

DEVELOPMENT OF MESO-SCALE FUEL REFORMER USING LOW TEMPERATURE CO-FIRED CERAMIC TAPE TECHNOLOGY

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Abstract: Application of LTCC tape technology to complex 3-D meso-scale microfluidic and reacting flow devices has drawn increasing attention. There are advantages to applying this technology for reacting flow systems due to the relatively superior material properties and ease of the fabrication procedures. Utilization of the technique on integrated devices has been demonstrated. LTCC tape technologies have also been used as the packaging substrate for catalyst in meso-scale reformers. It is found that the microfluidic channels and Pt catalytic layers in the LTCC reformer can be integrated by direct co-firing without additional processes. Performance characterization of the meso-scale fuel reformer is underway. Influences of parameters such as feeding rate of fuel, operating temperature, and pattern of the Pt catalyst on the reforming efficiency will be studied.

Keywords: LTCC, Fuel reformer

INTRODUCTION

In the past decades, meso-scale fuel cell based power systems have been extensively investigated due to the high energy efficiency [1-3]. Reformer is an important component of the fuel-cell-based power system. It converts hydrocarbon fuels to hydrogen. System integration is among the key issue of micro-scale reformer development [4, 5]. The recent progress in LTCC reacting flow devices has made LTCC a promising material and fabrication technology for developing highly integrated meso-scale fuel reformers. We present the development a meso-scale fuel reformer with co-fired Pt catalyst.

Application of LTCC tape technology to complex 3-D meso-scale microfluidic and reacting flow devices has drawn increasing attention [6-9]. The concept and methodology of this technology are based on packing layers of patterned LTCC tapes to form 3-D structures [6]. It is easier to fabricate structures with high aspect ratio and complexity using LTCC tape technology than using conventional materials and technologies, such as silicon-based MEMS technology. Since LTCC was originally developed for packaging microelectronic devices, it is compatible with screen-printing technology, which allows the integration of a variety of electronic elements and metal wirings.

Since LTCC tapes in green state are flexible, soft, and thin, various machining techniques, such as mechanical punching, CNC machining, and laser cutting, can be applied to fabricate vias, channels, cavities, and holes [7]. After sintering, the multi-layered green tapes turn into a solid block. It can be used to make microdevices with high rigidity and chemical stability, and such block with the nature of green tapes is resistant to harsh thermal and chemical environments. Utilizations of the technique on microcombustors [8] and chemical microthrusters [9, 10] have been shown in previous studies.

It has been demonstrated in several studies that LTCC tape technologies can be used as a packaging substrate and serve as a catalyst in meso-scale reformers [11, 12]. We demonstrate the feasibility of integrating platinum (Pt) catalytic layer in a meso-scale reformer using direct sputtering of Pt on green LTCC tapes. The design, fabrication, and preliminary testing of the meso-scale fuel reformer will be presented.

DESIGN AND FABRICATION OF THE LTCC REFORMER

The reformer consists of the upper and lower blocks with Pt sputtered on the inner surfaces, and a central block with fluid channels is shown in Fig. 1. The dimensions of the serpentine type rectangular flow channels are 500 μm \times 300 μm \times 934 mm. The thickness of each block is 300 μm . The total size of the LTCC reformer is 34 mm \times 34 mm \times 2.7 mm.

The process of fabrication is illustrated in Fig. 2. Each of the three blocks are made of three layers of 125 μm thick green tapes (ESL, 41020) using a hot-pressing lamination process (3000 psi at 70 $^{\circ}\text{C}$, 10 minutes), and a CO₂ laser (LaserPro, Mercury II M-60) is used to micro-machine the channels and via holes on pre-laminated LTCC blocks in Fig. 3. Thin films of platinum catalyst (99.99% Pt) are coated using an ion sputter (Hitachi, E-1010), and the catalytic layer is 10 nm thick.

After applying a viscous organic fluid composed of 15% deionized water and 85% honey (by weight) on the interfaces between these blocks, the blocks are stacked and undergo a second lamination step (300 psi at 75 $^{\circ}\text{C}$ for three minutes). The whole meso-scale fuel reformer is then diced with a chopper to remove unwanted edges followed by sintering (co-firing) in a programmed oven (Lindberg Blue, BF51728C). Due to the use of organic fluid in the second lamination, the

temperature profile for co-firing has to be modified from the standard process in Figure 4. The temperature in the oven is linearly raised from room temperature to 450 °C in three hours, and is maintained at the temperature for 120 minutes before being increased to 850 °C for sintering. The oven is allowed to cool down to room temperature naturally.

Finally, two 1/16 inch stainless steel tubes are glued in inlet/outlet ports of the LTCC reformer with the ceramic adhesive (Flexbar, Autocrete 15030). Figure 5 shows the photograph of the LTCC reformer.

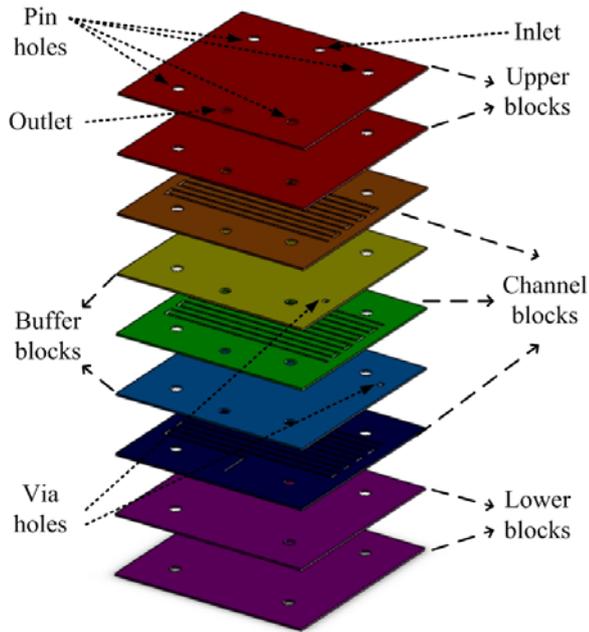


Fig. 1: Design of the LTCC meso-scale fuel reformer.

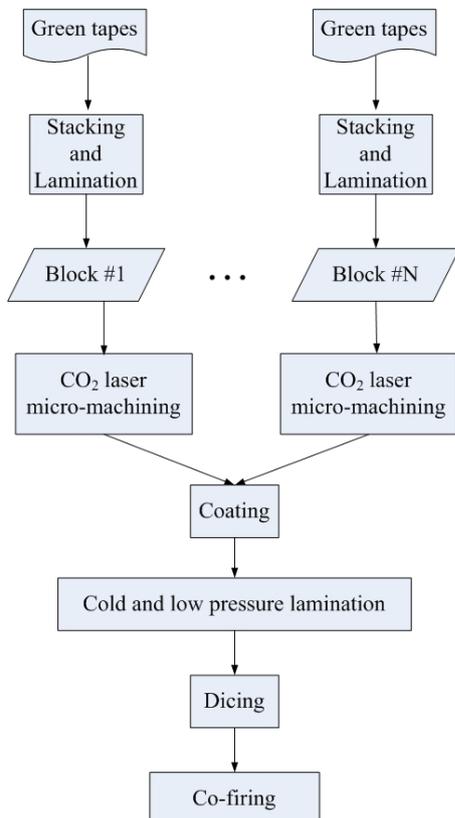


Fig. 2: Fabrication procedure of the LTCC meso-scale fuel reformer.

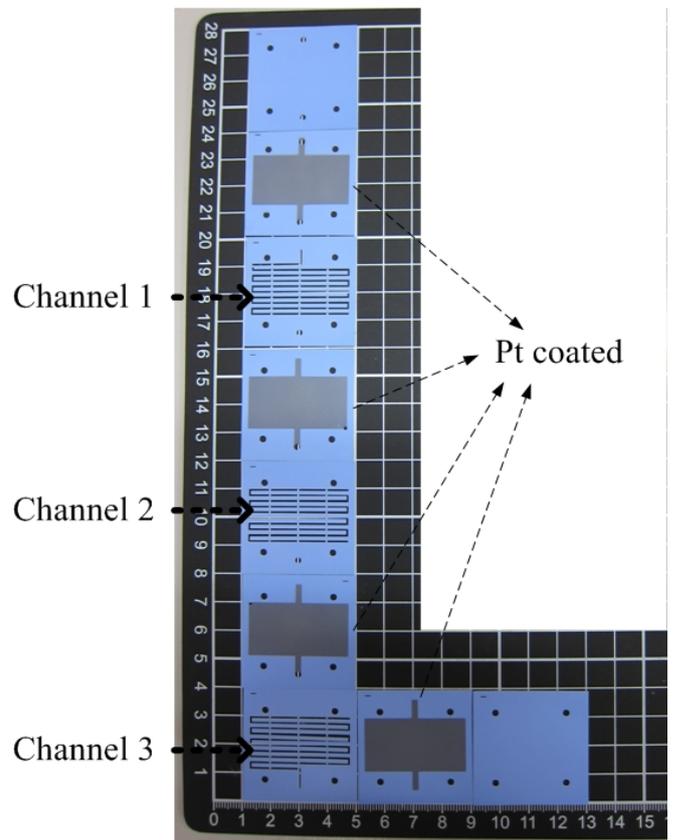


Fig. 3: Blocks after laser machining and Pt coated.

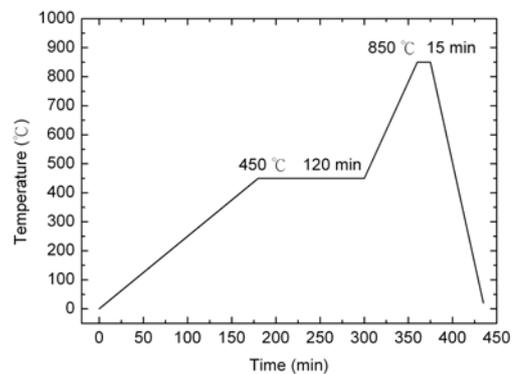


Fig. 4: Sintering profile of the LTCC.

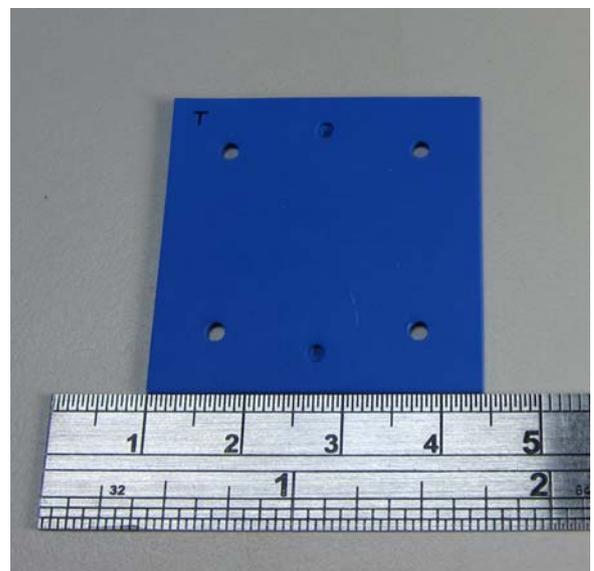


Fig. 5: Photograph of the LTCC reformer.

PRELIMINARY CHARACTERIZATION

The LTCC reformer was fabricated successfully. It has first been tested that there is no leakage in this reformer. Results also show that the sputtered Pt can be co-fired in the LTCC tape technology without inducing any morphing or distortion on the co-fired LTCC reformer.

In this study, methanol is used as a fuel to generate hydrogen. The reforming temperature of 500 K over Pt catalyst is relatively low. Moreover, methanol can be derived from a variety of source including biomass [13]. The reactions of methanol steam reforming to hydrogen are shown as the following:



Reactions (2) and (3) are represented methanol decomposition and water gas shift, respectively. Reaction (1) is the summation of reactions (2) and (3) and indicates the overall methanol steam reforming.

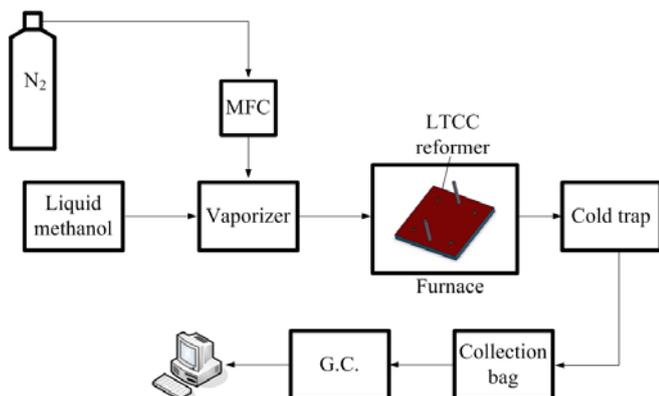


Fig. 6: Experimental setup.

A schematic diagram of experimental setup is shown in Fig. 6. The liquid methanol is heated at 110 °C in the vaporizer and the steam of methanol was transported with different flow rates (1, 10, 20 and 30 sccm) by nitrogen as carrier gas. The LTCC reformer was heated in the furnace at the temperature values of 150, 200, 250 and 300 °C. After reforming, the off-gas was collected in a collection bag through a cold trap. Finally, the compositions of the off-gas were analyzed using a gas chromatography (Agilent 7890A).

CONCLUDING REMARKS

A meso-scale fuel reformer is under development using LTCC tape technology. The overall size of the LTCC reformer is 34 mm × 34 mm × 2.7 mm. Serpentine type rectangular flow channel which is 500 μm wide, 300 μm deep, and 934 mm long has been fabricated. Platinum layers were successfully coated on inner top and bottom surfaces of the channel during green LTCC tape stage via sputtering. The 40 nm thick platinum will act as catalyst for methanol reforming in

the reformer. Performance characterization of the reformer is still underway. Different operating conditions such as feeding rate of a fuel, operating temperature, patterns of the Pt catalyst and the reforming efficiency will be tested.

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