Biological materials: the role of the tissue near the surface is important for friction mechanisms.[13, 14]. The complex adaptive multimode lubrication, including the biphasic lubrication of articular cartilage, has been partly studied in a recent report.[15]. Poly(vinyl alcohol) (PVA) hydrogel is a potential biomimetic articular cartilage material. In this study, PVA hydrogel was reinforced with PVA fibres to improve its frictional properties. By computational analysis, the position of the PVA fibre layer was examined with a migrating contact condition to reduce the friction coefficient. To improve the fibre reinforcement, a method for fibre placement was developed to retain the initial strain of the fibre in the hydrogel matrix. The experimental results showed that the fibre-reinforced PVA hydrogel with a surface fibre layer had a low friction coefficient of 0.031 in pure water. The fibre-reinforced PVA hydrogel successfully reduced friction coefficient.

1 Introduction

Articular cartilage is a soft gel material that has excellent frictional properties when used in a load-bearing system. Synovial joints are exposed to a wide weight range, supporting more than ten times the body weight during daily activities. The frictional speed sometimes decreases to nearly zero, with a low friction coefficient, i.e. <0.03. The first functionality of the soft joint surface might be the redistribution of contact stress and the improvement of frictional properties based on soft-elastohydrodynamic lubrication[1]. Under these severe and varied operating conditions, the superior frictional property is sustained by the synergistic cooperation of several frictional mechanisms, called ‘adaptive multimode lubrication’[2]. Cartilaginous gel tissue has a water content of up to 80 wt% in the proteoglycan matrix, which is enmeshed in a type II collagen network structure; this is mainly useful in tensile strain conditions. Its time-dependent compressive behaviour has been well explained by the ‘biphasic model’[3] and its collagen network structure results in a sharp reactional force peak in definite compression tests[4].

When the biphasic material is in contact with an impermeable counter surface, the interstitial fluid near the surface is not able to flow out from the contact area. In this situation, the contact load causes interstitial fluid pressurisation and the interstitial fluid presses the solid phase of the soft matrix in the direction of the pressure gradient. If the solid phase of the biphasic gel material is strengthened in the tensile direction by fibres, the fluid pressurisation effect would be improved, since the fibre-reinforced (FR) solid phase improves the ability to resist the pressurisation of the fluid phase. When the contact load is supported mainly by the fluid pressure, the resultant reduction of the solid-to-solid contact stress causes a decrease in the friction coefficient; this is called the ‘biphasic lubrication mechanism’[5], assuming that there is no friction between the solid phases. Computational methods have facilitated the investigation of the influence of biphasic cartilage functionalities on the inhomogeneity of the tissue[6], the migrating sliding condition[7, 8] and so on. As the bulk property of the tissue near the surface is important for friction mechanisms[9–12], the mechanical properties of the surface zone of the cartilaginous tissue influence the migrating sliding condition[13, 14]. The complex adaptive multimode lubrication, including the biphasic lubrication of articular cartilage, has been partly studied in a recent report[15]. Poly(vinyl alcohol) (PVA) hydrogel is a potential material that can be used as artificial cartilage; it can reproduce the adaptive multimode lubrication[16, 17] seen in the synovial joint mechanism. PVA hydrogels produced using a freeze-thawing (FT) method showed very low friction coefficients, i.e. <0.01, and minimal wear in specific simulated lubricants[18]. Another hybrid PVA hydrogel, fabricated using a FT and cast dry (CD) method, showed a low friction coefficient, which was ~0.01 in a saline solution[19]. Using a well-defined production procedure of PVA-CD and PVA-hybrid gelation, a friction coefficient of 0.006 in pure water was achieved[20].

While the boundary lubrication of PVA hydrogels has been improved by several studies, mainly using lubricants and production processes, there are other improvement strategies for natural articular cartilage. Several studies predicted that the collagen network and its arrangement in a synovial articular cartilage were effective for the improvement of interstitial fluid pressurisation and the reduction of the friction coefficient via a biphasic lubrication mechanism[8, 14]. If these computational predictions are correct, we can experimentally reproduce the same situation using artificial materials. In this study, a FR PVA hydrogel was proposed to mimic the peculiar structure of synovial articular cartilage. The effective fibre arrangements of FR-PVA hydrogel were examined using a biphasic finite-element (FE) analysis with a migrating contact sliding condition. Then, the prediction of the analyses was verified by experimental sliding tests of the FR-PVA hydrogels. The results showed that the fibre-reinforcement of PVA hydrogel successfully reduced the friction coefficient. Hence, the functional mechanism of the fibre-reinforced structure of articular cartilage was experimentally verified by using the FR-PVA hydrogel as a biomimetic artificial cartilage.

2 Materials and methods

2.1 PVA hydrogel properties

A PVA hydrogel sheet was prepared by a repeated FT method. PVA powder (Kishida Chemical Co., Ltd., Japan; Poval, 020-63185) was...
mixed with pure water at 20 wt% and dissolved at 95°C for 3 h in an autoclave. Then, the solution was stirred at 80°C for 2 h to ensure homogeneity. The casting mould for forming gel sheets of 2 mm thickness was an acrylic resin plate (Sumitomo Chemical Co., Ltd., Japan; Sumipex, t=5 mm), which was produced by a static casting method. The hot PVA solution was poured into the casting mould, which was pre-heated and kept on a hot plate at 55°C. The fibres were fabricated by PVA spinning (Kuraray Co., Ltd., Japan; Kuraron K-II WN8, #80/1, strength = 1.9 N, diameter = 0.05 mm) to secure the bonding between the PVA fibres and the PVA hydrogel matrix. The fibres were placed in a 130 mm square casting mould before pouring the PVA solution, as shown in Fig. 1. A quarter of the area of the casting mould consisted of a normal PVA gel region and another quarter of the area consisted of a cross-fibre mesh. This configuration enabled an exact comparison between two different specimens, i.e. normal PVA hydrogel and FR-PVA hydrogel, in a single production process. The cross-fibre region included two fibre layers; the total thickness was 2 mm and the fibre layer was built with a 1 mm mesh interval but was not weaved.

To generate the fibre layer in the gel matrix, a particular method was introduced, as shown in Fig. 2. A single long fibre was continuously wound around pins placed outside of the casting mould. This method improved the uniformity of the fibre tensions in a single layer. The PVA fibres shrink due to the temperature of the PVA solution; thus, this particular fibre-arrangement method generates self-tension in the mould. This low fibre tension in the PVA hydrogel matrix eliminates the initial looseness of the fibre-reinforcement network, which can then react to a compressive load without any lag in the bifphasic lubrication mechanism. The FT gelation process, which included a step at −20°C for 8 h and at 4°C for 16 h, was repeated five times. The white-coloured PVA hydrogel sheet was cut into 8 mm square pieces and stored in pure water. To verify the bonding between the fibres and the gel matrix, a preliminary tensile test was conducted using a material testing machine (Shimazu Co., Ltd., Japan; Autograph). A single uni-directional parallel fibre layer with a 2 mm interval was placed in the middle of a PVA hydrogel sheet, with a thickness of 2 mm, and cut into a standard dumbbell shape (JIS: Japanese Industrial Standard, No. 7, 2 mm width in the testing position). The tensile velocity was 2 mm/s.

The time-dependent compressive behaviour of the PVA hydrogel samples was measured using a self-made compressive tester equipped with a microscopic stage, as shown in Fig. 3. The compressive blocks are impermeable aluminium blocks. One side of the blocks generated the compressive motion, which was driven by a ball screw actuator (THK Co. Ltd., Japan; KR-15) with a DC servomotor (Yaskawa Electric Corp., Japan; Σ-V SGMMV, 20 W). The reactionary force of the compressive block on a linear ball slide (THK Co. Ltd., Japan; LS1365) was measured by a load cell (Kyowa Electric Instruments Co., Ltd., Japan; LUR-A-50NSA1). As the load cell involves a slight deformation of the sensor itself, the exact displacement of the compressive block was separately measured by an eddy current displacement sensor (Omega Corp., Japan; E2CA-X1R5A), which was also used as the position feedback control of the motion of the compressive block with a 1 μm accuracy. The overall sampling rate was 1000 Hz; the use of a modern control method with a disturbance observer enabled a high-speed precise motion control with a time constant of 10 ms. Normal PVA hydrogel specimens (N=8) were 8 mm squares. The test conditions for the normal PVA hydrogel included the application of 10 and 20% definite compression of material thickness in the first 1 s. Then, the position of the compressive block was maintained for 30 s by measuring the reactionary force in pure water. FR-PVA hydrogel specimens (N=8) were also compressed with the same configuration, i.e. 10% compression, to estimate the stiffness of the PVA fibres. The compression test was repeated eight times; hence, the variance of the experimental results was not due to regional differences of the material properties in one production sheet but rather from the initial contact conditions during the compression tests. The initial contact conditions of the compression test were determined by microscopic observation.

The bifphasic FE analysis of the experimental configuration was used to estimate the properties of the PVA hydrogel matrix via a curve-fitting method. A commercial FE analysis package,
ABAQUS V6, was used with CPE4RP elements (four-node bilinear displacement and pore pressure, reduced integration with hourglass control) with a two-dimensional model having a 0.05 mm square mesh. Fig. 4 shows the calculation conditions of the FE analysis. The friction coefficient of solid-to-solid contact, i.e. between the compressive plates and the hydrogel surface, in the FE analysis was set to 0.25 and the Poisson’s ratio of the hydrogel was set to 0, based on a previous measurement [19]. The surface seepage in the contact area was controlled with the FLOW user subroutine. The FE model of the compressive test showed a slight separation between the compressive plate and the hydrogel surface on the lateral edge. A recursive estimation of the curve-fitting procedure identified the properties of our PVA hydrogel material under two different compressive conditions (10 and 20% compression). Next, the stiffness of the PVA fibres was estimated by the fibre-reinforced FE model, as shown in Fig. 5. In the fibre-reinforced model, additional fibre layers were placed in the same position as in the experimental compression test. The fibre layer was represented by the SPRINGA element (axial spring between two nodes). The spring was a simple linear spring with no force generated in the compressive direction. The properties of the hydrogel matrix were the same as the normal PVA hydrogel. Hence, the fibre stiffness was estimated by the difference of the compressive behaviour between the normal PVA hydrogel and the FR-PVA hydrogel. The NLGEOM function in the calculation solver was used for geometrical non-linearity. The UTOL value for the threshold of automatic time length control was 10 kPa. The width of the PVA hydrogel model in the FE analysis was modulated using the average length of experimental specimens.

2.2 FE analysis of FR-PVA hydrogel with a migrating contact condition

A two-dimensional FE sliding analysis was conducted with a migrating contact condition, as shown in Fig. 6. In the experimental sliding setup, the geometrical rigid cylinder was slid over the PVA hydrogel with a 0.2 N/mm load at 8 mm/s and a 25 mm stroke, where each cycle takes 6.25 s. The friction coefficients of the middle 60% section in the second forward sliding cycle were compared to observe the influence of different fibre arrangements. In the corresponding FE analysis, the positions of the fibre layers were compared to find the effective fibre arrangements for subsequent experimental sliding tests. The relationship between the friction coefficient during migrating sliding and the depth position of the fibre layer was investigated in this study.

2.3 Experimental sliding test

From the findings of the sliding FE analysis, other fibre configurations were prepared for the experimental sliding test. Fig. 7 shows an example of a fibre arrangement. A single fibre layer was placed on the surface of the hydrogel matrix. To produce this specimen, a fibre sheet was first placed on the flat surface of the casting mould. Then, the PVA solution was poured over the PVA fibre sheet so the solution could be soaked by the fibre layer. After the gelation procedure, the specimen was cut into samples of 50 mm length and 20 mm width and attached to an acrylic resin plate with a cyanoacrylate adhesive (Konishi Co. Ltd., Japan, No.30424, AronAlpha Highspeed EX). A schematic drawing of the experimental sliding test is shown in Fig. 8. In the
ball-on-flat reciprocal sliding test, an alumina ceramic femoral head with a 13 mm radius (Kyocera, Japan) was slid over the PVA hydrogel sheet with a 6 N load at 8 mm/s and a 25 mm stroke using a frictional tester (Shinno Scientific Co. Ltd., Japan; Heidon Tribo Gear: Type 38). The experiment was conducted in a liquid bath at room temperature (23°C). To observe the effects of the fibre reinforcement on the PVA hydrogel, the lubricant used in this study was pure water. The friction coefficient was measured by averaging the middle of 60% section in the forward and backward sliding motion, similar to the sliding FE analysis.

3 Results

3.1 Identification of material properties by curve fitting

Before identifying the material properties by FE analysis, it was necessary to verify the bonding between the PVA hydrogel matrix and the PVA fibre. Fig. 9 shows the stress–strain curves of a fibre-reinforced PVA hydrogel and normal PVA hydrogel. Here, a uni-directional fibre layer with a 2 mm interval was used and the width of the testing piece was also 2 mm. Hence, the testing piece consisted of a single fibre in the PVA hydrogel. The tensile test along the perpendicular direction to the fibre orientation showed the same results as the normal PVA hydrogel. Although test was conducted as a trial preliminary experiment, the fibre-reinforced PVA hydrogel generated a steep increase in stress compared with the normal hydrogel. After 11% strain, the fibre gradually detached from the gel matrix in the experimental sample.

Fig. 10 shows the curve-fitting result for the material properties of the PVA hydrogel without fibre-reinforcement. The curve-fitting procedure successfully identified the properties of the PVA hydrogel matrix; the predicted results were in good agreement with the time-dependent behaviour obtained from the experimental compression tests. In synovial cartilage studies, it is well known that the articular cartilage shows a sharp reactional force peak just after the definite compression test [5]; this was reproduced in the curve-fitting analysis of the fibre reinforcement of the collagen network. As shown in Fig. 10, the experimental compression results without fibre reinforcement also showed a small peak just after the first compression period in each compression condition. If an additional material parameter was involved in an analytical model, the curve-fitting ability would be improved. However, the functionalities of each parameter might be complicated, and the orthogonality of the parameters would be affected. In this study, this peak just after the compression was represented using the viscoelasticity parameter because we could not reproduce this behaviour by any other parameter modulation and/or geometrical consideration. Although the use of a hyperelastic solid sponge model showed a slight improvement when fitting the experimental results, the difference was only of the order of a line width on a graph (not shown in this report). Hence, we decided to adopt the simple viscoelastic model to eliminate redundant components for comprehensive recognition. The two different time-dependent reactional forces obtained under different compressive conditions were reproduced by the single parameter set identified here in the FE calculations. The compaction effect on permeability was not required for an acceptable curve fitting.

The behaviour of the normal PVA hydrogel under 10% compression was predicted using a FE model. Using these results, the fibre-reinforced FE model was used to estimate the stiffness of the PVA fibre by fitting the time-dependent experimental results of the FR-PVA hydrogel under 10% compression. Fig. 11 shows the curve-fitting result of the fibre-reinforced FE model and the experimental compression test results of the FR-PVA hydrogel. In the FE model, the fibre layer was added to a normal PVA hydrogel model. Hence, the additional estimation parameter to fit the FR-PVA results represents the stiffness of the fibre layer. The fibre-reinforced FE model was able to reproduce the experimental time-dependent behaviour, including the inclination during the second half of the compression period. Table 1 lists the material properties identified in the analysis. Fig. 12 shows the typical behaviour of the FR-PVA hydrogel when the fibre layer is placed on the upper surface of the gel matrix. In the photograph, an experimenter pinched the specimen, but the gel specimen with the fibre layer remained straight and did not bend. This means that the superficial fibre layer effectively generated tensile stress in the specimen, as shown in Fig. 2.

3.2 Sliding analysis for fibre arrangements

In synovial articular cartilage, the collagen network is organised in a peculiar manner, with regard to the fibre density and orientation of the fibre network. The fibre arrangement in the gel matrix might

![Fig. 9](image1.png) Tensile test results to verify the bonding between PVA fibre and PVA hydrogel matrix

![Fig. 10](image2.png) Curve-fitting results to estimate the properties of PVA hydrogel

![Fig. 11](image3.png) Curve-fitting results to estimate the fibre stiffness of FR-PVA hydrogel. The result of normal PVA hydrogel, represented by the red line, is the same line in Fig. 10
be interesting to study from the viewpoint of biphasic lubrication and tribological properties. In this study, the effective position of the fibre layer was examined by biphasic FE sliding analysis, which is shown in Fig. 6. In addition, the model with no fibres and the model with two middle layers (Fig. 1) were compared. The friction coefficients obtained by the sliding FE analysis are shown in Fig. 13, where the horizontal axis is the depth of the fibre layer from the sliding surface (total thickness of 2 mm). In Fig. 13, the fibre position of 0 mm from the surface is an imaginary model because the actual fibre has a diameter and cannot realistically be located at the surface of the PVA hydrogel. With single-fibre configurations, the friction coefficient was reduced when the fibre layer approached the sliding surface. The model with two middle layers included fibre layers at depths of 0.65 and 1.35 mm, as shown in Fig. 1. The deeper layer of this model did not generate enough interstitial fluid load support on the sliding surface. Based on these results and the production cost, the most appropriate fibre arrangement was determined to be as close to the surface as possible, as shown in Fig. 7.

The internal field outputs of von Mises stress and pore pressure in the second forward sliding are shown in Fig. 14. In the contour plot, the middle two layers model (Fig. 14b) generated a high interstitial fluid pressure over the whole depth of the sample compared to the single surface layer model (Fig. 14c). Although the peak fluid pressure at the surface for the single surface layer model was not higher than that of the middle two layers model, the contact length of the single surface layer model is longer than that of the two layers model. The von Mises stress of the single surface layer model at the surface was lowest because the horizontal strain was constrained by the surface horizontal fibre. A long-term migrating sliding simulation was also conducted; the initial friction coefficient sustained its value for a sliding time of more than 200 s in all models (not shown in this report). With regard to the interstitial fluid support, the proportion of the load support in the fluid phase and solid phase in the contact area was calculated by the integration of the vertical component of the surface fluid pressure. Using the proportion of fluid load support, the effective friction coefficient $\mu_{\text{eff}}$ [5] was calculated based on the solid-to-solid friction coefficient $\mu_{\text{solid}}$. The surface layer model showed 88.4% of fluid load support in the migrating sliding condition, which led to an effective friction coefficient $\mu_{\text{eff}}$ of 0.029, while that of the no-fibre model was 0.063 and that of the middle two layers model was 0.047. The difference between the friction coefficients obtained from the sliding simulation and the calculated

| Table 1 Material properties of PVA hydrogel |
| Parameter | Nom. Value |
| --- | --- |
| Young’s modulus of solid phase* | $E$, MPa | 0.122 |
| Poisson’s ratio | $N$ | 0.35 |
| shear and volumetric relaxation modulus ratio (viscoelasticity of solid phase)* | $g$, k | 0.22 |
| relaxation time* | $T$, s | 1.0 x 10$^{-14}$ |
| permeability* | $K$, m$^3$/Ns | 0 |
| fibre stiffness (bulk modulus of solid phase)* | $K$, MPa | 4.2 |
| initial volumetric fraction of the fluid phase [18] | | 100% |
| surface seepage coefficient | $k_s$, mm$^3$/Ns | 1 in flowing, 0 in covered |
| friction coefficient of solid-to-solid contact [18] | $\mu_{\text{solid}}$ | 0.25 |

*Values identified in this study.

Fig. 12 Photograph of the FR-PVA hydrogel produced in this study

Fig. 13 Friction coefficient obtained by sliding FE analysis

Fig. 14 Internal field output of von Mises stress and fluid pore pressure in migrating sliding of
- No-fibre model
- Middle two layers model
- Single surface layer model
articular cartilage was represented by the collagen network structure. For synovial articular cartilage, more complex material parameters were required to capture the compressive behaviour under different compressive conditions, including the compaction effect on permeability and the non-linearity of collagen stiffness. The stiffness of the PVA fibre in this study was evaluated by using a simple elastic model to reproduce the reactional force of the PVA hydrogel compression. In this study, the hyperelastic solid sponge foam model with viscoelasticity was tested to improve the curve-fitting process and generate the small peak observed in Fig. 10. The model showed a slightly better result, with ~1% difference between the estimated material properties and those estimated by the normal viscoelastic model. Hence, we deduced that the small peak was derived from the viscosity of the PVA material. We could not generate the small peak behaviour without the incorporation of viscosity-related properties. In general, an additional parameter sometimes improves the curve-fitting ability. However, the orthogonality of the parameters in terms of the functionality would be affected. Thus, we believe that a simple model is able to represent the essential features of the PVA hydrogel.

It is well known that articular cartilage has a dense collagen network in the superficial tangential zone [22]. In this study, we compared the position of the PVA fibre layer in a PVA hydrogel matrix and considered the production cost. For the single-fibre layer configurations, the friction coefficient was reduced when the fibre layer approached the sliding surface. As the middle two layers model in this study was not so effective, determining the position of the second fibre layer would be interesting. In the full finite element analysis, the friction coefficient of the horizontal fibre model, where there are many horizontal fibre layers in each depth layer, was studied as an ideal model. Although this ideal model might not be a realistic structure, the full horizontal fibre model predicted a friction coefficient of 0.007 (not explained in this report). Moreover, the deep vertical fibril played an important role in improving the proportion of the fluid load support [23]. Despite problems with the production cost and technical limitations, a study on fibre arrangement would be interesting. The objectives of this study were not only the computational prediction of friction coefficients and the experimental frictional test, but also the development of a production process of artificial cartilage from an engineering viewpoint.

The functionality of the surface structure of articular cartilage was partly ascertained by the biomimetic artificial cartilage in this study. There are several aspects that can be improved in this study. The counter surface was limited to the alumina ceramic femoral head. An experimental sliding test with long-term and realistic load conditions is required to evaluate not only the friction coefficient, but also the effects of wear. A composite material sometimes undergoes structural failure and strength deterioration due to cyclic loading. The frictional behaviour in synovial lubricants and the chemical stability of the PVA fibres in PVA hydrogel has not yet been studied in practical applications. In the early stages of our study, we could not observe any effective results of the fibre-reinforcement of PVA hydrogel despite the predictions of computational FE simulations. The friction coefficient could not be reduced enough when the prepared fibre layer was simply placed in the casting mould. We found that the production process was an important factor to achieve the desired effects of the fibre reinforcement, i.e. it was necessary to prevent initial relaxation and preserve the initial tension of the embedded fibre. Hence, we developed a production method to strain the fibre in the casting mould, as shown in Fig. 2. Recently, it was reported that the initial tension of the synovial articular cartilage is effective in reducing the friction coefficient [24]. The synovial articular cartilage retains the tension of the collagen network via the hydrostatic pressure of the proteoglycan matrix. Based on our experiments and observations in this study, we believe that the initial tension of the synovial articular cartilage is a critical factor. Moreover, synovial articular cartilage has a three-dimensional fibre structure, including surface fibre orientation in the contact area, which would augment the biphasic lubrication mechanism. On the other hand, it was reported that the structure of articular cartilage is gradually built.
up during the growth process [25]. A future area of interest may be the self-organisation of artificial cartilage to preserve and enhance its functionalities. The findings of this study can provide useful insights not only for medical devices [26] but also tribology in general [27].

5 Conclusion

A fibre-reinforced PVA hydrogel was proposed to reduce the friction coefficient. The appropriate position of the fibre layer was researched by a sliding FE simulation. With a single-layer model, the surface fibre model showed the highest reduction of the friction coefficient. The experimental sliding test confirmed the prediction of FE analyses. The FR-PVA hydrogel had a friction coefficient of 0.031 in pure water. The functional mechanism of the fibre-reinforced structure of the articular cartilage surface was experimentally verified by using the FR-PVA hydrogel as biomimetic artificial cartilage.

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