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Article

Hysteresis-Free Near-Infrared Optical Hydrogen Sensor Based on Ti/Pd Bilayer Thin Films

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Abstract

Palladium (Pd) and titanium (Ti) exhibit opposite dielectric responses upon hydrogenation, with stronger effects observed in the near-infrared (NIR) region. Leveraging this contrast, we investigate Ti/Pd bilayer thin films as a platform for NIR hydrogen sensing—particularly at telecommunication-relevant wavelengths, where such devices remain largely unexplored. Ti/Pd bilayers, coated with Teflon AF (TAF) and fabricated via sequential electron-beam evaporation, were characterized using optical transmission measurements under repeated hydrogenation cycles. The Ti (5 nm) / Pd ($x = 2.5$ nm) / TAF (30 nm) architecture shows a 2.7 times enhancement in hydrogen-induced optical contrast at 1550 nm compared to Pd/TAF reference films, attributed to hydrogen ion exchange between the Ti and Pd layers. The optimized structure, with Pd thickness of $x = 1.9$ nm, exhibits hysteresis-free sensing behavior, a rapid response time ($t_{90} < 0.35$ s at 4% H_2), and a detection limit below 10 ppm. It also demonstrates excellent selectivity with negligible cross-sensitivity to CO_2 , CH_4 , and CO, as well as high durability, showing less than 6% signal degradation over 135 hydrogenation cycles. These findings establish a scalable, room-temperature NIR hydrogen sensing platform with strong potential for deployment in automotive, environmental, and industrial applications.

Keywords: optical hydrogen sensor; near-infrared (NIR) sensing; titanium; palladium; bilayer; thin film

1. Introduction

Hydrogen is a leading candidate for clean energy applications due to its high energy density, diverse production methods, and zero-emission combustion [1–4]. However, its flammability, wide explosive range, and lack of color or odor pose serious safety challenges, particularly during storage, transport, and end-use [4,5]. These risks underscore the need for compact, fast, and highly sensitive hydrogen sensors that operate reliably under ambient conditions [3,6].

Optical hydrogen sensors are well-suited for these demands, offering intrinsic safety by eliminating electrical components from the sensing region [7–9]. These devices detect hydrogen through reversible changes in the optical properties - typically transmittance or reflectance [10–12] - of hydrogen-responsive metal films [4]. Palladium (Pd) is widely used in such sensors due to its strong and reversible optical response at room temperature [13]. However, Pd-based systems often suffer from drawbacks such as hysteresis, slow kinetics near the $\alpha - \beta$ phase transition, and performance degradation over time due to cracking and delamination under cyclic stress [8,9,14,15].

To address these limitations, researchers have explored alloying Pd with other metals (e.g., Au, Ag, Co, Cu) and incorporating nanoscale features such as nanoholes, nanopatches or nanowires [9,16–20]. While effective, these strategies often require complex fabrication processes that limit scalability and increase cost. Alternatively, integrating Pd with another hydrogen-active material in

a bilayer configuration might offer a simpler and more scalable approach to enhance performance by leveraging complementary material properties. For example, Boelsma et al. achieved hysteresis-free sensing using Pd–Hf bilayers, although their devices required elevated temperatures due to slow hydrogen diffusion at room temperature [21].

Titanium (Ti) is a promising candidate for such bilayer designs. It forms stable hydrides, exhibits excellent mechanical stability, and adheres well to Pd- reducing film delamination [15,22–24]. More importantly, Ti and Pd exhibit opposing dielectric responses to hydrogenation [22], which are particularly pronounced in the near-infrared (NIR) range. This contrast opens the possibility of enhanced optical response through dielectric engineering. Despite this potential, Ti/Pd bilayers remain largely unexplored for hydrogen sensing at telecom wavelengths near 1550 nm, a spectral region attractive for fiber-optic integration due to its low loss and wide commercial infrastructure [25–27].

In this work, we introduce a NIR optical hydrogen sensor based on a Ti/Pd bilayer capped with a gas-permeable polymer, Teflon AF (TAF), known to enhance hydrogen transport [28]. By optimizing the sensor (5 nm Ti / 1.9 nm Pd / 30 nm TAF), we achieve hysteresis-free sensing, sub-second response times ($t_{90}(s) < 0.35$ s) at the lower flammability limit (4% H_2), detection limits below 10 ppm, and excellent long-term stability. The sensor also exhibits negligible cross-sensitivity to common interfering gases such as CO_2 , CH_4 and CO. These results establish the potential of Ti/Pd/TAF stacked films as a scalable, fiber-compatible, and high-performance platform for hydrogen sensing in safety and environmental monitoring systems applications.

2. Materials and Methods

2.1. Materials

High-purity palladium (Pd, 99.95 %), titanium (Ti, 99.99 %) were purchased from Kurt. J Lesker Company and used to fabricate bilayer sensors. Teflon AF 2400 (TAF) obtained from Dupont was used for polymer coating. Polymethyl methacrylate (PMMA), acetone ($\geq 99.5\%$), and 2-propanol ($\geq 99.9\%$) were purchased from Sigma-Aldrich. Deionized (DI) water (18 M Ω cm) was used for all cleaning processes.

2.2. Thin Film Deposition

Thin films were deposited in a custom-built electron beam evaporation chamber (Pascal Technology) under a base pressure of 1×10^{-6} Torr. Glass substrates (1 cm \times 1 cm) were cleaned using a sequential ultrasonic bath in acetone, 2-propanol, and DI water, each for 15 minutes, then dried under a nitrogen stream. A 5 nm Ti layer was deposited first, followed by a Pd layer with nominal thicknesses ranging from 1.9 to 2.5 nm. During deposition, the substrate was continuously rotated at 30 rpm to ensure uniform coverage. The deposition rate was maintained between 0.1 and 0.15 Å/s and monitored via a quartz crystal microbalance (QCM).

2.3. Teflon AF and PMMA deposition

After metal deposition, a uniform ~ 30 nm thick layer of Teflon AF 2400 (TAF) was applied using a Luxel vacuum evaporation organic furnace, at a pressure below $< 1 \times 10^{-6}$ mbar. For PMMA coating, a 10 mg/mL PMMA–acetone solution was spin-coated onto the bilayer sensor at 5000 rpm for 120 seconds, forming a uniform ~ 100 nm-thick film [29]. The coating thickness was selected based on prior studies, with the goal of providing complete coverage of the Pd surface without significantly hindering hydrogen diffusion [28–30]. TAF coating improves sensor performance by reducing the apparent activation energy for hydrogen absorption and desorption through interfacial chemical and electronic modification, while the PMMA layer improves selectivity by limiting the permeation of interfering gases such as water and CO [28,29].

2.4. Structural and Morphological Characterization

Surface morphology was characterized by scanning electron microscopy (SEM) using an ultra-high-resolution SEM (SU-9000, Hitachi) with a resolution of 0.4 nm at 30 kV. X-ray diffraction (XRD) measurements were performed using a powder diffractometer (XRD, Rigaku) with Cu-K α radiation ($\lambda = 0.154$ nm) to investigate the crystallographic structure of the films.

2.4. Optical Sensing and Measurement Setup

Hydrogen sensing performance was evaluated using a custom-built optical setup [31]. The sensors were placed inside a custom fabricated vacuum chamber equipped with dual quartz optical windows for in situ optical measurements during hydrogen exposure. The absolute pressure in the gas cell was monitored using three calibrated pressure transducers: two PX409-USBH transducers (Omega Engineering) and a Baratron gauge (MKS Instruments), enabling accurate readings from $\sim 2.7 \times 10^{-6}$ up to 1.1 bar. By using diluted 4% H₂ or 100 ppm H₂ balanced in N₂, we can control the partial H₂ pressure as low as ~ 4 ppm or ~ 100 ppb. In flow mode measurement, 4% H₂ in nitrogen mixture gases (Airgas) were diluted with ultra-high purity nitrogen gas from Airgas company to targeted concentrations by a commercial gas blender (GB-103, MCQ Instruments). The gas flow rate was kept constant at 400 ml/min for all measurements. An unpolarized, collimated halogen light source (HL-2000, Ocean Optics) was used to illuminate the sample at normal incidence. Transmitted light was collected into an optical fiber and directed to two spectrometers USB4000-VIS-NIR-ES (Ocean Optics, 400 – 1000 nm) and DWARF-STAR (StellarNet, 900 – 1650 nm), to record the transmission spectra across the visible and near-infrared regions. All the measurements were conducted at room temperature.

2.5. Finite-difference Time-domain Calculations

Finite-difference time-domain (FDTD) simulations was used to calculate the optical response of Pd and Ti/Pd bilayer samples using commercial software (Ansys Academic Lumerical FDTD research). The geometric parameters of Pd nanoparticles were obtained from SEM images and imported into the software. The mesh size of $2 \times 2 \times 2$ nm was chosen. The refractive indices of the glass and TAF was chosen to be 1.5 and 1.27 respectively. The optical parameters of Pd, PdH_x, Ti, and TiH_x were obtained from previously reported experimental data [22]

3. Results and Discussion

3.1. Film Morphology and Structure

The surface morphology of the deposited films (Pd only and Ti/Pd bilayer) was investigated using high-resolution SEM to understand the effect of the Ti underlayer on Pd film growth. The 2.5 nm Pd-only film directly deposited on a glass substrate (Figure 1a) exhibited a discontinuous granular morphology, characterized by isolated sub-10 nm particles shown in Figure 1a inset. In contrast, when a similar thickness of Pd was deposited on a 5 nm Ti layer, the film exhibited significantly improved continuity, with evidence of particle coalescence (Figure 1b). This morphological transition suggests enhanced wetting and surface diffusion enabled by the Ti adhesion layer, which likely reduces nucleation barriers and promotes lateral film growth. These findings are strongly supported by the work of Verma et al. [23], who demonstrated that the surface topography of the Ti underlayer can control Pd grain morphology, leading to smoother, more continuous films with enhanced columnar structure and reduced void formation.

X-ray diffraction (XRD) spectra (Figures 1c & 1d) of the 5 nm Ti-only and 5 nm Ti / 2.5 nm Pd bilayer films show broad, featureless patterns, indicating a lack of long-range crystalline order within the detection limits. The absence of additional peaks in the bilayer suggests that the Pd overlayer does not significantly modify the structural phase of the Ti layer. Such behavior has been observed in prior studies of ultrathin metal films, where limited adatom mobility at room temperature during vapor-phase growth promotes island-like or discontinuous morphology, leading to nanoscale grains

or amorphous structures that fall below the detection threshold of conventional XRD techniques [32,33].

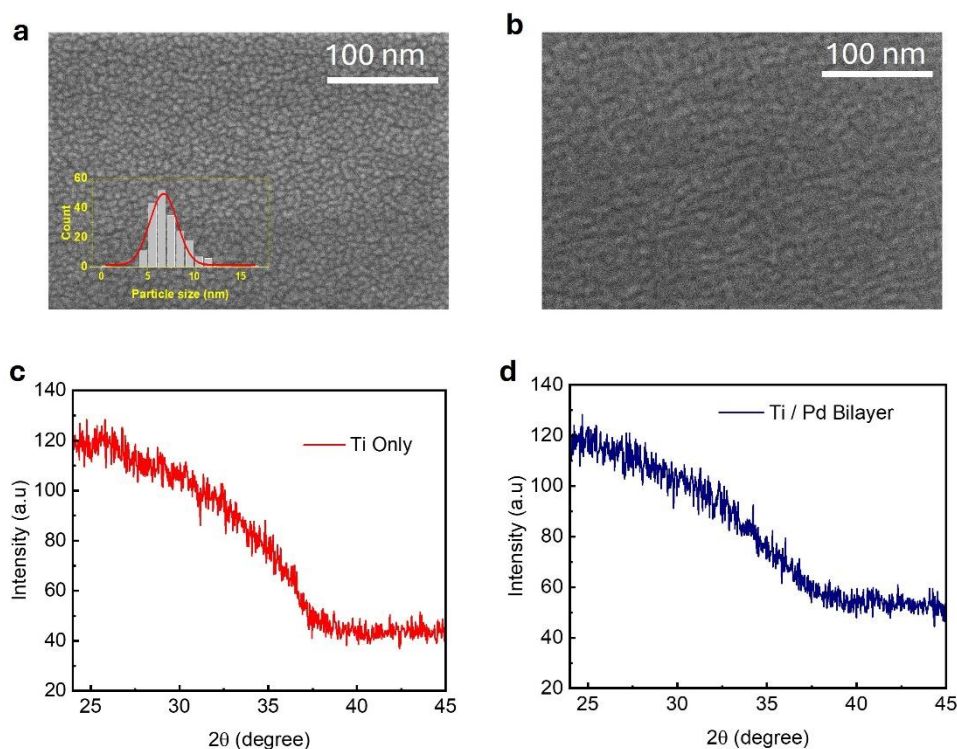


Figure 1. Scanning electron microscopy (SEM) images of (a) 2.5 nm Pd thin film with estimated particle sizes shown in the inset, and (b) 5 nm Ti / 2.5 nm Pd bilayer. X-ray diffraction (XRD) spectra of (c) 5 nm Ti film, and (d) 5 nm Ti / 2.5 nm Pd bilayer film.

3.2. Effect of Ti Underlayer in Pd/Ti Bilayer Films

3.2.1. Enhanced NIR Optical Response

The Ti/Pd bilayer exhibits a lower baseline transmission (T) than that of the Pd only reference film across the 400 – 1600 nm wavelength range due to additional absorption by the Ti layer (Figure 2a). This trend is also captured by FDTD simulations, which closely match the experimental results (Figure S1). Upon hydrogen exposure, both films show increased optical transmission, consistent with earlier Pd-based systems, however, earlier studies were limited to the wavelength shorter than 1000 nm [10,11,31]. The change in transmission intensity at $P_{H_2} = 1000$ and 0 mbar, defined as $\Delta T(\lambda) = T_{1000} - T_0$, is attributed to hydrogen induced changes in the electronic structure of the metals. Hydrogenation introduces extra electrons, raising the Fermi level and modifying the dielectric function. Palm et al. showed that this leads to the real and imaginary parts of the dielectric constant in Pd (Ti) decreases (increases), especially in the NIR – resulting in an increase (decrease) in optical transmission [34–37].

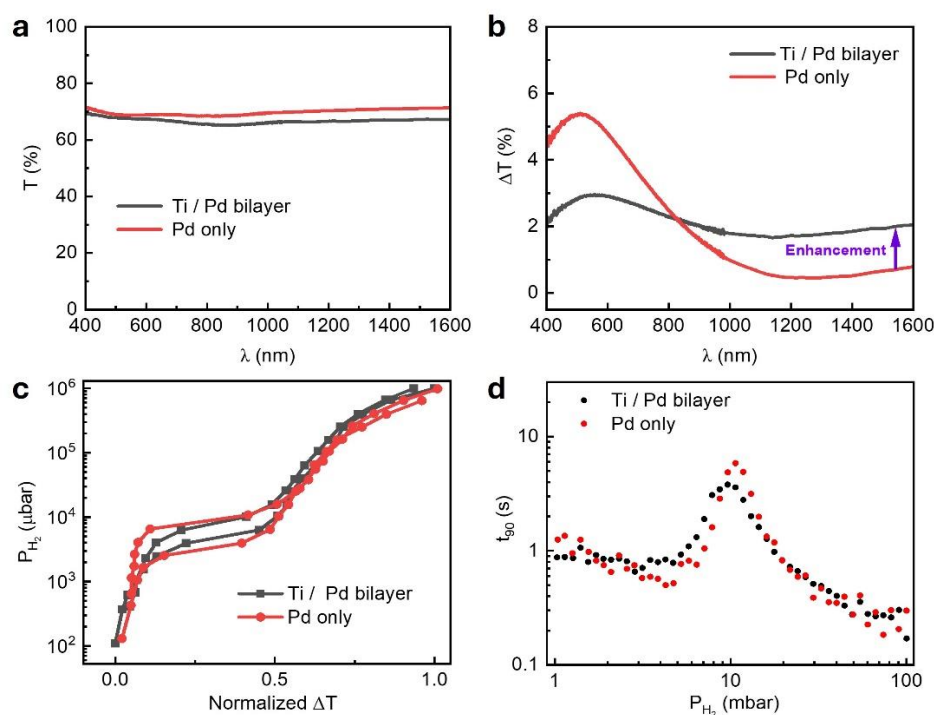


Figure 2. Optical transmission and sensing performance of 2.5 nm Pd / 30 nm TAF thin films with and without 5 nm Ti underlayer. (a) Experimental optical transmission spectra $T(\lambda)$ in vacuum (0 mbar H_2). (b) Hydrogen-induced change in transmission, $\Delta T(\lambda) = T_{1000 \text{ mbar}} - T_{0 \text{ mbar}}$, across 400 – 1600 nm spectral range. (c) Normalized ΔT sorption isotherms at $\lambda = 1550$ nm. (d) Response time (t_{90}) under hydrogen pressures ranging from 1 - 100 mbar.

The 2.5 nm Pd / 30 nm TAF film exhibits a strong ΔT centered around 550 – 600 nm but the response decreases rapidly with increasing wavelength, dropping below 1% beyond 1000 nm (Figure 2b)—following a trend previously reported in Pd films [11]. In contrast, the 5 nm Ti / 2.5 nm Pd / 30 nm TAF structure maintains $\Delta T \sim 2\%$ across the full spectral range. This behavior is further supported by FDTD simulations and a theoretical transmission calculation for an absorbing film (hydrogenated Pd), described by the equation [10,11]:

$$T = \frac{16 n_s (n^2 + k^2) e^{-\frac{4\pi k d}{\lambda}}}{[(n + n_s)^2 + k^2][(1 + n)^2 + k^2]} \quad (1)$$

where n and k are the effective refractive index and extinction coefficient of Ti/Pd bilayer calculated from (Figure S2a), d is the film thickness, λ is the wavelength, and n_s is the refractive index of the substrate. Both the simulated and calculated spectra capture the overall spectral trend and key features of the experimental response, although some deviations are observed in the magnitude and positions of peaks and dips (Figure S2b and S2c). Notably, at $\lambda = 1550$ nm, the calculated ΔT of 2.07% closely matches the experimental value of 2.05%. This corresponds to a 2.7-fold enhancement over the reference film ($\Delta T = 0.76\%$) and underscores the advantage of Ti/Pd for near-infrared sensing. This enhancement is particularly significant for sensing near 1550 nm - a telecom-standard wavelength offering low-loss optical propagation and compatibility with photonic integrated circuits (PICs). Previous studies have demonstrated sub-ppm sensitivity for gases such as NH_3 and CH_4 in this spectral region using PICs [27]. Our Ti/Pd/TAF sensor design extends this capability to hydrogen detection, offering a scalable and fiber-compatible optical sensing platform.

3.2.2. Reduced Hysteresis in the Optical Sorption Isotherm

Hydrogen isotherms were obtained by monitoring optical transmission at $\lambda = 1550$ nm during cyclic hydrogen exposure. While the data is presented at 1550 nm, plateau pressures can be extracted at any wavelength across the spectrum [9] (see also Figure S3a), their plateau pressures are essentially the same (Figure S3b), and their response time are identical (Figure S3c). The Ti/Pd/TAF structure exhibits a narrower hysteresis width—defined as the pressure difference between the absorption (P_{Abs}) and desorption (P_{Des}), $P_{\text{Hys}} = P_{\text{Abs}} - P_{\text{Des}}$ (3.4 mbar)—compared to the Pd/TAF reference film ($P_{\text{Hys}} = 5.5$ mbar), as shown in Figure 2c. This reduced hysteresis indicates a lower strain energy associated with the α - β phase transition in the Ti/Pd/TAF film. In pure Pd films, this transition involves abrupt lattice expansion generating internal stress that can lead to film deformation, including buckling, delamination, and degradation in hydrogen cycling performance [15]. The Ti underlayer mitigates these effects by acting as both a mechanical clamp and adhesion promoter. It constrains volume expansion in the Pd film and suppresses stress-induced deformation, resulting in smoother, more reversible hydrogen absorption, and less transmission change (ΔT) below 800 nm (Figure 2b). Nevertheless, its ΔT dominant at above 800 nm might be due to the stronger dielectric change in this region. The suppression of hysteresis is supported by previous electrical measurements by Kim et al. [38,39] and by SEM/XRD studies from Verma et al. [23], which underscore the role of Ti in modifying Pd film behavior. We attribute this improvement to both interfacial and microstructural modifications introduced by the Ti underlayer. In addition to enhancing adhesion, Ti likely acts as a strain-relieving buffer, reducing energy barriers during hydride phase transitions. Interdiffusion at the Ti-Pd interface may result in Pd-Ti alloy formation, which disrupts long-range crystalline and promotes grain refinement. The resulting increase in grain boundary density facilitates faster hydrogen diffusion. This interpretation is further supported by the smaller response time, t_{90} , peak observed in the Ti/Pd bilayer compared to Pd-only films (Figure 2d), reinforcing the role of interfacial engineering in enhancing hydrogen uptake and release kinetics. The response time (t_{90}), defined as the time needed for the optical signal to reach 90% of its total change.

3.2.3. Faster Response and Enhanced Hydrogen Kinetics

Kinetic measurements were conducted by tracking real-time changes in optical transmission at the same wavelength during stepwise hydrogen pressure pulses from 100 to 1 mbar. The response time (t_{90}) exhibited a peak near 10 mbar for both samples (Figure 2d) and corresponds to α - β phase transition region in the palladium hydride. This behavior was previously reported in Pd-based optical and electrical hydrogen sensors [39–42]. This peak reflects the slower kinetics due to phase coexistence and lattice rearrangement [17,43]. Notably, the 5 nm Ti / 2.5 nm Pd / 30 nm TAF structure reduces the peak response time to 3.8 s—about 35% faster than the 5.85 s observed for the 2.5 nm Pd / 30 nm TAF reference film at 10 mbar. The effect is more pronounced at thinner Pd layers: at 2.25 nm, the peak t_{90} drops from 5.19 s (Pd-only) to 2.52 s (Ti/Pd bilayer)—a 52% improvement (Figure S4c).

3.3. Optimization of Pd Thickness for High-Performance Sensing

To optimize the performance of the Ti/Pd/TAF sensor, we reduced Pd thickness to 2.25 nm and 1.9 nm while keeping the Ti underlayer and TAF capping thickness fixed. Among these configurations, 5 nm Ti / 1.9 nm Pd / 30 nm TAF sensor demonstrated the best overall performance in terms of sensitivity, hysteresis suppression, and response time. Detailed results for this optimized structure are presented in Figure 3, data for the other configurations are provided in the supplementary materials (Figure S4).

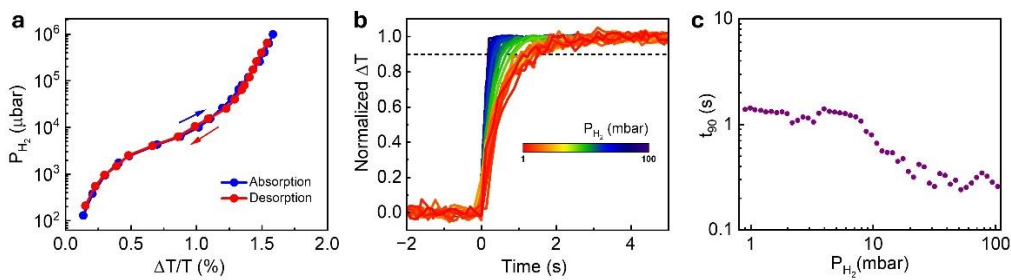


Figure 3. Optimized hydrogen sensing performance of 5 nm Ti / 1.9 nm Pd / 30 nm TAF film (a) Optical hydrogen sorption isotherms, and (b) Time-resolved normalized absorption response of the sensor under hydrogen pressure from 100 to 1 mbar at room temperature. (c) Extracted response times (t_{90}) of the sensors from panel (b) across the pressure range.

The hydrogen-induced transmission change of the optimized sensor (5 nm Ti / 1.9 nm Pd / 30 nm TAF) was examined as previously described (Figure S5a). The corresponding isotherm, shown in Figure 3a exhibits a minimal hysteresis width ($P_{Hys} < 0.3$ mbar), indicating hysteresis-free behavior across the measured pressure range. From the time-resolved data, we observe no response time peak—typically linked to the α - β phase transition in Pd—confirming that the transition is suppressed in this structure (Figures 3b and 3c). The sensor exhibits a rapid response ($t_{90} < 0.35$ s) at 4 % H_2 , the lower flammability limit, and maintains < 2 s response across the full 0.1 – 10 % ($1 - 100$ mbar) P_{H_2} range. This performance places the 5 nm Ti/ 1.9 nm Pd/ 30 nm TAF sensors among the faster thin film optical hydrogen sensors operating at telecommunication wavelengths, and comparable to more complex alloyed or nanostructured systems (see Table 1). Zeng et al. [39] similarly reported suppressed response time peaks in Ti/Pd electrical sensors with 2 nm Pd films, supporting the role of Pd thickness in mitigating the α - β phase transition; however, their devices exhibited significantly slower responses (>20 s) under comparable pressures.

The limit of detection (LOD) is a fundamental metric that represents the lowest hydrogen concentration a sensor can reliably detect. To determine the LOD of the optimized 5 nm Ti / 1.9 nm Pd / 30 nm TAF sensor, we extended measurements below 1 mbar - beyond range shown in Figure 3 – by gradually reducing the hydrogen concentration from 1000 ppm (0.1%) down to 10 ppm and recording the corresponding optical transmittance response (Figure 4a–c). For the concentrations below 100 ppm, the sensor was tested using 4% H_2 diluted in N_2 . Even at 10 ppm, the signal remained clearly detectable, with reversible on–off cycling confirming reliable performance at ultra-low concentrations. The pressure-dependent sensor’s response ($\Delta T/T$) followed Sievert’s law behavior, $\frac{\Delta T}{T} \propto (P_{H_2})^m$ [44], with the exponent m extracted from fitting the data. A slope of $m = 0.448 \pm 0.007$ was obtained for $P_{H_2} \leq 1000$ ppm (Figure 4d), while slightly lower value of $m = 0.37 \pm 0.009$ across the full measured P_{H_2} range (Figure S5b). These results indicate that hydrogen uptake in the Ti/Pd bilayer is governed by solubility-driven absorption dynamics, consistent with metal-hydride thermodynamics.

Table 1. To contextualize the performance of our Ti/Pd/TAF-based optical hydrogen sensor, we compared its key metrics—response time, limit of detection (LOD), and hysteresis behavior—with other state-of-the-art optical hydrogen sensors operating at room temperature. * n.a. = not addressed.

Sensing Platform	t_{90} (s) (@ 40 mbar)	t_{90} (s) (@ 1 mbar)	LOD (ppm)	Hysteresis Free?	Ref.
Pd ₈₀ Co ₂₀ NP	≤ 0.15	0.85	2.5	Yes	[9]
PdAu nano-particles @ PTFE/PMMA (100 x 25 (nm ²))	≤ 0.3	1	< 1000 (estimated)	Yes	[28]

Ti/Pd/TAF film	≤ 0.35	< 2	< 10 ppm	Yes	This work
Pd nano-disk array	> 10	-	50	n.a.	[42]
Pd bilayer lattices	~ 900	55	-	n.a.	[41]
PdAuCu nano-particles	0.4	-	5	Yes	[18]
PdAu nanostrucutres	40	-	-	yes	[47]
Pd strip	-	20	10	n.a.	[48]
PdY film	6	-	1000	n.a.	[49]
Pd/SiO2/Au	3	-	5000	n.a.	[50]
Pd/Au film	4.5	-	-	n.a.	[51]

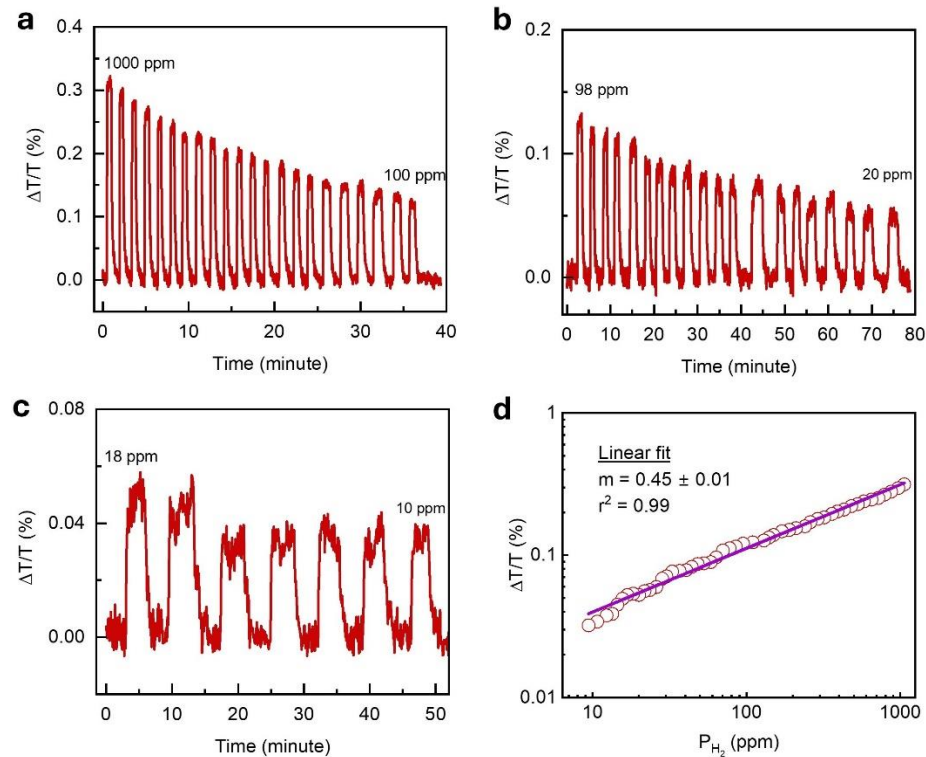


Figure 4. Limit of detection (LOD) measurement of the 5 nm Ti / 1.9 nm Pd / 30 nm TAF sensor. Transmission response $\Delta T/T$ at $\lambda = 1550$ nm measured under decreasing hydrogen concentrations ranging from (a) 1000 ppm to 100 ppm, (b) 98 ppm to 20 ppm, and (c) 18 ppm to 10 ppm (d) The measured optical sensitivity ($\Delta T/T$) as a function of low hydrogen concentration, derived from panels a-c, follows Sievert’s power law fit (solid line).

The long-term stability and durability of optimized Ti/Pd/TAF sensor were evaluated through over 135 consecutive hydrogenation and dehydrogenation cycles using 2% H_2 in N_2 under flow mode measurements. As shown in Figure 5a, the optical signal ($\Delta T/T$) at $\lambda = 1550$ nm remained stable over the entire 700-minute test duration, exhibiting no baseline drift or signal loss. A direct comparison of the response amplitude from the first 10 and last 10 cycles (Figure 5b) revealed less than 6% variation, with minimal error bars indicating strong reproducibility. This exceptional cycling stability is attributed to the Ti underlayer, which reinforces mechanical integrity by mitigating volume expansion in the Pd layer, thereby preventing delamination and preserving sensing performance over extended use [15].

Sensor selectivity—referring to the ability to detect a target analyte in the presence of other gases—and resistance to chemical poisoning are critical challenges in gas sensor development. An ideal hydrogen sensor should respond exclusively to hydrogen, without interference from other gases commonly present in the environment. To evaluate the selectivity of the optimized Ti/Pd/TAF sensor, we exposed it to 2% H_2 in N_2 , both alone and in a mixture with 5% CO_2 , 5% CH_4 , or 0.2% CO . As shown in Figure 5c and in the normalized response in Figure 5d, the sensor exhibited consistent

$\Delta T/T$ signals across all gas compositions, with no observable degradation during exposure to the mixed atmospheres. Although TAF is known for its high gas permeability and poor selectivity [45], the observed selectivity towards CO_2 and CH_4 may be attributed to the intrinsic resistance of Pd [9]. However, it cannot fully protect against CO [9,29]. To overcome this drawback, a PMMA layer of 100 nm thickness was coated on top of the Ti/Pd/TAF, as PMMA has been demonstrated as an excellent hydrogen-selective membrane [28,46]. Notably, the addition of the PMMA coating does not affect the response time compared to the uncoated sensor [29].

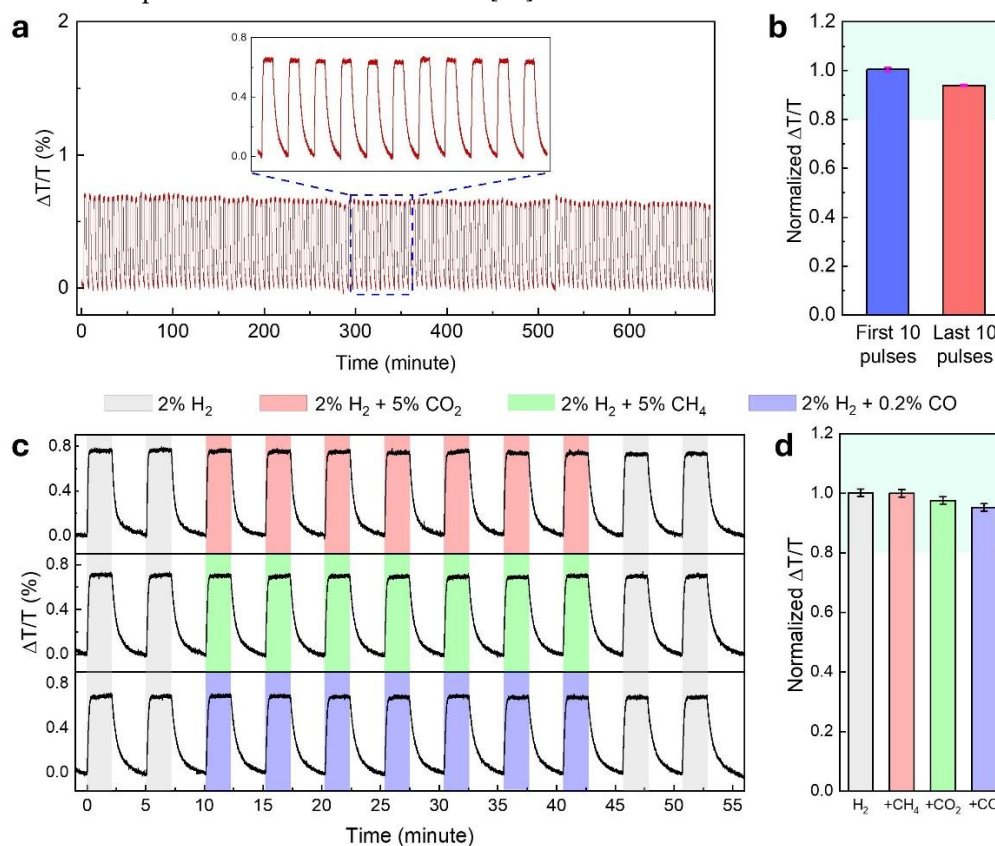


Figure 5. Long term stability and Effect of interfering gases on 5nm Ti/1.9nm Pd/30 TAF sensor. (a) Optical response ($\Delta T/T$) over 135 hydrogen cycles (2/3 minutes of loading/unloading) with 2% H_2 balanced in N_2 gas with a total flow of 400 ml/min. (b) Comparison of normalized response ($\Delta T/T$) amplitude for the first and last 10 pulses in figure (a). (c) Time-resolved optical response ($\Delta T/T$) of the sensor to 2 pulses of 2% H_2 followed by 7 pulses of 2% H_2 + 5% CO_2 , 2% H_2 + 5% CH_4 , 2% H_2 + 0.2% CO and a final 2 pulses of 2% H_2 in N_2 as a gas carrier. (d) Normalized sensor signal intensity compared to the reference signal obtained in 2% H_2 in N_2 gas flow extracted from (c). The error bars denote the standard deviation from 7 cycles.

5. Conclusions

We demonstrate a simple and effective approach to develop a hysteresis-free, fast response, highly sensitive and long-term stable hydrogen sensor without relying on complex alloying or nanostructuring. By exploiting the opposing dielectric responses of Ti and Pd to hydrogenation, the sensor achieves strong optical contrast at the telecom wavelength of 1550 nm. The optimized structure responds within 0.35 seconds at 4% hydrogen, detects concentrations below 10 ppm, and maintains consistent performance over 135 hydrogenation cycles with less than 6% signal degradation. The sensor also exhibits negligible cross-sensitivity to common interfering gases such as CO_2 , CH_4 , and CO. These findings highlight a promising path toward fiber-compatible, spark-free optical sensors for real-time hydrogen monitoring in safety-critical and industrial environments.

Supplementary Materials: The following supporting information can be downloaded at the website of this paper posted on Preprints.org

Author Contributions: A.T.M., T.A.N., and T.D.N conceived and designed the experiments. A.T.M. fabricated the samples, performed the sensing experiments, analyzed the experimental data, and wrote the first draft of the manuscript. H.M.L carried out the FDTD simulations. T.T.T.P and T.T conducted the XRD measurements. T.D.N. and Y.Z. supervised the project, coordinated research activities, and contributed to the development of manuscripts. T.A.N. and T.D.N. reviewed and revised all manuscript versions. All authors contributed to the final edits and approved the submitted version.

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Data Availability Statement: The data supporting the findings of this study are available from the corresponding author upon reasonable request.

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Abridged Disclaimer: The view expressed herein do not necessarily represent the view of the U.S. Department of Energy or the United States Government.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript: TAF, Teflon AF 2400; PMMA, Polymethylmethacrylate; SEM, scanning electron microscopy; XRD, X-ray diffraction, LOD; limit of detection.

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