Journal of the Korean Chemical Society
2024, *Vol. 68, No. 5*
Printed in the Republic of Korea
https://doi.org/10.5012/jkcs.2024.68.5.243

Communications

Adhesive-free Template Stripping of Gold Thin Films for Metallic Ultraflat Surface

Minju Park and Intek Song*
Department of Chemical and Biological Engineering, Andong National University,
1375 Gyeongdong-ro, Gyeongbuk 36729, Korea.
**E-mail: songintek@anu.ac.kr*

(Received July 8, 2024; Accepted August 10, 2024)

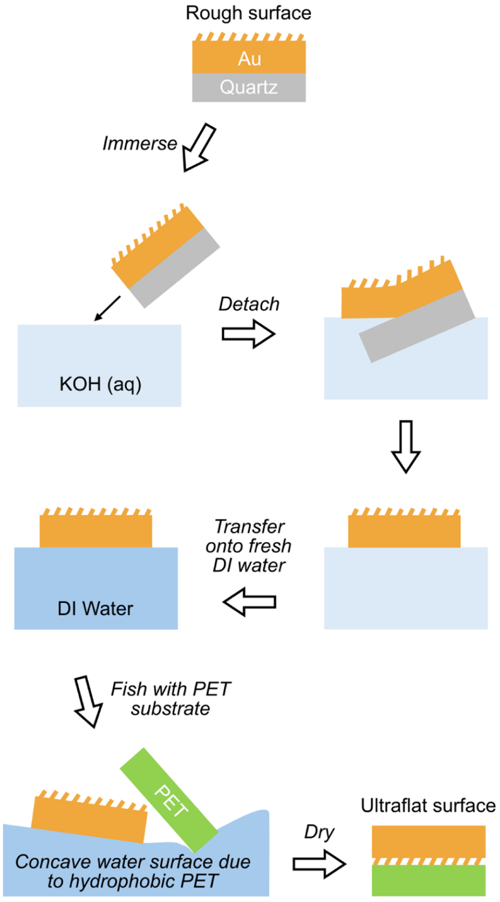
Key words: Template stripping, Transition metal, Thin film, Electrical characterization, Surface tension, Hydrophobicity

The preparation of ultraflat surfaces is critically important in surface chemistry and device fabrication due to their profound impact on the physical and chemical properties of materials. Ultraflat surfaces reduce nucleation sites for heterogeneous nucleation, enhancing crystal quality by allowing the growth of larger grains. Additionally, these surfaces simplify morphological analysis, facilitating clear differentiation between authentic surface features and anomalies such as defects or impurities.[1] The preparation of ultraflat surfaces is essential in surface chemistry and device fabrication due to their significant impact on material properties.[2,3] Ultraflat surfaces minimize heterogeneous nucleation sites, enhancing crystal quality by promoting the growth of larger grains. These surfaces also simplify morphological analysis, allowing clearer differentiation between genuine surface features and anomalies such as defects or impurities.

Traditional methods for preparing flat thin films, such as electron-beam (e-beam) evaporation and sputtering, have been refined to achieve a surface roughness of less than tens of nanometers, which is adequate for many applications. However, these techniques often do not yield surfaces as flat as meticulously polished substrates like silicon wafers or atomically flat mica. The clustering of evaporated metals often compromises the ultimate flatness of these surfaces.

An alternative, the template stripping technique, has proven highly effective for achieving ultraflat surfaces.[3–5] This method involves mechanically stripping a metal, such as gold, deposited onto flat substrates. The success of this technique hinges on the weak interaction between the gold and the substrate, allowing the gold's surface morphology to precisely mirror that of the substrate. Typically, gold surfaces prepared via this method exhibit a mean roughness of less than 5 Å across areas of 25 μm², demonstrating exceptional flatness.[5] Consequently, template-stripped metals are widely used in surface chemistry reactions, including self-assembled monolayers formation, and are particularly advantageous in flexible electronics.

Despite its benefits, traditional template stripping encoun-



***Scheme* 1.** A schematic diagram illustrating the simple transfer procedure of the adhesive-free template stripping method.

ters challenges due to the use of non-removable adhesives like epoxy, which limit the utility of the specimens, especially under conditions involving high temperatures or exposure to organic solvents.

This research presents a novel and simple transfer process for preparing ultraflat surfaces without relying on complex techniques or adhesives, thereby increasing the potential for future applications. The method utilizes meniscus behavior to capture floating gold thin films with hydrophobic substrates (*Scheme* 1). This study enhances the limitations of the previous method by enabling the use of a pristine polyethylene terephthalate (PET) substrate for transfer, which avoids the need for harsh HF treatment and simultaneously offers flexibility and electrical insulation.[6] It was observed that the specimens prepared using this method not only exhibit an ultraflat surface but also maintain the conductivity of the original gold thin film.

Initially, a gold thin film was deposited on an ultraflat substrate, either SiO_2(300 nm)/Si (single side polished, oxide uniformity below 5%, from Namkang Hi-Tech) or fused silica (double side polished, 0.5 mm thickness, from Hi-Solar) (*Fig*. 1a). The deposition was carried out using either an e-beam metal evaporator or a sputtering machine. In the case of the e-beam metal evaporator, the deposition rate was maintained at 1 Å/s, under conditions of less than 10^{-6} torr pressure, with the temperature not exceeding 80 ℃. For the sputtering method, utilizing a Cressington Sputter Coater 108auto, gold was sputtered for 80 seconds. Subsequent analysis of the thin films was conducted using atomic force microscopy (AFM) with a NanoscopeIII instrument (Digital Instrument Inc.). It was observed that the thin films achieved a thickness of 31 nm.

After deposition, the gold film on the substrate was partially immersed in a 1 M aqueous solution of either NaOH or KOH (*Scheme* 1). This basic solution causes the gold film to detach and float on the surface of the solution. The detached gold film was then transferred onto fresh deionized (DI) water to eliminate any residual base. This rinsing process was repeated twice to ensure thorough removal of all residues. Following the cleaning steps, the gold film was transferred to a fresh hydrophobic surface. Options for the hydrophobic substrate include a hydrogen-terminated silicon (H-Si) substrate or a PET substrate. Especially, the PET substrate was cleaned by sequentially rinsing it with deionized water, isopropyl alcohol prior to the transfer. Finally, the film was air-dried to enhance its adhesion to the substrate (*Fig*. 1b and c).

A distinctive feature of this method is the "flip" of the gold thin film when captured by hydrophobic surfaces. (*Scheme* 1) As the gold film contacts the water, the meniscus around the hydrophobic surface becomes convex, aiding in the film's capture without direct contact. This ensures that the rougher surface of the gold faces the hydrophobic substrate, exposing an ultraflat gold surface. Indeed, when a hydrophilic substrate was used as the fishing substrate, it always failed at flipping the floating film due to the concave surface made by the substrate.

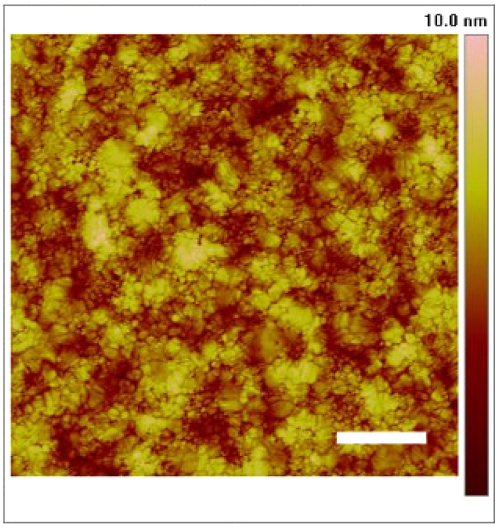


***Figure* 2.** An AFM image of the surface of the transferred gold thin film, with the roughness of is 0.981 nm. The lateral dimension is 5 μm × 5 μm. The scale bar denotes 1 μm.

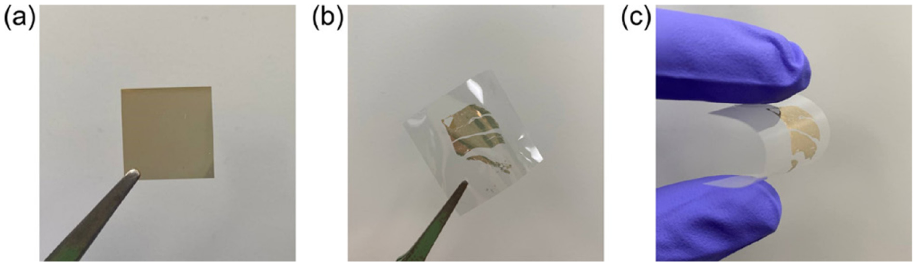


***Figure* 1.** Photographs of a gold thin film deposited on a quartz substrate (1.5 cm × 1.5 cm) before the transfer (a), and after the transfer onto a flexible PET (25 μm) substrate (b, c).

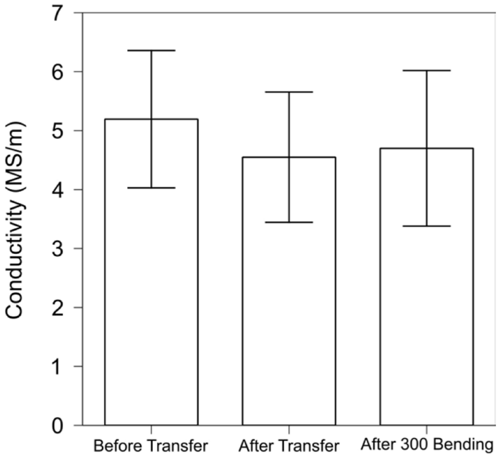


***Figure* 3.** Statistics of four-point-probe measurement data at room temperature: probe spacing of 1.27 mm, target current of 0.1 mA, maximum voltage of 1 V, voltage increment of 0.001 V, and sample thickness of 0.031 μm.

The surface roughness of the specimens was analyzed by atomic force microscopy (AFM; Nanoscope IIIa, Digital Instrument Inc.). The specimen prepared using this new method is 0.981 nm, which was 0.875 nm before the transfer (*Fig*. 2). Moreover, the ultraflat gold thin film retains flexibility once transferred onto a flexible PET substrate (*Fig*. 1c).

A key application of template-stripped gold is in electrodes for electrical devices, since conventional metal evaporation techniques can deteriorate the performance of the devices.[7] Leveraging the insulating and flexible properties of the PET substrate, the impact of the fabrication process on conductivity was examined (*Fig*. 3). The electrical properties of these devices were assessed through conductivity measurements using a four-point probe method, performed with an Ossila T2001A3 machine. The results revealed that the conductivity of the specimens prepared by the polymer-free template stripping method is comparable to that of the pre-stripped films. Furthermore, the conductivity was maintained after 300 cycles of repeated bending with displacement of 1.15 cm, facilitated by a custom-made machine.[8] This demonstrates that the stripping process preserves mechanical integrity and ensures robust adhesion between the gold thin film and the PET substrate. This confirms that the new method maintains the conductivity of the gold thin film, demonstrating its effectiveness in producing ultraflat yet highly conductive thin films suitable for future applications.

In conclusion, this novel method for preparing ultraflat surfaces by capturing floating gold thin films on hydrophobic substrates demonstrates advancements over traditional techniques. It simplifies the fabrication process, retains the conductivity and ultraflat morphology of gold films, and eliminates the use of adhesives, thereby enhancing the potential for broader applications, particularly in flexible electronics. Future work could focus on refining this technique for other metallic films and expanding its integration into technologies like sensors and optoelectronics, potentially increasing scalability and reproducibility.

Acknowledgments. This work was supported by a Research Grant of Andong National University.

REFERENCES

1. Hegner, M.; Wagner, P.; Semenza, G. *Surf. Sci.* **1993**, *291*, 39.
2. Song, I. *Bull. Korean Chem. Soc.* **2023**, *44*, 495.
3. Yoo, D.; Johnson, T. W.; Cherukulappurath, S.; Norris, D. J.; Oh, S. H. *ACS Nano* **2015**, *9*, 10647.
4. Vogel, N.; Zieleniecki, J.; Köper, I. *Nanoscale* **2012**, *4*, 3820.
5. Wagner, P.; Hegner, M.; Guentherodt, H. J.; Semenza, G. *Langmuir* **1995**, *11*, 3867.
6. Song, I. *Studies on the Surface Chemistry for the Controlled Growth of Two-Dimensional Nanomaterials*. Ph. D. Thesis, Pohang University of Science and Technology, Pohang, Korea, 2018.
7. Cho, U.; Kim, S.; Shin, C. Y.; Song, I. *ACS Omega* **2022**, *7*, 21220.
8. Jeong, B. Y.; Lee, S.; Shin, H. H.; Kwon, S.; Kim, S. H.; Ryu, J. H.; Yoon, S. M. *ACS Mater. Lett.* **2022**, *4*, 1944.