ARTICLE IN PRESS

INTERNATIONAL JOURNAL OF HYDROGEN ENERGY XXX (2017) 1–15

Available online at www.sciencedirect.com

ScienceDirect

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

Pellet size dependent steam reforming of polyethylene terephthalate waste for hydrogen production over Ni/La promoted Al_2O_3 catalyst

Bahador Nabgan [a], Tuan Amran Tuan Abdullah [a,b,*], Muhammad Tahir [b,c,1,1], Walid Nabgan [a,b,d], Sugeng Triwahyono [e], Aishah Abdul Jalil [a,b], Yahya Gambo [a], Maryam Ibrahim [a], Kamal Moghadamian [f]

[a] Centre of Hydrogen Energy, Institute of Future Energy, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

[b] Department of Chemical Engineering, Faculty of Chemical and Energy Engineering, Universiti Teknologi Malaysia (UTM), 81310 Skudai, Johor Bahru, Johor, Malaysia

[c] Chemical Reaction Engineering Group, Faculty of Chemical and Energy Engineering, Universiti Teknologi Malaysia (UTM), 81310 Skudai, Johor Baharu, Johor, Malaysia

[d] Catalysis Research Division, Research Institute of Petroleum Industry (RIPI), Tehran, Iran

[e] Centre for Sustainable Nanomaterials (CSNano), Department of Chemistry, Universiti Teknologi Malaysia (UTM), 81310 Skudai, Johor, Malaysia

[f] Faculty of Chemical Engineering, Islamic Azad University of Mahshahr, Mahshahr, Iran

ARTICLE INFO

Article history:
Received 14 February 2017
Received in revised form
15 July 2017
Accepted 15 July 2017
Available online xxx

Keywords:
H_2 production
Steam reforming
Polyethylene terephthalate
Ni–La/Al_2O_3 support
Pellet size
Catalyst stability

ABSTRACT

The effect of different pellet sizes of nickel (Ni) and lanthanum (La) promoted Al_2O_3 support on the catalytic performance for selective hydrogen production from polyethylene terephthalate (PET) plastic waste via steam reforming process has been investigated. The catalysts were prepared by impregnation method and were characterized using XRD, BET, TPD-CO_2, TPR, SEM, EDX, TEM and TGA. The results showed that Ni–La-co-impregnated Al_2O_3 catalyst has excellent activity for the production of hydrogen. Feed conversion of 88.53% was achieved over 10% Ni/Al_2O_3 catalyst which increased to 95.83% in the case of 10% Ni-5% La/Al_2O_3 catalysts with a H_2 selectivity of 70.44%. The catalyst performance in term of gas production and feed conversion was further investigated under various operating parameters, e.g., feed flow-rate, and catalyst pellet size. It was found that at 0.4 ml/min feed flow rate, highest feed conversion and H_2 selectivity were achieved. The Ni particles, which are the noble-based active species are highly effective, thus offered good hydrogen production in the phenol-PET steam reforming process. Incorporation of La as a promoter in Ni/Al_2O_3 catalyst has significantly increased the catalyst reusability with prolonged stability. The Ni–La/Al_2O_3 catalyst with larger size showed remarkable activity due to the presence of significant temperature gradients inside the pellet compared to

* *Corresponding author*. Centre of Hydrogen Energy, Institute of Future Energy, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia.

E-mail addresses: bnkabi62@yahoo.com (B. Nabgan), tuanamran@utm.my (T.A.T. Abdullah), bttahir@yahoo.com (M. Tahir), walid_nabgan_2008@yahoo.com, wnabgan@gmail.com (W. Nabgan).

[1] Permanent address: Department of Chemical Engineering, COMSATS Institute of Information Technology, Lahore, Punjab, Pakistan.

http://dx.doi.org/10.1016/j.ijhydene.2017.07.103
0360-3199/

Please cite this article in press as: Nabgan B, et al., Pellet size dependent steam reforming of polyethylene terephthalate waste for hydrogen production over Ni/La promoted Al_2O_3 catalyst, International Journal of Hydrogen Energy (2017), http://dx.doi.org/10.1016/j.ijhydene.2017.07.103

smaller size. Additionally, the catalyst showed only slight decrease in H_2 selectivity and feed conversion even after 24 h, although production of carbon nanotubes was evidenced on its surface.

Introduction

Due to the fast decline in fossil fuel reserves and associated environmental problems, there is a critical need for developing alternative sustainable energy resources [1,2]. As a promising candidate, hydrogen as a clean fuel option has received keen attention [3,4]. This clean energy carrier has found diverse applications especially in fuel cell devices. Due to the intrinsic problems of the fossil fuels, the global demand for renewable energy resources including hydrogen has been on the increase profoundly. In recent time, the hydrogen production on industrial scale is via steam reforming of natural gas [5,6], With the great prospects the hydrogen holds as a clean renewable fuel, research efforts are being focused towards its production from other alternatives involving conversion of waste materials such plastics into fuel. Via this approach, the environmental problems posed by plastic waste could be resolved with added advantage [7].

Among different types of plastic, polyethylene (PE) is widely used for numerous purposes, including packing and packaging materials with almost 63 wt% of the total generated plastic waste [8]. Polyethylene terephthalate (PET) is one of the main source of packing materials such as bottles for mineral water and soft drinks all around the world. One of the chemical methods used to recycle PET, is solvolysis, such as methoanolysis and glycolysis [9]. With the great advantage that a portion of PET can be recycled straight to virgin PET [10], or to raw constituents [11] via chemical alteration techniques (such as glycolysis and hydrolysis), yet the main problem of recycling PET is that a considerably big amount returns to waste dumps. Consequently, any novel implementation or artificial use of waste PET would be a momentous relief to the surroundings [12] and this is one of the main emphasis of the current study. Therefore, generation of hydrogen from plastics is indeed a promising technology environmentally and economically [13].

Among the catalysts, alumina (Al_2O_3) is the most commonly employed support in various thermal processes because of its chemical and mechanical stability, low cost and high surface area for metal dispersion. Although comparative studies [14] have shown better performance on basic supports, such as MgO and La_2O_3. The La_2O_3 and ZrO_2 as support materials have displayed a great vital catalyst-support interaction [15,16]. Bimetallic catalysts such as nickel and cobalt on Al_2O_3, ZrO_2 and La_2O_3 have been used for steam reforming [17] which are able to suppress the formation of coke. In addition to Al_2O_3, La_2O_3 has displayed prolong ability as reported by many researchers [18–20]. Lanthanum oxide could considerably improve chemical activities of highly distributed metal catalysts [21] due to its production of medium and strong basic sites, which could favor OH surface mobility and steam adsorption and then favor steam reforming [22]. In previous studies, lanthanum has been used as a promoter [23,24] or/and as a support [25,26]. Recently, it has been reported that lanthanum effectively enhanced the physicochemical properties and catalytic performance of Ni–Al_2O_3 aerogel catalyst compared to other second metals [27]. Therefore, a systematic investigation on finding optimum lanthanum addition to Ni–Al_2O_3 catalyst for hydrogen production by steam reforming of phenol-PET would be worthwhile. Ni is one of the most used metal in steam reforming processes due to its superb ability in facilitating cleavage of C–C, C–H, C–O bonds and dehydrogenation reactions [33].

On the other hand, the efficient use of heterogeneous catalyst surface to enhance reactions is limited by the rates of mass transfer inside the pellet pores [28]. This limitation affects reaction rates, due to differences between reactant and product phases (liquid or gas) compared with catalyst phase (solid), leading to slow reaction rates on the surface of catalyst and high product concentrations inside the porous pellets [29]. Consequently, to estimate the mass transfer limitations in different processes, significant studies have been carried out base on difference in pellet size [30,31]. However, a study of similar problem with regard to the influence of different pellet size of Ni–La/Al_2O_3, to the best of our knowledge, has not been carried so far. Therefore, it is of great importance to apply different pellet size of Ni/La co-modified Al_2O_3 composite for hydrogen production from PET plastic waste.

Steam reforming of phenol-PET solution for hydrogen production using Ni/Al_2O_3 and NiPd/Al_2O_3–La_2O_3 catalyst have been discussed in our previous studies [32,33]. Whereas, the present research aimed to explore catalytic PET conversion via steam reforming over different pellet sizes of Ni/La-based Al_2O_3 catalysts for selective hydrogen production in a fixed bed reactor. This provides further insight into ways of renewable and clean fuels production from waste feedstock. The catalysts were characterized using BET, XRD, SEM, TEM, EDX, TPD-CO_2, TPR and TGA. The effects of different operating parameters such as feed flow rate and pellet size of the catalyst on H_2 selectivity and conversion were critically discussed. The stability analysis of different pellet sizes of the Ni–La promoted Al_2O_3 catalyst was deliberated in order to reveal the function of the metals and support on catalyst activity.

Experimental

Catalyst preparation

Impregnation method was used for preparing Ni supported on γ-Al_2O_3. In order to prepare the catalyst, 10 wt % of active metal