Fabrication of terahertz hollow-glass metallic waveguides with inner dielectric coatings

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Hollow-core glass waveguides (HCWs) with inner coatings of silver and polystyrene (PS) have been fabricated for transmission of terahertz radiation. A liquid-phase chemical deposition process was used to deposit silver and PS thin film coatings inside glass tubing. The PS dielectric layer can substantially lower the loss of the HCW compared to the metal-only waveguide. A polymer coating was chosen because it is possible to deposit the dielectric film thickness required for operation in the terahertz regime. Specifically, the dielectric film thickness is proportional to the wavelength so the PS coatings need to be on the order of 10 to 15 μm to minimize transmission losses at terahertz frequencies. This is much thicker than the submicron thick dielectric coatings needed in the IR region. The thickness of the PS film depends on the concentration of PS in the coating solution and the coating rate. Both of these parameters are studied and related to the losses in the waveguides. The lowest loss of 0.95 dB/m at 119 μm (2.5 THz) was obtained for hybrid HE mode propagation in a waveguide with a 8.2 μm thick PS film deposited inside a 2.2 mm bore, 90 cm long glass tube.
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I. INTRODUCTION

Nonionizing terahertz radiation is enjoying a resurgence for a wide variety of applications including imaging, remote sensing and security screening, and spectroscopy.[1] None of these applications currently employs a terahertz fiber optic or waveguide, yet the functionality of some of these systems could be improved if a low-loss, flexible terahertz waveguide were available. For example, a low-loss, flexible terahertz waveguide could be used to deliver high-quality beams from terahertz sources such as quantum cascade lasers and photonic mixers. In particular, a hollow waveguide filled with dry gas or evacuated could transmit terahertz energy avoiding atmospheric absorption. Conversely, a low-loss hollow-core waveguide (HCW) could be filled with a gas for gas sensing or for nonlinear applications. Many approaches have been taken to fabricate a terahertz waveguide including solid-core polymer fibers,[2] photonic crystal fibers,[3–5] hollow polymer fibers,[6] Bragg fibers,[7–9] metal tubes,[10] sapphire fibers,[11] metal wires,[12] and porous polymer fibers.[13] Among the most promising waveguides for the transmission of terahertz radiation are hollow waveguides with inner metallic and metallic/dielectric thin film coatings.[14–16] Our recent work has focused on the use of Ag/polystyrene (PS) coatings deposited on the inside surface of silica tubing. The dielectric coating is needed to reduce the loss over that of a metal-only HCW. Specifically we measured a loss of 0.95 dB/m at 119 μm (2.5 THz) for a 2.2 mm bore, 90 cm long Ag/PS HCW.[15] By using low-loss dielectric coatings deposited over a metallic film to lower the loss, we are attempting to do for the terahertz regime what has been done so successfully for similar one-dimensional hollow waveguides in the IR region from 2 to 12 μm.[17]

The difficulty in developing efficient waveguiding technology for terahertz radiation results from the very high terahertz absorption in most dielectric materials and high Ohmic losses in metals. Specifically, the lowest absorption for polymers, which are often proposed for dielectric waveguides, is $\sim 0.1\ \mathrm{cm}^{-1}$ in the range from 0.1–0.5 THz. The absorption coefficient typically increases at higher frequencies. Even high-resistivity crystalline silicon, which offers one of the lowest losses in dielectrics throughout the terahertz band ($0.02\ \mathrm{cm}^{-1}$),[18] is too absorptive for transmission over several meters. Losses in metallic waveguides increase rapidly with frequency due to both the finite conductivity and surface roughness. As a result, neither dielectric waveguides, which perform well for optical waves, nor metallic waveguides, which are widely used for radio waves, offer a satisfactory solution for terahertz waves. This is evidenced by the reported high loss of ~4 dB/m (Ref. 2) for solid-dielectric fibers and an ~3.9 dB/m (Ref. 14) loss for a metal-only waveguide. The challenge for terahertz waveguides is to find very low-loss dielectric coating materials and to be able to deposit them uniformly inside small bore tubing with coating thicknesses of 10–20 μm, much greater than those typically encountered in the IR. The optimum dielectric layer thickness for metal/dielectric HCWs is proportional to the design wavelength, so the dielectric film thickness of a metal/dielectric HCW designed to transmit terahertz frequency radiation must be 10–100 times greater than those designed for IR radiation.

Numerous techniques have been used for fabricating IR

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transmissive, metallic/dielectric HCWs. These methods are well summarized in the book by Harrington.[17] In short they include nickel tubes with inner Ge and ZnSe dielectric coatings; Ag and PbF_2 layers on a thin metal strip rolled into a tube and inserted into a stainless steel sleeve; silver tubes with AgBr or AgCl films; Ag/AgI films deposited on the inner surface of polyethylene and Teflon tubing; and Ag/polymer coatings in hollow glass tubing. Today one of the most popular HCWs developed by our group is those made using either silica or polycarbonate tubing with an inner layer of Ag overcoated with a variety of dielectric materials including AgI, CdS, PbS, and PS. The measured loss for the large bore guides (>1000 μm) is less than 0.1 dB/m at 10.6 μm.

In this paper we extend the technology developed for IR transmitting HCWs into the terahertz regime. To realize a low-loss hollow waveguide we must be able to deposit dielectric films with very low absorption characteristics with a thickness of at least 5 μm or greater. For example, if we were to use AgI, CdS, and PbS these films would need to be 9.7, 9.3, and 5.4 μm, respectively, for operation at 119 μm. Such thick uniform films would be difficult to grow using our liquid-chemistry technique. Therefore, we have chosen to deposit a polymer, PS, as this material can be deposited with the requisite thickness. Furthermore, PS has a relatively low absorption coefficient in the terahertz region and the flexibility of PS will allow extension of this technology to flexible waveguide structures. We describe the fabrication process, physical characteristics, and terahertz transmission of HCWs with the PS coating ranging in thickness from 0 to 13 μm and with waveguide bore diameters varying from 1.6 to 2.2 mm.

II. METHOD OF WAVEGUIDE FABRICATION

The PS coatings deposited in this work follow the general procedures described by Bowden *et al.*,[15] which is similar to that used for the deposition of cyclic olefin polymer films on Ag by Abe *et al.*[19] PS is dissolved in toluene to form solutions with lower viscosity at high concentrations. Specifically, ACS grade toluene and 148G grade PS pellets purchased from Fisher Scientific and BASF Corporation, respectively, are combined to obtain solutions with concentrations ranging from 15 to 27.5 wt %. A TA Instruments AR 1000N rheometer is used to determine the viscosity of each solution. The viscosity data of the PS/toluene solutions have been fitted to the empirical relation

$$\eta = A \times 10^{BC}, \tag{1}$$

where C is the concentration and A and B are fitting parameters. The values of A and B are 2.6×10^{-3} and 0.079, respectively. This can be compared with the fluorocarbon polymer (FCP) films deposited inside silver coated fused silica tubing by Wang *et al.*[20] The viscosity of the PS solutions is much lower than that of the FCP solutions. For example, at 15 wt % the viscosity of the PS solution is about 73 times lower than the FCP solution. This is important because the lower viscosity of PS solutions allows thick films to be deposited under a more stable fluid flow condition than is possible using higher viscosity FCP solutions.

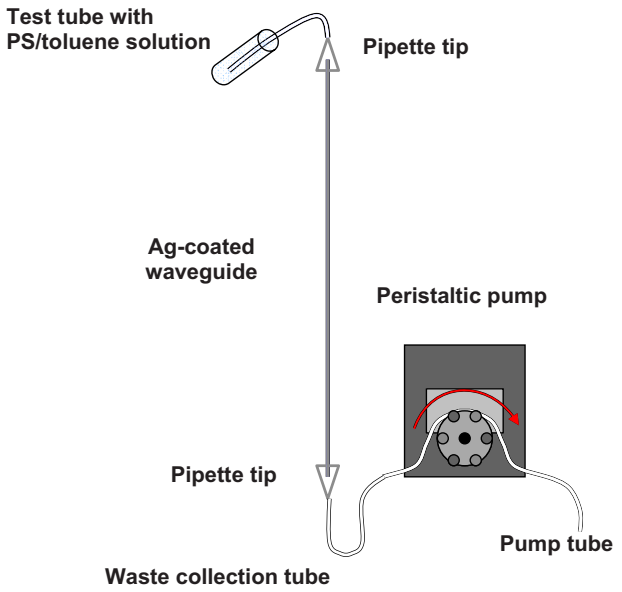

FIG. 1. (Color online) A schematic of the apparatus used to deposit polymer thin films by the liquid flow coating process.

The HCWs were prepared by first depositing an Ag film inside a glass substrate tube by combining a solution containing $AgNO_3$ and NH_4OH with a reducing solution containing dextrose and Na_2 ethylenediamine tetra-acetic acid as described in greater detail elsewhere.[21] The glass tubes had bore dimensions of 1.6, 1.7, and 2.2 mm and they were 120 cm long. The Ag film grows at a rate of approximately 1 μm/h and the Ag films were normally about 1 μm thick. This is much thicker than the skin depth for Ag at terahertz frequencies, which is 100 nm at 119 μm and, therefore, no terahertz radiation will penetrate the hollow waveguide wall.

The setup used to deposit the PS films over the Ag-coated guides is shown in Fig. 1. The setup is designed to draw the PS/toluene solution through the tubing at a constant coating velocity using a Masterflex L/S peristaltic pump configured with a microbore tubing pump head. A viton feed tube, which is chosen because it does not degrade when exposed to toluene, is connected to the upper opening of the waveguide and delivers the PS/toluene solution. The use of a peristaltic pump causes the coating velocity to pulsate. The use of a microbore peristaltic pump tube minimizes the amplitude of the pulsation. The pump creates a pressure difference between the atmosphere over the solution and the inside of the waveguide causing solution to be drawn into the feed tube. After a specific time interval the feed tube is removed from the vial while the pump runs continuously. This action forms a column of solution that passes through the waveguide at a constant coating velocity. The time interval between the insertion of the feed tube and its removal is adjusted for each combination of HCW bore diameter and coating velocity to produce a 10 cm long column of solution. As the column travels along the length of the HCW, a thin film of PS/toluene solution is deposited over the Ag coating. Once the column of solution exits the waveguide, the pump is stopped. The HCW is immediately transferred to a drying

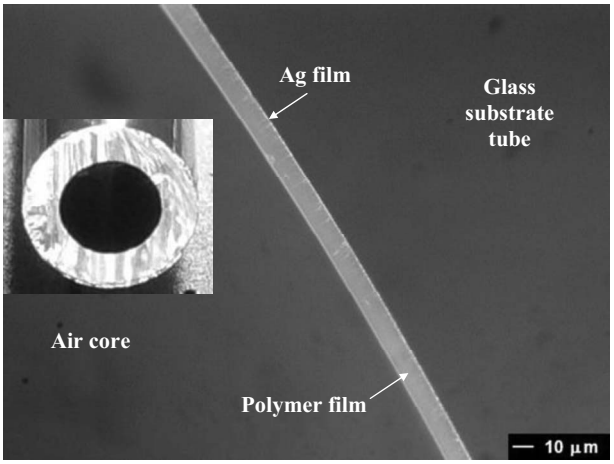

FIG. 2. An optical micrograph of a 2.2 mm bore diameter Ag/PS HCW. The insert shows the full cross section of the HCW.

apparatus that forces air through its core at a rate of 1 L/min. The guide remains on the drying apparatus for ~24 h to ensure complete evaporation of the solvent.

This process ensures uniform deposition of the PS coating with the required thickness up to 17 μm. An optical micrograph of the cross section of a 2.2 mm bore diameter Ag/PS HCW is shown in Fig. 2. The PS film, which is about 11 μm thick, is seen to be very uniform. This film was produced from a 25 wt % PS/toluene solution using a coating velocity of 4.5 cm/min. The PS film and Ag coating adhere well to each other and to the glass substrate tube.

Films with various thicknesses are obtained using a variety of combinations of solution concentration and coating velocity. The film thicknesses, calculated as described above, are plotted as a function of the square root of the coating velocity in Fig. 3. From Fig. 3 we see that the film thickness

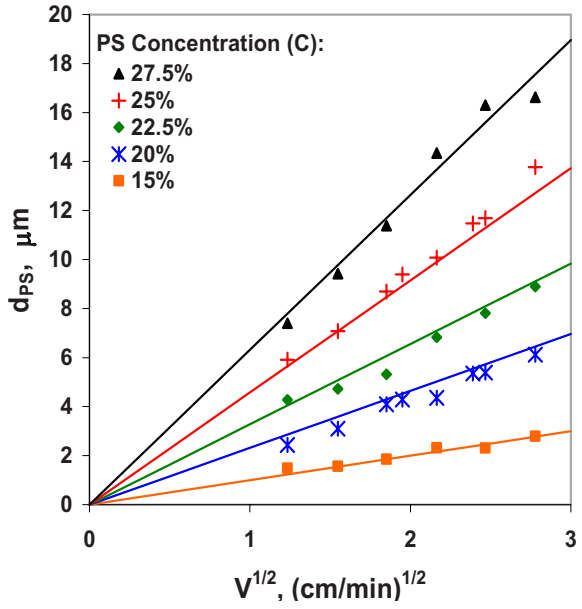

FIG. 3. (Color online) The thickness, d_{PS}, of PS films is plotted as a function of the square root of the coating rate, $V^{1/2}$, for various concentrations of PS in toluene. Markers indicate the measured data. Solid lines indicate the film thickness calculated from Eq. (3).

increases linearly as the square root of the coating rate increases for constant solution concentrations as predicted by the Fairbrother–Stubbs equation given by[22]

$$d_{PS} = A \times 10^{1/2BC} C V^{1/2}, \tag{2}$$

where d_{PS} is the thickness of the PS film; A is a constant that incorporates the surface tension of the PS/toluene solution, the viscosity proportionality constant, and the bore diameter of the waveguide; B is the constant that relates the viscosity of the polymer solution to its concentration, C; and V is the coating velocity. The thickest film is 16.6 μm achieved using a 27.5% PS/toluene solution and 7.7 cm/min coating velocity. The value of A and B are 0.042 and 0.019, respectively, as determined by least-squares regression. Thus, d_{PS} can be approximated by

$$d_{PS} = 0.019 \times 10^{0.042C} C V^{1/2}. \tag{3}$$

The solid lines in Fig. 3 correspond to the value of d_{PS} obtained by substituting the appropriate values of C and V into Eq. (3). Independent measurements of the viscosity give the value $B=0.079$, which is in good agreement with the value of $B=0.084$ determined by fitting the thickness data to Eq. (2). The agreement between Eq. (3) and nonoptical measurements of the film thickness is generally a good agreement.

Equation (3) has been determined for HCWs with a 1.7 mm bore size. The constant A is proportional to the bore diameter. Therefore, the PS film thickness can be predicted for any HCW with bore diameter, a in millimeter, by replacing the coefficient $A=0.019$ with $A=0.019(a/1.7)$.

III. PHYSICAL CHARACTERISTICS OF PS FILMS

HCWs were prepared with different bore diameters and PS film thicknesses. Parameters of the deposition process were adjusted in accordance with the thickness and uniformity of the PS film required for low-loss terahertz transmission. In particular, 60 cm long, Ag-coated glass tubes were coated using 15, 20, 22.5, 25, or 27.5 wt % PS/toluene solutions. For each concentration, waveguides were fabricated using coating velocities of 1.5, 2.4, 3.4, 4.7, 6.1, and 7.7 cm/min.

The thickness of the PS films was obtained from the Fourier transform infrared (FTIR) spectrum of the Ag/PS guide. The interference peaks occur at wavelengths, λ_m, given by

$$\lambda_m = \frac{d_{PS}(4\sqrt{n^2-1})}{m}, \tag{4}$$

where d_{PS} is the thickness of the PS film, n is the refractive index of the PS film, and m is a positive integer that corresponds to the order of the interference peak.[19] Rearranging Eq. (4), we obtain an expression for d_{PS} in terms of the difference in wave number, $\tilde{\nu}_m - \tilde{\nu}_{m-1}$, between any two adjacent interference peaks

$$d_{PS} = \frac{(\tilde{\nu}_m - \tilde{\nu}_{m-1})^{-1}}{4\sqrt{n^2-1}}. \tag{5}$$

The spectral response of the Ag/PS HCW in the near IR allows us to calculate the PS film thickness with high accu-

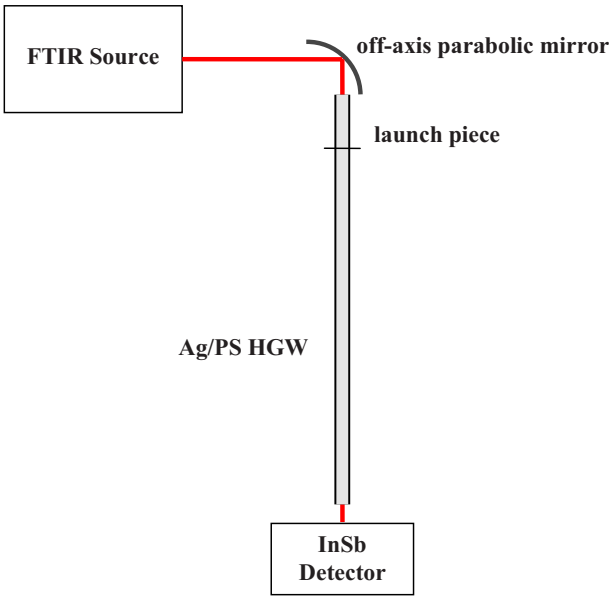

FIG. 4. (Color online) A schematic of the FTIR spectroscopy setup used to characterize Ag/PS HCWs.

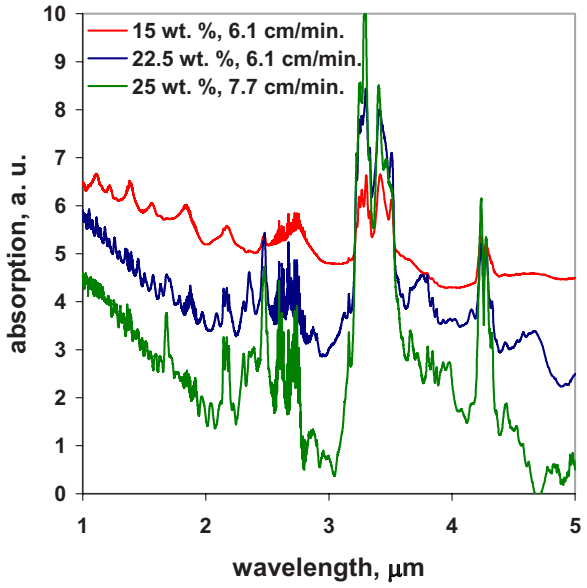

FIG. 5. (Color online) The near infrared spectra of 1.7 mm bore diameter Ag/PS HCWs with PS films deposited using various PS/toluene solution concentrations and coating rates. The vertical scale is offset for clarity. The curves correspond to 15 wt %, 6.1 cm/min (upper curve); 22.5 wt %, 6.1 cm/min (middle curve); and 25 wt %, 7.7 cm/min (lower curve).

racy. Some representative spectra and the calculation of the PS film thickness using Eq. (5) are given in the earlier paper by Bowden *et al.*[15]

The IR absorption spectrum is measured for each Ag/PS HCW using the FTIR spectroscopy setup shown schematically in Fig. 4. The external beam from the FTIR is focused into a Ag-only coated launch-tube HCW that conditions the mode for launching into the terahertz waveguide. A white light source and InSb detector are used to measure the absorption spectrum of the guides from 1 to 5 μm. Between 5 and 20 μm the absorption bands of PS are so strong that they obscure any interference effects from the PS film.

Figure 5 shows the absorption spectra of Ag/PS HCWs deposited using three combinations of solution concentration and coating velocity: 15%/6.1 cm/min, 22.5%/6.1 cm/min, and 25%/7.7 cm/min. From 2 to 5 μm the spectra are dominated by the intrinsic absorption bands of PS. Between 1 and 2 μm, where PS is transparent, interference peaks are clearly visible. The presence of sharp interference peaks indicates that the PS film is uniform along the length of the HCW. The spacing of the interference peaks is greatest for the 15%/6.1 cm/min waveguide and least for the 25%/7.7 cm/min waveguide, indicating that they have the thinnest and thickest PS films, respectively. The increase in absorption with decreasing wavelength seen in Fig. 5 is a result of the surface roughness of the coatings. The absorption peaks at 1.67, 2.14, and 2.46 μm are due to PS.

The thickness of the films is calculated using Eq. (5). By way of example, we use the data for the 22.5%/6.1 cm/min in Fig. 5 to calculate the thickness of the PS film. When the data are replotted from 4000 to 9000 cm^{-1} (2.5 to 1.1 μm) we see that there are 13 evenly spaced interference bands with an average spacing between adjacent peaks of 266 cm^{-1}. Assuming the refractive index of PS is 1.58 and substituting 266 cm^{-1} for $\tilde{\nu}_m - \tilde{\nu}_{m-1}$ in Eq. (5), the calculated PS film thickness is found to be 7.7 μm.

The THz transmission losses for the 2.2 mm bore HCWs with different thickness PS films are shown in Fig. 6. From the data it is clear that the loss decreases as the PS film thickness increases reaching a minimum attenuation of 0.95 dB/m with an 8.2 μm thick PS film. The loss measurements are performed in air at the normal humidity level. The water vapor absorption contributes ~0.5 dB/m to the total loss. The data in Fig. 6 are reproduced from Bowden *et al.*[15] They point out that while the measured loss is not as low as the calculated loss for the lowest-loss HE_{11} mode,[15] the trend toward lower loss with increasing film thickness is evident. Finally, it should be mentioned that the loss of 0.95 dB/m is still one of the lowest losses for any terahertz fiber optic measured in the terahertz regime.

IV. CONCLUSIONS

The terahertz waveguide technology pursued in this study is fundamentally an extension of the metallic/dielectric

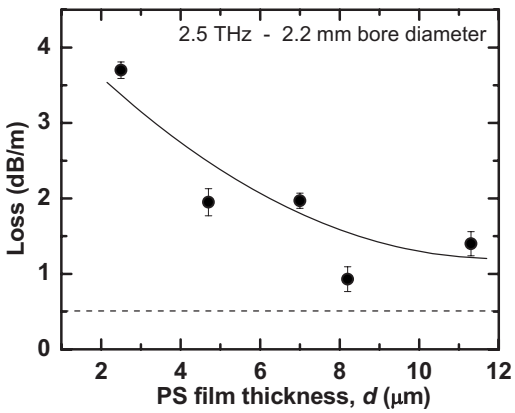

FIG. 6. Measured transmission loss at 2.5 THz for the 2.2 mm bore HCW as a function of PS film thickness. The trend is shown by the solid line and the approximate level of the water vapor absorption is shown by the dashed horizontal line. The lowest loss of 0.95 dB/m was measured for the 8.2 μm thick PS film. This data have also been reported in Ref. 15.

film HCW technology developed so successfully for IR applications. There are two major challenges to making a low-loss HCW for terahertz radiation. The first is to find truly transparent dielectric materials that can be deposited as thick films inside either glass or polymer tubing. In general there is not an extensive literature available on the optical properties of terahertz materials as there is for IR materials. Furthermore, some of the best known terahertz-transparent materials such as high-density polyethylene and silicon are not necessarily easy to deposit as uniform thick films inside our tubing. The second difficulty is the need to fabricate a waveguide with a dielectric film thickness of 10 to 15 μm, at least ten times greater than that which has been previously used in the IR region. We have chosen PS because it has a reasonably low loss at terahertz wavelengths and because it is possible to deposit smooth, uniform, well adhered PS films with thicknesses up to about 17 μm using a well established liquid-chemistry method. The key to the fabrication of such thick films is the high concentration yet low viscosity of the PS/toluene solution used to deposit them.

The Ag/PS coated HCWs have significantly lower loss for 119 μm radiation as compared to Ag-only HCWs. The improvement in the transmission characteristics is attributed to the change of the dominant transmission mode from TE_{01} for the Ag-only guides to HE_{11} for the Ag/PS HCWs.[23] The best waveguide demonstrated in this study is a 2.2 mm bore diameter Ag/PS HCW with an 8.2 μm PS film thickness. The measured transmission loss for this waveguide was 0.95 dB/m with a coupling efficiency of 81%.

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