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Simulation and experimental demonstration of CO2 detection using sequentially drilled and non-drilled open-air photonic fiber segments

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ABSTRACT

Many industrial processes currently require accurate and sensitive monitoring of CO_2 gas. To increase the sensitivity of CO_2 detection, open-air photonic fibers have been proposed. Open-air photonic fibers have a hollow core allowing the intercalation of gas into the fiber, allowing for the amplification of measurement techniques like absorption or Raman spectroscopy to detect CO_2 concentrations. We use conducted a variety of COMSOL based studies aimed at minimizing gas diffusion time throughout the hollow core. We investigate both pressure driven and diffusion based gas delivery methods, finding that both possess the ability to greatly reduce sensor response time. Preliminary experiments in a controlled environment validated the models and showed the ability to detect and control the uptake of CO_2 in open-air photonics fibers. This lays the foundation of a distributed chemical sensing system, particularly important for monitoring well integrity for carbon capture and storage, providing early warning for an incoming well failure and potential CO_2 leaking through it, potentially affecting proximal aquifers, a large public concern.

Keywords: Fiber, Environmental Sensing, Hollow-Core, CO_2, Raman, Spectroscopy

1. INTRODUCTION

Since their realization in the 1990s, microstructured optical fibers have steadily gained popularity in a wide variety of scientific disciplines. Notably, they are actively being used in quantum computing, gas spectroscopy, and laser systems. Though one type of microstructured fiber known as the hollow core photonic crystal fiber (HC-PCF) has become very popular in the field of spectroscopy. HC-PCFs, as seen in Fig. 1, have an entirely hollow core that is surrounded by a crystalline structure. The crystalline silica cladding causes light to be confined nearly exclusively to the hollow core. Thus, light propagates through the fiber in this hollow region. Naturally, these fibers are being employed as gas sensors as the fibers essentially act as gas cells. These fibers can serve as highly sensitive gas sensors because the fibers themselves can be very long, allowing for light-gas interaction to occur over great lengths.

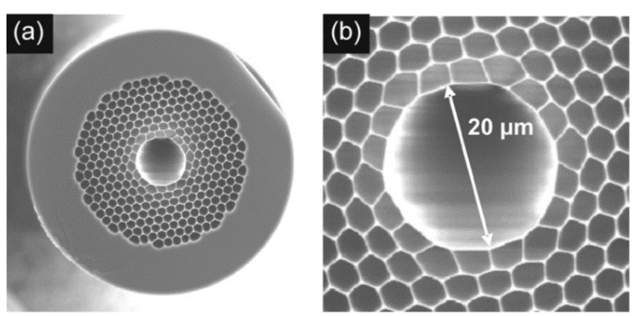

Figure 1: A scanning electron microscope image of a hollow core photonic crystal fiber [13]. Light propagates almost entirely in the hollow core of the fiber allowing for exceptional light-gas interaction capability.

Various configuration have been considered from the simplest experimental setup, diffusing gas through the ends of fibers [1-3], to alternative ones either involving pressurizing one end of the fiber [4, 5] or drilling holes into the side of the fiber to allow gas to flow into the core at several points along the fiber's length [6-8].

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Our critical target has been combining sensitivity/selectivity of Raman/IR with true distributed sensing for multipoint optical detection and scale-up capability. We have previously demonstrated the capability of IR and/or Raman interrogation in HF-PCFs of several volatiles in small volumes on short fiber stocks [9]. As light and chemicals overlap along the same hollow core, the increased light-matter interaction is increased by several order of magnitude enabling ppm level of detection [9]. Our research objectives are to demonstrate CO_2 detection with commercially available HF-PCF via diffusion or pressurization; enable rapid detection via optimized positioning of slotted apertures along the fiber side or at their edges; interleave with FBGs for spatial location and environmental monitoring when used with IR; optimize splicing with solid core fiber if needed for deployment along wellbore 100s-1000s meter stretches. We present some of our recent results and ongoing efforts.

Experimenting with HC-PCFs can be extremely expensive as they are highly specialized fibers, so the need for simulating and mathematically modeling gas flow through the fibers has become essential in designing experiments [10, 11]. In this manuscript, we present our detailed work on modeling and simulating a variety of different gas delivery methods for gas in HC-PCF fiber to eventually guide the experiments. Indeed, we also provide initial laboratory data on the detection and uptake of CO2 in hollow core fibers via Raman spectroscopy validating the model.

2. MODELING

To conduct the studies presented in this paper, we used COMSOL Multiphysics to model the physical processes. We based all the physical parameters of the models off the NKT Photonics HC-800-02 HC-PCF [12]. Additionally, we went through a validation process where we used our model to accurately reproduce experimental findings from three distinct experiments found in the literature.

In the present work, we introduce the average value of the gas concentration in the fiber relative to the concentration of the incoming gas. Hereafter, we refer to this quantity as the "average relative concentration" of the gas, and mathematically define the quantity as:

$$C_r(t) = \frac{1}{C_0 V} \iiint_V C(\vec{x},;t)dV \qquad (1)$$

where C_0 is the concentration of the incoming gas (assumed constant), V is the volume of the hollow region inside the fiber, and $C(\vec{x};t)$ is the concentration field inside the fiber. This metric allows us to monitor how the gas flows throughout the fiber, and ultimately will allow us to compare our models to experimental results. In our experiments, we use the 95% average relative concentration as a standard measure of comparison. In the paper, we refer to the amount of time for the fiber to reach 95% average relative concentration as the fiber "response time."

There are a multitude of ways to deliver gas to the core of the fiber for sensing via absorption or Raman spectroscopy, though they can naturally be divided into two broad categories. The first category deals with sensors which rely on gas diffusing through the hollow fiber. The second deals with sensors which use pressure to force gas through the fiber's core. We studied both gas delivery methods and present the results below. Additionally, for both the diffusion based and pressure driven gas delivery systems, we simulated two different physical setups. We first simulated gas flow through the end holes of the fiber only. Additionally, we simulated gas flow through side holes along the fiber. Note that in all the studies presented below, we simulated the diffusion/flow of CO_2 at 273 K through the fiber.

We also, at first used COMSOL to determine the optical modal properties. 2D optical mode distribution validated single mode operation with optical parameters was confirmed with the fiber providing company. Due to the periodic structure of the cladding, light is confined nearly entirely to the hollow core (filling factor f = 99%), allowing the gas and light active volumes to fully overlap, thus making the fiber an extremely sensitive sensor. The light can interact with the gas as if it were propagating through free space. The relative sensitivity defined as $r = f\, n_{gas}/n_{eff}$ which compares the light propagating through an optical fiber (n_{eff} is the mode effective index) with light propagating through free space is ~1 [1, 2]. This relaxes the absorption analysis to a 1D optical absorption/raman model that when paired with diffusion sufficed to validate literature experimental results and helped tremendously in accelerating any following configuration optimization, given a lighter COMSOL hybrid model with reduced memory requirements.

2.1 Diffusion driven filling

We first focused on studying how gas diffuses through the core of the fiber and what effects the amount of time it takes for the gas to diffuse. We simulated gas diffusing through the fiber end holes, and through side holes along the fiber.

2.1.1 End holes

The easiest sensor to design and setup experimentally is a sensor where gas simply diffuses through the ends of a fiber. However, while this is the most straightforward way to introduce gas into the core of the fiber, it is not the fastest way for gas to spread through the fiber. We conducted a simple set of simulations to demonstrate the shortcomings of relying on diffusion through end holes in such sensors. This served as motivation to seek alternative methods for introducing gas to the core of the fiber so that we can produce a sensor with a long length, though fast response time.

In this study we allowed CO_2 at 273 K and 1 atm to diffuse through both ends of the NKT HC-800-02. Between studies, we varied the length of the fiber and recorded the amount of time needed to reach 95% average relative concentration in the core of the fiber. The results are plotted in Fig.2 and illustrate that diffusion through end holes is not suited for use-cases which value quick sensor response time. For instance, a fiber of only half a meter long will take over an hour to reach 95% average relative concentration.

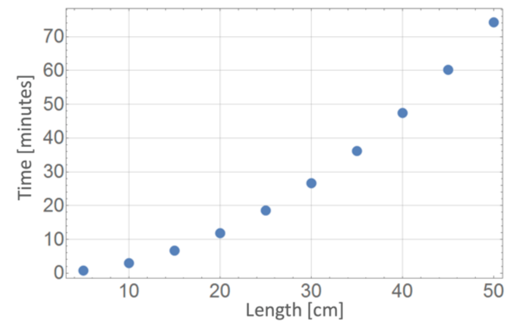

Figure 2: Time to 95% average relative concentration of CO_2 diffusing through both ends of varying lengths of HC-800-02.

Thus, this study served as motivation to explore alternative techniques for introducing gas into the core of the fiber as we seek to engineer a sensor that is capable of both quickly detecting gas and has a long length to increase the sensitivity of the device.

2.1.2 Side holes

The natural next step after simulating diffusion through the fiber was to simulate diffusion through side holes. These simulations are of particular interest because we expect reduced uptake time. The first suite of simulations that we conducted were aimed at investigating how the number of holes impacts the fiber's response time. In this study, we simulate gas diffusing through 1-5 side holes only in a 10 cm long fiber (Fig. 3).

It becomes clear that the response time lowers with each additional hole added, though the marginal benefit decreases. However, what is most important to note about this is that these findings can generalize to fibers of *any length*. If we

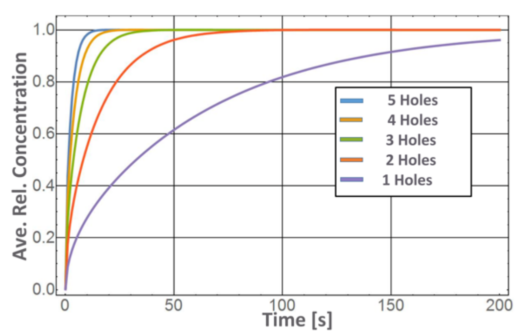

Figure 3: Average relative concentration as a function of time for varying number of holes on 10 cm segment of HC-800-02.

consider the spacing between holes, this becomes clear. To illustrate, we simulate diffusion of CO_2 through both ends of varying lengths of the same NKT Photonics fiber. The results, presented in Fig. 4, are identical to the previous simulation where we varied the number of holes.

Ultimately, this means that one can reduce the response time of an HC-PCF sensor arbitrarily low by introducing more holes. However, introducing holes comes at the cost of increasing optical losses due to scattering. Hoo et al. report that the optical loss from 7 holes on a section of 7cm fiber is 0.5 dB [6]. However, this relatively small loss/hole suggests that one could feasibly produce extremely sensitive HC-PCF sensors using several meters of fiber, though have response times on the order of seconds. While diffusion through side holes is a very promising method for minimizing sensor response time while maintaining high sensor sensitivity, the effects of side holes on optical losses has not been studied extensively which will be focus of our experimentations. Overall based on the model results, a separation of ~3cm provides at least an order of magnitude improvement in the uptake time (from ~200sec to ~20sec) which guided the design of our experiments.

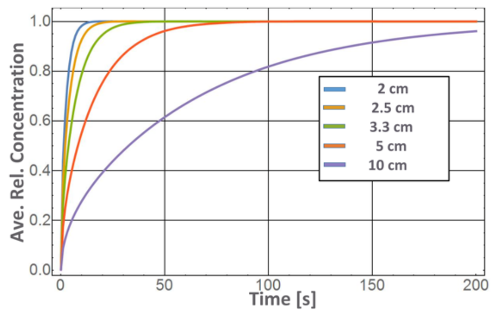

Figure 4: Average relative concentration as a function of time for five segments of HC-PBF of different length.

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2.2 Pressure driven filling

An alternative to using diffusion to introduce gas to the core of the fiber is to use pressure to force gas through the fiber. In principle, this technique would allow gas to fill the core very quickly. However, it can be very difficult to engineering and build such a device which uses pressure to fill the core. Again, we investigate pressure driven deliver in the two distinct physical setups: gas flow through end holes and through side holes.

The theory underlying diffusion-based gas flow versus pressure driven gas flow is similar. Both types of simulations require the Navier-Stokes equations describing fluid flow be coupled to the Convection-Diffusion equations describing a diffusing species (Fig. 5A)). However, the momentum term in Navier-Stokes equation is modified to include the shallow channel approximation term if its height is much smaller than its length like the ones of interest to us. In these micro-scale systems, there is significantly more drag due to the walls.

2.2.1 End holes

We first investigated how the length of the HC-PCF impacts fiber response time in pressure driven filling systems while maintaining a fixed pressure difference. We maintained the inlet of the fiber at 2 atm and the outlet at 1 atm and varied the length of the fiber. The results of the study are presented in Fig.5B).

This study shows that pressure driven gas delivery has the potential to drastically reduce the sensor's response time. For comparison, we predicted that a half meter long fiber will take over one hour to fill when the gas diffuses through both ends of the fiber. However, with a modest pressure difference of 1 atm, a half meter long fiber could fill in under 30 seconds. This is remarkable, though we also aimed to understand how the pressure difference impacts the device's response time. To do so, we maintained the fiber's length at 25 cm and varied the pressure at the inlet in increments of 01 atm. We present the results in the following Fig. 6. Naturally, by increasing the pressure at the inlet, the fiber will faster. However, as the inlet pressure increases, the marginal difference in response time dwindles. Regardless, if one can supply extremely high pressures, they would be able to fill the fiber arbitrarily quickly.

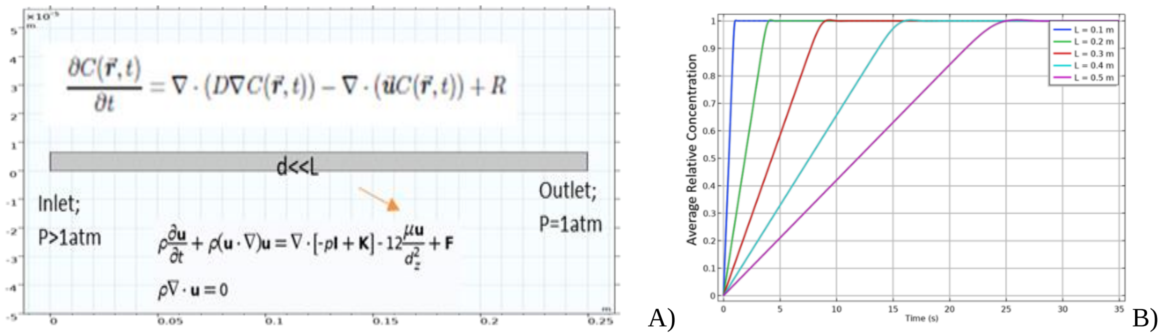

Figure 5: A) Convection-diffusion and Navier Stokes equations for pressurized system in the schematic; B) Pressure driven diffusion through the varying lengths of HC-PCF. For each case, the inlet was maintained at 2 atm and the outlet at 1 atm.

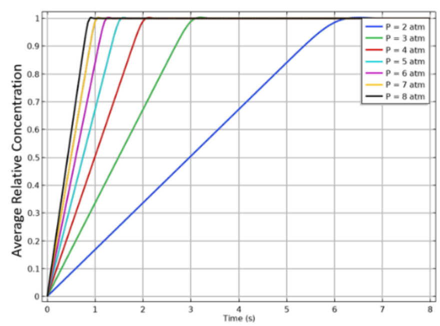

Figure 6: A 25cm long fiber with varying pressure applied at the inlet. In each study, the outlet was maintained at 1 atm.

2.2.2 Side holes

After exploring the basic properties of pressure driven gas flow through the ends of a fiber, we turned to fibers with side holes. We expected that these fibers would be advantageous because they would allow us to draw gas in through the side holes as opposed to forcing gas through the end holes. The first property we were interested in exploring was how the number of side holes impacted filling time. To investigate, we simulated four identical 25cm fibers with 1-4 holes. The results are found in the Fig.8. As more holes are introduced along the length of the fiber, the fiber does not necessarily draw more gas in through those holes. Instead, gas flows between the ends and the outermost holes. However, between the innermost side holes, gas simply diffuses. Thus, adding more holes does not cause the fiber to fill more quickly.

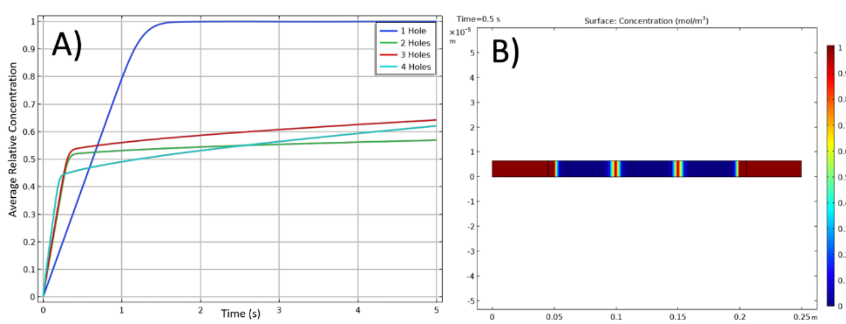

Figure 7: A) Varying the number of side holes in a 25cm length of HC-PCF while maintaining 0.1atm on one end holes and 1 atm on the side holes. B) In the figure, color indicates average relative concentration. Thus, as more holes are introduced, gas does not flow between the outermost and innermost holes. In this inner region, gas diffuses though doesn't flow due to a pressure difference.

Having discovered that a fiber with only one side hole will fill most quickly, we then investigated how both pressure and length impact the gas filling dynamics. We first studied the impact of length. To do so, we maintained the ends of the fiber at 0.1 atm and the hole at 1 atm. Additionally, to study the impact of outlet pressure on gas filling dynamics, we maintained a fiber length of 25cm with one side hole in the center and varied the pressure. These results are shown in the following Fig. 9. Changing the length from 50 cm to10 cm the time for equilibrium is reduced significantly from ~6sec to ~0.2secs at a pressure drop of 0.1atm. Maintaining fixed the length instead, the larger the pressure drop the longer the time for equilibrium, going from ~1.5 sec at 0.1atm to ~5sec for 0.attm.

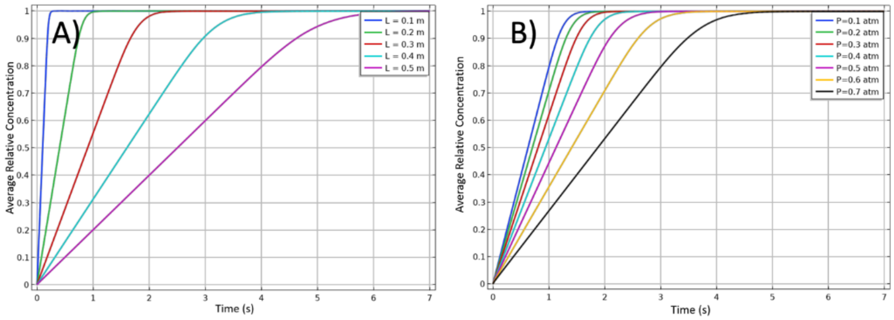

Figure 8: A) The average relative concentration in the fiber core as the length of the fiber with one hole in the middle varies. The end holes are maintained at 0.1 atm while the hole in the middle is held at 1 atm. B) The average relative concentration in the fiber core as outlet pressure varies. Between studies the fiber is maintained at a length of 25 cm.

3. TESTING

We have built several setups for Raman investigation at 785nm dedicated to diffusion processes studies (Figure 10 A), and at 532nm where we focus on pressure driven flows. The 785nm system has the advantage to reduce the fluorescence from background but it is weaker than Raman at 5332nm which instead suffers from fluorescence. Having excitation wavelength available will give as a way of downselecting the best performer.

The hollow core fiber HC-800-2, PMC-C-Green-26, and HC-1550-2 were procured and tested for throughput, but also cleaved into segment to validate the length dependent models and allow various novel fabrication tests. We were able to flow CO_2 in a controlled box at several pressures and measure CO_2 Raman signal dependence (Figure 10B)). This is a very promising result since the Raman signal is very weak and we are far from optimized as we have been limited in power and integration time: the power coupled into the PCF via the portable Intevac DeltaNu Inspector Raman gun is << 1mW, ~10cm^{-1} resolution, and we were limited to 1 minute integration only. Furthermore, additional filters have been installed to help cleaning up the from the signal and increase SNR of the CO_2 doublet (1276 cm-1 and 1293 cm-1) that is right on the shoulder of the Raman emitted by the silica of the fiber itself. We have been modifying the system to collect data continuously while introducing CO_2 rather than measuring just at end of flow. The first data on uptake of CO_2 were taken for 2" and 6" hollow core fibers and are aligned with the simulation data (Fig. 10C)). The 532nm system is made in-house and compact, composed of OEMLaser DPSS laser at 532nm with very low jitter, high power up to 200mW, a compact Ocean Optics QE Pro spectrometer with ~10cm-1 resolution and a GloPhotonics micro cell made of a 532nm HoF and two compact gas cell at both ends for controlled pressurized experiments (Figure 10b). This setup that required specialized optics for the specific wavelength was just completed and checked for alignment and throughput operation.

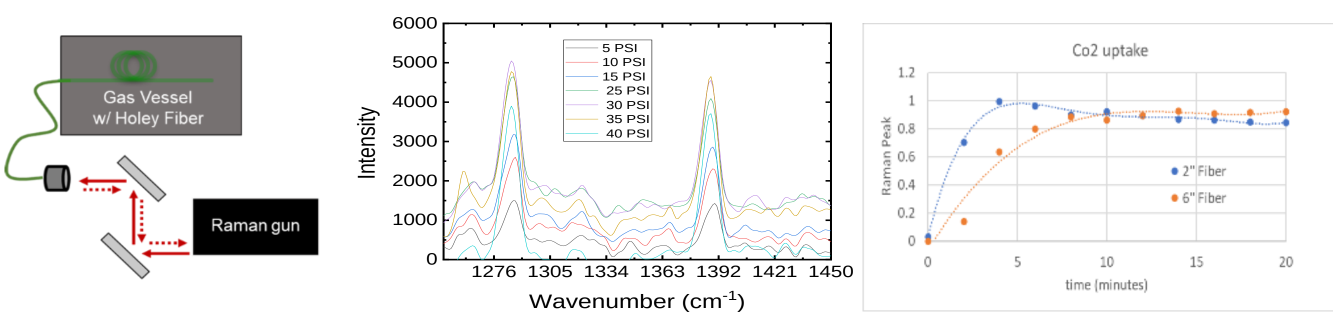

Figure 10. A) Current Raman system at 785nm with a Portable Raman gun and schematic; B) Raman spectra for CO2 at different pressure and C) CO2 uptake for 2' and 6" fibers

We selected and procured FBGs to investigate enhanced back reflection of the Stokes shifted signal (~830nm). For their use will be critical to enable splicing of the fibers to PCF fibers. In general, this will be a necessary capability for economically deploy PCFs for Kms downhole or subsurface, as they are currently quite more expensive than standard telecommunication fibers. Splicing or cleaving PCFs is delicate since given the holey nature and the lattice morphology they could not be stressed otherwise the fiber collapses on itself and there is no significant throughput. Nevertheless, we were able to achieve splicing and manage losses of ~10%. What is even more challenging is to create a join collar of only few 10s of μm that could hold the ends of both the PCF and solid core fiber together and maintain a gap to enable gas ingress. First attempt: 2% power throughput measured at 800nm via the collar splicing of 780nm transmitting solid core fiber to hollow core fiber (Figure 11A)). Although we successfully made the mechanical joint, it seems fragile and not able to sustain too much handling (easily breaks). We resorted to another simpler approach by just sliding the two ends into a mating sleeve that already comes with a large opening as an off-the-shelf item (Figure 11 right) – this setup guaranteed 50% throughput. The ingress pf CO_2 through the collar is being verified

In parallel we have been working on fabrication of holes in the PCFs by using a femtosecond laser that would drill multiple holes along the fiber length on reel-to-reel setup. First tests are promising for continuing tuning the laser power, focal length, and positioning. With a 40fs pulsed Ti-Sa 800nm laser 100 µJ, 20x @ 0.40 NA objective, 1 µm/s speed we were able to drill an array of 1-2 μm holes along 10cm fiber length (Figure 12)). We are about to put these technologies to the test in the diffusion experiments.

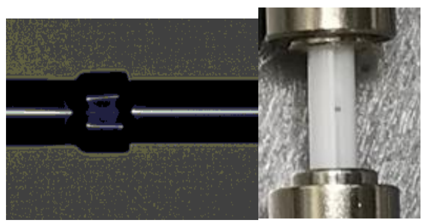

Figure 11. Hollow core to solid core fibers splices splice using (left) a C shaped collar of ~60um and (right) off-the-shelf mating sleeves.

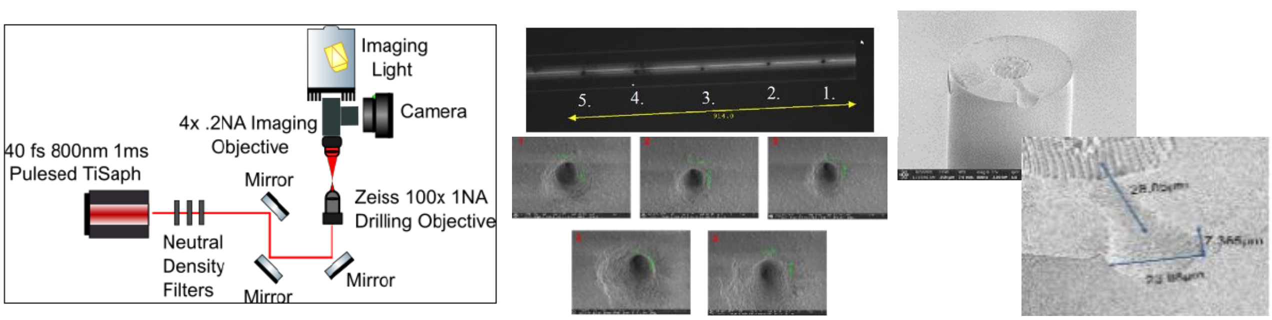

Figure 12. Setup for fs drilling; Image of 5 drilled holes in PCF; SEMs images of holes on the side and at cleaved facet showing etch depth control.

4. CONCLUSIONS AND FUTURE WORK

Ultimately, we were able to successfully demonstrate our modeling capabilities for a HC-PCF gas sensor in a variety of device setups and gas delivery methods. We investigated both diffusion based and pressure driven gas delivery methods to understand what the different methods offered. We found that diffusion through end holes is the most straight forward physical setup, though can lead to extremely long sensor response times as the fiber length increases. However, this issue can be mitigated by introducing side holes along the fiber, allowing for arbitrarily fast response times depending upon how many side holes are drilled. However, the side hole diffusion method is hindered by optical losses due to drilled side holes. Additionally, upon investigating pressure driven gas delivery, we found that pressure driven gas sensors can possess extremely short response times. We found that the response time depends on both the length of the fiber and the pressure difference between the inlet and outlet/s. In studying the impact of side holes in pressure driven systems, contrary to our intuition, we found that one side hole is the optimal number of side holes. However, while pressure driven gas delivery systems offer the potential for extremely fast response times, it depends entirely on what pressure differences one can experimentally achieve. Additionally, pressure driven systems are a significantly more challenging to engineer than diffusion based systems are. Depending on the application, it could be an appealing detection solution.

We aim to use these models and their results to design our experimental setup and aid in the design of our gas sensor. Initial experiments have indeed being guided and provided encouraging preliminary result validating the models trends. Additionally, we seek to compare the benefits of diffusion based and pressure based systems and assess which will suit our needs more appropriately. We are going to experimentally investigate the physical behavior which COMSOL predicts in the multi-hole pressure driven system at 532nm . It is possible that this behavior is an artifact of the initial conditions, though regardless would be interesting to study experimentally. We additionally seek to study the impact of side-holes on optical losses as this is essential in finding balance between sensor response time and sensitivity.

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