



Structured catalytic reactor for soot abatement in a reducing atmosphere



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ABSTRACT

Nanostructured Li-delfossite based catalysts were prepared via solution combustion synthesis (SCS) method and investigated under realistic conditions as catalysts for the carbon gasification in a reducing environment. LiFeO₂ catalyst was selected as the most promising candidate for the soot gasification catalyst on a soot trap application close coupled to a biogas autothermal reforming (ATR) reactor for syngas post-treatment process. LiFeO₂ was coated on a wall-flow monolith filter (15/20 μm mean pore size and 46% porosity) via solution combustion deposition (SCD) method and the coated filters' performance was evaluated during the soot particles loading. The pressure drop across the filters was very low (< 8 mbar), showing that the method of coating on the filters was successfully applied.

1. Introduction

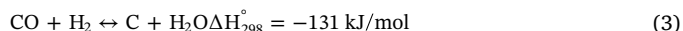
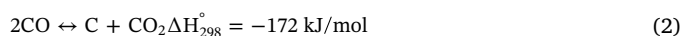
The ATR process couples the endothermic steam reforming (SR) and exothermic partial oxidation (POX) reactions, involving the main characteristics of these processes, a high H₂/CO ratio derived from SR and internal heating provided by the POX [1]. The ATR process also occurs in the presence of CO₂, combining the SR and POX with dry reforming (DR) process.

Biogas is a renewable resource, it is commercially produced from the anaerobic digestion of wastes such as municipal solid waste (MSW), manure and energy crops [2]. It is mainly composed of methane (CH₄) and carbon dioxide (CO₂) [3,4]. Biogas is an increasing waste valorization alternative instead of incineration or landfilling of untreated bio-wastes [5].

Hydrogen is an environmental friendly fuel. It is a clean energy carrier with zero carbon emissions, which can be produced in a very convenient way by the direct ATR of biogas (with no preliminary CO₂ separation) for its application in fuel cell cars [3,6,7]. An important and valuable feature of ATR is represented by the fact that heat is directly provided within the reactor, thus becoming a thermally self-sustained process, which reduces the need of heat exchangers and increases the flexibility of the plant [8–10].

Main constraints for optimum operation of the reformer are related to “secondary phenomena”, the formation of carbon [11]. Its formation is a major barrier for commercial application of biogas reforming process in the chemical industry [12]. Carbon formation cannot be

tolerated because it deposits on the surface of the catalyst which leads to the deactivation of the catalyst, increased pressure drop, and uneven flow distribution leading to overheating of the tubes locally as “hot spots” or totally as “hot tubes” [13,14]. Carbon formation can occur, both at the reactor inlet from methane decomposition (R. 1) and at the outlet (< 700 °C) by the Boudouard reaction (R. 2) or due to the reverse carbon steam gasification reaction (R. 3):



The principle of equilibrated gas predicts conditions in which carbon can be formed, but it does not guarantee that carbon is not formed if the principle predicts no potential in the equilibrated gas [11]. Formation of carbon can happen when the system is operating far from thermodynamic carbon limits [13]. It means that methane may decompose to carbon instead of reacting with steam to form the required syngas in spite of no potential for carbon in the equilibrated gas (R. 1). Moreover, this means also that carbon can be formed from the carbon monoxide decomposition (R. 2), or by the reaction between CO and H₂ (R. 3).

In the automotive industry, wall-flow filters are used for the efficient removal of particulate matter from gaseous streams [15]. These monoliths (commonly called Diesel Particulate Filters, DPFs) are ceramic porous structures that consist of many parallel channels of

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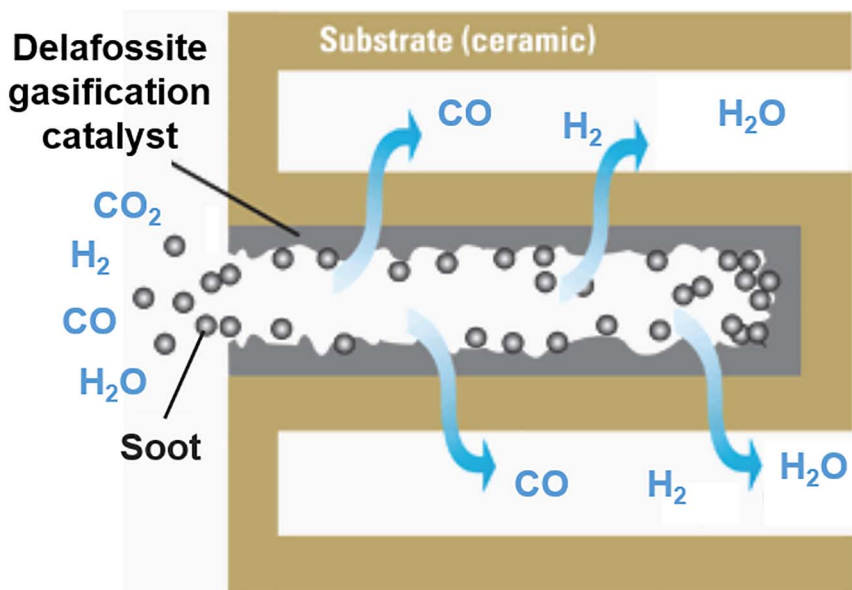


Fig. 1. Carbon gasification in a wall-flow monolith in a very reduced environmental.

rectangular cross section, with the adjacent channels alternately plugged [16]. A novel application of such filters has been developed by Raimondi et al. [17,18]. They have demonstrated that a trap located immediately downstream of a thermal partial oxidation (TPOX) reactor can block the soot there generated and thermally re-generate via soot gasification with the water and CO₂ contained in the syngas. This concept is implemented within the scope of this work using a nanostructured catalyst coating over the particulate trap, which could effectively improve the local catalyst-soot contact conditions (Fig. 1), a critical issue in this field.

The products of ATR reforming may consist of carbon dioxide, carbon monoxide, hydrogen, carbonaceous material (soot) and steam. The possibility of gasifying the soot in this specific environment can be achieved in-situ according to the reactions R. 2 and R. 3 (Fig. 1), creating a negligible pressure drop during the operation time of the processor.

This work deals with a detailed study of the carbon removal from inside a syngas environment (biogas reforming gases post-treatment process) by the use of a catalytic wall-flow trap based on transition metal catalysts. The functionality of the soot trap is firstly to trap the soot particles from the gas stream and then gasify in-situ these particles with the same syngas to avoid unacceptable back-pressure of the exhaust in the plant. Therefore, to ensure that the gasification occurs at the working temperature (around 700 °C), nanostructured delafossite catalysts have been synthesized and studied for this application. In addition, the work of coating and characterizing the wall-flow monoliths with respect to their performance has been carefully carried out.

2. Material and methods

2.1. Preparation of the catalysts

A series of Li–Cr delafossite catalysts (LiCrO₂, LiCr_{0.9}O₂, LiCr_{0.8}O₂, LiCr_{0.7}O₂, Li_{0.9}CrO₂, Li_{0.8}CrO₂, Li_{0.7}CrO₂), LiCoO₂, LiNiO₂ and LiFeO₂ were prepared via SCS method [19]. The method of catalyst synthesis is based on a highly exothermic and self-sustaining reaction [20], and it is particularly suited to produce nano-particles, homogeneous and crystalline powders [21,22]. According to the SCS technique, the precursors (metal nitrates and urea or glycine) are dissolved into distilled water in a beaker and then stirred for around 20 min until a completely homogeneous solution is obtained. During the stirring phase, the solution is heated at 40 °C. The concentrated aqueous solution with the precursors was placed in an oven at 650 °C for a few minutes in a crucible, so to

ignite the highly exothermic and self-sustaining reaction. Under these conditions, nucleation of metal oxide crystals is induced, but their growth is limited and nano-sized grains can be obtained.

2.2. Catalyst deposition

SiC filters with 15 and 20 μm mean pore size and 46% porosity were used. The filters were acquired from the ceramic filter manufacturer (Céramiques Techniques Industrielles S.A.). From the screening of the catalysts in the powder scale, LiFeO₂ catalyst was chosen as the most appropriate to be further deposited by in-situ SCD method directly over the wall-flow filters. The adopted procedure was similar to the one used for the catalyst powder preparation. The precursors (metal nitrates and urea or glycine) were mixed in a beaker with a volume of water and then agitated under a magnetic stirrer. The ceramic support was dipped in the aqueous solution with the precursors and then placed into an oven at 650 °C. The aqueous phase was rapidly brought to boil, the precursors mixture ignited and the synthesis reaction took place in situ. After total combustion, the filter is cooled to room temperature and then weighed. The procedure is repeated several times until reaching the desired weight of catalyst. Square monolithic segments monoliths (15 μm/46% and 20 μm/46%) with dimensions ~35 × 35 × 50 mm were coated with 10 g/l and 20 g/l of LiFeO₂.

2.3. Characterization of the catalyst

XRD and BET surface area analysis (Micrometrics ASAP 2010 BET analyzer) were performed to characterize the catalysts prepared. X-ray diffraction (XRD) patterns were obtained using an X'Pert Phillips diffractometer equipped with an X-ray source of Cu Kα radiation (λ = 0.15418 nm) at 40 kV and 30 mA. The specific surface area (SBET) of the catalyst prepared was determined with the Brunauer-Emmett-Teller (BET) method using N₂ physisorption at –196 °C on a Micrometrics ASAP 2020 instrument.

2.4. Catalytic activity test

The catalytic activity of the powder samples synthesized for soot gasification was analyzed by temperature programmed reaction (TPRe). The tests were carried out in a fixed-bed micro-reactor (a quartz tube with an inner diameter of 4 mm) placed in an electric, PID-regulated oven. A K-type thermocouple was put in contact with the packed bed to know exactly the temperature during the reaction. The catalytic bed

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