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Fabrication and fluidic characterization of static micromixers made of low temperature cofired ceramic (LTCC)

G.A. Groß[a,*], T. Thelemann[a], S. Schneider[a], D. Boskovic[b], J.M. Köhler[a]

[a]Institute for Micro- and Nanotechnologies, Technische Universität Ilmenau, Gustav-Kirchhoff-Str.7, D-98693 Ilmenau, Germany
[b]Fraunhofer Institute for Chemical Technology, P.O. Box 1240, D-76318 Pfinztal, Germany

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Abstract

Microreaction devices used for chemical synthesis must possess a high resistance against corrosive chemicals. Therefore, microreaction devices were made of glass, steel or ceramics. Photolithographic steps combined with etching processes as well as micropowder blasting or micromilling processes were applied for the formation of appropriate structures. The low temperature cofired ceramics (LTCC) technology combines easy structuring, assembling and packaging techniques with the high chemical resistance of a glass ceramic material. In contrast to the known ceramic technologies, the LTCC technology enables a fast and easy fabrication of microfluidic devices. Here, we present two micromixers made of LTCC and its fluidic characterization. Laser ablation was used for the structuring of green tapes which were layered and cofired to form the micromixers. X-type fluidic barriers were realized inside a 1 mm^2 squared meandered channel of about 160 mm length. A meandered channel mixer without X-type mixing structures was used as a reference. The pressure drop was measured for aqueous media with various viscosities and the friction factors were calculated. An exponential equation for the friction factor prediction is given. The residence time distribution was determined for both devices by pulse trace experiments and the dispersion model was used to describe the residence time distribution for low Reynolds numbers.

Keywords: Microreactor; Micromixing; Microfluidic; Pressure drop; Friction factor; Residence time distribution; RTD; Pulse trace; Lab on chip; Lab on substrate

1. Introduction

During the last decade, microreactors were developed for different purposes (Ehrfeld et al., 2000; Hessel, 2004, 2005; Imanaka, 2005; Geyer et al., 2006). In many cases, the chemical stability, pressure stability and the thermal stability are important factors for the choice of reactor material (Ehrfeld et al., 2000). Microreactors made of steel (Hessel, 2004, 2005), glass (Hessel et al., 2003) or silicon (Jensen, 2006; Kirner et al., 2004; Schwesinger et al., 1996; Tiggelaar et al., 2006) were used for applications with corrosive chemicals and pressure processes even at higher temperatures. In particular, ceramic materials combine high chemical stability with high thermal stability. There are few examples for the introduction of ceramic materials in microreactors (Mengeaud et al., 2002; Knitter and Liauw, 2004; Meschke et al., 2005; Freimuth et al., 1996).

Materials and technologies for the fabrication of microreactors must be selected with respect to the suitable material properties of reaction chambers and channel walls. In addition, the material choice and formation technologies have to match other functionalities like integration possibilities of actuators and sensors. The silicon based technology is very well suited for highly defined structures and integration of miniaturized electronics. Sometimes, the application of silicon for microreactors suffers from the non-transparency of the material. In principle, this problem can be overcome if transparent windows are installed on—or in—the silicon devices.

The refinement of conventional ceramic materials for microreactor formation is complicated due to the material hardness and the required efforts for structuring. Here, low temperature cofired ceramic (LTCC) is a very interesting alternative

* Corresponding author at: Department of Physical Chemistry and Microreaction Technology, Institute for Micro- and Nanotechnologies, Technische Universität Ilmenau, Weimarerstr. 32, D-98693 Ilmenau, Germany. Tel.: +49 3677 69 3152.
E-mail address: Alexander.gross@tu-ilmenau.de (G.A. Groß).

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which allows not only an easy structuring of substrates but also possesses unique packaging possibilities (Gongora-Rubio et al., 2001). The LTCC technology was developed for the assembling and packaging of microelectronic devices (Imanaka, 2005). Different types of transducers or other electronic elements can be integrated during the assembling process by screen printing of appropriate structures (Gongora-Rubio et al., 1999; Thelemann et al., 2002). The flexible "green" tapes are composed of glass and ceramic particles (e.g. aluminum oxide) combined with an organic binder which can be transformed into a rigid glass ceramic material using a two step tempering process (details see in Imanaka, 2005). First, the organic binder is removed by firing at a "low" temperature. Secondly, the remaining inorganic material is transferred into a rigid glass ceramic by heating up to the melting point of the glass. Hereby, the glass and ceramic particles are sintered. Ideally the ceramic particles are completely covered by glass and the surface roughness is conditioned by the size of the embedded particles. Remaining device possess an over-all closed glass surface. Green tapes are commercially available in various thicknesses and can be mechanically structured by cutting, embossing or laser ablation. Conductive paths, resistive heaters or other planar electronic elements can be printed by screen printing onto the green tapes by using appropriate printing pastes. Functional glass ceramic devices are received if various layers are packaged by lamination before firing. Conductive connections between the single layers can be realized by the so-called vias. Transparent windows were bonded onto LTCC-channels to enable optical and photometrical measurements (Mülln et al., 2007). Different LTCC devices with channels, trenches and electronic elements were realized and used for microfluidic applications before (Birol et al., 2005; Golonka et al., 2002, 2005, 2006; Peterson et al., 2005; Thelemann et al., 2007). Analogously to "Lab on Chip" devices, multifunctional LTCC devices with integrated fluidic-, electronic-, sensorial- and optical- functionalities enable the formation of "Lab on Substrate" devices.

Here, we describe the formation principle and fluidic characterization of two micromixers made of LTCC. X-type fluidic barrier elements were realized inside a 1 mm^2 meandered channel of about 160 mm length. A channel mixer without X-type barriers was used as a reference system. The friction factors for both devices as well as the residence time distributions were determined. Described data enable the prediction of the pressure drop and residence time distribution in a *Re* number range from 3 to 300.

2. Fabrication of the LTCC reactors

The LTCC technology and material system enables the easy creation of complex three-dimensional microfluidic structures through structuring and assembling before the material is transformed into a rigid glass ceramic device. "Green" substrates can be packaged easily by lamination. The single process steps (A–F) for the formation of microfluidic LTCC devices are shown in Fig. 1. Initially, green tapes were shaped in smaller pieces and equipped with aligning holes by punching them before use. Received tapes were positioned on a laser ablation system and structured by stepwise ablation (Service by TETEKERA-Gmbh, Ilmenau, Germany). The X-type fluidic barrier parts were made of a stack with five tape layers (about 0.2 mm thickness each) which were assembled by isostatic lamination (70 °C, 200×10^6 N/m^2) before structuring. This stack was double sided laser refined in order to form the fluidic structures (see Fig. 2). Double sided laser refinement was necessary to overcome the problem of undercutting. All structured and non-structured layers were stacked and laminated to a package which was transformed into a rigid glass ceramic module by firing following the two step process. During this "co-firing" process, the organic binder was first removed at about 450 °C within 1 h. Secondly, the sintering was done at about 850 °C within 10 min. During these firing steps the package volume shrink about 15%. The individual microfluidic devices were individualized by sawing. To prevent plugging of the fluidic channels by the sawing dust the final fluidic openings were made at the end through a final laser ablation. For the fluidic characterization experiments both mixing devices were connected to the fluidic periphery using PTFE flange connections.

Two inherent disadvantages of the LTCC material system are the non-transparency and the surface roughness compared to pure glass materials. Remaining devices possess a blue color.

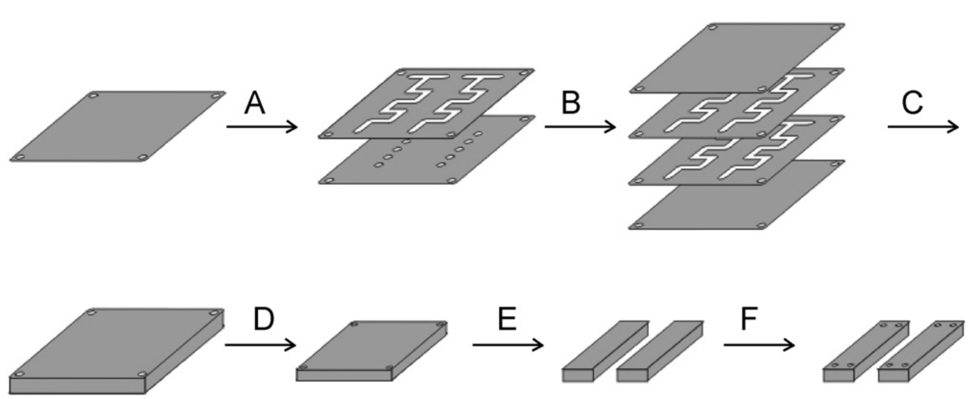

Fig. 1. Schematic preparation steps for LTCC microreactors. A: Mechanically structuring by punching, cutting or laser ablation. B: Stacking of construction layers. C: Isostatic lamination. D: Firing, shrinking takes place. E: Dicing. F: Fluidic openings by laser ablation.