Removal Mechanism of Artificial Dental Plaque by Impact of Micro-Droplets

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This paper presents the experimental evaluation of the impact of a single water droplet on a model of artificial dental plaque used as a biofilm. The instantaneous moment of the impact of the droplet on the biofilm was visualized using a high-speed imaging technique. It was found that the artificial plaque could withstand a shear stress of 10.5 kPa, and the normal stress resulting from the impact of the droplet was a dominant factor of the plaque-removal process investigated in our experiment. The actual impact of a single droplet on artificial plaque was precisely measured as the formation of a three-dimensional surface structure of a crater.

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The impact of droplets impinging on a surface is an interesting topic in both academic and industrial fields.1–3 For example, high-speed impacts on a solid surface have been studied to solve the problem of erosion by water drops on turbine blades, and the effect of droplet impact is utilized in ink-jet printing and spray cooling. In the field of semiconductor manufacturing, an important application of water droplet impact is the cleaning of a silicon wafer surface.4,5 After Worthington first studied the collision of a liquid droplet on a liquid film, a well-known phenomenon referred to as the milk-crown, droplet collision on a liquid surface has been a subject of many investigations.6–13

On the other hand, bacterial biofilms on teeth surfaces, also referred to as dental plaque, cause serious problems such as caries, which is the formation of cavities.14,15 The basic method to remove plaque is mechanical cleaning with a tooth brush. To perform the removal perfectly, an experienced technique is required, however. Another method is using a high-pressure water jet, but it has low efficiency. Moreover, the possibility of gum injury makes this method unsuitable for the elderly and persons in need of nursing care.

To resolve the above issues, impinging the biofilm with water micro-droplets has been proposed as a new method to remove dental plaque. This method does not harm intraoral skin because the micro-droplets have low momentum. Numerical and experimental analyses have been conducted for the design of a practical device to administer the film removal;16 however, because the impact of a single micro-droplet impinging on a biofilm takes place rapidly and on a small scale, it is difficult to conduct dynamical visualization of the phenomenon and the process is not fully understood. In particular, it is a challenge to optically observe the instantaneous moment of removal of an artificial plaque by the impact of the water droplet because the impact velocity of each droplet is higher than 100 m/s and the diameter of a droplet is about several tens of micrometers. From the reasons above, studying on the plaque removal mechanism by simply observing the phenomena of the plaque removal is extremely difficult. We need to know both of the detail information about the droplet and resulted plaque removal.

Therefore, in order to clarify the mechanism of biofilm removal by the impact of a single water droplet, we investigated the phenomenon via two approaches, namely, the observation of micro-droplets of water colliding with the water film, and three-dimensional surface measurements that focused on the shape of the crater generated by the impact of a water droplet.

Next, precise three-dimensional measurements of the crater generated by the impact of a single micro-droplet were carried out with a non-contact coordinate-measurement system. The shape of the crater was analyzed statistically to examine the characteristics. It was possible to reduce the droplet density by moving the stage at a continuous speed, and the measurements were obtained by focusing on the impact of a single droplet. Furthermore, to clarify the mechanism of the plaque removal, the shear stress was measured and its effect was analyzed.

Experimental Setup and Methods of Analysis

Imaging of a single droplet impinging on liquid surface.—In the first experiment, the handpiece of the device that generated the water droplets was used to direct a stream of droplets onto a water film. The instantaneous moment when a droplet impacted the film was captured by high-speed imaging, as shown in Fig. 1a. The top of the handpiece had two nozzles for air and one nozzle (diameter: 50 μm) for water, as shown in Fig. 1. Both air nozzles were oriented inward to focus at a point 6 mm below the top of the handpiece; the operating air pressure was 0.15 MPa gauge. Micro-droplets were produced owing to the instability of the water nozzle, and the droplets were accelerated and atomized by the high-speed air ejected from the air nozzles. The velocity of the generated mist was measured by particle image velocimetry (PIV). We used three types of the window size; 64×32, 32×16, and 16×8 pixels (horizontal direction: vertical direction) for the recursive cross-correlation method. The overlap percentage was 50% for both of horizontal and vertical direction. The droplets of the mists were used as the tracers. The spatial resolution was 9.4 μm/pixel. The maximum velocity of the mist at a position 8 mm downstream from the water nozzle was 171 m/s.

A high-speed camera (Hyper Vision HPV-X2, Shimadzu Corp., Japan) was set on the opposite side of the glass plate, separating it from the handpiece and laser. The impinging droplet was recorded through the glass plate at a recording speed of 10 mega-frames per second (Mfps) and an exposure time of 50 ns. A 532-nm laser (SSL-532-5000-10TM-D, Shanghai Sanctity Laser Technology Co. Ltd., China) was used to visualize the high-speed droplet motion. A large-distance microscopic objective lens (VHZ-50L, Keyence.) was mounted on the camera. The spatial resolution was 1.73 μm/pixel. The handpiece was set at a 60° orientation, and the distance between the nozzle and the glass surface was 8 mm to assume the practical use, meaning, using the hand piece to remove the plaque on teeth in a mouth. The observed area of 690 μm × 430 μm is enough small compared to the distance from the nozzle to the glass surface. Therefore, it is possible to assume that all the observed droplets fly to the surface with same angle of 60°. A spatially dispersed mist generated a continuous flowing water film on the glass surface; tap water was induced into the nozzle through the particle filter at a flow rate of 0.15 mL/min. The water film thickness is

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unknown. However, the thickness can be assumed as enough uniform in the observed area because the flow rate of water is constant in the experiment.

Analysis of surface shape formed by droplet impact.—In the second experiment, the removal of the artificial plaque by the impact of a single droplet was analyzed (Fig. 1b). Artificial plaque (Nissin Dental Products Inc., Japan) was deposited on the glass slide. After the artificial plaque dried, the glass slide was set on a stage that can move at a constant velocity. The glass slide passes by under the spray nozzle. Therefore, that can reduce the density of the impinging droplets on the one local area in order to avoid multi droplets strike a same crater.

The handpiece was placed 8 mm from the bottom surface of the artificial plaque. The mist generating conditions were the same as those for the experiment described in Experimental Setup section. In this experiment, the water film was formed on the artificial plaque due to the wettability of the material. As long as the impacting droplet collides on the water film formed on the artificial plaque, it is considered that the wettability of the plaque does not affect the removal property much. In the application, basically, it is a wet circumstance in the human mouth such is similar with our experiment. Three-dimensional information of the surface was obtained by using a non-contact coordinate-measurement system (KS-1100, Keyence Corp., Japan). that could measure the surface shape with a scanning laser.

Results and Discussion

Imaging of rapid impact of micro-droplet on water film.—Figure 2 shows the sequential images of a droplet impinging on a water film, obtained by the high-speed camera at a rate of 10 Mfps. The images were recorded from the back side of the glass. The droplet approaching the water film appears as a dark object, as circled and marked by a white arrow in Figs. 2a–2ii; the velocity just before the impact was then obtained as \(V_i = \frac{L}{\cos(60°)}\) \(\frac{dt}{d_i}\). Here the frame duration, \(dt\), is 0.1 \(\mu\)s when the recording speed is 10 Mfps. After the impact of the droplet on the water film, as shown in Figs. 2a–2iii, the droplet expanded radially, and the surface wave was observed as a dark edge in the images (Figs. 2a–2v) until it disappeared (Figs. 2a–2vi). The diameter of the observed wave right before it disappeared is defined as the characteristic diameter, \(d_a\), after the droplet impact.
Correlation coefficient of \( d = 0.70 \), respectively. Obviously, the expansion width after the droplet was sufficiently flat so that the shear stress on the surface of the artificial plaque was not observed. Here, a minimum distance \( d \) of 100.0 mm and width of 1.5 mm was set in a reservoir. The glass plate with the artificial plaque adhered on its surface was set on the side wall of the chamber. The distance \( d_{\text{disk}} \) could be controlled with 0.5 \( \mu \)m accuracy by using a micro-stage. The side surface of the rotating disk was sufficiently flat so that the shear stress on the surface of the artificial plaque could be assumed to be \( \tau_{\text{disk}} = \mu U_{\text{disk}}/d_{\text{disk}} \). In the experiment, no matter how much the distance was decreased, removal of artificial plaque was not observed. Here, a minimum \( d_{\text{disk}} \) of 1.0 \( \mu \)m yielded a maximum shear stress of 10.5 kPa.

Next, we conducted an experiment to elucidate the effect of the water film thickness on the cleaning of a surface.20 As reported in an earlier study, the shear stress caused by the droplet impact depends on the thickness. The average impact velocity was overestimated from the obtained images. After an impact, a shockwave can even be generated, causing a cavitation in the droplet surface.18

A typical three-dimensional image of a crater obtained from the high-speed imaging system is shown in Fig. 5. Based on the moving speed of the stage during the impact by the droplets, it is assumed that the crater was created by a single droplet because there was sufficient space between the droplets. In other words, the number of droplets in the observed area was enough small to not collide at the same point on the plaque.

The distances between the nearest neighbor craters were measured from the obtained data. The mean distance was 141 \( \pm \) 70 \( \mu \)m, that is enough larger than the mean diameter of the crater of 33 \( \mu \)m. There is a possibility to the more than two droplets collide on the same position.

Precise imaging of crater formed on artificial plaque by a single water droplet.—A typical three-dimensional image of a crater obtained with the non-contact coordinate-measurement system is shown in Fig. 5. Based on the moving speed of the stage during the impact by the droplets, it is assumed that the crater was created by a single droplet because there was sufficient space between the droplets. In other words, the number of droplets in the observed area was enough small to not collide at the same point on the plaque.

The high-speed liquid column strikes a solid material, very high pressure is generated as known as the “water-hammer effect.”20 The pressure of water hammer, \( P_{\text{WH}} \), is obtained as \( P_{\text{WH}} = \rho CV \), where, \( \rho \), \( C \), and \( V \) denote the fluid density, speed of sound in the medium and the velocity of the liquid. The mean velocity of the impacting droplet, \( V_i \), is 140 m/s. When we assume \( C = 1500 \) m/s, the speed of sound in water, \( P_{\text{WH}} = 998 \times 140 \times 1500 = 2 \times 10^8 \) (Pa).22 The value is extremely larger than the estimated shear stress. The followed normal stress generated in the artificial plaque after the impact varies moment by moment due to the deformation of the material and the droplet. However, this vertical pressure when the impact of droplet seems dominant factor of the plaque removal. The actual removal of artificial plaque by a single droplet explained in the next section supports this consideration.

![Figure 4. Schematics of experimental setup for measuring the shear stress.](image)

Figure 3a shows the distributions of \( d \) and \( d_i \). Note that the data only contains the information of observable droplets in the high-speed imaging. The average values of \( d \) and \( d_i \) were 14.1 and 50.5 \( \mu \)m, respectively.

The relationship between \( d \) and \( V_i \) and the relationship between \( d \) and \( d_i \) for each droplet are shown in Figs. 3b and 3c, respectively. The correlation coefficient of \( V_i - d_4 \) and \( d_i - d_4 \) were calculated as 0.39 and 0.70, respectively. Obviously, the expansion width after the droplet impact, \( d_4 \), was correlated with the droplet size before the impact, \( d_i \). However, the results indicate that the impact velocity also affected the expansion even though the correlation coefficient was relatively small. Note that, the fact of this correlation indicates that the assumption of the uniformity of the water film thickness is acceptable because \( d_4 \) should affected by the thickness. The average impact velocity was 140 m/s, and the highest velocity recorded in our experiment was 220 m/s. As a result of these high-speed droplets, the biofilm was subjected to large impacts. It has been reported that when the velocity of a droplet is higher than 100 m/s, a shockwave can even be generated, causing a cavitation in the droplet surface.15,18

When a thin water film spreads at such a high speed, it is expected to generate a high shear stress. This shear stress is one of the important factors in the cleaning of a surface.19 As reported in an earlier numerical study, the shear stress caused by the droplet impact depends on the velocity and viscosity of the liquid.16 We estimated the shear stress in our experiment from the obtained images. After an impact, a droplet spread owing to an inertial force and formed a thin disk-like film called a lamella on the solid surface. Although there was a water film in our experiment, we assumed that a sphere with a diameter of \( d \) was transformed to a cylinder with a diameter of \( d_4 \) and a height of \( h \). Based on the conservation of volume, \( h \) was calculated to be \( h = 2d_i^3/3d_4^2 \). When the velocity of the spreading droplet was assumed to be \( U_i = d_4/2dt \), the shear stress, \( \tau \), was calculated to be as \( \tau_{\text{drop}} = \mu U_i/h \). The average \( \tau \) obtained from the estimation was 46.5 kPa. Note that this assumed velocity, \( U_i \), was the highest conceivable value without considering the effect of the water film. If the viscous drag is taken into consideration, the velocity should decrease and the actual shear stress should be lower than our estimated value.

The result indicates that the shear stress was not a main factor in the removal of the artificial plaque. Although \( \tau_{\text{disk}} \) was lower than the estimated maximum shear stress, \( \tau_{\text{drop}} = 46.5 \) kPa, both values still have the same order of magnitude considering that \( \tau_{\text{drop}} \) was overestimated. Hence, the shear stress was not the dominant factor in removing artificial plaque in our experiment.

On the other hand, the impacting pressure can be also estimated from the experiment. When the high-speed liquid column strikes a solid material, very high pressure is generated as known as the “water-hammer effect.”20 The pressure of water hammer, \( P_{\text{WH}} \), is obtained as \( P_{\text{WH}} = \rho CV \), where, \( \rho \), \( C \), and \( V \) denote the fluid density, speed of sound in the medium and the velocity of the liquid. The mean velocity of the impacting droplet, \( V_i \), is 140 m/s. When we assume \( C = 1500 \) m/s, the speed of sound in water, \( P_{\text{WH}} = 998 \times 140 \times 1500 = 2 \times 10^8 \) (Pa).22 The value is extremely larger than the estimated shear stress. The followed normal stress generated in the artificial plaque after the impact varies moment by moment due to the deformation of the material and the droplet. However, this vertical pressure when the impact of droplet seems dominant factor of the plaque removal. The actual removal of artificial plaque by a single droplet explained in the next section supports this consideration.

![Figure 3. Impact of a single droplet: (a) distribution of diameter of a droplet before impact, \( d_i \) (blue bar), and diameter of a spreading droplet, \( d_4 \) (green bar). (b) velocity distribution of droplet before impact, \( V_i \) and \( d_4 \) and (c) relation between \( d_4 \) and \( d_i \).](image)
Figure 5. Three-dimensional image of a crater generated by a droplet on the artificial plaque.

Figure 6. Definition of the different areas of a crater shape as a function of depth. The blue line shows the cross section of a crater.

The results indicate that the surface around the crater was not flat. However, that should be small enough as the experimental errors in our experiment.

When we focused on the shape of craters, it appeared that the rim of the crater tended to be elevated compared to the surrounding area. Each color shown in Fig. 5 represents the distance from a specific reference point of the measurement, such as the surface of the glass slide. For example, the distance from the bottom of a crater is an important parameter for defining the characteristics of each crater. Here, the original film thickness remains unclear owing to the difficulty in creating a uniform artificial plaque film. Hence, we introduced the following definitions to quantitatively discuss the crater size. The description of a crater is divided into three parts as follows:

1. The depth value was binarized by 90% of the average depth. The equivalent diameter is defined as $D_{bi}$.
2. The area of $0.2 \times D_{bi}$ (m) inside the binarized area is defined as the bottom section. Therefore, we can ignore the steeper area shown in Fig. 6. Similarly, the area from $0.2 \times D_{bi}$ to $1.5 \times D_{bi}$ outside of the binarized area is defined as the rim section. Then, the area from $1.7 \times D_{bi}$ to $2.2 \times D_{bi}$ outside of the binarized area is defined as the surrounding section.
3. The difference between the average height of a bottom area and the average height of the rim or the surrounding is defined as the average crater height. The maximum crater height is obtained using the same method. In other words, in the rim section, Average height of rim = Average height in rim section – Average height in bottom section, and in the surrounding section, Average height of surrounding = Average height in surround section – Average height in bottom section.

Figures 7a and 7b show the distributions of the average and maximum height of the rim section and surrounding section, respectively; both plots show similar trends. Fig. 7a was obtained from the data of 104 samples. The maximum depth of the observed area was 60.3 μm; the median and average values were 16.1 and 22.1 μm, respectively.

The average difference between the rim section height and the surrounding section height (= mean height of surround part – mean height of rim part) was 2.5 μm, as shown in Fig. 8. This result indicates that the rim section tended to be higher than the surrounding section, which agrees with the mechanism of crater formation by the impact of a single droplet. As explained in Figs. 5b–5i to 5b–5iii, after the impact, the liquid pushed aside the plaque. It should be noted that artificial plaque is a viscoelastic material that can change the viscosity by interacting with water. In the process of plaque removal, the edge of the plaque forms an elevated rim, as observed in our experiment. If shear stress is a dominant mechanism of the removal, it is unlikely to yield a crater with an elevated rim because the shear stress is decreased as the water film spreads owing to the viscosity after the droplet collision. Therefore, the height of the plaque around the crater should increase radially toward the outside, unlike the case observed during our measurement.

Figure 9 shows the equivalent diameter of the bottom section of the craters, calculated from the area of the bottom section as $2 \times \left( \frac{\text{bottom area}}{\pi} \right)^{0.5}$. The obtained average diameter of the craters was $d_c = 32.6$ μm, which is larger than the average diameter $d_{bi}$, but smaller than the characteristic diameter $d_a$, as shown in Fig. 9. The droplet could not spread widely as in the case of the droplet impinging on the liquid.
film. In the removal of the artificial plaque, the dominant mechanism was probably that the impact of the droplet pushing the film aside, not that the shear stress breaking the coating.

**Conclusions**

A high-speed imaging technique was used to visualize the instantaneous moment of droplet impact. Furthermore, we obtained the precise three-dimensional image of a crater generated by the impact of a single droplet by using a non-contact coordinate-measurement system. An analysis was conducted to clarify the characteristics of a crater generated by the impact of a single droplet, and the main findings are summarized as follows:

1. The impact of a single droplet generated by a handpiece on a water film was visualized. The average impact velocity and the average diameter were 150 m/s and 14.1 μm, respectively. Compared with the experiment for measuring the endurance of the plaque against the shear stress, the shear stress generated by a droplet’s impact, estimated from the imaging (O ≈10 kPa), was not large enough to remove the artificial plaque.

2. The average equivalent diameter of the generated crater was larger than the impacting droplet diameter, $d_i$, but it was smaller than the characteristic diameter of spreading droplet, $d_d$.

3. The elevated rim around the crater accounted for the removal mechanism of the artificial plaque, i.e., the dominant mechanism of the removal was the impact of the dropping aside the plaque, not that the shear stress.

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