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Effect of loading bimetallic mixture of Ni and Pd on hydrogen storage capacity of MCM-41

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ABSTRACT

MCM-41 was produced by microwave irradiation, which allows high yield, improved product purity, increased reaction rate and crystallization. As transition metals enhance the hydrogen uptake, Pd and Ni were loaded on MCM-41 to increase the hydrogen storage capacity of the structure. The surface areas of the samples were measured by N_2 adsorption and it was observed that they had large surface area around 938–1369 m^2/g . The successful incorporation of metals into the structure was confirmed by characterization using X-ray diffraction and X-ray photoelectron spectroscopy. The hydrogen adsorption capacities of the samples were measured by the Intelligent Gravimetric Analyzer at room temperature and up to 10 bar pressure. The hydrogen storage capacity of MCM-41 was improved by increasing content of bimetallic mixture of Pd and Ni. The maximum hydrogen uptake was obtained as 0.98 wt% with 10:100 Pd–Ni:MCM-41.

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Introduction

Hydrogen is a light weight and environmentally friendly energy carrier that has a high energy density by weight. Although there is an increase in the amount of hydrogen production due to the demand for its potential, the low volumetric density of hydrogen limits its storage. Hence, the development of hydrogen storage technologies becomes crucial for the efficient usage of hydrogen as an energy carrier. Recent studies are mainly focused on materials having large pore size, high surface area and high affinity to hydrogen such as mesoporous and carbon based materials, and metal hydrides [1–3]. MCM-41 (Mobil Crystalline Material) is a member of family of zeolite materials and it is considered to be a good candidate for hydrogen storage as it is a silica based material with ordered hexagonal array of cylindrical pores, high surface area and

affinity to hydrogen [4]. Previously, Weitkamp et al. investigated the suitability of zeolite structures for hydrogen storage and concluded that zeolites can adsorb and desorb hydrogen [5]. The hydrogen storage mechanism of zeolites are strongly dependent on the cations as well as the framework, composition and the acidity-basicity of the structure [3]. The hydrogen storage capacity of zeolites at room temperature and 60 bar pressure was reported as 0.5 wt% [6]. Sheppard and Buckley concluded that MCM-41 had a maximum hydrogen uptake of 1.68–2.01 wt% at 77 K between 30 and 40 bar [7]. The hydrogen storage capacity of MCM-41 is also affected by its ability to stabilize the nanoparticles in the pores. Ramachandran et al. investigated hydrogen storage of metal oxide incorporated Al-MCM-41 systems and their studies indicated that hydrogen spillover was the main mechanism of hydrogen storage in Al-MCM-41 system [8]. MCM-41 production is commonly

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achieved by hydrothermal processing of the reactants in the autoclave. Recently, a good alternative to hydrothermal processing which is microwave synthesis is used for MCM-41 production [9]. Microwave synthesis allows higher yields and better purity and increased rate of reaction and crystallization when compared to the conventionally used hydrothermal method [10]. Jiang et al., has successfully synthesized transition metals loaded MCM-41 by microwave irradiation [11]. There are ongoing researches to better understand the hydrogen storage behavior of transition metal incorporated MCM-41. Prasanth et al. reported that hydrogen sorption capacities of mesoporous materials can be enhanced by in situ transition metal (Ni, Rh, Pd) incorporation to the structure [12]. In this study, the mesoporous materials were synthesized by hydrothermal processing and the metals were loaded during this process. However, the hydrogen storage behavior of MCM-41 synthesized by microwave irradiation has been seldom reported in the literature. Microwave irradiation is an extremely efficient method by means of time and energy in addition to the larger surface area of MCM-41 it provides [11]. Therefore, in this study microwave irradiation was selected for the synthesis of the samples. In order to increase the hydrogen uptake of MCM-41, the transition metals can be loaded to the structure either before or after the microwave irradiation. When the metals are loaded before the microwave irradiation, they fill the pores and it weakens the mesoporous order of the structure [11]. For this reason, we preferred an alternative method and impregnated the metals on the MCM-41 after the microwave irradiation. In this work, it was aimed to observe the effect of binary mixture of two transition metals (Pd and Ni) which are known to have hydrogen affinity on the hydrogen uptake of MCM-41. The effect of metal content on the structural properties and the hydrogen absorption behavior of MCM-41 was investigated.

Materials and method

Materials

An aqueous solution of sodium silicate with 27 wt% SiO₂ (AppliChem Inc.) was used as silica source and hexadecyltrimethyl ammonium bromide (HDTMABr) (98%, ABCR GmbH) was used as surfactant. Pd and Ni loaded MCM-41 was synthesized using Palladium (II) Chloride (99.99%, Aldrich Chem Co.) and Nickel Chloride 6 Hydrate (97.99%, Horasan Kimya), respectively.

Synthesis of MCM-41

6.6 gr of surfactant was slowly dissolved in 43 mL of deionized water at 313 K. 5.5 mL of sodium silicate was added drop wise to the surfactant and stirred until a homogenous mixture was obtained. After the pH of the mixture was adjusted to 11, it was microwave heated at 90 W for 30 min. The resultant product was filtered, washed with distilled water until the pH is neutralized and then solid residue was dried at 333 K for 24 h. The dried powder was then placed in a quartz reactor with quartz filters. The temperature was increased gradually and then the sample was calcined at 823 K for 6 h under the

flow of dry air to obtain MCM-41. After the calcination the system was gradually cooled down to room temperature.

Loading Pd and Ni mixture

The metal salts deposited were impregnated to the dried powder before calcination. Two different weight ratios (5:100, 10:100) of Pd–Ni metal mixture (with a Pd to Ni ratio of 1:1) to 1 gr of MCM-41 was stirred in a solution of 50 mL distilled water at room temperature for 24 h until the mixture was completely homogenized. After the centrifuge, it was dried at 333 K for 24 h and calcined at 823 K for 6 h under the flow of dry air, as described in the previous section.

Characterization

The surface area, pore size and adsorption capacity of MCM-41 were measured by N₂ adsorption using Brunauer–Emmett–Teller (BET) method. The samples were outgassed at 423 K overnight. The measurements were performed at 77 K using liquid nitrogen. The pore volume and distribution were determined with Barrett–Joyner–Halenda (BJH) and density functional theory (DFT) methods. The X-ray diffraction (XRD) analyses were conducted with a Bruker AXS diffractometer fitted with a Siemens x-ray gun using 0.15406 nm Cu K α radiation and the data was analyzed with Bruker AxS Diffrac PLUS software. The analyses were performed with a 2 θ range of 2–90° at 40 kV and 40 mA. The step size was set to 0.01°. The scanning electron microscopy (SEM) images of the samples were collected on Leo G34-Supra35 VP. Jeol 2000FX and JEM-ARM200CFEG UHR were used for transmission electron microscopy (TEM) images. The functional groups on the MCM-41 surface were determined using a ThermoScientific Nicolet iS10 Fourier transform infrared spectrometer (FTIR). The thermal stabilities of the samples were determined by a Netsch449C thermogravimetric analyzer. The samples were heated up to 1273 K with a rate of 10 K/min in nitrogen atmosphere. X-ray photoelectron spectroscopy (XPS) which is one of the most commonly used surface characterization techniques was used in order to examine the surface chemistry of the representative samples before and after hydrogen sorption. The XPS analyses were achieved by Thermo K-alpha X-ray photoelectron spectrometer with a monochromated Al K α supported by a low energy electron/ion flood gun for charge neutralization. The binding energy of the C was taken as 285.0 eV in the XPS measurements.

Hydrogen storage

The hydrogen adsorption capacity measurements were conducted at room temperature (298 K) and up to 10 bar pressure by the Intelligent Gravimetric Analyzer (IGA). The samples were degassed at 473 K and then cooled to room temperature before the hydrogen gas was purged into the system and storage capacities of samples were examined.

Results and discussion

The SEM images of synthesized MCM-41 in Fig. 1 which were obtained by secondary electron detectors show the

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