Selective gas sensor based on high-temperature stable piezoelectric resonators exemplified by CH_4 and C_2H_4
Selektiver Gassensor auf der Basis hochtemperaturstabiler piezoelektrischer Resonatoren am Beispiel von CH_4 und C_2H_4

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Abstract
A gas sensor based on high-temperature stable piezoelectric resonators and thin metal oxide sensor films is developed. The monitoring of changes in electrical and gravimetric properties of titanium oxide and praseodymium doped ceria sensor films enables the selective detection of methane and ethane gas mixtures at elevated temperatures.

Kurzfassung
Ein Gassensor auf der Basis hochtemperaturstabiler piezoelektrischer Resonatoren und dünnen Metalloxid-Sensorschichten wird entwickelt. Die Überwachung der Änderungen von sowohl elektrischen als auch gravimetrischen Eigenschaften von Titanoxid- und Praseodym dotierten Ceroxid-Sensorschichten ermöglicht es, Gasmischungen aus Methan und Ethan bei hohen Temperaturen selektiv zu detektieren.

1 Gas sensor
The gas sensor is based on high temperature stable piezoelectric $Ca_3TaGa_3Si_2O_{14}$ *y*-cut single crystals operated at about 5 MHz. They are coated with screen printed keyhole shaped platinum electrodes, which enables their operation in thickness shear mode by an applying ac voltage. Metal oxide sensor films are deposited on top of the electrodes via pulsed laser deposition.

The conductivity of metal oxides is impacted by the presence of reducing gas species and the adsorption/desorption of gas molecules or stoichiometry changes of lattice oxygen. Conductivity and gravimetric changes occur mostly simultaneously. However, the electrode and metal oxide sensor film layouts chosen here enable to monitor electrical and gravimetric effects separately. Therefore, the selectivity will increase compared to conventional metal oxidebased gas sensors.

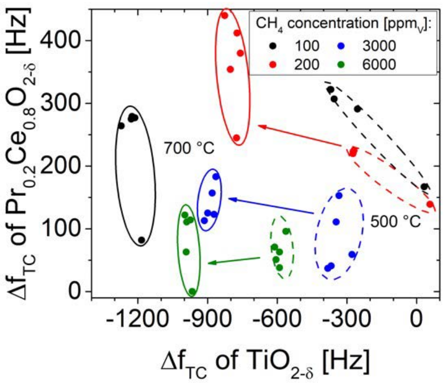

Figure 1 Resonance frequency shifts to evaluate the gas concentration in CH_4 and C_2H_4 gas mixtures.

In the conductivity mode, an increase in the sensor film conductivity results in an increased electrode diameter and resonance frequency shifts [1]. Resonators operated in the microbalance mode show shifts of their resonance frequency as a result of adsorption/desorption processes or release/incorporation of oxygen in the metal oxide [2]. The selectivity is determined by the chosen metal oxide concerning specific conductivity, conductivity mechanism (bulk, surface or grain boundary), non-stoichiometry and chemical selectivity. In addition, density, shear modulus and electromechanical losses of the piezoelectric material must be considered for data evaluation.

2 Selective detection of CH_4 and C_2H_4 gas mixtures
For the detection of CH_4 and C_2H_4, best selectivity is observed for $TiO_{2-\delta}$ and $Pr_{0.2}Ce_{0.8}O_{2-\delta}$ sensor films. Figure 1 shows the resonance frequency shifts Δf_{TC} of both resonators, plotted in dependence of each other and resulting in metrologically resolvable data clusters for different gas mixtures and temperatures. Such a plot can be used to determine CH_4/C_2H_4 concentrations.

3 Literature
[1] Fritze, H.; Richter, D.; Tuller H. L.: Simultaneous detection of atmosphere induced mass and conductivity variations using high temperature resonant sensors, Sensors and Actuators B 111-112 (2005), pp. 200-206

[2] Sauerbrey, G.: Verwendung von Schwingquarzen zur Wägung dünner Schichten und zur Mikrowägung, Zeitschrift für Physik 155 (1959), pp. 206-222.

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