Ceramic sensor elements for geothermal applications

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Abstract

The harsh environmental conditions of geothermal applications restrict the applicable sensor materials to a small number. This is an application area for ceramic multilayer technology, which, thanks to the possibility of 3D structuring of ceramic materials, offers the possibility of producing cost-effective harsh environment sensors suitable for mass production. This requires the stability of all materials used for the sensor, such as the ceramic and functional layers. For this purpose, we tested ceramic materials, metallization's, and inorganic cover layers regarding their suitability for use in geothermal environments and will present the stability of these materials under corrosive attack. Based on the material selection a ceramic piezoresistive high pressure sensory element was designed, manufactured, and characterized in laboratory environment and in application-specific test stands.

Key words

Geothermy, sensors, ceramic, Multilayer ceramic technology, LTCC, HTCC

I. Introduction

Geothermy is a renewable and sustainable source of energy and can provide weather independent heating and cooling. Geothermy requires drilling into the Earth's crust encountering harsh environmental conditions [1]. Especially in increased depth high temperature and high pressure-conditions exist. Furthermore, drilling involves vibration and contact with circulating fluids like drilling mud or salty water for cooling and removal of debris. The use of harsh environmental stable sensors can support the drilling process and allow for an effective geothermal extraction.

The harsh environmental conditions restrict the applicable sensor materials to a small number. Especially ceramic materials are of high interest since they are characterized by inherent material stability under high temperatures and aggressive media, while stability under high ambient pressures and vibration must be implemented by design.

This is an application area for ceramic multilayer technology, which, thanks to the possibility of 3D structuring of ceramic materials [2], offers the possibility of producing cost-effective harsh environment sensors suitable for mass production. This requires the stability of all materials used for the sensors, such as the ceramic and the functional layers. For this purpose, it was necessary to test suitable ceramic materials, metallization's, and cover layer materials with regard to their suitability for use in harsh environments.

II. Design of test specimens and manufacturing

Ceramic Multilayer Technology is an established method for the manufacturing of ceramic microcircuits [3]. The process (figure 1) uses ceramic green tapes, manufactured by tape casting, and functional pastes as semi-finished products. During a blanking step the ceramic tapes are cut into defined sizes (up to 8x8 sq in) and geometrically structured by punching (creation of via holes) or lasing. The via holes will afterwards be filled by stencil printing of a metallization paste, followed by screen printing of a predefined conductor layout. The individually structured and functionalized tapes will be stacked, laminated, and sintered. During the sintering process the laminated stack transforms into a highly robust monolithic ceramic body, while the functional structures can be buried into the ceramic. The process allows not only for the integration of conducting structures, but also for other passive functionalities like resistors, capacitors, or inductors. A novel technological approach is to combine mechanical and electrical functionalities within the ceramic offering the development of highly robust ceramic Micro-electro-mechanical-systems (MEMS) solutions to be used in demanding environments.

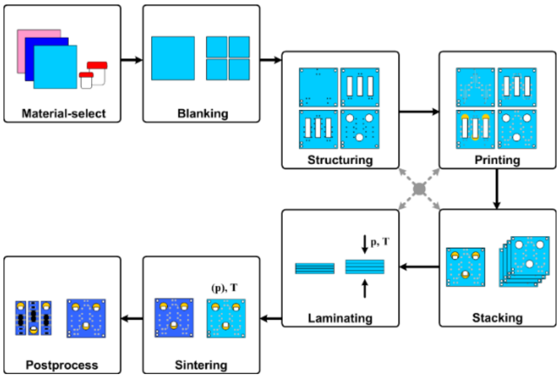


Figure 1 Multilayer ceramic manufacturing process

For the corrosion tests, commercial Low Temperature Cofired Ceramics (LTCC) and High Temperature Cofired Ceramics (HTCC) tapes were selected as base materials. The samples were manufactured using the ceramic multilayer technique. Depending on the expected z-shrinkage, several layers of 4" x 4" tapes were stacked on top of each other for a fired target thickness of approx. 1 mm and isostatically laminated at 70°C/200 bar. In the subsequent sintering step, the ceramic components were first debindered and then fired at 850°C (LTCC) or 1500/ 1550°C (HTCC). After firing, the samples were separated by wafer sawing to the sample size required for material characterization (5 x 50 mm²).

Uncoated ceramic substrates (Al_2O_3 96% and 99.6%, 3 YSZ, 2 types of LTCC) as well as substrates bearing functional layers were manufactured. As functional layers, contact metallization's, insulating cover layers and platinum structures were of particular interest. The layouts of the manufactured specimen with functional layers are shown in figure 2.

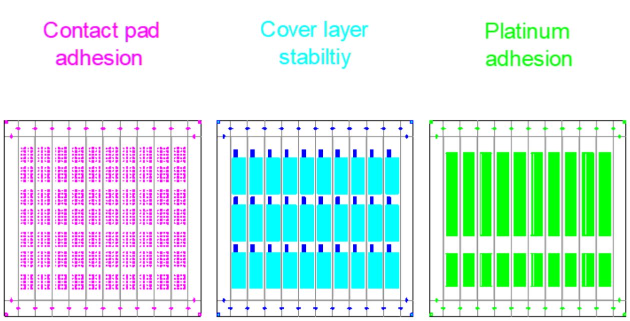
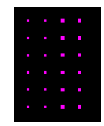
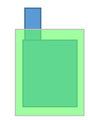


Figure 2 Layout of sample specimens (multiple panel layout)

Before singularizing the multiple ceramic panels, the layers listed in the table were deposited using screen printing. Depending on the material, the layers were sintered at 620°C, 850°C or 1500°C. An overview of material type, characterization strategy and layout give table 1.

Table 1 Overview of functional layers for corrosion tests

Material combination	Metallization type (thick film)	Characterization	Layout
LTCC substrate + Metallization	AgPt	Determination of peeled of metallization area (optical, microscopy) after storage test	 Contact pad adhesion layout - squares (250,500 µm edge length)
96% Al_2O_3 substrate + Metallization	Au, AgPt		
LTCC + Metallization + Insulating cover Layer	2 Variants of Glass, LTCC (double layer deposited)	Electrical measurement of cover layer porosity by deposition of metallization on insulating cover layer after storage test	 Cover layer stability layout - Metallized surface with cover that leaves the connection lug exposed
96% Al_2O_3 + Metallization + Insulating cover layer	Glass, Al_2O_3 (double layer deposited)		
LTCC + Platinum metallization	Pt	Measurement of conductor line resistance after storage test	 Pt adhesion test layout - Conductor lines (Width 100, 150, 200 µm) in 2 Lengths with pads
96% Al_2O_3 + Platinum metallization	Pt		

III. Conduction of corrosion tests

For the ageing of the manufactured samples, they were sent to the company Dustec Hochdrucktechnik GmbH (Wismar/Germany) for pressure testing in accordance with Directive 2014/68/EU (Pressure Equipment Directive). There, the samples were stored in a tube autoclave at 2000 bar for 4 hours. The test medium was water. The container was heated to 200 °C for some of the samples and to 300 °C for the others. At the end of the ageing period, the pressure was released, the autoclave cooled to room temperature and the samples removed.

A. Corrosion tests on uncoated ceramic substrates

For the corrosion tests on the ceramic substrates, 5 samples per material and ageing temperature were analyzed. To evaluate the hydrothermal ageing, the mass loss of the substrates was determined. For this purpose, the weight of the samples was determined before and after ageing and the mass loss was calculated from these values. The samples exposed to a pressure of 2000 bar at 200°C showed no measurable change in mass. The two LTCC materials showed a mass loss of up to 2% in the samples exposed to an ageing temperature of 300°C. For the HTCC ceramics, hardly any change in mass could be detected even at 300 °C.

Only for the Al_2O_3 96% a mass loss of ~ 0.5% was determined.

To gain a better understanding of the microstructural properties after degradation, additional high-resolution analyses were carried out on cross-sections of the various ceramics. The HTCC material 3YSZ showed a clear change in microstructure. A brittle structure was found on the outer edges directly exposed to the pressure effect, which was caused by decomposition of the material from the outside. In contrast, the Al_2O_3 99,6% ceramic showed no significant changes in the microstructure.

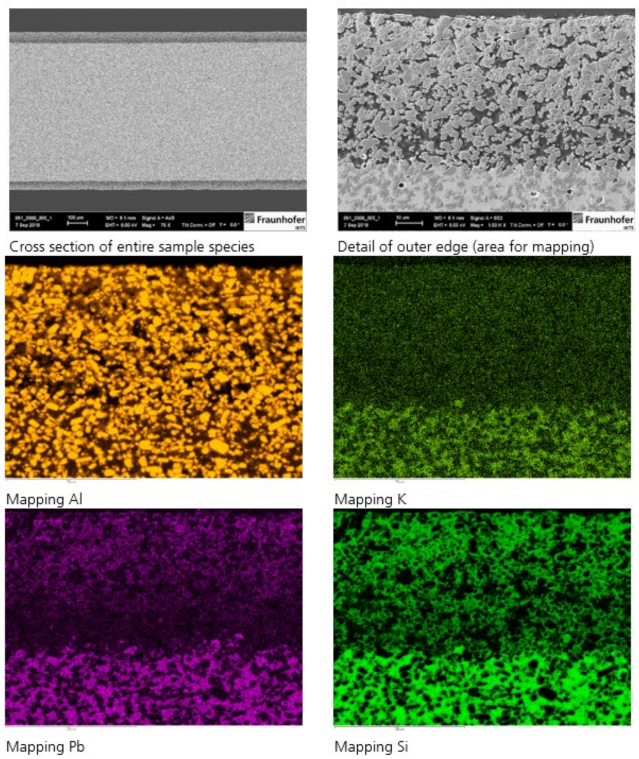


Figure 3 FESEM investigations on cross sections of LTCC type 1 (2000 bar 300 °C 4h)

The two LTCC ceramics also exhibited a significantly altered porous structure at the outer edges, which resulted from the dissolution of the glass matrix. This is shown for the LTCC type 1 in fig 3. The supplementary EDX mapping at the transition area between porous and unchanged microstructure clearly shows the dissolution of the glass matrix. In the upper, porous area, the glass components Si, K and Pb (selected as examples) can be found in significantly lower concentrations than in the unmodified microstructure. The Al_2O_3 particles are present in the same concentration in both areas.

B. Corrosion tests on functionalized ceramic substrates

To assess the hydrothermal ageing of the functional layers after the pressure tests, the samples were examined optically using a microscope. The contact pad materials (AgPt or Au metallization) showed no defects or detachment on either type of ceramic after 2000 bar, 200 °C, 4h. After the pressure tests at 300 °C, the surfaces of the pads were eroded and partially detached. The glass cover layers showed clear cracks at both temperatures. The figure below shows an example of the defects in the glass layers after the pressure tests at 300°C. In contrast, the cover variant with screen-printed LTCC material was defect-free and no short circuits to the metallization under the cover layer could be measured after the deposition of a metallization on the cover layer.

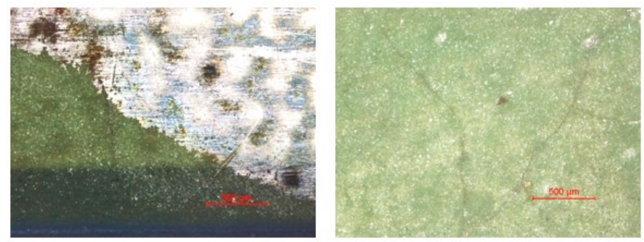


Figure 4 Glass layers over AgPt metallization with defects after 2000 bar, 300 °C, 4h storage time

III. Sensory elements and sensor integration

Based on the corrosion tests carried out, an optimum material pairing was selected for the manufacturing of ceramic sensor elements. That included sensory elements for temperature, impedance and piezoresistive high pressure measurements. The temperature sensor is shown in fig. 5 and includes a platinum-based resistor with a resistance of 30 Ω and a TCR of 3100 ppm/K. The resistor is protected by a ceramic cover layer and rewired using through-ceramic-vias to the metallization pads on the rear side of the sensor.

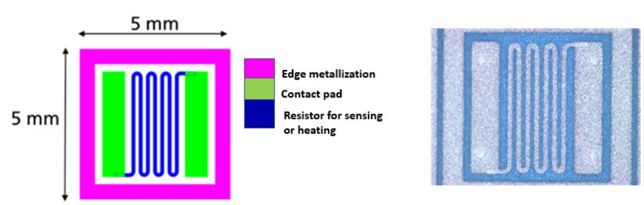


Figure 5 Layout (left) and manufactured ceramic temperature sensor (right)

The piezoresistive pressure sensing element is shown in fig. 6.

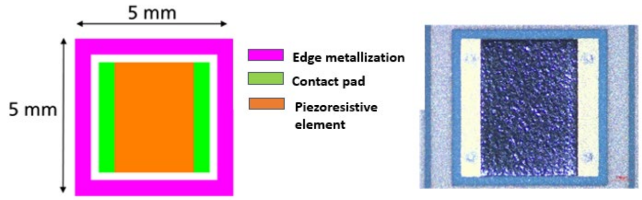


Figure 6 Layout (left) and manufactured piezoresistive pressure sensor (right)

Due to the high nominal pressures the sensing element was designed without a diaphragm. The inorganic piezoresistive sensing layer is protected by a ceramic cover layer and has a nominal resistance of 6 kΩ and a sensitivity of 2,9 Ω/bar. The rewiring is carried out in the same way as for the temperature sensor.

The ceramic sensory elements were finally combined in a signal module [4, 5]. Therefore, they were embedded in a PCB using prepreg materials with high glass transition temperatures. The ceramic sensors were placed in cavities located at the PCB-surface and electrically connected using silver sintering technology. The hermetic encapsulation was done using sputtering in combination with a layer build-up of galvanic copper and an ENIG-protection layer (Fig. 7).

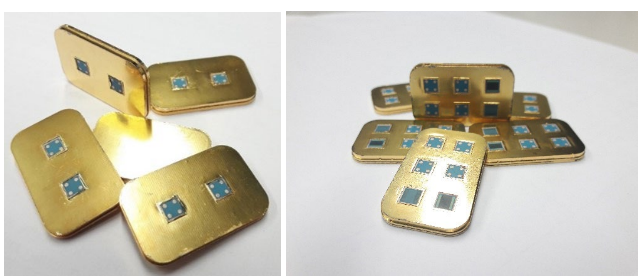


Figure 7 Ceramic sensory elements hermetically embedded in PCB-based signal modules

IV. Sensor characterization

For sensor characterization a small-volume component test stand was set up (fig. 8). The test stand can generate pressures of up to 2500 bar at temperatures of up to 200°C.

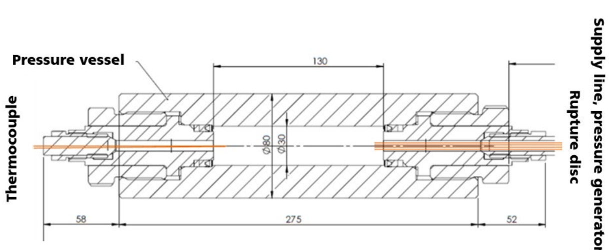


Figure 8 Water filled pressure vessel for sensor characterization under geothermal conditions

This allows electrical components to be tested in situ during the pressure tests and four-point measurements to be taken on sensors inside the pressure chamber. The electrical connection to the signal modules was realized via insulated high temperature cables and high temperature molding compound connected to the ceramic integrated vias.

Fig. 9 shows the results of a measurement campaign to characterize the ceramic sensor elements under geothermal conditions. The signal module is characterized outside and inside the container. After transfer into the water filled container, the pressure was increased from 1 to 1157 bar. The ceramic temperature sensor was used to continuously determine the media temperature. The piezoresistive resistance of the ceramic pressure sensor follows the applied pressure changes. Lowering the medium pressure leads to a reversible change in the measuring resistance.

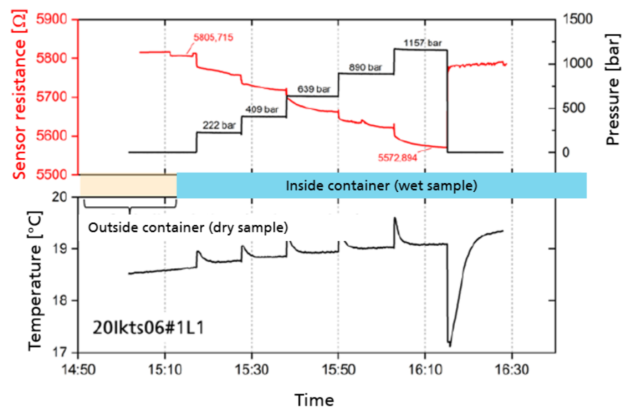


Figure 9 Measurement results of a pressure cycle of signal module within geothermal test stand

To increase the measuring accuracy (repeatability, linearity, and hysteresis), the sensor elements can be pre-aged. The usefulness of such measures in relation to the target application is the subject of future investigations.

V. Conclusions

As part of the investigations, the possibility of using multilayer ceramic technology for geothermal applications was examined. Initially, corrosion experiments were carried out to characterize and select specific ceramic and ceramic-compatible materials. Subsequently ceramic sensor elements were manufactured and characterized using the selected materials. Based on an innovative assembly and connection technology, the ceramic sensor elements were integrated into signal modules, which were characterized in suitable high-pressure and high-temperature test benches. The test results demonstrate the usability of multilayer ceramic sensor components under these extreme environmental conditions and can thus contribute to the digitization of the geothermal industry.

Acknowledgment

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