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Abstract. Low-loss hollow waveguides with a multiple dielectric layer are designed and fabricated for use in cavity ring-down spectroscopy (CRDS). Calculations of the waveguide design revealed that hollow waveguides for infrared CRDS system need to have transmission loss of 0.1 dB/m or less. We fabricated rectangular hollow waveguides with multiple dielectric layers to obtain low-loss properties. The waveguides are composed of four glass strips each with a dielectric multilayer deposited on the surface in advance. Experimental results show that the waveguides with double dielectric layers have lower losses at a target wavelength in the infrared than those with a single dielectric layer. The measured transmission loss of a 10-cm long waveguide deposited with a multilayer of Si/Al₂O₃/Ag for the transverse magnetic wave and Al₂O₃/Ag for the transverse electric wave was 0.24 dB at a target wavelength of 5.2 μ m. The loss of the waveguide will be reduced when only the lowest-order mode is excited in the waveguide. © The Authors. Published by SPIE under a Creative Commons Attribution 3.0 Unported License. Distribution or reproduction of this work in whole or in part requires full attribution of the original publication, including its DOI. [DOI: [10.1117/1.OE.52.10.106104](https://doi.org/10.1117/1.OE.52.10.106104)]

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1 Introduction

Exhaled breath contains various gases related to human diseases, so breath analysis that is a noninvasive diagnosis method may be utilized as a complement of blood or urine tests to increase diagnostic accuracy.^{1,2} Concentration of these disease-related components in the breath is usually in a parts-per-million (ppm) order or less, so a gas analysis system with high sensitivity and selectivity is required. However, the system represented by a mass spectrograph that meets these requirements tends to be complicated and expensive. A breath analysis system based on infrared absorption spectroscopy that detects vibrational modes of molecules is a candidate low-cost high-sensitivity system. To obtain high sensitivity, previous works have reported measurements using a multipath gas cell with multiple mirrors^{3,4} and cavity ring-down spectroscopy (CRDS). In a CRDS system, light is trapped in an optical cavity consisting of a pair of highly reflective mirrors, and decay time of leaking light that correlates with the absorption coefficient of the medium gas in the cavity is measured. Gas detection systems based on CRDS achieve parts-per-billion (ppb) sensitivity.^{5,6} However, the volume of the gas cell is large and this limits the analysis of exhaled breath. In this report, we design and fabricate low-loss hollow waveguides as a micro-volume gas cell for highly sensitive infrared CRDS systems.

2 Design

Unlike conventional CRDS systems with large volume gas cells,⁷ for hollow-waveguide based CRDS systems, transmission loss in the gas cell needs to be considered because the loss influences the intensity of light leaking from the cell. As shown in Fig. 1, the cavity in the CRDS system consists

of a hollow waveguide and a pair of high-reflection mirrors. When a laser pulse is injected into the cavity with an intensity I_{in} , the light pulse is repeatedly reflected between the mirrors while being attenuated by waveguide loss α_L and absorption of the medium gas in the cavity α . As a result, the light leaking from the cavity exponentially decreases in intensity. The intensity of the output light I_0 that has passed through the cavity once and that of the light after n -round trips I_n are derived as

$$I_0 = (1 - R)^2 \exp[-(2\alpha_L + 2\alpha C)L] I_{in}, \quad (1)$$

$$I_n = \{R \times \exp[-(2\alpha_L + 2\alpha C)L]\}^{2n} I_0, \quad (2)$$

where R is the reflectance of the mirrors, L is the cavity length, and C is the concentration of sample. For the waveguide cavity with a sample gas, Eq. (3) shows the time constant τ that corresponds to the time for I_0 to decrease by a factor of e . Similarly, the time constant for an empty cavity τ_0 is described by Eq. (4), where c is the speed of light

$$\tau = \frac{L}{c[(1 - R) + 2\alpha_L L + 2\alpha C L]}, \quad (3)$$

$$\tau_0 = \frac{L}{c[(1 - R) + 2\alpha_L L]}. \quad (4)$$

In CRDS systems, the decay time should be long enough to quantitate the absorption coefficient of sample gas that is directly derived from the decay time shown in Eq. (3). Figure 2 shows calculated decay time τ_0 when reflectance

Fig. 1 Schematic of hollow-waveguide based cavity ring-down spectroscopy cavity.

of mirrors is assumed to be $R = 0.995$, which is a common value for infrared high-reflection mirrors. Figure 2 shows that for all the cavity lengths, the decay time rapidly decreases when waveguide loss exceeds 0.1 dB/m. Therefore, we set our target waveguide loss as 0.1 dB/m. It is also shown that decay time τ_0 becomes large for long cavities because the influence of reflection losses at the mirrors is small. To fabricate long cavities, however, one needs a large apparatus for film deposition on long substrates. Therefore, in this article, we fabricated 0.1-m long waveguides to show the feasibility of fabricating such low-loss waveguides.

We fabricated rectangular hollow waveguides with ultra-low transmission loss by depositing multilayer dielectric films on the inside of waveguides.^{8–10} The waveguide is fabricated by assembling two pairs of glass strips that have multiple dielectric films deposited on a silver layer formed in advance on the strip. The dielectric multilayer is composed of an alternate stack of Al_2O_3 films as a low-index material and Si films as a high-index material.

When a rectangular hollow waveguide is excited by linearly polarized light, as shown in Fig. 3, the attenuation constant of the waveguide α is expressed by the sum of those of transverse electric (TE) mode α_{TE} and transverse magnetic (TM) modes α_{TM} in the corresponding slab waveguide

$$\alpha = \alpha_{\text{TE}} + \alpha_{\text{TM}}. \quad (5)$$

The attenuation constants of each mode in Eq. (5) are calculated by ray-optic method as

Fig. 2 Calculated decay time for an empty hollow waveguide cavity.

Fig. 3 Transmission modes in square hollow waveguides.

$$\alpha_{\text{TE,TM}} = \frac{u_0}{4k_0 T^2} (1 - R_{\text{TE,TM}}), \quad (6)$$

where u_0 is the normalized phase constant of mode ($\pi/2$ for the lowest-order modes), k_0 is the wavenumber in vacuum, and $2T$ is the length of each side. $R_{\text{TE,TM}}$ is the reflectances of plane waves that incident on the inner surfaces of waveguides with angles that are associated with the phase constant u_0 .⁸

As reported in Ref. 10, forming a multiple stack of a pair of dielectric films with low- and high-refractive indices on a metal substrate greatly reduces the losses of these modes. Optical thickness of the films should be set to a quarter of the target wavelength. As shown in Fig. 4, for TE mode, films functions must be an even number to reduce the loss, while for TM modes, they must be an odd number. This difference is due to the phase difference π between these modes at the reflection on the inner wall.

Figure 5 shows calculated losses of hollow waveguide with multiple dielectric layers with film numbers of $2m$ for TE waves and $2m + 1$ layers for TM waves. In this calculation, we assumed an inside dimension of 2×2 mm for the rectangular waveguide and target wavelength of $5.2 \mu\text{m}$, which corresponds to an absorption peak of nitric oxide known as a biomarker of asthma. For a metal layer, we assumed silver, which is easily deposited by a sputtering method, and we used the complex refractive indices shown in Ref. 11. In this figure, a low-loss region appears in the vicinity of $5.2 \mu\text{m}$ owing to the reflection enhancement by the dielectric multilayer. The wavelength giving the minimum loss can be changed to any desired wavelength, as far as the dielectric materials show low absorption, by changing the thickness of the layers and this enables measurement of

Fig. 4 Structure of multilayer in the waveguide.

Fig. 5 Calculated loss spectra of hollow waveguide with multiple dielectric layers.

Fig. 6 Calculated losses of hollow waveguides at a wavelength of 5.2 μm .

any other gases. By increasing film numbers, the width of the low-loss region becomes narrow and the minimum losses become lower.

Figure 6 shows calculated losses of the waveguide as a function of m value. We found that the losses of the waveguide are dramatically reduced by increasing the number of dielectric multilayers for both TE and TM modes. Figure 6 shows that the target loss of 0.1 dB/m will be cleared by a waveguide without dielectric films (see “Metal only” in the figure). However, the transmission losses of fabricated hollow waveguides are usually much higher than those in the theoretical estimation due to surface roughness on the inner wall of the waveguide and other structural imperfections. Therefore, inner dielectric films need to be formed to obtain waveguides with low losses. By forming a single dielectric layer on a pair of walls, which corresponds to the case $m = 0$, loss is expected to be greatly reduced.

3 Waveguide Fabrication

We used the sputtering equipment shown in Fig. 7 to deposit the metal and multiple dielectric layers. Metal and dielectric films are deposited on the 10-cm long glass strip while the strip is transferred by using a linear manipulator driven with a pulse motor. Owing to the ideally smooth surface of glass substrates, high smoothness is obtained for the surface of deposited films on the glass substrates.

Fig. 7 Schematic of sputtering machine for film deposition.

Fig. 8 A reflectance spectrum of Al_2O_3 -coated glass strip.

We measured reflectance spectra from visible to near-infrared to estimate longitudinal uniformity of dielectric layers deposited by sputtering. In the experiment, white light is incident on a glass strip at an oblique angle, and reflection spectra of the strip coated with a dielectric film were measured by a spectrum analyzer. In our experiment, the incident angle was fixed to 45 deg because of the limitation due to the detector size and easiness of optical alignment. Figure 8 shows a measured reflectance spectrum of a strip coated with Al_2O_3 and Ag. The vertical axis is reflectance that is normalized by that of an Ag-only coated glass strip. The dielectric film thickness d is derived from the wavelength of the m 'th peak counted from the interference peak appearing in the longest wavelength λ_m as

$$d = \frac{(2m-1)\lambda_m}{4\sqrt{n^2 - \sin^2 \frac{\pi}{4}}}, \quad (7)$$

where n is the refractive index of dielectric layer. In this measurement, we examined the longitudinal uniformity of the Al_2O_3 thickness and found that the thickness variation is $<5\%$ as shown in Fig. 9.

In Fig. 10, calculated losses of silver hollow waveguides with an inside dimension of 2×2 mm are shown. It is assumed that two inner surfaces facing up each other are coated with Al_2O_3 film. The figure shows loss spectra of waveguides with three different Al_2O_3 film thicknesses: ideal thickness giving the minimum loss at 5.2- μm wavelength and thickness $\pm 5\%$ shifted from the ideal value. It is seen that the loss at the target wavelength of 5.2 μm does not change with this level of thickness fluctuation although the interference peaks shift largely.

We fabricated 10-cm long rectangular hollow waveguides with a 2-mm inside dimension by assembling two pairs of

Fig. 9 Variation in calculated thickness of Al_2O_3 .

Fig. 10 Influence of variation in coating thickness of Al_2O_3 on the calculated losses of hollow waveguides.

glass strips. Figure 11 shows a cross section of the fabricated waveguide and Fig. 12 shows loss spectra of this waveguide measured by randomly and linearly polarized light. The loss peak at $10\ \mu\text{m}$ is due to abnormal dispersion of Al_2O_3 and other large peaks are due to interference effect of the dielectric layer. In the spectrum for random polarization, we observed the interference peaks of TE mode at 4.2 and 1.4 and that of TM mode at $2.1\ \mu\text{m}$. In contrast, for the perpendicularly polarized light, only peaks for the TE mode appear and a TM-mode peak appears for the horizontal polarization. At the target wavelength of $5.2\ \mu\text{m}$, the minimum loss is obtained for the horizontal polarization as designed.

Then, we fabricated a waveguide with multiple dielectric layers. For TE waves, we deposited the multilayer of $\text{Si}/\text{Al}_2\text{O}_3/\text{Ag}$ and $\text{Al}_2\text{O}_3/\text{Ag}$ for the TM wave and the loss spectra are shown in Fig. 13. These spectra are for

Fig. 11 Cross section of fabricated hollow waveguide.

Fig. 12 Loss spectra of the Al_2O_3 -coated waveguide.

Fig. 13 Loss spectrum of a waveguide with multiple dielectric layers compared with one with a single dielectric layer.

the horizontal polarization that gives low attenuation in the waveguide. It is shown that depositing multiple dielectric layers drastically reduces the transmission loss at the target wavelength of $5.2\ \mu\text{m}$.

In Fig. 13, the measured loss of the waveguide with multiple dielectric layers at $5.2\ \mu\text{m}$ is $0.4\ \text{dB}$ for the 10-cm long waveguide, which is higher than the target value of $0.1\ \text{dB/m}$. However, the relatively high loss is partly because of wide-spread incoherent light used in the experiment that excites a number of high-order modes in the waveguide. To estimate losses for laser light that has much smaller divergence that excites only low-order modes, we used a long coupling fiber as a mode filter. We used coupling fibers with two different lengths and the results are shown in Table 1. The loss is reduced by lengthening the coupling fiber because extremely high-order modes are eliminated in the fiber and the loss was $0.24\ \text{dB}$ with the 0.7-m long coupling fiber. This result shows that the loss of the waveguide will be reduced much more when only the lowest-order mode is excited in the waveguide.

Table 1 Measured losses of waveguide with long coupling fiber.

Coupling fiber length	Measured transmission loss (dB)
0	0.42
40 cm	0.29
70 cm	0.24

Table 2 Estimated losses of waveguides.

Incident beam	Surface roughness	Estimated loss
Gaussian beam with divergence of 7 deg FWHM	0	0.08 dB for 10 cm
	30 nm RMS	0.30 dB for 10 cm
Ray with transmission angle of 0.11 deg	0	1.6×10^{-4} dB/m
	30 nm RMS	7.4×10^{-4} dB/m

The above result of the 0.24-dB loss for 10-cm long waveguide may seem much larger than the losses expected from theory shown in Figs. 5 and 6. However, this is mainly because lossy high-order modes are excited in the waveguide even when using a 0.7-m long coupling waveguide. We can estimate a transmission loss of the lowest-order modes from that for an incident beam with the wide divergence angle by using a ray-optic calculation.^{12,13} In this method, the losses of rays transmitting in the waveguide are calculated as a function of transmission angle in the waveguide and the reflectance at the inner wall of the waveguide. The reflectance is affected by the surface roughness of the inner wall and this causes increase of transmission loss from the ideal value. First, we assumed an incident Gaussian beam with a divergence angle of 7 deg full-width half-maximum (FWHM) that was a measured value for the 0.7-m long coupling waveguide and a surface roughness of 30 nm in root mean square (RMS) that was an observed value for the waveguide sample by using an atomic force microscope. As shown in Table 2, the loss for the 10-cm long waveguide was calculated as 0.30 dB that was close to the measured loss of 0.24 dB. Then, we used the same roughness for the calculation of the loss for the lowest-order mode. It was assumed that the mode consisted of a single ray that has a transmission angle corresponding to the propagation constant of the mode. In this case, the angle was 0.11 deg for the waveguide with a 2-mm inside dimension and the estimated loss was 7.4×10^{-4} dB/m. Although this is much higher than the theoretical loss of the ideal waveguide due to the surface roughness of the waveguide, it is lower than the target loss of 0.1 dB/m. We expect that the effective transmission loss of the waveguide will be close to the estimated value when constructing a cavity because lossy high-order modes are eliminated in the resonator.

4 Conclusion

We designed and fabricated low-loss hollow waveguides as a micro-volume gas cell for highly sensitive infrared CRDS systems. In the calculation, we found that the hollow waveguides for infrared CRDS system require transmission loss that is 0.1 dB/m or less. We fabricated rectangular hollow waveguides with multiple dielectric layers to obtain low-loss properties. The measured transmission loss of a 10-cm long waveguide deposited with a multilayer of Si/Al₂O₃/Ag for TE wave and Al₂O₃/Ag for TM wave was 0.24 dB at a target wavelength of 5.2 μ m. The loss of the waveguide will be

reduced when only the lowest-order mode is excited in the waveguide.

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