Proceedings of the Institution of Mechanical Engineers, Part J: Journal of Engineering Tribology

http://pij.sagepub.com/

Superior lubricity in articular cartilage and artificial hydrogel cartilage

eruo Murakami, Seido Yarimitsu, Kazuhiro Nakashima, Tetsuo Yamaguchi, Yoshinori Sawae, Nobuo Sakai and Atsus Suzuki

roceedings of the Institution of Mechanical Engineers, Part J: Journal of Engineering Tribology published online 16 Ap 2014

DOI: 10.1177/1350650114530273

Special Issue Article

Institution of MECHANICAL ENGINEERS

Journal of ENGINEERING TRIBOLOGY

Superior lubricity in articular cartilage and artificial hydrogel cartilage

Proc IMechE Part J:
J Engineering Tribology
0(0) 1–13

DOI: 10.1177/1350650114530273
pij.sagepub.com
SAGE

Teruo Murakami[1], Seido Yarimitsu[1], Kazuhiro Nakashima[2], Tetsuo Yamaguchi[2], Yoshinori Sawae[2], Nobuo Sakai[3] and Atsushi Suzuki[4]

Abstract

In healthy natural synovial joints, the extremely low friction and minimum wear are maintained by their superior load-carrying capacity and lubricating ability. This superior lubricating performance appears to be actualized not by single lubrication mode but by synergistic combination of multimode mechanisms such as fluid film, biphasic, hydration, gel film and/or boundary lubrication. On the contrary, in most artificial joints composed of ultra-high molecular weight polyethylene against metal or ceramic-mating material, boundary and/or mixed lubrication modes prevail and thus local direct contact brings down high friction and high-wear problems. To extend the durability of artificial joint, the reduction in friction and wear by improvement in lubrication mechanism is required as an effective design solution. In this paper, at the start, the mechanism of superior lubricity for articular cartilage is examined from the viewpoints of biphasic and boundary lubrication mechanism. Subsequently, the proposal of biomimetic artificial hydrogel cartilage is put forward to improve the lubricating modes in artificial joints. The tribological behaviours in two kinds of poly(vinyl alcohol) hydrogels are compared with that of natural cartilage. The importance in lubrication mechanism in artificial hydrogel cartilage is discussed.

Keywords

Biotribology, articular cartilage, artificial cartilage, hydrogel, biphasic lubrication, boundary lubrication

Date received: 30 November 2013; accepted: 11 March 2014

Introduction

In human musculoskeletal system with the multi-modal smooth movements, the extremely low friction and minimum wear in natural synovial joints appear to be maintained not by a single lubrication mode but by the synergistic combination of various modes from fluid-film lubrication to boundary lubrication.[1–3] As important lubrication modes correspond to the severity in daily activities, micro-elastohydrodynamic lubrication,[4] biphasic,[5,6] hydration,[7,8] boundary,[9–11] gel-film lubrication[12] and others are expected to become adaptively effective. Therefore, this lubrication mechanism is called adaptive multimode lubrication.[3,13,14] In natural synovial joints, the synergistic mechanism of articular cartilage and synovial fluid brings about effective superior lubrication. The authors focus in particular the importance of adsorbed film formation on cartilage surface and biphasic lubrication. To clarify the influence of boundary lubrication by adsorbed film, they selected severer reciprocating condition with gradual increase in friction for articular cartilage under continuous loading. In reciprocating test of ellipsoidal articular cartilage against flat glass plate including restarting test after interruption and unloading, Murakami et al.[15] showed the effectiveness of coexistence of γ-globulin and hyaluronic acid (HA) in lowering friction through adsorbed film formation compared with HA solution. In contrast, the coexistence of albumin and HA increased to higher friction level than HA solution due to the interaction of albumin and HA. Even under rubbing lubricated with HA solution containing γ-globulin, however, the friction after each 36-m sliding attained to still high level of 0.1 as

[1]Research Center for Advanced Biomechanics, Kyushu University, Fukuoka, Japan
[2]Department of Mechanical Engineering, Kyushu University, Fukuoka, Japan
[3]Department of Applied Science for Integrated System Engineering, Graduate School of Engineering, Kyushu Institute of Technology, Kitakyushu, Japan
[4]Department of Materials Science and Research Institute of Environment and Information Sciences, Yokohama National University, Yokohama, Japan

Corresponding author:
Teruo Murakami, Research Center for Advanced Biomechanics, Kyushu University, 744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan.
Email: tmura@mech.kyushu-u.ac.jp

coefficient of friction. In a subsequent study,[16] it was shown that the HA solution containing phospholipid, albumin and γ-globulin as appropriate concentrations similar to synovial fluid could maintain superior low-friction level of 0.01 as coefficient of friction and minimum wear in reciprocating test of articular cartilage. This result clarified the importance of adsorbed film formation with superior lubricity in thin-film lubrication.

On the biphasic lubrication mechanism for reciprocating cylindrical indenter on flat articular cartilage, Sakai et al.[17] estimated time-dependent changes in interstitial fluid pressure and von Mises stress in cartilage based on the biphasic finite element (FE) analysis, in which model properties for depth-dependent compressive modulus,[18] addition of spring element[19] corresponding to collagen reinforcement in tensile strain and strain-dependent permeability[19] were included. The material properties in FE model were estimated in cylindrical indentation test under wide condition including physiologically high speed. They pointed out that higher proportion of fluid load support can be sustained by biphasic lubrication mechanism in cartilage under migrating contact condition even at low-sliding speed such as 4 mm/s where hydrodynamic effect lapses. Furthermore, the improvement of fluid load support by fibre reinforcement in sliding condition was indicated. In this paper, particularly the effectiveness of biphasic lubrication in natural synovial joints is described in relation to boundary lubrication.

On the contrary, in artificial joints composed of ultra-high molecular weight polyethylene (UHMWPE) and metal or ceramic material, the mixed or boundary lubrication mode functions predominantly in daily activities, some direct contacts between rubbing surfaces occur and thus increase friction and induce considerable wear. Although the durability of over 15 years becomes established for most cases in clinical application of joint replacements for hip and knee joints, the joint loosening caused by wear debris-induced osteolysis[20,21] has required considerable patients to undergo revision surgery. Additionally, it is noteworthy that high friction on rubbing surfaces accelerates the surface failure and the joint loosening. Therefore, the prevention of wear and the reduction of friction are strongly required to improve the longevity of joint prostheses in clinical application. Although several methods such as crosslinking,[22] addition of vitamin E[23] and surface treatment by phospholipid polymer[24] to UHMWPE have considerably reduced wear and extended the life of joint prostheses, there are still unsolved problems on wear. For hard-on-hard hip prostheses such as metal-on-metal and ceramic-on-ceramic, the upgrading in material properties with high-wear resistance, better surface finish and good design improved clinical performances accompanied with beneficial fluid-film lubrication, but in certain cases, severe problems occurred as stripe wear under edge loading and/or malposition, release of metallic ions, pseudotumor, breakage of ceramic components, squeaking and so on. Besides, the lowering of friction is not always satisfactory. Therefore, alternate method to establish minimum wear and low friction in artificial joints is required.

In order to fundamentally improve the lubrication modes in artificial joints, the applications of various compliant artificial cartilage materials as cartilage replacement have been developed. The application of appropriate compliant artificial cartilage materials with properties similar to articular cartilage is expected to duplicate the superior load-carrying capacity and lubricating ability of natural synovial joints. The lowering of elastic modulus as 1–30 MPa or so as rubbing compliant materials from 800 MPa or so of UHMWPE not only can reduce the contact stress level but also can enhance the fluid-film formation due to the significant elastic deformation effect. Dowson[25] indicated the importance of compliant material properties with proposal as cushion bearing or cushion form bearing. Unsworth et al.[26] showed clear difference in frictional behaviours between UHMWPE and polyurethane acetabular surfaces in hip-joint replacements in experimental simulator tests. For hip prostheses lined with polyurethane of appropriate compliance and thickness, the fluid-film lubrication could be achieved even with low-viscosity lubricants from 0.002 to 0.071 Pa s. In the authors' previous study[27] on walking simulator tests of knee prostheses to evaluate the fluid-film formation between metallic femoral and conductive tibial components with electric resistance method, the significant fluid-film formation except slight drops at flexion stroke ends during stance phase was confirmed for silicone rubber tibial layer with elastic modulus of 9.1 MPa lubricated with silicone oil of 0.1 Pa s. In contrast, intimate contact occurred during full walking phase for anatomical knee prosthesis with UHMWPE tibial component lubricated even with high-viscosity silicone oils of 10 Pa s. This fact indicates the superiority in fluid-film formation due to soft-EHL effect with compliant silicone rubber. With decreasing of lubricant viscosity to 0.01 Pa s, however, local intimate direct contact occurred in knee prosthesis with silicone rubber tibial component. It is generally pointed out that porous hydrogels as water-swollen crosslinked hydrophilic polymers give lower friction than non-porous silicone rubber and polyurethane at start-up or after breakdown of the fluid film. The articular cartilage in natural synovial joints is considered as one of natural hydrogel containing high-water content. The application of hydrogel as cartilage replacement is expected to mimic the lubricious articular cartilage.

In 1988, Sasada[28] reported that the hip prosthesis with poly(vinyl alcohol) (PVA) hydrogel layer prepared by repeated freeze-thawing method with

85–90 wt% water content showed quite similar low-frictional behaviour to natural synovial joint, but did not attain the sufficient durability for clinical application. In contrast, Oka et al.[29] had better wear resistance in uni-directional pin-on-disc (alumina) test by applying the PVA hydrogel prepared through other synthetic process with lower water content, but wear increased in reciprocating test including thin-film conditions at stroke ends. In a subsequent study,[30] the treatment such as cross-linking by γ-radiation or high-pressure injection moulding, the mechanical properties were improved, but the combination of PVA hydrogel with low-water content against itself showed high friction. Therefore, this hydrogel was applicable as a mating material in hemiarthroplasty and artificial meniscus because of its low-friction property against articular cartilage.

Murakami et al.[31] showed in simulator test that the knee prosthesis model with PVA hydrogel layer prepared by repeated freeze-thawing method with high-water content of 79 wt% and Young's modulus of 1.1 MPa exhibited superior lower friction in walking condition lubricated with hyaluronate (HA) solution containing serum protein than for the model with polyurethane layer with elastic modulus of 40 MPa. The minimum fluid-film thickness considering elastic deformation effect for PVA hydrogel is estimated as 0.75 µm during walking, while that for the polyurethane layer exhibits thinner film than 0.2 µm. Maximum coefficient of friction for PVA hydrogel during stance phase was less than about 0.01, but that for polyurethane became higher than 0.2. The superiority of PVA hydrogel should be considered from the viewpoints of surface and biphasic properties in addition to difference in fluid-film thickness.

In reciprocating tests under thin-film condition,[32,33] even PVA hydrogel of high-water content showed significant wear in HA solutions. In reciprocating tests for PVA hydrogel against itself,[33] the addition of single protein in HA solutions increased wear of PVA with increasing concentration. However, at appropriate concentration ratio and concentration of albumin and γ-globulin in lubricants, the wear was remarkably minimized, where the layered adsorbed film was formed as shown by fluorescent images of adsorbed films after testing.[33] At high-wear condition, heterogeneous adsorbed films were formed. Subsequently, the time-dependent adsorbed film formation during reciprocation was confirmed by in situ visualization.[34] In the observation by using total internal reflection fluorescence microscopy, it was shown that the bottom layer composed of γ-globulin plays an important role in stability of mixed protein adsorbed films under rubbing condition. Furthermore, the coexistence of γ-globulin with HA enhanced the adsorption of HA on PVA and glass surfaces. The formation of adsorbed films with optimum structure is an important factor to establish longer durability of hydrogel. The improvement in strength of hydrogel is still required.

To enhance the strength of hydrogels, the chemical crosslinking treatments have been applied. Various semi-interpenetrating network (SIPN) hydrogel materials were synthesized by polymerizing a mixture of lightly crosslinked, vinyl monomers around a reinforcing polymer. Corkhill et al.[35] developed SIPN hydrogels of N-vinylpyrrolidone (NVP) and methyl methacrylate (MMA) as candidate materials with similar strength to natural articular cartilage. The friction and wear properties were evaluated in simulator and reciprocating tests[32] for flat specimens of different reinforcing polymers of polyurethane (pellethane) and cellulose acetate (CA), i.e. NVP–MMA–pellethane (Young's modulus: $E = 21$ MPa, water content: 40 wt%) and NVP–MMA–CA ($E = 23$ MPa, water content: 50 wt%). SIPN hydrogel showed higher friction and higher wear than those for freeze-thawing PVA hydrogel with high-water content. These results suggest the existence of optimum conditions of elastic modulus and water content for low friction and minimum wear. Furthermore, the importance of surface lubricity should be noticed.

Double network (DN) hydrogel composed of two kinds of hydrophilic polymers is another candidate material with higher strength but high-water content. DN hydrogel consists of the combination of stiff and brittle first network and soft and ductile second network. The hydrogel materials have elastic modulus of 0.4 to 0.9 MPa and exhibit the compressive fracture strength as high as a few to several tens of MPa, in spite of high-water content (90%). The application for hemiarthroplasty is developed as possible application.[36]

Although various kinds of hydrogel as cartilage replacement have been developed, clinical application has not yet been completed. To extend the longevity of joint prostheses with artificial hydrogel cartilage, the improvement of tribological properties of artificial cartilage materials is required under various operating conditions in daily activities.

In this study, the mechanisms with superior lubricity in both natural and artificial cartilage materials containing high-water content are explored particularly from the viewpoints of biphasic and boundary lubrication. The tribological behaviours in two kinds of PVA hydrogel materials prepared by repeated freeze–thawing method and cast-drying method were compared with natural cartilage.

Materials and methods

Materials

As stationary ellipsoidal specimens against a reciprocating flat glass plate (a slide glass) in reciprocating test, the natural articular cartilage and two kinds of artificial hydrogel cartilage specimens were prepared as follows:

Articular cartilage. The intact ellipsoidal articular cartilage specimens with subchondral layer were prepared from the femoral condyle of porcine knee joints (6 to 7 months old) (Figure 1(a)). Before testing, cartilage specimen surfaces were washed with a saline solution in order to remove any eventual residue of synovial joint fluid.

Artificial cartilage. In this study, two kinds of PVA hydrogel materials prepared by repeated freeze-thawing method and cast-drying method were used as described below.

1. PVA hydrogel by repeated freeze-thawing method (Figure 1(b)).[33,34,37]
   PVA (Kishida Chemical Co. Ltd., Japan) used in this method has the polymerization degree of 2000 and an average degree of saponification of 98.4–99.8 mol%. PVA powder at 20 wt% was dissolved in pure water and heated at 120°C for 30 min in autoclave and cast in a mould in air to control the specimen thickness and then gelled by repeated freeze-thawing (for 10 h at −20°C to for 20 h at 4°C) method. Number of freezing-thawing cycles was five times as optimum condition to establish maximum tensile strength and stiffness.[37] The elastic modulus of PVA hydrogel is 1.2 MPa, and equivalent water content is 79%.
2. PVA hydrogel by cast-drying method (Figure 1(c)).[38,39]
   PVA (Kuraray Co. Ltd., Japan) used in this method has the polymerization degree of 1700 and average degree of saponification of 98–99 mol%. An aqueous PVA solution was obtained by dissolving 15.0 wt% of the PVA powder into deionized and distilled water at 90°C for more than 2 h. The PVA solution was poured into a plastic dish of polyethylene and left in air at room temperature (ca. 25°C). The solution became very viscous within several days and solidified within a week or more. The sample was usually prepared by the process of repeated water exchange with drying.

Reciprocating tests of articular cartilage and artificial cartilage

The reciprocating tests of stationary articular cartilage and artificial hydrogel cartilage specimens were conducted against reciprocating flat glass plate.

1. Articular cartilage specimens composed of ellipsoidal condyle with subchondral bone.
2. Artificial cartilage specimens composed of a 2-mm thick soft layer of PVA hydrogel adhered on acrylic ellipsoid (40 mm × 25 mm, in diameter).

In the reciprocating apparatus,[15,16] the sliding speed and stroke length were 20 mm/s and 35 mm, respectively. Applied load was 2.94 N or 9.8 N. Sliding distance for continuous test was 36 m or 140 m. In repeated test in interval of 36 m, the reciprocating sliding was started immediately after loading and interrupted after 514 cycles at sliding distance of 36 m, 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 of 36 m. The restarting processes were repeated three times. The lubricants are saline or saline solutions containing L-α-dipalmitoyl phosphatidyl-choline (DPPC) as liposomes, serum protein (albumin and γ-globulin) and/or HA (molecular weight: 9.2×10^5).[16]

Biphasic FE analyses for articular cartilage and artificial cartilage

By applying the method of biphasic FE analysis for articular cartilage in reciprocating rubbing by Sakai et al.,[17] two-dimensional FE analysis for cylindrical articular cartilage or PVA hydrogel cartilage was conducted under continuous loading by impermeable rigid plate in reciprocating motion as shown in Figure 2.[14] The thickness of soft layer is 1.5 mm and

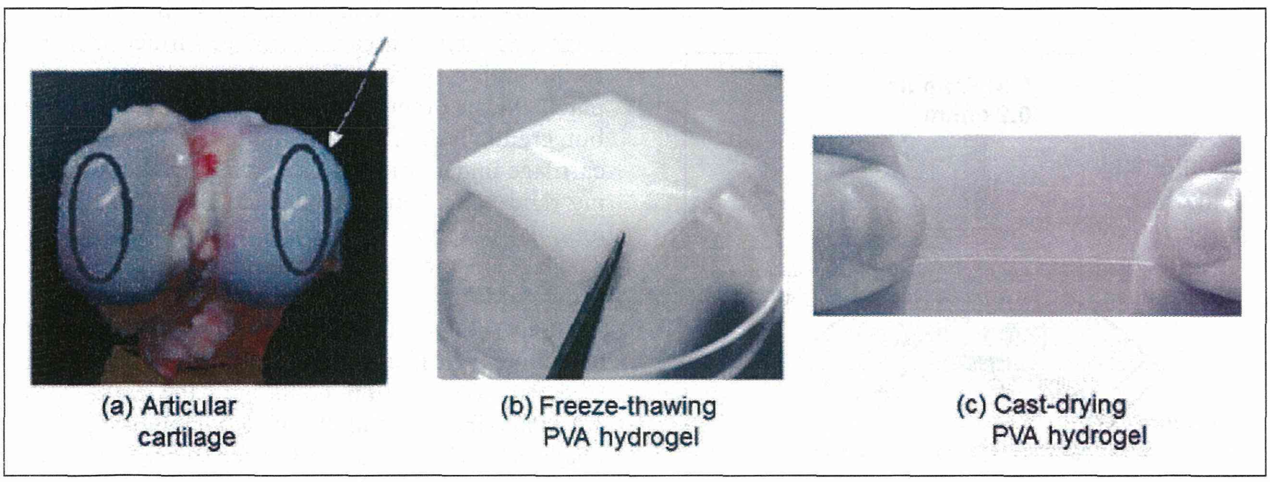

Figure 1. Articular cartilage (indicated by the arrow in (a)) and two kinds of PVA hydrogels as artificial cartilage. PVA: poly(vinyl alcohol).

its outer radius is 5 mm. A commercial package ABAQUS (6.8-4) was used in this study. The biphasic tissue was modelled by CPE4RP (four-node bilinear displacement and pore pressure, reduced integration with hour glass control) elements, and a mesh was chosen as 0.1-mm thick with 0.1 mm on surface. The bottom of the model was fixed and impermeable. The other surfaces were not fixed and basically permeable except for the contact region. The load of 0.5 N/mm (for articular cartilage) or 0.2 N/mm (for PVA) was applied at the centre of the cylindrical cartilage or PVA hydrogel surface with a ramp time of 1 s, and then the load was held constant for further 508 s (for cartilage) or 292 s (for PVA) during reciprocation motion. For stroke of 8 mm at period of 4 s, the sliding speed is 4 mm/s, where the hydrodynamic action diminishes. The changes in interstitial fluid pressure and stress in solid phase during reciprocating motion were examined by similar method to that in the previous paper for reciprocating rigid cylindrical indenter on flat cartilage plate.[17] The friction coefficient for solid-to-solid contact μ_{solid} between the geometrical rigid plate and the solid phase was set to 0.01 or 0.2.

Articular cartilage model. The biphasic FE analysis for articular cartilage was conducted by using inhomogeneous depth-dependent apparent Young's modulus of solid phase,[18] strain-dependent permeability (compaction effect)[19,40] and collagen reinforcement in tensile strain.[19] Material properties were specified by curve fitting comparing FE calculation with experimental time-dependent reaction force of the cylindrical indenter. The variables derived from the curve fitting on FE calculation were described in the previous paper.[14]

The horizontal and vertical fibrils were represented by spring element SPRINGA (axial spring between two nodes, whose line of action is the line joining the two nodes) of the software, in which the spring elements were configured to generate reaction force only in the tensile direction. The stiffness K of the spring elements was simplified to the uniform value over the tissue and both in horizontal and vertical direction.

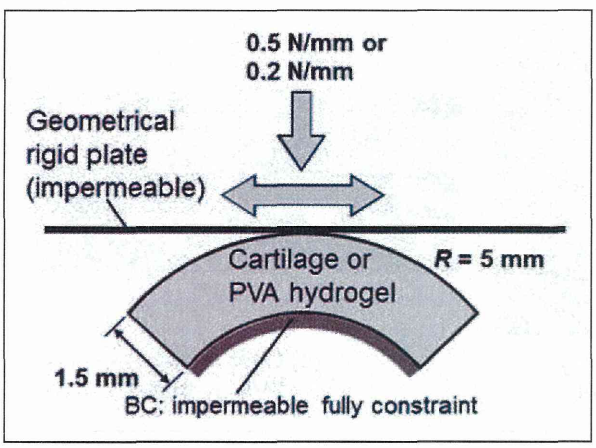

Figure 2. Two-dimensional model for biphasic FE analyses. FE: finite element.

Artificial hydrogel cartilage model. The material properties of PVA hydrogels were estimated from the stress-relaxation tests of cuboid (10 mm × 6 mm × 4 mm) under uni-axial compression at definite deformation after average stress of 0.1 MPa had been attained as indicated by Yamaguchi et al.[41] The compressive stress during stress relaxation $f(t)$ is shown as given below.

$$f(t) = \frac{3G\varepsilon_{z0}}{1 + G/3K(1 - \exp(-t/t_{\mathrm{rel}}))} \tag{1}$$

where G is the shear modulus of rigidity, K the bulk modulus of elasticity, ε_{z0} the compressive strain and t_{rel} the relaxation time. G, K and t_{rel} are estimated by fitting with this formula to stress-relaxation curve. The permeability k is estimated from the following formula

$$k = C\frac{W^2}{K\,t_{rel}} \tag{2}$$

where W is the shorter width of specimen and C the constant.

$$C = 3/16 \quad \text{for } K \ll G \tag{3}$$

$$C = 1/\pi^2 \quad \text{for } K \gg G \tag{4}$$

Results

Experimental results on frictional behaviours for articular cartilage on reciprocating glass plate

As mentioned in the previous papers[14–16] on biphasic lubrication for articular cartilage in reciprocating tests, low friction is sustained for reciprocation with migrating contact area on cartilage surface such as the reciprocation of rigid indenter on articular cartilage plate. On the other hand, the friction was low at start but gradually increased to higher level for articular cartilage under continuous loading on the reciprocating rigid plate. This time-dependent frictional behaviour is observed for friction of intact articular cartilage on the reciprocating glass plate in saline as shown in Figure 3. However, the level of final friction at 36-m sliding changes depending on the lubricant constituents.[16] If a simulated synovial fluid such as 0.5 wt% HA solution containing 0.01 wt% DPPC, 1.4 wt% albumin and 0.7 wt% γ-globulin was supplied as lubricant, very low friction was maintained until 36-m sliding (Figure 3), for which minimum wear of cartilage was observed. This lubricant contains main important constituents in natural synovial fluid. Thus, this result exhibits the superior lubricity in articular cartilage combined with synovial fluid as boundary lubricant. In this paper, the relationship between biphasic lubrication and boundary lubrication was examined on the basis of biphasic FE analysis in the following section.

Biphasic FE analysis on frictional behaviours for articular cartilage on reciprocating glass plate

The time-dependent changes in interstitial fluid pressure and von Mises stress of solid phase in simulated reciprocating tests for articular cartilage with $\mu_{solid} = 0.2$ were explained based on biphasic FE analysis in the previous paper.[14] In this paper, therefore, time-dependent behaviour of articular cartilage for $\mu_{solid} = 0.01$ was examined as shown in Figure 4. It is worth noting that at start immediately after loading, the interstitial fluid pressure is significantly high and von Mises stress is low, particularly the stress on the surface is extremely low. With further rubbing, the fluid pressure significantly subsides accompanied with further deformation at 508 s run under continuous loading. However, the stress level for $\mu_{solid} = 0.01$ (corresponding to simulated synovial fluid) is not so much high compared with the stress for $\mu_{solid} = 0.2$ (corresponding to saline).[14] The black area for fluid pressure at 508 s in Figure 4(a) corresponds to negative pressure, which facilitates the flowing in but may not contribute to the fluid load support.

Time-dependent frictional behaviours are estimated from biphasic FE analysis for two conditions for μ_{solid} as 0.01 and 0.2, as shown in Figure 5, in which the changes in partitioning of fluid load support are shown at start and at 507 s. For $\mu_{solid} = 0.2$, the fluid load support is changed from 90.5% to 27%.

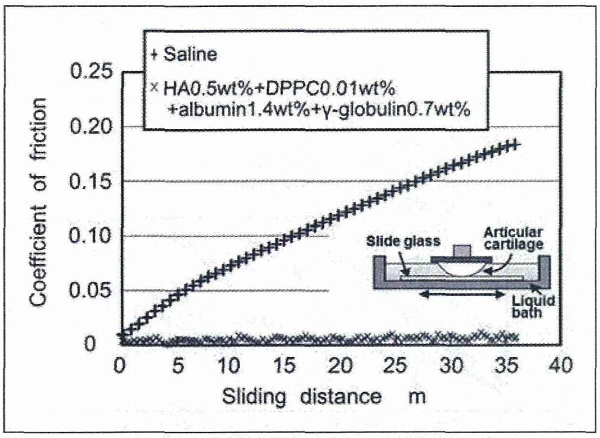

Figure 3. Influence of lubricants on changes in friction of intact articular cartilage against glass plate in reciprocating tests at 9.8 N.

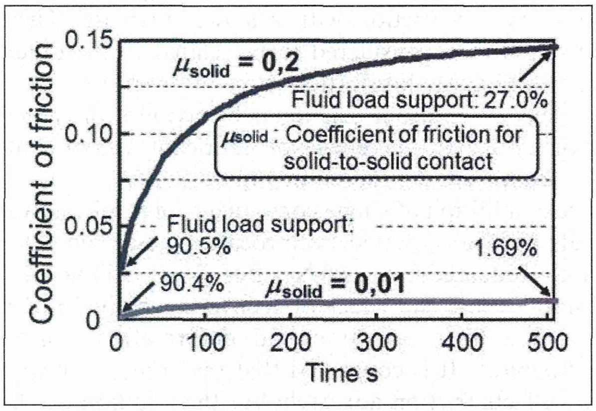

Figure 5. Influence of coefficient of friction for solid-to-solid contact on changes in friction of articular cartilage against glass plate (biphasic FE analysis). FE: finite element.

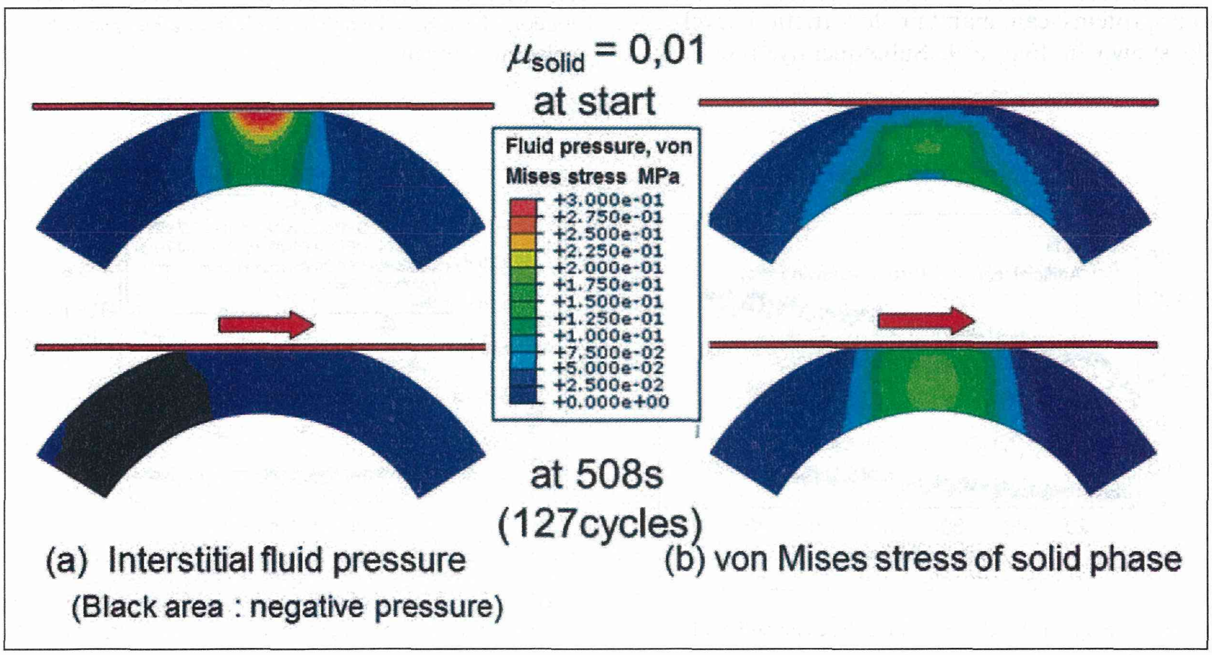

Figure 4. Changes in interstitial fluid pressure and von Mises stress in articular cartilage in reciprocating motion at low-friction condition (biphasic FE analysis). FE: finite element.

For $\mu_{solid}=0.01$, it is changed from 90.5% to 1.69%, but the coefficient of friction is sustained as about 0.01. This low friction for $\mu_{solid}=0.01$ indicates the importance of surface lubricity for low friction with boundary lubrication under continuous load, which corresponds to result for a simulated synovial fluid in Figure 3.

Experimental results on frictional behaviours for artificial cartilage on reciprocating glass plate

In reciprocating tests until 140-m sliding for articular cartilage and PVA hydrogel specimens against flat glass plate at continuous loading of 2.94 N, these specimens different in structures and properties revealed similar or different frictional behaviours in lubrication with saline as shown in Figure 6. The articular cartilage and freeze-thawing PVA hydrogel exhibited low-initial friction and gradual increase, where freeze-thawing PVA showed higher friction than that for cartilage. In contrast, cast-drying PVA exhibited very low friction with a slower increase. These differences are considered to be related to the difference in biphasic lubrication as mentioned later.

Next, to improve frictional behaviours in freeze-thawing PVA, the effect of lubricant constituents was examined. As shown in Figure 7, first, the influence of addition of single constituents in lubricant was evaluated in repeated reciprocating tests including 5 min unloading of 9.8 N after 36-m sliding and restart, where the effect of recovering of hydration, adsorbed film formation and deformation can be compared.[15] It is confirmed that the addition of protein affects friction adversely but the addition of HA and DPPC contributes to remarkable reduction in friction. Figure 8 shows the effect of HA solutions containing DPPC with and without proteins on friction. The combination of HA and DPPC with and without proteins can maintain low-friction level as clearly shown in Figure 9. Subsequently, the wear-resistance properties were examined. It is noticed that 0.5 wt% HA solution containing 0.01 wt% DPPC, 1.4 wt% albumin and 0.7 wt% γ-globulin showed minimum wear (Figure 9) similarly to articular cartilage. Thus, the reduction in both friction and wear for freeze-thawing PVA hydrogel is possible with the aid of synovia in human joint environment. However, changes in synovia constituent can happen in the diseased human body, and in such case, other lubrication mechanism becomes important as a robust system. Therefore, the role of biphasic lubrication is discussed in the next section.

Biphasic analysis on frictional behaviours for artificial cartilage on reciprocating glass plate

The mechanical properties of two PVA hydrogel specimens were estimated by the stress-relaxation test mentioned above as shown in Table 1.[41] It is noteworthy that the cast-drying hydrogel has lower permeability as two-order than freeze-thawing hydrogel.

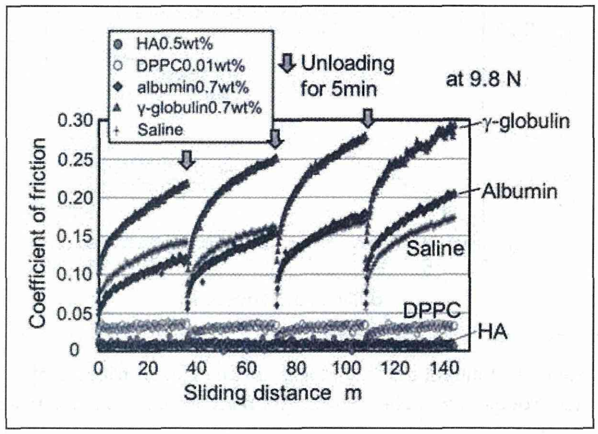

Figure 7. Influence of each synovia constituent on changes in friction of freeze-thawing PVA hydrogel against glass plate. PVA: poly(vinyl alcohol).

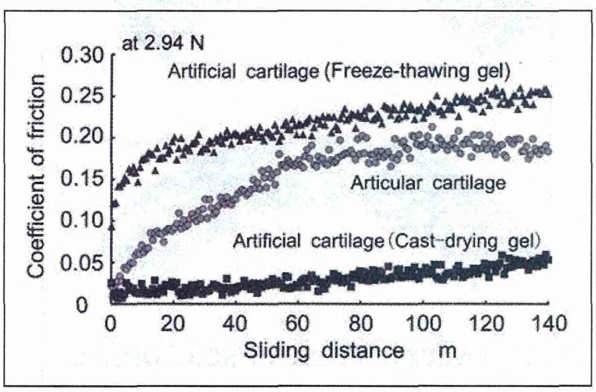

Figure 6. Changes in friction of intact articular and artificial cartilage specimens against glass plate in saline.

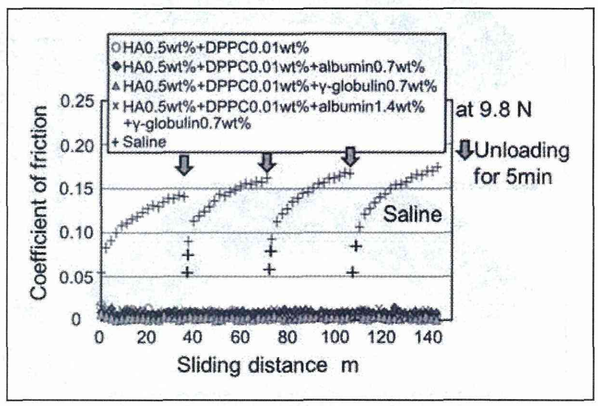

Figure 8. Influence of combination of synovia constituents on changes in friction of freeze-thawing PVA hydrogel against glass plate. PVA: poly(vinyl alcohol).

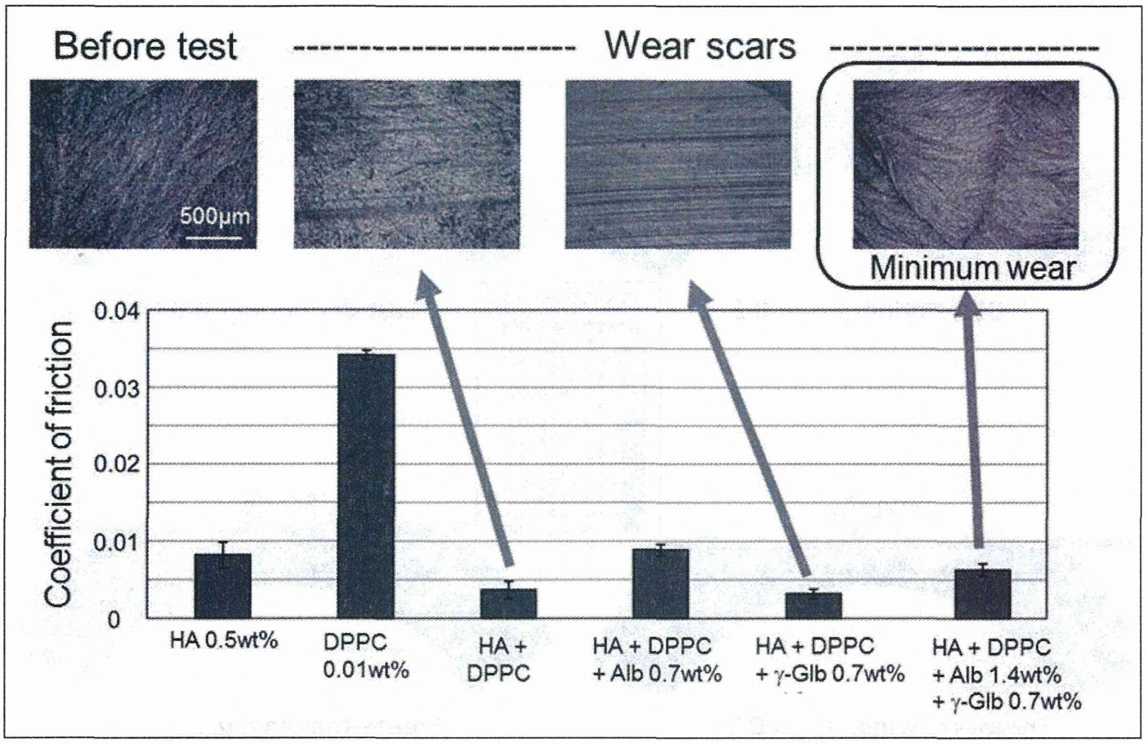

Figure 9. Influence of lubricants on average coefficient of friction after each 36 m sliding and worn surfaces of freeze-thawing PVA hydrogel in reciprocating tests at 9.8 N. PVA: poly(vinyl alcohol).

Table 1. Properties of PVA hydrogels.

Preparation method for PVA hydrogel	Permeability (m^4/Ns)	Young's modulus (kPa)	Poisson's ratio
Freeze-thawing	2.0×10^{-13}	110	0
Cast-drying	2.4×10^{-15}	190	0.41

PVA: poly(vinyl alcohol).

The permeability of cast-drying hydrogel has similar level to that of natural articular cartilage.

The interstitial fluid pressure after 292 s for two kinds of solid-to-solid friction conditions as 0.01 and 0.2 in reciprocating test is shown in Figure 10. The cast-drying PVA hydrogel maintains significant pressure even after 292 s, but in freeze-thawing PVA, the fluid pressure was remarkably reduced. For both PVA hydrogels, high friction of solid-to-solid sustained the fluid pressure levels to higher with larger deformation than those for low μ_{solid}. Therefore, caution is demanded because contrary to expectation, the lower surface friction accelerates an increase in solid-to-solid contact. The changes in load supports by fluid and solid phases are exhibited in Figure 11.

The estimated frictional behaviours for PVA hydrogels in reciprocating tests are shown in Figure 12. It is noticed that low-friction property of surfaces (solid-to-solid) maintains the sliding friction under significantly low level. Even for high μ_{solid}, cast-drying PVA can bring the gradual decreasing in friction, probably due to rising of fluid load support from 74% to 80% in biphasic analysis (Figure 11).

Discussion

In this paper, as one example of superior lubricity of articular cartilage, it was described that the addition of lubricant containing important lubricating constituents in synovial fluid can reduce friction (Figure 3) and minimize wear[16] for cartilage even under severe rubbing condition at continuous loading. Under this reciprocating condition, the role of fluid load support diminishes with rubbing (Figures 4 and 5), which means the loss of biphasic lubrication mechanism as indicated above. In natural synovial joints, it is expected that the other lubrication mechanism will alternate as an active one after weakening of biphasic lubrication effect, as suggested by the adaptive multimode lubrication mechanism.[3,13,14] The boundary lubrication based on appropriate adsorbed film formation on cartilage surface appears to become

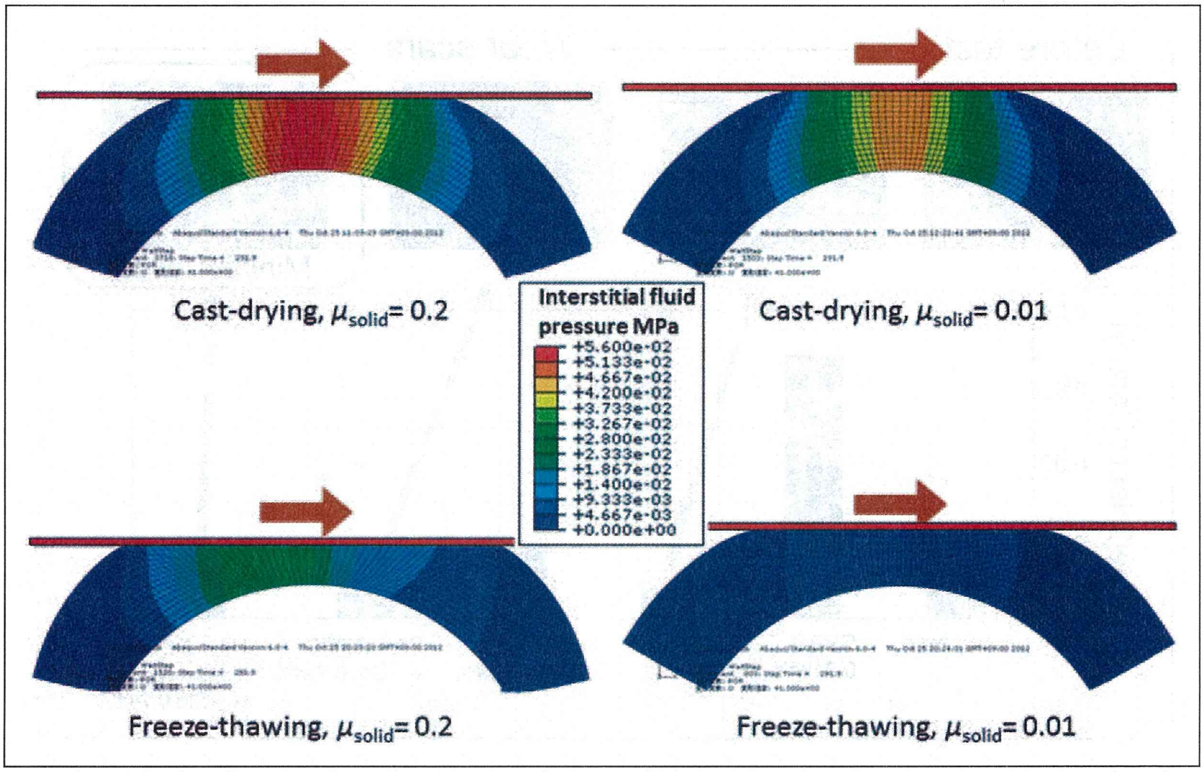

Figure 10. Interstitial fluid pressure in PVA hydrogels at 292 s in reciprocating tests (biphasic FE analysis). FE: finite element; PVA: poly(vinyl alcohol).

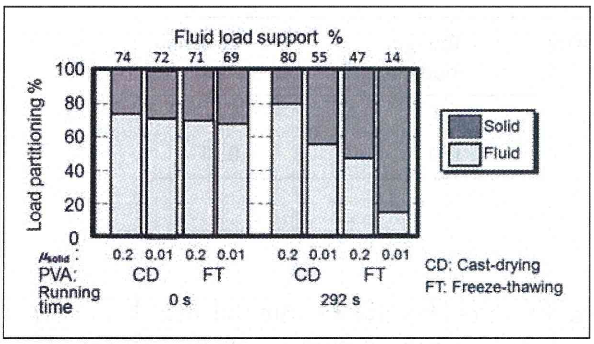

Figure 11. Changes in load support in PVA hydrogels (biphasic FE analysis). FE: finite element; PVA: poly(vinyl alcohol).

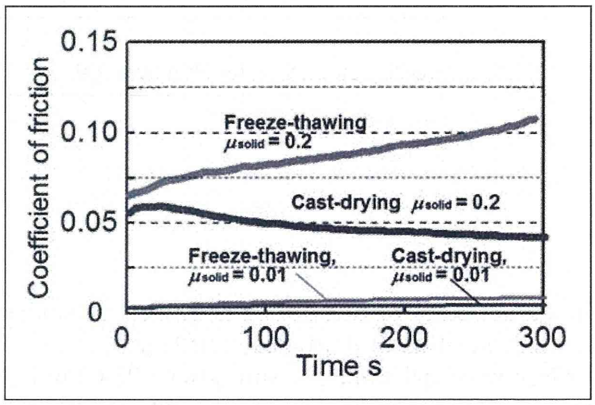

Figure 12. Changes in friction of PVA hydrogels (biphasic FE analyses). FE: finite element; PVA: poly(vinyl alcohol).

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% γ-globulin) into lubricant, which corresponds to low value of μ_{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,[42] 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