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Development of a Humidity Sensor Element based on sputter-deposited thin ZnO-Layers

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Abstract

A sensor concept for humidity measurement has been invented at the Institute of Micro Production Technology. The sensor element is manufactured on silicon oxide wafers with respect to the requirements of a new direct deposition process. This new process allows for thinner sensors with higher measuring accuracy. High purity 4N zinc oxide is used for the humidity sensing layer. Sensor properties and characteristics have been evaluated using X-ray diffraction and scanning electron microscopy. Finally, the evaluation electronic concept based on an embedded system with a field programmable gate array (FPGA) is presented.

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Keywords: humidity sensor; direct deposition; zinc oxide; ZnO; Collaborative Research Centre 653

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1. Introduction

Nowadays, technical systems are equipped with a variety of sensors. They are measuring the air flow velocity on wings of airplanes, the temperature within machines or the strain caused by external stress to mechanical components. Due to continuous optimisation and performance improvements, the number of sensors per technical system is increasing. Especially in the context of Industry 4.0, it is important to generate, collect and analyse data from various processes. New measurement tasks and measuring positions are making great demands on sensors. It is

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no longer sufficient to adapt existing sensor systems, hence it is important to develop new sensor manufacturing technologies.

Usually, sensors are fabricated on a carrier substrate, separated and glued to a measurement position. Within the subproject S1 “Modular, Multifunctional Micro Sensors” of the Collaborative Research Center 653, a new technology is investigated. This technology allows a direct deposition of sensors on components [1]. Thus intermediate layers, such as carrier substrates or glue, are no longer required. As a result, the measurement errors caused by these layers are eliminated and the sensors fabricated in that way get thinner and closer to the surface of the measurement object. Hence, the sensors can be used at new positions within machines for that for instance existing commercial sensors are too thick.

During the actual research periode, a humidity sensor has to be developed that can be directly deposited on a technical surface. Currently, a variety of humidity sensors are on the market working on various measurement methods (compare [2]). The commercially available sensors differ for instance in size, measuring accuracy, measuring range, design and the used hygroscopic material (compare section 3). Miniaturised sensors are mainly manufactured with micro-technological techniques. They are working on resistive or, more frequently used, capacitive measurement methods. Some sensors are simply designed without integrated electronics whereas more sophisticated sensors have different features, such as the ability of analog to digital signal processing. Some of them have for instance a data memory, an integrated temperature sensor or a bus interface [3, 4].

To allow the deposition of thin layers on components of arbitrary size a new patented sputter coating system has been developed [5, 6]. This system shall be used for depositing the humidity sensor and other sensors of the sensor family directly on the surface of components. Using sputtering for direct deposition has the advantages of a low process temperature and a high flexibility concerning layer materials, such as metals, ceramics and alloys. Moreover, it allows a high homogeneity and purity of the deposited material as well as a controlled and reproducible layer deposition. Further advantages are a high layer adhesion and the possibility to adjust the layer properties precisely. Moreover, the technology is suitable for industrial in-line manufacturing, as it is for instance insensitive to external influences like vibrations. In a first step, described in this paper, the sensor is manufactured on wafer-level in a commercial sputter device with standard thin-film processes. This enables in a second step a direct deposition with the new coating system.

2. Requirements

Compared to the manufacturing of commercial, wafer-based sensors, directly deposition-able sensors have to fulfill special requirements. Challenges are for instance the high surface roughness of technical components, the development of a sufficient electrical insulation layer [7] and an adequate layer adhesion on the components surface.

As the mentioned new coating system for direct deposition works on the principle of a sputter deposition process, the humidity sensor has to consist of a material that can be sputter deposited. Moreover, the sensor should consist of as less layers as possible. This has the advantage to minimise the number of required target changes and to reduce the process steps. Further general requirements for humidity sensors are listed in table 1.

Table 1. Overview of some important requirements the humidity sensor has to fulfill. The abbreviation RH stands for relative humidity and is described in section 3.1.

	Requirement	Value
Sensor specification	Detectable humidity	30 %RH to 80 %RH
	Sensitivity	High
	Discrimination threshold	Low
Measurement signal	Characteristic output curve	Preferably linear
	Long-term stability	Has to be guaranteed
Evaluation electronic	Output signal	Easy processable
	Data logging	Continuous
	Compatibility to standard IC-technology	Has to be guaranteed

3. Setup of the Sensor

3.1. Measurement Methods

Measuring humidity is more complex compared to the measurement of other physical measurands as it depends on further variables like actual pressure and temperature. Humidity can be specified by three different measurements: The specific humidity SH, the absolute humidity AH and the relative humidity RH. The specific humidity SH describes the ratio of the vapour mass to the mass of one kilogram of wet air normally expressed in g/ kg. The absolute humidity AH measures the mass of water in the air and is expressed in g/ m³. The relative humidity RH equals the absolute humidity AH relative to the saturation humidity (maximum humidity) SA (formula 1) [8, 9]. As the saturation humidity is depending on the temperature, the relative humidity is changing by definition with temperature.

$$RH = \frac{AH}{SA} \cdot 100\% \tag{1}$$

Weighing up the advantages and disadvantages of different humidity measurement methods [2], the resistive method is very promising to be used for directly deposition-able humidity sensors. The sensors working on that method have the advantages of cheap manufacturing in high quantities and great miniaturisation possibilities. Moreover, the manufacturing of such sensors can be realised by direct deposition. Furthermore, the method enables a high sensitivity and good long-term stability [10].

3.2. Setup of Resistive Sensors

Resistive sensors use the effect of humidity depending electrical properties of a hygroscopic material to determine the actual humidity. The main elements are the electrode structure and the hygroscopic material. Investigations are showing the high influence of the electrode structure on the functionality of the sensor element. For the sensor element presented in this paper, the following electrode structure is used (figure 1).

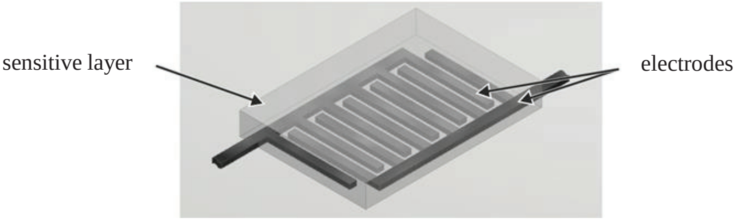


Fig. 1. Schematic view of an interdigital sensor structure.

In this so called interdigital structure the parallel fingers of both electrodes are arranged in a comb-like structure. The functional layer consisting of a hygroscopic material is sputter deposited onto the electrodes. The electrode structure is suitable for impedance measurements. The comb-like arrangement equals a parallel connection of both electrodes [11]. Moreover, it enables rapid response behaviour as the hygroscopic material is in direct contact with the ambient air. Furthermore, it is advantageous that no further process steps are required after depositing the functional layer. Hence, a contamination or negative impairment of the functional layer is avoided.

3.3. Suitable Functional Layer Materials for Humidity Measurement

Different materials can be used as a humidity sensitive layer. Polymers, as well as metal oxides, are commonly used as hygroscopic materials within resistive sensor systems [4]. However, both materials show a great dependence of their electrical properties on the air humidity. Besides polymers, a variety of other materials such as porous silicon [12], titanium oxide or complex alloys for instance MgCr2O4-TiO2 [13] have been investigated.

Furthermore, the mechanical strength, thermal capability, physical stability as well as the resistance of ceramic layers are promising [4].

Another suitable material is the II-VI semiconductor compound zinc oxide. Within recent publications the use of zinc oxide as hygroscopic layer has been described. The self-organisation of the zinc oxide is used to create large, defined surface structures that enable an adsorption of gas molecules. Recent research shows, that outstanding humidity sensing properties can be realised with zinc oxide layers in form of nanorods as well as nanowires [14] and further nanostructures [15]. For the manufacturing of such nanostructures various methods like molecular beam epitaxy (MBE), pulsed laser deposition (PLD) or chemical methods like hydrothermal synthesis can be used [16].

Besides the mentioned manufacturing methods, zinc oxide is also sputter-able and therefore useable for direct deposition. Recent publications are showing promising properties of sputter deposited zinc oxide for use as a humidity sensing layer [17, 18].

4. Manufacturing of the Sensor

The developed sensor described in this paper is measuring the resistance of a hygroscopic layer. For the sensor electrodes an interdigital structure is used and a thin layer of zinc oxide works as a hygroscopic functional layer. The deposition of the electrodes and the functional layer of the sensors are taking place in a commercial sputter system Senvac Z550 on a 525 µm thick silicon oxide wafer.

The targets have a 6.5” (165.1 mm) diameter. The distance between the target and substrate equals 75 mm. In a first step, the electrodes are sputter deposited by using a spin-coated and lithographically structured photoresist mask. The electrodes are made of a sputter deposited 250 nm thick nickel chromium layer.

Afterwards, the functional layer is sputter deposited above the electrodes. The zinc oxide target has a purity of 99.99 % (4N) and is used in combination with a magnetron. The base pressure before sputtering has been at 1.9 x 10^{-6} mbar, the pressure during sputtering at 4.9 x 10^{-3} mbar. The RF-power has been at 100 W. The substrate during sputtering was not heated. The flow-rate of argon was 10 sccm. To assure the stoichiometry oxygen at a ratio of 1:1 was added to the argon process gas. The manufactured humidity sensor element is shown in figure 2.

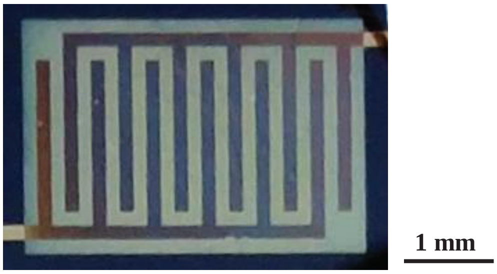


Fig. 2. Image of interdigital sensor structure. Size of the sensitive rectangular zinc oxide area 3.75 mm x 2.55 mm, width of electrodes 0.15 mm.

5. Investigations of the Deposited Layers

5.1. Measurement Setup

To characterise the humidity behaviour of the manufactured sensors predefined air humidity is generated in two different ways. One way is the use of saline solutions to create constant air humidity for static measurements within sealed containers. Depending on the salt solution, the relative humidity changes within a range from 11 %RF to approximately 86 %RF (table 2). The other way is to use a commercial climate chamber type SH-241 from ESPEC allowing to control temperature and air humidity.

The measurement of the actual humidity and temperature is realised with a commercial humidity sensor. The impedance measurement is carried out with a HAMEG HM8118 LCR-Bridge. LabVIEW is used for data processing.

Table 2. Different salt solutions and the corresponding equilibrium relative humidity [19].

Salt	Formula	Relative humidity in %RH	
		20 °C	25 °C
Lithium Chloride	LiCl	12	11
Magnesium Chloride	$MgCl_2$	33	33
Magnesium Nitrate	$Mg(NO_3)_2$	52	52
Copper(II) Chloride	$CuCl_2$	68	67
Potassium Chloride	KCl	86	86

5.2. Characterisation of Zinc Oxide Layer

Characterisation of the zinc oxide layers is carried out with a Jeol JSM-6400F scanning electron microscope (SEM) and a Bruker D8 Discover X-ray diffraction system (XRD) using a cobalt anode. To allow XRD-measurements, the layer thickness is enlarged. The results of the structural and morphological analysis obtained by SEM studies of the zinc oxide film are shown in figure 3.

The fracture plane presented in figure 3b indicates columnar crystallites resulting from the preferred growth direction of zinc oxide, which has hexagonal wurtzite structure. The growth velocity along the c-axis is larger than in other directions as the surface energies of the crystal planes are different [20]. This preferred orientation effects a strong basal fibre texture of the films confirmed by XRD measurements (figure 4). Like all of the XRD patterns obtained during research, the XRD pattern (compare figure 4) has one sharp peak corresponding to the (0002) plane of the wurtzite structure. No other diffraction peaks of any impurities are visible so the received film is estimated to appear in high purity grade.

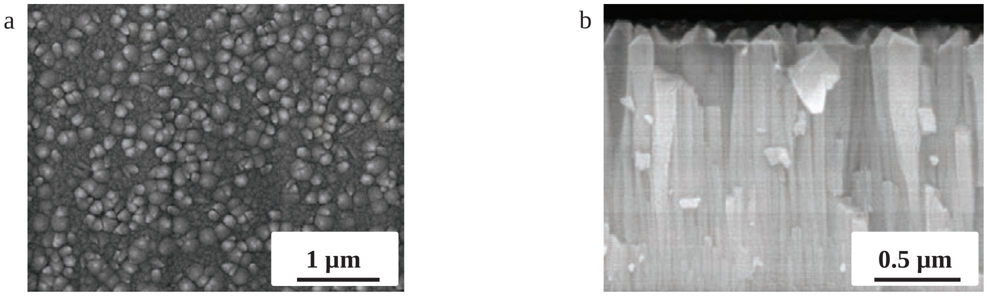
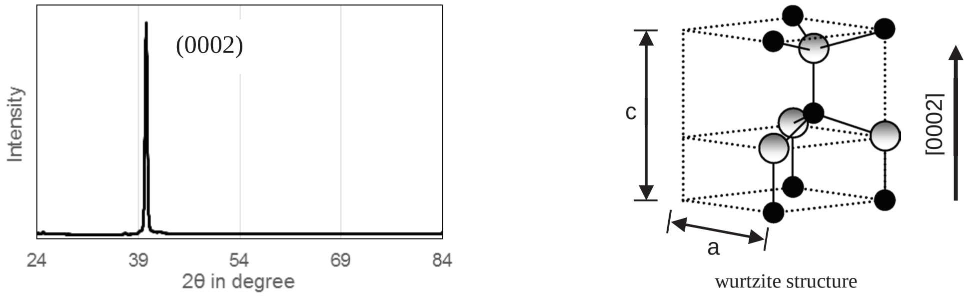


Fig. 3. SEM images of sputtered zinc oxide with (a) view on top and (b) view on fracture plane. The bottom of image b equals the silicon oxide surface.



wurtzite structure

Fig. 4. XRD pattern and crystallographic structure [16] of sputtered zinc oxide.

5.3. Humidity Sensing Behaviour

Impedance measurement technique is used to investigate the electrical properties of the sensing layer at various humidity levels as measuring under DC voltage could cause unwanted polarisation influences. Measurement frequency was set to 1 kHz for all investigations. Measurements were carried out with the LCR-Bridge at different humidity levels between 11 %RH and 85 %RH to obtain the characteristic curve of the produced zinc oxide sensing layer (compare figure 5a). To ensure a stable value the sensor was kept in an appropriate ambience for twelve hours and shielded against external influences, such as light, before measuring. The resistance decreases at higher humidity levels, so a clear humidity dependence of the material is observed. As the behaviour is non-linear, the resistance change at low humidity is smaller than in ranges above 50 %RH.

Dynamic response of the sensor is presented in figure 5b. Humidity is varied between 25 %RH and 85 %RH for several times. The sensor shows a reliable and reproducible response and recovery behaviour which corresponds to the ambient humidity change.

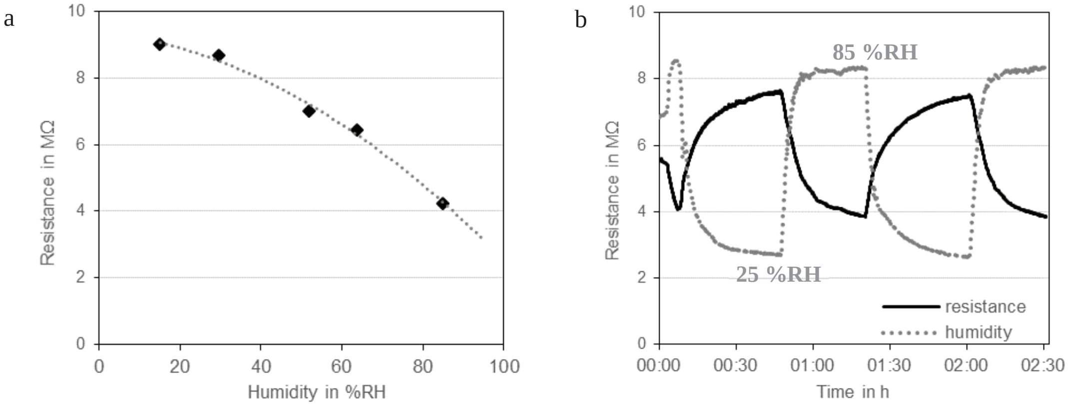


Fig. 5. (a) Resistance values at specific humidity levels and (b) dynamic response of the sensor.

The impedance of the layer changes with the humidity level as adsorption and desorption of water molecules on the filmsurface provoke different physical conduction mechanisms. At low relative humidity water is chemically adsorbed onto the surface as aqueous layer, where hydrogen (H^+) ions are the dominant carrier based on proton hopping transport. With increasing humidity subsequent water layers are adsorbed by physisorption and mainly dissociated, so conductivity gets influenced by hydronium (H_3O^+) ions, which pass a proton´s charge on to a neighbouring water molecule as described by the Grotthus mechanism. [4, 18]

6. The Evaluation Electronic Concept

For the completion of the presented sensor a suitable evaluation electronic has to be developed. The humidity dependent impedance of the sensor element has been measured at a constant temperature over the maximum humidity range from 11 %RH to 85 %RH. The characteristic output curve and sensor behaviour have been evaluated by using the LCR-Bridge (compare section 5.1).

In order to create an integrated sensor system a miniaturisation has been achieved by designing an evaluation electronic concept consisting of a circuit for the humidity sensor element as well as a field programmable gate array (FPGA). Up to now, not all features of the concept are realised. The principle setup of the concept is illustrated in figure 6. The FPGA is part of an external commercially available embedded system. As the sensor signal is disturbed by temperature, a sufficient compensation can be achieved by measuring the temperature. Therefore, a separate temperature sensor element has been added to the system that is evaluated by a developed circuit. The embedded system, the developed circuits and the sensor elements are connected by wire strands.

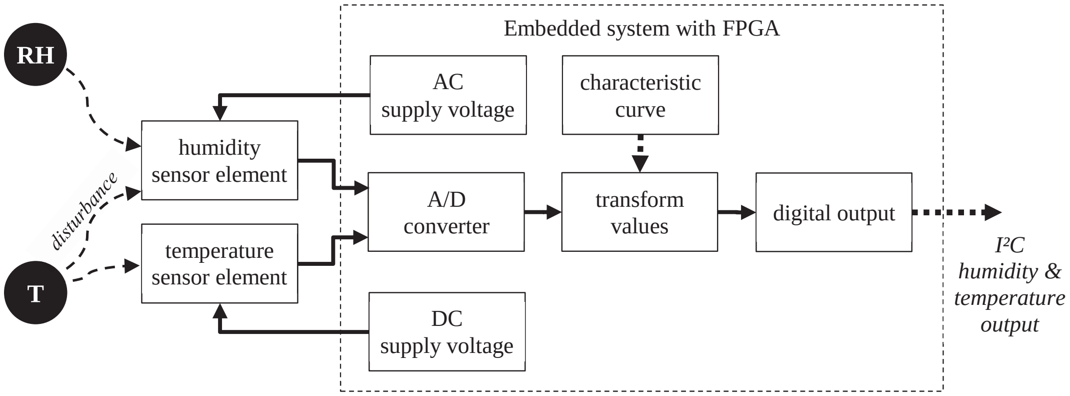


Fig. 6. Principle setup of the evaluation electronic concept.

The AC sine wave supply voltage for one of the circuits is generated with the FPGA device. The sine wave frequency can be set within a range from 100 Hz to 1,000 Hz. This allows adjusting the frequency with respect to the best measurement performance of the humidity sensor element. By measuring the response signal a characteristic output curve within the required humidity range can be generated by using the salt solutions or the humidity chamber. The measured values can be stored within the embedded system described above. During the measurements the acquired signal is transformed into the equivalent relative humidity according to the characteristic output curve of the sensor. Finally, the humidity as well as the temperature signals are converted into a digital signal to provide good sensor connectivity to superior systems. As a communication protocol the I²C (inter-integrated circuit) serial bus can be used. Digital communication allows a more reliable and influence resistant data exchange as well as good interoperability. Furthermore, it will be possible to send data and commands to the sensor system. This means for instance to request a single measurement value or to set specific sensing parameters.

7. Summary and Perspective

The development of a humidity sensor by using ZnO as the humidity sensing layer has been realised. The sensor is sputter deposited on a silicon oxide wafer taking into account requirements for the new direct deposition process in order to allow an easier adaption and direct deposition on technical surfaces with a new type of a sputter coating system. Investigations showed promising results with sensors based on a resistive measurement method. Another high influence on the sensor properties has the electrode structure. Best results have been achieved by using a comb-like structure. The sensor is usable within a range from at least 11 %RH to 85 %RH, exceeding the former range requirements. A temperature sensor element has been used to enable a compensation of the thermal influence on the relative humidity and to enable an investigation of the temperature dependence. An expandable evaluation electronic has been designed.

In a next step, the sensor has to be adapted for manufacturing on flexible polyimide foils and for the direct deposition with the invented and patented deposition system. A further research on the long-term stability is recommended as well as an optimisation of the sensitivity of the zinc oxide layer and the response behaviour. The evaluation electronic can be optimised and further miniaturised. Moreover, various electrode materials and the temperature behaviour of the sensor have to be investigated.

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