Vertical-Cavity Surface-Emitting Laser (VCSEL)-Based Ultrafast Photonic Sintering of

Solid Oxide Fuel Cells (SOFCs): Prospects for Time-Efficient/Two-Dimensional

Scalability to Large-Sized SOFCs

Jaehwan Kim,^{al}, Saeed Ur Rehman,^{bl} Myeong-Ill Lee,^{cl} Amjad Hussain,^b Youngsu Noh,^c Jiwon Oh,^a Wonseck Ku,^a Na-Eui Kwak,^a Dohyoung Kim,^a Heesu Hwang,^a Hee-Sung Yoon^d, Seungho Park *^c, Seung-Bok Lee*^{b,e}, and Jin-Ha Hwang*^{a,c}
^aDepartment of Materials Science and Engineering, Hongik University, Seoul 04066, South Korea
^bHigh Temperature Energy Conversion Laboratory, Korea Institute of Energy Research, Daejeon 34129, South Korea
^cDepartment of Mechanical Engineering, Hongik University, Seoul 04066, South Korea
^dHtoEn Co., Ltd., Pohang 37778, South Korea
^eDepartment of Advanced Energy and System Engineering, University of Science and Technology, Daejeon 34113, South Korea

¹These authors contributed equally to this work.

*Corresponding Authors: Seungho Park, Seung-Bok Lee and Jin-Ha Hwang

*Seungho Park Tel: 82-2-320-1632 Email: spark@hongik.ac.kr *Seung-Bok Lee Tel: 82-42-860-3466 Email: sblee@kier.re.kr *Jin-Ha Hwang Tel: 82-2-320-3069 Email: jhwang@hongik.ac.kr



Anode support green films lamination into anode support layer

Supplementary Figure S1. A schematic showing the fabrication of 4-layer anode-supported green half-cell by hot press lamination and warm isostatic pressing.



Supplementary Figure S2. (a) Thermal profile prediction incorporating exposure of the SiC susceptors to VCSEL illumination and comparison with the empirical temperature measurement as a function of the VCSEL laser power.



Supplementary Figure S3. Thermal profile calculation incorporating NiO with SiC susceptors subjected to VCSEL-based IR laser illumination. (SiC dimensions: 30 mm x 30 mm x 2 mm; NiO dimensions: 6.9 mm diameter and 0.93 mm thickness.) Simulated temperature profile of NiO positioned onto a SiC susceptor: (a) overall view and cross-sectional view. (c) Thermal profile prediction as a function of VCSEL laser power. (Dashed lines indicate the empirical temperatures obtained by applying the corresponding laser powers (31, 40, 51, & 63%) for targeted temperatures (1100, 1200, 1300, & 1400 $^{\circ}$ C).



Supplementary Figure S4. Conventional thermal furnace processing and VCSEL-based photonic sintering (PS) processing applied to the fabrication of SOFCs. (a) Cell-1 fabricated through conventional thermal sintering only (with GDC/LSC cathodes and LSC current collectors), (b) Cell-2 fabricated through VCSEL-based photonic sintering (with GDC/LSC cathodes and LSC current collectors), and (c) Cell-3 fabricated through VCSEL-based photonic sintering (with a GDC scaffold infiltrated with LSC)



(a)

(b)





Supplementary Figure S5. Examples of processing defects found in VCSEL-based heat treatments. (a) Bubbling, (b) cracks originating from the unoptimized binder burnout process stage, (c) significant warpage, (d) cracks, (e) undesired phase formation, and (f) powder residue induced by the sintering (or densification) and flattening process stages.



(a)



(b)

Supplementary Figure S6. Video clips obtained during the VCSEL-based sintering process of tapecast composite materials composed of NiO-YSZ (ASL: ~675 μ m)/NiO-ScCeSZ (AFL: 12 μ m)/ScCeSZ (electrolyte: 7 μ m)/GDC (BL: 2 μ m): (a) binder burnout and 1st sintering and (b) 2nd sintering with the flattening process.



Supplementary Figure S7. (a) Sample images before and after VCSEL-based photonic processing applied to the SOFC cathode formation stage (photonic sintering process applied to Cell-2 and Cell-3): (a) formation of GDC/LCS composites and LSC current collectors and (b) formation of GDC scaffolds followed by LSC infiltration.

Supplementary Table S1. Detailed heat treatment information employed in conventional sintering (Fig. 2c) targeted at fabrication of high-performance solid oxide fuel cells. The summarized information includes step serial number, step process name, step start temperature and step end temperature, step process duration time, and total elapsed time (cumulative).

Step Serial Number	Step Process Name	Start Temperature [°C]	End Temperature [°C]	Time [hr]	Total Elapsed Time (Cumulative) [hr]
1	Pre-Heating for Binder Removal	0	350	20	20
2	Binder Removal	350	350	5	25
3	Pre-Heating for Carbon Removal	350	600	10	35
4	Carbon Removal	600	600	4	39
5	Pre-heating To 1 st Sintering	600	1330	9	48
6	1 st Sintering	1330	1330	3	51
7	Cooling after1 st sintering	1330	0	9	60
8*	Pre-heating for 2 nd Sintering	0	1400	14	74
9*	2 nd Sintering	1400	1400	3	77
10*	Cooling after 2nd ^t sintering	1400	0	10	87
11*	Pre-Heating for Cathode Formation	0	950	9.5	96.5
12	Cathode Formation	950	950	3	99.5
13	Cooling After Cathode Formation	950	0	9.5	109

*Flattening Process Incorporated

Supplementary Table S2. Detailed heat treatment information employed in VCSEL-based photonic sintering (Fig. 2d) targeted at fabrication of high-performance solid oxide fuel cells. The summarized information includes step serial number, step process name, step start temperature and step end temperature, step process duration time, and total elapsed time (cumulative).

Step Serial Number	Step Process Name	Start Temperature [°C]	End Temperature [°C]	Time [sec]	Total Elapsed Time (Cumulative) [sec]
1	Pre-Heating for Binder Removal	50	350	3000	3000
2	Binder Removal	350	350	300	3300
3	Pre-Heating for Carbon Removal	350	600	250	3550
4	Carbon Removal	600	600	120	3670
5	Pre-heating To 1 st Sintering	600	1300	700	4370
6	1 st Sintering	1300	1300	180	4550
7	Cooling after 1 st sintering	1300	50	250	4800
8*	Pre-heating for 2 nd Sintering	50	1100	525	5325
9*	Pre-heating for 2 nd Sintering	1100	1200	100	5425
10*	Pre-heating for 2 nd Sintering	1200	1320	240	5665
11*	2 nd Sintering	1320	1320	300	5965
12*	Cooling after 2nd ^t sintering	1320	50	1270	7235
13	2 nd Pre-Heating for Cathode Formation (GDC Scaffold)	50	1050	500	7735
14	2 nd Pre-Heating for Cathode Formation (GDC Scaffold)	1050	1250	200	7935
15	Cathode Formation (GDC Scaffold)	1250	1250	180	8115
16	Cooling After Cathode Formation (GDC Scaffold)	1250	50	600	8715

*Flattening Process Incorporated