A novel method to measure the local internal structure and quantitative polymer concentration in polyelectrolyte hydrogels

T. Nishimura[1)], H. L. Guo[2)3)], R. Kiyama[1)], Y. Katsuyama[2)], J. P. Gong[2)4)], T. Kurokawa[*,2)]

[1)] Graduate School of Life Science, Hokkaido University, [2)] Faculty of Advanced Life Science, Hokkaido University, [3)] School of Chemical Engineering and Technology, Sun Yat-sen University, [4)] Institute for Chemical Reaction Design and Discovery (WPI-ICReDD), Hokkaido University

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Corresponding author*: kurokawa@sci.hokudai.ac.jp

Abstract

Polyelectrolyte gels have unique mechanical properties due to their large swelling ratio and heterogeneous internal structure. When we consider the mechanical properties of polyelectrolyte gels, it is essential to measure the charge density (polymer concentration) distribution of the polyelectrolyte chains that reflect the internal structure. However, in situ observation and quantitative measurement is still difficult. As a new approach to understand hydrogels, we have applied the microelectrode technique (MET), which has been used in the past for cell potential measurements, and constructed a method to analyze the polymer concentration distribution. Using this method for various hydrogels with heterogeneous internal structures, we succeeded in quantitatively evaluating their heterogeneous internal structures and polymer concentration distribution. This method is expected to be applied to investigate chemical adhesion principles at the adhesive interface of gels and substrates and for the structural analysis of biological tissues.

1. Introduction

Hydrogels have various properties such as flexibility, high water content, low friction, and biocompatibility. Their properties make them uniquely suitable for many biomedical applications, such as a substitute for articular cartilage. In order to apply hydrogels as new materials, it is essential to understand the internal structure and polymer concentration distribution of hydrogels, and how these characteristics influence the physical properties. Conventional methods such as x-ray scattering and observation by electron microscopy have been used to measure the internal structure of gels. However, these methods are insufficient for observing the local internal structure and quantitative polymer concentration distribution inside the hydrogel. Therefore, we developed a new method to evaluate these properties in polyelectrolyte hydrogels continuously and locally (sub-μm scale) from the gel surface by applying the MET to measure the cell potential of gels.[1)2)]. From testing with a variety of hydrogel systems, MET has shown strong potential as a method to characterize internal structure and polymer concentration.

2. Experiment

MET can measure the Donnan potential generated between the reference solution and the hydrogel by inserting a 150 nm tip diameter glass electrode at a constant speed into the gel (Fig. 1). It can convert the obtained Donnan potential to the activity of the counter ions around the polyelectrolyte chains inside the gel, and we can estimate the concentration of the counter ions around the polyelectrolyte chains by adjusting the converted activity with the

activity coefficient. Furthermore, since the counterion concentration is expected to be equal to the polymer concentration in the polymer chain based on the electrical neutrality condition, we considered that MET could be a new method to measure the local polymer concentration distribution quantitatively. Using the MET, we measured the potential of several types of hydrogels with heterogeneity to visualize the internal structure and estimate the polymer concentration distribution. In this work, we introduce the potential measurement results and polymer concentration distributions of two types of gels: one is a "particle inclusion gel" containing polyelectrolyte particles of about 50 μm in diameter, and the other is a "poor-solvent gel" in which the polyelectrolyte chains are phase-separated.

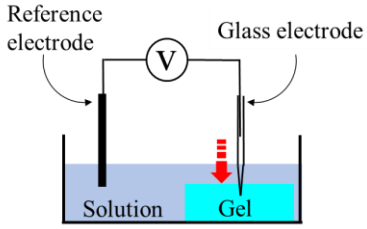

Fig.1 Microelectrode technique

3. Results and discussion

The optical images, potential profiles, and polymer concentration profile of the homogeneous gel, heterogeneous particle gel, and poor-solvent gel are shown in Fig. 2. From the potential profile of the particle gel, we found that MET can distinguish the two phases of particle and bulk portions from the potential measurement results and reflect the polymer's local internal structure. From the potential profile of the poor-solvent, we found that MET can measure the potential even in the internal structure of the gel, which has two phases: a very fine aggregate part (dense phase) and a sparse phase covering it. We also succeeded in quantitatively estimating the polymer concentration distribution in the two phases, sparse and dense, from the concentration profile.

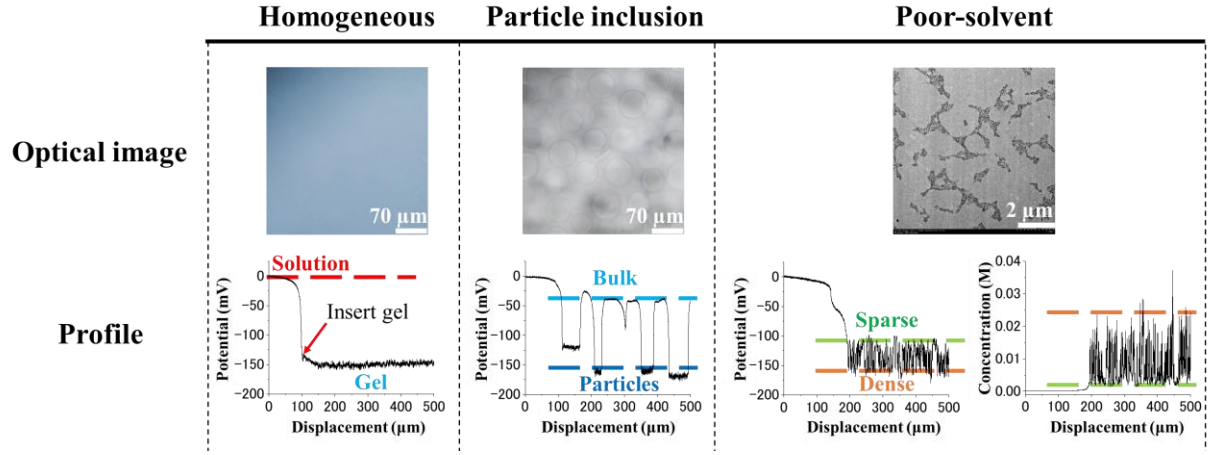

Fig. 2 The potential and polymer concentration profiles of various gels with optical images

4. Conclusion

By applying the microelectrode technique to gels, we have successfully constructed a method for local and quantitative measurement of the internal structure and polymer concentration distribution of polyelectrolyte gels.

References

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