Journal of Cleaner Production 162 (2017) 7–15

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Comparative life cycle assessment of Ni-based catalyst synthesis processes

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A R T I C L E I N F O

Article history:
Received 8 November 2016
Received in revised form
24 May 2017
Accepted 3 June 2017
Available online 3 June 2017

Handling Editor: Yutao Wang

Keywords:
Novel synthesis process
Life cycle assessment
Ni-based catalyst

A B S T R A C T

Ni-based catalysts supported on ceramics are particularly suitable for industrial applications, for instance reforming of hydrocarbons to produce synthesis gas or hydrogen and production of carbon nanofibers. Conventional synthesis processes for all metal/ceramic catalysts are impregnation, precipitation, co-precipitation and others. The authors have previously developed a novel process for the synthesis of Ni-based catalysts supported on reticulated ceramic foams, including impregnation of foams with ultrasonically generated aerosols of dissolved metal chlorides. By using appropriate multi-criteria analysis methods, the authors concluded that the novel process for the synthesis of Ni-based catalysts was superior in terms of economic and technological aspects. The aim of this research was to compare the novel synthesis processes for a Ni-Pd/Al_2O_3 catalyst and for other Ni-based catalysts by performing life cycle assessment and evaluating the environmental impacts of each synthesis process. Characterisation results showed that the dominant environmental impact results from production of palladium (II) chloride for the Ni-Pd/Al_2O_3 catalyst synthesis process, while the other catalyst synthesis process had large environmental impacts associated with high energy consumption. The final outcome, obtained from comparison of normalisation results, indicates that the novel Ni-Pd/Al_2O_3 catalyst synthesis process had the smallest environmental impact.

1. Introduction

Catalysts based on metals supported on ceramic materials are used in a wide range of heterogeneous catalytic processes. Nickel-based catalysts are mostly used for the production of highly efficient energy sources such as H_2 and synthesis gas ($CO + H_2$) (Calles et al., 2015; Tu et al., 2011) or advanced carbon materials (de Llobet et al., 2015; Takenaka et al., 2003). The Ni/Al_2O_3 system is the most common among those catalysts (Akande et al., 2005; Zhang et al., 2010) and suitable for dry methane reforming (Tu et al., 2011), oxidative methane reforming (Omata et al., 2008), ethanol steam reforming (Calles et al., 2015), production of carbon nanofibers (de Llobet et al., 2015; Takenaka et al., 2003), etc. Catalysts for the hydrocarbon reforming processes are usually modified by a small amount of a noble metal, for example 0.1 wt% of Pd, which prevents nickel deactivation (Fatsikostas et al., 2002; Profeti et al., 2009).

Conventional thermochemical processes for catalyst synthesis include impregnation, precipitation, co-precipitation, sol-gel and others. In order to obtain initial precursors for catalysts, those processes require suspending of Al_2O_3 or other oxide powder in aqueous solution of metal salts, preparation of mixed salt solutions or forming of gels from compounds that contain metallic ions (Akande et al., 2005; Yurdakul et al., 2016). Initial precursors are calcined at high temperatures (from 773 K (Sengupta et al., 2014) to 823 K (Sharifi et al., 2014)) until oxide precursor mixtures are synthesised. Reduction is a final step to obtain catalysts where only active particles are transferred to metallic state while support remains as an oxide. It is also performed at high temperatures, most commonly from 823 K (Sengupta et al., 2014) to 973 K (Sharifi et al., 2014). In some cases, the calcination step is avoided and

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http://dx.doi.org/10.1016/j.jclepro.2017.06.012
0959-6526/

catalytically active Pd and Ni can be obtained by low-temperature reduction of metal salts (at 573 K for 1 h) (Takenaka et al., 2003).

In a previous research, Nikolić et al. (2014b) have developed a novel, unconventional synthesis process to prepare a monolithic Ni-Pd/Al_2O_3 catalyst, where reticulated α-Al_2O_3 based foam was impregnated with ultrasonically generated aerosol of dissolved metal chlorides. The foam was produced according to a previously described method (Nikolić et al., 2014a). The developed catalyst synthesis process enabled technological simplification and energy savings (Nikolić et al., 2014b) because the calcination procedure was eliminated and the catalyst was reduced by hydrogen at a very low temperature, 533 K (Nikolić et al., 2014b). In an earlier work of these authors, the synthesis processes of Ni-Pd/Al_2O_3 catalyst and other Ni-based catalysts were evaluated by using three multi-criteria analysis methods (SAW, TOPSIS and PROMETHEE II). Research results indicated that the novel Ni-Pd/Al_2O_3 catalyst synthesis process ranked best in comparison with other processes (Nikolić et al., 2016).

Catalyst synthesis processes lead to formation of wastes that mainly comprise flue gases, liquid waste and a minor amount of solid waste. Environmental impacts are highly dependent on a synthesis process and starting materials. Most conventional thermochemical processes for catalyst synthesis require the preparation of a starting solution with a low concentration of dissolved chemicals. As an example, Abdedayem et al. (2015) have synthesised Cu/AC catalysts supported on activated carbon by impregnation process; the starting $Cu(NO_3)_2$ solution had a concentration of 0.001 mol L^{-1}. Jung et al. (2012) have prepared Ni/Al_2O_3 catalysts by using co-precipitation process; the concentration of $Ni(NO_3)_2$ and $Al(NO_3)_3$ in the starting solution was 0.085 mol L^{-1} and 0.098 mol L^{-1}, respectively. Therefore, the synthesis of 1 kg of catalyst requires a significant amount of starting solution, which usually becomes waste after use. Oxidative calcination of catalyst supports impregnated with metal salts, such as nitrates, leads to emission of NO_2 gas. Waste acid solutions are generated when mixtures containing metal nitrates and/or chlorides are reduced by hydrogen instead of calcined.

Life cycle assessment (LCA) is often used for comprehensive evaluation of process environmental impacts through the life cycle (Agarski et al., 2016; Curran and Young, 2014; Levasseura et al., 2016). To date, life cycle assessments of catalyst synthesis processes are scarce. Snowden-Swan et al. (2016) evaluated the greenhouse gas emissions of catalysts for hydrotreating of fast pyrolysis bio-oil with LCA. Yaseneva et al. (2014) performed a cradle-to-gate LCA study to compare potential water treatment processes based on two carbon nanofiber supported catalyst types. The environmental burden associated with the synthesis process of fine chemicals via TiO_2 (solar) photocatalysis was analysed with LCA and compared with the same reactions under thermal conditions by Ravelli et al. (2010). Fernandez et al. (2016) examined enzyme-catalysed processes for biodiesel production and effects on the environment with LCA. Comparative LCA of pharmaceutical wastewater treatment with heterogeneous catalysts was performed by Rodríguez et al. (2016). Considering the fact that catalysts in general can have considerable different properties and applications, in this research, comparative LCA of Ni-based catalyst synthesis processes are performed.

Based on previous studies on catalyst synthesis processes, the authors concluded that none of the research compared Ni-based catalyst synthesis processes by using LCA. In this study, comparative LCA was performed for five Ni-based catalyst synthesis processes. The novel process for synthesis of Ni-Pd/Al_2O_3 catalyst, developed by the authors, is compared with the other Ni-based catalyst synthesis processes; the environmental impacts of each process were evaluated and discussed.

2. Materials and methods

2.1. Goal and scope definition

The goal of this assessment is to perform comparative attributional LCA of five Ni-based catalyst synthesis processes and to assess their impacts on the environment. System boundaries include extraction of raw material from the environment, production and transport of semi-products and production of Ni-based catalysts (Figs. 1 and 2). Use phase and waste management of end

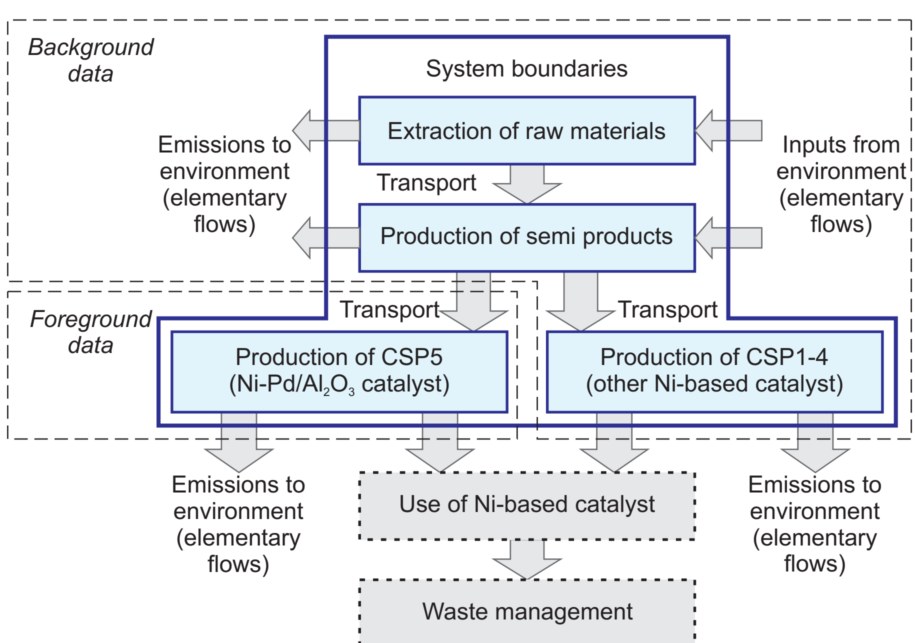

Fig. 1. System boundaries, foreground and background data for life cycles of CSP1-5.