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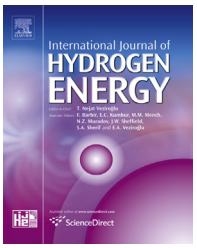

Attrition-resistant Ni–Mg/SiO_2–Al_2O_3 catalysts with different silica sources for fluidized bed syngas methanation

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ABSTRACT

Attrition-resistant Ni–Mg/SiO_2–Al_2O_3 catalysts for fluidized bed syngas methanation were prepared by combining co-precipitation and spray granulation using different silica sources including acidic silica sol (SS), sodium silicate (NS) and tetraethyl orthosilicate (TEOS). Air-jet attrition tests showed that the attrition strength of the resulting catalysts followed an order of C-10TEOS > C-33SS > C-10NS > C-10SS, where C-10TEOS, as an example, refers to the catalyst with 10 wt.% silica using TEOS as its silica source. Characterizations show that the porosity and skeletal structure have strong correlation with catalyst attrition strength. Simultaneous hydrolysis of TEOS and co-precipitation caused C-10TEOS to have dense and continuous skeletal structure to have the high strength of its Ni–Mg/SiO_2–Al_2O_3 precursor particles and thus the improved attrition resistance of sprayed catalyst. Silica sol filled in the voids among precursor particles resulted in a compact framework in C-33SS but its Ni–Mg/Al_2O_3 precursor had weak attrition resistance. Atmospheric syngas methanation over these catalysts at an SV of 600 NL g^{-1} h^{-1} in a fixed bed reactor clarified an activity order of C-10TEOS > C-10NS ≈ C-33SS. The TEOS also enabled highly dispersed metallic Ni and many surface active sites to facilitate the methanation activity and stability of this catalyst.

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Introduction

Due to the high mass and heat transfer efficiency [1–3], fluidized bed methanation for the production of synthetic natural gas (SNG) is currently attracting good attention and has also been well done at the laboratory and pilot scales [4–8]. In catalytic fluidized bed reactor, particles are subjected to attrition via collisions among particles and against the walls of reactor and cyclone [9,10]. This attrition would cause catalyst loss and vary particle size distribution, thus deteriorating the performance of catalyst [2,11]. Till now, there is not any commercial attrition-resistant syngas methanation catalyst worldwide.

Spray granulation has been well applied in preparations of attrition-resistant catalysts with spherical shape and uniform particle size distribution, the important catalyst properties required for fluidized bed reactors [10,12–14]. The typical

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examples are fluid catalytic cracking (FCC) catalyst, iron F-T catalyst and fluidized bed acrylonitrile catalyst [15–17]. Using additives such as binder or structural promoter is an effective way to improve the attrition strength and also catalytic activity and stability of the prepared catalyst. Kim et al. [18] prepared attrition-resistant SAPO-34 catalyst for MTO process using kaolin and alumina sol as binder. Sudsakorn [19] studied the effect of precipitated silica on the attrition resistance of F-T catalyst and found that the use of a small amount of precipitated SiO_2 can lead to high attrition strength and meanwhile good catalytic performance. Holland [20] found that FCC catalyst prepared using colloidal silica and alumina-doped colloidal silica as the binder resulted in good attrition resistance and catalytic activity.

The attrition to catalyst comes from both abrasion (removal of particle surface layers or corners) and fracture (fragmentation of particles) [11,21]. The added structural promoter or binder forms a robust skeletal structure to avoid catalyst particles from breakage and also to minimize abrasion in fluidized bed [17,19,22]. Silica was always chosen as the structural promoter and/or binder for making catalysts with high attrition resistance [22]. Goodwin and co-workers [17,19] investigated the effects of addition amount and method of silica on the attrition properties of the resulting catalyst. Bukur et al. [23] found that among their tested silica sources the colloidal silica enabled the highest attrition strength. Our previous study has prepared attrition-resistant Ni–Mg/Al_2O_3 methanation catalysts using different binders, and we found that the catalysts using silica sol, especially acidic silica sol, as binder showed tighter structure, fewer large pores and higher attrition resistance than that using alumina sol [24]. There are different types of silica, such as binder silica and precipitated silica. Different types of silica additive lead to the formation of differently formed SiO_2 networks so that the attrition strength and catalytic performance of the prepared catalysts vary [17,19,25,26]. There was no study on effects of the silica source on properties and catalytic performance of Ni-base fluidized bed methanation catalyst.

Therefore, this article is devoted to preparing attrition-resistant Ni–Mg/SiO_2–Al_2O_3 spherical catalyst for fluidized bed syngas methanation by co-precipitation and in turn spray granulation using different silica sources including acidic silica sol (SS), sodium silicate (NS) and tetraethyl orthosilicate (TEOS). The study also investigated the relationship between the catalyst attrition resistance and the structure properties of catalysts in their calcined state. These studies are expected to optimize the type of silica sources used for preparing the attrition-resistant methanation catalyst with low loss of catalytic activity for the applications to fluidized bed methanation.

Experimental

Catalyst preparation

All chemicals for catalyst preparation are in technical purity and used without further purifications. Catalysts were prepared in kilograms by co-precipitation and then spray granulation using silica sol (acidic silica sol with pH = 3.0) and precipitated silica (sodium silicate and tetraethyl orthosilicate) as silica sources. A detailed description of the co-precipitation method is available in our previous publication [24]. When using silica sol (SS), the Ni–Mg/Al_2O_3 precursor was prepared by co-precipitation of nickel nitrate, magnesium nitrate and aluminum nitrate through addition of a $NaAlO_2$ solution to maintain the precipitation pH at 11. The formed precipitate was further crystallized at 473 K for 10 h and in turn washed, dried, crushed and re-slurried by adding a preset amount of SiO_2 from a commercial acidic silica sol as the binder. The formed slurry was granulated by spray in a bench-scale spray dryer at an inlet temperature of about 513 K, which was followed by calcination at 873 K for 4 h in air to obtain the final catalyst. When sodium silicate (NS) or tetraethyl orthosilicate (TEOS) was used as the silica source, the catalyst precursor was prepared by co-precipitation of metal nitrates and NS/TEOS with $NaAlO_2$ at pH = 11. Then the precursor was similarly, in succession, washed, dried, crushed, re-slurried, spray granulated and calcined to get the catalyst.

In this study, two catalysts based on NS and TEOS as the silica source and two catalysts at different amounts of SS binder (10% and 33%) were prepared. The obtained catalysts were correspondingly designated as C-10NS, C-10TEOS, C-10SS and C-33SS according to their silica source and silica amount. Another catalyst 20Ni with its composition of 20Ni/10Mg/70Al_2O_3 (weight percent) but without silica additive was prepared following the same co-precipitation method shown above as a benchmark. Table 1 lists the NiO and SiO_2 contents of all prepared catalysts, showing that the NiO content of all catalysts are about 22 wt.%, and the SiO_2 content in C-10NS, C-10TEOS and C-10SS are about 10 wt.%, while that in C-33SS is about 32 wt.%.

Table 1 – Composition and attrition results of the catalysts prepared by spray granulation with different silica sources.

Catalyst	NiO content[a] (wt.%)	SiO_2 content[a] (wt.%)	Attrition index (%/h)	$d_{4,3}$ (μm)		ΔY (%)	Fines fraction (vol.%)
				fresh	attrited		
C-10NS	22.26	10.95	4.13	67.21	51.15	23.90	31.00
C-10TEOS	21.81	9.33	2.18	64.47	55.63	13.71	11.58
C-10SS[b]	21.64	10.82	31.65	65.04	42.34	34.90	54.54
C-33SS	21.45	32.08	2.41	65.28	59.40	9.00	12.93
γ-Al_2O_3	–	–	2.95	–	–	–	–
FCC	–	–	1.36	–	–	–	–
20Ni	21.19	–	–	–	–	–	–

[a] The NiO and SiO_2 contents were determined by XRF.
[b] C-10SS was tested for only 1 h in the attrition test.

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