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Continuous synthesis of molybdenum oxide microspheres by ultrasonic spray pyrolysis

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ABSTRACT

Molybdenum oxides microspheres with prominent MoO_2 phase were produced successfully by ultrasonic spray pyrolysis under reducing conditions. Ethylene glycol addition in the precursor solution was found to promote complete truncation of regular molybdenum oxides particle geometry into rectified spherical micro particles during spray pyrolysis. It also removed the MoO_3 and Mo_4O_{11} phases, while maintaining nanocrystalline MoO_2 phase structure. However, addition of EG reduced the surface area and increased the pore size rather significantly, which might adversely affect the catalytic performance of MoO_2 microspheres. Catalytic activity of the product molybdenum oxides was investigated for partial oxidation of *n*-dodecane.

Introduction

The road map on the use of hydrogen (H_2) to propel hydrogen economy in Japan was announced in 2014 [1]. Following this announcement, Tokyo has earmarked $350 million to subsidize and invest H_2 fuel cell cars and infrastructure such as H_2 fueling stations. The investment also includes 6000-unit Olympic village that functions exclusively on fuel cell power to initiate a transition to hydrogen powered society [2]. Although H_2 has been used as fuel to power rocket for decades, its application in commercial vehicles has not been materialized due to problem of storage and other issues related to H_2 production. There are many ways to produce H_2, among them include electrolysis, thermolysis, biomass gasification, bio-catalysis or fermentative hydrogen production. However, the most economically feasible technology for producing H_2 is still from fossil sources such as in a steam reforming of hydrocarbons and partial oxidation of paraffin.

In the partial oxidation of a paraffin namely *n*-dodecane to produce H_2, molybdenum oxide has been chosen as an active catalyst. Molybdenum oxide has also been extensively used in an area such as hydrocarbon decomposition [3,4] and it has been used as anode materials in fuel cells and lithium-ion batteries [5] because of its high catalytic activity, sulfur tolerance and high electrical conductivity [5,6]. Despite these advantages, commercial molybdenum oxide exhibits low surface area of 0.29 m^2/g. High active surface area is needed in order to achieve high catalytic performance. Higher surface area of 48 m^2/g was successfully achieved previously by a solvothermal method [7]. Attempts to improve the surface area in order to increase the H_2 yield to a higher level is expected to continue in the future.

Besides high surface area, there is also a need to synthesize molybdenum oxide that exhibits high purity, uniformity, strength, and chemical durability to produce hydrogen at extreme conditions. In order to achieve this quality, molybdenum oxide that consists predominately of MoO_2 phase should be obtained. MoO_2 is industrially produced from reduction of MoO_3 by H_2 at temperature between 450 °C and 600 °C [8], and through redox reaction between $Mo_2O_7^{2-}$ and $NH_2C_6H_4NH_2$ at low temperature [9]. Thus, a proper synthesis procedure and system should be made available to prevent the oxidation from taking place and to obtain a stable MoO_2 particle with a proper geometry. MoO_2 particle could exhibit a different geometry such as microsphere [9] and nanostructure [10]. MoO_2 could also be formed into other geometrical shapes such as nanoplate or nanosheets [10] and nanostars [11]. In this study, we synthesized MoO_2 microspheres by continuous ultrasonic spray pyrolysis under reducing conditions. The resulting particles were used as catalysts in partial oxidation of *n*-dodecane.

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