Contactless Conductivity Measurement of Ion Concentrations in Solutions

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In this work we present a device manufactured in low temperature co-fired ceramics (LTCC) technology for the contactless conductivity measurement of ion concentrations in aqueous solutions. The signal gets coupled into the microchannel across a ceramic plate of high permittivity. This allows better detection sensitivities as compared to other materials such as glass or PMMA. Finite element modeling (FEM) was carried out to underpin this advantage. First measurements with different concentrations of ion solutions were conducted.

Introduction

Contactless conductivity detection is a powerful and versatile sensing method for biochemical applications and is extensively employed for miniaturized analytical devices [1], [2]. It belongs, covering amperometric, potentiometric and contact conductivity detection, to the category of electrochemical detection. Common advantages in contrast to fluorescence- or absorbance-based systems are the relatively small dimensions of both the detector and read-out electronics. Compounds can furthermore be sensed without prior modification, and chip materials do not need to be transparent.

The working principle of contactless conductivity detection is based on capacitively coupling the excitation signal by either two or four electrodes into the microchannel [3] – [5]. Fluid and electrodes are not in direct contact with each other, thus preventing fouling processes and increasing life time of the device.

Low Temperature Co-fired Ceramics (LTCC) as a low-cost alternative for the fabrication of microanalytical devices is gaining increased interest. In its pre-fired state, LTCC is a flexible tape and can be easily structured using stamping, cutting or laser ablation techniques. Microchannel structures can so be fabricated in a multilayer arrangement [6].

Contactless conductivity detection in combination with PMMA or glass suffers from very low capacitive coupling into the channel. Employing ceramics with high values of permittivity (high-K ceramics) overcomes this drawback.

In our work a LTCC microfluidic device with a contactless conductivity detector has been fabricated. The detector consists of two planar electrodes separated from the channel by a high-K ceramics plate. Since LTCC tapes are not available with very high permittivities, a HTCC ceramic plate is used for the fabrication of the detector. Changes in the detection signal due to varying concentrations of an NaCl solution were measured. The benefit of high-K ceramics compared to low-K materials is pointed out using finite element modeling (FEM).

Device Fabrication

For realization of the device the A6-M LTCC-tape of FERRO with a thickness of 100 µm (unfired) has been applied. A stack of 7 layers of tape was laminated to provide mechanical stability. Micromachining of channels and holes has been conducted with a diode pointed NdYAG laser.

The channel cross-section is 100 µm x 100 µm, the length is 60 mm. Total thickness of the device is 700 µm. The top layer contains a detection window for inserting a planar detector after the firing process. Lamination was carried out at a pressure of 17 N/mm^2 and a temperature of 70 °C in an uniaxial press. Firing of the laminated stack has been conducted in a conventional belt furnace with a peak temperature of 850 °C and a total cycle time of 90 minutes. Figure 1 shows the fully assembled LTCC device.

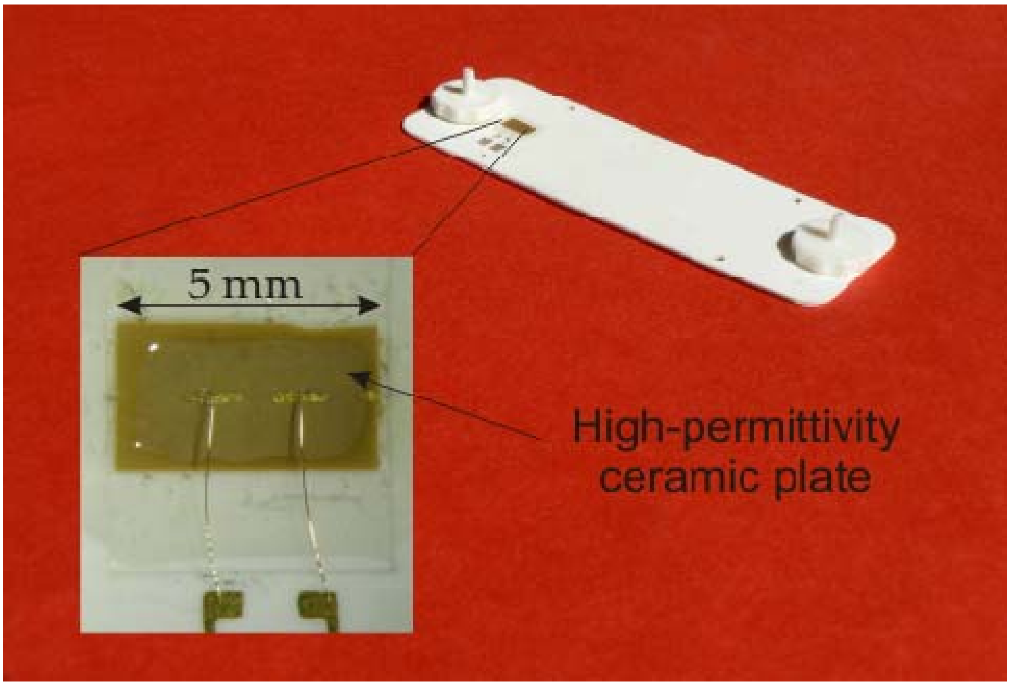

Fig. 1: LTCC device with planar detector-ceramic mounted in the recess above the channel (see inset). Electrode dimensions are 1 mm x 100 µm. Electrodes were wire-bonded to contact pads positioned on the LTCC substrate.

The planar detector was realized using a high-K HTTC ceramic plate with a permittivity of ε_r = 2000. It has a thickness of 300 µm. Gold electrodes were screen-printed on top using thick-film technology. After the firing process, the detector was glued into a cavity in the top layer of the LTCC device, enabling a direct contact between microchannel and high-K ceramic plate. The inset in Figure 1 depicts a detailed view of the detector. In order to reduce potential stray capacitance effects, the electrical connection of electrodes was not performed by conductor paths but by wirebonds.

Detector Simulation

A main benefit of the planar-detector arrangement is the ease of production, as electrodes need not be placed inside the channel. The high permittivity of the detector plate provides a better coupling of the excitation signal into the channel as compared to other low-K materials such as PMMA or glass.

Figure 2 shows FEM results of the real part of the electric current flowing into the detector at an excitation frequency of 200 kHz and a voltage of 5 V. For higher values of permittivity the inflowing current increases, indicating a higher coupling of the measurement signal with the channel. A problematic factor, however, is the increased stray capacitance across the ceramics short-circuiting the detection signal and thus lowering sensitivity. This can be tackled by optimizing electrode layout and measurement frequency.

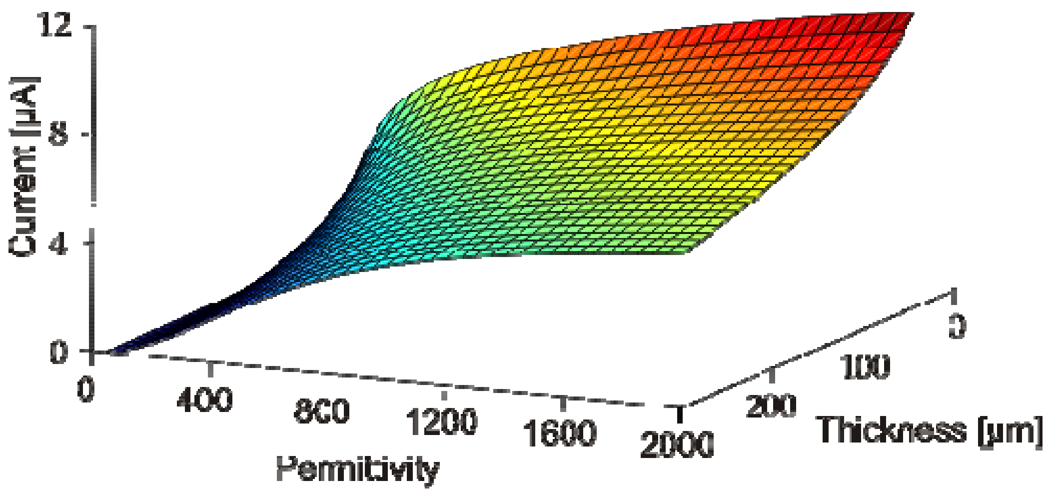

Fig. 2: FEM results of the electric current flowing into the detector for different values of ceramics permittivity and thickness. Simulation parameters for the channel were $\varepsilon_r = 80$ and electrical conductivity $\sigma = 0.1$ S/m. This corresponds to an ion concentration of about 10 mM for the mentioned channel dimensions.

The simulation results show that a decrease of ceramics thickness further improves detection sensitivity. In the proposed detector a plate with a thickness of 300 µm is used – a value that could not be further decreased due to mechanical stability issues. A possible workaround is the use of a thinner high-K LTCC tape instead of the HTCC plate used here. Such tapes are available at thicknesses of 100 µm in their unfired state; however, their permittivity is nevertheless much lower as compared to the HTCC plate used in this work.

Stray capacitance can be almost entirely suppressed using an opposite electrode setup as employed in [5]. Fabrication of such an arrangement, however, is somewhat more complex than that of the planar design, as stability and wiring issues have to be considered and photolithographic steps are necessary.

Results

Detector sensitivity was determined by measuring the change in impedance for different concentrations of an NaCl solution. Starting from and ideal infinite dilution (i.e. DI-water), solutions were pumped through the device with increasing concentrations (1, 2, 5 and 10 mM respectively). Figure 3 shows the resulting change in impedance and phase.

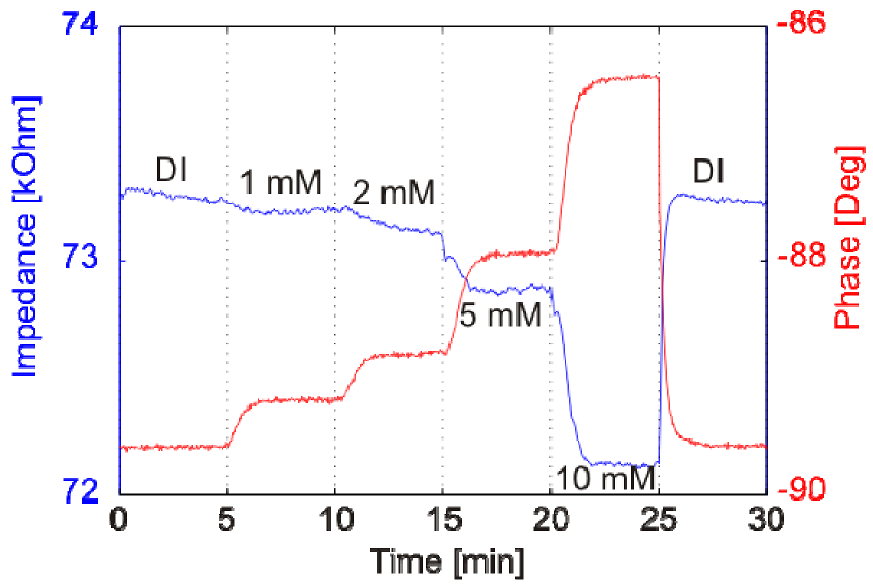

Fig. 3: Measured changes of impedance and phase over time for varying concentrations of an NaCl solution.

A Quadtech LCR-meter (Model: 7600) set to a measurement frequency of 200 kHz and a signal voltage of 5 V was used to obtain impedance and phase values.

In the case of the channel filled with DI-water, the device showed almost purely capacitive behavior. Due to resistive losses in the ceramics, however, the phase angle is somewhat bigger than the expected -90°. At intervals of 5 minutes the ion concentration was increased. The corresponding changes in impedance and phase did not happen immediately, but took place between 60 and 90 seconds to settle at a stable value. Increasing ion concentration of the solution contributes to a decrease in impedance as well as phase angle. The smallest concentration value that could be distinguished was 1 mM, with a corresponding change in impedance in the range of 1000 ppm.

Reproducibility of the measurement was confirmed by immediately changing from the 10 mM solution to DI-water and comparing the result with the value of the impedance recorded 30 minutes earlier. After a settling time of 90 seconds it reached the same value, indicating that the ceramics did not change its physical properties when in contact with the ion solution. Long-term effects such as possible changes in permittivity due to permanent contact with the fluid are being examined at the moment.

Conclusion

An LTCC microfluidic device with a contactless conductivity detector made of high-K ceramics has been introduced. Conductivity measurements were carried out with NaCl solutions ranging down to a concentration of 1 mM. The advantage of better coupling as compared to low-K materials was shown. FEM was employed to estimate the influence of different permittivities and thicknesses of the ceramic plate on detection sensitivity.

In microfluidic devices made of ceramics the material porosity is a serious matter that can influence the performance of the device. Long-term effects such as changes in permittivity due to contact with fluids in the channel are being examined at the moment and will be reported on.

References

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