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On the Applicability of Silicon Carbide Based Field Effect Sensors in the Control of Exhaust/Flue Gas After-Treatment Systems †

Mike Andersson *, Lida Khajavizadeh and Anita Lloyd Spetz

Division of Sensor- and Actuator Systems, Department of Physics, Chemistry, and Biology, Linköping University, SE-581 83 Linköping, Sweden; lida.khajavizadeh@liu.se (L.K.); spetz@ifm.liu.se (A.L.S.)

* Correspondence: mike.andersson@liu.se; Tel.: +46-76-3086422

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Abstract: The performance of Silicon Carbide based field effect gas sensor devices, modified to enable long-term reliable operation with improved sensitivity to e.g., NH₃ and CO at relevant temperatures for exhaust/flue gas emissions monitoring and control of combustion processes as well as after-treatment systems in automotive/stationary applications has been investigated with promising results.

Keywords: Field Effect Sensors; Silicon Carbide; MOSFETs; Combustion Control; Process Control; Emissions Monitoring; Exhaust after-treatment; Flue Gas Cleaning

1. Introduction

As emissions limits are tightened, both in relation to transportation and domestic/local heat/power production, and their fulfilment also under real-world conditions (not only for type approval) being enforced, close control of both the combustion process as such and the exhaust/flue gas after-treatment systems is required. In order to ensure efficient combustion and after-treatment system control under different conditions as well as its proper function over time, accurate and reliable monitoring of ever smaller concentrations of exhaust/flue gas components such as nitrogen oxides (NOₓ), hydrocarbons (HCs), carbon monoxide (CO), oxygen (O₂) and ammonia (NH₃) is required. Generally being fairly small-scale but large-volume applications the means, besides exhibiting accuracy and long-term reliable performance, also have to be cost-efficient, thereby rendering in-situ positioned solid-state sensors one of the currently most attractive options.

In addition to solid electrolyte based gas sensors, such as the lambda-, wide band oxygen- and NOₓ-sensors [1] in commercial use today, and the resistive-type metal oxide semiconductor sensors (e.g., the TiO₂ based MOS sensor applied as lambda-probe in automotive engine control) also Field Effect Transistor (FET) based sensors employing the wide band gap semiconductor Silicon Carbide (SiC), see Figure 1, have, at least among economically viable solid-state sensors, been considered for emissions monitoring in automotive applications (SiC FET sensors for combustion control are commercially available for certain applications) [2]. An example of a basic SiC FET gas sensor is displayed in Figure 1a. By applying as gate contact in the MOSFET (Metal Oxide Semiconductor Field Effect Transistor) device a material which upon interaction with one or more gaseous substances can give rise to a modulation of the gate to substrate electric field, FET based gas sensors can be fabricated. When, for example, a MOSFET device employing a platinum (Pt) gate is exposed to a hydrogen-containing substance, atomic hydrogen may be generated through dissociative adsorption on the catalytic metal surface and subsequently diffuse through the thin Pt film to adsorb on top of oxygen atoms in the surface of the oxide, see Figure 1b. The resulting hydroxide groups give rise to a change
in polarization of the metal/oxide interface, affecting the gate-to-semiconductor electric field. This hydrogen-mediated direct detection of hydrogen-containing substances, such as hydrocarbons and ammonia, is an attractive feature as the field-effect sensors, in contrast to sensors based on oxygen-mediated indirect detection of these substances (through red-ox reactions consuming oxygen anions), can be easily rendered insensitive to changes in e.g., exhaust and flue gas O$_2$- and NO$_x$-levels and so exhibit better selectivity to e.g., ammonia (facilitating better separation between ammonia and NOx than the solid electrolyte-based NO$_x$ sensor in widespread use today). As for the mechanisms behind the detection of CO employing FE sensors, see [2]. The change in gate-to-semiconductor electric field in turn modulates the number of charge carriers in the top part of the semiconductor and thus the Ids/Vds-characteristics of the device (Figure 1c). Either the change in drain current upon gas exposure when supplying a constant voltage or vice versa is measured.

Following developments over the last few years in improving the sensor device materials’ stability at elevated temperatures [3], the recent improvements in ammonia detection performance also at temperatures above 450 °C, and specifically a growing interest in selective ammonia monitoring for fine-tuning the control of the seemingly most viable NO$_x$ reduction measure—urea-SCR (Selective Catalytic Reduction), see Figure 2—currently implemented in both automotive, marine, and stationary applications, it has been of interest to explore the long-term performance/applicability of these SiC FET based NH$_3$ sensors in ammonia slip monitoring (downstream the SCR catalyst). Analogously, with the increased interest in sensor-based combustion control in small-scale boilers (/stoves) in local/domestic heating as well as the production of power/process-heat in small- to medium-scale systems for improved efficiency and reduced emissions, the long-term performance of the low-cost SiC FET-based CO sensor in the real environment of these applications has also been of interest to evaluate. This short report therefore aims at giving an overview of the long-term performance of the respective SiC FET NH$_3$ and CO sensors when evaluated in the real applications (the real environments) over periods of 6 months or more.

2. Materials and Methods

In order to evaluate the long-term performance of SiC-based field effect sensors in real-world (in-situ) on-line flue gas CO and exhaust NH$_3$ slip monitoring for the control of combustion in domestic heating systems/process-heat production as well as fine-tuning of the SCR NO$_x$ reduction
process, respectively, CO- and NH₃-sensor devices (transistor chips) have been encapsulated in a ceramic packaging and placed inside a protective housing, see Figure 2, to facilitate easy mounting in the exhaust system/flue gas channel. In total three NH₃ sensor units have been mounted downstream the SCR catalyst of a marine diesel engine on-board a ferry in continuous operation, and connected to the necessary driver and data acquisition electronics. Analogously three CO-sensor devices have been mounted in the flue gas channel of a wood-fired boiler for residential heating and another two CO-sensors in the flue gas channel of a burner for the production of process heat.

Figure 2. Displays in (a) the sensor chip (inset) encapsulated in an LTCC-based ceramics packaging, in (b) the sensor housing inside which the ceramic sensor element is residing, and in (c) the electronic unit for driving and acquiring the sensor data during the real-world field tests.

Two weeks after mounting of the sensors, the sensor units went through a zero-point calibration. Over a period of approximately 4 h, the signals from the respective sensor were at this point also recorded along with the readings from two separate reference instruments to confirm the CO/NH₃ sensing performance and establish the sensitivity of each sensor. For the ammonia slip measurements a near-infrared diode laser was used and for the CO measurements a Testo 340 instrument (based on electrochemical measurements of the CO concentration). After calibration and reference measurements the sensor signals for all the tested sensors were recorded over a period of at least 6 months and were evaluated regarding stability, both in terms of zero-signal level and the sensitivity (i.e., the change in sensor signal output as a result of the change in concentration of the respective substance). Even though the sensors were continuously operated during the entire test period, reference measurements were only made on some regular occasions during the tests.

3. Results and Discussion

As can be seen from Figure 3, the NH₃ slip sensor exhibited good, stable sensitivity to NH₃ in the 0–50 ppm range throughout the 6-month test, with good resolution in the single-digit ppm range. When temporarily switching off the NH₃ (urea) dosing, a situation displayed in Figure 3b, the NH₃ sensors exhibit basically no variation in the output signal, thus being unaffected by e.g., variations in exhaust NOₓ concentrations. From the measurements an uncertainty level of +/- 3 ppm for the whole test period could be calculated. Figure 4a displays the variation in sensor signal to variations in flue gas CO concentration, as recorded by the Testo reference instrument, over a period of approximately 20 min. As can be seen the sensor signals follow with fairly good accuracy the variations in CO concentration and with excellent response and recovery times. Figure 4b gives an account on the long-term stability of the CO-sensor over a period of 200 days during the last approximately 7 months of a 12-month test period. The sensitivity, as evaluated through comparisons between the sensor signal and the signal from the reference instrument, exhibits good stability over the whole evaluation period, while a slight change in the zero output signal can be observed. The change over the entire 200 day period would correspond to an approximate uncertainty of 30 ppm for the read sensor signal (the sensor reading would give an approximate 30 ppm lower concentration reading than the real CO concentration).
Figure 3. Displays in (a,b) the variation in sensor signal when the ammonia slip is building up downstream the SCR catalyst (in the range 10–40 ppm) during ~15 min constant urea dosing, 3 weeks and 4 months after sensor start-up, respectively. In (c) the variations in sensor signal during a period of no urea dosing are displayed.

Figure 4. Displays in (a) the variation in the output sensor signal for the SiC FE based CO sensor together with the output signal from the Testo reference instrument, and in (b) the long-term stability of the sensor performance as evaluated through observed changes in zero output signal and sensitivity over time.

4. Conclusions

Exhibiting promising long-term performance, especially regarding the excellent stability in sensitivity (the change in sensor output resulting from a certain change in gas concentration), over the 6- to 12-month test periods, and especially for ammonia slip monitoring in the SCR control application also showing good selectivity to ammonia over nitrogen oxides, Field Effect sensors may offer an alternative to other sensor technologies in small-scale combustion and exhaust after-treatment system control applications.
References


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