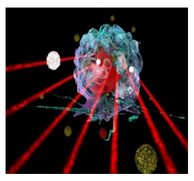


Wound dressing



Healing

Ophthalmic



Cancer therapy

Medicine

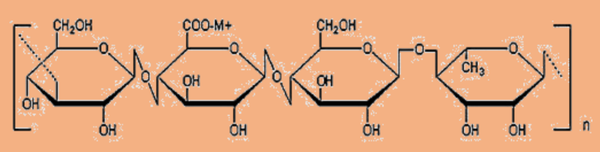
Food industry

Objectives

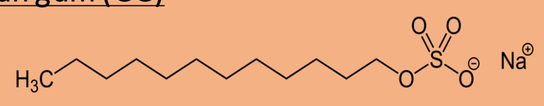
- ✓ To synthesise hydrogels with relatively homogeneous structure.
- ✓ To study the effect of surfactant addition on structure and dynamics of hydrogels

2. MATERIALS AND METHODS

Chemicals used

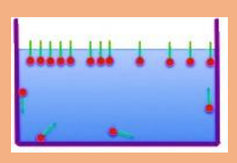


Gellan gum (GG)

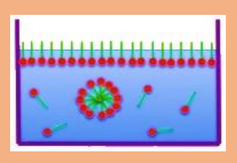


Sodium dodecyl sulfate (SDS)

CMC-Critical Micellar Concentration

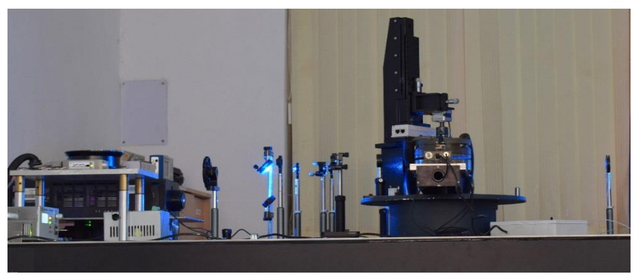


Below CMC



Above CMC

Dynamic light scattering (DLS)



DLS setup in lab

- Time averaged intensity correlation function (ICF) $g_T^{(2)}(q,t)$ was measured for 30 minutes at single position which also gives $\langle I(q)\rangle_T$.
- Ensemble averaged scattered intensity $\langle I(q)\rangle_E$ was measured by rotating the sample cell at a speed of 6rpm using a stepper motor.

The intermediate scattering function (ISF) $f(q,t)$ was determined from $g_T^{(2)}(q,t)$ by Pusey and van Megan method

$$f(q,t) = \frac{Y-1}{Y} + \frac{(g_T^{(2)}(q,t) - \sigma_I^2)^{1/2}}{Y}$$

Where σ_I^2 is the mean square intensity fluctuation & $Y = \langle I(q)\rangle_E / \langle I(q)\rangle_T$

The short time expansion is given by

$$f(q,t) = 1 + Dq^2t + ..$$

$$D = D_A\sigma_I^2/Y$$

D-corrected diffusion coefficient

D_A-apparent diffusion coefficient

3. RESULTS AND DISCUSSION

- The intermediate scattering function $f(q,t)$ was constructed using equation from measured time averaged ICFs.
- The ISF for all the hydrogel samples decays from an initial value of 1 at short time.
- However at long times, due to the presence of frozen-in structures a non-decaying component is seen and the value saturates to different values.
- This result is a clear indication of the non-ergodicity of the hydrogel samples.

- The long time ISF $f(q,\infty)$ which is a measure of frozen in component is plotted in above figure with varying surfactant concentration.
- The incomplete decay arising due to frozen in structures is clearly seen.
- They arise due to constraints imposed on the diffusion of particles by gel network.
- Below CMC the frozen in structures increases however above CMC the trend is reversed.

- The fast relaxation time is seen to shift to smaller values with increase SDS concentration but below CMC
- However above CMC, relaxation times increases.
- But for stretched exponential function a clear comparison is not possible due to the value of β. Nonetheless below CMC slow relaxation time decreases.

- The cooperative diffusion coefficient increases when surfactant is added below CMC.
- But after CMC on further increasing surfactant concentration the diffusion coefficient decreases.
- The slow diffusion coefficient also follows a similar trend but clear comparison is not possible because of β , however diffusion coefficient increases below CMC.

- The cooperative diffusion coefficient is related to the correlation length ξ by the equation $\xi = kT/6\pi\eta D_c$.
- The correlation length is found to decrease below CMC, however after CMC it increases.
- Since crosslinker is fixed in this study, the decrease of the correlation length may be due to increasing crosslinking efficiency.
- However after CMC, because of presence of micelles, crosslinking decreases hence increased correlation length.

4. CONCLUSION

- ✓ The dynamics of gellan gum hydrogels with addition of surfactant SDS was studied by dynamic light scattering.
- ✓ A non-ergodic treatment was used for analysis.
- ✓ Results reveal that CMC plays a vital role in determining diffusion coefficients.
- ✓ The present study highlights the scope of studying structural properties of gel so that they can be tailored to suit drug delivery applications.

5. FUTURE WORK

- ➢ The Effect of other surfactants on the dynamics of hydrogels can be studied.
- ➢ The mechanical properties of the prepared hydrogels can be evaluated by the rheological measurements.

6. REFERENCES

1. Peppas, N A et al. Eur J Pharm Biopharm. 2000, 50, 27-46.
2. Pusey, P.; Van Megan, W. Physica A. 1989, 157, 705-741.

7. ACKNOWLEDGEMENTS

SJ acknowledges BRNS for funding. MK acknowledges DST for INSPIRE fellowship.