High efficient hydrogen production from methane with a nanosized Fe/Al2O3 catalyst

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Accepted for publication on 16th April 2015

Clean hydrogen can be produced in the direct non-oxidative decomposition of methane over nanosized catalyst. Metal nanoparticles always appeared at the tip of the carbon product as carbon nanotube or carbon nanofilaments. We reported the use of a nanosized Fe catalyst to decompose methane, rather than the conventional choice of nickel based catalyst. We find the activity of metal catalyst is strongly influenced by the structure of carbon. Small Fe nanoparticles had very high activity for this reaction, due to the unstable iron carbide in nanosize. Smooth carbon diffusion resulted in the super long catalyst life time of individual 2-3 nm Fe nanoparticles at the temperature of 1000 ^{o}C. On the other hand, we still made great effort to decrease the reaction temperature of this process, which is much effective to increase the catalyst lift time and is favorable for the process control in large scale production. One effective way is to use highly active hydrocarbons to enhance the decomposition of methane. Thermal couple effect was observed in the coupled decomposition of ethylene or acetylene, which is exothermic reaction, with the decomposition of methane, which is an endothermic reaction. Finally we also discussed the reactor design concept for this special process. The accumulation of coke on the catalyst with rapid increase of product volume, although not resulting in the deactivation of the catalyst under careful control, changed the contact time of methane with the diluted metal catalyst inside carbon. Counter-current contact manner of catalyst with the methane in multistage fluidized is effective to realize the high efficient production of hydrogen and nanocarbon.

Keywords: Hydrogen; Methane; Non-oxidative decomposition; Nanosized Fe catalyst

4th International Symposium on
Energy Challenges & Mechanics
- working on small scales
11-13 August 2015
Aberdeen, Scotland, UK

利用纳米铁催化剂高效裂解甲烷制备清洁氢气

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无氧条件下使用纳米催化剂直接裂解甲烷是有效的制备清洁氢气的方法之一，因为其不产生 CO 与 CO_2。 这个过程中纳米金属催化剂常出现于碳产品（如碳纳米管或纳米碳纤维）的顶部。我们报道使用纳米铁基催化剂，而不是传统的镍基催化剂来裂解甲烷。我们发现纳米催化剂的活性严重依赖于伴生的碳产品的结构。小的铁纳米颗粒具有非常高的活性，因为小的碳化铁相非常不稳定。稳定的碳扩散可使单分散的 2-3nm 的铁颗粒催化剂可以在 1000℃保持非常长的寿命。当然，我们也致力于研究在高效裂氢气的前提下尽可能降低反应温度。因为这对于从根本上提高催化剂的寿命以及减化放大制备过程中的控制非常有益。一个有效的方法是利用高活性的碳源来促进甲烷的分解。比如使用裂解时放热的乙烯或乙炔可以促进甲烷的分解，因为后者的分解是吸热。在此存在着有趣的热能耦合效应。最后，我们讨论这个过程中适宜的反应器形式，因为体积不断增加的碳产品虽然在精细的控制下不致于使催化剂过程失活，但确实会降低在其中的纳米催化剂与甲烷的接触效率。逆流接触的多段流化床是有效的反应器形式，可以有效促进氢气与纳米碳的高效制备。

关键词：氢气；甲烷；无氧分解；纳米铁催化剂