



Structured catalysts with high thermoconductive properties for the intensification of Water Gas Shift process



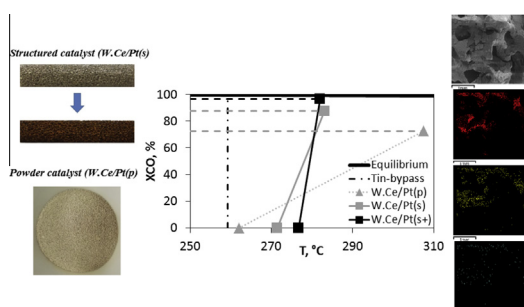
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HIGHLIGHTS

- Preparing structured catalysts over high thermal conductivity aluminum carriers.
- Strong adhesion of the catalytic washcoat layer to the metallic layer.
- Development of a nearly adiabatic laboratory system for the Water Gas Shift reaction.
- Redistribution of the heat of reaction with flattening of the thermal profile.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 22 April 2016

Received in revised form 22 June 2016

Accepted 24 June 2016

Available online 25 June 2016

Keywords:

Water Gas Shift
Process intensification
Aluminum foam
Structured catalyst
Washcoat
Heat transfer

ABSTRACT

The influence of the thermal properties of structured carriers on the performance of a highly active catalytic formulation, was investigated in the Water Gas Shift (WGS) process. Some aluminum foam structured catalysts were prepared by charging the carriers with a platinum/ceria/alumina-washcoat formulation; the catalytic performance were evaluated by providing a gaseous mixture, whose composition was chosen on the basis of a typical steam/carbon monoxide ratio at the outlet of a reformer. The activity of the structured catalysts and the thermal profile of the catalytic bed, were compared to those of a powder catalyst with the same composition of the washcoat formulation, in “quasi-adiabatic” condition, under equal conditions in terms of inlet temperature and weight hourly space velocity (WHSV). The experimental results showed much better performances for the structured catalysts, both in terms of CO conversion and temperature difference between the inlet and the outlet of the catalytic bed. This results were explained in terms of backdiffusion of the reaction heat throughout the structured catalysts bed, with a consequent flattening of the temperature profile, induced by the presence of the highly conductive aluminum foam carrier.

The foam was able to modify the adiabatic temperature profile, obtaining, with respect to a typical packed bed reactor, a higher temperature at the inlet section and a lowered temperature at the outlet bed section, overcoming respectively, the kinetic and thermodynamic limits, achieving so an increased CO conversion. Accordingly, a conventional double staged WGS reactors with intermediate heat exchange could be replaced with a single WGS reactor characterized by a “quasi isothermal adiabatic” temperature profile.

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1. Introduction

Process intensification is a way to optimize fixed and operating costs in an industrial plant. Thus, it is not only related to a

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minimization of the equipment's volume, but it is also related to the enhancement of both chemical and physical aspects of the process; this definition can be usefully interpreted as being a process development involving dramatically smaller equipment which leads to [1]:

1. Improved control of reactor kinetics giving higher selectivity/reduced waste
2. products.
3. Higher energy efficiency.
4. Reduced capital costs.
5. Reduced inventory/improved intrinsic safety/fast response times.

The importance of transport phenomena for process intensification in an industrial reactor is an important issue [2]. Moreover, further aspects could be also related to improvements in environmental impact, which is another critical issue that is constantly faced today [3].

The employment of catalysts with improved heat transfer properties may allow a safer control of a catalytic reactor, because they are able to prevent chemical phenomena such as thermal hotspots and runaway reactions, which can be crucial when treating of complex gas mixtures such as syngas [4,41].

One important process of the chemical industry which necessitates an intensification is the Water Gas Shift process (WGS) [5]. It is the first purification stage of a hydrogen production plant, and it involves the conversion of CO in CO₂ to increase the H₂ yield in syngas and to provide a H₂/CO mixtures with the appropriate ratio for the production of important chemicals, such as methanol or ammonia [6]. Considering a classical adiabatic reactor, the exothermicity and the equilibrium characteristics of the WGS reaction, together with the catalysts performances, reflects negatively on the CO conversion, disfavoring the reaction rate at the inlet of the catalytic bed, where the temperature is low, and limiting the very high conversion values at the outlet, where the temperature is high [7]. So, from the above considerations, the process intensification for the WGS has to be targeted to a modification both of the catalytic phase and the thermal profile along the catalytic bed. The first aim can be related to the development of an efficient catalyst in the low temperature range, where the CO equilibrium conversion theoretically achievable is higher [8]. The second aim is related to an improvement of the heat management in the whole system. This work is focused on the improvement of the heat transfer along the catalytic bed of the WGS adiabatic reactor. This intensification could be achieved by enhancing the heat conduction along the catalytic bed, which would also improve the overall reaction rate, by allowing a flatter temperature profile, approaching the outlet composition at a lower temperature. These two simultaneous approaches can lead to a very high CO conversion even in a single catalytic stage [9]. In this context, the thermal conductivity of the new catalyst plays a crucial role [10], the required catalyst should show not only high activity and selectivity but also high thermal stability and conductivity, low pressure drop and enhanced heat transfer [11]. An interesting alternative to the traditional catalysts is represented by structured ones [12]; in this case it is possible to exploit both the thermal conductivity of a metal structure, such as open cell foams, and the activity of a catalytic formulation in powder form, by coupling the two parts by means of a coating. Open cells foams can also enhance the mass transfer inside the catalytic bed, this being another important feature that makes them very interesting and promising for a process intensification [13].

Regarding the catalytic phase, in the literature many examples on the use of Au [14], Pt [15], Rh [16], Pd and Ru [17] are known, however the best performances are reported for Pt and Au. The lat-

ter in particular has shown excellent activity for LTS (Low Temperature Shift), while, at the same time, the tendency to deactivate rapidly because of sintering phenomena, since complex strategies are necessary to anchor the active phase to the support [18]. On the other hand, Pt based catalysts seem to be highly active and stable at the same time [19], by standing as the best choice for the preparation of a highly productive catalyst. Many studies on platinum based catalysts supported on reducible oxides are already known, for examples Tang et al. [20] have recently published the preparation by electrospinning of Pt/CeO₂ nanofibers with diameter of 80–120 nm, that show an excellent CO conversion in a temperature range of 320–360 °C. So, the aim of this work was to study the catalytic and thermal properties of a structured foam catalyst with regard to the WGS process intensification; the choice of the best foam material and the investigation on different platinum-based catalyst formulations in powder form were already reported in previous works [21–23]. The structured samples were characterized and tested in typical WGS reaction conditions. A comparison between the structured catalyst and the same catalyst in powder form was made. The stability of the structured catalyst was also investigated with a time on stream test. Finally, a nearly adiabatic system was set up, in order to compare the heat transfer influence on the whole system's performances.

2. Materials and methods

The structured catalysts are usually prepared by deposition of a catalytic formulation on the structured carrier, sometimes modified by the deposition of a washcoat formulation [24]. The use of a washcoat allows to increase the specific surface area of the carrier significantly [25] so, the chemical and physical affinity of the washcoat compounds with the structured support material is very crucial. For what regards the structured carrier, an Aluminum open cells foam (40 pores per inch, PPI, relative density = 6–8%) has been previously chosen as the best in terms of thermal properties between different types of foam investigated [22]. Therefore, the foam was loaded with an alumina-based washcoat, prepared as explained in the following section, while, for what regards the catalytic formulation, due to the results of previous works [21,23], a platinum/ceria based catalyst was chosen as the best compromise, from an economical point of view, between costs of materials and hydrogen productivity. In fact, platinum based catalysts allows to have high CO conversion and hydrogen yield even with very low metal loading [26]; on the other hand, the use of alumina reduces the amount of ceria which is needed for supporting the active species. In this way, the catalyst materials supply becomes cheaper [27].

2.1. Preparation of the washcoat

The washcoat slurry was prepared by suspending, under mechanical stirring, 15.4 wt% of γ -alumina (Puralox by Sasol), previously powdered to obtain a set of particle less than 10 μ m, in a colloidal solution of 4.6 wt% of pseudoboehmite (Pural SB by Sasol) and 1 wt% of metilcellulose (by Sigma Aldrich, viscosity: 4'000.00 cP) at pH = 4 [28]. The washcoat mixture has been finally vigorously stirred for about 24 h, to guarantee the perfect suspension of the solids content.

2.2. Preparation of the structured catalysts

The structured catalysts were prepared in two subsequent steps, the first involves a dip-coating procedure, the second an impregnation of the active phase. A cylindrical monolithic Aluminum foam (purchased by ERG Materials and Aerospace, 40 pores

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