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高镍负载量Ni/SiO_2和低镍负载量Ni-Ce/SiO_2催化CO甲烷化的比较研究

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A comparative study on the catalytic properties of high Ni-loading Ni/SiO_2 and low Ni-loading Ni-Ce/SiO_2 for CO methanation

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摘要 采用等体积浸渍法制备了高镍负载量的13%Ni/SiO_2(13Ni/Si)催化剂和低镍负载量的7%Ni-2%Ce/SiO_2(7Ni-2Ce/Si)催化剂.通过N_2物理吸附、XRD、FT-IR、TEM、H_2-TPR/TPD等技术对催化剂进行表征,在连续流动微反装置上考察了催化剂的CO甲烷化活性.结果表明,在7Ni-2Ce/Si催化剂中NiO、CeO_2和SiO_2之间产生的相互作用,改变了Ni-O-Si键的化学环境,促进了氧化镍物种的分散和还原,进而提高了催化剂的活性比表面积,同时在催化剂表面形成了新的中等强度的CO吸附中心.与高镍负载量的13Ni/Si催化剂相比,低镍负载量的7Ni-2Ce/Si表现出更高的CO吸附能力和甲烷化活性.常压下,在CO体积分数1%和空速7 000 h^{-1}的反应条件下,低镍负载量的7Ni-2Ce/Si催化剂上CO完全甲烷化最低温度为230 ℃,比高镍负载量的13Ni/Si低了30 ℃.

关键词: CO甲烷化 Ni-Ce/SiO_2催化剂 CeO_2助剂 镍负载量

Abstract: Two Ni-based catalysts of 13%Ni/SiO_2(13Ni/Si) and 7%Ni-2%Ce/SiO_2(7Ni-2Ce/Si, by weight) were prepared by the incipient-wetness impregnation method and characterized with N_2-sorption, XRD, H_2-TPR, FT-IR, TEM, H_2-TPD and CO-TPD techniques. It was shown that addition of Ce promoter generated an interaction among NiO, CeO_2 and SiO_2, which changed chemical environment of Ni-O-Si bond, enhanced the dispersion and reduction of NiO, and increased the active surface area. In particular, a new type of moderately strong CO adsorption sites was formed on the surface of the 7Ni-2Ce/Si catalyst. As a result, the low Ni-loading 7Ni-2Ce/Si catalyst exhibited higher CO adsorption capacity and CO methanation catalytic activity than the high Ni-loading 13Ni/Si. Under the reaction conditions of 1% CO (volume fraction in H_2 atmosphere), GHSV of 7 000 h^{-1} and atmospheric pressure, the temperature for complete conversion of CO over the 7Ni-2Ce/Si catalyst was 230 ℃, being 30 ℃ lower than that found over the high Ni loading 13Ni/Si catalyst.

Key words: CO methanation Ni-Ce/SiO_2 catalyst cerium promoter Ni loading

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