



Evaluation of a superior lubrication mechanism with biphasic hydrogels for artificial cartilage

Teruo Murakami^{a,*}, Nobuo Sakai^b, Tetsuo Yamaguchi^c, Seido Yarimitsu^a, Kazuhiro Nakashima^c, Yoshinori Sawae^c, Atsushi Suzuki^d

^a Research Center for Advanced Biomechanics, Kyushu University, 744 Motoooka, Nishi-ku, Fukuoka 819-0395, Japan

^b Department of Applied Science for Integrated System Engineering, Graduate School of Engineering, Kyushu Institute of Technology, Japan

^c Department of Mechanical Engineering, Kyushu University, Japan

^d Department of Materials Science & Research Institute of Environment and Information Sciences, Yokohama National University, Japan

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ABSTRACT

To extend the durability of artificial joints, biomimetic artificial hydrogel cartilage is proposed as a way of improving the lubrication mechanism in artificial joints. The application of hydrogels with properties similar to those of articular cartilage can be expected to duplicate the superior load-carrying capacity and lubricating ability of natural synovial joints. Frictional behaviors with three kinds of poly(vinyl alcohol) (PVA) hydrogels with high water content were examined in reciprocating tests. Interstitial fluid pressure, von Mises stress and fluid flow were compared in biphasic finite element analysis, and frictional behavior was evaluated in terms of biphasic lubrication and surface lubricity. Hybrid gel prepared by a combination of cast-drying and freeze-thawing methods showed superior low friction.

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1. Introduction

Clinical application of joint replacements to patients with severe joint diseases has brought the recovery of daily activities and the relief from serious pain. However, in certain cases, the revision operations have been conducted due to the loosening of joint prostheses which is mainly derived from wear debris-induced osteolysis [1]. Therefore, the reduction of wear is strongly required for improvement in longevity of artificial joints.

In most of the artificial joints composed of ultrahigh molecular weight polyethylene (UHMWPE) and metallic or ceramic components, the mixed or boundary lubrication mode appears to prevail because of thinner fluid film formation than surface roughness in daily activities [2,3]. To solve these problems, in this paper, the application of artificial compliant hydrogel cartilage with superior lubrication ability is proposed on the basis of bionic design concept to mimic natural articular cartilage. In natural synovial joints, various lubrication modes appear to function effectively to maintain low friction and minimum wear as synergistic coordinated action [4] in the adaptive multimode lubrication mechanism [5–8].

In order to substantially improve the lubrication modes in artificial joints, the applications of various compliant artificial cartilage

materials have been proposed as cartilage replacement, where the lowering of contact pressure due to enlargement of contact area and the increase of fluid film thickness due to soft elastohydrodynamic lubrication (EHL) and/or micro-EHL [9] effect were expected [10]. However, non-porous and hydrophobic polyurethane and silicone rubber tend to increase friction at start-up or under adverse conditions [11]. Therefore, the applications of hydrogels as water-swollen cross-linked hydrophilic polymers were investigated [12–15].

The application of hydrogel as artificial cartilage is expected to mimic the natural articular cartilage and to improve lubrication by combined effect of soft EHL, biphasic lubrication, hydration lubrication and boundary lubrication as pointed out by Murakami et al. [16]. Although various hydrogels have been developed and the tribological behaviors have been examined, poly(vinyl alcohol) (PVA) hydrogel is a prospective candidate for artificial cartilage material with biocompatibility, hydrophilic property and appropriate mechanical properties as physically or chemically cross-linked hydrogels [17–24]. So far, PVA hydrogels with high and low water content were evaluated.

Mabuchi et al. [18] and Sasada [19] showed quite low frictional behavior similar to natural synovial joint in hip prosthesis containing an artificial cartilage layer of PVA hydrogel prepared by a repeated freeze-thawing method with 85–90 wt% water content on inner surface of the acetabular cup, but it did not attain the sufficient durability for clinical application. In contrast, Oka et al. [20] prepared the PVA hydrogel with lower water content through another synthetic process and exhibited better wear resistance in

* Corresponding author. Tel.: +81 92 802 3243.

E-mail address: tmura@mech.kyushu-u.ac.jp (T. Murakami).

uni-directional pin-on-disc test, but it had a problem of increased wear in reciprocating test including thin film condition at stroke ends. In PVA hydrogels with low water content, the biphasic lubrication appears to be ineffective.

Murakami et al. [24] evaluated frictional behaviors of simplified knee prosthesis models composed of a tibial component with compliant artificial cartilage layer of 2 mm thickness and a cylindrical metallic femoral component during walking. It was found that the knee prosthesis model with PVA hydrogel (prepared by repeated freezing-thawing) with high water content of 79 wt% exhibited superior lower friction with HA solution containing serum protein than that for the model with polyurethane layer. During high loading stance phase the coefficient of friction was lower than 0.01 for PVA hydrogel but it was higher than 0.2 for polyurethane. The low friction for PVA appears to be brought due to protective adsorbed film formation in mild mixed lubrication mode.

However, in another reciprocating test of PVA hydrogel against itself lubricated with HA solution under thin film conditions at constant load, an increase in protein concentration in HA solution increased wear of PVA [25]. As it is, at certain combination of albumin and γ -globulin in lubricants, the wear was remarkably reduced, and the fluorescent images of adsorbed films after testing demonstrated the optimum adsorbed film formation as layered structure at minimum wear and low friction conditions [25]. Subsequently, the corresponding adsorbed film formation during reciprocation was confirmed by in situ visualization [26]. This fact indicates that the formation of adsorbed films with optimum structure for superior lubricity is an important factor to establish longer durability of artificial cartilage. In this low friction condition, the shearing zone with low resistance in the layered adsorbed film where protective layer covered the PVA surface appears to sustain low friction.

Blum et al. [27] indicated that boundary lubricant functionalized PVA hydrogel treated with derivative of organic fatty acid showed significantly lower friction than neat PVA hydrogel in boundary lubrication regime (at very low speed) in sliding experiment. Boundary lubricant functionalized PVA hydrogel exhibited decreases in both stiffness and friction coefficient and an increase in water content. Although they evaluated the mechanical behaviors based on a poro-elastic sliding finite element (FE) model, they proposed that friction reducing effect is due to the hydrocarbon chains pushing away from the fluid absorbed by the hydrophilic polymer. This mechanism seems to correspond to the severer condition where the load support by interstitial fluid pressure becomes infinitesimal.

Improving methods for performance of PVA hydrogel are to control the structures and properties at nanoscopic, microscopic and macroscopic levels, and thus to enhance biphasic properties for efficient lubrication and load-carrying property [16]. The PVA hydrogels based on hydrogen bonding network as physical bonding prepared by simple methods without any additives or irradiative stimulus such as the repeated freeze-thawing and cast-drying methods [28–30] have superiority in safety in human body. In this paper, the tribological behaviors of three PVA hydrogels prepared by the freeze-thawing, cast-drying and hybrid methods [31] are compared particularly from the viewpoint of biphasic lubrication. It is noteworthy that the tribological behaviors of PVA hydrogel of high water content can be controlled by biphasic properties of hydrogel in thin film lubrication under slow movement.

In natural synovial joints, articular cartilage has high water content from 70% to 80% and shows biphasic behaviors in which both solid and fluid properties should be considered as indicated by the biphasic theory [32]. Under slow movement conditions at which sliding speed is too low for hydrodynamic pressurization to sufficiently support the load, the interstitial fluid pressurization

is expected to support most of the load and reduce friction [33–35]. For reciprocating sliding under constant load, Pawaskar et al. [36] introduced sliding motion into their biphasic FE model and indicated the importance of migrating contact area for the sustainability of the biphasic lubrication. Sakai et al. [37] applied material properties of the articular cartilage at physiological high loading speed measured in high precision testing machine for the biphasic FE model. The local strain evaluated by fluorescent images of stained chondrocytes indicated that the apparent Young's modulus of solid phase changes as 20 times in depth direction. The depth-dependent elastic properties and the tensile reinforcement by spring elements representing the collagen network were effective in enhancement of interstitial fluid pressurization for articular cartilage. The compaction effect on permeability of solid phase was functional in a condition without the migrating contact. The biphasic FE analysis for reciprocating sliding of articular cartilage under continuous loading without migration exhibited time-dependent friction increase. At the next step, the biphasic FE analysis was applied for PVA hydrogels with simple material properties, and the correspondence between FE analysis and experimental measurement was confirmed [16]. In this paper, a similar method was applied for three kinds of PVA hydrogels.

For clinical application of PVA hydrogels, the biocompatibility and hydrophilic properties are preferable. PVA hydrogel is not degradable but stable in most physiological situations [38] and has considerable mechanical strength with lubricity. So far as known, PVA hydrogels have been successfully applied for cartilage replacement as repair implant for the focal chondral defects in human clinical studies [39]. It is indicated in follow-up periods of up to 2 years that isolated implants surrounded by high quality bone, a flush presentation, and about 10% radial compression improve outcome in vivo. PVA gave no apparent damage on opposing cartilage but misplacement of these implants was the major reason for dislocation and loosening.

Both the repair devices for focal chondral defects and the application as rubbing surface layer in total joint replacements or hemi-arthroplasty are considered as possible clinical applications of PVA hydrogels. In this study, the combination of PVA hydrogel surface layer and opposing flat rigid plate is evaluated in experiment and FE analysis.

2. Materials and methods

2.1. Materials

In this study, PVA hydrogels physically cross-linked with hydrogen bond were selected. Their structure and material properties are different depending on the preparation methods. As promising candidates for use as artificial cartilage, three types of PVA hydrogels were prepared via repeated freeze-thawing (FT), cast-drying (CD) and hybrid (a combination of FT and CD) methods as follows.

2.1.1. Repeated freeze-thawing (FT) method [17]

PVA (Kishida Chemical Co., Ltd., Japan) used in this method has the polymerization degree of 2000 and average degree of saponification of 98.4–99.8 mol%. PVA powder at 20 wt% was dissolved in pure water and heated at 95 °C for more than 3 h in autoclave and cast in a mold and packed to control the specimen thickness, and then gelled by the repeated freeze-thawing (for 10 h at –20 °C to for 20 h at 4 °C) method. To improve the structure and material properties of PVA hydrogel by the repetition of freeze-thawing process [21], the freeze-thawing cycles were repeated 5 times as optimum condition to establish maximum tensile strength and

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