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LOW-COST COMMERCIAL HYDROGEN SENSOR FOR MEASURING UNBURNED HYDROGEN IN ENGINE EXHAUST

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***Abstract*—**A commercial low-cost hydrogen sensor, based on the catalytic oxidation principle, is investigated for suitability in sensing emissions of unburned hydrogen from diesel dual-fuel engines. This sensor is insensitive to oxygen level over the range of 0–3 vol% hydrogen and 8–20% oxygen, exhibiting RMS error of 0.04 H_2% in this region. Linear cross-sensitivity to carbon monoxide at 0.392 H_2%/CO% is observed. Cross-sensitivity to propane, carbon dioxide, and nitrogen monoxide are deemed negligible. Once the effect of carbon monoxide is taken into account, readings of real engine exhaust match gas chromatography to RMS error of 0.02 H_2%, demonstrating that this sensor is suitable for measuring hydrogen in engine exhaust. *(Abstract-1)*

***Keywords-component*—Hydrogen; Dual-Fuel; Hydrogen Sensor**

I. INTRODUCTION

In the search to reduce worldwide greenhouse gas emissions, alternative carbon-free methods of powering economies and industries are being sought worldwide. While advances have been made in battery-electric and fuel cell technology, challenges remain [1]. Internal Combustion Engines (ICEs) running on a chemical fuel are likely to remain dominant in the transportation industry [2]. Currently, hydrogen is considered as a likely front-runner fuel for transportation applications [3].

Using hydrogen fuel in port-injected dual fuel mode is attractive for the current transportation industry. This technology can be retrofit to existing diesel vehicles [4]. With over 1.2 million medium and heavy-duty trucks in Canada [5] in 2022, replacing every truck engine would incur large economic cost. Further, when retrofitting an existing production diesel, adding a second hydrogen direct injector to the cylinder head can have challenges [6], making port-fuel injection attractive. Retaining the original diesel injectors would also allow seamless switching to diesel-only mode at any time if hydrogen is unavailable.

However, incomplete combustion of hydrogen fuel has been reported for port-injected dual-fuel diesel engines [7, 8, 9, 10]. Peak emissions have been reported on the order of 1% [7] and 2.5% [8] by volume in exhaust gas dependent on load. Similar results were reported for a port-fuel-injected spark ignition engine [11] and some fuel cells [12]. While molecular hydrogen is not toxic, global warming potential of hydrogen emissions due to effects on methane and ozone has been reported. Studies have estimated the 100-year warming potential of hydrogen to be 11.6 times that of carbon dioxide [13]. Hydrogen emissions will need to be controlled to realize the full greenhouse gas reduction potential of hydrogen fuel.

There are a great variety of methods used to quantify hydrogen in gas mixtures, with research ongoing [14, 15, 16, 17]. This topic is expected to become even more important with increasing usage of hydrogen in the world economy [14, 15, 16, 17].

This work focuses on the characterization and use of a commercial sensor, RKI Industries part number 752-04 (H_2 range 0–4%). It is based on the catalytic oxidation operating principle, intended for fuel cell applications, and claims no cross-sensitivity to several common gases in air [18]. Here the focus is on use with undiluted engine exhaust, it is tested for response to hydrogen and cross-sensitivity to several common species. Finally, real diesel engine exhaust is used to compare the sensor response model of CO sensitivity to results from gas chromatography.

II. SPECIES TESTED FOR CROSS-SENSITIVITY

Cross-sensitivity to several gases was considered, according to how likely they are to be in hydrogen-diesel dual-fuel exhaust. The list of test gases is presented in Table I. Water will be present in engine exhaust. To reduce the amount of water in the sample and to keep it consistent, exhaust samples will be cooled using an ice bath. Water vapor is therefore not included in this cross-sensitivity analysis. Carbon monoxide is well-known to affect some catalytic sensors [15, 14, 19,

20].There is a potentially large and variable amount of carbon dioxide present in dual-fuel exhaust due to combustion of injected diesel fuel. This gas is included for further testing. Hydrocarbons are known to affect some catalytic sensors [14, 15]. Unburned Hydrocarbons (UHC) are present in dual-fuel exhaust to varying degrees [21]. Here, a mixture of propane diluted in nitrogen is used to represent these emissions. Nitrogen oxides are present in dual-fuel exhaust in varying amounts [21, 22]. Here, a mixture of 1.96% NO in N_2 is used to represent NO_x emissions. The tested sensor is a catalytic combustion type. These are often affected by oxygen concentration [14]. Pure oxygen is mixed into the other gases to simulate oxygen-depleted engine exhaust. The remainder of the flow is made up of nitrogen gas.

TABLE. I
TEST GASES FOR FLOW RIG

Gas	Composition[a]
H_2	100% H_2
CO	9%CO, 45%CO_2, bal N_2
CO_2	100% CO_2
UHC	5060ppm C_3H_8, bal N_2
O_2	100% O_2
NO_x	1.96% NO, bal N_2
Diluent Gas	100% N_2

[a]Volume Basis.

III. CALIBRATION APPARATUS

To test the sensor for response to different gas mixtures, a flow rig setup capable of mixing up to 7 gases at different ratios was used. This flow rig contains seven mass-flow controllers and reads sensor response using an Arduino microcontroller. This equipment has been described in prior literature [23], but has been modified for automated testing.

MKS brand, GE50 series mass flow controllers are used, functioning on the thermal heat capacity measurement principle [24]. To correct for gas mixtures not included with the factory calibration, the data is later corrected to account for heat capacity of the gas mixture vs calibrated value. Seven controllers, ranging in full-scale flow of 200 Standard Cubic Centimeter per Minute (SCCM) to 10,000 SCCM, can be used in parallel. Each unit is rated to an accuracy of 1% of indicated flow, down to a minimum of 1% of full-scale [24]. Check valves are used on six of the gas streams to prevent backflow. This is omitted on the lowest-flow stream due to concerns that the cracking pressure (1/3 PSI) of the check valve would cause bumping and inconsistent flow rate. Tests are performed at 10,000 SCCM total flow. The lowest flow used is 2 SCCM of hydrogen, to characterize the deadband of the sensor. Depending on the test, inactive gas streams are isolated using motor-actuated ball valves. Each test point is held for approximately 2 minutes to allow the gas mixture and sensor to reach steady-state.

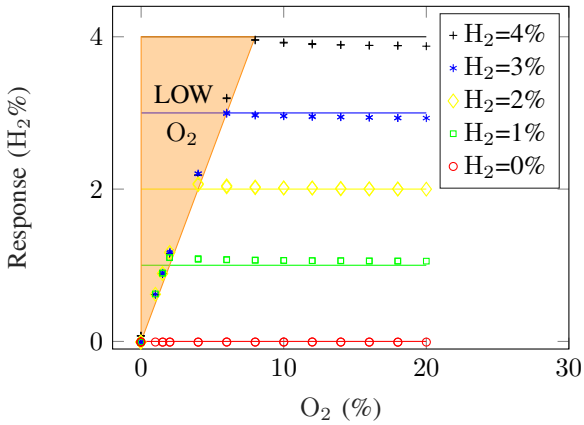

Figure. 1. Dependence of sensor output on oxygen concentration at different hydrogen concentrations

IV. RESULTS

A. Oxygen Depletion

Catalytic type sensors are affected by oxygen concentration [14]. This sensor is rated to give a correct reading between 8–20% oxygen, but is not rated to function properly under 8% oxygen [25]. The sensor readings for several, constant hydrogen concentrations at varying oxygen levels are presented in Fig. 1.

Oxygen depletion affects the maximum amount of hydrogen reported by the sensor. Lower concentrations (such as 1%) will report correctly even at relatively low (3%) oxygen. Higher hydrogen concentrations require more oxygen to report correctly. This effect can be seen in the leftmost edge of Fig. 1.

The purpose of testing this sensor, is for use in measuring hydrogen in engine exhaust. Studies have found peak H_2 exhaust emissions of 1% [8] and 2.5% [11] per volume. Restricting the operating range to 0–3% H_2 and 8–20% O_2, the RMS error of sensor output is 0.040% and maximum error is +0.069%. At the full scale of 4% H_2, error increases slightly (Fig. 1).

B. Full-Scale Hydrogen Response

Fig. 2 shows the response of the sensor to hydrogen concentrations from zero to full-scale (4 vol%). At all points, 17% oxygen is available for the catalytic reaction to take place. This sweep is performed once increasing hydrogen concentration, and once decreasing to give an idea of any sensor or setup hysteresis, which was not evident. Some non-linearity is present, as evidenced by a slight S-shape in the sensor response. Compared to a perfect response (IE 1% H_2 should read 1% H_2), RMS error is 0.056 H_2% and maximum error is 0.064 H_2%. This is similar to the RMS and maximum errors obtained by varying oxygen concentration between 8–20% O_2 with 0–3% H_2 in the previous section.

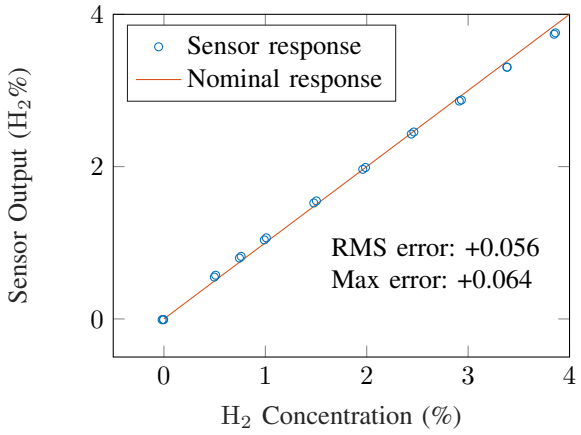

Figure. 2. 0-4% H_2 with 17% O_2

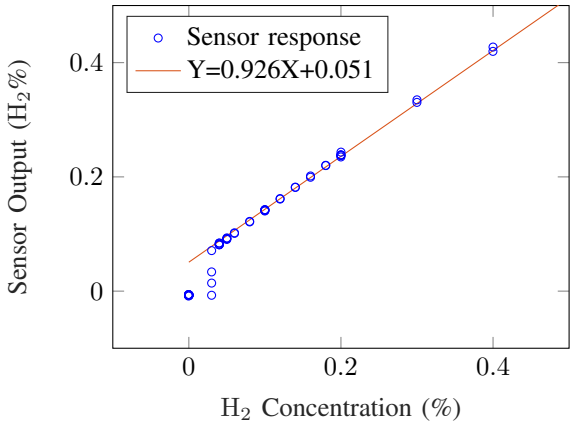

Figure. 3. Sensor Response, Low Range

C. Low Range Hydrogen Response

The sensor was also tested at low hydrogen concentrations of 0–0.5%. Hydrogen was added at 0–50 SCCM, with nitrogen and oxygen added to obtain a total flow of 10,000 SCCM with 14% oxygen, representative of low-load engine testing. The lower limit of the hydrogen mass flow controller is 2 SCCM (1% of full-scale) [24]. This lower limit corresponds to 0.02% hydrogen. Above this, the flow measurement accuracy is rated to 1% of reading. Ten voltage measurements are averaged at each point, and results are shown in Fig. 3. At zero hydrogen, the sensor reads approximately zero as expected. Of the points where the sensor indicates above 0.07%, which due to small offset occurs at actual H_2 concentrations above 0.03%, a linear response to hydrogen with a positive offset and slightly lower than unity response is observed. The presence of intermediate values under 0.07% is due to averaging ten samples at each point, and no instantaneous measurements between 0.000–0.047% were observed. Results are consistent with a sensor deadband, where small values are rounded down to zero, applied at the sensor output.

TABLE. II
SCREENING TEST LEVELS

Factor	Level (lo)	Level (hi)
H_2	0.1%	1.6%
CO	0ppm	1800ppm
CO_2	1%	11%
UHC (Propane)	0ppm	1000ppm
O_2	9%	17%
NO	0ppm	3000ppm

D. 1- and 2-Factor Interactions

Further screening for the effect of all 1- and 2-factor combinations of the 6 factors, using a 2-level experimental design, was performed. Each point was repeated 6 times for statistical significance. A 2-level experimental design was chosen to reduce the number of tests that need to be performed, relative to a 3-level design. Although a 2-level design cannot account for a nonlinear response to any individual factor, this was deemed acceptable for this screening test, as only oxygen depletion and low output ranges exhibited marked nonlinearity. At all times during this test there is a minimum of 0.1% H_2 and 9% O_2 to avoid these effects. Levels are given in Table II. For this cross-sensitivity test, the high level for hydrogen (1.6%) was chosen as representative of exhaust emissions [7], rather than the full scale of the sensor.

$$\hat{Y} = a_0 + a_1 X_1 + ... + a_{(1-2)} X_1 X_2 + ... + a_{(5-6)} X_5 X_6 \quad (1)$$

The data was fit, using Linear-Least-Squares regression, with all 2-factor interaction effects considered (Eqn.1). Here a_n represents the model coefficients, X_n represents normalized gas concentration, and $\hat{Y}$ represents normalized sensor output. Before regression, gas concentrations are normalized such that low range corresponds to zero and the high range corresponds to one. Sensor output is also normalized to the levels for hydrogen (0.1–1.6%). This allows a direct comparison of the magnitude of different model coefficients. The resulting model coefficients, with confidence intervals and P-values, are listed in Table III. RMS error, converted from normalized units to H_2%, is 0.003 H_2% and the maximum absolute error is 0.018 H_2%.

In Table III many of the model terms have P-values above 0.05, indicating low significance. Also, many terms have low magnitude, indicating a small effect on the sensor output. Sensor response is normalized to a 1.5% span (Table II) and therefore a normalized model coefficient of $a_n < 0.003$ results in a change in sensor response of under 0.005 H_2%, deemed negligible for the intended application. Those that have P<0.05 and $a_n < 0.003$ are: Offset, H_2, CO, H_2-CO_2, H_2-UHC, H_2-O_2, and H_2-NO_x. These cross-sensitivities and interaction effects are described below.

E. CO Sensitivity

As shown in Table III the main cross sensitivity is to Carbon Monoxide. Cross sensitivity of some types of catalytic sensors to carbon monoxide is well known [14, 19], although there

TABLE. III
SCREENING TEST MODEL COEFFICIENTS

Effect	Coefficient (Normalized)	Confidence (95%)	P Value
Offset	+0.030	+/-0.001	0.000
H_2	+0.967	+/-0.002	0.000
CO	+0.047	+/-0.002	0.000
CO_2	-0.002	+/-0.002	0.008
UHC	-0.000	+/-0.002	0.406
O_2	-0.001	+/-0.002	0.279
NO_x	-0.003	+/-0.002	0.001
H_2-CO	-0.003	+/-0.004	0.050
H_2-CO_2	-0.010	+/-0.003	0.000
H_2-UHC	+0.007	+/-0.003	0.000
H_2-O_2	-0.008	+/-0.003	0.000
H_2-NO_x	+0.021	+/-0.002	0.000
CO-CO_2	-0.002	+/-0.004	0.106
CO-UHC	-0.001	+/-0.004	0.336
CO-O_2	-0.000	+/-0.004	0.426
CO-NO_x	+0.001	+/-0.002	0.218
CO_2-UHC	+0.000	+/-0.003	0.393
CO_2-O_2	-0.000	+/-0.003	0.371
CO_2-NO_x	+0.000	+/-0.002	0.477
UHC-O_2	+0.000	+/-0.003	0.443
UHC-NO_x	-0.001	+/-0.002	0.260
O_2-NO_x	-0.000	+/-0.002	0.472

are some catalytic sensors that have low cross sensitivity to CO [26].

The effect of adding CO-containing calibration gas to a mixture containing hydrogen at various concentrations and consistent 14% oxygen is shown in Fig. 4. This gas also contained CO_2, as would real engine exhaust. The plotted results are the difference between sensor response at zero CO, and the results with added CO. Linear least-squares fits are presented in Table IV for different H_2 concentrations.

A linear cross-sensitivity to CO is seen in contradiction to the sensor datasheet [18], although this is limited to just over 0.1% H_2 at 3500ppm CO. Further, experience running a hydrogen-diesel dual-fuel engine shows that most useful points are under 1,000ppm CO. At 1,000ppm, the effect of CO on sensor reading is around 0.035 H_2%, under the 0.056% RMS error in Fig. 2. The cross-sensitivity varies with hydrogen concentration, indicating an interaction effect, but this is again considered small for the intended use, supporting the use of this sensor for detecting hydrogen in engine exhaust.

In the screening test (Table III), carbon monoxide has a model coefficient of 0.047. This is normalized to an expected sensor response of 1.5 H_2% and CO concentration of 1800ppm (Table II) and therefore the screening test results in a CO slope of 0.392 H_2%/CO%. This is similar to, but slightly different than Table IV. Observed interaction effects between CO-CO_2 from Table II are not enough to explain the discrepancy. However, at 1800ppm CO the difference in sensor response between these two scale factors can be calculated to be about 0.015 H_2% which is considered small for the intended engine application.

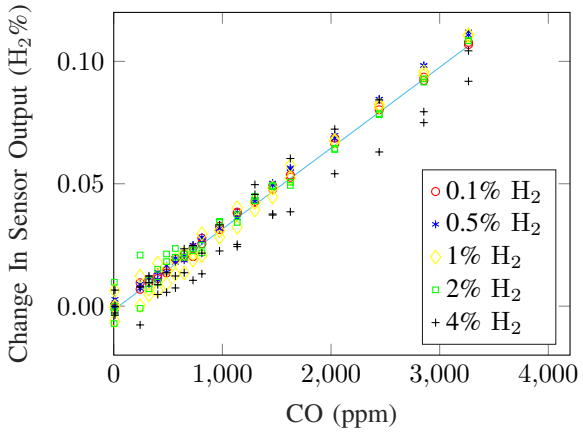

Figure. 4. Cross-sensitivity to CO at different hydrogen amounts and 14% O_2

TABLE. IV
CO CROSS-SENSITIVITY AT DIFFERENT H_2 LEVELS USING 9%CO + 45% CO_2 CALIBRATION GAS

H_2 (%)	CO **Slope** (H_2%/CO%)	**Confidence (95%)**	**P Value**
0.1	0.331	+/-0.004	0.000
0.5	0.347	+/-0.004	0.000
1	0.345	+/-0.013	0.000
2	0.323	+/-0.014	0.000
4	0.306	+/-0.026	0.000

F. UHC, CO_2, NO, H_2-NO_x *Response*

To further confirm a lack of sensitivity to UHC, CO_2, and NO, testing was performed where the concentration of each of these components was swept with a minimum of 0.1% H_2 present to avoid sensor deadband behavior. A slightly larger range for NO_x (3500ppm) and CO_2 (15%) was available for this test, and again UHC (propane) was varied 0–1,000ppm. The NO_x test was repeated with 1.6% H_2 to look for H_2-NO_x interaction as suggested by the screening test above. Similar to Fig. IV, the plotted values represent the difference with and without the gas under study, with results presented in Fig. 5. Response to UHC is slightly positive, but always within 0.005%. CO_2 creates a slight downward trend except for the last point, but is again bounded to 0.005%. NO_x at high hydrogen appears to go have a slight upward trajectory while at low hydrogen having a slight downward trajectory indicating the presence of a small, positive H_2-NO_x interaction. However the difference between maximum and minimum H_2-NO_x effect is only 0.025 H_2%, considered negligible for the intended application.

G. Simplified Linear Model

$$\hat{Y} = a_{\text{Off}} + a_{\text{H2}} X_{\text{H2}} + a_{\text{CO}} X_{\text{CO}} \tag{2}$$

A reduced-order model, only considering the effect of Offset, H_2, and CO, was fit to the same data as Table III. Again the data was normalized to the values in Table II. This model is

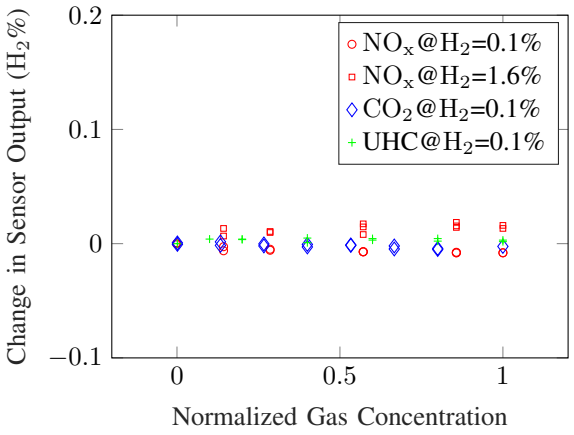

Figure. 5. Cross-sensitivity to: NO_x 0–3500ppm, CO_2 0–15%, UHC (Propane) 0–1000ppm

given in Eqn.2. Normalized coefficients are given in Table V. This dataset contains different combinations of gases, but the model only accounts for three variables. The effect of the other combinations of gases will therefore add to the observed error. In units of indicated hydrogen, a_{off} = 0.042 H_2%, and a_{CO} = 0.392 H_2%/CO%. RMS error is 0.009 H_2%, and the greatest single deviation from the model was 0.036 H_2%.

TABLE. V
SIMPLIFIED LINEAR MODEL COEFFICIENTS

Effect	Coefficient (Normalized)	Confidence (95%)	P Value
Offset	+0.028	+/-0.001	0.000
H_2	+0.975	+/-0.002	0.000
CO	+0.047	+/-0.003	0.000

H. Cross-Sensitivity to real engine exhaust

While this sensor shows limited cross-sensitivity to the tested gasses at engine-relevant concentrations, real engine exhaust can contain a wide variety of compounds. Therefore, this sensor was tested for response to real exhaust, and the results compared to the simplified linear model.

To test the sensor, a Cummins 4.5L naturally-aspirated engine was run with diesel only and no added hydrogen. Engine exhaust can include hydrogen, especially in spark-ignition engines [27], but also in diesel engines [28]. Therefore two samples were collected and analyzed using Gas Chromatography. No hydrogen was detected.

The engine was then run at several more points producing varying CO levels. Sensor response, relative to the CO level, is plotted in Fig. 6. The expected value from the reduced-order linear model, assuming zero hydrogen, is also plotted. At low CO levels, the sensor is within the deadband range and outputs zero. The points that are above deadband were compared to the reduced linear model, with results presented in Table VI. The RMS error is under 0.02 H_2% and the maximum absolute error is 0.0346 H_2%, considered acceptable for the intended application.

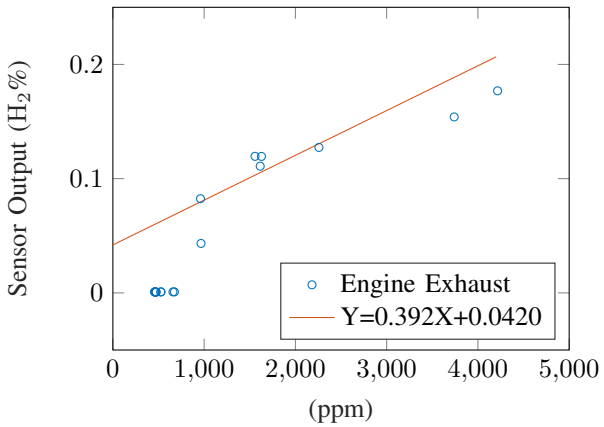

Figure. 6. Real Engine Exhaust Response

TABLE. VI
RESPONSE TO REAL ENGINE EXHAUST VS MODEL

Error	Magnitude (H_2%)
RMS	0.0194
Max (+)	0.0346
Max (-)	-0.0165

V. CONCLUSION

Sensing of hydrogen in the presence of other gases can be achieved by many different methods, and this field remains an area of active research. A commercial hydrogen sensor, based on the catalytic oxidation method, was comprehensively tested.

A high-accuracy flow rig was used to determine response to hydrogen and 5 other gases likely to be present in engine exhaust. Between 0–4% H_2, comparing the sensor output to actual concentration shows an RMS error 0.056 H_2%. This sensor also exhibits deadband, where sensor output values under 0.07 H_2% are rounded to zero. Due to the catalytic combustion operating principle, a minimum oxygen concentration is required for accurate reading. This minimum is dependent on the hydrogen concentration. Within the range of 8–20% O_2 and 0–3% H_2, the RMS error of sensor output vs actual hydrogen concentration is 0.040 H_2%.

A 2-level, 6-variable model with interaction effects found that many cross-sensitivities and interactions were not significant (P<0.05) or of negligible magnitude relative to the intended application. This was confirmed by further testing. A simplified linear model was then fit accounting for only sensor span, CO cross-sensitivity, and offset. This model achieved RMS error of 0.009 H_2% on the dataset. Linear CO cross-sensitivity was found. Despite the sensitivity of 0.392 H_2%/CO%, due to the expected values of CO this effect is small at most dual-fuel engine operating points. At 1,000ppm CO, using the simplified linear model, this will affect results by 0.04 H_2%.

The sensor was used to measure real diesel engine exhaust, under the tested assumption of zero hydrogen in the exhaust. Of the points where sensor response was above the sensor

deadband, RMS error between the simplified linear model and sensor response was under 0.02 $H_2\%$.

Based on the testing completed, the RKI 752-04 sensor is suitable for use for measurement of unburned hydrogen in engine exhaust. This will allow for evaluation of the amount of hydrogen slip in dual-fuel or 100% hydrogen engine exhaust, which can be an important contributor to total greenhouse gas emissions. This low-cost sensor is portable and could be used to monitor engine out hydrogen emissions as this technology is developed and deployed to the market.

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