Title	第二高調波発生顕微鏡による多糖類サクランの自己組織化構造の研究
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Citation	
Issue Date	2023-03
Type	Thesis or Dissertation
Text version	ETD
URL	http://hdl.handle.net/10119/18438
Rights	
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Abstract

The polysaccharide sacran is a supramolecular structure with many interesting natural properties such as super absorption of water, efficient adsorption of metal ions, and especially nonlinear optical susceptibility to the manifestations extraordinary strange. Moreover, the sacran molecule is a living molecule, its structure in an aqueous solution can change unpredictably due to its ability to self-assemble depending on the surrounding environment and solution concentration. This is really a big challenge for studying the structure using its nonlinear optical response. In this study, I have presented crucial research results from sonicated sacran molecules from initial 0.5 wt% sacran solutions with the methodology: combining SEM, EDX-SEM measurements, and SFG spectroscopy with SHG microscopy for the SHG-active sacran aggregates identified in sacran cast film made from ultrasonicated sacran solutions. The dissertation includes four chapters with the content:

Chapter 1: This chapter aims to give a brief overview of the theory of nonlinear optics and some most famous nonlinear optical effects commonly used in current research: sum- and difference frequency generation (S/DFG), second-harmonic generation (SHG), third-harmonic generation (THG), and coherent Raman scattering (CRS). Introduction to the background of surface second-harmonic generation (SSHG) and the SHG microscopy method used in this study of sacran molecular structure. Briefly summarize the sacran polysaccharide material used in this study and its crucial properties and raise the purpose and motivation for carrying out this study.

Chapter 2: This chapter includes an introduction to the laser system, the detector and receiver, the experimental setup of the SHG microscope, and the result of the SHG observational study that includes (i) power dependence of second-harmonic generation signal generated from sacran aggregates, (ii) polarization dependence of second-harmonic generation active sacran aggregates, (iii) nonlinear optical behavior and scavenging ability of sacran molecules, and conclusions.

Chapter 3: This section includes the combined experimental setup between SHG microscopy, SEM, EDX-SEM, and SFG spectroscopy. The results include SEM images and chemical element analysis maps of SHG-active sacran aggregates using EDX-SEM measurement and the SFG spectra of sacran cast film with very dense SHG spot distribution. In this chapter, I also pointed out the characterization of the SHG-sacran aggregates and modeled the structure of SHG-active sacran aggregates on the microscale.

Chapter 4 is a general conclusion about this study.

This study has added many points of insight into the sacran molecule whose SHG activity and nonlinear optical response at specific conditions depending on ultrasonication time and metal ion adsorption. I also first observed the SFG spectra of sacran molecules that can pave a new approach to studying this biomaterial.

I tentatively modeled the structure of the SHG-active sacran aggregates with positively charged sacran chains in the center of the SHG spot and aligned around by negatively charged sacran. On the other hand, water molecules are stored and well-aligned in the SGH-active sacran aggregates. Hence the hexagonal shape might come from an ice crystalline morphology like that of snow.

Keywords: Sacran, Second-harmonic generation (SHG) microscopy, Sum-frequency generation (SFG) spectroscopy, Scanning electron microscope (SEM), Energy-dispersive X-ray spectroscopy (EDS)