Abstract

Title: "Synthesis of blue hydrogen, CNTs, and graphene grafted CNTs from methane in fluidized bed reactor."

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Hydrogen is being considered as a clean energy carrier because of its high calorific value and non-polluting nature. Presently 80% of global hydrogen demand is concentrated in refineries and fertilizer industries (for ammonia production), most fulfilled through commercial Steam Methane Reforming (SMR). Challenges of SMR include its energy-intensive nature and generation of approximately 13.7 kg of CO_2 per kg of H_2 in the gas stream. Thus, the product must be purified to remove CO_x. An alternate way to produce hydrogen is through the Thermo-Catalytic Decomposition (TCD) of methane gas, in which hydrogen and carbon are the only products. The carbon can be produced in various valuable forms, i.e., CNTs, graphene, graphite, etc. The energy input requirement per mole of methane for the TCD process is appreciably lesser than that of SRM (75.6 and 165 kJ/mol CH_4, respectively).

The present study investigates the TCD of methane in a fluidized bed reactor. A significantly high amount of hydrogen and carbon was produced over selective nickel-based copper/zinc-promoted alumina-supported catalyst. The catalyst particle size range and minimum fluidization velocity were identified by the cold flow hydrodynamic study. The addition of a small amount of Cu and Zn enhances the reducibility and the thermal stability of the catalyst. It increases the dispersion and decreases the deactivation rate. The produced carbon was characterized by various analytical tools, which showed that it is in the form of multi-walled bamboo-shaped carbon nanotubes (CNTs). Low ID/IG from Raman spectroscopy confirmed the purity of produced CNTs. The separation of the CNTs and partial regeneration of the spent catalyst was performed using the non-destructive ultrasonication technique. The regeneration ability of the catalyst was examined by performing multiple reaction-regeneration cycles. The regenerated catalyst regained its full activity, and more than 90% methane conversion was achieved with high-quality CNTs (o.d 60-80 nm, length 5 µm).

This overall approach not only eliminates the CO_x production in the product stream but also captures and extracts the carbon in the form of valuable CNTs. The selling price of the produced CNTs can significantly improve the overall process economics. Thus, efforts have been made to improve the quality of CNTs by varying the active metals and their concentration in the catalysts. The produced CNTs were utilized to prepare ultralight, hydrophobic, and highly porous aerogels that can be used as a proficient and recyclable oil adsorbent during oil spill incidents.