JOURNAL OF MATERIALS SCIENCE: MATERIALS IN MEDICINE 4 (1993) 515–520

A comparison of friction in hydrogel and polyurethane materials for cushion-form joints

L. CARAVIA, D. DOWSON, J. FISHER*,
Department of Mechanical Engineering, University of Leeds, Leeds, LS2 9JT, UK
P. H. CORKHILL, B. J. TIGHE
Department of Chemical Engineering and Applied Chemistry, Aston University, Birmingham, B4 7ET, UK

The use of a thin layer of a compliant material as a bearing surface in human replacement joints has been found to promote fluid-film lubrication in the joint during continuous movement. However, during periods of continuous loading and little or no movement the fluid film may break down and the articulating surfaces may come into direct contact. This study considers the friction of three compliant materials under conditions of continuous loading and slow sliding velocities. The friction of a non-porous polyurethane and two porous hydrogel polymers, a terpolymer and a semi-interpenetrating network, was studied. Friction was measured between a hard spherical slider and a thin layer of the compliant material. Sliding was carried out in the presence of deionised water or new-born-calf serum as the lubricant. The friction was measured at the start up of motion, after prolonged periods of static loading, and during the subsequent sliding at a constant velocity. All three materials were found to give lower friction during steady-state sliding than at start up. Friction was also higher when serum was used as a lubricant. However, the porous hydrogels gave lower friction than the non-porous polyurethane, especially at start up. The terpolymer hydrogel gave the lowest friction values of the three materials, both at start up and in the steady state.

1. Introduction

The use of thin layers of compliant materials to form one or both of the bearing surfaces in total joint replacements is now being investigated in a number of research centres [1–3]. Theory predicts that, unlike existing joint designs, such cushion form joints will operate with fluid-film lubrication and this will give both low friction and low wear. However, when the fluid film breaks down, such as during periods of heavy loading with little movement or at the start of movement, there may be direct contact between the articulating surfaces, and in such conditions both the friction and wear may increase to an unacceptable level. The soft material which will form the bearing surface must therefore give low friction and have a low wear rate in conditions of mixed or boundary lubrication.

Polyurethane layers have been previously investigated for this application [1, 4] but most work has concentrated on the frictional characteristics in the fluid-film lubrication regime, under dynamic loading conditions with continuous reciprocating motion. Under fluid film lubrication conditions the coefficient of friction can be less than 0.01 and little or no wear occurs at the bearing surface [4, 5]. However, we have recently studied the tribological properties of these polyurethane layers sliding against stainless steel under conditions where we have deliberately produced a mixed or boundary lubrication regime. At start up after prolonged loading the coefficient of friction increased by over twenty times to 0.9, while under continuous sliding, at low velocities and constant loads, under a boundary or mixed lubrication regime, the friction coefficient was between 0.1 and 0.4 [6]. These levels of friction are much higher than in existing artificial joints with more rigid polymers (0.05 to 0.07 for ultra-high molecular weight polyethylene UHMWPE). In addition, the wear of polyurethane under boundary lubrication conditions has been shown to be up to two orders of magnitude higher than for UHMWPE [5]. While the compliant polyurethanes have shown remarkable tribological performance under fluid film lubrication conditions [4, 5], the higher levels of friction at start up and under boundary lubrication is a concern, and there is a need to consider other compliant materials which may give reduced levels of friction in the absence of a fluid film.

In recent years there has been some interest in the use of hydrogels as artificial articular cartilage [3, 7]. These materials have low coefficients of friction and are also attractive because of their biocompatibility. The problem in the past has been in producing hydrogels with the appropriate physical and mechanical properties to withstand the high levels of stress found

---

*Author to whom all correspondence should be addressed.

in both natural and artificial joints. Recently Corkhill *et al.* [8] have described novel materials, both hydrogel copolymers and interpenetrating networks which have enhanced properties for this demanding application.

The purpose of this study was to compare the frictional properties of two novel hydrogels with different network structures to medical-grade polyurethanes [6] in conditions of mixed or boundary lubrication. A hard spherical slider on a flat layer of the soft material was used to model the joint configuration. The effect of sliding in the presence of deionised water and bovine calf serum was investigated.

2. Materials and methods

The friction of three materials was studied: a hydrogel terpolymer (NVP–CHexMA–HexMA) of N-vinylpyrrolidone (NVP), cyclohexyl methacrylate (CHexMA), and hexylmethacrylate (HexMA), a semi-interpenetrating network (SIPN) hydrogel of NVP methyl methacrylate (MMA) and cellulose acetate (CA) and a medical-grade aliphatic poly(ether)urethane, Tecoflex 93A (Table I).

Polyurethanes have many applications in the medical field and are familiar elastomeric polymers with known properties. Tecoflex polyurethane is commercially available as pellets and these were injection moulded onto polyethersulphone substrates to yield layers 0.8 mm thick. The polyurethane was chemically bonded to the substrate during the manufacturing process.

Hydrogels are water-swollen, cross-linked, polymer networks and are unusual in that the introduction of three or more monomers can confer specific properties that are of benefit to the final product. When synthesizing the two types of hydrogel, the objective was to produce gels with a nominal elastic modulus of 20 MPa and a high water content. The mechanical properties of many typical hydrogels are inadequate for the demanding conditions in the joint. However, copolymers of NVP and CHexMA have previously been synthesized with a high water content (50%) and a nominal elastic modulus of 37 MPa [8]. The high elastic modulus was due to the introduction of the cyclohexyl ring into the polymer backbone. Terpolymer formation by the addition of hexylmethacrylate reduced the elastic modulus ($E = 20$ MPa) as the incorporation of the relatively long hexyl chain increased the freedom of rotation. This reduction in modulus was accompanied by a slight increase in water content (57%) but the hydrogel was still relatively brittle (with an elongation to break of 46%).

TABLE I. Material properties

Material	Elastic modulus (MPa)	Tensile Strength (MPa)	Elongation to break (%)
Polyurethane	20	49	423
Terpolymer hydrogel	20	4.1	46
SIPN hydrogel	23	6.3	193

Interpenetrating polymer networks (IPNs) have been described as a combination of polymers in network form at least one of which has been synthesized or cross-linked in the presence of the other [9]. Although there has been relatively little work in the field of hydrogel IPNs and related materials, it has been demonstrated that the mechanical properties of conventional hydrogels can be greatly enhanced by the synthesis of SIPNs [10, 11]. As a matrix for SIPN formation a NVP–MMA copolymer with a nominal elastic modulus of 12 MPa and a water content of 55% was chosen. Cellulose acetate was chosen as the reinforcing interpenetrant as it has been shown previously that the incorporation of stiff ring systems can improve mechanical properties [8]. SIPN formation enabled both the elastic modulus and the strain to failure of the matrix to be increased. By incorporation of 10% cellulose acetate into the system, a hydrogel with an elastic modulus of 23 MPa, a water content of 50% and an elongation to break four times that of the NVP–CHexMA–HexMA terpolymer was produced.

Hydrogel terpolymer membranes were prepared by co-polymerizing the monomers *in situ* between two glass plates clad with Melinex sheets and separated by a gasket [12]. Using this method, membranes with a thickness of 0.8 mm were fabricated. For SIPN synthesis a similar protocol was followed but the interpenetrant, cellulose acetate, was first dissolved in the solution of monomers.

Friction was measured between a spherical stainless-steel slider with a surface roughness of 0.05 μm sliding on 0.8 mm thick, nominally flat layers of polyurethane or hydrogel. All three compliant materials had elastic moduli of around 20 MPa. The contact stress between the indenter and the compliant layer was approx 2 MPa [13] and this fell well within the physiological range. Sliding was carried out at a constant velocity of 8 mm s^{-1}. This was also within the physiological range but was less than the peak velocities found in the natural joint. Hence, the fluid film generated was thinner than those likely to be found in cushion-form joints [13], and it was possible to study friction when the surface asperities penetrated the film.

Friction was measured on a sliding-friction machine (Fig. 1) adapted from a sledge microtome [14]. The compliant-material layer was fixed in position in a bath of lubricant which was driven by a motor to slide in one direction at a constant sliding speed. The slider was loaded onto the layer and was held on one end of a pivoted loading arm, while the other end of the loading arm was restrained by a piezoelectric force transducer. The sliding speed was monitored by a linear variable differential transducer. The signals from both transducers were fed into a microcomputer via an analogue-to-digital converter. The transducer signals were analysed by Unkelscope data-acquisition and-manipulation software. The software used previously determined calibration data to translate the transducer signals into values of friction force and sliding distance.

The effect on friction of the time of loading prior to sliding was investigated in order to simulate the conditions at the start of joint movement. The loaded slider

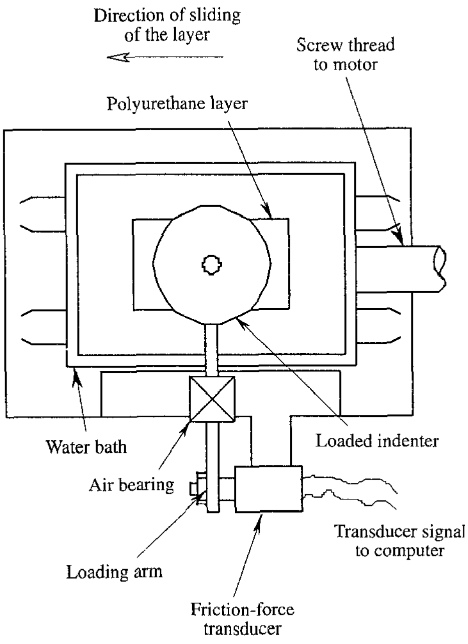

Figure 1 Sliding-friction apparatus.

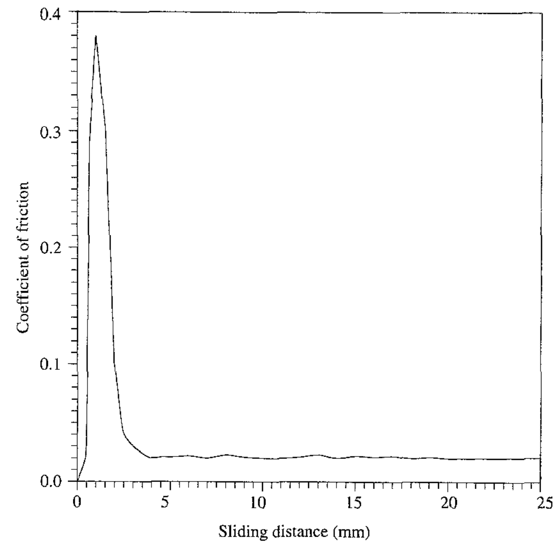

Figure 2 Typical friction trace from a single sliding test.

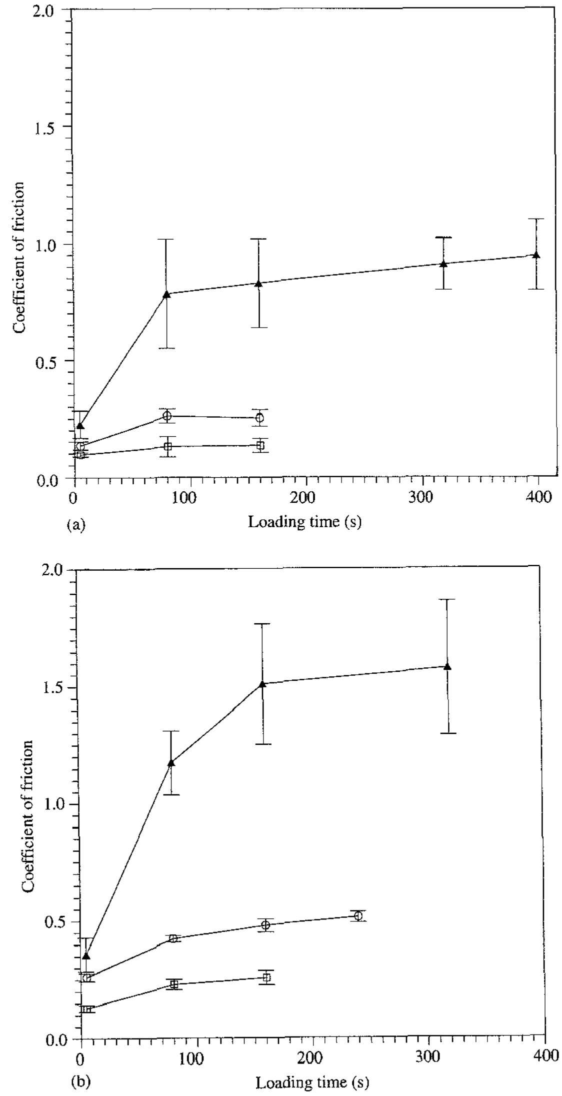

Figure 3 Start-up friction curves showing the dependence of the start-up friction on the loading time prior to sliding with (a) water and (b) bovine-calf serum as the lubricant.(▲–▲) Polyurethane, (○–○) SIPN hydrogel, and (□–□) terpolymer hydrogel.

was lowered onto the layer and a measured time period of between 5 to 400 s elapsed before sliding began. Friction was measured as sliding began (start-up friction) and continued for an average sliding distance of 30 mm (steady-state friction). Seven sliding tests were carried out for each material sliding in both fluids, and the mean friction and sample standard deviation both at start up and in the steady state were calculated. Statistical comparison was carried out using a one-way analysis of variance and a Student's t-test.

3. Results

Each friction test yielded two distinct friction values (Fig. 2). As sliding began, the friction increased rapidly to give a start up-peak. As sliding continued, the friction fell to give a much lower steady-state value.

3.1. Start-up friction

The start-up friction was found to be dependent on the duration of loading prior to sliding for all three materials (Fig. 3). As the loading period increased from 5 to 100 s, the value of the start-up friction peak increased and reached a maximum value. Increasing the length of the loading period to more than 100 s had little further effect on the maximum start-up friction value. The maximum start-up friction value obtained for the polyurethane when sliding in water was $\mu = 0.95$, and when sliding in serum was $\mu = 1.5$. The hydrogels gave significantly lower values of start-up friction, typically 0.1–0.3 in water and 0.1–0.5 in serum (probability of chance occurrence $p < 0.001$ for all values), and the friction continued to increase after 200 s of constant loading, although at a slower rate than in the initial 100 s. The terpolymer gave the lowest maximum start-up friction values of $\mu = 0.13$ in water and $\mu = 0.26$ in serum. The SIPN hydrogel gave values of 0.26 in water and 0.52 in serum which were higher than the terpolymer ($p < 0.001$) but still considerably lower than for the polyurethane.

The use of serum instead of deionised water as the lubricant caused the maximum value for start-up

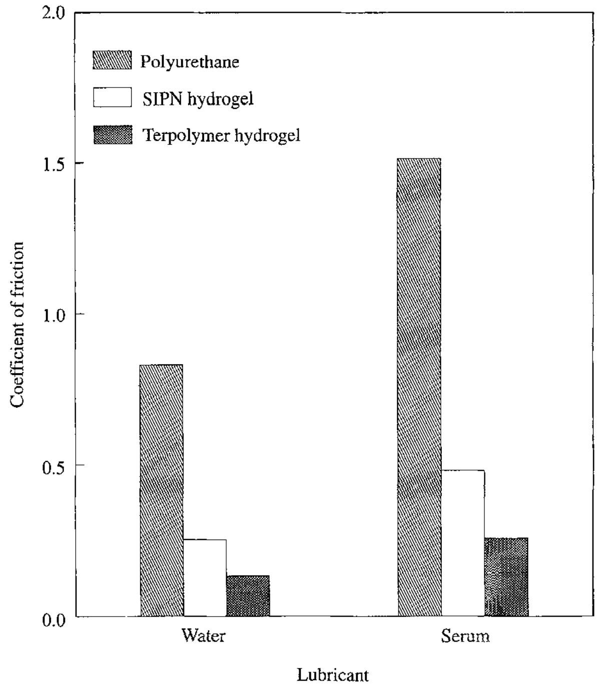

Figure 4 The effect of the lubricant on the maximum start-up friction (after 100 s of loading).

friction to increase ($p < 0.001$ for all values) for all three materials tested (Fig. 4).

3.2. Steady state friction

The steady-state sliding friction was much lower than the start-up friction and was independent of the loading period prior to sliding (Fig. 5).

The polyurethane gave the highest steady-state friction values with both water ($\mu = 0.10$) and with serum ($\mu = 0.15$) as the lubricant. The hydrogel terpolymer gave the lowest friction with coefficients of friction of 0.02 when sliding in water and 0.038 when sliding in serum. For both the polyurethane and terpolymer hydrogel, sliding in serum caused a slight increase in the recorded friction values ($p < 0.001$) as shown in Fig. 6.

The SIPN hydrogel gave friction values slightly higher than the terpolymer ($p < 0.001$) with coefficients of friction of 0.09 when sliding in water and 0.05 when sliding in serum. Unlike the polyurethane and the terpolymer the SIPN showed a reduction in the steady-state friction when sliding in serum.

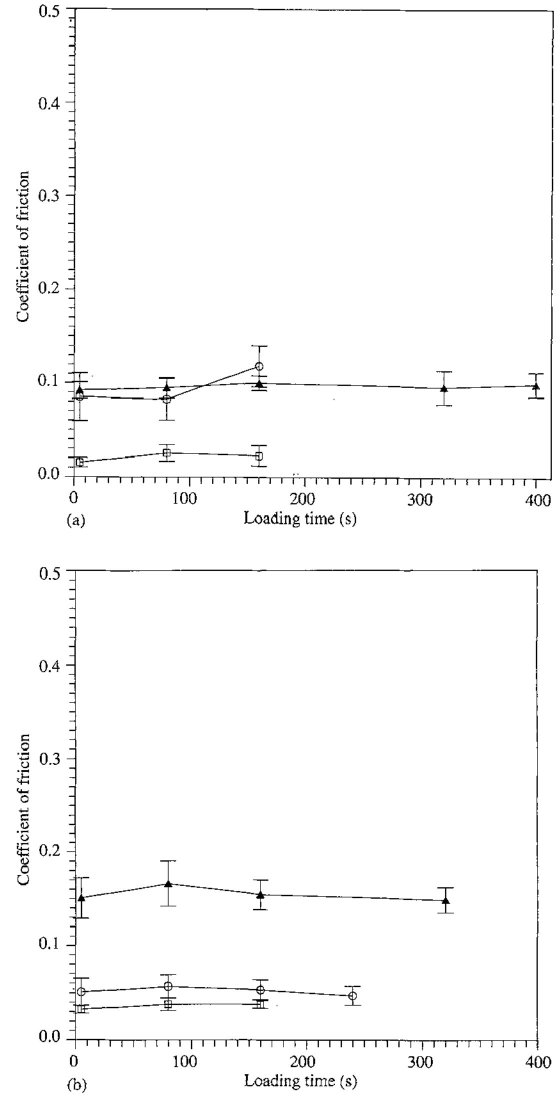

Figure 5 Steady-state friction curves when sliding with (a) water and (b) bovine-calf serum as the lubricant. (▲–▲) Polyurethane, (○–○) SIPN, and (□–□) terpolymer hydrogel.

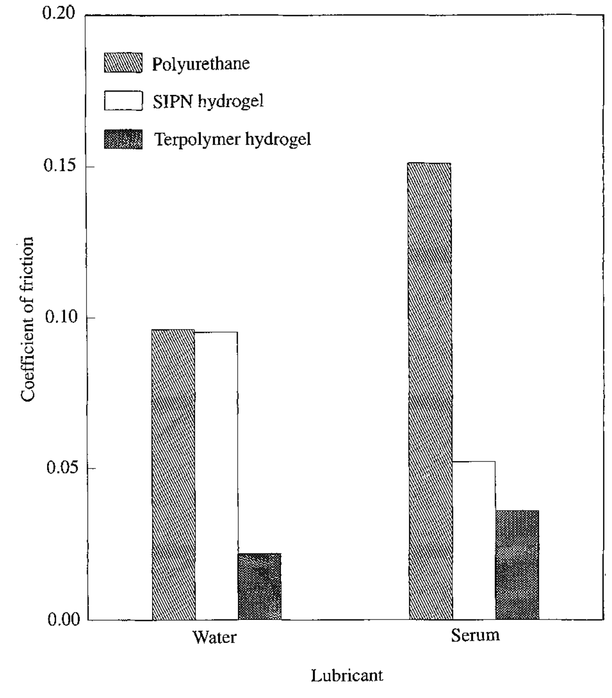

Figure 6 The effect of the lubricant on the steady-state sliding friction.

4. Discussion

The use of compliant materials as bearing surfaces in artificial joints has been shown to be beneficial in reducing friction and wear in laboratory tests [4] when operating under continuous cyclic motion and loading and fluid-film lubrication. It is clear from this study and our previous work [6], however, that such materials can give very high friction when the fluid film breaks down. Hydrogel polymers with a high water content may maintain low friction by releasing water from the polymer network which is then available to form a film and, in addition, as their water content is high, they may have reduced levels of friction when direct surface contact occurs.

The friction results show that as sliding began the friction was initially high but continued motion caused the friction to fall rapidly to give a relatively low value. The initial high friction values recorded in these tests at start up indicated that there was a substantial amount of contact between the slider and the compliant layer. As the loading time prior to sliding was increased, more fluid was squeezed out of the conjunction, there was more surface-to-surface contact and the friction was correspondingly higher. As sliding continued, the friction fell rapidly, due partly to the fact that new material was being deformed and partly to elastohydrodynamic entraining action drawing fluid into the conjunction. In the case of the hydrogels in particular, movement of the contact onto new material could have proved beneficial since the slider was moving onto a part of the polymer network from which the fluid had not been squeezed out.

4.1. Start-up friction

The polyurethane gave consistently higher friction than either of the hydrogels. In particular, very high maximum friction values of $\mu = 1.0$ or higher were recorded at start up for the polyurethanes compared to values of mostly less than 0.5 for the hydrogels. The very high friction values at start up shown by the polyurethane (which approached the dry-contact friction value) indicated that after the first 100 s or so of sliding much of the fluid had been squeezed out of the conjunction and a large amount of surface-to-surface contact was occurring. The friction values for the hydrogels at start up were still too high to indicate full fluid-film lubrication, but it was clear that a much smaller amount of surface contact was occurring than for the polyurethane and that fluid was still present in the conjunction. This large difference in the start-up friction values between the polyurethane and the hydrogels showed clearly that a high-water-content, porous, polymer structure bestows a distinct advantage in reducing friction in the absence of a continuous fluid film. The hydrogels had water contents of 57% for the terpolymer and 50% for the SIPN, and a proportion of this water was free to move within the polymer network. It is not clear from this experimental study whether the porosity of the hydrogel materials and their high water content will result in the fluid film being enhanced through fluid exudation on the application of load, as is suggested by the biphasic theory for porous materials such as cartilage [15], or whether the porosity will result in the fluid film being depleted [16]. However, it is clear that when fluid-film lubrication breaks down and direct contact between surfaces occurs, the porous hydrogels have greatly reduced values of friction compared to the polyurethane. The different surface energies of the three materials may also affect the strength of the layer/slider attraction forces and thus the friction. However, the presence of a fluid in the test means that surface-energy effects are largely attenuated [17]. It was interesting to note that the coefficient of friction of the hydrogel at start up was close to that reported for cartilage under very slow sliding and boundary lubrication [18].

4.2. Steady-state friction

In the steady state the friction values were much lower than at start up. The difference in friction between the three materials appeared less marked than at start up. However, these very low values of friction were close to the resolution of the friction measurement apparatus of $\mu \pm 0.015$. (This resolution of the friction measurement was limited by the large range of friction force which was measured during a single friction test, from as high as $\mu = 1.0$ or more at start up down to $\mu = 0.02$ in the steady state.) It was clear, however, that the friction during steady-state sliding was about an order of magnitude smaller than the maximum start-up friction and that the hydrogels generally exhibited lower friction than the polyurethane.

Serum contains long chain molecules which can normally act as boundary lubricants. The results show, however, that the friction for the polyurethane and the terpolymer hydrogel increased when sliding was carried out in the presence of serum rather than water. The calculated steady-state fluid-film thickness for these materials sliding under the test conditions was 0.05 μm; this was similar to the surface roughness of the metal slider and so was not sufficient to completely separate the surfaces. The friction values recorded were generally much higher than that expected for full fluid-film lubrication and, hence, were consistent with the theoretical predictions that a mixed lubrication regime would occur. However, the fact that the friction in the steady state was so much lower than at start up, indicates that fluid was causing greater separation of the slider and layer surfaces than at start up (mixed lubrication was occurring) and so the applied load was being partly supported by a fluid film. In such conditions, where fluid did separate parts of the two surfaces, the fluid film was extremely thin. Since the film was so thin, when serum was present the long molecule chains in the serum could become entangled in the contact area between the slider and the layer resulting in higher friction values. Serum has also been shown to give higher friction in hard polymer contacts [19]. It must be noted, however, that the friction measurements during steady-state sliding with the SIPN hydrogel were slightly contradictory to the other tests in that the friction with serum was lower than with water as a lubricant. There is no doubt that the complex surface interactions between porous materials and boundary lubricants require further study.

5. Conclusion

The friction results showed that high-water-content porous materials such as hydrogels can significantly reduce friction in compliant layered contacts compared to polyurethane under mixed or boundary lubrication conditions. This is especially important in the crucial moments of start-up of motion after a period of static loading. The terpolymer hydrogel gave the lowest friction but its mechanical properties may not be suitable for use in artificial joints. The SIPN, on the other hand, had mechanical properties which began to approach those required for this application whilst achieving much better frictional characteristics than the polyurethane.

It is not yet clear which elements of the hydrogel composition are important in determining good frictional properties, whether it is the monomers which make up the gel, the type of network or its water-releasing capabilities or a combination of these different elements. However, a hydrogel composition which gives low friction may not necessarily give good wear and durability properties, and a compromise composition may thus be required. The synthesis of SIPNs has improved the mechanical properties of hydrogels, but further work is required in order to determine hydrogel compositions for the demanding conditions in joint replacements.

Acknowledgements

This work was supported by the Wellcome Trust, the SERC, the ARC and the SmithKline Foundation.

References

1. A. UNSWORTH, M. J. PEARCY, E. F. T. WHITE and G. WHITE, *Engng. Med.* **17** (1988) 101.
2. D. D. AUGER, J. B. MEDLEY, J. FISHER and D. DOWSON, Proceedings of the 16th Leeds–Lyon Symposium on Tribology, Leeds 1992, "Mechanics of Coatings", edited by D. Dowson, C. M. Taylor and M. Godet (Elsevier, Amsterdam, 1990) p. 264.
3. M. OKA, T. NOGUCHI, P. KUMAR, K. IKEUCHI, T. YAMAMURO, S. H. HYON and Y. IKADA, *Clin. Mater.* **6** (1990) 361.
4. D. D. AUGER, D. DOWSON and J. FISHER, Proceedings of the 8th Meeting of European Society of Biomechanics, Rome. (1992), p.115.
5. Z. M. JIN, D. DOWSON and J. FISHER, *Wear* **162** (1993) 627.
6. L. CARAVIA, D. DOWSON and J. FISHER, *ibid.*, **160** (1993) 191.
7. J. C. BRAY and E. W. MERRILL, *J. Biomed. Mater. Res.* **7** (1973) 431.
8. P. H. CORKHILL, A. S. TREVETT and B. J. TIGHE, *Proc. Instn. Mech. Engs., Part H: J. Engng. Med.* **204** (1990) 147.
9. L. H. SPERLING, "Interpenetrating polymer networks and related materials (Plenum Press, New York, 1981).
10. P. H. CORKHILL and B. J. TIGHE, *Polymer* **31** (1990) 1526.
11. *Idem., J. Mater. Chem.* **2** (1992) 491.
12. P. H. CORKHILL, A. M. JOLLY, C. O. NG and B. J. TIGHE, *Polymer* **28** (1987) 1757.
13. D. DOWSON, J. FISHER, Z. M. JIN, D. D. AUGER and B. JOBBINS, *Proc. Instn. Mech. Engrs., Part H: J. Engng. Med.* **205** (1991) 59.
14. P. S. WALKER, D. DOWSON, M. D. LONGFIELD and V. WRIGHT, *Rheologica Acta*, **8** (1969) 234.
15. V. C. MOW, M. H. HOLMES, and W. M. LAI, *Trans. Orthop. Res. Soc.* **8** (1984) 262.
16. Z. M. JIN, D. DOWSON and J. FISHER, *Proc. Instn. Mech. Engrs., Part H: J. Engng. Med.* **206** (1992) 1171.
17. L. H. LEE, in "Advances in polymer friction and wear" (edited by L. H. Lee) (Plenum Press, New York, 1974) p. 31.
18. S. E. CLIFT, B. HARRIS, P. A. DIEPPE and A. HAYES, *Biomaterials* **10** (1989) 329.
19. P. KUMAR, M. OKA, K. IKEUCHI, K. SHIMIZU, T. YAMAMURO, H. OKUMURA, and Y. KOTOURA, *J. Biomedical Mater. Res.* **25** (1991) 813.

Received 18 December 1992
and accepted 12 January 1993