



Kinetics and deactivation of a dual-site heterogeneous oxide catalyst during the transesterification of crude jatropha oil with methanol

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Abstract

In this work, a dual-site heterogeneous catalyst was used to study the kinetics of the transesterification of crude jatropha oil with methanol in batch experiments. Experimental data were obtained between 150 and 182 °C at a constant molar ratio of alcohol to oil (11:1) and at a set catalyst concentration (3.32 wt% based on weight of oil). Kinetic modelling was performed with an assumed pseudo-first order to describe the rate and catalyst deactivation, which suitably fit the experimental data. The model parameters were determined for both the mass transfer controlled and reaction regimes. The triglyceride (TG) conversion, methyl ester (ME) formation, and system thermodynamics were also evaluated. Based on the calculated values, within an acceptable range, the equilibrium constant, $K_e = 1.42$; activation energy, $E_a = 161$ kJ/mol; and free energy, $\Delta G = -1286$ J/mol indicate that the developed model adequately described the methanolysis of the oil, which may be useful in reactor design and process simulation. The values that were calculated from the kinetic equations agree well with the experimental values, and the results help to understand and predict the behaviour of dual-site catalysts in practical applications for the production of biodiesel.

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1. Introduction

In recent years, the synthesis of fatty acid methyl esters (FAME) has become important, as research has intensified on the utilization of vegetable oils and animal fat derivatives for liquid fuel (biodiesel) production

[1–5]. FAME can be obtained from renewable feedstock, such as vegetable oils or animal fats, by the process of transesterification with alcohols in the presence of an acid or an alkaline catalyst. Biodiesel is a clean-burning fuel that is nontoxic and biodegradable and is considered the fuel of the future [6]. Biodiesel is referred to as neat fuel (B100) in its pure form or as the blended form (B20 or B80) when mixed with petroleum diesel. Biodiesel can be utilized in compression ignition engines under a variety of operating conditions. B100 contains no petroleum fuels and emits virtually no sulphur, aromatics, particulates, or carcinogenic compounds; thus it is a safer alternative to petroleum diesel. Biodiesel can be used in all conventional diesel engines, and its

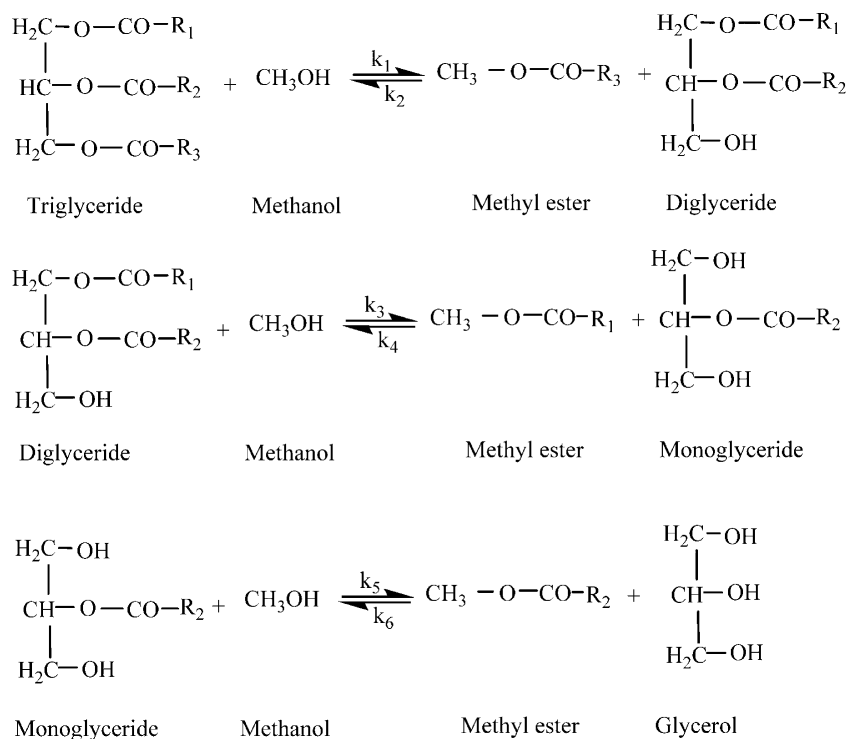
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Scheme 1. 3-Step reversible reactions of triglyceride, where R_1 , R_2 , and R_3 are long chains of carbon and hydrogen atoms (fatty acid chains).

performance and engine durability closely match those of petroleum diesel, requiring no modifications in fuel handling and delivery systems [7,8]. Three consecutive and reversible reactions are believed to occur during the transesterification of vegetable oils and fats, converting to intermediate products that are formed during the reaction: diglycerides (DGs), monoglycerides (MGs), and the final methyl ester product. The reaction scheme is shown in Scheme 1.

The various parameters that influence the synthesis of FAME have been investigated by many authors due to the increasing industrial application of methyl esters [7]. These parameters include the molar ratio of alcohol to vegetable oil, catalyst type and loading (wt% of oil), reaction temperature, agitation intensity and presence of impurities, such as free fatty acids and moisture content. There are a number of kinetic studies on the transesterification of esters with alcohol catalyzed by homogeneous base catalysts, but very little information is available on the kinetics of transesterification reactions by solid-base catalysts [9]. In particular, few studies have investigated the transesterification reactions of vegetable oils and FFAs with the application of a dual-site composite catalyst. Hattori et al. [10] proposed a mechanism for the transesterification of ethyl acetate with different alcohols using a variety of solid-base catalysts, particularly alkaline-earth-metal oxides. However, this mechanistic

study provided neither values of activation energies nor rate constants. Recently, a kinetic model was proposed to describe the transesterification of ethyl acetate with methanol catalyzed by a heterogeneous magnesium oxide catalyst [11,12]. This model was based on a three-step ‘Eley–Rideal’ type of mechanism applied in liquid phase, where methanol adsorption is assumed to be rate determining. The activation energy was 20 kJ/mol, with a methanol adsorption equilibrium coefficient of $3.13 \times 10^{-3} \text{ m}^3/\text{mol}$. The objective of this study was to evaluate the use of a dual-site heterogeneously catalyzed transesterification reaction in batch reactors under pre-determined conditions using the kinetic model based on the three-step ‘Eley–Rideal’ type mechanism assuming methanol adsorption as a rate-determining step. The kinetics and deactivation studies of a dual-site catalyst in the transesterification of CJO (non-edible stock) are important for determining reactor design parameters and advancing the stage of efficient biodiesel production.

In addition, a kinetic model for the reaction of triglyceride over solid catalyst was developed, and the deactivation characteristic during the transesterification and esterification reactions with methanol was aimed to be observed. The influence of various reaction parameters on the catalyst activity and kinetic rate were evaluated. Because the kinetics of transesterification and esterification reactions are increased using basic and

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