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Development of kerosene fuel processing system for PEFC

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Abstract

Several liquid fuels have been applied for fuel processing systems for polymer electrolyte fuel cells (PEFCs). In these fuels, kerosene is very attractive for the stationary fuel cells because of its low price and superior infrastructure in Japan. Relatively high sulfur content and high carbon atom number of kerosene, however, bring a disadvantage in reforming activity. To overcome such a technical disadvantage, both desulfurization adsorbent and steam reforming catalyst were developed, and furthermore a fuel processing system for kerosene is evaluated by simulation.

Keywords: Polymer electrolyte fuel cell; Kerosene; Reforming; Desulfurisation

1. Desulfurization technology

Petroleum-derived hydrocarbons are normally desulfurized by hydro-desulfurization at high temperature and high hydrogen pressure condition [1,2]. In a fuel cell system, however, desulfurization by adsorption would be more acceptable [1,2] in terms of efficiency and simplicity. This concept led us to develop a sulfur adsorbent for kerosene, working in a very mild condition of low temperature, atmospheric pressure, and without hydrogen addition [3–5]. A target life of the adsorbent is 4000 h, corresponding to 1 year on the assumption that the fuel cell system would be operated in daytime only.

The adsorptive desulfurization test of commercial catalysts and developed adsorbent were carried out under the condition of 100 times higher flow rate than standard. The developed Ni-based sulfur adsorbent reduced the sulfur content to 1 ppm, while commercial hydrogenation Ni catalysts, used as adsorbents here, lowered the sulfur content to 15–42 ppm (Fig. 1). Further study of preparation method and hydrogen reduction condition brought a very high dispersion of Ni, leading to the much better desulfurization performance (Fig. 2).

A nine times accelerated life test was conducted for the best adsorbent (named as IADS-147 shown in

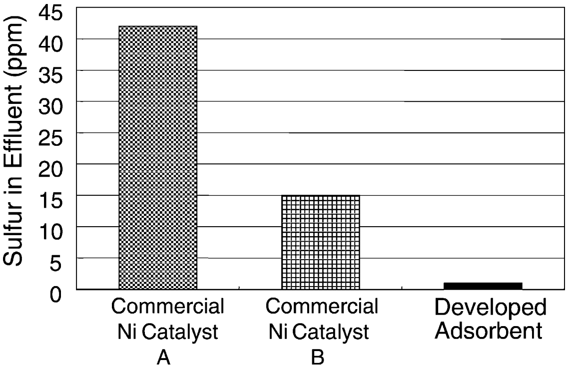

Fig. 1. Evaluation of sulfur adsorbent. Conditions—feed: commercial kerosene, JIS no. 1; *S*: 64 ppm; flow rate: standard × 100.

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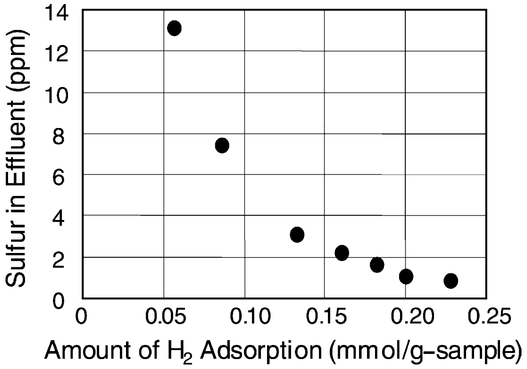


Fig. 2. Influence of Ni dispersion on performance. Conditions—feed: commercial kerosene, JIS no. 1; *S*: 48 ppm; flow rate: accelerated.

Fig. 2). As illustrated in Fig. 3, the sulfur in the effluent was below detection limit of 0.02 ppm during 4000 h, and the sulfur level reached 0.05 ppm at 5000 h. This result suggests that the target life time of 4000 h would be achieved.

2. Steam reforming technology

In terms of naphtha steam reforming, the authors reported that the Ru-based catalyst had much higher activity and stability than the Ni-based catalyst [6]. The Ru-based naphtha reforming catalyst (ISR-2G), however, was not applicable to kerosene steam reforming reaction because kerosene contains less active

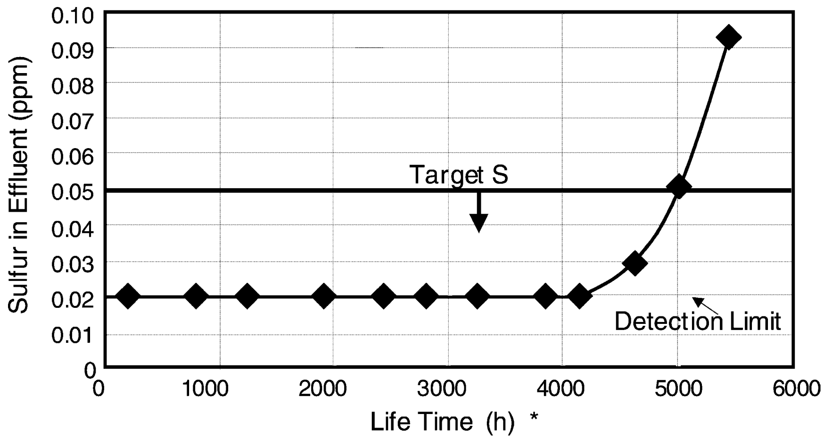


Fig. 3. Life test of developed sulfur adsorbent (IADS-147). ∗: calculated time, by considering nine times accelerated feed flow rate. Conditions—feed: commercial kerosene, JIS no. 1; *S*: 48 ppm; flow rate: standard × 9.

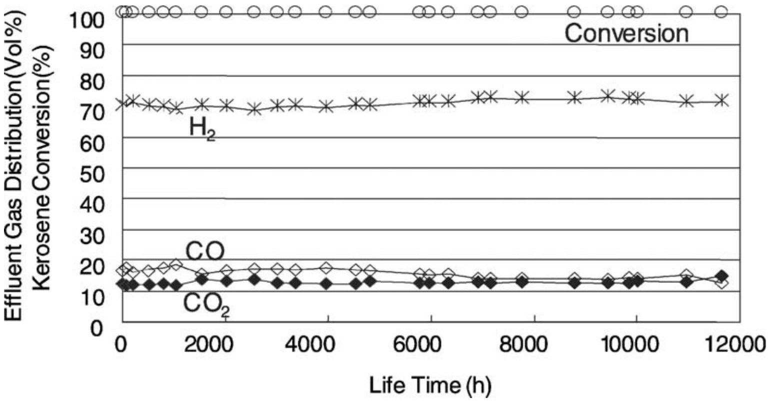


Fig. 4. Life test of developed reforming catalyst (ISR-7G). Conditions—outlet temperature: 730 °C; *S*/*C* molar ratio: 3; LHSV: 0.5 h^{-1}; feed: desulfurized JIS no. 1 kerosene ($S < 0.05$ ppm).

hydrocarbons, e.g. high molecular weight hydrocarbons and aromatic hydrocarbons. These hydrocarbons have also an inherent character to tend to bring much coke formation during the steam reforming reaction [7].

To overcome such difficulties, a catalyst (ISR-7G) for kerosene steam reforming was developed by using a carrier with higher surface area and higher heat resistance than was used for ISR-2G. Higher amount of Ru and a different additive were also adopted for ISR-7G. It showed 12,000 h catalyst life with 100% kerosene conversion and stable outlet gas composition (Fig. 4). This life test is still undergoing.

A target life of this steam reforming is 40,000 h, which corresponds to 10 years on the presupposition, as mentioned before, that the fuel cell would be operated in daytime only. Though ISR-7G has the potential to achieve the target life of 40,000 h, further improvement was attempted for a longer catalyst life. Thus, the obtained catalyst (ISR-8G) showed slower coke accumulation rate, and hence demonstrated higher stability than ISR-7G under an accelerated life test (Fig. 5).

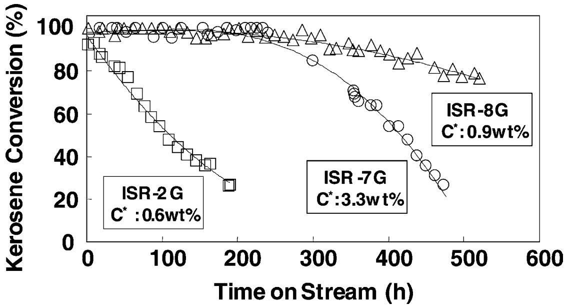
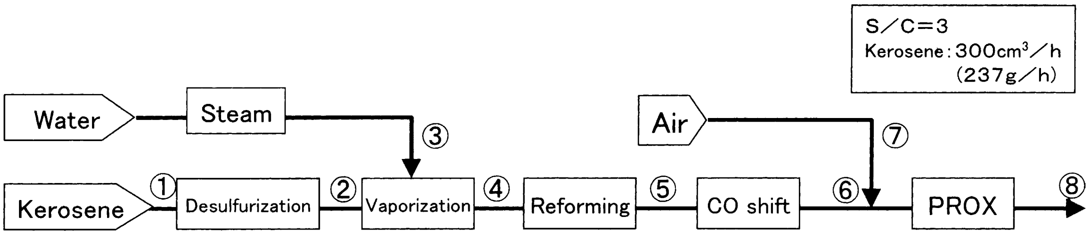


Fig. 5. Performance of improved catalyst ISR-8G. ∗: carbon accumulated during the reaction. Conditions: temperature: 730 °C; *S/C* molar ratio: 3; feed: desulfurized JIS no. 1 kerosene ($S < 0.05$ ppm).

3. Evaluation of kerosene fuel processing system

A kerosene fuel processing system was evaluated by simulation. Fig. 6 shows a process flow model and a material balance. The compositions at the outlet of desulfurization unit (②) and of reforming unit (⑤) were obtained according to the experimental results, using



	① g/h	② g/h	③ g/h	④ g/h	⑤ Nm^3/h	⑥ Nm^3/h	⑦ Nm^3/h	⑧ Nm^3/h
H_2					0.908	1.104		1.081
CH_4					0.005	0.005		0.005
CO					0.206	0.011		0.000
CO_2					0.170	0.365		0.376
H_2O(gas)			944.4	944.4	0.630	0.435		0.457
Kerosene(gas)				237				
Kerosene(liq)	237	237						
N_2							0.062	0.062
O_2							0.017	
Total					1.919	1.92	0.079	1.981
Temp.(°C)	R.T.	180			730	250	R.T.	150

Fig. 6. Simulation of kerosene processing system. ∗: ②, ⑤: from experimental results; ⑥: equilibrium; ⑧: estimation.

developed sulfur adsorbent and reforming catalyst, in independent bench scale reactors. The outlet composition of CO shift unit (⑥) was calculated from the chemical equilibrium at 250 °C. The composition in ⑧ was estimated on the assumption that, in PROX unit with O_2/CO molar ratio $= 1.5$, oxygen would convert all CO to CO_2 and excess oxygen would be all consumed by the reaction with H_2, producing H_2O.

The simulation at the above mentioned condition revealed that 300 cm^3/h (=237 g/h) of kerosene would supply 1.08 N m^3/h hydrogen. This suggests that 300 cc/h kerosene may be required for a 1 kW size fuel cell having 30% (HHV) power generation efficiency.

4. Conclusion

(1) Ni-based sulfur adsorbent (IADS-147) showed a lifetime longer than 4000 h for kerosene desulfurization.
(2) The developed kerosene steam reforming catalyst (ISR-7G) had the catalyst life of about 12,000 h. The life test is still undergoing.
(3) Kerosene fuel processing system was evaluated by simulation. It revealed that 1.08 N m^3/h hydrogen would be produced from 300 cm^3/h (237 g/h) of kerosene.

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