Journal of Power Sources 283 (2015) 320–327

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Effectiveness of paper-structured catalyst for the operation of biodiesel-fueled solid oxide fuel cell

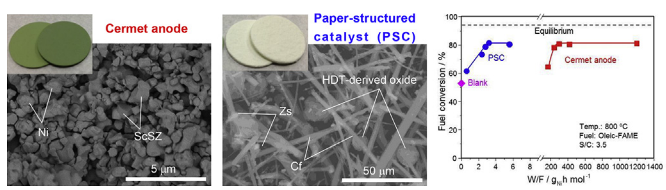
Tran Quang-Tuyen [a, b, *], Taku Kaida [a], Mio Sakamoto [a], Kazunari Sasaki [a, b, c, d], Yusuke Shiratori [a, b, c, d]

[a] Faculty of Engineering, Kyushu University, Motooka 744, Nishi-ku, Fukuoka 819-0395, Japan
[b] International Research Center for Hydrogen Energy, Kyushu University, Motooka 744, Nishi-ku, Fukuoka 819-0395, Japan
[c] International Institute for Carbon-Neutral Energy Research (WPI), Kyushu University, Motooka 744, Nishi-ku, Fukuoka 819-0395, Japan
[d] Next-Generation Fuel Cell Research Center, Kyushu University, Motooka 744, Nishi-ku, Fukuoka 819-0395, Japan

HIGHLIGHTS

- A model biodiesel fuel (BDF) was applied as an alternative fuel for SOFC.
- Novel paper-structured catalyst (PSC) containing Mg/Al-hydrotalcite was synthesized.
- PSC exhibited fuel conversion comparable to SOFC anode with less than 1/100 Ni weight.
- Long-term stability of SOFC fueled by BDF was achieved by the application of PSC.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 22 January 2015
Received in revised form
19 February 2015
Accepted 22 February 2015
Available online 26 February 2015

Keywords:
Paper-structured catalyst
Mg/Al-hydrotalcite
Steam reforming
Oleic-FAME
Solid oxide fuel cell

ABSTRACT

Mg/Al-hydrotalcite (HDT)-dispersed paper-structured catalyst (PSC) was prepared by a simple paper-making process. The PSC exhibited excellent catalytic activity for the steam reforming of model bio-diesel fuel (BDF), pure oleic acid methyl ester (oleic-FAME, $C_{19}H_{36}O_2$) which is a mono-unsaturated component of practical BDFs. The PSC exhibited fuel conversion comparable to a pelletized catalyst material, here, conventional Ni–zirconia cermet anode for solid oxide fuel cell (SOFC) with less than one-hundredth Ni weight. Performance of electrolyte-supported cell connected with the PSC was evaluated in the feed of oleic-FAME, and stable operation was achieved. After 60 h test, coking was not observed in both SOFC anode and PSC.

1. Introduction

High temperature solid oxide fuel cell (SOFC) can accept direct feed of hydrocarbon fuels. SOFC is expected to be the most efficient device for direct conversion of chemical energy into electricity [1,2]. Reforming of hydrocarbons can proceed in the porous cermet anode to produce H_2 and CO, which electrochemically oxidized to

* Corresponding author. Faculty of Engineering, Kyushu University, Motooka 744, Nishi-ku, Fukuoka 819-0395, Japan.
E-mail addresses: tran.tuyen@mech.kyushu-u.ac.jp, tqtuyen78@yahoo.com (T. Quang-Tuyen).

http://dx.doi.org/10.1016/j.jpowsour.2015.02.116
0378-7753/

Table 1
Composition of raw papers (RPs) after drying.

Type of RP	HDT/wt%	Cf/wt%	Zs/wt%	Cationic polymer/wt%	Anionic polymer/wt%	Pulp/wt%
HDT-free	0	86.1	8.6	0.5	0.5	4.3
HDT-dispersed	14.7	73.3	7.3	0.5	0.5	3.7

Table 2
Ni contents in the HDT-dispersed PSCs (for two pieces of each type of PSC with 16 mm in diameter and 1.1 mm in thickness) and pelletized Ni–ScSZ cermet anodes.

Paper-structured catalyst[a]	Ni loading/mg	Ni–ScSZ cermet anode[b] (Reference catalyst)	Ni loading/mg
PSC-1	0.6	Cermet-1	170
PSC-2	2.4	Cermet-2	240
PSC-3	2.8	Cermet-3	300
PSC-4	3.2	Cermet-4	430
PSC-5	5.6	Cermet-5	1200

[a] Weights of HDT, Cf and Zs in two pieces of each type of PSC are 18.9, 95 and 9.5 mg, respectively.
[b] Weight ratio of Ni:ScSZ is 1:1.

generate electricity and heat [3,4]. This operation is called direct internal reforming (DIR) operation, which brings us several advantages such as downsizing and cost reduction of fuel cell system and the reduction of energy consumption for the stack cooling by the supply of excess air due to strong endothermicity of the reforming reaction [5,6].

Nickel is commonly-used anode material for SOFC due to its excellent electro-catalytic properties. However, coking on Ni is a main drawback of DIR operation of SOFC fueled by light hydrocarbons [7–9] as well as heavy hydrocarbons [10–12]. Carbon deposition on and inside of porous anode material is one of the most crucial issues of DIR-SOFC, because it significantly degrades fuel cell performance as the results of the coverage of electrochemical and reforming reaction sites. To solve this problem, many approaches have been reported. The first approach is careful control of steam to carbon ratio (S/C). Excess amount of water is essential to promote reforming reaction, however high S/C ratio results in the reduction of electromotive force, furthermore deteriorates catalytic activity of the porous cermet anode due to oxidation of Ni [13,14]. The second approach is incorporation of coking-resistant catalyst materials such as copper [15,16], but low melting temperatures of copper and its oxides limit the use of them for SOFC anode [17,18]. Zhan et al. applied a thin Ru–CeO_2 catalyst layer on the top of a conventional Ni–YSZ anode [19]. They obtained stable cell operation without coking by the direct feed of fuel mixture of 5% iso-octane and 9% air diluted by 86% CO_2 to a SOFC single cell at 770 °C under current density of 0.6 A cm^{-2} for 50 h, although difficulties of fuel diffusion and current collection must be overcome.

Recently, flexible paper structured-catalyst (PSC) based on inorganic fiber network was developed to solve the problems associated with the DIR operation of SOFC mentioned above. The PSC exhibited excellent catalytic performance for dry reforming of methane [20,21]. Long term stability of SOFC fueled by pure oleic acid methyl ester (oleic-FAME), which is a main component of practical biodiesel fuels (BDFs), was confirmed by the connection of SOFC with PSC [22]. However, for the practical application of PSCs to SOFC systems, further improvement of catalytic performance is necessary. In this study, Ni loaded PSC with improved catalytic performance in which layered double hydroxide particles containing Mg^{2+} and Al^{3+} called Mg/Al-hydrotalcite (HDT) were dispersed in the paper-making process was tested for the stream reforming of oleic-FAME. Then, effectiveness of the HDT-dispersed PSC for the operation of biodiesel-fueled SOFC was demonstrated.

2. Experimental

2.1. Model fuel

Biodiesel fuel (BDF) is produced from biomass resources such as vegetable oil, waste cooking oil and animal fat by alkali catalyzed

Table 3
Component materials of anode- and electrolyte-supported cells prepared in this study.

	Anode			Electrolyte	Cathode	
	Current collector	Substrate	Functional layer		Functional layer	Current collector
Anode-supported[a]	NiO–ScSZ (30 μm)	NiO–ScSZ (800 μm)	–	ScSZ (22 μm)	LSM-ScSZ (30 μm)	LSM (30 μm)
Electrolyte-supported[b]	NiO–ScSZ (30 μm)		NiO–ScSZ (60 μm)	ScSZ (220 μm)	LSM-ScSZ (30 μm)	LSM (30 μm)

[a] Anode contains 550 mg of Ni metal.
[b] Anode contains 6.8 mg of Ni metal.

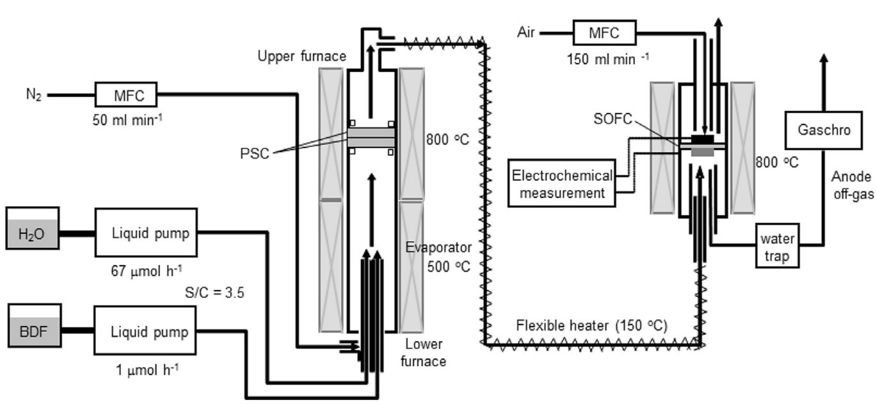


Fig. 1. Experimental setup for testing SOFC connected with PSC in the feed of oleic-FAME.