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As a technology that utilizes abundant natural gas resources, it is very important to develop catalysts that directly convert methane into high value-added chemicals. Stable molecular structure and high C-H bond dissociation energy require excessive thermal reaction conditions, and these harsh thermal system produces additional carbon dioxide due to overoxidation. Herein, we develop the novel SiO₂@V₂O₅@Al₂O₃ core@shell nanostructure as a catalyst for selective oxidation of methane to formaldehyde. The new core@shell catalysts are prepared by the hydrothermal synthesis and subsequent atomic layer deposition (ALD). The core SiO₂@V₂O₅ nanostructures are coated by Al₂O₃ shell through multiple ALD cycles, enhancing catalytic activity and thermal stability in the methane partial oxidation. The highest catalytic activity (CH₄ conversion = 22.2%; HCHO selectivity = 57.8%) at 600 °C is shown in the core@shell catalyst prepared by 50 ALD cycles. *In situ* XRD, H₂-TPR, Raman, and UV-Vis characterizations demonstrate that the new T_d monomeric vanadium species with V-O-Al bond is responsible for the high methane conversion.