

SCALING FUNCTION OF DIFFUSION COEFFICIENT OF PROBE MOLECULES IN GEL*

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When a spherical particle is immersed into a simple fluid, it fluctuates in time and space. The thermal fluctuation and the friction ζ between the particle and the fluid determine the diffusion coefficient of the particle.

$$D = \frac{kT}{\zeta} \quad 1)$$

Here, k and T are the Boltzmann's constant and the absolute temperature, respectively. The hydrodynamic friction of the fluid of the viscosity η that experienced by the particle of the hydrodynamic radius R_h is given by the following equation.

$$\zeta = 6\pi\eta R_h \quad 2)$$

The combination of above equations yields the celebrated Stokes-Einstein relationship [1].

The particle, when introduced into a gel, however, experiences the additional friction by the polymer network of the gel as well as the friction of the fluid. Thus the diffusion of the particle slows down because of the presence of the polymer network of gel. Extensive studies of the diffusion in the gel are done in various systems of the gels using many test particles (probe molecules) [2-4]. The experimental results obtained so far is, however, still a matter for discussion.

In modern statistical theory of the polymer systems, many physical quantities are expressed as a function of the relevant length of the system [5]. The relevant length scales of the probe diffusion process in the gel are the correlation length of the polymer network of the gel and the size of the probe molecule. It is, therefore, natural to expect the following scaling function $f(x)$ for the diffusion coefficient of the probe molecule in the gel [6],

$$\frac{D}{D_0} = f(x) = \exp(-x) \quad 4)$$

where D_0 is the diffusion coefficient of the probe molecule in the simple fluid and x denotes the relevant scaling variable of the form,

$$x = \frac{R_h}{\xi} \quad 5)$$

The correlation length of the polymer network depends on the concentration ϕ . It has been reported that the concentration dependence of the correlation length is well explained by the following power law relationship when the cross linking density is low enough [5].

$$\xi \propto \phi^{-3/4} \quad 6)$$

Besides this, a power law relationship can be expected for the size of the probe molecule R_h and the molecular weight of the probe molecule M if the probe molecules are compact.

$$R_h \propto M^{1/3} \quad 7)$$

Substitution of eqs. 6 and 7 into eq. 5 yields the scaling variable of the form

$$x = \frac{R_h}{\xi} \propto M^{1/3} \cdot \phi^{3/4} \quad 8)$$

Then, we expect the scaling function of the form

$$\frac{D}{D_0} \propto \exp\left(-M^{1/3} \cdot \phi^{3/4}\right) \quad 9)$$

In this paper the diffusion coefficients of probe molecules in poly(acrylamide) gel are measured using the pulsed field gradient nuclear magnetic resonance technique to study the scaling properties of the probe diffusion within the gel. The results shown in Figure 1 are analyzed in terms of the theory described above. It is found that all the reduced diffusion coefficients of probe molecules in the gel D/D_0 that obtained at different concentrations of acrylamide are well expressed by a single master curve as shown in Figure 2. It is also clear from this figure that results are in good agreement with the hydrodynamic theory. Further detailed study on this subject will promote the better understanding of the phenomenon.

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References

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- [1] A. Einstein, *Investigations on the Theory of Brownian Movement* (Dover, New York, 1956).
- [2] I.H. Park, C.S. Johnson Jr. and D.A. Gabriel, *Macromolecules* **23**, 1548 (1990).
- [3] S.J. Gibbs and C.S. Johnson Jr., *Macromolecules* **24**, 6110 (1991).
- [4] A.H. Muhr and J.M.V. Blanshard, *Polymer* **23**, 1012 (1982).
- [5] P. G. de Gennes, *Scaling Concepts in Polymer Physics* (Cornell University Press, Ithaca and London, 1979).
- [6] R. I. Cukier, *Macromolecules* **17**, 252 (1984).

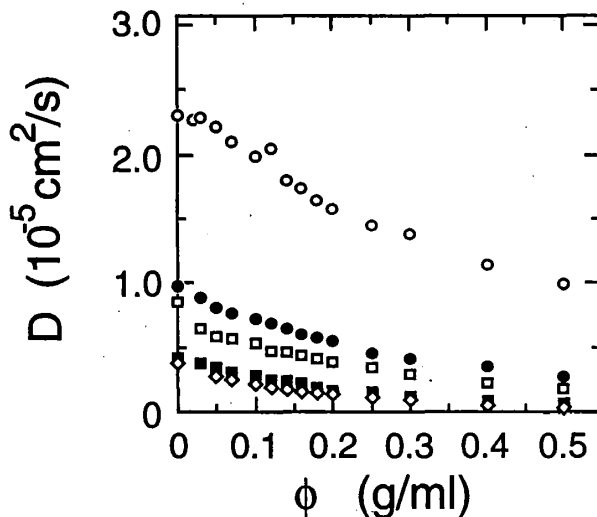


Figure 1

The dependence of the diffusion coefficients of probe molecules on the concentration of gel. The probe molecules are, water, ethanol, glycerin, PEG, and sucrose from top to bottom.

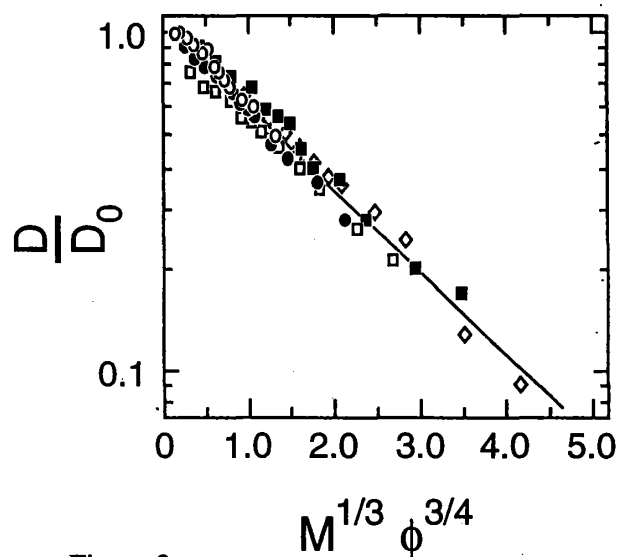


Figure 2

The scaling plot of the experimental results shown in Figure 1. The symbols are the same with that of Figure 1.