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In-situ high temperature CO/HC gas sensors for optimization of the firewood combustion in low-power fireplaces

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Abstract

In this paper, the combustion and emission situations of batch wise firewood fueled fireplaces is discussed. Difference in CO-emissions of a hand operated and automatically air stream controlled firewood combustion process is illustrated. An air stream control algorithm is introduced which directs all phases of the firing process: *ignition, high temperature* and *burn out* phase. The combustion air stream control concept is based on motor driven shutters combined with air mass stream sensors and on flue gas analysis by sensors for combustion temperature, residual oxygen concentration (ROC) and residual un- or partly combusted pyrolysis gas components (CO/HC) Different commercially available high temperature CO/HC sensors along with an indigenously developed metal oxide (MOG) sensor array are evaluated in batch firing experiments with reference to the data sampled by a HT-FTIR analysis system. Finally, the signal stability of the sensors was investigated by repeated exposure to CO/air gas mixtures.

Keywords: Firewood combustion, Firing process control, Mixed potential gas sensors, Metal oxide gas sensors

1 Introduction

Residential wood combustion is of widespread concern owing to its adverse impacts on air quality and human health, especially in the many developing countries where wood is regularly used for residential cooking and heating, however, also developed countries use wood as a cheap alternative for domestic heating mainly at the countryside. Considering Germany in particular, about 14 million low-power single room fireplaces are being operated. Even after the second version of the new German emission law (1stBImSchV) for low-power firewood fueled fireplaces has come into effect in 2015, the upper emission limits for single-room fire appliances (1250mg/m^3 for CO and 40mg/m^3 for particulate matter (PM)) are still stipulated much higher than the typical emissions from heating oil burners (approximately 50 mg/m^3 CO).

Under optimal conditions, combustion of wood/biomass results solely in the emission of water vapor and carbon dioxide (CO_2). However, under incomplete combustion conditions, numerous gaseous and aerosolized compounds are emitted from biofuels in addition to CO_2 and water. These toxic gas components include CO, partially oxidized hydrocarbons (HC), polycyclic aromatic HC (PAHs) and particulate matter (PM) [1].

A substantial reduction of those emissions is possible with proper automated heating strategies. The control of the air streams based on combustion temperature and residual oxygen concentration (ROC) is meanwhile the modern state of the art. However, our firing experiments conducted at various fireplaces have shown that it is essential not only to enable sufficient oxygen but also to avoid cooling effects in the post combustion chamber by excess air throughout the firing process. To approach the quality of the firing process to the level of complete combustion, control of combustion air streams must be optimized. This is possible at situations of high combustion kinetics, i.e. at high combustion temperatures, which is merely achieved in a relatively short phase of a firing process by means of optimal feeding with combustion air. These aspects indicate the necessity for a representative knowledge of the combustion situation at every time of the firing process by monitoring of combustion temperature, ROC and the evolved gas components [2].

In this paper, the combustion and emission situations of batch-wise firewood fueled fireplaces (stoves) are discussed and a new air stream control algorithm is introduced which directs all phases of a batch-wise firing process: *ignition, high temperature* and *burn out phase* [1]. The combustion air stream

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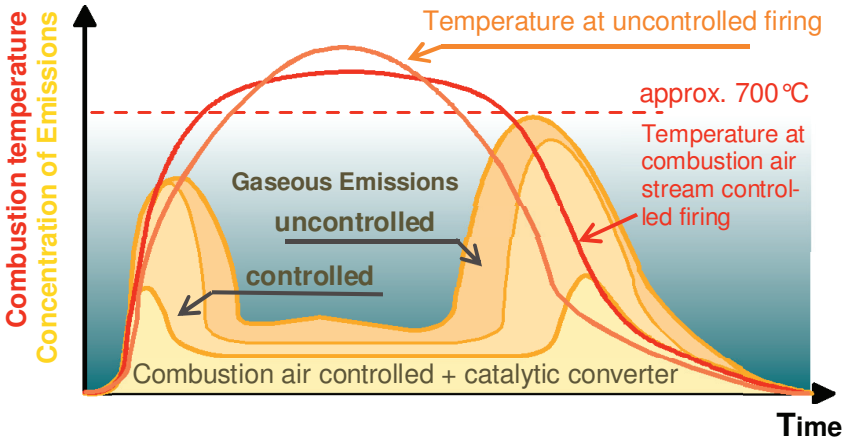

Fig. 1: Schematic representation of the characteristics of combustion temperature and gaseous emissions of a wood-log stack combustion under (i) uncontrolled, (ii) air stream controlled and (iii) air stream controlled with catalytically supported post-combustion conditions.

control concept is based on motor driven shutters combined with air mass stream sensors and on flue gas analysis by in-situ sensors for combustion temperature, ROC and residual un- or partly combusted pyrolysis gas components (CO/HC). Furthermore, different types of CO/HC-sensors are investigated to assess their suitability for in-situ application in exhaust gases of firewood combustion processes.

2 Concept of firing process optimization

Minimized emissions of un- or partly combusted gas components are the result of an optimized firing process. The log-wood combustion process is a batch process and can be classified into different phases as ignition phase (about 15-30min), high temperature combustion phase (about 30-40 min) and a burn out phase (up to about one hour). The characteristics of emissions and combustion temperature variation from a typical firing process are illustrated in Fig. 1. The emissions are at high concentration during ignition phase and burnout phase, and comparatively low in high temperature phase, at which the kinetics of reaction is fast enough for efficient oxidation of the pyrolysis gases.

Thus, one way of reducing the overall emissions of a batch process is shortening the ignition phase and prolonging the high temperature phase as much as possible. This is possible by proper stacking of the wood-logs and optimal dosing of combustion air. Improper dosing of combustion air can have two different effects. Insufficient combustion air results in (i) incomplete combustion and (ii) a decrease in combustion temperature and, consequently, in tremendous increase of emissions, whereas overdosing of combustion air cools down the combustion gases which again results in

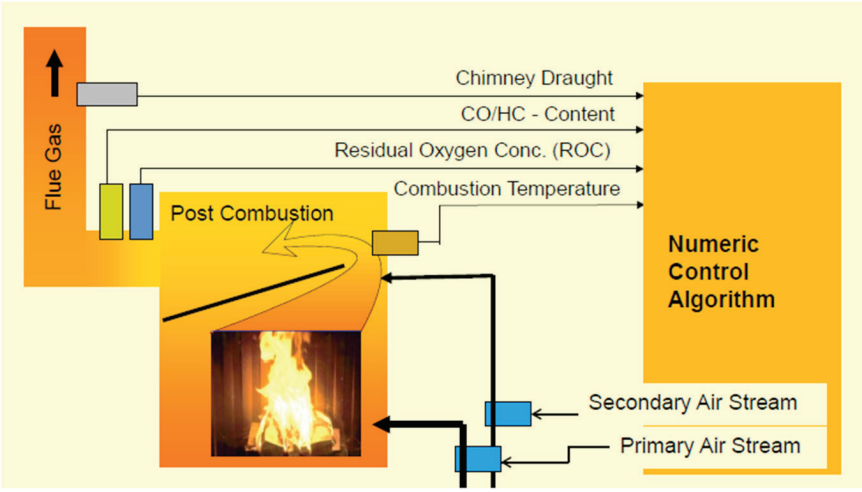

Fig. 2: Schematic concept of firewood combustion control

incomplete combustion due to slower oxidation kinetics [3].

Prerequisite of optimal dosing of the primary and secondary combustion air (Fig. 2) for process optimization is to monitor the actual firing process by in-situ sensors throughout the process [1]. Besides the above mentioned sensors for flue gas analysis, the signals of air mass stream sensors are used as input to a numeric control algorithm for setting of the combustion air streams by motor driven shutters (Fig. 2).

3 Experimental setup

Two kind of stoves were used in this study, Varia 2R (Spartherm GmbH, Melle, Germany) and SF10SK (Brunner GmbH, Eggenfelden, Germany). Both fireplaces were complemented by sensors and actuators and operated with the new air stream control algorithm (Chap. 2). The former fireplace was mainly used for validation of the control concept and the latter for in-situ evaluation of different types of CO/HC sensors.

For validation of the operating concept described above, the Varia 2R stove is operated at hand controlled and air stream regulated mode. In both cases, the combustion temperature, the residual oxygen concentration and the CO/HC-concentration are continuously measured by in-situ sensors. Combustion temperature was measured by a thermocouple positioned in the post combustion chamber (Fig. 3a) whereas the sensors for measuring of ROC (MFO10-O, Dittrich GmbH, Baden-Baden) and CO/HC-concentration are installed in the exhaust pipe. The air streams are measured by mass flow sensors and are adjusted to desired flow by control of the motor-driven shutter position by a separate algorithm. In case of hand controlled mode, only a total combustion air stream can be fed to the stove which is manually set by an adjustment lever.

For simultaneous evaluation of different types of CO/HC-sensors to check their suitability in in-situ operation, CarboSen 1K (CS1K) and CarboSen 10K (CS10K) (Lamtec Meß-und Regeltechnik für Feuerungen GmbH & Co. KG, Walldorf, Germany), an indigenously developed metal oxide (MOG) sensor array and a TGS 823 (Figaro Corp.) were operated in the flue gas tube-outlet of the SF10SK stove (Fig. 3c). Basically, CS1K and CS10K are different types of commercially available mixed potential gas sensors [4]. The MOG-sensor array consists of four different metal oxide gas sensitive layers (Fig. 4) [5].

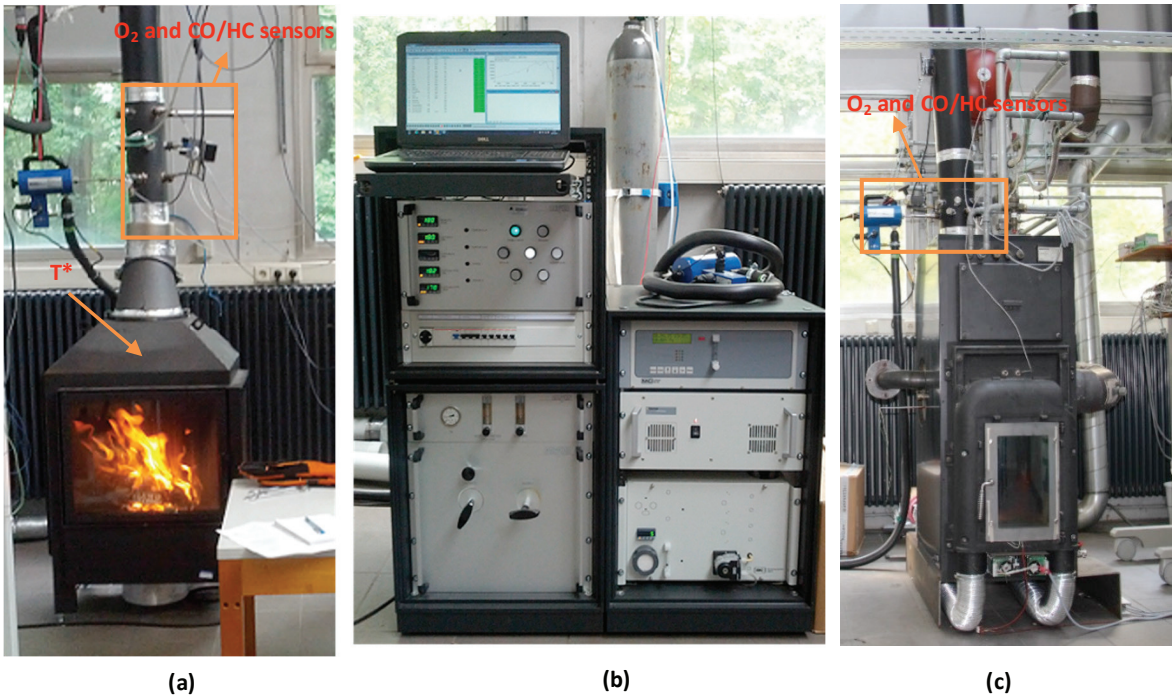

Fig. 3: (a) Varia 2R stove (Spartherm Feuerungstechnik GmbH, Melle) used for validation of the combustion process control, (b) HT-FTIR/paramagnetic oxygen exhaust gas analysis system (GASMET, Ansyco GmbH, Karlsruhe) and (c) water cooled SF10SK-fire place (Brunner GmbH, Eggenfelden) used to test various types of sensors in the exhaust gases.

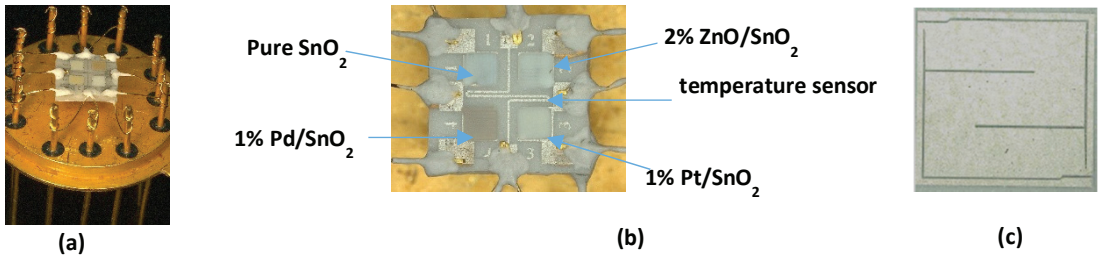

Fig. 4: Self developed metal oxide sensor array (a) comprising of an alumina chip (4x4mm²) with four different additive/SnO_2 gas sensitive layers and a resistive temperature sensor on the top side (b) and a Pt-thin-film heater at the reverse side (c).

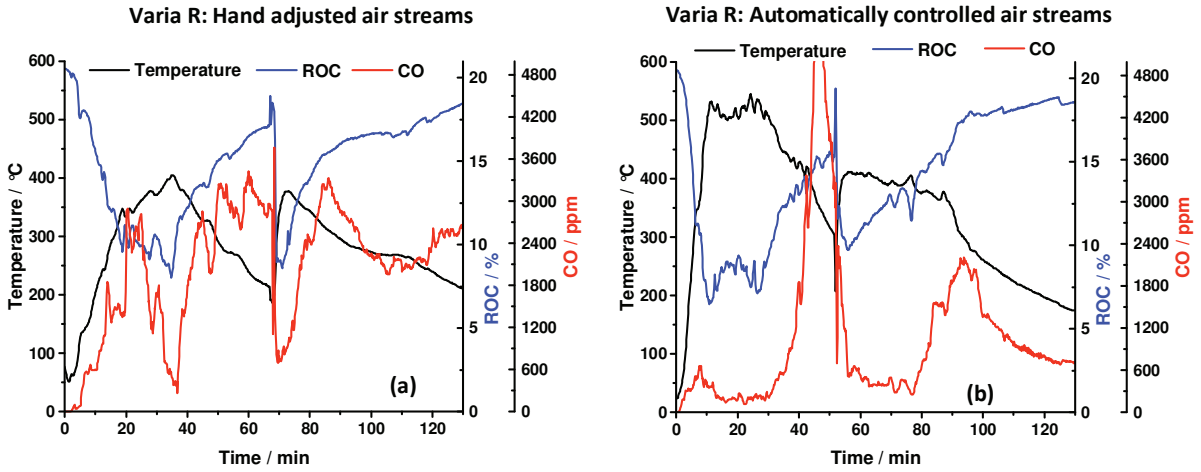

Fig. 5: Illustration of the difference in CO-emissions of a hand operated (a) and an automatically air stream controlled (b) firewood combustion process in a Varian R fireplace. Care was taken that wood-log-stacking was as similar as possible. At about minute 70 (a) and minute 50 (b), respectively, the fireplace was reloaded with two additional wood-logs on the glue-bed. Temperature sensor is located at position T (Fig. 3 a) and is assumed to be not located at the position of maximum combustion temperature. But the temperature of the controlled process (b) is recorded relatively higher compared to the hand-operated system. All CO-concentration values are original data, i.e. not related to 13%ROC.*

For the proof of the signal stability, the sensor elements were first calibrated in model gases (CO/synthetic air-mixtures), then operated in three batch firing experiments and finally calibrated again in the same model gases by using an automated sensor test-setup [6]. A high temperature FT-IR system (Fig. 3b) is used for analysis of the exhaust gas and referencing of the CO/HC sensor signals. It provides analysis of the IR-active flue gas components in time intervals of 12s.

4 Results

In this chapter, first the efficiency of the new combustion air stream control method based on the obtained sensor signals is reported by comparing the CO-emissions of Varia R stove in hand-operated mode with automated control mode. Second, different types of CO/HC-sensors are operated simultaneously to study their performance in the flue gas of a SF10SK tiled-stove fireplace dependent on flue gas composition over time of a combustion batch. The signal stability is monitored by calibration of the sensor set in CO/air – mixtures before and after operation in the exhaust gas of three firing experiments.

4.1 Validation of the control concept

To demonstrate the high efficiency of the new concept of firing process control, the most important parameters of two firing experiments,

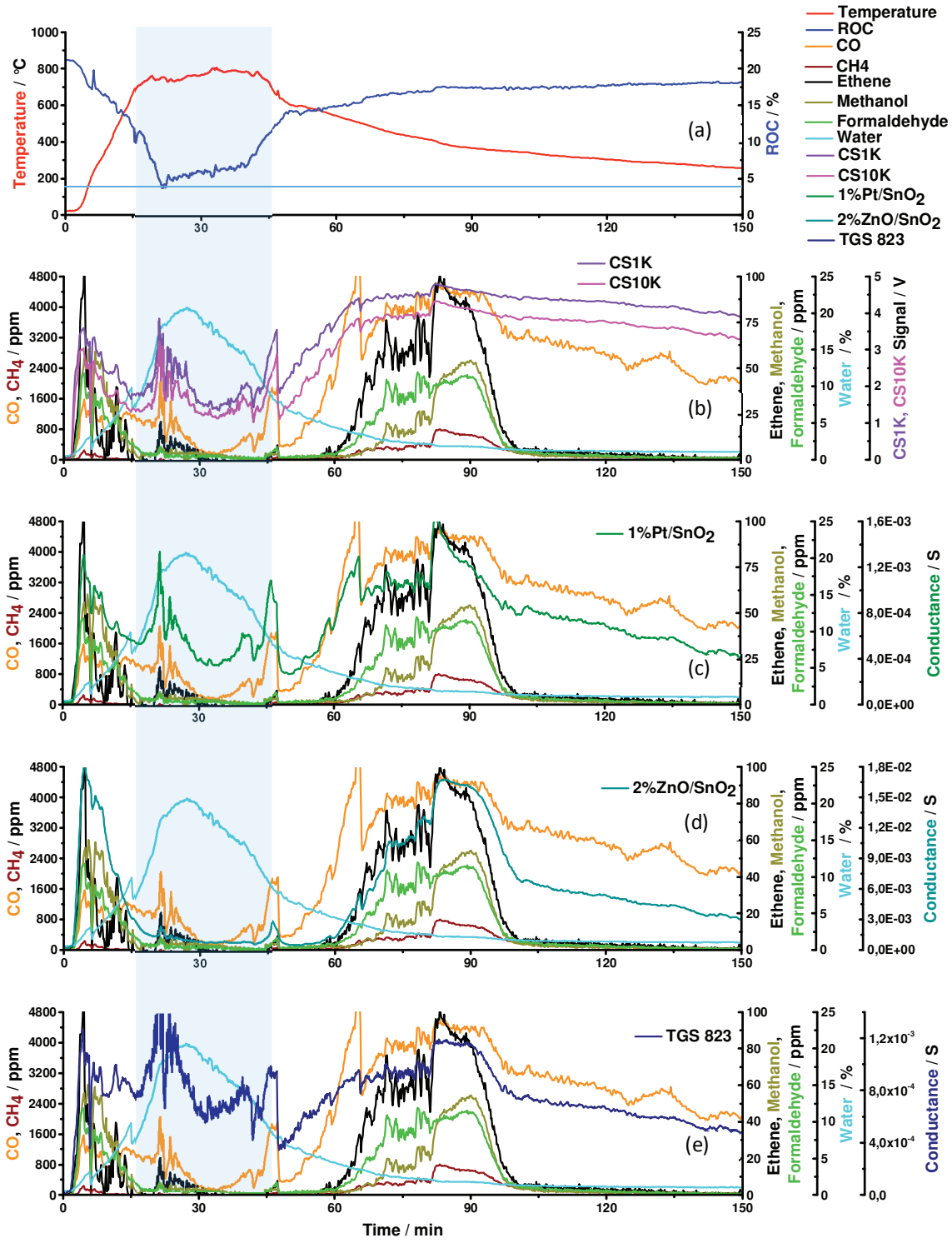

Fig 6: Batch firing process data over time recorded by operating the SF10SK tiled stove with indication of the high temperature phase in transparent blue. (a) Combustion temperature and ROC. The horizontal blue line indicates the minimum combustion temperature (about 4% ROC). (b-e) Signals of the two CarboSen-elements, the 1% Pt/SnO_2 and 2% ZnO/SnO_2 of the MOG sensor array and of the TGS823. In all diagrams the respective signals are related to the concentrations of CO, CH_4, formaldehyde, methanol, ethane and water vapour measured by the HT-FTIR analysis system.

one under hand-assisted process control and the other under automated combustion air control based on the sensor elements introduced above, are compared in Fig. 5. The diagrams show clearly that by automated air stream control the maximum temperature is reached faster after ignition and the relative maximum temperature is higher compared to the hand-assisted operation of the fireplace. CO emission is considerably low between min. 10 and min. 40 (Fig. 5b) due to higher combustion reaction kinetics up to the moment when temperature drops again below 400°C in burn-out phase. And, after re-loading of firewood on the glue bed, again the CO-concentration is reduced to values below 600ppm for more than 20min., which is not observed in the hand operated case.

4.2 Evaluation of CO/HC high temperature gas sensors

As an example, the sensor signals over time simultaneously measured during a firing experiment are given in Fig. 6 together with the as analysis data sampled by the HT-FTIR analysis system for referencing.

The following results are disclosed: Relatively low concentrations of un-combusted components can be expected at combustion temperatures above 600°C. However, this is only possible, if the ROC is not too low. There is a clear indication that even at temperatures higher than 700°C the emissions are relatively high, if the ROC is lower than about 6%. This is assumed to be due to imperfect admixing of the pyrolysis gases with the air in the post-combustion chamber. However, at ROC≥6% and a combustion temperature of about 800°C the concentrations of all of the emission gas components are recorded at values lower than 150 ppm (related to 13%ROC) over a time period of about 10min (Fig. 6).

Regarding the CO/HC-sensor signals, there are several characteristic differences in sensitivities to different gas components. The signals of CS1K and CS10K signals over time are perfectly following each other. The signals related to different gas component concentrations recorded by HT-FTIR are perfectly matching the CO-concentration over time at low CO-concentrations but are less matching the CO-concentration peaks due the general logarithmic dependency of the signal of mixed potential gas sensors on target gas concentration. The data show that the CS1K and CS10K sensor elements are more sensitive to CO than to other hydrocarbons and, as expected, the sensitivity is increased at decreased ROC.

In contrast, the signal of the 1% Pt/SnO_2 sensitive layer of the MOG sensor array (Fig. 6c) follows the CO-concentration over time and is clearly influenced by the variation of the hydrocarbon-concentrations as well. In addition, the sensitivity is considerably enhanced in the high-temperature combustion phase when ROC is reduced and water vapour concentration is enhanced. Further investigations will be necessary to analyze the contribution of both parameters, i.e. to which extend this cross sensitivity can be compensated by the ROC-value, which is sampled simultaneously. Similar behaviour is recorded by the TGS823-signal (Fig. 6e)

For comparison, the signal of the 2% ZnO/SnO_2 MOG-layer behaves clearly different to the other CO/HC-sensors discussed above. There is nearly no dependency on the CO- and water vapour concentration, however, the highest dependency on the hydrocarbon concentrations is observed. At this moment, it is still open whether there is a general high sensitivity of this sensor element on all the hydrocarbons or only to one of the components.

4.3 Sensor Signal stability

For investigation of the sensor stability and sensitivity, the sensors were calibrated at four different $p(O_2)$ according to the procedure described in Chap. 3.

The calibration data of CS1K and CS10K are shown in Fig. 7. The data sets of pre and re-calibration are denoted by Cal1and Cal2, respectively. The signals corresponding to the open-circuit-voltage (OCV) follow the well-known general equation for mixed potential gas sensors [7] and the differences of the calibration curves are very low or even not significant. This is a good evidence of the high stability of the signals.

Similarly, the absolute conductance values of the $1\%Pt/SnO_2$ and the 2% ZnO/SnO_2 were compared together with the TGS823-signals before and after operation in three firing experiments. In case of $2\%ZnO/SnO_2$ and TGS823, the absolute conductance value of the sensors dropped to rather low values. However, in case of $1\%Pt/SnO_2$, the absolute conductance value is increased. Further studies will demonstrate, whether these sensitivity values can be stabilized at further operation in flue gases and which role operation temperature plays.

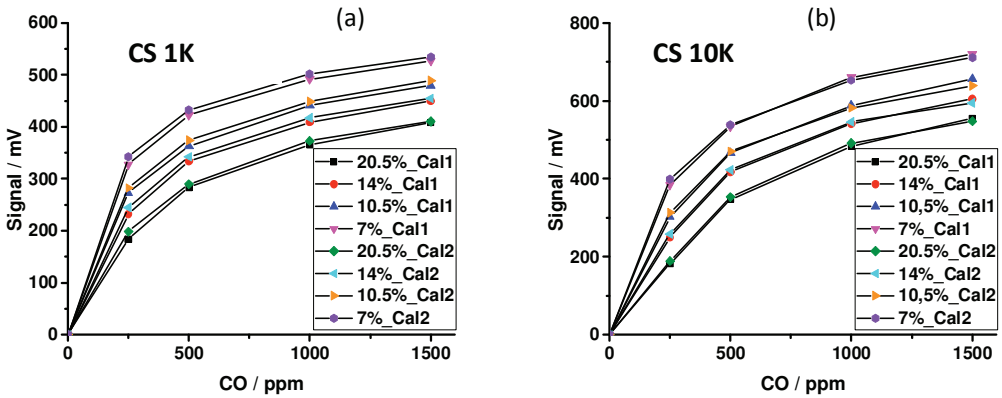

Fig. 7: OCV corresponding signals of CarboSen 1K (a) and CarboSen 10K (b) sensor element vs. CO concentration at different oxygen concentrations adjusted by mixing of fixed CO/synthetic air (5000ppm) with additional synthetic air and nitrogen. The oxygen concentration is given in %. Cal1 and Cal2 refer to calibrations before and after in-situ operation in the flue gas of three firing experiments (SF10SK).

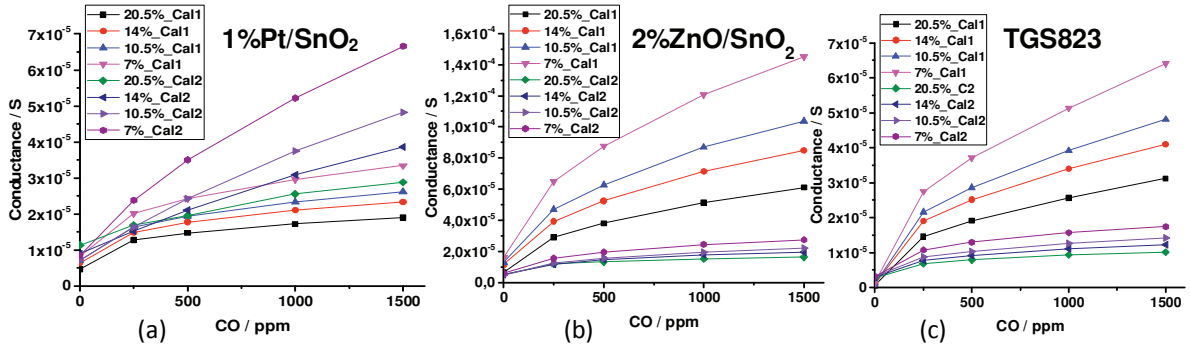

Figure 8: Conductance of two gas sensing layers of the MOG sensor array and TGS 823 vs. CO/synthetic air mixtures at different oxygen concentrations (same procedure as in Fig. 7). The calibration was done before (Cal1) and after in-situ measurements in the exhaust gas of three firing experiments in the SF10SK fireplace (Cal2).

5 Summary and Conclusions

Experiments with two different firewood stoves demonstrated that the use of in situ sensors for monitoring of the combustion temperature, the ROC and CO/HC-concentration as input to a control algorithm for primary and secondary air streams enables considerable reduction of the emission of CO and other unburned hydrocarbons in comparison to a hand-operated firing process. Obviously, automated air stream control allows more efficient combustion because it allows to optimize the dosage of the air into the post combustion chamber which enables maximizing of the combustion temperature. It was shown that by this kind of automated operation of the wood-log firing process CO-concentrations even lower than 150 ppm (or about 150mg/ m^3) in the high temperature phase of combustion are possible.

CarboSen 1K(CS1K) and CarboSen 10K (CS10K), 1%Pt/SnO_2 layer and 2%ZnO/SnO_2 layer of a MOG sensor array and TGS 823 sensors were evaluated in firing experiments by referencing the sensor signal to CO and other hydrocarbon signals from HT-FTIR analysis. CS1K and CS10K showed good correlation to the CO signal. 1%Pt/SnO_2 showed even better correlation to CO but suffers on some influence of water vapour whereas 2%ZnO/SnO_2 does not show significant cross sensitivity to water vapour. In contrast, it showed good correlation to hydrocarbons rather than to CO. Finally, TGS 823 signal correlates more to hydrocarbon signals but is also sensitive to CO. It also shows cross sensitivity to water.

Stability of the sensors was studied by comparing the sensor responses to model

gases (CO) before and after three firing experiments. CS1K and CS10K sensor signals were found to be stable. The sensitivity of 2%ZnO/SnO_2 and TGS823 to CO, however, is clearly reduced after the firing experiments. In contrast, 1%Pt/SnO_2 showed higher sensitivity.

With these studies on commercial mixed potential gas sensors of the CarboSen-type, it could be demonstrated that high stability at in-situ operation in wood-log fueled fireplaces is possible, however, the measurements on two different sensing layers of MOG sensor array and on TGS823 did not disclose uniform stability but showed very interesting sensing behaviour. Experiments will be continued to ensure these preliminary results.

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