Mechanical wear behavior of nanocrystalline and multilayer diamond coatings on temporomandibular joint implants

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We used microwave plasma chemical vapor deposition to deposit nanocrystalline and multilayer (nanocrystalline/microcrystalline/nanocrystalline) diamond thin films on Ti–6Al–4V substrates imitating the condyle and fossa components of the temporomandibular joint. We tested the condyle/fossa pairs for wear in a mandibular movement simulator for an equivalent of two years of clinical use. Analysis of the wear surfaces by optical microscopy, scanning electron microscopy (SEM), and Raman spectroscopy showed that damage in both the films was minimal, no loss of film occurred and the wear performance was superior for the multilayer film. Comparisons with an uncoated condyle/fossa pair showed that the coated temporomandibular joint pairs had improved wear performance.

1. Introduction

Temporomandibular disease is used to describe a variety of different disorders that affect the function and can lead to significant pain in the temporomandibular joint (TMJ) in humans [1]. The human TMJ is located in that part of the body that requires high performance from prostheses used to replace it. The poor quality of a TMJ implant can lead to an array of adverse bodily reactions that include inability to chew, osteolysis, and in some extreme cases, even blindness [2]. The main reason for this has been attributed to wear debris generation from the joint replacement. Consequently, an attempt was made to fabricate TMJ implants that have high wear resistance and longer service lifetimes by coating them with a nanocrystalline diamond (NCD) thin film [2–4]. Accomplished work towards this effort indicated that an NCD thin film could be successfully coated on a curved surface that resembles the geometry of the condylar bone of the TMJ [3]. The condylar simulants that were produced were then tested for wear under conditions that approximated a pin-on-disk. This testing condition represents a rather extreme evaluation for wear for this joint since the mode of articulation between the condylar bone and the fossa very rarely resembles a pin-on-disk motion. In this work, we report results of a study of the wear characteristics of condylar simulants that articulate against curved surfaces that resemble a portion of the fossa eminence bone. In addition, we also assess the wear performance of a nanocrystalline and a three-layer (nanocrystalline/microcrystalline/nanocrystalline) thin film that was previously fabricated but never evaluated for wear performance [5].

2. Experimental procedures

The condyle simulants were machined from a Ti–6Al–4V rod according to the dimensions measured from a condyle prosthesis explanted from a patient. A photograph of a condyle used in this study with its dimensions is shown in Fig. 1. The fossa simulants were machined from a Ti–6Al–4V plate. The present research is focussed on studying the wear behavior of NCD coating on a TMJ device using a mandibular movement simulator. The radius of the concave-curved portion of the fossa relative to the convex-curved area of the condyle was set at $R(\text{fossa}) = 11\,R(\text{condyle})$. A picture of this prosthesis simulant is also shown in Fig. 1.

We used Wavemat 6 kW microwave plasma chemical vapor deposition reactor to deposit thin films on polished condyle and fossa substrates. The root mean square (rms) value of roughness of these two types of substrates was estimated to lie in the range of 5–20 nm [2]. The gas flow rates we used for deposition are 500 sccm of H_2, 88 sccm of CH_4, and 8.8 sccm of N_2. This combination of gases results in 15% CH_4 fraction in H_2 and an N_2/CH_4 ratio of 0.10 [4]. We performed deposition at a chamber pressure

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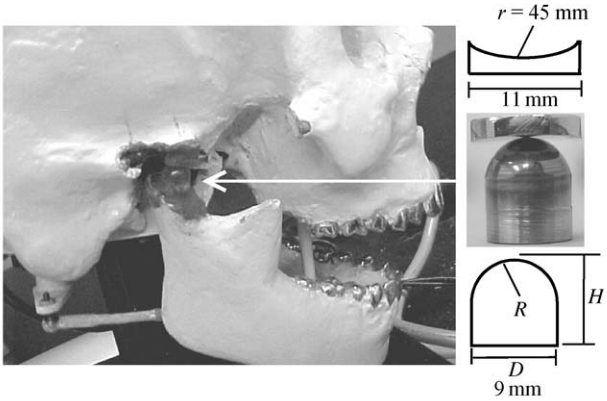

Figure 1 A picture of a custom-made mandibular simulator showing the mounting area for the coated condyle/fossa pair. The condyle dimensions are $R = 4.3$ mm, $H = 11$ mm, and $D = 9$ mm. The dimensions of the fossa are shown in the schematic diagram.

of 35 torr and a microwave power of 0.75 kW. The substrate temperature (T_S) before the start of deposition was maintained at 700 ± 5 °C at the condyle tip and the center of the fossa. We deposited two types of thin films on both the condyles and the fossas. In the first set of simulants, we deposited a single-layer nanocrystalline thin film with a thickness of approximately 3 μm for condyles, and approximately 2.25 μm for fossa substrates. In the second set, we continuously deposited a multilayer thin film that comprised of a nanocrystalline first layer, followed by a microcrystalline second layer, and finally another nanocrystalline layer at the surface. We achieved a multilayer thin film configuration by putting N_2 on for a third of the deposition time required to grow a desired film thickness (i.e. 3 μm and 2.25 μm for condyles and fossas, respectively), and having N_2 off for another third of the time in order to grow a microcrystalline layer, and having N_2 on again for a final third of the deposition time. We maintained the same thickness for both types of thin films by maintaining the same number of oscillations in the apparent sample temperature (T_{app}) recorded with a Mikron M-77S two-color optical pyrometer [6]. We performed micro-Raman spectroscopy using a Dilor XY modular spectrometer with a 514.2 nm Ar^+ laser operated at 300 mW output power. The laser beam passes through a microscope and is focussed on the target area using a 100 × objective lens. We performed X-ray diffraction (XRD) using a Phillips diffractometer at an incident beam angle of 5°. We used a Tencor Alpha-Step 500 diamond stylus profilometer to measure the depth of the wear tracks after testing.

We used two coated (i.e. single-layer and multilayer coated pairs) and an uncoated condyle/fossa pairs to evaluate wear properties after articulation in a custom-built mandibular movement simulator (MMS), shown in Fig. 1. This device reproduces a typical mandibular motion of a human jaw and in this set of experiments it simulates wear under cyclic chewing motion at a frequency of 1.2 Hz. For each test, the condyle/fossa pair was loaded in the simulator using wax. Thereafter, the wax was coated with a resin compound that hardens to firmly hold the pair in the required testing position. The fossa position was maintained at an angle of eminence of ≈ 25° for all tests. The condyle was mounted in such a way that it made full contact with the fossa before testing began. Thereafter, a low pressure (LW) Prescale Fuji Film was used to determine the pressure at the condyle/fossa point of contact after articulation for 3–6 cycles. The articulation load was approximated to be around 100 N. Each wear test was performed for 2.25×10^5 loaded cycles in air at ambient temperature. This number of cycles is equivalent to approximately two years of clinical use.

3. Results and discussions

Fig. 2 shows that we successfully deposited a single-layer nanocrystalline and a multilayer diamond thin film. In Fig. 2(b), the three layers that make up the multilayer are clearly visible and the middle layer, that is microcrystalline in character [5], appears to have more of a columnar morphology when compared with the two-nanocrystalline layers that bound it. The nanoindentation measurements we performed on the fossa coated with a single-layer nanocrystalline film at an indentation depth of 100 nm from the surface, showed that the film had a hardness of 56 ± 10 GPa and a modulus of 601 ± 10 GPa. Although we did not perform the same test for the multilayer film, we do not expect significant differences in these measured properties between these two types of films since the top layer of the multilayer film is nanocrystalline in character and has an approximate thickness of 1 μm. XRD spectra in Fig. 3(a) show

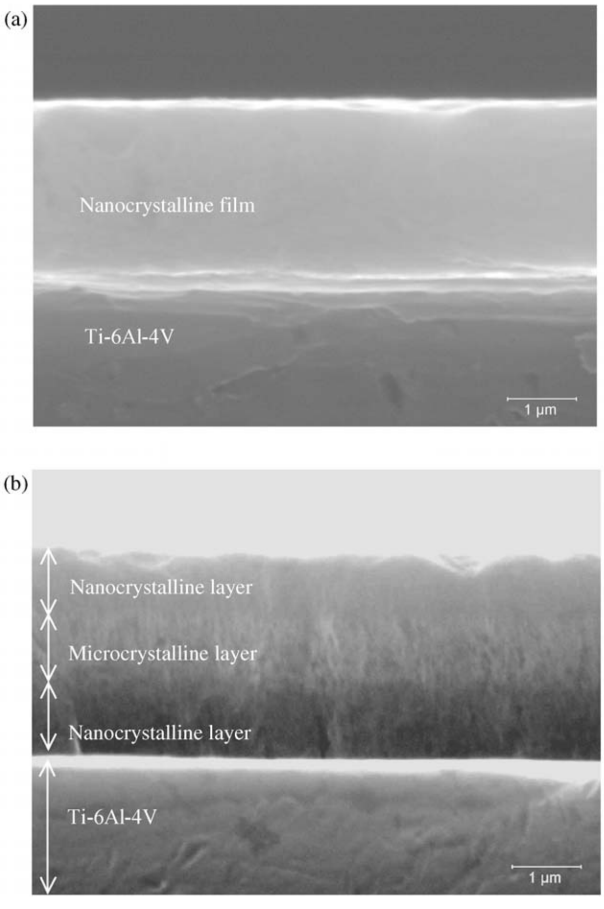

Figure 2 Cross-section of SEM micrographs of (a) a nanocrystalline diamond film coated fossa and (b) a nanocrystalline/microcrystalline/nanocrystalline multilayer film, deposited on a fossa.

that both films have a crystalline diamond component, as indicated by the (1 1 1) diffraction peak. However, all the diffraction peaks from the diamond phase are broadened considerably as compared to Ti-alloy substrate indicating the nanocrystalline nature of the coating. Fig. 3(b) shows the Raman spectra confirming that the diamond component in both films is nanocrystalline in character, as indicated the broad diamond peak at $\approx 1350\,\text{cm}^{-1}$ and a weak band ca. $1150\,\text{cm}^{-1}$. Both films also show broad G-Band peaks at approximately $1560\,\text{cm}^{-1}$ that is indicative of the presence of amorphous carbon [5].

Fig. 4 shows the wear surfaces on the multilayer-coated condyle/fossa pair. The condyle wear area in Fig. 4(a) has a near-elliptical shape and it appears to have polished-diamond morphology when compared with its rougher surrounding. Apart from what appears to be a film crack at the edge of the wear area, as seen in the inserted high magnification micrograph, there does not appear to be extensive damage of the film. The same polished-diamond morphology that was seen on the condyle (Fig. 4(a)) is also visible on the fossa, as seen in Fig. 4(b). This observation is clearly shown in the inserted high magnification micrograph taken at the boundary of the worn and as-deposited portion of the film.

There is no evidence of film cracking on the nanocrystalline film coated condyle, as shown in Fig. 5(a). We still see a polished-diamond wear area

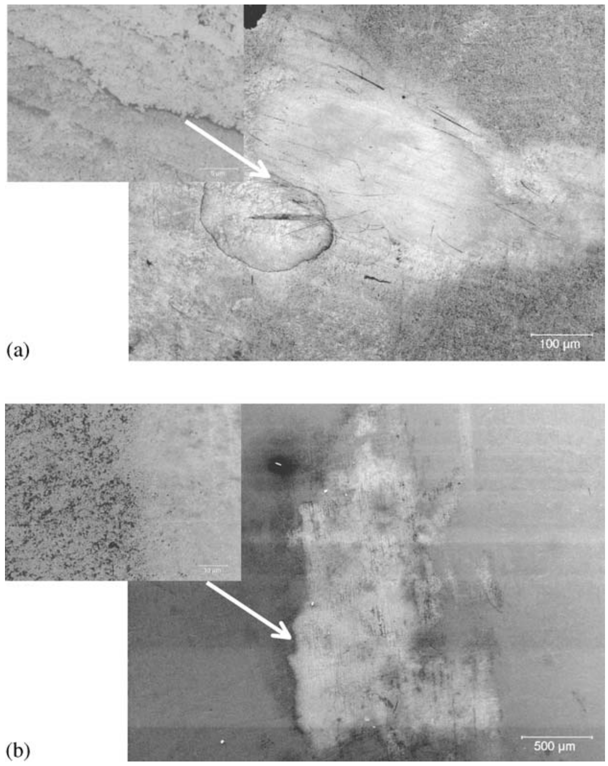

Figure 4 (a) An SEM micrographs of the multilayer condyle wear area. The high magnification micrograph at the edge of the wear area shows evidence of what appears to be thin film cracking. (b) An SEM micrograph of the multilayer fossa wear area. The high magnification micrograph shows the difference in the film texture between the as-coated and worn areas.

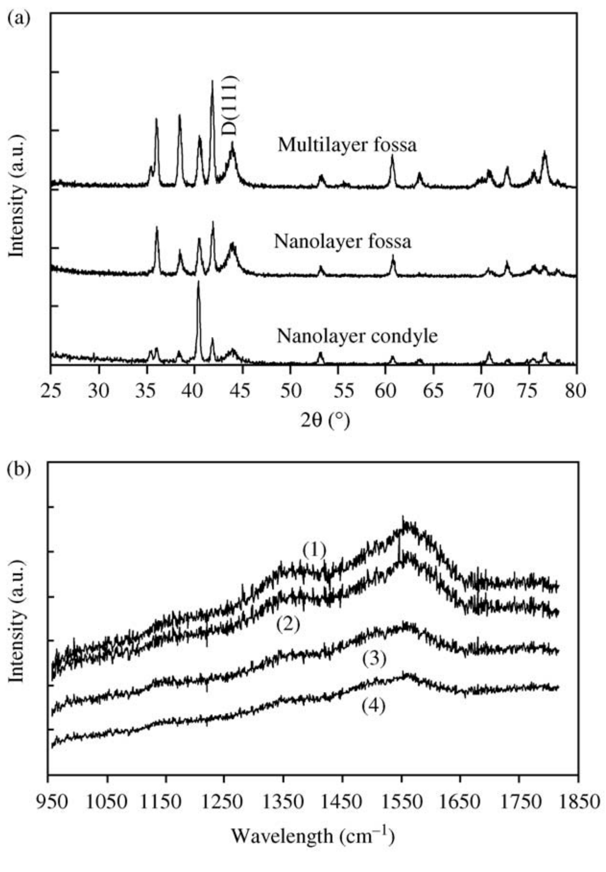

Figure 3 (a) XRD spectra of nanocrystalline film coated condyle and fossa and a multilayer film coated fossa showing the presence of crystalline diamond peak of (1 1 1) crystal structure; (b) shows micro-Raman spectra on fully coated areas of (1) nanocrystalline coated condyle, (2) multilayer film coated condyle, (3) nanocrystalline coated fossa, and (4) multilayer film coated fossa. The spectra show the presence nanocrystalline diamond at about $1350\,\text{cm}^{-1}$.

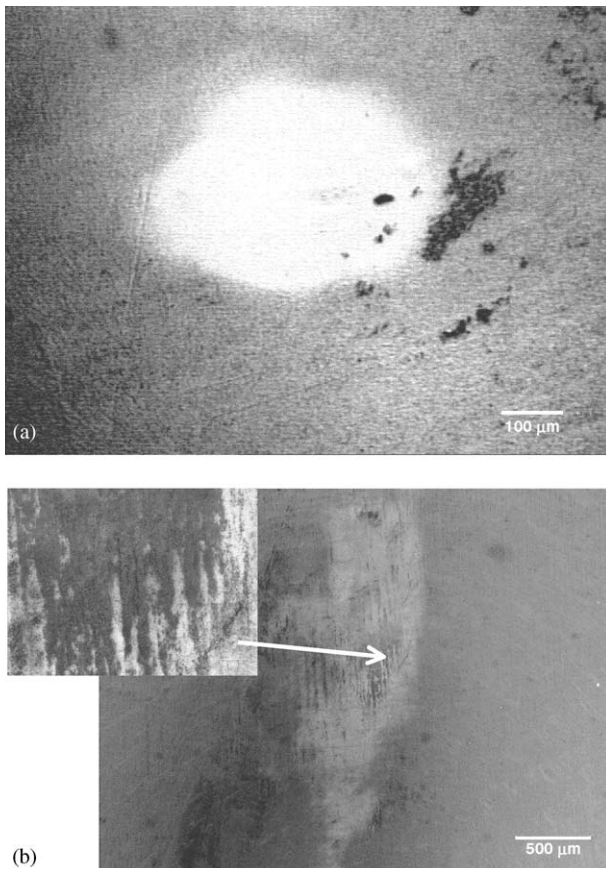

Figure 5 (a) An optical micrograph of the wear area of the nanocrystalline thin film coated condyle. The wear area appears as a bright near-elliptical spot. (b) An SEM micrograph of the fossa area with the high magnification inserted micrograph showing a crack in the nanocrystalline film.

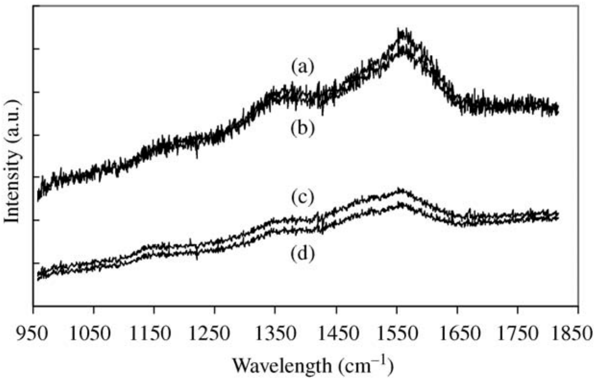

Figure 6 Post wear test micro-Raman spectra for (a) nanocrystalline film coated condyle, (b) multilayer film coated condyles, (c) nanocrystalline film coated fossa, and (d) multilayer film coated fossa.

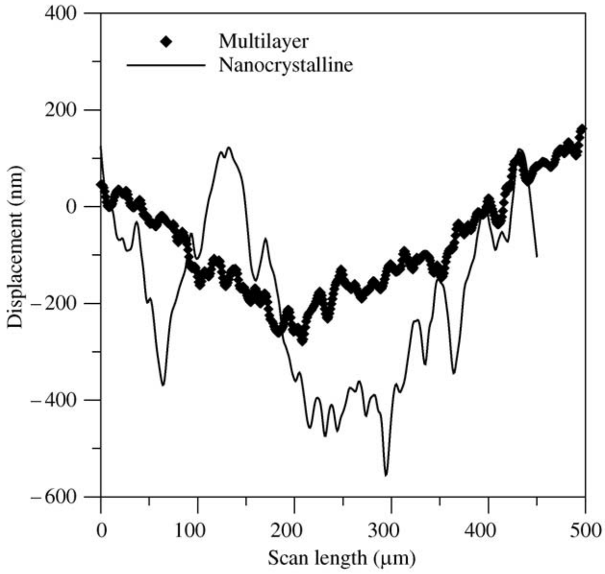

Figure 7 Profilometry scans across the wear tracks of the multilayer and the single-layer nanocrystalline diamond thin films. The maximum peak-to-valley height values were found to be 253 and 669 nm for the multilayer and single-layer nanocrystalline films, respectively.

surrounded by a rougher film. The small dark areas at the periphery of the wear area were not induced by the wear process under study, but by the action of a high-frequency drill that is used to extract the condyle fossa pair from the MMS after completion of the test. Micro-Raman scans on these areas showed a presence of NCD, which indicates that no film loss has occurred, but only extreme film damage. Fig. 5(b) shows that the wear action on the fossa also produced polished-diamond morphology. However, in this case, we see evidence of film cracking within the wear area, as shown by the inserted high magnification micrograph. Of all the coated samples we analyzed in this study, this is the only clear evidence of film cracking that we have seen within a wear area.

We performed Raman spectroscopy inside all the wear areas to ascertain film loss or retention after 2.25×10^5 cycles of mandibular wear action. The results in Fig. 6 show that we still have all the Raman peaks we saw in the as-deposited film conditions shown in Fig. 3(b) (i.e. G-Band ($1560\,cm^{-1}$), nanocrystalline diamond ($1350\,cm^{-1}$), and a broad peak at $1150\,cm^{-1}$). This proves that no film loss occurred in the wear areas of both condyle/fossa pairs after approximately two years of clinical use.

We performed profilometry studies on the wear tracks of the fossa specimens only since those of the condyles were too small to be measured with reliable accuracy. The profile scans presented in Fig. 7 are an average of two scans in the area analyzed. From Fig. 7 we can see that the maximum peak-to-valley height value of the single layer nanocrystalline film is more than twice that of the multilayer film. The values we measured were 235 nm and 669 nm for single-layer and multilayer films, respectively.

Based on the results presented here, we conclude that the multilayer appears to have superior wear properties than the single-layered film. The improved wear rates for multilayered films have been observed before by Ager *et al*. [7]. In this work, the reasons for this improvement were not fully explained since the multilayer film was found to be less hard than the monolithic film. In another work by Takeuchi *et al*. [8], the wear rates of the multilayer films were found to be worse than those of single-layered films, but had better bending strength. The improved strength for the multilayer films was attributed to the interface formed by the multilayer films, which was found to prevent crack propagation. In this work, the middle layer has higher diamond crystallinity and hardness than the two layers that bound it [5]. We suspect that the hardness of this highly crystalline film is responsible for the improved wear performance of the multilayer film. However, at this time we do not have enough evidence to conclude that this improvement may be attributed to the interface formed between the microcrystalline and the nanocrystalline layers of the film. On the other hand, it is interesting to note that the only film cracking that we observed occurred on the single-layered film in Fig. 5(b).

We have shown that coating TMJ implants with nanocrystalline and multilayer diamond thin films improved their service life spans. This was further made clear by comparing wear results of these coated implants with those of an uncoated condyle/fossa pair that was tested under the same wear conditions. The optical microscopy results after 2.25×10^5 cycles of articulation are shown in Fig. 8. Fig. 8(a) shows a wear area on the fossa created after articulation with an uncoated condyle. The area is very dark compared with the polished area that surrounds it. In addition, a shallow groove appears to have formed which indicates extensive wear at this area of articulation. On the contrary, Fig. 8(b) shows that there was minimal damage of the NCD thin film after an equivalent of two years of clinical use. We only see a wear area that appears to be smoother than the slightly rougher surrounding, but without any indication of loss of film.

4. Summary

In conclusion, we have shown that coating of simulated TMJ implants with either a single-layer nanocrystalline or a multilayer diamond thin film improved their service lifetimes. In this set of experiments, the multilayer appears to have superior wear properties than the single-layer nanocrystalline film. We believe that the service

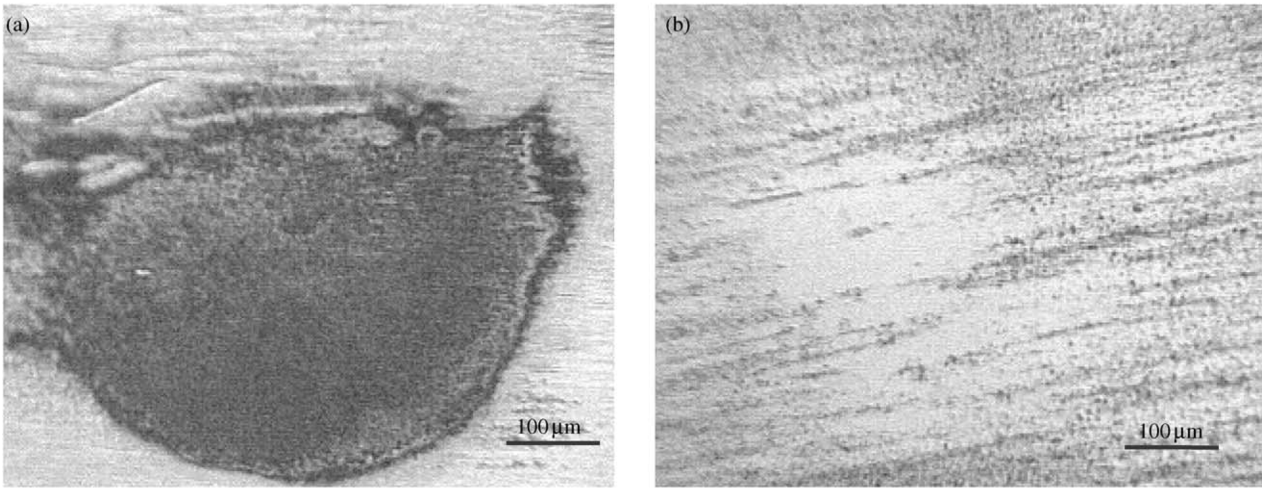

Figure 8 The picture (a) shows a micrograph of a wear area of an uncoated fossa. (b) shows a microstructure of a wear area of nanocrystalline diamond thin film coated fossa. The same experimental conditions were used for both samples. The surface damage on the uncoated surface on the right is extensive while the coated surface shows no wear damage.

lifetimes of other types of prostheses can be similarly improved. Other variables that are worth considering for future work include the effect of articulation environment and fossa geometry on the wear characteristics of the condyle/fossa pair.

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