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Lessons from the Lollipop: Biotribology, Tribocorrosion, and Irregular Surfaces

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Abstract Biotribology and tribocorrosion are often not included in numerical or computational modeling efforts to predict wear because of the apparent complexity in the geometry, the variability in removal rates, and the challenge associated with mixing time-dependent removal processes such as corrosion with cyclic material removal from wear. The lollipop is an accessible bio-tribocorrosion problem that is well known but underexplored scientifically as a tribocorrosion process. Stress-assisted dissolution was found to be the dominant tribocorrosion process driving material removal in this system. A model of material removal was described and approached by lumping the intrinsically time-dependent process with a mechanically driven process into a single cyclic volumetric material removal rate. This required the collection of self-reported wear data from 58 participants that were used in conjunction with statistical analysis of actual lollipop cross-sectional information. Thousands of repeated numerical simulations of material removal and shape evolution were conducted using a simple Monte Carlo process that varied the input parameters and geometries to match the measured variability. The resulting computations were analyzed to calculate both the average number of licks required to reach the Tootsie Roll® center of a Tootsie Roll® pop, as well as the expected variation thereof.

Keywords Biotribology · Tribocorrosion · Wear prediction · Wear evolution

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1 Introduction

Bio-tribocorrosion is a field at the interface of biotribology and tribocorrosion that unites these disciplines in an attempt to understand and characterize the effects of chemical and mechanical contributions to material removal and wear in a biological environment. Bio-tribocorrosion is well recognized as a critically important field of study that directly affects the health and function of human beings [1–5]. However, to date, there have been relatively few attempts to perform computational modeling that include bio-tribocorrosion effects in the predictions of component performance, evolution in geometry, or life. The Tootsie Pop® therefore provides an accessible platform from which to demonstrate techniques that may be used to observe, model, and study the coupling of biotribology and tribocorrosion in a highly variable but ultimately predictable wear system.

There has been some debate regarding the importance of understanding mechanisms of tribocorrosion versus simply

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being able to model and predict wear behavior [4]. In this manuscript, we focus on taking in vivo measurements of wear with all of the associated uncertainties and variability and propose a simple model for the evolution of geometry. In the case of the lollipop, this progression in wear lends a curious scientific perspective to the iconic question, "How many licks does it take to get to the Tootsie Roll® center of a Tootsie Pop®?" Though they did not previously recognize this question as a matter of bio-tribocorrosion, other groups have attempted to provide an answer through a variety of means. Via the creation of physical simulators, or statistical analysis of participant data, these studies resulted in a wide variation of predictions from 70 licks up to nearly 3,500 licks [6–9].

We began by studying the equatorial geometry of the lollipops to establish a baseline profile of the hard candy shell and to define the Tootsie Roll® center and the limits on wear. Of importance to this study were the variations between individual definitions of a lick or licking style, the differences in lollipop geometry, and the biological and chemical differences in the saliva of the participants. Despite the mechanistic complexity, the wear behavior of a Tootsie Pop® was easily analyzed using numerical methods and data from a large sample population. Thus, the problem of "How many licks?" was reduced to a simplified analysis of material removal, the accuracy of which depended critically on the appropriate inclusion of input parameters that matched the measured statistical variation. Despite remarkably different styles of licking and a wide variation in material removal rates among participants, the model predicted a nearly style-independent number of licks to reach the center.

2 Materials, Methods, and Mathematics

Regular-sized Tootsie Pops® (Tootsie Roll® Industries) were used for these experiments. The average initial mass of the lollipops (mass of the stick subtracted) was 17.51 ± 0.54 g. The average density of the hard candy shell of the lollipops was 1.54 ± 0.06 g/cm^3, and the average surface area was 2,803 ± 78 mm^2.

The wear rate of the candy shell under repeated licking with a dry tongue was found to be essentially negligible, producing <30 μg of material removal per lick. The dissolution of the candy shell proceeded at similar rates whether dissolving in a quiescent water bath, or in the mouth without abrasion from the tongue or palette, 7.7 ± 1.4 and 6.7 ± 1.5 mg/s, respectively. The rate of material removal measured for continuous licking or agitation in the mouth was measured to be 18 ± 2.5 mg/s, which clearly indicates a bio-tribocorrosion process due to the nearly threefold increase in material removal rate. This increase in mass removal rate could not be explained by fluid agitation alone. Lollipops exposed to mild agitation of 25 mL of water (which corresponds to the amount of saliva produced over a 5 min span while eating [10]) experienced a dissolution rate of only 9.0 ± 1.6 mg/s. Therefore, the wear of lollipops *in oris* may be best described as stress-assisted dissolution, which has been widely studied [11–14]. The bio-tribocorrosion problem considered here couples a corrosive environment (pH of saliva is ~6–7) with soft, rough biomechanical sliding (R_a tongue ~33 μm, E ~15 kPa) that is exceedingly difficult to replicate artificially [15–18].

Tootsie Pops® are not uniformly shaped and are definitely not round. In order to measure and model the Tootsie Pop®'s geometry, a selection of samples were sectioned equatorially and polished (Fig. 1). These cross-sections were photographed using a Nikon D3X DSLR camera with

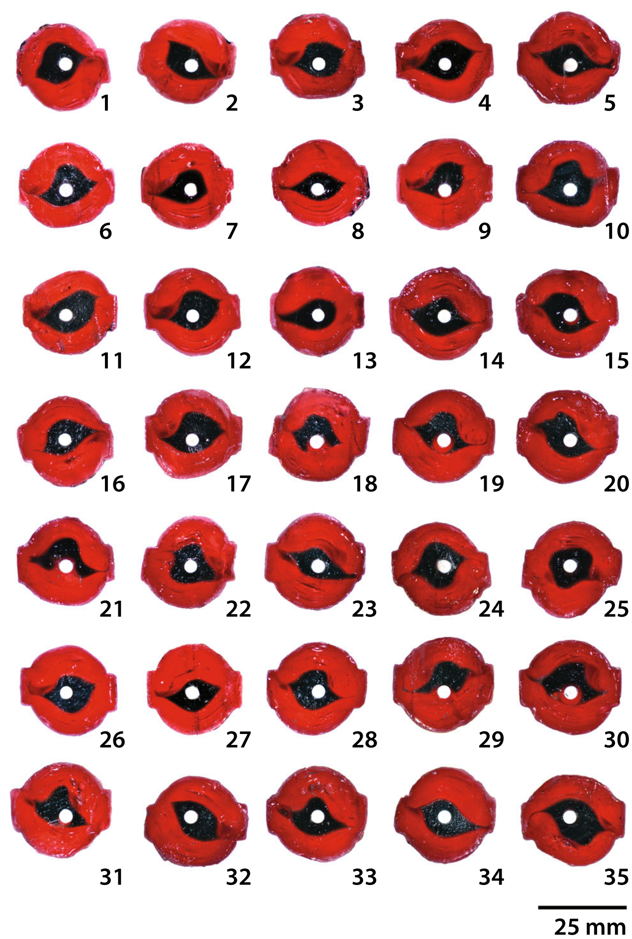

Fig. 1 Composite image of 35 equatorial cross-sections of randomly selected regular-sized cherry-flavored Tootsie Pops® illustrating the variation in shape and size, especially of the Tootsie Roll® centers. Although cherry-flavored lollipops are shown here, flavors during testing were allowed to vary as it was assumed this had little bearing on wear

a 3,072 × 4,068 CCD chip (12.5 megapixels) and analyzed with custom-written MATLAB software. Color RGB images were separated into three separate intensity maps, one for each color. An intensity-based threshold was used to segment the three different regions within the lollipop; the candy shell, the Tootsie Roll® center, and the stick. After noise reduction, the single-pixel-wide edges of the segmented regions were identified and used to measure the sizes and shapes of the regions of interest. The lollipop diameter was on the order of 1,500 pixels, and the intensity rise used as the threshold occurred over about 15 pixels, resulting in an experimental uncertainty for measuring the lollipop radius of about 2 %. This process was applied to 27 different lollipops. The average Tootsie Pop® shape and size was computed by aligning the surfaces of all lollipops in a single shared polar coordinate system (Fig. 2a). The average Tootsie Pop®, shown in Fig. 2b, preserved all general geometric features of most individual Tootsie Pops®.

Quantitative analysis of each lollipop was carried out individually and averages over the population were performed. The distance between the geometric center and the surface of the candy shell at each boundary pixel defined a local lollipop radius, R_c. Averaged over all locations and all 27 lollipops, we found $R_c = 13.8 \pm 0.5$ mm. The standard deviation of ±0.5 mm corresponds to a variation of about 4 %, or twice the experimental uncertainty. By contrast, the radius of the Tootsie Roll® center, R_t, measured from the geometric center exhibits a much larger variation of about 22 %: $R_t = 5.9 \pm 1.3$ mm. The origins of this large variability can be directly observed in Fig. 1. The Tootsie Roll® center generally exhibits an elongated cat's eye shape located off-center from the geometric center of the candy shell.

Multiple thickness metrics can be determined from the surface coordinates of the candy shell and the surface coordinates of the Tootsie Roll® center. One such metric is the shortest distance from each location on the candy surface to the tootsie surface, S_m (Fig. 2a). We found the average S_m to be 7.2 ± 1.2 mm over all surface points and all lollipops (Fig. 2c). The large standard deviation of S_m arises from large variations in Tootsie Roll® shape. This metric is important for wear processes occurring at single locations, randomly chosen from test to test. Alternatively, in cases where removal occurred uniformly over the entire candy surface, the controlling variable would be the

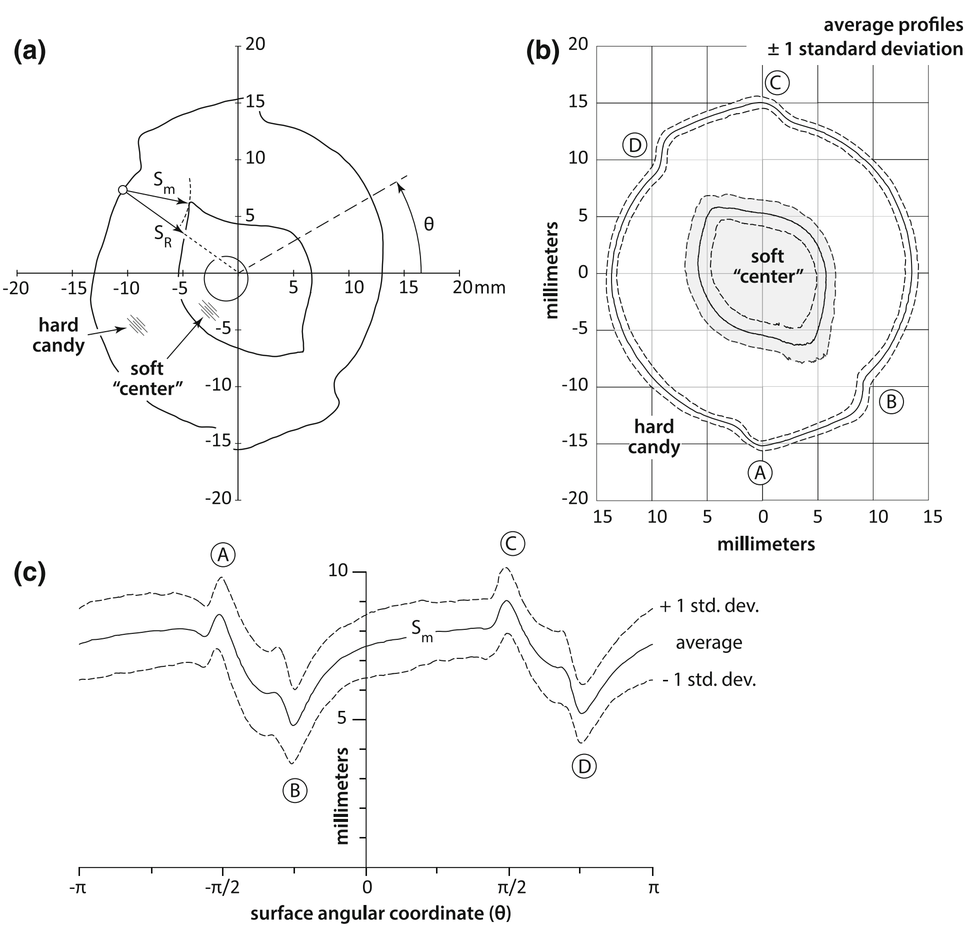

Fig. 2 **a** Equatorial profile of a real Tootsie Pop® cross-section (number 12 in Fig. 1). MATLAB image thresholding was used to isolate the hard candy shell, the Tootsie Roll® center, and the stick. S_m designates the minimum distance between any exterior point of the candy and the nearest Tootsie surface. S_R designates the distance from the candy shell to the Tootsie center as measured along the radius as defined from the area centroid of the entire cross-section. **b** The composite average Tootsie Pop® outline plus and minus one standard deviation for 27 lollipops. **c** S_m for each point of the exterior candy unwrapped radially around the area centroid with reference points *A*, *B*, *C* and *D*

minimum S_m, not the average S_m. We found that the minimum S_m, averaged over all lollipops, was just 4.3 ± 0.7 mm, suggesting that the mode of candy removal may strongly influence the number of licks it takes to get to the Tootsie Roll® center.

In order to measure a lumped parameter of material removal rate per lick, a group of participants were asked to perform repeated licking and weighing of Tootsie Pops® at regular intervals of ten licks. Each participant defined his or her own licking style and applied it consistently throughout the duration of each test. The effect of a single lick on the rate of surface recession was not measured during these experiments nor was the duration of each lick a measured or controlled parameter. Volume loss per lick and the associated uncertainty were calculated based on methods following Schmitz et al. [19].

The normal force of the tongue was estimated to be about 3 N, corresponding to a contact pressure on the order of a few kilopascals. This value is commensurate with other soft contacts within the human body and is well within previously measured values for maximum tongue force [20]. The sliding distance per lick was, maximally, the average lollipop circumference for full-surface and half this value for one-sided licking. Measured values for the average mass lost per lick, for each of the respective licking styles, was used to compute wear rates.

In order to validate the model predictions, an additional series of experiments were performed and dimensional measurements were collected as point cloud data using a 3D laser scanning device (Maker Bot® Digitizer). In these studies, scans and mass measurements were again performed at regular intervals. The resulting point clouds were converted into polygons by modeling software (SolidWorks® by Dassault Systemes®) from which volumetric data were computed. This method resulted in a measured volumetric uncertainty on the order of $\pm 100\ mm^3$ and an areal uncertainty of $\pm 24\ mm^2$. These uncertainties are entirely dominated by the accuracy of the scanning method, not the number and spacing of points, surface roughness, or irregularity as described by Carmignato et al. [21]. Using a calibration standard this technique was found to have uncertainties of less than 2 % in volume and less than 1 % in surface area.

3 Results and Discussion

Many problems in biotribology do not lend themselves readily to interrupted measurements in vivo. As such, engineers have often sought to simplify the problem using simulators to include factors believed to dominate the process. Historically, mechanical simulators become complex kinematic mechanisms with oversimplified contact conditions, biological solutions, and motions. The design of these instruments is to perform nearly identical and repeated motions during the course of the study with the goal of highly controlled and precise experiments. Simulators have been created and operated in studies on the wear of Tootsie Pops®, often attempting to measure wear as a function of the number of licks. Given the complexity and challenges with matching all of the various parameters that determine wear, it is perhaps not surprising that these previous in vitro efforts [9] underpredicted the rates of wear by nearly an order of magnitude compared to those found *in oris* [6].

The rate of mass loss per lick varied greatly within the sample population, as shown in Fig. 3. Although lick styles were not controlled, they could be broadly classified as either "one-sided" or "full-surface." The one-sided lick style yielded an average mass loss per lick of 17 mg with a standard deviation of 10 mg. In contrast, the "full-surface" lick resulted in an average mass loss per lick of 62 mg with a standard deviation of 17 mg. Although there was a large standard deviation within the population, the standard deviations of a single individual's mass removal rates were approximately four times lower. Mass removal measurements of 90 mg per lick or greater could be best described as a protracted mechanical agitation (eating) and were not included in the analysis of licking data.

The volume losses for both the one-sided and full-surface licking styles using the 3D laser scanner are illustrated in Fig. 4a, b. A marked difference in wear profiles is evident between the evolutions of the two representative surface profiles. The one-sided licking style clearly affected a smaller percentage of the Tootsie Pop® profile, whereas the full-surface licking style showed significant geometric changes over the entire profile. This is further illustrated by the calculation of an average equatorial wear

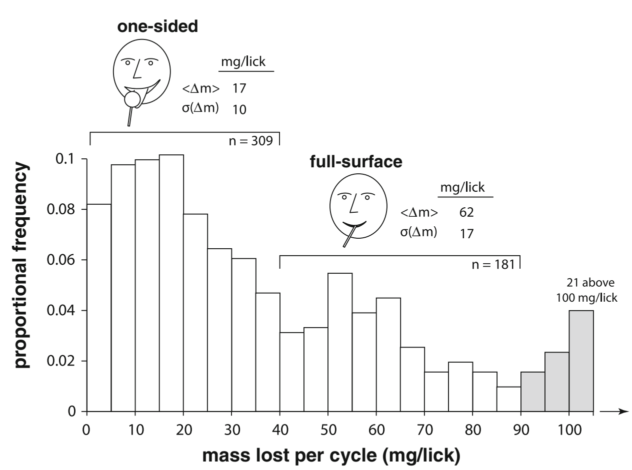

Fig. 3 Proportional frequency of mass loss per cycle as related to lick style. Wear rates above 90 mg/lick were not included in the analysis

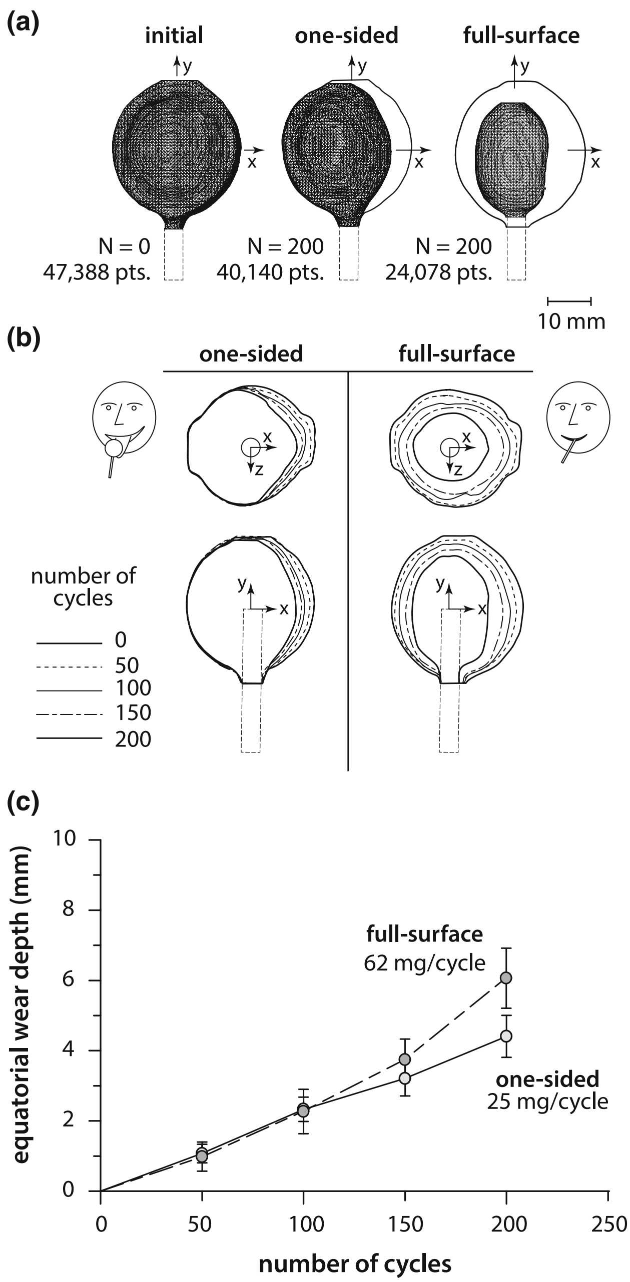

Fig. 4 **a** Representative digital scans of lollipops: initial unworn surface, one-sided licking after 200 cycles (with unworn outline), full-surface licking after 200 cycles (with unworn outline). **b** Outlines showing the progression of wear at 50 cycle intervals for one-sided and full-surface conditions. **c** Average equatorial penetration depth per cycle for each of the two licking styles

depth per lick for each of the two licking styles (Fig. 4c). It is interesting to observe that the measurements from the maximum region of equatorial wear for both the full-surface and one-sided removal approaches produced nearly identical results over the first 100 cycles. The average penetration rates were both measured to be 23 μm per cycle, with variations of ±6 μm for the full-surface and ±3 μm for the one-sided approach. These nearly identical rates of penetration occurred despite a fourfold difference in the overall volumetric material removal rates.

Given the wide variation in material removal rates from person-to-person, the irregularly shaped samples, and unique variations in geometry, the likelihood of developing simple closed form models for the evolution in geometry is low. However, this problem, like many other studies of the coupled evolution of wear and geometry, can be tackled using numerical methods. From orthopedic applications to dental wear, numerical approaches to predict the evolution of wear are well established [22–33]. For this bio-tribo-corrosion problem, we propose the use of a lumped parameter wear rate that includes wear due to both chemical and mechanical effects [34].

The modeling approach used here follows previous work [23–28, 33], which incrementally removes material normal to the surface (h_s) on a cycle-by-cycle basis according to the local contact pressure (P_s), sliding distance (d), and wear rate (K), Eq. 1.

$$h_s = K \cdot P_s \cdot d \qquad (1)$$

Assuming that an individual applies the same force, lick duration, and sliding distance per lick, the volume removed per lick can be easily converted into a lumped wear rate. This permits the computation of material recession over the engaged surface given a prescribed pressure distribution for each lick style: (1) a full-surface model assuming a uniform recession of material into the surface and (2) a one-sided model assuming smooth trigonometric functions along the surface normals such that removal rates are maximum at the equator and zero at the poles. The numerical simulation progresses on a cycle-by-cycle basis beginning with a representative geometry and proceeds until the model predicts contact with the Tootsie Roll® center. For any given simulation, a starting geometry is randomly constructed based on the measured variations in shape. Additionally, a single or full-surface wear rate from within the measured distributions is applied to the corresponding simulation. The simulations, which distribute wear across the generated surfaces, rapidly evolve the starting geometry into a worn geometry by keeping the volume of material removed constant between cycles.

The relative ease of computing the evolution in surface geometry allows us to perform Monte Carlo simulations of wear (5,000 different simulations) over the entire range of input parameters to give both average values of wear depth and of the associated uncertainties. The mean values from the simulations are shown in Fig. 5a for both models. Strikingly, the results overlap in a nearly linear region of wear depth recession over the first 150 cycles despite the two methods differing in average volumetric

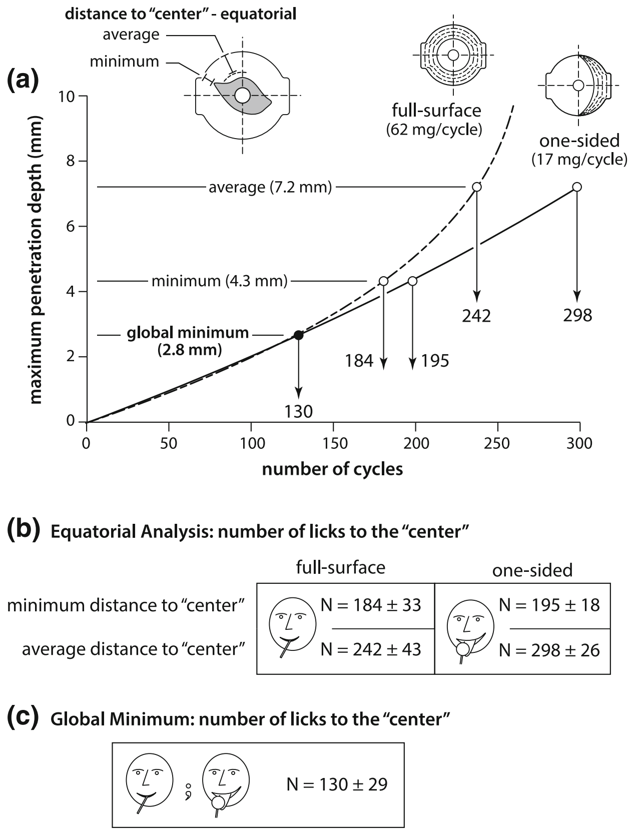

Fig. 5 **a** Model of penetration depth versus cycles for one-sided and full-surface licking styles. Both methods are approximately linear and equivalent over the first 80 cycles. **b** Wear model predictions of number of licks to reach the Tootsie Roll® center based upon data from equatorial cross-sections. **c** The global minimum penetration depth required to reach the "center" is frequently located off the equatorial region slightly toward the apex of the lollipop and has an average global minimum value of 2.8 ± 0.8 mm. Based on this global minimum value, an alternate number of licks to reach the "center" was calculated

removal rates by a factor of almost four. The reason for this is related to the difference in contact area between the two methods. The full-surface material removal method engages the entire exterior surface of the candy using the tongue, hard palate, and lips, while the one-sided approach uses only the tongue. This similarity in wear depth between the models is in good agreement with the experimental measurements shown in Fig. 4. The linear trend in the initial stages of wear is not surprising given the relatively small fraction of wear depth over the characteristic dimensions of the component and illustrates both the utility and potential accuracy of even simple linear extrapolations of wear depth over irregularly shaped geometries.

As described earlier in the manuscript, the question of success in this application is based on exceeding a particular wear depth and reaching the center of the lollipop. Along the equatorial region, the minimum and average wear depth required to reach the center were determined (Fig. 3). Based on the modeling approach shown in Fig. 5a, the number of cycles and uncertainties to reach the minimum and average locations of the center are given in Fig. 5b. For the single-sided approach, there is an additional element of chance, as the location of maximum wear depth may or may not be aligned with the minimum location, but this problem of alignment is alleviated in the full-surface approach as the entire surface is engaged in the material removal.

Finally, although the analysis of the geometry is based on the equatorial cross-sections, detailed examination of the lollipops indicates that the minimum candy shell thickness is rarely (if ever) located along the equator. A series of longitudinal cross-sections indicated that the global minimum thickness required to reach the center is frequently located off the equatorial region slightly toward the apex of the lollipop and has an average global minimum value of 2.8 ± 0.8 mm, which conveniently falls within the region where both of the methods overlap. Interestingly, in this region, the numerical analysis can offer a solution that is truly independent of the licking method: 130 ± 29 licks to reach the center. It is further refreshing to note that the experiments performed at Swarthmore College found 144 ± 45 cycles when the students were instructed to lick lollipops and simply record when they reached the center [6].

4 Concluding Remarks

Biotribological problems, and biology in general, do not always conform to simple geometries and often involve irregularity in surface shape. Even so, the evolution of wear may still be predicted using simple numerical methods that incrementally update the surface geometry on a cycle-by-cycle basis. As this problem has illustrated, biotribology and tribocorrosion problems can be described using classical tools and methods despite the complexity of combining an intrinsically time-dependent process (corrosion) with a mechanically driven process (wear). Here, a uniform per cycle material removal rate incorporates the coupled tribocorrosion effect into a wear model.

Biotribology and tribocorrosion processes are expected to show wide variability in material removal rates given a sample population of individuals. Traditionally, only average values are considered, but modern numerical methods can easily perform repeated simulations following statistical Monte Carlo approaches to include the expected range of input parameter variability in providing predictions of in vivo operation. Mechanical wear and tribocorrosion material removal processes actually lend themselves well to numerical extrapolation in surface geometry due to the relatively small dimensional changes, which do not generally distort the overall geometric form of the problem.

The inclusion of temporal material removal rates, such as stress-assisted dissolution or corrosion, can be lumped into mechanical removal rates on a per cycle basis for the purpose of numerical simulation under conditions in which the time periods between removal events are fairly matched to the application. In this work, wear and stress-assisted dissolution of the candy shell of a Tootsie Pop® were used as lumped material removal rate in a numerical simulation that could rapidly predict wear depth and shape evolution. The relative ease of performing these computations is in direct contrast to the risk associated with accelerated physical simulation, which inherently cannot match both mechanical removal rates with dissolution rates that are time dependent without altering the fluid environment.

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