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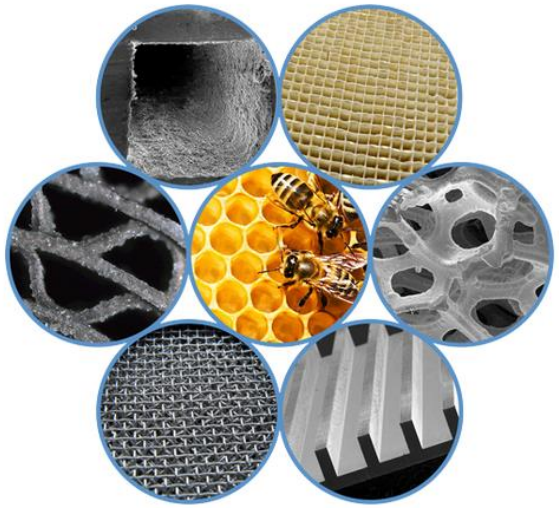

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STRUCTURED CATALYTIC REACTOR FOR SOOT ABATEMENT IN A REDUCING ATMOSPHERE

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ABSTRACT

This work deals with a detailed study of the soot removal from inside a syngas environment. Li-delafossite based catalysts were prepared via a solution combustion synthesis (SCS) method and investigated under realistic conditions as catalysts for the gasification of particulate matter retained in a soot trap downstream from a biogas autothermal reformer (ATR). The $LiFeO_2$ and $LiCoO_2$ catalyst exhibited the highest activity toward the catalytic gasification of soot in a synthesis gas mixture.

1. Introduction

To develop a robust and efficient biogas reformer aimed at covering a wide span of potential applications, from fuel cells feed up to the production of pure, PEM-grade hydrogen, the problem of carbon formation needs to be solved. To avoid particulate emissions leading to environmental problems as well as the need to prevent an undesirable deposition on the catalysts of the fuel cell, contributing a fast degradation of the component, a wall-flow soot trap has been provided downstream the reformer to retain most of the soot produced by the reactor. This work is being performed within the SP1-JTI-FCH.2-Collaborative Project 'BIOROBUR'.

The syngas produced by ATR reforming is constituted by carbon dioxide (CO_2), carbon monoxide (CO), hydrogen (H_2) and steam (H_2O). The possibility of gassifing the soot in this specific environment can be reached according to the following reactions for the carbon:

$$C + H_2O \rightarrow CO + H_2 \qquad \Delta H^{\circ}_{298} = 131.4 \text{ kJ/mol} \qquad (1)$$

$$C + CO_2 \rightarrow 2CO \qquad \Delta H^{\circ}_{298} = 172 \text{ kJ/mol} \qquad (2)$$

2. Experimental Set-Up

Li-delafossite based catalysts were prepared via a SCS method and were fully characherized (X-ray diffraction, BET and TPD analyses). The activity of the catalysts towards soot gasification was analyzed by temperature programmed reaction (TPRe), which was carried out in a fixed-bed micro-reactor. A flow of 100 ml/min of CO_2 (10,92%), CO (10,6%), H_2 (26,83%), H_2O (24,87%) and N_2 (26,76%) was sent to a fixed bed of 50 mg of a mixture of carbon (Printex U) and powdered catalyst (ratio 1:9 on a mass basis) with 150 mg of inert silica (to reduce the specific pressure drop and to prevent thermal runaways). The catalyst/carbon/SiO_2 mixture was inserted in the reactor and confined between two quartz-wool layers. The reactor was placed in a PID-regulated oven, and a K-type thermocouple was put in contact with the packed bed to know exactly the temperature during the reaction. A Gas hourly space velocity (GHSV) of 20.000 1/h and a heating rate of 5 °C/min until a set-point of 850 °C were used for the tests. Moreover, soot–CO_2 gasification reaction were studied by thermogravimetric analysis (TGA) on a Mettler Toledo TGA/SDTA 851e system within a temperature range of 25°C-850°C at atmospheric condition. The investigation was realized using a heating rate of 5 °C/min and a flow of 50 ml/min of CO_2 (10,6%) and Argon.

2.1. Catalytic monolith preparation and characterization

$LiFeO_2$ and $LiCoO_2$ catalysts that showed the best activity among the prepared catalysts were deposited by the *in situ* SCS method directly over the wall-flow filter, in a specific amount of 10 wt% with respect to filter weight. The support selected was a silicon carbide (SiC) filter produced by CTI company (pore size: 15 µm and 20 µm, porosity of channel walls = 46%). The morphology of the deposited catalyst layer was analysed by FESEM observation (Figure 1).

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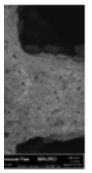

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f soot by syngas in TPRe runs,
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are represented in Figure 2. To
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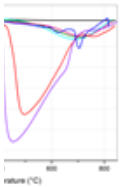

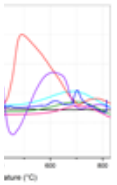

% Vol CO (Up-right), % Vol
eft) Vs Temperature (°C).

ıtures (600-850°C) for all the
more, to ensure that the reverse
es, thermogravimetric analysis
desorption (TPD) studies were
nd 850 °C. The results showed
oxygen species on the catalyst.
ce a high quantity of CO_2, CH_4
ethanation and water gas shift
orbed and influences the proper
fter the gasification reaction, a
to oxidize the amount of soot
promising, showing relatively
ance to gasification of carbon.
$LiFeO_2$ were used as catalysts
50°C. The same enocouraging
catalysts were almost retained.