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A review on all-optical fiber-based VOC sensors: Heading towards the development of promising technology

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Abstract:

The direct exposure of volatile organic compounds (VOCs) is widely linked with several adverse health effects. Optical fiber-based VOC sensors offering huge advancements in technologies to overcome several limitations observed from convectional VOC sensors that appeared over the past decade. Real-time monitoring and detection of VOCs are important to maintain a safe and healthy domestic/industrial environment. The optical fibers are highly-sensitive and widely applied to several ranges of parameters including gas sensing, and have also been used to monitor VOCs at the sub-ppm range. Recognizing the major developments in the field of optical fiber, this article aims to review the recent progress in VOC monitoring, resulting in a wide range of sensing configurations are being discussed. Additionally, the article highlights the advantage, limitations, and future possibilities in this area.

Keywords: Optical fiber, Sensor, Volatile organic compounds, *in situ*, Monitoring, Toxic.

1. Introduction:

Generally, making a fire, driving a car, house paint, building materials, cutting the grass, or simple breathing, results in the release of various organic compounds such as alcohols, carbonyls, esters, alkenes, alkanes, amides, and ethers. More commonly, we can say that almost every daily life activity of humans leads to the emission of organic compounds in the environment [1]. Since the development and revolution of industries, various activities, such as the petroleum industry, have emitted an excess amount of harmful pollutants including various kinds of toxic gases and volatile organic compounds (VOCs) [2], [3]. In general, VOCs are organic compounds which have a high vapor pressure at room temperature, i.e., they can easily get evaporated into the gaseous form at indoor temperature. The concentration of VOCs in the indoor environment is nearly 10 times higher than the outdoor. It has been estimated that approximately 50-300 VOCs have been detected in the indoor air at schools, homes, offices, and various shopping complexes and commercial buildings [4]. Moreover, the use of a broad range of personal care and consumable products can release a significant amount of toxic VOCs into the atmosphere [5]. Hence, VOCs level is used as a criterion to assess the air quality in the indoor environment in addition to other measures such as temperature and CO₂ levels [6]. VOCs can be found in both outdoor and indoor environments. For the outdoor environment, the common source of their presence is mainly arising by vegetation, incomplete combustion of fuel, minerals, and petroleum products. However, the emission from terrestrial and ocean sources are the natural source of their outdoor release. For the indoor VOCs, particleboard, plywood, gas, glues, wood, kerosene, etc. plays a major role in its emission. The source of VOCs and their contribution in percentage to the environment is shown in Fig.1 [7].

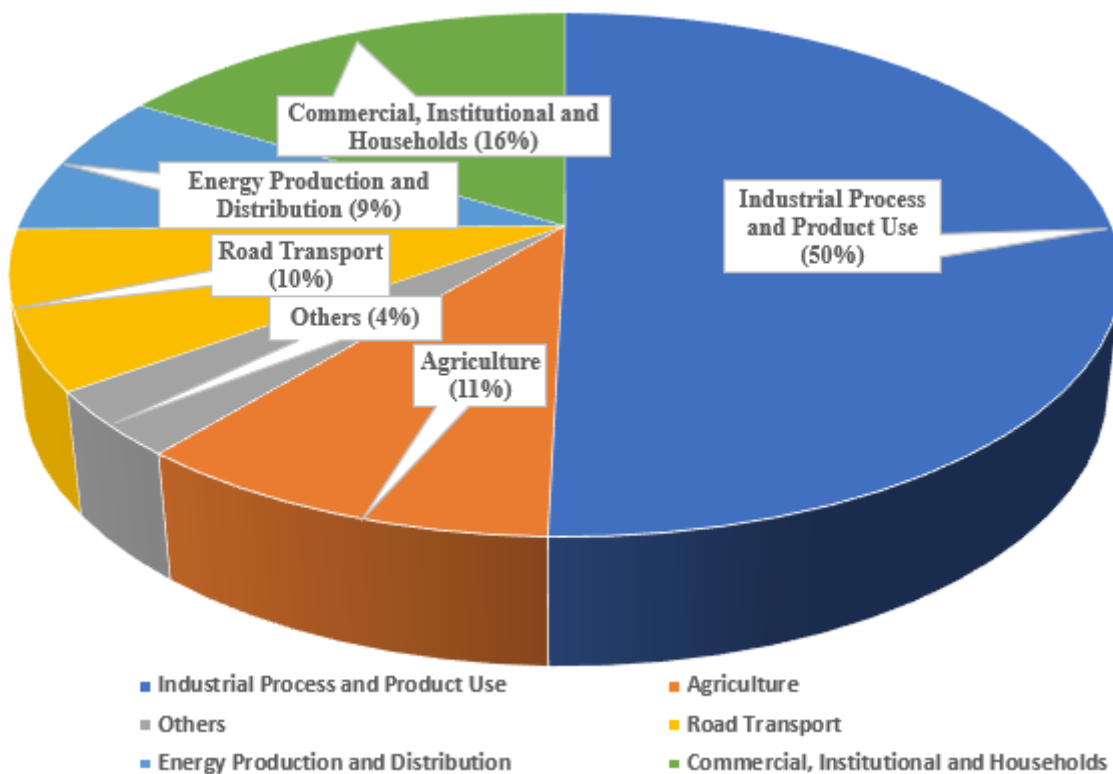


Fig. 1. Source of VOCs and their contribution to the environment.

Regarding the monitor and control risk of VOCs, numerous agencies, such as the Environmental Protection Agency, National Institute of Occupational Safety and Health, and European Agency for Safety and Health at Work has developed and issued guidelines to limit the exposure of humans to the potential VOCs in workplace air. This is because of their unfavorable effects on human health, even if it is available at low concentrations. Some VOCs have been identified as carcinogenic or highly toxic in nature and can have a long and short-term effect on human health as well as on the environment. The most common VOCs are acetone, acetaldehyde, benzene, ethyl acetate, carbon tetrachloride, heptane, isopropyl, hexane, alcohol, naphthalene, formaldehyde, and toluene. Most of them are toxic such as alcohols, aromatic compounds, aldehydes, and ketones. However, toluene, benzene, xylene, and ethylbenzene are the kind of aromatic hydrocarbons, and considered as the most harmful compounds among all the available VOCs with a high potential to affect humans both systematically (e.g., circulatory, the nervous, reproductive, cardiovascular, immune, and respiratory systems) and specifically (e.g., kidneys, liver, stomach, and spleen). Therefore, regular monitoring of VOCs is needed to control and measure the atmospheric environment and health of human beings.

Accordingly, highly sensitive and selective analytical methods including gas chromatography (GC) [8], spectrophotometry [9], and liquid chromatography [10] have been used since the past years for the accurate measurement and quantification of VOCs. These techniques utilize the ion-pair extraction of the analytes and quantification by mass spectrometry to detect the VOC concentrations as low as ppt. However, the most significant limitations of these techniques are typically time-consuming, expensive, need of skilled technicians, off-site analyses, and matrix-matched calibration standards that are not routinely determined. To overcome these issues, some other sensing techniques are proposed such as electrochemical gas sensors (EGS) [11], quartz crystal microbalances (QCM) [12], metal oxide semiconductor (MOS) [13], non-dispersive infrared gas sensors (NDIGS) [14], colorimetric gas sensors (CGS) [15], etc. Each reported method has its advantages and limitations depending

upon the environment and the nature of molecules. The EGS have issues related to zero drift and aging while the MOS-based sensors operate on high temperature and show high sensitivity to gasses and external humidity.

During the past decades, massive advancement has been performed in the field of optical-based sensors for the application in various fields including biosensing, chemical sensing, gas sensing, etc. [16]–[28]. The sensors based on optical techniques can detect gasses at the ppt level which is more sensitive and selective compared to conventional sensors [29]–[31]. The optical sensors are robust in nature with a fast response time without getting affected by drift. Various kinds of optical detection techniques are available with direct spectrometry, in which the reagent/film/polymer based optical sensor are the most common types of detection technique used in sensing applications. In a later type of sensing system, the change in the optical response of the intermediate agent is utilized to quantify the analytes absorbed on the surface and then to deduce the concentration of gas once its calibration is performed. Since the invention of optical fiber, optical detection techniques are mostly centric toward the development of optical fiber-based sensing devices. The optical fiber-based sensing device appears as the most promising candidate for gas detection due to its small size, high sensitivity, and immune to electromagnetic interference, chemical inertness, multiplexing capability, and robustness to harsh environments. Various kind of optical fiber sensors has been reported till date in order to improve their advancement, and sensitivity enhancement [32]–[43].

In this review article, a detailed study has been carried out on the classification, progress, and research gap of the various sensing mechanisms used to develop the optical fiber-based VOC sensors. The review article is organized as followed; in section 2, a study of the most popular and toxic VOCs and their adverse effect on human health is provided; section 3 illustrates the applications of VOC sensors in our daily life; section 4 contains the detailed discussion over various sensing configurations and materials used to develop the most compact and low-cost sensing structures for VOCs and SVOCs monitoring. Finally, sections 5 and 6 emphasize future challenges and the summary of the review.

2. VOC: General definition, classification, and properties:

An organic compound refers to the molecular structure of any chemical compound consisting of a minimum one hydrogen and one carbon atom in its molecular structure. VOCs are the specific organic compounds which are presenting in the environment as atmospheric gases but sometimes can also be found in the form of solid or liquid at room temperature and pressure. These organic compounds can be classified according to its rate of evaporation such as VOCs (more than 95% by weight), non-volatile organic compounds (NVOs) (less than 5% by weight), and semi-VOCs (between 5 to 95% by weight). Each VOCs has different physical properties. However, they are generally, have very low boiling points, colorless and most of them are highly reactive and are often mixed with interfering atmospheric gases as shown in Table.1. The adverse effect of some popular VOCs on human health are listed, in terms of their threshold limit value.

Table.1. The name of VOCs, their chemical formula, immediate effect on health and threshold values of some selected VOCs [44].

VOC name	Formula	Immediate effect on health	Threshold value
Acetylene	C_2H_2	Dizziness, fatigue, headache, tachycardia, nausea, vomiting	N/A
Acetone	C_3H_6O	Headache, fatigue, mouth dryness, nausea, and dizziness	750 ppm
Cyclohexane	C_6H_{10}	Irritation to the eyes, throat, respiratory tract	300 ppm
Benzene	C_6H_6	Narcotic effects, convulsion, respiratory failure	1 ppm
Formaldehyde	$HCHO$	Eye irritation, shortness of breath, chest tightness or pain	0.75 ppm
Ethanol	C_2H_5OH	Headache, irritation of eyes, drowsiness	1000 ppm
Methanol	CH_3OH	Airway narrowing, vertigo, headache, bronchial constriction, nausea	200 ppm
Toluene	C_7H_8	Serious damage to liver, skin, kidneys, and the central nervous system	100 ppm
n-butanol	C_4H_9OH	Dizziness, drowsiness, damage eyes, and skin due to its stimulating nature	100 ppm
2-propanol	C_3H_8O	dizziness, drowsiness, nausea	400 ppm

The detail of adverse effect of several VOCs on human health is discussed below

2.1. Acetylene (C_2H_2)

Acetylene is popularly known as an extremely dangerous gas which is associated with its intrinsic instability, specifically when it is heated, pressurized, liquefied, or mixed with air. The consequence from the combustion of acetylene produces a huge amount of heat which can explode beyond the absolute pressure of gas i.e. 103 kPa [45], [46]. Direct exposure to acetylene may lead to dizziness, fatigue, headache, tachycardia, nausea, and vomiting, and in case of exposing with high concentrations of acetylene may cause consciousness and death.

2.2. Acetone (C_3H_6O)

Acetone is a metabolite of human fat and glucose, which can be found in human breath. The concentration of acetone in exhaled breath is associated with the concentration of blood sugar [47]. Acetone is very harmful to human health. Its direct inhalation may lead to irritation in the nose, eyes, and throat, even for a short time exposure around 5 min at a low concentration of about 300-500 ppm. In high concentrations, it can lead to headache, fatigue, dryness in the mouth, nausea, loss of coordinated speech, and dizziness [48].

2.3. Cyclohexene (C_6H_{10})

Cyclohexene is a nonpolar cycloalkane and mostly used in the synthesis of maleic acid, adipic acid, and butadiene, inorganic synthesis, and in oil extraction as a stabilizer material for high octane gasoline. Similarly, to acetone, its inhalation at a low concentration also creates irritation to the eyes, throat, and respiratory tract. Exposure to the high concentrated cyclohexene can cause drowsiness [49].

2.4. Benzene (C_6H_6)

Benzene is a natural component of both refined and crude petroleum. However, its 98% part is produced from the petrochemical and petroleum refining industries [50]. In the urban area, it is produced from exhaust fumes of the vehicles which is a particular threat to the cities. In 1994, benzene was listed in the top 20 most abundantly produced toxic chemicals in the United States [51]. After that, the maximum allowed exposure limit has been set to $16.25 \mu\text{g}/\text{m}^3$ to avoid health and environmental issues [52]. Additionally, benzene is one of the most widely

used compounds in the synthesis of various resins, polymers, and synthetic fibers. At a low concentration around 200 ppm, the inhalation of benzene may result in the narcotic effects in humans while the inhalation at high concentration may lead to convulsion, and its fatal for humans due to the respiratory failure. It can also because of leukaemia and lymphomas for long exposure [53].

2.5. Formaldehyde (CH₂O)

Peoples are mostly influenced to formaldehyde by inhalation as it is one of the widespread gases at room temperature. Even the inhalation of its low concentration may lead to various damages such as throat and eye irritation, shortness of breath, and chest tightness or pain. [54]. Moreover, a high concentration level of formaldehyde results in serious damage to human organs which is the cause of death. Although formaldehyde is highly toxic and harmful to health, it is still used extensively as a raw material in the textile, wood, chemical, and pharmaceutical industries [55]. Therefore, the organizational body in many countries has set the limit of the gaseous formaldehyde level, specifically in the indoor environment to be 0.08 ppm according to the World Health Organization (WHO) to control the concentration of formaldehyde in the air [56], [57].

2.6. Ethanol (C₂H₅OH)

Although ethanol is considered as one of the least hazardous volatile compounds, its vapor can lead to several health problems including headache, irritation of eyes, drowsiness, and difficulty in breathing [58]. Recently, the use of the blended mixture of ethanol-gasoline in vehicles has been reported as a new trend to save money. However, this can lead to the excess release of VOCs in the environment [59].

2.7. Methanol (CH₃OH)

Methanol is considered as one of the highly toxic VOCs, as it may cause serious health issues to human body, such as airway narrowing, vertigo, headache, bronchial constriction, nausea, and even visual impairment [60]. Even it has high toxicity, it is used widely in the synthesis of automotive fuel, dyes, drugs, formaldehyde, perfumes, etc [61]. It is also widely used in the manufacturing of various alcohols which are distributed worldwide. However, due to structural similarity, the co-existence of different aliphatic alcohols makes the detection of methanol challenging.

2.8. Toluene (C₇H₈)

Toluene with molecular formula C₇H₈, is an important industrial VOC which is intensively employed for the synthesis of other chemical intermediates e.g. cosmetics, adhesives, solvents, dyes, pharmaceuticals, medicines, explosive, and agricultural chemicals [62]. The toxicity level of toluene is similar to benzene. The human organs such as liver, skin, kidneys, and the central nervous system become damaged after exposure to the toluene at high concentrations [49]. Therefore, the WHO has set the guideline for an indoor air concentration of 0.07 ppm for toluene at 25 °C to control and monitor air quality.

2.9. n-Butanol (C₄H₉OH)

N-butanol also known as Butan-1-ol is a primary alcohol with the molecular formula C₄H₉OH is widely used as an extraction agent, solvent, preparing plasticizer, esters, solvent, and various industrial products [63], [64]. Exposure to n-butanol vapor may lead to dizziness and drowsiness resulting in the damage of the respiratory system, eyes, and skin due to its narcotic and stimulating nature [65]. Photophobia, lacrimation, and blurred vision can also occur from prolonged human exposure to higher concentrations.

2.10. 2-propanol (C₃H₈O)

2-propanol is widely used in the production of isopropyl halides and acetone and is also employed in the various applications of industrial products such as paints, insecticides, and polishes, rubbing alcohol, etc. [66]. At high concentration around 400-5000 ppm, it causes deterioration of the ciliary activity in guinea pigs. Inhalation of it causes dizziness, drowsiness,

nausea, and irritation to the nose and eyes, while inhalation for a longer time may lead to coma [49].

3. Applications of VOC sensors

The application of VOC monitoring has gained interest in various areas such as indoor air quality, environmental monitoring, fire detection, industrial safety, and health care applications due to the environmental regulations and health concerns. In all of these applications, the problems are the wide range and low concentrations of target molecules along with the complex matrix comprising a large number of organic and inorganic interferences. Although, benzene, tetrachloroethylene, naphthalene, and formaldehyde are the most common chemicals with high significance due to their ubiquitous presence and several adverse effects on human health. Other VOCs can be more of interest based on the particular environment situations and target health applications. Long-time exposure to VOCs is potentially harmful to human health which involves damage to the organs, skin, and the respiratory system[67] leading to sick building syndrome [68], [69]. In addition to these unspecific adverse health consequences, some VOCs are considered to be carcinogenic such as benzene or are believed to be carcinogenic such as formaldehyde. The limited target quantities are the main challenge of VOC measurements. The guidelines have been issued from WHO and European Union to control the VOCs for some particular substances like indoor air, are 0.01 mg/m³ for naphthalene and 0.1 mg/m³ for formaldehyde, and 5 µg/m³ for benzene as guidelines issued from [70], [71]. A high selective measurement is the main requirement to differentiate the highly carcinogenic and highly toxic organic gasses from the inorganic gasses e.g., hydrogen (H₂), ozone (O₃), nitrous oxide (NO_x), and carbon monoxide (CO) which are generated naturally or by other sources. Various applications of VOCs measurement mentioned above can be illustrated as shown in Fig. 2.

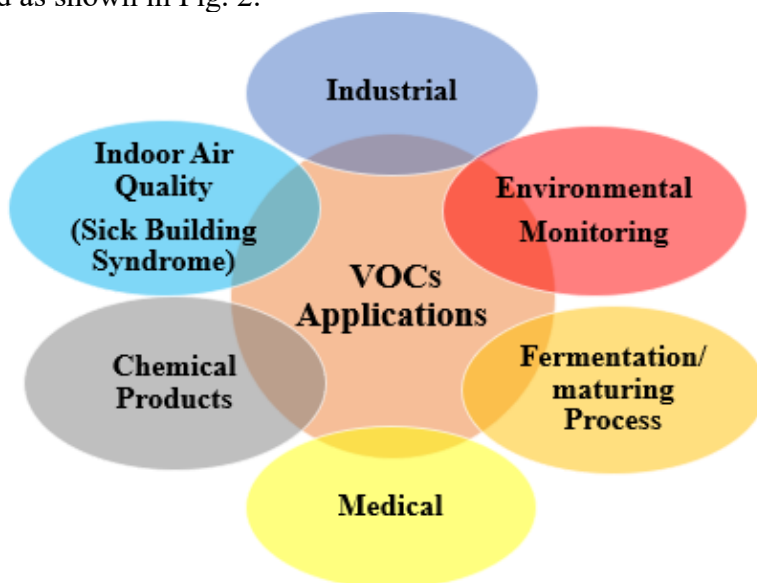


Fig.2. Some potential applications of VOC sensors.

The chemical associated with VOCs presenting in human's exhaled breath can be utilized as a biomarker for the detection of disease and point of care diagnosis [72]. Apart from the detailed study of toxic exposure, the VOCs exhaled in human breath can provide insights information of health conditions and diseased metabolic states, and can perform as a potential biomarker for recognizing various diseases noninvasively. A significant emphasis has been provided to continuous and selective detection of numerous pathophysiological and physiological conditions of living organisms through e-nose technique and VOC sensors [73]. The non-invasive monitoring of VOCs has been achieved from the biomatrices of exhaled breath, saliva, and sweat. The VOCs from saliva and sweat can be detected continuously in

real-time. The sensing methods for VOC detection include chemiresistive, optical, and electrochemical techniques. In this article, a detailed study has been provided on optical detection techniques to monitor various VOCs.

4. Measurement method

With the introduction of optical fibers, a significant level of research has been focused on the development of optical fiber-based VOC sensors. Even though they are considered secondary devices they have beneficial features compared to the gold standard techniques such as gas chromatography-mass spectroscopy (GCMS) as they are low cost, small in size, multiplexing, immune to electromagnetic interference, and able to operate remotely. A key concept of optical fiber sensors is to be able to detect the signal change at the output when there is a change in the environment by using light interaction at the interface of surrounding and optical fiber. The measurement setup includes a light source, detector, and optical fiber along with the sensing head as shown in Fig 3. The emitted light from the source is first coupled into the fiber and then propagated along the fiber by total internal reflections (TIR) which leads to the change in the property of light in the presence of surroundings. In the present era, the VOC sensors and their sensitivity enhancement are mostly performed based on the basic phenomenon of evanescent wave excitation and its interaction with the surrounding environment through sensing material deposited over fiber [74]. The investigation results obtained by these sensors are not affected by the bulky enticement because the penetration depth of the evanescent field ranges from ten to several hundred nanometres. In general, for VOC sensors the optical fiber is coated with some additional sensing and highly selective material, the sensor goes through the intimate coupling of fibers and sensitive material, which is the basic principle of the evanescent wave sensing technique. Therefore, when the propagated light interacts with the deposited material, its property gets changed in terms of intensity, wavelength, phase, and polarization in presence of various concentrations of VOCs. The technique is considered to be an attractive way for sensitivity and selectivity enhancement and lower the limit of detection of fiber-optic sensors. The output signal with the sensing information is then carried out to the photodetector for the signal demodulation. The measured VOC concentration can be obtained from signal processing after detecting the variation in the output signal with different surrounding concentrations. Hence, the sensing area of the sensor is the most important element in terms of sensitivity and selectivity that determines the efficiency of the VOC sensors. Generally, the optical fiber is made of silica cladding and germanium-doped silica core. The core of the fiber has a slightly high refractive index compared to the cladding to allow the light to be strongly confined inside the fiber core and propagated along with the fiber with minimum transmission loss according to the TIR phenomenon. In order to monitor the VOCs concentration, one of the most effective methods is to deposit the VOC sensitive materials on top of the fiber, in which its property such as refractive index, conductivity, and volume can change with the presence of VOCs at different concentrations. The change in properties of the sensing material affects the light propagated inside the fiber causing the variation in terms of intensity, phase, or wavelength which can be detected as the output signal.

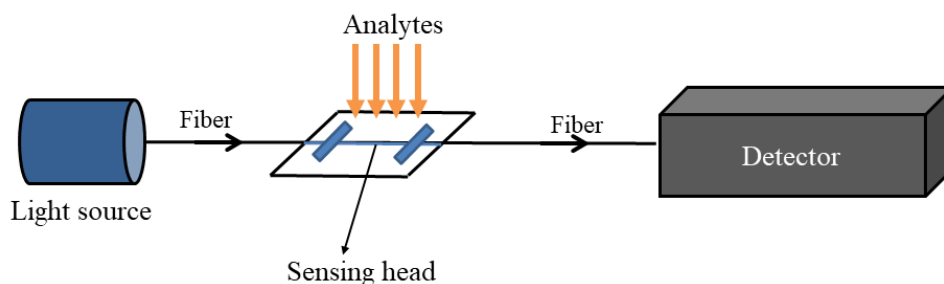


Fig. 3. The schematic diagram for the optical fiber sensor.

The optical fiber-based sensors for detecting VOCs can be categorized into four levels based on sensing techniques involving i). interferometry based, ii). Bragg grating based, iii). microstructured based, iv). absorption based. In each category of the fiber-based sensors, the working principle, sensing structure, sensing characteristics, advantages, and limitations of each technique are reviewed intensively and provided in the following sub-sections.

4.1. Interferometric based VOC sensors

Generally, in the fiber optic sensors based on interferometry, the propagated light is divided into at least two arms of the interferometer and at least one part of the light interacts with the surrounding environment [75], [76]. Interferometry is a well know investigation method extensively used over the various sensing applications for precise and accurate monitoring [77]. In the past decades, this technique has been applied to many applications in nano-engineering and technology for its unique features such as high sensitivity due to being *in-situ*, contactless, and non-destructive. Recent advances in computation, data interpretation, and data acquisition, have made it possible to use this technique to perform exceptionally reliable and sensitive measurements. The monitoring principle of interferometric fiber sensors is the phenomenon of splitting and recombining two or more propagated coherent lights. With the particular structured design, the emitted light from the source split into two or more beams at the tapered/spliced region and then recombined at another tapered/spliced region. As the propagated light travels down along various transmission paths, constructive or destructive interference takes place once they meet and recouple with each other. The target substances can be monitored using various modulations of detection such as phase, intensity, wavelength, polarization, etc. For the VOC sensing applications, various interferometer configuration has been utilized to achieve high selectivity and good sensitivity such as Fabry-Perot interferometry (FPI), Mach-Zehnder interferometry (MZI), Michelson interferometry (MI), and Sagnac interferometry (SI). The difference in their arrangements is shown in Fig. 4.

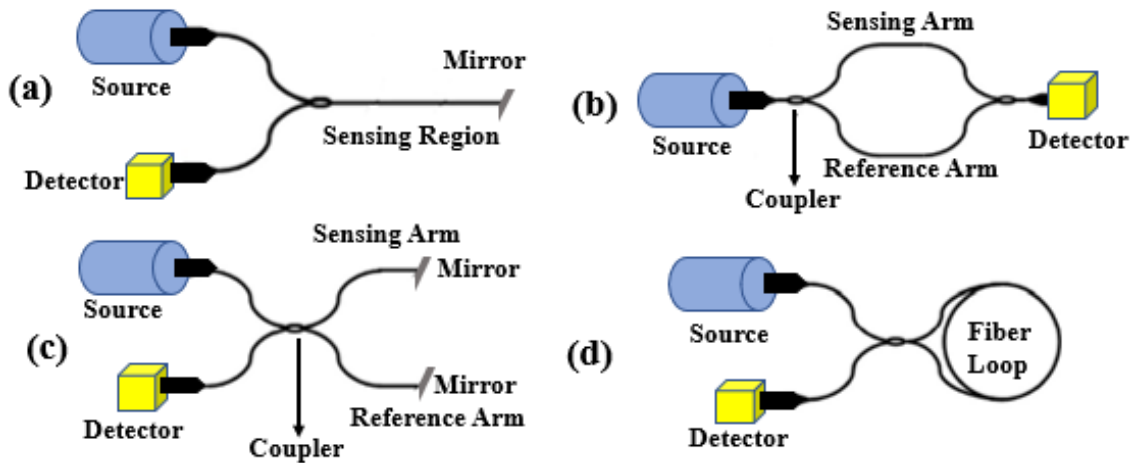


Fig. 4. Optical fiber-based various interferometers, (a) FPI (b) MZI (c) MI, and (d) SI.

Figure 4 (a). illustrates the FPI configuration, it has been considered as the simplest configuration for sensing. In FPI, the propagated light reflected from the mirror-dielectric interface and travelled along the same path through which it was launched. In FPI, the reflectors, forming a cavity, are generally separated by an air gap, and a cleaved fiber is used to launch the light. While, in the case of MZI based fiber sensors, it is followed by inherent complexity due to their configuration as shown in Fig. 4 (b), but still being utilized in the majority of sensors. In an MZI based sensor, the launched laser beam is injected into the optical fiber which splits equally into two arms, one is known as the sensing arm and the other is the reference arm as shown in Fig. 4 (b). After leaving the MZI, the two beams from both arms recombine to form an interference pattern at the exit point. It has been observed that the adjustments in the

length of the sensing arm and the surrounding material, lead to the change in the output interference pattern. Figure 4 (c), illustrates the arrangement of MI, as it appears the MI arrangement is quite similar to the MZI, since it uses the same reflection modes from the mirrors, instead of analogous or transmission fabrication methods and operating principles. Figure 4 (d), is the configuration of SI, which is considered to be the simplest arrangement like FPI, however, the SI, consists of a fiber loop along which the two beams propagate in opposite directions with different polarization states. SI is one of the configurations used widely due to its insensitivity to the environment [75]. Optical fiber-based interferometers have gained huge success in sensing applications e.g. the SI techniques can be used as a rotation measurement for both military and civilian applications [56, 57]. The latest trend in the optical fiber interferometer is to improve and modify it for micro-scale sensing applications. Therefore, convectional bulky components for convectional interferometry such as combiners, beam splitters, and objective lenses have been replaced rapidly by small-sized optical fiber devices which enable the sensors to perform on fiber scales. The fiber interferometry offers a micro-scale detection which is very suitable for VOC sensing applications, which supports high coupling, easy alignment, and high stability and is found to be ideal for real monitoring applications.

Theoretical modal of measurement: The sensing response of optical waveguide sensors is being measured either by wavelength interrogation technique or intensity interrogation technique. The basic mechanism of both interrogation techniques is discussed below:

(i). Wavelength interrogation technique: To analyze the transmission spectra, an external light source, or the combination of a spectrometer and a broadband source, is required. The cost of the wavelength measurement system is therefore significant. The sensing configuration monitors the change in the output spectra corresponding to the change in the surrounding environment [80]. The shift in wavelength spectra is required to measure the sensitivity of the sensor.

(ii). Intensity interrogation technique: Another sensing scheme used for the measurement is popularly known as the intensity interrogation technique, in conjunction with a cos-effective light source and a photo detector. The wavelength interrogation technique needs wavelength information. The change in the refractive index of surrounding media can be examined from relative intensity measurement without any spectral measurement, and it has a broad tolerance for the power stability of the light source [80].

Several interferometric sensors have been developed utilizing optical fibers [81]–[83]. The technology benefited from the salient features of optical fiber to provide a miniature VOC sensor. Taking the field forward, Carlos *et al.* reported an idea of monitoring the VOC gases based on interferometry techniques in 2010 [84]. The proposed sensor is based on the intensity interrogation technique. In this work, the sensor utilizes polydimethylsiloxane (PDMS) sensing film over a glass substrate. PDMS is associated with unique features to change its refractive index or swell when it interacts with various VOCs leading to the fringe shift in a Pohl interferometric configuration. The experimental setup used for the characterization is shown in Fig. 5 in which the light reflected (I) from three interfaces i.e., air–substrate (I_1), substrate–PDMS (I_2), and PDMS–air (I_3), and the interference pattern on the screen is defined as:

$$I = I_1 + I_3 + 2\sqrt{I_1 I_3} \cos(\delta) \quad (1)$$

With:

$$\delta = 2y k \sin\theta - \Delta\theta \quad (2)$$

where y represents the position of fringe on the screen and k wavenumber of propagated light, θ is the angle of interference between I_1 and I_3 , and δ is the phase difference. The pattern of interference fringes evaluated from Eq. (1) in the function of the position on the screen y (cm) is illustrated in parallel Fig. 5 (b). The fringe shift calculated for a PDMS layer coated over the

glass substrate is shown by a continuous line, while the shift in fringe pattern arises due to the swelling, and change in the refractive index of PDMS is shown by dotted and dashed lines.

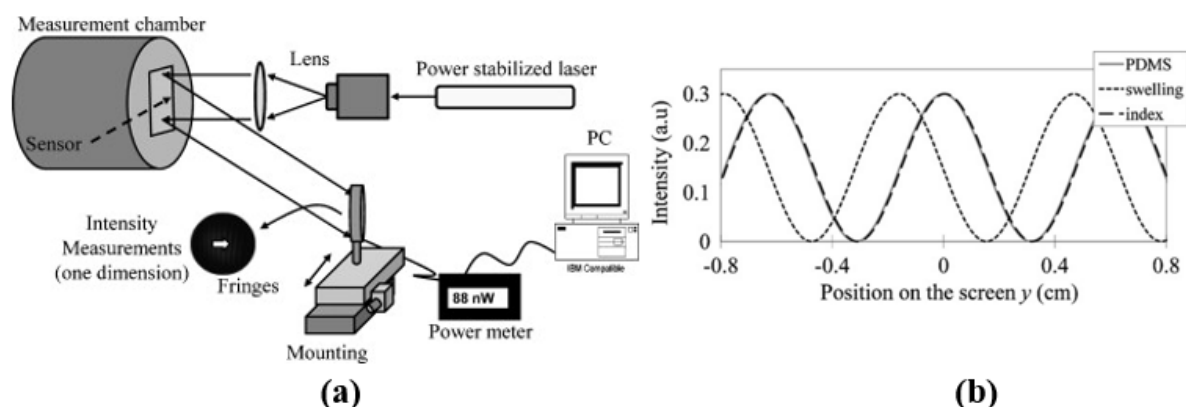


Fig. 5. (a). Pohl interferometric system for VOC detection (b). fringe shift due to swelling of PDMS [Reprinted/Adapted] with permission.

After PDMS was proposed as a potential sensing material for VOC sensors, various sensing devices have been developed and modified to achieve maximum sensitivity for volatile compounds using PDMS. In 2013 Raphael *et al.* developed an on-chip VOCs sensor based on the FPI utilizing PDMS-polydiphenylsiloxane [85]. The deformable Fabry–Perot based sensing configuration as shown in Fig.6 was coated with PDMS-polydiphenylsiloxane copolymer and PDMS and tested with cyclohexane and m-xylene as VOCs.

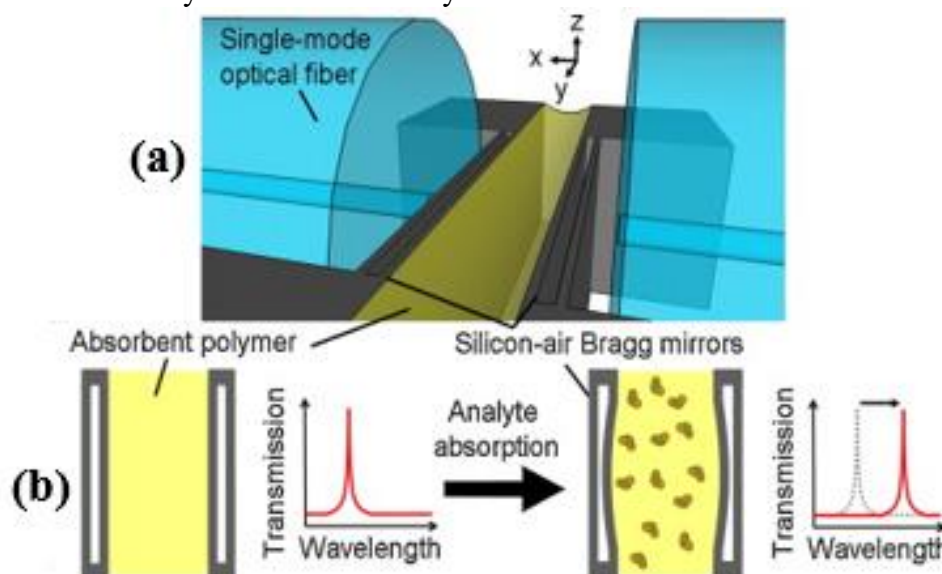


Fig. 6. (a) Schematic representation of the deformable interferometric sensor (b) Schematic representation of the sensor operation principle. [Reprinted/Adapted] with permission.

From Fig.6, it is clearly seen that the Fabry-Perot configuration of the sensor was constructed by using silicon-air Bragg reflectors along with a sensing polymer as the dielectric region between the two vertical reflecting surfaces from the single-mode fiber (SMF). The fabricated device was coated with PDMS and PDMS- polydiphenylsiloxane for comparative study. The sensor is based on wavelength interrogation technique. The device deposited with PDMS- polydiphenylsiloxane was found to be 93% more sensitive to cyclohexane and 64% sensitive to m-xylene in comparison to PDMS coated sensor. The main drawback of this device is its selectivity of PDMS-polydiphenylsiloxane copolymer over mixed VOCs. The integration of a microfluidic system with fiber simplifies the parallel functionalization of several

interferometers, utilizing various polymers, to a specific class of gases. Additionally, the fiber grooves allow the *in-situ* monitoring of the interferometers along with passively aligned traditional SMF, besides free space light alignment. However, the sensor has been examined for two VOCs only which may limit its application for the places containing mixed volatile environment. There are many other materials besides PDMS that have been utilized as sensitive material integrated with fiber for VOC sensing to attain a good sensing response. In 2016, Xiangping *et al.* demonstrated the ability of zeolite as a VOC sensing material based on wavelength interrogation technique [86]. Zeolites are micro porous aluminosilicate crystals covered with uniform nanometer-scale pore systems. The nonporous zeolites film has the ability to selectively adsorb VOC molecules based on the shape selectivity or size exclusion which primarily depends on the framework Si/Al ratio, crystal structure, and type of cations. In this work, a thin sensing layer was deposited on spherical end facet optical fiber for the detection of trace isopropanol vapors. The intuitive view of the spherical sensing probe is shown in Fig. 7 (a) while Fig. 7 (b) illustrates the experimental setup used in the characterization of the device. Light launched from the light source was reflected by the Fabry-Perot cavity which includes the spherical fiber facet coated with thin zeolite film. The phase shift (ϕ) of round-tip propagation in the device can be defined as

$$\phi = \frac{4\pi nL}{\lambda} \quad (3)$$

Where n and L represent the refractive index and thickness of sensing film, while λ is the wavelength. Although the reported sensor shows good sensitivity and strong selectivity toward isopropanol, it was not tested over other mixed VOCs which limits its application in the industrial area. Zeolite SBA-15 and MCM-41, has nanopores, that can possibly adsorb any gaseous molecules of the corresponding size, hence the material needs to be optimized for VOC sensing.

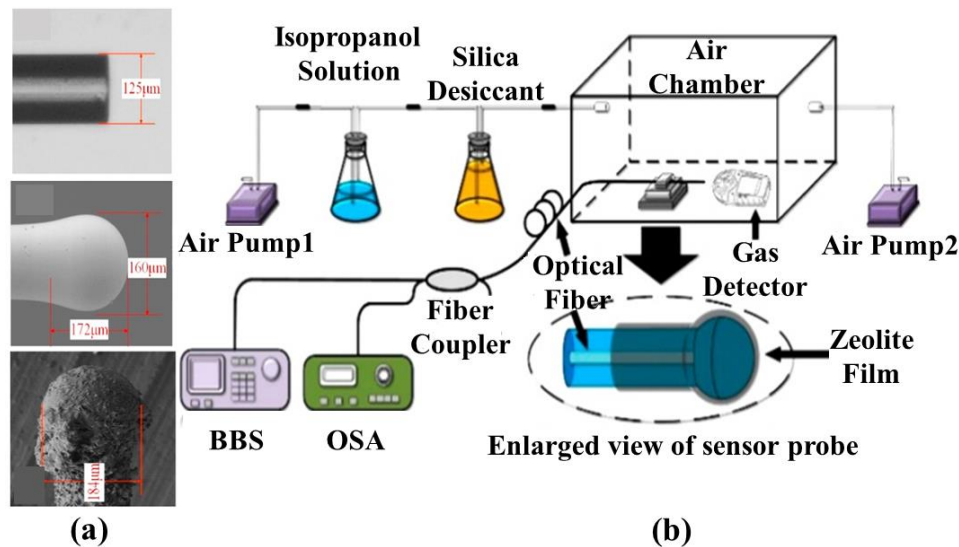


Fig. 7. (a) SEM images of zeolite coated spherical end-facet fiber sensors (b) Schematic diagram of the experimental setup. [Reprinted/Adapted] with permission.

To enhance the sensitivity and selectivity of VOC sensors, Cai-bin *et al.* proposed another compact interferometric device based on wavelength interrogation technique, in 2017 [87]. This device comprises SMF covered with polymethyl methacrylate (PMMA) as sensing material. Generally, PMMA reacts with most of the organic compounds, as they have a similar atomic bonding structure of $-\text{CH}_2\text{C}-$. Hence, the interaction of VOC molecules and PMMA leads to the penetration of VOC molecules through the large molecular gap of the PMMA layer, resulting in the swelling effect. The author has utilized the property of PMMA to develop a VOC sensor based on the swelling effect and refractive index change of the sensing material.

The swelling of the material affects the cavity length of the Fabry-Perot structure leading to a wavelength shift of the resonance dip. The reported sensor achieves a high selectivity and exhibited good sensitivity of 2.7 pm/ppm and 2.17 pm/ppm for ethanol and acetone, respectively. The schematic of the sensing configuration is shown in Fig. 8. The reported sensor can be considered as a potential candidate in VOC monitoring due to its special feature to distinguish between VOCs and other toxic gasses as it had low sensitivity towards other gasses such as CO, NH₃, and H₂. Although the sensors exhibit good selectivity, however, it lacks giving accurate information about the response and recovery time of the sensor. Additionally, the optimization of thickness of PMMA (300 nm) is highly required to avoid poor response and recovery time of the sensor.

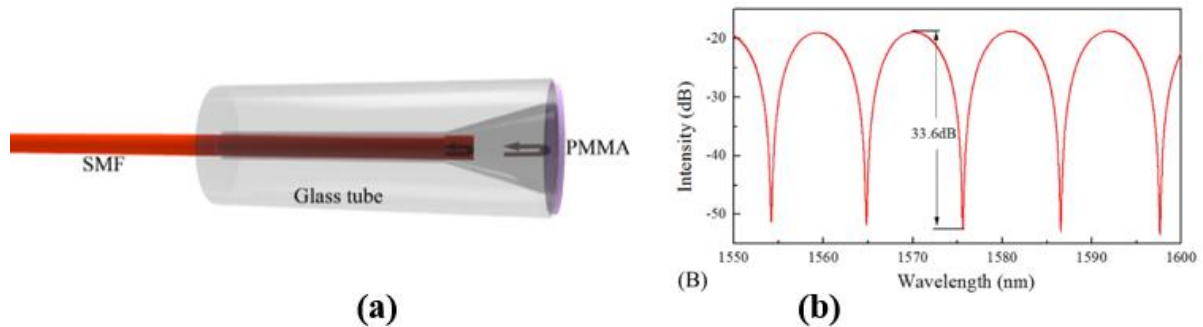


Fig. 8 (a) The schematic diagram of the FPI sensor. (b) The reflective interference spectrum of the FFPI sensor. [Reprinted/Adapted] with permission.

The sensitivity and selectivity of the VOC sensors play the most important role in its commercialization. Many researchers keep modifying the sensing structure in order to make it applicable on a large scale. Recently, Rajesh *et al.* developed a novel axicon tip VOC sensor filled with PDMS, as shown in Fig. 9 (a) [88]. The sensor is based on wavelength interrogation technique. The group has demonstrated the fabrication and the potential of the sensor over various VOC gases. The chemical etching process was used to fabricate a refractive-reflective negative axicon cavity inside a cladding mode offset photosensitive SMF as shown in Fig. 9 (b), the basic principle of the device includes the two steps reflection i.e., (i). the interface of fiber and PDMS (R_1) and (ii). The interface of PDMS and air (R_2), as can be seen in Fig. 9 (b). The main advantage of this device is its tiny size and robustness that allows only tip to interact with the surrounding for the VOC monitoring.

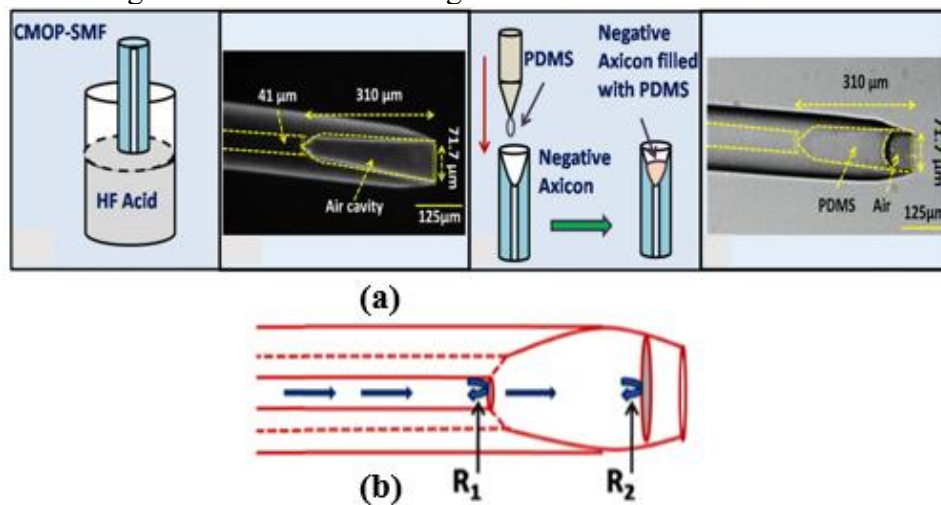


Fig. 9. (a) Schematic of the fabrication step for negative axicon tip (b) Light propagation and interaction with PDMS material through. [Reprinted/Adapted] with permission.

The authors have considered PDMS for cavity coating due to its swelling properties in presence of VOCs gaseous molecules and capability to perform effectively at room temperature. This cavity interacts physically with the VOC molecules, changing the length of the cavity and the RI of PDMS due to the oxidation/reduction interaction. The cavity filled with the PDMS as a sensing layer is formed between two reflecting surfaces R_1 and R_2 . The launched light propagated along with the core gets reflected from R_1 while the rest of the light will be propagated through PDMS and reflected from R_2 . The interference between R_1 and R_2 occurs because of the change in path difference between these two beams that occurs due to the cavity length. The sensitivity of the sensor was analyzed with wavelength shift in comparison to extinction ratio (ER). The maximum sensitivity of 3.2 pm/ppm, 8.8 pm/ppm, 17.8 pm/ppm, 9.1 pm/ppm, 21.6 pm/ppm, and 23.7 pm/ppm are obtained for various concentrations ranging between 5-200 ppm of methanol, isopropanol, hexane, acetone, toluene, and chloroform gases, respectively. The sensor was tested over 6 VOCs and reported a good sensitivity and selectivity for all. The major advantage of the device is its possible realization. The group has demonstrated a light guiding phenomenon via numerical investigation to validate the sensing principle of the device. Although, the fabrication of negative axicon tip is a bit tricky, which depends on the concentration of HF and the time of etching. The author didn't study the effect of varying the etching process to attain the maximum coupling of light. The proposed configuration required the optimization of etching time, chemical concentrations, and filling techniques to achieve the desired result.

An emerging optical fiber-based sensing structure in past decades has attracted many researchers towards the D-shaped fiber sensors. Based on the interferometry configuration, the D-shaped fiber is mostly used to develop refractive index sensors [89], humidity sensors [90], and temperature sensors [91]. However, in 2020, Huadan *et al.* demonstrated a D-shaped optical fiber sensor for refractive index sensing with various residual thickness of cladding and also illustrated its application in VOC sensing based on wavelength modulation technique [92]. The sensor was made of standard SMF fabricated by using ultra-precise polishing technology in order to enhance the sensitivity. The polishing of the fiber leads to maximum leakage i.e., the evanescent field of propagated light from the core and interacts with surroundings. A maximum refractive index sensitivity of 10243 nm/RIU for the analytes with the refractive index varying from 1.430-1.444 was achieved. The sensing configuration of this proposed D-shaped fiber sensor is shown in Fig 10 (a). The propagation of light in D-shaped fiber is shown in Fig. 10 (b). The efficiency of the multimodal interference (MI) in the D-shaped SMF is related to the residual thickness of the cladding. The D-shaped SMF with different residual thickness results in various angles of incidence. The evolution of the optical field along with the flat section was defined by the interference between the excited higher order modes.

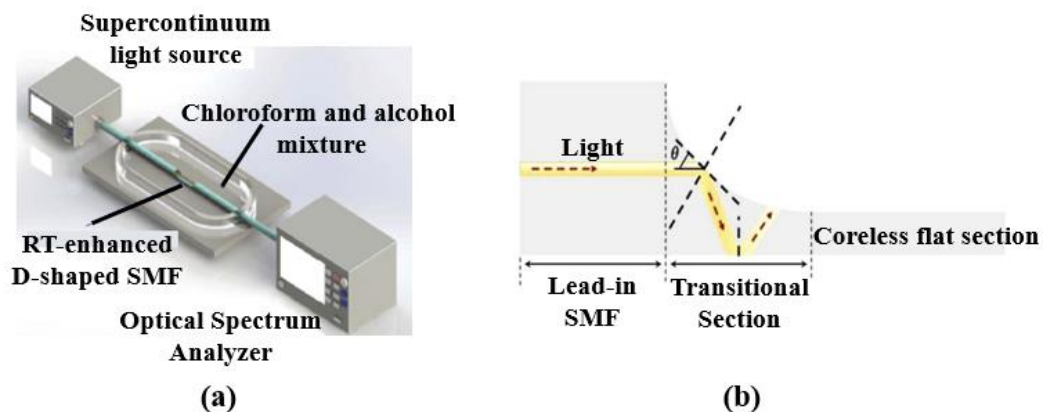


Fig. 10. Schematic diagram of (a) the experimental setup to evaluate the performance of the sensor (b) polished surface to excite the highest order mode in the cladding.
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This sensor was implemented for monitoring the evaporation of the alcohol and chloroform. The refractive index of the mixture between 99% chloroform and 1% alcohol was examined to be around 1.44. A clear variation in transmission during the evaporation of chloroform with a sharp redshift to > 1550 nm in 30 min and then moderate blue shift around 1450 nm in 2 hours was observed during the experiment. This phenomenon was explained by the change of the effective refractive index in the MI in the presence of alcohol and chloroform mixtures due to the evaporation. From the proposed design authors have demonstrated the potential of D shape fiber for VOC sensing, but the unavailability of any specific sensing material for VOC molecule trapping limits its use for real-world realization. Additionally, the lack of information related to the response time and selectivity might be another loophole for such devices. Various interferometric VOCs fiber sensors including the principle of the sensor, target VOCs, their sensitivity, detection limit, and their operating range are summarized and provided in Table 2.

Table 2: Comparative table showing various interferometric based optical fiber VOC sensors

Ref.	Principle	Target VOCs	Sensing Layer	Sensitivity	Limit of Detection (LOD)	Operating range
[93]	FPI	Methanol, Hexanol, and acetone	Norland adhesive (NA) and Polyethylene Glycol (PEG)	3.53 pm/ppm for methanol using PEG and 0.1 pm/ppm using NA	1ppm with PEG 10 ppm with NA	-
[94]	FPI	Toluene, methanol, decane, And Dimethylmethylphosphate (DMMP)	PDMS and PEG	4.75 mV/ng for decane and 77 mV/ng for DMMP	50 pg (105.6 ppb)	-
[95]	FPI/SI	Ethanol and 2 propanol	PDMS	9.02×10^{-4} to 5.14×10^{-4} nm/ppm for ethanol and 2.71×10^{-3} to 7.78×10^{-4} nm/ppm for 2-propanol	-	0-6800 ppm
[86]	FPI	Isopropanol	Zeolite	0.91 nm/ppm in range of 0 to 70 ppm	-	0-90 ppm
[96]	FPI	Formaldehyde (FM) and isopropanol (IPA)	Zeolite	For formaldehyde: 281.9 pm/ppm and for isopropanol: 4.99 pm/ppm	-	IPA: 0-70 ppm FM: 0-1000 ppm
[87]	FPI	Acetone and ethanol	PMMA	For acetone: 2.17 pm/ppm, For ethanol: 2.7 pm/ppm	-	0-1800 ppm
[97]	SI	Isopropanol	PDMS	1.03 pm/ppm in range of 0-6000 ppm	19.3 ppm	0-6000 ppm
[88]	FPI	Chloroform, Isopropanol, Hexane, Acetone, Methanol and Toluene.	PDMS	23.7, 8.8, 17.8, 9.1, 3.2, 21.6 pm/ppm	0.84, 2.26, 1.11, 2.19, 6.10, 0.92 ppm	0-200 ppm
[92]	multi-mode interference	Chloroform and ethanol	Based on residual thickness	RI of the mixture of 99% chloroform and 1% ethanol is measured to be ~ 1.44 . the evaporation has been evaluated with the shift in wavelength.	-	1.430-1.444 RI
[98]	MI	Benzene and ethanol	No sensing film	0.006 pixels/ppm for ethanol 0.015 pixels/ppm for benzene.	-	Ethanol: 1611-32210 ppm Benzene: 964-19290 ppm

[99]	Spectral interferometry	Pentane, hexane, octane, heptane, chloroform, trichloroethane, Tetrachloroethene, Dichloromethane	Polysiloxane, dimethylpolysiloxane (DMPS)	1000 ppm for heptane. 104 ppm for perfluoroethylene 2600 ppm for TCE	100 ppm	Few ppm-1000 ppm
[84]	Pohl interferometer	Methanol (Me), ethanol (Et), propanol (Pp), octane (Oct), heptane (Hep), methyl acetate (MA), Propyl acetate (PA)	PDMS	0.14 nW/ppm for PA 0.12 nW/ppm for Oct 0.04 nW/ppm for Hep 0-0.02 nW/ppm for rest	1500 ppm	0-24,000 ppm
[100]	FP	Isopropanol	PDMS	20pm/ppm	-	0-500 ppm
[101]	FP	Acetone, ethanol, isopropanol, methanol, toluene	Graphene/PMMA	3.57±0.001 pm/(μ L/L), 2.81±0.0004 pm/(μ L/L), 3.62±0.0008 pm/(μ L/L), 6.14±0.0004 pm/(μ L/L), 1.86 ± 0.0007 pm/(μ L/L) 1.76±0.00008pm/(μ L/L) For methanol, acetone, chloroform, ethanol, toluene and isopropanol, respectively	5.6 μ L/L	5-200 μ L/L
[102]	MZI	Acetone, ethanol, methanol	Liquid crystals	-83 pm/ppm-methanol -31 pm/ppm-acetone -101.4 pm/ppm-ethanol	-	0-2000 ppm
[81]	FPI	Xylene Toluene	Carbon nanotubes	8.3×10^{-4} ppm ⁻¹ xylene 3.5×10^{-4} ppm ⁻¹ toluene	-	3-39 ppm 54-93 ppm
[82]	FPI	Toluene	Carbon nanotube	-	-	0-160 ppm
[83]	FPI	Methanol Ethanol Isopropanol	Au ₂ Ag ₂ (C ₆ F ₅) ₄ (C ₆ H ₅ N) ₂	Sensitive of methanol	-	21.5 mmol/l 43 mmol/l 86 mmol/l
[103]	FPI	Acetone	Polyvinyl alcohol	43.9 pm/(μ L/L)	0.45 μ L/L	0-80 μ L/L
[104]	MZI	Ethanol	ZIF-8	41 pm/ppm-1	740 ppb	600-1000 ppm
[105]	MZI	Ethylene	Copper (I)	60 pm/ppm	~4.7 ppm	0-140 ppm
[106]	FPI	Toluene acetone	SU-8 and PDMS	2900 V/ng for toluene using PDMS. 4 u V/ng for acetone using SU-8	202 ppm for toluene 2336 ppm for acetone	0-800 ng

4.2. Fiber grating-based VOC sensors

Among the broad area of optical fiber sensors, the Fiber Bragg Grating (FBG) based sensors are emerging as the most promising technology to monitor small variations in surrounding media. The FBGs are manufactured by introducing the periodically varied refractive index of the core along the length of optical fiber, which resonates at a wavelength, popularly known as Bragg wavelength. In short, the FBG works as an optical filter allowing only a certain wavelength to propagate along the waveguide whereas the rest are reflected [107]–[109]. Ever since the development of fiber Bragg grating (FBG) from the late 1970s, it has been extensively used for the development of various optical fiber-based sensors. Based on their grating period, they can be categorized into two major classes: short-period or also known as Fibre Bragg Gratings (FBGs), and Long Period Gratings (LPG). More detailed information regarding the fabrication and principle of these fibers can be found in previously reported articles [110], [111]. This section comprises various FBG and LPG based VOC sensors reported in past years.

Most of the grating sensors which has been developed and available in the market are configured for temperature and strain monitoring. However, in 2010 Toplissa *et al.* reported a novel design of VOC sensors using LPG based on wavelength interrogation technique [112].

They have used the coating of calixarene over the LPG fiber which exhibits good sensitivity to the aromatic compounds such as benzene and toluene while being comparatively insensitive to the other aliphatic hydrocarbon like hexane. The nanoscale cavities available in the calixarene molecule entrap the exposed VOC molecule leading to a change in the refractive index of the calixarene which affects the transmission spectrum of LPG. Using this configuration, a maximum sensitivity of 231ppmv was achieved for toluene however a fast-reversible response of 15s was attained. The sensing response of the device in presence of various VOCs is shown in Fig. 11. The result indicates that the fabricated device attains a high selectivity for toluene. The sensing response of the sensor coated with a calixarene in presence of toluene vapor is shown in Fig. 11 (a). The sensor was investigated at various concentrations varying from 0 to 61.2 kppmv. The sensing response was analyzed by measuring the spectral shift of two resonant bands. A similar experiment has been performed for other VOCs too, the shift in spectra is shown in Fig. 11 (b). Later the selectivity experiment has been performed and the result exhibits a high selectivity for toluene as shown in Fig. 11 (c). It's clearly seen that at the level up to 10 kppmv concentration, the sensitivity of the device to hexane is only 7.5% which is low in comparison to toluene. While for cyclohexane, the observed sensitivity is 14.8% lower than the toluene. The device reported high selectivity among several VOC.

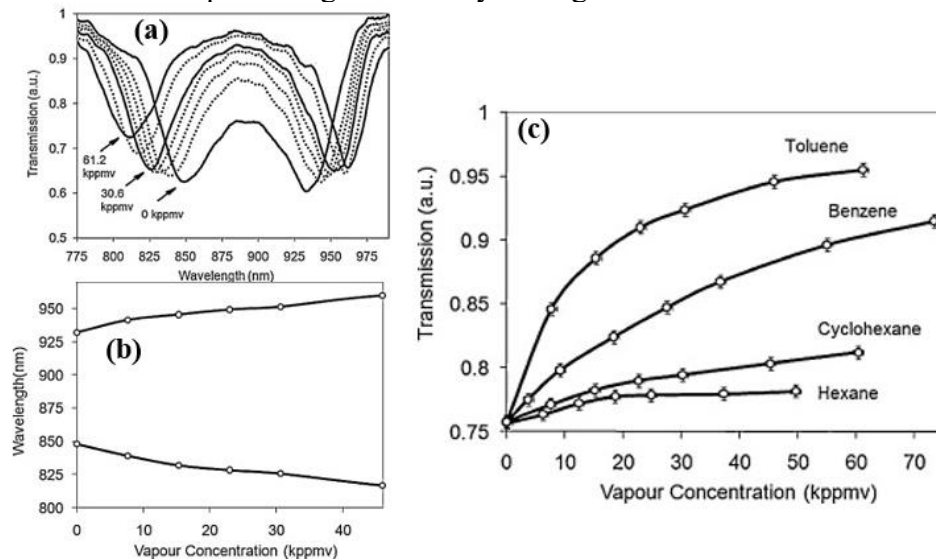


Fig. 11. Volumetric sensitivity of the sensor to the vapor's toluene, benzene, hexane and cyclohexane. [Reprinted/Adapted] with permission.

In the same year, Park *et al.* reported a novel selective chemical sensing approach by deposition of PDMS on FBG structure for the monitoring of various VOCs [113]. The sensor was based on wavelength interrogation technique utilizing the swelling effect of the PDMS, which is coated on FBG, in the presence of VOCs. The swelling effect provides a tensile force to the FBG. The amount of this tensile force implemented on the fiber highly depends on the swelling ratio of the sensing material. Therefore, the type and the concentration of exposed VOCs can be determined accurately and selectively by analyzing the shift of Bragg wavelength of the PDMS deposited FBG. The schematic diagram of the reported sensor is shown in Fig. 12 (a). The sensor was characterized over methanol, 1-propanol, 2-propanol, and acetone. Fig. 12 (b) illustrates a good wavelength shift corresponding to each VOC.

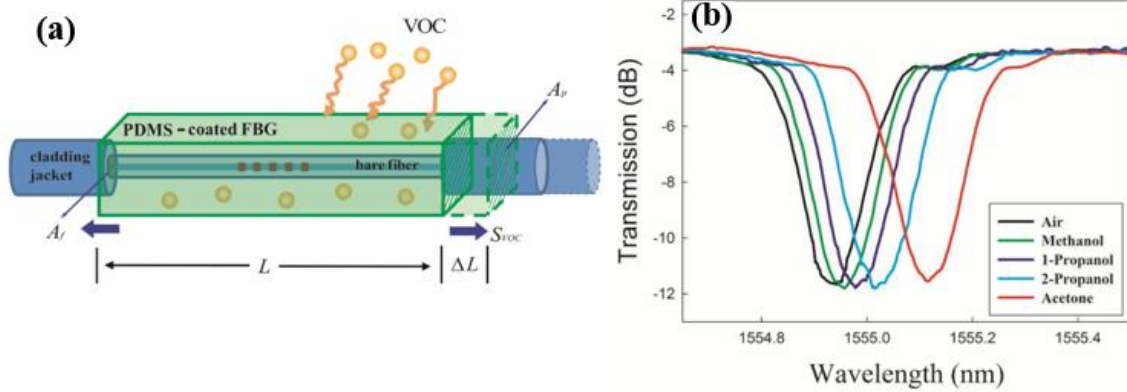


Fig. 12. (a). Schematic diagram of PDMS coated FBG sensor, (b) spectral response of sensor for each VOCs. [Reprinted/Adapted] with permission.

Later in 2012 Maria *et al.* reported a novel idea of utilizing zinc oxide (ZnO) nanorod as an outer cladding of LPG to enhance sensing response to ethanol vapor [114]. The nanorods have a high surface-to-volume ratio which offers a higher sensitivity compared to a ZnO film [115]. When the fabricated sensor was exposed to various concentrations of ethanol vapor, its optical properties such as refractive index and/or absorption loss were altered accordingly. These alterations in the optical properties can be investigated from the cladding mode of the LPG structure which is manifested as strength and/or peak wavelength changes in its transmission spectrum. The sensor was based on wavelength interrogation technique. In order to verify the growth of ZnO nanorods, ZnO-coated LPG was examined by scanning electron microscopy (SEM) as shown in Fig.13. The reported article reveals that the conditions of ZnO growth, which clearly define the Opto-geometrical feature of the random nanostructures overlayer, dominate the output spectral response of the LPG device. This unique dependency was confirmed by a detailed investigation of ethanol response on ZnO covered LPG device under various growth conditions and by comparing their spectral response with available simulation analysis. Figure 13 (b) illustrates the changes in the LPG notch strength ΔS , while (c) illustrates the variation in wavelength upon exposure of ethanol vapor with the exposure time. The solid line in Fig. 12 (b) corresponds to the relative ethanol concentration measured by commercially available PASCO probe ((PS-2194). Fig. 12 (c) exhibits the spectral effect of the ethanol vapor on the LPG attenuation band after 95min of exposure and corresponding recovery. The reported devices show the potential of ZnO to achieve high sensitivity for ethanol. Additionally, authors have observed the change in the grating strength of ~ 0.5 dB is easily measurable from the developed device, which makes it possible to monitor the small concentrations of ethanol. However, the limit of detection, selectivity, and response time has not been considered into account during the device development which is one of the major limitations for its applicability.

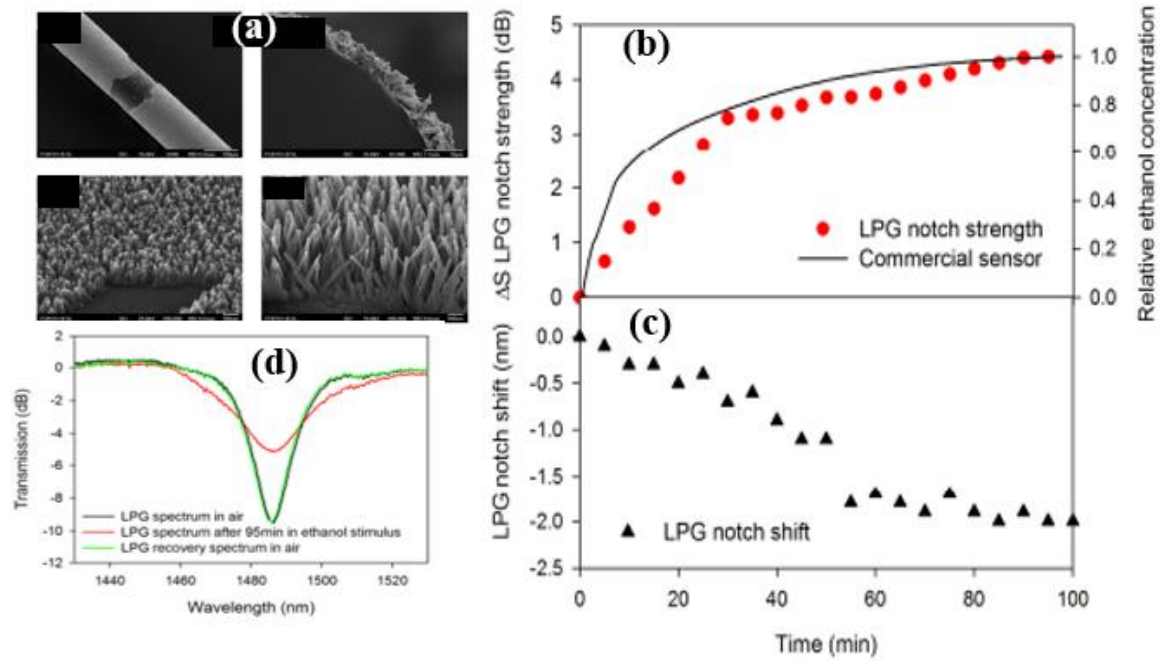


Fig. 13. (a) SEM images of growth of ZnO nanorods (b). changes in the LPG notch strength ΔS and (c). wavelength upon exposure to 50Torr ethanol vapor versus exposure time (d). spectral effect of the ethanol vapor on the LPG attenuation band after 95min of exposure and corresponding recovery behaviour to air. [Reprinted/Adapted] with permission.

In 2018, Jiri *et al.* proposed a highly sensitive VOC sensor using an LPG coated with ZIF-8, a sensing material obtained from the zeolite imidazolate framework family [116]. The proposed sensor was based on wavelength interrogation technique. The fabricated sensor was exposed to various VOCs such as ethanol, acetone, and methanol over its high concentration range up to ppm level. Two sensors with different grating periods of 109.0 and 109.5 μm were developed and referred as sensor A and sensor B, respectively. Figure 14 illustrates the transmission spectra of sensor A and sensor B with bare LPG and coated LPG along 5 layers of ZIF-8 is shown in Fig. 14. It can be seen from Fig. 14 (a) that the “U” shaped band of the LP_{019} cladding mode splits into two attenuation bands after the coating of the sensing film. While in Fig. 14 (b), no attenuation band can be observed corresponding to the LP_{019} mode before the coating of sensing film as it is developed only after the deposition of the sensing film. The maximum sensitivity has been achieved when the LPG operates precisely at the phase matching a turning point in which the bandwidth of the U-shaped attenuation band is measured. For ethanol, this optimization process results in the calculated limit of detection reducing from 163 ppm to 5.6 ppm. The optimized sensor exhibits a high sensitivity of 0.018 ± 0.0015 and 0.015 ± 0.001 nm/ppm with a limit of detection (LOD) of 5.56 and 6.67 ppm for ethanol and acetone, respectively.

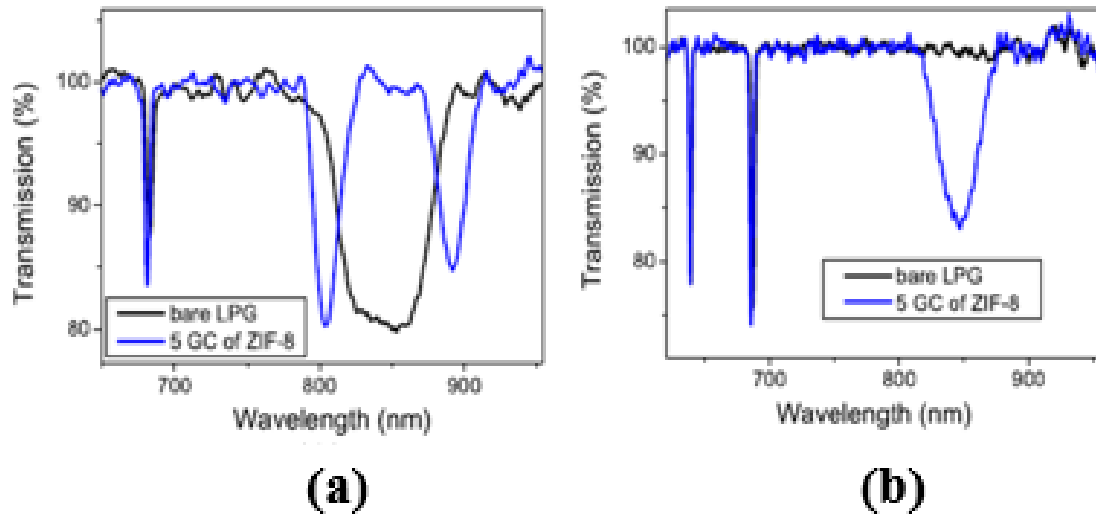


Fig.14. The transmission spectrum of an LPG with a period of (a) 109.0 μm –Sensor A and (b) 109.5 μm –Sensor B measured in the air. **[Reprinted/Adapted] with permission.**

In 2019, Raquel *et al.* proposed a new material for the detection of methanol vapor using etched FBG based on wavelength interrogation technique [117]. The fabricated sensor was coated with diphenylalanine nanotubes (DNT) to make the sensors capable of methanol in methanol-ethanol mixed vapors. The reported sensor analyzed two phases of the diphenylalanine nanotube: orthorhombic and hexagonal. With a presence of a methanol-ethanol mixed environment, the hexagonal phase results in a redshift, while the orthorhombic phase illustrates a blueshift. Moreover, it was reported that the response of the sensor was reversible. In comparison to the hexagonal phase, the orthorhombic phase provides a higher sensitivity of $(-7.3 \pm 0.8) \text{ pm}/(\% \text{v/v})$ with the same response time.

As the latest device, Konstantin *et al.* reported a VOC sensor for methanol detection using LPG coated with a new sensing material Silk Fibroin [118]. The sensor consists of a thin layer of sensing film of silk fibroin formed over LPG through the drop-casting technique. The experimental setup consists of a light source, gas chamber, and optical spectrum analyzer to investigate the sensing response of the sensing device. The sensor was based on wavelength interrogation technique. In the setup, the gas chamber was equipped with a sensitive manometer to record the variation in pressure due to the evaporation of the methanol which was injected in liquid form through the inlet. The sensor was based on the basic phenomenon of wavelength shift which was attributed due to the binding of injected methanol to the fibroin leading to the changes in its intermolecular H-bonds. The reported sensor illustrates a high sensitivity of 0.22 nm/mbar for the methanol varying between 80 to 100 mbar. This was the first time that the role of fibroin protein has been explored in the sensing of VOCs with a good reversible response. Additional grating-based VOC sensors are listed in table 3.

Table.3: Comparative table showing various grating-based optical fiber VOC sensors

Ref.	Target VOC	Type of Grating	Sensing Material	Sensitivity	LOD	Operating range
[112]	Toluene, cyclohexane, hexane and benzene	LPG	Calixarene	-	-	0 to 80 kppmv
[113]	methanol, 1-propanol, 2-propanol, acetone	FBG	PDMS	$1.63 \times 10^{-3} \text{ nm}/\% \text{ at } A_p = 2.25 \text{ mm}^2$ $2.9 \times 10^{-3} \text{ nm}/\% \text{ at } A_p = 4 \text{ mm}^2$	-	0-1 relative volume ratio
[116]	Ethanol, Acetone, and methanol	LPG	Zeolite imidazolate framework (ZIF8)	$0.015 \pm 0.001 \text{ nm}/\text{ppm}$ for Acetone and	6.67 ppm for acetone	987- 19,700 ppm for acetone

				0.018 ± 0.0015 nm/ppm for ethanol	5.56 ppm for ethanol	1240 to 24,800 ppm for ethanol
[117]	Methanol	FBG	Diphenilalanine nanotubes	(-7:3±0:8) pm/(%v/v)	-	
[118]	Methanol	LPG	Fibroin	0.22 nm/mbar	-	
[119]	Xylene and cyclohexane	LPG	PDMS	19 nm/50% for xylene	300 ppm for Xylene	
[120]	cyclohexane, m-xylene, trichloroethylene, and commercial gasoline	LPG	Copolymer PDMS/polymethyl-octylsiloxane (PMOS)		Xylene:134ppm Trichloroethylene: 3300 ppm Cyclohexane: 1850 ppm Gasoline: 10500ppm	
[121]	Acetone	LPG	PDMS	$9.4 \times 10^{-4} \text{ ppm}^{-1}$	170 ppm	
[122]	Toluene	LPG	calix [4] resorcin arene	-	~100 ppm	1-400 ppm
[114]	Ethanol	LPG	ZnO nanorod	Measure in RI variation	-	100 min exposure time
[123]	Ethanol Acetone Toluene	TFBG	Molecularly imprinted polymer	0.44 pm/ppm 0.38 pm/ppm 0.28 pm/ppm	Few ppm	0-17 ppm
[124]	Hydrocarbons	4 FBG	hydrophobic siloxane co-polymer	-	-	-
[125]	Ethanol	FBG	PMMA	Linear response for 3% concentration	-	-
[126]	Dichloromethane Acetone	D shape FBG	PDMS	4000 ppm 6000 ppm	-	0-90000 ppm

4.3. Microstructured optical fiber (MOF) VOC sensors

The development of sensing devices based on microstructured optical fiber (MOF) has drawn huge attention of the scientific community due to the market niches that can be benefited from their specific features [127]–[129]. The main difference between the standard fibers and the MOF is related to their structures. The standard fibers such as single-mode fibers (SMFs) and multi-mode fibers (MMFs) are made of Ge doped solid-core enclosed by a solid fused silica cladding allowing the total internal reflection (TIR) phenomenon due to the refractive index difference between core and cladding. On contrary, the MOFs are made out of multiple holes distributed along the length of the fiber. This distribution determines the effective refractive index of the structure as well as specific propagation modes. The modes can be confined in the MOF core or forced to travel along with the cladding or the air holes. There are several ways to categorize the MOF besides the type of material such as dimensions, geometries, shape, and distribution of air holes. The detailed categorization is shown in Fig.15 [130], [131].

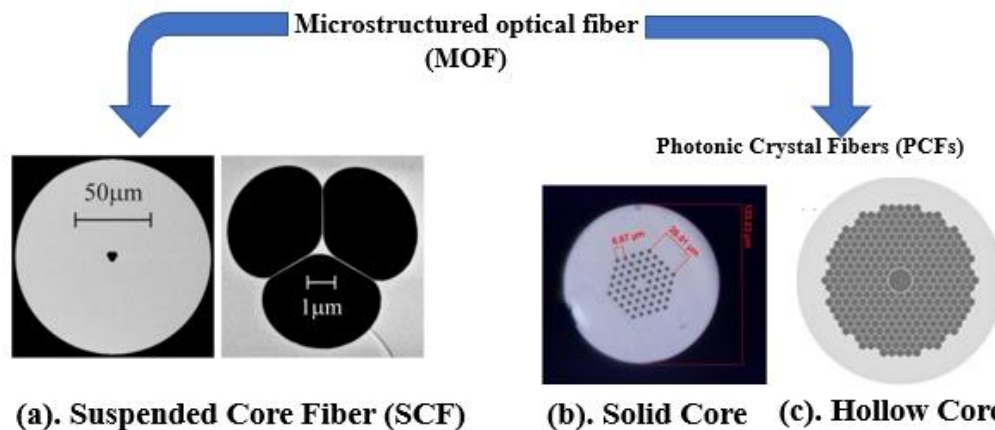


Fig.15. Classification of MOFs (a) Suspended core [132] (b) Solid core [133] (c) Hollow core [134]. [Reprinted/Adapted] with permission.

Photonic crystal fiber (PCF) is a kind of specialty optical fiber consisting of built-in microstructures, mostly small air holes in glass. The first PCF term was proposed by Russell *et al.* in 1997 [135], which was characterized by an aperiodic distribution of air holes along the length of the fiber. The structural parameters that define the geometrical arrangement of the distributed holes are diameter of the core (ρ), distance between the centers of two consecutive air cavities (Λ), and diameter of the holes (d). MOF has been explored on large scale in the application of VOC detection and other gases. The development of MOF based sensors to detect gases and VOCs has been increased promisingly because (i) there are some gasses available in the environment which are highly toxic and poisonous for living species and the environment, and (ii) several industries need to identify the toxic gasses and VOCs to track maturation, fermentation processes, and other harmful chemical reactions inside the industry. Hence, various scientific efforts and contributions have been proposed to use the special features of MOFs to develop the VOC sensors, some of the latest reports have been discussed below.

Detection of VOCs has got a lot of attention due to their significant effect on living species as well as ecological balance. These VOC sensors also play a vital role in the applications related to biomedicine which are gaining huge interest from researchers. One of its major work is to diagnosis of diseases by detecting certain indicators [136] i.e. VOC molecules available in the exhaled breath of the patient. Due to the possibility of making the sensors more sensitive and selective to various VOC concentrations, MOFs are proposed and considered as a potential candidate. In addition, MOFs are particularly useful for sensing VOCs because of their structural dimensions and design, which allows the diffusion of VOC molecules to travel through them while the air holes act as a cell for VOC vapors. One of the major differences between the detection of VOCs and other gasses is that the absorption bands for VOC molecules are not found in the range of telecommunication windows (1300 and 1550 nm) [137]. Thus, the sensing mechanism of MOF based VOC sensors is mostly due to the basic phenomenon of detecting changes in the effective refractive index of its surrounding. All the reported MOF based VOC sensors are based on wavelength interrogation techniques.

In 2009, J. Villatoro *et al.* reported an in reflection two-mode PCF interferometry to significantly reduce the difficulty of light coupling. The sensor exhibits a high sensitivity to tetrahydrofuran (THF) and acetonitrile (CH_3CN), without the requirement of any additional sensing material [138]. The reported sensor was consisting of a PCF spliced with a standard SMF as shown in Fig. 16. During the splicing process, the air holes of PCF are fully collapsed to form a standard silica glass which allows the splitting and recombination of propagated core modes. The other end of the PCF is cleaved perpendicularly, and the holes were left open so that the propagated light can interact with VOC molecules through these holes. Experimental tests were carried out for THF and CH_3CN with a maximum shift of 820 and 150 pm, respectively. The sensing head can monitor these compounds without any additional functionalization or sensing material which is considered to be the main advantage of this device.

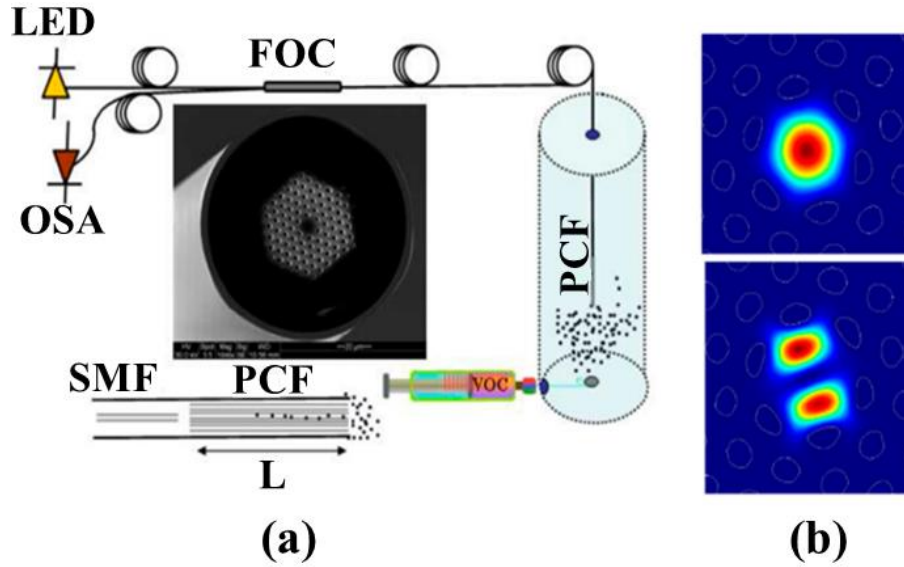


Fig. 16. (a) Diagram of the experimental setup, a micrograph of the PCF used, and drawing of the interferometer, and (b) Transverse component of the electric field of the LP01- and LP11-like modes. [Reprinted/Adapted] with permission.

In 2014, Kim *et al.* reported a VOC sensor with hollow-core photonic crystal fiber (HC-PCF) based on reflection mode for the detection of acetone vapor without using any additional sensing material [139]. In this work, one end of HC-PCF was spliced with self-phase modulation (SPM) and another end was spliced to a solid-core photonic crystal fiber (SC-PCF), as shown in Fig. 17. The sensor works on the basic principle of FP interferometer which occurs due to the three interference of various refractive indices that existed simultaneously. As the micro-cavity was filled with acetone vapors, the interference spectrum originating from the reflected light on the silica and air interfaces get changed and shows a good wavelength shift. The final sensitivity of the designed sensor has been analyzed using the fast Fourier transform (FFT) technique and exhibits a shift of 1.3 rad.

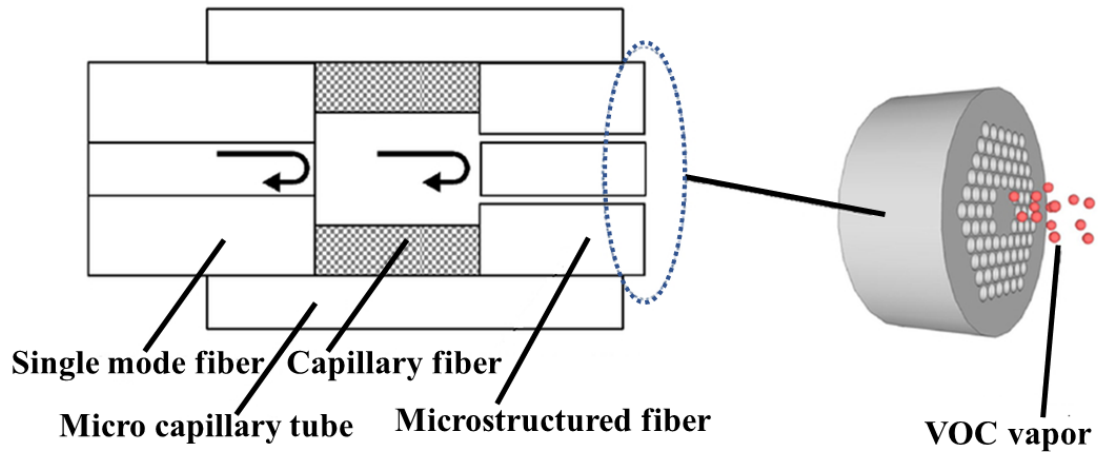


Fig. 17. Sensing principle of fabricated MOF sensor. [Reprinted/Adapted] with permission.

Later in the same year, Niu *et al.* reported a sensor for ethanol detection based on the same configuration [140]. However, the HC-PCF was worked on the transmission phenomenon based on the Rayleigh scattering effect in the HC-PCF instead of reflection. In the development of the sensor, both ends of HC-PCF were aligned with standard SMFs without splicing. A very short space between the fibers has been left to allow the diffusion of the VOC molecule through

it. The arrangement of the device is illustrated in Fig. 18. As the various concentration of ethanol is released in the gas chamber, the ethanol molecules interact with the air holes of HC-PCF through the space between SMF and HC-PCF leading to the excitement of Rayleigh scattering to produce an intensity loss of the transmitted light. When the propagated light reaches the end of the fibers, the modes recombine to form the fundamental mode. This mechanism is similar to an MZI. The device exhibits a maximum sensitivity of ~ 0.022 dB/ppm for ethanol concentration with a ~ 5.1 cm long HC-PCF.

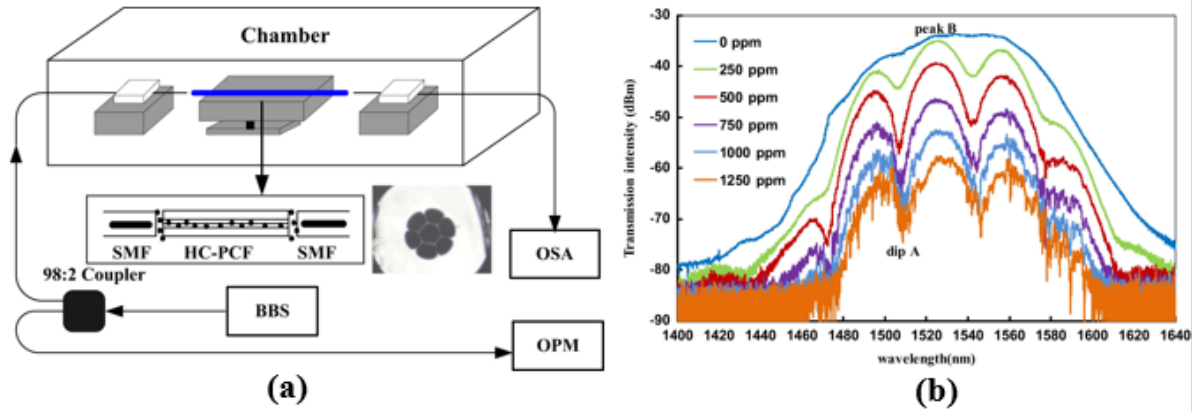


Fig. 18. (a) illustrates the schematic of experimental set-up and (b). represent the variation of wavelength corresponding to various concentration of ethanol. **[Reprinted/Adapted] with permission.**

The major limitation of these MOF based VOC sensors is the selectivity due to the lack of functionalized layer for specificity. To solve this issue Diego *et al.* developed an SCF-based VOC sensor utilizing a thin layer of ITO inside its microcavity in 2018 [141], [142]. In the proposed device, six Fabray-Perot sensing heads were multiplexed. Each one of them was fabricated by splicing at different lengths of suspended core fiber (SCF) to the SMF in order to achieve specific interferometric patterns for every device. Among all the sensing heads, three of them were deposited by ITO thin film to enhance the sensitivity for ethanol whereas two were deposited with SnO_2 to show a different response. The last sensing head was left without any metal deposition for temperature measurement. The sensing response of the sensor was carried out by analyzing the FFT phase variations in the interferometric pattern that occurred for each sensor. The device achieves a maximum phase shift of 0.31π rad for methanol atmospheres. The SCF and the setup for sensor characterization are shown in Fig. 19. This article concludes that the final sensitivity was highly influenced by the geometry cavities, but mostly by one of the cores. The SCF with an air hole in the middle of its core exhibits 5 times higher sensitivity in comparison to the others. Figure 19 (e) illustrates the response of each MOF in presence of ethanol vapor. Among which MOF-1 exhibits the lowest sensitivity with a maximum variation of 0.685 radians, while MOF-3 gives the highest sensitivity with a maximum variation of 5 radians.

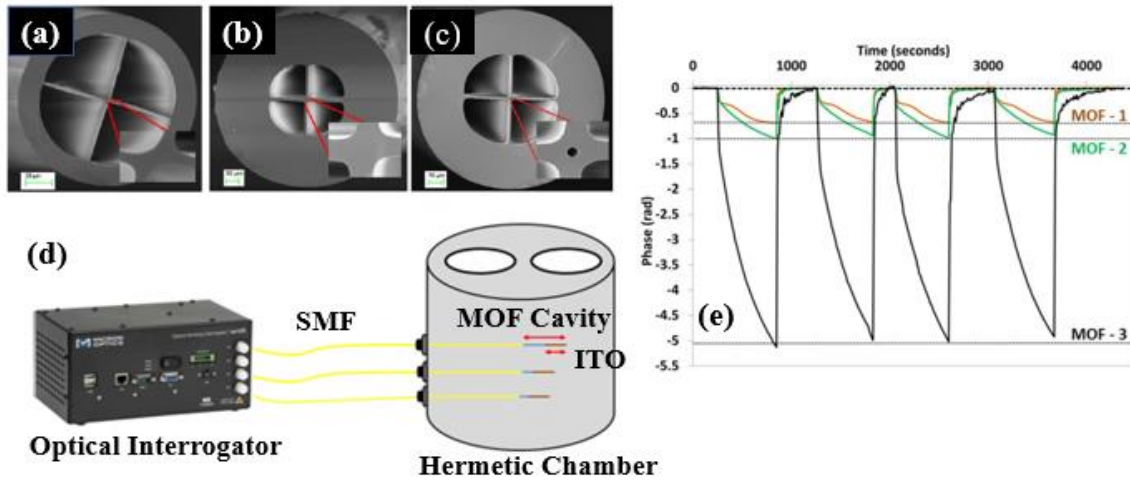


Fig.19. Cross-section of (a) MOF-1, (b) MOF-2, (c) and MOF-3, and (d) experimental setup used to analyze the response of the sensors. (e). sensing response of the sensor
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Nowadays, the role of MOF has not been fully explored yet for VOC monitoring in terms of their selectivity and sensitivity. Even though the non-functionalized surface sensor illustrates a good sensing response, there might be a selectivity issue when the device is placed in a mixed environment. The air holes along the length of MOFs play a key role because the holes allowed the host gas or VOC molecules to pass through it and improve the kinetics; moreover, the interaction between guided light and gaseous molecules is improved leading the good sensitivity and resolution of the devices. The splicing of MOF with SMF required the air hole to collapse during the splicing which may distort the actual guided mode. Additionally, its fabrication step itself has the limitations of difficulty to precisely control the length of the MOF, specifically, the length of FP cavity, required manual alignment during the splicing of MOF with SMF. Another limitation of such devices might occur due to their hole size, which needs to be optimized before the fabrication for successful passing of VOC gases through the holes, as each VOC molecules have a different size. Some of the reported MOF-based VOC sensors are listed in Table 4.

Table.4: Comparative table showing various MOF based VOC sensors

Ref.	Target VOC	Type of fiber	Sensing material	Sensitivity
[138]	THF and Acetonitrile	PCF	-	-
[139]	Allyl alcohol	PCF	-	A shift of 1.3 rad
[140]	Ethanol	Hollow core PCF	-	0.022 dB/ppm
[143]	Ethanol	Hollow core PCF	-	Shift 2.56 nm
[142]	Methanol	PCF	-	Shift 0.3π rad
[141]	Ethanol	Suspended core	-	Nearly 5π rad
[144]	Ethanol	Hollow core PCF	-	Relative sensitivity 97.28%
[145]	Ethanol Water Benzene	Hexagonal PCF	-	94.47% for water, 96.32% for ethanol, 99.63% for benzene
[146]	Ethyl chloride	Hollow core PCF	-	
[147]	Ethanol	Suspended core	-	3.9 pm/wt. %
[148]	Hydrocarbons	Suspended core	-	1.20 ± 0.04 dB/mol%

4.4. Absorption-based VOC sensors:

In the absorption-based optical fiber sensors, the detection is performed by monitoring the variation in the intensity of the light due to the interaction with VOCs [149], [150]. The sensing mechanism can be explained for two configurations, such as; (i) when the altered cladding has a lower refractive index than the core, then the variation in intensity is caused by absorption of light of the evanescent wave which is guided along with the cladding region, and (ii). if the modified cladding region has a higher refractive index than the core, part of the

optical power is refracted into the cladding, and another part is reflected into the core. The optical power reflected in the core and the light traveling through the cladding is both affected by the cladding's optical characteristics which are altered in presence of VOCs [151]. Hongyi *et al.* has reported a sensor based on the reflectance phenomenon using wavelength interrogation technique, in 2011 [152]. The sensor consisted of polypyrrole conducting polymer to detect VOCs, as shown in Fig. 20 (a). The polypyrrole exhibits excellent redox property, good environmental stability, high electrical conductivity, and water-soluble which makes it a favourable sensing material for gas monitoring. In the reported article author observed that the reflectance response of the sensor is decreased as the concentration of methanol vapor increases because of the absorption of methanol vapor. The reflectance of blue light ($\lambda = 436$ nm) gives the highest change than any other wavelengths [152]. Figure 20 (b) illustrates the response of the sensor with and without polypyrrole and Fig. 20 (c). illustrates the sensing response for a various concentration of methanol. The proposed sensor illustrates a fast response time of only a few tens of seconds while the lowest LOD of 1 ppm for methanol and 10-30 ppm for other VOCs. The proposed sensor needs additional care during the installation because the polypyrrole film with quartz base is connected mechanically on the end face of the fiber. The additional care may limit its use in the hazard or industrial environment.

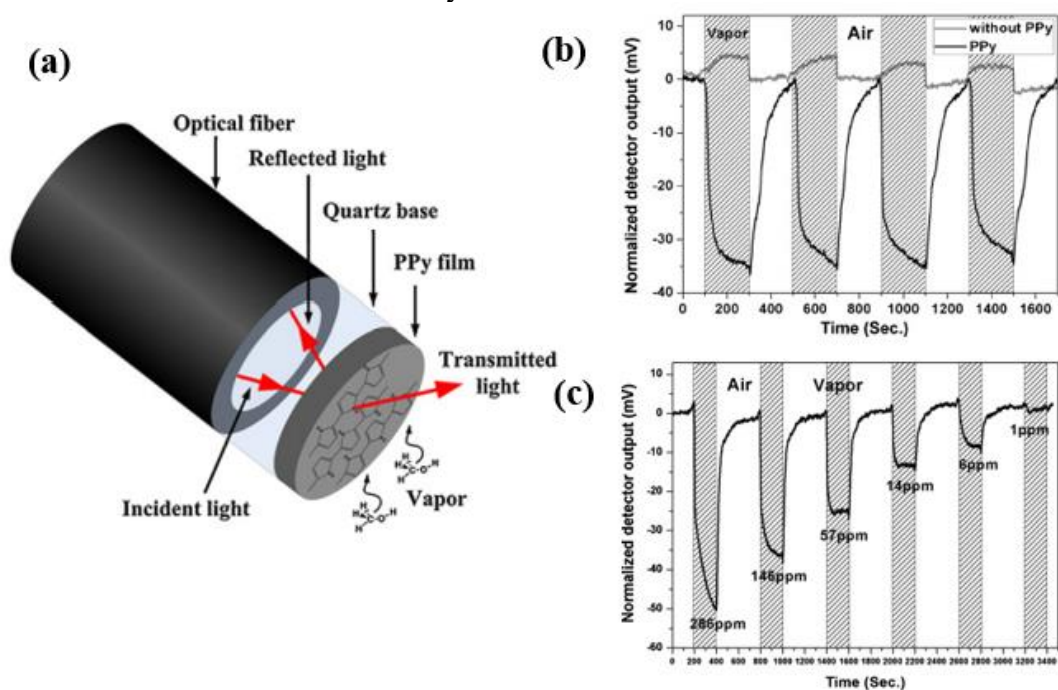


Fig. 20. (a). Schematic representation of reported sensor; response of the sensor (b). The effect of polypyrrole thin film on optical fiber for the methanol vapors (c). various concentration of methanol [Reprinted/Adapted] with permission.

In 2013 Elosua *et al.* reported another absorbance-based optical fiber sensor to detect VOCs using wavelength interrogation technique [153]. The reported sensing configuration comprises the deposition of the organometallic compounds on the uncladded plastic fiber. The sensing organometallic compound illustrates a reversible change in its optical properties such as absorption, refraction index, etc. in presence of VOCs. The sensing material was deposited on a 1.5 cm long uncladded fiber with a layer-by-layer assembling method. The reported sensor was exposed to various concentrations of methanol, ethanol, and isopropanol vapors. It exhibits a maximum sensitivity of 0.520, 0.417, and 263 nm/ppm, for methanol, ethanol, and IPA, respectively. Temperature insensitivity is its major advantage over other VOC sensors however, lack of discussion over response time and the limit of detection can be considered as a limitation of the device. In the same year, Khan *et al.* developed another wavelength interrogation base

D-shaped optical fiber sensing configuration for the detection of low concentrations of VOCs as shown in Fig. 21 (a) [154]. The sensor was based on the pulse width modulation technique. The sensing head was immobilized with Nile red and polyvinylpyrrolidone as a sensing membrane and tested with the VOCs ranging from 0 to 100 ppb. The sensing material utilizes the solvatochromic property of Nile red in changing the optical characteristics of the material after VOC absorption. The sensing response of the proposed device was linear and also exhibited a good reproducibility of the system. The maximum sensitivity of 7.79 ns/ppb for benzene was achieved. Fig. 21 (b) illustrates the SEM image of the fabricated sensor, while Fig. 21 (c) represents the fabricated device.

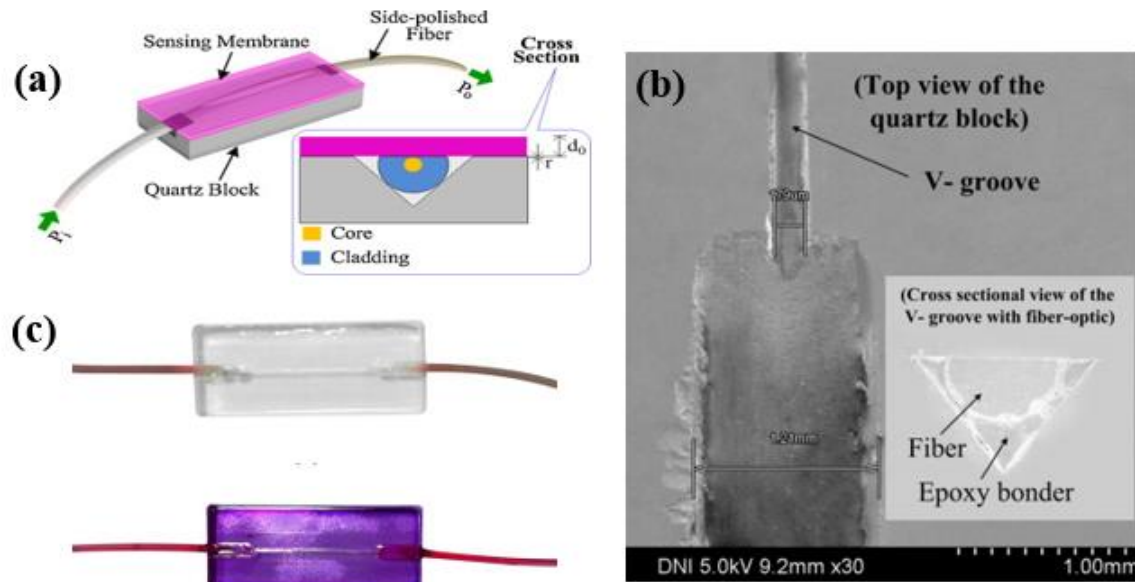


Fig. 21. Schematic of the reported side-polished optical fiber sensor for VOC detection.
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In 2015, Yeom *et al.* reported a D-polished fiber sensor for VOC detection using wavelength interrogation technique [155] with enhanced sensitivity, selectivity, and dynamic range. The sensing head consists of a standard SMF functionalized with a thin layer of the sensing membrane which works as a planer waveguide [156]–[158]. It requires five types of solvatochromic dye which are used in synthesizing the sensing membrane. The fabricated sensor was investigated over five kinds of VOCs, which are dimethylamine, benzene, ethanol, acetic acid, and toluene. The sensitivity of the membrane deposited with Reichardt's dye for Dimethylamine gas was higher than other VOCs. The malononitrile-coated sensing membrane achieved a higher sensitivity for ethanol in comparison with other dye-coated sensors. Although, the D-shaped fiber is considered a a potential candidate for sensing applications and achieves high sensitivity, however, a precise D shape polishing of fiber might be a complicated task in practical. The polishing of fiber needs to be optimized first to get maximum coupling of the core guided light with the surrounding environment, resulting a high sensitivity of the sensor. Additionally, the polishing of fiber may make it fragile for multiple monitoring.

Recently in 2020, Yunhe *et al.* also reported an absorption-based sensor to monitor VOCs [159]. The reported sensor utilizes a novel cholesteric liquid crystal (CLC) film-based dual-probe optical fiber sensor for the detection of acetone and ethanol. When the CLC film interacts with VOC molecules, it's considered as the host-cavities engulfing analyte molecules in an enzyme-like mechanism. The short-range interactions between the liquid crystal (LC) molecules are similar to that interaction between VOC and LC. Hence, the penetration of such gaseous molecule into the CLC will lead to a change in pitch, consequently the shift in reflection wavelength. The schematic of the fabricated device is illustrated in Fig. 22 (a). The

developed sensor consisted of a 2×2 multimode fiber coupler. The two output fiber ends were covered by two types of cholesteric liquid crystal film (CLCF) with different pitches. The redshift of CLCF reflection spectra occurred with the increase in the concentration of VOCs. The fabricated device achieved a good sensitivity of the two CLCFs which are 8.435 nm·L/mmol and 14.867 nm·L/mmol to acetone, and 14.586 nm·L/mmol and 29.303 nm·L/mmol to ethanol, respectively. Figure 22 (b) exhibits that the wavelength shift depends on the total concentration of the mixed gas at various ratios of ethanol and acetone, both CLCFs achieve a higher sensitivity to the mixed gaseous environment. The main advantage of the proposed structures is their selectivity. The sensor can successfully distinguish the difference between the mixture of two VOCs. The dual-probe VOC sensor can also distinguish the concentrations of acetone and ethanol in their mixture. The coupler-based sensor also helps to minimize the size of the sensor and facilitates the sensor installation for real-world realization. However, a reflection-based sensor needs a reflecting metallic surface (such as Ag, Au, etc.) at the end facet to effectively reflect maximum light from the interface. The unavailability of metallic coating may cause poor reflectivity and hence the sensing response. Table 5 illustrates the comparison between various absorption-based VOC sensors.

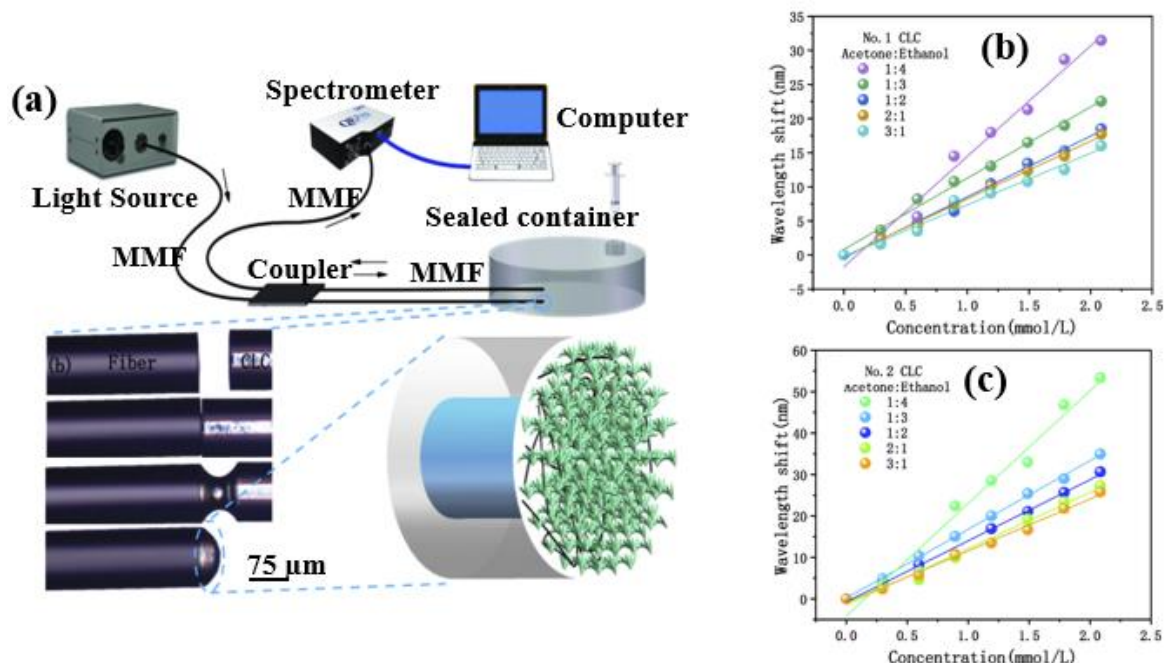


Fig.22. (a) Schematic diagram of the experimental device; dependence of the CLC wavelength shift on the total concentration of the mixed gas at different mixing ratios of acetone to ethanol (a) No.1 CLCF and (b) No.2 CLCF. [Reprinted/Adapted] with permission.

Table.5: Comparative table showing various absorption based optical fiber VOC sensors

Reference	Target VOC	Sensing material	Sensitivity	LOD	Operating range
[152]	Methanol, ethanol, acetone, toluene, chloroform, isopropyl alcohol	Polypyrrole	-	10-30 ppm	1-286 ppm
[153]	Methanol, Ethanol, Isopropanol	organometallic compound [Au ₂ Ag ₂ (C ₆ F ₅) ₄ (C ₆ H ₅ C≡CC ₆ H ₅) ₂] _n	0.520nm/ppm, 0.417 nm/ppm 263 nm/ppm	4 ppm 16 ppm 10 ppm	0-180 ppm
[154]	Benzene	Nile red and polyvinylpyrrolidone	7.79 ns/ppb	-	0-100 ppb
[155]	Dimethylamine, benzene, ethanol, acetic acid, toluene	C ₂₀ H ₁₄ N ₄ O malononitrile dye Reichardt's dye	Highly sensitive for ethanol Highly sensitive for Dimethylamine	1 ppb	0-15 ppb

[159]	Acetone Ethanol	cholesteric liquid crystal film	8.435 nm·L/mmol and 14.867 nm·L/mmol to acetone 14.586 nm·L/mmol and 29.303 nm·L/mmol to ethanol	-	0-2.98 mmol/L 0-1.798 mmol/L
[160]	Acetone	Nanocrystalline copper bromide (γ -CuBr)	Acetone exhibits 14.3% superior Sensitivity to ammonia gas, and 7.4% then ethanol gas	-	~800 ppm
[161]	Ethanol	Magnesium cobalt oxide (MgCo_2O_4)	81×10^{-3} k/Pa for Ethanol	-	0-500 ppm
[162]	Ethanol Methanol	Silica based Nile red	-0.130 nm/ppm for ethanol -0.036 nm/ppm for methanol	~77 ppb ~281 ppb	0-18 ppm
[163]	Methanol	Epoxy/Au/MoS ₂	-0.0184 pm/ppm	-	-
[72]	Isopropanol	Ag/P-TiO ₂ composite	1.35 nm/%	-	0-100% concentration
[164]	Acetone, Ethyl acetate Cyclohexane Isopropanol	2D nanosheet of MoS ₂	0.0195nm/ppm- acetone 0.0143nm/ppm- ethyl acetate 0.0072nm/ppm- cyclohexane 0.0058nm/ppm-Isopropanol	0.51 ppm 0.70 ppm 1.39 ppm 1.72 ppm	0-1000 ppm
[165]	Ethanol	quantum-dot gel	-3.3%/ppm	-	0-30.6 ppm
[166]	Ethanol	WO ₃ and WO ₃ /gC ₃ N ₄ Composite	62.5/counts	-	0-500 ppm
[167]	Ammonia, ethanol, methanol	samarium oxide (Sm ₂ O ₃)	7×10 ³ counts/kPa for ethanol 4×10 ³ counts/kPa for methanol 483 counts/kPa for ammonia	-	0-500 ppm
[168]	Tetrahydrofuran Acetone Methanol	cholesteric liquid crystal film	7.08 nm·L/mmol, 3.46 nm·L/mmol 0.52 nm·L/mmol	-	0-14 mmol/L
[169]	Ethanol	polypyrrole-polyethylene oxide	3.89	-	0-10 ppm
[170]	Methanol Ethanol Ammonia	Nanocrystalline bismuth oxide	212×10^{-3} /kPa 109×10^{-3} /kPa 15×10^{-3} /kPa	-	400 ppm

4.5. Semi-VOC optical fiber sensors:

In addition to VOC, the semi-VOCs (SVOCs) are also a matter of concern. Even the SVOCs are less likely to vaporize, this doesn't make them less dangerous. Indoor dust is a major sink for SVOCs, which are commonly found in consumer products. The WHO classified SVOCs as indoor organic pollutants with a high boiling point varying between 240/260 and 380/L [171]. Some of the major SVOCs compounds are benzoic acid, benzyl alcohol, organochlorine pesticides, hydrocarbons, phenolic compounds, etc. Several attempts have been taken into account since 1993, to precisely detect such kinds of SVOCs in air and water samples using the mid-infrared (MIR) detection technique [172]–[179].

The inherent selectivity and sensitivity of MIR spectroscopy provide access to fingerprint spectra of organic compounds and LOD utilizing Fourier-transform infrared attenuated total reflectance (ATR) based sensors in the ppb levels [180]–[182]. In 2013, Rui *et al.* reported ATR based sensor for in-situ and simultaneous detection of multiple chlorinated hydrocarbons mixed in water [183]. The analysis was performed for 1,2-dichlorobenzene, 1,3-dichlorobenzene, monochlorobenzene, trichloroethylene, chloroform, and perchloroethylene. The sensor comprises polycrystalline silver halide fiber possessed a unique cylindric geometry deposited with an ethylene/propylene copolymer matrix for the solid-phase extractor. The fabricated fiber sensors greatly enhanced the analytical signal and provide higher sensitivity. This analysis exhibits a high sensitivity towards the mixture of various chlorinated hydrocarbons at a wide concentration range varying from 5~700 ppb. Fig. 23 illustrates the sensing configuration along with the schematic of experimental setup used to characterize the sensor. The silver halide sensing head was used as transducer, attached with fiber optic extensions at both ends. The evanescent field analysis was performed by focusing the IR beam into the silver halide coated sensing head via cylindrical extension using a gold-coated, off-

axis, parabolic mirror. In the mid-IR region, various chemical bonds absorb various infrared frequencies based on the atoms connected and surrounding molecules.

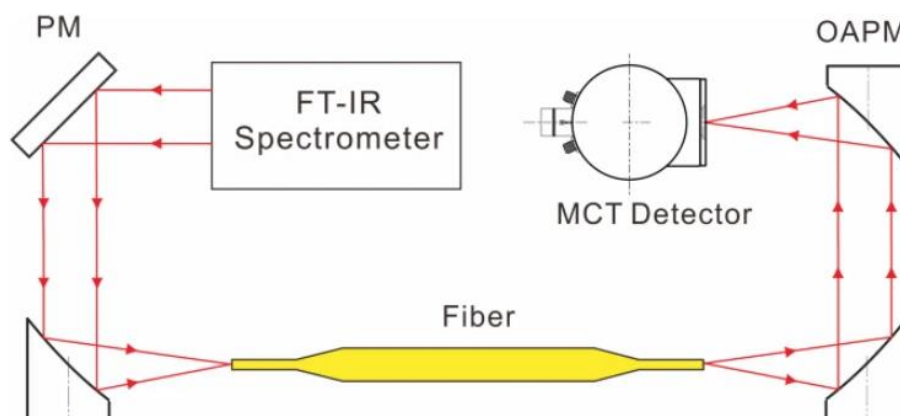


Fig. 23. Schematic of experimental setup. [Reprinted/Adapted] with permission from

The same year witnessed the improved sensing configuration for the detection of monocyclic and polycyclic aromatic hydrocarbons in water mixture developed by Pejčic *et al.*[184]. The sensing configuration comprises a zinc selenide optical waveguide coated with a thin film of poly(isobutylene). The analysis was performed over several VOCs among which the sensor accurately quantifies the toluene, benzene, ethylbenzene, xylenes, and naphthalene concentrations in an oil-water mixture. In 2015, Lu *et al.* have utilized flattened silver halide fibers along with 2D correlation spectroscopy by investigating the heat-induced denaturalization of bovine serum albumin [185]. The author has used the silver halide fiber sensor in ATR spectroscopy because its broad IR-transparent window allows tunable flexible geometry. The use of fiber-optic as a sensing element leads to the enhancement of the analytical signal and hence the sensitivity.

Later, in 2016, the same group of Lu *et al.* and collaborators has developed another ATR-based sensor utilizing copolymer material for rapid quantification of SVOCs in water [186]. The experimental setup used in characterization was somehow similar to Fig. 23. The group has developed this protocol utilizing a similar optical fiber waveguide that comprises a coating of silver halide ($\text{AgCl}_x\text{Br}_{1-x}$) and hydrophobic ethylene/propylene copolymer (60/40). The sensing head was attached with cylindrical extensions at both ends as shown in Fig. 23, in order to increase the number of TIR. The deposited polymer traps the VOCs mixtures and excludes water. Depending on the VOC and monitoring scenario, IR spectra with specific frequency windows, scan times and spectral resolutions are obtained, from which concentration information for each VOC was derived. This developed protocol allows the detection of multiple VOCs simultaneously, at a very low concentration of 10 ppb.

In 2018, Zhong *et al.* exploited the photochemical plastic optical fiber sensors to determine phenol concentration and its photocatalytic degradation rate [187]. The sensing head was fabricated by depositing $\text{Er}^{3+}:\text{YAlO}_3/\text{SiO}_2/\text{TiO}_2$ on the uncladded portion of the plastic optical fiber. The sensing material contains the up-conversion luminescence agent $\text{Er}^{3+}:\text{YAlO}_3$ to generate UV light. As a sensing mechanism, the UV light is absorbed by TiO_2 particles in order to generate electron-hole pairs to monitor the degradation of phenol. In particular, the refractive index of the fiber core is lower than the sensing material due to the presence of TiO_2 and $\text{Er}^{3+}:\text{YAlO}_3$. The authors have observed that the increase in the RI between the fiber core and the sensing material can enhance the evanescent wave penetration depth, surface luminous intensity, and a number of total reflections on the sensing head, leading to the enhancement of

the excitation light for the degradation of phenol in aqueous solution. Fig. 24. illustrates the sensing configuration of the developed sensor.

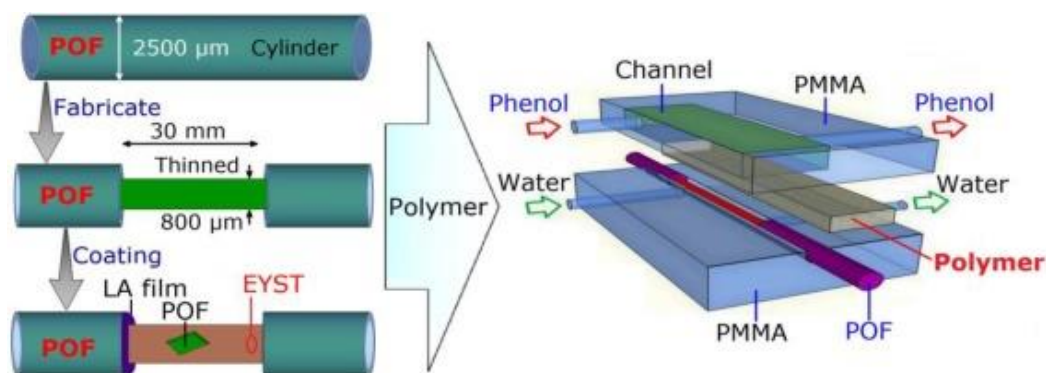


Fig. 24. Schematic representation of the fabrication of the photochemical fiber-optic sensor. [Reprinted/Adapted] with permission.

Later in 2019, Zhong *et al.* reported a multiparameter lab-on-a-chip device for monitoring the degradation of phenolic compounds using microalgal biofilm [188]. The sensor consists of lab-on-a-chip integrated a photocatalytic D-type plastic fiber, D-type plastic fiber with a glass fiber membrane (in order to distinguish the solution from *Chlorella Vulgaris*), normal D-type plastic fiber sensor, and microchannel, as shown in Fig. 25, (a). The developed sensor accurately monitored the phenol degradation and progression of a microalgal biofilm, as shown in the microscopic image, Fig. 25 (b).

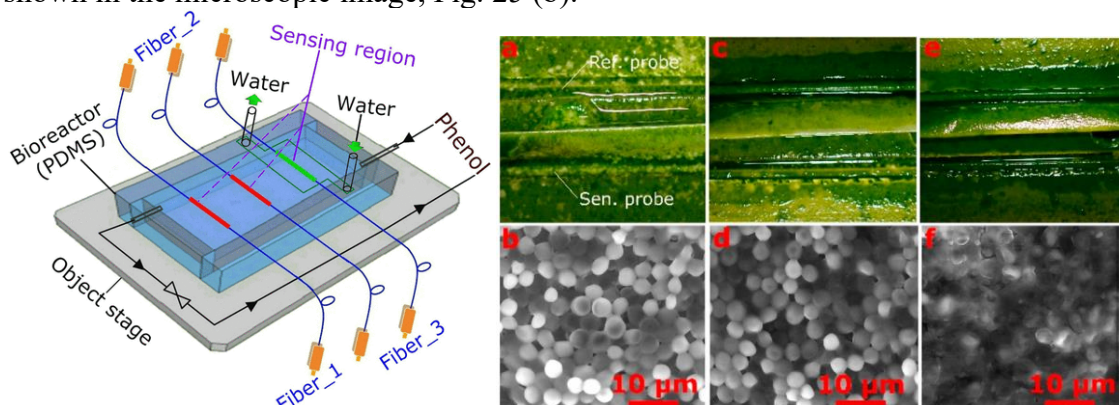


Fig. 25. (a). Schematic diagram of the developed device (b) Microscope image of the biofilm at different culture times. [Reprinted/Adapted] with permission.

The same group of Zhong *et al.* reported similar kinds of VOC sensors for the detection of phenolic compounds in water in 2019 [189], [190]. The group has demonstrated the potential of plastic optical fiber in SVOC monitoring. Fig. 26 illustrates the schematic of one of the SVOC sensors reported by the group [189]. The sensor is based on the change in the property of the reflected light from the fiber and sensing material interface. The sensing head consists of a polymer membrane, gold mirror, and SiO₂/nitrogen-doped TiO₂. The gold film was used as a reflecting mirror for the incident light in order to enhance the number of TIR and hence improve the decay of evanescent waves. While SiO₂/nitrogen-doped TiO₂ was used to enhance the light absorption because the nitrogen dopant can improve the relatively large bandgap of the TiO₂ and enhance the absorption of visible light of the TiO₂. The composition of SiO₂ helps in the improvement of the quantum and photocatalytic efficiency of the TiO₂. Additionally, the sensing head contains an inlet and outlet micro-channel between the fiber and sensing layer to provide the injection and outflow of the water. The light ray, after penetrating the fiber core, directly traverses the core-SNT interface where it is diffracted and transmitted into SNT coating due to the difference in RIs between the fiber core and SiO₂/nitrogen-doped TiO₂. The sensor

exhibits a high selectivity and sensitivity for phenol of $0.294 \times 10^{-3} \text{ (mg} \cdot \text{L}^{-1})^{-1}$ along with the low LOD 30 $\mu\text{g/L}$. The selectivity of the device was tested over pure phenol analyte and mixed solutions with FeCl_3 , CuSO_4 , and glucose.

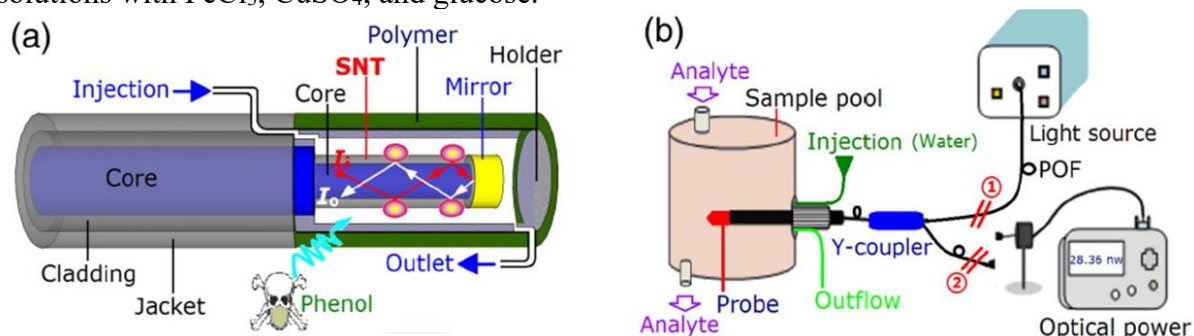


Fig. 26. (a) Schematic diagram of the sensing configuration, and (b) experimental setup used to monitor phenolic compound. **[Reprinted/Adapted] with permission.**

As most recent work published in 2020, Mishra *et al.* reported a phenolic-compounds sensor utilizing a long period of grating immobilized with an enzyme entrapped in polyacrylamide gel [191]. The author has used tyrosinase (polyphenol oxidase) as an enzyme with a thickness of 900 nm deposited around the sensing configuration. The basic principle of the sensor was based on the change in the refractive index of tyrosinase with the interaction of phenolic compounds leading to the shift in the grating wavelength. The results exhibit a good blue shift with an increasing concentration of the phenolic compounds in an aqueous solution. The fabricated device achieved a good sensitivity of 0.0088 nm/ μM at a pH value of 7.0 in the concentrations range of 0–800 μM of aqueous solution with a LOD of 6.8 μM . The selectivity of the sensor was also investigated using 4-chlorophenol, m-cresol, and phenol. The schematic of the developed device and the experimental setup is shown in Fig. 27. In addition to the discussed devices, there are several sensors have been reported in past decades, shown in Table. 6.

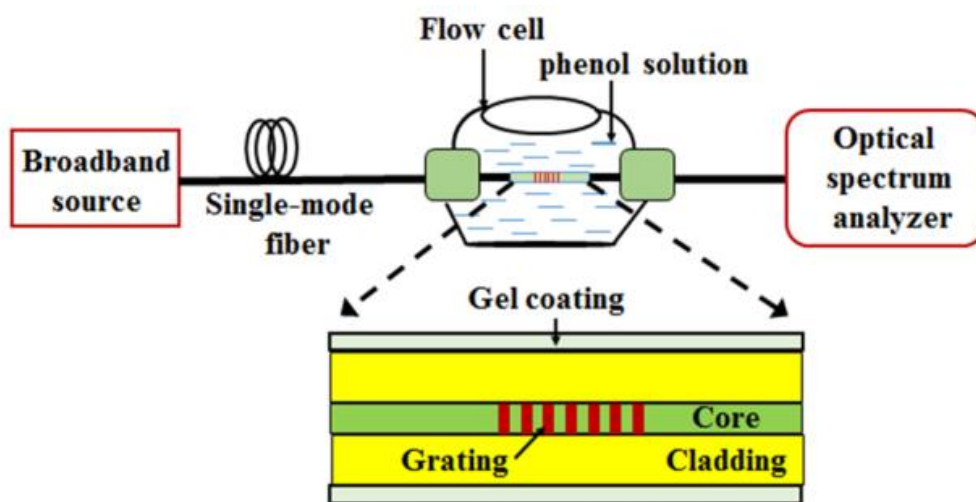


Fig. 27. Schematic of a sensing head and the experimental setup.

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Table.6: Comparative table showing various Mid-IR optical fiber based SVOC sensors.

Reference	Target VOC	Sensing material	Sensitivity	LOD	Operating range
[183]	Chlorinated hydrocarbons	polycrystalline silver halide	-	mid	5~700 ppb
[180]	Chlorinated hydrocarbons	poly(vinylchloride) (PVC)	Linearity > 0.96	0.3, 0.6, and 0.5 mg L ⁻¹	-
[184]	VOC compounds and hydrocarbons	poly(isobutylene) (PIB)	Linearity > 0.99	-	Few ppb

[192]	Chlorinated hydrocarbons	Teflon	Linearity > 0.90	250–800 ppb	TOC: 0-6000 mgL ⁻¹ COD: 0-6000 mgL ⁻¹
[186]	VOC	Silver halide	-	Mid to low ppb	0-1000 ppb
[190]	Phenol	Er ₃ +YAlO ₃ /SiO ₂ /TiO ₂	-0.39 (mg·mL ⁻¹) ⁻¹	15 µg·L ⁻¹	10-100 mg/L
[189]	Phenol	SiO ₂ /N-doped TiO ₂	0.294×10 ⁻³ (mg·L ⁻¹) ⁻¹	30 µg·L ⁻¹	0 to 100 mg/L
[187]	Phenol	Er ₃ +YAlO ₃ /SiO ₂ /TiO ₂	-0.195 × 10 ⁻³ (mg/L) ⁻¹	40 µg/L	0-100 mg/L
[191]	Phenolic compounds	tyrosinase (polyphenol oxidase)	0.0088 nm/µM	6.8 µM	0-800 µM

5. More relevant applications and future prospects:

Sensing material, configuration, sensitivity and selectivity, and detection range are the most essential requirement of any gas sensor. For commercial purposes, the reviewed sensing configurations must need to comply with certain standard requirements [193]

5.1. Selection of sensitive materials:

Remarkable progress in the sensing performance of VOC sensors has been achieved in the past decade by utilizing several kinds of sensing material such as polymer, nanomaterial, zeolite, epoxy, etc. However, each material possesses unique properties for the detection of VOC vapors, depending on the sensing configuration and the type of VOCs. Hence, in order to develop a highly selective and sensitive VOCs/SVOCs sensor, one needs to understand the role of sensing material. The detailed discussion over the selection of various sensing materials has been discussed below:

Adsorption material: There are several sorbents have been introduced and explored in past years due to their excellent ability to trap the VOC molecules from the environment such as PDMS, polyethylene, ethylene/propylene, etc [194]. Among which PDMS was exploited by several researchers after Baltussen *et al.* described the versatility of PDMS as a potential sorbent material for VOCs [195]. Unlike most sorbent materials, PDMS retains its charge through dissolving rather than adsorption. Unlike other solid sorbent sensing materials, the retention on PDMS occurs resulting in the dissolution rather than adsorption. Additionally, it also possesses several advantages over other polymer which makes it a potential sorbent for VOC monitoring. For example, (i). the PDMS is more inert compared to another sorbent and, hence, less reactive with surrounding [195]. (ii). Several researchers demonstrated the selectivity of PDMS for several VOCs hence it can be considered as an effective polymer to trap polar VOCs such as organic acids [195] (iii). Compared to other sorbent sensing materials, it requires low thermal desorption temperatures [195] (iv). it does not introduce or absorb additional residue or water into the measurand [196]. In order to improve the adsorption selectivity of VOCs, Liu *et al.* modified PDMS with activated carbons [197]. The result illustrates that hydrophobicity plays a key role in the water adsorption of activated carbons. These findings confirm that the polarity of the molecule is a key element in adsorption on the PDMS hydrophobic surface. In addition to PDMS, hyper crosslinked polymeric resins and Zeolite are also considered as the main sensing materials for the detection of VOCs/SVOCs. Zeolites are widely used in environmental protection applications such as adsorption of VOCs and heavy metal ions due to their excellent hydrophobicity, tunable pore size, superior thermal stability. Zeolites are aluminosilicates with a highly organized pore structures [198]. The typical adsorption model of zeolites is monolayer adsorption at low-pressure. With the increase in the relative pressure of VOCs, the capillary condensation phenomenon occurs in their pore structure leading to a sharp increase in the adsorption capacity. The most widely used zeolites for VOC monitoring are SBA- 15 and MCM-41 [199], [200]. Zeolites are considered as a more stable sensing material in chemical and thermal environments. The basic mechanism of VOCs adsorption from the adsorbents normally consists of the electrostatic attractions, the interaction between the hydrophilic sites and the polar VOCs, and hydrophobic sites and nonpolar VOCs.

The average adsorption capacity of VOCs has a good linear relationship with the average specific surface area and pore volume. Generally, the large size of VOC molecules exhibits a lower adsorption capacity while the volume of narrow micropores adsorbent material governs the adsorption of low concentration VOCs such as the adsorption of benzene.

Metal oxides nanostructures: Since the sensing capability of metal oxides was established in the 1960s, VOC sensors based on these materials have attracted considerable attention from the both scientific and industrial points of view. From a survey of literature discussed above, it can be concluded TiO_2 , ZnO , MoO_3 , SnO_2 , In_2O_3 , etc., illustrates several nanostructured morphologies e.g., nanotubes, nanoparticles, nanofibers, nanorods, nanowires are considered as the most promising metal oxide nanostructures for selective detection of VOCs with LOD of ppb. Among the several VOCs, the detection of acetone and ethanol has been widely studied using such oxide nanostructures, whereas a very limited number of reports about other VOCs such as benzene and acetylene can be found too. The major drawback of such material is its selectivity with the mixed volatile environment, which may be the drawback of using oxide nanostructures as VOC sensing material. However, some researchers observed that alloying the oxide metals may improve the selectivity which can be tuned for the VOCs by tuning the composite material. For example, Sukhwinder *et al.* reported the composition of $\text{MoS}_2/\text{TiO}_2$ to be highly selective for ethanol (1ppm) and methanol (2ppm) [201], while Shao *et al.* observed that graphene quantum dots/ TiO_2 composition gives better selectivity for isopropanol [202]. Hence the tuning of composition with metal oxide may be one of the best ways to achieve a high selectivity using such materials.

5.2. Response time: Another important requirement is the response time of the sensor, which is defined as how fast the sensor works with the change in the environment. It mainly depends on the sensing material, specifically in terms of its density, physical structure, and the thickness of deposited material. Chemical vapors must be diffused through the sensitive film quickly to achieve a fast response time, which is only possible with an optimized thickness of the sensing layer. Besides the response time and selectivity, the recovery time is another important requirement that has to be fulfilled by any sensor before it can be commercially utilized. In general, the recovery time of the sensor is defined as the time for a sensor to return to the baseline value after the step removal of the measured variable [203]. In order to attain high accuracy and precise detection, the effect of external factors e.g., humidity, pressure, temperature, and gas dynamics around the sensor must be well-understood.

5.3. Sensitivity and selectivity: Among several, sensitivity and selectivity are the topmost essences for any gas sensor. Selectivity is defined as the capability of the sensor to distinguish specific VOC among the mixture of various VOCs. The selectivity of optical fiber-based VOC sensors is mainly governed by light-matter interaction and their selective absorption on the surface [72], [112], [155]. The physical and chemical properties of the sensing matrix work together to evaluate the selectivity of the sensing device. The application of highly selective and real-time detection of VOCs has generally been demanded for highly toxic and explosive gases e.g. hydrogen [204], methane [205], nitrogen oxide [206], chloroform [207], dichloromethane [208], etc. In this review article, a detailed study has been carried out on various sensing configurations and materials to enhance the sensitivity and selectivity of the device. On the other hand, there are some sensors available with poor selectivity, i.e., they respond to only some specific vapors. This feature of the sensor is also useful in situations that require the analysis of gas mixtures. For example, several beverages and food emit various VOCs during their fermentation maturing process, which can be studied to see if the process is completed correctly. Hence, in those applications which required a more rigorous analysis, or in the case where the mixture of organic vapor has to be identified, an association of non-selective fiber-based sensors can be implemented, which is generally known as a sensor array. In these sensing systems, the response of the device is mathematically evaluated in order to

identify a given sample, e.g., employing Artificial Neural Networks (ANN); another possibility is to use Principal Components Analysis (PCA) analysis. For the case of wine, the developed sensors are sensitive to acetic acid and alcohols and helpful in determining whether or not the product has been prepared in the correct way.

Further possibility: The selection of appropriate material and the sensing configuration can combinedly enhance the sensing response of any sensor. Many technological advancements could be done along with optical fiber and the sensing material to facilitate real-time detection and improve their sensitivity. Most of the reported sensing configuration is either complex or failed to achieve good response time and selectivity and hence limits their implementation. Through this detailed review, it is investigated that some sensing configuration exhibits good sensitivity but the LOD, selectivity, and response time has not been considered. However, some sensor achieves good selectivity but failed to reach a high sensitivity and LOD, etc. Each sensing configuration has its own limitation and advantages. A standard fiber can be helpful to enhance the sensitivity by reducing the fiber diameter up to tens of nanometer by tapering process and utilizing surface acoustic wave to achieve high sensitivity and fast response time. The VOC sensing material along with plasmonic metals might be a potential candidate to enhance the sensitivity of the device by utilizing the surface plasmon resonance (SPR) phenomenon. SPR technique has been used for several decades in order to enhance the sensitivity of any gas sensor and achieved a good response [209]–[211]. It has been reported by several researchers that reducing the diameter causes the acoustic wave to move into the cladding and reach to surrounding media cladding interface, hence enabling it to interact externally with the guided wave [212]. When such nanofibres will be coated with a VOC sensing material, the propagated acoustic wave will differential delay in velocity due to the variation in density or elasticity of sensing material such as PDMS, PANI, etc., in the presence of VOC molecules, fundamental to its sensing, which will be utilized to estimate the sensing response of sensor [213]. Although, nanofibre is capable of enhancing the sensitivity, however, the excessive reduction of the diameter may lead to decreases in the mechanical stability of such devices. This technique attracts researchers due to its tremendous advantages of highly sensitive detection properties and wide range of applications.

6. Conclusion:

There are various optical detection techniques have been developed and employed for VOC detection and monitoring in past decades. In this review article, the role of various kinds of optical fibers and their detection method, sensor design, sensing material, working principle, and sensing performance including selectivity, sensitivity, and linearity are presented in detail. Several sensing materials and additional coating have been employed to enhance the sensitivity and target specific or mixed VOCs. Each sensor comprises some advantages and some limitations which are outlined in the discussion. The new idea and approaches for improving the sensing performance of devices concerning the data acquisition, selection of sensing material, and analysis are suggested. Today, the increasing growth in the fields of optics, electronics, materials science, and data analysis has opened up a new path for the study which can be expanded to include these principles for high precision and accuracy.

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