Optical properties of Al$_2$O$_3$ thin films grown by atomic layer deposition

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We employed the atomic layer deposition technique to grow Al$_2$O$_3$ films with nominal thicknesses of 400, 300, and 200 nm on silicon and soda lime glass substrates. The optical properties of the films were investigated by measuring reflection spectra in the 400–1800 nm wavelength range, followed by numerical fitting assuming the Sellmeier formula for the refractive index of Al$_2$O$_3$. The films grown on glass substrates possess higher refractive indices as compared to the films on silicon. Optical waveguiding is demonstrated, confirming the feasibility of high-index contrast planar waveguides fabricated by atomic layer deposition. © 2009 Optical Society of America

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1. Introduction

Finding a material platform for low-loss, high-index contrast optical waveguides has been an important task in integrated optics since its inception. Integrated optical devices and systems are applied in optical communication and optical sensors, as well as emerging applications, such as on-chip optical clock distribution and optical chip-to-chip interconnects [1]. These applications require strong light confinement to a waveguide core, which can be achieved using a large index step between the core and the claddings. For telecommunication devices, strong confinement is needed to achieve a high degree of integration and allow for sharp bends of channel waveguides. For sensors, it is important to have a strong field at the waveguide surface to attain high sensitivity. Many materials and thin-film manufacturing technologies have been tried, but the quest is far from over. Among all-dielectric waveguide systems, the strongest confinement is found in silicon-on-insulator (SOI) waveguides [2], which have a silicon core (refractive index $n_{Si} = 3.48$) and silica ($n_{SiO2} = 1.45$), or even air, claddings. The SOI waveguides so far seem to be an ideal technical solution for planar photonic integrated circuits. Besides perfect optical characteristics, this material system is absolutely compatible with the complementary metal-oxide-semiconductor (CMOS) fabrication process in microelectronics. However, because of the method by which the SOI wafers are manufactured (wafer bonding followed by cleaving), it appears to be difficult to implement a more sophisticated photonic circuitry [4] in SOI systems in which, for example, multiple optical layers are required. The architecture of some integrated optical devices, such as large optical switches, may require waveguide crossovers with negligible cross talk, which, again, is hardly realizable in a single-layer photonic circuit. Also, SOI has limited applicability in optical sensors, since silicon is not transparent in the visible range. Another CMOS-compatible material system for optical waveguides is silicon oxynitride [5], which is a mixture of silicon oxide and silicon nitride typically manufactured by plasma-enhanced chemical vapor
deposition. With a high nitrogen content in the film, the refractive index may reach \( n_{\text{Si3N4}} = 2.2 \). High nitrogen content films, however, tend to show high optical loss. With low nitrogen content, the index step is reduced and strong light confinement is no longer possible. Magnetron deposition [6] and various sputtering techniques [7,8] can also be used for thin-film manufacturing. They are successfully used in multilayer dielectric mirrors, but as far as high-index optical waveguides are concerned, the roughness of the film surface is often too large, which leads to strong light scattering. Among non-CMOS technologies, sol-gel [9] and ion exchange [10] have been employed. Ion exchange is probably one of the oldest and best-optimized waveguide manufacturing technologies. The index step in ion-exchange waveguides is small. Sol-gel waveguides generally show good performance with large index steps in systems such as TiO\(_2\)/SiO\(_2\), but incompatibility with CMOS limits applications of this technology.

In the present work, we study the optical properties of alumina films prepared by atomic layer deposition (ALD) [11–15]. Because of the self-limiting nature of ALD growth, the films typically show perfect thickness uniformity across the entire wafer. With carefully prepared substrates, the top surface of the film is expected to be practically atomically flat. This eliminates, or at least greatly reduces, light scattering at surface imperfections—the major source of optical losses in thin-film waveguides. ALD, compared to other thin-film fabrication techniques, has low growth rates, since film growth occurs through the sequential pulsing of two or more different precursor gases, separated by inert gas purges. The ALD mechanism ensures that growth is self-limited, resulting in smooth, uniform films. However, the exceptional quality of the ALD films makes this technology quite promising for fabrication of integrated optical devices and systems. Here we present data on the refractive index of ALD alumina films in the spectral range from 400 to 1800 nm deduced from the reflectance spectra, and demonstrate planar optical waveguides.

2. Experimental and Theoretical Details

\( \text{Al}_2\text{O}_3 \) films were grown on silicon (100) and soda lime substrates in a Picosun R-75 ALD reactor using trimethylaluminum (TMA) and water [15]. The deposition temperature was 300°C. Films with nominal thicknesses of 400, 300, and 200 nm were grown with 4400, 3300, and 2200 deposition cycles, respectively. Each cycle consisted of a 0.1 s TMA pulse, followed by a 3.0 s nitrogen purge, a 0.1 s water pulse, and, finally, another 3.0 s nitrogen purge.

The reflection spectra have been measured using a DigiKrom 240 monochromator (Spectral Product) and a halogen lamp as a light source. Fused quartz was used for normalizing intensity, since the refractive index of fused quartz is well known. The incident angle was 5.6° and the beam size was 0.5 mm. The substrate dimensions were approximately 39 mm \( \times \) 25 mm and the optical properties of the \( \text{Al}_2\text{O}_3 \) films were investigated by measuring the reflection spectra at five different positions on each sample 5 mm apart from each other. MATLAB curve fitting tools were used for analysis of the reflection spectrum data. The experimentally measured reflection spectra were numerically fit to the expected calculated spectra [16], assuming the Sellmeier formula for alumina. The fitting results provided the thickness and refractive index of the film in the entire spectral range of interest. Using the Lavenberg–Marquardt algorithm, the experimental data were analyzed by minimizing the mean-square differences between the measured reflection spectra and the calculated spectra, which are generated from the fitting model at the corresponding wavelength, angle of incidence, and substrate and cover refractive indices at corresponding wavelengths. The refractive indices for silicon and soda lime glass were taken from Refs. [17,18], respectively.

The fitting algorithm also included adjustable factors to account for slow drift of the intensity of the light source used in the measurements. Reflection spectra were taken from the range of 400 to 1800 nm, so, for this wide range of wavelength, the light source has some drift in intensity, which introduces error in the reflection spectra since the effect of drift of light source depends upon the wavelength. So, including the effect of drift of light source, we used a first-order correction in the calculated reflection spectrum:

\[
R_{\text{exp}} = R_{\text{Calu}}(C_0 + C_1(\lambda - C_2)),
\]

where \( C_0 \) and \( C_1 \) are adjustable parameters, \( \lambda \) is the wavelength, the value of \( C_2 \) is 100 nm, which includes the effect of short wavelength, and \( R_{\text{Calu}} \) is the calculated reflection spectrum.

The fitting parameters were Sellmeier coefficients, film thickness, \( C_0 \), and \( C_1 \). The fits were carried out for each position for each sample and, then, for the refractive index of alumina, the results from the fitting were averaged for the same substrate.

3. Optical Properties

A. Refractive Index

The films on silicon substrates possess a very high-index contrast and, thus, show a very strong interference in the reflection spectra. As long as the refractive index of the alumina film is much lower than that of the silicon substrate, the reflection spectrum minima contain information about the refractive index and film thickness [16], while the spectrum maxima are defined by the optical properties of the silicon substrate. The reflection minima are more clearly pronounced in the logarithmic scale. Therefore, to obtain more accurate information about the refractive index and thickness of the \( \text{Al}_2\text{O}_3 \) film on the silicon substrate, the logarithm of the reflection spectrum was fitted to the logarithm of the
calculated reflection spectrum. The error analysis of fitting justifies the choice of the logarithmic scale for structures of this type—low-index film on a high-index substrate. Figures 1(a) and 1(b) show the measured and calculated reflection spectra of Al$_2$O$_3$ films on silicon and soda lime glass substrates, respectively.

The quality of the fit was investigated in detail by the mean-squared error (MSE) [14], defined as

$$\text{MSE} = \frac{1}{N} \sum_{i=0}^{N} \left( \frac{R_i^{\text{exp}} - R_i^{\text{calc}}}{\sigma_i} \right)^2,$$

where $N$ is the total number of measurements and $\sigma_i$ is the standard deviation of experimental data points. The MSE values for Al$_2$O$_3$ films on silicon and soda lime glass are 0.0006 and 0.0070, respectively. The positions of maxima and minima in the reflection spectrum depend on the product of film refractive index and film thickness, and there is strong anticorrelation between them. By introducing the scaling factor (Sn) in refractive index and film thickness, we construct the contour plot for MSE. The scaling factor (Sn) introduced a controllable error in refractive index and film thickness to calculate and contour plot the MSE. Figure 2 shows the contour plot for MSE with respect to the scaling factors for the refractive index and film thickness, and the film refractive index is equal to the product of scaling factor (Sn) and film refractive index ($n_f$) given by Table 1. The error plot is constructed in such a way that the minimum error is achieved when both scaling factors are equal to unity for the film thickness and its refractive index. The contour closest to the minimum corresponds to doubling the fitting error. An order of magnitude with larger error in the case of a film on a soda lime glass substrate is due to the substantially smaller index contrast and, consequently, much weaker interference in the reflection spectrum.

### Table 1. Sellmeier Coefficients for Al$_2$O$_3$ Films

<table>
<thead>
<tr>
<th></th>
<th>$A_0$</th>
<th>$A_1$</th>
<th>$\lambda_1$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicon</td>
<td>1.31838</td>
<td>1.32052</td>
<td>121.594</td>
</tr>
<tr>
<td>Soda lime glass</td>
<td>1.33247</td>
<td>1.40684</td>
<td>119.992</td>
</tr>
</tbody>
</table>

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spectra. The fitting results are excellent in comparison to those of Kim et al. [14] and Toyoda et al. [19].

In the numerical fit of the reflection spectra, the film refractive index was defined according to the Sellmeier-like dispersion formula:

\[ n_f(\lambda) = \sqrt{A_0 + \frac{A_1\lambda^2}{\lambda^2 - \lambda_1^2}} \]

where the \( A_0 \) term accounts for the contribution of short wavelength absorption to the refractive index at a longer wavelength, while \( A_1 \) and \( \lambda_1 \) are the relative oscillator strength and the resonant wavelength, respectively. Table 1 gives the values of \( A_0, A_1, \) and \( \lambda_1 \) for Al\(_2\)O\(_3\) films on silicon and soda lime glass deduced from analysis of the reflection spectra shown in Fig. 1.

Figure 3 shows the dispersion curves of the refractive index of Al\(_2\)O\(_3\) films on silicon and soda lime glass in the wavelength range from 400 to 1800 nm. The refractive indices at 633 nm were 1.64 and 1.67 for Al\(_2\)O\(_3\) films on silicon and soda lime glass, respectively. The refractive indices are in good agreement with those of Räisänen et al. [20] and Ott et al. [21]

Table 2. Range of Film Thickness, Mean Thickness, and Standard Deviation Obtained from Fit

<table>
<thead>
<tr>
<th>Substrate</th>
<th>Experimental Film Thickness (nm)</th>
<th>Range of Film Thickness (nm) Obtained from Fit</th>
<th>Mean Thickness (nm)</th>
<th>Standard Deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicon</td>
<td>400</td>
<td>415–403</td>
<td>410.56</td>
<td>6.41</td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>312–304</td>
<td>307.58</td>
<td>6.29</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>204–199</td>
<td>201.82</td>
<td>4.31</td>
</tr>
<tr>
<td>Soda lime glass</td>
<td>400</td>
<td>402–377</td>
<td>392.63</td>
<td>3.65</td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>302–279</td>
<td>296.54</td>
<td>2.03</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>192–178</td>
<td>181.87</td>
<td>1.43</td>
</tr>
</tbody>
</table>

The error introduced by the substrate refractive index in film refractive index was \( \pm 0.001 \) for silicon substrate and \( \pm 0.003 \) for soda lime glass substrate. The refractive index of the Al\(_2\)O\(_3\) films on silicon compared to that of films on soda lime glass is apparently lower, by about 0.03, which is much higher than the error introduced by the discrepancy of substrate refractive index. Since Si wafers typically have a 2–3 nm layer of native SiO\(_2\), this may contribute to the reduced overall index of the film as it is extracted from the reflection spectra. The observed difference, however, is about an order of magnitude larger than what may be caused by the existence of the native oxide, and it is about the same for the film thickness from 200 to 400 nm. The different optical properties, already mentioned in Ref. [14], apparently indicate somewhat different film densities on different substrates.

B. Film Uniformity

The thickness of the Al\(_2\)O\(_3\) films was measured at 10 different points, which were 2.5 mm apart from each other on each sample. The results show that the film thickness is uniform over the entire substrate. Table 2 provides the range of film thickness, mean film thickness, and the standard deviation obtained from fitting. Figure 4 shows the thicknesses of Al\(_2\)O\(_3\) films on silicon and soda lime glass, which are close to the nominal values of 400, 300, and 200 nm.

4. Thin-Film Waveguide

The waveguide properties were investigated for the Al\(_2\)O\(_3\) films on soda lime glass. For the theoretical calculation, the effective refractive index [23] for different modes was calculated for all three samples and the refractive index and film thickness were taken from the fitting results. For experimental verification, the prism coupling method [24–26] was used for measuring the effective refractive index. The effective refractive index for each mode is directly related to the angle of incidence at which incidence light was guided through the waveguide. The effective refractive index is defined as

\[ n_{\text{eff}}(\lambda) = \sqrt{\frac{n_f^2(\lambda) - n_g^2(\lambda)}{n_f^2(\lambda) - n_i^2(\lambda)}} \]

where \( n_f(\lambda) \) is the refractive index of the film at wavelength \( \lambda \), and \( n_g(\lambda) \) and \( n_i(\lambda) \) are the refractive indices of the substrate and the incident medium, respectively.
where $n_P$ is the prism refractive index, $\varphi$ is the base prism angle, and $\theta_m$ is the angle at which incidence light is guided through the waveguide. Figure 5(a) shows the schematic diagram of our experiment.

$$n_{\text{eff}} = n_P \cdot \sin\left(\varphi + \arcsin\left(\frac{\sin(\theta_m)}{n_P}\right)\right), \quad (4)$$

A He–Ne laser emitting at a wavelength of 632.8 nm and a prism with a vertex angle of 60.19° and a refractive index of 1.7520 were used. Figure 5(b) shows the guided mode for the 400 and 300 nm thick Al$_2$O$_3$ films on soda lime glass. Table 3 gives the experimental and calculated effective refractive indexes for the Al$_2$O$_3$ films on soda lime glass. At 632.8 nm, the 400 and 300 nm thick films support fundamental TE- and TM-polarized modes and 200 nm thick films support only the fundamental TE-polarized mode.

For characterization of the waveguide losses, the intensity of the guided mode tracks was measured as a function of the distance. The losses in waveguide are defined as

$$I = I_0 \exp(-\alpha x), \quad (5)$$

where $\alpha$ is defined as losses in waveguide. To calculate losses in dB/cm,

$$\alpha_{\text{db}} = (10/L)\log_{10}(\alpha), \quad (6)$$

where $L$ is length in cm travel by the guided light in waveguide. Figure 6 shows the natural log of intensity as a function of distance (millimeters). The slopes of the intensity curves correspond to the losses in the range from 7 to 11 dB/cm.
5. Conclusion

In summary, the ALD technique was employed to fabricate Al$_2$O$_3$ films on silicon and soda lime glass substrates. The refractive index of the films was found by fitting the reflection spectra measured in the 400–1800 nm spectral range and assuming the Sellmeier formula with MSE values of 0.0006 and 0.0070 for Al$_2$O$_3$ films on silicon and soda lime glass, respectively. The refractive index of Al$_2$O$_3$ films on soda lime glass was found to be higher than that in the case of the silicon substrate. High-index contrast optical waveguides are demonstrated.

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References


