Modern Physics Letters B, Vol. 20, No. 27 (2006) 1763–1769

World Scientific
www.worldscientific.com

PHASE FORMATION AND SUPERCONDUCTIVITY OF Fe-TUBE ENCAPSULATED AND VACUUM-ANNEALED MgB_2

K. P. SINGH[*,†], V. P. S. AWANA[*,§], MD. SHAHABUDDIN[†], M. HUSAIN[†],
R. B. SAXENA[*], RASHMI NIGAM[*], M. A. ANSARI[*], ANURAG GUPTA[*],
HIMANSHU NARAYAN[*,‡], S. K. HALDER[*] and H. KISHAN[*]

[*]*National Physical Laboratory, K. S. Krishnan Marg, New Delhi 110012, India*
[†]*Physics Department, Jamia Millia Islamia University, New Delhi 110025, India*
[‡]*Department of Physics and Electronics,*
National University of Lesotho Roma 180, Kingdom of Lesotho, Southern Africa
[§]*awana@mail.nplindia.ernet.in*

Received 15 September 2005

We report optimization of the synthesis parameters viz. heating temperature (T^H), and hold time (t^{hold}) for vacuum-annealed (10^{-5} Torr) and LN_2 (liquid nitrogen) quenched MgB_2 compound. These are single-phase compounds crystallizing in the hexagonal structure (space group $P_{6/mmm}$) at room temperature. Our XRD results indicated that for phase-pure MgB_2, the T^H for 10^{-5} Torr annealed and LN_2-quenched samples is 750°C. The right stoichiometry i.e., MgB_2 of the compound corresponding to 10^{-5} Torr and T^H of 750°C is found for the hold time (t^{hold}) of 2.30 hours. With varying t^{hold} from 1–4 hours at fixed T^H (750°C) and vacuum (10^{-5} Torr), the c-lattice parameter decreases first and later increases with t^{hold} (hours) before a near saturation, while the a-lattice parameter first increases and later decreases beyond a t^{hold} of 2.30 hours. The c/a ratio versus t^{hold} plot showed an inverted bell-shaped curve, touching the lowest value of 1.141, which is the reported value for perfect stoichiometry of MgB_2. The optimized stoichimetric MgB_2 compound exhibited superconductivity at 39.2 K with a transition width of 0.6 K. In conclusion, the synthesis parameters for phase pure stoichimetric vacuum-annealed MgB_2 compound are optimized and are compared with widely-reported Ta tube encapsulated samples.

Keywords: MgB_2; Fe-encapsulations; superconductivity; and vacuum synthesis.

1. Introduction

The discovery of superconductivity at 39 K in MgB_2 has attracted a lot of attention from both condensed matter experimentalists and theoreticians.[1] The appearance of superconductivity at 39 K seems to be just at the limit of BCS theory. In fact, clear evidence for the isotope effect has already given an indication that phonons play an important role in the pairing mechanism in this compound.[2] The compound had yet been studied rigorously in terms of its crystal structure, thermal and electrical conduction,[3–5] specific heat,[6,7] isotope effect[2,8] and doping.[8–10] Based on various

[§]Corresponding author.

physical property measurements, important critical parameters of the compound viz., critical superconducting temperature (T_c), coherence length (ξ), penetration depth (λ), critical current (J_c) and lower/upper critical fields H_{c_1}/H_{c2} have yet been determined and reviewed.[11,12] In fact the superconductivity of MgB_2 is much simpler in comparison to the yet mysterious HTSc (High T_c superconductivity) cuprates. In particular MgB_2 has a simpler structure, low anisotropy and larger coherence length. Most interestingly MgB_2 possess higher quality grain boundaries,[13] which permit excellent current transport. Higher quality grain boundaries and better superconducting critical parameters of MgB_2 provide an edge over HTSC cuprates. A higher T_c of 40 K and better material properties respectively in comparison to so-called inter-metallic BCS-type superconductors and HTSc cuprates makes MgB_2 a unique superconductor. Very recently an avalanche of activity had taken place in terms of high quality MgB_2 tapes and wires.[14,15] Less anisotropy and high quality grain boundaries are further improved by nano-sized particle doping. Various nano-sized dopants prefer to stay at low angle grain boundaries and thus improve dramatically the transport properties of the compound.[16,17]

Interestingly enough, though the importance of the MgB_2 superconductor is realized by both theoreticians and experimentalists, the quality of the material in terms of its phase formation, right stoichiometery, grains size/connectivity and porosity etc. still warrants improvements. Pure phase MgB_2 can be synthesized in an oxygen-free environment from 700°C to 1400°C, respectively, in vacuum (10^{-5} Torr) to high pressure of argon gas.[18,19] We report here the optimization of the synthesis parameters, viz. heating temperature (T^H), and hold time (t^{hold}) for vacuum-annealed (10^{-5} Torr) and LN_2 (liquid nitrogen) quenched Fe-tube encapsulated MgB_2 compound. The optimized stoichimetric MgB_2 compound exhibited superconductivity at 39.2 K with a transition width of 0.6 K.

2. Experimental Details

Various MgB_2 compounds were synthesized using high quality Mg and B powders, by mixing them in stoichiometeric ratio. The mixed and ground powder were further palletized. The pellets were then put in a closed end soft iron (SS) tube. The SS tube which contains the raw MgB_2 pellet was then sealed inside a quartz tube at high vacuum of 10^{-5} Torr (see Fig. 1). The encapsulated raw MgB_2 pellet is then heated at desired heating temperatures (T^H) with a hold time (T^{hold}) and is finally quenched in liquid nitrogen (LN_2). X-ray diffraction (XRD) patterns were obtained at room temperature using Cu-K_α radiation. Resistivity measurements were made in the temperature range of 12 to 300 K using a four-point-probe technique on a close cycle refrigerator (CCR).

3. Results and Discussion

Figure 2 depicts the room temperature XRD (X-ray diffraction) patterns of various MgB_2 compounds being synthesized at a fixed T^H of 750°C and different

Fig. 1. Photograph for SS–Fe tube-inserted and quartz tube-encapsulated MgB_2 raw compound.

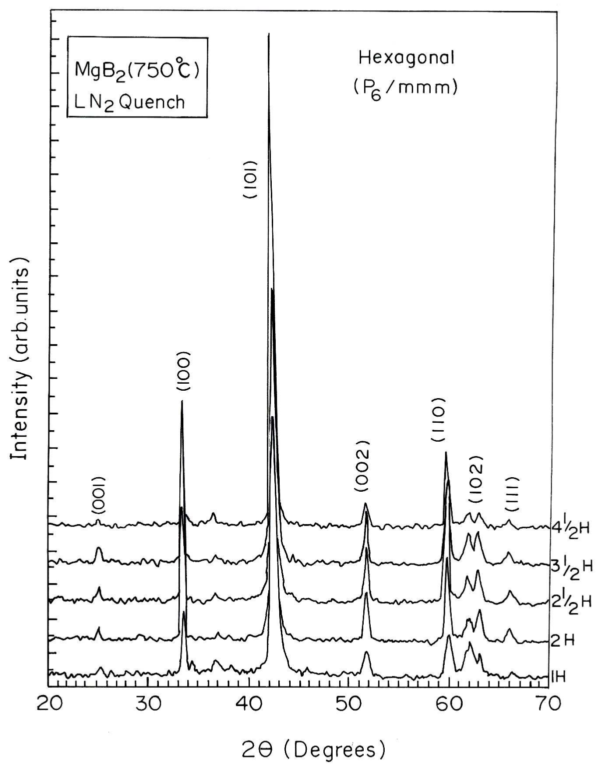

Fig. 2. X-ray diffraction patterns for 750°C and 10^{-5} Torr vacuum-annealed MgB_2 samples with different $T^{\rm hold}$.

$T^{\rm hold}$ ranging from 1 hour to 4.30 hours. All the samples crystallized in hexagonal ($P_{6/mmm}$) structure without any noticeable impurity. What is worth mentioning is the fact that for a $T^{\rm hold}$ of either less/more than 1 hour/5 hours with T^H of 750°C, the resultant material were not single-phase. In fact, a very small impurity

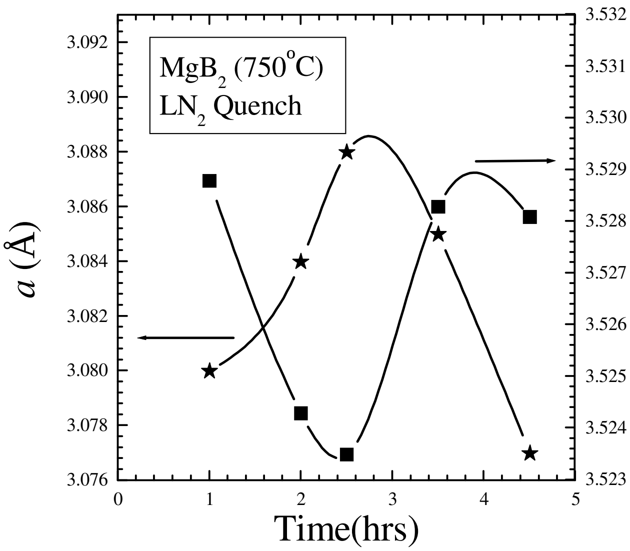

Fig. 3. Lattice parameters a and c versus T^{hold} plot for 750°C and 10^{-5} Torr vacuum-annealed MgB_2 samples.

of Mg at 2 (theta) = 37 degrees is seen for the samples having T^{hold} of either 1 hour or 4.30 hours, along with some amount of MgO at 63 degrees in all samples. Also we found that with either higher or lower T^H than 750°C, the resulting MgB_2 samples were not single-phase. What is worth mentioning is the fact that T^H of 750°C corresponds to vacuum annealing of 10^{-5} Torr. In fact, it is reported that pure phase MgB_2 can be synthesized in an oxygen-free environment from 700°C to 1400°C, respectively, in vacuum (10^{-5} Torr) to high pressure of argon gas.[18,19] Here in this article we focus our attention on the optimization of vacuum-annealed (10^{-5} Torr) MgB_2 in its single phase region.

Lattice parameters a and c for MgB_2 compounds being synthesized at a fixed T^H of 750°C and different T^{hold} ranging from 1 hour to 4.30 hours are plotted in Fig. 3. It is seen that, the c-lattice parameter decreases first and later increases with t^{hold} (hours) before a near saturation, while the a-lattice parameter first increases and later decreases beyond a t^{hold} of 2.30 hours. The c/a ratio is plotted in Fig. 4, which shows an inverted bell-shaped curve, touching the lowest value of 1.141. Interestingly, the 1.141 (c/a) value corresponds to the t^{hold} of 2.30 hours. It is reported earlier through various experiments regarding the phase formation of MgB_2 that the c/a ratio of this compound touches the value 1.414 for perfect stoichiometry of MgB_2, without any deficiency of Mg.[20] This shows that we have achieved perfect stoichiometry of MgB_2 without appreciable Mg deficiency at a T^{hold} of 2.30 hours and T^H of 750°C in a vacuum of 10^{-5} Torr.

Resistance (R) versus temperature (T) plots for 750°C and 10^{-5} Torr vacuum-annealed samples with different T^{hold} of 1 hour to over 3 hours are given in

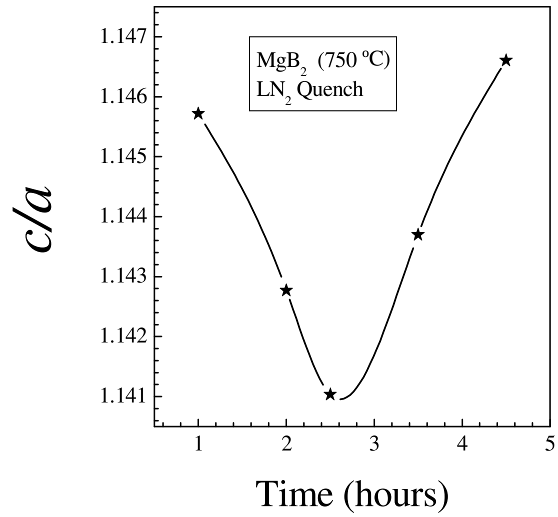

Fig. 4. c/a versus T^{hold} plot for 750°C and 10^{-5} Torr vacuum-annealed MgB_2 samples.

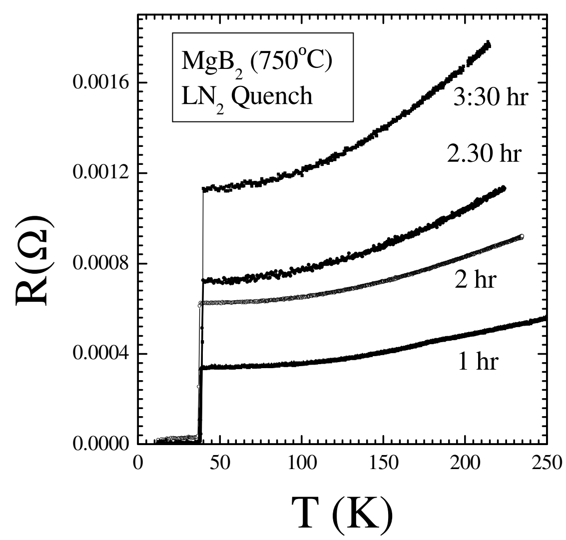

Fig. 5. $R(T)$ plots for 750°C and 10^{-5} Torr vacuum-annealed MgB_2 samples with different T^{hold}.

Fig. 5. Generally speaking, all the samples are metallic with their superconducting transition temperature ($T_c^{R=0}$) in the range of 37 K to 39.2 K. The optimum superconductivity is achieved for a T^{hold} of 2.5 hours with $T_c^{R=0}$ of 39.2 K and transition width ($T_c^{\text{onset}} - T_c^{R=0}$) of around 0.6 K.

As far as normal state (above T_c^{onset}) behavior is concerned, it is in general agreement with reported data on MgB_2 polycrystalline samples. The $R(T)$ has a near constant metallic slope down to say 100 K, and later it seems to follow the power law with a much less positive slope. The ratio of resistance (RR), which is generally defined as $R_{300}/R_{\rm onset}$ is close to 3.0. In literature, the RR has been found up to above 16.0 in good quality MgB_2 samples.[11,21] RR is found to be lower for disordered samples, for example in $MgB_{2-x}C_x$ compounds, the RR comes down to say 1.5.[21] It seems that though we achieved phase pure stoichiometric MgB_2 as indicated by a c/a ratio of 1.141 for the optimized sample, the disorder being created by a possible Fe inclusion in the material cannot be ignored. Generally, the encapsulation done in the Ta tube instead of the SS–Fe tube helps in achieving the better quality of MgB_2. Our results indicate that SS–Fe encapsulation gives rise to the Fe-induced disorder in MgB_2 and hence is not the right choice for attaining disorder-free MgB_2. However, the present disorder induced pinned samples could be the right choice for high critical current (J_c) applications.[22]

In conclusion, the synthesis parameters for phase pure stoichimetric vacuum-annealed MgB_2 compound are optimized and its superconducting properties are compared with the widely reported Ta tube-encapsulated samples.

Acknowledgment

Authors from the National Physical Laboratory appreciate the interest and advice of Professor Vikram Kumar (Director) in the present work.

References

1. J. Nagamatsu, N. Nakagawa, T. Muranaka and J. Akimitsu, *Nature* **410** (2001) 63.
2. S. L. Budko, G. Lapertot, C. Petrovic, C. E. Cunninghan, N. Andersen and P. C. Canfield, *Phys. Rev. Lett.* **86** (2001) 1877.
3. B. Lorenz, R. L. Meng, Y. Y. Xue and C. W. Chu, *Phys. Rev. B* **64** (2001) 52513.
4. J. S. Ahn, E. S. Choi, W. Kang, D. J. Singh, M. Han and E. J. Choi, *Phys. Rev. B* **65** (2002) 214534.
5. P. A. Sharma, N. Hur, Y. Horibe, C. H. Chen, B. G. Kim, S. Guha, M. Z. Cieplak and S.-W. Cheong, *Phys. Rev. Lett.* **89** (2002) 167003.
6. F. Bouquet, R. A. Fisher, N. E. Phillips, D. G. Hinks and J. D. Jorgensen, *Phys. Rev. Lett.* **87** (2001) 47001.
7. F. Bouquet, Y. Wang, I. Sheikin, T. Plackowski and A. Junod, *Phys. Rev. Lett.* **89** (2002) 257001.
8. D. G. Hinks, H. Claus and J. D. Jorgensen, *Nature* **411** (2001) 457.
9. W. Mickelson, J. Cunings, W. Q. Han and A. Zetti, *Phys. Rev. B* **65** (2002) 52505.
10. A. Tampieri, G. Celotti, S. Sprio, D. Rinaldi, G. Banicca and R. Caciuffo, *Solid State Commun.* **121** (2002) 497.
11. D. K. Finnemore, J. E. Ostensen, S. L. Budko, G. Lapertot and P. C. Canfield, *Phys. Rev. Lett.* **86** (2001) 2420.
12. O. F. de Lima, R. A. Ribeiro, M. A. Avila, C. A. Cardoso and A. A. Coelho, *Phys. Rev. Lett.* **86** (2001) 5974.
13. S. B. Samanta, H. Narayan, A. Gupta, A. V. Narlikar, T. Muranaka and J. Akimitsu, *Phys. Rev. B* **65** (2002) 92510.
14. R. Flukiger, H. L. Suo, N. Musolino, C. Beneduce, P. Toulemonde and P. Lezza, *Physica C* **385** (2003) 286.

15. K. Q. Ruan, H. L. Li, Y. Yu, C. Y. Wang, L. Z. Cao, C. F. Liu, S. J. Du, G. Yan, Y. Feng, X. Wu, J. R. Wang, X. H. Liu, P. X. Zhang, X. Z. Wu and L. Zhou, *Physica C* **386** (2003) 578.
16. S. Soltanian, M. J. Qin, S. Keshvarazi, X. L. Wang and S. X. Dou, *Phys. Rev. B* **68** (2003) 134509.
17. G. J. Xu, J.-C. Grivel, A. B. Abrahamsen and N. H. Andersen, *Physica C* **406** (2004) 95.
18. C. M. Franco, B. Ferreira, C. A. M. Santos, L. Ghiveldar, H. J. I. Filho and A. J. S. Machado, *Physica C* **408** (2004) 130.
19. N. N. Kolesnikov and M. P. Kulakov, *Physica C* **363** (2002) 166.
20. D. G. Hinks, J. D. Jorgensen, H. Zheng and S. Short, *Physica C* **382** (2002) 166.
21. A. Bharathi, S. J. Balaselvi, S. Kalavathi, G. L. N. Reddy, V. S. Sastry, Y. Hariharan and T. S. Radhakrishnan *Physica C* **370** (2002) 211.
22. V. P. S. Awana, R. Rawat, A. Gupta, M. Isobe, K. P. Singh, A. Vajpayee, H. Kishan, E. Takayama-Muromachi and A. V. Narlikar, *Solid State Communications* **139** (2006) 306.