COMPARISON OF TWO PREPARATION METHODS OF SUPPORTED Li-Ni CATALYST ON ALUMINA FOR SELECTIVE OXIDATION OF METHANE

Mitsunobu Ito, Tomohiko Tagawa and Shigeo Goto*
Department of Chemical Engineering, Nagoya University,
Chikusa, Nagoya 464-8603, Japan

Received March 23, 1999
Accepted July 20, 1999

Abstract

Lithium-added nickel catalysts on alumina were prepared for CO₂ reforming of methane by two methods; precipitation and impregnation. Performances of the catalysts were investigated by TG, CO-adsorption and SEM analysis. The catalyst with ratio of Li/Ni=1.0 prepared by precipitation method has high nickel dispersion, catalytic activity and stability for CO₂ reforming of methane.

Keywords: Carbon dioxide reforming, nickel catalyst, precipitation, impregnation

INTRODUCTION

Carbon dioxide reforming of methane is a particularly efficient process for producing synthesis gas with low H₂/CO ratio which is efficient for producing oxygenates such as alcohol or aldehyde [1].

*Corresponding author. Tel. & Fax. : (+8152)789-3261.
E-mail : goto@park.nuce.nagoya-u.ac.jp
Several supported transition metal catalysts were used for these reactions [2]. Nickel based catalysts were mainly used in industry due to their low cost. However, carbonaceous deposition occurred easily followed by catalyst deactivation [3]. Alkaline metals were reported to be effective as co-catalysts, suppressing carbonaceous deposition [4] and improving C2 selectivity in oxidative coupling of methane [5].

We reported the reaction mechanism and performance of several industrial nickel catalysts for carbon dioxide reforming [6, 7]. Nickel catalyst was modified by addition of alkaline metal with impregnation method and lithium added catalysts showed high activity and stability [8].

Precipitation is another preparation method. Therefore, in this study, two methods to prepare the lithium-added nickel catalyst on alumina, viz. impregnation and precipitation are compared for CO2 reforming of methane. Thermogravimetric (TG) analysis, nickel surface measurement (CO-adsorption method) and SEM analysis are conducted to clarify the nature of these catalysts.

EXPERIMENTAL

Catalyst preparation

Impregnation method

Catalysts were prepared by impregnating nickel and lithium on alumina (20 to 32 mesh size) with the aqueous solution of nickel and lithium nitrate. These catalysts were dried in the oven at 383 K and were calcined with air flow at 603 K for 5 h. Nickel contents were 30 wt.%. The mole ratios of Li/Ni were 0, 0.2, 0.5 and 1.0.

Precipitation method

Nickel and lithium were precipitated on the alumina (20 to 32 mesh size) by titrating an aqueous solution of sodium carbonate into that of nickel and lithium nitrate at 353K. The catalysts were separated from the solution by filtration and were washed with hot water to remove excess salts. They were dried overnight in a oven at 383 K and were calcined with airflow at 603 K for 5 h. Nickel contents were 30 wt.%. The mole ratios of Li/Ni were 0, 0.333, 0.5 and 1.0.