WIDE AREA AND DISTRIBUTED HYDROGEN SENSORS

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ABSTRACT

Recent advances in optical sensors show promise for the development of new wide area monitoring and distributed optical network hydrogen detection systems. Optical hydrogen sensing technologies reviewed here are: 1) open path Raman scattering systems, 2) back scattering from chemically treated solid polymer matrix optical fiber sensor cladding; and 3) schlieren and shearing interferometry imaging. Ultrasonic sensors for hydrogen release detection are also reviewed. The development status of these technologies and their demonstrated results in sensor path length, low hydrogen concentration detection ability, and response times are described and compared to the corresponding status of hydrogen spot sensor network technologies.

1.0 INTRODUCTION

Hydrogen detection is often considered an essential part of the overall safety of hydrogen fuel facilities. For example, NFPA (National Fire Protection Association) 52 [1] requires "equipment used for the compression, processing, dispensing, storage, and generation of hydrogen" to have gas detectors such that gas can be detected at any point on the equipment. Similarly, the International Fire Code [2] requires hydrogen fueled vehicle repair garages to have an approved flammable gas detection system and to have the mechanical ventilation system interlocked with the hydrogen detection system.

Some indoor facilities with dedicated high point air exhausts are well suited for conventional spot type hydrogen detectors since there are well defined locations to place the sensors. However, many other hydrogen facilities are either outdoors or are only partially enclosed without any clear location for hydrogen accumulation or exhaust. The installation of spot sensors in these outdoor or partially enclosed facilities becomes problematic at best, such that many facilities cannot comply with the codes and standards requirements for hydrogen detection using conventional spot sensors. Other indoor facilities may either be so large or have complex partitioned ceilings so as to require an excessive and unachievable number of spot detectors. This gap between the current code requirements and the available hydrogen detection technology has been identified in an April 2008 report by the Fire Protection Research Foundation [3]. The report was written in support of the U.S. Department of Energy research activities and ranked the need for further developments in wide area sensing technology among its top priorities.

The need for wide area or widely distributed hydrogen sensors in outdoor and partially enclosed facilities is directly analogous to the need for similar sensors for hydrocarbon gas detection in hydrocarbon processing and refueling facilities. Wide area optical scanning sensors have been developed and are now used in many of these facilities. However, many of the hydrocarbon optical scanning sensors are based on infrared gas absorption properties that are not applicable to hydrogen detection.

Many rocket propulsion and aerospace programs have had a hydrogen leak detection need for a long time, and there have been studies of available technologies for wide area leak detection for such applications. Sellar and Wang of the Florida Space Institute reported on the results of such an assessment for NASA [4]. They assessed eleven optical and acoustic technologies for remote leak sensing and assigned NASA Technology Readiness Levels (TRL) [5] to these technologies to characterize their technological maturity and development status. Four of the eleven technologies were assigned a TRL of 4 or higher, indicating that they have been validated for hydrogen or helium detection in at least a laboratory environment. Those four technologies are: passive acoustic (ultrasonic), spontaneous Raman scattering, Schlieren imaging, and shearography.

This paper provides a review of the development status of these four technologies and compares their advantages and disadvantages for various hydrogen leak sensing applications. We also provide a similar review and comparison for distributed detection hydrogen sensing technologies in which the sensor consists of a length of special optical fiber that responds to the presence of hydrogen anywhere along its length. All of these technologies respond with either an electrical signal or image that can be used for remote detection and possible leak location. There are other potential wide area sensors that provide a visual indication of the presence of hydrogen, but no electrical signal or remote image for alarm and automatic shutdown. Hoagland et al. [6] described one such device that provides a color change indication of hydrogen for manual detection.

2.0 WIDE AREA RAMAN SCATTERING DETECTORS

Light incident upon a gaseous hydrogen molecule induces both a vibrational and a rotational energy change in the molecule. These energy changes produce shifts in the frequency of some of the photons, with the frequency and corresponding wave length shifts called vibrational Raman scattering and rotational Raman scattering. When the Raman scattering entails a shift to larger wave lengths (with energy losses from the photon), it is called Stokes scattering. The shift to a lower wave length is called the anti-Stokes Raman scattering beam. A Raman scattering hydrogen detector measures the intensity of scattered laser beam signals in the narrow frequency and wave length bands corresponding to vibrational and/or rotational Raman scattering. Backscattering is usually used so that the detector and laser can be co-located.

Laboratory Raman scattering hydrogen detection systems have been described by Ball [7] and by Ninomiya et al. [8]. Both systems used Neodymium-doped Yytrium-Aluminum-Garnet (Nd:YAG) lasers as the light source. The Ninomiya et al. pulsed Nd:YAG laser was operated at the third harmonic wave length of 355 nm. Ball operated his Nd:YAG laser in both frequency-doubled and frequency-tripled configurations to generate wave lengths of 532 nm and 355 nm, respectively. The Raman scattering cross-section at 355 nm is seven times as large as the cross-section at 532 nm [7].

Ball's Raman scattering detection system is shown schematically in Figure 1. The laser produced a 5 cm diameter beam of variable pulsed energy. After filtering, the Raman scattered signal is collected on a spectrometer and photomultiplier tube and recorded on an oscilloscope. Descriptions of all components are provided in the Ball thesis [7]. Pre-2005 Raman scattering hydrogen detection studies by other researchers with other experimental configurations are also referenced and summarized by Ball [7].

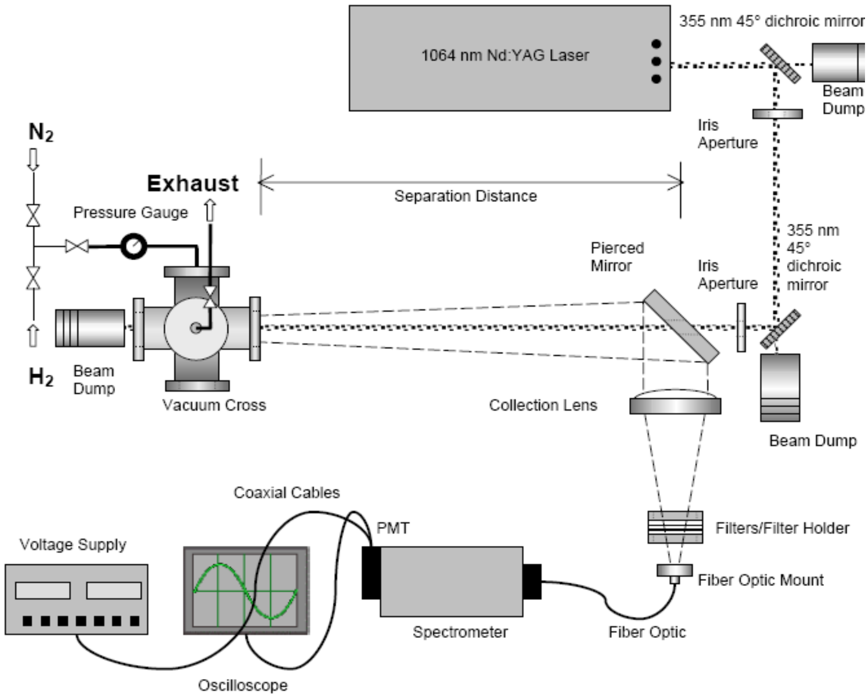

Figure 1. Laboratory Raman scattering apparatus used by Ball [7] for hydrogen detection.

The Ninomiya et al. [8] Shikoku Research Institute Raman scattering system is shown schematically in Figure 2. The 60 mJ, 20 Hz beam from the Nd:YAG laser is transmitted along the axis of a Newtonian telescope of aperture 212 mm. The primary and secondary mirrors of the telescope are UV coated, with reflectivity >90% from 270 nm to the visible region. Laser backscatter at all wave lengths is collected by the telescope, collimated, and split into two beams by a beam splitter. Each beam passes through a narrowband interference filter and is directed into a photomultiplier tube (PMT). In addition, a laser line edge filter is installed in front of the beam splitter to reject stray laser light, Rayleigh scattering, and Mie scattering.

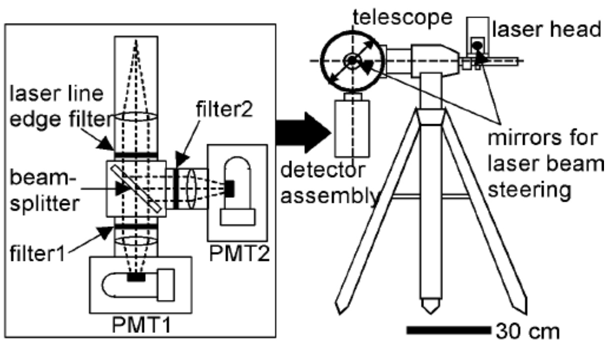

Figure 2. Raman scattering system used by Ninomiya et al. [8]

The hydrogen induced Raman shifts of the 355 nm incident wavelength produce a vibrational scattered Stokes wavelength of 416 nm and a rotational scattered Stokes wavelength of 362 nm [8]. The narrowband filters in the Ninomiya et al. system allow detection of both backscattered signals. Ninomiya et al. also state that their narrowband filters can also monitor spurious signals at neighboring wavelengths that may be generated by broadband fluorescence, so that the spurious signals can be discriminated from the Raman backscattered signal induced by hydrogen.

Ball [7] conducted hydrogen detection laboratory experiments using laser-mirror separation distances of 6.6 m and 2.3 m and at both laser wave lengths. The hydrogen-scattered signal using the 532 nm wave length was much weaker than from the 355 nm wave length source beam, but both sources produced linear correlations of scattered signal strength with hydrogen concentration in hydrogen-nitrogen mixtures. The lower hydrogen concentration limit of detection depended on laser pulse energy and collection lens diameter. The Raman scattered signal strength for a given hydrogen concentration and distance from the source varied linearly with the product of the pulse energy and the square of the lens diameter. Lower limit hydrogen concentrations (as determined by statistically significant deviations from the signal linear correlation) reported by Ball [7] for his signal filtering are shown in Table 1.

Table 1. Lower hydrogen concentration detection limits obtained by Ball [7]

Laser Energy Pulse (mJ/pulse)	Lens Diameter (cm)	Lower H_2 Detection Limit (v%)
125	10	1.94
250	10	0.76
125	5	3.12

Ninomiya et al. [8] have developed their Raman scattering detection system sufficiently to conduct experiments in outdoor conditions using the setup shown in Figure 3. As indicated in the drawing, hydrogen was released from a nozzle situated at a distance of 10 to 50 m from the laser and signal processing instrumentation. Hydrogen flow rates from the 2 mm orifice nozzle were in the range 10 to 50 lpm in the tests reported in reference 8. The laser beam was about 1 cm in diameter at 10 m and about 3 cm at a distance of 30 m. The beam was aligned so that it passed 3-5 cm above the nozzle orifice.

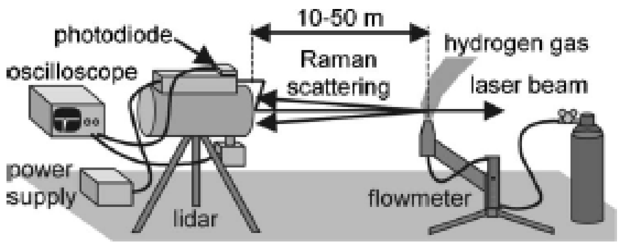

Figure 3. Ninomiya et al. [8] hydrogen detection field experimental arrangement.

The Raman backscatter signal-to-noise ratio decreased with distance from the detection system, such that the ratio was greater than 50 at a distance of 15 m, and greater than 5 at a distance of 50 m. Since the telescope was focused at a distance of 30 m, the signal strength was greatest at that distance. There was only minor variation of the signal strength with hydrogen flow rate at the beam location near the nozzle.

Ninomiya et al. [8] also conducted two-dimensional imaging experiments by installing a beam scanner between the laser head and the mirror located on the telescope axis. The scanner was operated with 10 horizontal positions and 10 vertical positions, with a complete scan every 30 seconds. Scattered beam detection signals above a nominal threshold were recorded on the image from a visual camera so that the two-dimensional image of the hydrogen jet could be superimposed on the image of the nozzle. Figure 4 shows one such image of the detected jet above the nozzle/burner.

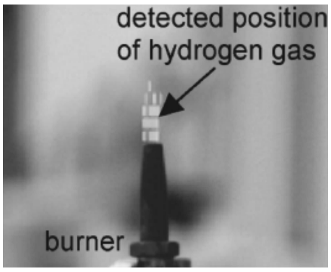

Figure 4. Hydrogen jet image from Ninomiya et al. Raman scattering demonstration [8].

Both the Ball [7] and Ninomiya et al. [8] results reported to date indicate that Raman scattering provides potential for wide area monitoring, but concerns remain about low hydrogen concentration detection without using powerful lasers. There are also concerns about effects of signal noise due to fluorescence from equipment and structures in congested facilities as well as personnel safety issues associated with deployment in occupied areas. Both the University of Florida and the Shikoku Research Institute are pursuing further development of their systems. The University of Florida has also been pursuing laser induced breakdown spectroscopy for hydrogen detection [9]. However, since laser induced breakdown spectroscopy involves beam focusing on a single spot and sufficiently high energies to generate plasma, it represents a potential ignition source and is not suitable for wide area detection.

3.0 DISTRIBUTED FIBER OPTIC SENSORS

A large number of organizations have been working on various technologies for fiber optic detection of hydrogen. Most of this work so far has involved spot detection of hydrogen with the actual sensor located at the end of the fiber. Although these sensors can be deployed in some type of multiplexed network with remote or central signal processing, a large number of sensors/fibers would be needed in large unenclosed areas, and the problems of uncertain detector location would still be applicable. The types of fiber optic sensors that are inherently more applicable to the hydrogen detection needs addressed in this paper have hydrogen sensing capability over all or multiple segments of the fiber optic cable. Thus each fiber optic cable provides for distributed linear hydrogen detection.

Intelligent Optical Systems (IOS) has been developing fiber optic multiplexed spot and distributed sensors for hydrogen detection. The IOS hydrogen sensing is associated with hydrogen reversible reaction with tungsten oxide and a palladium catalyst, resulting in color changes to the oxide compound and associated changes in optical properties. The basic sensor design and operation with the color changing mixture applied to a porous glass rod at the end of a double fiber is described in a 2003 U.S. patent [10], and the generic design approach for distributed fiber optic sensing is described in a 2006 U.S. patent [11].

Figure 5 shows a schematic of the IOS distributed sensor fiber design. The fiber has a fused silica core, but instead of glass cladding there are polymer cladding layers that serve multiple purposes: 1) the polymer cladding layer has a low refractive index to minimize loss as the light travels through the fiber core; 2) the polymer is gas-permeable to allow hydrogen to diffuse into the polymer matrix upon exposure; and 3) the polymer matrix contains tungsten oxide plus palladium chemistry formulations that are designed to respond and react to hydrogen upon exposure.

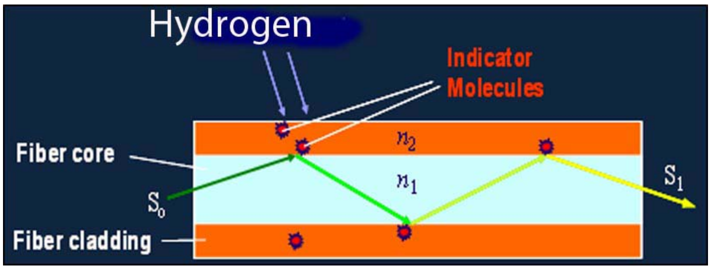

Figure 5. IOS distributed sensor fiber design schematic [12]

The light guiding properties of the IOS optical fiber are altered when hydrogen reacts with the embedded chemical indicator. The change in the transmission properties of the light received at the distal end of the fiber is detected using signal processing algorithms designed to maximize sensitivity and minimize false alarms.

Fiber response to hydrogen exposure is a function of both hydrogen concentration and amount of exposed fiber. For example, to a first order, a longer IOS fiber, exposed to a low concentration of analyte, will respond similarly to a shorter fiber, exposed to a higher concentration. The response of 1 meter of exposed hydrogen sensing fiber (handmade prototype) over a range of hydrogen levels is shown in Figure 6. The step responses to hydrogen concentrations starting at 4% hydrogen and decreasing to 0.5% are shown at different optical wavelengths. Current response times are on the order of several seconds.

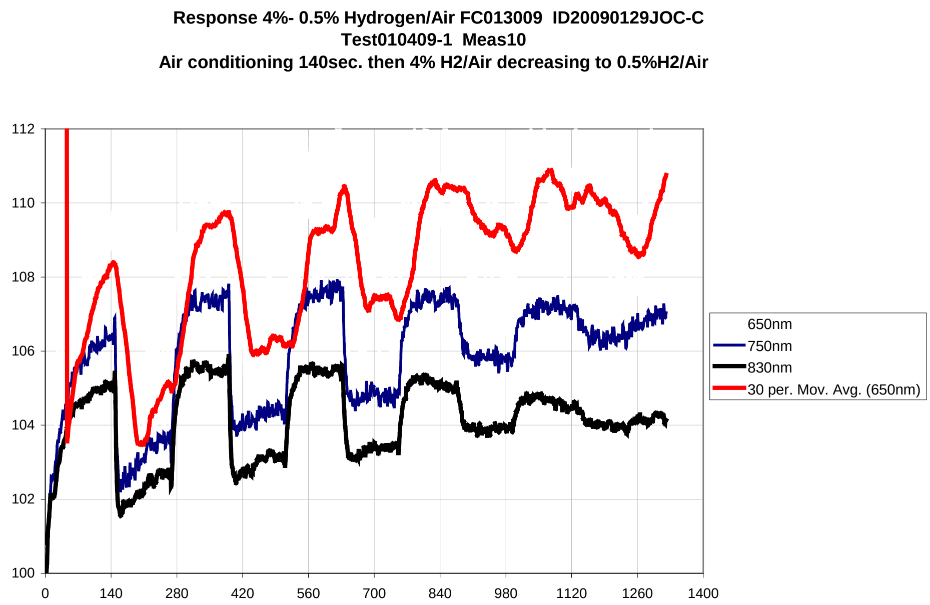

Figure 6. Response (% transmittance vs time) of IOS distributed sensor cable to hydrogen [12]

Hydrogen detection sensitivity for this type of sensor is affected by humidity. Development work in recent years has focused on improved, humidity resistant, polymeric coatings for the fiber and simple, less expensive fabrication methods. Testing through fiscal year 2007 [13] showed that the sensor could respond at humidity levels up to 95%, albeit with weaker signals at the higher humidity.

Sumida et al. [14] developed a similar distributed fiber optic sensor using a platinum supported thin film of tungsten trioxide. The sensor has an optical signal attenuation response to hydrogen exposure that varies linearly with sensor length (at a rate of 62 dB/m) for lengths up to about 20 cm. The Sumida et al. multi-sensor optical fibers had sampling lengths of 15 cm in some tests, and 10 cm in other tests. The system tested with three 15 cm length sensors spliced into a long length of optical fiber is illustrated in Figure 7. The light source was a 1.3 μm pulsed LED operated with a 20 ns pulse. The back scattered light was recorded on an optical time domain reflectometer (OTDR).

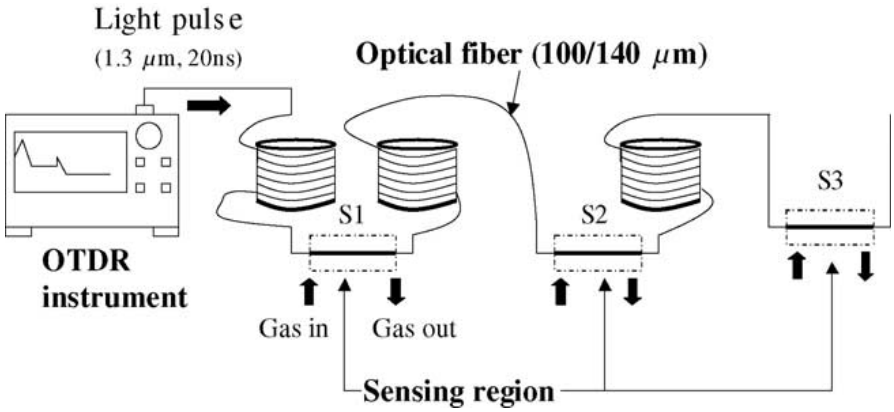

Figure 7. Three sensor optical fiber test setup of Sumida et al. [14].

The OTDR signal from the Sumida et al. sensor responding to 20 ns source pulses extended over an equivalent distance of 5 m. This suggests that spatial resolution with this arrangement is limited to a few meters, and spacing of the sensors in the optical fiber should be greater than 5 m. They reported good results with three 10 cm length sensors situated at distances of 200, 550, and 880 m from the light source and OTDR.

Bévenot et al. [15] have described a multi-sensor optical fiber hydrogen detector constructed with a thin palladium layer deposited on the bare core of multimode fiber. Their light source is a collimated beam with non-normal incidence on the input end of the fiber. The experimental arrangement for a fiber with two 20-cm length, 12 nm thick Pd layer sensors is shown in Figure 8. Successful responses to hydrogen concentrations as low as 0.8 v% were reported, with fiber lengths as long as 200 m.

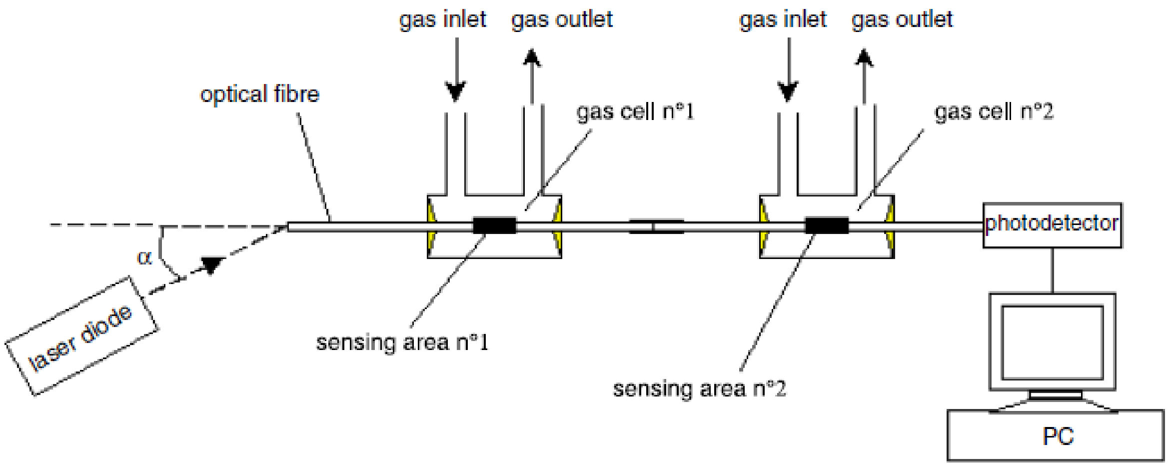

Figure 8. Double sensor optical fiber detection system of Bévenot et al. [15].

Although the distributed sensor optical fiber hydrogen detection systems described above have only been tested in the laboratory to date, IOS has developed similar distributed fiber sensor systems equipped with chemically treated fiber cladding for chemical agent detection, and has started deploying these prototype systems for field testing at an operating aircraft control center and a mass transit station. Further development of the IOS hydrogen distributed sensor continues with U.S. Department of Energy funding. Sumida et al. [14] also indicate that work is continuing on their distributed hydrogen sensor, with emphasis on extending the sensor length and optimizing the optical properties of the cladding and the refractive index profile of the fiber core.

4.0 ACOUSTIC SENSORS

Hydrogen jet releases associated with breaches in pressurized piping and valves produce ultrasonic turbulent pressure fluctuations as well as audible emissions. The peak intensities of these ultrasonic fluctuations usually occur around 40 kHz [4]. Acoustic gas detectors are designed to receive and process these ultrasonic signals, and to discriminate a sudden ultrasonic signal from the normal ultrasonic signals generated by process equipment.

Commercially available ultrasonic sensors are currently used for gas leak detection at petrochemical plants. Figure 9 shows one of these detectors installed above gas piping. According to the manufacturer’s literature, the detector has a detection radius for hydrogen up to 8 meters at a leak rate of 0.01 kg/s. The ultrasonic signal intensity depends on hydrogen pressure as well as leak rate or orifice equivalent diameter. Reported data [16] shows that the ultrasonic signal from a 1 mm orifice at 5.5 MPa, corresponding to a 0.003 kg/s release, decays almost linearly with distance from the source. At a distance of 8 m, the ultrasonic sound level was approximately 86 db.

Figure 9. Ultrasonic leak detector above gas piping (from Gassonic) [16].

Portable ultrasonic leak detectors have also been utilized to locate the source of hydrogen leaks. NASA has used both commercial and specially fabricated prototype portable ultrasonic detectors with mixed success in finding the source of fuel leaks. A 1990 success story locating a relatively small main propulsion system leak at a Kennedy Space Center rocket launch site is described in a NASA report [17]. The prototype detector in that case was a 40 kHz piezo transducer installed at the focus of a sound collecting horn, and a special processing module to transform the ultrasonic signal into the audio frequency range.

5.0 IMAGING SYSTEMS

Several different types of imaging systems are at least conceptually applicable to wide area hydrogen leak detection. Schlieren systems and shearing interferometry systems for this application are described briefly in [4]. The schlieren systems utilize optical beam refractions associated with gas density gradients produced when hydrogen is released into air. Shearography systems produce images from the interference patterns produced by two laser beams that traverse paths with different indices of refraction and path lengths.

Fraunhofer ICT [18] has developed a background oriented schlieren (BOS) system for hydrogen release visualization research. The Fraunhofer BOS system focuses a digital camera on a background pattern of distributed dots in the field of view. As density gradients develop in this field of view, the camera records virtual displacements of the dots. Special software is used to calculate displacements for the entire field of view based on the displacements of the background dots. Fraunhofer used their BOS system to produce colorful high-speed images of the hydrogen release and combustion products generated during the explosion of a hydrogen-air mixture in and around a 1 m^3 enclosure. Although their system is not designed for monitoring hydrogen facilities, it may be possible to extend this technique for such an application.

Common path shearing interferometry uses a polarized collimated laser beam and a Wollaston prism in combination with an analyzer to generate a camera image of a field with density (index of refraction) variations. Ambrosini et al. [19] have recently reviewed the state of the art of shearing interferometry technology and other imaging techniques for flow visualization and diffusion measurements. They classify common path shearing interferometry as one of the most robust methods with relatively small-to-moderate expense and easy construction. Of course, the applications they were envisioning were laboratory experiments as opposed to wide area hydrogen detection. Nevertheless, this method has some potential for the applications envisioned in this paper.

If the various imaging methods are going to be used for hydrogen detection as well as hydrogen release imaging, they will need to be used with imaging analysis software. This type of software is being developed intensively and being commercialized for applications such as smoke and fire detection, so that it should not be difficult to extend it to hydrogen leak detection and alarm.

6.0 COMPARISON OF VARIOUS SENSOR TECHNIQUES

The hydrogen detection technologies discussed above have different capabilities in terms of sensing path length, low concentration detection limit, response time, and system monitoring area. The latter cannot be characterized yet since most of the systems are still under development. The first three parameters can be roughly characterized based on the results reported in the references cited here. Table 2 provides such a comparison with references and also lists the development status of the technologies. The last technology listed in Table 2 is a network of hydrogen spot sensors, such as the systems described in references [20-22].

Table 2. Detection system parameter comparison

Technology	Demonstrated Sensing Length (m)	Lowest H2 Concentration Detected (v%)	Best Response Time (s)	Development Status
Open Path Raman Scattering	50 [8]	0.8 [7]	< 1	Outdoor Demonstration [8]
Distributed Optical Fiber	Sensor: 1 [12] Fiber Length: 880 [14]	0.5 [12]	2 – 5 [12]	Laboratory System
Ultrasonic	8 [16]	NA	< 1	Commercially Available
Imaging	≈ 4 [18]	1 [23]	< 1	Components Only
Networked Spot Sensors	≈ 0.1	< 0.05 [20, 24, 25]	Varies	Field Demonstrated [21, 24]

The demonstrated sensing length listings in Table 2 indicate that the open path Raman scattering technology has demonstrated the longest sensing length to date, but the distributed optical fiber has been demonstrated with limited sensor lengths spaced over long fiber lengths. This suggests that the open path Raman scattering has the potential for installation over continuous hydrogen piping and facility boundaries, whereas the distributed optical fiber has the potential for installation with the sensors situated on and around flanges, valves, and other prospective leak sites. Ultrasonic sensors are also typically situated near potential leak sites, but at a standoff distance rather than in contact with the piping components themselves.

Networked spot sensors provide the best capability for detecting low hydrogen concentrations providing at least one of the sensors is situated within about 10 cm of the outer perimeter of the hydrogen-air plume, jet or ceiling layer. Several types of spot detectors have been shown to respond clearly to concentrations that are an order-of-magnitude lower than the lowest concentrations detected by optical wide area and distributed sensors. Response times of spot detectors vary with the type of sensor. Networked spot sensors are also the most developed technology with the possible exception of ultrasonic sensors. Imaging technologies are clearly the least developed for hydrogen detection, but they offer the advantage of showing an image to allow location of the hydrogen release as well as its presence.

Some of the other factors that influence the relative performance and reliability of various sensor technologies are cross-sensitivity to other gases, contamination vulnerability, and operability at different temperatures and humidity levels. The optical fiber and open path Raman scattering technologies are now being tested for these effects and it is too early to tell if there will be any significant limitations in this regard. The ultrasonic sensors would also respond to other gas jet releases other than hydrogen. Several different types of spot sensors have significant drawbacks in these areas as categorized by Boon-Brett et al. [25].

The comparisons shown in Table 2 represent a snapshot based on information available to the authors. Several of the sensor technologies are rapidly developing (open path Raman scattering, distributed optical fibers and imaging), while others seem to be quite mature. Therefore, it is too early to speculate about the long term competitiveness of the various wide area hydrogen sensing technologies. It is also possible that another technology not discussed here, such as carbon nanotubes, may also become a viable candidate for future applications.

7.0 CONCLUSION

The wide area hydrogen sensor technologies described in this paper offer the potential to provide detection for facilities where spot detectors are not feasible because of the difficulty of providing complete coverage and appropriate spot sensor locations. Pursuing the development of these detectors, and in particular the Raman Scattering and Distributed Fiber Optic Sensors, because these technologies can potentially cover very large areas, could ultimately help hydrogen facilities better satisfy or exceed the detection requirements of consensus safety codes and government regulations.

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