Birefringence of silica hydrogels prepared under high magnetic fields reinvestigated

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Abstract

Birefringence is an indicator of structural anisotropy of materials. We measured the birefringence of Pb(II)-doped silica hydrogels prepared under a high magnetic field of various strengths. Because the silica is diamagnetic, one does not expect the structural anisotropy induced by a magnetic field. In previous work (Mori, Kaito and Furukawa 2008 Mater. Lett. 62 3459–61), we prepared samples in cylindrical cells made of borosilicate glass and obtained a preliminary result indicating a negative birefringence for samples prepared at 5 T with the direction of the magnetic field being the optic axis. We have measured the birefringence of Pb(II)-doped silica hydrogels prepared in square cross-sectional cells made of quartz and overturned the previous conclusion. Interestingly, the magnetic-influenced silica hydrogels measured have been classified into four classes: two positive birefringent ones, a no birefringent one, and a negative birefringent one. Proportionality between birefringence and the strength of magnetic field is seen for the former two.

Keywords: non-crystalline materials, sol-gel growth, high magnetic field, birefringence

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1. Introduction

1.1. General

Birefringence is a measure of structural anisotropy of materials. For example, the measure of birefringence is proportional to the nematic order parameter in nematic liquid crystals [1]. Form and flow birefringence reflects the structural anisotropy of polymers [2]. Stress of transparent materials can be evaluated through measurement of stress birefringence [3, 4].

Birefringence itself can be a function of materials. The polarization state of light is transformed through birefringent media [3, 4]. In particular, a piece of birefringent material with a definite retardation, such as a quarter wave plate and a half wave plate, plays a special role in optical systems. That is, transformation between linear polarization and a circular one is carried out through a quarter wave plate, and that between right- and left-circular polarizations is carried out through a half wave plate. Solid materials are suitable for stabilizing the value of retardation.

On the other hand, birefringent soft materials have a potential for functional materials owing to their flexibility. One of the characteristics of soft materials is their stimuli-sensitivity. For example, nematic order induced by an external field such as an electric field can be controlled by the strength of field. One can change the direction of fast/slow axis as well as the magnitude of retardation without exchanging the piece relying on this property. Liquid-crystal variable retarders are now widely used, such as in references [5–12]. The variable focal liquid-crystal lens [13] is another example of applications, where the average birefringence is controlled through the variable nematic order induced by an external field. In turn, we shift our focus to gels. If one successfully prepares birefringent gels with a definite birefringence $\Delta n$, because the gels can easily be cut in an arbitrary size (say, $d$), one can make a piece with a desired retardation $\Gamma = \Delta nd$. Of course, there is a tradeoff between the shape’s stability and its flexibility.

The effect of a magnetic field applied during the gelation on the network structure of organic polymer gels was studied. Alignment perpendicular to the magnetic field occurred for poly(N-isopropylacrylamid) gels [14] and agarose gels [15, 16]. In cases where the side-chain group prefers the parallel alignment, polymer chains align perpendicularly to the magnetic field because the side-chain group is basically normal in the main chain. On the other hand, in cases where a group has a magnetic moment, a part including such a group prefers an orientation parallel to the magnetic field [17]. One can intuitively understand the mechanism of magnetic alignment for such polymer gels.

We have, so far, focused on the magnetic alignment of silica hydrogels. In particular, we have studied Pb(II)-doped silica hydrogels prepared in a magnetic field. In contrast to the organic polymer gels, because silica as well as Pb(II) is diamagnetic, one would not expect the magnetic alignment of silica hydrogels. However, as will be described in section 1.2, surprisingly, the magnetic alignment of silica hydrogels did occur. It is possible that there is an entirely new mechanism in the magnetic alignment, which may open a new field of science. Of course, now that the new mechanism has been revealed, new techniques to produce the alignment of materials whose magnetic alignment has not so far been expected, will be developed. One of the advantages of magnetic alignment is that the operation is simple compared to a programmed process such as thermal treatment. Anisotropic gels themselves must possess anisotropic transportation of materials within them. Such materials can be used as
new media in gel permeation chromatography. If anisotropic silica hydrogels can be solidified, e.g., by drying and then baking them, keeping the anisotropy, improvement of mechanical strength in certain direction is to be expected. Anisotropically dried silica gels can be used as new media for separation such as column chromatography. As suggested by the result of previous work [18], oriented-nanocrystallite dispersed silica materials be expected. Such materials can function in nanophotonics as well as fields of quantum confinement.

In this paper, we reinvestigate birefringence of Pb(II)-doped silica hydrogels prepared in magnetic fields. A previous conclusion of negative birefringence [19] will be reverted. This incorrect conclusion might have be due to the use of borosilicate glass cells. In appendix A, renewed results of birefringence measurement for borosilicate glass cells are presented to supplement section 1.2. The main purpose is to present the results of birefringence in samples prepared in quartz cells.

1.2. Background

In 2006, we grew lead (II) bromide (PbBr$_2$) crystals using silica hydrogels as a medium for crystal growth [18, 20]. The silica hydrogels were made from sodium metasilicate (Na$_2$SiO$_3$·9H$_2$O) aqueous solution by adding a concentrated acetic acid solution. Na$_2$SiO$_3$-based silica hydrogels were often used as media for crystal growths [21–25]. As a source of Pb(II) we added a lead (II) nitrite solution. This mixed solution was settled in a magnetic field of $B = 5$ T to prepare the silica hydrogels. We observed an ordered array of PbBr$_2$ nanocrystallites with their crystallographic axes aligned to the direction of the magnetic field as a result of the crystal growth. The fact that no magnetic field was applied during the crystal growth suggests that a structural anisotropy was formed in the silica hydrogels. To detect the anisotropy in the silica gels, in reference [19] we measured birefringence $\Delta n$ and performed scanning microscopic light scattering [26] (SMILS). Though improvement of equipment of the Sènarmont measurement is now in progress (thus, the values will be revised), at that time we detected a negative birefringence of magnitude on the order of $10^{-6}$ with the direction of the magnetic fields being as the optic axis. In addition, those values were certainly affected by the sample cells of borosilicate glass, as described in the next paragraph concerning our successive study [27]. However, the results of SMILS did certainly indicate ordering; the width of the distribution of relaxation time of the autocorrelation function of the electric fields of scattered light was smaller for samples exhibiting birefringence than for ones exhibiting no birefringence. Even if one is suspicious of the results of birefringence, we can say that the samples are classified into two: one has a narrow size distribution of the gel network and the other a wide one.

We extended birefringence measurement of Pb(II)-doped silica hydorgels, which were prepared in magnetic fields up to 10 T [27]. As sample cells, square cross-sectional cells of both borosilicate glass and quartz were used. A photograph of a cell is shown in figure 1 with an illustration of the cross-section. We note that special care should be taken inputing the light at the center of the cell in the case of cylindrical cells. Otherwise, the result will be inaccurate. This is a geometrical situation, independent of the cell materials. The method of birefringence measurement was the same as in a previous study [19]. In this paragraph, we pick up the results of birefringence measurement of samples prepared in borosilicate glass cells in order to clarify the reason why we were reached to an incorrect conclusion of negative birefringence. The results, i.e. native data, of [19, 27] are plotted together in figure 2. The data of figure 3 of reference [19] (closed circles) and those of figure 1 of reference [27] (open circles) are plotted...
together. For the former, we have drawn the standard deviation as the full length of error bars, while the half length was the standard deviation in [19]. The scale of vertical axis is the same as in [19], and thus different from that in reference [27]. Nevertheless, one can still confirm a negative slope for the open circles. A negative slope for the data of [27] is, nevertheless, still seen. We have added data for borosilicate glass cells (appendix A), but this trend remains. In reference [27], the slope was magnified, and thus we hurried to a conclusion that a decreasing property of \( \Delta n(B) \) and the negative birefringence had been reproduced.

In our previous work [27], as mentioned immediately above, we insisted on the negative birefringence and struggled to explain the results indicating the negative birefringence for borosilicate glass cells as the effect of the cell wall surface. However, if a surface effect is involved, the behaviors for both sample cells at high magnetic fields, where the field effect overcomes the surface effect, should exhibit the same trend. That is, the intercept on \( B = 0 \) is merely shifted due to the surface effect and the asymptotic behavior for the high magnetic field should be the same. The slopes of \( \Delta n(B) \) for borosilicate glass cells and quartz cells were entirely different from each other. In this respect, we infer that unexpected effects are involved in samples prepared in the borosilicate glass cells. Borosilicate glass cells are not suitable for birefringence measurement.

Figure 1. A photograph and cross-sectional dimension of cells used in [27] and the present study.

Figure 2. Data of figure 3 of reference [19] (closed circles) and those of figure 1 of reference [27] (open circles).
Before proceeding to the next section, we would like to add a note on the stimuli-sensitivity of soft materials. In section 1.1 we introduced the variable retarders and variable focal lens. These are examples of the stimuli-sensitivity after the preparation of materials. On the other hand, the present concern is on the stimuli-sensitivity during materials preparation. In this respect, stability of the structure constructed in a magnetic field may be a concern. That is, the magnetically aligned state is not one of true equilibrium. We had already confirmed that the appearance of the gel samples did not change for years if the vessels were tightly sealed and the same measured birefringence property remained. This means that the evaporation of solvent degrades the samples. To test the thermal stability, one should raise the temperature without any evaporation of the solvent.

2. Materials and methods

2.1. Materials

At first, 50g Na$_2$SiO$_3$·3H$_2$O was resolved in 103 ml distilled water as described in reference [18]; and then 8 ml of this aqueous solution was diluted with 8 ml distilled water. 16 ml concentrated acetic acid aqueous solution was added with stirring and then 6.4 ml of 1 M lead (II) nitrate aqueous solution was added. This solution was stirred for 2 hours. Next, the mixture was separated into test tubes (a photograph and an illustration of the cross-section are shown in figure 1) and sealed. After that, the test tubes were placed in equipment with a high magnetic field, a superconducting magnet (JMTD-10100 M, Japan Magnet Technologies Co.). Five different magnetic fields (3, 5, 7, 9, and 10 T) were applied by setting the samples at different positions. The samples were gellated by settlement for a week. All treatments of materials were done at room temperature (298 K).

2.2. Birefringence measurement

The intensity $I(\theta)$ of light emerging from the analysis in the Sénarmont method was measured to determine the extinction angle. A spectrometer (Ocean Optics, USB4000) was mounted on a conventional polarization microscope (Nikon OPTIPHOT2-POL) to obtain the intensity $I$ as a function of the rotation angle $\theta$ of the analyzer. By virtue of obtaining the function $I(\theta)$, instead of detecting the extinction angle through the means of the naked eye as is usually the case in the Sénarmont method, using different options to a conventional polarization microscope, we could detect small extinction angles of less than a degree. If there is no sample, the transmitted light extincts for the configuration of the crossed polarizers. If a birefringent sample is inserted with the axis pointing 45° with respect to the transmitting axis of the polarizer, the extinction angle changes. We fitted the intensity by $I = A \cos (2\theta - \delta) + C$ to determine the extinction angle $\delta$ [rad] = $2\pi\Gamma/\lambda$, where $\lambda$ is the wavelength of light source, commonly the mercury light, whose $\lambda$ is 546 nm. In the present study, a mercury light was employed. All measurements were done at room temperature (298 K). Photons were counted per 100 ms for a few minutes to obtain a time-averaged intensity $I$ at one $\theta$, and the standard deviation was also calculated. For the fitting of $I(\theta)$, 19 measurements at every 10° were taken for one point in a sample. Therefore, it took one hour or more to obtain $I(\theta)$’s measurements for one point in a sample. This method was very time-consuming; thus, the number of measured data was limited. The measure of birefringence, $\Delta n$, is obtained from $\Gamma = \Delta nd$, where $d$ is the thickness of the sample.
3. Results and discussion

As mentioned in our previous work [27], we selected cells whose optical path difference (retardation) $\Gamma$ without a sample was on the order of $10^{-1}$ nm or less (for details of $\Gamma$, see section 2.2). Results obtained according to the method described in section 2.2 are listed in table 1. At first, averages over individual samples were taken. We found that those results can be classified into four groups (two groups of positive birefringence, one with no birefringence, and one with negative birefringence). We took averages over each group. In the sample average, data belonging to different groups were treated as different ones. We included data from previous work [27]. We plot these results in figure 3.

### Table 1. Data for quartz cells, newly measured in this study.

| Sample | 10Q1 | 10Q2 | 10Q3 | 10Q4 | 10Q5 |
|--------|------|------|------|------|------|
| Position | 1    | 1    | 2    | 1    | 2    |
| $10^7 \Delta n$ | 3.84 | 0.284 | 0.714 | 8.08 | 4.79 |

| Sample | 10Q6 | 10Q7 | 10Q8 | 9Q1 | 9Q2 | 9Q3 |
|--------|------|------|------|-----|-----|-----|
| Position | 1    | 1    | 1    | 2   | 2   | 3   |
| $10^7 \Delta n$ | -0.537 | -7.96 | 10.7 | 4.92 | 2.07 | 0.261 |

| Sample | 9Q4 | 7Q1 | 7Q2 | 7Q3 | 7Q4 | 7Q5 |
|--------|-----|-----|-----|-----|-----|-----|
| Position | 1    | 2   | 2   | 3   | 1   | 1   |
| $10^7 \Delta n$ | 1.40 | 6.82 | 2.59 | 8.53 | 6.80 | 0.261 |

| Sample | 9Q6 | 7Q7 | 9Q1 | 9Q2 | 9Q3 |
|--------|-----|-----|-----|-----|-----|
| Position | 1    | 1   | 2   | 2   | 3   |
| $10^7 \Delta n$ | 1.11 | -0.174 | -0.118 | 6.29 | 2.44 | 1.30 |

| Sample | 5Q4 | 5Q5 | 5Q6 | 5Q7 | 5Q8 | 5Q9 | 5Q10 |
|--------|-----|-----|-----|-----|-----|-----|------|
| Position | 1    | 2   | 3   | 1   | 1   | 1   | 2    |
| $10^7 \Delta n$ | 1.24 | -0.752 | 0.485 | 1.92 | 2.72 | 2.62 | 1.89 | -0.214 | 0.845 | 0.397 | 0.632 |

| Sample | 3Q1 | 3Q2 | 3Q3 | 3Q4 | 3Q5 | 3Q6 |
|--------|-----|-----|-----|-----|-----|-----|
| Position | 1    | 2   | 1   | 2   | 3   | 1   |
| $10^7 \Delta n$ | -10.6 | -9.76 | -11 | 0.611 | 0.112 | -0.135 | -0.543 | 0.769 | 1.55 | 5.62 | 4.53 | 3.36 |

| Sample | 3Q7 |
|--------|-----|
| Position | 1    |
| $10^7 \Delta n$ | -9.99 | -3.72 |
As shown by the dotted lines, surprisingly, there appears to be good proportionality for the two positive birefringence groups. Points are distributed over entire lines for both branches. Also, no birefringence data are distributed up to 10 T. Data for the negative birefringence group are scattered and are accompanied by large errors. Thus, there is a possibility that the negative birefringence did not originate in the magnetic alignment. We wish to exclude those data from further discussion. There may be three phases in Pb(II)-doped silica hydrogels prepared in magnetic fields. In general, one phase is stable and the others are metastable. Birefringence of Pb(II)-doped silica hydrogels prepared in quartz cells is plotted in figure 3. As mentioned immediately above, data are classified into four groups: two positive birefringence ones (‘posi#1’ and ‘posi#2’), one no birefringence (‘no’), and one negative birefringence (‘nega’). In previous work [19], we made an image of the first-order phase transition from the no birefringence phase to the birefringence one as the strength of the magnetic field increased. On the other hand, on the basis of the present results, one can expect a successive transition from the no birefringent phase to the low-positive birefringence phase and then to the high-positive birefringence phase as the strength of the magnetic field increases. In the low-\(B\) region the stable phase should be the no birefringence one, in the middle-\(B\) region the stable phase should be the low-positive one, and in the high-\(B\) region the stable phase should be the high-positive birefringence one. In usual first-order phase transition, there are coexistence regions and at the edge of the coexistence region the curve of state equation of metastable phase bends. Along with the surprisingly good proportionality, the wideness of the metastable regions is also remarkable.

The existence of two aligned phases in magnetically-aligned Pb(II)-doped silica hydrogels may be a new discovery. As opposed to the smectic phase, where plenty of subphases exist, the nematic phase exhibits no such abundance—in this respect, the discovery of the transparent nematic phase was striking [28]. There was, however, controversy over reproducibility and then it was said that the transparent nematic phase is a metastable phase; controversies still remain such as [29]. However, from the fact that data points are distributed over a wide \(B\) range, two ‘nematic’ phases in the magnetically aligned Pb(II)-doped silica hydrogels have enough stability.

4. Concluding remarks

We have reverted a conclusion of our previous work [19] on the appearance of birefringence of Pb(II)-doped silica hydrogels prepared in magnetic fields. By preparing samples in quartz cells, we have obtained positive birefringence samples along with no birefringence samples. Besides
the no birefringence samples (phase), we were able to classify positive birefringence samples into two groups on the basis of the proportional coefficient between the birefringence $\Delta n$ and the strength of magnetic field $B$. Surprisingly, the proportionality is good for both phases. One can expect a successive phase transition among, at least, three phases. We wish to postpone the accurate determination of the proportional coefficients. Along with the accurate proportional coefficients, if the phase transition points are determined, one can develop a method to process silica hydrogels possessing desired $\Delta n$ values.

Difference in the structures of gel network of those phases will be helpful in characterizing the phase transition. Difference in two positive birefringence phases are especially of interest. Ordering of the gel network occurs in the birefringence phases. The degree of the order, and thus the structure, should be different in these phases. In applications such as separation, controlling the structure is key.

In our previous work [19], perpendicular alignment of the silica gel network to the magnetic field was speculated upon. We will reconsider the direction of the magnetic alignment of Pb(II)-doped silica hydrogels in the very near future. Fractal property was found in silica hydrogels [30, 31]. If anisotropy is introduced, self-affine analysis should be done. This will be one of our future studies. Given the small birefringence, the structural anisotropy must be small. Preparation in a stronger magnetic field will bring about an appreciable anisotropy.

Table 2. Added data for borosilicate glass cells.

| $B$ [T] | 10B1 | 10B2 | 10B3 |
|---------|------|------|------|
| sample  | 1    | 2    | 3    |
| position| 1    | 2    | 3    |
| $10^4\Delta n$ | -1.92 | -1.40 | -2.08 |

| $B$ [T] | 7    | 5    |
|---------|------|------|
| sample  | 7B1  | -    |
| position| 1    | 2    |
| $10^4\Delta n$ | -0.218 | -0.986 |

| $B$ [T] | 5    |
|---------|------|
| sample  | 5B1  |
| position| 1    |
| $10^4\Delta n$ | 8.97 |

$\Delta n$ values.

Figure 4. New results of birefringence for borosilicate glass cells.
To achieve stability against the evaporation of solvent, the solvent of the gel can be replaced with a non-voltaic one. This will be another of our future studies. Improvement of thermal stability can also be expected. It will enable a test on the effect of thermal treatment.

Acknowledgments

The samples were prepared using the equipment of the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University. It is noted that permission of reuse of published figures has been obtained from the publisher of [27] (regarding [19], the authors retain a right to reuse figures published in their own paper) although figures 2 and 3 as well as figure 4 were not exact reuse.

Appendix A. Birefringence for borosilicate glass cells renewed

We have added data for borosilicate glass cells to those shown in figure 2. Added data are listed in table 2. Samples 5-Ref [19] and 3-Ref [19] are data which were not shown in figure 3 of reference [19] (out of range). We were not interested in the positive branch at that time. Those data are included in the new results of figure 4. With the aim of drawing the curve of the negative branch in the high-\(B\) region, samples prepared in \(B = 5, 7,\) and 10 T were investigated. Unfortunately, we could not obtain results belonging to the negative branch. Instead, data for the positive branch were obtained for 5 T samples (positions 1 and 2 of 5B1 sample and position 1 of 5B2 sample). As for quartz cells, cells whose \(\Gamma\) without sample was on the order of 10\(^{-1}\) were selected. To make plots of figure 4, at first we took averages over individual samples, and then averages were taken for three groups (positive, negative, and no birefringence)—as mentioned immediately above, we had no added data for the negative branch. In the sample average, data belonging to different branches were treated as different. Also, data which were out of range of figure 3 of reference [19] are shown in table 2 and included in figure 4.

Figure 4 is essentially the same as figure 2, except for the positive branch (squares at 3 and 5 T). Thus, we can still insist on the negative birefringence. However, a possibility has arisen that the positive branch is true behavior and the point indicating the negative birefringence (triangle) is an exceptional one. That is, for example, deformation after the sample preparation may cause unexpected birefringence behavior. We wish to postpone the conclusion on the sign of birefringence for borosilicate glass cells.

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