

Supplementary Information

Alumina Coated with Titanium Dioxide Supported Iron for Hydrogen Production and Carbon Nanotubes Via Methane Decomposition.

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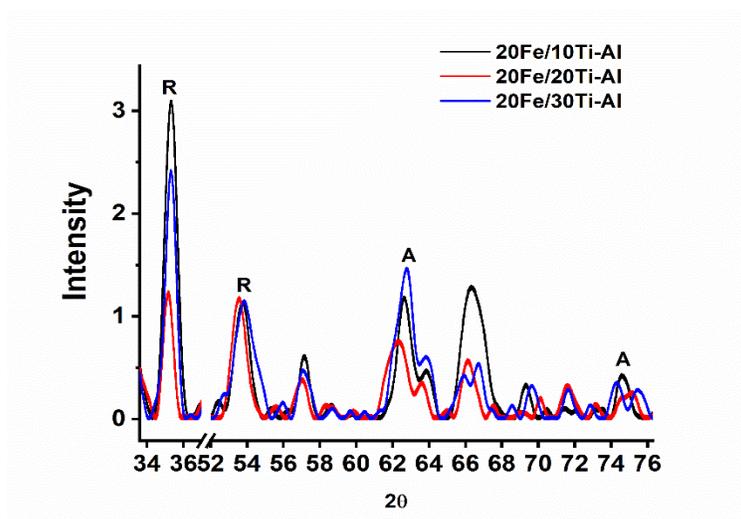


Figure S1. The X-ray diffraction of the catalyst in the selected Bragg's angle. Note: Here, R and A signify for Rutile and Anatase phases.

Catalytic Reactor Setup S1

The experimental setup for the catalytic decomposition of methane is illustrated in Figure S1. The reaction involved two distinct gas sources: CH_4 , and N_2 . These gases were directed from their storage cylinders to a common mixing point. CH_4 initiated the hydrogen production process. N_2 functioned as the standard dilution stream; in addition, before the reaction occurred hydrogen gas was utilized to convert the metal oxide into its metallic state to serve as the active catalyst. The stainless-steel fixed-bed with 9 mm inner diameter and 300 mm length was used as the reactor inserted in a vertical furnace with well-temperature control, the temperature readings were taken from a K-type thermocouple attached to the fixed-bed reactor wall. The gases after the reaction were analyzed using a gas chromatograph. A Thermal Conductivity Detector (Shimadzu GC 2004) was connected to the reactor outlet. Argon was used as the gas carrier for the Gas-Chromatography (GC).

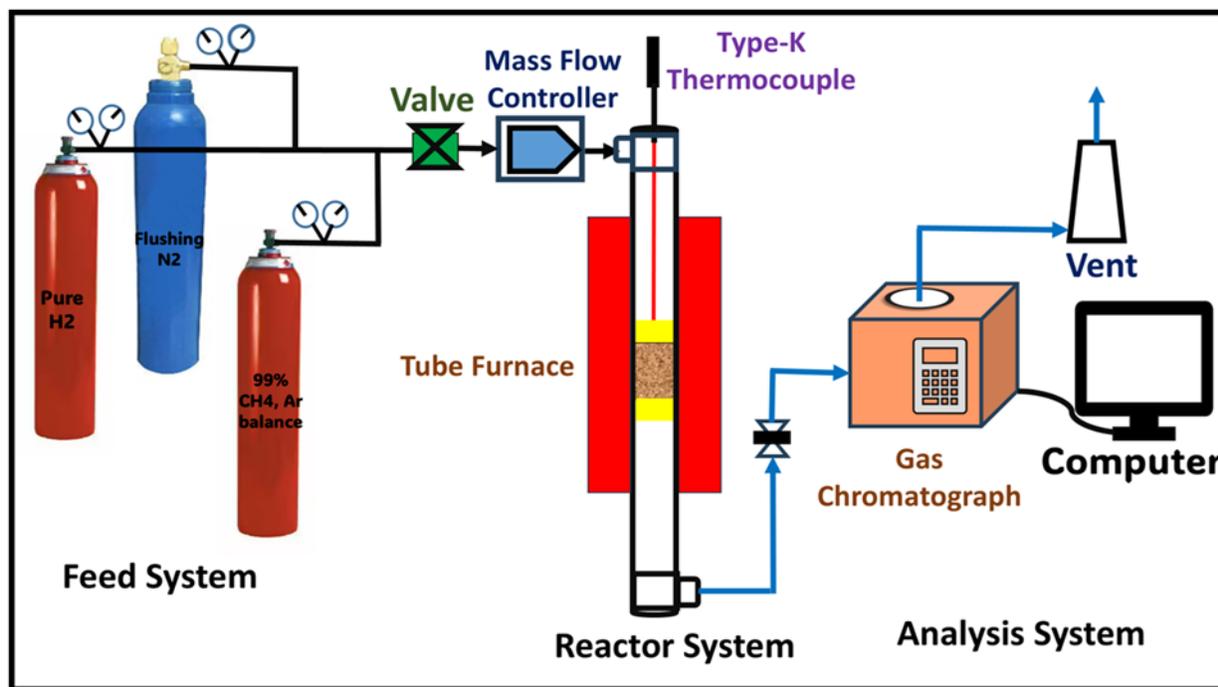


Figure S2 Schematic diagram of the experimental setup for catalytic hydrogen production.

Catalyst Characterization S2

BET

The Brunauer-Emmett-Teller (BET) was used to determine the specific surface area and the porosity of the fresh catalyst with an analyzer called Micro-meristic Tristar II 3020. For each analysis, 0.2g of the catalyst was degassed at 250°C for three hours to eliminate any moisture content and other adsorbed gases.

TPR

Temperature-programmed reduction (TPR) is an analytical technique used to understand reducibility and the behavior of metal oxides of fresh catalysts. The Micromeritics Auto Chem II 2920 was used, and a tube was filled with 70 mg of the catalyst samples then An H₂/Ar mixture flowing at 40 ml/min was introduced as the furnace temperature was raised to 1000°C at 10°C/min. A cold trap within the machine removed the water produced during the reduction, while a thermal conductivity detector recorded the H₂ consumed.

XRD

X-ray diffraction (XRD) is a highly effective tool for determining the crystalline phases in a fresh catalyst and for identifying any impurity phases that could impact the catalyst's performance. The Miniflex Rigaku diffractometer was used with Cu Ka X-ray radiation operating at 40 kV and 40 mA. The diffraction 2θ angle range was set to 10-80 with a step size of 0.01. The raw data file from the instrument was analyzed using X'pert high score plus software. The JCPDS data bank was used to match different phases with their corresponding scores.

TGA

Thermogravimetric Analysis (TGA) is a crucial technique used in the characterization of catalysts. It measures changes in a material's mass as a function of temperature or time under a controlled atmosphere. A Shimadzu Thermo-gravimetric analyzer (TGA) determined the quantity of carbon deposits. The process involved heating 10-15mg of the used catalysts from room temperature to 1000°C at a heating rate of 20°C/min. The machine recorded the weight difference.

TPO

Temperature-programmed oxidation (TPO) is significantly valuable for obtaining information about the nature of various carbonaceous species present on the catalyst surface. The Micromeritics Auto Chem II 2920 will be used. The spent catalysts underwent the same pretreatment as in TPR, and the analysis was conducted over a temperature range of 50-1000°C under the flow of 10% O₂/He mixture at 40 ml/min.

Raman

Raman spectroscopy and X-ray diffraction (XRD) is an effective analytical technique for identifying and characterizing carbon deposits on the used catalysts. It can differentiate between various forms of carbon (e.g., amorphous carbon, graphitic carbon) and offer insights into the composition of these deposits. The type of carbon deposited and the graphitization degree were determined using a JASCO laser Raman spectrometer from Tokyo, Japan. The spectrometer used an excitation beam with a wavelength of 532 nm. In addition, the crystalline carbon is determined by using XRD analysis by the method described in the previous section.

SEM&TEM

SEM and TEM measurements were conducted on the fresh and spent samples to inspect the morphology of the dispersion of active metal on the support for the fresh catalyst and the deposited carbon types for the spent catalyst. The JEOL (JEM-2100F, JEOL Ltd, Tokyo, Japan) transmission electron microscope was used for this purpose, operated at an accelerating voltage of 120 KV. Before TEM imaging, the samples were prepared by dispersing them in absolute ethanol and sonicating them for 30 minutes. Afterward, the dispersion was drop-casted on a TEM grid coated with a sample and left to dry at room temperature.