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MgB_2 coated conductors directly grown on flexible metallic Hastelloy tapes by hybrid physical–chemical vapor deposition

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ABSTRACT

MgB_2 coated conductors (CCs), which can avoid the low packing density problem of powder-in-tube (PIT) processed wires, can be a realistic solution for practical engineering applications. Here we report on the superior superconducting properties of MgB_2 CCs grown directly on the flexible metallic Hastelloy tapes without any buffer layer at various deposition temperatures from 520 to 600 °C by using hybrid physical–chemical vapor deposition (HPCVD) technique. The superconducting transition temperatures (T_c) are in the range of 38.5–39.4 K, comparable to bulk samples and high quality thin films. Clear (101) and (002) reflection peaks of MgB_2 are observed in the X-ray diffraction patterns without any indication of chemical reaction between MgB_2 and Hastelloy tapes. From scanning electron microscopy, it was found that connection between MgB_2 grains and voids strongly depend on the growth temperature. A systematic increase in the flux pinning force density and thereby the critical current density with decreasing growth temperature was observed for the MgB_2 CCs. The critical current density (J_c) of $J_c(5\ \text{K}, 0\ \text{T}) \sim 10^7\ \text{A/cm}^2$ and $J_c(5\ \text{K}, 2.5\ \text{T}) \sim 10^5\ \text{A/cm}^2$ has been obtained for the sample fabricated at a low growth temperature of 520 °C. The enhanced J_c (H) behavior can be understood on the basis of the variation in the microstructure of MgB_2 CCs with growth temperature.

1. Introduction

The shortage of liquid helium and resulting soaring price are indeed a serious issue nowadays, which demands the development of cryogen-free superconducting magnets more than ever. The fascinating magnesium diboride (MgB_2) superconductor is a promising candidate material for the development of cryogen-free superconducting magnets and even for low-cost microelectronic devices. Its remarkably high critical temperature (T_c) of 39 K [1] is 2–4 times higher than conventional metallic superconductors, such as NbTi and Nb_3Sn, the prime materials for current superconductor industries [2]. Higher critical current density (J_c) at higher temperature under higher magnetic field is definitely beneficial for any practical application [3]. MgB_2 superconducting wires and tapes have been fabricated by powder-in-tube (PIT) process [4,5]. But PIT-processed conductors face low packing density problem (below 50%), which hampers further progress toward the full potential of this material [6].

On the other hand, dense MgB_2 tapes with high J_c values of $10^5\ \text{A/cm}^2$ at 4.2 K and 10 T have been realized, though only a little work has been reported, through coated conductor approach [7]. For example, Abe et al. reported the transport J_c of 25,000 A/cm^2 at 5 K and 0 T for MgB_2 films grown on stainless steel substrates using the molten-salts electroplating (MSEP) technique [8]. Chen et al. obtained $J_c(10\ \text{K}, 0\ \text{T})$ over $10^6\ \text{A/cm}^2$ for MgB_2 films grown on three different metallic substrates, stainless steel, copper and niobium substrates [9]. The J_c value of $5.5 \times 10^6\ \text{A/cm}^2$ at 10 K in self-field is reported for MgB_2/Niobium substrates by Zhuang et al. [10]. But the J_c of these samples decreased rapidly as the field was increased because of their poor flux pinning properties. In our previous work [11], high self-field J_c values of the order of $10^7\ \text{A/cm}^2$ at 5 K for MgB_2/Cu (001) superconducting tapes have been demonstrated, but the high reactivity of Cu tape with Mg reduced the superconducting cross sectional area and also a decrease of J_c in applied field was observed. Recently, He et al. reported the fabrication of MgB_2 films grown on molybdenum substrates, but the films showed very low $J_c(5\ \text{K}, 0\ \text{T})$ of $6.5 \times 10^5\ \text{A/cm}^2$, probably due to the Mg voids and the poor grain connectivity [12].

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The selection of substrate material is very crucial. Due to the reactivity of the substrate with MgB_2, the presence of secondary or insulating phases strongly affect the overall J_c performance of MgB_2 [13]. Komori et al. could obtain very high in-field transport J_c for MgB_2 grown on yttria-stabilized-zirconia (YSZ) buffered Hastelloy tapes without any chemical reactions in-between MgB_2, YSZ and Hastelloy tapes [7]. Hastelloy tape is widely used as a substrate material for cuprates high-T_c superconductors because of its good material properties, such as good flexibility, lower corrosiveness and most importantly, low AC loss characteristic [14]. In this work, we report that Hastelloy tape without any buffer layer can be a good substrate for MgB_2 CCs. No chemical reaction has been found between MgB_2 and Hastelloy tapes. Excellent superconducting properties, such as T_c, J_c, and H_{c2} of superconductors, can be obtained, which are strongly influenced by heat treatment conditions. The changes in microstructure and superconducting properties of MgB_2 CCs fabricated by hybrid physical–chemical vapor deposition (HPCVD) technique as the growth temperature varies from 520 to 600 °C are discussed.

2. Experimental

Grain boundaries of MgB_2 are not a barrier for transport current flow [15] and are also act as effective flux-pinning centers [16]. Direct deposition of MgB_2 onto a substrate could generate a lot of grain boundaries, the effective pinning centers. If we do not need to deposit buffer layers, the overall fabrication process is pretty simple and at the same time reduces the fabrication cost. Polycrystalline Hastelloy tapes of 45 μm thickness, cut into sizes of 1 cm × 1 cm, were used as flexible metallic substrates. MgB_2 coated conductors (CCs) were fabricated by coating MgB_2 films directly on the Hastelloy tapes by using HPCVD technique. The Hastelloy tape is placed on the top surface of a susceptor surrounded by Mg chips. The reactor was evacuated to a base pressure of 10^{-3} Torr and then purged several times by flowing high purity argon and hydrogen gases. Prior to the film growth, the susceptor was inductively heated towards the set temperature under a reactor pressure of 200 Torr in H_2 atmosphere, so that the Hastelloy tape and Mg chips can be heated together. After reaching the set temperature, a boron precursor gas, B_2H_6 (5% in H_2), was introduced into the reactor to initiate the film growth. The flow rates were 90 sccm for the H_2 carrier gas and 10 sccm for the B_2H_6/H_2 gas mixture, respectively. Finally, the fabricated MgB_2/Hastelloy CC was cooled down to room temperature in a H_2 carrier gas flow. The MgB_2 CCs were fabricated at various set temperatures in the range of 520–600 °C. As the growth temperature was increased from 520 to 600 °C, the film thickness decreased from 1.12 to 0.55 ± 0.05 μm. The changes in thickness might be due to relatively low sticking coefficient of Mg at higher deposition temperatures [17]. More details on the HPCVD technique can be found elsewhere [18,19].

The crystal structures of MgB_2 CCs were investigated by X-ray diffraction (XRD) using Cu Kα as an X-ray source. Surface morphologies and thicknesses were investigated by scanning electron microscopy (SEM). Magnetization hysteresis (M–H) measurements were carried out for all MgB_2/Hastelloy CCs with magnetic property measurement system (MPMS, Quantum Design). Resistivity measurements were carried out by using the standard four probe method. The temperature dependence of resistance under an applied field up to 7 T was measured with physical property measurement system (PPMS, Quantum Design).

3. Results and discussion

The X-ray diffraction patterns for pure Hastelloy tape and MgB_2/Hastelloy coated conductors fabricated at different temperatures of 520, 540, 560, and 600 °C are shown in Fig. 1. The clear (101) and (002) reflections of MgB_2 observed in the XRD patterns indicate the polycrystalline nature of MgB_2 CCs. The c-axis lattice parameter 3.520 ± 0.003 Å was determined from the (002) reflection of the tapes and is comparable to that of MgB_2 bulk samples [20]. There is no indication of chemical reaction between MgB_2 and Hastelloy tapes, any impurity or secondary phases, such as Mg and MgO, reported in other previous works [8,12]. We argue that Hastelloy tape is a better substrate compared to other metallic substrates, such as Cu and so on, which can avoid chemical reactions at the interface.

The plane and cross-sectional views of MgB_2 CCs, examined by scanning electron microscopy, are presented in Fig. 2. Fig. 2a–d corresponds to the surface morphologies of CCs grown at 520, 540, 560, and 600 °C, respectively. It is clear that grains are relatively well connected and voids are smaller at lower temperature. At a lower growth temperature of 520 °C, relatively dense surface is observed as compared to a much more porous surface at 600 °C. The microstructure seems to be much more degraded at higher temperatures and thus low growth temperature is preferable in order to obtain higher J_c. The innermost areas of MgB_2 CCs have been investigated as well by cross-sectional SEM study. The cross-sectional images for the MgB_2/Hastelloy CCs prepared at 520 and 600 °C are shown in Fig. 2e and f, as an example. Quite interestingly, for both samples, we can observe that the voids are initiated from the interface region, which more likely attributed to high deposition rate of 0.1 μm/min.

The temperature dependence of normalized resistance (R/R_{40K}) is shown in Fig. 3. Among all samples, the MgB_2 CC fabricated at a temperature of 560 °C shows the highest T_c of 39.4 K, which is comparable to the best reported data [12,21]. It is higher by 2 K than we obtained for the MgB_2/Cu tapes [11], much higher than that of the MgB_2/Cu plates, 33 K [22] or that of the MgB_2/YSZ/Hastelloy tapes, 29 K [7]. The high T_c is most probably due to the high purity of our samples deposited directly on Hastelloy tapes. The T_c increase with raising the growth temperature, reaches its maximum value of 39.4 K for 560 °C MgB_2 CC, and then, starts to decrease. The growth temperature dependent increase in T_c seems to be related to the improvement in crystallinity, as was reported in our previous work [11]. On the other hand, the transition width, ΔT_c, is found to be monotonically broadened as the growth temperature increased. The ΔT_c for the sample deposited at 520 °C was 0.36 K and at 540 °C – 0.55 K, 560 °C – 0.61 K and at 600 °C – 0.91 K, respectively. The homogeneity of the MgB_2 CCs seems to be degraded with the

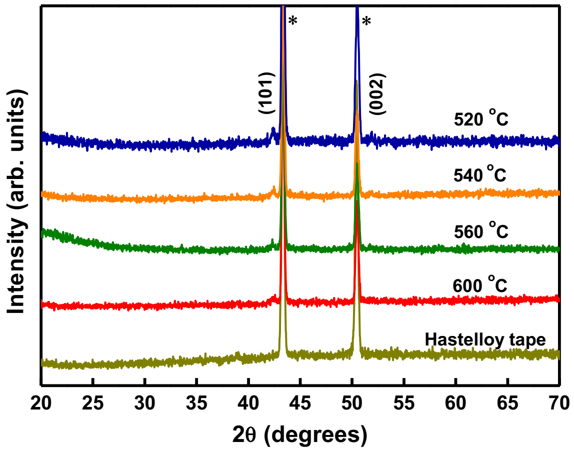

Fig. 1. X-ray diffraction patterns for pure Hastelloy tape and MgB_2/Hastelloy coated conductors fabricated at various temperatures of 520, 540, 560, and 600 °C.