

Contents lists available at [ScienceDirect](http://www.sciencedirect.com)

South African Journal of Chemical Engineering

journal homepage: <http://www.journals.elsevier.com/south-african-journal-of-chemical-engineering>

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Thermodynamic investigation of waste cooking oil based hydrogen generation system with chemical looping process

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ARTICLE INFO

Article history:

Received 9 October 2015

Received in revised form

17 April 2016

Accepted 18 April 2016

Keywords:

Hydrogen

Waste cooking oil

Chemical looping reforming

Thermodynamic analysis

Thermoneutral

ABSTRACT

The thermodynamic features of hydrogen production via chemical looping reforming of waste cooking oil were studied at atmospheric pressure using Gibbs free energy minimization, accounting for the possibility of coke formation. The synthesis gas composition was determined as a function of reforming temperature (400–1000 °C), steam to carbon ratio (S/C, 1–14) and NiO to carbon ratio (NiO/C, 0–2). To evaluate the thermodynamically plausible products, the study started with an expanded product set of approximately 30 by-products.

The results show that coke formation can be thermodynamically inhibited by increasing the S/C ratio and/or the NiO/C ratio. The conditions that maximize hydrogen production, minimize methane and carbon monoxide content as well as avoid coke formation at thermoneutral conditions were found to be S/C = 5, T = 600 °C and NiO/C = 0.493. Under these conditions, a hydrogen yield of 144.3 mol/kg of soybean waste cooking oil can be obtained, which appears to be an attractive result for starting experimental research.

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1. Introduction

Increasing energy demand coupled with the depletion of fossil fuel resources and increasing environmental pollution has stimulated increasing interest in using H₂ as a clean fuel. In fact, H₂ has been proposed as a potential energy source due to its abundance, cleanliness and high energy yield. Unfortunately, the majority of H₂ is produced from fossil fuels; only 4% of H₂ is derived from other renewable sources (Parthasarathy and Narayanan, 2014). Because fossil fuels will be depleted by 2050, it is prudent to search for a sustainable

and eco-friendly source of H₂ generation (Ashkuzzaman and Jiang, 2014). Renewable technologies to produce H₂ for fuel cell applications not only safeguard the environment but also provide a sustainable source of H₂.

Among the various H₂ production methods, chemical looping reforming (CLR) seems to be a promising and environmentally friendly alternative (de Diego et al., 2009). In fact, CLR has been considered an alternative to catalytic autothermal steam reforming (de Diego et al., 2009; Ortiz et al., 2011). The main advantage of the CLR route is that the oxygen carrier (OC) is an oxide (usually a metal oxide (MeO)) (Pröll

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<http://dx.doi.org/10.1016/j.sajce.2016.04.001>

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et al., 2010; Wang, 2014). As a result, air may be used instead of pure O₂, and N₂ is never mixed with H₂. A CLR system consists of two interconnected reactors, designated the air (AR) and fuel reactors (FR) (Dueso et al., 2012). In the FR, fuel and steam are burned with an OC to form a synthesis gas (SG) (mixture of CO, H₂, CO₂, CH₄, H₂O, etc.) while OC particles are reduced to a metal (Me). The reduced metal is transferred into the AR, where it is oxidized with air. The regenerated material is then ready to start a new cycle. The major advantage of this process is that the heat needed for converting fuel to H₂ can be supplied without costly O₂ production, without mixing air with carbon containing fuel gases and without using part of the H₂ produced in the process (de Diego et al., 2009). A N₂-free gas stream containing concentrated H₂ and CO is obtained from the reformer, avoiding dilution of the H₂ with N₂. If the H₂ produced is to be used in fuel cells, it should be noted that the dilution of the H₂ stream by N₂ results in an increased anode overpotential during the operation of a proton exchange membrane fuel cell (PEMFC) (da Silva et al., 2012). H₂ production via CLR of various fuels is currently being investigated intensively, and several papers on this subject have already been published. Moldenhauer and co-workers (Moldenhauer et al., 2012) performed a reaction between a nickel-based OC and liquid kerosene in a CLR reactor with continuous particle circulation. An injection system was constructed in which sulfur-free kerosene was evaporated, mixed with superheated steam and fed directly into a lab scale CLR reactor. Moldenhauer et al. showed that it is possible to use liquid fuel in a continuous CLR process and to achieve nearly complete fuel conversion. Kai et al. (Kai et al., 2012) implemented a process of separating H₂ from SG through the chemical looping of a Fe-based catalyst as an oxygen-transfer material and a modified calcium oxide (CaO) as a CO₂ sorbent in a fixed-bed reactor. Kai et al. achieved a hydrogen purity above 99.5% and a yield approaching 27.91 mmol/g Fe catalyst.

Due to the emergence of various food industries, restaurants all over the world, the amount of waste cooking oil (WCO) required for disposal is a major concern. Currently, about 29 million tons of WCO are generated annually in the world (Maddikeri et al., 2012). The base materials of WCO are plant-based lipids, such as soybean oil, corn oil, palm oil, or animal-fats. Chemically, the major components of WCO are triglycerides (tristearin, trimyristin, tripalmitin, tripalmitoleic, trioleate, etc.), with minor amounts of mono and diglycerides (Dale et al., 2008). Until recently, WCO was a significant environmental problem, and the management of this waste was a significant challenge. However, WCO cannot be discharged into sewers as its discharge will lead to blockages, odor or vermin problems and may also pollute watercourses, causing problems for wildlife (Lee et al., 2012). In this context, many scientists have intensively investigated WCO valorization in recent years. The biodiesel production via WCO transesterification remains by far the main route of WCO reuse (Al-Hamamre and Yamin, 2014; Mohammad et al., 2014; Chen et al., 2009). Recently, Amani and co-workers (Amani et al., 2014) investigated the transesterification of waste cooking palm oil with methanol into fatty acid methyl esters (FAMES) using solid acidic mixed oxide catalysts Mn_{3.5x}Zr_{0.5y}Al_xO₃ prepared via coprecipitation. The authors showed that the catalyst achieved a FAME content of more than 93%, and the optimal reaction conditions were as follows: reaction temperature of 150 °C, reaction time of 5 h, molar methanol-to-WCPO ratio of 14:1, and catalyst loading

of 2.5 wt.%. Hamze et al. (Hamze et al., 2015) studied the transesterification of the WCO with response surface methodology (RSM) based on Box–Behnken design. The results revealed that the catalyst concentration is the most important parameter and the maximum biodiesel yield under the optimized conditions was 99.38 wt.%. It should be noted, here, that even with all the advantages, there are still some disadvantages associated with the use of biodiesel in combustion engine. Biodiesel has a 12% lower energy content than fossil diesel, which leads to an increase in fuel consumption of approximately 2–10% (Atabani et al., 2012). Moreover, biodiesel has higher cloud and pour points as well as higher nitrogen oxide emissions than fossil diesel. Biodiesel also has lower volatilities, which lead to soot formation in engines due to incomplete combustion (Atabani et al., 2012). In the past few years, some researchers have turned their interest toward fatty material-based H₂. Converting WCO into H₂ is a three-win alternative, simultaneously addressing pollution, food security, and energy security. Various processes have been proven theoretically and experimentally by many research groups. Pimenidou et al. (Pimenidou et al., 2010a) used a CLR process to produce H₂ from WCO using a nickel-based OC. High purity H₂ was produced by adding calcined dolomite as a CO₂ sorbent into the reactor catalytic bed (Pimenidou et al., 2010b). Dupont et al. (Dupont et al., 2007) studied a novel process of H₂ production called unmixed steam reforming (USR) using methane and sunflower oil. Dupont et al. showed that both methane and sunflower oil are suitable fuels for the USR process and that the thermal decomposition of the fuel played a significant role in early H₂ production concurrent with coking conditions.

This paper explores an innovative application of soybean WCO (SWCO) for H₂ production via a CLR process. We believe that SWCO is a promising feedstock for renewable H₂ production because of SWCO's low O₂ content and high potential yield of H₂. Moreover, SWCO is a potential alternative for H₂ production due to its highly centralized generation in restaurants, various eating outlets and food industries as well as its historically low prices. SWCO may therefore have energy, environmental, and economic advantages that could be exploited. H₂ obtained from SWCO has been proposed to be a low-risk end use for SWCO derived from livestock that have been removed from the food chain. Because of the complex chemical structure of SWCO, the unavailability of physical and chemical properties and the multitude of chemical reactions that can occur, no thermodynamic investigation of H₂ production by CLR of SWCO has been considered in the past. Generally, an investigation of thermodynamic equilibrium is an important tool preceding experimental work. This investigation identifies thermodynamically favorable process operating conditions and predicts the equilibrium product composition. In addition, this investigation is an aid in reactor modeling, in examining kinetic schemes and reaction mechanisms, and in identifying rate-controlling processes (Shi et al., 2001). This paper aims to identify thermodynamically favorable operating conditions at which SWCO may be converted to H₂ by CLR. An expanded product set is used to examine the plausible appearance of the various species in the CLR system. Coke deposits are also investigated to determine coke forming and coke-free regions. It should be noted that the thermodynamic equilibrium investigation conducted here did not consider any kinetic constraints such as

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