Characterization of Lubricity of Mucins at Polymeric Surfaces for Biomedical Applications

Seunghwan Lee

Abstract---The lubricating properties of commercially available mucins originating from different animal organs, namely bovine submaxillary mucin (BSM) and porcine gastric mucin (PGM), have been characterized at polymeric surfaces for biomedical applications. Atomic force microscopy (AFM) and pin-on-disk tribometry have been employed for tribological studies at nanoscale and macroscale contacts, respectively. Polystyrene (PS) was employed to represent 'rigid' contacts, whereas poly(dimethylsiloxane) (PDMS) was employed to represent 'soft contacts'. To understand the lubricating properties of mucins in correlation with the coverage on surfaces, adsorption properties of mucins onto the polymeric substrates have been characterized by means of optical waveguide light-mode spectroscopy (OWLS). Both mucins showed facile adsorption onto both polymeric substrates, but the lubricity was highly dependent upon the pH change between 2 and 7.

Keywords—Bovine submaxillary mucin (BSM), Porcine Gastric Mucin (PGM), lubricity, biomedical.

I. INTRODUCTION

MUCINS are high-molecular-weight ($>10^6$Da) glycoproteins that constitute slimy mucus secretion covering epithelial cell surfaces and are known to be primarily responsible for the protective and lubricious properties of viscoelastic mucous layers [1,2]. Despite obvious slipperiness of mucus gels observed from daily-life experiences, systematic researches on the lubricating properties of mucin and mucus gels are surprisingly rare to date [3-7]. The significance of understanding the lubricating properties of mucins and mucus gels is firstly related to the health status of the internal organs that mucin/mucus gels are designed to protect, as seen from dry mouth syndrome (as known as "xerostomia" [8]), or hypersecretion of mucus under inflammation [9]. The significance of systematic understanding of lubricating properties of mucin/mucus gels is also related to biomedical engineering area, in particular, tissue-contacting devices, such as stents, catheters, and endoscopes [10]. These devices require the movement along the surfaces of internal organ in their operation, and counter-surface is generally soft biological tissues that are covered with mucus gels. Smooth gliding of these tubular devices can lead to lower pain and easier operation. On the contrary, recent development of capsule-style endoscopy attempts to exploit adhesive properties between the devices and gastrointestinal mucosal tracts for smooth locomotion because

Seunghwan Lee is with the Department of Mechanical Engineering, Technical University of Denmark, Kgs. Lyngby, DK-2800, Denmark (phone: 45-4525-2193; fax: 45-4593-6213; e-mail: seele@ mek.dtu.dk).

unless self-powered, locomotion can start based on reliable "griping" by the devices on the mucosal wall. Various approaches to improve adhesive properties between the devices and mucus gels have been developed, including micropatterning [11], mucoadhesive films [12], and combination of both [13]. Examples mentioned above dictate that the application of biomedical devices or biosensors that include moving parts in human body require a thorough understanding of the tribological properties of mucus gels for optimum design and operation.

To date, mucin coatings for biomedical applications have been most actively studied with polymeric biomaterials, owing to the surfactant-like, amphiphilic characteristics of mucins; unglycosylated, 'naked' polypeptide regions are believed to readily interact with hydrophobic surfaces, and thus facilitate the spontaneous adsorption from aqueous solutions, whereas the glycosylated regions tend to stretched into bulk water for favorable interaction with water. Schematic illustrations for the molecular structure of mucins and their conformation at water/hydrophobic interfaces are presented in Fig. 1.
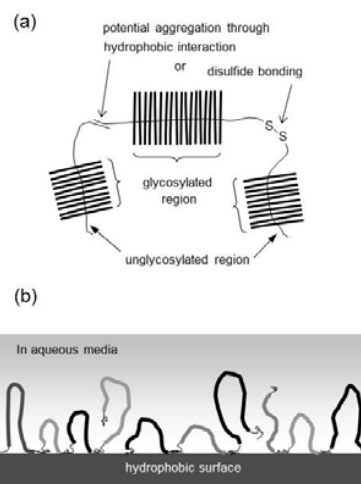

Fig. 1 Schematic illustrations of (a) mucin molecules and aggregates (b) proposed conformation on hydrophobic surfaces in aqueous media

Previous studies of mucin coatings on polymeric substrates have shown favorable biocompatibility, such as hydrophilization of polymeric surfaces [14], suppression of bacterial adhesion [15,16], and suppression of immune cell activities [17-19].

In a previous paper [5], the author and coworkers have

studied that the aqueous lubricating properties of porcine gastric mucin (PGM) in dilute regime (1 mg/ml) at a soft sliding contact by employing poly(dimethylsiloxane) (PDMS) as tribopair. In various aqueous solutions with a broad range of pH (2 to 12) and ionic strength (ca. 0 to 1 M), PGM has shown the most effective lubricating performance at acidic pH (e.g. pH 2), as long as the ionic strength is lower than ca. 0.1 M. The superior lubricating behavior of PGM at acidic pH than at other pHs was attributed to the pronounced aggregation of PGM through the hydrophobic interaction between unglycosylated regions, and consequently increased *adsorption strength* than *adsorbed amount* on PDMS surfaces. In the present study, it is aimed to extend the investigation of lubricating properties of mucins at polymeric surfaces to another commercially available mucin, namely bovine submaxillary mucin (BSM), as well as to another type of polymer substrate, namely Polystyrene (PS). While both PDMS and PS are known to reveal surface hydrophobicity, the mechanical properties of them are significantly different in that PDMS can represent compliant, rubber-like materials (Young's modulus, 1 – 2 MPa) and thus provide "soft" tribological contacts, whereas PS is much stiffer (Young's Modulus, 3,000 – 3,600 MPa) that it can provide "rigid" contacts. It is further noted that the two mucins studied in this work were used as received from the manufacturer without any purification. As is well known [20], these commercial mucin products contain many non-mucinous "contaminants", including cell debris and smaller proteins.

II. MATERIALS AND METHODS

A. Mucins and Chemicals

Commercially available porcine gastric mucin (PGM: type III, partially purified, bound sialic acid content ~1 %, M1778-10G, Sigma Aldrich, St. Louis, MO) and bovine submaxillary mucin (BSM: type I, purified, bound sialic acid content ~5 %, M-4503, Sigma Aldrich) were used as received from the manufacturer without further treatment. The received mucins were dissolved in two aqueous buffer solutions with pH 7 or pH 2 at the concentration of 1 mg/ml. The pH of the aqueous buffer solutions was adjusted by addition of HCl or 6 M NaOH into the distilled water containing 10 mM 4-(2-hydroxyethyl)piperaine-1-ethanesulfonicacid (HEPES, Sigma-Aldrich) and 140 mM NaCl. For AFM experiments (see below), HEPES buffer solution without 140 mM NaCl was used.

B. Optical Waveguide Lightmode Spectroscopy (OWLS)

The adsorption properties of mucins onto water/polymer interface have been investigated by means of optical waveguide light-mode spectroscopy (OWLS). OWLS is based on the grating-assisted in-coupling of a He-Ne laser into a planar waveguide (200-nm-thick $Si_{0.25}Ti_{0.75}O_2$ waveguiding layer on 0.5-mm-thick AF 45 glass, MicroVaccum Ltd, Budapest, Hungary), and allows for a direct online monitoring of the optical ("dry") mass of macromolecule adsorption.

Detailed information on the operational principles of OWLS is available in literature [21].

In this work, to emulate the tribological interface comprised of polymeric materials, OWLS waveguides were coated with ultra-thin polystyrene films (ca. 25 nm) or PDMS films (ca. 30 nm) by means of spin-coating. For spin-coating, OWLS waveguides were cleaned firstly by piranha solution (H_2SO_4: ca. 30% $H_2O_2 \approx 3:1$), thoroughly rinsed with water, dried with nitrogen gas, and finally oxygen-plasma-treated for 1 min. For PS coating, PS (M.W. 192,000, from Sigma-Aldrich) was dissolved in toluene at a concentration of 5 mg/ml, and used for spin-coating. For PDMS coating, the base and curing agent of a commercial silicone elastomer kit (see section D. Pin-on-disk tribometry below for further details) were dissolved in hexane at a ratio of 10:3 and the total concentration of 0.5 % (w/w). Both coatings were cured in an oven at 70 °C overnight. Further details on the PDMS film coatings and their properties are available in previous publications [21].

All OWLS experiments were carried out in a BIOS-I instrument (ASI AG, Zürich, Switzerland) using a flow-through cell (8 × 2 × 1 mm, Dupont, Wilmington, DE) equipped with a Kalrez O-ring. The OWLS waveguides were first exposed to buffer solution (pH 7.4, HEPES 10 m plus 140 mM NaCl) until a stable baseline was obtained. Then, mucin solution was injected into the flow-cell. The change in refractive index reflects the adsorption of mucin, yet includes the contribution from the refractive index of bulk polymer solution itself. The adsorbed mass was thus measured only after the flow cell was rinsed with buffer solution. When a stable signal was obtained after buffer rinsing, adsorbed mass density data were calculated according to de Feijter's equation [22]. The 0.182 cm^3/g was used for the refractive index increment (dn/dc) value for both BSM and PGM.

C. Atomic Force Microscopy (AFM)

A conventional beam-deflection-based atomic force microscope (AFM) was employed to characterize the lubricating properties of the mucins adsorbed on PS surfaces on nanometer scale in aqueous environment. A commercial SPM scan head (Nanoscope® IIIa, MultiMode™, Veeco Instruments Inc.) equipped with a liquid cell/tip holder (Veeco Instruments Inc.) was used, and the fine movement of the sample, placed on top of the piezo scan tube, was controlled by SPM 1000 electronics and SPM 32 software (RHK Technology, Inc., Troy, MI), using an SPM Interface Module (RHK Technology, Inc., Troy, MI) to interface the scan head with the controller. A colloidal tip with PS particle (1 μm in diameter, Novascan Technologies Inc., Ames, IA) was used as the counterface to the PS-coated SiO_2 surfaces. The PS coating on SiO_2 substrate was carried out according to the same procedure for the coating on OWLS waveguides. To minimize potential damages to AFM probe during cleaning procedure, AFM tip-cantilever assembly was immersed in EtOH and then distilled water, 5 min. each, and then dried by evaporation, prior to use. The PS substrates were incubated in the mucin solutions (1 mg/ml) for 30 min, rinsed with 10 mM

HEPES buffer solution and then dried under a nitrogen stream. All measurements have been performed in 10 mM HEPES buffer solution.

The nanotribological properties of mucin have been characterized by the acquisition of "friction-vs-load" plots in a number of areas on each sample [23]. Briefly, the sample was laterally scanned relative to a stationary tip-cantilever assembly position in a line scan mode. Simultaneously, the sample was ramped up and then down in vertical direction to provide a variation in normal load. Both normal and lateral deflection of the tip-cantilever assembly generated from the interaction between the probe tip and the sample surface were detected by a position-sensitive detector, and they were interpreted as the normal load (converted based on the manufacturers' normal spring constant value, $k_N = 0.58$ Nm^{-1}) and the interfacial friction forces (raw photodiode signals, as shown in arbitrary units), respectively. By plotting the friction forces as a function of normal load, "friction-vs-load" plots were obtained. To ensure a valid comparison of the frictional properties of various samples, the same tip-cantilever assembly was used for all the measurements. Between the measurements on different samples, the tip/cantilever assembly was immersed in a surfactant solution, distilled water, for 5 min each, prior to dry by evaporation.

D. Pin-on-Disk Tribometry

The macroscopic scale lubricating properties of mucin solutions have been characterized by means of pin-on-disk tribometry (CSM, Neuchâtel, Switzerland) [5]. In this approach, a loaded pin was placed on a disk and the interfacial friction forces between them were measured while rotating the disk at controlled speeds. The load was controlled by dead weight and the sliding speed was controlled by a motor underneath the disk. The friction forces generated during sliding contacts were monitored by a strain gauge. The raw data consist of friction forces measured as a function of time or the number of rotations over a fixed sliding track.

In this work, sliding friction forces from the self-mated sliding contacts of PDMS lubricated by mucin solutions have been measured at various speeds (from 0.25 to 100 mm/s). The applied load was set at 1 N (corresponding to 0.51 MPa maximum Hertzian contact pressure) throughout this work. For the experiments to obtain μ (μ = friction/load) vs. speed plots (Fig. 7, below), the average friction force over a defined number of rotations (20) was obtained at each speed.

PDMS was employed for both pin and disk materials by employing a commercial silicone elastomer kit, SYLGARD® 184 elastomer kit (Dow Corning, Midland, MI, USA). The base and curing agent were thoroughly mixed at a 10:1 ratio (w/w), and the foams generated during mixing were removed using a mechanical pump. Then, the mixture was transferred into the molds for pin and disk and cured in an oven at ~70 °C overnight. A commercial polystyrene cell-culture plate with round-shaped wells (96 MicroWell Plates™, NUNCLON™ Delta Surface, Roskilde, Denmark) served as the mold for hemispherical PDMS pins, and a home-machined aluminum plate with flat wells was used as the mold for flat PDMS disks. The dimensions of the pin and disk fabricated in this way were 3 mm (radius) for the pins and 30 mm (diameter) × 5 mm (thickness) for the disks. Finally, it is noted that one pair of PDMS was used for only one measurement to avoid cross-contamination.

III. RESULTS AND DISCUSSION

A. Adsorption Properties of Mucins

Surface adsorption properties of lubricant additives, both amount and binding strength, are critical to determine the lubricating properties of lubricants, especially in boundary lubrication regime. Thus, the adsorbed masses of the mucins from 1 mg/ml solutions onto the polymeric surfaces were characterized first. An example of the adsorption profiles of BSM and PGM onto PS surfaces is shown Fig. 2.
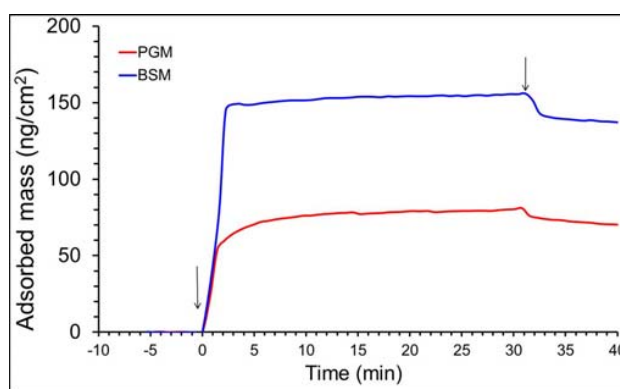

Fig. 2 Adsorption profiles of BSM and PGM onto PS-coated waveguides as measured by OWLS. The arrow at $t = 0$ indicates the injection of mucin solutions into OWLS flow cell, and the arrow at $t \sim 30$ min indicates the rinsing with buffer solution

As shown in Fig. 2, the adsorption of both mucins onto PS surfaces occurred very rapidly; a majority of the final adsorbed masses (the values obtained after rinsing with buffer solution at $t \sim 30$ min) was obtained within the first a couple of minutes. The adsorbed mass from the data shown in Fig. 2 are ca. 137 ng/cm^2 for BSM and ca. 70 ng/cm^2 for PGM. It is noted that the adsorbed mass of BSM is substantially higher than PGM under physiological condition.

The same experiments were carried out for the adsorption properties of the mucins onto PDMS surfaces. It was of interest in this work to study the comparative lubricating properties of the mucins at pH 7 and 2, since a superior lubricity was observed from PGM at pH 2 than at pH 7 in a previous study [5]. Thus, the adsorption properties of the mucins were also obtained from both pH conditions, pH 2 and pH 7. The surface adsorption profiles of the mucins onto PDMS surfaces were similar to those onto PS surfaces, i.e. rapid initial adsorption followed by very slow further increase in mass, and the final results only are given in the Fig. 3.
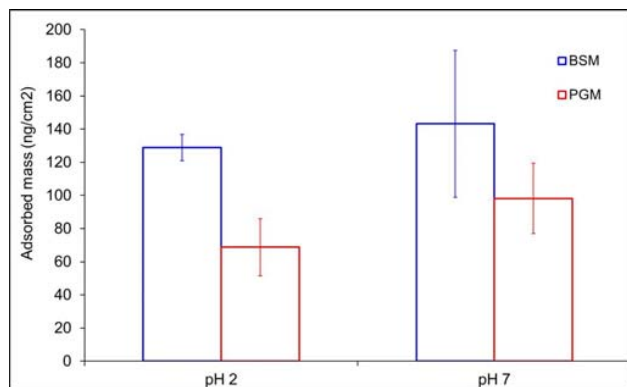
Fig. 3 The adsorbed masses of BSM and PGM onto PDMS surfaces at pH 2 and pH 7 as determined by OWLS

For BSM, 143.3±44.3 and 128.9±7.9 ng/cm^2 were obtained at pH 7 and 2, respectively, whereas for PGM, 98.2±21.3 and 68.7±17.2 ng/cm^2 were obtained at pH 7 and 2, respectively (error bars obtained from the standard deviations over three measurements for each sample). Thus, higher adsorbed mass from BSM than PGM is consistent for both polymeric surfaces as well as for both pH conditions.

B. Lubrication with Mucin Coatings by AFM

In this work, the assessment of the lubricating properties of the mucins has been carried out for a number of sliding contact situations that can occur for moving biomedical devices; (a) nanoscale contacts with mucin coatings (b) macroscale contacts with injectable lubricants (c) macroscale contacts with lubricant solutions. In Fig. 3, the lubricating properties of the two mucin *coatings* on PS surfaces as characterized by AFM are presented.
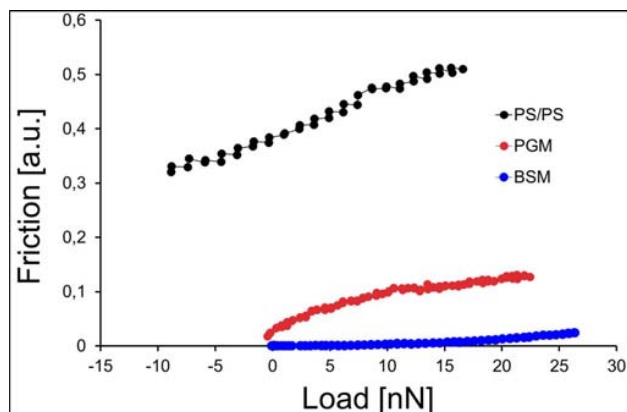

Fig. 4 Friction vs. load plots from the sliding contacts between PS/PS surfaces in buffer solution, either between bare substrates (black symbols) or coated with PGM (red symbols) or BSM (blue symbols)

Both mucin coatings showed significant reduction in friction forces compared to bare PS/PS substrates, which suggest that these mucins have a potential as a lubricant coating under nanoscopic scale contacts, for example, insertion of biosensors made of polymeric materials into

human body. It is further noted that BSM is more efficient than PGM in reducing the frictional forces between PS surfaces. This is consistent with higher adsorbed mass achieved by BSM than PGM as shown in Fig. 2. The AFM experiment on PDMS surface was not performed due to too high adhesive forces involving bare PDMS surface.

C. Lubrication by Means of Injection

Very fast surface adsorption kinetics of the mucins as observed from OWLS studies (Fig. 2 and 3) implies that these mucin solutions have a potential to be used as injectable lubricants where rapid lubrication of polymeric biomaterials is necessary. A rapid lubricating effect following rapid adsorption of mucin solution was verified by a control experiment of injecting highly concentrated mucin solutions into the tribocup where the sliding contacts between PDMS/PDMS is already taking place in buffer solution. An example is shown in Fig. 5.
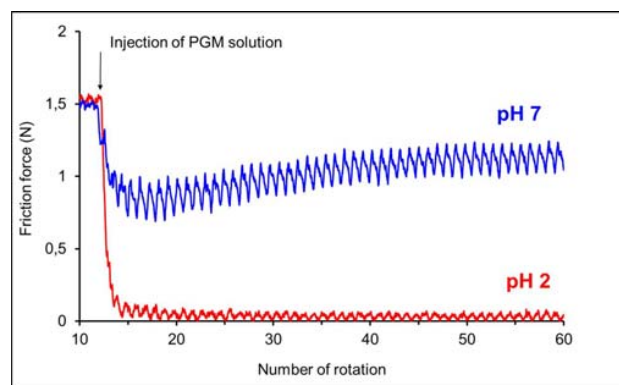

Fig. 5 Friction vs. number of rotations for the sliding contacts between PDMS/PDMS surfaces in buffer solution, followed by injection of mucin solutions (at ~ 12th rotations)

As shown in Fig. 5, the reduction of friction forces between PDMS/PDMS surfaces upon injection of PGM solution (the final concentration by dilution in buffer solution is 1 mg/ml) into the tribocup occurred immediately after the injection (within a couple of rotations). Further, the data shown in Fig. 5 confirms that the superior lubricating properties of PGM at pH 2 compared to pH 7 [5] has a characteristics of very fast surface adsorption kinetics.

D. Lubrication with Solution

Finally, the lubricating properties of both PGM and BSM at both pH 7 and 2 have been characterized by employing 1 mg/ml of the mucin solutions. Generally, the lubrication by solution is significantly different from the lubrication by a coating film in that excess molecules in solution can be readily adsorbing onto the area where pre-adsorbed molecules are worn-off by tribostress, and thus "self-heal" the lubricating efficacy [24]. This type of lubrication is ideal for repeated, cyclic tribostress is applied, such as in articulating joint implants. However, a basic requirement for "self-healing" mechanism is fast surface adsorption kinetics by

lubricant additives. For the mucins studied in this work, this behavior is confirmed in Fig. 5. The μ vs. speed plots are shown in Fig. 7.

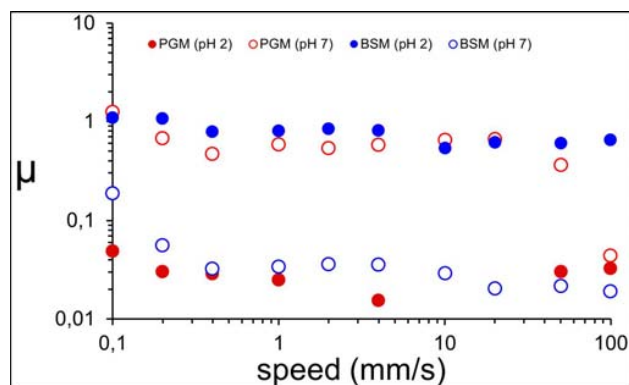
Fig. 6 The μ vs. speed plots obtained from PDMS/PDMS sliding contacts lubricated with PGM and BSM solutions (1 mg/ml) at pH 2 and pH 7. Some missing data points for PGM at pH 2 (for example, 10 and 20 mm/s), the μ values were lower than 0.01

As shown in Fig. 6, the lubricating effects by BSM and PGM were manifested differently under different pH conditions. For PGM, for example, superior lubricating behavior was observed at pH 2 than pH 7, consistently with the previous study [5]; at pH 2, extremely low μ values of from ca. 0.04 to lower than 0.01 (some missing data points in Fig. 10 and 20 mm/s), which represents the reduction in μ by approximately two orders of the magnitude on average compared to the buffer solutions (data not shown)). Meanwhile, the influence of pH on the lubricating properties of BSM solutions was observed to be opposite to those of PGM solutions; excellent lubricating properties from the BSM solution were observed from at pH 7, but not at pH 2.

The results in Fig. 7 firstly indicate that both PGM and BSM have optimized their lubricating functions according to physiological demands. However, the pH dependence of the two mucins shown in Fig. 7 cannot be explained simply by the adsorbed masses in each corresponding condition, as shown in Fig. 3. Despite contrasting pH dependence of the lubricating properties of BSM and PGM, adsorbed mass was consistently higher at pH 7 than pH 2 for both types of mucins. As was discussed in the previous study [5], this observation further supports that adsorbed mass is not the sole parameter to determine the lubricating efficacy of the mucins, and yet the strength of adsorption may play more significant role in boundary lubrication. Further studies to understand the composition, structure, and conformational changes of these two types of mucins in order to understand the complex lubricating behavior as a function of pH are required in the future.

IV. CONCLUSION

In this work, it was demonstrated that commercially available mucins have a potential to be applied as slippery coatings for polymeric biomaterials, such as tissue contacting devices or biosensors for its excellent lubricity at hydrophobic surfaces. Despite that these commercial mucins are known to contain various non-mucinous components due to incomplete purification process by the manufacture, they are economical and easy to use. Most of all, they are of biological origin that favorable biocompatibility is expected. For PS/PS contacts, both mucins showed feasible lubrication on nanoscale contacts as characterized by AFM. This might be helpful for applying small-scale contacts, such as micron-sized sensors etc. Macroscale, soft contacts provided by PDMS/PDMS contacts have shown that the lubricating properties of the two mucins, BSM and PGM, are substantially different in efficacy at pH of 2 and 7, BSM being more slippery at pH 7 and PGM being more lubricious at acidic pH, which are correlated with their genuine physiological conditions requiring lubrication.

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