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Influence of the support in selective CO oxidation on Pt catalysts for fuel cell applications

Technical communication

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Abstract

One crucial requirement for the proton exchange membrane fuel cells (PEMFC) is to feed clean hydrogen to the anode, which is rapidly poisoned by traces of CO present from the upstream hydrocarbon reforming and water–gas shift processes. The removal of CO can be achieved by using catalysts able to selectively oxidize CO in the presence of excess hydrogen. Herein we report the effect of the support on Pt catalysts for total and selective oxidation (SELOX) of CO. The catalysts supported on ceria and zirconia presented higher activity than alumina and silica supported catalysts in SELOX reaction at low temperatures, but with lower CO conversions.

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1. Introduction

Fuel cells are receiving increased recognition as alternatives to existing power sources because of their theoretical high fuel efficiency and low-environmental impact [1,2]. The most promising fuel cell technology for transport applications appears to be the proton exchange membrane fuel cell (PEMFC) fueled by hydrogen, due to the fast start-up, high power density and low-temperature operation [2,3]. In order to avoid storing high pressure hydrogen on a vehicle, it has been proposed that hydrogen for PEMFC can be produced in an onboard fuel processor [4].

The production of hydrogen in onboard reformers actually occurs in three catalytic processes in series: (i) autothermal reforming of a hydrocarbon or alcohol for producing synthesis gas (fuel + O_2 + $H_2O \rightarrow CO_x$ + H_2), (ii) water–gas shift (WGS) reaction (CO + $H_2O \rightarrow CO_2$ + H_2), which maximizes H_2 yield and lowers the CO concentration; and (iii) selective oxidation (SELOX) of CO in the H_2 -rich reformate in order to reduce CO concentration to less than 10 ppm [5]. CO impurities contained in the reformer gas $(60-65\%H_2, 10-15\%H_2O, 1-2\%CO, balance CO_2)$ are known to poison commonly used anode catalysts (Pt, PtRu) and decrease the efficiency of the fuel cell to an unacceptable level [6].

The crucial requirements for the SELOX reaction are high CO oxidation rate and, equally important, high selectivity. Selectivity is crucial considering the competing H₂ oxidation that may lead to a decrease in the overall fuel cell efficiency. Besides oxidation of both CO and H₂, methanation and WGS reactions can also take place simultaneously. Thus, the catalyst should fulfill several requirements, namely: (i) to possess high CO oxidation activity, (ii) to exhibit high selectivity with respect to the undesired H₂ oxidation and, additionally, (iii) to function at the temperature region defined by the fuel processing unit (250–300 °C) and the operation temperature of PEMFC (80–100 °C).

Several catalytic systems have been tentatively tested in the SELOX reaction. Supported noble metal catalysts, such as Pt, Ru and Rh, were found to be the most appropriate for this reaction: CO can be totally converted to CO_2 but a noticeable amount of H₂ is also simultaneously consumed in the presence of excess O₂ [7–11]. For low temperatures, highly dispersed gold on an oxide support showed high activity and

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