High purity hydrogen generation via catalytic partial dehydrogenation of kerosene Jet A-1

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Abstract

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High purity hydrogen generation via catalytic partial dehydrogenation of kerosene Jet A-1

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ABSTRACT

One of the objectives of the aviation industry is to provide a greener aircraft fleet environment. In this context a new generation of more electrified aircraft (MEA) is developed. This technology is partly based on the integration of a PEM fuel cell replacing the Auxiliary Power Units. To run the fuel cell, on-board hydrogen generation is an appropriate solution because it can be produced from kerosene JET-A1. In this work, the generation of hydrogen on board the aircraft is based on the concept of partial catalytic dehydrogenation of kerosene (PDh). The modified kerosene is then fed back into the fuel pool. Catalyst based on Pt-Sn supported on γ-Al2O3, were synthesised in this work and their evaluation in the PDh reaction indicate a good selectivity towards hydrogen (purity ≥ 97.8%) and rather high activity using Jet A-1 kerosene feed. Introduction In the context of a generation of a more electrified aircraft (MEA), fuel cells are a very attractive technology due to their high efficiency. One of the technologies of the MEA roadmap is a fuel cell secondary power generation unit to replace the Auxiliary Power Unit (APU). In order to supply the PEMFC with the required hydrogen on-board an aircraft, on-board hydrogen generation is the appropriate solution as it can produce the fuel from kerosene JET-A1, but is only feasible if the process is able to produce sufficient pure hydrogen in a compact, light-weight, and in a low-carbon footprint, sustainable way. Partial dehydrogenation (PDh) of kerosene JET-A1 on-board the aircraft could be a convenient way. The concept of PDh is a potential field of hydrogen generation, and only extracts hydrogen from hydrocarbons essentially yielding only the hydrogen and respective dehydrogenated hydrocarbon species that can be recycled to the fuel tanks (1). Given the appropriate catalyst and operating conditions (temperature, pressure, flow rate) the process should yield no additional by-products and in addition no other reagents are needed, like H2O or O2, unlike the other reforming concepts. The choice of the catalyst is crucial for the partial dehydrogenation process; it must produce H2 without compromising the original fuel properties. An ideal catalyst must be sulfur tolerant, generate sufficient hydrogen of high purity, be selective to dehydrogenation and avoid cracking reactions responsible for coke deposition and catalyst deactivation. Starting from catalyst based on Pt-Sn supported on γ-Al2O3 (2, 3), the effect of addition of In (4) and the influence of the nature of the support with the synthesis of a new alumina support with enhanced porosity properties have been investigated in this work. The evaluation of the catalytic materials in the PDh reaction indicates a good selectivity towards hydrogen (purity ≥ 97.8%) and rather high activity using Jet A-1 kerosene feed. Experimental A series of catalysts has been prepared by incipient wetness impregnation (IWI) of γ-Al2O3 (Sasol SCFa40 calcined at 500 °C-2 h) using aqueous solutions of H2PtCl6·xH2O, SnCl2·2H2O and InCl3. The first step is In impregnation followed by the co-impregnation of Pt-Sn solution (catalyst loading 1% Pt-1% Sn w/w in each case). Catalysts are finally calcined at 560 °C for 2 h. Catalysts were labelled as Cat Inx% where X is the In weight percent (0.25 %, 0.5 %, 0.75 %, 1 % w/w) while catalyst Cat. Ref. contains only Pt-Sn. A new alumina support with enhanced porosity properties has been synthesised using sugar as template: a water solution of AlCl3·6H2O (Sigma–Aldrich) was prepared and sucrose (Sigma–Aldrich) was added with a molar ratio Al:sugar:H2O 0.5:1:75. A solution of NH3 (30% wt.) was added drop-wise to adjust the pH to 5 while stirring at 500 rpm. The resulting gel was heated at 60 °C until dry and calcined at 600 °C for 6 h, using a ramp rate of 2 °C min⁻¹. The active phase (1% wt. Pt - 1% wt. Sn) was added by IWI. The resulting material obtained was labelled ALUSUC. Catalysts were characterized with a range of techniques including XRD (PANanalytical X Pert), surface area and pore distribution were determined from N2 adsorption-desorption isotherms (ASAP2020), TEM (JEOL 1200 EX), NH3-TPD and H2-TPR (Autochem 2910), TGA/DSC (Nietzsch STA409PC), GC (Agilent 7890A) dual column HP- PLOT molesieve 5A with TCD and HP- PLOT/Q with FID. The catalysts were tested for the dehydrogenation of kerosene in a stainless steel fix-bed tubular reactor. Catalysts were reduced at atmospheric pressure under a H2/Ar flow (4:6 v/v) at 350 °C for 2 h. After the activation step a 93% vol. vapour flow of kerosene (Jet A-1 Ultra-Low Sulfur Kerosene, having sulphur content < 3 ppm) with a 7% vol. of H2 (to simulate a recycle), was fed to the reactor containing pelletized catalyst (d =1-0,85 mm) at 450 °C and 10 bar. The flows were regulated in order to obtain a contact time of = 2 s (calculated at STP). After condensation of effluent partially dehydrogenated kerosene, hydrogen production was calculated by measuring the gas out-flow with a digital mass flow meter (Brooks S860S1 Smart II mass flow meter) and registering the data using LabView 8.2 environment. The productivity is expressed as NL h⁻¹ kgcat⁻¹, NL being the volume in normal conditions (273 K, 1 atm). Hydrogen purity was determined using a gas chromatograph (Agilent 7890A) equipped with a dual column system. Results and discussion Materials optimisation can be achieved either by modification of the active phase or by modification of the catalyst support. Modification of the active phase. The addition of a third additive to the active phase composed of Pt and Sn is an interesting option to improve the catalytic performances, therefore a study on the effect of indium on Pt-Sn/γ-Al2O3 has been carried out. The H2 productivity plot (Fig.1) represents the activity of the catalysts with different loadings of In. Figure 1: catalysts H2 productivity for different load of In. The amount of In strongly affects the performances. The reducibility of the active phase is improved and the activity towards the dehydrogenation reaction is increased. The number of strong acid sites is reduced, so avoiding undesired side reactions such as cracking, and consequently decreasing carbon coke formation (table 1) and improving catalyst stability. The presence of indium also decreases the amount of coke formed in the proximity of Pt particles, which serves to stabilise the catalyst activity. The catalyst Cat-Inx5% with metal ratio of 0.5:1:1 In:Pt:Sn has been identified as the best compromise between catalyst activity and stability. This catalyst allows the production of a hydrogen flow (purity 97.8%) of 2900 NL h⁻¹ kgcat⁻¹ correspond to 86.7 NL Lkerosene⁻¹ that can generate around 2.9 kW electricity (assuming a 50% efficiency for PEM fuel cell). The hydrogen yield is about 6.5% and is generated mainly from dehydrogenation of cycloalkanes to aromatics. Table 1: Catalyst performance and characterization data. Modification of the catalyst support. Surface area and porosity of the support are also key factors for the activity of the catalyst. A novel process for the synthesis of γ-Al2O3 has been carried out. The enhanced porosity displayed by this new alumina is obtained using glucose as template. The new catalyst material has a BET surface area of 226 m² g⁻¹ with a narrow pore distribution centered at 4,4 nm. The total acidity is 96 μmolNH3 g⁻¹. Figure 2: ALUSUC catalyst H2 productivity The hydrogen productivity is of 3500 NL h⁻¹ kgcat⁻¹ and the
stability is also consistently enhanced with a lifetime of 79 h. The amount of carbon coke formed after 6 hours reaction is 3.8 % wt. TGA/DTA analysis showed that the two exothermic peaks related to the coke combustion are shifted towards lower temperature, meaning that the carbon formed on the new catalyst is less strongly bonded to the surface and can be eliminated easily at lower temperature. Also, the first peak area at lower temperature is reduced considerably with the new catalyst respect the old one. In line with previous studies on carbon coke deposits (5), it’s hypothesised that the first peak is related to coke formed on the active metal phase and the second peak to coke formed on acid sites of the alumina support. Therefore, the stability of the ALUSUC is enhanced because the carbon coke deposited on the active phase is reduced. Conclusion In effect and modification of the alumina support on acidity, reducibility, activity and deactivation has been studied observing the correlation between those parameters. 97.6-98.2% pure H2 flow is produced with a yield range of 5.5-6.5% and the catalyst materials allow a production of H2 estimated to be sufficient for ≈3 kW electric power generation (assuming 1 kg cat. and 50% PEM efficiency). Partial dehydrogenation of hydrocarbons could be an interesting way for on board H2 generation and its applications. References [1] P. Janker, F. Nischke, C. Wolff. US Patent No. 20090274615A1, assigned to Airbus Deutchland GmbH (2009) [2] C. Lucarelli, S. Albonetti, A. Vaccari, C. Resini, G. Taillades, J. Rozière, et al., Catal. Today. 175 (2011) 504–508. [3] M. Taillades-Jacquin, C. Resini, K.E. Liew, G. Taillades, I. Gabellini, D. Wails, J. Rozière, D.J. Jones, Appl. Catal., B 142-143 (2013) 112-118. [4] E. Gianotti, Á. Reyes-Carmona, M. Taillades- Jacquin, G. Taillades, J. Rozière, D. J. Jones, Applied Catalysis B: Environmental, Volumes 160–161. [5] K.M. Hardiman, C.G. Cooper, A. a. Adesina, R. Lange, Chem. Eng. Sci. 61 (2006) 2565–2573

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