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<td>Author(s)</td>
<td>MARUYAMA, T; ARAI, S</td>
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<tr>
<td>Citation</td>
<td>APPLIED PHYSICS LETTERS (1992), 60(3): 322-323</td>
</tr>
<tr>
<td>Issue Date</td>
<td>1992-01-20</td>
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<tr>
<td>URL</td>
<td><a href="http://hdl.handle.net/2433/43524">http://hdl.handle.net/2433/43524</a></td>
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Type | Journal Article |

Textversion | publisher |

Kyoto University
Aluminum oxide thin films prepared by chemical vapor deposition from aluminum acetylacetonate

Toshiro Maruyama and Susumu Arai

Department of Chemical Engineering, Faculty of Engineering, Kyoto University, Kyoto 606, Japan

(Received 27 August 1991; accepted for publication 4 November 1991)

Amorphous aluminum oxide thin films were prepared on glass and silicon (100) substrates by a low-temperature atmospheric-pressure chemical vapor deposition method. The raw material was aluminum acetylacetonate, which is nontoxic and easy to handle. The substrate temperature could be lowered to 250 °C by the thermal decomposition of aluminum acetylacetonate in air.

Among various chemical vapor deposition (CVD) methods for preparing aluminum oxide (Al₂O₃) thin film, metalorganic chemical vapor deposition (MOCVD) of alkylaluminum, such as the Al (CH₃)₃ - O₂ system, gives the lowest deposition temperature (350-500 °C). However, obtaining the metalorganic source materials can be difficult. They are usually expensive or toxic and are not readily available. Alternatives to the metalorganic source materials are Al(i-OC₃H₇)₃ (aluminum-tri-isopropoxide, 420-600 °C),¹ aluminum acetylacetonate (420-450 °C),²-⁴ and aluminum 2-ethylhexanoate (500-600 °C).⁵

Korzo et al.² prepared amorphous Al₂O₃ film by vacuum pyrolysis of aluminum acetylacetonate on silicon substrate which was heated at a temperature above 450 °C. They reported that the temperature at which the film begins to form increased with increasing resistivity of the silicon. Ajaya et al.³,⁴ prepared amorphous Al₂O₃ film by atmospheric-pressure pyrolysis of aluminum acetylacetonate on glass substrate which was heated at 420 °C. Aluminum acetylacetonate powder as the source material was carried by argon carrier gas into a working chamber.

This letter will show that the substrate temperature in the CVD of Al₂O₃ film can be lowered to 250 °C by the thermal decomposition of aluminum acetylacetonate in air.

Aluminum acetylacetonate [(Al(C₅H₇O₂)₃) Nihon Kagaku Sangyo Co., Ltd.] of reagent grade was used as the source material. It is nontoxic and easy to handle in solid form (powder) at room temperature. It was heated at a temperature of 150 °C and the generated gas was entrained by nitrogen carrier gas. The flow rate of the carrier gas was 0.3-1.0 s/min.

Borosilicate glass plates, quartz glass plates, and silicon (100) single-crystal wafers were used as the substrates. The substrate was placed on a temperature-controlled electric heater. The substrate temperature ranged from 250 to 600 °C. The depositions were carried out at atmospheric pressure in air atmosphere.

The composition of the film was measured by x-ray photoelectron spectroscopy. The infrared spectra were obtained by means of a Fourier transform infrared spectrometer (Shimadzu FTIR-4300). The samples were prepared by depositing films on silicon single-crystal substrates. The crystallinity of the film was analyzed by x-ray diffraction with Cu Kα radiation. The optical transmittance of the film was obtained by means of a multipurpose recording spectrophotometer with a blank glass substrate inserted into the reference beam path of the spectrophotometer.

The films were transparent and showed no apparent peeling. The lower limit of the reaction temperature was 250 °C, which is very low compared to those reported by Korzo et al.² (450 °C) and Ajaya et al.³,⁴ (420 °C). They prepared films in inert atmosphere, whereas in this study the film was grown in air atmosphere. The x-ray photoelectron spectroscopy showed that the oxides were near-stoichiometric Al₂O₃. For the film obtained in an inert atmosphere, however, a carbon contamination and a little excess of oxygen (O/Al = 1.75-1.85) were observed in preliminary experiments. The carbon contamination and an excess of oxygen were reported also by Ajaya et al.³,⁴ for deposition in an inert atmosphere.

Figure 1 shows the Arrhenius plot of deposition rates, which were obtained at a source temperature of 150 °C and a N₂ flow rate of 0.3 s/min. The straight lines in this figure show that the activation energy is ca. 28.0 kJ/mol (0.29 eV/molecule) which is comparable to that of CVD of Al₂O₃ from 2-ethylhexanoate.⁵

Figure 2 shows infrared absorption spectra of films deposited at four different reaction temperatures. The spectra are very characteristic of Al₂O₃ films; i.e., broad absorption bands at about 1100-500 cm⁻¹ represent the vi-
FIG. 2. IR transmission spectra of 220, 180, 350, and 970 nm thick films deposited at substrate temperatures of 250, 350, 450, and 600 °C. (The wave number scale is linear with a scale change at 2000 cm⁻¹.)

Absorptions of Al₂O₃. Absorption peaks at about 2000-1250 and 700-400 cm⁻¹ represent vibrations of water in gas phase. Absorption peaks at about 3400-3200 cm⁻¹ represent O—H bond vibration. The O—H bond is inferred to belong to the water adsorbed to the film surface, because the surface of Al₂O₃ becomes active at higher substrate temperatures (>450 °C). Absorption peaks at about 1100-1050 cm⁻¹ represent Si—O bond vibration. The Si—O bond is attributable to a thermal oxidation of the surface of the Si substrate. The x-ray diffraction pattern of the film showed that the films were amorphous.

Figure 3 shows the optical transmittance for the 970-nm thick film deposited on the quartz glass substrate at 600 °C. The optical transmittance was more than 95% in the wavelength range of 400-2600 nm. The refractive index obtained from the curve measured with blank reference was 1.56.

FIG. 3. Optical transmittance of a 970-nm thick film deposited on quartz glass substrate at 600 °C.

In conclusion, an amorphous aluminum oxide thin film was prepared on glass and silicon (100) substrates by a low-temperature atmospheric-pressure chemical vapor deposition method. The raw material was aluminum acetylacetonate, which is nontoxic and easy to handle. The substrate temperature could be lowered to 250 °C by the thermal decomposition of aluminum acetylacetonate in air.

This work was supported by the Japan Securities Scholarship Foundation, Nissan Science Foundation, Iketani Science and Technology Foundation, Miyashita Research Foundation for Materials Science, Yazaki Science and Technology Foundation, and Nippon Sheet Glass Foundation.