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Aqueous-phase Fischer-Tropsch synthesis over rhodium catalysts for simultaneous synthesis and separation of hydrocarbons and oxygenates

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PURPOSE OF THE ABSTRACT

Direct syntheses of C2+-oxygenates such as alcohols, aldehydes and organic acids from syngas generated from renewable sources (biomass, organic waste) have been extensively developed recent years in order to design sustainable alternatives to the relevant petroleum-based products. However, the selective catalytic conversion of syngas to oxygenates remains challenging, and no commercial process exists today although extensive efforts have been made [1]. So far, rhodium-based catalyst exhibits the most practical application due to its good selectivity for the synthesis of ethanol and other oxygenates from carbon monoxide and hydrogen [2].

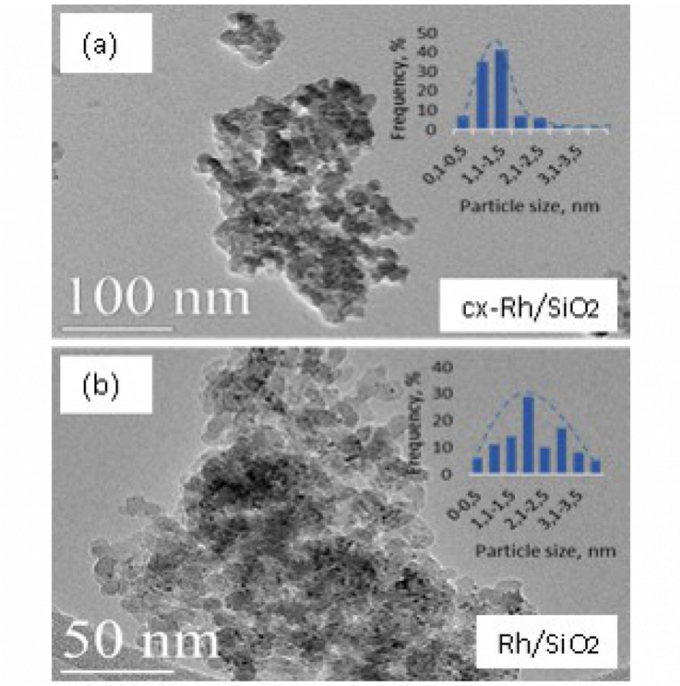
The present work focuses on the design of highly dispersed rhodium catalysts using organometallic complexes for direct synthesis of hydrocarbons and oxygenates from syngas. To the best of our knowledge, aqueous-phase Fischer-Tropsch synthesis with rhodium catalysts has not been reported to date. In addition, carrying out the reaction in a batch reactor allows selective separation of hydrocarbons and oxygenates into two phases. Light hydrocarbons are collected in the gaseous phase, while oxygenates are present in the liquid aqueous phase.

The Rh-based catalyst were prepared by incipient wetness impregnation of commercial silica with an aqueous solution of rhodium precursor to achieve a metal weight loading of 10%. The use the rhodium Schiff base complex as a precursor for preparation Fischer-Tropsch catalyst allows to obtain highly dispersion of metal on the support and to control the particle size distribution. Comparing with the catalyst - Rh/SiO2, prepared from traditional precursor (rhodium chloride), the catalyst - cx-Rh/SiO2 based on complex has a much narrower particle size distribution. It contained only ruthenium nanoparticles with sizes from 1.5 to 2.5 nm (Figure 1).

The catalytic performance of the rhodium-based catalysts was tested in aqueous-phase Fischer-Tropsch synthesis. The products of syngas conversion over the rhodium catalysts were separated in situ in two phases. The gas phase contained methane, C2-C8 light hydrocarbons and carbon dioxide and the aqueous phase contained dissolved oxygenates. Most of oxygenates are C1-C4 alcohols. The highest selectivity to oxygenates of around 30% was found on the catalyst based on rhodium Schiff base complex,

The aqueous-phase fischer-Tropsch synthesis demonstrated its high efficiency in the simultaneous synthesis and separation of hydrocarbons and oxygenates. In addition, the use organometallic Schiff base complexes as catalyst precursor provides excellent opportunities for the design of catalysts for Fischer-Tropsch synthesis with high selectivity to C2-oxygenates.

FIGURES



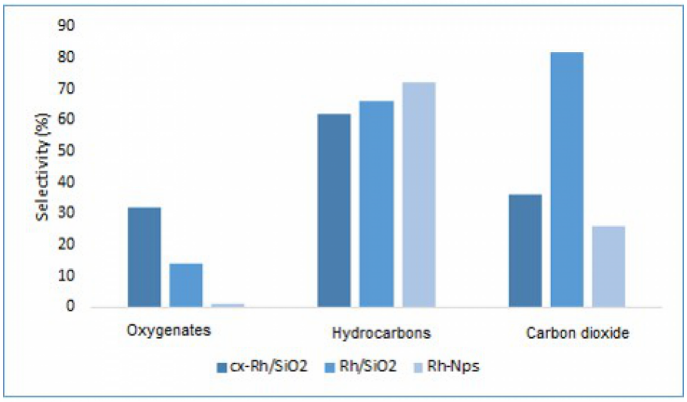


FIGURE 1

Figure 1

TEM images of cx-Rh/SiO2 (a) and Rh/SiO2 (b) catalysts

FIGURE 2

Figure 2

Product selectivity over Rh catalysts (reaction conditions: conversion=70-80%, T=200 o C, P=30 bar and H2/CO=2)

KEYWORDS

syngas | oxygenates | rhodium nanopartciles | process combination

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