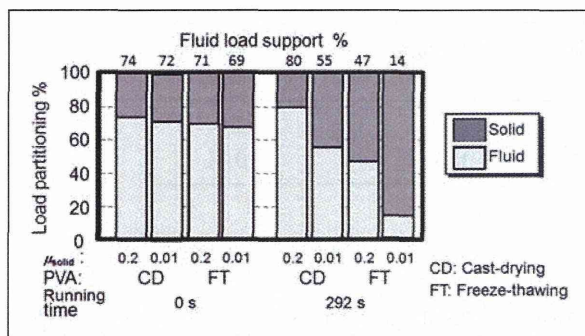
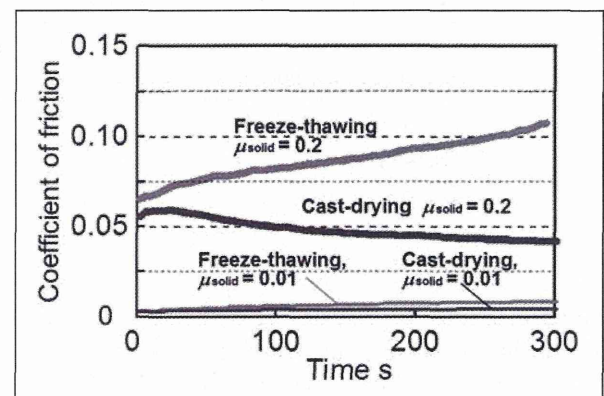


**Figure 10.** Interstitial fluid pressure in PVA hydrogels at 292 s in reciprocating tests (biphasic FE analysis). FE: finite element; PVA: poly(vinyl alcohol).



**Figure 11.** Changes in load support in PVA hydrogels (biphasic FE analysis). FE: finite element; PVA: poly(vinyl alcohol).



**Figure 12.** Changes in friction of PVA hydrogels (biphasic FE analyses). FE: finite element; PVA: poly(vinyl alcohol).

effective by supplying of important lubricating constituents of appropriate concentration and ratio (0.5 wt% HA, 0.01 wt% DPPC, 1.4 wt% albumin and 0.7 wt%  $\gamma$ -globulin) into lubricant, which corresponds to low value of  $\mu_{\text{solid}}$ . In this paper, the possibility in which similar mechanism is expected to be applicable for artificial hydrogel cartilage is discussed as below.

As artificial hydrogel cartilage containing high-water content, two kinds of hydrogel showed quite different time-dependent frictional behaviours in reciprocating tests in saline (Figure 6). In the measurement of time-dependent changes of contact area in static loading test,<sup>42</sup> freeze-thawing PVA hydrogel showed initial abrupt deformation and subsequent

steady state. Cast-drying PVA showed similarly initial abrupt deformation and maintained steady state but its compressive deformation was lower than freeze-thawing PVA. In contrast, natural articular cartilage showed gradual increase in deformation, as suggested by usual biphasic model. These phenomena are likely to depend on the permeability and elastic modulus of hydrogel. Detailed analysis for difference in deformation behaviours between PVA hydrogels and articular cartilage should be examined in further study.

The high friction and constant compressive deformation of freeze-thawing PVA hydrogel is considered to be due to biphasic properties with high permeability (Table 1). The interstitial fluid pressure in

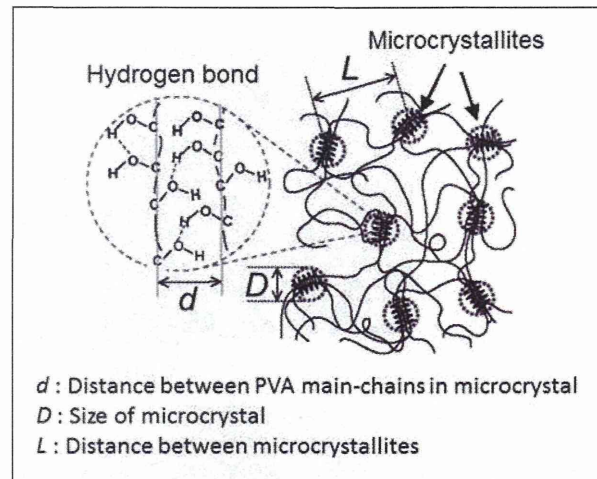


freeze-thawing PVA remarkably diminished after 292 s running as shown in Figure 10, and therefore the fluid load support became small (Figure 11). The friction in saline is expected to increase gradually as indicated by the curve for  $\mu_{\text{solid}} = 0.2$  (Figure 12). Even in case of low fluid load support, it was demonstrated in experiment that another lubrication mechanism as boundary lubrication with important synovia constituents enables to reduce friction remarkably (Figures 7 to 9) and minimize wear (Figure 9). The comparison of experimental results (Figures 6 to 8) with biphasic FE analysis (Figure 12) clearly demonstrates the acceptable correspondence of both cases for high or low friction by considering appropriate selection of 0.2 or 0.01 as  $\mu_{\text{solid}}$ . Therefore, it is indicated that the usage of freeze-thawing PVA lubricated with a simulated synovial fluid is a possible solution for clinical application.

Next, the friction behaviours of the cast-drying PVA hydrogel should be considered. As shown in Figure 6, the low friction of cast-drying PVA hydrogel during sliding duration in saline for a long time is very attractive. This property appears to be derived from low permeability (Table 1) of biphasic material. As shown in Figure 10, the interstitial fluid pressure sustained even after 292 s running for cast-drying PVA. Its fluid load support increased after 292 s running for  $\mu_{\text{solid}} = 0.2$  (Figure 11). Therefore, friction can be reduced after 292 s running for  $\mu_{\text{solid}} = 0.2$  in biphasic analysis (Figure 12). However, the frictional behaviour in experiment (Figure 6) is different from the estimated curves for  $\mu_{\text{solid}} = 0.2$  or 0.01. The appropriate selection for  $\mu_{\text{solid}} = 0.05$  or so may bring favourable correspondence.

Thus, it is worthy of remark that the permeability regulated by the material properties and structures of hydrogel will mainly control the biphasic fluid flow behaviour. The material properties and structures in both PVA hydrogels appear to depend on the structure controlled by hydrogen bond. The model for microstructure in PVA hydrogel was proposed by Otsuka and Suzuki<sup>38</sup> as shown in Figure 13.

The formation of hydrogen bonds and microcrystallites was identified using X-ray diffraction (XRD) technique, Fourier transform infrared (FTIR) spectroscopy and measurements of the swelling ratio under repeated water exchanges.<sup>43</sup> In Figure 13,  $d$ ,  $D$  and  $L$  are deduced from the diffraction peaks at XRD spectra. Small-angle scattering indicates that the distance between microcrystallites  $L$  in freeze-thawing PVA hydrogel is shorter than in cast-drying PVA hydrogel. As shown in Figure 1, the transparency of cast-drying hydrogel indicates its uniform network structure, and the milky white appearance of freezing-thawing hydrogel suggests the heterogeneous network structure. In Figure 14, the schematic models based on personal communication from E Otsuka and A Suzuki<sup>42</sup> for these hydrogels are shown accompanied with atomic force microscopy (AFM) surface images of their swollen states in water. The network



**Figure 13.** Network structure of PVA hydrogels crosslinked by microcrystallites.<sup>38</sup>

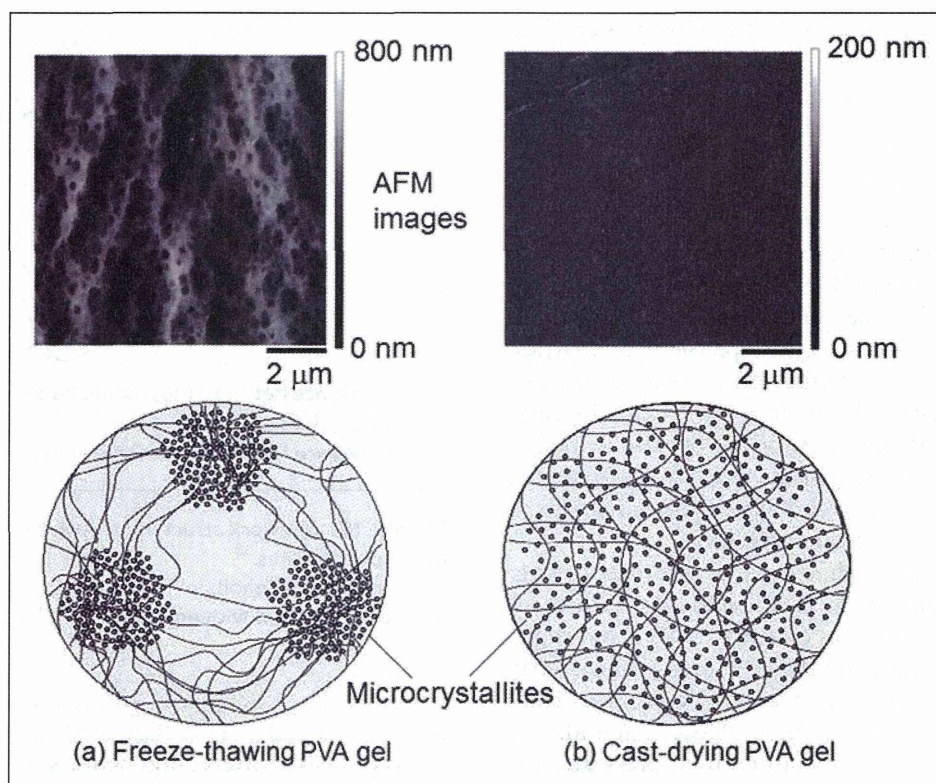
PVA: poly(vinyl alcohol).

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is generally composed of microcrystallites and amorphous zones. It is suggested that the fluid flows out rapidly from freeze-thawing PVA hydrogel, which is composed of microcrystallites and considerable amorphous zones, as indicated by high permeability (Table 1), and porous texture in AFM image (Figure 14). On the contrary, in cast-drying PVA hydrogel with low permeability, the fluid retained inside of gel with slow flows out, and thus the interstitial fluid pressure maintains for longer time under reciprocating tests.

Friction for biphasic materials under lubricated conditions is estimated from a combination of the friction for solid-to-solid contact and the negligibly low friction for fluid load support, as indicated in formula for friction by Ateshian et al.,<sup>44,45</sup> where high partitioning of load support by fluid phase contributes to lowering in friction. Changes in load support in compliant hydrogels estimated on the basis of biphasic FE analysis for reciprocating test under continuous loading are shown in Figure 11, which depend on not only permeability but also friction level for solid-to-solid contact. In saline where the friction for solid-to-solid contact is high, the higher load support by interstitial fluid pressure maintained with low permeability for cast-drying gel gives lower friction, while the limited load support by low-fluid pressure attributable to faster fluid exudation caused by higher permeability for repeated freeze-thawing gel gives high friction, as shown in Figure 12. The lower friction for cast-drying gel in saline (Figure 6) appears to be brought about by superior surface lubricity as  $\mu_{\text{solid}} = 0.05$  or so as mentioned above. The smooth surface of cast-drying PVA may contribute to the reduction of friction in saline by suppressing interaction between rubbing surfaces. The rough and porous surface of freeze-thawing PVA may enhance interaction between rubbing surfaces in saline and





**Figure 14.** AFM images in water and schematic models<sup>42</sup> for two kinds of hydrogel structures. AFM: atomic force microscopy.

increase friction, but improve the adsorbed film formation under lubricated condition with a simulated synovial fluid and reduce friction (Figures 8, 9 and 12). Elucidation of the exact lubrication mechanism for hydrogel should be continued in further studies.

Other improving methods for properties of PVA hydrogel are to additionally control the nanoscopic structure, to reinforce the structure with fibrous element or other method and thus to enhance biphasic properties for efficient lubrication and load-carrying property. The freeze-thawing and cast-drying PVA hydrogels based on hydrogen-bonding network as physical bonding prepared without any additives or irradiative stimulus have superiority in safety in human body. The possibility of restoration with hydrogen bonding in case of local fracture is a favourable property.

The main purpose of this study was to examine friction differences between two kinds of hydrogels, but wear differences become more important in clinical application. In authors' related research on reciprocating tests with an alumina ball against flat PVA hydrogel plates in pure water (Suzuki et al.<sup>46</sup>), cast-drying gel with low permeability showed lower wear and lower friction than freeze-thawing gel with high permeability. The superiority of cast-drying gel appears to be caused by its better biphasic property in water. However, in this study, freeze-thawing gel exhibited minimum wear in simulated synovial lubricant as HA solution containing appropriate concentration of albumin,  $\gamma$ -globulin and DPPC in

reciprocating tests under continuous loading. This indicates the effectiveness of surface protection by adsorbed film formed on freeze-thawing gel surface. The wear behaviour of cast-drying gel in various lubricants should be examined in future studies.

For the clinical application of PVA hydrogel, durability, synergistic interaction with lubricants, ease of production, formability, appropriate fixation methods and other properties are required. In particular, the establishment of appropriate design in consideration of a multimode lubrication mechanism is required not only for low friction but also for zero-wear and high-fatigue resistance in the human body. To evaluate predictively the in vivo behaviours of two kinds of hydrogels, the various tests such as reciprocating tests and simulator tests should be conducted under biomechanically simulated environmental conditions. The desirable in vivo performance of PVA hydrogel implanted in rabbit knee joint in ongoing tests with biomedical research group suggests the feasibility of clinical application. In the next step, the authors plan to apply a hybrid hydrogel (Suzuki et al.<sup>46</sup>) composed of cast-drying PVA on freeze-thawing PVA as another candidate material for the artificial cartilage with superior lubricity.

## Conclusion

To explore the superior lubricity of articular cartilage and artificial hydrogel cartilage, time-dependent frictional behaviours in natural articular cartilage and



two kinds of PVA hydrogels prepared by repeated freeze-thawing method and cast-drying method were examined in reciprocating tests and biphasic FE analyses.

Articular cartilage showed initial low friction and gradual increase in saline lubrication where biphasic fluid load support mechanism subsides with rubbing as indicated by biphasic FE analysis. However, the addition of a simulated synovial fluid enabled the superior lubrication by appropriate adsorbed film formation in boundary lubrication mode.

In artificial cartilage materials in saline, cast-drying PVA with low permeability clearly showed significantly lower friction than freeze-thawing PVA with high permeability. For freeze-thawing PVA hydrogel in which biphasic lubrication mechanism diminished with rubbing, the supply of appropriate synovial constituents improved friction and wear properties in boundary lubrication mode.

It was shown that the synergistic combination of biphasic lubrication and boundary lubrication becomes effective to sustain superior lubricity in articular cartilage and PVA hydrogels even at slow movement under continuous loading.

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## Appendix

### Notation

$C$	constant
$f(t)$	compressive stress during stress relaxation
$G$	shear modulus of rigidity
$k$	permeability
$K$	bulk modulus of elasticity
$t$	time
$t_{rel}$	relaxation time
$W$	shorter width of specimen
$\epsilon_{z0}$	compressive strain
$\mu_{solid}$	friction coefficient for solid-to-solid contact



# Influence of synovia constituents on tribological behaviors of articular cartilage

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**Abstract:** The extremely low friction and minimal wear in natural synovial joints appear to be established by effective lubrication mechanisms based on appropriate combination of articular cartilage and synovial fluid. The complex structure of cartilage composed of collagen and proteoglycan with high water content contributes to high load-carrying capacity as biphasic materials and the various constituents of synovial fluid play important roles in various lubrication mechanisms. However, the detailed differences in functions of the intact and damaged cartilage tissues, and the interaction or synergistic action of synovia constituents with articular cartilage have not yet been clarified. In this study, to examine the roles of synovia constituents and the importance of cartilage surface conditions, the changes in friction were observed in the reciprocating tests of intact and damaged articular cartilage specimens against glass plate lubricated with lubricants containing phospholipid, protein and/or hyaluronic acid as main constituents in synovial fluid. The effectiveness of lubricant constituents and the influence of cartilage surface conditions on friction are discussed. In addition, the protectiveness by synovia constituents for intact articular cartilage surfaces is evaluated.

**Keywords:** articular cartilage; synovial fluid; synovial joint; lubrication; biotribology

## 1 Introduction

In various biotribological systems, it is widely known that the healthy synovial joints maintain superior load-carrying capacity and lubricating properties with extremely low friction and minimal wear even in heavily loaded hip, knee and ankle joints. The synovial joints are prominent natural bearings different in geometric congruity depending on joint positions/movements and are in general covered with soft layers of biphasic articular cartilage lubricated with synovial fluid containing appropriate lubricating constituents. The superior tribological properties of synovial joints appear to be established by a well-suited combination

of articular cartilage and synovial fluid. However, the detailed cooperative and/or interactive behaviors between articular cartilage and synovial fluid under various rubbing conditions have not yet been clarified. In this paper, we will focus on the influence of main synovia constituents such as phospholipid, protein and hyaluronic acid on tribological behaviors of articular cartilage different in surface conditions particularly as related to lubrication mechanism.

The operating conditions in human synovial joints change under variable loading and motions including sliding and rolling depending on joint types in various daily activities. Therefore, the superior lubricating performance of natural synovial joints is likely to be actualized not by a single lubrication mode but by the synergistic combination of various modes from fluid film lubrication to boundary lubrication [1, 2].

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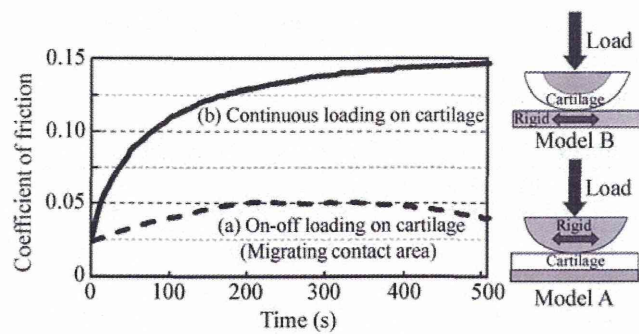
Other specific lubrication mechanisms such as weeping lubrication [3], boosted lubrication [4], biphasic lubrication [5], micro-elastohydrodynamic lubrication (micro-EHL) [6] and so on have been proposed. The ingenious lubrication mechanism as the synergistic combination of various modes depending on the severity of operating conditions was called the adaptive multimode lubrication mechanism [7, 8]. For example, during normal walking, fluid film lubrication mechanisms such as soft-EHL and/or micro-EHL play major roles to maintain low friction and minimize wear. In contrast, in thin film conditions such as at slow motion or at movement after standing for a long time, it is expected that adsorbed films [9–12], surface gel films [13], hydration lubrication [14] and polymeric brush-like layers [15, 16] contribute to keep friction low and protect rubbing surfaces.

Another new development in lubrication theory is the elucidation of the biphasic lubrication mechanism. Since an experimental finding [17] and a proposal of boundary friction model based on biphasic lubrication by Ateshian [18], the important phenomena on the effectiveness of biphasic lubrication with interstitial fluid pressurization have been demonstrated on the basis of the biphasic finite element (FE) analyses and experimental observations [19, 20]. The articular cartilage has high water content from 70% to 80% in tissue as porous media composed of type II collagen, proteoglycan and chondrocytes, and thus exhibits a time-dependent biphasic behavior due to the simultaneous coexistence of solid and liquid phases [21]. When articular cartilage as biphasic material with low permeability is applied by compressive load, the fluid content in the tissue is trapped within contact area and the collagen matrix network resists interstitial fluid pressure in aggregate solid matrix. Thus, the interstitial fluid pressure supports significant proportion of total load in contact area and this situation consequently causes the reduction of contact force of solid phase for a considerable time. The time-dependent change in load support by interstitial fluid pressure in biphasic cartilage depends on the extent of exudation from cartilage tissue and rehydration of cartilage. If the fluid load support is maintained at high level for a long time, the low friction is maintained because of low level for solid-to-solid contact [20].

For reciprocating sliding under constant load, Pawaskar et al. [22] introduced sliding motion into their FE model and indicated the importance of migrating contact area for the sustainability of the biphasic lubrication in their biphasic FE analysis. Sufficient stroke for rehydration of cartilage tissue in reciprocating motion maintained the high level of load support by interstitial fluid pressure. Sakai et al. [23] examined the compressive response of the articular cartilage by high precision testing machine with a feedback-controlled servomotor and estimated material properties in physiological condition for the biphasic FE model, which included (1) the depth-dependence of apparent Young's modulus of solid phase, (2) strain-dependent permeability as compaction effect, and (3) collagen reinforcement in tensile strain. These properties (parameters) were estimated by the curve fitting between the experimental time-dependent compressive behavior and simulation in indentation tests for cartilage specimens with cylindrical rigid indenter of 5 mm radius. In the reciprocating test, the load of 0.5 N/mm was applied at the center of the cylindrical indenter in 1 s and then the reciprocating motion was introduced with the speed of 4 mm/s over a stroke length of 8 mm. FE analyses using commercial package ABAQUS (6.8-4) showed that the tensile reinforcement by spring elements representing the collagen network and the depth-dependent elastic properties improved the proportion of the fluid load support especially in the sliding condition. The compaction effect on permeability of solid phase was functional in a condition without the migrating contact area, whereas under sliding condition the compaction effect showed a little effect in terms of the proportion of the fluid load support.

In the next stage, the influence of operating conditions on the effectiveness of biphasic lubrication in reciprocating sliding was examined. The differences in frictional behaviors between the reciprocation with migration of contact zone, i.e., at on-off loading on articular cartilage (model A) as described above, and without migration of contact zone, i.e., at continuous loading on cartilage (model B), shown in Fig. 1 were compared in FE analysis [24]. In this simulation of reciprocating test with similar method to the previous study [23], the load of 0.5 N/mm was applied by





**Fig. 1** Time-dependent frictional behaviors estimated by biphasic theory for cartilage.

the rigid cylindrical indenter against flat cartilage specimen or by the rigid flat plate against cylindrical cartilage specimen with a ramp time of 1 s and then the load was held constant during reciprocation. The reciprocation of rigid cylinder or flat plate at 4 mm/s was started immediately after loading and continued for 508 s, 127 cycles at period of 4 s. The initial fluid load support percentages are very high as 90% and 91% for models A and B, respectively. After 127 cycles, it is noticed that the high percentage of fluid load support (83%) was maintained even after 508 s in model A, but the percentage of fluid load support was remarkably decreased to 27% in the model B. The time-depending changes in friction coefficient  $\mu_{\text{eff}}$  were estimated for  $\mu_{\text{eq}}$  as coefficient of friction for solid-to-solid contact using the following formula by Ateshian et al. [20, 25].

$$\mu_{\text{eff}} = \mu_{\text{eq}} (1 - (1 - \Psi) W_p / W) \quad (1)$$

where  $W$  is the total load support,  $W_p$  the load support by fluid pressure and  $\Psi$  the fraction of contact area of solid phase.

In Fig. 1, the time-dependent changes in friction estimated from total traction force in biphasic FE analysis for assumption of  $\mu_{\text{eq}} = 0.2$  [24] are shown. It is worth noting that the lower friction level is maintained due to the sustainability of interstitial fluid pressure in the reciprocating sliding for model A. In contrast, significant gradual increase to high level in friction is observed in reciprocation for model B. It is supposed that the tribological problems are more likely to occur for model B with high friction level and thus the method to suppress friction increase is required.

In this study, the combination of cartilage-on-glass was used to simplify the frictional condition, although articular cartilage is rubbed against cartilage or meniscus in natural synovial joints. The glass plate has very smooth, hard and non-porous/impermeable surface compared with articular cartilage but hydrophilic surface with negatively charged property similar to proteoglycan on superficial cartilage layer in wet condition [12]. The adsorption of synovia constituents on glass plate appears to be considerably similar to boundary film formation on intact cartilage as shown by *in situ* observation for fluorescent images of adsorbed molecules during reciprocating rubbing process [26], while the interaction to the smooth, hard and non-porous/impermeable glass surface may induce certain different behaviors. Smooth glass surface minimizes ploughing resistance, but may enhance the adhesive resistance by interaction with adsorbed protein molecules at intimate contacts in very thin film condition. However, the intrinsic tribological properties of compliant and biphasic articular cartilage are expected to be reflected appropriately in the effectiveness of lubricant constituents even in sliding pair of articular cartilage and glass plate. As a matter of course, the difference in tribological behaviors between for cartilage-cartilage and cartilage-glass combinations should be explored. The influence of glass plate on frictional behaviors is discussed in Section 4. Thus, the frictional behaviors in a sliding pair of ellipsoidal articular cartilage specimens and reciprocating glass plate were examined in the sliding condition for model B without migration of contact zone for cartilage.

## 2 Materials and methods

The reciprocating test for the sliding pair of the upper stationary ellipsoidal articular cartilage specimen and the lower reciprocating flat glass plate was conducted in the reciprocating tester shown in Fig. 2. The continuous loading condition without migration of contact zone for articular cartilage corresponds to the severe operating condition for cartilage (model B) as described above in related to the biphasic FE analysis.

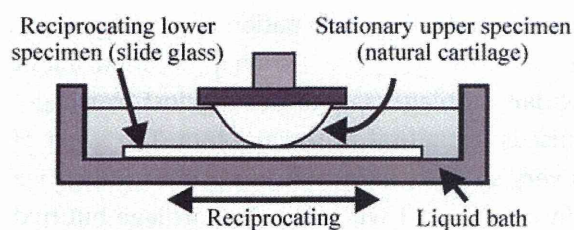


Fig. 2 Reciprocating apparatus.

## 2.1 Materials

An upper intact cartilage specimen with subchondral layer was prepared from a femoral condyle in a porcine knee joint (6 to 7 months old). The damaged cartilage specimen was prepared by wiping 15 times with a wiping tissue (Kimwipe), where the partial removal of surface gel layer was confirmed by observation with atomic force microscopy (AFM). AFM images in tapping mode (in Dimension Icon, Bruker Corporation, USA) in saline solution for intact and damaged specimens are shown in Fig. 3. On the damaged cartilage surface, the partial removal of surface gel-like layer is recognized with some exposed collagen fibers. The glass plate as a lower specimen is a slide glass.

The lubricants are saline solution containing 0.15 M NaCl (Otsuka Pharmaceutical Factory Inc., Japan), saline solution of 0.5 wt% sodium hyaluronate (HA, molecular weight:  $9.2 \times 10^5$ ), HA solutions containing 0.7 wt% or 1.4 wt% bovine serum albumin (Wako Pure Chemical Industries Ltd., Japan) and/or 0.7 wt% human serum  $\gamma$ -globulin (Wako Pure Chemical Industries Ltd., Japan) and/or 0.01 wt%  $L\alpha$ -dipalmitoyl phosphatidylcholine ( $L\alpha$ -DPPC) as an phospholipid liposome. In order to prevent bacterial growth in protein solutions as lubricant, 0.3 wt% sodium azide

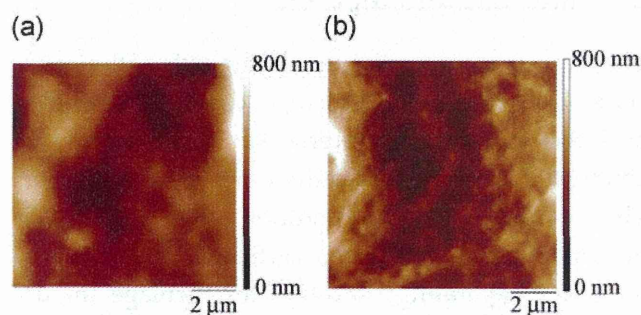


Fig. 3 AFM images of articular cartilage surfaces in saline solution: (a) intact cartilage and (b) damaged cartilage.

was added for protein solutions. The combinations of lubricant constituents used in reciprocating tests are shown in Table 1.

Table 1 Compositions of lubricants (wt%) as saline solutions.

	HA:sodium hyaluronate	DPPC	Albumin	$\gamma$ -globulin
1	0	0	0	0
2	0	0	0.7	0
3	0	0	0	0.7
4	0.5	0	0	0
5	0.5	0	0.7	0
6	0.5	0	0	0.7
7	0	0.01	0	0
8	0	0.01	0.7	0
9	0	0.01	0	0.7
10	0.5	0.01	0	0
11	0.5	0.01	0.7	0
12	0.5	0.01	0	0.7
13	0.5	0.01	1.4	0.7

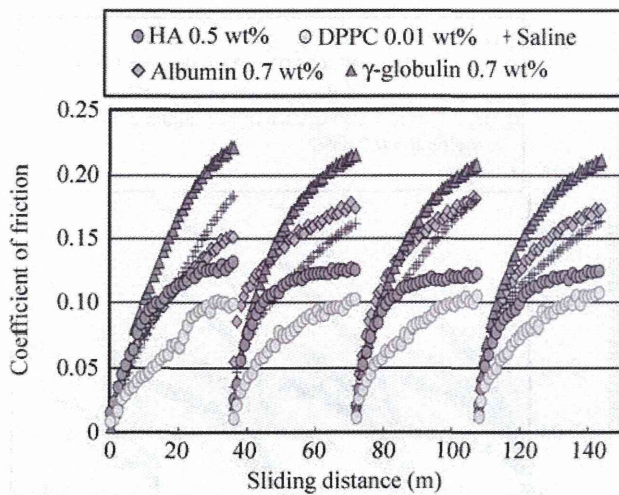
## 2.2 Experimental methods

The reciprocating test was conducted at a sliding speed of 20 mm/s for rectangular reciprocating mode and at a stroke of 35 mm at a constant load of 9.8 N. The glass plate was cleaned ultrasonically in a solution of 0.5 vol% Triton X-100, distilled water and ethanol, and then dried. The lubricants were supplied in liquid bath. At room temperature, the reciprocating sliding was started immediately after loading, and interrupted after 514 cycles at sliding distance of 36 m for running time of 30 min, and then the unloading state was maintained for 5 min. Subsequently, the reciprocating test was restarted immediately after reloading and continued for a further sliding distance 36 m. The restarting processes after unloading were repeated three times. The changes in friction force were continuously monitored to compare the differences in time-dependent frictional behaviors. The number of tests under the same condition was three.

## 3 Results

Time-dependent frictional behaviors for intact cartilage lubricated with saline, saline solutions of albumin,  $\gamma$ -globulin, HA and DPPC are shown in Fig. 4. It is noted that the initial friction is very low between 0.01





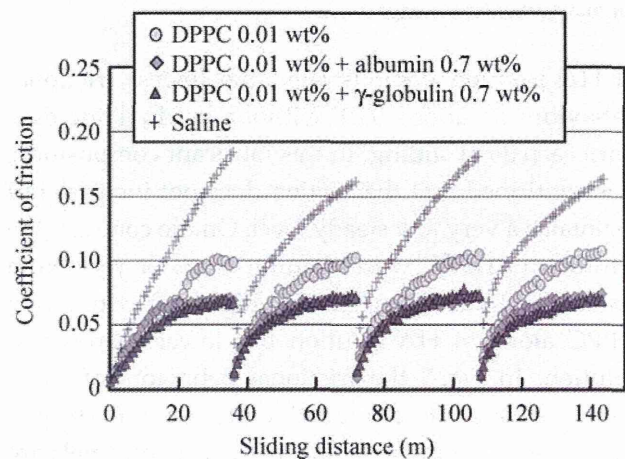
**Fig. 4** Influence of lubricant constituents on frictional behaviors for intact articular cartilage.

and 0.02 as coefficient of friction for all lubricants, as typical for intact natural articular cartilage. However, the friction gradually increases with sliding distance until the sliding stops at 36 m. The final values are different as the order of  $\gamma$ -globulin > saline > albumin > HA > DPPC. The addition of a single constituent into saline usually reduced the friction level at the final stage except  $\gamma$ -globulin. For  $\gamma$ -globulin, initial friction is lower than saline but the friction gradually increases to a higher level with thinning of lubricating film. At reloading-restarting after 5 min unloading at 36 m sliding, the restarting friction is remarkably reduced from the previous high level at interruption, but it is slightly higher than the initial friction, as reported by Murakami et al. [26, 27]. This friction reduction was considered to be brought by the recovery of both the hydration and some deformation of articular cartilage, in which the hydration lubrication and biphasic lubrication becomes partly effective accompanied with adsorbed film formation although initial adsorbed film may have been partly removed. In the second reciprocating sliding process, the friction again gradually increases with sliding distance. In the second to fourth processes where the cartilage surface was partly injured, albumin showed higher friction than the saline (Fig. 4).

The results mentioned above indicate the limitation of effectiveness of single additive for improvement of steady or final friction at each 36 m sliding. Therefore, it is required to examine the possibility in which the

combination of different synovia constituents should be effective. As reported by our previous study [26], on the reduction of final friction at each 36 m sliding, the synergistic effect of  $\gamma$ -globulin and HA was confirmed, but the coexistence of albumin and HA showed the adverse interaction for intact and damaged cartilage. It was considered that the combination of HA and  $\gamma$ -globulin form adsorbed film cooperatively, and furthermore HA as a viscosity improver is likely to alleviate the friction resistance by its viscous property to improve the fluid film formation in a mixed lubrication regime. It was pointed out for the combination of albumin and HA that the repulsive properties of negatively charged molecules prevented the lubricating adsorbed film formation. In this study, the lubricity in the combination of DPPC and albumin or  $\gamma$ -globulin was examined for intact and damaged cartilage specimens. As shown in Fig. 5 for intact articular cartilage, the coexistence of DPPC and albumin or  $\gamma$ -globulin reduced friction compared with DPPC alone in saline. In contrast, some interaction between DPPC and proteins brought increase in friction for damaged roughened cartilage surfaces with partially removed gel-like layer as shown in Fig. 6.

Next, the effectiveness of DPPC to HA solutions with and without proteins is evaluated. Figure 7 shows frictional behaviors for intact cartilage. It is noteworthy that even the addition of 0.01 wt% DPPC alone to HA solution exhibited a remarkable reduction in friction. Furthermore, the addition of 0.01 wt% DPPC accompanied with 1.4 wt% albumin, 0.7 wt%  $\gamma$ -globulin



**Fig. 5** Influence of DPPC and proteins on frictional behaviors for intact articular cartilage.