

RESEARCH ARTICLE

Material Balance Study of Humid-Air Platinum Nanodisk Hydrogen Sensor Response at 80%RH and 80°C

J. L. Burnd^{1*}, C. Song² and E. J. Wallace¹

¹Clarendon Laboratory, Department of Physics, University of Oxford, Oxford, U.K.

²OXION Initiative in Ion Channels and Disease, University of Oxford, Oxford U.K.

*Corresponding author

Corresponding author:
brndjeol@bioch.ox.ac.uk



Abstract

Hydrogen sensing in humid air can not be assessed on the basis of concentration labels since the sensor surface is subjected to a mixture of hydrogen, oxygen, water vapor and purge gas flow. This study provides quantitative assessment of the catalytic plasmonic platinum nanodisk hydrogen sensor record in order to answer one research question: if the 80%RH and 80°C operational conditions remain chemically well fed and analytically separable once they are translated into water feed mass, hydrogen dose, oxygen requirement, reaction-water formation, purge volume and detection floor margin. The sensor consists of a quasi-randomly distributed Pt nanodisk array deposited on fused silica with a mean disk diameter of 215 ± 10 nm, height of 25 nm and optical reference maximum of 710 nm. The operational record includes synthetic air with 20.5% oxygen concentration, total flow rate of 200 mL min^{-1} , water feed rates of 0.081, 0.206 and 0.333 g h^{-1} at 20, 50 and 80%RH, hydrogen pulses ranging from 0.06 to 1.26 vol%, pulse intervals of 300 s, purges of 600 s, a 114 h humid core period within a 143 h stability run and detection floors of 30–50 ppm. One way of calculation is employed in all cases: water and hydrogen delivery is calculated based on the thermo-hygroscopic dose transduction technique, and oxygen supply reserve, product water upper bounds and purge-to-dose separation are calculated by means of the co-reactant balance analysis technique. The 80%RH humid core is associated with the water mass of 37.96 g and carrier gas flow of 1368 L. Every 1.26 vol% hydrogen pulse requires 205 mL oxygen, but only 6.30 mL of oxygen are required for complete oxidation; thus the minimum oxygen reserve at the maximum dose equals 32.5. The water formed in reactions is small relative to the external water: 33.7 mg during ten pulses active period against 832.5 mg external water feed, 0.641 g during 114 h humid core period against 37.96 g external water. Hydrogen is programmed during 15.83 h, purge during 31.67 h and humid carrier flow outside pulse-purge blocks during 66.50 h of the humid core period. Calculated balances provide a direct answer to the research question: the selected humid operation conditions are unfavorable in terms of low water exposure, favorable in terms of high water throughput together with excess oxygen, small product water contribution, good purge volume separation and detection floors substantially lower than both the 600 ppm repeated pulse and the 1000 ppm comparison value.

Keywords: hydrogen sensor, platinum nanodisk, catalytic plasmonics, humid air, co-reactant balance, purge duty, water burden, optical gas sensing

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1. Introduction

Hydrogen is becoming widely applied as an energy carrier, and its safe use requires timely concentration detection. Sensor development is not limited to dry laboratory gases only but involves stationary and mobile hydrogen infrastructure exposed to ambient air and variations

in temperature, humidity and purging cycles. General hydrogen safety investigations consider trueness, precision, response and recovery dynamics, stability, selectivity and detection range among application-specific performance characteristics [1]. The same applies to vehicle and fuel quality investigations, which prove the importance of humidity, temperature and gas mixture evaluation under realistic operational

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conditions [2,3]. Test guidelines are also consistent with the requirement that the impact of environmental parameters be assessed as a component of the intended use and not as standalone specifications. Laboratory test protocols form a reasonable basis for such evaluation [4]. It is important for low concentration warning since a sensor can exhibit appropriate sensitivity to hydrogen in dry air and lose any sense of recorded signal due to water presence and change in adsorption, local dielectric or recovery dynamics.

The existing scientific literature contains several mature sensing routes to hydrogen detection. Metal oxides' chemiresistive devices use changes in surface charge and adsorbate related to oxygen [5,6]. Catalytic and thermal-conductivity sensors utilize heating and gas transfer differences, electrochemical devices operate through electrode reactions and Pd-based devices utilize hydrogen absorption and lattice expansion associated with changes in optical or electrical properties [7,8]. Optical hydrogen sensors deserve special attention for hazardous environment applications since their sensing head can be remotely interrogated and free from potential spark generation. Optical hydrogen detection reviews emphasize the selection of active metals, alloy stability, polymer layers and plasmonic enhancement [9,10]. The fiber-integration studies also demonstrate the practical significance of coating design and geometry [11]. These studies prove that not only sensitivity but also the surrounding gas mixture and exposure history influence the ability to interpret the recorded signal.

Plasmonic hydrogen sensing introduces optical readout into this problem. Localized Surface Plasmon Resonance is sensitive to nanoscale dielectric and electronic environment changes close to metal surface [12,13]. Nanostructure geometry determines its optical properties [14]. The palladium-based plasmonic systems have been used for sensitive and fast hydrogen detection due to hydrogen interaction with optical properties of the metal [15]. Alloying and hybridization with metal-polymer layers increase speed and stability of response [16,17]. Whole spectrum data analysis and inverse optical design have also been applied for hydrogen detection limits minimization and improvement of performance in humid air [18,19]. Catalytic-plasmonic hydrogen detection based on platinum nanodisks differs from hydride-based detection in that platinum provides catalytic surface for hydrogen activation in oxygen containing air [20,21]. For this reason oxygen supply, water exposure and purge volumes become important for understanding the detected signal.

Humidity is commonly presented as %RH value, but %RH is not an absolute water load. With fixed flow rate, the same %RH value corresponds to different water amounts depending on exposure duration, as it can be derived from the principles of humidity measurement [22]. Water introduces additional cross-sensitivity and changes heat transport, affecting the measured response in hydrogen detection [23,24]. It is especially important for catalytic-plasmonic detection since water can change local surface organization, adsorbate population and optical contrast without damaging the structure. Long high humidity stability experiment should be therefore read as throughput of material: what amount of water passes through the reactor, how many liters of gas carry that water, what amount of hydrogen is supplied, how much oxygen and purge gas are present in the feed sequence.

The numerical basis is a platinum nanodisk operating record, where all significant exposure variables are listed. The sensor is made of quasi-random Pt nanodisks on fused silica substrate with average diameter of 215 ± 10 nm, nominal height of 25 nm and LSPR peak at 710 nm. The following gas program was used: synthetic air containing 20.5% oxygen with 200 mL min^{-1} flow; water feeds of 0.081, 0.206 and 0.333 g h^{-1} at 20, 50 and 80% RH; hydrogen pulses of 0.06, 0.13, 0.28, 0.60 and 1.26 vol%; 300 s pulse interval; 600 s purges; and 114 h high-humidity core in 143 h stability record [20]. These numbers allow the gas-and-water balance to be estimated without any extra spectral points, calibration factors and laboratory procedures.

The research question is limited compared to general performance

review: when 80% RH and 80°C working conditions are expressed through water throughput, hydrogen concentration, oxygen availability, reaction water upper limit, purge filling degree and detection floor separation, does the operating record reflect a chemically plausible and analytically separate humid-air operating conditions? Such an answer is important since the same optical response can be interpreted as a humidity-enhanced response, oxygen-limited catalytic state, water product accumulation and recovery artifact due to insufficient purge. The calculation allows separating these alternatives with the help of the given flow, humidity, time and concentration values.

The Pt nanodisk surface shown in Figure 1 makes the material balance calculation start from the catalytic-plasmonic interface and not from abstract gas cell. The macroscopic balances derived below will reflect the stream delivered to this interface, while the local optical and catalytic response will occur within the humid Pt near-field environment. It is useful because it prevents misunderstanding of gas feed calculations as a microscopic kinetic model.

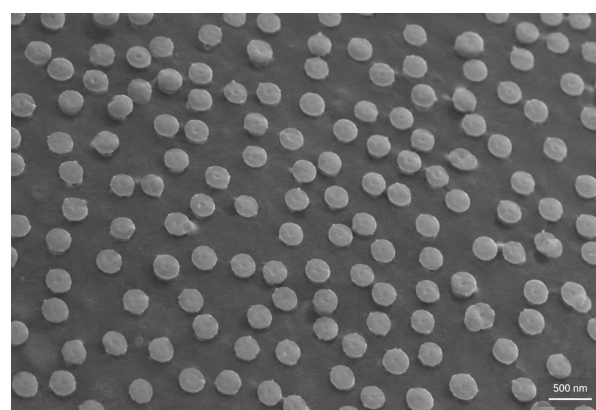


Figure 1: Platinum nanodisk operating surface.

2. Materials and Methods

2.1. Sensor Operating Record and Calculation Variables

The numerical base is according to the platinum nanodisk hydrogen-sensor operating record of Theodoridis, Andersson, Nilsson, Fritzsche and Langhammer [20]. The numerical record provides the nanodisk dimensions, optical resonance position, carrier-gas composition, flow rate, humidity conditions, water-feed rates, temperature range, hydrogen-pulse concentrations, pulse and purge times, humid stabilities and detection floor levels. These values are transformed into gas, water and co-reactant amounts without including any spectral signatures, wavelength shifts points, zero-hydrogen noise levels and calibration factors except the numerical record given.

The carrier gas is the synthetic air that contains 20.5% O_2 in nitrogen. The total flow is 200 mL min^{-1} in the optical reactor which equals to 12 L h^{-1} . The hydrogen is delivered in a ladder of 0.06, 0.13, 0.28, 0.60 and 1.26 vol% that corresponds to 600, 1300, 2800, 6000 and 12600 ppm. Every hydrogen pulse lasts for 300 seconds and every purge period lasts for 600 seconds. At 200 mL min^{-1} flow rate, each hydrogen pulse delivers 1000 mL of total gas and every purge delivers 2000 mL synthetic air. Humid stability program includes 114 h of humid core of 80% RH and 80°C within 143 h of total measurement. In terms of cycles, the humid core can be considered as 19 dose-equivalent cycles in which there are five concentrations that occurs twice. It yields to 10 hydrogen pulses per cycle and 190 hydrogen pulses in the humid core. Table 1 shows the physical and operational input data, while Table 2 presents the pulse program transformations that will be used in further calculations.

Table 1: Operating record.

Descriptor	Value used in the calculation
Sensing element	Quasi-random Pt nanodisk array on fused silica; measured mean diameter 215 ± 10 nm; nominal height 25 nm.
Optical reference	Broad LSPR maximum near 710 nm before gas-program analysis.
Carrier atmosphere	Synthetic air containing 20.5% O ₂ in N ₂ .
Total volumetric flow	200 mL min ⁻¹ through the optical reactor.
Humidity settings	0–80% RH; explicit water-feed rates of 0.081, 0.206 and 0.333 g h ⁻¹ for 20, 50 and 80% RH.
Temperature grid	33, 50, 80 and 100°C for humid titration and hydrogen exposure.
Long humid section	80% RH and 80°C for 114 h within a 143 h stability programme.
Low-concentration floor	30 ppm for 0–50% RH at 80°C and 50 ppm at 80% RH.
Safety comparison value	1000 ppm H ₂ target used for detection-floor interpretation.

Table 2: Pulse-program conversions.

Input	Value	Direct conversion	Use in the calculation
Hydrogen concentrations	0.06, 0.13, 0.28, 0.60 and 1.26 vol%	600, 1300, 2800, 6000 and 12600 ppm	Defines the concentration ladder for pulse-dose and oxygen-demand calculations.
Pulse duration	300 s	5 min	Sets the total gas volume admitted during each hydrogen pulse.
Purge duration	600 s	10 min	Sets the synthetic-air volume used to clear each pulse.
Gas volume per pulse	200 mL min ⁻¹ × 5 min	1000 mL	Converts concentration into delivered H ₂ and available O ₂ .
Gas volume per purge	200 mL min ⁻¹ × 10 min	2000 mL	Defines purge-to-hydrogen separation strength.
Dose-equivalent set	Five concentrations, each occurring twice	10 hydrogen pulses plus 10 purges	Provides the common unit for humid-core accounting.
Number of wet sets	19 dose-equivalent sets	190 hydrogen pulses	Converts single-set balances into 114 h humid-core balances.

These two tables together give a complete operating record for the computation. Table 1 shows the actual physical sensor and carrier gas, and humidity input and long-term exposure. The other table gives the conversion of repeated pulse program to volume of gas. This division is required since there is no way of representing the humidity level, hydrogen input, and purge recovery in one concentration axis.

2.2. Thermo-Hygrometric Dose Transduction

The thermo-hygrometric dose transduction considers the sequence of exposures to be a record of delivered doses. With hydrogen concentration denoted by c in vol%, the delivered volume of hydrogen for a single pulse is

$$V_{H_2}(c) = \frac{Q\tau_p c}{100}, \quad (1)$$

with $Q = 200$ mL min⁻¹ and $\tau_p = 5$ min. Therefore, the total volume of gases for a hydrogen pulse will be 1000 mL, which gives us $V_{H_2} = 10c$ mL when c is measured in vol%. This conversion is necessary due to the fact that the ladder of concentrations is not linear with respect to delivered dose: a 1.26 vol% pulse provides twenty-one times more hydrogen than a 0.06 vol% pulse with the same duration. Humidity has been transduced from relative measure to a mass burden. With $Q_L = 12$ L h⁻¹ as a volume of carrier gas per hour, the mass of water per litre of carrier gas for humidity level H is

$$B_w(H) = \frac{\dot{m}_w(H)}{Q_L}, \quad (2)$$

with $\dot{m}_w(H)$ being the rate of water feed. The cumulative mass of water supplied during time period t is

$$M_w(H, t) = \dot{m}_w(H)t. \quad (3)$$

It is not assumed here that each individual water molecule is bound to the platinum surface or stays in the reactor. This describes the amount

of water load supplied via the gas flow which should be used as the first-order criterion for long operation under high humidity. Detection-floor analysis is performed based on the ratio of the detection floor to a 1000 ppm comparison value and to the smallest hydrogen pulse:

$$\Psi_{\text{tar}} = \frac{C_{\text{tar}}}{C_{\text{LoD}}}, \quad \Psi_{\text{pulse}} = \frac{C_{\text{low}}}{C_{\text{LoD}}}, \quad (4)$$

where $C_{\text{tar}} = 1000$ ppm, $C_{\text{low}} = 600$ ppm and C_{LoD} takes the 30–50 ppm range.

2.3. Co-Reactant Balance Analysis

co-reactant balance analysis quantifies whether the gas stream is chemically sufficient for complete hydrogen oxidation under the programmed conditions. The global reaction is



For synthetic air containing $x_{O_2} = 0.205$, the oxygen volume admitted during a hydrogen pulse is

$$V_{O_2, \text{pulse}} = x_{O_2} Q \tau_p. \quad (6)$$

Since $Q\tau_p = 1000$ mL, every pulse admits 205 mL oxygen. The stoichiometric oxygen demand is

$$V_{O_2, \text{req}}(c) = \frac{1}{2} V_{H_2}(c), \quad (7)$$

and the oxygen-excess ratio is

$$\Xi_{O_2}(c) = \frac{V_{O_2, \text{pulse}}}{V_{O_2, \text{req}}(c)}. \quad (8)$$

If the value exceeds unity significantly, the feed bulk is relatively oxygen-enriched. The ratio does not guarantee that each site is oxygen-enriched and microkinetic limitations are absent but excludes a deficiency of oxygen in admitted gas volume.

The amount of product-water generated was assumed as an upper bound under a hypothesis that all hydrogen delivered to the fuel cell is oxidized to water. Molar volume V_m was assumed as equal to 24.88 L mol⁻¹ based on the approximate 30°C and 1 atm humidification conditions which were assumed for gas volume calculations. The amount of product water is

$$m_{\text{rxn}}(c) = \frac{V_{H_2}(c)}{1000V_m} M_{H_2O}, \quad (9)$$

where $M_{H_2O} = 18.015$ g mol⁻¹. The upper bound was adopted intentionally conservatively. The imposed water feed cannot be exceeded by product water generation if the upper bound is small compared with water feed.

2.4. Purge Duty and Humid-Core Segmentation

The calculation of purge duty takes into account the volume that is cleared after each hydrogen dose. The purge-to-hydrogen volume ratio in a single dose is

$$\Pi_{\text{purge}}(c) = \frac{Q\tau_g}{V_{H_2}(c)}, \quad (10)$$

with $\tau_g = 10$ min and $Q\tau_g = 2000$ mL. Although this is not an actual measure of microscopic recovery of the surface, it is a measure of how well the programme distinguishes between consecutive doses in terms of carrier gas volume. At the set level, ten purges and ten hydrogen pulses offer a common point of comparison to repeat the procedure.

The 114 h humid core was partitioned into programmed hydrogen exposure, scheduled purge and wet carrier flow outside explicit pulse-purge blocks. With $N_s = 19$ dose-equivalent sets, $N_p = 10$ hydrogen pulses per set, $\tau_p = 5$ min and $\tau_g = 10$ min,

$$t_{H_2} = N_s N_p \tau_p, \quad (11)$$

$$t_{\text{purge}} = N_s N_p \tau_g, \quad (12)$$

$$t_{\text{wet, other}} = t_{\text{core}} - t_{H_2} - t_{\text{purge}}, \quad (13)$$

where $t_{\text{core}} = 114$ h. Such intervals help separate active hydrogen challenge from wet conditioning of longer duration. This separation is valuable since, in some cases, the stability test can be predominantly carrier-exposure despite having many analyte pulses.

2.5. Operating Decision Limits

The operating condition is specified by four transparent decision limits. It is oxygen-abundant if $\Xi_{O_2} > 10$ in the largest hydrogen pulse, water-product subordinate if upper-bound reaction water is below 10% of external water in the same time frame, strongly purge-separated if the ratio of purge and hydrogen volume at set level exceeds 100, and sufficiently removed from the detection floor if $\Psi_{\text{tar}} > 10$ and $\Psi_{\text{pulse}} > 5$. This set of criteria is specific to the operation record being analyzed. Different reactor sizes, surface types, flow designs, and detection limits would each have their own criteria.

3. Results and Discussion

3.1. Water-Load Conversion and Hydrogen-Dose Structure

The first finding from the data analysis is the material significance of the humid test. The water loading rates at 200 mL min^{-1} carrier flow rate equate to 6.75 mg L^{-1} at 20% RH, 17.17 mg L^{-1} at 50% RH and 27.75 mg L^{-1} at 80% RH. Thus, the difference between humid tests with 20% and 80% RH is not just a matter of nomenclature; it is an almost fourfold difference in the water dose per liter of carrier gas. With 80% RH, the 114 h humid core provides

$$0.333 \text{ g h}^{-1} \times 114 \text{ h} = 37.96 \text{ g} \quad (14)$$

the water to the reactor stream. The same interval passes 1368 L total carrier gas. In other words, the stability test could be said to represent a long-term wet throughput test, where the sensor faces not only high instantaneous water activity but also a much larger amount of water mass transported than the mass of hydrogen introduced by the pulses. The water-burden plate shown in Figure 2 translates the humidity settings into the corresponding water mass. An important measure here is the amount of water burdened at 80% RH: 37.96 g of water goes through the reactor stream during 114 hours of the humid core. Thus, the sensor experiences sustained water transport, rather than a particular value of the relative humidity label.

The hydrogen ladder is strongly dose-structured too. It introduces 0.6, 1.3, 2.8, 6.0 and 12.6 mL H_2 of hydrogen per 300-s pulse. For example, a dose-equivalent set that includes each concentration two times introduces 46.6 mL H_2 . Nineteen such sets introduce 885.4 mL H_2 of hydrogen throughout the humid core. In particular, one single pulse contains the hydrogen mass equal to twenty-one pulses of 0.06 vol% of hydrogen for the same pulse duration. Thus, the ladder consists of five steps that are non-equidistant with respect to the hydrogen dose. This is particularly relevant to humid air sensing, where high-dose pulses may generate more demanding recovery conditions than lower-dose ones despite their stable total programme.

The dose ladder in Figure 3 reveals the different degrees of severity among the five hydrogen exposure doses. While the highest 1.26 vol% dose includes 12.6 mL of H_2 in 300 s, the smallest dose of 0.06 vol% includes only 0.6 mL of H_2 . Therefore, the 600 s purge time after each

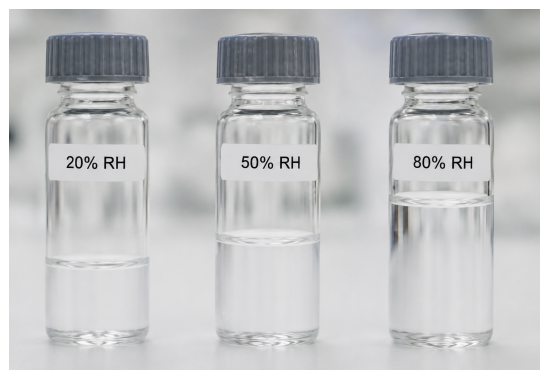
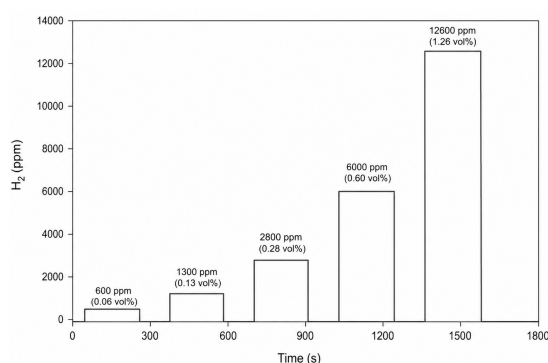


Figure 2: Water burden at 20–80% RH.

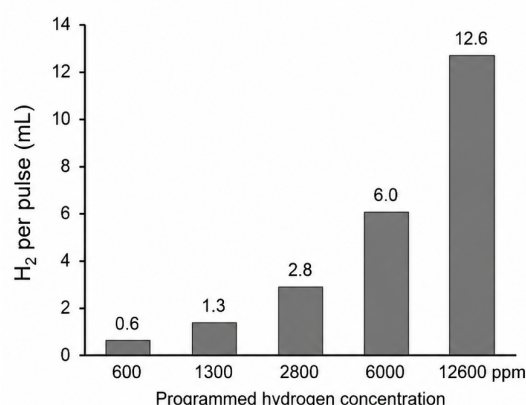
pulse becomes an important parameter in the dose design since it is the means to provide volume separation for non-equivalent hydrogen exposures.



(a) Concentration sequence.



(b) Pulse-purge timing.



Hydrogen (mL) Pulses (n)

One set	46.6	10
19 sets	885.4	190

(c) Dose-volume summary.

Figure 3: Hydrogen pulse ladder.

The retained thermo-hygrometric dose transduction conversions in Table 3 distinguish two different parameters combined into one exposure dose. Humidity doses are used to describe the water loading, and hydrogen doses are used to describe the catalytic exposure dose. This allows recognizing the humid environment operation as a two-parameters exposure test. The general assessments of the hydrogen sensors performance consider the gas matrix and temperature as integral parts of the device performance [25, 26]. The same approach is taken by the application-related research works on hydrogen sensors [2, 7].

Table 3: THDT conversions for humid burden and hydrogen dose.

RH (%)	Water feed (g h ⁻¹)	Water burden (mg L ⁻¹)	H ₂ level (vol%)	Equivalent (ppm)	H ₂ per pulse (mL)
20	0.081	6.75	0.06	600	0.6
50	0.206	17.17	0.13	1300	1.3
80	0.333	27.75	0.28	2800	2.8
–	–	–	0.60	6000	6.0
–	–	–	1.26	12600	12.6

In terms of Table 3, 80% RH at 80°C provides the most severe yet controllable exposure condition. The latter condition produces the largest water load while keeping the detection threshold at 50 ppm in case of 80% RH. In spite of the use of the most conservative 50 ppm value, the lowest programmed pulse at 600 ppm will still be 12 times higher than the detection threshold while a 1000 ppm comparison value will be 20 times higher than the threshold. In the case of the lower-humidity portion of the 80°C history with the 30 ppm floor, the ratio between the programmed pulses and the detection threshold will be 20 and 33.3, correspondingly. In other words, the optical system retains a certain concentration difference while being loaded with a relatively large amount of water.

3.2. Oxygen Sufficiency for the Concentration Ladder

The co-reactant balance analysis calculation indicates that there is no oxygen limitation in the humid air programming scenario. In case of each hydrogen pulse, the synthetic-air flow provides 205 mL oxygen. The stoichiometric oxygen requirement varies within the range of 0.30 mL for the 0.06 vol% pulse and 6.30 mL for the 1.26 vol% pulse. Thus, the excess ratio of oxygen relative to the programmed pulses varies within the range of 683.3 for 600 ppm and 32.5 for 12600 ppm. In both cases, the excess ratio is larger than one and exceeds 10 at any level. In case of the equivalent dose set, ten pulse intervals provide 2050 mL oxygen, while oxidation of 46.6 mL hydrogen set requires 23.3 mL oxygen, see Table 4.

In this regard, Figure 4 oxygen reserve plates directly visualize the oxygen conclusion. The amount of oxygen excess decreases with the increase of concentration, as the admitted oxygen volume is still 205 mL, while hydrogen pulses grow. However, the highest possible pulse has more than thirty times the required oxygen to completely oxidize hydrogen atoms. That limits the possible interpretations: changes at high humidity cannot be associated with oxygen deficiency in the macroscopic mixture. Possible sources of such changes include local coverage competition, water-induced changes in Pt interface, heat and mass transfer around nanodisks and hydrogen activation on the wet catalytic surface. Water is known to create localized ordered structures on platinum interfaces [27, 28]. Oxygen electrocatalysis on platinum has similar structural sensitivity at nanoscale level [29, 30]. Therefore, the local chemistry can become complicated in spite of macroscopically simple gas composition.

In this way, the oxygen calculation clearly differentiates platinum catalytic-plasmonic sensing from purely hydride-based sensing. Palladium hydride-based sensors are usually analyzed in terms of hydride equilibrium properties [31]. Hydride thermodynamics can also depend on nanoparticle sizes and materials [32, 33]. In the case of platinum nanoparticles, catalytic hydrogen oxidation and dielectric sensing are

Table 4: CRBA quantities for oxygen demand and product-water formation.

H ₂ (vol%)	H ₂ dose (mL)	O ₂ admitted (mL pulse ⁻¹)	O ₂ required (mL)	O ₂ excess ratio	Product water (mg)
0.06	0.60	205	0.30	683.3	0.43
0.13	1.30	205	0.65	315.4	0.94
0.28	2.80	205	1.40	146.4	2.03
0.60	6.00	205	3.00	68.3	4.34
1.26	12.60	205	6.30	32.5	9.12

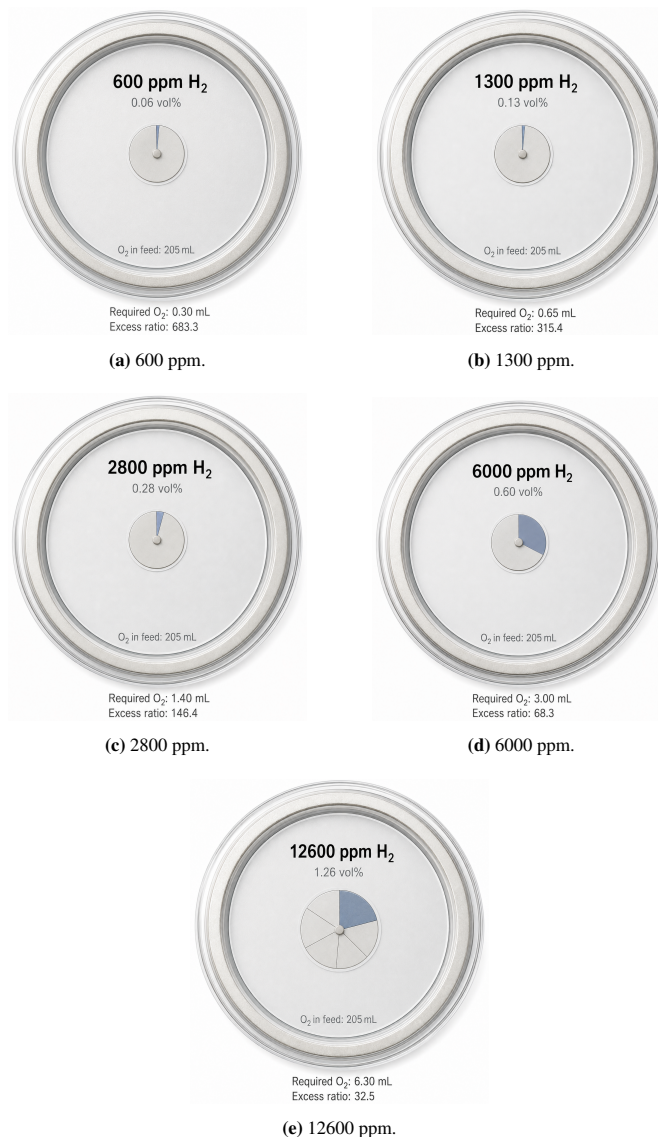


Figure 4: Oxygen reserve across hydrogen pulses.

crucial processes, so the oxygen availability must be checked explicitly. Nevertheless, this does not exclude the necessity of kinetic measurement; the kinetic limitation, however, would be related to surface or transport effects, rather than stoichiometric lack of O₂ in the admitted gas.

3.3. Reaction-Water Apportionment

The product-water calculation shows that the reaction water is orders of magnitude lower than the external humidity input at reactor scale. Complete oxidation of 1.26 vol% pulse will give rise to approximately 9.12 mg of water. Complete oxidation of the ten-pulse equivalent dose will lead to 33.7 mg of water. In 2.5 h duration of active set at 80% RH, the humidifier will provide 832.5 mg of water. Hence, reaction

water represents merely 4.05% of total reactor-scale water input in active set. On the level of the 114 h humid core, complete oxidation of 885.4 mL hydrogen dose will lead to 0.641 g of water. Meanwhile, the humidifier will provide 37.96 g of water. The core-level product-water share is 1.69%.

The results for water origin in Table 5 and Figure 5 exclude a possible misinterpretation by splitting imposed humidification from upper-bound water generated through hydrogen oxidation. Hydrogen oxidation generates water, which implies the assumption that the catalytic reaction plays the dominant role in generating water in the humid environment. The data refute this assumption for 200 mL min⁻¹ flow rate and 0.06–1.26 vol% hydrogen concentration range. Generated water has a chemical meaning on-site and could potentially change adsorbate population, but this water is not the main water source introduced into the reactor. Humidifier provides the majority of water, and therefore the sensor design must focus on imposed humidity, surface hydration, and purge recovery, and not just product water reduction.

Table 5: Water-origin apportionment for the active set and the complete humid core.

Interval	External water feed (g)	Theoretical product water (g)	Product-water share (%)
One 2.5 h dose-equivalent set at 80% RH	0.8325	0.0337	4.05
Nineteen sets distributed over 114 h	37.96	0.641	1.69

Share of active set product water is greater than the share of core-level product water, since the active set concentrates hydrogen doses within 2.5 h, whereas humid core has extended periods of humid carrier flow out of explicitly organized pulse–purge block. This difference shows necessity of using time partitioning. Short period around the pulses is reaction-rich, while long 114 h period of humid core is conditioning-rich. Stability of the sensor during the experiment therefore can be seen as the ability to tolerate the extended period of external water throughput with intermittent catalytic reactions, and not as high-rate constant hydrogen oxidation.

3.4. Purge-Duty Partitioning of the Humid Core

2000 mL of synthetic air purge follows each hydrogen pulse. Because hydrogen dose varies with concentration ladder, purge-to-hydrogen ratio varies from 3333 for 600 ppm pulse to 159 for 12600 ppm pulse. For the set level, the dose-equivalent set comprises 20 L of purge gas and 46.6 mL of hydrogen, so the purge-to-hydrogen volume ratio is 429. This is high enough to qualify the program as strongly purge-separated by the internal decision rule. The result does not imply that the surface site resets to its initial state every time after each pulse; however, it means that the gas program has significant macroscopic capacity of surface reset.

Humid-core partition provides another operational point of view. 19 dose-equivalent sets consist of 190 hydrogen pulses. Hydrogen exposure takes 950 min, i.e., 15.83 h. Purge duty takes 1900 min, i.e., 31.67 h. Wet synthetic-air duration without explicitly organized pulse–purge blocks takes 66.50 h. These times make up 13.9% of 114 h humid core, 27.8% of 114 h, and 58.3% of 114 h respectively. Table 6 reports the partition of times and volumes.

It can be seen from Figure 6, however, that pulse counting does not fully cover the stability record of 143 hours of operation. Over half of the humid core represents wet carrier flow outside explicit blocks of pulse–purge steps, and more than one quarter of time is dedicated to purge steps. Such design makes it possible for the gas stream to clean out any hydrogen accumulated between exposures in humid background all the time. The operational meaning of this setup can be understood as testing of recovery from isolated hydrogen pulses within permanent wet conditioning of the sensor. It is more specific

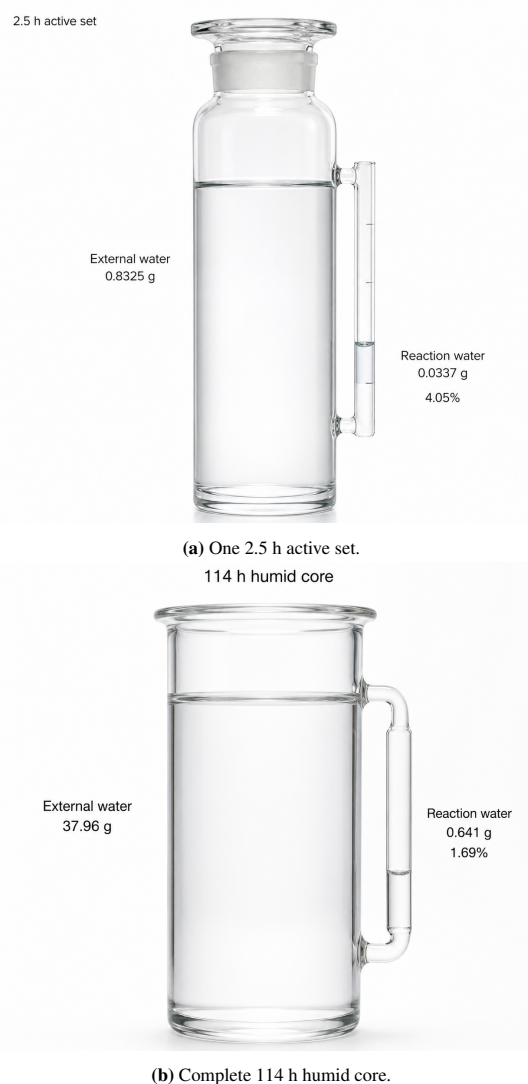


Figure 5: External and reaction water at 80% RH.

Table 6: Time and gas-volume partitioning inside the 114 h humid core.

Core component	Time (h)	Fraction of humid core (%)	Total carrier gas volume (L)
Programmed hydrogen-pulse intervals	15.83	13.9	190
Scheduled synthetic-air purge intervals	31.67	27.8	380
Humid carrier outside explicit pulse–purge blocks	66.50	58.3	798
Total humid core	114.00	100.0	1368

than just mentioning stability at high humidity.

Analysis of purge duty also gives a handle on the interpretation of the large-dose pulses. The smallest purge-to-hydrogen ratio among pulses is 159 for the 1.26 vol% pulse. If there were any problems in the recovery after the largest pulses, it should not be associated with an insufficient purge volume at the macroscopic scale, but more probably with surface retention, water organization, heating of nanodisks, slow desorption of intermediates or zero-hydrogen optical relaxation. It is worth noting this distinction in order to plan future experiments since the solution to the purge-volume problem differs from the solution to the surface kinetics problem.

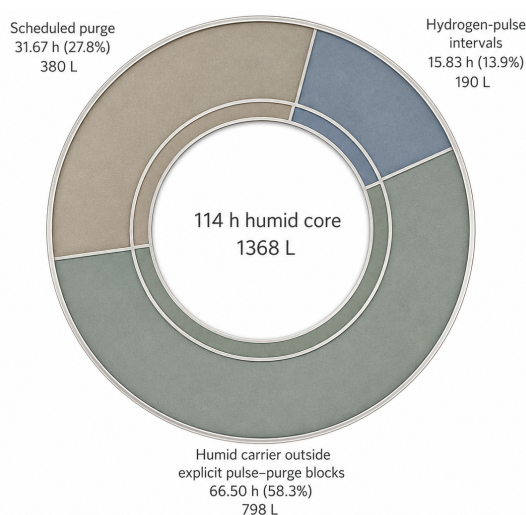


Figure 6: Humid-core time occupancy.

3.5. Detection-Floor Separation Under Humid Co-Reactant Balance

After the conversion to margins, the detection-floor record stays favourable. Detection floor of 30 ppm gives a target margin of 33.3 relative to 1000 ppm and the lowest-pulse margin of 20.0 relative to 600 ppm. Detection floor of 50 ppm gives the margins of 20.0 and 12.0. Both values satisfy internal detection-separation limits. The margins are not regulatory claims. This is a method of demonstrating that the low-concentration floor stays sufficiently separated from the comparison concentration and the smallest programmed regular pulse. From Figure 7 it can be seen how the low-concentration floor appears on the same scale as programmed hydrogen ladder. There is still a significant separation even under the conservative value of 50 ppm floor, which means that the 80% RH condition maintains sufficient clearance between the detection capability and the repeated 600 ppm pulse.

The converging integrated plane in Figure 8 represents the operating condition as an outcome of a convergent balance result. Evidence does not depend on a single response on the plot; it comes from simultaneous excess of oxygen, high externally added water load, small contribution of product-water, good purge separation and concentration-floor clearance.

Operating interpretation in Table 7 and Figure 8 indicates that 80% RH and 80°C operating point is beneficial due to several independent factors. 30–50 ppm C_{LoD} values are way below the test concentrations, oxygen abundance is high along the ladder, externally added humidification contributes to most of the water mass and purge stream is relatively large compared to the hydrogen dose.

These results support each other. A high optical response would be less convincing in case of oxygen deficiency, in case product water contributed predominantly to the reactor-level water load or purge was not enough to separate successive pulses. The calculated record contains none of these features.

Interpretation gives the understanding of the role of temperature. The operating point of 80°C is not considered here as a simple thermal setting but as a part of a coupled condition where catalytic activity, humid optical contrast, adsorption dynamics and detection floor are involved. Water on metal surfaces may cause reorganization of adsorption states and interfacial structures [27, 28]. Also, platinum catalytic behavior is structure sensitive [29]. Material-balance calculation does not explain the mechanism but puts constraints on it. Having sufficient oxygen and purge gases, the explanation of humid stability will be mostly related to the interface between water-covered platinum and plasmonic optical

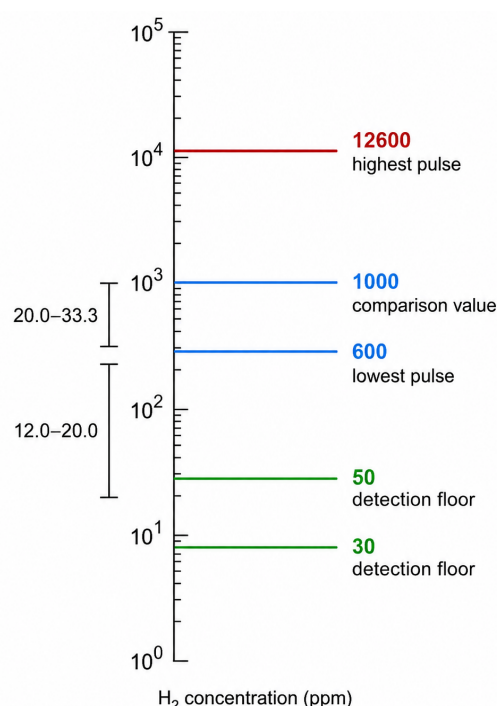


Figure 7: Detection-floor clearance.

Table 7: Combined operating interpretation at 80% RH and 80°C.

Criterion	Calculated result	Interpretation
Target detection margin	20.0–33.3	The detection floor remains well below the 1000 ppm comparison value.
Lowest-pulse detection margin	12.0–20.0	The 600 ppm pulse remains clearly above the 30–50 ppm detection floor.
Largest-pulse oxygen excess	32.5	The gas programme is oxygen-rich even at 1.26 vol% H_2 .
Product-water share in active set	4.05%	Reaction water is locally relevant but subordinate in the reactor-scale water balance.
Product-water share in humid core	1.69%	The long-run water burden is dominated by externally supplied humidity.
Set-level purge-to-hydrogen ratio	429	The purge programme provides a large clearing volume relative to hydrogen dose.
Humid-core active H_2 occupancy	13.9%	The 114 h run combines prolonged wet conditioning with repeated, separated hydrogen events.

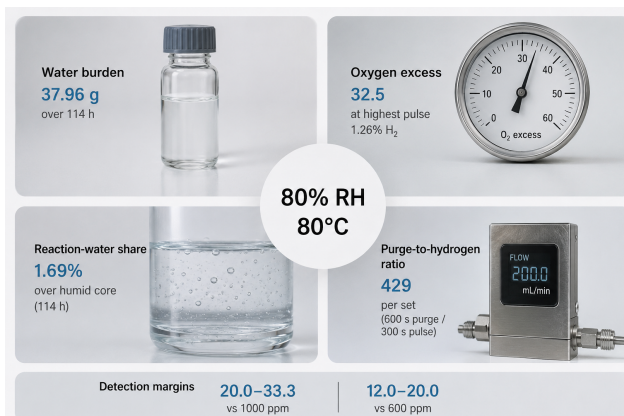


Figure 8: Integrated operating balance.

modes.

Further, platinum nanodisk record should be analyzed in connection

with other hydrogen sensing techniques. Sensing by means of material expansion induced by hydrogen was reported for palladium mesowires and cracked palladium film [34,35]. Nanostructured metal hydrides and fiber-integrated Pd/WO₃ coating are similar optical and optomechanical techniques [11,15]. More modern designs include alloying and metal-polymer structures for improved reaction speed and material stability [16,17]. Full-spectrum readout and inverse optical design may improve humidity tolerance and detection limits [18,19]. Hot-electron techniques and other optical sensors expand the design space beyond conventional hydride readout [9,21]. The platinum nanodisk record is unique in a sense that the primary problem is not a hydride phase transition hysteresis but the catalytic-plasmonic behavior in the presence of oxygen-containing humid air. It makes the co-reactant balance essential in addition to optical responses analysis.

3.6. Reporting Implications for Humid-Air Catalytic-Plasmonic Tests

Firstly, a practical implication is that hydrogen sensor papers based on humid operation should contain sufficient information for the material balance interpretation. The values of relative humidity, temperature and concentration are important, but flow rate, water feed rate, pulse duration, purge duration, carrier composition and long-run exposure time are no less important. Without these values, it is difficult to say whether the two experiments have comparable water burden or gas-dose severity. The platinum nanodisk record is especially convenient for this conversion since it contains both humidity and feed rate values. Secondly, the product water formation should be distinguished from external humidity. At low percent hydrogen pulses and high humidification, the reaction water contribution may be small compared with externally added water. However, under other conditions, such as higher hydrogen concentration, lower external humidity, smaller reactor volume or slower carrier flow, product water could become important. A simple upper bound calculation may indicate whether it is the case or not prior to building a detailed kinetic model. It is important since this approach avoids incorrect interpretation of the product water formation under the dominance of externally added humidity.

Thirdly, the purge procedure should be described by its ratio to the gas dose. A ten minute purge can be excessive or insufficient depending on flow rate, reactor volume and gas dose. In this operating record, the purge gas volume is large compared to the hydrogen volume, so the purge-based interpretation of recoveries should be focused on surface and optical relaxation rather than the gas-exchange deficit. Such a transferable description of the purge procedure needs purge gas volume and estimated reactor volume exchange number.

Finally, the detection floor should be interpreted together with the lowest programmed pulse. The low C_{LoD} is good but its practical significance depends on the distance to the lowest tested concentrations. Here the 600 ppm pulse is 12-20 times higher than the floor at the humid operating point, making the detectability clear under the specified conditions. In other cases, the low C_{LoD} may be less significant when the repeated test does not confirm stable response around that level.

4. Conclusions

The research question considers whether the 80% RH and 80°C platinum nanodisk hydrogen-sensor operating point retains credibility when it is re-interpreted as a delivered gas-and-water record, not as an RH value. The answer is affirmative, in terms of the stated operating record. The requirement is severe in the sense that it passes 37.96 g externally supplied water through the reactor over 114 h, not in the sense that hydrogen consumption depletes available oxygen or forms enough reaction water to prevail in the humid air. This is the key conclusion.

Using the water and dose calculations, one learns that the 80% RH

core is made up of 27.75 mg water per liter of carrier gas and 1368 L carrier flow. Ten-pulse equivalent dose delivers 46.6 mL H₂. The 19 wet sets carry 885.4 mL programmed H₂ through the humid core. Even the lowest repeated hydrogen pulse stays 12.0–20.0 times above the 30–50 ppm detection floor, and the 1000 ppm comparison value stays 20.0–33.3 times above the floor. In other words, the operating point maintains concentration separation despite carrying significant water burden.

The co-reactant calculation provides the answer to the second half of the research question. Each pulse allows 205 mL O₂ from the synthetic air. Even the largest 1.26 vol% pulse consumes only 6.30 mL O₂ completely, thus giving an oxygen excess ratio of 32.5 even in the case of maximum hydrogen pulse. Also, water-product formation is relatively small in relation to the externally imposed humid feed: even complete conversion over a single active set produces 33.7 mg water against 832.5 mg external water and 0.641 g reaction water against 37.96 g externally supplied water over 114 h of humid core. These numbers exclude oxygen exhaustion and reaction water accumulation in the reactor as the main explanation for the favorable humidity effect record.

Time-partition calculation shows why the stability test must be interpreted as a recovery under humidity experiment. 15.83 h of the humid core is taken by programmed hydrogen, 31.67 h by programmed purge and 66.50 h by wet carrier flow not belonging to explicit pulse-purge blocks. Purge-to-hydrogen volume ratio of a single set is 429. The final interpretation is specific: 80% RH and 80°C operating point is characterized by a high water throughput, oxygen surplus, minor product water formation and significant purge separation with detection floors much lower than repeated hydrogen concentration values. Humid-air catalytic plasmonic hydrogen-sensor papers must report water feed rate, carrier gas composition, pulse duration, purge duration and long-run exposure time in order to evaluate water burden, oxygen reserve, product water contribution and purge separation.

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