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Compatible materials with $YBa_2Cu_3O_Y$

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High-temperature superconducting materials have been investigated to make thin films for device applications. Crystallization and oxygen annealing up to 900 °C is necessary to obtain a superconductivity in $YBa_2Cu_3O_Y$ thin film. $YBa_2Cu_3O_Y$ is gradually hydrolysed in air [1]. Its overcoating is required to prevent the hygroscopic nature. The coating materials should be non-reactive with $YBa_2Cu_3O_Y$ in these high-temperature treatments. They should also be transparent to visible and infrared light in optical sensitive device applications. Non-reactive materials are necessary also as substrates.

Most of the simple oxides (e.g. SiO_2, ZrO_2 and Al_2O_3) form reaction products with $YBa_2Cu_3O_Y$ [2, 3]. The crystal orientation of $YBa_2Cu_3O_Y$ can be epitaxially controlled on $SrTiO_3$ substrate. However, its transition to superconductor is not sharp after high-temperature annealing, due to a reaction with the substrate [4]. Most of the reaction products with these substrates are compounds containing Ba, which diffuse from $YBa_2Cu_3O_Y$ [5]. These reaction products may be stable against further reactions with $YBa_2Cu_3O_Y$. BaF_2 has been used as a flux for crystal growth [6]. Alkaline-earth fluorides may also be stable against $YBa_2Cu_3O_Y$.

In the present investigation, reactions with $YBa_2Cu_3O_Y$ powder were studied on alkaline-earth fluorides, MgF_2, CaF_2 and BaF_2, and on Ba compounds, Ba_2TiO_4, $BaAl_2O_4$, $BaZrO_3$ and $Ba_3Zr_2O_7$, to find possible candidates for non-reactive overcoating and substrate. Powders of these materials are white and their dense body may be transparent to visible light.

$YBa_2Cu_3O_Y$ was prepared by firing a mixture of Y_2O_3, $BaCO_3$ and CuO at 900 °C for 3 h. After grinding it was further heated at 940 °C for 5 h. The product was mixed well with 5 wt % of the candidate materials, i.e. the alkaline-earth fluorides and the Ba compounds. The mixtures were heated at 900 °C for 3 h and for a further 9 h. Powder X-ray diffraction patterns were compared in each firing step to study the reactivity with these candidates. The electrical conductivity was measured for the samples sintered at 920 °C for 7 h using the four-point probe method.

The product heated with MgF_2 at 900 °C for 3 h was contaminated with BaF_2, Y_2BaCuO_5, CuO and MgO as shown in Fig. 1a. Relative X-ray intensities between these components were almost the same

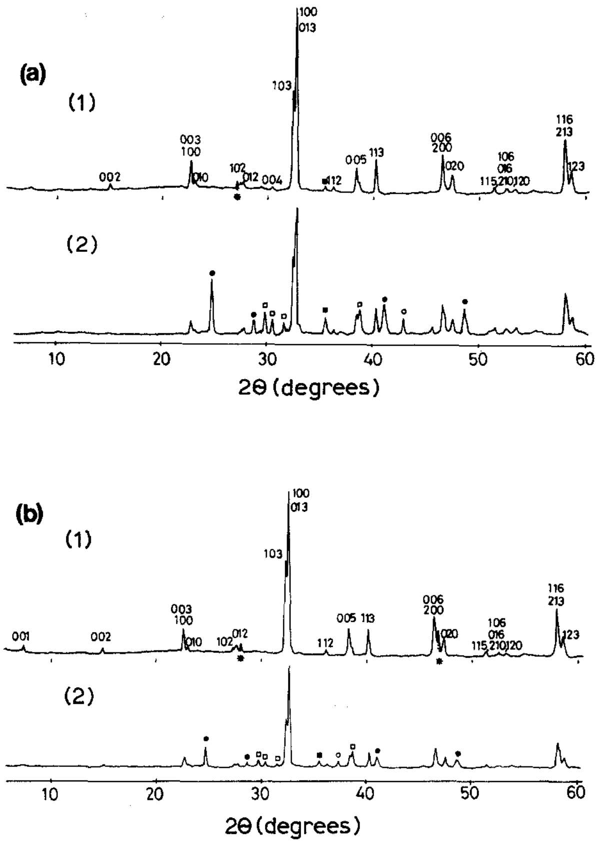

Figure 1 Powder X-ray diffraction patterns (CuK_α radiation) for the mixture of (1) $YBa_2Cu_3O_Y$ with (a) MgF_2 and (2) their reaction products. Similar patterns are also shown for (b) CaF_2. (*, ▧) CaF_2, (●) BaF_2, (□) Y_2BaCuO_5, (■) CuO and (○) CaO.

even after further heating at 900 °C for a further 9 h. The starting $YBa_2Cu_3O_Y$ powder had a small amount of CuO. The above-mentioned contaminants except for CuO were formed by a reaction between $YBa_2Cu_3O_Y$ and MgF_2. The products with CaF_2 were also mixtures of $YBa_2Cu_3O_Y$, BaF_2, Y_2BaCuO_5, CuO and CaO as depicted in Fig. 1b. The situation is quite similar to the above-mentioned case for the heated products with MgF_2. No marked reaction was observed on X-ray diffraction of the products with BaF_2. Ba_2TiO_4 disappeared with the heating and any reaction products were observed on X-ray diffraction. There have been no reports on the presence of compounds containing more barium than Ba_2TiO_4 in a BaO–TiO_2 binary. The compound might form a solid solution with $YBa_2Cu_3O_Y$ but no remarkable change was detected on X-ray diffraction. Any other barium compounds, $BaAl_2O_4$, $BaZrO_3$ and $Ba_3Zr_2O_7$, showed no obvious reactions in their heated products.

The electrical conductivity was measured on sintered bodies of the reaction products with BaF_2, $BaAl_2O_4$ and $BaZrO_3$. $YBa_2Cu_3O_Y$ had a resistivity of 5.5×10^{-3} Ω cm at room temperature, a metallic temperature dependence and a sharp superconducting transition (T_c (0) = 91 K) as shown in Fig. 2. The reaction products with the barium compounds had resistivities around 10^{-2} Ω cm at room temperature and semiconducting temperature dependences due to the weak link between the $YBa_2Cu_3O_Y$ grains. The products with BaF_2 and $BaZrO_3$ showed a relatively sharp superconducting transition and $T_c(0) \approx 82$ K. The product with $BaAl_2O_4$ showed a very broad transition to superconductor and $T_c(0) \approx 69$ K. The transition temperature at zero resistance has been reported to be reduced from 90 to 50 K with increasing x in $(Al_xY_{1-x})Ba_2Cu_3O_{6.5+\delta}$ [7]. X-ray diffraction of the product did not show any marked change in the present investigation.

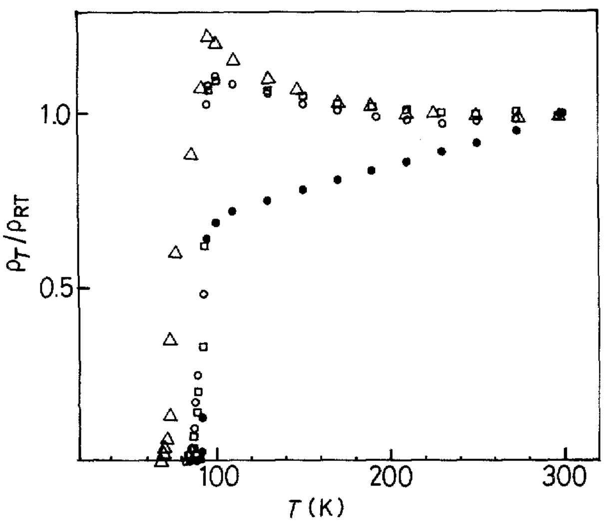

Figure 2 Superconductivities of $YBa_2Cu_3O_Y$ itself and its mixtures with BaF_2, $BaAl_2O_4$ and $BaZrO_3$.

Symbol	Sample	T_c (0)(K)	ρ_{RT} (Ω cm)
●	$YBa_2Cu_3O_Y$	91	5.5×10^{-3}
○	BaF_2 5 wt % addition	82	1.5×10^{-2}
△	$BaAl_2O_4$ 5 wt % addition	68.5	1.2×10^{-2}
□	$BaZrO_3$ 5 wt % addition	82.5	2.4×10^{-2}

In summary, BaF_2 and $BaZrO_3$ are promising candidates as overcoating and substrate materials. $BaZrO_3$ has a perovskite-type crystal structure and both an epitaxity with $YBa_2Cu_3O_Y$ and also an oxygen diffusivity can be expected. The resistivity for humidity and possibility for oxygen annealing have to be further studied for an overcoating application.

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References

1. M. F. YAN, R. L. BARNS, H. M. O'BRYAN, JR, P. K.GALLAGHER, R. C. SHERWOOD and S. JIN, *Appl. Phys. Lett.* **51** (1987) 532.
2. H. KOINUMA, K. FUKUDA, T. HASHIMOTO and K. FUEKI, *Jpn. J. Appl. Phys.* **27** (1988) L1216.
3. T. KOMATSU, O. TANAKA, K. MATUSHITA, M. TANAKA and T. YAMASHITA, *ibid.* **27** (1988) L1025.
4. X. D. WU, D. DIJIKKAMP, S. B. OGALE, A. INAM, E. W. CHASE, P. F. MICELI, C. C. CHANG, J. M. TARASCON and T. VENKATESAN, *Appl. Phys. Lett.* **51** (1987) 861.
5. D. DIJIKKAMP, T. VENKATESAN, X. D. WU, S. A. SHAHEEN, N. JISRAWI, Y. H. MIN-LEE, W. L. McLEAN and M. CROFT, *ibid.* **51** (1987) 619.
6. B. R. PAMPLIN, in "Crystal growth" (Pergamon Press, Oxford, 1975) p. 268.
7. J. P. FRANCK, J. JUNG and M. A.-K. MOHAMED, *Phys. Rev.* **B36** (1987) 2308.

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