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Modeling studies of an impinging jet reactor design for hybrid physical–chemical vapor deposition of superconducting MgB_2 films

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ABSTRACT

An impinging jet reactor was developed for the deposition of superconducting MgB_2 thin films by hybrid physical–chemical vapor deposition, a technique that combines Mg evaporation with the thermal decomposition of B_2H_6 gas. A transport and chemistry model for boron film deposition from B_2H_6 was initially used to investigate the effect of carrier gas, Mg crucible temperature and gas flow rates on boron film growth rate and uniformity. The modeling studies, which were validated experimentally, demonstrated a reduction in B_2H_6 gas-phase depletion and an increased boron film growth rate using an argon carrier gas compared to hydrogen. The results were used to identify a suitable set of process conditions for MgB_2 deposition in the impinging jet reactor. The deposition of polycrystalline MgB_2 thin films that exhibited a transition temperature of 39.5 K was demonstrated at growth rates up to ~50 μm/h.

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1. Introduction

The discovery of superconductivity in MgB_2 (T_c~39 K) [1] has stimulated interest in developing in situ growth techniques to facilitate the fabrication of Josephson junctions for superconducting electronics as well as thick coatings on conductive wires and tapes for magnetic resonance imaging (MRI) magnets, RF cavities and undulators for advanced light sources. The main challenge associated with the development of a suitable process is that high overpressures of Mg vapor must be present for MgB_2 phase stability at the elevated temperatures desired for epitaxially oriented films (~720 °C) [2]. Hybrid physical–chemical vapor deposition (HPCVD) is a promising technique that has been shown to produce epitaxial MgB_2 thin films on (0 0 0 1) SiC substrates with a superconducting transition temperature greater than that of the bulk material (T_c~42 K) and low residual resistivity [3,4]. The HPCVD technique, described in detail previously [3,5], combines the thermal evaporation of high-purity Mg pellets and the thermal decomposition of diborane (B_2H_6) gas to deposit MgB_2 thin films at elevated substrate temperatures (~700 °C) and increased reactor pressures (~100 Torr) compared to high vacuum deposition methods.

In the original reactor geometry used for HPCVD [3,5], the Mg pellets were placed on the same heated susceptor as the substrate. This arrangement limited the operational flexibility of the process by prohibiting independent temperature control of the Mg source and substrate. It also restricted the amount of Mg that could be placed within the reactor during a growth run which limits the deposition time and layer thickness. Consequently, there is strong interest in further developing the HPCVD process to expand the current capabilities and to introduce a greater degree of process flexibility. Recent reports have successfully demonstrated alternative reactor geometries for the HPCVD growth of MgB_2 that incorporates separate heaters for the Mg source and substrate and enables an expanded range of substrate temperatures and deposition times [6–10]. Our initial dual-heater HPCVD design [6] used a resistive heater for substrate temperature control and

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an inductively heated crucible for Mg temperature control. Although this design addressed the lack of independent temperature control in the original HPCVD configuration, the MgB_2 growth rate was significantly reduced in this configuration due to substantial upstream depletion of B_2H_6 from the gas phase resulting from pre-reaction on heated surfaces and gas flows near the substrate surface. Our prior studies [11] demonstrated that the growth rate of MgB_2 is limited by the B_2H_6 concentration in the reactor; consequently, reactor designs that enhance the gas-phase transport of B_2H_6 to the substrate surface are expected to yield increased growth rates. We recently demonstrated high growth rates for MgB_2 (~110 μm/h) using an impinging jet reactor configuration in which B_2H_6 was introduced into the reactor via a water-cooled tube with the gas flow perpendicular to the substrate surface [8]. The high gas velocities obtainable in an impinging jet configuration are expected to reduce the gas residence time and therefore the extent of parasitic gas-phase decomposition of B_2H_6 upstream of the susceptor.

In this study, a transport model of the impinging jet reactor was developed and was used to predict the gas-phase temperature and velocity profiles in this reactor geometry. Given the lack of information on the gas phase and surface chemistry of Mg and B_2H_6 and the associated reaction kinetics, it was not possible to develop a chemistry model for MgB_2 deposition. Instead, a chemistry model for boron film deposition from B_2H_6 was used in combination with the transport model of the impinging jet to carry out a detailed study of the effect of process conditions and carrier gas on the growth rate and uniformity of boron thin films. The model was used to identify process conditions that yield reduced B_2H_6 gas-phase depletion, high boron film growth rates and uniform film thickness, which were subsequently verified experimentally. The results of the boron film growth studies were used to identify a suitable range of conditions for the deposition of MgB_2 films in the impinging jet configuration.

2. Experimental procedure

A schematic of the impinging jet configuration for HPCVD growth of MgB_2 is shown in Fig. 1. The substrate was mounted on a resistive heater, which was encased in a custom housing to protect the heater leads and thermocouple from Mg vapor. The Mg pellets were placed in a stainless steel crucible that was inductively heated. The crucible had a hole in the middle where the B_2H_6 inlet tube was located, which permitted the direct delivery of a mixture of B_2H_6 and carrier gas, either H_2 or Ar, to the substrate. A cone-shaped cap was placed on top of the crucible to help direct the flow of Mg vapor to the substrate and reduce Mg deposition on the inner diameter of the reactor vessel. The B_2H_6 was introduced into the reactor via a water-cooled quartz tube to reduce pre-heating of the gas. An alumina sleeve (or cap) was placed between the crucible and the water-cooled quartz inlet tube to reduce thermal gradients at the top of the inlet tube. In addition to the B_2H_6 jet inlet, an additional purge gas, either H_2 or Ar, was used to reduce the back-diffusion of gases in the region upstream of the Mg crucible.

Boron thin films were deposited on (0 0 0 1) sapphire substrates using the impinging jet reactor configuration shown in Fig. 1 but without the addition of Mg pellets. Diborane (5% B_2H_6 in H_2) was used as the boron precursor with ultra-high-purity H_2 or Ar as the carrier gas. The reactor pressure was 25 Torr, the purge gas flow rate was 400 sccm and the substrate temperature was set at 675 °C in all the experiments. The parameters varied in the experiments included the Mg crucible temperature (80–900 °C) and the inlet jet flow rate (100–825 sccm). The inlet jet mole fraction of B_2H_6 was held constant at 0.005 as the total jet flow rate was varied. The boron film thickness was measured using scanning electron microscopy of the film/substrate cross-sections.

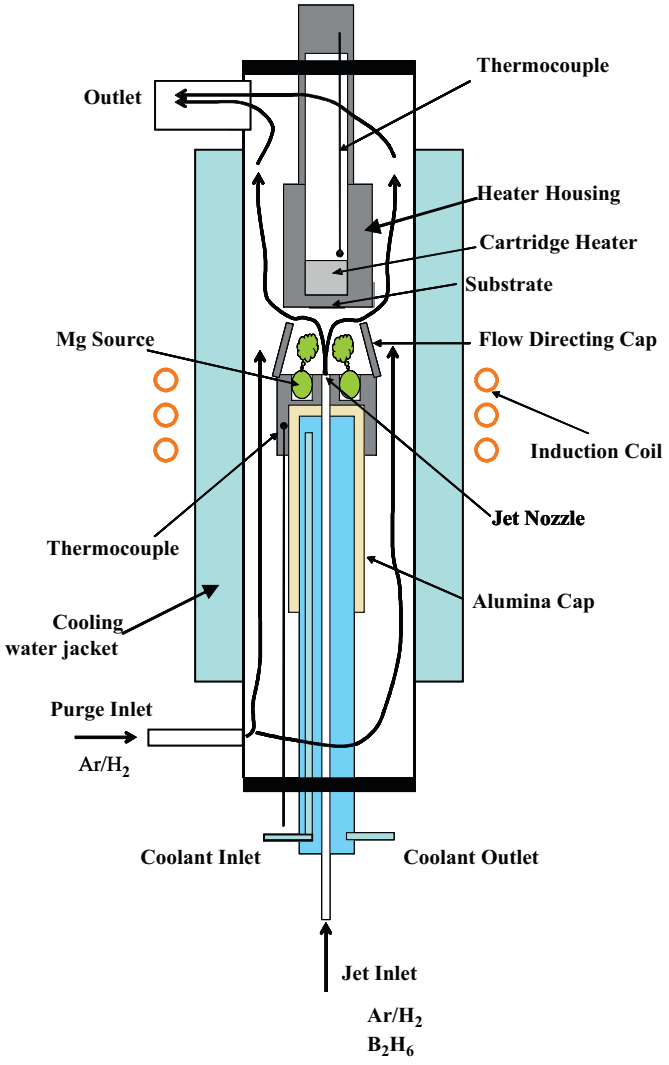

Fig. 1. Schematic of the impinging jet HPCVD reactor.

MgB_2 films were also grown on (0 0 0 1) sapphire substrates in the impinging jet configuration using an Ar carrier gas and conditions similar to those described above for the boron films. In this case, however, the substrate and Mg crucible temperatures were both held constant at 765 °C and the total inlet jet flow rate was varied from 125 to 225 sccm. The MgB_2 film thickness was determined by measuring the height of a step that was etched into the film using HCl. X-ray diffraction measurements were performed using a Scintag model X2 θ–2θ diffractometer with a Cu X-ray tube and a Si(Li) peltier solid state detector. Resistivity measurement of the MgB_2 films was performed using a van der Pauw geometry in a custom cryostat system over a temperature range of 300–30 K.

3. Modeling procedure

Numerical process modeling was used to simulate the reactive flow conditions and predict the boron film deposition rate and film uniformity in the impinging jet reactor configuration. The computational model is similar to that previously developed for boron film deposition in the original HPCVD reactor [12]. The commercial software package CFD-ACE+ (ESI-CFD, Inc., Huntsville,

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