



Study on hydrogen storage mechanisms of mesoporous materials at low temperature and low pressure



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ABSTRACT

MCM-48, metal doped MCM-48 and MCM-41 have been synthesized by hydrothermal method, respectively. The specific hydrogen storage amount of prepared materials at 77 K below 100 kPa are recorded in order to explore their hydrogen storage mechanisms. Experimental results indicate that pore size is the key factor that dominates their hydrogen storage capacity. Adsorption plays an important role from 0 to 35 kPa and mesoporous materials with smaller pore size seem to be better candidates for hydrogen storage. Diffusion will play a dominant role in the pressure range of 35–100 kPa, and mesoporous materials with larger pore size exhibit higher hydrogen storage capacity.

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1. Introduction

Decreasing supply of fossil fuels and increasing concerns over environmental issues have made hydrogen an ideal alternative to conventional fossil-fuel resources [1]. However, safe storage of hydrogen remains the bottleneck against the widespread application of hydrogen due to its low volumetric density and limited rate of adsorption and desorption [2]. Therefore, great attention has been paid to the quest of efficient, cost-effective and stable medium for hydrogen storage.

At present, most of the research on zeolites for hydrogen storage is mainly focused on microporous materials. And some studies about the hydrogen storage mechanisms have been reported. Chang et al. [3] demonstrated that AIPOs with small pore showed high adsorption capacity due to the increased interaction between AIPOs pore walls and hydrogen. Long et al. [4] stated that very small pores led to high binding affinity due to the increased van der Waals contact area. Chang et al. [5] demonstrated that zeolites with strong electrostatic fields and narrow pores without intersections were beneficial for high hydrogen uptake. Nevertheless, the potential use of mesoporous materials such as MCM-48, MCM-41 in hydrogen storage have not been extensively studied yet. And H₂ adsorption studies in these mesoporous silica are scarce.

Previous studies indicated that hydrogen sorption characteristics of mesoporous materials can be improved by the incorporation of transition metals into mesoporous structures [6], but the specific hydrogen storage mechanisms are still unknown and there are few reports on that study. Thus, further work is required to demonstrate the hydrogen storage mechanisms of mesoporous materials at low temperature and low pressure so as to give further direction to the synthesis of different mesoporous materials with high hydrogen storage capacity. In this work, the hydrogen adsorption behavior for MCM-48, metal doped MCM-48 and MCM-41 at low pressure (0–100 kPa) and 77 K is investigated in detail. Moreover, the relationship between pore size and hydrogen storage capacity was further elucidated. Finally, an adsorption-diffusion mechanism was proposed to account for the pore size-hydrogen storage capacity relationship.

2. Experimental details

MCM-48 was synthesized according to the literature [7] with slight modification. KF aqueous solution was added before the addition of TEOS and the hydrothermal growth was conducted at 423 K for 12 h. The final molar composition of the sol was 1TEOS/0.2CTAB/0.5NaOH/70H₂O/0.1KF. For Cu²⁺ or Ni²⁺ doped MCM-48, the Cu²⁺ or Ni²⁺ aqueous solution was added to the solution after TEOS has been added completely and the molar composition is 1TEOS/0.2CTAB/0.5NaOH/70H₂O/0.1KF/0.005Cu²⁺(Ni²⁺).

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Mesoporous silica MCM-41 was synthesized according to the literature [8] with slight modification. Briefly, 16.5 g $\text{Na}_2\text{SiO}_3 \cdot 5\text{H}_2\text{O}$ was dissolved in 80 mL deionized water. Then, 4.32 g CTABr was dissolved in 70 mL deionized water. Finally two aqueous solutions were mixed together under stirring. pH value of the mixed solution was adjusted to 11 by controlled addition of 6 M H_2SO_4 . The hydrothermal reaction was conducted at 393 K for 48 h.

XRD patterns were recorded on a Rigaku D/Max 2400 diffractometer with a Cu $K\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$) source over the range of $2\theta = 1\text{--}10^\circ$. Scanning electron microscopy (SEM) images were obtained on a QUANTA 450 instrument with an acceleration voltage of 30 kV. Hydrogen storage capacity was measured using a Cahn-2000 high-vacuum electronic balance testing system.

3. Results and discussion

The small-angle XRD results in Fig. 1a indicated a long-range ordered hexagonal mesoporous structure for the synthesized MCM-41 and a well-ordered three-dimensional pore structure for the synthesized MCM-48. Fig. S2 showed the average pore size of prepared MCM-41 and MCM-48 powders calculated from BJH model. Obviously, the mean pore diameter of MCM-41 is larger than that of MCM-48. Fig. 1b showed H_2 adsorption excess isotherms of MCM-48 and MCM-41. It is interesting to note that MCM-48 exhibited higher hydrogen storage capacity in the

pressure range of 0–35 kPa. However, with further increase of operating pressure, hydrogen storage capacity of MCM-41 became higher. It should be noted that pore geometries of MCM-41 and MCM-48 are very similar to each other so that pore size may be the key factor dominating the unique H_2 adsorption phenomenon.

Cu^{2+} and Ni^{2+} were then doped into MCM-48 in an attempt to accurately manipulate their pore size so that the relationship between pore size and hydrogen storage capacity can be more straightforward. Fig. 2a shows the XRD patterns of MCM-48, Cu/MCM-48, Ni/MCM-48. All samples exhibited strong $\{2\ 1\ 1\}$ diffraction peak and weak $\{2\ 2\ 0\}$, $\{4\ 2\ 0\}$, $\{3\ 2\ 2\}$ diffraction peaks. Compared with pure MCM-48 phase, XRD patterns of metal-doped MCM-48 shifted slightly to a slightly lower angle, which can be attributed to the size discrepancy in metal ions: the radii of Cu^{2+} (Pauling radius = 73 pm) and Ni^{2+} (Pauling radius = 69 pm) are larger than Si^{4+} (Pauling radius = 42 pm), causing perceivable expansion of framework and therefore, the increase of inter-planar spacing. The order of pore size for prepared samples follows the sequence: Cu/MCM-48 > Ni/MCM-48 > MCM-48. In addition, prepared samples were further characterized by SEM. As shown in Fig. S1, the morphologies of Cu/MCM-48 and Ni/MCM-48 are quite similar with pure MCM-48. In Fig. 2b, it is interesting to observe that, at low pressure (<35 kPa), pure MCM-48 exhibited higher hydrogen adsorption capacity. With further increase of the pressure (up to 100 kPa), however, the hydrogen adsorption capacity

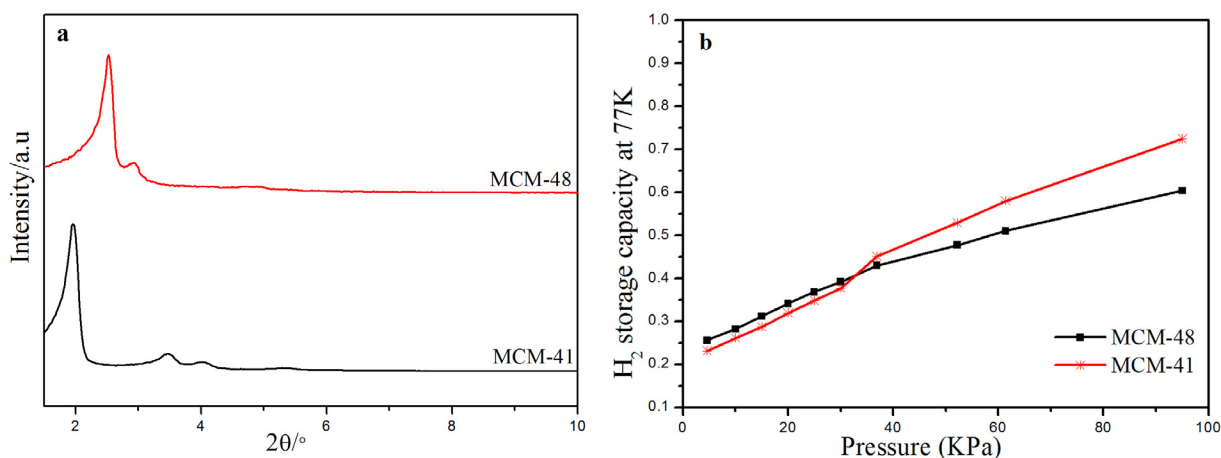


Fig. 1. (a) XRD patterns and (b) Hydrogen storage capacity at 77 K of MCM-48 and MCM-41.

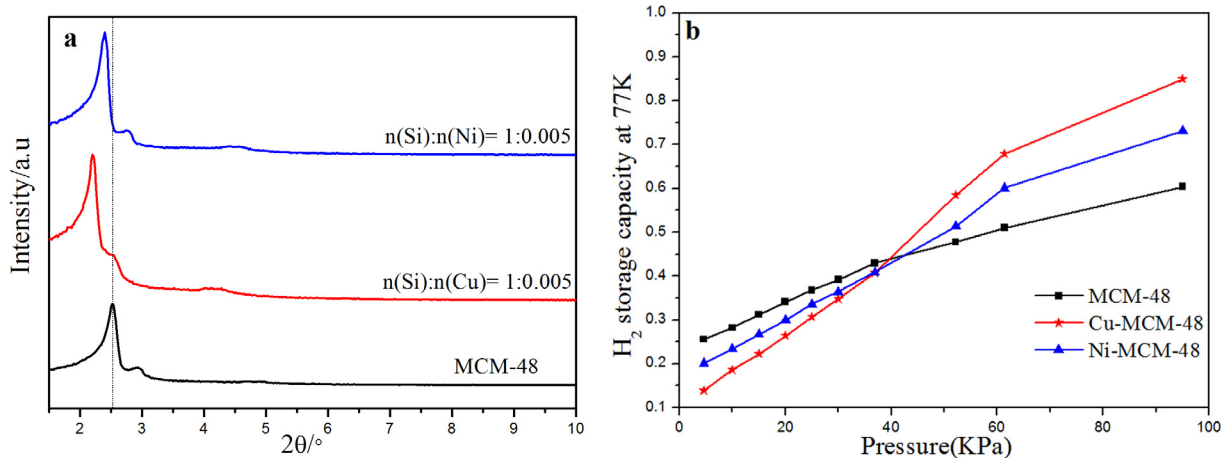


Fig. 2. (a) XRD patterns and (b) Hydrogen storage capacity at 77 K of MCM-48, Cu/MCM-48 and Ni/MCM-48.

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