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Carbon nanostructures grown on 3D silicon carbide foams: Role of intermediate silica layer and metal growth

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HIGHLIGHTS

- Carbon nanostructures (CNSs) were grown on 3D silicon carbide foams.
- SiC foams were coated with intermediate mesoporous silica layers.
- Effect of metal growth on the morphology of CNSs were studied.
- Uniformity of CNSs can be obtained at a mild temperature of 700 °C.

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ABSTRACT

Carbon nanostructures (CNSs) in the form of fibers and spheres have been successfully grown on 3 dimensional (3D) macroscopic silicon carbide (SiC) foams with the aid of mesoporous silica-films as interfacial layer via ethylene decomposition on nickel and iron as growth catalysts using chemical vapor deposition (CVD). The effect of growth temperature from 600 to 900 °C on morphology, diameter and specific surface area of as-grown CNSs was studied. The as-grown CNSs were characterized by field emission scanning electron microscope (FE-SEM) and transmission electron microscope (TEM) to examine the morphology, diameter, microstructure and defect. Crystallinity and degree of graphitization were determined by X-ray diffraction and Raman spectrometer. Textural properties such as specific surface area, pore size and pore volume were measured by nitrogen gas adsorption–desorption. Uniform distribution of CNSs on 3D SiC-foam was obtained at 700 °C for short synthesis time (30 min), as well as high surface area of above 100 m^2/g. Degree of graphitization of CNSs grown on modified SiC substrates by using Ni as growth catalyst was higher than the one with the Fe-growth catalyst.

1. Introduction

Structured catalysts offer better solution for some classical problems in heterogeneous catalysis such as heat and mass transfer limitations, pressure drop, hot spot formation, low dispersion of active metals and low surface area of catalysts [1–4]. The main challenge in structured catalysts is the incorporation of active catalysts on 3D solid foams, monoliths and microchannels [5–12]. Recently, there have been great interests to incorporate large surface area carbon nanostructures (CNSs) onto 3D solid foams and microchannels [3,13,14]. Carbon nanostructures are promising graphitic catalyst and catalyst supports in heterogeneous catalysis, due to its excellent properties such as high surface area, better corrosion resistance, high chemical and thermal stability, low mass density, and high thermal conductivity [8,10,15]. In addition, CNSs in the form of fibers, tubes and spheres are inert catalyst support without acidity and significant strong metal-support interactions (SMSI), unlike alumina and zeolites. With inert materials such as carbon nanostructures, the competition between the active nanoparticles with the reactive catalysts supports can be avoided [7,16,17].

Carbon nanostructures (CNSs) have been grown via laser ablation [18], arc discharge method [19], and chemical vapor deposition method (CVD). The later received much attention due to higher growth rate and cheaper cost for large-scale production [3]. Typically, CNSs formations via CVD occur in the temperature range of 400–1000 °C by decomposition of various carbon-sources

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(CH_4, C_2H_6, C_6H_6, CO, CO + H_2) in the presence of transition metal catalysts (Fe, Co and Ni) [20,21].

In general, CNSs formation involves three main steps (i) adsorption and decomposition of carbon source on active surface of growth catalyst, (ii) followed by dissolution of carbon to form metal-carbide, and (iii) the diffusion of dissolved carbon through active metal nanoparticle and finally CNSs growth [1,22].

The scope of this present work is to grow uniform CNSs on macroscopic 3-D silicon carbide (3D-SiC) foam at high temperatures of 600–900 °C with the aid of mesoporous silica as interfacial oxide via catalytic thermal chemical vapor deposition (C-TCVD) using nickel and iron as growth catalysts. However, distribution and uniformity of CNSs on solid support foam depends on various parameters that includes; source and purity of carbon-containing precursor, the reduction ability and availability of active catalysts, process temperature, interaction between the active catalyst and the support, the nature of mesoporous interfacial oxide to act as the anchorage site (Figs. 1 and 2).

The choice of 3D-SiC foam is to retain the excellent properties of as-grown CNSs on the support and to solve the challenges of handling and high pressure drop pose by the use of powdery support. The chemical inertness of the SiC-based supports allows the recovery of both the active phase and the support after the end-life of the catalyst by a simple acid or basic treatment. It is expected that the possibility to regenerate the active phase and the support will significantly reduce the economic feasibility of the process [7,8].

It is expected that the as synthesized CNSs on macroscopic 3-D SiC foam with high specific surface area, and less microporosity will be a potential catalyst supports for efficient mass and heat transfer of reactant(s) and product(s) in heterogenous catalytic applications [2,23]. These include conversion of ethylbenzene to styrene, [24] oxidation of H_2S to elemental sulfur, and decomposition of hydrazine and NH_3, [8] and structured catalyst in dry reforming of methane.

2. Experimental

2.1. Materials and reagents

β-SiC foam (60 PPI, 8 cm diameter cylinder) having BET surface area of 34 m^2/g was obtained from SiCAT (Germany), and diced into suitable pieces, $Fe(NO_3)_3{\cdot}9H_2O$, $Ni(NO_3)_2{\cdot}6H_2O$, tetra ethyl orthosilicate (TEOS), 1-propanol, 1-butanol, HNO_3, and CTAB were purchased from Sigma–Aldrich. All gases used are of high purity (H_2, C_2H_4, Ar, N_2, Air) 99.999%, supplied by Saudi industrial gas company (SIGAS).

2.2. Foam modification

Calcination of β-SiC foam was performed in high temperature electric furnace at 900 °C to create nanoscopic layer of silica. The temperature was ramped at 1 °C/min to 120 °C, held for 2 h to remove any moisture, other impurities, and the foam was heated to 900 °C at 10 °C/min, and held for 3 h.

2.3. Synthesis of mesoporous silica layer by evaporation induced self-assembly (EISA)

Evaporation induced self-assembly (EISA) was used in preparation of mesoporous silica layer [25] by addition of 28.06 g of 1-propanol to 15.04 g of TEOS and stirred for 5 min, followed by 1.04 g of concentrated nitric acid (1 M) and 3.98 g of de-ionized water, stirred for additional 60 min. Silica solution was obtained after

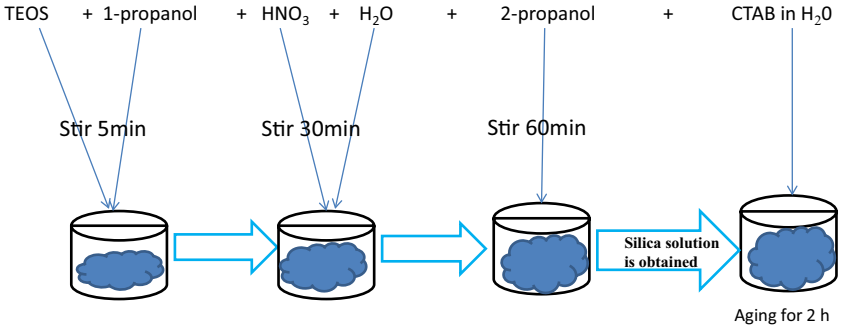

Fig. 1. Schematic representation of evaporation induced self-assembly of mesoporous silica film.

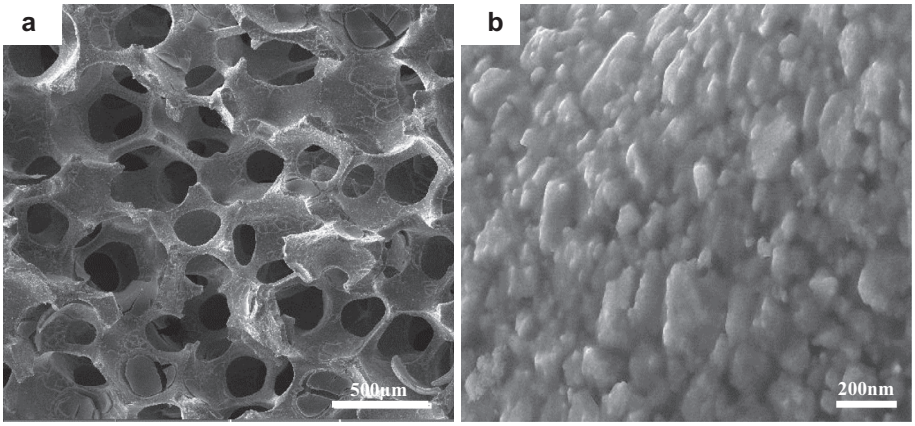

Fig. 2. (a) FESEM image of as-received SiC-foam. (b) SEM image of mesoporous silica deposited on SiC-foam.