

Free-space laser interrogation of Tantalum-Palladium Hydrogen sensor

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Summary: A free-space laser interrogation scheme of Hydrogen sensitive $Ta_{1-y}Pd_y$ thin film layers is presented. We focus on the design of the optical setup, by isolating systematically the potential sources of error and improving signal-to-noise ratio as well as stability of the signal, with the goal of optimizing remote Hydrogen sensors for industrial applications.

Keywords: Hydrogen sensing, hydride, laser, reflectivity, resistivity, Tantalum-Palladium

Background and Motivation

The rapid expansion of Hydrogen technologies across energy, industrial, and power-system applications has intensified the demand for reliable H_2 leak detection. Hydrogen is, however, intrinsically difficult to sense by conventional laser absorption methods because the H_2 molecule lacks strong vibrational-rotational absorption bands in the infrared and ultraviolet spectral regions. As a result, established free-space spectroscopic techniques such as TDLAS or open-path FTIR—successfully deployed for gases like methane or ammonia—are fundamentally unsuited for direct Hydrogen detection. Consequently, most optical Hydrogen sensors rely on interrogation of Hydrogen-sensitive materials, rather than on direct optical absorption by the gas itself.

Among these approaches, fiber-optic interrogation of Palladium-based sensing elements currently exhibits the highest technology readiness. Palladium and its alloys reversibly absorb H_2 to form hydrides, inducing changes in lattice constant, mechanical strain, and optical properties. Pd sensors can have issues with stability (microcracking, delamination), modest sensing range (difficult to detect low H_2 concentration) and intrinsic hysteresis. Alloying reduces this problem, but at the price of a reduced sensitivity. In fiber-optic Hydrogen sensors, these changes are transduced into intensity, phase, or wavelength shifts that can be read remotely through optical fibers. Reviews indicate that such systems have progressed from laboratory demonstrations to extended field trials and niche deployments, corresponding to TRL 4–6, depending on architecture and application.[1]

Fiber-based schemes benefit from intrinsic electrical isolation, immunity to electromagnetic interference, and compatibility with distributed

sensing concepts. Representative implementations include Fabry–Pérot cavities formed by suspended palladium membranes at fiber tips, where Hydrogen-induced membrane deformation results in measurable interference shifts with ppm-level sensitivity, or direct measurement of optical reflectance at fiber tip (so-called micro-mirror configuration). In parallel, free-space laser interrogation, typically based on optical reflectometry or scattering from palladium-coated surfaces, represents a less mature but conceptually simpler alternative. In these schemes, a laser probes a Hydrogen-sensitive film directly, and Hydrogen exposure is inferred from changes in reflectance caused by Hydrogen-induced deformation or modified optical constants. While attractive for their simplicity and potential low cost, free-space interrogation concepts remain largely confined to laboratory environments and early validation experiments, corresponding to TRL 2–3. Their sensitivity to alignment, environmental contamination, and line-of-sight constraints poses significant barriers to deployment in industrial environment.[2]

Recently it was demonstrated that thin film $Ta_{1-y}Pd_y$ with high Ta content is an optical sensing material with extremely wide sensing ranges covering at least 7 orders of magnitude in Hydrogen pressure.[3] Nanoconfinement of the $Ta_{1-y}Pd_y$ layer suppresses the first-order phase transitions present in bulk and ensures a sensing response free of any hysteresis. It therefore represents a more appropriate alternative to pure palladium or high-content Pd thin film for Hydrogen sensing in a wide range of concentrations, also as a fiber optic sensor.[4]

Objective

In this contribution, we explore the potential of a free-space laser interrogation scheme of

Hydrogen sensitive $Ta_{1-y}Pd_y$ thin film layers. We focus on the design of the optical setup, with the goal of isolating the potential sources of error and improving signal-to-noise ratio as well as stability of the signal. Figure 1 shows the bench-top optical setup used to measure single-wavelength optical reflectance at 976nm. Besides the direct reflection path from the laser reflected into Ta-Pd sample (8) into the main detector (6), we divert a part of the light intensity into reference detector (7). Additionally, another part of the light is coupled after reflection on the Hydrogen sensitive layer to a camera (5).

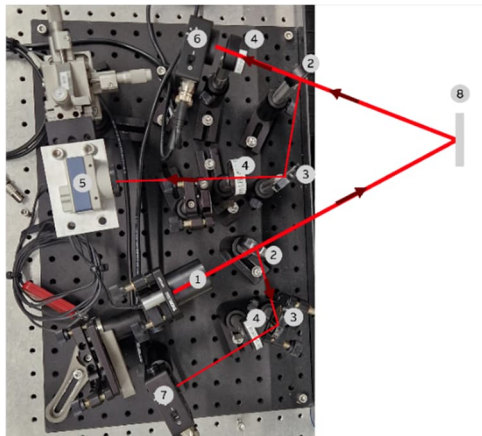


Figure 1. Initial optical setup: 1. IR Laser + neutral density filter: class 1 output at 976 nm (< 0.4 mW) (depicted: Thorlabs CPS980; a temperature-stabilized (with LDM9LP) LP980-SF15 fiber laser is used in a second stage. 2. Beamsplitters, 3. Mirrors, 4. Focus lenses 5. Teledyne Dalsa G3-GC10-C1280IF Genie Nano 1GigE Camera. 6. Main detector. 7. Reference detector. 8. Ta-Pd Sample.

Results

Figure 2 shows a comparison between the optical reflectance (top) and the electrical 4-point resistance (bottom) for 50, 100, 250, 500, 1000 and 10000 ppm in a nitrogen background for $Ta_{88}Pd_{12}$ Hydrogen sensing layers. At $2.3 \cdot 10^5$ s, the laser has been switched off and on again, resulting in an intensity change that can be compensated with the reference signal (see inset). Between the cycles of Hydrogen application, samples are flushed with technical air or Nitrogen. Both readout schemes allow an accurate measurement of the hydrogen concentration in this range. The sensor response is slightly larger in the Nitrogen flushing case, indicating that presence of Oxygen in the environment reduces durably H_2 sensitivity.

Conclusion and Outlook

This free-space laser interrogation readout for Hydrogen sensing is valuable to test and improve remote H_2 sensing readout schemes. The simultaneous 4-point measurement of electrical resistance provides a local reference and

enables isolating various sources of error in the optical readout scheme. Next step is to add laser beam tracking by camera (5) to compensate for misalignments inherent to remote optical interrogation.

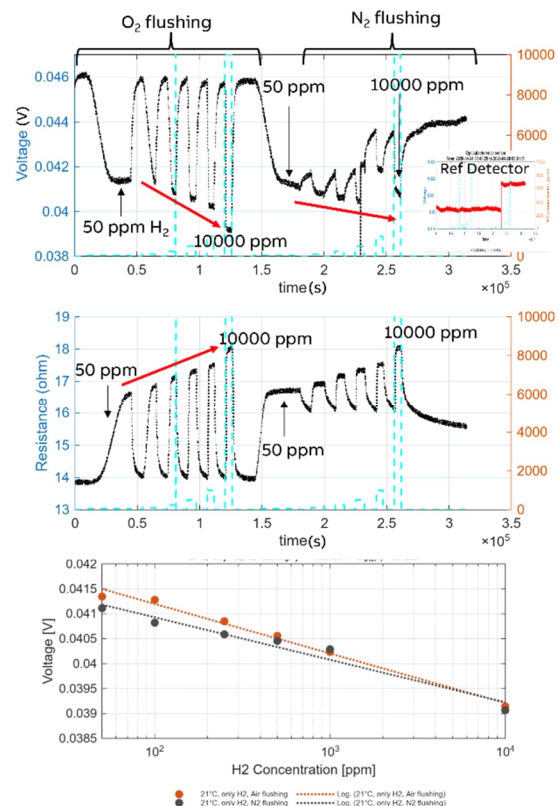


Figure 2. $Ta_{88}Pd_{12}$ response to various H_2 concentration in N_2 . Top: 4-point resistance, Middle: reflectance. First series: flushing with technical air (21% O_2 in N_2), second series: flushing with N_2 . Bottom: corrected optical detector voltage as function of H_2 concentration.

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References

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