Introduction

Gold is not oxidized in air or in molecular oxygen, even at high temperatures, because its oxide (Au₂O₃) is unstable. However, gold is oxidized in highly reactive chemical environments, e.g., during reactive sputtering in an oxygen atmosphere, ozone, and oxygen plasma. Oxides were also prepared using an air corona discharge and by laser deposition in oxygen. Gold oxide formation and decomposition on the gold electrodes of microelectromechanical system devices after the release of an organic sacrificial layer via exposure to an oxygen plasma have also been studied. Gold oxides decompose after 36 h at room temperature and decompose immediately at temperatures exceeding 117°C. They also decompose under ultraviolet light irradiation (254, 302, and 365 nm) and decompose more rapidly in water vapor at the shorter wavelengths. The thicker nature of the gold oxides is advantageous for their preservation, and they were preserved in their oxidized state for 196 d in anhydrous dodecane in a dark atmosphere.

Keywords Gold oxide, oxygen-dc glow discharge, gold discharge ring, thermal decomposition, UV light decomposition, water molecule, dodecane, preservation, XPS

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plasmon resonance (SPR)-based optical waveguide sensing system and XPS in our previous work. The decomposition reactions of these thin gold oxide layers on gold films in aqueous solutions of acetaldehyde, hydrochloric acid, and sodium hydroxide were monitored and the oxide decomposition rates were also obtained via the SPR-based optical waveguide sensing system for the first time.

In this work, thicker gold oxide layers were prepared by varying the duration of the oxygen-dc glow discharge while using a gold discharge ring, in contrast to the use of an aluminum discharge ring in the previous work. The decomposition of these gold oxides, which can be caused by heating in air, UV light irradiation and the passage of time, under a variety of atmospheres, was studied by XPS. The angular dependences of the XPS spectra provided information on the chemical species involved and on the surface structures of the gold oxides on the gold films. These results were compared with those obtained when using an aluminum glow discharge ring to clarify the behavior of this unique oxide metal. The preservation method of immersing the gold oxide on the gold film in a saturated hydrocarbon with low water content at room temperature in a dark atmosphere was performed using anhydrous dodecane because the thicker nature of these gold oxides is expected to be advantageous for their preservation.

### Experimental

#### Gold oxide preparation

Gold (>99.99%; Tanaka Kikinzoku Kogyo, Tokyo, Japan) was deposited on mica substrates (dimensions: 9 × 18 × 0.1 mm; Nissin EM, Tokyo, Japan) at room temperature under high vacuum (<6.7 × 10⁻⁴ Pa) conditions to form 50-nm-thick films. These gold films were then oxidized at room temperature using an oxygen-dc glow discharge (10 Pa, 5 mA, 0.17 – 30 min) from a high-purity oxygen (>99.999%; Japan Fine Products, Kawasaki, Japan) flow in a vacuum evaporator equipped with a negatively biased (to approximately 400 V) gold (>99.99%) electrode.

#### Heating and UV light irradiation of gold oxides

The gold oxide layers on the gold films were heated at various temperatures (93 – 184 °C) for times of 0.5 and 3 h in an electric dryer (SOFW-300, AS ONE, Osaka, Japan). An iron-constantan thermocouple (Nippon Netsudenci Seisakusho, Tokyo, Japan) was placed in contact with the sample and used to measure the temperatures of the oxides; the measurement uncertainty was estimated to be ±3 °C.

The gold oxide layers were then irradiated for 6 h at room temperature under various atmospheres through a quartz window in another vacuum chamber equipped with UV lamps (operating wavelengths of 254, 302, and 365 nm, 6 W; Ultra-Violet Products UVM-57, Funakoshi, Tokyo, Japan). The atmospheres were air (1.0 × 10⁻¹⁴ Pa), water vapor (1.5 × 10⁻¹⁴ Pa), and a high vacuum (1.8 – 4.7 × 10⁻⁴ Pa).

#### Preservation of gold oxides

The gold oxide layers were preserved for up to 197 d at room temperature in a dark atmosphere. The oxides were also preserved for up to 196 d in sealed 30 mL glass bottles that contained anhydrous dodecane and approximately 150 mg molecular sieves (4A 1/8, Fujifilm Wako Pure Chemical, Osaka, Japan) at room temperature in a dark atmosphere.

### XPS measurements

The XPS spectra of the gold oxide layers on the gold films were measured at a resolution of less than 0.6 eV using a photoelectron spectrometer (Axis-Ultra DLD, Shimadzu, Kyoto, Japan) with monochromatic Al Kα (1486.6 eV) radiation. These spectra were measured at angles of 90, 30, and 10° from the surfaces (dimensions: 0.7 × 0.3 mm) at data collection intervals of 0.1 eV. The spectra that were measured at 90° are shown in this work, unless a different measurement angle is specified. The O 1s spectra were accumulated several times. Various locations were measured on the oxide layer surfaces as part of a series of measurements. Voigt functions (Gaussian and Lorentzian convolution functions) were used, after subtraction of the Shirley background, for deconvolution of the Au 4f and O 1s peaks. The intensity ratio of the Au 4f to O 1s was 4/3 and the Au 4f 7/2 line at 84.0 eV was used to calibrate the binding energy. Use of a low-energy electron flood gun for charge compensation was not required. Impurities that originated from neither the gold used (e.g., Ag, Cu, Ca, Si) nor the glow discharge (e.g., Al, Fe, F) were detected on the gold oxide surfaces. Carbon impurities that incorporated oxygen atoms from the air were detected. Their effects on the O 1s spectra of the gold oxides prepared by the oxygen-dc glow discharge (10 min) were estimated to be less than 2%.

The thicknesses (d) of the gold oxide layers on the gold films were determined from the relative intensities (Iₓ/Iₒ) of both pairs of oxide (ox) peaks with respect to the metal (m) peaks of Au 4f in the spectra that were measured at θ = 90° using the following equation:

$$d = \lambda_{ox} \times \sin \theta \times \ln \left( \frac{I_{ox}}{I_{m}} \right) \left( \frac{D_{ox}}{D_{m}} \right) \frac{\lambda_{ox}}{\lambda_{m}} + 1 \right),$$

where \(\lambda_{ox}(1.937 \text{ nm})\) and \(\lambda_{m}(1.781 \text{ nm})\) are the photoelectron inelastic mean free paths in Au₂O₃ and Au, respectively.

Atomic densities of \(D_{m}\) (19.32 g/cm³) and \(D_{ox}(13.675 \text{ g/cm³})\) for Au and Au₂O₃ were used, respectively. The atomic ratio \([O]/[Au]\) of the gold oxide (Au₂O₃) was calculated based on the intensities of oxide component III of O 1s and those of the Au 4f oxide peaks using the XPS spectrometer’s software. The peak positions for Au 4f and O 1s and the full width at half

| Peak       | Energy/eV | FWHM/eV |
|------------|-----------|---------|
| Au 4f₂₂ (metal) | 84.0 | 0.8 | 0.9 |
| Au 4f₂₂ (oxide) | 85.8 | 1.6 | 1.2 |
| Au 4f₂₂ (metal) | 87.7 | 0.8 | 0.9 |
| Au 4f₂₂ (oxide) | 89.5 | 1.6 | 1.2 |
| O 1s (component I) | 531.9 | 2.1 | 2.1 |
| O 1s (component II) | 531.1 | 1.5 | 1.5 |
| O 1s (component III) | 530.0 | 1.7 | 1.7 |
| O 1s (component IV) | 530.2 | 1.2 |       |

The spectra were measured at angles of 10, 30, and 90° to the surfaces. Component I for 20 and 30 min is too weak to observe at angle of 90°. The numbers of data are 10 for component IV and more than 20 for the others and the uncertainties are less than ±0.1 eV.
maximum (FWHM) values of the gold oxides obtained in this study are shown in Table 1. The values are averages of those of 10 samples for component IV and those of more than 20 samples for the others, and uncertainties are less than ±0.1 eV. The FWHM values for O 1s are independent of the discharge time. However, the intensities of both the component III and the pair of Au 4f peaks increase, but the intensity of component I decreases as the discharge duration increases.

Results and Discussion

Formation of gold oxides by an oxygen-dc glow discharge

The XPS spectra of Au 4f and O 1s for gold oxide layers prepared from gold films by an oxygen-dc glow discharge for various periods (0.17, 1, 5, 20, and 30 min) at room temperature. O 1s components I, II, and IV are present, but neither O 1s component III nor the pair of oxide Au 4f peaks is present for the film prepared with the 0.17 min period. Component IV changes into component III, which appears along with the pair of oxide Au 4f peaks for the film prepared with the 1 min period. The intensities of both the component III and the pair of Au 4f peaks increase, but the intensity of component I decreases as the discharge duration increases.

The Au 4f and O 1s spectra for the gold oxide layer prepared by the oxygen-dc glow discharge for a period of 0.5 min and measured at angles of 90, 30, and 10° relative to the surface. The relative intensity of O 1s component I becomes stronger while that of component IV becomes weaker as the measurement angle decreases. The oxygen species of components I and II are present in this order from the top surface of component IV.

The Au 4f and O 1s spectra for the gold oxide layer prepared by the oxygen-dc glow discharge over various periods (0.17, 1, 5, 20, and 30 min) are shown in Fig. 1. The Au 4f spectra show the 4f7/2 and 4f5/2 pair of strong metal peaks at 84.0 and 87.7 eV, respectively, and their FWHM values are the same as those of the metal peaks observed in the XPS spectra of gold. The O 1s components I, II, and IV are present at 531.9, 531.1, and 530.2 eV, respectively, but neither O 1s component III nor the pair of oxide Au 4f peaks is present for the discharge durations of 0.17 and 0.5 min. The intensity of component I is stronger than the other components during these early formation periods. Component IV at 530.2 eV changes into component III at 530.0 eV after a discharge duration of 1 min, at which the pair of oxide Au 4f peaks at 85.8 and 89.5 eV also appears. The intensities of both component III and the pair of Au 4f peaks increase, but the intensity of component I decreases as the discharge duration increases. The intensity of component I becomes weak for 5 min and it disappears after 20 min. The FWHM values of the O 1s components remain independent of the discharge time, but the FWHM values for the Au 4f metal increase slightly, while those of the oxide decrease after the discharge time of 30 min, as shown in Table 1.

The Au 4f and O 1s spectra for the gold oxide layer prepared by the oxygen-dc glow discharge for a period of 0.5 min at room temperature and measured at angles of 90, 30, and 10° relative to the surface. The relative intensity of O 1s component I becomes stronger while that of component IV becomes weaker as the measurement angle decreases. The oxygen species of components I and II are present in this order from the top surface of component IV.
period of 30 min and measured at angles of 90°, 30°, and 10° from the surface are shown in Fig. 3. The relative intensities of the oxide peaks of Au 4f becomes stronger while that of O 1s component III becomes weaker as the measurement angle decreases. O 1s component I is observed at the angle of 30° and the intensity becomes slightly stronger at the angle of 10°. The oxygen species of components I and II are present in this order from the top surface of component III.

The thicknesses of the gold oxide layers and the relative intensities of O 1s components I, II, III, and IV prepared by an oxygen-dc glow discharge for periods of up to 30 min are shown in Fig. 4. The thicknesses of gold oxide layers prepared by an oxygen-dc glow discharge using an aluminum discharge ring are also shown for comparison purposes. The spectra were measured at an angle of 90° to the surfaces. The relative intensity of component I is stronger than the other components during the early formation periods of 0.17 and 0.50 min. Component IV changes into component III after the discharge duration of 1 min. The thicknesses of the gold oxide layers correlate well with the relative intensities of oxide component III. The thicknesses of gold oxide layers prepared by the oxygen-dc glow discharge using the gold discharge ring increased significantly with increasing discharge time after 10 min, in contrast to the results obtained when using the aluminum ring.

| Discharge time/min | Gold oxide thickness/nm | Atomic ratio [O]/[Au] | Ratios of O 1s components |
|--------------------|-------------------------|-----------------------|--------------------------|
| 5                  | 0.56 ± 0.11             | 1.30 ± 0.07           | 0.13 ± 0.03, 0.16 ± 0.02, 0.71 ± 0.04 |
| 30                 | 2.02 ± 0.25             | 1.32 ± 0.09           | 0 ± 0, 0.21 ± 0.03, 0.79 ± 0.03 |

The numbers of data for discharge times of 5 and 30 min are 7 and 17, respectively, and the uncertainties are the standard deviations.

When using the aluminum ring, it is considered that the gold oxide layer produced on the gold film acts as a barrier and hinders further oxide growth when using the aluminum discharge ring. No aluminum contamination from the aluminum ring was detected. In contrast, the gold oxide layer formed from the gold ring by sputtering of oxygen accumulates over time and its thickness increases when using the gold discharge ring.

The thickness and the atomic ratio [O]/[Au] of the gold oxide
(Au₂O₃) layers prepared by the oxygen-dc glow discharge for the 5 min duration were 0.56 ± 0.11 and 1.30 ± 0.07 nm, respectively. The corresponding values for the 30 min period were 2.02 ± 0.25 and 1.32 ± 0.09 nm, respectively. These results are presented along with the ratios of the O1s components in Table 2. The value of the atomic ratio is close to the value of 1.5 for Au₂O₃. These findings indicate that the Au 4f oxide peaks and component III of the O 1s spectra can be assigned to those of Au₂O₃. The gold oxide layers can be prepared by the simple oxygen-dc glow discharge treatment and the layer thicknesses on the gold films can be controlled easily using the treatment duration, as shown in Fig. 4(a).

Thermal decomposition of gold oxides

The XPS spectra of Au 4f and O 1s for the gold oxide layers that were prepared by the oxygen-dc glow discharge over the period of 30 min and then preserved for various periods (1, 7, 15, and 191 d) at room temperature in a dark atmosphere are shown in Fig. 5. The spectra measured within 1 h after preparation (0 d) of the layers are also shown. The gold oxide decomposes after 15 d and the spectra show neither O 1s component III nor the pair of oxide Au 4f peaks. The O 1s spectrum contains components I and II. The O 1s spectra of the gold oxides obtained after the longer periods (141 – 197 d) only contain component I.

The XPS spectra of Au 4f and O 1s from the gold oxide layers that were prepared by the oxygen-dc glow discharge over the 30 min period and then heated for 3 h at various temperatures (93, 117, 142, and 162°C) in a dark atmosphere are shown in Fig. 6. The corresponding spectra of a gold oxide layer that was not subjected to heating and was instead preserved in a dark atmosphere for the same period are also shown for comparison purposes. The gold oxides decomposed at temperatures above 117°C and the spectra show neither the Au 4f oxide peaks nor oxide component III.
not heated and was instead preserved in a dark atmosphere for the same period are also shown for comparison purposes. No oxide peaks are present in the Au 4f spectra at temperatures above 117°C, thus indicating that the decomposition of the oxide layers begins at this temperature. However, the O 1s spectra show the peaks of components I and II. The peak of component I appears as a result of decomposition of the gold oxide above 93°C. The gold oxides decomposed completely after heating for 0.5 h at 142, 162, and 184°C, but only decomposed in an incomplete manner during heating for 0.5 h at 117°C. The decomposition of the gold oxide prepared by an oxygen-dc glow discharge for a 10 min period using the aluminum discharge ring was observed to begin at 121°C in our previous studies. These temperatures are effectively the same within the range of the experimental error. The gold oxides in these studies that were heated at 250, 300, 400, and 500°C for 3 h in a high vacuum decomposed completely and the O 1s spectra contained only components I and II. Subtraction of components I and II from the O 1s spectrum of the unheated gold oxide then provides component III of the oxide peak, as shown in Table 1.

The O 1s components I and II of the gold oxides in this study are both present on oxide component III and remain sufficiently stable for 15 d and during heating at temperatures up to 184°C in air. Component I is also present on the gold surface, even for long periods of up to 197 d. These findings are consistent with the properties of the hydroxyl groups on the model of the oxide surface; the oxide film consists of an outermost layer composed of chemisorbed water, a hydroxylated region, and then an innermost portion composed of the oxide film. These findings are also consistent with the results that were obtained in theoretical studies of the reaction of water on oxygen-covered gold surfaces of Au (111) and Au (100). The energy barrier for water decomposition decreases sharply on the pre-covered oxygen atoms on the gold surface and these oxygen atoms promote dehydrogenation of the water molecules. In contrast, dissociation of the hydroxyl group on the gold surface is difficult and it is thus adsorbed stably on the gold surface.

**UV decomposition of gold oxides**

The XPS spectra of Au 4f and O 1s for the gold oxides that were prepared by the oxygen-dc glow discharge for the period of 30 min and subsequently irradiated using UV light (254, 302, and 365 nm) for 6 h in a dark atmosphere in a vacuum chamber (1.0 × 10⁵ Pa) are shown in Fig. 7. The corresponding spectra of a gold oxide layer that was not irradiated and was instead preserved in a dark atmosphere for the same period are also shown for comparison purposes. The gold oxide layer that was
The number of data was either 6 or 12 and the uncertainties are the standard deviations. The ratios were calculated based on the intensities of the Au 4f peaks and O 1s component III, even after 196 d. No contaminants were observed. The XPS spectra of Au 4f and O 1s for the gold oxides that were prepared by the oxygen-dc glow discharge for 30 min and irradiated at 302 nm for 6 h under various atmospheres are shown in Fig. 8. The corresponding spectra of a gold oxide that was not irradiated using UV light and was instead preserved in a dark atmosphere for the same period are also shown for comparison purposes. The irradiated gold oxide decomposed completely in the water vapor atmosphere. The oxide also decomposed in air, although incompletely, but showed almost no decomposition under the high vacuum. The gold oxide that was preserved in the water vapor atmosphere without UV irradiation for the same period showed no decomposition.

The ratios of the gold oxide layers after UV light irradiation at wavelengths of 254, 302, and 365 nm for 6 h in various atmospheres are presented in Table 3. These ratios were calculated based on the intensities of the Au 4f peaks of the irradiated gold oxide layers and on those of the gold oxide samples that were preserved for 6 h in the dark atmosphere. The number of data was either 6 or 12 and the uncertainties shown are the standard deviations. Table 3 clearly shows that the presence of water and UV light irradiation both promote the decomposition of the gold oxides. The table also illustrates the wavelength dependence of the decomposition process, where the gold oxides decompose more quickly after irradiation at shorter wavelengths. It was concluded that the reactions of the gold oxides decompose more quickly after irradiation at shorter wavelengths. The present study has shown that water and UV irradiation both promote the decomposition of the gold oxides. Therefore, gold oxides can be preserved for longer periods by removing both water and UV irradiation. Gold oxide samples were thus immersed in anhydrous dodecane (melting point: –10°C; 84% water content: <0.003%35) at room temperature in a dark atmosphere. The corresponding spectra of a gold oxide that was not immersed in anhydrous dodecane and was measured within 1 h after initial preparation (0 d) are also shown. The gold oxide was preserved in the oxidized state for 196 d in anhydrous dodecane.

The gold oxides were shown to decompose slowly at room temperature in a dark atmosphere. However, they decompose rapidly in a water vapor atmosphere under UV light irradiation (254, 302, and 365 nm) and also demonstrated faster decomposition under UV irradiation at shorter wavelengths. The present study has shown that water and UV irradiation both promote the decomposition of the gold oxides. Therefore, gold oxides can be preserved for longer periods by removing both water and UV irradiation. Gold oxide samples were thus immersed in anhydrous dodecane (melting point: –10°C; 84% water content: <0.003%35) at room temperature in a dark atmosphere. The corresponding spectra of a gold oxide that was not immersed in anhydrous dodecane and was measured within 1 h after initial preparation (0 d) are also shown. The gold oxide was preserved in the oxidized state for 196 d in anhydrous dodecane.

**Preservation of gold oxides in dodecane**

The gold oxides were shown to decompose slowly at room temperature in a dark atmosphere. However, they decompose rapidly in a water vapor atmosphere under UV light irradiation (254, 302, and 365 nm) and also demonstrated faster decomposition under UV irradiation at shorter wavelengths. The present study has shown that water and UV irradiation both promote the decomposition of the gold oxides. Therefore, gold oxides can be preserved for longer periods by removing both water and UV irradiation. Gold oxide samples were thus immersed in anhydrous dodecane (melting point: –10°C; 84% water content: <0.003%35) at room temperature in a dark atmosphere. The corresponding spectra of a gold oxide that was not immersed in anhydrous dodecane and was measured within 1 h after initial preparation (0 d) are also shown. The gold oxide was preserved in the oxidized state for 196 d in anhydrous dodecane.

**Conclusions**

Four oxygen species, comprising O 1s components I, II, III, and IV, were present in the XPS spectra of gold oxide layers with thicknesses ranging over 0.1 – 2.0 nm that were prepared from gold films by an oxygen-dc glow discharge using a gold oxidation discharge. These atmospheres included a high vacuum (1.8 – 4.7×10⁻⁴ Pa), air (1.0×10⁻⁴ Pa), and water vapor (1.5×10⁻⁴ Pa). The XPS spectra of Au 4f and O 1s for the gold oxides that were prepared by the oxygen-dc glow discharge for 30 min and irradiated at 365 nm showed almost no decomposition. The gold oxides thus decompose more rapidly under UV irradiation at shorter wavelengths in the atmosphere.

The gold oxides that were prepared by the oxygen-dc glow discharge over the 30 min period were then irradiated using UV light (254, 302, and 365 nm) for 6 h under a variety of atmospheres. These atmospheres included a high vacuum (1.8 – 4.7×10⁻⁴ Pa), air (1.0×10⁻⁴ Pa), and water vapor (1.5×10⁻⁴ Pa). The XPS spectra of Au 4f and O 1s for the gold oxides that were prepared by the oxygen-dc glow discharge for 30 min and irradiated at 302 nm for 6 h under various atmospheres are shown in Fig. 8. The corresponding spectra of a gold oxide that was not irradiated using UV light and was instead preserved in a dark atmosphere for the same period are also shown for comparison purposes. The irradiated gold oxide decomposed completely in the water vapor atmosphere. The oxide also decomposed in air, although incompletely, but showed almost no decomposition under the high vacuum. The gold oxide that was preserved in the water vapor atmosphere without UV irradiation for the same period showed no decomposition.

The ratios of the gold oxide layers after UV light irradiation at wavelengths of 254, 302, and 365 nm for 6 h in various atmospheres are presented in Table 3. These ratios were calculated based on the intensities of the Au 4f peaks of the irradiated gold oxide layers and on those of the gold oxide samples that were preserved for 6 h in the dark atmosphere. The number of data was either 6 or 12 and the uncertainties shown are the standard deviations. Table 3 clearly shows that the presence of water and UV light irradiation both promote the decomposition of the gold oxides. The table also illustrates the wavelength dependence of the decomposition process, where the gold oxides decompose more quickly after irradiation at shorter wavelengths. It was concluded that the reactions of the gold oxides decompose more quickly after irradiation at shorter wavelengths. The present study has shown that water and UV irradiation both promote the decomposition of the gold oxides. Therefore, gold oxides can be preserved for longer periods by removing both water and UV irradiation. Gold oxide samples were thus immersed in anhydrous dodecane (melting point: –10°C; 84% water content: <0.003%35) at room temperature in a dark atmosphere. The corresponding spectra of a gold oxide that was not immersed in anhydrous dodecane and was measured within 1 h after initial preparation (0 d) are also shown. The gold oxide was preserved in the oxidized state for 196 d in anhydrous dodecane.

**Table 3** Ratios of gold oxide layers prepared from gold films by an oxygen-dc glow discharge (10 Pa, 5 mA, 30 min) using a gold discharge ring after UV light irradiation at wavelengths of 254, 302, and 365 nm for 6 h under various atmospheres at room temperature.

| Wavelength/nm | 254  | 302  | 365  |
|---------------|------|------|------|
| Water vapor (1.5×10⁻⁴ Pa) | 0 ± 0 | 0 ± 0 | 0.91 ± 0.12 |
| Air (1.0×10⁻⁴ Pa) | 0.10 ± 0.04 | 0.23 ± 0.05 | 0.91 ± 0.08 |
| High vacuum (1.8 – 4.7×10⁻⁴ Pa) | 0.46 ± 0.03 | 0.63 ± 0.04 | |
discharge ring for various periods (0.17 – 30 min). Components I and II were both stable and were assigned to hydroxyl groups, and these oxygen species were present in this order from the top surface of the gold oxide (component III). Component IV is a gold oxide precursor component that occurs during the early formation periods, and subsequently changes into the gold oxide component III. The gold oxides of component III decomposed after 15 d at room temperature and decomposed immediately at temperatures exceeding 117°C in a dark atmosphere. The presence of water and UV light irradiation both accelerated the decomposition of these gold oxide layers in air. The oxide decomposition is more rapid under UV light irradiation at shorter wavelengths. The gold oxides can be preserved in the oxidized state for 196 d by immersing them in anhydrous dodecane in a dark atmosphere. This preservation method is very simple and easy to perform; the gold oxides are immersed in hydrocarbons with low water contents at room temperature in a dark atmosphere. The gold oxide can subsequently be removed from the hydrocarbon before use and can be dried without any surface contamination. This simple preservation method for gold oxide for long periods in an anhydrous hydrocarbon prevents the decomposition reaction of this oxide with water and under UV light exposure in air, and also will aid in enabling practical use of these oxides in future.

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References

1. G. C. Bond, Gold Bull., 2001, 34, 117.
2. J. J. Pireaux, M. Liehr, P. A. Thiry, J. P. Delrue, and R. Caudano, Surf. Sci., 1984, 141, 221.
3. C. R. Aita and N. C. Tran, J. Vac. Sci. Technol., A, 1991, 9, 1498.
4. D. E. King, J. Vac. Sci. Technol., A, 1995, 13, 1247.
5. A. Krozer and M. Rodalh, J. Vac. Sci. Technol., A, 1997, 15, 1704.
6. N. Saliba, D. H. Parker, and B. E. Koel, Surf. Sci., 1998, 410, 270.
7. A. Y. Klyushin, T. C. R. Rocha, M. Hävecker, A. Knop-Gericke, and R. Schögl, Phys. Chem. Chem. Phys., 2014, 16, 7881.
8. B. Koslowski, H.-G. Boyen, C. Wilderrotter, G. Kästle, P. Ziemann, R. Wahrenberg, and P. Oelhafen, Surf. Sci., 2001, 475, 1.
9. H. Tsai, E. Hu, K. Perng, M. Chen, J.-C. Wu, and Y.-S. Chang, Surf. Sci., 2003, 537, L447.
10. K. Raiber, A. Terfort, C. Benndorf, N. Krings, and H.-H. Strehlow, Surf. Sci., 2005, 595, 56.
11. A. I. Stadnichenko, S. V. Koshcheev, and A. I. Boronin, Moscow Univ. Chem. Bull., 2007, 62, 343.
12. P. Fuchs, Appl. Surf. Sci., 2009, 256, 1382.
13. A. I. Stadnichenko, S. V. Koshcheev, and A. I. Boronin, J. Struct. Chem., 2015, 56, 557.
14. R. W. Bigelow, Appl. Surf. Sci., 1988, 32, 122.
15. E. Irisou, M.-C. Denis, M. Chaker, and D. Guay, Thin Solid Films, 2005, 472, 49.
16. T. Sakata, H. Ishii, N. Sato, T. Shimamura, K. Kuwabara, K. Kudou, and K. Machida, Jpn. J. Appl. Phys., 2006, 45, 5646.
17. T. Sakata, Y. Okabe, K. Kuwabara, N. Sato, K. Kudou, K. Machida, and H. Ishii, IEEJ Trans. Sens. Micromach., 2007, 127, 253.
18. T. Sakata, Y. Okabe, K. Kuwabara, N. Sato, K. Ono, N. Shimoyama, K. Machida, and H. Ishii, Jpn. J. Appl. Phys., 2009, 48, 026501.
19. T. Sakata, H. Ishii, M. Nagase, K. Takagahara, K. Kuwabara, K. Ono, N. Sato, and K. Machida, Jpn. J. Appl. Phys., 2012, 51, 066501.
20. M. Yamamoto, E. Higurashi, T. Suga, R. Sawada, and T. Inho, Jpn. J. Appl. Phys., 2018, 57, 04FC12.
21. M. Yamamoto, T. Matsumae, Y. Kurushima, H. Takagi, T. Suga, T. Itoh, and E. Higurashi, Micromach., 2019, 10, 119.
22. H.-G. Boyen, G. Kästle, F. Weigl, B. Koslowski, C. Dietrich, P. Ziemann, J. P. Spatz, S. Riettmüller, C. Hartmann, M. Möller, G. Schmid, M. G. Garnier, and P. Oelhafen, Science, 2002, 297, 1533.
23. D. C. Lim, I. Lopez-Salido, R. Dietsche, M. Bubek, and Y. D. Kim, Surf. Sci., 2006, 600, 507.
24. D. C. Lim, I. Lopez-Salido, R. Dietsche, M. Bubek, and Y. D. Kim, Angew. Chem., Int. Ed., 2006, 45, 2413.
25. D. C. Lim and Y. D. Kim, Appl. Surf. Sci., 2006, 253, 2984.
26. L. K. Ono and B. R. Cuencya, J. Phys. Chem., C, 2008, 112, 4676.
27. M. Higo, Y. Matsubara, Y. Kobayashi, M. Mitsushio, T. Yoshidome, and S. Nakatake, Thin Solid Films, 2020, 699, 137870.
28. M. Higo, M. Mitsushio, T. Yoshidome, and S. Nakatake, Gold Bull., 2020, 53, 77.
29. M. Higo, K. Ono, K. Yamaguchi, M. Mitsushio, T. Yoshidome, and S. Nakatake, Anal. Sci., 2020, 36, 1081.
30. D. Briggs and M. P. Seah, "Practical Surface Analysis by Auger and X-ray Photoelectron Spectroscopy", 1988, John Wiley & Sons Ltd., Sussex, UK.
31. E. McCafferty and J. P. Wigtman, Surf. Interface Anal., 1998, 26, 549.
32. R. Liu, Comput. Theor. Chem., 2013, 1019, 141.
33. Z. Jiang, M. Li, T. Yan, T. Fang, Appl. Surf. Sci., 2014, 315, 16.
34. Hydrocarbons—Physical Data. The Engineering Toolbox, https://www.engineeringtoolbox.com/hydrocarbon-boiling-melting-flash-autoignition-point-density-gravity-molecularweight-d_1966.html, accessed 14 September 2019.
35. Dodecane. The Good Scents Company, http://www.thegoodscentscompany.com/data/rw1242151.html, accessed 14 September 2019.