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Low-power sensor node for the detection of methane and propane

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Abstract. The detection of flammable gases is necessary to avoid explosive atmospheres. For this reason, low-cost pellistors are frequently used. However, such commercial pellistors require an operation temperature of 450 °C or more for the detection of methane and a correspondingly high power consumption. We present a novel wireless low-power catalytic gas sensor system based on non-precious metal catalyst for the detection of methane and propane operated at 350 °C. The combination of a microelectromechanical system (MEMS)-based sensor with a low-power radio system provides the opportunity to monitor complex infrastructure without using a power grid as power supply. The sensor system has been characterised extensively under the exposure to methane and propane at concentrations between 2000 and 8000 ppm, as these gases are the common test gases for pellistors in industry. Methane is the main component of natural gas; propane is an important component of liquified petroleum gas (LPG). In addition, the influence of changes in humidity on the sensor response to methane was examined in more detail. Due to the planned operation of the sensor and radio system in different application scenarios, short (3 s) and long (60 s) sampling rates were used for investigations.

1 Introduction

The early detection of flammable gases or explosive gas mixtures is extremely important to avoid endangerment of people and damage to infrastructure. Catalytic sensors for flammable gases, so-called pellistors, are sold in high numbers and used at gas supply facilities and filling stations, as well in the private sector for gas heaters and installations. Due to their continuous energy consumption between 300–500 mW (Bíró et al., 2014), conventional pellistors can only be operated on the mains supply, making the installation of a sensor network complex and expensive. In large industrial plants, the major cost drivers are wiring and maintenance not the sensor costs themselves (Somov et al., 2013). Significantly lower energy consumption would enable sensor nodes for the detection of flammable gases or explosive gas mixtures independently of the power grid. The energy consumption of pellistors can be lowered by changing the sensor design from classical "bead-type" sensors to microelectromechanical system (MEMS)-based planar devices. Conventional bead-type pellistors with their large thermal mass require a long heat-up phase until they reach steady-state conditions and are ready for the measurement task. Due to the lower power consumption and smaller thermal mass, several research groups have already investigated MEMS-based planar pellistors. Some publications described the reduction of power consumption (Bársony et al., 2009; Su et al., 2013; Trautweiler et al., 2012) and others present the possibility of a non-steady-state measurement (Fricke et al., 2014). Nevertheless, all MEMS sensors described so far must be operated at temperatures ≥ 400 °C, and they use conventional noble metal catalysts for the detection of combustible gases. However, the working temperature is an important additional factor for reducing the response time and power consumption of a catalytic sensor. Thus, at high operation tempera-

tures (> 400 °C), the heat loss from the sensor to the environment due to thermal conduction of the air is one of the main reasons for the high power consumption of pellistors (Ma et al., 2013); this also includes micro pellistors. Therefore, in addition to the MEMS approach the presented sensor was operated at lower operation temperature by use of a noble metal-free catalyst. Among the transition-metal oxide catalysts, cobalt oxide catalyst exhibits the highest activity in catalytic combustion of hydrocarbons (Stein et al., 1960). For the experiments described below, nickel-doped cobalt oxide with an Ni to Co molar ratio of 1 to 2 (termed as $NiCo_2O_4$) was used as the functional material, which is also reported to have excellent properties for the low-temperature oxidation of methane and propane (Tao et al., 2015; Ren et al., 2016). Of course, the power consumption of the evaluation electronics and wireless communication must also be as low as possible. In the following, we present our work on a novel combination of a long-range low-power radio system with a newly developed low-temperature planar pellistor for combustible gas detection.

2 Wireless gas sensing system

2.1 Description of the radio system

The main challenge of the gas sensor system is connection of the sensor to the energy management system and the radio system (Fig. 1), which should be as efficient as possible in order to achieve long service life, real-time capability, less maintenance, long-range capability, robustness, and mesh network capability.

For this purpose, a radio system was developed and evaluated, which uses a unique wake-up strategy to ensure that all sensors can be reached by radio permanently and in real time, but still has a very low energy consumption (Gamm et al., 2010). In contrast to current wireless communication systems (WLAN, Bluetooth, ZigBee), which minimise their energy consumption through periodic deactivation and activation (Kajikawa et al., 2016), this system allows for continuous real-time accessibility with low latencies. Since the individual sensor nodes also communicate with each other, measurement data can be recorded in star or multi-hop topology, if required. This offers the opportunity for an almost unlimited distance coverage in networks and opens the way for a significant reduction of costs. The radio system is robust, simple, efficient, maintenance free, and could be energy self-sufficient by using energy-harvesting technology. Even the radio protocol has been designed to be particularly robust and immune to interference for use in a safety-critical environment, where each measurement value is confirmed bidirectional at the receiver (Saez et al., 2017). The novel combination of these features with the low-power gas sensor enables the installation of the measuring system in inaccessible places as well as usage in a battery-powered mobile environment.

2.2 Gas sensor and catalytic material

The second elementary component is the gas sensor. By using a MEMS-based hotplate, the power consumption of the sensor module (one active and one passive sensor) decreases to approximately 110 mW (Walden et al., 2015). This enables the use of the sensor in mobile applications. Because the suspended MEMS hotplate reaches the working temperature in less than 15 ms, a fast cycling or a fast thermal response is possible. This provides the opportunity for a deep sleep and a fast sensor wake-up, if necessary. The sensor is operated in a Wheatstone bridge to compensate disturbances such as airflow, ambient temperature, and changes in the thermal conductivity of the surrounding medium. One leg of the circuit is set by two 10 kΩ low-temperature-coefficient passive resistors. The other leg consists of two sensing MEMS pellistors. One pellistor is coated with the catalyst and the other serves as reference. A low-noise instrumentation amplifier amplifies the differential output voltage of the bridge.

In addition to the reduction of the power consumption by using a MEMS-based suspended hotplate, the working temperature, which is currently between 400 and 500 °C for commercially available sensors, could be decreased to 350 °C by the use of a novel catalyst material. Additionally, Walden et al. (2015) have characterised the sensor platform in detail, and they could show that lowering the working temperature by 100 K reduces the power consumption by 16 mW. The sensor requires about 71 mW at 450 °C and 55 mW at 350 °C. This reduces the power consumption by almost 23 %. The approximately linear behaviour in this temperature range depends on the dominant thermal conduction and the negligible thermal radiation. Commercial pellistors often use noble metal catalysts (Pd, Pt) dispersed on alumina supports with working temperatures exceeding 450 °C. This is necessary to reliably detect methane, which is the most inert combustible gas. In contrast to conventional pellistors, we used a non-precious metal oxide as catalyst. Tao et al. (2015) and Shao et al. (2017) described the use of $NiCo_2O_4$ as a good candidate for a complete methane oxidation in a temperature region between 300 and 400 °C. The gas-sensitive element consists of a low-temperature catalyst without using any precious metal. $NiCo_2O_4$ with a Ni : Co molar ratio of 1 : 2 was synthesised by co-precipitation according to the report by Tao et al. (2015), whereby the catalyst with a nanosheet morphology (Fig. 2, grey) was obtained as verified by scanning electron microscopy (SU-70 DEM). Subsequently, the $NiCo_2O_4$ catalyst was wet ground for 10 h in a steel jar with 2 mm steel balls to obtain an inkjet-printable material. The ground catalyst contains both the original nanosheets and the crushed particles (Fig. 2, blue), which should be beneficial to obtain a high surface area. A laser particle size analysis (LB-550, Horiba) confirmed the partial decreasing in particle size due to the grinding process of about 35 %.

The inkjet printing on the MEMS-based hotplate was performed with a DMP 2831 printer (Dimatix FUJIFILM,

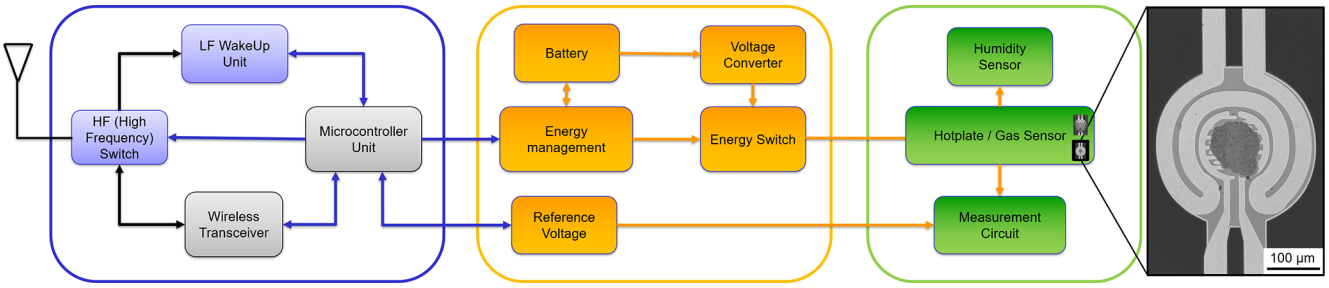

Figure 1. Detailed view of the measurement system. The radio module (blue part) manages the communication to the host, controls the energy management (orange part), and activates the sensor module (green part) if needed. The right image shows a magnified view of the sensor. The MEMS-based pellistor has a diameter of 320 µm with a meandering platinum heater strip located at the outer area of the hotplate. The black dot in the centre of the hotplate represents the inkjet-deposited catalytic material.

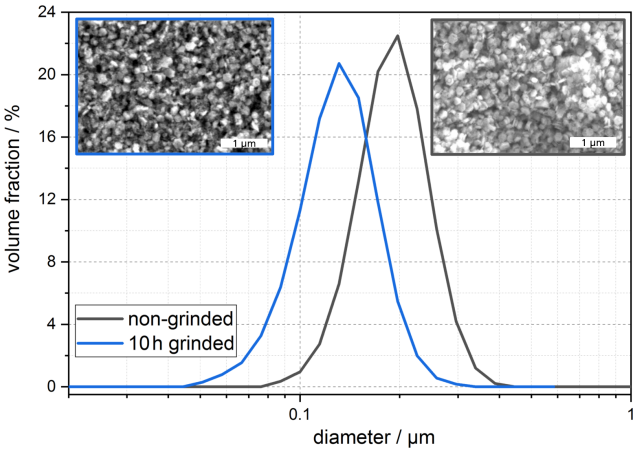

Figure 2. SEM images of (grey) the synthesised $NiCo_2O_4$ catalyst and (blue) after grinding for 10 h. The grinding is necessary to obtain a particle size distribution compatible with the inkjet printer used. The graph shows the shift to smaller particle sizes after the grinding. The peak maximum shifts from about 0.2 to 0.13 µm.

2021). As shown in Fig. 1, only parts of the hotplate were covered with the catalytically active ink.

3 Results

In this section, the results of the different sensor experiments are presented. The experiments focus on the sensor sensitivity, the optimisation of the battery lifetime, and possible sensor readout strategies concerning the measurement frequency. Furthermore, the effect of humidity on the sensor response is discussed.

3.1 Sensor sensitivity

For the monitoring of flammable gases, a reliable detection of the lower explosion limit (LEL) is necessary. To report leakages or to evacuate people from harmful areas, 10 % of the LEL of methane should be detected. Here we present a gas-sensing device optimised for methane detection. Figure 3a presents the measured data for three different gas concentrations below the LEL of methane of 4.6 % (de Smedt et al., 1999). Figure 3b demonstrates that the sensor responds also to propane. The response to propane at the same concentration is about 18 % higher than to methane due to higher combustion enthalpy of propane. At a low working temperature of 350 °C (Fig. 3a), the sensor shows a stable baseline, and due to the high signal-to-noise ratio (SNR), concentrations far below the LEL of methane (< 5 % LEL) could be detected. The measurements were performed in a gas-measuring chamber with a volume of 1 L and a gas flow of 250 sccm. A general description of the gas measurement station is given by Kneer et al. (2014). The t_{on} time of the sensor was 500 ms with a sleep time of 3 s. Therefore, the duty cycle is about 14 %.

The sensor shows a response of approximately 5.5 mV to 8000 ppm CH_4 in dry synthetic air. This is equal to a response of 6.9 mV per % CH_4. Independent of supply voltages and mechanical boundaries, the literature describes sensor responses at higher temperatures. Xu et al. reported about 2 mV per % CH_4 at 400 °C utilising 15 wt % Pd-Pt catalyst on γ-Al_2O_3 (Xu et al., 2011). The response of MEMS sensors with Rh_2O_3-Al_2O_3 catalyst at 400 °C is in the same range, presented by Su et al. (2013). As a result of lower working temperature, the probability that the pellistor will serve as an ignition source is substantially reduced (Papp et al., 2013). These gases were chosen because they are the common test gases for pellistors in industry. Methane is the main component of natural gas; propane is an important component of liquified petroleum gas (LPG) (Jessel, 2001).

3.2 Battery lifetime optimisation

After receiving qualitatively good results from the gas measurements, further optimisation of the battery lifetime was carried out. With a t_{on} time of 500 ms for the gas sensor, the sensor node needs about 74 mAs of electric charge per measurement at a battery voltage of 3.2 V. This equals 237 mWs. The share of the gas sensor is 86 % (205 mWs) of the whole

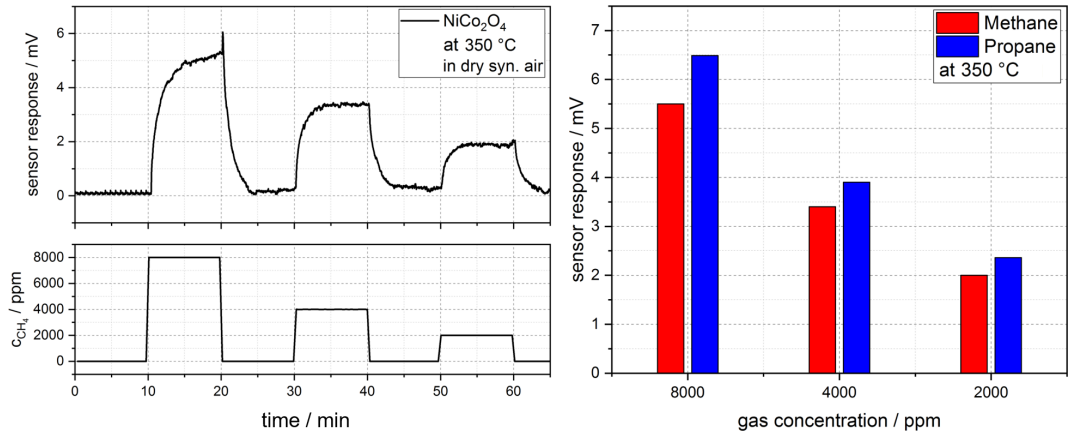

Figure 3. **(a)** Response of the gas sensor to different methane concentrations below the LEL in dry synthetic air at an ambient temperature of 25 °C. Even 2000 ppm methane (< 5 % LEL) could be clearly identified. The graph shows the sensor response without amplification. **(b)** Comparison of the sensor response to methane and propane in dry synthetic air. Under the same conditions the sensor shows an about 18 % higher response to propane.

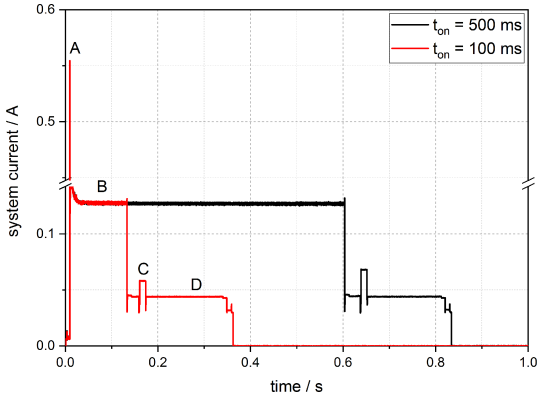

Figure 4. Comparison of different t_{on} times of the gas sensor. Due to the fast heating of the MEMS-based hotplate, the t_{on} time could be decreased to 100 ms. This increases the battery lifetime significantly. The runtime of one measurement cycle can be divided into four stages. (A: system start up; B: sensor heating time; C: data processing; D: communication with the gateway)

energy consumption. Only 14 % (32 mWs) is required for the microcontroller, the radio, and the peripheral circuits. This gives us the opportunity to measure more than 116 000 cycles with the built-in 2400 mAh (7.68 Wh) battery. With a sampling rate of 3 s, the system can work for more than 4 d. Commercially available portable pellistor sensors achieve several hours of measurement time to date. The sensor's battery lifetime could be further increased by setting the t_{on} time to a smaller value. Due to the short heating time of the hotplate, the t_{on} time could be set to 100 ms, which guarantees a stable supply voltage of the sensor (after 33 ms) and a readout of the analogue-to-digital converter (ADC). Figure 4 shows the difference in the energy consumption of the sensor system.

With a t_{on} time of 100 ms, only 51 mWs are required to heat up the gas sensor. The energy consumption for the other parts remains constant at 32 mWs. The total demand of electrical energy can be reduced to 83 mWs, which is only 35 % of the initial value. A 2400 mAh (7.68 Wh) battery would now last more than 332 000 cycles. This equals an operation time of more than 11 d with a measuring interval of 3 s.

The runtime of one measurement cycle can be divided into four stages.

- A: system start up with higher current of the voltage controller;
- B: heating time of the gas sensor with readout of the ADC;
- C: processing and transmitting of the collected data;
- D: handshake with the gateway.

The stages A, C, and D are fixed in their power consumption for this sensor scenario. Due to the time variation of B, the power consumption could be decreased. For some application scenarios (e.g. non-safety applications such as environmental sensing), the measurement interval could easily be changed from seconds to minutes. In Fig. 5, we present an exemplary measurement of 8000 ppm CH_4 in dry synthetic air, which equals ≈ 20 % of the LEL, with two different sampling rates of 3 and 60 s.

The sensor response is not affected by the duty cycle change of the system. The different sampling rates show almost the same results and raise time to the changing gas matrix. With this knowledge, it is possible to increase the battery lifetime by adapting the duty cycle factor. If the measurement interval is doubled, the battery lifetime increases by a factor of 2 as well. A measurement interval of 60 s leads to a sensor node runtime of more than 220 d and shows no loss

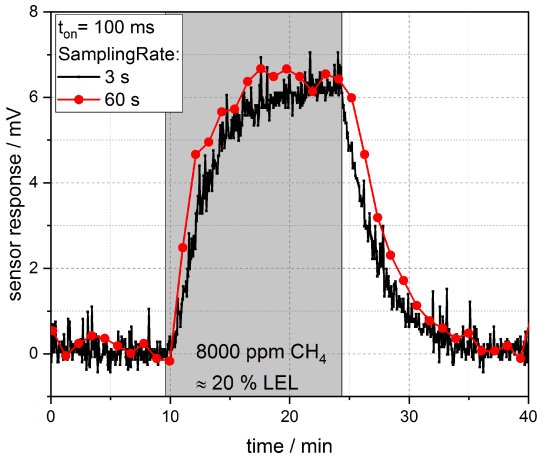

Figure 5. Sensor response to 8000 ppm methane at a sensor temperature of 350 °C and a sampling rate of 3 and 60 s in dry synthetic air at an ambient temperature of 25 °C. The implementation of different application scenarios in relation to the required measurement interval and the desired battery lifetime is shown.

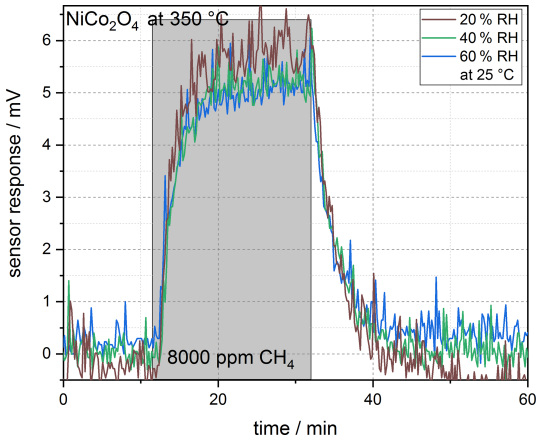

Figure 6. The influence of different relative humidity levels on the gas sensor response to 8000 ppm CH_4 in synthetic air at an ambient temperature of 25 °C. The variation of the measurement results is quite low, so the change in humidity has only a minor influence on the sensor signal.

of information for each single measurement compared to the measurement interval of 3 s. In this way, different work scenarios can be covered: a fast-repeating measurement cycle for safety applications or a long runtime mode for air quality sensing were less measurement cycles are required.

3.3 Effects of humidity

The humidity in the gas matrix is a major factor influencing the pellistor sensor response. Water molecules compete with the reaction partners for the catalytic surface, which reduces the active surface and thus the sensor response. In Fig. 6, the influence of three different relative humidity levels on the sensor response is shown. For these measurements, the t_{on} time was set to 100 ms and the sampling rate to 3 s. The operation temperature was 350 °C.

The measurements results show that, at an exposure of 8000 ppm CH_4 in synthetic air, the influence of a change in relative humidity is not significant. Within a relative humidity level change between 20 % and 60 % at 25 °C, the response decreases by 10 %, which corresponds in an equivalent methane concentration of 723 ppm. Only a slight baseline change of < 0.5 mV for 20 % relative humidity could be observed after the gas treatment. The absolute sensor signal amplitude is in the same range as without having humidity in the gas matrix (cf. Fig. 3). Apparently, the non-precious metal catalyst material is not significantly affected by ambient humidity variations.

4 Conclusion

In our work, we designed and characterised a novel wireless gas sensing system for the detection of methane and propane. A catalytic sensor, a so-called pellistor, was used as the sensor element. To ensure a low power consumption of the pellistor, it was implemented in MEMS technology. In addition, the operating temperature of the sensor could be reduced to 350 °C for methane detection, whereby a new type of non-precious metal oxide catalyst was used. By combining the low-power gas sensor with the efficient radio system, the possibility of a stand-alone warning system could be demonstrated. The variation of the sampling rate from 3 to 60 s and a reduced t_{on} time of 100 ms enables a runtime of more than 220 d with a 2400 mAh (7.68 Wh or 27.6 kJ) battery. Measurements at 2000–8000 ppm CH_4 have shown that the sensor system is able to detect concentrations at least down to < 5 % of the LEL of methane at an operation temperature of 350 °C. The measurements performed on 2000–8000 ppm propane confirm the activity of the metal oxide catalyst towards further hydrocarbons besides methane. Compared to the state-of-the-art of 435 °C (Bársony et al., 2009), the operation temperature could be lowered by 85 °C, which considerably reduces the probability that the pellistor will serve as an ignition source. By applying different moisture levels to the sensor between 20 %–60 % relative humidity at room temperature, the influence of the major cross-sensitivity factor was investigated. The results show a minor influence below 10 % relative to the sensor signal amplitude.

Data availability. The underlying measurement data may be requested from the authors if required.

Author contributions. BB wrote the original draft and did the gas measurement experiments. DG performed the battery consumption measurements and was responsible for the wireless data transmis-

sion. OY carried out the synthesis of $NiCo_2O_4$ sample, and performed the SEM investigations. LE prepared the $NiCo_2O_4$ samples and conducted the particle size examinations. The sensor readout procedure was developed by BB, DG and HFP in close discussion with MJ. JW and LR created the concept for the sensor node and were responsible for the funding acquisition. All authors contributed to the reviewing and editing of the final paper.

Competing interests. The authors declare that they have no conflict of interest.

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