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Decomposition-adsorption-deposition: An effective and novel technique for synthesis of hexagonal boron nitride microsheets

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ABSTRACT

Magnesium diboride as a precursor with dual role of Nitrogen introduces a simple "decomposition-adsorption-deposition (DAD)" technique for the synthesis of Hexagonal Boron nitride microsheets (BNMSs) on silicon (Si) substrate at 1100 °C. The synthesized BNMSs has the apparent morphology

like the white dispersed feathers on a plate surface with diameter in the range of 3–15 µm and lengt greater than 30 µm. All the BNMSs has the characteristics of *h*-BN lattice with an interlayer spacing of 0.34 nm. Boron and Nitrogen elemental compositions and *h*-BN phase of the synthesized BNMSs are verified from its characterization by X-ray photoelectron spectroscopy (XPS), Fourier transformed infrared (FTIR) spectroscopy and Raman.

1. Introduction

Hexagonal boron nitride (*h*-BN) consists of sp^2-bonded boron and nitrogen atoms in two dimensional (2D) layers. It is most commonly known as "white graphite". However, Unlike graphite, it is a wide band gap semiconductor [1] with a direct band gap of 5. 97 eV [2]. It is famous for its excellent properties. These include a high melting point of nearly 2600 °C, high temperature stability up to 1000 °C in the air and up to 2200 °C and 2400 °C in the argon and nitrogen atmosphere respectively. Beside this, it has high thermal conductivity, mechanical strength, low dielectric constant and corrosion resistance (i.e. offers resistance to corrosion in acidic as well as in molten metals) [3]. Based on the fore mentioned properties, *h*-BN has applications in various fields of electronics, chemistry, high temperature technology and metallurgy etc. Heat sinks, CVD crucibles, vacuum melting crucibles, substrates, boron doped silicon wafers in semiconductor processing, sputtering targets and high temperature furnaces are some of the typical uses of *h*-BN [3].

Reduction in size of materials from bulk to nano (10^{-9}) or micro (10^{-6}) substantially improve its properties. It happens for the low dimensional materials due to electrons and holes confinement [4], surface effects, and geometrical confinement of the phonon. As a result, it make the materials suitable for a variety of its potential applications in modern technology as electronic and structural material. These applications lie in the field of hydrogen storage, fuel cells, drug delivery, insulation, information technology, composite and microelectronic mechanical systems (MEMs) etc. [5].

Layered sheets of *h*-BN in low dimensional (nano & micron size) can

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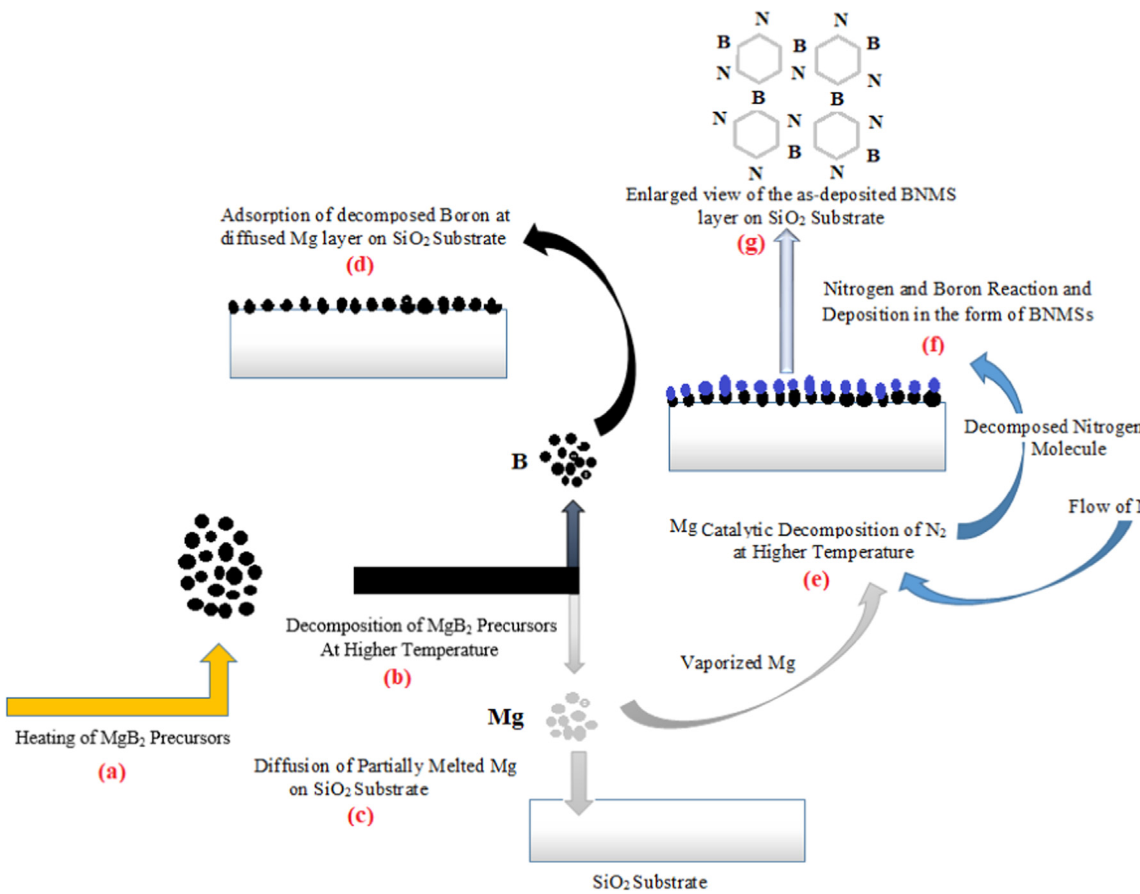

Fig. 1. Sketch (a - g) shows an illustration of Decomposition-Adsorption-Deposition (DAD) technique used during the synthesis of BNMSs.

be used in far-ultraviolet light emitting diodes and as a charge leakage barrier layer in electronic equipment due to its nature as a large band gap semiconductor [6]. Layered structures of h-BN are useful in edge control of graphene domains [7], growth of single-crystalline graphene [8] and aligned growth of graphene [9]. Crystalline layered of *h*-BN have ^{10}B contents. The ^{10}B has a larger cross-section for thermal neutron (3840b) [10]. It makes ~ 20% of the natural B whereas the rest of ~ 80% consist of boron-11 (^{11}B) [11,12]. The ^{10}B contents in BNMSs produce charged daughter nuclei when interact with thermal neutron. The bond of B and N makes a semiconductor layers. These layers emit electron-hole pairs by the interaction of charged nuclei within the same material. The detected electron-hole pairs are counted for the strength of neutron flux. Therefore, *h*-BN layered structures of BNMSs can be a good choice for making a solid state neutron detector with greater detection efficiency [10].

Micron size sheets of *h*-BN (BNMSs) has crystalline and multilayered structure with larger surface area. In addition, the B-N bonds has a dipolar nature. Such a characteristics of BNMSs make it a good choice to be used as a potential hydrogen storage element [13].

Though layered structures of *h*-BN have excellent properties for a variety of applications in modern technologies, however, it synthesis in the desired size, quality, shape or morphology is still a challenge for the researchers working in the same field. Some of them have developed mechanical [14] or chemical procedures [15]. In these techniques, *h*-BN bulk crystals have been used as precursors. As a result, a few layered *h*-BN structure has been synthesized by exfoliated it from the bulk crystals of *h*-BN [14]. However, the size of the synthesized product was found to demerits its use in further applications [6]. Chemical vapor deposition (CVD) produces comparatively pure structure of carbon and related materials in the form of thin film on a suitable substrate. The usefulness of this technique also found it ways in the synthesis of *h*-BN structures in the form of thin film on silicon (Si) substrate. In the beginning, most of the CVDs were run with BF_3/NH_3 [16], BCL_3/NH_3 [17], or B_2H_6/NH_3 [18] as precursors. It has been found that these precursors have not only affected the deposition rate but also unable to maintain boron and nitrogen stoichiometric ratio in the final product. Borazine ($B_3N_3H_6$) has also been used as a precursor in deposition of few layer thin film of *h*-BN in atmospheric pressure chemical vapor deposition (APCVD). The as-used $B_3N_3H_6$ as a precursor was not only toxic but also resulted in a few layer thin film of *h*-BN (5 – 50 nm) [6].

In our previous studies, we developed techniques for the synthesis of multilayered micro-flakes of *h*-BN [19] and vertically aligned BNNTs [20]. In the first technique, dual role of N_2 (i.e. as inert atmosphere and nitrogen source) has been discovered during the synthesis of *h*-BN microflakes [19] whereas in the second technique, Magnesium diboride (MgB_2) has been developed as an effective precursor for the synthesis of vertically aligned BNNTs [20]. The logics of N_2 (as a reactive atmosphere and nitrogen source) and MgB_2 (as a precursors) have been combined in the present study. The combination of these logics resulted in the concept of “decomposition-adsorption-deposition (DAD)” technique for the synthesis of BNMSs. The details of all these logics resulted in DAD and the as-produced products are fully discussed in the coming sections.

2. Experimental details

Magnesium diboride (MgB_2) in the form of micron size crystals along with Nitrogen gas of 99.99% purity were bought as precursors and a single zone quartz tube furnace as a major part of the experimental set up. The precursor's powder in 100 mg of weight was taken in alumina boat. Monocrystalline Silicon wafer with IC Grade, Conductive type: P, Dopant: Boron and Size: 3"- 8" [21] has been bought, cut (in