Magnetoresistance properties of MgB$_2$ thick films on YSZ substrate using spray pyrolysis method

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Abstract. The MgB$_2$ thick films have been fabricated on the polycrystalline YSZ substrates using spray pyrolysis technique. Structural/microstructural, electrical and magnetic properties were investigated by XRD, SEM-EDX and R-T measurements. The films fabricated showed dense and homogenous structural formation with strong grain connections. The electrical properties of MgB$_2$ thick films were investigated under different magnetic fields. It was seen that resistive transition broadened with increasing the applied magnetic fields. It was believed that the broadening is due to the thermally activated flux flow (TAFF). The activation energy, $U(H)$, of the MgB$_2$ films was calculated using Arrhenius law.

Introduction

The superconductivity at 39 K discovered in MgB$_2$ among binary alloys attracted much interest in its fabrication techniques and practical applications [1]. MgB$_2$ does not exhibit weak-link at grain boundaries. Low anisotropy, high critical current density, $J_c$, due to strongly linked grains, long coherence length and high critical fields ensure rapid processing in making thin/thick films, wires or nanowires and single crystal. Especially, the low processing temperature in thin/thick film formation offers important electronic applications such as junction. However, MgB$_2$ films processed in the low processing temperature show $T_c$ values between 34 and 37 K, which are relatively low values compared to the $T_c$ value of pure MgB$_2$ material. The incorporation of MgO could be the possible reason for this observed low $T_c$.

H.J.Kim et al reported that the vortex phase diagram of MgB$_2$ films is similar to High-$T_c$ systems and similarity of phase diagrams of MgB$_2$ and $HT_c$ materials leads us to explain the pinning properties in MgB$_2$ easily. MgB$_2$ films show 3D vortex glass state of flux vortices, which is possible origin of high current density, $J_c$ [2,3].

In this paper, we have fabricated MgB$_2$ thick films on the YSZ substrate using spray pyrolysis technique. The effects of high and low heat processing temperature on the structural/microstructural and electrical properties were investigated by XRD, SEM and R-T. Flux dynamics of MgB$_2$ thick films on YSZ substrate are studied by temperature dependence of resistance under magnetic fields up to 4 T. The activation energy, $U(H)$ was calculated using Arrhenius law.
Experimental

High purity MgB$_2$ powder was mixed in absolute ethanol with concentration 0.01 mol/liter. The solution with MgB$_2$ were mixed for 0.2 h and poured into nozzle container. The tip of the nozzle is 0.1 mm diameter. YSZ substrates were put in a glass container and sprayed with the solution. The spraying process was repeated four-five times. Fabricated films were heated at 600$^0$C and 900$^0$C for 24 h under Ar atmosphere in the molybdenum tube.

The films’ microstructure and phase analysis were characterized by XRD (Rigaku RadB) and SEM (Leo Evo 40 VPX). Electrical properties of the films fabricated were investigated by temperature dependence of resistance under under magnetic fields between 0-4 T using VSM (Crygonic) magnetometer.

Results and Discussions

XRD patterns of the MgB$_2$ thick films fabricated are presented in Figure 1. Films heated treated at 600$^0$C and 900$^0$C for 24 h showed the single phase MgB$_2$. The crystal symmetry of both films was found to be hexagonal and the unit cell parameters of the films heat treated at 600$^0$C and 900$^0$C for 24 h were calculated to be $a=3.0881$ and $3.0898$ Å, and $c=3.5341$ and $3.5359$ Å, respectively, which are close to previously obtained results [4,5]. However, it was found that the peak intensities of the film heat treated at 900$^0$C increased. Although the main phase was obtained to be MgB$_2$, some MgO impurity phases were detected due to oxidation of films which are believed to be due to small amounts of oxygen in the non-spectroscopic Ar gas used during the heat treatment cycles.

![Figure 1. XRD pattern of films fabricated at 900$^0$C and 600$^0$C.](image)

Surface morphology of the films and microanalysis of the phases have been investigated by SEM and EDX analysis. The microstructure of the films fabricated at 600$^0$C and 900$^0$C for 24 h is shown in Figure 2.a and b, respectively. It was found that the film fabricated at 600$^0$C consisted mainly of the MgB$_2$ phase, Figure 2.a, which is consistent with XRD pattern. The white colored grains on the surface of the film were identified to be the MgO phase. The surface morphology changed with increasing heat treatment temperature from 600$^0$C to 900$^0$C. More compact grain formation without porosity was observed, Figure 2.b. This is typical structure of MgB$_2$ material which was previously obtained by many research groups [6].
Temperature dependence of resistance in MgB$_2$ films heat treated at 600°C and 900°C was given in Figure 3. The films fabricated under low temperature heat treatment condition showed superconducting transition at 37 K, $T_c$, and the zero resistance temperature, $T_0$, was reached at 20 K, Figure 3.a. The sample shows a metallic behavior in the normal state. However, the magnetic field influenced strongly $T_c$ and $T_0$. Transition broadened with increasing the applied magnetic field and $T_c$ and $T_0$ decreased with the applied magnetic field as given Table 1. Transition broadening ($\Delta T = T_c - T_0$) was found to be 17 K at the magnetic field of 0 T and increased with increasing the applied magnetic field.

Resistance versus temperature curves of the MgB$_2$ films heat treated at 900°C for 24 h were shown in Figure 3.b. It was observed that the $T_c$ and $T_0$ values at 0 T was 38 K and 33 K, respectively. Although the superconducting transition at zero magnetic field is sharp ($\Delta T = 5.6$ K), the resistive broadening in R-T with increasing the magnetic field curves was obtained, Table 1. The broadening effect is similar to that in HT$_c$ systems. Thermally activated flux flow (TAFF) model is generally used to explain such a broadening behavior in the HT$_c$ materials [7]. According to the TAFF model, flux lines are depinned with the help of thermal activation while $T_{irr} < T < T(H_{c2})$. During the experiments, the applied current ensured that only small driving forces exist in the vortex system of MgB$_2$ and therefore we believed that the TAFF theory is suitable to interpret the magnetoresistance data.
Figure 3. Temperature dependence of resistance of MgB$_2$ films at under magnetic field of 0, 2 and 4 T. MgB$_2$ films heat treated at a) 600°C for 24 h and b) 900°C for 24 h.

Table 1. Superconducting Properties of films fabricated with different heat treatment cycles

| Heating Temp.(°C) | $T_c$(K) | $T_\phi$(K) | $\Delta T$(K) | $U(H,T)/k_B$ |
|------------------|---------|------------|--------------|--------------|
|                  | 0 T     | 2 T        | 4 T          | 0 T          | 2 T        | 4 T          | 0 T          | 2 T        | 4 T          |
| 600              | 37.7    | 33.8       | 29.1         | 20.8         | 12.3       | 12.8         | 16.9         | 17.5       | 11.3         |
| 900              | 38.3    | 34.8       | 29.7         | 32.7         | 25.3       | 15.3         | 5.6          | 9.5        | 14.4         |
|                  |         |            |              |              |            |              | 220          | 111        | 79           |
|                  |         |            |              |              |            |              | 2230         | 760        | 185          |

The activation energy $U(H,T)$ plays an important role for the determination of the flux dynamics in the superconducting materials. $U(H,T)$ also reveals the height of the energy barriers of superconducting materials which control the vortex motion in the vortex liquid [8]. In the Thermally Activated Flux Flow (TAFF) region, the thermally activated resistivity is defined as [9],

$$R = R_0 \exp \left(- \frac{U(J,H,T)}{k_B T} \right)$$

(1)

where $R_0$ is the pre-exponent factor, $k_B$ the Boltzmann constant and $U$ the activation energy for the flux motion which depends on the temperature, $T$, the current density, $J$, and the applied magnetic field, $H$. At the constant current, the dependence of $U(H,T)$ is believed to give information about the dissipation mechanism. $U(H,T)$ can be given as temperature independent in the narrow temperature region of the low resistivity part. The activation energy $U(H)$, can be calculated from the slope of the plot of $\ln(R)$ versus $1/T$ as shown in figure 4. The calculated activation energy values of the films fabricated under high and low temperature heat treatment conditions are given in Table 1.

The activation energy, $U(H)$, decreased rapidly with increasing magnetic field. The variation of $U(H)$ with the applied magnetic field is explained as an increase of the magnetic field and also flux trapping. When the magnetic field was increased, the fluxes from the externally applied magnetic field penetrate into the samples and are trapped there leaving the internal field almost unchanged.
Conclusion

In this study, we have fabricated MgB$_2$ thick film on the YSZ substrate by using Spray Pyrolysis technique. The films fabricated were heat treated under low (600°C) and high temperature (900°C) conditions. The XRD and SEM analysis showed that heat treatment temperature played an important role on the crystallization and grain connections in the films. R-T measurement pointed out that the film processed at low temperature showed the broadened superconducting transition. This problem was solved with high temperature heat cycle. Activation energy, $U(H)$ was calculated using Arrhenius law. $U(H)$ decreased strongly with increasing the magnetic fields which is attributed to the flux motion.

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